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**ABSTRACT**

Extra virgin olive oil from Calabria Region (South Italy), pomace olive oil, palm oil and soybean oil were heated at 180 and 220°C for 30, 60 and 120 mins. Emission of volatile organic compounds produced during the heat treatment was evaluated by the solid phase micro-extraction of the head space technique and analysed by gas chromatography with mass spectrometric detector. The results after oil heating showed a significant variation in the volatile composition with temperature and time increasing. Twenty-five compounds have been identified, and alkanals, alkenals, and alkadienals were the most represented classes. Results from volatile profile evolution suggest that specific aldehyde compounds can be used as markers both for the evaluation of the organoleptic properties and for the health characteristics of the studied oils.

**Keywords**

Deep-fried oil, volatile organic compounds, SPME, GC-MS.

## 1. Introduction

One of the many uses of an edible vegetable oil is in deep-frying processes in food preparation. Deep-frying is one of the oldest cooking methods (Jaarin & Kamisah, 2012) which give a characteristic taste to the food. It is highly valued by the consumers and easy and fast to do. When a vegetable oil is heated, changes in physical appearance occur such as darkening in colour and increasing in viscosity due to the alteration of the fatty acid composition (Rani et al., 2010). Heating causes some chemical reactions in the oil such as oxidation, hydrolysis and polymerization (Choe & Min, 2007). During these processes, many oxidative products such as hydroperoxides and aldehydes are produced, which can be absorbed into the fried food (Choe & Min, 2006).

In Italy, one of the main agricultural crops is represented by the olive tree, mainly for extra virgin olive oil (EVOO) production. The flavour in an olive oil is very important and it was widely studied (Vichi et al., 2003; Cavalli et al., 2004; Baccouri et al., 2007; Baccouri et al., 2008;). It is characterised by a large number of carbonyl substances, which includes linear saturated and unsaturated aldehydes and alcohols, esters and hydrocarbons (Flath et al., 1973; Esposto et al., 2009;). This composition can be affected by geographical origin (Ouni et al., 2011), by ripening degree (Konavouras et al., 2005) and by extractive processing method (Dabbou et al., 2012). Olive pomace is the most important by-product in the olive oil extraction industry, in particular when the three-phase technology is used (Nasopoulou et al., 2011). The three phases system (oil, olive pomace and olive mill waste water) produces an olive pomace with a low moisture content which facilitates the pomace olive oil (PO) solvent extraction by *n*-hexane  (Ciftci et al., 2009; Mele et al., 2018). Palm oil (P) and soybean oil (SO) are rich in antioxidants such as tocopherols and tocotrienols (Kamisah et al., 2005; Clemente & Cahoon, 2009) and are the most commonly used vegetable oils in the household and industrial deep frying purposes. Each oil produces different volatile emissions in different cooking methods (Peng et al., 2017). Many studies were conducted on these and others vegetable oils about the variation in the chemical composition during heating, in particular on its oxidative stability (Saad et al., 2007; Maggio et al., 2011; Jaarin & Kamisah, 2012; Poyato et al., 2014; Qing et al., 2016; Giuffrè et al., 2017a; Giuffrè et al., 2017b; Giuffrè et al., 2018). A minor number of studies exist on their volatile profile after heating (Katragadda et al., 2010; Peng et al., 2017).

The most rapid and accurate technique to study the volatile fraction of an oil is the SPME (Song et al., 1997) introduced since 90s for the aroma analysis of various food products (Arthur & Pawliszyn, 1990). It is based both on the capacity of the silica fibre to absorb the analytes released in head space from the sample and to desorb the same molecules in an instrument, such as gas chromatography with mass spectrometric detector (GC-MS).

VOCs produced by cooking were also found to be an indoor pollutant both in household and in food factory, in particular if the ventilation is not enough (Jones, 1999). Outdoor cooking is a problem in volatile organic compounds (VOCs) emission even if lower than for indoor cooking (Edwards et al., 2017). Studies on Chinese women have demonstrated that the exposure to fumes of cooked oils is associated with the lung cancer (Zhao et al. 2018). For this reason is important to know the emission composition in VOCs during oil frying.

The aim of this work was to study the effect of temperature and heating duration on the volatile organic compound (VOC) composition of extra virgin olive oil, olive pomace oil, soybean oil and palm oil.

## 2. Materials and method

### 2.1. Materials

EVOO and PO were produced in the Calabrian region (South of Italy) in the harvest year 2016-2017. SO and P were purchased in a supermarket EVOO was obtained by a mechanical extraction with a three phase apparatus. PO, SO and P were obtained by solvent (*n*-hexane) extraction.

Nine 1L bottles of the each oil were randomly taken for analysis. After this, three representative batches each one composed by the mixed content of nine bottles were prepared for each oil to analyse VOCs in triplicate. At this point, from each batch were prepared 7 glass pyrex beakers, containing 150 g oil each one; 6 out of 7 contained the oil to be heated for the six different treatments: 180 °C (30, 60 and 120 min), 220°C (30, 60 and 120 min) and 1 out of 7 contained the oil to be analysed as a fresh oil (control). Beakers containing oils were placed on a heating plate and temperature was adjusted and controlled by a thermometer.

### 2.2. Solid phase micro-extraction of the head space (HS-SPME)

The most rapid and accurate technique to study the volatile fraction of an oil is the HS-SPME (Song et al., 1997) introduced since 90s for the aroma analysis of various food products (Arthur & Pawliszyn, 1990). A SPME fibre coated with poly-dimethylsiloxane (PDMS, 100 µm) from Supelco (Bellafonte, PA, USA) was used to absorb the VOC's with the procedure described by Temime et al. (2006) modified as follow. A 10 g aliquot of each heated and unheated (fresh) oil was placed in a 20 ml glass vial with septum cup and provided with a magnetic stirrer. The fibre was conditioned before using at 270°C for 30 mins as recommended by the manufacturer. Sample was placed in a thermostatic bath at 45°C in stirring (500 rpm). The fibre was inserted into the vial's HS. Absorption was stopped after 20 mins and fibre was thermally desorbed for 5 mins in a gas chromatographer in split-less mode. All SPME operations were conducted manually.

### 2.3. GC-MS analysis

A GC Thermo Trace 1310 apparatus (Waltham, MA, USA) equipped with Single Quadrupole Mass Spectrometer ISQ LT system and a fused-silica capillary columns (30 m length, 0.25 mm i.d., 0.25 µm film thickness, Thermo Scientific, Whaltam, MA, USA), TG-5MS 5% phenyl phase was used for GC-MS analysis. The oven temperature was programmed at 60°C held for 10 mins, then from 60°C to 220°C at 5°C/mins, and held isothermally at 220°C for 10 mins. Injector, MS transfer line and ion source temperatures were respectively 250, 270 and 260°C. Mass range was from 45 to 500 atomic mass unit (amu). Gas carrier was helium at 1.5 mL/mins flow rate. The tentative identification of the VOCs was based on the comparison of spectra with those of NIST/EPA/NIH Mass spectral library Version 2.0.

### 2.4. Statistical analyses

Each sample was analysed in triplicate and volatile profiles aroma data were submitted to analysis of variance (ANOVA) using SPSS 20.0 software for Windows (SPSS Inc., Chicago, IL, U.S.A.). Wherever F values were significant, Tukey's test was used to separate the mean effects (different temperatures for each oil type). Significance was defined at $P < 0.001$. Graphics were done using Excel 2010 software for Windows (Microsoft Corp., Redmond, WA, USA).

## 3. Results and discussion

### 3.1. Volatile organic compounds profile

The highest number of compounds, i.e., twenty-one was found in EVOO (Table 1). A similar result was obtained both by Temime et al. (2006) in Tunisian extra virgin olive oils, in which were found twenty-five compounds and by Cavalli et al. (2004) in nine virgin olive oils from France and Spain, in which were found a number of VOCs ranging between 23 and 25. A lower number of compounds (twenty) was found in three Algerian EVOOs (Cherfaoui et al., 2018). It has to be pointed out that other authors, in different cultivars, have identified a greater number of compounds with the same technique (Hammouda et al., 2017) or with a different technique (Lukić et al., 2019).

The E-2-hexenal has been found to be the most represented constituent (28.3% in EVOO) (Table 1), in fact it is considered as a quality fresh-marker of an olive oil (Cavalli et al., 2004), it gives the characteristic sensory properties of sweet, fragrant, almond, green and leafy to the olive oil (Kanavouras et al., 2005). During heating a very high significant decrease in E-2-hexenal content was observed to the lowest value (1.7%) in the EVOO 220°C-120 min sample. The same decreasing trend in E-2-hexenal was found in almost all the studied oils with the exception of PO 180°C-30 min (Tables 1-4). The higher the heating temperature and the longer the heating duration, the lower the E-2-hexenal content. Other compounds characterising the volatile profile of EVOO were α-farnesene (15.3%), α-cubebene (14.4%) and nonanal (8.9%) (Table 1). They are characteristic for an EVOO composition and it was demonstrated to be affected by the olive cultivar (Reboredo-Rodríguez et al., 2012) and the olive growing area (Temime et al., 2006). Some aldehyde compounds such as Z-2-heptanal, E-2-octenal, 2-nonenal and 2-decenal (Z), which were not in EVOO, were formed during heating, due to oleic acid oxidation (Belitz et al., 2009), and could be used as indicators of the oil heating treatment. The Z-2-decenal alkenal was not detected in EVOO, however it was found in high quantity in all the heated samples, and increased constantly with heating to 16.3% in EVOO 180°C-30 min and to 30.8% in EVOO 220°C-30 min (Table 1). Other minor compounds were not in fresh EVOO but they were identified after heating: butyl-cyclopentane (found in EVOO heated at 220°C), Z-2-heptenal, 1-heptanol, 1-octen-3-ol (in EVOO 220°C-60 min), 5-hepten-2-one, 6-methyl- (2.0 and 2.2% in both EVOO heated at 180°C-60 min and 180°C-120 min), E-2-octenal (2.5 and 2.6% in EVOO 180°C-120 min and 220°C-120min respectively), 1-octanol (in highest quantity, 1.6%, in EVOO 220°C-60 min and in EVOO 220°C-120 min), 2-nonenal which was mainly produced at 220°C after 120 min heating. This was due to the oxidation mechanism of the oil, accelerated by the heating (Fullana et al., 2004; Lin & Liou, 2000), that causes a chemical primary oxidation and the consequent production of hydroperoxides (Choe & Min, 2006), and its successive degradation to different class of compounds (aldehyde, ketons, alcohols, etc.).

Aldehydes can possess toxic properties, in particular, many studies have been conducted on E,E-2,4 decadienal (Chang et al., 2005; Chang & Lin, 2008; Halvorsen & Blomhoff, 2011), considered either carcinogens or suspected carcinogens. It was categorized into group 2A by the International Agency of Research on Cancer, which estimated a daily limit in human diet to 5 mg/kg of body weight (Conklin et al., 2010; Wang et al., 2008).

The major constituent detected in the PO volatile fraction was decanal (42.54%), followed by E-2-hexenal (16.01%), 2-undecenal (15.70%) and nonanal (12.43%) (Table 2). In PO there was a smaller decrease in E-2-hexenal content than in EVOO, which was lowest (7.76%) in PO180°C-120 min. Interesting was the highly significant decrease in decanal in all the heated samples, which achieved a value of 2.24% in PO 220°C-120 min and which can be taken into account as the oil temperature stress index parameter. A similar conclusion, but in reverse, can be done for Z-2-decenal passed from 5.02% in PO to 21.67% in PO 220°C-120 min.

In P were found seven volatiles compounds: the highest was nonanal (32.75%) and the lowest was Z-2-decenal (6.82%) (table 4). In P heated samples, heptanal, octanal and decanal showed a highly significant decrease. These compounds passed respectively from 14.41%, 13.12% and 10.84% to 1.37% (P 180°C-120 min), 1.92% (P 220°C-60 min and P 220°C-120 min) and 1.47% (P 220°C-60 min). Nine new compounds were formed during P heating. The E,E-2,4-decadienal was the most represented in all the heated samples, mainly in P180°C-120 min (32.35%), followed by Z-2-heptenal in P 180°C-30 min (8.68%) and E,E-2,4-decadienal in P 180°C-120 min (6.44%), (Table 3). The formation of these compounds is due to the oxidation after heating of the unsaturated fatty acids: the higher the unsaturated fatty acid content, the higher the aldehydes formation (Peng et al., 2017). This because poly-unsaturated fatty acids such as linoleic and linolenic acids are more susceptible to oxidation because of presence of multi double-bonds (Schauer et al., 2002). In fact, SO, considered a linolenic acid-rich oil showed higher concentration in the aldehyde compounds, with E,E-2,4-dodecadienal found as the highest (58.73%) in SO 180°C-120 min (Table 4). Nonanal (34.97%) and decanal (34.19%) were the most represented VOCs in fresh SO. Decanal constantly decreased with heating and with heating duration, whereas nonanal showed an initial increase at 180°C-30 min and a subsequent decrease with heating and with heating duration (Table 4). Furthermore, fresh SO was the one with the lowest number of identified compounds, moreover it showed a greater number of VOCs in the heated samples than in the fresh ones, confirming the high instability of this vegetable oil under heating (Table 4). The SO instability was also confirmed by the lowest total phenolic content (15 mg gallic acid/kg) and by the lowest antioxidant activity quantified with both the lowest values in the ABTS assay (51.8 μM TE/100g) and the lowest values in the DPPH hydrophilic assay (6.6 μM TE/100g) measured in the same samples studied by Giuffrè et al., (2018).

### 3.2. Total alkanals, alkenals and alkadienal

The identified aldehyde compounds have been grouped as alkanals, alkenals and alkadienals to study the relationship between the heating temperatures, the heating time and their production in the studied vegetable oils.

The total aldehydes rate varied significantly during heating of all of the four studied oils both at 180°C and 220°C (Figure 1a,b). This was due to the presence of some alkans (α-cubebene, α-muurolene and α-farnesene) (Table 1) in the EVOO, the only oil obtained from a mechanical process which minimally alters the olive original volatile profile, while they are totally absent in all the other three studied unheated vegetable oils obtained by solvent extraction. In figures 1a and 1b a significant increment is shown by the total alkenals, which increased from 32.1% in EVOO to 60.9% and 68.8% respectively in EVOO 180°C-120 min and EVOO 220°C-120 min, confirming how the formation of these compounds is strictly correlated to the oleic acid oxidation, the major acid constituent in olive oil.

The PO heated at 180°C showed a significant difference in the total aldehydes with respect to PO control with a decrease at 180°C-30 min and a tendency increase at 180°C-60 min and 180°C-120 min (Figure 2a), this was probably due to an initial degradation of some aldehydes and a subsequent formation of new ones, while the 220°C heated samples showed a significant difference after 30 mins heating with a decrement in total alkanals (-57.1%) and an increment in total alkenals (+58.6%) after 120 mins heating (Figure 2b).

The total aldehyde profile of P showed a similar trend in both heating temperature, with a significant increment in total alkadienal compounds, from 0% in P to 38.79% and 25.40% respectively in P 180°C-120 min and P 220°C-120 min, and a decrement in total alkanal compound, from 71.13% in P to 29.81% and 28.16% respectively in P 180°C-120 min and P 220°C-120 min (Figure 3a,b). Total alkanals and total alkenals of P varied significantly with heating and with heating duration (Figure 3a,b).

The total aldehydes in SO showed a trend similar to P, in fact total alkanals decreased when total alkadienal increased during heating time at each heating temperature (figure 4a,b). A very significant difference was found in the sample before heating and after heating at 180°C for 60 mins, with a decrement in alkanals (-80.0%) and an increment in alkadienal of +867.2%; while the sample heated at 220°C showed the highly difference from its unheated oil immediately after 30 mins, -90.1% and +916.2% respectively in total alkanals and total alkadienals.

## 4. Conclusion

Results showed that temperature and heating duration have a very high significant influence in the volatile organic compounds content of edible vegetable oils. EVOO showed the highest number of components with E-2-hexenal as the highest in quantity in fresh oil and Z-2-decenal and 2-undecanal as the highest in quantity in heated extra virgin olive oils. PO (control) contained decanal in higher quantity and after heating Z-2-decenal and 2-undecanal prevailed as for extra virgin olive oil in addition to nonanal.

Nonanal prevailed in both fresh P and SO whereas E,E-2,4-dodecadienal prevailed in both P and SO oils after their heating. Heating completely varied the volatile organic compounds content of four of the edible vegetable oils mostly used in the world. By the results of this study, the oil user can decide the temperature and the heating duration to be applied to preserve flavours and to reduce off-flavours which are produced during heating in the EVOO, PO, P and SO.

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**References**

Arthur, C.L., & Pawliszyn, J. (1990). Solid phase microextraction with thermal desorption using fused silica optical fibres. *Analytical Chemistry,* 62, 2145–2148.

Baccouri, B., Temime, S., Campeol, E., Cioni, P., Daoud, D. & Zarrouk, M. (2007). Application of solid phase microextraction to the analysis of volatile compounds in virgin olive oils from five new cultivars. *Food Chemistry*, 102, 850-856.

Baccouri, O., Guerfel, M., Baccouri, B., Carretani, L., Bendini, A., Lercker, G., Zarrouk M., & Daoud Ben Miled D. (2008). Chemical composition and oxidative stability of Tunisian monovarietal virgin olive oils with regard to fruit ripening. *Food Chemistry ,* 15, 743–754.

Bardhan, J., Chakraborty, R., & Raychaudhuri, U. (2011). The 21st century form of vitamin E - Tocotrienol. *Current Pharmaceutical Design,* 17, 2196–2205.

Belitz, H.D., Grosch, W., & Schieberle, P. (2009). Food Chemistry. (4th ed.). *Lipids.* (pp. 158–247). Berlin: Springer.

ben Hammouda, I., Freitas, F., Ammar, S., Gomes Da Silva, M.D.S., & Bouaziz, M. (2017). Comparison and characterization of volatile compounds as markers of oils stability during frying by HS–SPME-GC/MS and Chemometric analysis. *Journal of Chromatography B,* 1068–1069, 322–334.

Cavalli, J.F., Fernandez, X., Lizzani-Cuvelier, L., & Loiseau, A.M. (2004). Characterization of volatile compounds oh French and Spanish virgin olive oils by HS-SPME: Identification of quality-freshness markers. *Food Chemistry,* 88, 151–157.

Chang, L.W., Lo, W.S., & Lin, P. (2005). Trans trans-2,4-decadienal, a product found in cooking oil fumes, induces cell proliferation and cytokine production due to reactive oxygen species in human bronchial epithelial cells, *Toxicological Sciences,* 87, 337–343.

Chang, Y.C., & Lin, P. (2008). Trans,trans-2,4-decadienal induced cell proliferation via p27 pathway in human bronchial epithelial cells. *Toxicology and Applied Pharmacology,* 228, 76–83.

Cherfaoui, M., Cecchi, T., Keciri, S., & Boudriche, L. Volatile compounds of Algerian extra-virgin olive oils: Effects of cultivar and ripening stage. *International Journal of Food Properties*, 21, 36-49.

Choe, E., & Min, D.B. (2006). Chemistry and reactions of reactive oxygen species in foods. *Critical Reviews in Food Science and Nutrition,* 46, 1–22.

Choe, E., & Min, D.B. (2007). Chemistry of deep-fat frying oils. *Journal of Food Science,* 72, 77–86.

Ciftci, O.N., Fadiloglu, S., & Gogus, F. (2009). Conversion of olive pomace oil to cocoa butter-like fat in a packed-bed enzyme reactor. *Bioresource Technology,* 100, 324–329.

Clemente, T.E., & Cahoon, E.B. (2009). Soybean oil: genetic approaches for modification of functionality and total content. *Plant Physiology,* 151, 1030–1040.

Conklin, D.J., Barski, O.A., Lesgards, J. F., Juvan, P., Rezen, T., Rozman, D., Prough, R. A., Vladykovskaya, E., Liu, S., Srivastava, S., & Bhatnagar, A. (2010). Acrolein consumption induces systemic dyslipidemia and lipoprotein modification. *Toxicology and Applied Pharmacology,* 24, 1-12.

Dabbou, S., Issaoui, M., Brahmi, F., Nakbi, A., Chehab, H., Mechri, B., & Hammami, M. (2012). Changes in volatile compounds during processing of Tunisian-style table olives. *Journal of the American Oil Chemists' Society,* 89, 347–354.

Edwards R., Princevac M., Weltman R., Ghasemian M, Arora N-K., Bond T. (2017). Modeling emission rates and exposures from outdoor cooking. *Atmospheric Environment,* 164, 50-60. http://dx.doi.org/10.1016/j.atmosenv.2017.05.029

Esposto, S., Montedoro, G., Selvaggini, R., Riccò, I., Taticchi, A., Urbani, S., & Servilli, M. (2009). Monitoring of virgin olive oil volatile conpounds evolution during olive malaxation by an array of metal oxide. *Food Chemistry,* 113, 345–350.

Flath, A.R., Forrey, R.R., & Guadagni, D.G. (1973). Aroma components of olive oil. *Journal of Agricultural and Food Chemistry,* 21, 948–952.

Fullana, A., Carbonell-Barrachina, A.A., & Sidhu, S. (2004). Comparison of volatile aldehydes present in the cooking fumes of extra virgin olive, olive and canola oils. *Journal of Agricultural and Food Chemistry,* 52, 5207–5214.

Giuffrè, A.M., Zappia, C. & Capocasale M. (2017a). Effects of high temperatures and duration of heating on olive oil properties for food use and biodiesel production. *Journal of the American Oil Chemists' Society* 94, 819-830.

Giuffrè, A.M., Capocasale, M., Zappia, C. & Poiana M. (2017b). Influence of high temperature and duration of heating on the sunflower seed oil properties for food use and bio-diesel production. *Journal of Oleo Science* 66, 1193-1205.

Giuffrè, A.M., Caracciolo, M., Zappia, C., Capocasale, M., & Poiana, M. (2018). Effect of heating on chemical parameters of extra virgin olive oil, soybean oil and palm oil. *Italian Journal of Food Science* 30, 715-739. http://dx.doi.org/10.14674/IJFS-1269

Halvorsen, B.L. & Blomhoff, R. (2011). Determination of lipid oxidation products in vegetable oils and marine omega-3 supplements. *Food and Nutrition Research,* 55, 1654-1661.

IARC, Agents Classified by the IARC Monographs, IARC, Lyon CEDEX, France, 2012.

Jaarin, K., & Kamisah, Y. (2012). Lipid peroxidation. *Repeatedly Heated Vegetable Oils and Lipid Peroxidation*. (Chapter 10).

Jones A.P. Indoor air quality and health. (1999). *Atmospheric Environment,* 33, 4535-4564.

Kamisah, Y., Adam, A., Wan Ngah, W. Z., Gapor, M. T., Azizah, O., & Marzuki, A. (2005). Chronic intake of red palm olein and palm olein produce beneficial effects on plasma lipid profile in rats. *Pakistan Journal of Nutrition,* 4, 89–96.

Kanavouras, A., Kiritsakis, A., & Hernandez, R.J. (2005). Comparative study on volatile analysis of extra virgin olive oil by dynamic headspace and solid phase micro-extraction. *Food Chemistry,* 90, 69–79.

Katragadda, H. R., Fullana, A., Sidhu, S., & Carbonell-Barrachina, A. A. (2010). Emissions of volatile aldehydes from heated cooking oils. *Food Chemistry,* 120, 59–65.

Klein, F., Platt, S.M., Farren, N. J., Detournay, A., Bruns, E.A., Bozzetti, C., Daellenbach, K.R., Kilic, D., Kumar, N.K., Pieber, S.M., Slowik, J. G., Temime-Roussel, B., Marchand, N., Hamilton, J. F., Baltensperger, U., Prevot, A.S., & El Haddad, I. (2016). Characterization of gas-phase organics using proton transfer reactiontime-of-flight mass spectrometry: cooking emissions, *Environmental Science and Technology,* 50, 1243–1250.

Lin, J.M., & Liou, S.J. (2000). Aliphatic aldehydes produced by heating Chinese cooking oils. *Bulletin of Environmental Contamination and Toxicology,* 64, 817–824.

Lukić, I., Carlin, S., Horvat, I., & Vrhovsek, U. (2019). Combined targeted and untargeted profiling of volatile aroma compounds with comprehensive two-dimensional gas chromatography for differentiation of virgin olive oils according to variety and geographical origin. *Food Chemistry*, 270 , 403–414.

Maggio, R.M., Valli, E., Bendini, A., Gomez-Caravaca, A.M., Toschi, T.G., and Cerretani, L. (2011). A spectroscopic and chemometric study of virgin olive oils subjected to thermal stress. *Food Chemistry,* 127, 216–221.

Mele, M.A., Islam, M.Z., Kang, H.M. & Giuffrè A.M. (2018). Pre-and post-harvest factors and their impact on oil composition and quality of olive fruit. *Emirates Journal of Food and Agriculture*, 30, 592-603. doi: 10.9755/ejfa.2018.v30.i7.1742

Nasopoulou, C., Stamatakis, G., Demopoulus, C.A., and Zabetakis, I. (2011). Effects of olive pomace and olive pomace oil on growth performance, fatty acid composition and cardio protective properties of gilthead sea bream (*Sparus aurata*) and sea bass (*Dicentrarchus labrax*). *Food Chemistry,* 129, 1108-1113.

Ouni, Y., Flamini, G., Issaoui, M., Nabil, B.Y., Cioni, P.L., Hammami, M., Douja, D., & Zarrouk, M. (2011). Volatile compounds and compositional quality of virgin olive oil Oueslati variety: Influence of geographical origin. *Analytical Methods,* 124, 1770-1776.

Peng, C.Y., Lan, C.H., Lin, P.C., and Kuo, Y.C. (2017). Effect of cooking method, cooking oil, and food type on aldehyde emission in cooking oil fumes. *Journal of Hazardous Materials,* 324, 160-167.

Petersen, K., Kleeberg, K., Jahreis, G., Busch-Stockfisch, M., & Fritsche, J. (2012). Comparison of analytical and sensory lipid oxidation parameters in conventional and high-oleic rapeseed oil. *European Journal of Lipid Science and Technology,* 114, 1193-1203.

Poyato, C., Ansorena, D., Navarro-Blasco, I., & Astiasaràn, I. (2014). A novel approach to monitor the oxidation process of different types of heated oils by using chemometric tools. *Food Research International,* 57, 152-161.

Qing, G., Shan, G., Yanwei, S., Yunfeng, G., Xiaoran, W., & Zesheng, Z. (2016). Antioxidant efficacy of rosemary ethanol extract in palm oil during frying and accelerated storage. *Industrial Crops and Products,* 94, 82–88.

Rani, A.K.S., Reddy, S.Y., & Chetana, R. (2010). Quality changes in trans and trans free fats/oils and products during frying. *European Food Research and Technology,* 230, 803-811.

Reboredo-Rodrìguez, P., Gonzàlez-Barreiro, C., Cancho-Grande, B., & Simal-Gàndara, J. (2012). Dynamic headspace/GC-MS to control the aroma fingerprint of extra-virgin olive oil from the same and different olive varieties. *Food Control,* 25, 684-695.

Saad, B., Wai, W.T., Lim, B.P., & Saleh, M.I. (2007). Flow injection determination of anisidine value in palm oil samples using a triiodide potentiometric detector. *Analytica Chimica Acta,* 591, 248-254.

Schauer, J.J., Kleeman, M.J., Cass, G.R., & Simoneit, B.R.T. (2002). Measurement of emissions from air pollution sources. 4. C1–C27 organic compounds from cooking with seed oils. *Environmental Sciences and Technology*, 36, 567–575.

Sjaastad, A.K., Jorgensen, R.B., & Svendsen, K. (2010). Exposure to polycyclic aromatic hydrocarbons (PAHs), mutagenic aldehydes and particulate matter during pan frying of beefsteak. *Occupational and Environmental Medicine,* 67, 228-232.

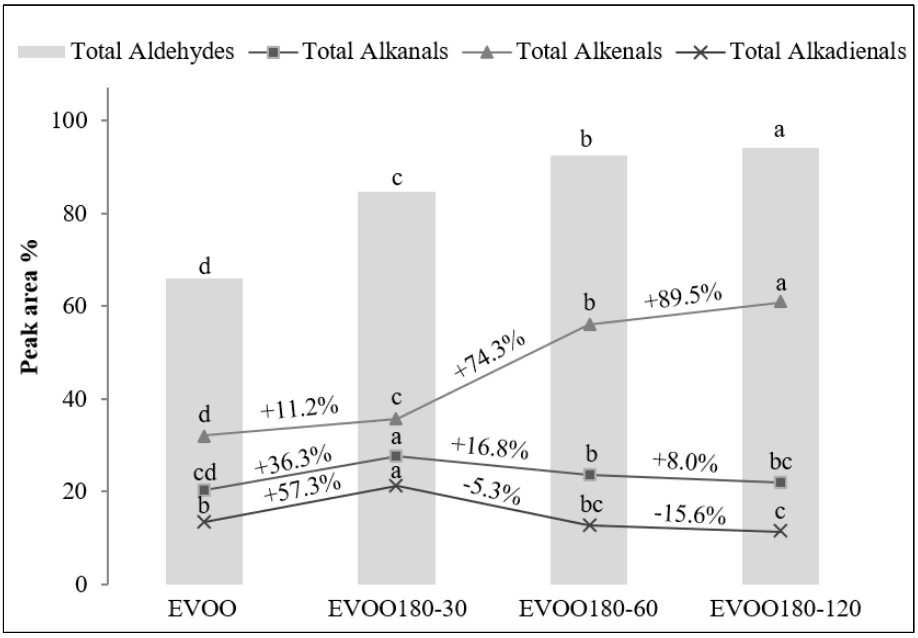
Song, J., Gardner, B.D., Holland, J.F., & Beaudry, R.M. (1997). Rapid analysis of volatile flavor compounds in apple fruit using SPME and gc/time-of-fight mass spectrometry. *Journal of Agriculture and Food Chemistry,* 45, 1801-1807.

Temime, S.B., Campeol, E., Cioni, P L., Daoud, D., & Zarrouk, M. (2006). Volatile compounds from Chétoui olive oil variations induced by growing area. *Food Chemistry*, 99, 315-225.

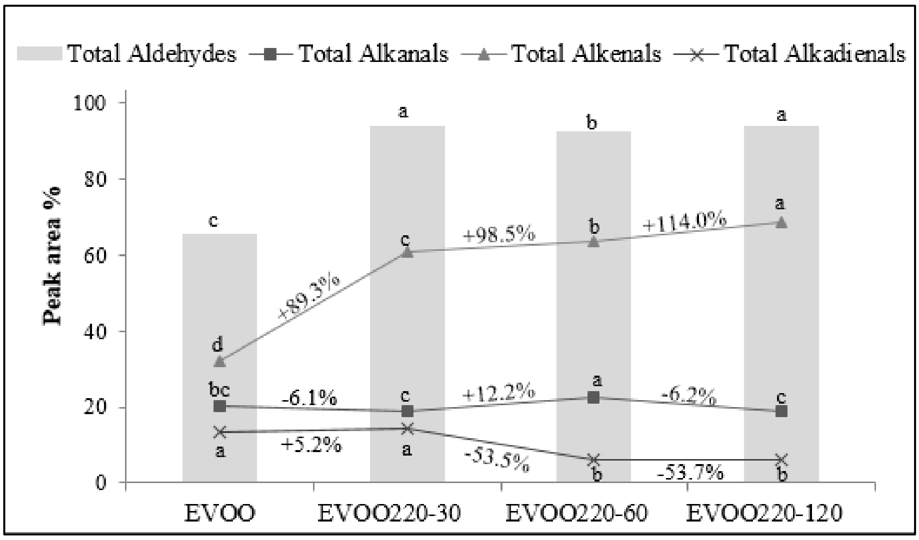
Vichi, S., Pizzale, L., Conte, L.S., Buxaderas, S., & Lopez-Tamames, E. (2003). Solid-phase microextraction in the analysis of virgin olive oil volatile fraction: modifications induced by oxidation and suitable markers of oxidative status. *Journal of Agricultural and Food Chemistry,* 51, 6564–6571.

Wang, G.W., Guo, Y., Vondriska, T.M., Zhang, J., Zhang, S., Tsai, L.L., Zong, N.C., Bolli, R., Bhatnagar, A., & Prabhu, S.D. (2008). Acrolein consumption exacerbates myocardial ischemic injury and blocks nitric oxide-induced PKCε signaling and cardioprotection. *Journal of Molecular and Cellular Cardiology,* 44, 1016-1022.

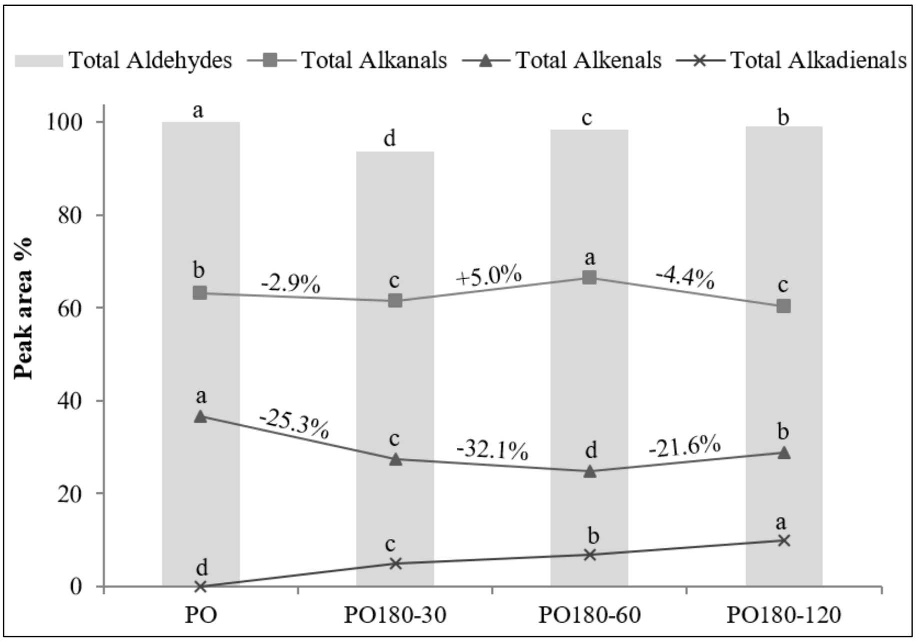
Zhao Y., Zhao B. Emissions of air pollutants from Chinese cooking: A literature review. (2018). BUILD SIMUL 11, 977-995. https://doi.org/10.1007/s12273-018-0456-6

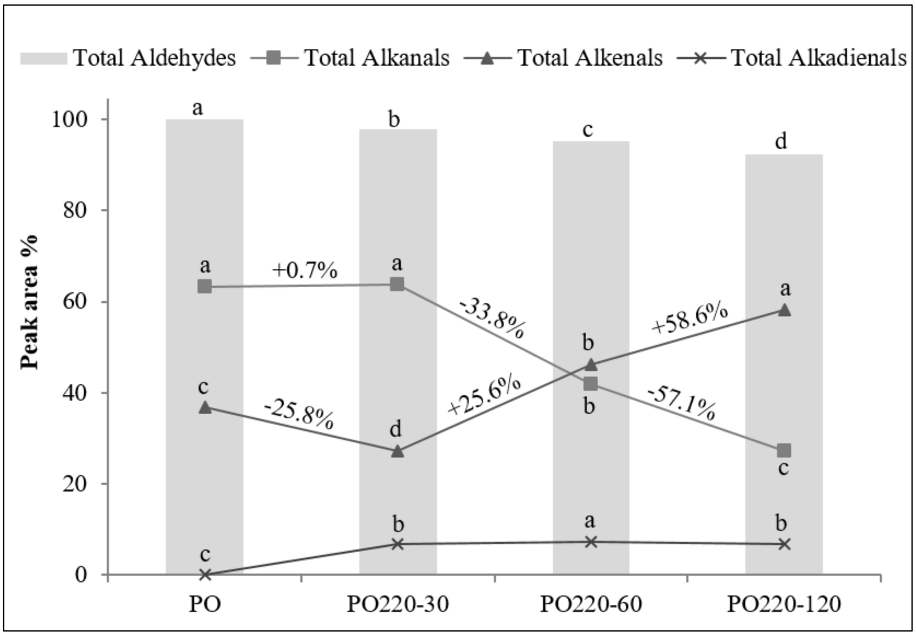


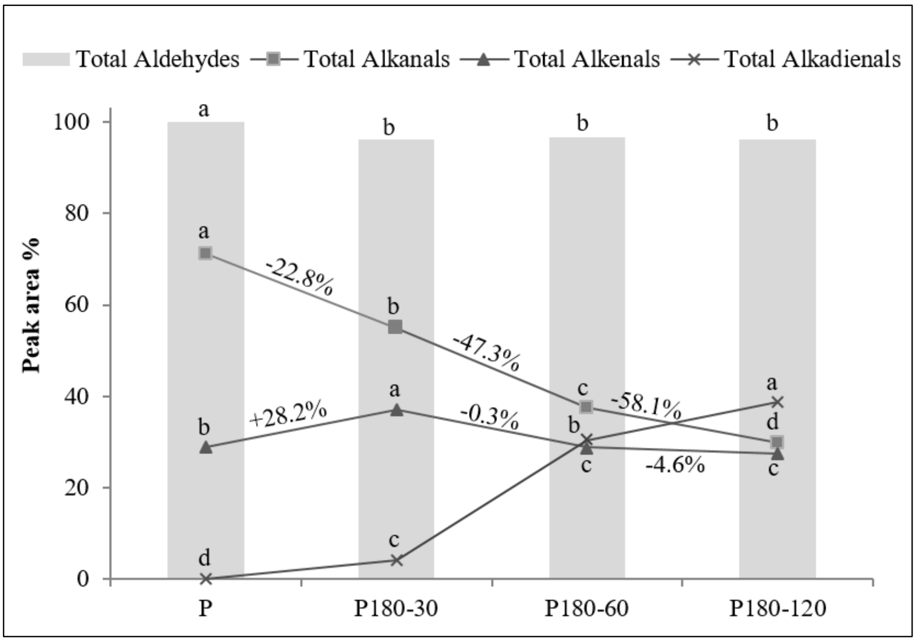
Total Aldehydes
Total Alkanals
Total Alkenals
Total Alkadienals
Peak area %
0
20
40
60
80
100
EVOO
EVOO180-30
EVOO180-60
EVOO180-120
d
c
b
a
+11.2%
+74.3%
+89.5%
+36.3%
+16.8%
+8.0%
+57.3%
-5.3%
-15.6%
cd
bc

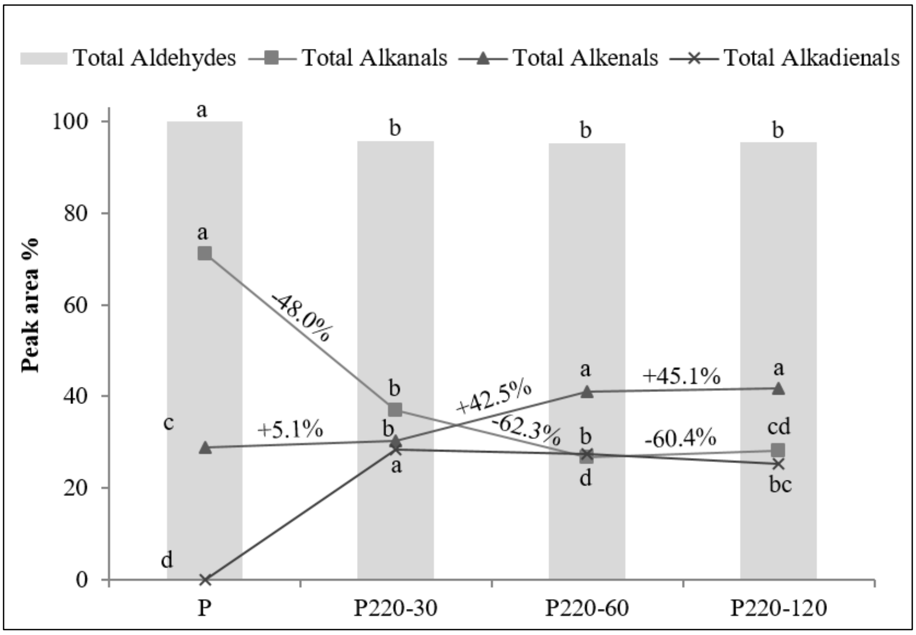
Total Aldehydes
Total Alkanals
Total Alkenals
Total Alkadienals
Peak area %
0
20
40
60
80
100
EVOO
EVOO220-30
EVOO220-60
EVOO220-120
+89.3%
+98.5%
+114.0%
-6.1%
+12.2%
-6.2%
+5.2%
-53.5%
-53.7%
bc


Total Aldehydes
Total Alkanals
Total Alkenals
Total Alkadienals
Peak area %
100
80
60
40
20
0
PO
PO180-30
PO180-60
PO180-120
-2.9%
+5.0%
-4.4%
-25.3%
-32.1%
-21.6%

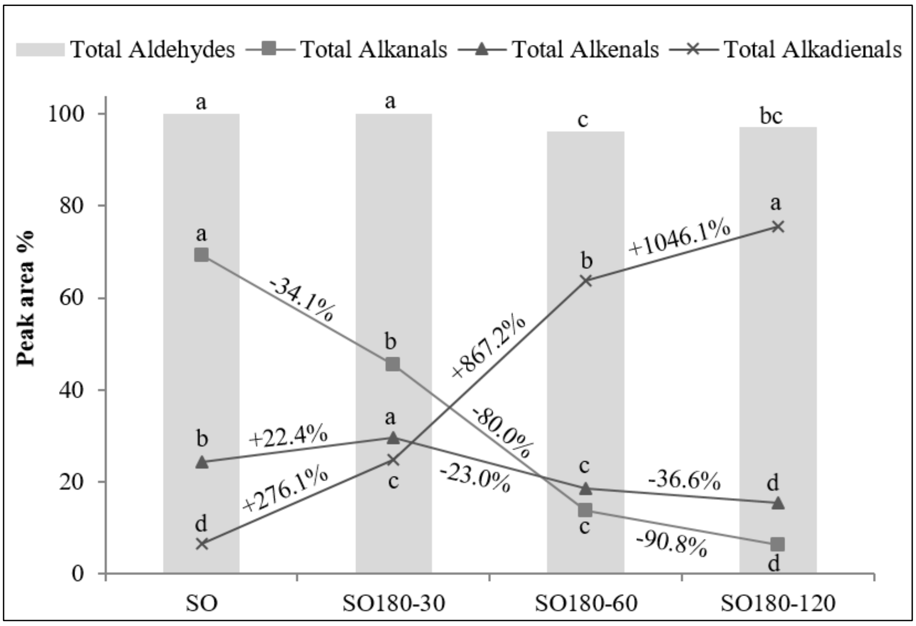

Total Aldehydes
Total Alkanals
Total Alkenals
Total Alkadienals
Peak area %
100
80
60
40
20
0
PO
PO220-30
PO220-60
PO220-120
+0.7%
-33.8%
-57.1%
-25.8%
+25.6%
+58.6%

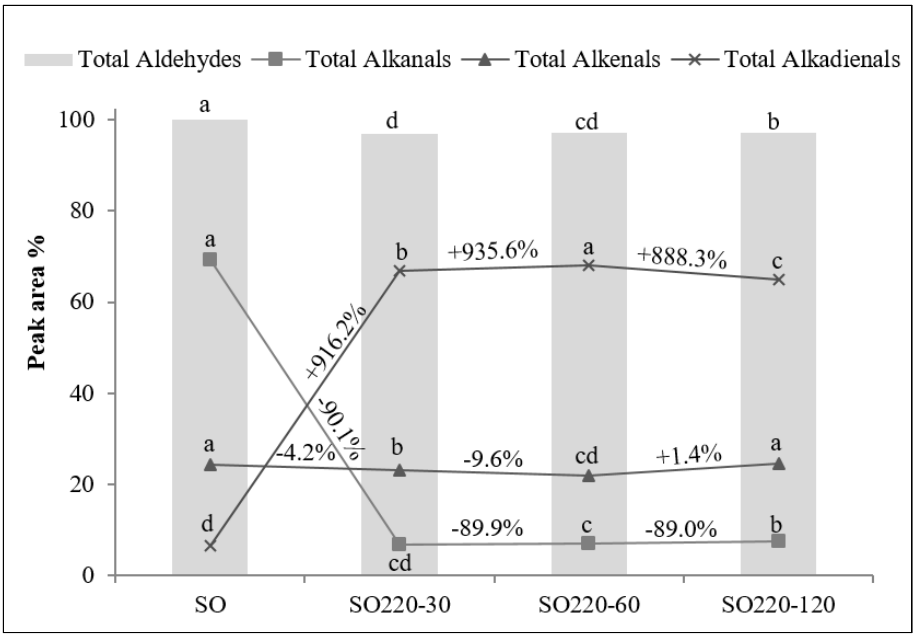


Total Aldehydes
Total Alkanals
Total Alkenals
Total Alkadienals
Peak area %
100
80
60
40
20
0
P
P180-30
P180-60
P180-120
-22.8%
-47.3%
-58.1%
+28.2%
-0.3%
-4.6%

Total Aldehydes
Total Alkanals
Total Alkenals
Total Alkadienals
Peak area %
100
80
60
40
20
0
P
P220-30
P220-60
P220-120
-48.0%
+42.5%
+45.1%
+5.1%
-62.3%
-60.4%


Total Aldehydes
Total Alkanals
Total Alkenals
Total Alkadienals
Peak area %
SO
SO180-30
SO180-60
SO180-120
-34.1%
+867.2%
+1046.1%
-80.0%
+22.4%
+276.1%
-23.0%
-36.6%
-90.8%


Total Aldehydes
Total Alkanals
Total Alkenals
Total Alkadienals
Peak area %
SO
SO220-30
SO220-60
SO220-120
+935.6%
+888.3%
+916.2%
-90.1%
-4.2%
-9.6%
+1.4%
-89.9%
-89.0%