



National Institute for Public Health  
and the Environment  
*Ministry of Health, Welfare and Sport*

## **Mineral Oils in food; a review of occurrence and sources**

RIVM Letter report 2019-0048  
D. Buijtenhuijs | B.M. van de Ven





National Institute for Public Health  
and the Environment  
*Ministry of Health, Welfare and Sport*

## **Mineral Oils in food; a review of occurrence and sources**

RIVM Letter report 2019-0048

D. Buijtenhuijs | B.M. van de Ven

## Colophon

© RIVM 2019

Parts of this publication may be reproduced, provided acknowledgement is given to the: National Institute for Public Health and the Environment, and the title and year of publication are cited.

DOI 10.21945/RIVM-2019-0048

D. Buijtenhuijs (author), RIVM  
B.M. van de Ven (author), RIVM

Contact:

Daan Buijtenhuijs

Volksgesondheid en Zorg\Centrum Voeding, Preventie en  
Zorg\Voedselveiligheid

Daan.buijtenhuijs@rivm.nl

This investigation was performed by order, and for the account, of  
NVWA, within the framework of RBT, assignment 9.1.70

This is a publication of:

**National Institute for Public Health  
and the Environment**

P.O. Box 1 | 3720 BA Bilthoven

The Netherlands

[www.rivm.nl/en](http://www.rivm.nl/en)

## Synopsis

### **Mineral Oils in food; a review of occurrence and sources**

Mineral oils may be present in food because they have been added or they may have ended up in foods as contaminants. Measures have been taken and enforced over recent decades that have reduced the quantities in foods. Based on the levels that are known currently, the National Institute for Public Health and the Environment (RIVM) does not anticipate any adverse health effects in the Netherlands.

This is concluded based on a review of the occurrence and sources of mineral oils in foodstuffs. Mineral oils are used in many stages of the production, preparation, distribution and storage of food. This may be as a crop protection agent, for example, or as a lubricant for food processing machinery, a food additive or an additive to plastic packaging materials. The composition of the mineral oils used is different for each application.

Mineral oils consist of two groups of compounds: saturated hydrocarbons (MOSH) and aromatic hydrocarbons (MOAH). The potential adverse health effects of these two groups are different. MOSH and MOAH compounds in foodstuffs come primarily from refined oils. MOAH from insufficiently refined oils can be carcinogenic, even at low exposure levels. For that reason, they are not allowed to be used in the food production chain. Cocoa beans, rice and nuts may for example not be imported if they are packed in jute bags that have been treated with non-refined oils.

Despite the general drop in the levels of mineral oils in foodstuffs, high levels are still sometimes measured. To find out where these come from and what foods comprise the major sources of exposure, the European Commission called upon its member states in 2017 to measure the levels of mineral oils in food products. In the Netherlands, this task is being carried out by the Food and Consumer Products Safety Authority (NVWA). The measurements can be used for determining which food products lead to the highest mineral oils intake. It is then also possible to investigate the sources of these mineral oils. The possibility for further measures can then be examined.

Keywords: mineral oil hydrocarbons, MOSH, MOAH, source, risk assessment, toxicity, food contact materials, foodstuffs.



## Publiekssamenvatting

### **Minerale Oliën in voedsel; een overzicht van het voorkomen en de bronnen**

Minerale oliën kunnen in voedsel zitten doordat ze eraan zijn toegevoegd, of er als verontreiniging in zijn terechtgekomen. Door maatregelen en handhaving zijn de hoeveelheden in voedsel de laatste decennia afgenomen. Op basis van de gehalten die tot nu toe bekend zijn, verwacht het RIVM in Nederland geen schadelijke gezondheidseffecten.

Dit blijkt uit een evaluatie van beschikbare kennis over minerale oliën in voedsel en bronnen van waaruit minerale oliën in voedsel terecht kunnen komen. Minerale oliën worden in verschillende stappen van de productie, bereiding, distributie en opslag van voedsel gebruikt. Bijvoorbeeld als gewasbeschermingsmiddel, als smeerolie voor voedselverwerkende machines, als voedseladditief, of als toevoeging in plastic verpakkingsmateriaal. Per toepassing is de samenstelling van minerale oliën anders.

Minerale oliën bestaan uit twee groepen stoffen: verzadigde koolwaterstoffen (MOSH) en aromatische koolwaterstoffen (MOAH). De mogelijke schadelijke gezondheidseffecten van deze groepen verschillen. MOSH en MOAH in voedsel zijn voornamelijk afkomstig van gezuiverde oliën. MOAH uit onvoldoende gezuiverde oliën kunnen al bij een lage blootstelling kankerverwekkend zijn. Daarom mogen deze oliën in de voedselketen niet worden gebruikt. Zo mogen bijvoorbeeld cacao, rijst en noten niet worden geïmporteerd als deze in juten zakken zijn verpakt die met ongezuiverde oliën zijn behandeld.

Ondanks de algemene daling van minerale oliën in voedsel, worden er soms nog hoge gehalten gemeten. Om te achterhalen waar ze vandaan komen en welke levensmiddelen een belangrijk aandeel leveren in de blootstelling, heeft de Europese Commissie in 2017 lidstaten opgeroepen gehalten van minerale oliën in voedselproducten te meten. In Nederland wordt dit uitgevoerd door de Nederlandse Voedsel- en Warenautoriteit (NVWA). Op basis van de verzamelde meetgegevens, en wat de belangrijkste bronnen lijken te zijn van waaruit de minerale oliën in de producten terechtkomen, kan worden onderzocht welke maatregelen mogelijk zijn.

Kernwoorden: minerale olie koolwaterstoffen, MOSH, MOAH, bronnen, risicobeoordeling, toxiciteit, voedselcontactmaterialen, voedsel.





## Contents

### **Summary — 9**

- 1 Introduction — 13**
- 2 Literature search — 15**
- 3 Occurrence in food and migration from materials — 17**
  - 3.1 Concentration of mineral oils in food products — 17
  - 3.2 Dietary exposure assessment the Netherlands — 22
  - 3.3 Sources of mineral oils in food — 24
  - 3.4 Identifying the source — 28
  - 3.5 Conditions of influence on the migration of MOH into foods — 29
- 4 Analysis techniques — 41**
- 5 Risk assessment of concentrations MOSH and MOAH found in monitoring — 43**
  - 5.1 Toxicity and risk assessment of MOSH — 43
  - 5.2 Toxicity and risk assessment of MOAH — 45
  - 5.3 Conclusion toxicity and risk assessment MOSH and MOAH — 47
- 6 Discussion — 49**
- 7 Recommendations — 51**
- 8 Acknowledgements — 53**
- 9 References — 55**



## Summary

In 2012, the European Food Safety Authority (EFSA) published a scientific opinion on mineral oil hydrocarbons (MOH) in food (EFSA, 2012). Although, due to insufficient data, no tolerable daily intakes (TDI) for MOH could be established, it was concluded that exposure to MOH via food intake in Europe was of potential concern. MOAH may act as genotoxic carcinogens, while some MOSH can accumulate in human tissue and may cause adverse effects in the liver. To better understand the relative presence of MOSH and MOAH in food commodities that are major contributors to dietary exposure, the European Commission issued in 2017 a “Recommendation for monitoring of MOH in food and in materials and articles intended to come into contact with food” (EC, 2017). In the Netherlands, this monitoring is done by the ‘Netherlands Food and Consumer Product Safety Authority’ (Nederlandse Voedsel- en Warenautoriteit; NVWA).

In order to explain the concentrations of MOH found in the monitoring and to assess the risks of MOH in food, the NVWA requested the National Institute for Public Health and the Environment (Rijksinstituut voor Volksgezondheid en Milieu; RIVM) to: 1) evaluate the current knowledge on migration of MOH to food and 2) give propositions to assess the risks of MOH found in the monitoring. In response to this request the RIVM screened the literature on the migration of mineral oil published since the EFSA opinion of 2012, focusing on the concentration of MOH in food products, the sources of MOH in foods and conditions influencing the migration of MOH to foods. In addition, it proposed an approach for the risk assessment of MOH in food.

### *Concentration data*

Concerning concentrations of MOH in foods, only a few new studies were found. Most of the reported MOSH concentrations are below 10 mg/kg, with some exceptions including pasta (133 mg/kg) and sweets (84 mg/kg). For MOAH, most measured concentrations are below 0.5 mg/kg with some exceptions exceeding to 2-3 mg/kg. The highest mean concentrations of MOSH were reported for pasta, cacao powder, coffee, tea, chocolate flakes and sweets. For MOAH, concentrations were reported highest in pasta, vegetable oils, chocolate flakes, cocoa and coffee beans. No conclusions could be drawn on the source of the contaminations found in foods.

### *Dietary intake assessment*

To address the question as to which food groups contribute the most to mineral oil exposure via food, a dietary intake assessment of the RIVM (Van de Ven *et al.*, 2018) was included. Here, a median (P50) and high (P95) level of exposure was calculated for persons aged 2 to 6 years old and 7 to 69-year olds. Exposure to the younger age group was approximately a factor two higher than in the older age group, with the high exposure resulting in MOSH and MOAH exposures of 0.40 and 0.028 mg/kg bw per day in 2 to 6-year olds. For this age group, the food groups contributing most to the total mineral oil exposure via food intake were ‘confectionary’ (non-chocolate), ‘pasta’, ‘ice and desserts’

and 'vegetable products'. For persons aged 7 to 69 years old the food groups contributing the most to mineral oil exposure were 'pasta' and 'herbs, spices and condiments'. The contribution of foods packed in recycled paperboard to the total exposure via food intake was 15% (2 to 6-year olds) and 18% (7 to 69-year olds) calculated for the high exposure level (P95).

#### *Sources*

Regarding the potential sources of MOH in food, it can be concluded that sources listed by EFSA in 2012 all still seem relevant. However, due to the identification of sources of mineral oil contaminations and measures taken for mitigation, an overall decline of mineral oil contaminations since the 1990s has been observed. Examples of successfully reduced sources of mineral oil contaminations include the use of jute bags treated with batching oil, the application of (white) mineral oils as glazing agents, release agents in industrial bakeries, additions to animal feed, different contaminations of edible oils and the migration from paperboard packaging. Because of this decline, the relative contribution of environmental contamination may have increased. Due to accumulation and excretion by plants and animals the composition of MOH may change along the way up the food chain. Because of this, humans could be exposed to MOH that are well absorbed and accumulated. Whether this is a significant problem is not clear since animal products are estimated to have a low contribution to the total exposure via food whereas this contribution of vegetable products is higher. Exact data on what sources are causing mineral oil contamination in food today is missing and therefore it remains important to invest in a systemic investigation of these sources.

#### *Conditions of influence of migration*

As for the conditions of influence on the migration of mineral oil to foods, most studies have been done on the migration from paperboard packaging to dry foods. Migration here occurs for the more volatile MOH compounds, with a chain length up to C24. Migration is mainly influenced by temperature, with higher temperature leading to faster migration and a broader mass range migrating from packaging to food. Also, most migration occurs during the early stages of storage. Storage configuration has a large influence on migration. Other factors of importance are the mineral oil content in the ambient air, food structure (a higher fat content leading to a higher migration) and the physicochemical properties of a barrier (polyethylene was shown to be a weak barrier, in contrast to polypropylene), if present. Cooking was shown not have a large influence on the mineral oil content in the food, apart from rice. In rice, up to 50% of mineral oil can be removed during normal boiling times in water.

#### *Analytical techniques*

The standard method of choice for the analysis of MOSH and MOAH is on-line-coupled chromatography with gas chromatography and flame ionization detection (LC-GC-FID). Due to the large variety of structurally similar compounds of both MOSH and MOAH, it is impossible to identify individual compounds using LC-GC-FID. Therefore, in case that more information is needed on which types of MOSH and MOAH are present in a sample, additional analytical methods such as GCxGC-FID/MS, GC-MS

or nuclear magnetic resonance (NMR) spectroscopy are used. A technical guidance document on sampling and analysis of MOSH and MOAH in food and food contact materials was published by the Joint Research Centre (JRC) in 2019. This document is intended to be used by the stakeholders involved in the determination of mineral oil hydrocarbons in foods and food contact materials.

#### *Risk assessment*

The risk assessment of mineral oil hydrocarbons is subject to gaps of knowledge and so far, no tolerable daily intake levels (TDI's) for MOSH or MOAH have been set. Literature published since EFSA's opinion of 2012, shows that the concern on the potential risks for human health for MOSH has somewhat decreased. The toxicological effects of MOSH that have been demonstrated in rats and which are used to derive a toxicological Reference Point by EFSA, are questioned in their relevance for human health. Also, based on a dietary intake assessment for the Netherlands, the RIVM concluded that with the current exposure to MOSH, no adverse health effects are to be expected.

For MOAH, by lack of dose-response data, no toxicological Reference Point is available to calculate a margin of exposure (MOE). MOAH remains a concern due to its potential to cause mutagenic and carcinogenic effects. However, not all types of MOAH are mutagenic and carcinogenic. The mutagenicity of MOAH is considered to be caused mainly by polycyclic aromatic hydrocarbons (PAH) that contain three or more, non- or simple-alkylated, aromatic rings. These PAH are mainly present in combusted or heated mineral oils. Highly purified mineral oils have been shown to be non-mutagenic due the absence of mutagenic MOAH. The standard analytical method used to determine MOAH content does not provide information on which type of MOAH is found and therefore, for the risk assessment of MOAH, it is important to distinguish between the different sources that caused MOAH to be present in food.

#### *Discussion*

There are many different sources from which MOH can end up in foods. Although MOH are present in many foods, the levels have generally declined in the last decades. The chemical profile of MOH can vary greatly between foods, depending on the source. Also, the migration of mineral oils to food can be strongly influenced by a number of conditions.

Based on the available literature the following is concluded on the toxicity of MOH:

- MOSH and MOAH have no acute toxic effects but effects are possible after chronic exposure.
- For MOSH, the Reference Point for food grade/white oils is higher than for oils of lower grade.
- For the Dutch population, no adverse health effects are expected with the current exposure to MOSH via food.
- For MOAH, the 1- and 2-rings MOAH are of limited health concern.
- MOAH from combusted or heated oils or oils that are not sufficiently refined can be mutagenic and carcinogenic.
- Regarding mutagenic carcinogenicity, the RIVM considers food grade oils brought on the market of no concern.

The risk assessment of MOH is subject to gaps of knowledge which explains the lack of harmonized European or national limits for MOH in food. To obtain some guidance about which levels of MOH in food may be too high, the Belgian Scientific Committee of the 'Federal Agency for the Safety of the Food Chain' proposed 'action thresholds' for MOSH in food. In response to a request of the NVWA the RIVM evaluated the method for deriving these thresholds and came to the conclusion that these thresholds could not be endorsed.

For MOSH, the action thresholds are derived per broad food group, not taking into account the exposure from all sources together. Also, the action thresholds could be unnecessary strict for food products that only contribute little to the overall intake. But most importantly, the action thresholds for MOSH do not take into account what is reasonably feasible for the specific food products.

For MOAH, the Belgian committee discussed the use of the detection limit as an action threshold but considered the available data to be too limited to propose an action threshold. The RIVM agrees with that.

As an alternative, the RIVM recommends a different approach to reduce the concentrations of MOSH and MOAH found in food.

#### *Recommendations*

The RIVM has the following recommendations:

- As a first step, the relative contribution of the various foods to the total exposure to MOSH and MOAH could be determined.
- Foods that contribute significantly can be used to identify the source(s) of MOH and provide leads for mitigation measures to reduce the MOH content.
- The chemical profile of the MOH mixture may give information about the source of contamination as well as the toxicity. Identifying sources can be especially important for MOAH, since the presence or absence of the mutagenic/carcinogenic MOAH is partly determined by the source. Analyzing this could be considered when possible.
- As no adverse health effects are expected for the Dutch population with the current exposure to MOSH while MOAH can be mutagenic and carcinogenic, RIVM recommends to focus on MOAH.
- Mineral oils that are authorized as food additives are virtually free of MOAH and are therefore of lesser concern.
- Understanding the different conditions that influence migration of MOH is relevant to explain the concentrations found in the monitoring and may help identify avoidable contaminations.
- If action thresholds are being set for MOH in food, in line with the approach that is used for other contaminants, action thresholds could be derived from the 95<sup>th</sup> percentiles (P95) of measured concentrations in the foods. It is noted that for MOH that are authorized as food additive or plant protection product, specific limits apply.

## 1 Introduction

Mineral oils (MOH; mineral oil hydrocarbons) are complex mixtures of hydrocarbons, derived from crude oil. They consist of two fractions: 1) mineral oil saturated hydrocarbons (MOSH) and 2) mineral oil aromatic hydrocarbons (MOAH). MOSH consist of straight and branched open-chain alkanes (paraffins) and alkylated cycloalkanes (naphthenes). MOAH consist of (poly)aromatic hydrocarbons, that are generally alkylated.

Due to its numerous applications in food harvesting and food production processes, MOH can end up in food leading to consumer exposure. MOH have a large variation in both carbon number and structure, resulting in a very broad range of different chemical structures that can appear in food. For food, the hydrocarbons containing 10 to about 50 carbon atoms are relevant. MOH can end up in our food as a contamination or an intended addition. Sources of contamination include MOH containing food contact materials, lubrication oils from machinery and fuel oil. Examples of authorized additions of MOH to food include paraffinic waxes used for the surface treatment of confectionary and certain fruits, and paraffin oils and waxes used as active substance and additives in pesticide formulations. Other examples of MOH that are directly applied to food are refined 'white' mineral oils used as release agents for bakery ware and as anti-dusting agents for grain stored in silos. These uses have no authorization in the EU.

In 2012, the European Food Safety Authority (EFSA) published a scientific opinion on MOH in food (EFSA, 2012). Due to the lack of data on specific structural groups of MOH and consensus on the toxicological data that was available, no definite toxicological evaluation could be made. Also, there was little data available to accurately estimate the exposure to MOH in food. Hence, no 'tolerable daily intakes' (TDI) for MOH could be proposed and up till now, no harmonized European Regulation exists for MOH nor any national legislation. However, based on the available data, EFSA concluded that MOH exposure via food consumption in Europe was of potential concern (EFSA, 2012). The potential sources of MOH in food were listed and of these sources migration of MOH from recycled paperboard packaging into food was put forward as a possible significant contributor to the total exposure to MOH.

To address the problem of a lack of concentration data of MOH in food, the European Commission issued a Recommendation for monitoring of MOH in food, and in materials and articles intended for contact with food in 2017 (EC, 2017). In the Netherlands, this monitoring is done by the 'The Netherlands Food and Consumer Product Safety Authority' (Nederlandse Voedsel- en Warenautoriteit; NVWA). In order to explain the concentrations of MOH in food products and food packaging, and to assess the risks of MOH in food, the NVWA requested the National Institute for Public Health and the Environment (Rijksinstituut voor Volksgezondheid en Milieu; RIVM) to address the following questions:

- What is the current knowledge on migration of MOH to food?
- How could the risks be assessed of the concentrations MOSH and MOAH found in monitoring?

### *Approach*

In order to answer the first research question, literature on the migration of MOH to food, published since the 2012 EFSA report, was screened and summarized (see 'Literature search'). The focus was on 1) relevant food products, considering the concentrations of MOH and consumer exposure (based on concentration and food consumption), 2) sources of MOH in food and 3) conditions of influence on the migration of MOH to food.

While drafting this report, another RIVM-report was published on the same subject in 2018 (Van de Ven *et al.*, 2018). This RIVM 2018 report summarized the toxicity data published since the EFSA opinion in 2012 and included an intake assessment of MOSH and MOAH in the Dutch population. As part of the intake assessment, also the contribution of specific foods often packed in paperboard packages to the total dietary intake of MOSH and MOAH was estimated. This 2018 RIVM-report has been used to address both research questions. The dietary intake assessment was included to answer the first research question. This was complemented by a literature search on the migration of MOH to food, including literature on concentration data, potential sources of MOH in food and conditions of influence on the migration of MOH to food. Because of the complicated analytical determination of MOSH and MOAH in food, a brief overview of the analytical techniques is included in this report.

Concerning the second research question, the 2018 RIVM report included an overview of the toxicological studies performed, since the publication of the 2012 EFSA opinion up until mid-2017. Therefore, for the second research question, no new literature search on the toxicology of MOH was performed as part of the current report. Instead, the toxicological studies described in the 2018 RIVM report are summarized and complemented with a report of the Bundesinstitut für Risikobewertung (BfR, 2018) and a review article by Grob (2018b).



## 2 Literature search

A literature screening was performed to answer the first research question using the search engine of SCOPUS. Search criteria were established in line with the 2018 RIVM report on MOH in food, using the following words: 'mineral oil' OR 'MOSH' OR 'MOAH' in the title of the publication. Relevant studies investigating the migration of MOH, including sources and conditions of influence, or reporting on concentration data of MOH in food were selected based on the title and abstract. The search was restricted to publications published between June 2012 (publishing of EFSA opinion) and July 2018. However, a few articles published outside this time-frame that were considered relevant were also included.

In addition, a number of scientific reports were used as an extra source of information:

- RIVM report 2018: 'Mineral oils in food; a review of toxicological data and an assessment of the dietary exposure in the Netherlands' (Van de Ven *et al.*, 2018)
- Report 2018 of Wageningen University and Research: 'Levensmiddelen-verpakkingen gemaakt van oud-papier en karton: migratie van minerale oliën' (Thoden van Velzen *et al.*, 2018)
- Opinion 2017 of the French Agency for Food, Environmental and Occupational Health and Safety (ANSES): 'Opinion on the migration of mineral oil compounds into food from recycled paper and cardboard packaging' (ANSES, 2017)
- Advice 19-2017 of the 'Scientific Committee of the 'Federal Agency for the Safety of the Food Chain': 'Action thresholds for mineral oil hydrocarbons in food' (SciCom, 2017)
- BfR (Bundesamt für risicobewertung) Updated opinion No. 008/2018 of 27 February 2018. Highly refined mineral oils in cosmetics: Health risks are not to be expected according to current knowledge.



### 3 Occurrence in food and migration from materials

Migration from food contact materials can lead to the occurrence of mineral oil hydrocarbons (MOH) in food, but the occurrence of MOH in food is not always the result of migration from food contact materials. These subjects cannot easily be separately discussed and therefore this chapter deals with both.

#### 3.1 Concentration of mineral oils in food products

Data on the occurrence of MOH in food that has been published since the EFSA opinion in 2012 is restricted to concentration data of the Non-Governmental Organisation (NGO) Foodwatch (2015; 2016a; 2016b) and concentration data from a Belgian survey in response to the request of the European Commission in 2017 (Van Heyst *et al.*, 2018). These data are described in this report. At the time of writing the current report, no data of other EU Member States had been published in response to the EU request of 2017.

##### *Foodwatch 2015*

In 2015, Foodwatch published concentrations of MOSH and MOAH in food products packaged in cardboard and available on the market in three European countries: Germany, France and the Netherlands (Foodwatch, 2015). Concentrations of MOSH and MOAH were analysed in both the food and its packaging.

The products, some of which had a (plastic) interior lining, were selected based on their long shelf life and therefore having a greater probability of the migration of MOH from the packaging into the food. Also, the products were foods that are commonly consumed and had been shown to contain MOH. In total, 120 foods packaged in cardboard were sampled and tested for the presence of MOSH and MOAH. Results showed that 100 foods were contaminated with MOSH, while 51 contained MOAH. For foods that were sampled in all three countries mean concentrations of MOSH and MOAH are presented in Table 1.

*Table 1. Mean concentrations of MOSH and MOAH (mg/kg) in rice, pasta and cornflakes (food and packaging) sampled in the Netherlands, Germany and France. Measurements below LOD were counted as a concentration of zero.*

	The Netherlands		Germany		France	
	MOSH	MOAH	MOSH	MOAH	MOSH	MOAH
Rice food	1.3	0.2	1.4	0.2	1.6	0.3
package	24	0	55	9.8	162	22
Pasta food	28	1.1	1	n.d.	0.8	0.2
package	505	76	73	2.8	196	33
Cornflakes food	1.2	0.3	0.3	n.d.	1.7	0.5
package	257	66	297	88	315	100

n.d.: not detected in all samples

Below, a short description is given of these concentrations found in the foods analysed per country.

Different brands of rice, pasta, semolina and breakfast cereals (cornflakes) were sampled on the German market. In addition, one brand of lentils, couscous, breadcrumbs, oatmeal, baking mix, starch, cacao, chocolate flakes, powdered sugar and pudding powder were sampled. The concentrations of MOSH found in these foods ranged from <0.2 mg/kg (limit of detection; LOD) in various products to 3.8 mg/kg in one brand of rice. For MOAH, the corresponding figures were <0.2 mg/kg (LOD) in several products and 0.7 mg/kg in another brand of rice. The highest concentration of MOSH was found in the cardboard packaging of semolina (537 mg/kg) and the lowest concentration in the packaging of one brand of breakfast cereals (8 mg/kg). The concentrations of MOAH in cardboard packaging ranged from <5 mg/kg (LOD) for a number of products to 145 mg/kg in both a brand of breakfast cereals and chocolate flakes.

The products sampled in France included different brands of rice, pasta, breakfast cereals (cornflakes), couscous and lentils. Furthermore, one brand of cacao, cacao powder, biological cacao powder, chocolate cookies, corn starch, mashed potato flakes and a wheat grain product were sampled. Concentrations of MOSH found in the foods ranged from <0.2 mg/kg (LOD) in several products to 12.8 mg/kg in one brand of cacao. For the concentrations of MOAH, results ranged from <0.2 mg/kg (LOD) in several products to 2.7 mg/kg in one brand of lentils. In the cardboard packaging, MOSH concentrations ranged from <5 mg/kg (LOD) in one brand of rice to 625 mg/kg in chocolate cookies and MOAH concentrations ranged from <5 mg/kg (LOD) in various products to 159 mg/kg in one brand of breakfast cereals.

Sampled products from the Dutch market included different brands of rice, pasta, breakfast cereals (cornflakes) and chocolate sprinkles, as well as one brand of oatmeal, cacao powder and flavoured sprinkles. Furthermore, one brand of the following cereal products was sampled: breadcrumbs, corn starch, couscous, semolina and a whole wheat grain product. The concentrations of MOSH ranged from <0.2 mg/kg (LOD) in several products to a high concentration of 133 mg/kg in one brand of white pasta. The concentrations of MOAH ranged from <0.2 mg/kg (LOD) in a number of products to 5 mg/kg in another brand of white pasta. In the cardboard packaging, MOSH concentrations ranged from <5 mg/kg (LOD) in the package of one brand of chocolate sprinkles to 1008 mg/kg in one brand of pasta packaging. The corresponding concentrations for MOAH were <5 mg/kg (LOD) in several products to 213 mg/kg in the package of one brand of pasta (same product as the one with the highest MOSH concentration).

Table 2. Mean MOSH and MOAH concentrations per food group, found in food samples in all 3 countries by Foodwatch.

Food group	Concentration (mg/kg)	
	MOSH	MOAH
Breakfast cereals	1.5	0.6
Cacao powder	8.7	1.1
Cereal products	2.2	0.7
Chocolate	5.1	1.2
Chocolate sprinkles	2.6	0.8
Pasta	13.8	1.5
Rice	1.5	0.6

For some foods, there was a reasonable correlation between concentrations of MOSH and MOAH found in the food and in the packaging, but not for all. The same applies to the ratio between the reported MOSH and MOAH concentrations in a food. MOSH concentrations could be high in a food product where MOAH concentrations were low, but this was not true for all foods. Foodwatch describes the recycled cardboard packaging and storage boxes as suspected sources of MOH contaminations in food. However, Foodwatch also states that no clear conclusions can be drawn on this based on the analysed concentrations of MOSH and MOAH.

In addition to the food survey in 2015, Foodwatch published in 2016 concentrations of MOSH and MOAH in aluminium packed chocolate bunnies and chocolate Santa Clauses, available on the German market (Foodwatch, 2016a; 2016b). The MOSH and MOAH concentrations found ranged from 0.6 to 21.2 mg/kg and <0.5 mg/kg (LOD) to 2.9 mg/kg, respectively. No LOQs (limit of quantification) were reported.

#### *Belgian survey 2018*

In 2018, a Belgian study was conducted into the presence of MOH in food, both with and without cardboard packaging, sold on the Belgian market (Van Heyst *et al.*, 2018). In total, 217 food samples were selected based on a suspicion of contamination with MOH (based on previous market surveys) and their consumption frequency. In this way, the most important contributors to exposure could be identified within each food category. Samples were classified using FoodEx2, version 2 of the EFSA food classification and description system for exposure assessment (EFSA, 2011). Because the extraction methods were not suitable for all foods, only 198 food samples could be analysed. Concentrations of MOSH were detected in 142 samples, with the maximum concentrations for products ranging from 0.74 mg/kg in couscous up to 85 mg/kg in sweets. The other 56 samples had concentrations below the limit of quantification (LOQ; 0.5 mg/kg). Concentrations were particularly low in fish and meat samples, with only one meat sample containing MOSH (17 mg/kg). Similar to fish and meat, concentrations of MOSH were always below the LOQ for carrot, mushroom, onion and tomato. According to the authors, the low (or absent) MOSH concentrations could be due to measures taken by food packaging producers to reduce the migration of MOH to foods from packaging, such as a reduced use of recycled fibres for cardboard packaging, use of migration barriers between food and packaging, and

the use of migration-poor and/or migration-free ink when printing packaging.

Relatively high concentrations of MOSH were found in coffee and tea, ranging from 1.9 mg/kg to 7.4 mg/kg. The authors suggest that this was because of MOH containing jute bags in which these raw products are transported. Jute bags are often treated with batching oil containing a high-boiling mineral oil fraction. Among the food category 'sugar and similar, confectionery and water-based sweet desserts' products showed varying results. No sugar samples contained MOH, whereas chocolate flakes had an average MOSH concentration of 6.6 mg/kg. Again, the authors suggest that this was due to the contamination from jute bags in which cacao beans are transported. Furthermore, chocolate flakes are in direct contact with (recycled) cardboard packaging, without a functional barrier present. Relatively high amounts of MOSH were found in sweets (up to 85 mg/kg). Since MOH containing food additives are allowed for surface treatment of sweets, this could explain the high amounts of MOSH found according to the authors. However, no such additives were listed on the product labels.

In the study, a potential link between the MOH concentrations in food and the type of packaging was evaluated. Although no overall conclusions could be drawn, it appeared that all pudding powder samples, which were all in contact with paper and board packaging, contained MOSH. Also, all peas and lentils dry packed in cardboard contained MOSH, in contrast to those oil packed in cans that did not contain MOSH at detectable levels. The packaging material would have to be analysed in order to confirm the potential link with MOSH concentrations found in the foods.

In total, 23 samples contained MOAH concentrations above the LOQ (0.5 mg/kg), ranging from 0.6 to 2.24 mg/kg. These samples were nearly all vegetable oils and chocolate flakes. MOAH was also found in one out of the six coffee samples. The occurrence of MOAH in cocoa and coffee beans was, like for MOSH, considered to result from their migration from the batching oil in the jute bags. Other samples with MOAH concentrations above the LOQ were pudding powder, lasagna sheets, oatmeal, couscous, wholemeal and white rice, which were all packed in direct contact with cardboard. As for MOSH, analysis of the packaging is needed to identify the packaging as the source of contamination.

The MOSH and MOAH concentrations in food were compared to action thresholds proposed by the Scientific Committee of the Belgian Food Safety Agency. These action thresholds ranged from 5 to 150 mg/kg for MOSH depending on food type, and were set on the analytical detection limit (LOD) of 0.5 mg/kg for MOAH (SciCom, 2017). Only one sample had a MOSH concentration above the action threshold, whereas for MOAH, 23 samples were above the threshold (i.e. limit of quantification). Results are summarized in Table 3.

Table 3. MOSH and MOAH concentrations obtained from Van Heyst 2018 compared with action thresholds proposed by the Scientific Committee of the Belgian Food Safety Agency (SciCom).

Food category	Action threshold of SciCom MOSH / MOAH (in mg/kg)	[MOSH/MOAH] < action threshold (# samples)	[MOSH/MOAH] > action threshold (#samples)
Animal and vegetable fats and oils	100 / 0.5	9 / 5	0 / 8
Grain and grain-based products	15 / 0.5	99 / 95	0 / 7
Vegetables and vegetable products (incl. fungi)	20 / 0.5	13 / 11	0 / 1
Legumes, nuts and oilseeds	150 / 0.5	29 / 31	0 / 0
Snacks, desserts and other foods	20 / 0.5	10 / 3	0 / 2
Sugar and similar, confectionary and desserts	30 / 0.5	24 / 17	1 / 5
Fish and other seafood	60 / 0.5	7 / 7	0 / 0
Meat and meat products (incl. edible offal)	30 / 0.5	6 / 6	0 / 0
<b>Total</b>		<b>197 / 175</b>	<b>1 / 23</b>

The concentrations found in this study were lower compared with surveys conducted previously (Vollmer *et al.*, 2011; EFSA, 2012; Biedermann *et al.*, 2013). This could be an indication that adaptations made by packaging industries are effective in reducing the migration of MOH to food.

#### *Summary new studies on concentration data of MOH in food*

Few data have been published on the occurrence and concentrations of MOH in food since the 2012 EFSA opinion. The concentration data show varying MOSH and MOAH concentrations for a number of food groups. In the report of Foodwatch of 2015 dry foods available on the German, French and Dutch market packed in both recycled and fresh paperboard were measured. This dry food included among others breakfast cereals, cacao powder, cereal products, chocolate (sprinkles), pasta and rice. Out of 120 products tested, 100 were shown to contain MOSH, and 51 were shown to contain MOAH. Paperboard packaging was sometimes a plausible source, like for most pasta samples; where for other foods this relation was not that clear. The ratio between the MOSH and MOAH concentration in the food products was varying. Mean concentrations of MOSH were reported highest in pasta (13.8 mg/kg) and cacao powder (8.7 mg/kg). For MOAH the differences between the food groups were smaller, with pasta containing the highest amount (1.5 mg/kg).

In a recent Belgian survey, MOSH and MOAH concentrations were analysed in 198 food samples. Most MOSH and MOAH concentrations were below 10 and 0.5 mg/kg, respectively. High MOSH concentrations

were mostly reported for coffee, tea, chocolate flakes and sweets. Vegetable oils, chocolate flakes, cocoa and coffee beans contained the highest MOAH concentrations. Potential sources named for elevated MOH concentrations were jute bags treated with batching oil, contaminated (recycled) paperboard packaging and the use of mineral oil as food additives in the food production process. However, no real conclusions could be drawn on this since these potential sources of contamination were not analysed.

### 3.2 Dietary exposure assessment the Netherlands

Besides concentration data, consumption data are needed to answer questions concerning the actual exposure of humans to MOH in food. In 2018, a dietary exposure assessment of MOH in the Netherlands was reported (Van de Ven *et al.*, 2018). In this study, Dutch food consumption data of persons aged 2 to 69 years were combined with concentration data in food products. Concentrations used were those published by Foodwatch in 2015 and 2016 (Foodwatch, 2015; 20126a; 2016b), and by EFSA (2012) for those foods that were not analysed by Foodwatch. Data from the EFSA opinion did not include concentrations of MOAH in food. Therefore, it was assumed, as done by EFSA (2012), that the MOAH concentrations were 15% of the MOSH concentrations. Concentration data was classified according to level 1 of the FoodEx classification system (EFSA, 2011).

A median (P50) and a high (P95) long-term exposure to MOSH and MOAH was calculated for children aged 2 to 6 and persons aged 7 to 69 for the total diet and for paperboard packaged foods. Both the median and high levels of exposure to MOSH and MOAH in 2 to 6-year olds were approximately a factor 2 higher than in persons aged 7 to 69. The highest estimated level (the upper bound estimate of the P95) of exposure to MOSH and MOAH was 0.40 and 0.028 mg/kg bw per day in 2 to 6-year olds for the total diet, respectively.

When the intake of MOSH and MOAH for dry food packed in paperboard packaged foods was compared with the intake for the total diet, dietary exposure to MOSH and MOAH for paperboard packaged foods contributed only 2% to the exposure for the total diet at the median level in both age groups. At the high exposure level, this percentage increased to 15% for 2 to 6-year olds and 18% for 7 to 69-year olds; this was largely due to the consumers of pasta.

In order to explore which products were mainly responsible for the exposure to MOH, the contribution of different food groups to the total exposure to MOSH was investigated (Fig. 1).

For children aged 2 to 6, food groups that contributed at least 10% to the total exposure to MOSH were 'confectionary' (non-chocolate)' (17%), 'pasta' (14%), 'ice and desserts' (12%) and 'vegetable products' (10%). The contribution of the food group 'confectionary (non-chocolate)' could mainly be attributed to sour sweets and wine gums, contributing 8% and 5% respectively. For the food group 'pasta', this was white pasta (13%); for 'ice desserts', this was ice water and vanilla ice cream (6% and 3%); and for 'vegetable products', the main contributors were apple sauce (4%) and fried spinach (cooked) (1%).



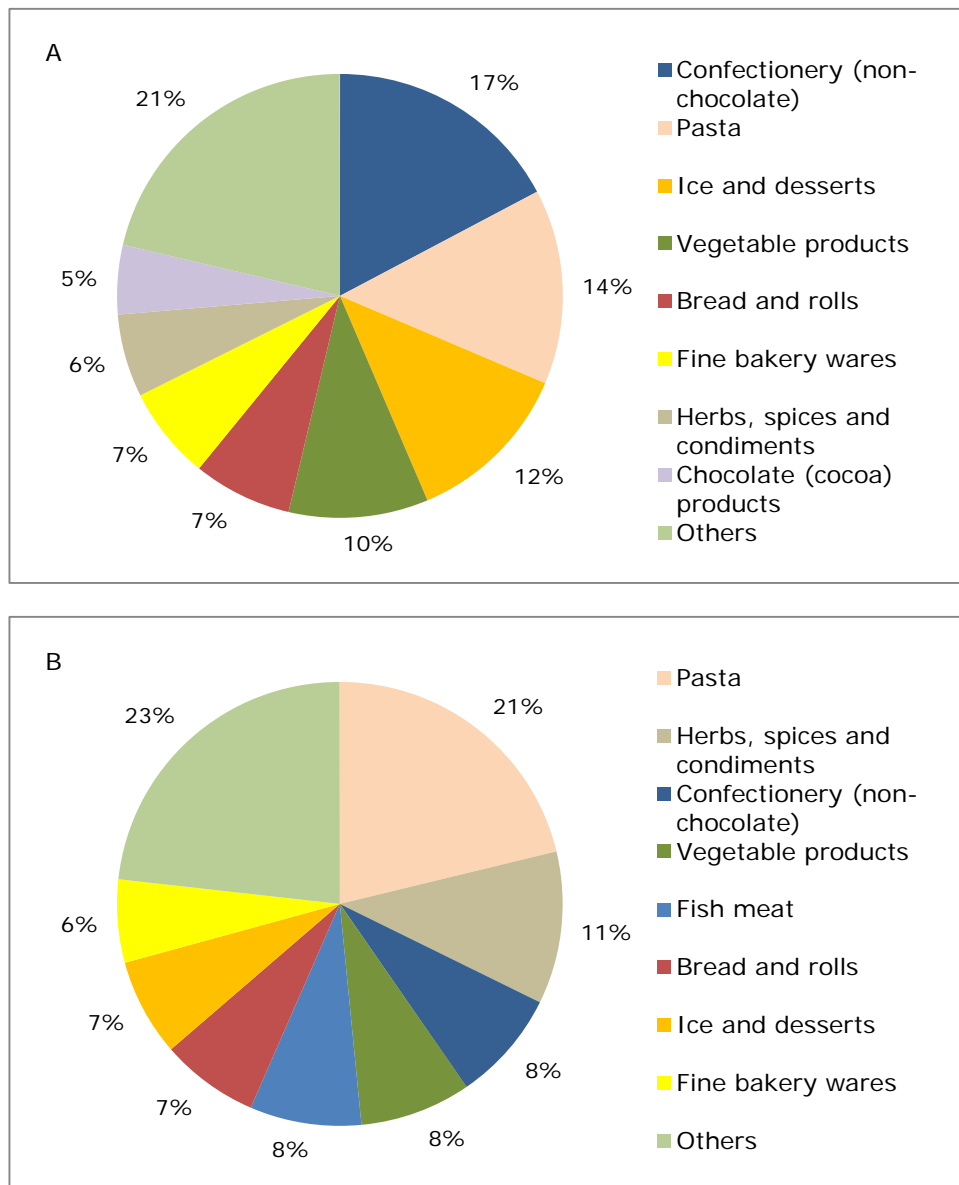


Figure 1. Contribution (%) of food groups, with a contribution of at least 5%, to the total exposure distribution to MOSH in children aged 2 to 6 (A) and persons aged 7 to 69 (B) via the total diet (Figure as taken by Van de Ven et al., 2018).

Food groups contributing more than 10% to the total exposure to MOSH for persons aged 7 to 69 were 'pasta' (21%) and 'herbs, spices and condiments' (11%). Similar to the contribution to exposure for 2 to 6-year olds, it was white pasta that mainly caused the high contribution of 21% of the food group 'pasta'. This is considerably higher than the total contribution from paperboard packed dry foods (2%), but can be explained by the fact that not all consumed pasta is packaged in paperboard. Pasta can also contain significant amounts of MOH, derived from other sources, such as 'white mineral oils' used as release or spraying agents. The contribution of the food group 'Herbs, spices and condiments' was due to many different food products, each with only a small contribution, and mostly from instant soups.

### *Summary dietary intake assessment MOH*

A dietary intake assessment of the RIVM showed the relative contributions of different food groups to the total exposure to mineral oils via food intake. Exposure to the age group 2-6 was approximately a factor two higher than in the older age group 7-69, with the high exposure scenario (P95) resulting in MOSH and MOAH exposures of 0.40 and 0.028 mg/kg bw per day in 2 to 6-year olds. For 2 to 6-year olds the food groups contributing most to the total mineral oil exposure were 'confectionary' (non-chocolate), 'pasta', 'ice and desserts' and 'vegetable products'. For persons aged 7 to 69 years old the food groups contributing the most to mineral oil exposure were 'pasta,' and 'herbs spices and condiments'. The contribution of foods packed in recycled paperboard to the total exposure was 15% (2 to 6-year olds) and 18% (7 to 69-year olds) calculated for the high exposure level (P95).

### **3.3 Sources of mineral oils in food**

One of the major obstacles to reduce MOH contamination of food is the identification of the source of the contamination. Identifying the sources and their impacts on food contamination is of special importance for the mapping of the routes of contamination along the production chain. Also, when sources of MOH in food are known, food surveys could be more targeted. In its 2012 report, EFSA listed many different sources of the presence of MOH in food. A short overview of this list is given below, distinguishing between sources in 'food processing' and sources in 'food contact materials', with the addition of a few other potential sources of MOH in food.

#### *Food processing*

During food processing, MOH can enter food via different ways, depending on their application during the process. The different sources that contribute to the transfer of MOH to food during food processing are:

- Release agents: MOH used mainly in the production of baking ware, sugar products and pasta by being sprayed onto all sorts of surfaces (funnels through which dough has to glide, cutting devices to portion dough, etc.).
- Anti-dusting agents: MOH used as a dust control agent during the transport and processing of grains, such as rice.
- Machine oils: MOH used in machines for food harvesting or processing. This includes diesel oil, lubrication oils and oils used for cleaning and maintenance of machinery.
- Coating of foods: white mineral oils used as glazing agents for rice and mineral waxes used for the coating of vegetables, fruits and confectionary.

#### *Food contact materials*

MOH are present in food contact materials and can be released from these materials, primarily food packaging, entering the food. Concerning food contact materials, EFSA stated the following sources of MOH migrating to food:

- Jute and sisal bags, treated with batching oil and used for transport of primarily rice, hazelnuts, chocolate and coffee.

- Waxed packaging materials: mineral waxes used for paper or paperboard to render it more water-resistant. Products packed in waxed paper packaging include cheese, bakery wares and candies.
- Plastic materials: white mineral oils and waxes are allowed to be added to plastic food contact material in the EU, without a specific migration limit, i.e. up to the overall migration limit of 10 mg/dm<sup>2</sup> food contact surface.
- Lubricating oils for cans: MOH may be used in the can making process
- Printing inks: the use of conventional offset printing inks containing 20-30% MOH on food packaging material. This source has been addressed in numerous studies.
- Recycled board: recycled paper and board can be contaminated with MOH since they can be produced from printed paper, possibly containing adhesives and solvents.
- Adhesives: glues and adhesives can contain MOH components to render them sticky. Migration from bags, boxes and labels containing adhesives is therefore possible.

Other sources of MOH in food stated by EFSA (2012) include pesticides (as additive in the formulation as well as the active substance), the addition of MOH to feed, and environmental contamination.

It is important to note that some of the sources of MOH in food concern the intentional addition of MOH to food (such as glazing agents and coatings). In those cases, there is no contamination.

In this report, the literature published since the 2012 EFSA opinion was screened to establish whether any new sources of MOH contamination have been identified or whether known sources are no longer of relevance due to their elimination. The few articles available showed that the identification of the sources of contamination is still a key problem when addressing MOH contamination in food. Overall, no new sources have been identified, but some of the sources listed by EFSA have been eliminated for specific products, as will be discussed below. In a major part of the studies conducted since the 2012 EFSA opinion the migration of MOH from recycled paper and board is studied. In recycled paper and board, MOH are contained in offset printing inks, adhesives and solvents that are present in paper fed into the recycling process. A description of the literature on sources of MOH in food published since the 2012 EFSA opinion is given below.

*Lommatzsch et al., 2016*

The contribution of hot-melt adhesives (glues) used to close paperboard packaging to the amount of MOH in food was studied by Lommatzsch *et al.* (2016). The hot-melt raw materials investigated consisted mainly of paraffinic waxes, hydrocarbon resins and polyolefins. The hydrocarbon resins were the predominant source of the hydrocarbons of sufficient volatility to migrate into dry foods. Migration of substances of a hot-melt was estimated to be 1 mg/kg food, depending on different factors such as the amount of substances volatile enough to migrate to the food (eluted from the GC between n-C<sub>16</sub> up to n-C<sub>24</sub>), the hot-melt surface, the amount of food and contact time.

*Brühl, 2016*

In a study conducted by Brühl (2016), the different sources that could contribute to the MOH contamination of oilseeds and vegetable oils were discussed. Overall, the same sources were named as those listed by EFSA (2012), including insecticide formulations containing MOH (microcrystalline wax free from MOAH and refined paraffin fractions) and the use of refined MOH (free from MOAH) as anti-dusting agents. Also, lubricants for the machinery, thermal heating fluids and mineral oils used in can production and packaging material were mentioned. In addition, environmental contamination was considered a substantial source. Environmental contamination of food with MOH can be attributed to incomplete combusted compounds in exhaust gases from power plants, motor engines and heating burners, and to debris from tires and road tar as dust.

*ANSES, 2017*

Another recent report addressing the potential sources of MOH in food was published by the French Agency for Food, Environmental and Occupational Health and Safety, ANSES (ANSES, 2017). In this report, the characterization and risks of MOH migrating into food from recycled paper and cardboard packaging were addressed. Regarding sources of MOH in food, ANSES was in line with the 2012 EFSA opinion, focusing on the importance of recycled paper and cardboard as a significant source. Hot-melt glues and adhesives used to stick paper were also mentioned as possible migration sources, as well as secondary cardboard boxes used as food containers.

*WUR, 2018*

In a recent report of 2018 from Wageningen University & Research (WUR) on the migration of MOH from recycled paper and board to food, the sources listed in the 2012 EFSA opinion were discussed (Thoden van Velzen *et al.*, 2018). Emphasis was put on the difference between foods being contaminated with MOH and those to which MOH are intentionally added via food additives containing MOH. Sources of contamination should be identified as they probably impose higher risks, as these MOH are likely of low grade, containing concentrations of carcinogenic MOAH.

*Grob, 2018a*

An overview of the main sources that have been identified since the discovery of food contaminations with MOH was published by Grob (2018a). Grob (2018a) summarizes the findings concerning sources of MOH contamination in food up to 2010 and describes how part of the contamination due to these sources has (almost) been successfully reduced, resulting in the overall reduction of MOH food contamination since the 1990s. These sources include the jute bags treated with batching oil, the application of white mineral oils as glazing agents, its use as release agents in industrial bakeries, additions of MOH to animal feed, contamination of edible oils (e.g. frying oils) with MOH and the migration of MOH from paperboard packaging. Although contaminations have been reduced, it is clear that all sources listed by EFSA still have to be taken into account when considering the potential sources of MOH contaminations in food.

Grob (2018a) identified the absence of a systemic investigation of sources as a major gap for addressing the problem of food contaminated with MOH. As mentioned by Brühl (2016), Grob (2018a) describes that the environmental contamination with MOH can eventually end up in the food chain due to the uptake of MOH by plants. For this, Grob (2018a) refers to a study describing air samples containing MOH with a mass distribution corresponding to that of MOH from particulate matter from the exhaust of diesel engines and engine (lubricating) oil (Brandenberger *et al.*, 2015). In another study mentioned by Grob, MOH concentrations in air ranged from 0.03 µg/m<sup>3</sup> in rural areas to 5 µg/m<sup>3</sup> in a road tunnel (Neukom *et al.*, 2002). The air contaminations could eventually settle on the soil and subsequently be taken up by plants. In Neukom *et al.* (2002), also different environmental samples were shown to contain MOH concentrations ranging from 0.5 mg/kg in humus in a town garden to 38 mg/kg in compost, indicating accumulation of MOH in rotting plant material. This shows that plants grow in a MOH-contaminated environment which could eventually result in human exposure to mineral oils via consumption of these plants. Besides direct consumption of MOH contaminated plants, animals fed MOH contaminated plant material could lead to human exposure to MOH due to the consumption of animal products. Animals that consume MOH contaminated plant material will only excrete a part of the MOH and some the excreted MOH will end up in milk or eggs. The MOH that is not excreted will be taken up and can accumulate in the meat that we consume. Since this concerns MOH that were prone to accumulate in animals, they might accumulate in humans as well. In other words, along the food chain the MOH content could be modified in the direction of MOH that are well absorbed and accumulated by humans. However, considering the small relative contribution of animal products to the total intake of MOH, as described in Figure 1, the significance of this contamination seems very small. For vegetables this could be different. Although concentrations of MOH have been shown to be low by Van Heyst *et al.* (2018), in the dietary intake assessment by RIVM (Van de Ven *et al.*, 2018) the contribution of vegetable products was estimated to be 8 and 10% for 2-6 year olds and 7-69 year olds.

#### *Summary of new studies on sources of MOH in food*

Overall, the potential sources of contamination of food with MOH listed in the 2012 EFSA opinion are still relevant. It seems that the literature published since the 2012 EFSA opinion still regards recycled paperboard packaging as one of the main sources of food contamination with MOH. This is not entirely supported by the previously mentioned report of RIVM (Van de Ven *et al.*, 2018) in which a dietary intake assessment showed the relatively low contribution of recycled paperboard packaging to MOH intake, at least via dry foods (see paragraph 3.2) which excludes pizza boxes, fast food packaging etc.

Also, a number of the sources listed by EFSA have been identified and eliminated, mainly the jute and sisal bags treated with batching oil and MOH used as release agent in the production of mainly baking ware and sugar products. Due to the complexity of the international food production chain, the identification of a food contamination source remains a major problem. The (partly) elimination of certain sources of MOH in food has resulted in an overall decline of MOH contamination since the 1990s. With the overall level of contamination decreasing, the

(non-decreasing) environmental contamination could become relatively more important. Environmental contamination is however difficult to avoid.

Exact data on the sources that result in MOH contamination in food today are missing. Therefore, it is difficult to conclude which sources should be given priority when taking measures against food contamination and surveying the routes of contamination along the chain of food production and storage remains an important but challenging task. Moreover, considering the overall decrease in MOH contaminations in food the necessity for mitigation measures to reduce the source should be considered.

### 3.4 Identifying the source

Despite the difficulties of identifying the source of contamination of foods with MOH at the end of the production process, there may be ways to obtain some indications as to the origin of a contamination. For example, the ratio between MOSH and MOAH is different in crude oils, which contain MOAH concentrations up to 35%, as compared to oils from the more refined 'technical grade', which have relative low MOAH content, and the 'food grade' or 'white mineral oils', which are virtually free of MOAH. Therefore, in food containing only MOSH and no MOAH, the source will most likely be a white mineral oil used as food additives or food processing agents, where foods containing MOH in a relative high MOAH to MOSH ratio, are likely contaminated with less refined oils.

As white mineral oils are added deliberately to our food, the source of it is likely more easily identified than MOH of a lower grade resulting from contamination, having a wider range of possible sources. Another typical feature of MOH that can help to identify their source is that those used as a release agent during the 1990's were shown to be commonly centred at n-C<sub>21</sub> to n-C<sub>23</sub> (Grob, 2018a) and that the typical profile of paraffin waxes (used to improve the water resistance of paperboard) shows a chromatographic hump around n-C<sub>27</sub> to n-C<sub>28</sub> (Barp *et al.*, 2015b). These examples suggest that the chemical profile of the MOH present in food could provide information about the source of the contamination, whether it is the identification of a source or its exclusion.

In addition, the simultaneous analysis of other contaminants can also provide information about the origin of MOH contamination. An example is the presence of DIPN (diisopropyl naphthalene), which is typical for contamination from recycled paper that contains DIPN because of its use in carbonless copy paper (Sturaro *et al.*, 1994).

When characterizing MOH contaminations, usually only a distinction is made between MOSH and MOAH. The importance of the characterization of MOAH in a more detailed manner is of importance in terms of toxicological relevance. Biedermann and Grob (2010) described the differences in specific MOAHs between crude batching oil and an extract of more refined oil from ink in a newspaper. A GCxGC-FID plot showed that, despite a roughly equal total MOAH content, the amount of MOAH containing 3 and 4 rings (which have more carcinogenic potential than

1- or 2-ringed MOAH) was much higher in the crude batching oil than in the oil extracted from a newspaper. In this way, the characterization of the MOAH provides both knowledge on the toxicological relevance of the contamination and its origin. The characterization of Biedermann and Grob (2010) also showed that even though the amount of MOAH in food may be substantial, it does not have to be of carcinogenic potential.

*Mineral oil toolbox – BLL, 2018*

A useful tool has been developed by the 'Bund für Lebensmittelrecht und Lebensmittelkunde' (BLL) in Germany: "Toolbox for Preventing the Transfer of Undesired Mineral Oil Hydrocarbons into Food". It is free accessible on the internet and provides a comprehensive overview of the routes of entry of mineral oils into food. The toolbox includes sources and tools for mitigation for MOSH, MOAH or the analogue substances that may end up in food. It distinguishes between mineral oil entering food through migration from (secondary) food packaging, contamination from lubricants (of different technical grade) used in the food production chain and mineral oils used as additives/processing aids. It has the aim to enable companies to review their food handling processes and take appropriate product-related mitigation measures to reduce avoidable sources of mineral oil in food (BLL, 2018).

### **3.5 Conditions of influence on the migration of MOH into foods**

The migration of MOH from recycled paper into food may take place via two mechanisms: direct (wetting) contact and transfer through the gas phase, where MOH move from the packaging material to the food surface via a process of evaporation and re-condensation. For dry solid foods packaged in paper materials, the migration of volatile contaminants through the gas phase is usually more relevant than that via direct contact. At ambient temperature, only MOSH and MOAH up to C<sub>24</sub> are volatile enough to migrate through the gas phase in substantial amounts.

MOH components can migrate through various layers of packaging, such as in the case of MOH migrating from an outer corrugated cardboard transport box into the packed food inside this box. The speed and extent of migration depend on several factors, such as physicochemical properties of the migrating substance, the packaging material and the food. However, the most important factors that influence migration are conditions, such as time, temperature and the ratio between the surface area of the food contact material and the foodstuff. Last, the presence of a barrier and its physicochemical properties are important.

Most of the studies into the conditions that influence the migration of MOH into food concern the migration from recycled paper and board. Recycled paper and board contain MOH from different sources such as newspapers and other printed papers entering the recycling process. Also, not necessarily due to the recycling process, adhesives and solvents used as carriers for binders and additives, and paraffin waxes added to the packaging material to improve water resistance can contain MOH. The literature assessing the conditions influencing MOH migration into food published since the 2012 EFSA opinion, is summarized below.

Lorenzini *et al.*, 2013

In 2013, Lorenzini *et al.* studied the migration behaviour of MOSH, MOAH and diisopropyl naphthalenes (DIPN) from paper materials to dry foods (Lorenzini *et al.*, 2013). DIPN is present in recycled paper due to its presence in carbonless copypaper. Migration from two commercial products packed in recycled paperboard, i.e. muesli and egg pasta, were monitored up to the end of their shelf life (1 year). Muesli was contained in an internal bag of unprinted polyethylene (PE) whereas the egg pasta was kept in an external bag of printed polypropylene (PP). The influence of time, storage conditions, food packaging structure and temperature were studied.

### Temperature

In the two food models, migration from recycled paperboard (packs wrapped individually in aluminium foil) during 1 year at 20 °C reached MOSH concentrations of 20 mg/kg in muesli and 14 mg/kg in egg pasta, representing 45% and 53% of the migration potential (the potential maximum amount of MOH that could migrate from packaging to food (i.e.  $< n-C_{24}$ ), respectively (Fig. 2). Migration of mineral oils to food was shown to be fast, with 5 mg of MOSH migrating in 1 week at 20 °C. This amount doubled when kept for two weeks at the same temperature. Storage at 30 °C caused a transfer of 15.1 mg/kg to the pasta in about one month.

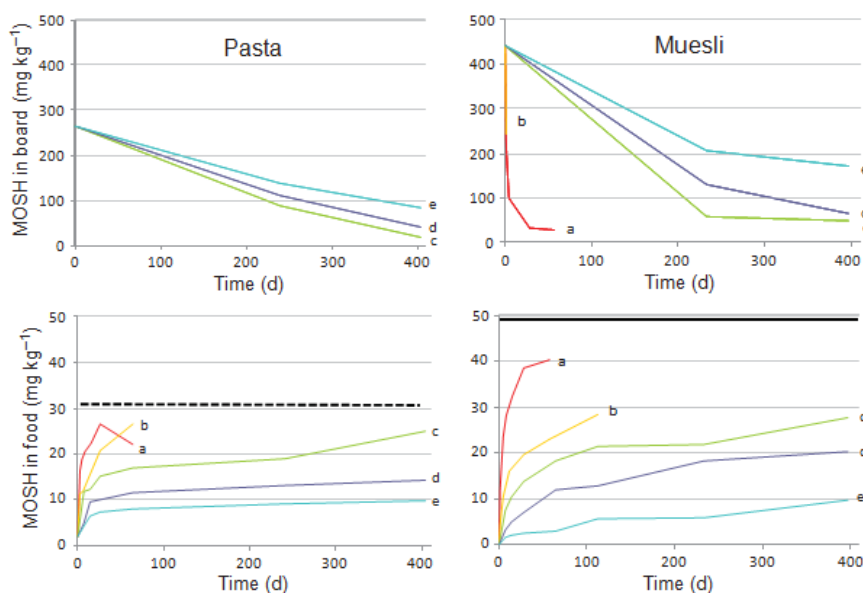


Figure 2. Migration of MOSH  $< n-C_{24}$  into pasta and muesli at different temperatures: 60°C (a), 40°C (b), 30°C (c), 20°C (d) and 4°C (e). Lines (a) and (b) are missing for pasta paperboard due to incomplete data. The black dashed in food graphs lines represent the potential migration at RT (Figure as taken from Lorenzini 2013).



An increase in temperature not only caused higher migration levels, it also broadened the molecular mass range that migrated from the packaging to the food (Fig. 3; migration up to C19 at 4 °C, C25 at 30 °C and C28 at 60 °C). Migration at refrigerator temperature (4 °C) was lower but still substantial (after one year 50% of that at 20 °C) and migration was slower and more restricted to highly volatile compounds.

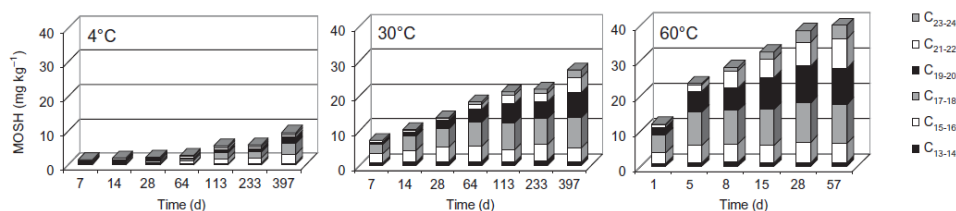


Figure 3. MOSH migration to muesli as a function of carbon numbers at different storage temperatures. At higher temperature and prolonged storage, higher molecular mass hydrocarbons migrate (Figure as taken from Lorenzini 2013).

### Storage configuration

The way the food pack is positioned appeared to be very important, since boxes standing alone on a shelf lose volatile hydrocarbons into ambient air, whereas this is barely the case of products packed into larger cardboard boxes or products piled up on pallets. For packs stored in shelved and free packs at room temperature, the concentration of MOSH in muesli reached 22.5 and 18.9 mg/kg (51% and 40% of the potential), respectively (Fig. 4). Packs stored in corrugated board boxes showed a MOSH concentration of 35 mg/kg (77% of the potential). A similar influence of storage condition on the migration was seen for egg pasta. Here, concentrations of MOSH in shelved and free packs after one year were 18.3 (68%) and 15.6 mg/kg (58%). For the egg pasta in boxed packs, this concentration was 24.3 mg/kg (91%).

Maximum MOAH concentrations in muesli were reached after 8 months storage for all storage configurations: 5.2, 4.6 and 3.6 mg/kg for boxed packs, shelved packs and free packs, respectively (Fig. 4). DIPN concentrations were highest after 12 months of storage: 1.2, 0.9 and 0.6 mg/kg for boxed, shelved and free packs respectively (Fig. 4).

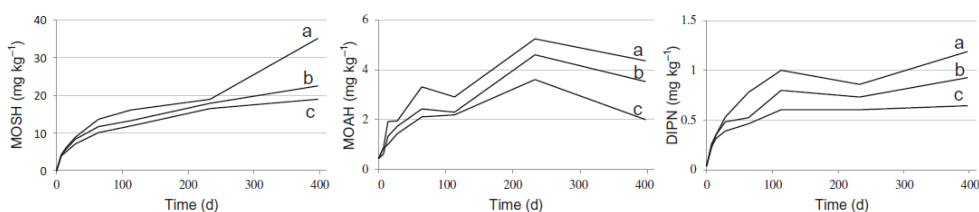


Figure 4. Migration of MOSH (left), MOAH (centre) and DIPN (right) into muesli for different configurations of storage at RT. The maximum migration to food is observed for the packs inside cardboard boxes (a), followed by piled packs (b) and free standing packs (c) (Figure as taken from Lorenzini 2013).

For egg pasta, the highest MOAH concentrations were 4.1, 4.2 and 4.5 mg/kg for boxed packs, shelved packs and free packs, respectively, reached after 8 months of storage too (data not shown). Here, DIPN concentrations were also highest after 8 months of storage and reached

15.8 (46%), 15.6 (41%) and 14.8 (32%) mg/kg for boxed, shelved and free packs.

### Plastic barrier

The polyethylene internal bag containing the muesli and the polypropylene external bag containing the egg pasta were examined for their influence on migration. The polyethylene bag acted as a sink, firstly adsorbing hydrocarbons and then partly releasing them to the food. The polypropylene bag was a much more efficient barrier, strongly limiting the migration towards the atmosphere and thus giving rise to the highest level of food contamination.

*Biedermann et al., 2013*

In a similar study in 2013, Biedermann *et al.* reported results of a migration study that was conducted with six dry foods (including chocobiscuits, polenta, noodles, rice, breadcrumbs and oatmeal) packed into unprinted recycled paperboard with and without internal packing (Biedermann *et al.*, 2013).

### Migration in different foods over time

Stored for 9 months at ambient temperature, food directly packed in paperboard absorbed on average 69% of MOSH and 50% of MOAH <n-C<sub>24</sub> from the paperboard, resulting in concentrations ranging between 30 and 52 mg/kg for MOSH (Fig. 5) and between 5.5 and 9.4 mg/kg for MOAH. Differences in migration between the foods were relatively small: mostly less than a factor of 2 between the extremes. Migration was shown to be more influenced by the porosity of the food than the fat content. Also, most of the migration occurred in the early stages of storage, with MOSH migration ranging between 22% and 57% of the migration potential after 2 months of storage. Migration of MOAH was slower and remained at a lower percentage of the migration potential after 9 months of storage when compared to MOSH.

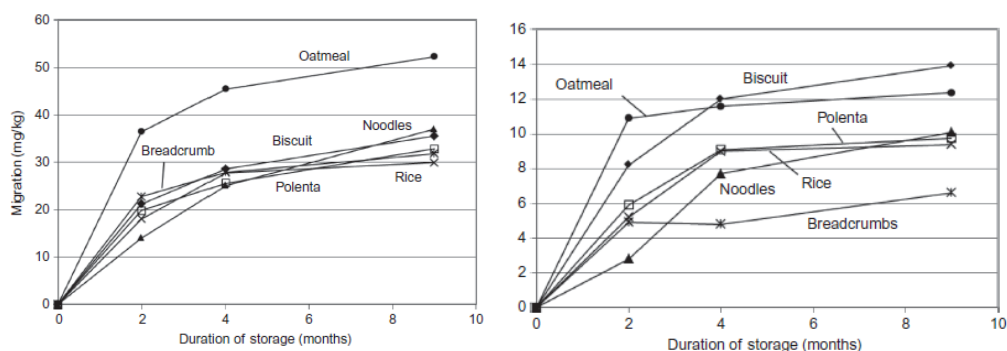


Figure 5. Left: migration of MOSH from recycled paperboard into foods packed without an internal tray and plastic film. MOSH, mineral oil saturated hydrocarbons. Right: migration of MOSH from the recycled paperboard into the foods packed into the tray and the PE film over 9 months. MOSH, mineral oil saturated hydrocarbons; PE, polyethylene (Figures as taken from Biedermann *et al.* 2013).

### **Plastic barrier**

Different internal plastic barriers were tested. Confirming the results published by Lorenzini in 2013, a polyethylene film had little effect on the migration rate, but did act as a sink. A barrier of polypropylene, on the contrary, proved to strongly slow migration: the highest migration of saturated hydrocarbons after 9 months (2.3 mg/kg) corresponded to only 3% of the content in the paperboard and included migrated polyolefin oligomeric saturated hydrocarbons (POSH). Polypropylene with an acrylate slowed migration even further. However, migration from the paperboard was still detectable in four of the six samples. Last, polyethylene terephthalate (PET) was shown to be a tight barrier over the full period of 9 months storage.

#### *Barp et al., 2015a*

Barp *et al.* (2015a) monitored the migration of MOSH, MOAH and DIPN from recycled paperboard boxes in direct contact with semolina and egg pasta during shelf life (up to two years) at room temperature. For paperboard boxes sealed with a hot-melt adhesive, the contribution due to the migration of polyalphaolefins (PAO) from this adhesive was also evaluated. Similar to Lorenzini's study in 2013, three different storage conditions were tested: packs wrapped in aluminium (to prevent any influence from the surrounding environment), pack standing on shelves (simulating storage in a supermarket) and packs stored in corrugated cardboard boxes (simulating storage in a warehouse) to investigate the migration from secondary packaging (only for semolina pasta).

### **Semolina: storage configuration**

For semolina packed in recycled paperboard boxes closed with hot-melts, the total MOSH and MOAH concentrations reached were highest in the packs wrapped in aluminium: 6.8 mg/kg and 1 mg/kg respectively (Fig. 6). This was followed by the samples stored in the transport box consisting of corrugated board with MOSH and MOAH concentrations of 4.1 mg/kg and 0.7 mg/kg, respectively. Migration was lowest for the samples stored on shelves, where migration reached values of 3.2 mg/kg for MOSH and 0.6 mg/kg for MOAH. The increased concentrations of MOH observed for samples stored in transport boxes seemed to be mainly due to the reduced losses towards the ambient air in combination with a migration phenomenon from the transport box.

### **Semolina: contribution adhesives**

For the concentrations of MOSH in semolina, the contribution of the adhesives used to glue the recycled cardboard was shown to be 1.3 mg/kg food corresponding to an increase of 30 % compared to migration from recycled paperboard without adhesives. For MOAH, the hot-melt contribution was not evident.

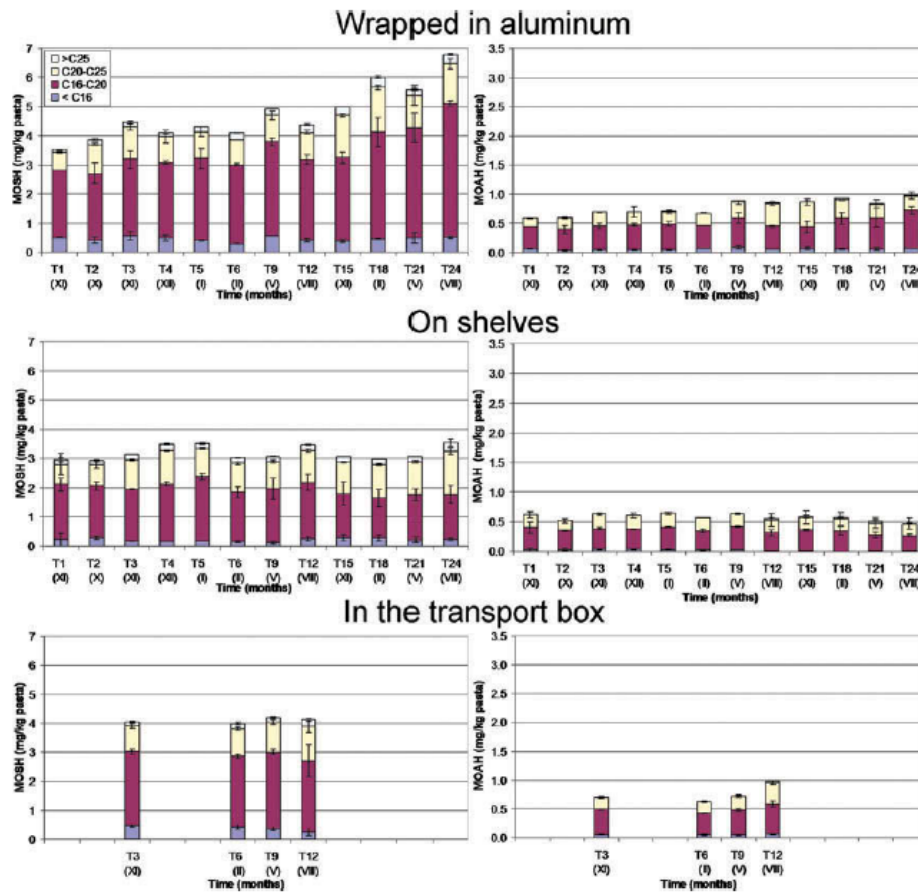


Figure 6. MOSH (left) and MOAH (right) in semolina pasta packed in recycled paperboard boxes closed with hot melts stored at different conditions. The values are divided into four different ranges of volatility. The sampling time specifying the month of the year (roman numeral) is reported on the x-axis. Results are expressed as the average of four replicates (bars) and standard deviations (vertical lines) (Figure as taken from Barp et al. 2015a).

**Egg pasta: storage configuration**

For the egg pasta packed in boxes wrapped in aluminium a constant contamination during storage was observed, with MOSH and MOAH concentrations around 8.0 and 1.5 mg/kg, respectively (Fig. 7). In contrast to the semolina samples, samples stored on shelves ended up having higher concentrations than the samples wrapped in aluminium (14.5 and 2 mg/kg for MOSH and MOAH, respectively). Samples stored on shelves were also shown to have and increase in contamination over the period of storage. This seemed to be an important contribution coming from the external environment.

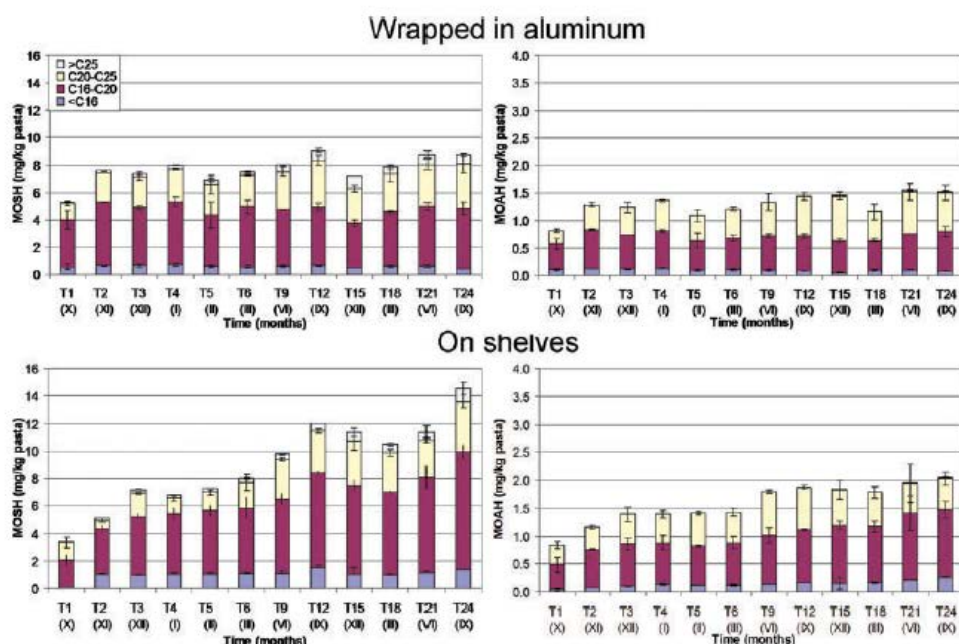


Figure 7. MOSH (left) and MOAH (right) in egg pasta packed in recycled paperboard (without hot melt) stored at different conditions. The values are divided into four different ranges of volatility. The sampling time specifying the month of the year (roman numeral) is reported on the x-axis. Results are expressed as the average of four replicates (bars) and standard deviations (vertical lines) (Figure as taken from Barp et al. 2015a).

### Migration semolina compared to egg pasta

By keeping the packed food in aluminium foil the concentration of MOSH, MOAH and DIPN that was lost from the paperboard could be compared to the concentration that was taken up by the pasta. MOH contents reached during and at the end of the shelf-life period were remarkably higher for egg pasta (MOSH: 8 mg/kg; MOAH: 1.5 mg/kg) than for semolina (MOSH: 4.2 mg/kg; MOAH: 0.6 mg/kg). Possible reasons for this mentioned were the higher fat content in the egg pasta promoting migration of the hydrophobic compounds, both in velocity and amount. Also the fact that the egg pasta was stored amongst several recycled paperboard boxes could have played a role, since the hydrocarbons could have migrated from these surrounding boxes into the food.

### MOSH lost by paperboard

For MOSH, an increasing trend was observed for the amount lost by the paperboard over the whole shelf life. However, at the end of shelf life, only 30% of the amount lost by the paperboard was calculated to have migrated to the pasta, where this was 50% for MOAH. The increasing trend for MOSH lost by the paperboard depended mostly on the heavier fraction ( $C_{20-25}$ ), where the more volatile components ( $<C_{20}$ ) remained constant after three months of storage. For MOAH, no clear correlations between the amount lost by the paperboard and the amount in the pasta could be found. For DIPN, a good mass balance was shown for the amount lost by the paperboard and the amount that migrated to the food over the whole shelf life (24 months).

In conclusion, the recycled paperboard boxes were shown to contaminate the semolina pasta with a (lower) contribution of hot melts used to close the boxes. MOSH and MOAH levels did, however, never exceed 3.6 and 0.6 mg/kg, respectively. Secondary packaging gave a contribution to the total contamination of 25%. The external environment was not shown to contribute to the contamination of semolina pasta. This environmental contribution was suggested for egg pasta, since these samples showed an increasing trend for the MOSH content up to the end of shelf life (14.5 mg/kg) whereas samples wrapped in aluminium reached a constant MOSH content around 8 mg/kg.

*Barp et al., 2015b*

In 2015 Barp *et al.* conducted a second similar study, investigating the influence of conditions on migration of mineral oils to pasta in direct contact with virgin paperboard or polypropylene flexible film (Barp *et al.*, 2015b). Migration from virgin fibre was low and not significantly affected by the food matrix, reaching constant levels of 0.5 to 0.6 mg/kg. Thanks to these low levels of contamination from the virgin paperboard packaging, it was possible to study the influence of other potential sources of contamination.

**Semolina: storage configuration**

Regarding other sources than the paperboard packaging, migration from the secondary packaging or from the surrounding environment was only slightly visible in semolina pasta, with contamination levels of increasing trend during storage: 0.7, 0.8 and 1 mg/kg MOSH <C<sub>25</sub> after 3, 9 and 12 months of storage. This increasing trend could not be seen in pasta wrapped in aluminium nor in the boxes stored on shelves.

**Storage configuration and surrounding environment**

Migration from hot melts used to close the box and from the surrounding environment was well evident in egg pasta. For samples with hot melts, contamination was characterized by a narrow hump centered around n-C<sub>18</sub> in the MOSH fraction, with no MOAH and DIPN. The contribution of the hot melts to the total hydrocarbon contamination increased during storage: 1.5 mg/kg after 3 months of storage and 5 mg/kg at the end of the shelf life. The surrounding environment's contribution (evident in the range below n-C<sub>16</sub>) increased as well during storage: 3 mg/kg after 3 months of storage and 10 mg/kg at the end of shelf life, corresponding to one third and half of the total contamination, respectively (Fig. 8).

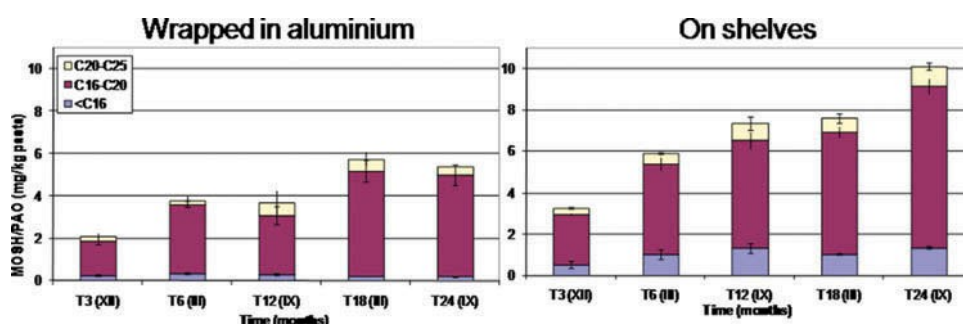


Figure 8. MOSH/PAO ( $\text{mg kg}^{-1}$  pasta) in egg pasta packed in virgin paperboard boxes with hot melts wrapped in aluminium and kept on shelves. The values are divided into three different ranges of volatility. The sampling time specifying the month of the year (roman numeral) is reported on the x-axis. The results are expressed as the average of four replicates (bars) and standard deviations (vertical lines) (Figure as taken from Barp et al. 2015b).

### Plastic barrier

Pasta samples packed in polypropylene (PP) film had amounts of polyolefin oligomeric saturated hydrocarbons (POSH) from the film below  $2 \text{ mg/kg}$  (Fig. 9). The polypropylene film showed good barrier properties toward migration from the surrounding environment over the whole shelf life of pasta, with no MOH or DIPN being detected over 24 months of storage for samples stored on shelves. However, its barrier effect to prevent migration is limited to 9 months if the pasta pack remains closed in a high hydrocarbon tension ambient, such as can be a secondary box of corrugated cardboard. This was probably due to the fact that hydrocarbons accumulate in the plastic matrix to the point of being able to cross it, travelling towards the inside of the pack.

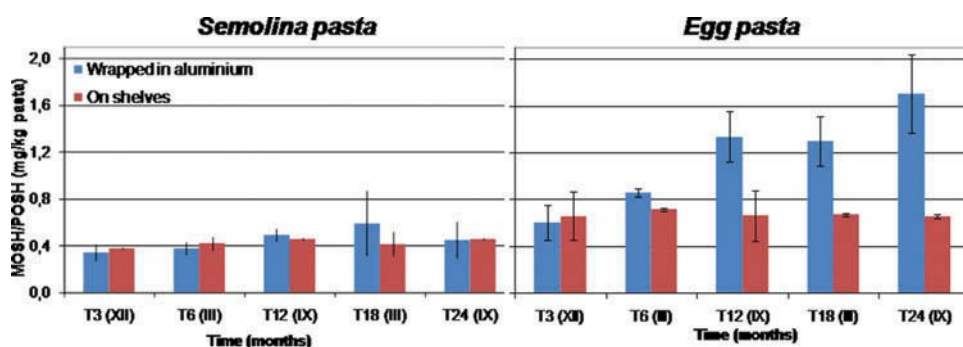


Figure 9. Contamination ( $\text{mg kg}^{-1}$  pasta) found in semolina and egg pasta packed in PP film stored wrapped in aluminium and kept on shelves. The sampling time specifying the month of the year (roman numeral) is reported on the x-axis. The results are expressed as the average of four replicates (bars) and standard deviations (vertical lines) (Figure as taken from Barp et al. 2015b). Biedermann-Brem and Grob, 2011

The studies described above concern the migration of MOH from the food packaging material to the packed (dry) food but do not necessarily tell us anything about the concentrations of MOH in the food once it is cooked. As mentioned before, the transfer of MOH to food from packaging material almost exclusively occurs through the gas phase. Migration at ambient temperature has been found to be substantial up to the alkane  $n\text{-C}_{24}$  and this suggests a high removal during cooking

should be possible because of evaporation into the vapor bubbles leaving the pan. To investigate this, Biedermann-Brem and Grob studied the removal of MOH that migrated from paperboard packaging during boiling in water for different foods, including noodles, rice, couscous and chocolate powder (Biedermann-Brem and Grob, 2011).

For rice, around 50% of MOSH and MOAH was shown to be removed by regular cooking (i.e. as one would do at home). The removal was dependent on the type of rice (raw or parboiled), duration of cooking and whether or not an excess of water was used. However, for the other foods it was shown that cooking as indicated on the label (i.e. as consumers would actually cook the food) did not result in a significant removal of MOH. For couscous and chocolate powder, only excessively long boiling times lead to a higher removal but these boiling times were not seen as realistic. For noodles, even these excessively long boiling times did not remove MOH from the food up to a significant amount. The low removal rates are explained by the authors by the water moving into the pores of the food and then acting as a barrier, preventing the MOH from transferring to the vapour bubbles. This study showed that overall cooking in boiling water does not efficiently remove mineral oils of a volatility that migrated from paperboard.

*Summary of new studies on conditions of influence on migration mineral oil to food*

The reviewed literature in this report has shown that conditions during the food production process and subsequent storage of the food products can have a major influence on the MOH concentrations in dry foods that are paperboard packed. The different conditions of influence, such as temperature during storage, storage configuration, storage duration, food structure and the physicochemical parameters of barrier (if present) could help to explain MOH concentrations in food. They could also be used to target those products that are kept under conditions leading to elevated migration of MOH to food.

The studies assessing the influence of different conditions on the migration of MOH from packaging to food show similar results which could be summarized in more detail as follows:

- The reported migration levels of MOSH differ between studies, showing different qualities of recycled paperboard; most reported migration levels are below 10 mg/kg but one study showed MOSH migration levels of 30-52 mg/kg (packed food wrapped in aluminium foil).
- At room temperature (20 °C) migration of MOSH and MOAH occurs for the volatile compounds of mass range up to n-C<sub>24</sub>.
- Temperature is the main factor influencing migration in terms of speed and extent: a higher temperature leads to an acceleration of migration and broadens the mass range of migrating substances (above room temperature hydrocarbons with a mass >n-C<sub>24</sub> migrate from packaging to food); at low temperature (i.e. 4 °C) migration occurs slower and is more restricted to highly volatile substances but migration is still substantial.
- The major part of migrating compounds transfer in the early stages of storage.



- The storage configuration is of high influence on migration: migration is highest when food packs are stored in secondary packaging (i.e. cardboard boxes), followed by when packs are piled up together. Migration is lowest when packs are free standing. This is explained by the opportunity of releasing volatile hydrocarbon into the ambient air for free standing packs, where this is not the case for the other two storage configurations. However, MOH present in the ambient air could increase MOH concentrations in the packed food.
- Regarding food structure, the fat content seems to be of influence on migration with higher fat content leading to a higher migration, both in velocity and amount. Porosity is also of importance for migration: a higher porosity leads to more absorption of hydrocarbons.
- Hot-melt adhesives can have a significant contribution to migration of MOSH. This was not evident for MOAH.
- Regarding barriers, polyethylene was shown to be a weak barrier with minor influence on migration, acting as a sink for hydrocarbons and then releasing these compounds to the food. Polypropylene was shown to be an effective barrier, although it loses efficiency at storage for longer periods in a cardboard box due to a high vapour tension. Polyethylene terephthalate proved to be a tight barrier. Using virgin paperboard shows surrounding secondary boxes and the ambient air to be significant sources from which migration can occur. The use of virgin paperboard is often mentioned as a measure against mineral oil contamination in (paperboard packed) food. However, the fact that virgin paperboard is a weak barrier means that surrounding secondary boxes and the ambient air could still cause significant contaminations in the packed food.
- Apart from rice, boiling dry foods in water as one would prepare food at home, does not efficiently remove mineral oils that migrated into the food from paperboard packaging.



## 4 Analysis techniques

Although it is not the focus of this report, some attention is given to the analytical methods for the determination of MOSH and MOAH, including their advantages and limitations.

Literature shows that since the mid 90's, on-line-coupled liquid chromatography with gas chromatography and flame ionization detection (LC-GC-FID) has been the method of choice and is considered the gold standard for routine MOSH-MOAH analysis. The combination of chromatography, which separates MOSH from MOAH, and GC-FID for quantification allows for an appropriate determination of the MOSH and MOAH content. Further fractions with different molecular masses can be defined, based on the retention time of the corresponding n-alkanes under the same chromatographic conditions. However, the large variety of structurally similar single compounds makes it impossible to identify the individual compounds.

The reason why LC-GC-FID is considered ideal for the analysis of MOH mixtures is because it combines the high sample capacity and wide range of separation of the LC-system and the high separation efficiency along with a variety of selective detection methods of the GC-system. In comparison with off-line techniques the on-line-coupled LC-GC system offers advantages such as high reproducibility, high sample throughput, less susceptibility to carry over effects, a high level of automation and robustness as well as a higher sensitivity of the multidimensional chromatography since no sample material is wasted. However, the LC-GC-FID method has disadvantages too. It only delivers irregular humps of unresolved compounds which require experience and expertise to be correctly interpreted and false positive results are difficult to recognize. False positive results can be caused by compounds such as natural n-alkanes, polyolefin oligomeric hydrocarbons (POHs), polyalphaolefins (POA), and lipids, which includes the risk of overestimating the amount of MOSH and MOAH.

Therefore, in cases of positive samples, a confirmatory method could be used (Spack *et al.*, 2017). Furthermore, to obtain more information about which types of MOSH and MOAH are present, additional analytical methods are needed to be used, such as GCxGC-FID/MS (Biedermann, 2015), GC-MS (Populin *et al.* 2004) or nuclear magnetic resonance (NMR) spectroscopy, (Lachenmeier *et al.* 2017).

Methods for analysis of MOSH and MOAH have recently been comprehensively reviewed by Weber *et al.* (2018). It contains an overview of analytical methods used for MOH in food, food contact materials, cosmetics and animal tissues. As an alternative to chromatography, nuclear magnetic resonance (NMR) spectroscopy was suggested, especially with the possibility of detecting only the toxicologically relevant aromatic rings. From the reviewed methods, NMR appears to be best suitable to provide a rapid screening of mineral oil products, thereby having potential to be a substitute for the IP346

method. The IP346 method is the current industry standard to assess carcinogenicity of mineral oil raw materials.

In 2019, JRC published a technical guidance document on sampling and analysis of MOSH and MOAH in food and food contact materials, in the frame of Recommendation (EU) 2017/84 for the monitoring of mineral oils (Bratinova and Hoekstra, 2019). The guidance is intended to be used by all stakeholders involved in the determination of mineral oil hydrocarbons in food and food contact materials, i.e. food inspectors, official control laboratories, laboratories in industry and laboratories of non-governmental organisations.

## 5 Risk assessment of concentrations MOSH and MOAH found in monitoring

The second research question of this report concerns the risk assessment of MOH concentrations found in the monitoring. Before a risk assessment can be performed, the data on toxicity has to be considered. For this, the 2012 EFSA opinion was used as a starting point, as well as the 2018 RIVM report. In the 2018 RIVM report, an overview is given of the toxicological studies performed since the 2012 EFSA opinion up until mid-2017, which was considered sufficiently recent, and therefore no new literature search on the toxicology of MOH was performed for the current report. Only a published review (Grob, 2018b) and a risk assessment by the Federal Institute for Risk Assessment in Germany (Bundesinstitut für Risikobewertung) (BfR, 2018) have been additionally considered.

A summary of the 2012 EFSA opinion (EFSA, 2012), the 2018 RIVM report (Van de Ven *et al.*, 2018) and additional literature is presented below, for MOSH and MOAH separately.

### 5.1 Toxicity and risk assessment of MOSH

#### *EFSA, 2012*

The absorption from oral intake of the straight chain (n-) alkanes is high for C14-C18 ( $\pm$  90%) and decreases with increasing carbon number, being only 25% for C26-C29, where, because of their low absorption, MOSH above n-C35 are considered less relevant for monitoring. Absorption of cyclo-alkanes (naphthenes) seems similar to that of n-alkanes, where absorption of branched alkanes is slightly less. Alkanes are generally biotransformed to fatty acids, which is more rapid for n-alkanes than for branched- and cyclo-alkanes. MOSH have no genotoxic properties. MOSH have low acute oral toxicity and was considered not relevant given the level of exposure from food.

The effects of exposure to MOSH are only seen after longer exposure duration. MOSH are shown to bioaccumulate in various tissues both in humans and rats, including fat, lymph nodes, spleen and liver. In rats, 90-days of exposure to MOSH led to the formation of microgranulomas in lymph nodes and the liver. The microgranulomas in lymph nodes were, by lack of progress to adverse lesions, considered as non-adverse, but the microgranulomas in the liver of rats are associated with inflammatory reactions. In humans exposed to MOSH, granulomas have been observed in liver and other organs, but these changes were not associated with inflammatory reactions or other adverse consequences. There was however no information on exposure levels at which these effects occur in humans. Due to insufficient information, the hepatic microgranuloma formation associated with inflammation as seen in rats was considered possibly relevant to humans by EFSA and was identified as the most critical effect. The NOAEL was 19 mg/kg bw/d in a 90-day oral rat study. Although n-alkanes bioaccumulate to a much lesser extent than branched- and cyclic-alkanes, the waxes (containing > 90%

n-alkanes) were the most potent mixtures tested. The lowest LOAEL was 161 mg/kg bw/d in Fischer 344 rats.

In the EFSA opinion, some concern was expressed on the potency of MOSH to alter immune function by oral exposure, as intradermal and intraperitoneal injections of high doses of certain MOSH induced autoimmune responses in arthritis-prone rodent models. Given the deficiencies in the toxicity database and in the absence of toxicological studies on MOSH mixtures typical of those humans are exposed to, it was considered inappropriate to establish a health based guidance value (like an ADI) for MOSH. Instead, the use of an MOE approach was proposed. The NOAEL of 19 mg/kg bw was used as Reference Point for background exposure to MOSH.

For some specific uses, like MOSH used as release agents for bread and rolls and for spraying of grains leading to 'high exposures', white oils are known to be used. The range of MOSH grades for these white oils is more restricted than that for the background exposure. Therefore the CONTAM Panel used the highest NOAEL below the lowest LOAEL for these grades of MOSH, 45 mg/kg bw/d as Reference Point. Based on the intake assessment with the occurrence data available, EFSA identified a potential concern for both the 'background exposure' and the 'high exposure' (EFSA, 2012).

#### *RIVM 2018 report*

New studies on the toxicity of mineral oils, published after the EFSA opinion have been described and discussed in the report of RIVM (Van de Ven *et al.*, 2018). The main conclusions are summarized below.

Two new sub-chronic oral immunotoxicity studies in rats showed that MOSH mixtures had no impact on the immune response (Cravedi *et al.* 2017, Andreassen *et al.* 2017), indicating that the immunotoxic effect seems to be route specific and also that immunotoxicity is not a relevant effect for long term dietary intake of MOSH.

A new study in Fischer 344 rat of Barp *et al.* (2017b) showed that, even though branched-and cycloalkanes (C<sub>25</sub>-C<sub>30</sub>) are more prone to accumulate in the liver, it was the n-alkane fraction (chain length >C<sub>25</sub>) that seemed to induce the hepatic microgranulomas. n-Alkanes are hardly found in human liver tissues, nor in other human tissues (Biedermann *et al.* 2015). This suggests that these n-alkanes are not absorbed very well and/or are efficiently metabolised and eliminated in humans, at the dietary intake levels.

Evaluation of an analysis on the mode of action/human relevance framework (MoA/HRF) for MOH-induced epithelioid granulomas in the Fischer rat, using modified Bradford Hill considerations, led to the conclusion that the mode of action was not relevant to humans (Adenuga, 2017).

Results of the new studies should however be considered in coherence with the studies that have already been evaluated by EFSA. Therefore, no proposal for revision of the Reference Point of 19 mg/kg bw (NOAEL) was made (RIVM 2018). For the risk assessment of MOSH intake by the

Dutch population, the dietary intake calculations resulted in MOEs of 190 in children aged 2 to 6, and 450 in persons aged 7 to 69; the corresponding MOEs for the high (P95) exposure were 90 and 160, respectively. It was concluded that, considering the questionable relevance of liver microgranulomas for humans, the estimated exposure levels to MOSH were of no health concern for the Dutch population (Van de Ven *et al.*, 2018)

For white mineral oils, which are used as release agents for bread and rolls and for spraying of grains, a higher NOAEL of 45 mg/kg bw/d was defined as Reference Point in the MOE calculation. These uses were identified as of potential concern by EFSA (2012) and this conclusion was supported for 'bread and rolls' by a preliminary assessment for the Dutch population (Van de Ven *et al.*, 2018). It was suggested that mineral release agents are no longer used for bakery ware (Barp *et al.* 2017b, Grob 2018a).

#### *BfR, 2018*

In the year that the RIVM report was finalised, the Federal Institute for Risk Assessment in Germany (Bundesinstitut für Risikobewertung; BfR) published an opinion on the use of mineral oils in cosmetics (BfR, 2018). It was concluded that "Inflammatory granulomas were detected exclusively in the liver of Fischer 344 rats during experiments after high doses of low-viscosity mineral oils had been administered. These lesions differ morphologically from the non-inflammatory lipogranulomas observed in the liver of autopsy patients in correlation with increased MOSH levels. Practically no lipogranuloma was found in the liver in another study conducted on autopsy patients with low MOSH levels. The toxicological relevance of the lesions observed in the Fischer 344 rats for humans is doubtful at the moment." (BfR, 2018).

In conclusion, based on the available literature, the concern for potential risks of exposure to MOSH seems to have diminished. The relevance of toxicological effects seen in rats for humans is questioned and the current exposure of consumers to MOSH via food intake does not seem to be of concern.

## **5.2 Toxicity and risk assessment of MOAH**

Limited data on MOAH toxicokinetics suggest that MOAH compounds are well absorbed, extensively biotransformed, and that they do not bioaccumulate in mammals. MOAH have low acute oral toxicity and therefore acute toxicity was considered not relevant given the level of exposure from food (EFSA, 2012).

The main potential concern for MOAH is genotoxicity. Mutagenicity of MOAH is considered to be caused mainly by polycyclic aromatic hydrocarbons (PAH) that contain three or more, non- or simple-alkylated, aromatic rings. Non-alkylated PAH are mainly formed by heating of the oil, and are a minor fraction of MOAH (EFSA, 2012).

The common approach for the risk characterisation of genotoxic carcinogens is to calculate a margin of exposure (MOE), but neither for MOAH mixtures, nor for mineral oils, dose-response data on the

carcinogenicity is available. Therefore it is not possible to establish a Reference Point that could be used for the calculation of the MOE (EFSA, 2012). Since the EFSA opinion, no dietary animal toxicity studies or any *in vivo* dose-response toxicity data on MOAH have been published, so this conclusion has not changed.

EFSA concluded that all crude mineral oils are mutagenic, unless they are treated to specifically remove MOAH. Removing MOAH is an important step in the refining process of mineral oils, which is done by solvent extraction. In addition, aromatic rings of MOAH are converted into cyclic alkanes by saturating the aromatic rings with hydrogens e.g. by hydrogen treatment under high pressure. The remaining MOAH in refined mineral oils are mainly 1-2 ring highly alkylated structures (Concawe, 2016).

Indeed, highly purified oils have been shown to be non-mutagenic, both *in vitro* and *in vivo* (EFSA, 2012). This would lead to the conclusion that the use of highly purified (i.e. food grade) oils in all processes of the food production chain would overcome the concern on carcinogenicity from MOAH from this source. Codes of practices for the processing of food like HACCP (Hazard Analysis and Critical Control Points) prescribe that lubricants used in machinery for food processing should contain only mineral oils that are not classified as carcinogenic. The same applies to printing inks used on food contact materials. MOAH from printing inks almost exclusively have 1–2 aromatic rings (Grob, 2018b).

Carcinogenic potential of mineral oil fractions can be tested by use of the IP346 assay, a gravimetric method used by oil refineries, which is based on good solubility of the aromatic fraction in DMSO. Mineral oils that contain less than 3% w/w of substances extractable with DMSO do not have to be classified and labelled as 'carcinogenic' according to the CLP Regulation (EC) No. 1272/2008 (EC, 2008). The IP346 assay has been validated by mouse dermal carcinogenicity tests, performed in two mouse-strains that were dermally exposed several times per week for up to two years (Concawe, 2016). The genotoxic species of MOAH are removed by refining mineral oils to the point that the IP346 test is <3%. This way, technical oils on the European market have a low potential of genotoxicity. In the production of "food-grade" oils, MOAH are further reduced, usually by hydrogenation, but not fully eliminated.

It was noted that non-mutagenic refined mineral oils may become mutagenic during use (EFSA 2012). High temperature processes, e.g. >800 °C can convert non-mutagenic mineral oil components into mutagenic PAH, and motor oils in addition to thermal decomposition may also pick up engine combustion products such that after use they may contain several orders of magnitude higher concentrations of PAH (EFSA 2012). It is not known to what extent this will contribute to the formation of mutagenic components and their contamination in food. Contribution of pyrogenic sources is however included in the PAH monitoring programmes in food. In 2008, PAH were of low concern to the average consumer, whereas for high level consumers, there was a possible need for risk management actions (EFSA, 2008). Apart from heated mineral oils, PAH in food can result from other sources as well, like smoke flavourings and burning of food.



In conclusion, for the risk assessment of MOAH found in food, it is important to distinguish between different sources of MOAH. MOAH derived from crude oils or oils not sufficiently purified, as well as MOAH from combusted (or heated) mineral oils are carcinogenic and therefore, these contaminations should be avoided as much as possible. MOAH from highly refined oils or waxes are of lesser concern.

### **5.3 Conclusion toxicity and risk assessment MOSH and MOAH**

Reviewing the literature that has been published since the 2012 EFSA opinion showed that the concern for human health effects from the exposure to MOSH has decreased. The toxicological effects in rats have so far not been shown to be relevant for humans, and with the current exposure to MOSH in food no health effects are expected in the Netherlands. For MOAH, the main concern is that they can be mutagenic and carcinogenic, due to the PAHs that contain three or more aromatic rings. These PAHs can be present in MOH due to insufficient purification of the oil, or they can be formed by heating the oil. By use of the standard analysis method for the determination of MOAH content, no information can be obtained on which type of MOAH has been found. Methods to discriminate between different MOAH in mixtures are currently not available. That is why it is important to distinguish between the different sources of contamination for MOAH, as some contain harmful MOAH, where others do not.



## 6 Discussion

In this report, an overview is provided on the sources and occurrence of mineral oils in food. As many different sources are identified, it is not surprising that MOH are present in many foods, although the levels in food have generally declined in the last decades. The sources of mineral oils are numerous and the chemical profile, although hard to discover, can vary greatly between sources. Also, the migration of mineral oil to food can be strongly influenced by a number of conditions (e.g. storage, presence of a barrier). Although a systemic investigation of all sources of mineral oils in food is still missing, this report could be used to contribute to an overview of the relevant sources of mineral oil contaminations nowadays.

Concerning the toxicity of MOH, the available literature was reviewed which resulted in the following important points:

- MOSH and MOAH have no acute toxic effects but effects are possible after chronic exposure
- For MOSH, the Reference Point for food grade/white oils is higher than for oils of lower grade
  - For the Dutch population, no adverse health effects are expected with the current exposure to MOSH via food
- For MOAH, the 1- and 2-rings MOAH are of limited health concern
- MOAH from combusted or heated oils or oils that are not sufficiently refined can be mutagenic and carcinogenic
- Regarding carcinogenicity, the RIVM considers food grade oils brought on the market of no concern

The risk assessment of MOH is subject to gaps of knowledge. The toxicological relevance of effects shown for MOSH in rats for humans is questioned. For MOAH, with a possible mutagenic and carcinogenic potential, there are no dose response data available. In addition, the characterization of MOH remains a problem, especially for MOAH: some MOAH mixtures may be mutagenic and carcinogenic while others are relatively harmless. A review of the literature published since the 2012 EFSA opinion showed that these gaps in knowledge still form an obstacle for assessing the risks of MOSH and MOAH for human health (see paragraph 5.1 and 5.2). The lack of harmonized European or national limits for MOH (MOSH and MOAH) in food products confirms this.

To obtain some guidance on which levels of MOH in food may be too high, the Belgian Scientific Committee of the 'Federal Agency for the Safety of the Food Chain (FAVV)' has proposed 'action thresholds' for MOH in food (SciCom, 2017). These threshold values are "concentrations above which the origin of the contamination should be determined and measures may be needed to reduce or eliminate the contamination" (FAVV, 2017).

For MOSH, the committee derived threshold values for 14 different broad food product groups, by dividing the 'acceptable or tolerable' daily intake through the Belgian consumption values for the total intake by that food group, at the 97.5th percentile (P97.5). This was done for two populations: adults (18 - 64 years) and children (3 - 9 years). An

'acceptable' intake of 0.19 mg/kg bw/day was derived by dividing the NOAEL of 19 mg/kg bw/day by 100, assuming 100 to be a safe margin of exposure. The action thresholds for MOSH (C16-C35) for the 14 food groups ranged from 5 mg/kg (milk and dairy products) to 150 mg/kg (legumes, nuts and oilseeds, and eggs and egg products).

For MOAH, as no safe intake could be established, the Belgian committee suggested that the detection limit of 0.5 mg/kg could be used as an action threshold. However, the data was considered too limited to propose an action threshold. Therefore, it was recommended to limit the presence of MOAH in food as much as possible.

In response to a request of the NVWA, the RIVM evaluated the Belgian approach (report not published). For MOSH, the conclusion of this evaluation was that the RIVM does not endorse the method for deriving action thresholds, for several reasons. First, the Belgian action thresholds are derived per broad food group and do not take into account the exposure from all sources together. This way, the concentration of MOSH could be below the relevant threshold values for all foods, while the overall exposure to MOSH is above the acceptable intake. Another disadvantage is that for food products that only contribute little to the overall intake of the food group to which they belong, the limit could be unnecessary strict and no adverse health effects are to be expected from levels higher than the action threshold limit set for these products. Moreover and most importantly, the action thresholds do not take into account what is feasible for the specific food products, which can result both into threshold values unattainably low, as well as, for other food products, unnecessarily high.

For MOAH, the Belgian committee discussed the use of the detection limit of 0.5 mg/kg as an action threshold, but considered the available data to be too limited to propose an action threshold. RIVM agrees with that. Contaminations with MOAH can be above 0.5 mg/kg food, but if they originate from a source that only contains non-mutagenic MOAH, this would provide little cause for concern. The LC-GC-FID method used for routine analysis of MOAH content in food does not characterize the specific types of MOAH, and therefore the risk of the presence of MOAH concentrations cannot be estimated if no information on their origin is available.

As an alternative approach to assess the risks of mineral oil in food, a number of recommendations are provided in this report (see below).

## 7 Recommendations

The RIVM has identified possible approaches to reduce mineral oil contaminations and a number of guiding considerations:

- As a first step, the relative contribution of foods to the total exposure to MOH via food intake could be determined. The dietary intake assessment of RIVM (Van de Ven *et al.*, 2018) can be used for this, complemented with occurrence data collected in response to the EC-recommendation for monitoring in food present on the Dutch market (EC, 2017), as well as the latest food consumption data of the Dutch National Food Consumption Survey 2012-2016 published in November 2018 (VCP 2012-2016).
- Foods that contribute significantly can be investigated to identify the source(s) of MOH and provide leads for mitigation measures to reduce the MOH content.
- Since the chemical profile of a MOH mixture (e.g. the ratio between MOSH and MOAH, concentration the main chain length of the MOSH fraction, or the amount of rings in the MOAH-fraction) may give information about the source of contamination as well as about the toxicity, analyzing this could be considered when possible. Inversely, knowledge on the source of a contamination can give information about the chemical profile and potential health risks of the contamination.
- Since no adverse health effects are expected for the Dutch population with the current exposure to MOSH while MOAH can be mutagenic/carcinogenic, RIVM recommends to focus on MOAH.
- Authorized uses of MOH (e.g. food additives, glazing agents, pesticides) entail 'white' mineral oils virtually free of MOAH. Therefore, these sources require less attention and concern.
- Understanding the different conditions that influence migration of MOH is relevant to explain the concentrations found in the monitoring and may help identify avoidable contaminations.
- The RIVM recommends that the ALARA (As Low as Reasonably Achievable) principle is applied, in particular for MOAH, since mutagenicity is assumed to be a non-threshold effect. It is noted that for MOH that are authorized as food additive or plant protection product, specific limits apply.
- If action threshold are being set for MOH in food, an option is to derive them from the 95<sup>th</sup> percentile of the measured concentrations in the food. New monitoring data of MOH in food can be used for this.



## 8 Acknowledgements

The authors thank Wim Mennes, Gerrit Wolterink, Anton Rietveld and Polly Boon of the RIVM for their valuable comments on an earlier version of the letter report.





## 9 References

Adenuga D, Goyak K, Lewis J, 2017. Evaluating the MoA/human relevance framework for F-344 rat liver epithelioid granulomas with mineral oil hydrocarbons. *Critical Reviews in Toxicology* 7:9, 750-766. Available online: <https://doi.org/10.1080/10408444.2017.1319336>

ANSES, 2017. Opinion on the migration of mineral oil compounds into food from recycled paper and cardboard packaging. Report 2015-SA-0070 of 8 March 2017. Available online: <https://www.anses.fr/en/system/files/ESPA2015SA0070EN.pdf>

Barp L., Suman M., Lambertini F., Moret S., 2015a. Migration of selected hydrocarbon contaminants into dry pasta packaged in direct contact with recycled paperboard. *Food Additives and Contaminants*, 2015; 32 (2): 271-283. Available online: <https://doi.org/10.1080/19440049.2014.999259>

Barp L., Suman M., Lambertini F., Moret S., 2015b. Migration of selected hydrocarbon contaminants into dry semolina and egg pasta packed in direct contact with virgin paperboard and polypropylene film. *Food Additives and Contaminants*, 2015; 32 (9): 1542-1551. Available online: <https://doi.org/10.1080/19440049.2015.1075176>

BfR, 2018. Highly refined mineral oils in cosmetics: Health risks are not to be expected according to current knowledge. Updated BfR Opinion No. 008/2018 of 27 February 2018. Available online: <https://www.bfr.bund.de/cm/349/highly-refined-mineral-oils-in-cosmetics-health-risks-are-not-to-be-expected-according-to-current-knowledge.pdf>

Biedermann M. and Grob K., 2010. Is recycled newspaper suitable for food contact materials? Technical grade mineral oils from printing inks. *European Food Research and Technology*, 2010; 230: 785-796. Available online: <https://doi.org/10.1007/s00217-010-1223-9>

Biedermann-Brem S. and Grob K., 2011. Removal of mineral oil migrated from paperboard packing during cooking of foods in boiling water. *European Food Research and Technology*, 2011; 232: 1035-1041. Available online: <https://doi.org/10.1177/1177390118777757>

Biedermann M., Ingenhoff J., Zurfluh M., Richter L., Simat T., Harling A., Altkofer W., Helling R., Grob K., 2013. Migration of mineral oil, photoinitiators and plasticisers from recycled paperboard into dry foods: a study under controlled conditions. *Food Additives and Contaminants*, 2013; 30 (5): 885-898. Available online: <https://doi.org/10.1080/19440049.2013.786189>

Biedermann M. and Grob K. (2015) Comprehensive two-dimensional gas chromatography for characterizing mineral oils in foods and distinguishing them from synthetic hydrocarbons. *Journal of Chrom. A* 1375, 146. Available online at <https://doi.org/10.1016/j.chroma.2014.11.064>

BLL, 2018. Toolbox for Preventing the Transfer of Undesired Mineral Oil Hydrocarbons into Food. Available online: <https://www.bll.de/download/toolbox-for-preventing-the-transfer-of-undesired-mineral-oil-hydrocarbons-into-food>

Brandenberger S., Mohr M., Grob K., Neukom H.P., 2005. Contribution of unburned lubricating oil and diesel fuel to particulate emission from passenger cars. *Atmospheric Environment*, 2005; 39: 6985–6994. Available online: <https://doi.org/10.1016/j.atmosenv.2005.07.042>

Bratinova S., Hoekstra E. (Editors), 2019. Guidance on sampling, analysis and data reporting for the monitoring of mineral oil hydrocarbons in food and food contact materials. Report of the Joint Research Centre of the European Commission. Luxembourg: Publications Office of the European Union, 2019 ISBN 978-92-76-00172-0, doi: 10.2760/208879, JRC115694. Available online at <http://publications.jrc.ec.europa.eu/repository/handle/JRC115694>

Brühl L., 2016. Occurrence, determination, and assessment of mineral oils in oilseeds and vegetable oils. *European Journal of Lipid Science and Technology*, 2016; 118: 361-372. Available online: <https://doi.org/10.1002/ejlt.201500528>

Concawe 2016. Critical review of the relationship between IP346 and dermal carcinogenic activity. Report 6/16, March 2016. Available online: <https://www.concawe.eu/wp-content/uploads/2017/01/report-6-16-final-version-3.pdf>

EC, 2008. Regulation (EC) No 1272/2008 of the European Parliament and of the Council of 16 December 2008 on classification, labelling and packaging of substances and mixtures. Available online (consolidated version dec 2018): <http://data.europa.eu/eli/reg/2008/1272/2018-12-01>

EC, 2017. Commission Recommendation (EU) 2017/84 of 16 January 2017 on the monitoring of mineral oil hydrocarbons in food and in materials and articles intended to come into contact with food. *OJ L* 12/95. Available online: <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32017H0084&from=EN>

EFSA, 2008. Polycyclic aromatic hydrocarbons in food. Scientific opinion of the panel on contaminants in the food chain. *EFSA Journal* 742: 1-114. EFSA, 2012. Available online: <https://www.efsa.europa.eu/en/efsajournal/pub/724>

EFSA, 2011. Evaluation of the FoodEx, the food classification system applied to the development of the EFSA comprehensive European food consumption database. EFSA J. 9:1970. Available online: <https://www.efsa.europa.eu/en/efsajournal/pub/1970>

EFSA, 2012. Scientific Opinion on Mineral Oil Hydrocarbons in Food. EFSA Panel on Contaminants in the Food Chain (CONTAM). EFSA Journal 2012; 10 (6): 2704. [185 pp.] doi:10.2903/j.efsa.2012.2704. Available online: <https://www.efsa.europa.eu/en/efsajournal/pub/2704>

FAVV (2017). Inventaris acties en actiegrenzen en voorstellen voor harmonisering in het kader van de officiële controles. Deel 1. Online beschikbaar: <http://www.favv-afsc.fgov.be/thematischepublicaties/inventaris-acties.asp>

Foodwatch, 2015. Besmetting van ons voedsel met minerale oliën. Resultaten van de foodwatch Test. Foodwatch Nederland, oktober 2015. Available online: [https://www.foodwatch.org/fileadmin/foodwatch.nl/Onze\\_campagnes/Sc\\_hadelijke\\_stoffen/Documents/Rapport\\_Besmetting\\_van\\_ons\\_voedsel\\_m\\_et\\_mineraleolie\\_testresultaten.pdf](https://www.foodwatch.org/fileadmin/foodwatch.nl/Onze_campagnes/Sc_hadelijke_stoffen/Documents/Rapport_Besmetting_van_ons_voedsel_m_et_mineraleolie_testresultaten.pdf)

Foodwatch, 2016a. Mineröl in Schokoladen-Osterhasen. Foodwatch Deutschland, März 2016. Available online: [https://www.foodwatch.org/fileadmin/foodwatch.nl/Onze\\_campagnes/Sc\\_hadelijke\\_stoffen/Vitamin\\_research/Documents/Final\\_Schoko-Hasenreport.pdf](https://www.foodwatch.org/fileadmin/foodwatch.nl/Onze_campagnes/Sc_hadelijke_stoffen/Vitamin_research/Documents/Final_Schoko-Hasenreport.pdf)

Foodwatch, 2016b. Mineröle in Schokoladen-Weihnachtsmännern. Foodwatch Deutschland, December 2016. Available online: [https://www.foodwatch.org/fileadmin/Themen/Mineraloel/Dokumente/2016-12\\_Test\\_Mineraloele\\_Weihnachtsschokolade.pdf](https://www.foodwatch.org/fileadmin/Themen/Mineraloel/Dokumente/2016-12_Test_Mineraloele_Weihnachtsschokolade.pdf)

Grob K., 2018a. Mineral oil hydrocarbons in food: a review. Food Additives and Contaminants, 2018; Part A, Available online: <https://doi.org/10.1080/19440049.2018.1488185>

Grob K., 2018b. Toxicological assessment of mineral hydrocarbons in foods: state of present discussions. Journal of Agricultural and Food Chemistry, 2018; 66 (27): 6968-6974. Available online: <https://doi.org/10.1021/acs.jafc.8b02225>

Lachenmeier DW, Mildau G, Rullmann A, et al. Evaluation of mineral oil saturated hydrocarbons (MOSH) and mineral oil aromatic hydrocarbons (MOAH) in pure mineral hydrocarbon-based cosmetics and cosmetic raw materials using <sup>1</sup>H NMR spectroscopy. F1000Res. 2017;6:682. <https://doi.org/10.12688/f1000research.11534.2>

Lommatzsch M., Biedermann M., Grob K., Simat T. J., 2016. Analysis of saturated and aromatic hydrocarbons migrating from a polyolefin-based hot-melt adhesive into food. Food Additives and Contaminants, 2016; 33 (3): 473-488. Available online: <https://doi.org/10.1080/19440049.2015.1130863>

Lorenzini R., Biedermann M., Grob K., Garbini D., Barbanera M., Braschi I., 2013. Migration kinetics of mineral oil hydrocarbons from recycled paperboard to dry food: monitoring of two real cases. *Food Additives and Contaminants*, 2013; 30 (4): 760-770. Available online:

<https://doi.org/10.1080/19440049.2013.766765>

Neukom H. P., Grob K., Biedermann M., Noti A., 2002. Food contamination by C20-C50 mineral paraffins from the atmosphere. *Atmospheric Environment*, 2002; 36: 4839-4847. Available online:

[https://doi.org/10.1016/S1352-2310\(02\)00358-8](https://doi.org/10.1016/S1352-2310(02)00358-8)

Populin T. , Biedermann M., Grob K., Moret S., Conte L. (2004). Relative hopane content confirming the mineral origin of hydrocarbons contaminating foods and human milk Relative hopane content confirming the mineral origin of hydrocarbons contaminating foods and human milk. *Food Additives and Contaminants* 21(9), 893. Available online at <https://doi.org/10.1080/02652030400001164>

SciCom (2017). Actiedrempels voor minerale olie koolwaterstoffen in levensmiddelen. ADVIES 19-2017 van 22 september 2017. Available online:

[http://www.afsca.be/wetenschappelijkcomite/adviezen/2017/\\_document s/Advies19-2017\\_SciCom2016-15\\_mineraleoliekoolwaterstoffen.pdf](http://www.afsca.be/wetenschappelijkcomite/adviezen/2017/_document s/Advies19-2017_SciCom2016-15_mineraleoliekoolwaterstoffen.pdf)

Spack L, Leszczyk G , Varela J, Simian H, Thomas, Gude T, Stadler R (2017): Understanding the Contamination of Food with Mineral Oil: The Need for a Confirmatory Analytical as well as Procedural Approach. *Food Addit & Contam* 34(6): 1052-1071. Available online at

<https://doi.org/10.1080/19440049.2017.1306655>

Sturaro A, Parvoli G, Rella R, Bardati S, Doretti L. 1994. Food contamination by diisopropylnaphthalenes from cardboard packages. *International Journal of Food Science and Technology*, 1994; 29: 593–603. Available online: <https://doi.org/10.1111/j.1365-2621.1994.tb02102.x>

Thoden van Velzen E.U. Leeman W.R., Krul L., 2018. Levensmiddelenverpakkingen gemaakt van oud-papier en karton: migratie van minerale oliën. WUR rapport 1789, 29 maart 2018. Available online:

<https://doi.org/10.18174/444489>

Van de Ven B.M., Fragki S., Te Biesebeek J.D., Rietveld A.G., Boon, P.E., 2018. Mineral oils in food; a review of toxicological data and an assessment of the dietary exposure in the Netherlands. RIVM report 2017-0182. Bilthoven, the Netherlands: National Institute for Public Health and the Environment. Available online:

<https://www.rivm.nl/bibliotheek/rapporten/2017-0182.pdf>

Van Heyst A., Vanlancker M., Vercammen J., Van den Houwe K., Mertens B., Elskens M., Van Hoeck E., 2018. Analysis of mineral oil in food: results of a Belgian market survey. *Food Additives and Contaminants*, 2018; 35 (10) Available online:

<https://doi.org/10.1080/19440049.2018.1512758>

VCP 2012-2016 Voedselconsumptiepeiling. Available online at:

<https://www.rivm.nl/voedselconsumptiepeiling/overzicht-voedselconsumptiepeilingen/vcp-basis-1-79-jaar-2012-2016>

Vollmer A, Biedermann M, Grundböck F, Ingenhoff JE, Biedermann-Brem S, Altkofer W, Grob K., 2011. Migration of mineral oil from printed paperboard into dry foods: survey of the German market. *European Food Research and Technology* 232: 175–182. Available online:

<https://doi.org/10.1007/s00217-010-1376-6>

Weber, S., Schrag, K., Mildau, G., Kuballa, T., Walch, S.G., Lachenmeier, D.W. (2018) Analytical Methods for the Determination of Mineral Oil Saturated Hydrocarbons (MOSH) and Mineral Oil Aromatic Hydrocarbons (MOAH)—A Short Review. *Analytical Chemistry Insights* 13: 1–16. Available online at

<https://doi.org/10.1177/1177390118777757>

