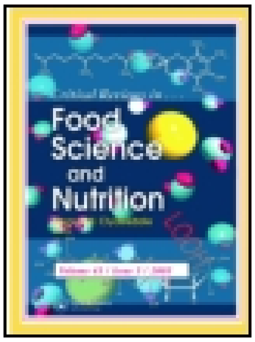

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REVIEW

The occurrence of 3-monochloropropane-1,2-diol esters and glycidyl esters in vegetable oils during frying

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ABSTRACT

3-monochloropropane-1,2-diol esters (3-MCPDE) and glycidyl esters (GE) are processed-developed contaminants presence in vegetable oils after undergo refining process under excessive heat. Refined oils are extensively used in various frying applications, nevertheless, the reservation against their quality and safety aspects are of major concern to consumers and food industry. Realizing the importance to address these issues, this article deliberates an overview of published studies on the manifestation of 3-MCPDE and GE when vegetable oils undergo for frying process. With the modest number of published frying research associated to 3-MCPDE and GE, we confined our review from the perspectives of frying conditions, product properties, antioxidants and additives, pre-frying treatments and frying oil management. Simplicity of the frying process is often denied by the complexity of reactions occurred between oil and food which led to the development of unwanted contaminants. The behavior of 3-MCPDE and GE is closely related to physicochemical characteristics of oils during frying. As such, relationships between 3-MCPDE and/or GE with frying quality indices – i.e. acidity in term of free fatty acid or acid value); secondary oxidation in term of *p*-anisidine value, total polar compounds and its fractions, and refractive index – were also discussed when oils were subjected under intermittent and continuous frying conditions.

KEYWORDS

3-monochloropropane-1,2-diol esters; glycidyl esters; contaminants; frying; quality indices

Introduction

3-monochloropropanediol esters (3-MCPDE) and glycidyl esters (GE) are heat-induced processed contaminants presence in refined oils. The former compound was first detected in different types of vegetable oils and animal fats back in 2006 (Zelinkova et al. 2006). In principle, 3-MCPDE is developed when triacylglycerols (TAG) react with chloride ion (which is the precursor of 3-MCPDE) in the presence of excessive heat. Oils in their native or unrefined form do not contain 3-MCPDE and instead, this contaminant is presence in most of refined oils (Weißhäar 2008). Unlike 3-MCPDE, the prevalence of GE in refined oils much coincides with diacylglycerols (DAG) content in crude oils (Craft et al. 2012).

The issues on 3-MCPDE in edible oils were first brought up by the Federal German Institute for Risk Assessment (BfR) in December 2007. Research done by the International Life Science Institute (ILSI) showed that 3-MCPDE can be detected in all refined vegetable oils and thermally processed foods (ILSI 2009). In March 2008, the European Food Safety Authority (EFSA) reported the consensus reached between the scientific committee of Contaminants in the Food Chain (CONTAM) and the BfR hypothesis on full conversion of 3-MCPDE into unbounded or free form (3-MCPD) (Fiebig 2011).

All refined oils generally face the issues on heat-induced contaminants when exposed to excessive heat (Kushairi, Singh, and Ong-Abdullah 2017). In September 2013, the European Food Safety Authority (EFSA) disseminated the data on 3-MCPDE content in various foods produced in the European Union (EU) from 2009 and 2011 as well as the establishment of tolerable daily intake (TDI) of 2 µg for every kg of body weight (bw) (European Food Safety Authority (EFSA)) 2013). Palm oil becomes the spotlight of food safety concerns associated to 3-MCPDE and GE over the years, and heightened in May 2016 when EFSA published a comprehensive report on the considerable amount of 3-MCPDE and GE presence in refined palm oil (EFSA 2016).

According to the International Agency for Research on Cancer (IARC), 3-MCPDE is classified as nephrotoxic while GE is denoted as probably carcinogenic to human (EFSA 2016). This clearly indicates that GE is more detrimental than 3-MCPDE particularly when these artifacts exist in their free form. Both of 3-MCPDE and GE are detectable in most vegetable oils and fats with considerable amount of these compounds are presence in palm oil (EFSA 2016).

The European Commission (EC) has published the Regulation (EU) 2018/290 on the threshold of GE in food on 26th February 2018 through the Official Journal of the European Union. The maximum limit for GE in vegetable

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oils and animal fats is established at 1 ppm. The GE allowance of 0.5 ppm is set for refined oils used to formulate food for young children and infants. Stringent GE limits are imposed for refined oils used to formulate food for special medical purposes for young children and infants, i.e. 0.075 ppm for powder form and 0.006 ppm for liquid form. The EU implementation of the maximum limits for GE has entered into force on 19th March 2018.

In early October 2018, the EC has proposed for two maximum levels of 3-MCPDE in refined oils. The proposal underwent deliberation and voting at the Standing Committee as well as public consultation and comment before EC came out with the finalized draft proposal on 30th June 2019. Vegetable oils including coconut, corn, rapeseed, sunflower, soybean, palm kernel and olive oils (consist of refined olive oil and virgin olive oil) are grouped under lower tier (1.25 ppm). Palm oil, which falls under the category of “other vegetable oils” is clustered at the higher level (2.5 ppm) together with pomace olive oil, fish oil and oils from other marine organisms. The European Commission (EC) has published the Regulation (EU) 2020/1322 on the maximum allowance of 3-MCPDE in food on 23rd September 2020 through the Official Journal of the European Union. This legislation is set to become mandatory by 1st January 2021.

Frying process

Frying is an ancient and well-established process in food preparation. A great demand in fried food consumption in the recent years is reflected by the capitalization of frying industry with usage of considerable amount of the global oils and fats. In 2019, the oils and fats consumption rose tremendously by almost three-and-the-half folds higher (from 61,704,000 tonne to 210,356,000 tonne) as compared to 1990 (Oil World Annual 2020). Moreover, 85% of the total oils and fats production is dedicated for food industry of which more than half of the value is channeled for cooking and frying sectors.

Frying is essentially a technique to remove excessive water from the food through immersion into heated oil at temperature above boiling point of water, as high as 190 °C, to produce fried food with distinctive sensory attributes, (i.e. texture, flavor and appearance), palatable and desirable (Aydınkaptan and Barutçu Mazı 2017). This process shares similar principle as baking where a brownish crust is developed on the food outer layer which contribute to its fried taste and flavor (Berger 2005). However, frying generally cooks food faster than baking because of its higher heat transfer coefficient (Tuta and Palazoğlu 2017). Owing its convenient, simplicity and economic viable operation, frying boosts the extensive sales of a large variety of fried products worldwide. As considerable amount of oil is integrated by fried food, the oil used for frying provides high source of energy of up to 9 kcal g^{-1}, lipid soluble vitamins (A, D, E and K) and essential fatty acids (i.e. oleic, linoleic and linolenic acids).

However, consumers are becoming more concern about their health and trending toward healthy food including the amount of oil intake in fried food. Susceptibility of frying oil to excessive heating in the presence of oxygen (air) and moisture leached out from food leads to multiple series of chemical reactions comprising hydrolysis, oxidation and polymerization. The mechanism of oil reactions during frying has been elaborated in detail by Nayak et al. (2016).

Hydrolysis primarily occurs when the moisture from food interacts with oil during frying. The leached water decomposes TAG bonds into lower-molecular-weight components such as DAG, monoacylglycerol (MAG), glycerol and free fatty acids (FFA) (Nayak et al. 2016). Some of these breakdown compounds can also be presence through partial hydro-peroxidation at excessive temperature in the presence of air (Bensmira et al. 2007). Acceleration of hydrolytic reaction is prominent with the increase of frying temperature and high moisture content in fried food. Additionally, the degree of hydrolysis is determined by the chain length of fatty acids. (Choe and Min 2007).

Oxidative reaction can be clustered into three categories namely auto-, thermal- and photosensitized oxidation (Nayak et al. 2016). This reaction prevails through removal of hydrogen from fatty acids in the presence of heat, trace metals and/or light. The alkyl radicals subsequently propagate with oxygen to generate peroxyl radicals, and subsequently eliminates hydrogen from different fatty acids to form hydroperoxides and new alkyl groups (Nayak et al. 2016; Dueik and Bouchon 2011). Secondary oxidation occurs when hydroperoxides are further decomposed into carbonyl compounds (i.e. aldehydes and ketones), alcohols, hydrocarbons and acids (Chatzilazarou et al. 2006). The number of double bonds presence in the fatty acids structure at the glycerol backbone influences the severity of oil oxidation (Sangle and Daptare 2014). Apart from oil stability, oxidation could also affect the sensorial properties of fried food such as aroma, flavor, color and nutrient content (Dana and Saguy 2001).

Polymerization is an advance stage of secondary oxidation where constituents such as carbonyls, alcohols and fatty acids will further react with oxygen (air) and produces higher-molecular weight compounds namely polymer compounds (Gupta 2005). In fact, this reaction could even take place in the absence of oxygen when the oil undergoes thermal alteration of TAG molecules when exposed to excessive heat. Formation of polymer compounds is often associated with increase in oil viscosity and color, tendency of foaming during frying and impart bitter taste to fried food (Nayak et al. 2016).

The complexity of all reactions is pronounced when oil breakdown products interact with food and changes the properties of frying oil (Dueik and Bouchon 2011). The tendency of reaction pathways is manifested in reliant to the composition of oil used for frying (Olivero-David et al. 2014), food morphological properties and ingredients (Karoui et al. 2011), frying procedures (Aladedunye and Przybylski 2009a), and process conditions (Aladedunye and Przybylski 2009b).

Since fried food absorbs considerable amount of oil, quality deterioration associated to frying would not only exerts undesired fried food quality (Gertz and Behmer 2014) but also produces unwanted breakdown constituents that can affect human health (Zribi et al. 2014). Such consumer requirements have compelled industry to search for alternative frying methods to produce fried food with least oil content without affecting its sensory attributes, and at the same time, to protect and minimize oil deterioration during frying. Moreover, food industry could even gain advantages to increase the mass production of fried food without necessarily increasing the operation cost considering reduced amount of oil needed for replenishment as a result of lower oil absorption and oil deterioration. The food safety challenges encountered by the frying sectors are such as the presence of oxidized and polymerized compounds (Venkata and Subramanyam, 2016), acrylamide (Kuek et al. 2020), fumes and volatile organic compounds (Chen et al. 2018), polycyclic aromatic hydrocarbons (An, Liu, and Liu 2017) and *trans*-fatty acids (Song et al. 2015). As the global edible oils and fats producers are currently tasked with the heat-induced contaminants namely 3-MCPDE and GE, the issues would also affect the frying industry.

Factors to consider in the manifestation of 3-MCPDE and GE during frying

Over the last five years, researchers have started to investigate the fate of 3-MCPDE and GE in refined oils used in food processes including frying process. To date, more than ten publications have reported the transients of 3-MCPDE and GE in different vegetable oils including palm oil and its liquid fraction namely as palm olein. The behaviors of 3-MCPDE and/or GE are influenced by several factors such as type of frying media (Hammouda et al. 2017; Aniołowska and Kita 2015b), frying methods (Belkova et al. 2018; Dingel and Matissek 2015), frying temperature (Turan, Solak, and Keskin 2019; Wong et al. 2017a; Wong et al. 2017b; Aniołowska and Kita 2016), antioxidants and additives (Ahmad Tarmizi et al. 2019; Wong et al. 2019; Yıldırım and Yorulmaz 2018), salt impregnation (Bognár et al. 2020; Turan, Solak, and Keskin 2019; Arisseto et al. 2019; Arisseto et al. 2017; Wong et al. 2017a; Wong et al. 2017b), product morphological properties and cultivars (Arisseto et al. 2019; Arisseto et al. 2017; Aniołowska and Kita 2015a), pre-frying treatments (Arisseto et al. 2019; Merkle et al. 2018a, Merkle et al. 2018b), and adsorbent and filtration (Merkle et al. 2018a). Compilation of literatures that discussed on the occurrence of 3-MCPDE and GE in vegetable oils during frying is summarized in Table 1.

Type of frying media

Selection of frying oil is often associated with the ability of oil to withstand excessive heat. On that note, the degree of saturation is one of the key criteria for choosing the right oil for frying. Oil with significant amount of saturated fatty acids commonly imposes high thermal resistance when compared to typical liquid oils owing the number of double bonds presence in fatty acid structure that distinguishes the rate of oil degradation (Gupta 2005).

The endogenous formation of 3-MCPDE and GE in vegetable oils are perceived to have association with their origins. Seed oils such as refined sunflower, soybean, canola and corn oils are mostly reflected by their lower 3-MCPDE and GE contents while refined palm and olive oils, which are classified as fruit oils, gave relatively higher levels. This can be seen in the study by Hammouda et al. (2017) when comparing the frying performance of palm oil (3-MCPDE of 1.3 ppm, GE of 4.0 ppm), olive pomace oil (3-MCPDE of 1.1 ppm, GE of 1.5 ppm) and their blends (75:25, 50:50 and 25:75). Since the levels of 3-MCPDE for both oils were somewhat comparable, all the blends contained 3-MCPDE within 1.2 ppm and 1.3 ppm. However, the GE content in all blends showed a broader range between 2.3 and 4.0 ppm. Enhancement of the blends with palm oil appeared to yield insignificant lower 3-MCPDE content (0.5 ppm to 0.7 ppm) compared to blended oil containing 75% of pomace olive oil. The decomposition of GE was more apparent than 3-MCPDE as proven by lower final GE content in all (0.2 ppm to 0.5 ppm) after 16 h of frying.

Aniołowska and Kita (2015b) specifically quantified the amount of GE based on the individual fatty acids consisting of palmitic acid (C16:0), stearic acid (C18:0), oleic acid (C18:1) and linoleic acid (C18:2). Four oils comprising rapeseed oil, palm oil, palm olein and mixture of palm olein with high oleic sunflower oil were evaluated for 5 days of intermittent frying. Fresh rapeseed oil only showed trace amounts of GE in C18:1 and C18:2 with the sum concentration of 0.8 ppm. The total GE in fresh palm oil were detected at 5.8 ppm while fresh palm olein and fresh oil mixture gave remarkable overall GE content of 25.3 ppm and 20.5 ppm, respectively. It is noted from the total GE that C16:0 reflected by 34% to 39% while 46% to 49% represented by C18:1. Considerable amount of GE attached to C18:1 and C16:0 in palm oil products is closely related to nature of their fatty acid compositions (Weißhaar and Perz 2010). In fact, MacMahon, Begley, and Diachenko (2013) obtained the GE of 0.1 ppm to 13.3 ppm for C16:0 and 0.8 ppm to 22.5 ppm for C18:1 when quantifying GE contents in 14 palm oil samples.

Prominent GE reduction by more than three-quarter was observed in the case of intermittent frying in palm oil and palm olein while two-third GE was shaved off for the oil mixture while rapeseed oil demonstrated a decrease of just below 50% (0.4 ppm) (Aniołowska and Kita 2015b). The authors also examined the GE content in oil taken up by the fried product. Finish frying of pre-fried French fries in rapeseed oil encountered two-fold higher GE content in extracted lipid compared to other frying medium. The amount of GE in French fries fried in the remaining oils were 50 to 83% lower than those detected in the frying media (Aniołowska and Kita 2015b). Higher GE content in the extracted lipid containing rapeseed oil could be associated with the quality deterioration of absorbed oil.

It is worth mentioning that the quantification of GE reported in all the studies done by Aniołowska and Kita

Table 1. Publications on the fates of 3-MCPDE and/or GE in oils during frying.

Oils	Products	Frying mode and conditions	Research findings	References
PO	Potato chips, French fries and potato pallets	Intermittent frying, T = 180 °C, t = 4 min for potato chips and French fries, t = 30 s for potato pallets, 16 frying cycles daily for 5 days, daily oil replenishment	• 95% GE reduction for frying potato pallets (from 35.9 ppm to 2.0 ppm) • 93% GE reduction for frying French fries (from 35.9 ppm to 2.5 ppm) • 87% GE reduction for frying potato chips (from 35.9 ppm to 4.7 ppm)	Aniołowska and Kita (2015a)
RSO, PO, POo HOSFO + POo	French fries	Intermittent frying, T = 180 °C, t = 4 min, 16 frying cycles daily for 5 days, daily oil replenishment	Oils with greater GE content showed higher reduction rate after frying for 40 h • 78% reduction in PO (from 5.8 ppm to 1.3 ppm) • 76% reduction in POo (from 25.3 ppm to 6.0 ppm) • 67% reduction in HOSFO + POo (from 20.5 ppm to 6.8 ppm) • 48% reduction in RSO (from 0.8 ppm to 0.4 ppm)	Aniołowska and Kita (2015b)
HOSFO	Potato chips	Continuous frying, T = 160 °C to 188 °C, t = 100 s to 200 s, 24 h of nonstop frying, continuous oil replenishment	Consistent 3-MCPDE and GE contents were obtained after 24 h of frying • 3-MCPDE varied from 0.3 ppm to 0.5 ppm in oil and fried product • GE varied from 0.2 ppm to 0.4 ppm in oil and fried product	Dingel and Matissek (2015)
POo	Potato chips	Intermittent frying, T = 150°C, 165 °C and 180 °C, frying time varies until product moisture content <2%, 16 frying cycles daily for 5 days, daily oil replenishment	Increase in frying temperature accelerated the reduction rate of GE after 40 h of intermittent frying • 87% reduction at 180°C (from 39.0 ppm to 5.0 ppm) • 66% reduction at 165°C (from 39.0 ppm to 13.2 ppm) • 62% reduction at 150°C (from 39.0 ppm to 14.7 ppm)	Aniołowska and Kita (2016)
Blends of PO + OPO	French fries	Intermittent frying, T = 175 °C, t = 4 min, 16 frying cycles for 16 h, without oil replenishment	Depreciation rate of 3-MCPDE was lower than GE after 16 h of frying Increase in PO ratios in the oil blends have almost comparable 3-MCPDE contents before (1.2 to 1.3 ppm) and after frying (0.5 to 0.9 ppm) but not for the case of GE (1.5 to 4.0 ppm and 0.2 to 0.5 ppm, respectively)	Hammouda et al. (2017)
POo	Potato chips	Intermittent frying, T = 160 °C and 180 °C, t = 2.5 min, 5 frying cycles daily for 5 days, without oil replenishment	Degradation rate of 3-MCPDE decreased after 5 days of frying when: • Higher frying temperature (180°C compared to 160°C) • Higher NaCl (5% compared to 3% and 1%) Formation rate of GE rose after 5 days of frying when: • Lower frying temperature • Higher NaCl (5% compared to 3% and 1%)	Wong et al. (2017a)

Oils	Products	Frying mode and conditions	Research findings	References
POo	Chicken meat	Intermittent frying, T = 160 °C and 180 °C, t = 2.5 min, 5 frying cycles daily for 5 days, without oil replenishment	Decomposition rate of 3-MCPDE decreased after 5 days of frying: ● Higher frying temperature (180 °C compared to 160 °C) ● No additional of NaCl Formation rate of 3-MCPDE increased after 5 days of frying when: ● Higher frying temperature (180 °C compared to 160 °C) ● Higher NaCl (5% compared to 3% and 1%) Decomposition rate of GE increased after 5 days of frying when: ● Higher frying temperature (180 °C compared to 160 °C) ● Higher NaCl (5% compared to 3% and 1%) Formation rate of GE rose after 5 days of frying when: ● Lower frying temperature (160 °C compared to 180 °C) ● No additional of NaCl	Wong et al. (2017b)
PO	Rice ball, beef patty, potato, garlic, onion, banana, polenta and cassava	One-off frying, T = 160 °C to 190 °C and t = 2 to 9 min depending to product types	Frying in PO containing 1.6 ppm of 3-MCPDE yielded fried products with 3-MCPDE within 0.1 ppm and 0.3 ppm Chloride presence in oil and food did not significantly affect 3-MCPDE in oil during frying Good correlation between 3-MCPDE and moisture content and oil content, respectively	Arisseto et al. (2017)
RSO	Potato chips	Intermittent atmospheric (conventional frying, T = 165 °C, t = 3 min, 36 frying cycles for 17 h Intermittent vacuum frying, T = 125 °C, P = 10 kPa, t = 6 min, 48 frying cycles for 17 h	Vacuum frying (165 °C, 10 kPa) slowed down the degradation of 3-MCPDE compared to conventional frying (165 °C)	Belkova et al. (2018)
SFO	Potato chips	One-off frying, T = 180 °C, t = 3 min Intermittent frying, T = 180 °C, t = 3 min, 8 frying cycles daily for 3 days	3-MCPDE and GE in oil were not significantly affected by NaCl (1 to 5%) and rosemary extract (500 ppm to 2000 ppm) but significant 3-MCPDE in fried product during one-off frying Degradation rate of 3-MCPDE and GE were significantly higher upon addition of rosemary extract (2000 ppm) in oil and fried products during intermittent frying	Yıldırım and Yorulmaz (2018)

Table 1. (Continued).

Oils	Products	Frying mode and conditions	Research findings	References
SFO and RSO	Pre-fried fish product	*Effect of NaCl, frying time and temperature:* A daily frying (8 h) in SFO, T = 185 °C and 200 °C, t = 25 s and 40 s, frying interval = 1 h, 4 h and 7 h *Effect of MAG and DAG:* A daily frying (8 h) in RSO, T = 195 °C, t = 34 s, frying interval = 1 h, 4 h and 7 h *Effect of adsorbent, filtration and L-cysteine:* Intermittent frying in RSO, T = 195 °C, t = 34 s, 6 frying cycles daily for 10 days	Formation rate of 3-MCPDE increased after 7 h of par-frying in SFO when: • Higher NaCl (1.5%) at 185 °C • Lower NaCl (0.16%) at 200 °C • Effect of temperature is more prominent than NaCl No significant changes in GE during par-frying in SFO for 7 h regardless to frying interval, NaCl and frying temperature Formation rate of 3-MCPDE increased after 7 h of par-frying in RSO due to addition of MAG + DAG between 3 and 5% Formation rate of GE decreased after 7 h of par-frying in RSO due to addition of MAG + DAG between 3 and 5% Application of calcinated zeolite (adsorbent) increased the depreciation rate of 3-MCPDE and GE Oil filtration only decelerated 3-MCPDE formation but insignificant for GE L-cysteine did not show consistency to reduce 3-MCPDE and GE	Merke et al. (2018a)
RSO	Pickled herring product	Intermittent frying, T = 170°C, t = 3 min, 8 daily for 9 days, daily production = 2600 kg fried product	Effect of frying time: • Significant 3-MCPDE increase in fried product (from <0.1 ppm to 0.2 ppm) and oil (from 0.5 ppm to 4.1 ppm) after 9 days of frying • Relatively low GE in fried product (<0.1 ppm) and oil (0.2 ppm) after 9 days of frying Effect of breading (0.1% and 1% NaCl) before frying: • Different NaCl contents yielded negligible effect on 3-MCPDE and GE in both fried product and oil Effect of marinade (3.2% NaCl) after frying and packaging: • Marinade lowered 3-MCPDE and GE contents in fried product Effect of pasteurization time (30 min, 40 min and 50 min) after marinade: • Pasteurization at any time yielded negligible effect on 3-MCPDE and GE in fried product	Merke et al. (2018b)

PO, palm oil; POo, palm olein; RSO, rapeseed oil; HOSFO, high oleic sunflower oil; SFO, sunflower oil; OPO, olive pomace oil; CO, corn oil; HVF, hydrogenated vegetable fat; NaCl, sodium chloride; KCl, potassium chloride; $CaCl_2$, calcium chloride; $FeCl_3$, ferric chloride; NH_4Cl, ammonium chloride; TBHQ, tert-butylhydroquinone; BHA, beta-hydroxy acid; BHT, butylated hydroxytoluene; MAG, monoacylglycerols; DAG, diacylglycerols; T, frying temperature; t, frying time.

POo	Potato sticks made of Agata, Asterix and Markies cultivars	One-off frying, T = 180 °C, t = 6 min	3-MCPDE in fried products correlated with amount of oil content reflected by different potato cultivars 3-MCPDE in fried products was 33% and 8% higher than control when pre-blanching at 85 °C for 5 min and 98 °C for 3 min, respectively Combination between pre-blanching at 98 °C for 3 min and pectin coating reduced 3-MCPDE by 10%	Arisseto et al. (2019)
PO	Potato chips	Intermittent frying, T = 180 °C, t = 2.5 min, 5 frying cycles daily for 3 days, without oil replenishment	Antioxidants increased the decomposition rate of 3-MCPDE and GE based on the following effectiveness order: TBHQ > rosemary extract > sage extract > BHA > BHT	Wong et al. (2019)
POo	French fries	Intermittent frying, T = 180 °C, t = 4 min, 16 frying cycles daily for 5 days, daily oil replenishment	Depreciation rate of 3-MCPDE across frying time was lower than GE Additional of anti-clouding agent retarded the disintegration of 3-MCPDE and GE Chloride presence in oil did not onset the formation of new 3-MCPDE during frying	Ahmad Tarmizi et al. (2019)
SFO	Leavened dough	Daily frying, T = 160 °C, 180 °C and 200 °C, t = 1 min, 3 min, 5 min, salt content = 0%, 1%, 2%, 50 frying cycles daily, frying interval = 4 min	Optimum frying conditions for the lowest 3-MCPDE in frying oil and dough, and free 3-MCPD in defatted dough: • T = 172.6 °C to 173.3 °C (<175 °C), t = 2.4 min to 2.6 min, without NaCl • T = 172.7 °C to 173.3 °C (<175 °C), t = 1 min, 2% NaCl	Turan, Solak, and Keskin (2019)
HOSFO	Without product	Simulated frying, T = 175 °C, heating interval = 1 h, 2 h, 4 h, 6 h and 8 h, oil fortified with different chloride sources: (a) 3% NaCl, (b) 3% KCl, (c) 3% $CaCl_2$, (d) 0.1% $FeCl_3$, (e) 0.1% NH_4Cl	Absence of chloride source did not onset the formation of new 3-MCPDE during heating (averaging at 0.3 ppm) Addition of chlorinated salts increased the formation rate of 3-MCPDE based on the following effectiveness order: $FeCl_3$ > NH_4Cl > $CaCl_2$ > KCl > NaCl Different chloride sources affected the rate of 3-MCPDE formation after 8 h of heating: • NaCl increased from <1 ppm to 1.6 ppm • KCl increased from <1 ppm to 2.4 ppm • $CaCl_2$ increased from <1 ppm to 49 ppm • NH_4Cl increased from <1 ppm to 79 ppm • $FeCl_3$ increased from 150 ppm to 242 ppm	Bognár et al. (2020)

PO, palm oil, POo, palm olein, RSO, rapeseed oil, HOSFO, high oleic sunflower oil, SFO, sunflower oil, OPO, olive pomace oil, CO, corn oil, HVF, hydrogenated vegetable fat, NaCl, sodium chloride, potassium chloride, KCl, calcium chloride, $CaCl_2$, ferric chloride, $FeCl_3$, ammonium chloride, NH_4Cl, TBHQ, tert-butylhydroquinone, BHA, beta-hydroxy acid, BHT, butylated hydroxytoluene, MAG, monoacylglycerols, DAG, diacylglycerols, T, frying temperature; t, frying time

(2015a, 2015b, 2016) were notably higher than other literatures that reported GE during heating and frying. The variations in results can be explained from the basis of different sample preparation procedure and instrument used to detect GE in oil matrices (Becalski et al. 2012). Unlike majority of frying research articles that quantified GE and 3-MCPDE simultaneously using a gas chromatography-mass spectrometry (GC-MS), Aniołowska and Kita (2015a, 2015b, 2016) measured GE using a liquid chromatography fitted with tandem mass spectrometry (LC-MS-MS). The LC-MS-MS offers the advantages of higher sensitivity to detect and quantify a broader range of GE in oils based on individual fatty acids, i.e. C16:0, C18:0, C18:1 and C18:2, and only requires minimal sample preparation without the need of derivatization step. In addition, LC-MS-MS exclude sample volatilization stage which further minimizes chemical breakdown and tendency to develop new compounds at high temperature (Perez et al. 2016).

Frying methods

In this review, most of the discussion on 3-MCPDE and GE transients confined to conventional or open frying under discrete batch and atmospheric pressure. Only a single publication that discussed on how 3-MCPDE and/or GE manifested when frying process was undertaken using industrial continuous flow multizone fryer, and also one published research that applied vacuum fryer. Continuous frying of potato chips for 24 h displayed a consistent 3-MCPDE content in the oil (between 0.3 ppm and 0.5 ppm) and fried product (between 0.4 ppm and 0.5 ppm) across sampling intervals between 2 and 4 h (Dingel and Matissek 2015). Similar trend was also observed for GE where both oil and product gave the values hovering at 0.2 ppm to 0.4 ppm and 0.2 ppm to 0.3 ppm, respectively. Irrespective to frying interval, the result differences observed during continuous frying were insignificant between the amount of these artifacts detected in the oil and fried product. This can be explained by the nature of continuous frying operation where the oil inside the fryer, which is taken up by the fried product and evaporates during frying, is continuously replenished with fresh or new good quality oil resulting in consistency in the formation of unwanted constituents (Ahmad Tarmizi, Kuntom, and Ismai). This is supported by several published studies (Ismail 2005; Ahmad Tarmizi and Ismail 2008; Ahmad Tarmizi and Ismail 2014; Ahmad Tarmizi and Ahmad 2015) that observed equilibrium phases for oil acidity, p-anisidine value, polar and polymer compounds, induction period (or oxidative stability index), and color when frying was undertaken using industrial- and pilot-scale continuous frying systems.

In recent years, vacuum frying has received increasing demand due to its advantages over conventional frying in the respect of preserving nutritional value, natural color and sensory attributes, and minimizing oil deterioration and acrylamide formation (Dueik, Robert, and Bouchon 2010). Based on these privileges, Belkova et al. (2018) have taken steps to study the effect of vacuum frying on the behavior of 3-MCPDE and further compared with conventional frying at 165 °C. In this research work, vacuum frying was carried out at lower temperature (125 °C) and reduced pressure (10 kPa, water saturation temperature of 46 °C). The deceleration of 3-MCPDE was faster in conventional frying because of higher frying temperature applied throughout 17 h of frying. The study concluded that the impact of frying temperature was stronger even though vacuum frying at lower temperature with minimal exposure to air (oxygen) has successfully reduced the incidence of TAG oxidation and polymerization, and the release of volatile organic compounds. Several publications opined that oxidized and polymerized oil is inversely linked with the rate of 3-MCPDE and/or GE deformation (Ahmad Tarmizi et al. 2019; Merkle et al. 2018a; Aniołowska and Kita 2015a, 2015b, 2016).

Frying temperature

Selection of the right frying temperature should take into consideration of food properties, oil turnover time and food-to-oil ratio. By owing optimum frying temperature, it helps to keep the oil deterioration at a lowest rate, ensuring the product oil uptake remains minimal, and thus reduces the operation cost and oil utilization (Mehta and Swinburn 2001). Furthermore, exposing the oil at frying temperature inevitably led to intricate chemical reactions in the oil as well as starch gelatinization and protein denaturation which contribute to distinctive sensory attributes to the fried products.

Three research works have specifically determined the impact of frying temperature on the transient of 3-MCPDE and GE in oil. Aniołowska and Kita (2016) have intermittently fried potato chips at three different temperatures, i.e. 180 °C, 165 °C and 150 °C, and found that highest frying temperature remarkedly fastened GE degradation by 87%. Frying at reduced temperatures (165 °C and 150 °C) did not show obvious GE reduction despite the temperature gradient of 15 °C was significant (66% and 62% of GE reduction, respectively). From this finding, the researchers opined that the effect of frying time was more relevant as compared to frying temperature. The reduction in GE is caused by destabilization of GE during frying where high reactivity of epoxide ring has led to transformation into other constituents such as glycidol propanediol or polymerized monoacyl esters (Shimizu et al. 2013).

Wong et al. (2017a) compared the fates of 3-MCPDE and GE when performing intermittent frying of carbohydrate-based product (potato chips) for 5 consecutive days at 160 °C and 180 °C. Frying at higher temperature accelerated the disintegration of 3-MCPDE which was found similar with their separate study using chicken meat breast (Wong et al. 2017b). Such coincidence is supported by their TAG results where frying at 180 °C gave lower values over frying time which further enhances 3-MCPDE formation in the presence of notable amount of acyloxonium ions. It is also plausible that high frying temperature promotes the conversion of 2-MCPDE into 3-MCPDE through glycidol intermediate (Rahn and Yaylayan 2011; Hamlet et al. 2011). On the other hand, GE development is more apparent when frying of potato chips and chicken meat breast were performed at lower temperature. This can be evidenced from low level of MAG and DAG detected in oil heated at 160 °C which confirms the conversion of both acylglycerols into GE (Shimizu et al. 2013).

Antioxidants and additives

Antioxidants are often added into the oils to primarily induce their shelf-life stability during storage frying. Moreover, the role of antioxidants in preserving oil quality is also deliberated in many scientific papers. Fortification of synthetic antioxidants like tert-butylhydroquinone (TBHQ), butylated hydroxytoluene (BHT) and butylated hydroxyanisole (BHA) have shown their inhibitory effect to supress oil deterioration; nevertheless, the usage of synthetic antioxidants has raised some concerns on the chemical substances being added into food that can adversely affect human health (Sharayei et al. 2011). Natural antioxidants such as sage, oregano, thyme and rosemary extracts has gain considerable interest albeit the premium that need to be considered in comparison to chemically-synthesised counterpart (Berger 2005).

A frying research on the effect of antioxidant fortification in vegetable oil toward the behaviors of 3-MCPDE and GE was first published in 2018 (Yıldırım and Yorulmaz 2018). Addition of rosemary extract demonstrated a plateauing 3-MCPDE trend at a range between 0.4 ppm and 0.8 ppm which did not significantly change with the controlled oil. However, the carry-over of 3-MCPDE in fried product was more apparent when the lipid extract contained somewhat lower 3-MCPDE (0.2 ppm to 0.6 ppm) for potato chips fried in oil containing rosemary extract when compared to unfortified oil (0.6 ppm to 0.9 ppm). This observation suggests the inhibitory role of antioxidant used during excessive heating. In general, the amount of 3-MCPDE presence in the oil used for frying is generally reflected by the level of 3-MCPDE in the oil absorbed by the fried product. Inclusion of antioxidant in the oil does not only capable of reducing the degree of oil degradation but also suppresses the precursor of 3-MCPDE i.e. chloride radicals, and minimize the formation of cyclic acyloxonium free radicals (CAFR) intermediates (Zhang et al. 2016).

The absence of rosemary extract caused the endogenous GE to increase in the oil, which somewhat identical with the results obtained by Wong et al. (2017b) when they fried protein-based product albeit most of literatures showed the otherwise GE trend during frying. Rosemary extract, on the other hand, significantly increase the degradation rate of GE in oil from 0.4 ppm to 0.1 ppm, but the amount of 3-MCPDE retained in potato chips was averaging at 0.2 ppm. The GE content was found lower because of reduced temperature during frying (180 °C) while GE would start to increase rapidly when the oil temperature surpasses 230 °C as demonstrated during the edible oil refining process (Matthäus and Pudel 2015; Zelinkova et al. 2006).

In 2019, there are two published papers investigated whether the incorporation of anti-clouding agent, which is polyglycerol fatty acid ester (PGE), (Ahmad Tarmizi et al. 2019) and different antioxidants (i.e. tert-butylhydroquinone (TBHQ), beta-hydroxy acid (BHA), butylated hydroxytoluene (BHT), oleoresin rosemary and sage extract) (Wong et al. 2019) would impact to the fates of 3-MCPDE and GE during extensive frying of potato products in palm olein.

Progressive 3-MCPDE reduction was only visible when there was no or only trace of PGE (0.1%) presence in the oil. Slower 3-MCPDE retardation in the case of oil added with PGE as high as 0.4% coincided with the increase in the percentage of polar compound fractions, particularly oxidized triacylglycerols (OxTAG) and polymerized triacylglycerols (PTAG). Based on this observation, higher rate of TAG degradation over frying time seemed to promote the newly developed 3-MCPDE, and thus moderate the overall 3-MCPDE disintegration in the oil added with PGE. The absence of PGE resulted in rapid loss in GE after 5 days of frying; nevertheless, PGE hold GE reduction and hence retained their initial GE between 39% and 56% even after the completion of frying session. Slower rate of 3-MCPDE and GE degradation in the oil can be explained from the perspective of PGE polarity in term of its wide range of hydrophilic-lipophilic balance (HLB) values between 6 and 11 (Tadros 2013). Discrepancy in PGE polarity distinguishes the degree of glycerol polymerization and types of esterified fatty acids attached to TAG backbones. The PGE with higher HLB value has the likelihood of hydrophilicity and thus enhances the polarity of oil particularly during frying (Norn 2015). The authors concluded that the addition of PGE is deemed unnecessary, and even if the PGE addition is required, only adequate amount should be established to retard oil cloudiness at realistic storage time.

The synergistic effect of antioxidants used in the study conducted by Wong et al. (2019) significantly influenced the rate of 3-MCPDE and GE disintegration throughout frying period. The ability of antioxidants to disregard the formation of radical intermediate is utmost critical. The nature of TBHQ (which contains dihydroxylphenols) exhibited advantages over BHA and BHT (which contain monohydroxylphenols) in providing a protective mean to the oil by slowing down lipid peroxidation and thus obstruct the initiation of 3-MCPDE formation during frying. In general, TBHQ has almost comparable performance to oleoresin rosemary while BHA and BHT have more or less similar antioxidant capability with sage extract. Similar to 3-MCPDE, GE also showed a downward trend but at slower rate following the degree of synergistic capability of each antioxidant. The authors recorded the lowest GE retention in TBHQ (67%) followed by oleoresin rosemary (70%), sage extract (73%), BHA (74%) and BHT (77%). The inclusion of antioxidants, at some degrees, could minimize TAG fragmentation into DAG and suppressed GE formation throughout the frying course.

Salt impregnation

Salting or curing refers to applying salt to food for the purpose of seasoning and preservation. One of the routes of salt impregnation is osmotic dehydration which principally involves product immersion is saline or brine solution at predetermined time (Ziaiifar et al. 2008). In this process, some of the moisture retained in the product diffuses into the brine solution caused by osmotic pressure, and at the same time, solute from the solution migrates into the product (Andrés-Bello, García-Segovia, and Martínez-Monzo 2011). In the light of frying, osmotic dehydration enabled to reduce the amount of oil content in fried product by lowering its initial moisture content before frying and shorter frying time.

Two publications by Wong et al. (2017a, 2017b) have systematically conferred the effect of immersing potato slices and chicken breast meat, respectively, in brine solution containing 1% (10,000 ppm) to 5% (50,000 ppm) of sodium chloride (NaCl). The transient of 3-MCPDE during frying of potato slices in the presence of NaCl slowed down the rate of 3-MCPDE degradation (Wong et al. 2017a). It is believed that excessive amount of NaCl could adequately source out chloride ions to react with TAG via intermediate acyloxonium ions to generate new 3-MCPDE. However, the amount of 3-MCPDE formation during frying is lower than the rate of its fragmentation, and this gave net declining trend in 3-MCPDE.

This observation appeared contradict with the frying experiments using chicken meat breast (Wong et al. 2017b). Unlike the control experiment, addition of NaCl displayed an upward trend of 3-MCPDE across frying time. An increase of 13% to 18% was observed when potato slices were immersed in 1% to 5% of brine solution prior to intermittently fried at 160 °C. More prominent rate of 3-MCPDE formation was found (17% to 42%) when similarly treated potato slices were fried at higher temperature (180 °C). Combination effect between NaCl and temperature also agrees with the finding observed by Velíšek et al. (2003). The formation of 3-MCPDE in abundant has exceeded the rate of 3-MCPDE deformation which resulted in net increase in 3-MCPDE over frying time. Divergence in 3-MCPDE behavior during frying of carbohydrate- and protein-based products can be associated with product structure which determines salt capability to diffuse or adheres on the surface due to product compactness (Ahmad Tarmizi et al. 2019).

In the light of GE behavior, frying of salted potato slices yielded an inverse trend to that of 3-MCPDE (Wong et al. 2017a). Contrariwise, brined chicken breast meat exhibited a depreciation of GE during 5 day of consecutive frying. Bakhiya et al. (2011) opined that GE formation follows one of the 3-MCPDE formation pathways where acyloxonium ions derived from MAG react with chloride in the oil. This hypothesis appeared relevant to describe GE breakdown when the oil was used for frying chicken breast meat.

Arisseto et al. (2017) proved that frying of various products containing NaCl between 4.9 ppm up to 119.0 ppm in corn oil with undetectable 3-MCPDE (less than 0.05 ppm) did not onset the formation of 3-MCPDE after frying. This observation disapproved the outcome from the work on frying model systems which did not consider the effect of oil interaction with actual food products (Svejkovská, Velíšek, and Doležal 2006). Since Arisseto et al. (2017) conducted the study as one-off frying, shorter heating time is likely not sufficient to promote the formation of 3-MCPDE in the oil. Similar effect of single frying also evidenced by Yıldırım and Yorulmaz (2018) who noted unseen variations in both 3-MCPDE and GE after frying. Pre-frying of salted fish fingers for 7 h influenced the rate of 3-MCPDE formation in the oil at lower frying temperature (185 °C) but insignificant when pre-frying was performed at higher temperature (200 °C) (Merkle et al. 2018a). Nevertheless, the GE content in the oil was found similar irrespective to salt concentrations.

The impact of chlorinated salts on the behavior of 3-MCPDE under simulated frying conditions has been confirmed by Bognár et al. (2020). The study showed that the formation of 3-MCPDE were relatively lower in both NaCl and potassium chloride (KCl) (less than 3 ppm) while $CaCl_2$ portrayed higher 3-MCPDE content (49 ppm) in the heated oil containing 3% of aforementioned salts. Surprising, much lower dosage (0.1%) of ferric chloride ($FeCl_3$) and ammonium chloride (NH_4Cl) in the oil gave considerable 3-MCPDE contents of 79 and 242 ppm, respectively. In the presence of excessive heat, Fe^{3+} and NH_4^+ ions promote higher radical generation that enables chloride to immediately react with acyloxonium acyloxonium intermediate to develop 3-MCPDE (Li et al. 2016; Zhang et al. 2015). Thus, it can further suggest that the catalytic effect of cation presence in the chlorinated salts determines the rate of 3-MCPDE formation upon interaction with oil exposed to excessive temperature.

It is important to note that the presence of chloride in oil used for frying is does necessarily cause the formation of 3-MCPDE albeit chloride is known as the precursor for 3-MCPDE. Ahmad Tarmizi et al. (2019) observed that consistent level of chloride in the oil over frying time did not onset endogenous formation of new 3-MCPDE. This finding is in agreement with Arisseto et al. (2017) where the presence of natural chloride in potato did not initiate further formation of new 3-MCPDE. Merkle et al. (2018b) did not encounter any development of 3-MCPDE in both fried herring fillets and oil over frying time despite the breading ingredient contained notable NaCl contents of 0.1% and 1%. The researchers also observed that marination of fried and packed product in brine solution containing 3.2% NaCl yielded lower 3-MCPDE as compared to unmarinated product. It is plausible that marination leads to osmotic effect which further dilutes the 3-MCPDE concentration due to water migration from marinade to the product.

Product morphological properties and cultivars

Product morphological properties and cultivars essentially determine the extent of frying oil deterioration and the amount of oil migrated into fried product. The first study by Arisseto et al. (2019) examined the prevalence of 3-MCPDE in eight food products from five different sources of raw materials, i.e. meat (beef patty), cereals (rice ball, polenta), fruit (banana), bulbs (garlic, onion) and tubers (potato, cassava) when frying was undertaken in palm oil containing 1.6 ppm of 3-MCPDE. The amount of 3-MCPDE presence in all fried products varied between 0.1 ppm to 0.3 ppm which exhibits the indication of 3-MCPDE carry-over from the frying medium. This argument was proven by undetectable 3-MCPDE in all products before frying. Fried garlic yielded the highest 3-MCPDE among others (0.3 ppm) which can be explained from the basis of nearly 80% of moisture loss and greater oil uptake (17.7%).

Arisseto, Marcolino, and Vicente (2015) experienced similar observation when commercially fried chopped garlic contained 1 ppm of 3-MCPDE, which was the highest compared to other commercial fried products. Most of fried products that encountered slower rate of moisture loss and reduction in oil content had relatively lower 3-MCPDE content (0.1 ppm). Associations between moisture loss and oil content with regard to 3-MCPDE were considerably strong with the correlation of coefficient (r) exceeding 90%. The researchers also found that most of the theoretical 3-MCPDE content in the fried products – which are calculated based on the percentage of oil uptake and 3-MCPDE concentration in the frying media – resulted in relatively small variation with the actual 3-MCPDE content.

The latest research by Arisseto et al. (2019) depicted that different potato varieties namely as Agata, Asterix and Markies determines the amount of 3-MCPDE presence in fried product. The lowest 3-MCPDE content was shown by Agata cultivar by taking into consideration of its reduced moisture content and notable dry solid matters (before subjected to frying) when compared to other varieties. This observation was also evidenced by Ilko et al. (2011) who noted a distinctive 3-MCPDE content in two potato varieties as well as similar potato varieties harvested from different provinces. The variation in 3-MCPDE content was also correlated with the amount of oil taken up by the fried products. Agata cultivar yielded the least amount of oil content (6.4%) as opposed to Asterix (8.3%) and Markies (8.94%) cultivars. The results suggested that higher moisture retention in the potatoes would lead to higher rate of oil absorption because of the emptied voids as a result of water evaporation are subsequently filled with oil (Ziaiifar et al. 2008).

Aniołowska and Kita (2015a) distinguished three types of potato products with different moisture contents and degree of dehydration on the formation of GE across 5 days of intermittent frying. Frying in palm oil containing 3-MCPDE of 35.93 ppm led to GE decomposition predominantly in potato pallets (95% reduction) and French fries (93% reduction) whereas potato chips only displayed 87% reduction. As frying is essentially a surface phenomenon, product with higher surface roughness and surface-to-volume ratio has the likelihood of absorbing more oil (Ziaiifar et al. 2008). Additionally, the porosity and microstructure of the product determines the amount of oil taken up after frying.

Pre-frying treatments

Many scholars reported the feasibility of pre-frying treatments in reducing the rate of oil absorption in fried food. The pretreatments include coating, blanching, drying and osmotic dehydration. Indeed, many studies have even combined multiple pretreatments to the raw products before frying. In this review, pre-frying treatments focuses on food coating and blanching to mitigate occurrence of 3-MCPDE and GE in both oil and fried food. Coating is considered as a surface treatment which is not limited to add value to the product, but also enhances the water binding capacity, reduces surface porosity and creates good barrier against oil absorption during frying (Ziaiifar et al. 2008; Miranda and Aguilera 2006). The product is simply coated by applying either a thin and 'invisible' layer or thick batter and breading formulations comprising flour and water mixture (Mellema 2003). Blanching, on the other hand, involves plunging the product into hot water to deactivate enzymes and microorganisms (Andrés-Bello, García-Segovia, and Martínez-Monzo 2011; Mehta and Swinburn 2001). This technique is mainly responsible to prevent enzymatic browning (Shyu and Hwang 2001), minimizes non-enzymatic browning during frying (Fan, Zhang, and Mujumdar 2005) and improves the color and texture of fried product (Shyu, Hau, and Hwang 2005).

The first study on coating application to retard on 3-MCPDE and GE formation was done by Merkle et al. (2018a). The impact of emulsifier E417 incorporation in the wet breading ingredient for fish fingers was investigated on the behaviors of 3-MCPDE and GE prior to pre-frying process. Pre-frying the breaded fish fingers recorded a rapid 3-MCPDE built-up as opposed to GE which shown a moderate increment. Additional of 1% and 3% emulsifier not only produced pre-fried products with higher oil uptake (7.9% and 8.5%, respectively in comparison to the control sample, 7.7%), but also induced the amount of 3-MCPDE and GE in the oil throughout the pre-frying session. The presence of these artifacts in the oil was also reflected in par-fried products.

The 3-MCPDE and GE acceleration can be associated with the emulsifier E471 content which is essentially a mixture of MAG and DAG (Merkle et al. 2018a). This observation supports Destaillats et al. (2012) opinion where the formation of 3-MCPDE and GE takes place in different pathways consisting of partial acylglycerols (MAG and DAG) and chloride source: (1) 3-MCPDE formation occurs at temperatures greater than 180 °C via nucleophilic substitution of which TAG acts as the precursor, and (2) GE formation prevails at temperatures above 200 °C in the presence of MAG and DAG through elimination of protonated hydroxyl group, intra-molecular rearrangement of acyloxonium intermediate and removal of fatty acid. Simulation studies done by Hamlet et al. (2011), Freudenstein, Weking, and Matthäus (2013), Destaillats et al. (2012), and Matthäus et al. (2011) also confirmed the relationship between the formation of 3-MCPDE and GE with the presence of MAG and DAG.

Pre-frying of dry-breaded fish fingers with L-cysteine, which is a semi-essential proteinogenic amino acid (Sameem, Khan, and Niaz 2019), was also investigated by Merkle et al. (2018a) at different dosages (0.02%, 0.04% and 0.08%); nevertheless, the data generated seemed not convincing enough to reduce 3-MCPDE and GE during frying. All dosages gave significantly higher 3-MCPDE content in oil than the control experiment beyond 30 h albeit L-cysteine at 0.08% significantly reduced 3-

MCPDE for the first 7 h of heating. This confirms that the functionality of L-cysteine to mitigate 3-MCPDE is only workable for the model system containing NaCl and glycerol (Velíšek et al. 2011). They also hypothesized that the interaction between L-cysteine and 3-MCPDE generates S-(2,3-dihydroxypropyl)-cysteine which could further limit the formation of 3-MCPDE in the model system. The nature of L-cysteine as a hydrophilic amino acid explains its polarity behavior (Jan et al. 2015) particularly in the oil, which is associated with the formation of 3-MCPDE and GE during frying (Ahmad Tarmizi et al. 2019).

A recently published research articles examined whether blanching of potato sticks prior to frying is effective in slowing down the development of 3-MCPDE and GE in oil (Arisseto et al. 2019). Pre-blanching potato sticks at 98 °C for 3 min and subsequent coating with pectin exhibited 3-MCPDE reduction of 10% in fried product. The study also concluded that the influence of blanching alone, i.e. pre-blanching at 85 °C for 5 min and pre-blanching at 98 °C for 3 min, seemed to promote higher 3-MCPDE by 33% and 8%, respectively compared to untreated potato sticks. The transient of 3-MCPDE content for potato sticks that underwent for different pre-frying treatments was coincided with the amount of oil content in the fried products. Combination of blanching and pectin coating lowered the product oil content by 18% as compared to the control sample. Pectin is responsible to stabilize the potato tissue structure by constructing calcium-pectate linkages that helps strengthening the cell wall to resist high frying temperature and thus obstructs oil migration into fried food (Khalil 1999). The functionality of ingredients used in the coating material has proven to obstruct new formation of 3-MCPDE and GE during frying.

Adsorbent and filtration

Utilization of adsorbent and oil filtration at the end of daily frying operation is often practiced by established fast food outlets and industrial snack food manufacturers to optimize the usability of oil for frying. For that reason, Merkle et al. (2018a) has initiated a research work on the imposition of those treatments to mitigate the presence of 3-MCPDE and GE during pre-frying process. In this study, calcinated zeolite powder (aluminium silicate) was used as adsorbent while oil filtration was accomplished using a filter paper with the pore size of 20 μm. Inclusion of higher dosage of adsorbent (9%) remarkedly lowered 3-MCPDE and GE content in the oil. Strong dependency on adsorbent dosages is also portrayed by Strijowski, Heinz, and Franke (2011) who also applied as high as 10% adsorbent to achieve maximum 3-MCPDE and GE deformation during oil refining.

The effect of calcinated zeolite powder (9%) during intermittent pre-frying of fish fingers significantly fastened the decomposition of 3-MCPDE and GE by at least two-fold as compared to oil without adsorbent. The application of calcinated zeolite powder and synthetic magnesium silicate was also shown to be effective for GE removal of up to 40% (Strijowski, Heinz, and Franke 2011). The researchers concluded from their study that filtering-off solid debris from oil at the end of each day of frying only curbed the formation of 3-MCPDE while GE did not show much improvement with the unfiltered oil. Food particles literally cause unnecessary oil breakdown during frying which can curtail fry-life, insufficient heat transfer between oil and food and develop burnt-flavor in fried food (Berger 2005). Furthermore, the presence of trace elements such as iron, zinc and chloride in the food residues not only expedite oil deterioration, but also contribute to excessive development of 3-MCPDE in the oil during frying.

Relationship between 3-MCPDE and GE on frying quality indices

Compilation of literatures that deliberated on the transient of 3-MCPDE and/or GE in relation to physico-chemical changes in vegetable oils subjected to prolonged frying – i.e. intermittent and continuous frying under atmospheric pressure, and vacuum frying – is listed in Table 2. From Table 2, less than half of the research papers reported the relationship between 3-MCPDE and/or GE with selected quality parameters including acidity (expressed as FFA) and acid value (AV)), *p*-anisidine value (AnV), total polar compounds (TPC), PTAG, OxTAG, DAG and refractive index (RI). The strength of association between two variables during prolonged frying session is described by Pearson's correlation coefficients (r) (Akoglu 2018; Dancey and Reidy 2017). Table 3 summarizes the studies that underwent Pearson's correlation test for oil quality assessment.

In all studies, none of the parameters showed weak correlation with 3-MCPDE and GE ($r < 0.4$). Except for Aniołowska and Kita (2015 b), strong correlations were detected between quality parameters and 3-MCPDE and/or GE, respectively ($r \geq |0.7|$). This can be explained by the inclusion of all data generated from different frying media for correlation assessment, which in turn, resulted in moderate relationship with GE ($|0.4| \leq r < |0.7|$). It was also reported that the effect of different frying temperatures for frying potato chips in palm olein gave strong negative correlation ($r > -0.9$) for most parameters and almost perfect correlation ($r \approx |1|$) between TPC and GE (Aniołowska and Kita 2016).

It is previously discussed that DAG is the precursor for GE and thus yielded positive correlation with between these two variables as evidenced by Aniołowska and Kita 2015a; 2015b; 2016. Nevertheless, this was not the case of Ahmad Tarmizi et al. (2019) who obtained negative correlation between DAG and GE ($r = -0.70$) when evaluating the influence of anti-clouding agent in palm olein during 80 cycles of frying French fries. It is also noted from Table 3 that only one study by Ahmad Tarmizi et al. (2019) has performed Pearson correlation test between 3-MCPDE and quality parameters. In general, all parameters perceived strong negative correlation with 3-MCPDE ($r \geq -0.8$).

Conclusions and recommendations

This review focuses the manifestation of 3-MCPDE and GE in refined vegetable oils during frying. As palm oil is perceived to have higher levels of 3-MCPDE and GE, majority of the publication on frying emphasized on palm oil as

Table 2. Publications on 3-MCPDE and/or GE in relation to quality indices during prolonged frying.

Food	Oil type	3-MCPDE	GE	TCC	Acidity	AnV	K_{252}	K_{268}	FAC	IV	RI	TPC	PCF	ACYL	Color	VOC	WC	References
Intermittent frying																		
Potato chips, French fries, potato pallets	PO		X		X	X			X			X	X		X		X	Aniołowska and Kita (2015a)
French fries	RSO, PO, POo, HOSFO + POo		X		X	X			X		X	X	X		X			Aniołowska and Kita (2015b)
Potato chips	POo		X		X	X			X		X	X	X		X			Aniołowska and Kita (2016)
French fries	Blends of PO + OPO	X	X		X				X	X					X			Hammouda et al. (2017)
Potato chips	POo	X	X		X	X	X	X						X				Wong et al. (2017a)
Chicken meat	POo	X	X		X	X	X	X						X				Wong et al. (2017b)
Potato chips	SFO	X	X						X									Yıldırım and Yorulmaz (2018)
Pre-fried fish product	RSO	X	X									X			X			Merke et al. (2018a)
Potato chips	PO	X	X		X	X	X	X						X				Wong et al. (2019)
French fries	POo	X	X	X	X	X						X	X					Ahmad Tarmizi et al. (2019)
Continuous frying																		
Potato chips	HOSFO	X	X															Dingel and Matissek (2015)
Vacuum frying																		
Potato chips	RSO	X											X			X		Belkova et al. (2018)

PO, palm oil; POo, palm olein; RSO, rapeseed oil; HOSFO, high oleic sunflower oil; SFO, sunflower oil; NaCl, OPO, olive pomace oil; 3-MCPDE, 3-monochloropropanediol esters, glycidyl esters, GE, TCC, total chloride content, Acidity, expressed as free fatty acid (FFA) and acid value (AV), AnV, *p*-anisidinve value; K_{252}, conjugated dienes; K_{268}, conjugated trienes; FAC, fatty acids composition; IV iodine value; RI, reflective index; TPC, total polar compounds; PCF, polar compound fractions, i.e. polymerized triacylglycerols (PTAG), oxidized triacylglycerols (OxTAG) and diacylglycerols (DAG); ACYL, acylglycerols, i.e. triacylglycerols (TAG); diacylglycerols (DAG) and monoacylglycerols (MAG); VOC, volatile organic compounds; WC, water content.

frying media. In principle, there is no fixed trend of 3-MCPDE and GE when the oils are subjected to frying. Most of the research works prevail decomposition of these contaminants particularly under extended frying operation. It is well-established that excessive heat triggers the formation of unwanted compounds. Higher frying temperature generally accelerates the fragmentation of 3-MCPDE and GE, however, there are some cases show unpredictable observation when the oil is experiencing a combined effect of frying temperature, food composition and/or salt content. The use of MAG and DAG as coating ingredient also induces the formation of 3-MCPDE and GE in the oil. The presence of chloride in the oil used for frying and raw materials such as meat, cereals, fruit, bulbs and tubers did not onset the formation of 3-MCPDE and GE. The impact of frying methods could also distinguish the pattern of 3-MCPDE and GE contents in the oil. An equilibrium state was achieved for both compounds when the oil undergoes continuous frying, and this contradicts with intermittent frying which is performed under discrete batch. The pattern of 3-MCPDE is also unexpected when vacuum frying at lower temperature slows down the 3-MCPDE depreciation.

From the review, it is important to note that higher amount of 3-MCPDE and GE detected in the oil does not necessarily results in higher retention of these compounds at the end of frying operation. Utilization of adsorbent to mitigate 3-MCPDE and GE in used oil seems effective, however, filtering the oil at the end of frying operation only applicable in reducing 3-MCPDE. Antioxidants have shown the ability of lowering the levels of 3-MCPDE and GE in the oil depending on their degree of synergistic capability. Contrariwise, incorporation of additive such as anti-clouding agent decelerates the disintegration of 3-MCPDE and GE during frying. The quality of oil used for frying food is essential since considerable amount of oil migrated into the finished products. There are studies concluded that the rate of moisture loss and oil content coincide with the amount of 3-MCPDE and GE presence in fried food. Furthermore, the selection of crop varieties and applying pretreatments to the products prior to frying would determine the level of these contaminants in the finished products.

From the modest published frying studies related to 3-MCPDE and GE, one can conjecture that the impact of frying does not have a definite behavior on 3-MCPDE and GE occurrences contributed by various compounded factors and complexity of the frying process. Hence, more studies should be undertaken to fill up the gap to comprehend the mechanistic behaviors of 3-MCPDE and GE in particular to frying process. Many of the established hypotheses for 3-MCPDE and GE development are only applicable for oils subjected to refining temperatures without considering the presence of food under various frying conditions.

More exploration in this field should also be extended to the effects of frying procedures, i.e. food load, frying cycles and alternate frying, as well as unconventional frying techniques such as vacuum-, pressure- and air-frying on the fates of 3-MCPDE and GE. The findings are useful to further establish the predicted models on the transients of 3-

Table 3. Significant correlation coefficients between selected quality parameters and 3-MCPDE and/or GE.

Food	Oil type	3-MCPDE							GE							References
		Acidity	AnV	TPC	PTAG	OxTAG	DAG	RI	Acidity	AnV	TPC	PTAG	OxTAG	DAG	RI	
Potato chips, French fries, potato pallets	PO	–	–	–	–	–	–	–	−0.87	−0.94	−0.98	−0.94	−0.80	0.98	–	Aniołowska and Kita (2015a)
French fries	RSO, PO, POo, HOSFO + POo	–	–	–	–	–	–	–	−0.43	0.51	–	−0.45	−0.67	0.68	−0.49	Aniołowska and Kita (2015b)
Potato chips	POo	–	–	–	–	–	–	–	−0.97	−0.86	−0.99	−0.96	−0.95	0.94	−0.98	Aniołowska and Kita (2016)
French fries	POo	−0.80	−0.96	−0.87	−0.82	−0.89	−0.88	–	−0.86	−0.72	−0.80	−0.83	−0.80	−0.70	–	Ahmad Tarmizi et al. (2019)

PO, palm oil; POo, palm olein; RSO, rapeseed oil; SFO, sunflower oil; NaCl, 3-MCPDE; 3-monochloropropanediol esters, glycidyl esters; GE, Acidity, expressed as free fatty acid (FFA) and acid value (AV), AnV, *p*-anisidinve value, TPC, total polar compounds; PTAG, polymerized triacylglycerols; OxTAG, oxidized triacylglycerols; DAG, diacylglycerols; RI, refractive index

MCPDE and GE, and recommend the optimal frying procedures and methods for specific products. The availability of such information could facilitate the food industry on how to manage their operation in ensuring the finished product produced is safe and of good quality.

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