Effective physical refining for the mitigation of processing contaminants in palm oil at pilot scale

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ABSTRACT

This study aimed to develop a mitigation strategy for the formation of 2-monochloropropane-1,3-diol esters (2-MCPDE), 3-monochloropropane-1,2-diol esters (3-MCPDE), and glycidol fatty acid esters (GE) during palm oil refining. Single physical refining was the starting point (the control) for this study. Experimental treatments including a double refining repeating the entire single refining process (T1), double refining with a high-low deodorization temperature (T2), and double deodorization (T3) with similar temperature settings as T2 were performed. Compared with the control experiment, T2 successfully reduced the formation of GE by 87%; in particular, the second degumming and bleaching were crucial for eliminating GE. Both 2- and 3-MCPDE were formed prior to the deodorization process in all treatments. MCPDE concentrations remained stable throughout the refining process and, hence, they require a different mitigation approach as compared to GE. These results provide useful insights which can directly be implemented by the oil industry.

1. Introduction

2-monochloropropane-1,3-diol fatty acid esters (2-MCPDE), 3-monochloropropane-1,2-diol fatty acid esters (3-MCPDE), and glycidol fatty acid esters (GE) are process contaminants that can be found in refined vegetable oils and fats. Occurrence data showed that palm oil samples can have high concentration of these contaminants compared to other oil types and consequently 2-, 3-MCPDE, and GE were also found in food products containing palm oil as ingredient (Becalski, Zhao, Feng, Lau, & Zhao, 2015; Cheng, Liu, Wang, & Liu, 2017; Kuhlmann, 2016; MacMahon, Begley, & Diachenko, 2013). In the last decade, several mitigation strategies for the formation of 3-MCPDE and GE have been published, as recently reviewed by Oey et al. (2019). The source, the quality of the crude oil, and potential enzymatic hydrolysis of acylglycerols occurring between harvesting and processing play a role in the potential formation of 2-, 3-MCPDE, and GE during oil refinery (Matthäus & Pudel, 2013). These parameters need to be well regulated to achieve high quality vegetable oils with low 2-, 3-MCPDE and GE contents. However, full control over all the previously mentioned parameters is difficult to achieve. Optimizing the refining conditions would therefore be a more accessible strategy.

2-, 3-MCPDE and GE are mainly formed during processing of the crude oils in refineries where the oils are treated at elevated temperatures for an extended amount of time. On one hand, reaction mechanisms proposed by Šmidrkal et al. (2016), Destaillats, Craft, Sandoz, & Nagy (2012), and Hamlet et al. (2011) show the important role of chlorine as precursor element and cyclic acyloxonium ion reaction intermediates in the formation of 3-MCPDE and 2-MCPDE. On the other hand, GE is formed at high temperatures via an intramolecular rearrangement that does not involve chlorine or chlorine precursors (Cheng, Liu, & Liu, 2016; Cheng et al., 2017; Destaillats, Craft, Dubois, & Nagy, 2012).

In recent years, 2-, 3-MCPDE and GE received great attention after the report of the Joint FAO/WHO Expert Committee on Food Additives (JECFA) meeting in 2016, the scientific opinion of the European Food Safety Authority (EFSA) in 2016 and the updated scientific opinion of the latter in 2018 (EFSA CONTAM Panel, 2016; EFSA CONTAM Panel et al., 2018; JECFA, 2017). In 2018, a maximum limit (ML) for the presence of GE in vegetable oils and infant formulae has been established by the European Commission (European Commission, 2018). This ML was set at 1000 μg/kg for vegetable oils and fats, either for direct consumption or as ingredient. For vegetable oils and fats that are going

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to be used for the production of baby food or cereal-based food for young children, the ML for GE was set at 500 μg/kg (European Commission, 2018) pushing the research toward development of effective mitigation strategies. Currently, there are no maximum levels in place for 2- and 3-MCPDE in the EU. However, the European Commission are currently discussing new maximum limits regarding 3-MCPDE in various types of oils (European Commission, 2020).

Oey et al. (2019) concluded that most of the mitigation strategies do not act at multiple refining steps, which could potentially be the most effective strategy. Ramli et al. (2011) and Zulkurnain et al. (2012), have published mitigation strategies focusing on the degumming or bleaching process, respectively. Ramli et al. (2011) achieved a reduction of 64% MCPDE, while Zulkurnain et al. (2012) achieved a 67% reduction for 3-MCPDE. More recently, Sim et al. (2018) reported the successful combined effect of phosphoric acid and acid-activated bleaching earth with acidic pH for the reduction of GE in palm oil (<0.20 mg/kg), but 3-MCPDE mitigation required other conditions. New insights reporting that each type of bleaching earth has its own optimal dosage for the mitigation of 3-MCPDE and GE was provided by Hew et al. (2020). To date, mitigation strategies for 2-MCPDE have not been reported yet.

The aim of this study is to investigate the effects of various time- and temperature profiles in physical refining through pilot plant oil refining experiments, to reduce the formation of 2-, 3-MCPDE, and GE simultaneously. Three treatments, double refining, double refining with a high-low deodorization temperature, and single refining with double deodorization are compared against a single refining control. Pilot plant scale experiments are considered to mimic more realistically oil refining conditions in practice than laboratory scale experiments. Moreover, the implementation of the mitigation strategies to a full-scale refinery can be made with less effort due to the smaller scale differences.

2. Materials & methods

2.1. Materials

All chemicals used for the experiments and analysis are described in the supplemental material as well as the materials used for the pilot plant experiment. In all pilot plant treatments, the following materials and quantities were used. 0.5% w/w of a 25% v/v citric acid solution was used for the degumming process. 1.0% w/w Pure-Flo® B80 natural bleaching earth, 0.1% w/w Cabot Norit® SA 4 PAH-HF activated carbon, and 0.2% w/w Dicalite® 478 perlite filter aid was used during the bleaching process. An Eaton CLEARGAF polyester needlefelt filter bag was used to physically removed all solid, insoluble additives prior to the deodorization process. Each treatment used 100 kg crude palm oil (CPO)

from a single batch as starting each material. 30 kg of the same CPO batch was used to flush the entire pilot plant with 'fresh' CPO prior to each experiment.

2.2. The pilot plant

An experiment on physical refining of CPO was performed at the pilot plant located in the refining factory of Special Refining Company (SRC) in Zaandam, The Netherlands. Fig. 1 shows the schematic drawing of the pilot plant. The pilot plant consists of two main reaction tanks which each can hold 100 kg of oil for the pre-treatment (degumming and bleaching) and for the deodorization. Fully digital time- and temperature control and monitoring are available. The built-in sample collection valves in both tanks allow for sample collection at any moment for any duration of time and/or volume. More details of the pilot plant's design and functionalities are described in the supplemental materials.

2.3. Experimental design

To investigate the temperature–time effect and the impact of the individual refining processes, three physical refining treatments were conducted. The control was a single physical refining to simulate a regular palm oil refining at full scale. A second, identical refining cycle was performed after the control treatment. Immediately after the control treatment, the oil was pumped back into the first tank (Tank X03) and was refined for the second time creating the T1 treatment. After performing this second identical cycle of physical refining, the produced oil was called a double physical refined oil. The temperature–time diagrams of all treatments can be seen in Fig. 2; more specific Fig. 2A shows both the control and T1. Note that the control experiment was the same as the first refining step of T1 (left side of the dashed line in Fig. 2A), and therefore combined in the same experiment. All samples collected during the second refining process (combined with the samples of the control treatment) were used to study the effect of the T1 treatment.

The second treatment (T2) was similar to T1, except that the second deodorization was at a lower temperature (Fig. 2B). The choice to perform the second deodorization at 220 °C originates from previous in-house research performed by our industrial project partner. That temperature is the proverbial border where the formation of GE during the second deodorization can still be kept at a minimum, without compromising other processing parameters such as the vacuum, the strip-steam flow, and reaction times (data not shown). The first refining process of T2 can be directly compared to the control. After the first refining process, the oil was pumped back to Tank X03. This was immediately followed by the start of the second refining process. The same types and
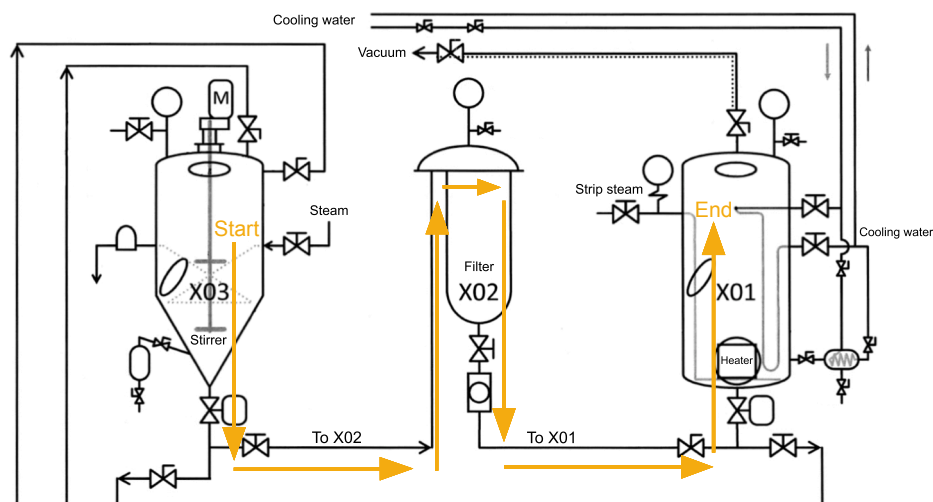

Fig. 1. Schematic overview of the major elements of the pilot plant. The thick orange arrows indicates the flow direction of the oil during refining. Tank 'X03' is the pre-treatment tank for the degumming and bleaching of the oil. Tank 'X01' is the deodorization tank. 'X02' is the filtration unit. The experiment starts after loading the CPO in tank X03 and the refined product is obtained after deodorization in tank X01 ('end'). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
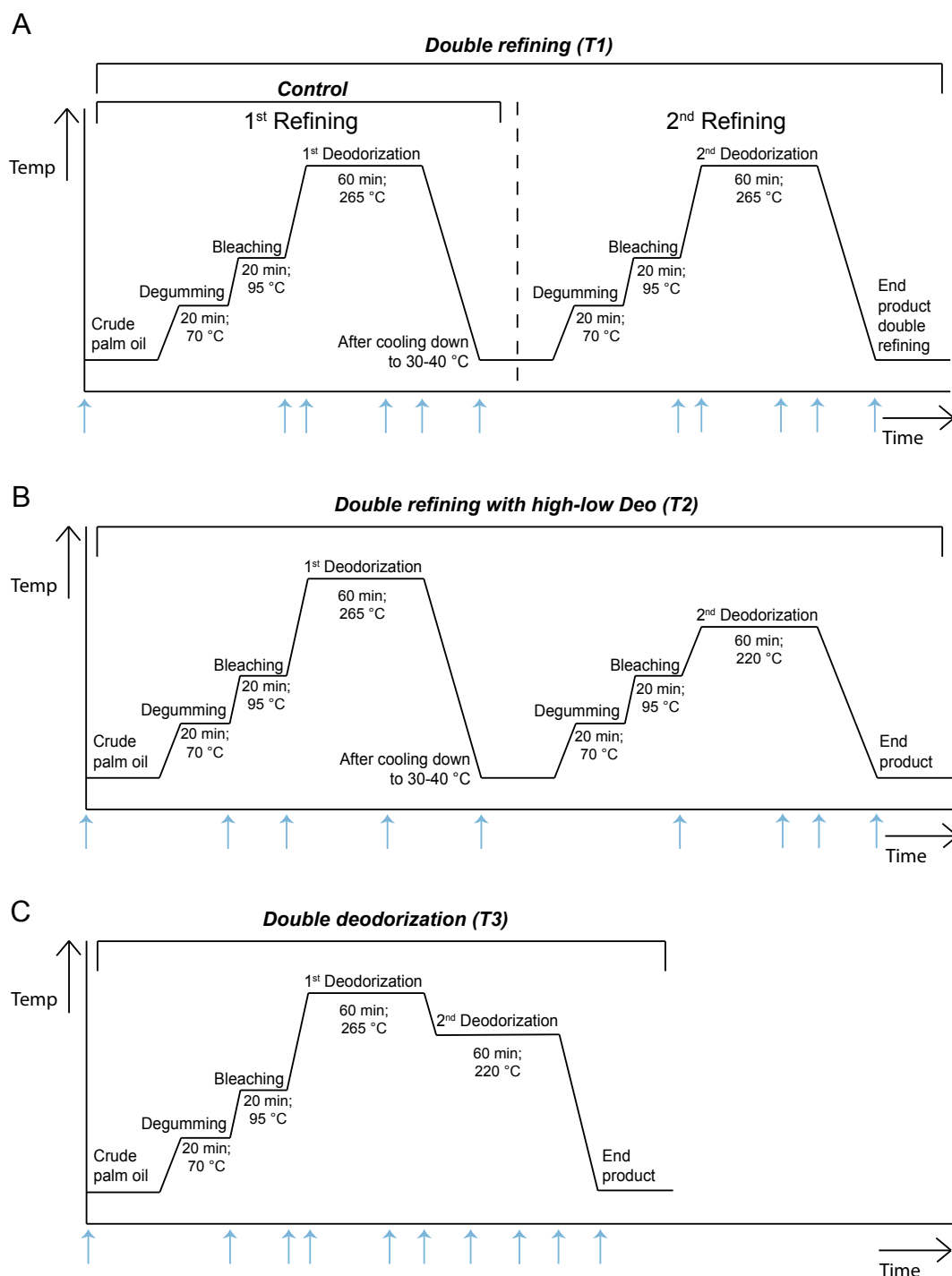
Fig. 2. Time-temperature profile and sampling moments (indicated with blue arrows) of the control experiment (Control, 2A-left), regular double refining (T1, 2A-right), double refining with high-low deodorization temperatures (T2, 2B), and double deodorization experiment (T3, 2C). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

amounts of additives used in the control was also used in T2. The design of the experiments considered that whenever food-grade oil has come in contact with bleaching earth or other non-sterile additives, it must undergo a secondary heat treatment to re-sterilize the oil. Therefore, the secondary heat treatment does not need to be executed at the same temperature as the first deodorization temperature.

The third treatment (T3) was a double deodorization experiment (Fig. 2C). In this experiment, the degumming, bleaching, and filtering processes were performed only once, followed by a deodorization at 265 °C for 60 min. Immediately after 60 min, the oil was cooled down

from 265 °C to 220 °C. The deodorization was then continued for another 60 min. Each treatment and the control were refined once.

2.4. Analytical determinations

Analysis of the oil samples for the concentrations of 2-, 3-MCPDE, and GE were performed using a modified AOCS Cd29a-13 method (Ermacora & Hrncirik, 2013). Some modifications to the AOCS Cd29a-13 method were made to improve the accuracy and sensitivity of the method. The modifications concerned the addition of a separate, third

internal standard for 2-MCPDE (1,3-dipalmitoyl-2-chloropropanediol-d5; PP-2-MCPD-d5; CAS: 1426395–62-1) and the implementation of a multiple reaction monitoring (MRM) MS-mode for the phenylboronic acid (PBA) derivatives of 2-MCPD, 3-MCPD, 2-monobromopropane-1,3-diol (2-MBPD), and 3-monobromopropane-1,2-diol (3-MBPD). Analysis was performed on a Agilent 7010B Triple Quadrupole GC–MS system (Agilent, USA) with an Agilent DB-35MS UI GC column (30 m × 0.250 mm × 0.25 μm) (Agilent, USA). A complete description of the analytical method can be found in the supplemental material together with the in-house validation summary. In general, 100–110 mg of each sample were weighed individually as the starting amount. Internal standards are added and tetrahydrofuran (THF) was used as the main solvent. Conversion of GE into 3-MBPD monoester was performed with an acid aqueous solution of 3 mg/mL sodium bromide and 5% v/v sulfuric acid. After neutralization and clean-up, transesterification of all esters was performed with 1.8% v/v sulfuric acid in methanol at 40 °C for 16 h. Prior to derivatization with PBA, the mixture was neutralized and cleaned up by a series of liquid–liquid extractions to remove the fatty acid methyl esters. Sample analysis for the quantification of the MCPDE and GE were performed in singlets after thorough method validation.

The analytical method validation was performed according to the Dutch NEN 7777 standard. Linearity validation was performed on 8 different days with independent calibration curves. The method accuracy was determined on different days with a set of 8 different samples of vegetable oils (palm and olive oils, three replicates) with known levels of the contaminants. The deviations (e.g. RSD%) were determined by analysis of 8 different oil samples with levels between the limit of detection (LOD) and 3.5 mg/kg. Each sample was analyzed in duplicate on a single day, and another (single) measurement was done on another day. Finally, the LOD and limit of quantification (LOQ) were determined by analyzing 8 spiked oil samples between 0.2 and 0.3 mg/kg. Our quantification method was proven to be linear for the used calibration range for all three contaminants (>0.990 r^2; intercept < 0.009). The average accuracy for 2- and 3-MCPDE, and GE were 99%, 98%, and 105% respectively. The limit of quantification (LOQ) for 2- and 3-MCPDE, and GE were 0.07, 0.10, and 0.07 mg/kg respectively. Additionally, the limit of detection (LOD) for 2- and 3-MCPDE, and GE were 0.04, 0.05, and 0.03 mg/kg respectively. Z-scores from recent proficiency test (FAPAS 2657 in 2019) were −0.2, 0.0, and 0.1 for 2- and 3-MCPDE, and GE respectively.

Additionally, the free fatty acid (FFA) concentration and the color of the oil were analyzed. These analyses were performed on site at SRC. The FFA concentration was determined by titration and were expressed as '% oleic'. The color of the oil samples was determined manually with a visual colorimeter and a 5¼" cuvette. Since the color of the oil is only important for the final samples of each refining, only the last three samples of each treatment were analyzed. The color of the oil samples is expressed in the Lovibond® Red, Yellow, Blue, and Neutral color scale. Sliders with various filters can be arranged in all kind of configuration until the best color match with the tint of the sample is obtained.

3. Results and discussion

3.1. Control & regular double refining (T1)

The results of the control treatment and T1 (unmodified double physical refining) are shown in Fig. 3A. At the end of the control treatment, 2-MCPDE concentration was 0.82 mg/kg, while 3-MCPDE concentration was 1.62 mg/kg. GE concentration was 3.73 mg/kg. After the second full refining process, the final samples of the T1 treatment showed a 2-MCPDE and 3-MCPDE concentrations which are respectively 15% and 17% higher. This is a minor increase compared to the final sample of the control. However, the GE concentration increased by 126%. The vast majority of 2- and 3-MCPDE was already formed before the start of the deodorization (at 265 °C) and remained stable at that level. This finding is similar to the deodorization experimental

results of Hrncirik and van Duijn (2011). They have reported that 3-MCPDE are already being formed at temperature as low as 180 °C for 1 h and were not much affected by further temperature increase up to 230 °C. More recently, Li et al. (2016) showed that sample heated up to 130 °C did not show significant differences in their 3-MCPDE concentrations and that the largest increase in 3-MCPDE concentration was observed between 150 and 180 °C.

GE on the other hand continues to increase during the entire deodorization process until the oil was actively cooled down after 60 min of deodorization. This indicates that MCPD Esters have a lower formation temperature than GE. The formed 2- and 3-MCPDE cannot be removed by a single or double refining process. It seems that 2- and 3-MCPDE are thermally stable and that they remain stable after the addition of 0.5% w/v citric acid. These results confirmed that 2- and 3-MCPDE have a different mechanism of formation than GE.

GE concentration was reduced by 51% during the second pre-treatment (after the second degumming and bleaching process) to 1.82 mg/kg, and further decreased until the start of the second deodorization step, followed by large increase upon exposure to high temperatures during the second deodorization. The formation of GE during the first refining was not surprising since GE is formed at high temperatures. Destaillats, Craft, Dubois, & Nagy (2012), Hrncirik & van Duijn (2011), and Pudel et al. (2011) performed various experiments on the effect of temperature on the formation of GE during palm oil refining. They reported GE formation temperatures between 200 and 230 °C depending on the reaction time. However, the increased formation rate (from 0.19 to 8.59 mg/kg) observed during a second deodorization under the same temperature and duration as the first deodorization has never been reported before.

One of the requirements for the formation of GE is to have either mono- or diglycerides as starting components (Cheng et al., 2016, 2017). Triglycerides in oils can be hydrolyzed under several conditions such as high or low pH environments (Frankel, 2012). A second exposure to citric acid during the second degumming process can cause the hydrolysis of a small part of the triglycerides, forming mono- and/or diglycerides. On its turn, the mono- and/or diglycerides can be converted into GE during the second deodorization process. In a lab scale testing, Cheng et al. (2016) reported a GE concentration of approximately 3.4 mg/kg after deodorization at 260 °C for 1 h. That GE concentration is quite similar to what we have found after our first deodorization at 265 °C for 1 h in the control. As a double refining strategy has not been reported before, there is not much reference for the observations after the second deodorization.

The FFA concentrations in the refined palm oil samples of both the control and the regular double refining experiment (T1) are shown in Table S1 of the supplemental material. The FFA concentration in the final sample after the control treatment and the second refining (T1) are respectively 0.73% and 0.25%. The color of the final sample of the control was 3.1 Red(R)/31.0 Yellow(Y) and the color of the final sample of T1 was 2.8 R/27.0 Y. For large-scale refining, the industry usually strives for a maximum FFA concentration of 0.10% and a color with a maximum of 3.0 R for neutralized/refined, bleached, and deodorized palm oil (Malaysia Department of Standards, 2007). However, as this is still a pilot plant experiment, we do not implement those limits to our results, but we are aware of its existence. The focus of this study was put on the comparison of the different treatments in an almost ideally simulated refining condition.

3.2. Double refining with high-low deodorization (T2)

Like the first experiments above, the majority of the 2- and 3-MCPDE were formed after the filtration step and remained stable throughout the remaining refining process until the end of the second refining process (Fig. 3B). The final concentrations for 2-MCPDE and 3-MCPDE are similar compared to the control treatment.

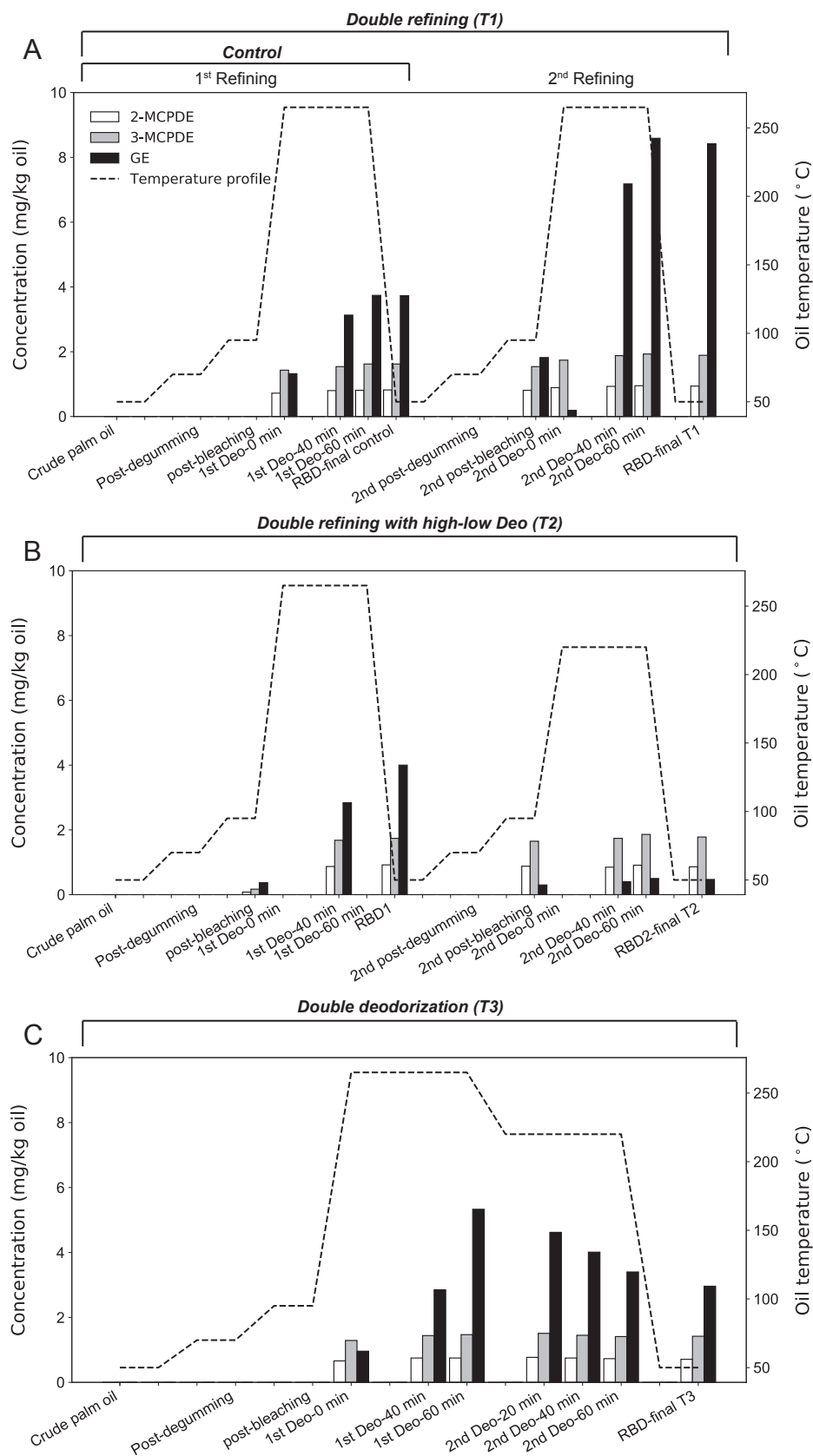After the second pre-treatment process (degumming & bleaching),

Fig. 3. Results of the control experiment (Control; 3A-left), the regular double refining experiment (T1; 3A-right), the double refining experiment with high-low deodorization temperature (T2; 3B), and the double deodorization experiment (T3; 3C). The oil temperature profile for all experiments are shown as a reference (dashed line). The samples were collected and displayed chronologically. These graphs correspond respectively with Table S1–S3 in the supplemental material.

GE was reduced by 93% compared to the 4.00 mg/kg observed after one full cycle of physical refining. This very low GE concentration was maintained throughout the second deodorization. This resulted in an 87% reduction compared to the single refined oil from the control experiment. As this reduction happened before the second deodorization process, the degradation or removal of GE must be correlated to the secondary pre-treatment processes (degumming & bleaching). As GE has a higher formation temperature than 2- and 3-MCPDE, a lower deodorization temperature of 220 °C prevented additional formation of GE. The final sample of T2 have an FFA concentration of 0.39% and a color of 1.9 R/19.0 Y. Full list of FFA concentration and oil color results of T2 can be found in Table S2. Epoxides, as can be found in GE, are unstable in low or high pH environment and high temperatures (Cheng et al., 2016). Exploiting the instability of epoxide under certain conditions could potentially be used to degrade GE.

3.3. Double deodorization (T3)

The double deodorization treatment (T3) results provide insights into the effects of the secondary pre-treatment used during T2, as well as the isolated (dual) temperature effect during deodorization. Fig. 3C shows the concentrations of the three contaminants in the collected samples during experiment T3. The concentration of 2-MCPDE after 60 min deodorization was 0.75 mg/kg and the concentration of 3-MCPDE was 1.47 mg/kg, and these remain stable throughout the experiment, like both T1 and T2 experiments. In terms of mitigating 2- and 3-MCPDE, it is essential to prevent formation in the first place. However, from a practical point of view this can be challenging because there are still many uncertainties regarding the precursors and perhaps even physical limitations of certain refineries to implement a good mitigation strategy. The GE concentration after 60 min of deodorization was 5.33 mg/kg. After the second deodorization process, the GE concentration in the final sample was 2.96 mg/kg. This is a 21% reduction against the final sample of the control, which is less than the reduction achieved in the T2 experiment.

This result once again shows the importance of the second degumming and/or the bleaching process in the mitigation of GE, which was part of T2, but not included in T3 (see Fig. 2). Although a substantial GE reduction was achieved in T3, it is clear double deodorization with a high-low temperature scheme is not sufficient to produce refined palm oil that meets the GE maximum limits.

Two publications have reported results of double deodorizations (Matthäus & Pudel, 2013; Shimizu, Weitkamp, Vosmann, & Matthäus, 2013). Matthäus and Pudel (2013) reported that a double deodorization performed at 200 °C for 120 min, followed by a brief 5 min at 250 °C was able to achieve the lowest amount of GE and 3-MCPDE, both at approximately 1 mg/kg. This was a reduction of approximately 50% for both GE and 3-MCPDE. When the second high-temperature phase was increased to 270 °C, the final GE concentration was approximately 3 mg/kg. In contrast to the findings of Matthäus and Pudel (2013), the 3-MCPDE concentrations in our experiments remained stable once being formed. Shimizu et al. (2013) performed a two-step heating test (240 °C for 60 min followed by 180 °C for 240 min) with artificially added chloride into their oil and reported a reduction of approximately 67% for GE (from 105 mg/kg to 35 mg/kg). Similar to Shimizu et al. (2013), we were able to observe a reduction in GE concentration with the high-low double deodorization temperature approach. However, a direct comparison between the results of previous studies and our study is not possible due to the many differences in refining methodology.

The experimental design of the control, first refining of T2, and up to the first deodorization (at 265 °C) of T3 had identical temperature–time settings and same amounts and batch of chemicals, crude oil and other additives. However, our data still show a certain degree of variation for all three contaminants. In average, 3-MCPDE varied with ±8.4%, 2-MCPDE with ±10.5%, and GE with ±24.5%.

Unlike the control and the T2 treatment, T3 was able to achieve a low

FFA concentration and a less red-colored oil. The low FFA concentration of 0.08% in the final sample may be explained by a prolonged deodorization process in which more FFA are distilled and/or degraded without the addition of additives that might induce formation of FFA. However, the exact reason for the variation of observed FFA concentrations amongst all the experiments remains unclarified. Nonetheless, as mentioned before, upscaling the pilot plant refining methods into full-scale resulted in a refined palm oil with good color and FFA concentration. A recent study on the type and amount of bleaching earth usage by Hew et al. (2020) did not show any obvious correlation between the type or amount of bleaching earth and the FFA concentration. Full list of FFA concentration and oil color results of T3 can be found in Table S3.

Due to the current regulatory EC maximum limits for GE in vegetable fats and oils, refineries must focus on meeting those limits. These mitigation strategies will lead to increased total costs per weight refined palm oil than for example a traditional single physical refining. However, exact financial statements and calculations of the costs of the proposed refining scheme are beyond the aim of this study.

4. Conclusions

Double refining with a high-low deodorization temperature strategy (T2) showed great potential to mitigate GE formation during oil refining at pilot plant scale experiments; it resulted in an 87% reduction as compared to single physical refining while double deodorization (T3) was less effective. Further contaminant reductions may be achieved by varying refining other oil refining parameters, that now have been kept the same, such as the type of acid and/or BE and their respective concentrations.

From the double refining with a high-low deodorization temperature treatment, bleaching and degumming appeared to be the key steps to achieve the GE reduction. Both the double refining with a high-low deodorization temperature and the double deodorization treatments were not able to reduce the presence of 2- and 3-MCPDE. Once these contaminants are formed, they remain in the oil at a constant concentration. Therefore, their formation should be prevented as much as possible before the physical refining.

CRediT authorship contribution statement

Sergio B. Oey: Methodology, Investigation, Writing - original draft, Visualization. **H.J. Fels-Klerx:** Conceptualization, Writing - review & editing, Supervision, Funding acquisition. **Vincenzo Fogliano:** Conceptualization, Writing - review & editing, Supervision. **Stefan P.J. Leeuwen:** Conceptualization, Validation, Writing - review & editing, Supervision, Project administration.

Author contributions

S.B. Oey proposed the experimental design, analyzed the samples, compiled data in tables and graphs, drafted the manuscript, and

corrected comments. H.J. van der Fels-Klerx, V. Fogliano, and S.P.J. van Leeuwen contributed to scoping the study, maturing the experimental design, reviewing draft manuscript versions, and providing editorial corrections.

Author disclosures

All authors have read and approved the manuscript. Authors declare to not have any conflict of interest.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodres.2020.109748.

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