Mineral oil hydrocarbons in minimally processed nutraceutical oils

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Abstract

The presence of unintended chemicals in food products and supplements may impact consumers' health negatively. Mineral oil hydrocarbons (MOHs) in particular are gaining research attention and have been detected and quantified in food products and supplements in the past. The aim of this study was to analyze encapsulated, and bulk minimally processed marine oils for MOHs and to evaluate the probable sources of these compounds. Hydrocarbons in supplement oils were extracted via saponification and analyzed by gas chromatography with both flame ionization and mass spectral detection. While no mineral oil aromatic hydrocarbons (MOAH) were detected in any sample, the analysis revealed the presence of mineral oil saturated hydrocarbons (MOSH) in 9 out of 10 minimally processed encapsulated oils. The MOSH appeared on the chromatograms as an unresolved complex mixture (UCM) with concentrations ranging from 376 ± 49 to 3831 ± 414 mg kg-1. These values are well below the maximum allowable limits for MOH in encapsulated products set by the United States Food and Drug Administration. Therefore, all the tested products are compliant with the US regulations. Moreso, the bulk oil samples did not contain detectable levels of MOH. This study suggests that MOH accumulation in encapsulated products is likely due to the use of lubricants during encapsulation, rather than environmental sources such as oil spills since MOAH that are characteristic of weathered petroleum products were not identified in the UCM.

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Abstract

The presence of unintended chemicals in food products and supplements may impact consumers' health negatively. Mineral oil hydrocarbons (MOHs) in particular are gaining research attention and have been detected and quantified in food products and supplements in the past. The aim of this study was to analyze encapsulated, and bulk minimally processed marine oils for MOHs and to evaluate the probable sources of these compounds. Hydrocarbons in supplement oils were extracted via saponification and analyzed by gas chromatography with both flame ionization and mass spectral detection. While no mineral oil aromatic hydrocarbons (MOAH) were detected in any sample, the analysis revealed the presence of mineral oil saturated hydrocarbons (MOSH) in 9 out of 10 minimally processed encapsulated oils. The MOSH appeared on the chromatograms as an unresolved complex mixture (UCM) with concentrations ranging from 376 ± 49 to 3831 ± 414 mg kg^{-1}. These values are well below the maximum allowable limits for MOH in encapsulated products set by the United States Food and Drug Administration. Therefore, all the tested products are compliant with the US regulations. Moreso, the bulk oil samples did not contain detectable levels of MOH. This study suggests that MOH accumulation in encapsulated products is likely due to the use of lubricants during encapsulation, rather than environmental sources such as oil spills since MOAH that are characteristic of weathered petroleum products were not identified in the UCM.

Keywords: Nutraceutical oils, Mineral oil hydrocarbons, unresolved complex mixtures, fish oils

Introduction

Dietary supplements have become part of day-to-day life, especially in the developed world as consumers begin to recognize their health benefits. With many people becoming more aware of their nutritional needs, it is not surprising that consumption of dietary supplements is on the increase (Hamulka et al., 2021). However, the presence of unintended materials in dietary supplements has recently gained attention (Mathews, 2018) and may pose a potential threat to the upward trajectory in the utilization of these health products. The occurrence of mineral oil hydrocarbons (MOH) is of particular interest because they have been detected in some dietary supplement oils (Arena et al., 2021; Reid & Budge, 2015).

MOH are a complex mixture of hydrocarbons (HC) that originate primarily from crude oil (Alexander et al., 2012). In food and oil supplements, they may arise from various sources, including packaging materials, the environment, processing aids, and lubricants. They are defined as molecules containing between 10 to 50 carbon atoms and are categorized into two groups, namely, mineral oil saturated hydrocarbons (MOSH) and mineral oil aromatic hydrocarbons (MOAH). MOSH include the straight chain and branched alkanes, as well as cycloalkanes, while the MOAH group consists mainly of the alkylated mono- and polycyclic aromatic hydrocarbons (PAHs) which are the predominant class of MOH found in food. About 16 priority PAHs have been listed by the Environmental Protection Agency for monitoring in the US based on their prevalence and toxicity (Zelinkova & Wenzl, 2015). MOAH are typically not present in food grade mineral oils (white oils) but are found in technical grade oils used for machinery lubrication. The MOSH fraction is not carcinogenic, but may act as a tumor promoter at high concentrations (Alexander et al., 2012). MOAH fractions pose the most concern over carcinogenicity (Xie et al., 2019) and genotoxicity, prompting the recent announcement by the

European Union member states (2022) of new limits on their content in foods (https://food.ec.europa.eu/system/files/2022-05/reg-com_toxic_20220421_sum.pdf).

A number of different organizations have set limits on MOH in food. For instance, the United States Food and Drug Administration (FDA) allows mineral oil at levels up to 0.6% in encapsulated oil products (21CFR 172.878), while the Joint FAO/WHO Expert Committee on Food Additives (JECFA) set an acceptable average daily intake (ADI) for MOSH at 0.01 mg kg^{-1} body weight (Alexander et al., 2012); based on this ADI, a limit of 0.6 mg kg^{-1} was derived for MOSH in food by Biedermann and Grob (2012). Furthermore, the European Union Commission set the legal limit for total MOH in sunflower oil at 50 mg kg^{-1} in 2008 (Grob, 2008). In 2012, a special European Food Safety Authority (EFSA) panel estimated that MOSH exposure from food in adults ranged between 0.03 to 0.3 mg kg^{-1}, while children experience higher exposure, and concluded that MOAH exposure is about 20% of total MOSH (Alexander et al., 2012). Thus, amount of contact and allowable limits in edible products varies widely for MOSH.

MOH have been quantified in a variety of food products. For instance, Canaver et al. (2018) reported the presence of MOH in dry foods including rice, corn flakes, sea salt, and oatmeal. They found that 29% of the products tested contained over 1.0 mg kg^{-1} MOAH, with oatmeal having the greatest amount at 2.72 mg kg^{-1}. Similarly, in a MOH survey of 51 infant formulae based on cow and goat milk sold in China market, 17 out of 51 samples analyzed were confirmed to contain MOSH, albeit at trace levels of less than 0.7 mg kg^{-1}. However, a goat milk-based infant formula had the highest concentration of MOSH at 3.5 mg kg^{-1} (Sui et al., 2020). The migration of MOH from packaging materials to food products/simulant has also been reported which further established that packaging materials such as printed paper may be a risk factor in overall MOH accumulation in food, especially in long term storage applications (Pan et al., 2021). The presence of MOH in omega-3 dietary supplements has also been reported. Reid and Budge (2015) measured the levels of weathered petroleum HC, a form of MOH that appears on chromatograms as unresolved complex mixtures (UCM), in unrefined and refined salmon, and refined sardine, anchovy, mackerel supplement oils. The fully refined oils did not contain UCM, suggesting that refining reduces MOH in oil supplements. More recently, Arena et al. (2021) evaluated 17 omega-3 supplements (fish, vegetable, and microalgae oils) and found MOSH and MOAH at varying concentrations but there was no source attribution.

The environment is an obvious source of MOH in food products and oil supplements. Oil spills in large bodies of water are an example of common environmental pollution. The spilled oil generally degrades over time to form weathered petroleum HC that accumulate in the tissues of marine animals. Most edible oils undergo several refining steps for purification and to improve stability and, as such, most contaminants including MOH are removed. This implies that refined oils should be free from such environmental pollutants (Reid & Budge, 2015); however, this might present a significant source of MOH in unrefined marine oils. MOH may also arise in foods from the use of additives such as antioxidants and stabilizers since these are added after refining. Furthermore, the use of MOH-based processing aides and lubricants in the post-refining steps such as encapsulation and bottling present a considerable risk of HC accumulation but are often overlooked. For instance, Reid and Budge (2015) found MOH in encapsulated oil products, but they only considered the oil and the capsule material as the potential source of MOH, rather than the encapsulation process itself. Therefore, the aims of this study were to determine if commercially available, minimally processed oil supplements contained detectable levels of petroleum HC and to evaluate the probable sources. To further explore potential sources, HC were determined in several fully refined oils in both encapsulated and bulk form.

Experimental Procedures

Oils and sample preparation

Encapsulated minimally processed marine oils (n=12) were purchased from online retailers, while bulk versions (n=4) of the same oils were kindly provided by their manufacturers. All were derived from marine fish or copepods. Two additional fully refined fish and algae oils were provided by the manufacturer and were also analyzed in their encapsulated and bulk forms.

The exterior of all capsules was rinsed with dichloromethane to remove any lubricating agents, flavours and other substances that could interfere with analysis. Chemicals and solvents were purchased from Fisher Scientific Company (Guelph, ON, Canada) unless otherwise stated. Prior to use, all glassware was washed with soap and water, rinsed with acetone, dried at 100 °C, and rinsed with dichloromethane.

Extraction of hydrocarbons via saponification

A condenser was pre-cleaned by refluxing with dichloromethane for 30 minutes. Approximately 0.2 g of capsule contents were added to a 250 ml round bottom flask spiked with 20 ug of pentacosane (C25) (Sigma-Aldrich, Oakville, ON, Canada). Samples were analyzed in triplicate. Boiling chips and 25 ml of freshly prepared 2M KOH in 95% ethanol was added to the round bottom flask and the contents were saponified by refluxing for 2 hours.

Isolation of non-saponifiable material

The glassware was allowed to cool, and 25 ml of hexane was poured through the condenser into the round bottom flask and the contents were transferred to a 250 ml separatory funnel. The round bottom flask was rinsed once more with 10 ml hexane and pooled with the rest of the extract in the separatory funnel. The extract was washed with 80 ml deionized water (dH_2O) and 20 ml saturated NaCl several times to get rid of saponifiable material, removing the lower phase to waste each time. The upper phase was poured off the top of the first separatory funnel into a second separatory funnel and washed several more times with 80 ml dH_2O and 20 ml saturated NaCl. After removing the lower phase to waste for the final time, the upper phase was poured out the top into a 40 ml centrifuge tube. The tube was centrifuged for 15 minutes. The hexane and non-saponifiable matter were carefully transferred through anhydrous sodium sulphate and filter paper into a clean 40 ml centrifuge tube, taking care not to transfer any particulate that collected during centrifuging. The collected hexane with non-saponifiable material was then evaporated under nitrogen to approximately 1 ml.

Isolation of hydrocarbons from other non-saponifiable material

A 6-cc silica Sep-Pak (Waters, Milford, MA, USA) was rinsed with 20 ml dichloromethane and then 20 ml hexane and the non-saponifiable material were quantitatively transferred to the Sep-Pak with 3 x 2 ml rinses of hexane. The eluents and any unsaponifiable material not retained in the silica were collected in a 15 ml round bottom test tube. The Sep-Pak was rinsed with 6 ml 2:3 dichloromethane:hexane (v/v) into the same round bottom test tube. The collected unsaponifiable material was then evaporated under nitrogen to approximately 1 ml for analysis by GC-FID. A mineral oil was carried through this procedure to confirm that the UCM was successfully isolated with this process.

Analysis by GC-FID and GC-MS

Analysis of sample extracts was performed on a Bruker Scion 436-GC with a flame ionization detector (FID) and a Zebron ZB-5 capillary column (5% Phenyl 95% Dimethylpolysiloxane, 30 m x 0.25 mm i.d.; Phenomenex, Torrance, CA, USA). The injector temperature was set to 250 °C and splitless injection was used. The oven temperature was initially held to 60 °C for 15 mins, then ramped at 13 °C min^{-1} to 280 °C, held for 5 minutes, and finally ramped 50 °C min^{-1} to 300 °C, holding for 22.68 minutes. The FID temperature was set to 300 °C. To further understand the composition of the samples, analysis was also performed on a Thermo Scientific Trace 1310 GC with ISQ 7000 Single Quadrupole Mass Spectrometer with the same column type and temperature program as the GC-FID analysis. Using the mass of oil analyzed and C25 as an internal standard, total concentration of petroleum HC (as UCM) was determined by summing peak areas under the range of the UCM. A blank sample was extracted and analyzed to ensure that HC were not added during sample work-up. Spectra were evaluated for the presence of ions at m/z 78, 91 and 120 to indicate benzene, methylated benzene and alkylated (C3) benzene (Wang et al 2002). The presence of ions associated with other aromatics, including m/z 128 (naphthalene), 156 (dimethylnaphthalenes) and 178 (phenanthrene) (Kao et al. 2015), was also assessed.

Results and Discussion

Of the minimally processed and encapsulated oils evaluated, 9 of the 10 products contained an obvious UCM (Table 1; Fig. 1) with HC content ranging from 376 to 3831 mg kg^{-1}. Encapsulated herring roe (supplement brand I-1) did not contain UCM. Furthermore, two of the encapsulated oils (supplement brands I-2 and J-2), that were also evaluated in bulk form, did not contain a UCM. Note that herring oil, whether encapsulated or in bulk form, did not show a

UCM. For comparison, HC were determined in encapsulated forms of a fully refined fish oil and a fully refined algal oil (Table 2) and similar HC contents were found in both (~1100 mg kg^{-1}). Bulk oils of the same refined products did not contain a detectable UCM (Table 2; Fig. 2).

All of the chromatograms were similar, with the UCM ending at ~35 min, ~2 mins after pentacosane had eluted. The UCM was evaluated for ions with masses associated with MOAH (i.e.,78, 91, 120, etc; see Methods) and none were identified, indicating that the MOH in the samples were MOSH. When the amounts of detected MOH were compared to regulatory limits put in place by the FDA, it was found that all were below the allowable limits, indicating that there was no safety risk associated with these products and that they comply with regulatory requirements.

All fish oils contained sterols and squalene, a biosynthetic precursor of sterols; the algal oil also contained a clear squalene peak. Pristane was a prominent peak in bulk and enapsulated calanus oils; it was also present in herring roe samples. These compounds were expected as they are all relatively non-polar biogenic compounds that elute with HC during column clean-up. National Institute of Standards and Technology (NIST) library matches suggested that the other sharp and well-resolved peaks in the chromatograms were saturated HC.

Previous assessments of areas affected by crude oil spills have found MOH in sediment and marine organisms. For instance, Lance et al. (2012) reported the presence of polycyclic aromatic hydrocarbons (PAHs) in the sediments and marine life of Nelson Lagoon, Alaska. They found that the tissues of blue mussels had absorbed high levels of PAHs, particularly benzo (a) pyrene. Page et al. (2004) also confirmed the presence of PAHs in fishes sourced from the eastern Gulf of Alaska. This suggests that UCM detected in the supplement oils could be due to oil spills, leading to environmental pollution. The MOH found in the current study were initially thought to be derived from past oil spills based; however, such crude oils would be expected to contain MOAH, and none were identified by mass spectrometry of the UCMs. The lack of aromatics suggests that the UCM here are unlikely to be derived from environmental sources such as oil spills. Additionally, supplements brand K-1 made from algae also contained UCM. The algae cultures used in the production of the supplement oil were grown in a controlled facility that was not exposed to the outside environment, and therefore the UCM is clearly not from an environmental source in this sample. So, while we have shown that all but one of the minimally processed and encapsulated supplement oils tested here contained detectable levels of HC, the lack of MOAH in the samples indicates that the MOH are not a result of a previous crude oil spill. This leaves additives and processing aides as the potential sources of the UCM.

Marine oils are subjected to different types and extents of processing by manufacturers based on the intended finished product. As expected, this difference in processing may affect the quality and composition of the supplement oil. At a basic level, some oils are not subjected to further processing steps after the “first press” and, thus, the resulting product is dubbed “minimally processed”. On the other hand, expressed oils could be subjected to different refining process steps including degumming, neutralization, bleaching, dewaxing, winterization and steam deodorization, resulting in a highly refined finished product (Gharby, 2022). Fish oils are commonly refined through molecular distillation, a process involving heating oils at temperatures between 130^{o}C to 150^{o}C in a column under a high vacuum (Rossi et al., 2012), leading to the purification and concentration of target distillation products such as omega-3 fatty acids. Regardless of the processing level applied, additives, such as flavors and antioxidants, are often added to nutraceutical oils as a final step before encapsulation or bottling. Thus, these additives could be a source of the HC detected in both the minimally processed and fully refined oil samples tested in this study. According to the available label information, additives such as oregano extract, tocopherols, and rosemary extract were included in the formulation of several of the supplement oils evaluated here, presumably to prevent oxidation. Some of the supplement brands did not contain any additives. Given that different additives were used in the tested samples and not all oils contained additives, it seems unlikely that additives could be the source of HC. Further, antioxidants and stabilizers are normally added to oils at the ppm level (Barrett et al., 2011; Budilarto & Kamal-Eldin, 2015; Mihaylova et al., 2020) and, in some samples, HC were quantified in the same range; if the additive was the sole source of HC, it would have to consist entirely of HC to generate that concentration in the oil. Additives are typically produced by third party suppliers and such a gross error in composition seems exceedingly unlikely.

UCM was not detected in any bulk oil analysed in this study, yet all but one encapsulated oil contained UCM (Table 1 & 2). The UCM detected in the encapsulated products were unlikely to be from environmental sources or additives, making processing aides the remaining probable source of the HC. Encapsulation is the only step that differs between bulk oils and encapsulated products. At that stage, processing aides such as white mineral oil are used as a release agent and lubricant on gelatin sheets that form the capsules (Gullapalli, 2010). The formed capsules are then tumbled in a dryer with adsorbent towels to remove lubricant from the exterior of the capsules (Gullapalli, 2010). White mineral oil is approved by the FDA for use as a processing aide at a maximum level of 0.6% and all tested products were below this level, demonstrating that supplement manufacturers are adhering to regulatory guidelines and producing products that are both safe and compliant. Since the bulk oils did not contain UCM but the encapsulated version of the same oil did, logic suggests that UCM detected in the encapsulated versions were introduced during the encapsulation step.

This present study has further confirmed the presence of UCM in commercial dietary supplement oils, consistent with previous studies (Arena et al., 2021; Reid & Budge, 2015); however, no MOAH were identified. As knowledge around the potential health risks associated with MOAH increases, regulatory bodies around the world have begun to set maximum allowable limits for certain MOH, specifically MOAH (Alexander et al., 2012), with which the tested products were compliant. While most of the manufacturers confirmed on their product labels and/or websites that they test for chemical contaminants like mercury, dioxins, and PCBs, regrettably, none made mention of HC testing, likely because it is not yet required for regulatory purposes in all markets. As regulatory bodies begin to study MOH levels in food products and implement maximum allowable levels for MOAH, it is recommended that manufacturers take the pre-emptive step of including analysis for MOH as part of routine testing before releasing their finished products for sale. Additionally, more research is still required to evaluate the effects of MOH on human health since most of the identified negative implications are directly related to marine organisms.

Conclusion

Our study has demonstrated that encapsulated supplement oils often contain MOSH that are apparent in GC chromatograms as UCM and have concentrations ranging from ~ 400 – 4000 mg kg^{-1}. These levels are all well within regulatory limits for MOH content and do not pose a safety risk. No MOAH were detected in any encapsulated or bulk oil tested here. Though possible sources of MOSH include the environment, additives, and processing, our research suggests that the encapsulation step is likely the source of MOH found in the analyzed supplement products. Moreover, this study suggests that the encapsulation step during processing leads to varying amounts of MOSH in oils but also that supplement manufacturers are well aware of the regulatory limits surrounding the use of white mineral oil and are adhering to these requirements. To avoid the presence of MOSH in encapsulated oils, manufacturers could consider the use of lubricants other than mineral oil.

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Authorship

JSR and SB conceived and designed the study and supervised the analysis. CB conducted the laboratory analysis. All four authors contributed to data analysis. OA led the writing of the manuscript, with contributions from JSR and SB.

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Conflict of interest statement

The authors declare they have no competing interests

Figure legends

Fig. 1 GCFID chromatogram of non-polar non-saponifiable components in salmon oil (Supplement brand A)

Fig. 2 Comparison of nonpolar fraction of non-saponifiable material in encapsulated and bulk oils: A) encapsulated and minimally refined calanus oil (Brand J-1); B) encapsulated and refined fish oil (Brand L-1); C) encapsulated and refined algal oil (Brand K-1); D) minimally refined bulk calanus oil (Brand J-2); E) refined bulk fish oil (Brand L-2); and F) refined bulk algal oil (Brand K-2)

Tables

Table 1. UCM content of minimally processed oil supplement (mean +/- sd; n=3)

Supplement Brand	**Source**	**UCM (mg kg^{-1})**	**Others**	**Type**
A	Salmon	2498 ± 235	SQ	Encapsulated
B	Salmon	3344 ± 561	SQ	Encapsulated
C	Salmon, menhaden	1411 ± 20	SQ, ST	Encapsulated
D	Anchovy, sardine, jack mackerel	376 ± 49	SQ, ST	Encapsulated
E	Krill	3831 ± 414		Encapsulated
F	Krill	2511 ± 109		Encapsulated
G	Krill	845 ± 250		Encapsulated
H	Salmon	2428 ± 579	SQ, ST	Encapsulated
I-1	Herring roe	ND	SQ, ST, PR	Encapsulated
I-2	Herring roe	ND	SQ, ST, PR	Bulk
J-1	Calanus	3156 ± 827	PR	Encapsulated
J-2	Calanus	ND	PR	Bulk

SQ – Squalene; ST – Sterol; PR – Pristane; ND – Not detected

Table 2. UCM content of fully refined oil supplements (mean +/- sd; n=3).

Supplement Brand	**Source**	**UCM (mg kg^{-1})**	**Others**	**Type**
K-1	Algae	1054 ± 55	SQ	Encapsulated
K-2	Algae	ND	SQ	Bulk
L-1	Anchovy	1162 ± 21	SQ, ST	Encapsulated
L-2	Anchovy	ND	SQ, ST	Bulk

SQ – Squalene; ST – Sterol; ND – Not detected