Refinement of dietary exposure assessment using food supply chain information

Dissertation

zur Erlangung des Grades eines Doktors der Naturwissenschaften – Doctor rerum naturalium (Dr. rer. nat.) –

> vorgelegt dem Department Sport und Gesundheit der Fakultät für Naturwissenschaften der Universität Paderborn

> > von Carolin Fechner

Paderborn, April 2020

Gutachter: Prof. Dr. Helmut Heseker (Universität Paderborn) Prof. Dr. Matthias Greiner (Bundesinstitut für Risikobewertung, Berlin und Stiftung Tierärztliche Hochschule Hannover)

Disputation: 25.09.2020

Eidesstattliche Erklärung

(gemäß § 10 der Promotionsordnung der Fakultät für Naturwissenschaften der Universität Paderborn)

Hiermit erkläre ich, dass ich die eingereichte Dissertation zum Thema "Refinement of dietary exposure assessment using food supply chain information" selbständig verfasst habe. Alle genutzten Quellen und Hilfsmittel wurden in Zitaten angegeben.

Ich habe gleichzeitig an keiner anderen Stelle eine Eröffnung des Promotionsverfahrens beantragt. Die vorliegende Dissertation wurde von keinem anderen Fachbereich bzw. keiner anderen Fakultät zurückgewiesen.

Declaration of academic honesty

(according to § 10 of the doctoral regulations of the Faculty of Science of the Paderborn University)

I hereby declare that I have prepared the submitted dissertation on the topic "Refinement of dietary exposure assessment using food supply chain information" independently. All references or aids used are stated in citations.

I have not applied for the initiation of the doctoral procedure at another official place simultaneously. The present dissertation has not been rejected by another department or another faculty.

Paderborn, 30.04.2020

Carolin Fechner

Acknowledgements

I would like to thank all persons, who supported me in the progress of my work, as they provided data and resources for the research, they spent time to discuss ideas with me and to proof-read manuscripts.

Since 2016, I had the chance to work on approaches to the refinement of dietary exposure assessment using food supply chain information. The project was issued by the German Federal Institute for Risk Assessment (BfR), Department Exposure, Unit Dietary Exposure and Aggregated Exposure. The BfR doctoral programme provided the opportunity to conduct this research and gain additional methodological skills. My doctoral project matched perfectly with my experiences and interests, as I studied Ecotrophology with focus on food technology and afterwards, I gained practical experience in food production. The work related to dietary exposure assessment gave me the chance to combine my experiences and interests in the food sector in an interdisciplinary context and gain new abilities.

Prof. Dr. Matthias Greiner of the BfR and the University of Veterinary Medicine Hannover, Foundation and Dr. Oliver Lindtner of the BfR initiated the doctoral project and had the idea to consider dietary exposure assessment in relation to global food supply. I am deeply grateful that they gave the chance to me to work on this interdisciplinary project. I would like to thank Prof. Dr. Matthias Greiner for the project administration, the continuous supervision offered and the preparation of the second opinion on the dissertation. I thank Dr. Oliver Lindtner who provided his expertise in dietary exposure assessment, statistics and project implementation, offered continuous supervision and helped me with his experience to develop new approaches.

I convey my thanks to Prof. Dr. Helmut Heseker of the Paderborn University, who offered university cooperation, invested time in the project and supported the work. Prof. Dr. Helmut Heseker was my doctoral adviser giving continuous supervision and being responsible for the project administration at the university.

During my doctoral project I spent one year at the Norwegian Scientific Committee for Food and Environment (VKM) in Oslo financed by the European Food Safety Authority (EFSA) as part of the European Food Risk Assessment fellowship programme (EU-FORA) 2018/2019. I would like to thank Dr. Inger Therese L. Lillegaard, Dr. Gro Haarklou Mathisen and Gisle Solstad of the VKM, Dr. Oliver Lindtner of the BfR, Dr. Sylvia Frantzen of the Institute of Marine Research in Norway (IMR), and Prof. Dr. Lene Frost Andersen of the University of Oslo (UiO) for giving me the chance to cooperate with different institutes and gain experience in dietary exposure assessment using Norwegian and German data.

Finally, I would like to thank my family and friends, who always supported me during my doctoral project with their views and input. Special thanks go to Catherine Murray-Küfner and Catrin Piotter for English language support.

Table of Contents

Eidesstattliche ErklärungII
Declaration of academic honesty II
AcknowledgementsIII
Table of ContentsIV
Abbreviations
List of tablesVII
List of figures
Abstract1
Kurzfassung2
Summary
1 Introduction
1.1 Dietary exposure assessment and influence of global food supply7
1.2 Objectives
2 Publications
2.1 Food Origin Information in Times of Global Food Supply – Basis for the Refinement of
Dietary Exposure Assessment12
2.2 Refinement of dietary exposure assessment using origin-related scenarios
2.3 Dietary exposure assessment of aluminium and cadmium from cocoa in relation to cocoa origin
2.4 Influence of the geographical origin on substance concentrations in herring as basis for dietary exposure assessments
2.5 Dioxins and dioxin-like PCBs in Atlantic herring: A Norwegian – German comparison of the influence of fishing area on country-specific dietary exposure
3 Methods and data
3.1 Data sources
3.2 Selection of case studies19
3.3 Dietary exposure
3.4 Statistical analyses and evaluations23

4 Results	24
4.1 Information on the geographical origin of food	24
4.2 Dietary exposure and geographical origin of food	26
4.3 Country-specific dietary exposure	27
5 Discussion	31
5.1 Dietary exposure and geographical origin of food	31
5.2 Country-specific dietary exposure	32
5.3 Data limitations, uncertainties and future requirements	33
6 Conclusion and future challenges	36
References	VIII
Appendix: Poster	. XIV
Poster 1	. XIV
Poster 2	. XVI
Poster 3	XVIII

Abbreviations

ADI	Acceptable daily intake
BfR	Bundesinstitut für Risikobewertung
	(German Federal Institute for Risk Assessment)
BSE	Bovine spongiform encephalopathy
BVL	Bundesamt für Verbraucherschutz und Lebensmittelsicherheit
	(Federal Office of Consumer Protection and Food Safety)
BW	Body weight
DL-PCBs	Dioxin-like polychlorinated biphenyls
EFSA	European Food Safety Authority
EU-FORA	European Food Risk Assessment fellowship programme
FAO	Food and Agriculture Organization of the United Nations
IMR	Institute of Marine Research in Norway (Havforskningsinstituttet (HI))
LB	Lower bound
LOD	Limit of detection
LOQ	Limit of quantification
Mintel GNPD	Mintel Global New Product Database
MLB	Modified lower bound
MRI	Max Rubner-Institut
	(Bundesforschungsinstitut für Ernährung und Lebensmittel / Federal
	Research Institute of Nutrition and Food)
n	Sample number
NVS II	German National Nutrition Survey II
P50	50 th percentile
P95	95 th percentile
PCDDs	Polychlorinated dibenzo-p-dioxins
PCDFs	Polychlorinated dibenzofurans
TEQ	Toxic Equivalents
TWI	Tolerable weekly intake
UB	Upper bound
UiO	Universitetet i Oslo (University of Oslo)
UN	United Nations
VKM	Vitenskapskomiteen for mat og miljø
	(Norwegian Scientific Committee for Food and Environment)
WHO	World Health Organization

List of tables

Table 1: Data sources used for dietary exposure assessment	17
Table 2: Data sources used to extract additional information on the geographical origin of f	ood
	. 18
Table 3: Standard scenarios $(1 - 4)$ and origin-related scenarios $(5 - 8)$ used to evalu	Jate
dietary exposure for the case studies selected	21

List of figures

Figure 1: Standard scenarios for deterministic dietary exposure assessment (Sarvan et al.,
2017) (left) and origin-related scenarios (right). Same colour shows corresponding scenarios
(standard (1–4) and origin-related (5–8)). While standard scenarios use the 95 th percentile for
high concentrations, origin-related scenarios use mean concentrations of different origins
(Fechner <i>et al.</i> , 2019c)9
Figure 2: Potential influences within the food supply chain on substance concentration and
dietary exposure (Fechner <i>et al.</i> , 2019d)11
Figure 3: Excerpt of data for food consumption, substance concentrations and resulting chronic
dietary exposure for the case studies selected

Abstract

Food supply is organised globally and the geographical origin of food could influence substance concentrations. Consumption and concentration data were used to assess the dietary exposure to substances in standard scenarios. Additional resources gave further food origin information which was used for origin-related scenarios to refine exposure estimates. The impact of origin was not covered by standard scenarios for bromide from tomatoes, as the origin-related scenario based on high tomato consumers and higher concentrations in Italian tomatoes resulted in the highest exposure of 0.015 mg/day/kg BW. However, origin influences were covered by standard scenarios, as these resulted in the highest estimate based on high consumers and high concentrations for the case studies ethephon from pineapples (0.9 µg/day/kg BW), aluminium from kiwifruits (0.02 mg/week/kg BW), aluminium and cadmium from cocoa powder (0.15 mg/week/kg BW and 0.4 µg/week/kg BW). German high consumers of herring were exposed to the total sum of dioxins and dioxin-like polychlorinated biphenyls with 26.5 pg WHO-2005-TEQ/kg BW/week based on Baltic Sea samples while Norwegian high consumers had a lower exposure of 6.5 pg WHO-2005-TEQ/kg BW/week based on Norwegian Sea samples. Considering the geographical food origin refined exposure estimates for selected case studies but more data are needed for future improvements.

Kurzfassung

Lebensmittelversorgung erfolgt global und die geografische Lebensmittelherkunft kann den Stoffgehalt beeinflussen. Verzehrs- und Gehaltsdaten wurden genutzt, um in Standardszenarien die Exposition zu Stoffen über Lebensmittel zu bewerten. Zusätzliche Quellen gaben weitere Informationen zur Lebensmittelherkunft, was für Herkunftsszenarien genutzt wurde, um Expositionsschätzungen zu verfeinern. Der Herkunftseinfluss wurde nicht von Standardszenarien abgedeckt für Bromid aus Tomaten, da das Herkunftsszenario beruhend auf Tomatenvielverzehrern und höheren Gehalten aus italienischen Tomaten die höchste Exposition von 0.015 mg/Tag/kg KG ergab. Jedoch wurden Herkunftseinflüsse auch in Standardszenarien abgedeckt, da diese zur höchsten Schätzung führten basierend auf Vielverzehrern und hohen Gehalten für die Fallstudien Ethephon aus Ananas (0.9 µg/Tag/kg KG), Aluminium aus Kiwi (0.02 mg/Woche/kg KG), Aluminium und Cadmium aus Kakaopulver (0.15 mg/Woche/kg KG and 0.4 µg/Woche/kg KG). Deutsche Heringvielverzehrer waren der Summe an Dioxinen und dioxinähnlichen polychlorierten Biphenylen mit 26.5 pg WHO-2005-TEQ/kg KG/Woche ausgesetzt basierend auf Ostseeproben, während norwegische Hochverzehrer eine niedrigere Exposition von 6.5 pg WHO-2005-TEQ/kg KG/Woche hatten basierend auf Proben der Norwegischen See. Die Betrachtung der geografischen Lebensmittelherkunft verfeinerte Expositionsschätzungen für gewählte Fallstudien aber mehr Daten sind nötig für zukünftige Verbesserungen.

Summary

Background

Food risk assessments are the basis for the risk management. In risk assessments, the dietary exposure assessment is an important part to evaluate the consumers' intake of substances. Global trade and food supply are organised in a complex network, which enables the consumption of food from various geographical origins. Besides many other influences, the geographical origin of food may be related to the content of substances like contaminants in specific food items. If the consumer focuses specific geographical origins when purchasing food, origin-related substance concentrations may influence dietary exposure assessments and have increasing or decreasing effects. Including the geographical origin of food in dietary exposure assessments may refine estimates, reduce uncertainties and be an important factor in the evaluation of food safety.

Objectives

The objective of the current dissertation was to develop approaches to refine dietary exposure assessment by integrating information on the geographical origin of food in the analysis of the chronic dietary exposure. It should be considered if consumption and concentration data as typical data sources for dietary exposure assessment could be used to extract geographical origin information. It should be evaluated if additional data sources could complement the information on the market situation related to the geographical origin of food.

Methods

Geographical origin information provided in food monitoring data was checked for selected agricultural products and processed food items, and Food and Agriculture Organization of the United Nations (FAO) data were used to check the credibility of origin indications in food monitoring data (chapter 2.1). Based on geographical origin information and information on variable substance concentrations available in food monitoring data, bromide from tomatoes, ethephon from pineapples and aluminium from kiwifruits (chapter 2.2), aluminium and cadmium from cocoa powder (chapter 2.3) as well as polychlorinated dibenzo-p-dioxins (PCDDs)/polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (DL-PCBs) from Atlantic herring (*Clupea harengus*) (chapters 2.4 and 2.5) were selected as case studies to refine dietary exposure assessment using information on the geographical origin of food.

Country-specific food consumption amounts and country-specific data on substance concentrations in food were combined in standard scenarios to evaluate the substance intake without consideration of the geographical origin. In standard scenarios, average or high food

consumption as well as average or high concentrations in food were considered. Standard scenarios were compared to additional origin-related scenarios. Origin-related scenarios used origin information from food monitoring data to group lower mean concentrations attributed to Origin A and higher mean concentrations attributed to Origin B. For cocoa powder, plausible origin information was missing in food monitoring data and concentrations were grouped by a sensitivity analysis instead, as origin relations in aluminium and cadmium concentrations were included in evaluations by using specific approaches. These approaches were based on so-called lower bounds (LB), modified lower bounds (MLB) or upper bounds (UB). Therefore, non-quantified values were replaced by zero or by limits reported in food monitoring data. For exposure estimates shown here, MLB or LB concentrations were used, as for the substances considered, most of the concentrations were quantified and differences to UB concentrations were minor. Exposure estimates were conducted for consumers of the food items and, in the case of herring, all participants of the country-specific consumption surveys were integrated.

The UN Comtrade database, the Eurostat database and the Mintel Global New Product Database (GNPD) were used to generate additional information for cocoa products and herring to evaluate the market situation on the geographical origin of food.

Results

The available origin information in food monitoring data was clearly limited by food labelling and reporting (chapter 2.1). The availability of origin data differed markedly among different food items and unspecified origin information masked more details, as then specific origins were not visible. The check with FAO data showed, that food monitoring data provided sufficient origin information to be used for origin-related dietary exposure assessment for the agricultural products tomatoes, pineapples and kiwifruits as well as for olive oil and herring. However, origin data for cocoa powder, tomato juice and apple juice, meat cuts and tuna were limited in food monitoring data, as information was unspecific or implausible and not suitable for further investigations. In this case, FAO data were not suitable to fully evaluate the credibility of origin information in food monitoring data. As only for a few food items origin information was sufficient in food monitoring also cocoa powder was selected later on for further investigations as example with insufficient origin data.

Dietary exposure was evaluated for selected case studies and showed specific origin-related exposure situations. For bromide from tomatoes, the most conservative origin-related scenario 8 based on Italian tomatoes with higher mean bromide concentrations (identified Origin B) and high consumption amounts resulted in the highest exposure of 0.015 mg/day/kg BW (chapter 2.2). The impact of origin on exposure was not covered by the conservative standard scenario 4 based on high concentrations and high consumption (0.006 mg/day/kg BW). For ethephon

4

from pineapples, the highest intake estimate was obtained with the conservative standard scenario 4 resulting in $0.9 \,\mu$ g/day/kg BW and based on high concentrations and high consumption (chapter 2.2). The same applied to aluminium from kiwifruits (chapter 2.2), as well as aluminium and cadmium from cocoa powder (chapter 2.3), as the highest estimates were also obtained in the conservative standard scenario 4 and amounted to 0.02 mg/week/kg BW, as well as 0.15 mg/week/kg BW and 0.4 μ g/week/kg BW. Here, standard scenarios covered influences of origin estimated in origin-related scenarios and were sufficient to include possible consumption situations in a conservative standard estimate.

The exposure to PCDD/Fs and DL-PCBs from herring was estimated for Norway and Germany using country-specific data (chapters 2.4 and 2.5). Using disaggregated consumption amounts and LB concentrations, highest intake estimates for the total sum of 17 PCDD/Fs and 12 DL-PCBs from herring were estimated for consumers of herring. Estimates for Norwegian consumers of herring were clearly lower than for German consumers of herring. For Norway, 6.5 pg WHO-2005-TEQ/kg BW per week was estimated in the origin-related scenario 6, which was based on Norwegian Sea samples and high consumption. For Germany, 26.5 pg WHO-2005-TEQ/kg BW per week was derived as highest estimate in the origin-related scenario 8, which was based on Baltic Sea samples and high consumption. The higher substance intake for German consumers of herring was related to higher Baltic Sea concentrations and higher consumption amounts in comparison to Norwegian consumers of herring. However, when considering all participants in the consumption surveys, exposure estimates were low for Germany and for Norway and origin relations were less relevant. This is related to the low number of consumers of herring in the population, as less than 6 % of the participants in consumption surveys consumed herring.

Additional data from trade databases and the Mintel GNPD gave more origin information for specific food items, which were considered there because of a lack of information in food monitoring. Cocoa products on the German market originated mainly from Côte d'Ivoire, high amounts were traded via the Netherlands, and origin indications on the packaging of cocoa powder showed different geographical origins, but were also limited to a few packages (chapter 2.3). Missing information for cocoa powder in food monitoring data was complemented using additional data sources, which helped to evaluate the exposure estimates in origin-related scenarios. In the case of herring, it was also possible to generate additional information from trade data and the Mintel GNPD (chapter 2.5). According to trade data, herring was imported to Germany and Norway, and the labels of packaged herring in the Mintel GNPD showed additional fishing areas other than the Baltic Sea for Germany and the Norwegian Sea for Norway. Consequently, herring from other fishing areas is relevant for the two target markets Germany and Norway.

Discussion

The integration of geographical origins into exposure scenarios displayed different exposure situations, which was shown for the case studies conducted. Food can originate from various geographical regions because of global trade. Integrating geographical origin information of food into dietary exposure assessments is relevant, if a) substance concentrations vary geographically, b) food items are supplied from different geographical origins to the target market and c) consumers focus on specific geographical origins of food at the purchase. More data on geographical origins of food, supply channels and consumer behaviour are required to assess the need of origin-related exposure assessment for individual food items and specific substances in general and to go beyond particular case studies. Different outcomes of originrelated exposure scenarios were shown by the current research. On the one hand, it is possible that standard scenarios do not cover influences of origin and consequently, origin-related scenarios are needed to derive the most conservative estimate. On the other hand, originrelated scenarios might provide a refined lower estimate than standard scenarios, which could be of special interest in cases, in which health-based guidance values are exceeded. The refinement of country-specific dietary exposure assessments to undesired substances integrating relevant geographical information could be especially relevant in future European risk assessments.

1 Introduction

1.1 Dietary exposure assessment and influence of global food supply

To evaluate the food safety, risk assessments are carried out as basis for risk management decisions. Dietary exposure assessment is an important part of food risk assessment, which evaluates the consumers' intake of substances via food and drinking water (Lindtner *et al.*, 2013; Sarvan *et al.*, 2017; WHO & FAO, 2009b). Typically, the exposure to undesired substances like environmental contaminants or pesticide residues is evaluated as focused in the current research, but the intake of nutrients could also be an object under investigation in dietary exposure assessment. Knowledge on the exposure to undesired substances is required to give characterisation of risks comparing the exposure level to health-based guidance values (WHO & FAO, 2009a). These values describe substance concentrations considered as safe for human consumption (WHO & FAO, 2009a).

For evaluating dietary exposure, data on food consumption and data on substance concentrations in food are combined (WHO & FAO, 2009b). Representative consumption data are generated in national surveys such as the German National Nutrition Survey II (NVS II) (Brombach et al., 2006; Krems et al., 2006; Sarvan et al., 2017; WHO & FAO, 2009b) or the Norwegian Norkost 3 (Totland et al., 2012). Different methods like the dietary history interview, 24-h recalls or food records are used to determine the short-term or the long-term food consumption for defined population groups (Brombach et al., 2006; Krems et al., 2006; Sarvan et al., 2017; Totland et al., 2012; WHO & FAO, 2009b). Data on concentrations of substances like contaminants in food are generated in national food monitoring programmes for unprocessed food items, and food composition databases exist for the nutritional content, while total diet studies provide information on mean substance concentrations in processed food, which can be used for dietary exposure assessment (Lindtner et al., 2013; Sarvan et al., 2017; WHO & FAO, 2009b). Standard exposure scenarios are used to model different consumption situations and evaluate the acute or chronic substance intake. Summarised in four standard scenarios, average or high food consumption as well as average or high substance concentrations in food are usually considered in different combinations (Sarvan et al., 2017). This is a deterministic evaluation, which means the estimates are based on statistic parameters like the mean of consumption and concentration data. The standard scenarios can be supplemented by scenarios related to the geographical origin used for the current research (Figure 1). A brand-loyal consumer behaviour is modelled in standard scenarios considering higher substance concentrations, as people who always consume processed food items of a specific brand containing higher substance concentrations could have a higher exposure (Sarvan et al., 2017; Tennant & Bruyninckx, 2018; WHO & FAO, 2009b). In this way, influences on substance concentrations linked to food production are included in considerations, but not all standard scenarios are performed for all cases every time (Sarvan *et al.*, 2017). The geographical origin of food could be considered in a similar way as done for brand loyalty, as lower and higher concentrations from specific geographical origins could be integrated in scenarios. Besides deterministic modelling, probabilistic evaluations using distributions of consumption and concentration data instead of statistical parameters are applied to refine dietary exposure assessment (Sarvan *et al.*, 2017; WHO & FAO, 2009b). The European Food Safety Authority (EFSA) focusses on the European situation of substance intakes using country-specific consumption data and pooled European substance concentrations to display regionally different exposure situations (EFSA, European Food Safety Authority, 2011).

The occurrence of the Bovine Spongiform Encephalopathy (BSE) in the 1990s caused a food crisis in Europe, which led to an improvement of the feed and food safety legislation introducing the concept of complete traceability of food in the supply chain to enable tracing affected foods in future crisis situations and react rapidly (Aruoma, 2006; Brambilla, 2019; Eur. Parliament & the Council, 2000, 2002). Thus, the geographical origin of food was recognised as an important influence on substance concentrations in food crisis situations and the tracking of the origin of foods was introduced (Aruoma, 2006; Beulens et al., 2005; Brambilla, 2019; EFSA, European Food Safety Authority et al., 2018b; Eur. Parliament & the Council, 2000, 2002; Weiser et al., 2016; Xiu & Klein, 2010). However, in times of global supply and networking, food items and raw materials for food production are sourced globally (Aruoma, 2006; FAO, 2017) and substance concentrations could generally vary related to the geographical origin of food. For example, a general geographical variation was observed for aluminium and cadmium in cocoa products (Abt et al., 2018; BfR, 2007; Kruszewski et al., 2018; Mounicou et al., 2003; Ofori-Frimpong et al., 1999) or for organic contaminants in fish from different fishing areas (Azad et al., 2019; Karl & Lahrssen-Wiederholt, 2013; Sunderland et al., 2018). Thus, the geographical origin of food could be relevant to evaluate dietary exposure to undesired substances in all situations, not only in crisis situations. In dietary exposure assessment, different geographical origin of food is usually not part of the scenarios and country-specific data are used if possible, but it could be important to take different geographical origins available on the country market into consideration to reduce uncertainties and refine the estimates, as already done for a possible brand-loyal consumer behaviour (Sarvan et al., 2017; Tennant & Bruyninckx, 2018; WHO & FAO, 2009b). To focus on both a specific brand or a specific geographical origin of foods could be important, as both could expose a consumer to lower or higher substance concentrations. Origin-related substance concentrations could have an increasing or decreasing effect on dietary exposure estimates, if the consumer focuses on a specific geographical origin of food.



Figure 1: Standard scenarios for deterministic dietary exposure assessment (Sarvan et al., 2017) (left) and origin-related scenarios (right). Same colour shows corresponding scenarios (standard (1–4) and origin-related (5–8)). While standard scenarios use the 95th percentile for high concentrations, origin-related scenarios use mean concentrations of different origins (Fechner *et al.*, 2019c)

1.2 Objectives

The main objective of this work was to develop approaches to the refinement of chronic dietary exposure assessment through the integration of food supply chain information. In times of global food supply, geographical origin is a major factor in a set of influences on the composition of our food, as substance concentrations could be linked to the geographical origin of food. This is not part of standard exposure assessments. To consider long-term effects of the geographical origin of food, chronic dietary exposure is relevant.

Several sub-objectives were defined:

- (1) It should be considered if consumption data and concentration data as typical data sources for dietary exposure assessment could be used to extract geographical origin information on food items to integrate these data into exposure scenarios. Especially the relation between variable substance concentrations and the geographical origin of food should be considered in specific case studies based on individual food items and single substances.
- (2) Additional data sources should be identified to gain more information on geographical origins of food available on a target market like Germany or Norway, which could be integrated in dietary exposure assessment.

The working hypotheses were:

- (1) Depending on the individual food item and the substance considered, varying substance concentrations in food are linked to the geographical origin of food making it relevant for dietary exposure assessment.
- (2) To cover geographical influences, it is not sufficient to work with standard dietary exposure scenarios, which
 - a) are based on average substance concentrations and do not integrate a geographical influence of origin.
 - b) integrate high substance concentrations (the 95th percentile (P95)) to include the influence of an unspecific geographical.
- (3) For dietary exposure assessments at a European level, the use of country-specific substance concentrations in combination with country-specific consumption is needed to model regionally different exposure situations related to different geographical origins of food available on the target markets. In contrast, the usage of pooled European concentrations could mask regional differences.
- (4) Even from limited data on the geographical origin of food, important insights can be derived into a refined dietary exposure assessment.

General assumptions made:

- (1) Agricultural products used as raw materials for food production originate from various geographical origins through global food supply.
- (2) The flow of an individual food item to a target market like Germany or Norway is composed of a set of geographical origins in a relatively stable proportion over time. Seasonal variations could occur. Starting from a specific geographical origin, each food item passes by specific geographical locations in the food supply chain, which is considered a specific supply channel. A combination of various supply channels results in the total food supply of a target market.
- (3) In the single steps of each supply channel (e.g. further processing, storage), there are different potential influences on substance concentrations (Fechner *et al.*, 2019d) (chapter 2.1). Different substance concentrations result depending on the specific geographical origin and on the supply channel connected to this origin. This has an influence on dietary exposure (Figure 2).

- (4) Consumers focus on specific geographical origins of food
 - a) directly, as they choose specific origins at the place to shop enabled by origin labelling of food.
 - b) indirectly, as they purchase food items mainly at the same place to shop or they focus on specific brands. Specific places to shop or brands could have food or ingredients available from one specific geographical origin and one specific supply channel, as stable contracts to specific supply partners exist.



Figure 2: Potential influences within the food supply chain on substance concentration and dietary exposure (Fechner *et al.*, 2019d)

2 Publications

2.1 Food Origin Information in Times of Global Food Supply – Basis for the Refinement of Dietary Exposure Assessment

(Fechner et al., 2019d)

The article was published in BfR-Wissenschaft 04/2019, a journal of the German Federal Institute for Risk Assessment (BfR) on the special issue "Feed and food safety in times of global production and trade" and is used for the cumulative dissertation by Carolin Fechner without any modifications or adaptations. The free download of the publication is available via the following link: <u>https://www.bfr.bund.de/cm/350/feed-and-food-safety-in-times-of-global-production-and-trade.pdf</u>.

The usage of this article for the dissertation by Carolin Fechner was authorised by the BfR, Unit Press and Public Relations, Department Risk Communication on 26.11.2019.

Carolin Fechner:	Conceptualization,	Formal	analysis,	Methodology,	Software,
	Visualization, Writing	g – origina	l draft		
Matthias Greiner:	Conceptualization, N	Aethodolog	gy, Project a	administration	
Helmut Heseker:	Conceptualization, F	Project adr	ninistration		
Oliver Lindtner:	Conceptualization, N	Nethodolog	gy, Resourc	es, Software, Su	pervision

2.2 Refinement of dietary exposure assessment using origin-related scenarios

(Fechner et al., 2019c)

The article was published in the Journal of Exposure Science & Environmental Epidemiology in 2019 and is used for the cumulative dissertation by Carolin Fechner without any modifications or adaptations.

Licence of the publication: "This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made" (Fechner *et al.*, 2019c).

Carolin Fechner:	Conceptualization,	Formal	analysis,	Methodology,	Software,
	Visualization, Writing	g – original	draft		
Matthias Greiner:	Conceptualization, N	/lethodolog	jy, Project a	dministration	
Helmut Heseker:	Conceptualization, F	Project adm	ninistration		
Oliver Lindtner:	Conceptualization, N	/lethodolog	y, Resource	es, Software, Sup	pervision

2.3 Dietary exposure assessment of aluminium and cadmium from cocoa in relation to cocoa origin

(Fechner *et al.*, 2019b)

This article was published in the journal PLOS ONE in 2019 and is used for the cumulative dissertation by Carolin Fechner without any modifications or adaptations.

Licence of the publication: "This is an open access article distributed under the terms of the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are credited" (Fechner *et al.*, 2019b).

Carolin Fechner:	Conceptualization,	Formal	analysis,	Methodology,	Software,
	Visualization, Writing	ı – original	draft		
Matthias Greiner:	Conceptualization, M	lethodolog	y, Project a	dministration	
Helmut Heseker:	Conceptualization, Project administration				
Oliver Lindtner:	Conceptualization, N	lethodolog	y, Resource	es, Software, Sup	pervision

2.4 Influence of the geographical origin on substance concentrations in herring as basis for dietary exposure assessments

(Fechner *et al.*, 2019a)

This article was published in the EFSA Journal in 2019 and is used for the cumulative dissertation by Carolin Fechner without any modifications or adaptations.

Licence of the publication: "This is an open access article under the terms of the Creative Commons Attribution-NoDerivs License, which permits use and distribution in any medium, provided the original work is properly cited and no modifications or adaptations are made" (Fechner *et al.*, 2019a)

Carolin Fechner:	Conceptualization,	Formal	analysis,	Methodology,	Software,
	Visualization, Writin	g – original	draft		
Sylvia Frantzen:	Conceptualization, N	Methodolog	y, Resource	es	
Oliver Lindtner:	Conceptualization, N	Methodolog	y, Resource	es	
Gro H. Mathisen:	Conceptualization, N	Methodolog	y, Supervisi	on	
Inger Therese L.					
Lillegaard:	Conceptualization,	Methodolog	gy, Project a	administration, F	Resources,
	Software, Supervisio	on			

2.5 Dioxins and dioxin-like PCBs in Atlantic herring: A Norwegian – German comparison of the influence of fishing area on country-specific dietary exposure

(Fechner *et al.*, 2020)

This article was submitted to a scientific journal for an open access publication.

Author contributions:					
Carolin Fechner:	Conceptualization,	Formal	analysis,	Methodolog	gy, Software,
	Visualization, Writin	g – origin	al draft		
Sylvia Frantzen:	Conceptualization, I	Methodolo	ogy, Resou	irces	
Lene Frost Andersen:	Methodology, Resources				
Oliver Lindtner:	Conceptualization, Methodology, Resources				
Gro H. Mathisen:	Conceptualization, Methodology, Supervision				
Inger Therese L.					
Lillegaard:	Conceptualization,	Metho	dology,	Project	administration,
	Resources, Softwar	e, Superv	ision/		

3 Methods and data

3.1 Data sources

Table 1: I	Data sources i	used for dietary	exposure	assessment

Data type	Data source	Data owner		
Consumption data Germany (Brombach <i>et al.</i> , 2006; Krems <i>et al.</i> , 2006)	German National Nutrition Survey II (Nationale Verzehrsstudie II (NVS II))	Max Rubner-Institut (MRI) Federal Research Institute of Nutrition		
	Full data were made available by the data owners for free.	and Food (Bundesforschungsinstitut für Ernährung und Lebensmittel)		
	Data are available in a scientific use file: https://www.mri.bund.de/de/institute/ernae hrungsverhalten/forschungsprojekte/nvsii/s cientific-use-file/	Haid-und-Neu-Str. 9 76131 Karlsruhe (Germany) Telephone: +49 721 6625-0 E-mail: <u>kontakt@mri.bund.de</u>		
Consumption	Norkost 3	University of Oslo (UiO)		
(Totland <i>et</i>	Full data were made available by the data owners for free.	Faculty of Medicine		
<i>al.</i> , 2012)	Summarised data are available in the report: https://www.helsedirektoratet.no/rapporter/ norkost-3-en-landsomfattende- kostholdsundersokelse-blant-menn-og- kvinner-i-norge-i-alderen-18-70-ar-2010- 11/	Postboks 1110 Blindern 0317 Oslo (Norway) Telephone: +47 22851081 E-mail: <u>ekspedisjon@basalmed.uio.no</u>		
Concentration	German Food Monitoring (GFM)	Federal Office of Consumer Protection		
data Germany	Full data were made available by the data owners for free.	And Food Safety (Bundesamt fur Verbraucherschutz und Lebensmittelsicherheit (BVL))		
	Summarised data are available in tables, reports and manuals: <u>https://www.bvl.bund.de/EN/Tasks/01_Foo</u> <u>d/01_tasks/02_OfficialFoodControl/04_LM</u> <u>Monitoring_en/LM_Monitoring_EN_node.</u> <u>html</u>	Bundesallee 35 38116 Braunschweig (Germany) Telephone: +49 531 21497-0 E-mail: <u>poststelle@bvl.bund.de</u>		
	https://www.bvl.bund.de/DE/Arbeitsbereich e/01_Lebensmittel/01_Aufgaben/02_Amtli cheLebensmittelueberwachung/04_Monito ring/Im_monitoring_node.html			
Concentration	Norwegian Seafood database	Institute of Marine Research (IMR)		
uata inorway	Full data were made available by the data owners for free.	(Havforskningsinstituttet (HI)) P.O box 1870 Nordnes		
	Summarised data are available: https://sjomatdata.hi.no/#search/	NO-5817 Bergen (Norway) Telephone: +47 55 23 85 00 E-mail: <u>post@hi.no</u>		

Data type	Data source	Data owner
Primary	FAOSTAT primary production data	Food and Agriculture Organization of
production data	Full data are available: <u>http://www.fao.org/faostat/en/#data</u>	the United Nations (FAO)
Trade data international	UN Comtrade Database – International Trade Statistics Database	United Nations
	Full data are available: https://comtrade.un.org/data	
Trade data	Eurostat: International Trade	European Commission
Europe	Full data are available: http://epp.eurostat.ec.europa.eu/newxtweb /mainxtnet.do	
	Data access via Warenstrom-Info: https://foodrisklabs.bfr.bund.de/warenstro m-info/	German Federal Institute for Risk Assessment (Bundesinstitut für Risikobewertung (BfR))
Market data	Global New Product Database (GNPD)	Mintel International Group Ltd.
on products available	Full data are available having an account: https://www.mintel.com/global-new- products-database	

Table 2: Data sources used to extract additional information on the geographical origin of food

Dietary exposure assessment was based on consumption and concentration data displayed in Table 1. The NVS II, conducted during 2005 – 2006 is a comprehensive database, on the food consumption representative for Germany covering participants aged between 14 and 80 years (Brombach *et al.*, 2006; Krems *et al.*, 2006). For dietary exposure assessments, 15,371 dietary history interviews (Fechner *et al.*, 2019b, 2019c) (chapters 2.2 and 2.3) or 13,926 24-h recalls for two days per person (Fechner *et al.*, 2019d; Fechner *et al.*, 2020) (chapters 2.4 and 2.5) were used. The survey Norkost 3 was conducted during 2010 – 2011 including Norwegian participants aged between 18 and 70 years and documenting the food consumption in Norway (Totland *et al.*, 2012). For dietary exposure assessments, 1,787 24-h recalls for two days per person were used (Fechner *et al.*, 2019a; Fechner *et al.*, 2020) (chapters 2.4 and 2.5). Substance concentrations were derived from German Food Monitoring and Norwegian Seafood monitoring programmes (Table 1) and used for dietary exposure assessment (Fechner *et al.*, 2019a; Fechner *et al.*, 2020) (chapters 2.2 – 2.5). Information on the geographical origin of food was partly provided in food monitoring data and related to substance concentrations to be used for dietary exposure assessments.

To derive additional information on the geographical origin of food, data sources displayed in Table 2 were used. FAO primary production data (FAO, 2020) showed possible geographical origins of food and were compared with indications in food monitoring data in a preliminary investigation (Fechner *et al.*, 2019d) (chapter 2.1). This was used for a plausibility check of origin indications for case studies considered in food monitoring data. Additionally, other data

sources were used to generate more food origin information and to evaluate the market situation for the case studies investigated. The databases UN Comtrade and Eurostat gave access to export and import volumes of worldwide traded goods, which are indicated in HS codes as identifiers for commodities in trade statistics (Eur. Commission, 2020; UN, 2020). These data sources were used to track supply steps and conclude on possible supply channels of food items to target markets. Newly launched packaged food products are documented in the Mintel Global New Product Database (GNPD) (Mintel International Group Ltd., 2020). Ingredients lists and packaging photos are collected there, which gave more detailed information on possible geographical origins of food and complemented data indicated in food monitoring.

3.2 Selection of case studies

Food items and substances considered in case studies (Fechner *et al.*, 2019a; Fechner *et al.*, 2019b, 2019c; Fechner *et al.*, 2020) (chapters 2.2 - 2.5) for origin-related dietary exposure assessment were:

- (1) Bromide in tomatoes
- (2) Ethephon in pineapples
- (3) Aluminium in kiwifruits
- (4) Aluminium and cadmium in cocoa powder
- (5) PCDD/Fs and DL-PCBs in Atlantic herring (Clupea harengus)

All case studies are summarised in Figure 3. Case studies (1) to (4) were based on data from Germany, while case study (5) included additional data from Norway. The selection of case studies (1) - (3) for the agricultural products tomatoes, pineapples and kiwifruits was based on food monitoring data between 2005 and 2015 (Fechner et al., 2019c) (chapter 2.2). These food items were chosen, as comprehensive and plausible origin indications in food monitoring data were found in the preliminary investigation (Fechner et al., 2019d) (chapter 2.1). For the first step of substance selection from food monitoring data, two criteria (i) and ii)) were set to enable the search for geographical variability in quantified substance concentrations. The substances were selected, if i) minimum 40 samples were available and ii) more than 50 % of those samples had quantifiable concentrations (Fechner et al., 2019c) (chapter 2.2). A list of substances resulted from the first step of selection and for each agricultural product, the substance with the highest variation was selected in a second step using the coefficient of variance (ratio of standard deviation and mean). Origin-related variability of substance concentrations was checked using means and boxplots per country and decided for bromide in tomatoes, ethephon in pineapples and aluminium in kiwifruits (Fechner et al., 2019c) (chapter 2.2).

Aluminum and cadmium in cocoa powder were selected as case study (4) using German Food Monitoring data, as other investigations already supported geographical variation in concentrations (Abt *et al.*, 2018; BfR, 2007; Kruszewski *et al.*, 2018; Müller *et al.*, 1998; Ofori-Frimpong *et al.*, 1999) and bitter chocolate was found to be one of the main contributors to dietary aluminium exposure (Tietz *et al.*, 2019). Food monitoring data between 2005 and 2015 were checked for analyses of aluminium and cadmium in cocoa powder, more than 40 samples were available and all samples had quantifiable concentrations (Fechner *et al.*, 2019b) (chapter 2.3). However, plausible origin information was rare in German Food Monitoring data, which was already shown in the preliminary investigation of origin indications (Fechner *et al.*, 2019d) (chapter 2.1) and a sensitivity analysis had to be used for origin grouping later on. As only for a few food items origin information was sufficient in food monitoring also cocoa powder was selected for further investigations as example with insufficient origin data.

As part of the European Food Risk Assessment fellowship programme (EU-FORA), Norwegian and German consumption and concentration data were accessible to conduct a country-specific dietary exposure assessment (Fechner *et al.*, 2019a; Fechner *et al.*, 2020) (chapters 2.4 and 2.5). Atlantic herring (*Clupea harengus*), called herring from here on, was selected, as this fish species was consumed (Brombach *et al.*, 2006; Krems *et al.*, 2006; Totland *et al.*, 2012) and investigated for substances in Norway and Germany, respectively. PCDD/Fs and DL-PCBs were investigated in both countries and related to the Baltic Sea for German samples and to the Norwegian Sea for Norwegian samples (Fechner *et al.*, 2019a) (chapter 2.4). The European exposure situation to PCDD/Fs and DL-PCBs is of high interest for human health, as EFSA reduced the tolerable weekly intake (TWI) from the total diet to 2.0 pg WHO-2005-TEQ/kg BW per week (EFSA, CONTAM Panel *et al.*, 2018a).

3.3 Dietary exposure

Four standard exposure scenarios (1 - 4) (Sarvan *et al.*, 2017) using general substance concentrations in food were extended by four origin-related scenarios (5 - 8) using substance concentrations of specific food origins to determine chronic dietary exposure based on consumption amounts related to the individual body weight (Table 3). Evaluations for tomatoes, pineapples and kiwifruits were done using consumption and concentration parameters (mean and P95) and resulted in a point estimate of the dietary exposure, which is a deterministic approach (Sarvan *et al.*, 2017) (chapter 2.2). Evaluations for cocoa powder (chapter 2.3) and herring (chapters 2.4 and 2.5) were based on consumption distributions, which were combined with concentration parameters (mean and P95) and resulted in exposure parameters (the 50th percentile (P50) and the P95). This represents a semi-probabilistic approach (Sarvan *et al.*, 2017). For herring, origin-related scenarios were considered only, as the study was based on country-specific data of Germany and Norway to compare country-

specific dietary exposures (chapters 2.4 and 2.5). Scenarios with Norwegian herring data representing lower concentrations correspond to the origin-related scenarios 5 and 6 used for the other case studies, while scenarios based on German herring data representing higher concentrations correspond to origin-related scenarios 7 and 8. Units of exposure estimates were orientated on the particular health-based guidance value for each substance considered.

Scenario	Consumption	Concentration	Exposure	Food items investigated
1	Mean Distribution	Mean all samples	Point estimate P50	tomato, pineapple, kiwifruit cocoa powder
2	P95 Distribution	Mean all samples	Point estimate P95	tomato, pineapple, kiwifruit cocoa powder
3	Mean Distribution	P95 all samples	Point estimate P50	tomato, pineapple, kiwifruit cocoa powder
4	P95 Distribution	P95 all samples	Point estimate P95	tomato, pineapple, kiwifruit cocoa powder
5	Mean Distribution	Mean Origin A	Point estimate P50	tomato, pineapple, kiwifruit cocoa powder, herring
6	P95 Distribution	Mean Origin A	Point estimate P95	tomato, pineapple, kiwifruit cocoa powder, herring
7	Mean Distribution	Mean Origin B	Point estimate P50	tomato, pineapple, kiwifruit cocoa powder, herring
8	P95 Distribution	Mean Origin B	Point estimate P95	tomato, pineapple, kiwifruit cocoa powder, herring

Table 3: Standard scenarios (1 - 4) and origin-related scenarios (5 - 8) used to evaluate dietary exposure for the case studies selected

Scenarios are based on literature (Sarvan *et al.*, 2017; WHO & FAO, 2009b) and were further developed in publications related to the current research (Fechner *et al.*, 2019a; Fechner *et al.*, 2019b, 2019c; Fechner *et al.*, 2020) (chapters 2.2 – 2.5). In origin-related exposure scenarios, only mean concentrations of Origin A and Origin B were used. In contrast, P95 concentrations were not used, as it was considered too conservative to assume both, firstly, that consumers could focus only on one specific geographical origin (here indicated as Origin A or Origin B) and secondly, that consumers could focus only on the highest concentrations (expressed by the P95) of the focused geographical origin (Table 3). Thus, only the focus on a specific geographical origin with lower (Origin A) or higher mean concentrations (Origin B) was used for modelling.

Definitions:

"All samples" all available substance concentrations from food monitoring data "Origin A" samples of a geographical origin having lower substance concentrations "Origin B" samples of a geographical origin having higher substance concentrations Bromide in tomatoes: Origin A – all regions except Italy, Origin B – Italy Ethephon in pineapples: Origin A – America, Origin B – Africa Aluminium in kiwifruits: Origin A – EU countries, Origin B – non-EU countries Aluminium and cadmium in cocoa powder: Origin A and B – modelled by a sensitivity analysis PCDD/Fs and DL-PCBs in herring: Origin A – Norwegian Sea, Origin B – Baltic Sea

<u>Abbreviations</u>: P50 50th percentile, P95 95th percentile

Consumption data used were derived from national surveys. Long-term consumption of tomatoes, pineapples and kiwifruits was extracted from dietary history interviews of the NVS II (Brombach et al., 2006; Krems et al., 2006) using disaggregated household recipes (Fechner et al., 2019c) (chapter 2.2). Dietary exposure assessment was carried out only for consumers of the food items considered and data showed that 97 % of the survey participants were consumers of tomatoes, 18 % consumed pineapples and 30 % consumed kiwifruits. Long-term cocoa consumption was derived from dietary history interviews of the NVS II using a completely disaggregated data version (household recipes and composite industrial products disaggregated) (Fechner et al., 2019b) (chapter 2.3) created for the LExUKon project (Blume et al., 2010; Schneider et al., 2014; Schwarz et al., 2014). Dietary exposure assessment was based on cocoa consumers only and data showed that 81 % of the participants were consumers. Long-term herring consumption was derived from two 24-h recalls of the NVS II using disaggregated household recipes for Germany (Brombach et al., 2006; Krems et al., 2006), while for Norway, the two 24-h recalls of Norkost 3 (Totland et al., 2012) were used giving aggregated and completely disaggregated consumption amounts based on household recipes and composite industrial products (Fechner et al., 2019a; Fechner et al., 2020) (chapters 2.4 and 2.5). Most disaggregated consumption amounts available were used for the generation of the herring results shown here. These data showed that 4.3 % of the German participants and 5.5 % of the Norwegian participants consumed herring, which was only a small part of the population (Fechner et al., 2019a) (chapter 2.4).

Substance concentrations used were derived from national food monitoring data. In standard exposure scenarios, the use of mean concentrations modelled the contact of consumers with regularly mixing low and high concentrations and the use of P95 concentrations displayed the consumers' contact to regular high concentrations, which could express brand loyalty (Fechner et al., 2019c; Sarvan et al., 2017) (chapter 2.2). Concentration datasets could contain results, which are below the limit of detection (LOD), so-called non-detected values, or which are below the limit of quantification (LOQ), so-called non-quantified values (Sarvan et al., 2017; WHO & FAO, 2009b). Such non-quantifiable concentrations are included in exposure evaluations using lower bound (LB), modified lower bound (MLB) or upper bound (UB) approaches (Sarvan et al., 2017; WHO & FAO, 2009b). In the current case, non-detected values were replaced by zero for the LB approach and the MLB approach or they were replaced by the LOD for the UB approach. Non-quantified values were replaced by zero for the LB approach, by the LOD for the MLB approach or by the LOQ for the UB approach (Fechner et al., 2019c) (chapter 2.2). This kind of replacement was not necessary for aluminium and cadmium in cocoa powder, as all samples had quantifiable concentrations (Fechner et al., 2019b) (chapter 2.3). Norwegian fish monitoring data did not provide the LOD and thus, the LB was used for PCDD/F and DL-PCB concentrations in herring for Norway and Germany (Fechner et al., 2019a; Fechner et al.,

22

2020) (chapters 2.4 and 2.5). For exposure estimates shown here, MLB or LB concentrations were used, as for the substances considered, most of the concentrations were quantified and differences to UB concentrations were minor.

To perform origin-related exposure assessment for each case study, lower mean concentrations were attributed to Origin A and higher mean concentrations were attributed to Origin B (Table 3) (Fechner et al., 2019c) (chapter 2.2). For aluminium and cadmium in cocoa powder, data on the geographical origin of the samples were not sufficiently reported in food monitoring data and could not be used for origin grouping (Fechner et al., 2019b) (chapter 2.3). Therefore, a general method called sensitivity analysis was used to group concentrations into three segments to obtain lower and higher concentrations (Frey & Patil, 2002; Heinemeyer et al., 2015). Two different grouping approaches were applied, the first one used sorted concentrations to generate three segments, each with the same number of values, and the second grouping approach used the 25th percentile and the 75th percentile to divide sorted concentrations into three segments (Fechner et al., 2019b) (chapter 2.3). The segment with the lowest concentrations was then attributed to Origin A and the segment with the highest concentrations was attributed to Origin B. In this way, the limited origin information in German Food Monitoring data was substituted by theoretical concentration groups (Frey & Patil, 2002; Heinemeyer et al., 2015) to simulate influences of origin. Estimates shown here used concentrations of grouping approach 1 (Fechner et al., 2019b) (chapter 2.3). For all other case studies, origin indications in food monitoring data were used to group lower and higher mean concentrations and to attribute them to Origin A and Origin B. In origin-related exposure scenarios, only mean concentrations of Origin A and Origin B were used. In contrast, P95 concentrations were not used, as it was considered too conservative to assume both, firstly, that consumers could only focus on one specific geographical origin (here indicated as Origin A or Origin B) and secondly, that consumers could only focus on the highest concentrations (expressed by the P95) of the focused geographical origin (Table 3). Thus, only the focus on a specific geographical origin with lower (Origin A) or higher mean concentrations (Origin B) was used for modelling. Seasonal variations were not investigated, as sample numbers were already quite low in the grouping by different geographical origins and information on production dates was not available instead only the sampling dates were stated.

3.4 Statistical analyses and evaluations

Statistical analyses of consumption and concentration data as well as exposure evaluations were carried out using IBM SPSS Statistics version 21 and version 25. The parameters mean, standard deviation and the percentiles were computed using the CTABLES command. Grouped substance concentrations were checked for normal distribution using the Kolmogorov-Smirnov test. As only some groups were normally distributed, the non-parametric

23

Kruskal-Wallis test in combination with the post-hoc Dunn-Bonferroni test with a significance level of $P \le 0.05$ were used to check for significant differences of substance concentrations between different origins grouped as Origin A and Origin B and all samples. Microsoft Excel 2010 and 2016 were used for graphical depiction and for the analysis of information in the FAO database, the Eurostat database, the UN Comtrade database and the Mintel GNPD.

4 Results

4.1 Information on the geographical origin of food

Comprehensive data are needed to include the geographical origin of food in dietary exposure assessment. For exposure assessment, consumption and concentration data displaying the situation within a certain population are needed. Consumption surveys used for the current investigations did not contain data on the geographical origin of food items eaten. Therefore, it was not possible to integrate geographical origin of food in exposure assessment using consumption data.

The food monitoring data normally used to derive substance concentrations of food for dietary exposure assessment (Sarvan et al., 2017) provided information on the geographical origin of food. However, a preliminary investigation on German Food Monitoring data showed, that the information content depended on the food item considered and it was possible to have included the information on processing stages instead of the geographical origin of food (Fechner et al., 2019d) (chapter 2.1). For the agricultural products considered, i.e. tomatoes, pineapples and kiwifruits, and for olive oil, unspecified origin indications ranged between 4 and 9 % in German Food Monitoring data and Germany was not listed as the main specific origin (Fechner et al., 2019d) (chapter 2.1). Other food items considered in German Food Monitoring data were cocoa powder, tomato juice, apple juice, and beef and pork cuts, which had a percentage of unspecified origins between 2.6 % and 25.9 % (Fechner et al., 2019d) (chapter 2.1). Germany was stated as the main geographical origin for these food items. The German Food Monitoring data contained the two variables "fishing area" and "country of origin" for the fish species herring and tuna (Fechner et al., 2019d) (chapter 2.1). These variables showed that the herring has a smaller global fishing area than tuna. The herring data on origin were comprehensive and sufficient for origin-related exposure assessment, while there was a lack of data for tuna. Unspecified information on the fishing area and the country of origin ranged between 23 % and 69 % for herring and tuna. The amount of unspecified information showed a more or less comprehensive lack of data on specific origins depending on the food item considered in food monitoring data, which generally could limit the integration of geographical origin information into exposure assessment (Fechner et al., 2019d) (chapter 2.1).

FAO primary production data (FAO, 2020) made it possible to check German Food Monitoring data for plausibility, as a processing or packaging stage could be indicated instead of a geographical origin of food. This was used for the preliminary investigation of the situation (Fechner et al., 2019d) (chapter 2.1) and for the evaluation of selected case studies for exposure assessment (Fechner et al., 2019b, 2019c) (chapters 2.2 and 2.3). Various other regions were stated for food items in the German Food Monitoring data and Germany was not indicated as the main origin (i.e. tomatoes, pineapples, kiwifruits, olive oil), while FAO data identified most of the origins indicated as plausible (79 – 97%) and this origin information would be sufficient for origin-related exposure assessment. In contrast, the situation varied for food items in the German Food Monitoring data with Germany indicated as the main origin (i.e. cocoa powder, tomato juice, apple juice, and beef and pork cuts). FAO data showed that cocoa was not harvested in Germany and most of the origin information on cocoa powder in food monitoring data became implausible, as a processing or packaging stage was documented instead (Fechner et al., 2019d) (chapter 2.1). Therefore, origin information on cocoa powder in the German Food Monitoring data was not sufficient for origin-related exposure assessment and other approaches would be needed to generate origin information. Germany was the main origin documented for tomato juice, apple juice, and beef and pork cuts in the German Food Monitoring and FAO data confirmed a possible primary production in Germany, as well. Here, the FAO data did not allow the discrimination between Germany as a plausible geographical origin and Germany as a labelled packaging stage (Fechner et al., 2019d) (chapter 2.1). Thus, the usage of FAO data alone was not sufficient to evaluate the plausibility of geographical origins stated in the German Food Monitoring data for these food items. Origin information would not be sufficient for origin-related exposure assessment, as Germany was mainly indicated and thus origin-related different concentrations were not visible or did not exist. FAO data helped to evaluate the usability of the German Food Monitoring data to integrate geographical origin information in dietary exposure assessment depending on the food item investigated and on the origin information given in the German Food Monitoring data (Fechner et al., 2019d) (chapter 2.1). However, sufficient origin information was mostly limited to agricultural products and the origin information indicated for most of the processed food items would not be sufficient to be used for origin-related dietary exposure assessment. This was identified as clear limitation in food monitoring data.

Trade data were used to generate more information for the specific case studies considered. It showed that tomatoes, pineapples, and kiwifruits were supplied to Germany by various countries and that the origin indications in the German Food Monitoring data were supported by trade data (Fechner *et al.*, 2019c; UN, 2020) (chapter 2.2). Italy was a great distributor of tomatoes and, New Zealand and Italy were the biggest kiwifruit exporters which showed the importance of different geographical origins in global trade (Fechner *et al.*, 2019c; UN, 2020)

25

(chapter 2.2). Cocoa beans, cocoa powder and cocoa mass on the German market were mainly sourced in the Côte d'Ivoire (Africa) and traded via the Netherlands according to trade data (Eur. Commission, 2020; Fechner *et al.*, 2019b) (chapter 2.3). This complemented the missing origin information of the German Food Monitoring data. The export and import volumes of herring in Germany and Norway were documented for both countries (Fechner *et al.*, 2020; UN, 2020) (chapter 2.5). Even if trade statistics did not provide information about available fishing areas on the local markets, the import of herring showed the possibility of different fishing areas on the target markets.

Information of the Mintel GNPD was extracted for packaged products (Mintel International Group Ltd., 2020) to fill the data gaps for specific case studies considered. In the case of cocoa powder on the German market, the Mintel GNPD showed that an origin declaration was available in some cases only (Fechner *et al.*, 2019b) (chapter 2.3). However, the labelling itself and the wording were not standardised which complicated the extraction of information. Declared origins were located in Africa, South America and Asia (Mintel International Group Ltd., 2020), which confirmed that most of the origin indications in the food monitoring data were implausible (Fechner *et al.*, 2019b) (chapter 2.3). Investigations for herring showed that fewer herring products were available on the Norwegian market in comparison to Germany and the geographical origin labelling was more detailed for Germany (Fechner *et al.*, 2020) (chapter 2.5). Packaged herring in Norway mainly originated form the Norwegian Sea and the larger area of Northeast Atlantic, while in Germany the focus was on the North Sea, the Norwegian Sea and the Northeast Atlantic and some indications listed the Baltic Sea (Fechner *et al.*, 2020; Mintel International Group Ltd., 2020) (chapter 2.5).

4.2 Dietary exposure and geographical origin of food

Figure 3 gives an overview of food items and substances considered in case studies. It shows an excerpt of food consumption data of consumers, substance concentration data as well as some of the resulting exposure scenarios. Not all performed scenarios are shown and only the considerations for consumers of the food items are displayed, as further information can be found in the related publications (Fechner *et al.*, 2019a; Fechner *et al.*, 2019b, 2019c) (chapter 2). The overview should summarise the findings using exemplary data on the most conservative exposure estimates to compare the outcomes of conservative standard scenarios with origin-related situations. Substance concentrations used for the exposure assessment had significant differences between Origin A and Origin B and between Origin B and all samples for all case studies.

For bromide from tomatoes, the highest exposure estimate was obtained in the origin-related exposure scenario 8 and amounted to 0.015 mg/day/kg BW (Figure 3). This was well below the acceptable daily intake (ADI) of 1.0 mg/day/kg BW, as only tomatoes were considered as

an individual food item (Fechner *et al.*, 2019c) (chapter 2.2). In scenario 8, the estimate was based on high consumption and mean concentrations of Origin B (here: Italy). In this case, the influence of origin on concentrations was not covered by standard exposure scenarios, which are based on average or high concentrations derived from all samples available.

For the following case studies, the standard exposure scenario 4, based on high consumption and high concentrations derived from all samples available, resulted in the highest exposure estimate and covered the influence of higher concentrations grouped as Origin B (i.e. Africa for pineapples, non-EU-countries for kiwifruits and a theoretically modelled Origin B for cocoa powder) (Fechner *et al.*, 2019b, 2019c) (chapters 2.2. and 2.3). The highest exposure estimate for ethephon from pineapples and aluminium from kiwifruits amounted to 0.9 µg/day/kg BW and 0.02 mg/week/kg BW, which was well below the ADI of 0.03 mg/day/kg BW for ethephon and the tolerable weekly intake (TWI) of 1.0 mg/week/kg BW for aluminium (standard scenario 4). For aluminium and cadmium from cocoa powder 0.15 mg/week/kg BW and 0.4 µg/week/kg BW were obtained as the highest estimates in the standard exposure scenario 4. Here, the aluminium exposure from cocoa powder amounted to 15.2 % of the related TWI (1.0 mg/week/kg BW) and for cadmium, 14.5 % of the TWI (2.5 µg/week/kg BW) was reached based on standard scenario 4. However, it would be necessary to perform all standard scenarios to cover possible influences of origin, as exposure estimates of scenario 3 were already lower than estimates in the most conservative origin-related scenario 8.

4.3 Country-specific dietary exposure

The exposure to PCDD/Fs and DL-PCBs from herring was evaluated for Norway and Germany using country-specific data, which is also summarised in Figure 3. Results shown here are related to consumers of herring, which represent only a small part of the population, as the percentage of consumers was below 6 % for Germany and Norway, respectively. All participants of consumption surveys were investigated, additionally (Fechner *et al.*, 2020) (chapter 2.5). Evaluations used German samples from the Baltic Sea considered as Origin B with significantly higher concentrations and Norwegian samples from the Norwegian Sea considered as Origin A (Figure 3).

Estimates for Norwegian consumers of herring were clearly lower than for German consumers of herring, but the TWI of 2.0 pg WHO-2005-TEQ/kg BW per week for the total sum of 17 PCDD/Fs and 12 DL-PCBs set by EFSA (EFSA, CONTAM Panel *et al.*, 2018a) was exceeded in most of the scenarios for both countries (all P50 and P95 scenarios for Germany and all P95 scenarios for Norway but not the P50 scenarios). Using disaggregated consumption amounts and lower bound concentrations, the highest estimate for the total sum of 17 PCDD/Fs and 12 DL-PCBs from herring was obtained for German consumers of herring amounting to 26.5 pg WHO-2005-TEQ/kg BW per week in the origin-related scenario 8. Here, high consumption of

herring and mean concentrations in herring from the Baltic Sea (Origin B) were combined. The exposure to the total sum of 17 PCDD/Fs and 12 DL-PCBs was lower for Norwegian high consumers of herring, amounting to 6.5 pg WHO-2005-TEQ/kg BW per week in the originrelated scenario 6, based on herring samples from the Norwegian Sea (Origin A). The higher intake for German consumers was related to higher Baltic Sea concentrations and higher consumption amounts. A clearly different situation for German and Norwegian consumers of herring was shown using country-specific data for evaluations. However, when considering all participants in the consumption surveys, exposure estimates were low for Germany and for Norway not exceeding the TWI and origin relations had not a clear impact on P50 or P95 exposures. This is related consumption data only, as less than 6 % of the participants in consumption surveys consumed herring. Thus, Norwegian P95 exposures for all participants resulted in an estimate in comparison to German P95 exposures, which were 0. This was only related to the fact, that 5.5 % of the Norwegian survey participants consumed herring and a P95 consumption could be derived. In Germany, less than 5 % of the participants consumed herring and neither a P95 consumption nor a P95 exposure could be derived (Fechner et al., 2019a; Fechner et al., 2020) (chapters 2.4 and 2.5).

Food item				000				
		Tomato	Pineapple	Kiwifruit	Сосоа	powder	Herring (Clup	ea harengus)
Country				Germany			Germany	Norway
Food consumption (here: consumers on	survey		NV	S II: dietary his	story ^a		NVS II: 24-h recalls ^b	Norkost 3: 24-h recalls ^c
Percentage consume	ers	97	18	30		81	4.3	5.5
Consumption [g/day/	kg BW] – mean	0.6	0.3	0.2	<	0.1	0.9	0.3
Consumption [g/day/	kg BW] – P95	1.7	1.1	0.7	0.1		2.2	1.2
Substance		Bromide	Ethephon	Aluminium	Aluminium	Cadmium	Total sum of and 12 D	17 PCDD/Fs DL-PCBs
Monitoring period				2005–2015			2012-	-2017
Unit of concentration	s			mg/kg			pg WHO-2	005-TEQ/g
Substance concen-	All samples	1.3 × (n=714)	0.2 × (n=213)	1.0 × (n=163)	146 × (n=167)	0.2 × (n=166)	-	-
tration: mean	Origin A	0.8 × (n=674)	0.1 × (n=176)	0.5 ^x (n=121)	90 ^{d,y} (n=56)	0.1 ^{d,y} (n=55)	-	0.8 × (n=100)
(here: MLB or LB)	Origin B	8.6 ^y (n=40)	0.8 ^y (n=24)	2.9 ^y (n=36)	199 ^{d,z} (n=56)	0.3 ^{d,z} (n=56)	1.8 ^y (n=47)	-
Chronic dietary exp	osure (here: consumers	only)						
Unit		mg/day/kg BW	µg/day/kg BW	mg/weel	k/kg BW	µg/week/kg BW	pg WHO-2005-T	EQ/week/kg BW
	· · · · · · ·							

Average consumption, high concentrations of all samples (standard scenario 3)	0.002	0.2	0.01	0.02	0.1	-	-
High consumption, high concentrations of all samples (standard scenario 4)	0.006	0.9	0.02	0.15	0.4	-	-
High consumption, mean concentrations of Origin A (origin-related scenario 6)	0.001	0.1	<0.01	0.06	0.1	-	6.5
High consumption, mean concentration of Origin B (origin-related scenario 8)	0.015	0.8	0.01	0.14	0.2	26.5	-

Figure 3: Excerpt of data for food consumption, substance concentrations and resulting chronic dietary exposure for the case studies selected

Legend Figure 3:

Data are derived from publications related to the current research (Fechner *et al.*, 2019a; Fechner *et al.*, 2019b, 2019c; Fechner *et al.*, 2020) (chapters 2.2 – 2.5).

Chronic dietary exposure was shown only for consumers of food items considered and most disaggregated consumption amounts available were used for evaluations here. For tomatoes, pineapples and kiwifruits, consumption parameters (mean and P95) were used to estimate the dietary exposure, while for cocoa and herring the whole consumption distributions were used to derive dietary exposure in the parameters P50 and P95.

Additional information:

^a NVS II National Nutrition Survey II (2006): dietary history for 15371 participants (14-80 years)

^b NVS II National Nutrition Survey II (2006): two 24-h recalls for 13926 participants (14-80 years)

^c Norkost 3 (2010): two 24-h recalls for 1787 participants (18-70 years)

^d Concentrations for Aluminium and Cadmium in cocoa powder derived from grouping approach 1 of the sensitivity analysis (Fechner *et al.*, 2019b) (chapter 2.3). A sensitivity analysis was used to group concentrations into three segments to obtain lower and higher concentrations (Frey & Patil, 2002; Heinemeyer et al., 2015). Then, the segment with the lowest concentrations was attributed to Origin A and the segment with the highest concentrations was attributed to Origin B. In this way, the limited origin information in German Food Monitoring data was substituted with theoretical concentration groups to simulate influences of origin.

^{x, y, z} Significant differences in substance concentrations: Grouped substance concentrations were checked for normal distribution using the Kolmogorov-Smirnov test. As only some groups were normally distributed, the non-parametric Kruskal-Wallis test in combination with the post-hoc Dunn-Bonferroni test with a significance level of $P \le 0.05$ were used to check for significant differences of substance concentrations between Origin A, Origin B and all samples. For each case study, significant differences between Origin A, Origin B and all samples are indicated using different superscript letters x, y and z.

Definitions:

"All samples" all available substance concentrations from food monitoring data "Origin A" samples of a geographical origin having lower substance concentrations "Origin B" samples of a geographical origin having higher substance concentrations Bromide in tomatoes: Origin A – all regions except Italy, Origin B – Italy Ethephon in pineapples: Origin A – America, Origin B – Africa Aluminium in kiwifruits: Origin A – EU countries, Origin B – non-EU countries Aluminium and cadmium in cocoa powder: Origin A and B – modelled by a sensitivity analysis PCDD/Fs and DL-PCBs in herring: Origin A – Norwegian Sea, Origin B – Baltic Sea

<u>Abbreviations</u>: PCDDs polychlorinated dibenzo-p-dioxins, PCDFs polychlorinated dibenzofurans, DL-PCBs dioxin-like polychlorinated biphenyls, WHO World Health Organization, TEQ Toxic Equivalents, MLB modified lower bound approach (for tomatoes, pineapples, kiwifruits), LB – lower bound approach (for herring), N sample number, BW body weight, ADI acceptable daily intake, TWI tolerable weekly intake, P95 95th percentile

Food icons used were designed by Freepik.com.

5 Discussion

5.1 Dietary exposure and geographical origin of food

Approaches to the refinement of chronic dietary exposure assessment were developed integrating information on the geographical origin of foods in origin-related scenarios. This main objective and related sub-objectives were achieved by working with selected case studies, based on individual food items and single substances, which showed geographical variability in concentrations. Information on geographical origin was basically extracted from the food monitoring data of substance concentrations in food. Additional data sources related to primary production (FAO, 2020), trade (Eur. Commission, 2020; UN, 2020) and information labelled on packaged food products (Mintel International Group Ltd., 2020) complemented the origin information of the food monitoring data. The combination of information derived from all data sources allowed first insights into origin-related exposure assessment based on specific cases and the research objectives set were met.

In connection with the research objectives several hypotheses were formulated, which needed to be tested. Hypothesis (1) suggested that geographically varying substance concentrations can be identified, which has relevance for dietary exposure assessment. For specific case studies, geographically varying substance concentrations were identified or modelled based on food monitoring data and used for exposure assessments (Fechner *et al.*, 2019a; Fechner *et al.*, 2019b, 2019c; Fechner *et al.*, 2020) (chapters 2.2 - 2.5). Origin-related scenarios were developed and provided additional information on the origin-related exposure to substances. Food items sampled from the German market and originating from different geographical origins were considered (tomatoes, pineapples, kiwifruits, cocoa powder) and country-specific assessments on herring from Germany and Norway were carried out based on different geographical origins available. Origin-related considerations were relevant to evaluate the exposure situation and confirmed hypothesis (1).

Hypothesis (2) stated that standard dietary exposure scenarios based on a) average substance concentrations or b) high substance concentrations (P95) do not cover the influence of origin displayed in origin-related scenarios. This hypothesis was confirmed for the case study on bromide exposure from tomatoes (Fechner *et al.*, 2019c) (chapter 2.2), as the highest exposure estimates for tomato consumers were derived from the origin-related exposure scenario 8 exceeding the values of the standard scenarios 1 - 4. This case study showed that the integration of the geographical origin of food in exposure assessment would be reasonable, if consumers focused on a specific origin. In contrast, for the case studies ethephon from pineapples, aluminium from kiwifruits as well as aluminium and cadmium from cocoa powder, the highest exposure estimate for consumers of these food items was obtained in standard scenario 4 (Fechner *et al.*, 2019b, 2019c) (chapters 2.2 and 2.3). The influence of origin-related

31

higher concentrations displayed in origin-related scenario 8 was covered in standard scenario 4. Thus, part b) of the hypothesis was not confirmed for these case studies, as it was sufficient to estimate the exposure using P95 concentrations not related to specific origins. The highest and most conservative estimate was carried out in standard scenarios, as suggested in methodological approaches for exposure assessment (EFSA, European Food Safety Authority, 2011; Sarvan *et al.*, 2017; WHO & FAO, 2009b) and additional origin-related scenarios are at least not needed to obtain the most conservative estimate. However, part a) of the hypothesis was confirmed for these case studies, as estimates of the origin-related scenario 8 exceeded standard scenarios 1 - 3. Thus, the use of average concentrations or consumption was not sufficient to cover possible influences of origin. Consequently, all standard scenarios would need to be performed to cover influences of origin.

For the case studies considered here, health-based guidance values were not exceeded in standard or origin-related exposure scenarios (Fechner et al., 2019b, 2019c) (chapters 2.2 and 2.3). Origin-related scenarios helped to refine the estimates integrating the geographical origin of food as an important influence showing lower or higher exposures to substances in originrelated consumption situations in comparison to standard scenarios. Such a refinement can be of special interest, if the total dietary exposure to a substance from the consumption of various food items is considered and health-based guidance values are exceeded by adding up the intake from different food sources. Under the precondition that consumers focus on specific geographical origins of food, it could be even more precise to base evaluations on known origin-relations to model refined and more realistic exposure scenarios instead of deriving the most conservative estimate. However, integrating data on food origin is also timeand resource-intensive and if data on available food origins is not sufficient, additional uncertainty could be introduced to exposure models. Thus, the objective of the exposure estimate should be clear. That means origin relations could be integrated as an additional factor, if a refined estimate is wanted, but if the most conservative estimate should be derived, often standard scenarios are sufficient.

5.2 Country-specific dietary exposure

Hypothesis (3) assumed that the use of pooled European concentrations in combination with country-specific consumption is not sufficient to model country-specific exposure situations and instead country-specific substance concentrations are needed to refine the estimates for different regions. The case study on PCDD/F and DL-PCB exposure from herring was used to perform a country-specific exposure assessment for the countries Germany and Norway (Fechner *et al.*, 2019a; Fechner *et al.*, 2020) (chapters 2.4 and 2.5). In a European context, the German situation modelled for consumers of herring could be seen as a conservative estimate because significantly higher Baltic Sea concentrations and a higher consumption

were used as data basis in comparison to Norway, where the scenarios were based on the quite low concentrations from the Norwegian Sea and lower consumption providing a lower estimate. Results shown here in detail are related to consumers of herring, which represent only a small part of the population, as the percentage of consumers was below 6 % for Germany and Norway, respectively (Fechner et al., 2019a) (chapter 2.4). Different PCDD/F and DL-PCB concentrations were related to specific fishing areas of herring and an obviously higher P95 LB exposure of 26.5 pg WHO-2005-TEQ/kg BW per week for German consumers of herring resulted origin-related scenario 8 (Fechner et al., 2020) (chapter 2.5). In contrast, exposure estimates for all participants of consumption surveys were generally low for Germany and Norway. Consequently, the integration of influences of origin was more relevant for the small sub-group of consumers of herring in the population, as for them clearly different exposure levels were estimated based on country-specific data. This case study confirmed hypothesis (3), as the use of country-specific concentrations clarified the situation of the consumers. In contrast, pooled European concentrations used by EFSA were even higher than German Baltic Sea concentrations used here (EFSA, CONTAM Panel et al., 2018a) and would lead to higher estimates for Norwegian and German consumers of herring. Especially if healthbased guidance values are exceeded, origin-related scenarios can help to refine the estimate and provide data for risk management decisions adapted to specific regions in a European context. Nevertheless, the focus on the small number of consumers represented a quite conservative view and a focus on all participants in consumption surveys showed less relevance of additional origin information. The integration of origin information into exposure estimates would be more relevant for food items, which are consumed by more people, as a larger part of the population would be affected.

5.3 Data limitations, uncertainties and future requirements

The publications connected to this research describe the typical limitations and uncertainties related to data and scenarios in dietary exposure assessments (Fechner *et al.*, 2019a; Fechner *et al.*, 2019b, 2019c; Fechner *et al.*, 2019d; Fechner *et al.*, 2020) (chapters 2.1 - 2.5). However, on top of that, further limitations and uncertainties are relevant for the current and future research related to the geographical origin of food. Hypothesis (4) supposed that even limited data on the geographical origin of food provide key insights into the refinement of dietary exposure assessment using food supply chain information. This was confirmed, as case studies were identified and used for the refinement of dietary exposure assessment integrating limited information on the geographical origin of food. Using the information available in the food monitoring data and complementing it with other data sources made it possible to generate approaches to the refinement of dietary exposure assessment and gain first insights into origin-related intake situations but further information would be needed for deeper insights.

33

More detailed data are needed for future research in the refinement of dietary exposure assessment using food supply chain information. The data requirement is displayed in assumptions, which were set for the current research. These assumptions showed a lack of data related to information on the geographical origin of food in the following subjects:

- (1) Supply channels of food from production countries to target markets and the structure of geographical origins of food on target markets (proportion)
- (2) Geographical influences on substances leading to varying concentrations
- (3) Consumer behaviour in relation to the geographical origin of food

Related to subject (1), information on the geographical origin of foods and the market situation are limited causing uncertainties in origin-related exposure assessment. Typical geographical origins of diverse food items and their proportion on the market and typical supply channels are completely unknown. Sampling in the food monitoring programmes was not representative for geographical origins of food available on the market (BVL, 2020), while data sources used for complementing the food monitoring information only partly explained the possible supply channels. For example for the herring case study, the food monitoring programmes sampled the Baltic Sea for Germany and the Norwegian Sea for Norway, while data of the Mintel GNPD indicated further relevant fishing areas for packaged herring from both countries (Fechner et al., 2019a; Fechner et al., 2020) (chapters 2.4 and 2.5). Information in the Mintel GNPD (Mintel International Group Ltd., 2020) could give geographical origin information on packaged products, but the unpackaged foodstuffs are not part of the evaluations and brands launching constantly new versions are overrepresented. Furthermore, primary production data (FAO, 2020) and trade data (Eur. Commission, 2020; UN, 2020) only provide aggregated information, which can be used to evaluate the general situation in comparison to the information documented in the food monitoring data, but it cannot directly be linked to substance concentrations. Thus, the real market situation on geographical origins of food available might not be depicted sufficiently in the data sources available for origin-related dietary exposure assessment at the moment. Additionally, unspecific and implausible geographical origin information complicated the exposure assessments. This could be related to the legal situation in Europe, as the required labelling is limited to some food items (mostly agricultural products) and the regulations do not exist for many processed food items like cocoa powder (D'Elia et al., 2011). Thus, sufficient origin data is missing in food monitoring programmes for most of the food items. A standardised and comprehensive geographical origin labelling of food would be the most important basis for more origin information, helping to evaluate origin-related substance concentrations and to perform dietary exposure assessment using this information. For example, since 2018 in Australia the geographical origin has to be labelled for most of the packaged food products and for unpackaged agricultural products in a standardised way using

symbols (Government of Australia, 2016). Such an initiative would help providing more comprehensive origin information in Europe as well. For example, the food labels of cocoa powder and herring documented in the Mintel GNPD proved that current labelling was not standardised (Fechner et al., 2019b; Fechner et al., 2020) (chapters 2.3 and 2.5) and it could complicate the extraction of origin information correctly from the packaging for both consumers and authorities. For a standardised and more automated transmission of information, technologies like applications using barcode information could be interesting to document available information automatically. The blockchain documentation of all stages in the supply chain (Creydt & Fischer, 2019) could also help to complement missing data, making geographical origins of food transparent and traceable for consumers and authorities. Consequently, it would be possible to generate comprehensive knowledge on the proportion of geographical origins of food available on a target market. Relevant supply channels could be identified and constantly monitored. This newly generated knowledge could be used for an origin-related representative sampling of food items in the food monitoring programmes that are used for the retrieval of information on substance concentrations. At the moment, weekly market reports for fruit and vegetables document geographical origins of products, which arrive in six German cities (BLE, 2020). This gives a first insight but not a complete market overview of geographical origins available. This could be a good starting point for future analyses and data collection. Country-specific concentrations related to the available geographical origins of food could be derived based on a comprehensive data collection, and used for refined and more realistic dietary exposure estimates in a European context.

There is a lack of information focusing on geographical relevant influences on substance concentrations in subject (2). The actual factors leading to the geographical variation of substance concentrations are not completely known. The reasons for the geographically varying substance concentrations were only assumed for the case studies considered by using the published results of specific studies of other researchers in other fields. On the one hand, influences could be directly related to the production conditions and practices at the geographical origin, as for example suggested for higher bromide concentrations in Italian tomatoes, which could be related to natural sources like the Mediterranean Sea (D'Alessandro et al., 2008; Davis et al., 2004; Sander et al., 2003) or higher cadmium concentrations in cocoa from South America in comparison to West Africa, which could be related to cadmium concentrations in soil (Abt et al., 2018; Bertoldi et al., 2016; Chavez et al., 2015; Fechner et al., 2019b, 2019c; Mounicou et al., 2003; Vītola & Ciproviča, 2016) (chapters 2.2 and 2.3). On the other hand, indirect relations to the geographical origin could be caused by the conditions when supplying food as suggested for higher aluminium concentrations in kiwifruits from non-EU countries because the kiwifruits could be packaged in cardboard produced using aluminium compounds (Robertson et al., 2014). Influences in the food supply chain would be hard to track

35

in the system of complex networks and various possible supply channels. The geographical origin of a food represents a proxy of various factors in the supply chain that may have an impact on substance concentrations. More information on geographical origins derived from extended food labelling can help to identify substances with geographical variations, which are not known up to now and hidden by the current sampling strategy in the food monitoring programmes. Based on this identification, similar research on the reasons of variable substance concentrations in food associated with the geographical origins could be promoted.

The consumers' attitude towards the geographical origin of the food purchased is an important factor for dietary exposure assessment and a lack of data was identified as subject (3). Origin-related estimates of substance intakes are needed only if the consumer focuses on certain origins. It was assumed that consumers focus on the geographical origin of food, as they a) consciously purchase food items of a specific origin enabled by food labelling or b) they have a stable purchasing behaviour related to specific places to shop or brands, which could be supplied with food and raw materials of a specific geographical origin only. It would be important to know more about the consumer behaviour in relation to the geographical origin of food to construct more realistic exposure scenarios. This would be of high interest if the geographical origin is part of the purchase decision and if the origin is more important for some food groups than for others. In a next step it would be important to know which geographical origins are in focus of respective food items and additionally, it would be interesting if consumers stick to specific places to shop or brands for different food groups. This information could be collected in a specific survey on geographical origins of food on a target market to gain first insights. Ideally, these research questions on consumer behaviour could be integrated in future consumption surveys to have a direct link to the food consumption reported.

6 Conclusion and future challenges

Approaches to the refinement of dietary exposure assessment using information on the geographical origin of food were performed based on origin-related scenarios and conducted for case studies based on individual food items and specific substances with geographically varying concentrations. The most conservative estimate was obtained for ethephon from pineapples, aluminium from kiwifruits, and aluminium and cadmium from cocoa powder in the standard exposure scenarios and influences of origin were covered already. However, in these cases, origin-related scenarios could give a refined lower estimate, which is of special interest, if health-based guidance values are exceeded. The influence of origin was not covered by standard exposure scenarios for bromide from tomatoes, as origin-related scenarios gave the most conservative estimate. In this case, origin-related scenarios are needed to evaluate the intake, if consumers focus on special origins. European exposure assessments are usually conducted by EFSA, and are based on country-specific consumption information and pooled

European concentrations. A country-specific exposure assessment on PCDD/Fs and DL-PCBs from herring consumption included country-specific concentrations and resulted in higher substance intakes for German consumers than for Norwegian consumers. In this way, country-specific concentration data related to available geographical origins on the country markets helped to evaluate the country-specific exposure situation for consumers. This could be of interest for future European exposure assessments.

The data available and used for the current research generated first insights into origin-related exposure situations for the case studies selected even if several limitations and uncertainties were identified. Food monitoring data were not representative for geographical origins of food available on the market and relations between substance concentrations in food and the geographical origin could not be fully evaluated. For a comprehensive data basis on geographical origins in future research, labelling of geographical origins of food would need to be extended and standardised or technologies need to be improved for the tracing of geographical origins of food. This would make it possible to extract information from food samples to generate a more comprehensive data basis for a representative sampling of geographical origins available on the market in food monitoring programmes. Consequently, it would be possible to find origin-related substance concentrations in various food items and to evaluate in which cases the integration of geographical origins of food is relevant for dietary exposure assessment. Integrating information on the geographical origin of food in dietary exposure assessments is relevant, if a) substance concentrations vary geographically, b) food items are supplied from different geographical origins to the target market and c) consumers focus on specific geographical origins of food at the purchase. For all these fields, the data collection would need to be improved and extended. Approaches to a refined dietary exposure assessment developed in the current research provide procedures to integrate food origin information in deterministic or semi-probabilistic exposure scenarios. Limitations and uncertainties were identified and need to be reduced in future research.

References

- Abt E, Fong Sam J, Gray P, Robin LP (2018). Cadmium and lead in cocoa powder and chocolate products in the US Market. *Food Additives & Contaminants: Part B Surveillance, 11* (2): 92-102. doi: 10.1080/19393210.2017.1420700
- Aruoma OI (2006). The impact of food regulation on the food supply chain. *Toxicology*, 221 (1): 119-127. doi: 10.1016/j.tox.2005.12.024
- Azad AM, Frantzen S, Bank MS, Nilsen BM, Duinker A, Madsen L, Maage A (2019). Effects of geography and species variation on selenium and mercury molar ratios in Northeast Atlantic marine fish communities. *Science of The Total Environment*, 652: 1482-1496. doi: 10.1016/j.scitotenv.2018.10.405
- Bertoldi D, Barbero A, Camin F, Caligiani A, Larcher R (2016). Multielemental fingerprinting and geographic traceability of Theobroma cacao beans and cocoa products. *Food Control, 65*: 46-53. doi: 10.1016/j.foodcont.2016.01.013
- Beulens AJM, Broens D-F, Folstar P, Hofstede GJ (2005). Food safety and transparency in food chains and networks - Relationships and challenges. *Food Control, 16* (6): 481-486. doi: 10.1016/j.foodcont.2003.10.010
- BfR, Bundesinstitut für Risikobewertung (German Federal Institute for Risk Assessment), Bundesinstitut für Risikobewertung (2007). BfR schlägt die Einführung eines Höchstgehalts für Cadmium in Schokolade vor. Stellungnahme Nr. 015/2007 des BfR vom 31.01.2007. 15/2007: 1-18 (18 pp.). Retrieved from http://www.bfr.bund.de/cm/343/bfr_schlaegt_die_einfuehrung_eines_hoechstgehalts fuer_cadmium_in_schokolade_vor.pdf – accessed: 2020-03-23.
- BLE, Bundesanstalt für Landwirtschaft und Ernährung (Federal Office for Agriculture and Food) (2020). *Bundesinformationszentrum Landwirtschaft Marktakteure Obst und Gemüse*. Website: Bundesanstalt für Landwirtschaft und Ernährung, Bonn, DE. <u>https://www.ble.de/DE/BZL/Daten-Berichte/Obst-Gemuese/obst-gemuese_node.html</u> accessed: 2020-04-02.
- Blume K, Lindtner O, Heinemeyer G, Schneider K, Schwarz M (2010). Aufnahme von Umweltkontaminanten über Lebensmittel (Cadmium, Blei, Quecksilber, Dioxine und PCB) Ergebnisse des Forschungsprojektes LExUKon. *BfR-Informationsbroschüre*, p. 1-56, 60 pages. Berlin DE: Bundesinstitut für Risikobewertung (BfR), Fachgruppe Expositionsschätzung und -standardisierung. Retrieved from <u>http://www.bfr.bund.de/cm/350/aufnahme von umweltkontaminanten ueber lebens</u> <u>mittel.pdf</u>.
- Brambilla G (2019). Impact of Downstream Use for the Food Chain and Downstream Use for Consumer Exposure. In: Heinemeyer G, Jantunen M, Hakkinen P (Eds.), *The Practice of Consumer Exposure Assessment*, Ch. 4.12. Cham, CH: Springer Nature Switzerland AG. doi: 10.1007/978-3-319-96148-4.
- Brombach C, Wagner U, Eisinger-Watzl M, Heyer A (2006). Die Nationale Verzehrsstudie II. *Ernährungs-Umschau, 53* (1): 4-9. Retrieved from <u>https://www.ernaehrungs-umschau.de/fileadmin/Ernaehrungs-Umschau/pdfs/pdf 2006/01 06/EU01 04 09.pdf</u>.
- BVL, Bundesamt f
 ür Verbraucherschutz und Lebensmittelsicherheit (Federal Office of Consumer Protection and Food Safety) (2020). The monitoring programme. Website: Bundesamt f
 ür Verbraucherschutz und Lebensmittelsicherheit, Brunswick, DE.

https://www.bvl.bund.de/EN/Tasks/01 Food/01 tasks/02 OfficialFoodControl/04 LM Monitoring en/LM Monitoring EN node.html – accessed: 2020-04-02.

- Chavez E, He ZL, Stoffella PJ, Mylavarapu RS, Li YC, Moyano B, Baligar VC (2015). Concentration of cadmium in cacao beans and its relationship with soil cadmium in southern Ecuador. *Science of The Total Environment,* 533: 205-214. doi: 10.1016/j.scitotenv.2015.06.106
- Creydt M & Fischer M (2019). Blockchain and more Algorithm driven food traceability. *Food Control, 105*: 45-51. doi: 10.1016/j.foodcont.2019.05.019
- D'Alessandro W, Bellomo S, Parello F, Brusca L, Longo M (2008). Survey on fluoride, bromide and chloride contents in public drinking water supplies in Sicily (Italy). *Environmental Monitoring and Assessment, 145* (1): 303-313. doi: 10.1007/s10661-007-0039-y
- D'Elia G, Alpigiani I, Bonardi S, Bacci C, Lanzoni E, Brindani F (2011). Food labelling in Europe: mandatory "country of origin" extended to more foods. *Annali della Facoltà di Medicina Veterinaria, Università di Parma, 31*: 65-79. Retrieved from https://www.cabdirect.org/cabdirect/abstract/20133207428.
- Davis SN, Fabryka-Martin JT, Wolfsberg LE (2004). Variations of Bromide in Potable Ground Water in the United States. *Groundwater*, *42* (6): 902-909. doi: *10.1111/j.1745-6584.2004.t01-8-.x*
- EFSA, CONTAM Panel , Knutsen HK, Alexander J, Barregård L, Bignami M, Brüschweiler B, Ceccatelli S, Cottrill B, Dinovi M, Edler L, Grasl-Kraupp B, Hogstrand C, Nebbia CS, Oswald IP, Petersen A, Rose M, Roudot A-C, Schwerdtle T, Vleminckx C, Vollmer G, Wallace H, Fürst P, Håkansson H, Halldorsson T, Lundebye A-K, Pohjanvirta R, Rylander L, Smith A, van Loveren H, Waalkens-Berendsen I, Zeilmaker M, Binaglia M, Gómez Ruiz JÁ, Horváth Z, Christoph E, Ciccolallo L, Ramos Bordajandi L, Steinkellner H, Hoogenboom LR (2018a). Risk for animal and human health related to the presence of dioxins and dioxin-like PCBs in feed and food. *EFSA Journal, 16* (11): 5333. doi: 10.2903/j.efsa.2018.5333
- EFSA, European Food Safety Authority (2011). Overview of the procedures currently used at EFSA for the assessment of dietary exposure to different chemical substances. *EFSA Journal, 9* (12): 2490 (*33 pp.*). doi: *10.2903/j.efsa.2011.2490*
- EFSA, European Food Safety Authority, Reich H, Triacchini GA (2018b). Occurrence of residues of fipronil and other acaricides in chicken eggs and poultry muscle/fat. *EFSA Journal, 16* (5): e05164. doi: *10.2903/j.efsa.2018.5164*
- Eur. Commission (2020). Eurostat Your key to European statistics. In: Eurostat, Luxembourg,
LU: European Commission. Retrieved from
http://epp.eurostat.ec.europa.eu/newxtweb/mainxtnet.do accessed: 2020-03-21.
- Eur. Parliament & the Council (2000). Regulation (EC) No 1760/2000 of the European Parliament and of the Council of 17 July 2000 establishing a system for the identification and registration of bovine animals and regarding the labelling of beef and beef products and repealing Council Regulation (EC) No 820/97. Official Journal of the European Communities, 204: 1-10. Retrieved from <u>http://eur-lex.europa.eu/legalcontent/EN/TXT/PDF/?uri=CELEX:32000R1760&rid=1</u>.
- Eur. Parliament & the Council (2002). Regulation (EC) No 178/2002 of the European Parliament and of the Council of 28 January 2002 laying down the general principles and requirements of food law, establishing the European Food Safety Authority and

laying down procedures in matters of food safety. *Official Journal of the European Communities,* 31: 1-24. Retrieved from <u>http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32002R0178&rid=1</u>.

- FAO, Food and Agriculture Organization of the United Nations, Food and Agriculture Organization of the United Nations (2017). The future of food and agriculture – Trends and challenges. 1-163. Retrieved from <u>http://www.fao.org/3/a-i6583e.pdf</u> – accessed: 2020-03-23.
- FAO, Food and Agriculture Organization of the United Nations (2020). FAOSTAT. In: FAOSTAT, Rome, IT: Food and Agriculture Organization of the United Nations. Retrieved from <u>http://www.fao.org/faostat/en/#data</u> – accessed: 2020-03-21.
- Fechner C, Frantzen S, Lindtner O, Mathisen GH, Lillegard ITL (2019a). Influence of the geographical origin on substance concentrations in herring as basis for dietary exposure assessments. *EFSA Journal*, *1*7. doi: *10.2903/j.efsa.2019.e170904*
- Fechner C, Greiner M, Heseker H, Lindtner O (2019b). Dietary exposure assessment of aluminium and cadmium from cocoa in relation to cocoa origin. *PLOS ONE*, 14 (6): e0217990. doi: 10.1371/journal.pone.0217990
- Fechner C, Greiner M, Heseker H, Lindtner O (2019c). Refinement of dietary exposure assessment using origin-related scenarios. *Journal of Exposure Science & Environmental Epidemiology*. doi: 10.1038/s41370-019-0117-6
- Fechner C, Heseker H, Lindtner O, Greiner M (2019d). Food Origin Information in Times of Global Food Supply – Basis for the Refinement of Dietary Exposure Assessment. *Feed* and food safety in times of global production and trade, Vol. 4/2019 Ch. 6 Exposure to Humans, p. 201 - 213. Berlin: Bundesinstitut für Risikobewertung. doi: 10.17590/20190930-102623.
- Fechner C, Frantzen S, Andersen LF, Lindtner O, Mathisen GH, Lillegaard ITL (2020). Dioxins and dioxin-like PCBs in Atlantic herring: A Norwegian – German comparison of the influence of fishing area on country-specific dietary exposure. *submitted 2020*.
- Frey HC & Patil SR (2002). Identification and Review of Sensitivity Analysis Methods. *Risk Analysis,* 22 (3): 553-578. doi: 10.1111/0272-4332.00039
- Government of Australia (2016). Country of Origin Food Labelling Information Standard 2016. *Australian Consumer Law (ACL) - Competition and Consumer Act 2010, section 134* (2): 42. Retrieved from <u>https://www.legislation.gov.au/Details/F2017C00920</u>.
- Heinemeyer G, Mosbach-Schulz O, Kreienbrock L, Schümann M, Filter M, Greiner M, Herzler M, Lindtner O, Kurzenhäuser S, Röder B, Bundesinstitut für Risikobewertung (BfR) (2015). Guidance document on uncertainty analysis in exposure assessment. Recommendation of the Committee for Exposure Assessment and Standardisation of the Federal Institute for Risk Assessment (BfR). Website *BfR-Wissenschaft* (03/2015) (56 pp.). Retrieved from http://www.bfr.bund.de/cm/350/guidelines-on-uncertainty-analysis-in-exposure-assessments.pdf accessed: 2018-02-28.
- Karl H & Lahrssen-Wiederholt M (2013). Factors influencing the intake of dioxins and dioxinlike PCBs via fish consumption in Germany. *Journal für Verbraucherschutz und Lebensmittelsicherheit - Journal of Consumer Protection and Food Safety, 8* (1-2): 27-35. doi: 10.1007/s00003-013-0805-4

- Krems C, Bauch A, Götz A, Heuer T, Hild A, Möseneder J, Brombach C (2006). Methoden der Nationalen Verzehrsstudie II. *Ernährungs Umschau*, *53* (2): 44-50. Retrieved from <u>https://www.ernaehrungs-umschau.de/fileadmin/Ernaehrungs-</u> Umschau/pdfs/pdf 2006/02 06/EU02 44 50.pdf.
- Kruszewski B, Obiedziński MW, Kowalska J (2018). Nickel, cadmium and lead levels in raw cocoa and processed chocolate mass materials from three different manufacturers. *Journal of Food Composition and Analysis, 66*: 127-135. doi: 10.1016/j.jfca.2017.12.012
- Lindtner O, Berg K, Blume K, Fiddicke U, Heinemeyer G (2013). The German approach to estimating dietary exposures using food monitoring data. In: Moy GG & Vannoort RW (Eds.), *Total Diet Studies*, Ch. 53, p. 521-530. New York, US: Springer. Retrieved from https://link.springer.com/chapter/10.1007/978-1-4419-7689-5_53/fulltext.html.
- Mintel International Group Ltd. (2020). Global New Product Database (GNPD) (Web page). In: Global New Product Database (GNPD): Mintel International Group Ltd. Retrieved from http://www.gnpd.com/sinatra/home/ – accessed: 2020-03-21.
- Mounicou S, Szpunar J, Andrey D, Blake C, Lobinski R (2003). Concentrations and bioavailability of cadmium and lead in cocoa powder and related products. *Food Additives & Contaminants, 20* (4): 343-352. doi: *10.1080/0265203031000077888*
- Müller M, Anke M, Illing-Günther H (1998). Aluminium in foodstuffs. *Food Chemistry*, 61 (4): 419-428. Retrieved from https://www.sciencedirect.com/science/article/pii/S030881469700085X.
- Ofori-Frimpong K, Asamoah G, Appiah M (1999). Distribution of free and total aluminium in some cocoa-growing soils of Ghana. *Ghana Journal of Agricultural Science, 32* (1): 101-108. Retrieved from https://www.ajol.info/index.php/gjas/article/view/1920.
- Robertson K, Garnham M, Symes W (2014). Life cycle carbon footprint of the packaging and transport of New Zealand kiwifruit. *The International Journal of Life Cycle Assessment, 19* (10): 1693-1704. doi: *10.1007/s11367-014-0775-5*
- Sander R, Keene WC, Pszenny AAP, Arimoto R, Ayers GP, Baboukas E, Cainey JM, Crutzen PJ, Duce RA, Hönninger G, Huebert BJ, Maenhaut W, Mihalopoulos N, Turekian VC, Van Dingenen R (2003). Inorganic bromine in the marine boundary layer: a critical review. *Atmospheric Chemistry and Physics*, 3 (5): 1301-1336. doi: 10.5194/acp-3-1301-2003
- Sarvan I, Bürgelt M, Lindtner O, Greiner M (2017). Expositionsschätzung von Stoffen in Lebensmitteln: Die BfR-MEAL-Studie – die erste Total-Diet-Studie in Deutschland [Dietary exposure assessment of substances in foods: The BfR MEAL study – The first German total diet study]. Bundesgesundheitsblatt – Gesundheitsforschung – Gesundheitsschutz, 60 (7: Kontaminanten in Lebensmitteln): 689-696. doi: 10.1007/s00103-017-2566-1
- Schneider K, Schwarz MA, Lindtner O, Blume K, Heinemeyer G (2014). Lead exposure from food: the German LExUKon project. Food Additives and Contaminants, Part A: Chemistry, Analysis, Control, Exposure and Risk Assessment, 31 (6): 1052–1063. doi: 10.1080/19440049.2014.905875
- Schwarz MA, Lindtner O, Blume K, Heinemeyer G, Schneider K (2014). Cadmium exposure from food: The German LExUKon project. *Food Additives and Contaminants, Part A:*

Chemistry, Analysis, Control, Exposure and Risk Assessment, 31 (6): 1038–1051. doi: 10.1080/19440049.2014.905711

- Sunderland EM, Li M, Bullard K (2018). Decadal Changes in the Edible Supply of Seafood and Methylmercury Exposure in the United States. *Environmental Health Perspectives, 126* (1): 017006. doi: 10.1289/EHP2644
- Tennant DR & Bruyninckx C (2018). The potential application of European market research data in dietary exposure modelling of food additives. *Food Additives & Contaminants: Part A, 35* (3): 412-424. doi: 10.1080/19440049.2017.1400187
- Tietz T, Lenzner A, Kolbaum AE, Zellmer S, Riebeling C, Gürtler R, Jung C, Kappenstein O, Tentschert J, Giulbudagian M, Merkel S, Pirow R, Lindtner O, Tralau T, Schäfer B, Laux P, Greiner M, Lampen A, Luch A, Wittkowski R, Hensel A (2019). Aggregated aluminium exposure: risk assessment for the general population. *Archives of Toxicology*, 93 (12): 3503-3521. doi: 10.1007/s00204-019-02599-z
- Totland TH, Melnæs BK, Lundberg-Hallén N, Helland-Kigen KM, Lund-Blix NA, Myhre JB, Johansen AMW, Løken EB, Andersen LF (2012). Norkost 3. En landsomfattende kostholdsundersøkelse blant menn og kvinner i Norge i alderen 18–70 år, 2010–11. Vol. IS-2000. Oslo, NO: Universitetet i Oslo, Mattilsynet, Helsedirektoratet. Retrieved from <u>https://www.helsedirektoratet.no/rapporter/norkost-3-en-landsomfattendekostholdsundersokelse-blant-menn-og-kvinner-i-norge-i-alderen-18-70-ar-2010-11/</u> – accessed: 2020-02-04.
- Tuomisto JT, Asikainen A, Meriläinen P, Haapasaari P (2020). Health effects of nutrients and environmental pollutants in Baltic herring and salmon: a quantitative benefit-risk assessment. *BMC Public Health, 20* (1): 64. doi: 10.1186/s12889-019-8094-1
- UN, United Nations (2020). UN Comtrade Database International Trade Statistics Database (Web page). In: United Nations. Retrieved from <u>https://comtrade.un.org/data</u> accessed: 2020-03-21.
- Vītola V & Ciproviča I (2016). The effect of cocoa beans heavy and trace elements on safety and stability of confectionery products. *Rural Sustainability Research,* (No.35): 19-23. doi: 10.1515/plua-2016-0003
- Weiser AA, Thöns C, Filter M, Falenski A, Appel B, Käsbohrer A (2016). FoodChain-Lab: A Trace-Back and Trace-Forward Tool Developed and Applied during Food-Borne Disease Outbreak Investigations in Germany and Europe. *PLOS ONE, 11* (3): 1-11. doi: 10.1371/journal.pone.0151977
- WHO, World Health Organization & FAO, Food and Agriculture Organization of the United Nations (2009a). Dose-response assessment and derivation of health-based guidance values. In: IPCS, Inter-Organization Programme for the Sound Management of Chemicals (Ed.), *Principles and Methods for the Risk Assessment of Chemicals in Food*, Series: *Environmental Health Criteria*, Vol. 240, Ch. 5, p. 5.1-5.61. Geneva, CH: World Health Organisation. Retrieved from http://apps.who.int/iris/bitstream/10665/44065/8/WHO EHC 240 8 eng Chapter5.p df?ua=1 accessed: 2020-03-23.
- WHO, World Health Organization & FAO, Food and Agriculture Organization of the United Nations (2009b). Dietary exposure assessment of chemicals in food. In: IPCS, Inter-Organization Programme for the Sound Management of Chemicals (Ed.), *Principles* and Methods for the Risk Assessment of Chemicals in Food, Series: Environmental Health Criteria, Vol. 240, Ch. 6, p. 6.1-6.95. Geneva, CH: World Health Organisation.

Retrieved

from http://apps.who.int/iris/bitstream/10665/44065/9/WHO EHC 240 9 eng Chapter6.p <u>df?ua=1</u> – accessed: 2020-03-23.

Xiu C & Klein KK (2010). Melamine in milk products in China: Examining the factors that led to deliberate use of the contaminant. Food Policy, 35 (5): 463-470. doi: https://doi.org/10.1016/j.foodpol.2010.05.001

Appendix: Poster

Poster 1

Fechner C, Blume K, Greiner M, Sommerfeld C, Lindtner O (2017). Refinement of Dietary Exposure Assessment in the Context of Global Food Supply – Case Study: Aluminium in Kiwi fruits. Joint International Symposium: Global Past, Present and Future Challenges in Risk Assessment – Strengthening Consumer Health Protection, 30/11 – 01/12/2017. Abstracts, p. 139, Abstract No. 14 (poster abstract). Berlin, Germany: German Federal Institute for Risk Assessment (BfR).

Refinement of Dietary Exposure Assessment in the Context of Global Food Supply – Case Study: Aluminium in Kiwi fruits



Carolin Fechner, Katrin Blume, Matthias Greiner, Christine Sommerfeld, Oliver Lindtner

Background

Food supply is becoming more global (FAO, 2015). Agricultural products are available from different countries of origin for the consumer and for further processing in food industry. In connection with dietary exposure assessment, as an important base for risk assessment in food safety and consumer protection (Sarvan *et al.*, 2017), the relation between substance concentration and geographical origin of food is of special interest. In the food supply chain there are different influences on substances and finally on dietary exposure possible (Fig. 1)

On one hand limited obligation to label the country of origin especially for ingredients in processed foods (D'Elia et al., 2011) is a challenge while connecting food origin with substance concentrations (contaminants or residues) in a refined dietary exposure assessment.

On the other hand more comprehensive information is expected from activities to better describe the global food chain in terms to prevent and to be prepared for food crisis as an example. The influence of origin-related substance concentrations in deterministic dietary exposure assessment has been considered using data for aluminium in kiwi fruits from German Food Monitoring 2010 and consumption data from German National Nutrition Survey II (NVS II).



Fig. 2: Standard scenarios for deterministic dietary exposure assessmer adapted to origin-related scenarios for aluminium from kiwi fruits ent (according to Sarvan et al., 2017)

Results

For aluminium in kiwi fruits the mean concentration of 2.875 mg/kg from Non-EU region is 2.9-fold and significant higher than 0.990 mg/kg from all samples (Tab. 1).

The chronic dietary aluminium exposure for consumers of kiwi fruits for average consumption and mean concentration considering all samples without distinction of origin is 0.002 mg/week/kg bw and the most conservative standard scenario (95th percentile consumption and concentration) results in intake estimates of 0.024 mg/week/kg bw.

When using the mean concentration of that origin with highest concentrations aluminium exposure of 0.014 mg/week/kg bw is calculated (Fig. 3). The highest calculated exposure represents 2.4 % of the tolerable weekly intake (TWI) of 1 mg/week/kg bw (EFSA, 2008).

Conclusions

The most conservative standard scenario results in the highest exposure value and covers the origin-related influence on aluminium concentrations in kiwi fruits. For chronic aluminium exposure from kiwi fruits origin-specific scenarios do not have to be performed if all standard scenarios are considered. However, origin-specific scenarios provide more detailed information on possible origin-related influences

References

D'Elia G, Alpigiani I, Bonardi S, Bacci C, Lanzoni E, Brindani F (2011). Food labelling in Europe: mandatory "country of origin" extended to more foods. Annali della Facoltà di Medicina Veterinaria. Università di Parma, 31: 65-79

EFSA, European Food Safety Authority (2008), Safety of aluminium from dietary intake - Scientific Opinion of the Panel on Food Additives, Flavourings, Processing Aids and Food Contact Materials (AFC). EFSA Journal, 6 (7): 754-n/a.

FAO, Food and Agriculture Organization of the United Nations (2015). The State of Agricultural Commodity Markets. Trade and food security: achieving a better balance between national priorities and the collective good. Food and Agriculture Organization of the United Nations. Retrieved from.

Krems C, Bauch A, Götz A, Heuer T, Hild A, Möseneder J, Brombach C (2006). Methoden der Nationalen Verzehrsstudie II [Methods of the National Nutrition Survey II]. Ernährungs Umschau, 53 (2): 44-50.



Fig. 1: Possible influences within the food supply chain on substance concentration and dietary exposure

Methods

Data from the German Food Monitoring 2010 was used to obtain aluminium concentrations in kiwi fruits in connection with information on origin (Sieke et al., 2008). A modified lower bound approach was applied for the determination of mean, standard deviation and percentile 95 replacing non-detects with zero and non-guantified values with the limit of detection (LOD) (WHO & FAO, 2009)

Aluminium concentrations were grouped origin-related and examined for significant differences to Auminium concentrations were grouped ongin-related and examined for significant dimerences to compare all samples with samples from origins having higher alluminium concentrations. To obtain consumption data the dietary history interview (DISHES) of NVS II was used (Krems *et al.*, 2006). Recipe codes *xy* were decoded previously by German Federal Institute for Risk Assessment (BR) using Bundeslebensmittlesholtissel (BLS) recipes (wersion II.4) to derive consumption amounts of unprocessed food items which were used in household recipes.

Typically applied standard scenarios of deterministic dietary exposure assessments were adapted to origin-related questions and calculated by combining different distribution parameters of substance concentration and food consumption (Sarvan et al., 2017) (Fig. 2). Statistical analyses were carried out using SPSS version 21 and Microsoft Excel 2010

Tab. 1: Aluminium concentrations in kiwl fruits grouped by geographic origin

samples grouped by origin	nq	n uantifiable	mean *	standard deviation *	percentile 95 *
all **	163	112	0.990	1.735	4.773
EU region ***	121	79	0.470	0.871	1.58
Non-EU region ****	36	31	2.875	2.572	9.86
THE Chile (n = 7) Now	Zealand (n	- 20)		Accessed.	
mean consumption and mean concentration e all samples -	0.002	- 28)			photo by treepik co

chronic dietary aluminium exposure for consumers of kiwi fruits [mg/week/kg bw]

Fig. 3: Chronic dietary exposure assessment for aluminium from kiwl fruits using two standard scer In comparison to one origin-related scenario

Sarvan I, Bürgelt M, Lindtner O, Greiner M (2017). Expositionsschätzung von Stoffen in Lebensmitteln: Die BfR-MEAL-Studie - die erste Total-Diet-Studie in Deutschland [Dietary exposure assessment of substances in foods: The BfR MEAL study - The first German total diet study]. Bundesgesundheitsblatt - Gesundheitsforschung - Gesundheitsschutz, 60 (7: Kontaminanten in Lebensmitteln): 689-696.

Sieke C, Lindtner O, Banasiak U (2008). Pflanzenschutzmittelrückstände, Nationales Monitoring - Abschätzung der Verbraucherexposition: Teil 1 [German food monitoring - Refined design for consumer exposure assessment: Part 1]. Deutsche Lebensmittel-Rundschau, 104 (6): 271-279. WHO, World Health Organization & FAO, Food and Agriculture Organization of the United Nations (2009). Dietary exposure assessment of chemicals in food. In: IPCS, Inter-Organization Programme for the Sound Management of Chemicals (Ed.), Principles and Methods for the Risk Assessment of Chemicals in Food, Series: Environmental Health Criteria, Vol. 240, Ch. 6, p. 6.1-6.95. Geneva, CH: World Health Organisation.

German Federal Institute for Risk Assessment • Max-Dohrn-Str. 8-10 • 10589 Berlin, GERMANY • Phone +49 30-18412-0 • Fax +49 30-18412-4741 • bfr@bfr.bund.de • www.bfr.bund.de/en

Poster 2

Fechner C, Lindtner O, Mathisen GH, Lillegaard ITL (2019): Food origin as an uncertainty in dietary exposure assessment. International Conference on Uncertainty in Risk Analysis – Challenges and Advances in Assessing, Managing and Communicating Uncertainty, 20 – 22/02/2019. Abstracts, p. 99, Abstract No. 2.4.2 (poster abstract). Berlin, Germany: German Federal Institute for Risk Assessment (BfR), European Food Safety Authority (EFSA).





) trout, cod, plaice

Federal Office of Cons

progra

Protection and Food Safety (Berlin, Germany)

German National Nutrition Survey II (NVS II) (2006): • 2 24-h recalls by teleph within 6 weeks

survey

Figure 2: Norwegian and German data sources for origin-related

dietary exposure assessment of contaminants in fish.

13926 participants aged 14-

80 years fish items were part of the

lin, Germany) German Food Monitoring

stitute of Marine Research

m fou

Food cons

(Bergen, Norway)
 Monitoring progra farmed fish

Seafood datab

Norkost 3 (2010/2011): 2 24-hour recalls by telephone at least one month apart

1787 men and women aged 18-70 years 97 different fish and fish

containing foods were

reported

Food origin as an uncertainty in dietary exposure assessment

Carolin Fechner (1), Oliver Lindtner (2), Gro Haarklou Mathisen (1), Inger Therese Laugsand Lillegaard (1)

(1) Norwegian Scientific Committee for Food and Environment (VKM), (2) German Federal Institute for Risk Assessment (BfR

Key message:

The origin-related grouping of contaminant concentrations in fish could be used as a basis for a refined dietary exposure assessment referring to both the Norwegian and the German situation.

Introduction

The EU-FORA fellowship programme gives the possibility for international cooperation as VKM (Norwegian Scientific Committee for Food and Environment) in Norway and BfR (German Federal Institute for Risk Assessment) in Germany are part of the 2018/2019 cohort. The project launched in this cooperation is concerned about food origin as source of uncertainty in dietary exposure assessment. Fish as a food is supplied from different catching areas (Figure 1). Since fish contains various contaminants, and relations between catching area and contaminant concentration are observed in different studies. fish is chosen as example (1 - 4).

Methods

Norwegian and German data on contaminant concentrations are considered for selected fish species (Figure 2). Contaminant concentrations are grouped by catching areas of fish to identify if different contamination levels exist for different areas.

Results

Differences in available fish from various catching areas between Norway and Germany are evaluated. This project will investigate how, and to what extent, uncertainty can be introduced to dietary exposure assessment if fish origin influences contaminant concentrations significantly, but originrelated arouping is not considered in the exposure estimate.

Discussion

The origin-related grouping of contaminant concentrations in fish using catching areas could be used as a basis for a refined exposure assessment referring to both the Norwegian and the German situation. Previous investigations on agricultural products show influences of origin grouping on dietary exposure (5), which could be also relevant for fish from different catching areas. This is of special importance if European risk assessments are carried out combining concentration data recorded in several countries without taking food origin into account.



Figure 1: Conceptual supply chain of fish to the target markets Norway and Germany.

References

- 1. Azad AM, Frantzen S, Bank MS, Nilsen BM, Duinker A, Madsen L, et al. Effects of geography and species variation on selenium and mercury molar ratios in Northeast Atlantic marine fish communities. Science of the
- Azad AM, Frantzen S, Bank MS, Nilsen BM, Duinker A, Madsen L, et al. Effects of geography and species variation on selenium and mercury molar ratios in Northeast Atlantic marine fish communities. Science of Total Environment. 2019; 652: 1482-96.
 Julshamn K, Duinker A, Nilsen BM, Frantzen S, Maage A, Valdersnes S, et al. A baseline study of levels of mercury, arsenic, cadmium and lead in Northeast Arctic cod (Gadus morhua) from different parts of the Barents Sea. Marine Pollution Bulletin. 2013; 67 (1-2): 187-95.
 Karl H, Lahrssen-Wiederholt M. Factors influencing the intake of dioxins and dioxin-like PCBs via fish consumption in Germany. Journal of Consumer Protection and Food Safety. 2013; 8 (1-2): 27-35.
 Sunderland EM, Li M, Bullard K. Decadal changes in the edible supply of seafood and methylmercury exposure in the United States. Environmental Health Perspectives. 2018; 126(1): 017006-1--6.
 Fechner C, Greiner M, Heseker H, Lindtner O. Refinement of dietary exposure assessment using origin-related scenarios. J Expo Sci Environ Epidemiol. 2019; accepted 11 January 2019.

Norwegian Scientific Committee for Food and Environment (VKM), PO Box 4404 Nydalen, N-0403 Oslo, Norway. Phone: +47 21 62 28 27, E-mail: Carolin.Fechner@vkm.no, Internet: vkm.no

Poster 3

Fechner C, Frantzen S, Andersen LF, Lindtner O, Mathisen GH, Lillegaard ITL (2019). The influence of fish catching area on dietary exposure. Science Conference "The Science of Food Safety – What's our Future?", 21 – 22/08/2019. Abstracts (poster abstract). Dublin, Ireland: Food Safety Authority of Ireland (FSAI).





The influence of fish catching area on dietary exposure

Carolin Fechner (1), Sylvia Frantzen (2), Lene Frost Andersen (3), Oliver Lindtner (4), Gro Haarklou Mathisen (1), Inger Therese L. Lillegaard (1)

(1) Norwegian Scientific Committee for Food and Environment (VKM), Oslo, Norway (2) Institute of Marine Research (IMR), Bergen, Norway (3) University of Oslo, Oslo, Norway (4) German Federal Institute for Risk Assessment (BfR), Berlin,

Key message:

Dietary exposure assessment is refined using origin-related concentrations of dioxins and DL-PCBs in herring, as they are 2-fold higher from the Baltic Sea than from the Norwegian Sea. This exemplifies the importance of food origin in exposure assessment.

Introduction

Relations between the catching area of fish and the contaminant concentration in fish were shown in several studies (1 - 4). Previous investigations on agricultural products show influences of origin grouping on dietary exposure (5), which could also be relevant for fish. We aim to refine dietary exposure assessment using information on food origin. Herring has been chosen as a case study because it is supplied from different catching areas and consumed in Norway and Germany. The present work is a collaboration between VKM (Norwegian Scientific . Committee for Food and Environment) in Norway and BfR (German Federal Institute for Risk Assessment) in Germany initiated by the EU-FORA fellowship programme.

Methods

To investigate if typical herring stocks from Norway and Germany have different contaminant concentrations, monitoring data between 2011 and 2017 is considered. Norway provides data of commercial catching areas of the Norwegian Spring Spawning herring stock while Germany has market data available. Contaminant concentrations are grouped by catching areas of existing herring stocks. Country-specific consumption data of two 24hour recalls and assumptions on fish origin are used to calculate the dietary exposure. Extrapolating long-term consumption from two 24-hour recalls causes an overestimation for high consumers of herring.

Results and Discussion

Effects of origin-related concentrations and country-specific consumption on dietary exposure are evaluated. For herring, the Norwegian Sea and the Baltic Sea are catching areas of typical stocks in Norway and Germany and contaminant concentrations in herring from these catching areas are used for a refined exposure assessment (Figure 1).

Considering the food origin is of special importance if European risk assessments are carried out combining concentration data recorded in several countries. If substance concentrations are influenced by food origin, this relation should be taken into account in exposure assessment for food with different origins eaten in different countries.

References

- 1. Karl, H. and M. Lahrssen-Wiederholt (2013). "Factors influencing the intake of dioxins and dioxin-like PCBs via fish consumption in Germany." Journal of Consumer Protection and Food Safety 8(1-2): 27-35 2. Frantzen, S. and A. Måge (2016). Fremmedstoffer i villfisk med vekt på kyst nære farvann. Brosme, lange og bifangstvann. Gjelder tall for prøver samlet inn i 2013-2015. Bergen, NO, Nasjonalk institutt for
- Karl, H. and M. Lahrssen-Wiederholt. (2013). "Factors influencing the intake of dioxins and dioxin-like PCBs via fish consumption in Germany." Journal of Consumer Protection and Food Safety 8(1-2): 27-35.
 Franzten, S. and A. Máge (2016). Fremmedstoffer i villfisk med vekt på kyst nære farvann. Brosme, lange og bifangstvann. Gjelder tall for prøver samlet inn i 2013-2015. Bergen, NO, Nasjonalt institut for ernærings- og sjømatforskning (NIFES): 1-116.
 Sunderland, E. M., M. Li and K. Bullard (2018). "Decadal changes in the elible supply of seafood and methylmercury exposure in the United States." Environmental Health Perspectives 126(1): 017006-017001-017006-017006.
 Azad, A. M., S. Frantzen, M. S. Bank, B. M. Nilsen, A. Duinker, L. Madsen and A. Maage (2019). "Effects of geography and species variation on selenium and mercury molar ratios in Northeast Atlantic marine fish communities." Science of the Total Environment 652: 1482-1496.
 Fechner, C. M. Greiner, H. Heseker and O. Lindmer (2019). "Refinement of dietary exposure assessment using origin-related scenarios." Journal of Exposure Science & Environmental Epidemiology.
 FESA, CONTAM Panel (EFSA Panel on Contaminants in the FOod Chain), H. K. Knutsen, J. Alexander, L. Barregfard, M. Bignami, B. Brüschweiler, S. Ceccatelli, B. Cottrill, M. Dinovi and L. Edler (2018). "Risk for animal and humar health related to the presence of dioxins and dioxin." FEPSA Journal 16(11): 5333.
 Photoss: food and seagull pixabay.com; school of herring public/domainpictures.net

Norwegian Scientific Committee for Food and Environment (VKM), PO Box 4404 Nydalen, N-0403 Oslo, Norway. Phone: +47 21 62 28 27, E-mail: Carolin.Fechner@vkm.no, Internet: vkm.no



Figure 1: Origin-related dietary exposure to the sum of PCDD/Fs (dioxins) and DL-PCBs by herring consumption calculated using Norwegian and German data. PCDDs polychlorinated dibenzo-p-dioxins, PCDFs polychlorinated dibenzofurans, DL-PCBs dioxin-like polychlorinated biphenyls, P50 percentile 50, BW body weight, TWI tolerable weekly intake.