

Dietary fishmeal levels affect the volatile compounds in cooked muscle of farmed large yellow croaker *Larimichthys crocea*

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Funding information

the Fundamental Research Funds for the Central Universities of Ocean University of China, Grant/Award Number: 201562017; National Natural Science Foundation of China, Grant/Award Number: 31372542; Academician Workstation for Aquatic Animal Nutrition and Feed in Guangdong Evergreen, Grant/Award Number: 2014B090905014

Abstract

A comparative study on the volatile compounds in cooked muscle of wild and farmed large yellow croaker (LYC) was conducted. The two farmed LYC groups were fed with diets containing 44% (CF) and 25% (LF) of fish meal (FM) respectively. Results showed that 48 volatiles, including aldehydes, alcohols, ketones, hydrocarbons, aromatics, acids, esters, furans and miscellaneous compound, were detected in cooked fillets. The LF group had significantly lower amounts of total aldehydes and ketones, higher content of miscellaneous compound in cooked fillets than that in the CF and wild groups ($p < .05$). Compared with the wild group, the LF group had significantly lower amounts of total alcohols, acids and esters, while the CF group had significantly lower amounts of total aldehydes, higher content of total ketones in cooked muscle ($p < .05$). According to the principal component analysis (PCA), some volatiles (propanal, nonanal, etc.) could be considered as sensitive indicators to classify cooked muscle samples. In conclusion, differences in the volatiles in the cooked muscle between the wild and farmed LYC have been found. Low level of dietary FM (25%) changed the volatile profiles in cooked fillets of farmed LYC. A PCA may be useful to screen potential volatiles to classify cooked muscle samples in this study.

KEYWORDS

cooked muscle, Fish meal, large yellow croaker, principal component analysis, volatile compounds

1 | INTRODUCTION

The odour is one of the most significant factors determining the character and quality of fish species. Many researchers have studied the aroma-active components of various fish species. Cayhan and Selli (2010) analysed the key aroma compounds in cooked grey mullet (*Mugil cephalus*). Sun et al. (2013) studied the changes in the volatile profiles of bigeye tuna (*Thunnus obesus*) meat before and after various heat treatments.

With 148,616 metric tons of production in 2015 (China Fishery Statistical Yearbook, 2016), large yellow croaker (*Larimichthys crocea*; LYC) is a highly appreciated and first major mariculture fish species in China. Compared with wild LYC, however, farmed one showed the fattier body, whiter skin colour, softer muscle, higher fishy odour and odour intensity, and lower taste feeling. This results in low market price and poor consumer acceptability of farmed LYC. Grigorakis, Taylor and Alexis (2003) found that wild gilthead sea bream (*Sparus aurata*) contained a higher number of volatiles than that of farmed counterpart. Moreover, rearing conditions, diet composition and handling processes may also influence fish muscle volatiles (Hallier,

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Prost & Serot, 2005; Moreira et al., 2014; Zhou, Chong, Ding, Gu & Liu, 2016). Concerning the diet composition, Moreira et al. (2014) and Alexi, Fountoulaki and Grigorakis (2016) found that dietary plant protein and oil sources significantly affected the contents of some volatiles (1-penten-3-ol, α -pinene, etc.) in fillets of Senegalese sole (*Solea senegalensis* Kaup, 1858) and gilthead sea bream respectively.

Fish meal (FM) was used as the main protein source in compound aquafeed for mariculture fish. However, increasing demand, uncertain availability and high price of FM with the expansion of aquaculture made it essential to search alternative protein sources. In previous study, Mu et al. (2017) found that compared with the control diet (44% FM), low dietary FM (25% FM) significantly influenced the volatiles in the raw muscle of farmed LYC. Meanwhile, in China, people usually consume cooked rather than raw LYC muscle. The types and amounts of volatiles could be different between raw and cooked fish muscle (Moreira, Valente, Castro-Cunha, Cunha & de Pinho, 2013; Sun et al., 2013). So, the aims of this study were to evaluate possible effects of low dietary FM content on volatile compounds in the cooked fillets of LYC, and to compare the differences in muscle volatiles between wild and farmed fish. Moreover, to screen the potential and sensitive volatile indicators in cooked muscle, the volatiles of LYC fillets were statistically interpreted by principal component analysis (PCA).

2 | MATERIALS AND METHODS

2.1 | Experimental animals and diets

There were two groups of fishmeal-based diets farmed LYC with 267.47 ± 5.88 g and 251.46 ± 3.92 g of body weight respectively. The two diets were supplemented with 44% (CF) and 25% (LF) FM respectively. And the formulation and proximate compositions of the experimental diets were listed in the previous study (Mu et al., 2017). The third group was the wild LYC (296.15 ± 6.55 g) obtained at Xiangshan bay of Ningbo, Zhejiang Province, China. There were 10 LYC per group, and four fish per group were used for the subsequent analysis.

2.2 | Chemicals

Propanal, 3-methyl-butanol, pentanal, decanal, undecanal, dodecanal, 3-methyl-1-butanol, 1-pentanol, 1-hexanol, 1-undecanol, acetone, 2, 3-pentanedione, cyclohexanone, 2-undecanone, decane, undecane, dodecane, *p*-xylene, *o*-xylene, *m*-xylene, hexadecanoic acid, octadecanoic acid and 2, 4, 6-trimethyl-pyridine standards were purchased from Sigma-Aldrich. All reagents used in this work were of analytical reagent grade.

2.3 | Analysis of volatile compounds

2.3.1 | Sample preparation

Frozen fillets were thawed at 4°C just before analysis and then were sliced into small pieces, placed in a plate and cooked in a boiling

water bath for 5 min (Moreira et al., 2013). After being drained and cooled, the cooked fillet was minced and mixed. Five grams of cooked muscle and 0.25 g of sodium chloride were weighted and placed into a 15 ml headspace vial. Then 33 μ l of an aqueous solution of 91.82 μ g/ml of 2, 4, 6-trimethyl-pyridine (Sigma-Aldrich) was added as an internal standard (IS). Headspace vial was sealed with a screwcap fitted with a septum.

2.3.2 | Headspace solid-phase microextraction

Volatile compounds in cooked muscle were extracted according to the headspace solid-phase microextraction (HS-SPME) method of Iglesias and Medina (2008) with slight modifications. The manual HS-SPME device (Supelco Inc., USA) equipped with a 50/30 μ m divinyl benzene-carboxen-polydimethylsiloxane fibre (DVB/CAR/PDMS; Supelco Inc., USA) was used for extraction of volatiles from the fish muscle. The sample in vial was equilibrated at 60°C for 20 min followed by HS-SPME exposure at the same temperature. The volatiles in the headspace were absorbed onto SPME fibres for 40 min.

2.3.3 | GC-MS analysis of volatile compounds

After the headspace collection of volatiles in fish muscle, GC-MS analysis was performed with a GCMS-QP2010 (Shimadzu, Japan), which was equipped with a Rxi-1MS capillary column (30 m \times 0.25 mm id, 0.25 μ m film thickness; Shimadzu, Japan). The method of GC-MS analysis was described by Mu et al. (2017).

2.4 | Statistical analysis

The differences in the volatile profiles in cooked muscle among the wild and two farmed LYC groups were analysed by one-way analysis of variance (ANOVA) followed by Tukey's multiple range test. Differences in the volatiles amounts between the raw and cooked muscle were analysed by *t* test. The level of significance was chosen at $p < .05$, and data are expressed as mean values ($n = 4$) accompanied by the standard errors of means. Above data analyses were performed by SPSS 17.0 for windows.

The muscle volatile profiles were also analysed using PCA to understand the similarities and dissimilarities among variables. The data have been preprocessed by normalization prior to the PCA. The analysis was carried out using the software of SIMCA-P 11.5 (Umetrics AB, Malmö, Sweden).

3 | RESULTS AND DISCUSSION

Table 1 lists the 48 volatiles detected in the cooked muscle of LYC. In which, 37, 38 and 45 volatile compounds were found in the LF group, CF group and wild group respectively. All the volatiles detected in cooked muscle can be divided into nine groups, including 11 aldehydes, eight alcohols, seven ketones, nine hydrocarbons, six aromatics, two acids, two esters, two furans and one miscellaneous

TABLE 1 Volatile compounds ($\mu\text{g/g}$ muscle) in the raw and cooked muscle of large yellow croaker including the farmed fish fed low fish meal diet (LF), farmed fish fed control diet (CF) and wild fish (WF)

Code ^a	Volatile compounds (42/48) ^b	LF		CF		WF	
		Raw muscle ^c	Cooked muscle	Raw muscle ^c	Cooked muscle	Raw muscle ^c	Cooked muscle
a	Aldehydes (9/11)						
a1	Propanal	0.10 \pm 0.04	nd.	0.32 \pm 0.05*	nd.	0.38 \pm 0.07*	nd.
a2	3-methylbutanal	nd.	nd. ^A	nd.	nd. ^A	nd.	0.23 \pm 0.04 ^{B*}
a3	Pentanal	nd.	0.25 \pm 0.01 ^{B*}	nd.	0.11 \pm 0.02 ^{A*}	nd.	0.11 \pm 0.02 ^{A*}
a4	Hexanal	0.35 \pm 0.06*	0.14 \pm 0.03 ^A	0.96 \pm 0.11*	0.43 \pm 0.07 ^B	1.03 \pm 0.17	0.76 \pm 0.06 ^C
a5	(Z)-4-Heptenal	nd.	0.03 \pm 0.00 ^{A*}	nd.	0.04 \pm 0.01 ^{A*}	nd.	0.06 \pm 0.00 ^{B*}
a6	Heptanal	0.09 \pm 0.02	0.07 \pm 0.01 ^A	0.26 \pm 0.05	0.15 \pm 0.03 ^A	0.99 \pm 0.29	0.40 \pm 0.06 ^B
a7	Benzaldehyde	0.44 \pm 0.10*	0.06 \pm 0.01 ^A	0.27 \pm 0.12	0.08 \pm 0.01 ^{AB}	nd.	0.11 \pm 0.01 ^{B*}
a8	Octanal	0.06 \pm 0.01	0.03 \pm 0.01 ^A	0.11 \pm 0.02	0.05 \pm 0.01 ^A	0.84 \pm 0.08*	0.14 \pm 0.03 ^B
a9	Nonanal	0.14 \pm 0.03	0.08 \pm 0.01 ^A	0.41 \pm 0.05*	0.13 \pm 0.01 ^A	0.41 \pm 0.08	0.41 \pm 0.07 ^B
a10	Decanal	0.11 \pm 0.01*	nd.	0.14 \pm 0.03*	nd.	nd.	nd.
a11	Undecanal	0.02 \pm 0.00*	nd. ^A	0.02 \pm 0.00*	nd. ^A	nd.	0.06 \pm 0.01 ^{B*}
a12	Dodecanal	0.02 \pm 0.00*	nd. ^A	0.03 \pm 0.01*	nd. ^A	nd.	0.04 \pm 0.01 ^{B*}
a13	Pentadecanal	nd.	nd. ^A	nd.	nd. ^A	nd.	0.01 \pm 0.00 ^{B*}
	Subtotal	1.33 \pm 0.12*	0.67 \pm 0.02 ^A	2.51 \pm 0.20*	0.99 \pm 0.06 ^B	3.39 \pm 0.30*	2.33 \pm 0.13 ^C
b	Alcohols (7/8)						
b1	3-methyl-1-Butanol	nd.	nd.	nd.	nd.	0.19 \pm 0.03*	nd.
b2	1-Penten-3-ol	0.05 \pm 0.01	0.04 \pm 0.01 ^B	0.17 \pm 0.03*	0.03 \pm 0.01 ^{AB}	0.14 \pm 0.04*	0.01 \pm 0.00 ^A
b3	1-Pentanol	0.06 \pm 0.00*	0.02 \pm 0.00	0.19 \pm 0.03*	0.04 \pm 0.01	0.13 \pm 0.04	0.04 \pm 0.01
b4	(Z)-2-Penten-1-ol	0.07 \pm 0.00	0.05 \pm 0.01 ^B	0.22 \pm 0.03*	0.03 \pm 0.01 ^{AB}	0.18 \pm 0.04*	0.02 \pm 0.00 ^A
b5	1-Hexanol	nd.	0.01 \pm 0.00 ^A	nd.	0.03 \pm 0.00 ^{AB*}	nd.	0.05 \pm 0.01 ^{B*}
b6	1-Heptanol	0.02 \pm 0.00	0.02 \pm 0.00 ^A	0.03 \pm 0.01	0.03 \pm 0.00 ^B	nd.	0.04 \pm 0.00 ^{B*}
b7	1-Octen-3-ol	nd.	nd. ^A	nd.	nd. ^A	nd.	0.03 \pm 0.01 ^{B*}
b8	1-Octanol	0.01 \pm 0.00	0.01 \pm 0.00 ^A	0.01 \pm 0.00	0.03 \pm 0.00 ^{B*}	nd.	0.04 \pm 0.00 ^{B*}
b9	Undecanol	0.02 \pm 0.00*	0.01 \pm 0.00 ^A	0.03 \pm 0.00	0.02 \pm 0.00 ^{AB}	nd.	0.04 \pm 0.01 ^{B*}
	Subtotal	0.22 \pm 0.01*	0.16 \pm 0.01 ^A	0.66 \pm 0.07*	0.21 \pm 0.02 ^{AB}	0.60 \pm 0.04*	0.27 \pm 0.03 ^B
c	Ketones (6/7)						
c1	Acetone	nd.	nd. ^A	nd.	0.73 \pm 0.06 ^{B*}	nd.	nd. ^A
c2	2,3-Pentanedione	nd.	nd.	nd.	nd.	0.20 \pm 0.06*	nd.
c3	2-Hexanone	0.07 \pm 0.01	0.11 \pm 0.01 ^{A*}	0.26 \pm 0.02	0.20 \pm 0.01 ^B	0.18 \pm 0.04	0.19 \pm 0.01 ^B
c4	Cyclohexanone	0.22 \pm 0.05*	nd.	nd.	nd.	nd.	nd.
c5	2-Heptanone	nd.	nd. ^A	nd.	nd. ^A	nd.	0.07 \pm 0.00 ^{B*}
c6	2,3-Octanedione	0.02 \pm 0.00	0.02 \pm 0.00 ^A	0.05 \pm 0.00*	0.02 \pm 0.00 ^{AB}	0.07 \pm 0.01*	0.04 \pm 0.01 ^B
c7	6-methyl-5-Hepten-2-one	0.03 \pm 0.00*	nd.	0.02 \pm 0.00*	nd.	nd.	nd.
c8	Acetophenone	nd.	nd. ^A	nd.	nd. ^A	nd.	0.04 \pm 0.01 ^{B*}
c9	2-Nonanone	nd.	nd. ^A	nd.	nd. ^A	nd.	0.02 \pm 0.00 ^{B*}
c10	2-Undecanone	0.01 \pm 0.00*	tr. ^A	0.01 \pm 0.00	0.01 \pm 0.00 ^A	nd.	0.03 \pm 0.00 ^{B*}
	Subtotal	0.34 \pm 0.05*	0.13 \pm 0.00 ^A	0.33 \pm 0.03	0.97 \pm 0.05 ^{C*}	0.43 \pm 0.04	0.39 \pm 0.00 ^B
d	Hydrocarbons (10/9)						
d1	Hexane	0.26 \pm 0.11	nd.	0.68 \pm 0.20*	nd.	1.11 \pm 0.26*	nd.
d2	Decane	nd.	0.03 \pm 0.00 ^{C*}	nd.	0.01 \pm 0.00 ^{B*}	nd.	nd. ^A
d3	Undecane	0.02 \pm 0.00	0.06 \pm 0.01 ^{A*}	0.07 \pm 0.03	0.11 \pm 0.01 ^B	0.16 \pm 0.05	0.12 \pm 0.01 ^B
d4	Dodecane	0.04 \pm 0.00	0.06 \pm 0.01 ^A	0.23 \pm 0.06	0.11 \pm 0.01 ^B	0.29 \pm 0.07	0.10 \pm 0.00 ^B
d5	1-Tridecene	0.01 \pm 0.00*	nd.	0.02 \pm 0.00*	nd.	nd.	nd.

(Continues)

TABLE 1 (Continued)

Code ^a	Volatile compounds (42/48) ^b	LF		CF		WF	
		Raw muscle ^c	Cooked muscle	Raw muscle ^c	Cooked muscle	Raw muscle ^c	Cooked muscle
d6	Tridecane	0.03 ± 0.00	0.05 ± 0.01*	0.08 ± 0.01	0.07 ± 0.01	0.10 ± 0.03	0.07 ± 0.02
d7	Tetradecane	0.05 ± 0.01*	0.01 ± 0.00 ^A	0.10 ± 0.02*	0.03 ± 0.00 ^{AB}	0.11 ± 0.04	0.04 ± 0.01 ^B
d8	Pentadecane	0.29 ± 0.03*	0.10 ± 0.01 ^B	0.30 ± 0.06*	0.07 ± 0.01 ^A	0.05 ± 0.01	0.04 ± 0.01 ^A
d9	Hexadecane	0.03 ± 0.01*	tr. ^A	0.03 ± 0.01*	0.01 ± 0.00 ^{AB}	nd.	0.02 ± 0.00 ^{B*}
d10	Heptadecane	0.21 ± 0.04*	0.09 ± 0.02 ^B	0.24 ± 0.07	0.07 ± 0.01 ^{AB}	0.02 ± 0.00	0.05 ± 0.00 ^{A*}
d11	2,6,10,14-Tetramethyl-Pentadecane	0.04 ± 0.00*	0.01 ± 0.00 ^A	0.03 ± 0.01	0.02 ± 0.00 ^{AB}	0.02 ± 0.00	0.04 ± 0.01 ^B
	Subtotal	0.97 ± 0.09*	0.43 ± 0.03	1.78 ± 0.32*	0.51 ± 0.03	1.86 ± 0.37*	0.49 ± 0.05
e	Aromatics (6/6)						
e1	Toluene	0.04 ± 0.01	0.07 ± 0.01	0.32 ± 0.09*	0.04 ± 0.01	0.40 ± 0.12*	0.05 ± 0.01
e2	Ethylbenzene	0.12 ± 0.02	0.09 ± 0.01	0.37 ± 0.11	0.14 ± 0.03	0.95 ± 0.13*	0.14 ± 0.04
e3	Styrene	0.09 ± 0.01*	0.02 ± 0.00 ^A	0.21 ± 0.05*	0.05 ± 0.01 ^{AB}	0.43 ± 0.14*	0.10 ± 0.03 ^B
e4	<i>p</i> -Xylene	0.09 ± 0.01*	0.02 ± 0.00	0.22 ± 0.01*	0.03 ± 0.00	0.34 ± 0.05*	0.03 ± 0.01
e5	<i>o</i> -Xylene	0.12 ± 0.01	0.27 ± 0.08	0.31 ± 0.09	0.24 ± 0.06	0.65 ± 0.19*	0.14 ± 0.03
e6	<i>m</i> -Xylene	0.02 ± 0.00*	0.01 ± 0.00 ^B	0.12 ± 0.04*	tr. ^A	0.21 ± 0.07*	nd. ^A
	Subtotal	0.49 ± 0.03	0.48 ± 0.10	1.55 ± 0.15*	0.50 ± 0.10	2.40 ± 0.31*	0.47 ± 0.06
f	Acids (1/2)						
f1	Hexadecanoic acid	0.01 ± 0.00	0.03 ± 0.01 ^{A*}	0.03 ± 0.01	0.05 ± 0.00 ^{AB*}	nd.	0.06 ± 0.01 ^{B*}
f2	Octadecanoic acid	nd.	0.01 ± 0.00 ^{A*}	nd.	0.02 ± 0.00 ^{AB*}	nd.	0.03 ± 0.01 ^{B*}
	Subtotal	0.01 ± 0.00	0.04 ± 0.01 ^{A*}	0.03 ± 0.01	0.07 ± 0.00 ^{AB*}	nd.	0.09 ± 0.01 ^{B*}
g	Esters (2/2)						
g1	Butyl propanoate	0.03 ± 0.01*	0.01 ± 0.00 ^A	0.03 ± 0.01	0.01 ± 0.00 ^A	nd.	0.02 ± 0.00 ^{B*}
g2	Diethyl phthalate	0.24 ± 0.02*	0.08 ± 0.01 ^A	0.62 ± 0.08*	0.26 ± 0.08 ^{AB}	0.01 ± 0.00	0.36 ± 0.07 ^{B*}
	Subtotal	0.27 ± 0.02*	0.09 ± 0.01 ^A	0.65 ± 0.08*	0.27 ± 0.08 ^{AB}	0.01 ± 0.00	0.38 ± 0.07 ^{B*}
h	Furans (0/2)						
h1	2-ethylfuran	nd.	nd. ^A	nd.	nd. ^A	nd.	0.34 ± 0.02 ^{B*}
h2	2-pentylfuran	nd.	nd. ^A	nd.	nd. ^A	nd.	0.06 ± 0.02 ^{B*}
	Subtotal	nd.	nd. ^A	nd.	nd. ^A	nd.	0.41 ± 0.03 ^{B*}
i	Miscellaneous compounds (1/1)						
i1	1-methylene-1 <i>H</i> -Indene	0.05 ± 0.01	0.13 ± 0.02 ^{B*}	0.04 ± 0.01	0.06 ± 0.01 ^A	0.02 ± 0.00	0.05 ± 0.01 ^A
	Total	3.67 ± 0.17*	2.12 ± 0.15 ^A	7.55 ± 0.24*	3.58 ± 0.15 ^B	8.71 ± 0.41*	4.88 ± 0.28 ^C

nd., not detected; tr., traces (<0.01 µg/g muscle). Data within the same row with different superscript uppercase letters are significantly ($p < .05$) different; *indicate a significant ($p < .05$) difference between raw and cooked muscle volatile compounds.

^aCompound code as it appears in Figure 1a.

^bThe number of volatiles in the raw/cooked muscle.

^cData published by Mu et al. (2017).

compound. Aldehydes, hydrocarbons and aromatics were the major volatile components. Aroma descriptors, odour threshold values and possible origins of some volatile compounds found in fish are presented in Table 2.

3.1 | Volatile compounds in cooked muscle

3.1.1 | Aldehydes

In total, seven aldehydes were identified in the cooked fillets (Table 1). Compared with CF group, the LF group had a significant

lower concentration of hexanal, and higher pentanal ($p < .05$). There were no significant differences in the levels of (Z)-4-heptenal, heptanal, benzaldehyde, octanal and nonanal in the cooked muscle between the two farmed fish groups ($p > .05$). Similar result was obtained in cooked muscle of barramundi (*Lates calcarifer*) and Senegalese sole, in which hexanal was quantitatively the most abundant compound (Frank, Poole, Kirchhoff & Forde, 2009; Moreira et al., 2013). Nonanal has been detected in cooked grey mullet and barramundi as an aroma-active compound (Cayhan & Selli, 2010; Frank et al., 2009).

TABLE 2 Odour descriptions, thresholds ($\mu\text{g}/\text{kg}$) and possible origins given by the literature of volatile compounds identified in large yellow croaker muscle

Volatile compounds	Odour description ^a	Threshold ($\mu\text{g}/\text{kg}$) ^b	Possible origin ^c
Aldehydes			
Propanal	Acetaldehyde-like, pungent	15.1	Oxidation of linolenic acid
3-methylbutanal	Pungent, solvent, almond, nutty	1.1	Strecker degradation of leucine
Pentanal	Almond, malt, fruity, pungent, acetone, sweet, wine	1.57	Oxidation of n-6 PUFA
Hexanal	Garlic, fresh, green, grassy, pungent, tallow, fat, green beans, fishy	4.5–5	Oxidation of n-3, n-6 and n-9 PUFA
Z-4-Heptenal	Boiled potato, cooked fish	0.8	Retro-aldol condensation of 2,6-nonadienal
Heptanal	Green, floral, fatty, pungent, fishy, dry fish, citrus fruit	2.8	Oxidation of oleic and linoleic acid
Benzaldehyde	Almond, fruity, creamy, nutty	350–3,500	Strecker degradation of phenylglycine
Octanal	Sweet, orange, floral, spicy, citrus, green, fatty	0.7	Oxidation of oleic and linoleic acid
Nonanal	Geranium, raw fish, plastic, marine, citrus, green, fatty	1	Oxidation of oleic and linoleic acid
Decanal	Marine, cucumber, floral, fat, orange peel, tallow, green	2.71	Oxidation of n-9 PUFA
Undecanal	Minty, fruity	5	Oxidation of oleic acid
Dodecanal	Lily, fat, citrus	2	
Alcohols			
3-methyl-1-Butanol	Balsamic	4	Oxidative deamination of leucine
1-Penten-3-ol	Plastic, green, pungent, solvent, vegetal, burnt, meaty	358.1	Oxidation of eicosapentaenoic acid by 15-lipoxygenase and hydroperoxide lyases
1-Pentanol	Mushroom, earthy, green, wax	150.2	
(Z)-2-Penten-1-ol	Grilled hazel nut		
1-Hexanol	Green, grassy	5.6	
1-Heptanol	Green, savoury, fermented, fresh, nutty	5.4	
1-Octen-3-ol	Mushroom, fermented, earthy, herbaceous, spicy, mushroom, fishy, grassy	1.2	Oxidation of arachidonic acid by 12-lipoxygenase and degradation of linoleic acid hydroperoxide
1-Octanol	Fatty, green, floral	125.8	Oxidation of oleic acid
Ketones			Thermal degradation, lipid oxidation, amino acid degradation, microbial oxidation and Maillard reaction
2, 3-Pentanedione	Sweet, buttery, caramel, fruity	30	
2-Hexanone	Green, fruity, floral		
2-Heptanone	Fruity, floral, green, pungent		
2, 3-Octanedione	Savoury, cooked		
6-methyl-5-Hepten-2-one	Green, sweet, fruity	68	
Acetophenone	Sweet rose floral		
2-Nonanone	Fruity, rot, creamy	0.04	
2-Undecanone	Tallow, musty, fruity, floral, woody	5.5	
Hydrocarbons			Saturated alkanes could come from decarboxylation and splitting of carbon-carbon chain of higher fatty acids
Dodecane	Cheese		
2, 6, 10, 14-tetramethyl-pentadecane	Floral, woody, green, cooked		

(Continues)

TABLE 2 (Continued)

Volatile compounds	Odour description ^a	Threshold (µg/kg) ^b	Possible origin ^c
Aromatics			
Toluene	Plastic		
Ethylbenzene	Ethereal, floral, concrete-like	2,205.25	
Styrene	Plastic	730	
<i>p</i> -Xylene	Pungent, phenolic	450.23	
<i>o</i> -Xylene	Geranium, oily, pungent		
<i>m</i> -Xylene	Phenolic		
Acids			
Hexadecanoic acid	Fresh, fruity		
Furans			
			Products derived from Maillard and Strecker degradation of amino acids and sugars, oxidation of fatty acids
2-ethylfuran	Coffee-like, burnt, sweet, rubber, pungent	2.3	The product of linolenate oxidative degradation
2-pentylfuran	Green bean, sweet, spicy	5.8	

^aOdour descriptions from literatures: Frank et al. (2009), Grigorakis et al. (2003), Hallier et al. (2005), Kawai (1996), Varlet et al. (2007), Tanchotikul & Hsieh (1989), Girard & Durance (2000), Giri et al. (2010), Prost et al. (1998); Vejaphan et al. (1988), Cayhan & Selli (2010), Sérot et al. (2002), Selli et al. (2006).

^bOdour thresholds from literatures: Frank et al. (2009), Giri et al. (2010), Guadagni, Buttery & Okano (1963), Refsgaard, Haahr & Jensen (1999).

^cPossible origins from literatures: Kawai (1996), Varlet et al. (2007), Silva, Valente, Castro-Cunha, Bacelar & de Pinho (2012), Girard & Durance (2000), Giri et al. (2010), Chung et al. (2007), Alasalvar, Taylor & Shahidi (2005), Josephson & Lindsay, (1986), Sérot et al. (2002), Duflos et al. (2006).

3.1.2 | Alcohols

A total of seven alcohols were detected in the cooked fillets of farmed LYC (Table 1). Among these alcohols, the levels of 1-heptanol and 1-octanol were significantly lower in the LF group ($p < .05$). The 1-heptanol has been described as giving green, savoury, fermented, fresh and nutty odour (Table 2; Frank et al., 2009; Giri, Osako & Ohshima, 2010; Hallier et al., 2005). Fatty, green and floral are some odour descriptors used to define the aroma of 1-octanol in different literatures (Giri et al., 2010; Sérot, Regost & Arzel, 2002). This alcohol has been considered as deriving from the oxidation of oleic acid (Table 2; Sérot et al., 2002).

3.1.3 | Ketones

In total, three and four ketones were identified in the cooked fillets of LYC fed the LF and CF diet respectively (Table 1). Acetone was detected in the cooked flesh of the CF group, not the LF group. Meanwhile, the CF group had a significant higher level of 2-hexanone ($p < .05$). Tanimoto, Kitabayashi, Fukusima, Sugiyama and Hashimoto (2015) found that the level of acetone in cooked ordinary muscle of yellowtail *Seriola quinqueradiata* was significantly lower than that in the cooked dark muscle. Furthermore, they pointed out that acetone in ordinary muscle had a high correlation with its lipid oxidation indices (peroxide values [PVs] and thiobarbituric acid-reactive substances [TBARS] values). The content of acetone in longissimus muscle of pigs tended to decrease with the decrease in dietary linoleic acid content (Larick, Turner, Schoenherr, Coffey &

Pilkington, 1992). Thermal degradation, lipid oxidation, amino acid degradation, microbial oxidation and Maillard reactions were the possible mechanisms for ketone formation (Chung, Yeung, Kim & Chen, 2007). Therefore, more acetone detected in the CF group could indicate more biochemical changes in fish muscle of the CF group. But further studies needed to explain this finding.

3.1.4 | Hydrocarbons

Nine kinds of hydrocarbons including alkenes and alkanes were detected in the cooked fillets of the two farmed LYC groups (Table 1). The LF group had significant higher amounts of decane and pentadecane, and lower levels of undecane and dodecane in the cooked muscle ($p < .05$).

3.1.5 | Aromatics

Six aromatics were detected in the cooked fillets of farmed LYC (Table 1). Compared with the CF group, a significant higher content of *m*-xylene in the cooked flesh was found in the LF group ($p < .05$). The *m*-xylene was identified with a phenolic odour in crayfish waste (Tanchotikul & Hsieh, 1989).

3.1.6 | Acids

There were two kinds of acids identified in the cooked fillets (Table 1). The differences in the levels of hexadecanoic acid and octadecanoic acid between these two farmed fish groups were not

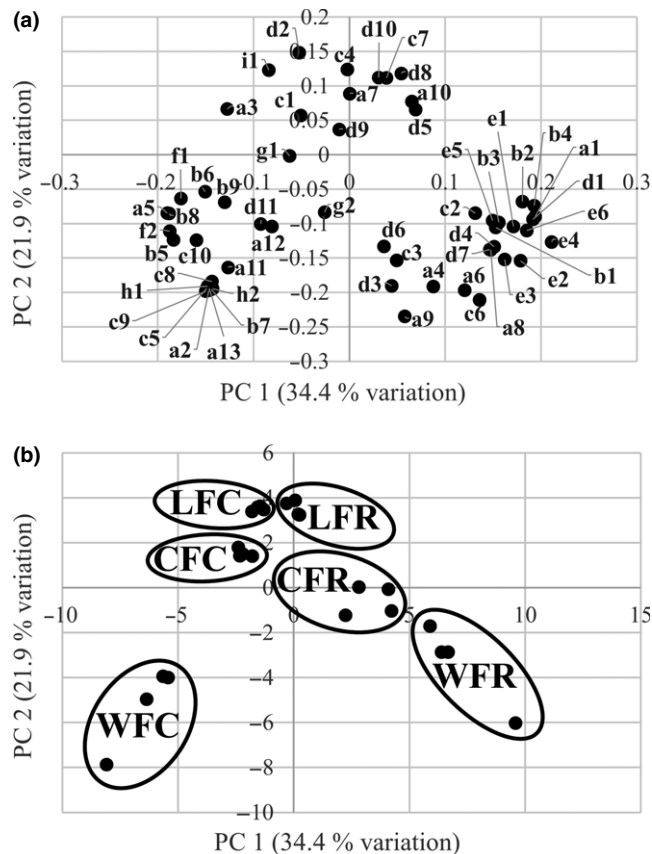


FIGURE 1 Loading plot (a) for the principal component analysis carried out with the volatile compounds (code as indicated in Table 1) in all raw (Mu et al., 2017) and cooked muscle samples of large yellow croaker, corresponding to the score plot in (b). LFR, raw muscle of fish fed the low fish meal diet. LFC, cooked muscle of fish fed the low fish meal diet. CFR, raw muscle of fish fed the control diet. CFC, cooked muscle of fish fed the control diet. WFR, raw muscle of wild fish. WFC, cooked muscle of the wild fish

significant ($p > .05$). Hexadecanoic acid was also found in cooked rainbow trout (*Oncorhynchus mykiss*) and described as the compound that provides fresh and fruity odour (Selli, Rannou, Prost, Robin & Serot, 2006).

3.1.7 | Esters

Two kinds of esters were detected in the cooked fillets of the two farmed LYC groups (Table 1). There were no significant differences in the amounts of butyl propanoate and diethyl phthalate in the cooked muscle between the LF and CF groups ($p > .05$). Most esters could provide major contribution to sweet and fruity aroma (Tanchotikul & Hsieh, 1989). Phthalates have been also determined in various food, such as turbot (*Psetta maxima*; Xu et al., 2014), scallops (*Patinopecten yessoensis*; Chung, Yung, Ma & Kim, 2002), heated beef and sheep fats (Watkins, Rose, Warner, Dunshea & Pethick, 2012). These compounds have been in worldwide production as plasticizers and, with their frequent use and application, have become ubiquitous in the environment (Xu, Liang &

Zhang, 2007). In this study, diethyl phthalate could be a contaminant from sea water, package, environment and so on. But there was significant difference in the content of muscle diethyl phthalate between the farmed and wild LYC groups. So, further studies are needed to explain this result.

3.1.8 | Furans

No furan was detected in the cooked muscle of farmed LYC (Table 1).

3.1.9 | Miscellaneous compounds

There was only one kind of miscellaneous compound identified in the cooked muscle of farmed LYC (Table 1). Compared with the CF group, the LF group had a significant higher concentration of 1-methylene-1*H*-indene in the cooked fillets ($p < .05$).

3.2 | Comparison of volatiles in cooked muscle between the farmed and wild LYC

3.2.1 | Aldehydes

In total, seven and 11 aldehydes were detected in the cooked fillets of farmed and wild LYC respectively (Table 1). In the cooked muscle of all the fish, 3-methylbutanal, undecanal, dodecanal and pentadecanal were detected only in the wild fish. The cooked fillets of wild fish had significantly higher amounts of 3-methylbutanal, hexanal, (*Z*)-4-heptenal, heptanal, octanal, nonanal, undecanal, dodecanal and pentadecanal than the farmed fish ($p < .05$). Heptanal and nonanal have been considered as being of great important for cooked rainbow trout aroma (Selli et al., 2006).

3.2.2 | Alcohols

Among the volatiles identified in the muscle of LYC, seven and eight alcohols were detected in the cooked fillets of farmed and wild fish respectively (Table 1). In the cooked muscle of all the fish, 1-octen-3-ol was detected only in the wild fish. It is known to be one of the major volatile alcohols in cooked muscle of Senegalese sole and bigeye tuna (Moreira et al., 2013; Sun et al., 2013). It has been suggested that 1-octen-3-ol could be generated from the oxidation of arachidonic acid by 12-lipoxygenase (Duflos, Coin, Cornu, Antinelli & Malle, 2006). In addition, the cooked muscle of wild fish had significantly higher levels of 1-hexanol, 1-heptanol, 1-octanol and undecanol, and lower amounts of 1-penten-3-ol and (*Z*)-2-penten-1-ol than the cooked fillets of the LF group ($p < .05$). Significantly lower concentration of 1-penten-3-ol was also found in muscle of wild barramundi than the farmed counterpart (Frank et al., 2009). However, there were no significant differences in the levels of these above six alcohols in cooked fillets between the CF and wild groups ($p > .05$).

3.2.3 | Ketones

There were three, four and six ketones identified in the cooked fillets of LF group, CF group and wild group respectively (Table 1). Acetophenone, 2-heptanone and 2-nonanone were detected only in the wild fish, while acetone was found only in the CF group. In addition, the cooked muscle of wild fish had a significantly higher content of 2-undecanone compared with the cooked fillets of the two farmed fish groups ($p < .05$; Table 1). Prost, Serot and Demaimay (1998) also concluded that 2-undecanone had a significantly higher flavour dilution factor in cooked wild turbot than in farmed one and it may positively contribute to wild turbot flavour. Compared with the wild LYC, significantly lower amounts of 2-hexanone and 2, 3-octanedione were observed in the cooked fillets of the LF group ($p < .05$). However, no significant differences were observed in the levels of 2-hexanone and 2, 3-octanedione in the cooked muscle between the CF and wild groups. Similar content of 2, 3-octanedione characterized savoury and cooked aroma also was detected in farmed and wild barramundi muscle (Frank et al., 2009).

3.2.4 | Hydrocarbons

Nine and eight hydrocarbons were detected in the cooked muscle of farmed and wild LYC respectively (Table 1). In the cooked fillets, decane was only detected in the two farmed groups. Moreover, the cooked muscle of wild fish had significantly lower levels of pentadecane and heptadecane, higher amounts of undecane, dodecane, tetradecane, hexadecane and 2, 6, 10, 14-tetramethylpentadecane than the cooked fillets of LF group ($p < .05$). However, there were no significant differences in the levels of these above seven hydrocarbons in the cooked fillets between the CF and wild groups ($p > .05$).

3.2.5 | Aromatics

There were six and five aromatics identified in the cooked muscle of farmed and wild LYC respectively (Table 1). The *m*-xylene was detected only in the farmed fish. The wild fish had significantly higher content of styrene than the LF group ($p < .05$), not the CF group. Compared with the wild, the concentrations of styrene and *m*-xylene were higher in farmed barramundi (Frank et al., 2009).

3.2.6 | Acids

There were two kinds of acid identified both in the farmed and wild LYC (Table 1). The cooked fillets of wild fish had significantly higher amounts of hexadecanoic acid and octadecanoic acid than the cooked muscle of LF group ($p < .05$), not the CF group ($p > .05$).

3.2.7 | Esters

Among the volatiles identified in the muscle of LYC, two esters were detected both in the cooked muscle of farmed and wild fish respectively (Table 1). The wild fish had a significantly higher content of

butyl propanoate ($p < .05$). Moreover, the concentration of diethyl phthalate in the cooked flesh was significantly higher in wild fish than that of the LF group ($p < .05$), not the CF group ($p > .05$).

3.2.8 | Furans

No furan was identified in the two farmed fish groups, while 2-ethylfuran and 2-pentylfuran were detected in the cooked muscle of wild LYC (Table 1).

3.2.9 | Miscellaneous compounds

Only 1-methylene-1*H*-indene was detected in the cooked muscle of farmed and wild LYC (Table 1). The content of 1-methylene-1*H*-indene was significantly higher in the LF group than the wild group ($p < .05$).

3.3 | Comparison of volatiles between the raw and cooked muscle

3.3.1 | Aldehydes

After cooking, the types of aldehydes identified in the LYC muscle dropped by 2 in farmed fish groups, while increased by 6 in wild fish compared with their raw fillets (Mu et al., 2017) respectively (Table 1). In regard to the raw and cooked muscle in farmed LYC, pentanal and (Z)-4-heptenal were detected only in the cooked muscle, while propanal, decanal, undecanal and dodecanal were identified just in the raw fillets. In regard to the raw and cooked muscle of wild fish, 3-methylbutanal, pentanal, (Z)-4-heptenal, benzaldehyde, undecanal, dodecanal and pentadecanal were detected only in the cooked muscle, while propanal was identified just in the raw fillets (Table 1). Sun et al. (2013) had the similar findings in bigeye tuna. The cooked fillets in LF group had significantly lower amounts of hexanal, benzaldehyde, decanal, undecanal and dodecanal, while higher levels of pentanal and (Z)-4-heptenal compared with its raw muscle ($p < .05$). In the muscle of CF group, the levels of pentanal and (Z)-4-heptenal significantly increased, while propanal, hexanal, decanal, undecanal and dodecanal significantly decreased, after cooking ($p < .05$). Meanwhile, cooking significantly increased the amounts of 3-methylbutanal, pentanal, (Z)-4-heptenal, benzaldehyde, undecanal, dodecanal and pentadecanal, and decreased the levels of propanal and octanal in the wild LYC fillets ($p < .05$). It is proved that Maillard and Strecker degradation reactions play crucial roles in formation of the meaty aroma of cooked seafood (Kawai, 1996). The formation of 3-methylbutanal and benzaldehyde could originate from Strecker degradation of leucine and phenylglycine respectively (Varlet, Prost & Serot, 2007; Silva, Valente, Castro-Cunha, Bacelar, and de Pinho, 2012). Cooking significantly increased the amounts of these two aldehydes only in the wild fish muscle (Table 1). The 3-methylbutanal has been described as imparting pungent, solvent, almond and nutty odour (Girard & Durance, 2000; Giri et al., 2010). Benzaldehyde has been regarded as a major flavour component in cooked grey mullet (Cayhan & Selli, 2010; Vejaphan, Hsieh &

Williams, 1988). It has been concluded that (Z)-4-heptenal could be formed from the retro-aldol condensation of 2, 6-nonadienal (Josephson & Lindsay, 1986). The formation of (Z)-4-heptenal is accelerated with increased temperatures. This aldehyde has been detected as an important aroma-active compound in cooked grey mullet, wild and farmed turbot (Cayhan & Selli, 2010; Prost et al., 1998). (Z)-4-heptenal was present at content higher than its odour threshold (0.8 µg/kg) (Frank et al., 2009) in all the cooked fillets (Table 1) and could be a main characteristic aroma component in cooked muscle of LYC. This aldehyde has been considered as having a boiled potato and cooked fish flavour (Hallier et al., 2005).

3.3.2 | Alcohols

After cooking, the types of alcohols identified in LYC muscle increased by 1 in the two farmed fish groups, increased by 4 in the wild fish compared with their raw fillets (Mu et al., 2017) respectively (Table 1). In regard to the raw and cooked muscle of farmed LYC, 1-hexanol was detected only in the cooked muscle. In regard to the raw and cooked muscle of wild LYC, 1-hexanol, 1-heptanol, 1-octen-3-ol, 1-octanol and undecanol were detected only in the cooked fillets, and 3-methyl-1-butanol was identified just in the raw flesh. The cooked fillets of LF group had significantly lower amounts of 1-pentanol and undecanol compared than its raw muscle ($p < .05$). Similar result was obtained in boiled scallop (*Patinopecten yessoensis*), which had a lower concentration of n-pentanol than that of the raw fillets (Suzuki, Ichimura & Etoh, 1990). In the muscle of CF group, the levels of 1-hexanol and 1-octanol significantly increased, while 1-penten-3-ol, (Z)-2-penten-1-ol and 1-pentanol significantly decreased after cooking ($p < .05$). Similarly, Sun et al. (2013) concluded that the relative amounts of 1-penten-3-ol and 1-octen-3-ol decreased rapidly in fillets heated to 100°C compared with raw bigeye tuna. In addition, cooking significantly increased the amounts of 1-hexanol, 1-heptanol, 1-octen-3-ol, 1-octanol and undecanol, while decreased the levels of 3-methyl-1-butanol, 1-penten-3-ol and (Z)-2-penten-1-ol in the wild LYC fillets ($p < .05$). The same is true in cooked Senegalese sole (Moreira et al., 2013).

3.3.3 | Ketones

After cooking, the types of ketones identified in the LYC muscle decreased by 2 in LF group, remained constant in CF group, while increased by 3 in wild group compared with their raw fillets (Mu et al., 2017) respectively (Table 1). In regard to the raw and cooked muscle in farmed LYC, 6-methyl-5-hepten-2-one was detected only in the raw fillets, while cyclohexanone was identified only in the raw flesh of LF group, and the acetone was detected just in the cooked muscle of CF group. In regard to the raw and cooked muscle of wild LYC, 2-heptanone, acetophenone, 2-nonanone and 2-undecanone were detected only in the cooked fillets, while 2, 3-pentanedione was identified just in the raw flesh. The cooked fillets of LF group had significantly lower amounts of cyclohexanone, 6-methyl-5-hepten-2-one and 2-undecanone, higher content of 2-hexanone

compared with its raw counterpart ($p < .05$). In the muscle of CF group, the levels of acetone significantly increased, while 2, 3-octanedione and 6-methyl-5-hepten-2-one significantly decreased after cooking ($p < .05$). Meanwhile, cooking significantly increased the amounts of 2-heptanone, acetophenone, 2-nonanone and 2-undecanone, while decreased the levels of 2, 3-pentanedione and 2, 3-octanedione in the wild fish fillets ($p < .05$). Bigeye tuna fillets heated to 100°C had higher relative concentrations of 2-heptanone and 2-undecanone, while lower relative content of 2-nonanone compared with raw muscle (Sun et al., 2013). In general, the methyl ketones have a distinct green and fruity odour, contributing more floral notes as chain length increases (Tanchotikul & Hsieh, 1989).

3.3.4 | Hydrocarbons

After cooking, the types of hydrocarbons identified in the LYC muscle decreased by 1 in farmed fish groups, remained constant in wild compared with their raw fillets (Mu et al., 2017) respectively (Table 1). Among the farmed LYC, decane was detected only in the cooked fillets, and hexane and 1-tridecene were identified just in the raw fillets. In regard to the wild LYC, hexadecane was detected only in the cooked muscle, while hexane was identified just in the raw muscle. Compared with the raw fillets, cooked fillets of all farmed LYC had significantly higher content of decane, while lower amounts of 1-tridecene, tetradecane, pentadecane and hexadecane ($p < .05$). Similar results showed that smoked rainbow trout had lower concentrations of some alkenes (1-pentadecene, heptadecene) and alkanes (pentadecane and hexadecane) than that of the raw flesh (Guillén & Errecalde, 2002). In LF group, the levels of undecane and tridecane were significantly higher, while heptadecane and 2, 6, 10, 14-tetramethyl-pentadecane were significantly lower in the cooked fillets than that of the raw muscle ($p < .05$). Meanwhile, cooking significantly decreased the content of hexane in the fillets of CF group ($p < .05$). In the wild fish muscle, the levels of hexadecane and heptadecane significantly increased, while hexane significantly decreased, after cooking ($p < .05$) (Table 1). Similarly, Guillén and Errecalde (2002) concluded that smoked black bream had a higher content of heptadecane compared with raw flesh.

3.3.5 | Aromatics

After cooking, the types of aromatics identified in the LYC muscle remained unchanged in farmed fish groups, but decreased by 1 in wild fish compared with their raw fillets (Mu et al., 2017) respectively (Table 1). The levels of styrene, *p*-xylene and *m*-xylene in all the farmed fish muscle significantly decreased after cooking ($p < .05$). In addition, the cooked muscle of CF group had a significantly lower content of toluene than its raw fillets ($p < .05$). In regard to the wild LYC, *m*-xylene was detected only in the raw fillets. Cooking significantly decreased the amounts of all the aromatics in the wild fish fillets ($p < .05$). However, cooking had no significant effects on the levels of ethylbenzene and *o*-xylene in all the farmed fish muscle, and toluene in the LF group ($p > .05$).

3.3.6 | Acids

After cooking, the types of acids detected in the LYC muscle increased by 1 in farmed fish groups and increased by 2 in wild fish compared with their raw fillets (Mu et al., 2017) respectively (Table 1). Among the farmed LYC, octadecanoic acid was identified only in the cooked fillets. Similarly, Sun et al. (2013) reported that some acids mainly presented in heated bigeye tuna meats (100 and 150°C). In regard to the wild LYC, hexadecanoic acid and octadecanoic acid were all newly detected in the cooked fillets (Table 1). Cooking significantly increased the amounts of hexadecanoic acid and octadecanoic acid in all the fish fillets ($p < .05$). Similar results showed that a great number of acids in higher levels presented in smoked muscle of black bream than in raw fillets (Guillén & Errecalde, 2002). The higher temperature could contribute to the formation of these acids in different studies.

3.3.7 | Esters

After cooking, the types of esters identified in the LYC muscle remained unchanged in farmed fish groups, while increased by 1 in wild fish compared with their raw fillets (Mu et al., 2017) respectively (Table 1). The levels of butyl propanoate and diethyl phthalate in the fillets of LF group significantly decreased after cooking ($p < .05$). In the cooked fish fed the CF diet, the content of diethyl phthalate was significantly lower compared with its raw muscle ($p < .05$). In regard to the wild LYC, butyl propanoate was detected only in the cooked muscle. Cooking significantly increased the amounts of butyl propanoate and diethyl phthalate in the wild fish flesh ($p < .05$) (Table 1). Similarly, the concentration of ethyl octanoate was significantly higher in cooked Senegalese sole than that of the raw flesh (Moreira et al., 2013). Acetic acid, cyclohexyl ester detected in bigeye tuna tend to mainly present in the heated muscle (100 and 150°C) (Sun et al., 2013).

3.3.8 | Furans

After cooking, the types of furans detected in the LYC muscle remained zero in farmed fish groups, while increased by two in wild fish compared with their raw fillets (Mu et al., 2017) respectively (Table 1). The 2-ethylfuran and 2-pentylfuran were identified only in the cooked muscle of wild fish. Cooking significantly increased the levels of these two furans in the wild fish fillets ($p < .05$) (Table 1). Similar results also indicated that 100°C heat-treated fillets had higher concentrations of 2-ethylfuran and 2-pentylfuran compared with raw bigeye tuna meat (Sun et al., 2013).

3.3.9 | Miscellaneous compounds

After cooking, only 1-methylene-1H-indene was also identified in the muscle of farmed and wild LYC (Table 1). Its content significantly increased in the fillets of LF group after cooking ($p < .05$).

3.4 | Principal component analysis

A PCA was used for a simplified view of the relationship among the volatile profiles identified in the raw (Mu et al., 2017) and cooked muscle of LYC. Three factors explained 74.2% of total variance. The representation of the variables and samples applying the first two PCs is shown in Figure 1a, b. The first principal component (PC1), which explained 34.4% of total variance, was positively correlated to *p*-xylene, propanal, etc., and negatively related to (Z)-4-heptenal, 1-octanol, etc. The second principal component (PC2), which accounted for 21.9% of total variance, was positively related to decane, cyclohexanone, etc., and negatively related to nonanal, 2,3-octanedione, etc.

In the PC score plot (Figure 1b), there were six separate groups of points, corresponding to different muscle samples of LYC. Obviously, PC1 separated the raw and cooked fillets of all the fish. Similar results also showed that cooked muscle of Senegalese sole tends to separate of raw flesh by PC1 (Moreira et al., 2013). There were 11 compounds (propanal, *p*-xylene, etc.) whose percentage accounted by PC1 was >60.0% in Table 3. These volatiles contributed greatly to the differences in the characteristics of volatile profiles between the raw and cooked fillets of LYC and could be considered as sensitive indicators which could differentiate the raw and cooked muscle in the present study. Figure 1b shows that PC2 discriminated the samples of three different fish groups. On the PC2 part, the sequence of both the raw and cooked fillets of three groups from top to bottom was LF group, CF group and wild group respectively. The raw and cooked muscles of LF group were placed in the positive PC2. It means that they were similar and presented a high levels in volatiles positively related to this axis. The raw and cooked fillets of wild fish were positioned in the negative PC2, showing low amounts of volatiles positively related to this axis and high levels of compounds negatively related to this axis. However, the raw and cooked fillets of CF group tended to situate in the middle of the plane. Consequently, muscle volatiles of wild fish were separated from farmed fish in Figure 1b, showing that they had different volatile profiles in the raw and cooked fillets. However, it is obvious that compared with LF group, both the raw and cooked fillets of CF group were located closer to the wild group in the score plot (Figure 1b). It is indicated that lower dietary FM content could affect the LYC muscle volatiles. There were 11 compounds (nonanal, 2, 3-octanedione, etc.) whose percentage accounted by PC2 was greater than 45.0% in Table 3. These volatiles contributed greatly to the differences in the characteristics of muscle volatile profiles of those three fish groups and could be considered as sensitive markers which could differentiate the muscle volatiles of different fish groups in the present study.

Based on the above results, a new PCA model for each two samples of the raw or cooked muscle was separately conducted to discover sensitive volatiles which could distinguish the raw or cooked muscle of every two fish groups in the present study. Figure 2 shows the score plots after PCA of different variables in the cooked muscle of each two fish groups separately by PC1 and PC2. The

TABLE 3 Results of the principal component analysis on the volatile compositions in different muscle samples of large yellow croaker showing the most important loadings and the percentage variance accounted for by the first two principal components (PC1 and PC2)

All samples ^a (PC1)		All samples ^a (PC2)	
Compound code (11) ^b	R _x ² (>60%)	Compound code (11) ^b	R _x ² (>45%)
a1	0.745	a9	0.683
e4	0.736	c6	0.504
d1	0.728	c9	0.486
a5	0.715	a2	0.481
b8	0.702	a6	0.481
f2	0.696	b7	0.474
e6	0.680	h2	0.469
b5	0.668	c5	0.468
b2	0.647	a13	0.465
b4	0.616	a4	0.461
f1	0.612	h1	0.453
LFC versus CFC (PC1)		LFC versus WFC (PC1)	
Compound code (11) ^b	R _x ² (>70%)	Compound code (31) ^b	R _x ² (>70%)
d7	0.927	c9	0.981
c1	0.924	a4	0.967
c3	0.893	a6	0.963
a3	0.836	c5	0.951
d2	0.815	a11	0.942
b9	0.812	b8	0.940
d4	0.765	h1	0.934
d3	0.759	a2	0.931
b8	0.749	b6	0.929
b6	0.724	a5	0.918
a9	0.719	d3	0.914
		b7	0.912
		g1	0.911
		a13	0.904
CFC versus WFC (PC1)		h2	0.892
Compound code (15) ^b	R _x ² (>70%)	g2	0.869
c9	0.967	d9	0.858
a11	0.951	d2	0.856
a2	0.950	a9	0.844
b7	0.942	a8	0.828
c5	0.903	c10	0.826
a6	0.903	b5	0.825
a13	0.890	d4	0.815
c1	0.858	d11	0.812
a9	0.832	c8	0.810
a8	0.831	c3	0.783

(Continues)

c8	0.752	f2	0.783
a4	0.738	e6	0.781
a5	0.726	a7	0.757
b5	0.722	a12	0.753
a12	0.706	a3	0.736

^aAll samples refer to all the raw (Mu et al., 2017) and cooked muscle of large yellow croaker.

^bCompound code as indicated in Table 1. R_x², variance explained. LFC, cooked muscle of fish fed the low fish meal diet. CFC, cooked muscle of fish fed the control diet. WFC, cooked muscle of the wild fish.

most important loadings and the percentage accounted by PC1 and PC2 after PCA are shown in Table 3.

The score plot of volatiles in the cooked muscle of two farmed LYC groups, explaining 66.1% of the total data variance by the first two principal components (PCs) of the PCA, is given in Figure 2a. PC1 accounted for 51.9% of total variability, whereas PC2 explained 14.2%. PC1 separated the cooked fillets of two farmed fish groups. There were 11 compounds (tetradecane, acetone, etc.) whose percentage accounted by PC1 greater than 70.0% in Table 3. These volatiles contributed greatly to the differences in the characteristics of volatiles in the cooked muscle of two farmed fish groups and could be regarded as sensitive markers. It is generally accepted that factors influencing the lipid composition of fish fillets may determine its volatile profiles. The lipid composition has been suggested as being affected by fish species, origin, cultured conditions (diet compositions, farming conditions, etc.) among many other factors (Frank et al., 2009; Grigorakis et al., 2003; Hallier et al., 2005; Timm-Heinrich, Eymard, Baron, Nielsen & Jacobsen, 2013). It is concluded that the high levels of dietary plant protein sources affected the muscle volatile composition of Senegalese sole (Moreira et al., 2014), fatty acid compositions in diets and fillets of rainbow trout (Timm-Heinrich et al., 2013). So the changes in muscle volatile composition of LYC upon the decrease in dietary FM could be due to changes in fatty acid compositions of the diets in the present study. However, it is regrettable that there are no data on the fatty acid compositions of the diets and fish muscle in the present study. But according to the formulation of the present experimental diets (Mu et al., 2017), it is found that compared with the control diet (CF, 44% FM), the content of FM decreased about 20%, while the level of fish oil and soya bean meal increased 1.5% and 25%, respectively, in the low dietary FM diet (LF, 25% FM). The crude lipid content of soya bean meal is very low (about 1.5%). So the differences in the fatty acid compositions between these two diets could be minor. However, it is found that the specific growth rate of the LYC in LF group was lower than that in the CF group (unpublished data). Therefore, the changes in growth and physiological responses of LYC upon the decrease in dietary FM could result in the changes in lipid metabolism and the fatty acid retention in muscle. These could further affect the muscle volatiles. Dietary FM replacement by plant protein sources has been suggested to influence fatty acid retention and lipid metabolism in Atlantic salmon (*Salmo salar* L.) (Pratoomyot, Bendiksen, Bell

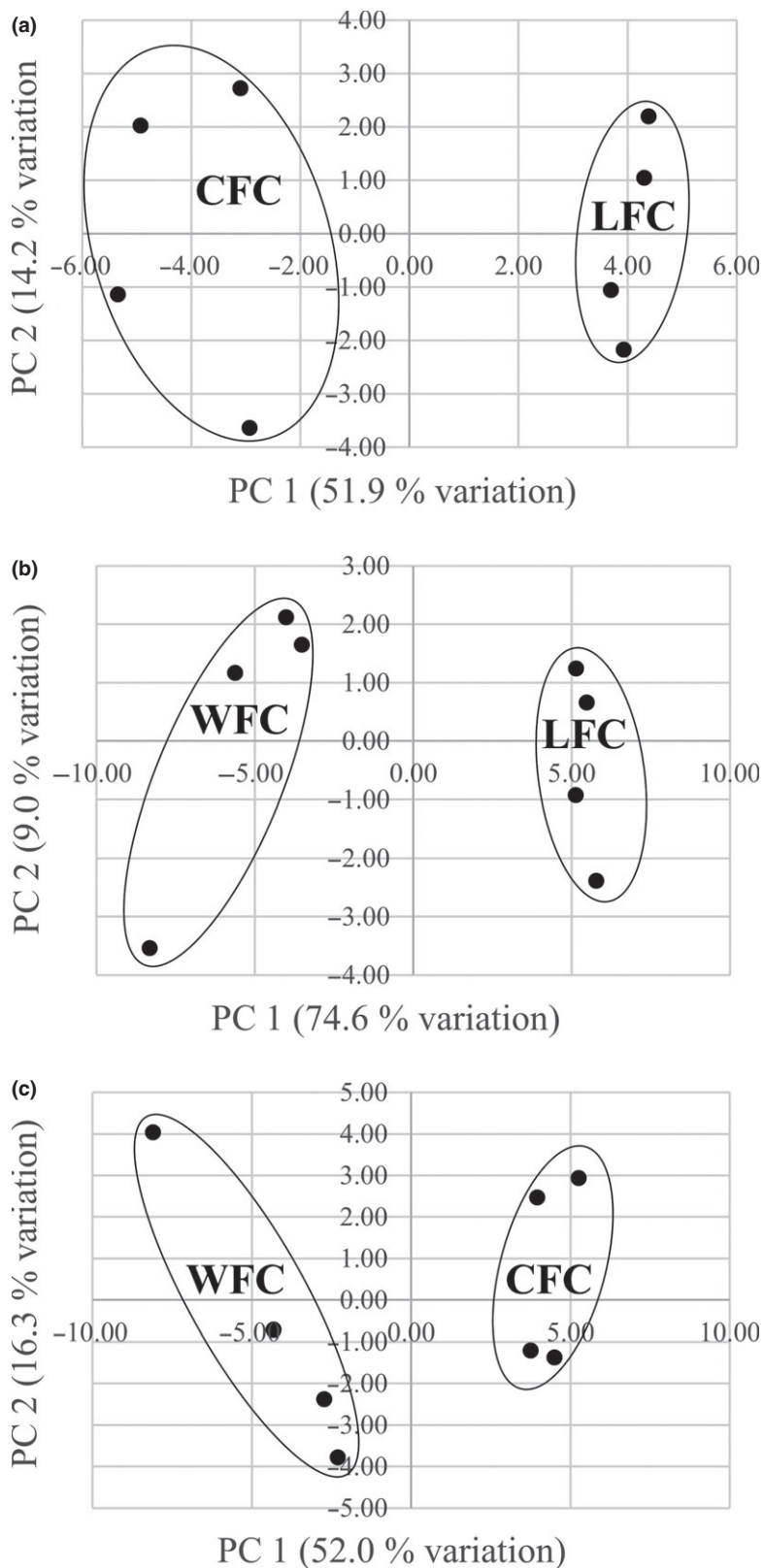


FIGURE 2 Score plots for the principal component analysis carried out with the volatile compounds in cooked muscle of large yellow croaker fed the low fish meal diet and control diet (a), in cooked muscle of large yellow croaker fed the low fish meal diet and wild fish (b), in cooked muscle of large yellow croaker fed the control diet and wild fish (c) respectively. LFC, cooked muscle of fish fed the low fish meal diet. CFC, cooked muscle of fish fed the control diet. WFC, cooked muscle of the wild fish

& Tocher, 2010) and Senegalese sole (Moreira et al., 2014). Further studies are necessary to make it clear in LYC.

The score plot of volatiles in the cooked muscle of LYC fed the LF diet and wild fish group is given in Figure 2b. The first two PCs of the PCA accounted for 83.6% of the total data variability. PC1

explained 74.6% of total variance, whereas PC2 accounted for 9.0%. Figure 2b shows that PC1 discriminates the cooked fillets of those two fish groups. There were 31 volatiles (2-nonanone, hexanal, etc.) whose percentage accounted by PC1 greater than 70.0% in Table 3. These substances contributed greatly to the differences in the

characteristics of volatiles in the cooked muscle of those two fish groups.

Figure 2c indicates the score plot of volatiles in the cooked muscle of LYC fed the CF diet and wild fish group. The first two PCs of the PCA explained 68.3% of the total variance. PC1 explained 52.0% of data variability, whereas PC2 accounted for 16.3%. It is obvious that PC1 differentiates the cooked fillets of these two fish groups (Figure 2c). In total, 15 volatiles (2-nonanone, undecanal, etc.) whose percentage explained by PC1 >70% were found (Table 3). These compounds contributed greatly to the differences in the characteristics of volatile profiles in the cooked muscle of those two fish groups and could be regarded as sensitive markers.

4 | CONCLUSION

In conclusion, 48 volatiles were detected in the cooked muscle of LYC. These volatiles were divided into nine groups, including 11 aldehydes, eight alcohols, seven ketones, nine hydrocarbons, six aromatics, two acids, two esters, two furans and one miscellaneous compound. The cooked muscle of LF group had significantly lower amounts of total aldehydes and ketones, and significantly higher content of miscellaneous compound than CF and wild groups ($p < .05$). The CF group had significantly lower content of total aldehydes, and significantly higher amount of total ketones in the cooked muscle than the wild group ($p < .05$). According to the PCA, some volatiles could be considered as sensitive indicators to classify muscle samples in the present study.

ACKNOWLEDGMENTS

This research was financially supported by grants from the Fundamental Research Funds for the Central Universities of Ocean University of China (No. 201562017), the National Natural Science Foundation of China (No. 31372542) and the Academician Workstation for Aquatic Animal Nutrition and Feed in Guangdong Evergreen (2014B090905014).

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How to cite this article: Mu H, Wei Z, Yi L, et al. Dietary fishmeal levels affect the volatile compounds in cooked muscle of farmed large yellow croaker *Larimichthys crocea*. *Aquac Res.* 2017;00:1–14. <https://doi.org/10.1111/are.13405>