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Fluorescence spectroscopy as a rapid and non-destructive method for monitoring quality and authenticity of fish and meat products: Impact of different preservation conditions

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ABSTRACT

Fish and meat are highly perishable food products, requiring both proper technologies for quality preservation and rapid methods for analysis. From a technological point of view, several preservation methods have been applied to extend shelf life of fish and meat products while from an analytical perspective, fluorescence spectroscopy has been extensively used to investigate its potential as a rapid and non-destructive technique compared to traditional analytical methods.

In this review, the most commonly applied techniques to preserve the quality of fish and meat products are first presented. The main methods used to assess both quality and authenticity of such products are then discussed. A special focus is placed on the fluorescence spectroscopy as a highly sensitive and selective techniques compared to other spectroscopic techniques. Moreover, attention has been paid to the effect of different preservation methods on the fluorescence measurements of fish and meat products. Finally, certain limitations are discussed, and some recommendations are suggested for future studies.

1. Introduction

Fish and meat and their products are highly recommended as part of a healthy diet, providing a variety of vital components such as proteins, polyunsaturated fatty acids, vitamins, and minerals. However, it is well-known that fish and meat are highly perishable food products and their quality declines rapidly during *postmortem* storage due to microbial growth, oxidation, and enzymatic autolysis (Dave & Ghaly, 2011; Ghaly, Dave, Budge, & Brooks, 2010). Therefore, it is a matter of utmost importance to ensure that adequate preservation methods are applied to such products in order to maintain their safety and quality and extend their shelf life as much as possible. In this context, traditional techniques such as temperature-based methods, chemical preservatives, vacuum and modified atmosphere packaging, and more recent methods such as high pressure processing, super chilling, essential oils, ionisation and active packaging have been investigated in order to maintain quality of fish and meat products (Sampels, 2015a,b; Zhou, Xu, & Liu, 2010).

During the last decades, food quality and authenticity have become increasingly of great importance for consumers, governments, and food industry due probably to changes in eating habits, consumer behaviour, industrialization of the food supply chains, and increase in food fraud (Christensen, Nørgaard, Bro, & Engelsen, 2006; Ropodi, Panagou, & Nychas, 2016; Xiaobo, Xiaowei, & Povey, 2016). Quality is a complex term, comprising many properties or characteristics like physical, compositional, and microbial features. Quality can be also related to modifications induced by technological processes or storage, nutritional value, and safety (Pathare, Opara, & Al-Said, 2013; Trimigno, Marincola, Dellarosa, Picone, & Laghi, 2015). Moreover, quality of products of animal origin is even more complicated to define on a common scale since it is affected by several intrinsic and extrinsic factors, such as animal species, age, origin, season, nutritional status, etc. (Hassoun & Karoui, 2017; Hocquette et al., 2012).

An important parameter related to food quality is authenticity process, whose the aim is to verify that a food is in compliance with its label description in respect to geographical origin, production method

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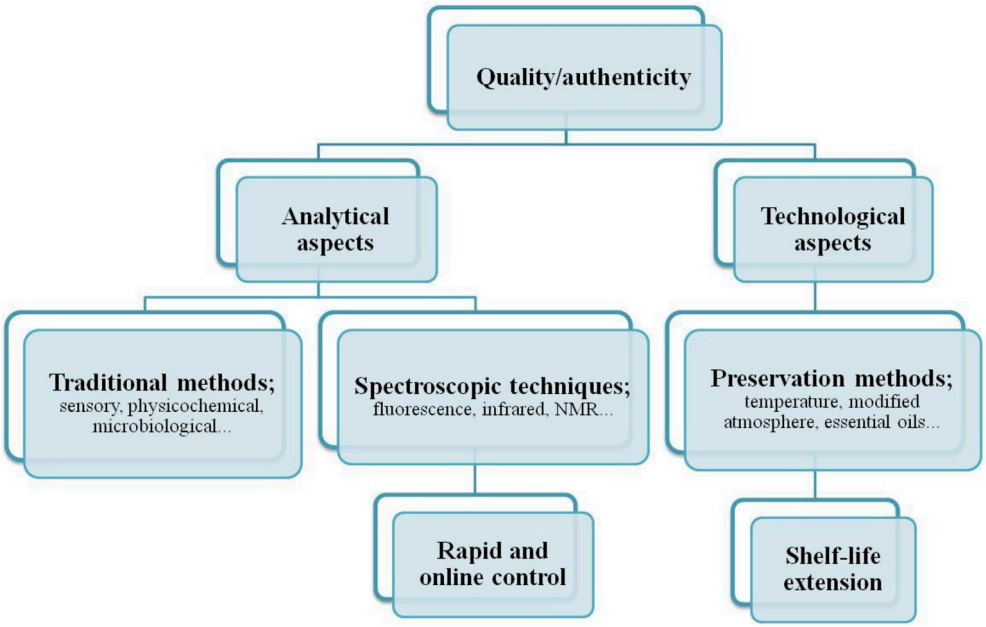

Fig. 1. Schematic illustration of quality and authenticity concepts and related relevant topics for fish and meat products.

(e.g., conventional, organic, wild, farmed …), processing technologies (e.g., fresh or frozen/thawed products), among others (Abbas et al., 2018; Danezis, Tsagkaris, Camin, Brusic, & Georgiou, 2016). Additionally, adulteration and food fraud create technical, logistical, and financial challenges, especially with increased international trade in food products. In a recent review, Sørensen and co-workers defined adulteration as the undeclared introduction of an additional cheaper substance to foods, while food fraud was referred as an intentional misrepresentation of foods using prohibited substances, contamination of the product and other non-compliances to product descriptions (Sørensen, Khakimov, & Engelsen, 2016).

Due to the vulnerability and perishability of fish and meat products, it is essential to develop effective and rapid techniques for monitoring quality changes and determining authenticity of these products (Fig. 1). Several traditional techniques including sensory evaluation, microbial analysis, chromatography, and physicochemical measurements have been used for quality and authenticity analyses. Despite the importance of these standard and reference methods, they are destructive, sometimes expensive, time-consuming, and often susceptible to large sources of variation (Cheng et al., 2013; Cheng & Sun. 2015). Over the last few years, several methods have been developed and tested to overcome these limitations. In this regard, spectroscopic methods, including mid-infrared (MIR), near-infrared (NIR), nuclear magnetic resonance (NMR), and Raman spectroscopies represent a very successful alternative to traditional techniques due to their desirable features such as rapid detection, objectivity, reliability, ease of use, and minimal or no sample preparation (Cozzolino & Murray, 2012; He & Sun, 2015; Xiaobo et al., 2016). More interestingly, due to its high selectivity and sensitivity, fluorescence spectroscopy has been reported to be an efficient and promising tool to control food quality and authenticity (Airado-Rodríguez, Skaret, & Wold, 2010; Kumar, Tarai, & Mishra, 2017; Sádecká & Tóthová, 2007). Indeed, the use of fluorescence spectroscopy for applications related to fish and/or meat has increased significantly during the last decade to reach about 300 scientific papers in the year 2015 (Fig. 2).

A number of review papers on the application of spectroscopic techniques have been published in the last years, focussing on the use of NIR and MIR spectroscopies (Cozzolino & Murray, 2012), NMR (Trimigno et al., 2015), and hyperspectral imaging (Feng, Makino, Oshita, & García Martín, 2018; Siche et al., 2016). Some reviews have

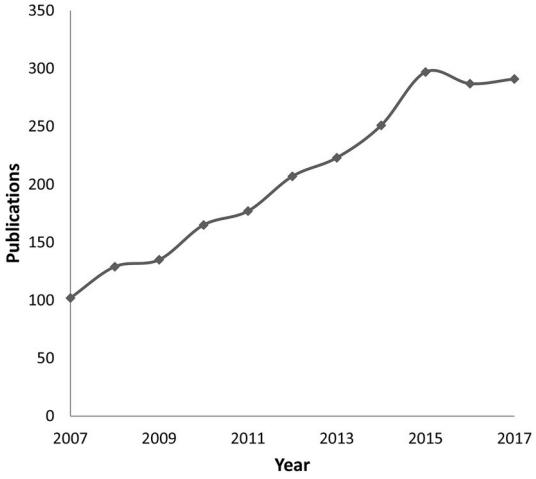

Fig. 2. Publications of scientific articles on the application of fluorescence spectroscopy in fish and/or meat. Information obtained from the database Scopus (search criteria: ALL FIELDS: fluorescence spectroscopy, and ARTICLE TITLE, ABSTRACT, KEYWORDS: "fish and/or meat). The data were obtained from Scopus in August 2018.

reported the applications of fluorescence spectroscopy for quality evaluation of general food products (Christensen et al., 2006; Karoui & Blecker, 2011; Sádecká & Tóthová, 2007). However, to date, we are unaware of any review to distinctively present the current research and potential application of fluorescence spectroscopy in measuring quality and authenticity of fish and meat products stored under various preservation methods. Moreover, the large number of published papers in this field (Fig. 2) requires a continuous update. Thus, the objective of this paper is to review, from a technological point of view, the most used methods to preserve quality and increase shelf life of fish and meat and their products. On the other hand, the applications of fluorescence spectroscopy, as a rapid and non-destructive method, compared to traditional techniques, for quality evaluation and authenticity control of fish and meat are presented.

2. Fish and meat preservation methods

Many preservation techniques have been applied to retard deterioration and extend the shelf life of fish and meat products. Food preservation methods are usually based on the control of temperature, available oxygen, water activity, microbial loads, or several of these parameters at the same time (Dave & Ghaly, 2011; Zhou et al., 2010).

2.1. Temperature-based preservation methods

Low-temperature techniques, including chilling, super chilling, and freezing are the most common preservation methods of fish and meat and their products. Chilling (or cooling) storage involves keeping product temperature between 0 and 4 °C, while super chilling (or super cooling) refers to lowering the temperature below the usual freezing point (between −0.5 °C and −2.8 °C) without the product freezing (Kaale, Eikevik, Rustad, & Kolsaker, 2011; Sampels, 2015b; Stonehouse & Evans, 2015). During the super chilling process, only a part of the internal water of the product is frozen, offering several advantages, such as inhibition of growth of harmful microorganisms and maintaining freshness and high food quality, compared to the normal cold storage (Kaale et al., 2011; Stonehouse & Evans, 2015).

Cooking of fish and meat is one of the most important processing operations since, in the modern society, such products are almost always cooked prior to consumption. Cooking affects the molecular structures of the cooked products due to its impact on protein denaturation, lipid oxidation, Millard reaction, and the formation of flavour substances (Gatellier, Santé-Lhoutellier, Portanguen, & Kondjoyan, 2009; Miao, Liu, Bao, Wang, & Miao, 2017). Other processing methods, such as smoking, salting, and drying have been very commonly used as traditional preservation process since antiquity to improve sensory characteristics and extend the shelf life of food products (Sampels, 2015b).

2.2. Modified atmosphere packaging

Modified atmosphere packaging (MAP) can be defined as the enclosure of the food product in a package in which the percentage of the three principal gases (%CO_2, %O_2, and %N_2) is modified to provide an optimal atmosphere for retarding microbiological growth and deteriorative chemical processes. This technique has been extensively used to extend the shelf life of many food products, especially fish and meat (Sivertsvik, Jeksrud, & Rosnes, 2002). Generally speaking, the change in the normal composition of atmospheric air can be achieved by reducing the oxygen content and increasing the levels of carbon dioxide and/or nitrogen (Mastromatteo, Conte, & Del Nobile, 2010; Sivertsvik et al., 2002; Zhou et al., 2010). However, to optimize the effectiveness of MAP, the optimum level of each gas must be determined for each food product. For example, to preserve the bright red colour of meat, high percentages of oxygen should be kept in the package, while gas mixtures without oxygen are generally recommended for fatty fish to avoid oxidation. To minimize the oxidative deteriorative reactions and reduce aerobic bacteria growth, vacuum packaging has been widely used in fish and meat products (Sampels, 2015b; Zhou et al., 2010). In fact, the vacuum packaging could be considered as a special case of MAP.

2.3. High-pressure processing

In recent years, high-pressure processing (HPP) has attracted great attention as environmentally-friendly and non-thermal food processing technologies due to its potential of maintaining organoleptic properties and nutritional quality of treated products. Consequently, this preservation method could be considered as an interesting solution to extend the shelf life of food products by inhibiting bacterial growth without resorting to heat (Huang, Wu, Lu, Shyu, & Wang, 2017; Rode & Hovda, 2016). This technique has been extensively reviewed in the last few years. Indeed, interested readers can refer to recent reports focusing either on the food product in general (Huang et al., 2017) or on fish (Alves de Oliveira et al., 2017) and meat (Guyon, Meynier, & de Lamballerie, 2016) products in particular.

2.4. Use of natural compounds

The use of salts, such as potassium nitrite, sodium nitrite, sodium lactate, sodium citrate, sodium acetate, etc, as well as synthetic antioxidants, such as butylated hydroxytoluene, butylated hydroxyanisole, and propyl gallate have been reported as chemical preservatives of fish and meat products (Dave & Ghaly, 2011; Ghaly et al., 2010; Zhou et al., 2010). Nevertheless, consumers increasingly demand healthier food products, which are free of chemical additives or any substances that could be harmful to their health. With this in mind, natural preservatives such as edible films and coatings and essential oils (Dehghani, Hosseini, & Regenstein, 2018; Hassoun & Emir Çoban, 2017; Jayasena & Jo, 2014; Ribeiro-Santos, Andrade, Sanches-Silva, & de Melo, 2018) have been increasingly applied to fish and meat products in order to enhance their antimicrobial and antioxidant stability. More recently, active food packaging, including the incorporation of essential oils and other natural compounds to package films, has been developed in order to enhance antimicrobial and antioxidant properties and extend the shelf life of perishable food products such as fish and meat (Ribeiro-Santos, Andrade, Melo, & Sanches-Silva, 2017).

3. Conventional techniques used for quality and authenticity control of fish and meat

Sensory assessment has been widely used as one of the most traditional and efficient tools for quality analysis in many food sectors. The sensory analysis can be performed either traditionally by well-trained panellists or untrained subjects according to the objective of analysis, or by using instrumental techniques. One example of the traditional sensory methods is “quality index method”, which is widely used to assess freshness of many raw fish species. Even though human test panels are expensive, time-consuming, and cannot be used as a routine procedure, they are still preferred to instrumental sensory methods. These latter, referred as biomimetic sensors, have been developed recently to mimic the olfactory (e-nose), gustatory (e-tongue), and visual (e-eye) systems of humans (Ghasemi-Varnamkhasti, Mohtasebi, & Siadat, 2010; Ropodi et al., 2016).

Many traditional physicochemical methods, such as colour, texture and structure measurements, oxidation products and volatiles compounds, have been commonly used and considered as references methods to assess food quality (Cheng, Sun, Han, & Zeng, 2014; Hassoun & Karoui, 2017; Pathare et al., 2013). Microbial analysis is another conventional method, which plays an important role in food quality evaluation (He & Sun, 2015). Additionally, gas chromatographic mass spectrometric has been developed for the detection of food authenticity by measuring volatile and semi-volatile compounds (Cuadros-Rodriguez, Ruiz-Samblas, Valverde-Som, Pérez-Castano, & Gonzalez-Casado, 2016; Danezis et al., 2016). However, in practice, very few techniques meet the requirements of industry, since most of these methods have several disadvantages being destructive, subjective, expensive, and most of all time-consuming (Cheng, Cheng, Sun, & Pu, 2015; Elmasry & Nakauchi, 2016; ElMasry, Nakazawa, Okazaki, & Nakauchi, 2016).

4. Spectroscopic methods

During the last few decades, the potential of many spectroscopic techniques, especially fluorescence and vibrational spectroscopy, have been confirmed in many applications.

4.1. Fluorescence spectroscopy

Fluorescence is the emission of light by a fluorophore following the absorption of UV or VIS light. Fluorophores are fluorescent molecules composed of several conjugated aromatic rings or linear and cyclic molecules which possess one or more conjugated bonds. These molecules may be hydrophilic, hydrophobic or even amphiphilic.

The geometry of the sample illumination and the optical density of the sample have a great impact on the apparent fluorescence intensity and spectral distribution of a sample. Commonly, the geometry used in fluorescence spectroscopy is the right-angle observation of the center of a centrally illuminated cuvette. In cases where the sample is very thick and dense, the measurements using a right-angle are difficult to perform. High optical densities at excitation and emission wavelengths may decrease the real fluorescence intensity of the sample and distort the fluorescence spectra. With the usual right-angle observation, the instrument only detects the fluorescence emitted from the central part of the exciting beam and, therefore, when the concentration of the sample is high, a significant part of the incident light is absorbed before reaching the central part of the cuvette. In addition, optical properties i.e., scattering and absorption, of biological samples, such as fish and meat, may decrease fluorescence intensity due to the increased path length and reabsorption of fluorescence (Airado-Rodriguez, Høy, Skaret, & Wold, 2014; Lakhal, Acha, & Aussenac, 2014). To overcome this problem, front-face fluorescence spectroscopy can be used. By using this method, the sample is excited at the cuvette surface, in a way that any displacement of the excitation light through the sample to the cuvette center is avoided. Therefore, the fluorescence spectra (in excitation and emission) using front-face mode is not distorted. Using this technique, the excitation light is focused to the front surface of the sample, and the fluorescence emission is collected from the same region, using an angle that decreases the reflected and scattered light. This technique is adequate for highly concentrated and opaque or solid samples. However, one should keep in mind that the sample surface must be representative of the quality attributes under investigation (Albani, 2007; Christensen et al., 2006).

Usually, the data obtained from the fluorescence spectroscopy is presented as emission spectra. This spectrum is measured as the light emitted (fluorescence) across a wide range of wavelengths upon excitation at a fixed wavelength, giving a plot that displays the fluorescence intensity versus the emission range, in terms of wavelength (nm) or wavenumber (cm^{-1}). Alternatively, an excitation spectrum can also be obtained by measuring the emission at one fixed wavelength while exciting the molecule over a wide range of wavelengths (Lakowicz, 2006). The measurements at only one excitation or emission wavelength has been most commonly used in many applications of fluorescence spectroscopy and will be referred to classical mode in our review. However, in food science analysis it is very common to use fluorescence Excitation-Emission Matrix (EEM), also known as fluorescence landscape, which is further investigated through multivariate data analysis. To obtain the fluorescence data in landscape, several emission spectra at different excitation wavelengths (or vice versa) must be measured, thus creating an excitation-emission map that covers the total area of fluorescence. This structure has the advantage of detecting analytes or interferences present in different areas, and visualizing all fluorophores that exist in the sample (Kumar et al., 2017; Lakhal et al., 2014). However, the collection of fluorescence EEM using commercially available scanning fluorimeters requires about 30 min, while only few seconds are enough to acquire a classical spectrum at one excitation or emission wavelength.

Synchronous fluorescence spectroscopy (SFS) (Fig. 3) is a good compromise between the fluorescence EEM spectroscopy and fluorescence single spectroscopy (the classical mode). The SFS has the advantage of being able to characterize several fluorophores from a single spectrum but in a shorter time. The use of this screening mode is very advantageous when the sample chemical composition in unknown. Constant-wavelength SFS is the most frequently used in food analysis (Kumar et al., 2017; Li et al., 2012). Recent investigations have confirmed the potential of SFS for studying various quality parameters (Aït-Kaddour, Loudiyi, Ferlay, & Gruffat, 2018; Liu et al., 2012; Sahar, Rahman, Kondjoyan, Portanguen, & Dufour, 2016).

4.2. Other spectroscopic methods

Apart from the fluorescence spectroscopy, other spectroscopic techniques, including NIR, MIR, Raman, and NMR methods have been widely investigated in the last decade.

Based on measurement of changes in the spectral characteristics over a wavenumber range from 14000 to 4000 cm^{-1} for NIR (often coupled with visible near infrared; VIS/NIR) or from 4000 to 400 cm^{-1} for MIR, numerous studies have focused on the use of infrared spectroscopy for the estimation of quality and authenticity in fish and meat products (Cozzolino & Murray, 2012; Troy, Ojha, Kerry, & Tiwari, 2016; W.; Wang, Peng, Sun, Zheng, & Wei, 2018). Recent advances and accessories such as the development of Fourier transform infrared (FT-IR) and the use of attenuated total reflection allowed this technique to be extended further, providing fast analytical tool as compared with traditional infrared. Fourier transform near-infrared (FT-NIR) and Fourier transform mid-infrared (FT-MIR) spectroscopies have emerged as effective and reliable analytical techniques for identification and quantification of various quality attributes in fish and meat products. The FT-IR was introduced in the year 1991 for microbiological characterization (Naumann, Helm, & Labischinski, 1991), and since then the technique has gained increasing attention due to its versatility, allowing various applications in many fields. In recent years, several applications of this technique were published including differentiation between fresh and frozen/thawed chicken (Grunert, Stephan, Ehling-Schulz, & Johler, 2016) and tuna (Reis et al., 2017), characterization of frauds in bovine meat (Nunes, Andrade, Santos Filho, Lasmar, & Sena, 2016), identifying and quantifying fraud in fresh, cooked and frozen turkey meat (Alamprese, Amigo, Casiraghi, & Engelsen, 2016), and studying of different authenticity issues related to fish (Alamprese & Casiraghi, 2015).

More recently, hyperspectral imaging technique has been developed as rapid, non-destructive, smart, and promising analytical tool in order to generate spatial and spectral information of the tested sample simultaneously (Chen, Sun, Cheng, & Gao, 2016; Cheng, Nicolai, & Sun, 2017). Many recent reports have demonstrated the potential of this technique to predict several quality and authenticity issues of fish and meat products, such as microbial spoilage, texture and colour attributes, discrimination between fresh and frozen/thawed products, etc (Cheng & Sun, 2015; Elmasry & Nakauchi, 2016; Kamruzzaman, Makino, & Oshita, 2015; Ropodi, Panagou, & Nychas, 2017; Washburn, Stormo, Skjelvareid, & Heia, 2017). However, the application of hyperspectral imaging in the fluorescence mode is still very limited (Lee, Kim, Lee, & Cho, 2018). Therefore, the technique was investigated for monitoring cooking temperature in cod (*Gadus morua*) fillets processed with three different temperatures (30, 50, and 70 °C). A partial least square regression (PLSR) model was built and used to predict cooking temperatures in each pixel of the spectral images, and the resulting images were displayed with a linear colour scale, varying from blue, for the low temperature treated-samples to yellow, for the high temperature-treated ones (Fig. 4). Our preliminary results showed that fluorescence hyperspectral imaging had a huge potential for classification and visualisation of fish status during thermal processing.

Raman is one of emerging vibrational spectroscopy that provides detailed chemical information and characterisation and identification of heterogeneous foods and food ingredients by measuring the wavelength and intensity of inelastically scattered light from molecules (Reid, O'Donnell, & Downey, 2006; Troy et al., 2016; Xiaobo et al., 2016). This technique has been successfully used to determine the content of horse meat in its mixture with beef (Zajaç, Hanuza, &

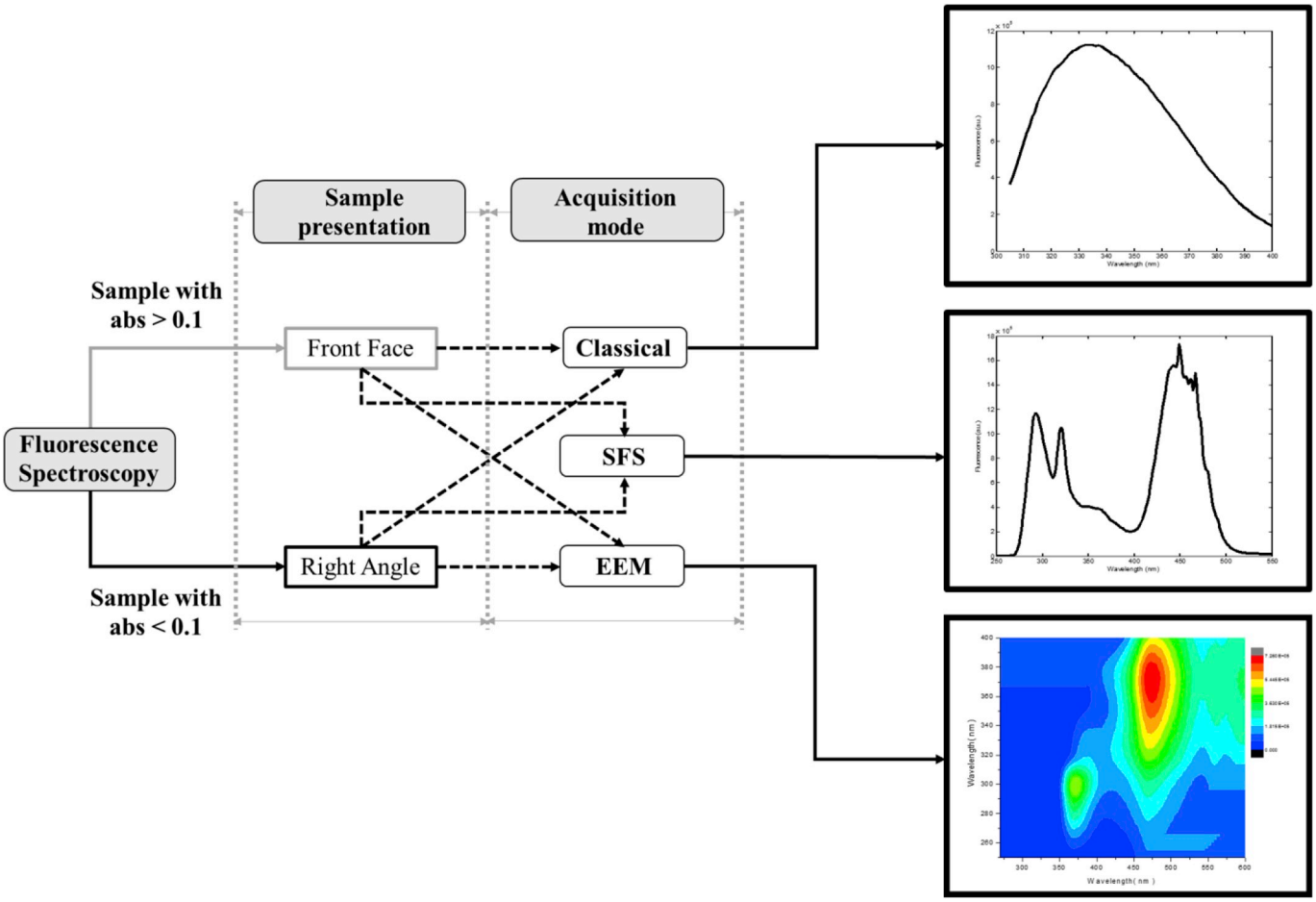

Fig. 3. Principal acquisition modes of fluorescence spectroscopy (see the text for the abbreviations).

Dymińska, 2014) and to discriminate between species and fresh and frozen/thawed fish (Velioglu, Temiz, & Boyaci, 2015). In recent times, Raman imaging has emerged as another novel technique that combines the advantages of Raman spectroscopy and digital imaging to be used in food quality and safety evaluation (Yaseen, Sun, & Cheng, 2017).

NMR spectroscopy is an important analytical technique based on the analysis of the energy absorption in the radio-frequency range of the electromagnetic spectrum by atomic nuclei with non-zero spins (nuclei that contain odd numbers of protons or neutrons, such as ^{1}H and ^{13}C) in the presence of a magnetic field (Damez & Clerjon, 2013; Reid et al., 2006; Xiaobo et al., 2016). Traceability, authenticity, and safety of food are the main application fields of this fingerprinting method, providing "high-throughput" spectroscopic and structural information on a wide range of molecular compounds (Danezis et al., 2016; Laghi, Picone, & Capozzi, 2014; Trimigno et al., 2015). Good results were obtained using this technique as a tool for discriminating between wild and farmed sea bream (Melis, Cappuccinelli, Roggio, & Anedda, 2014), discriminating between beef samples according to their race and sex (Santos et al., 2014), and monitoring metabolites and other *postmortem* changes in Atlantic salmon (Shumilina, Ciampa, Capozzi, Rustad, & Dikiy, 2015).

5. Applications of fluorescence spectroscopy for quality and authenticity analysis

5.1. Fish and other seafoods

Fish and seafood products could be considered as complex multi-fluorophoric systems, containing several molecules with conjugated double bonds, such as aromatic amino and nucleic acids, nicotinamide adenine dinucleotide (NADH), vitamin A, riboflavin, and oxidation products (Kumar et al., 2017; Sádecká & Tóthová, 2007; Xiaobo et al., 2016). Some examples of relevant studies documenting the use of fluorescence spectroscopy for quality and authenticity control of fish and other seafoods are shown in Table 1.

5.1.1. Use of fluorescence spectroscopy for monitoring freshness

One of the pioneer investigations that tackle the potential of FFFS for monitoring fish freshness was conducted by Dufour and co-workers on two fatty fish species, namely mackerel (*Scomber scombrus*) and salmon (*Salmo salar*), and two lean fish species; cod and whiting (*Micromesistius poutassou*), stored in ice for 1, 5, 8, and 13 days. In this study, the fluorescence signals were measured after excitation set at 250 nm (aromatic amino and nucleic acids), 290 nm (tryptophan), and 336 nm (NADH) (Dufour, Frencia, & Kane, 2003). The results showed some differences in the shape of the fluorescence spectra according to the storage time, suggesting that this technique could be considered as fingerprints allowing the discrimination between fresh and aged fish fillets.

In recent years, a few papers have been published by the research team of one of us (Hassoun & Karoui, 2015, 2016; Karoui & Hassoun, 2017), demonstrating the potential of the fluorescence spectroscopy to assess fish freshness as a function of storage time and storage condition. In more details, the potential of FFFS in classical acquisition mode was evaluated for monitoring freshness of 4 groups of lean fish species; whiting (*Merlangius merlangus*) fillets stored up to 12 days under different conditions of light and vacuum packaging: i) dark/partial vacuum; ii) dark/total vacuum; iii) light/partial vacuum; and IV) light/total vacuum (Hassoun & Karoui, 2015). From a technological

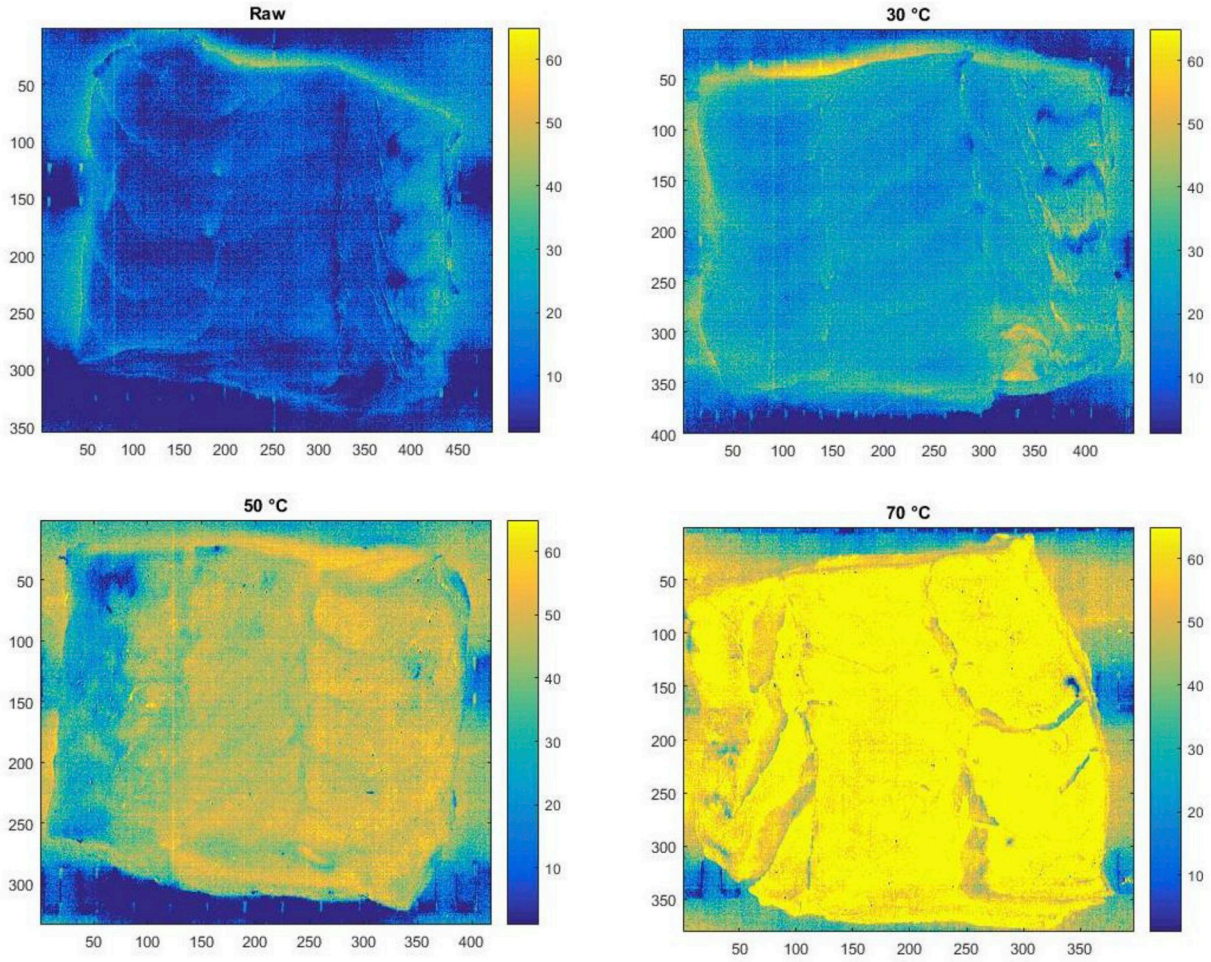

Fig. 4. Visualization of cod sample heated with different temperatures using PLSR model.

perspective, the authors demonstrated that fish fillets packed in a total vacuum and kept in the dark had higher quality attributes compared to fillets stored under the other preservation conditions. Regardless of storage condition, it was noticed that fluorescence intensities around the emission maximum of tryptophan (excitation set at 290 nm) and NADH (excitation set at 340 nm) tended to increase with storage time. Moreover, at a specific storage time, fluorescence intensities around the emission maximum were higher for samples exposed to light and packed in partial vacuum, which was attributed to the formation of fluorescent oxidation products. The same authors investigated the ability of the FFFS for monitoring quality changes of whiting fillets stored under two different conditions of MAP (50% N_2/50% CO_2 and 80% N_2/20% CO_2) in comparison with storage in normal air (Hassoun & Karoui, 2016). The findings of this study demonstrated that the use of MAP, particularly that of 50% CO_2 could extend the shelf life and improve the quality of fish fillets, which in turn could be evaluated by the fluorescence spectroscopy as a rapid and non-destructive technique compared to some traditional analytical methods. More recently, the effects of rosemary and basil essential oils on the preservation of quality and the shelf life of Atlantic mackerel (*Scomber scombrus*) fillets, as a fatty fish, were evaluated using the FFFS in comparison with some traditional measurements (Karoui & Hassoun, 2017). When viewed from a technological point of view, the results showed that fish fillets treated with these essential oils had higher antioxidant and antimicrobial stabilities compared to control samples. Moreover, from an analytical angle, the application of chemometric tools, such as principal component analysis (PCA) and factorial discriminant analysis (FDA) demonstrated that only the combination of fluorescence and traditional measurements allows discrimination between the fish groups, indicating that the use of fluorescence spectroscopy cannot eliminate the need for more detailed physicochemical analysis.

However, given the limited number of samples analysed, the findings of the previous studies should be confirmed on a larger number of samples as well as on other fish species. Additionally, the increase in shelf life achieved using MAP and essential oils should have been validated by means of microbiological and sensory analysis. Besides, even though these investigations exhibited different fluorescence properties, much effort is still needed to verify exactly which fluorophores were responsible for such fluorescence signals. Moreover, it is worth noting that, the use of fluorescence spectroscopy in classical mode (at only one excitation or emission wavelength) may restrict the efficiency of this technique in detecting several fluorophores.

5.1.2. Evaluation of lipid and protein oxidation by fluorescence spectroscopy

Fish are characterized by their high content of polyunsaturated fatty acids and high content of proteins and enzymes, making them highly vulnerable to oxidative changes and other alteration processes (Shi et al., 2019; Xu, Riccioli, & Sun, 2015). Many authors have suggested the use of fluorescence spectroscopy as a rapid technique for evaluating oxidation and other changes and determining quality.

One of the first reported applications of fluorescence spectroscopy for monitoring quality of fish was published in 1982 by Davis and Reece, when it was revealed that fluorescence emission from frozen fish occurs as a result of reactions between free amino groups and carbonyl compounds (Davis & Reece, 1982). Several earlier studies, conducted

Table 1
Some examples of relevant studies concerning the applications of fluorescence spectroscopy for quality and authenticity control of fish and other seafoods.

Fish or other seafoods	Fluorescence acquisition mode	Fluorophore (s)	Aim of the study	Reference
Sardine	Single excitation/emission wavelength	Interaction compounds formed between damaged lipid and protein compounds	Determination of the impact of chilled and frozen storage on some traditional quality parameters and fluorescence measurements	Aubourg and Medina (1997)
Blue whiting	Single excitation/emission wavelength	Interaction compounds formed between damaged lipid and protein compounds	Monitoring lipid oxidation during chilled storage using lipid indices, TVB-N, and fluorescent interaction compounds	Aubourg et al. (1998)
Salmon and cod	Classical mode	Collagen type I and type V	Monitoring changes in fluorescence properties and investigate correlations with texture measurements	Andersen and Wold (2003)
Cod, mackerel, salmon, and whiting	Classical mode	Tryptophan, NADH, aromatic amino and nucleic acids	Investigation the potential of fluorescence spectroscopy for monitoring fish freshness	Dufour et al. (2003)
Salmon pâté	Classical mode	Oxidation products	Detection of early lipid oxidation in complex food matrix	Olsen et al. (2006)
Japanese dace (*Tribolodon hakonensis*)	EEM	Amino acids and uric acid	Monitoring freshness of fish by investigating fluorescence properties of eye fluid	Liao, Suzuki, Shirataki, Kuramoto, and Kondo (2018)
Color carp (*Carpio color*)	Synchronous	Pyrene	Determination concentrations of pyrene in fish gills	Liu et al. (2012)
Shrimp	EEM	Tryptophan and other fluorophores	Determination of geographical origin of two shrimp species	Eaton, Alcivar-Warren, and Kenny (2012)
Cod caviar paste	Classical mode	Oxidation products, Protoporphyrin, photoprotoporphyrin	Determination of lipid auto-oxidation and photo-oxidation	Airado-Rodriguez et al. (2014)
Horse mackerel (*Trachurus japonicus*)	EEM	Fluorescent oxidation products, tryptophan	Estimation of the initial loss of freshness of frozen fish	ElMasry et al. (2016)
Whiting	Classical mode	Tryptophan, Schiff-base	Monitoring of freshness of fish stored under MAP	Hassoun and Karoui (2016)
Sea bass (*Dicentrarchus labrax*)	Classical mode	Tryptophan, NADH, riboflavin, vitamin A	Discrimination between fresh and frozen/thawed fish and determination of the initial freshness status of fish before freezing	Karoui et al. (2017)
Hake (*Merluccius hubbsi*)	EEM	Tryptophan, NADH	Investigate correlation between fluorescence measurements and some classical quality parameters	Risso, Crovetto, Ávila, and Gutiérrez (2017)

EEM: Excitation– Emission Matrices; MAP: Modified Atmosphere Packaging; TVB-N: Total Volatile Basic Nitrogen; NADH: Nicotinamide Adenine Dinucleotide.

by Spanish researchers headed by Santiago P. Aubourg, have investigated the potential of fluorescence spectroscopy as a rapid technique to assess lipid oxidation in several fatty and lean fish species, including sardine (*Sardina pilchardus*), mackerel, and whiting during different storage and processing conditions (e.g., refrigerated storage, frozen storage, canning …) (Aubourg & Medina, 1997; Aubourg, Medina, & Gallardo, 1998; Aubourg, Sotelo, & Gallardo, 1997). Although interesting results were obtained since a fluorescence shift towards a higher wavelength maximum was detected because of lipid oxidation, this shift was calculated as the ratio between only one excitation/emission wavelength pair (393/463 nm and 327/415 nm) which may cause some loss of information contained in fish samples.

In recent years, the oxidation process and other quality indicators such as concentration of volatiles and sensory properties were investigated on cod caviar paste samples stored at 4 °C with different concentrations of O_2 in the presence or absence of light, using FFFS after excitation set at 382 nm (Airado-Rodríguez et al., 2010). A broad and intense peak in the 410–500 nm regions, especially for samples exposed to light, was observed and ascribed to products arising from reactions of unsaturated aldehydes with proteins, while the peak noticed at about 470 nm was attributed to oxidation products. These results were confirmed later in a similar but more detailed study by the same research team, using multispectral imaging of fluorescence, providing more details information about such heterogeneous samples at microscopic levels (Airado-Rodriguez et al., 2014).

Evaluation of protein conformational changes and modifications in secondary and tertiary structures in protein during processing can be investigated by fluorescence spectroscopy (K. Wang, Sun, Pu, & Wei, 2017). In a recent study (Hu et al., 2017), the authors used Schiff-base structures, which are formed during cooking as a result of interaction between myofibrillar proteins and lipids, as indicators of protein oxidation of farmed sturgeon (*Acipenser gueldenstaedtii*). The emission spectra obtained after excitation set at 360 nm displayed various patterns of spectra as a function of cooking methods (roasting, frying, boiling, steaming, and microwaving). Indeed, fluorescence intensity obtained from samples heated by the extreme cooking methods (roasting and frying) was found to be much higher than fluorescence intensity resulted from samples heated by the other cooking methods. More recently, the same search team used a similar approach to study the impact of roasting time and *in vitro* digestion on protein oxidation (Hu et al., 2018). An increase in fluorescence intensity was observed with increasing cooking time, which was attributed to the accumulation of Schiff-base structures and formation of carbonyl derivatives.

5.1.3. Application of fluorescence spectroscopy for monitoring fish authenticity

Fish authenticity is related to several aspects of great importance for consumers, including mainly the discrimination between fresh and frozen/thawed fish, the determination of geographical origin, and the distinguishing between fish species. However, our review of the literature shows that applications of fluorescence spectroscopy in the determination of fish species and geographical origin are still lacking.

Freezing is an efficient method of fish preservation, but this technique could affect fish structure and texture, and increase protein denaturation and lipid oxidation. Several techniques have been investigated to determine the impact of freeing and frozen storage on fish quality. In this regards, the potential of FFFS was investigated to discriminate between fresh and frozen thawed sea bass (*Dicentrarchus labrax*) (Karoui, Hassoun, & Ethuin, 2017). Four fluorophores including NADH (excitation at 340 nm), tryptophan (excitation at 290 nm) riboflavin (excitation at 380 nm), and vitamin A (emission set at 410 nm) were examined. The results showed that the technique was able to discriminate not only between fresh and frozen thawed fish, but also between frozen fish of different quality of raw material before storage. In another study, ElMasry and co-workers have developed a rapid method based on EEM coupled with some chemometric tools for screening fish products, even in their frozen state, in order to determine the initial freshness of these products before they are frozen (ElMasry et al., 2016, 2015; Higashi, ElMasry, & Nakauchi, 2016). For example, EEM in the range of 250–800 nm were recorded on horse mackerel (*Trachurus japonicus*) samples previously stored for 1, 4, 7 and 12 days before freezing. Some prominent and changeable fluorescence peaks were observed at some excitation and emission range. Although the authors did not give clear assignments of these fluorescence signals to specific fluorophores, they succeeded in obtaining a reasonable prediction ($R^2 = 0.89$) of freshness index (*K-value*).

Among the earliest applications of fluorescence spectroscopy to fish authenticity issues was the determination of contamination level of fish oils by dioxin and polychlorinated biphenyls (Pedersen, Munck, & Engelsen, 2002). In this study, the authors applied fluorescence spectroscopy in EEM mode to 88 fish oils samples and evaluated their results using some multivariate chemometric tools, such as PARAFAC and PLS regression. The results showed that the fluorescence spectroscopy was able to predict the dioxin contamination with a similar prediction error (below 1 ng/kg) as the references methods.

It should be stressed that research studies involving the use fluorescence spectra acquired in the EEM or SFS modes for the characterization of quality and authenticity of fish and other seafoods are limited. One of the rare studies reporting about the use of EEM fluorescence was conducted by Svensson and Andersen (2014) in the 260–650 nm range to assess brine composition of salted herring (*Clupea harengus*) during the ripening process (Svensson & Andersen, 2014). The decomposition of the fluorescence landscapes using parallel factor analysis (PARAFAC) as multi-way analysis revealed the presence of four fluorophores in the brine (two states of tryptophan, vitamin B6, and riboflavin). Partial least square (PLS) regression and N-PLS applied on unfold landscapes and raw fluorescence landscapes, respectively allowed the authors to predict protein concentrations in brine with the same accuracy as NIR (Svensson & Andersen, 2014).

5.2. Meat and meat products

Meat contains many fluorescent compounds such as tryptophan residues in protein, NADH, porphyrin, pyridinoline in collagen, lipid oxidation products, vitamin A and riboflavin (Sahar & Dufour, 2015). Overall, most studies concerning meat quality evaluation by fluorescence spectroscopy have focused on microbial spoilage detection and prediction of some quality parameters such as collagen, connective tissue, tenderness, fatty acids content, among other, while little has been published regarding meat authenticity.

5.2.1. Application of fluorescence spectroscopy for evaluating several quality parameters

Studies have been reported on the use of fluorescence spectroscopy to detect collagen contents in adipose and connective tissues of meat having significant importance for meat tenderness and textural properties. For example, in a study conducted by Egelandsdal and coworkers, fluorescence spectroscopy was used to detect collagen quantity in three different muscles namely pork *glutens medius*, beef masseter, and *latissimus dorsi* muscle (Bjørg Egelandsdal, Dingstad, Tøgersen, Lundby, & Langsrud, 2005). PLS regression analysis of spectral data revealed that collagen contents were present in the range of 1.3–4.0% with prediction error of 0.55%. In a previous trial, the same research team studied the potential of fluorescence spectroscopy (emission wavelength range = 360–500 nm, excitation wavelength range = 332–380 nm) to measure tenderness of beef *longissimus dorsi* muscles and revealed a moderately good (R = 0.45–0.84) correlation between Warner–Bratzler peak values and fluorescent emission spectra (Bøjrg Egelandsdal, Wold, Sponnich, Neegård, & Hildrum, 2002). The study concluded that fluorescent spectroscopy has moderately good potential to measure the tenderness of beef muscles. Likewise, Allais, Viaud, Pierre, and Dufour (2004) postulated that fluorescent

spectroscopy of tryptophan has the potential to monitor textural properties of meat emulsions and frankfurters prepared with different fat/lean ratios. Using canonical correlation analysis, the authors showed that fluorescent spectra of batter and frankfurters were highly correlated ($R^2 = 0.91$), and concluded that this technique can be effectively used to predict the texture of meat products (Allais et al., 2004).

Beside the texture, other quality parameters were evaluated using fluorescence spectroscopy. For example, the content of total volatile basic nitrogen (TVB-N), as a quality indicator of meat, was investigated using fluorescence hyperspectral imaging during cold storage of pork (Lee et al., 2018). The results showed an increase in fluorescence intensities with increasing TVB-N content, indicating a loss of meat quality during sytorage. For further understanding of quality degradation during storage, the authors created a distribution map with various colours representing the increase in TVB-N content with increasing storage time.

5.2.2. Use of fluorescence spectroscopy for monitoring thermal changes

When meat is subjected to thermal treatments, many changes occur such as denaturation of protein, degradation of some fluorophores (tryptophan residues in proteins, vitamins), and development of some new fluorophores (Maillard-reaction products, heterocyclic amines), which consequently changes the fluorescence signals of meat. In this context, SFS in combination with PARAFAC was used to investigate the effect of cooking kinetics on bovine meat samples taken from *Longissimus dorsi* muscle (Sahar, Boubellouta, Portanguen, Kondjoyan, & Dufour, 2009). Fluorescence spectra were recorded for meat cooked at 237 °C for 1–10 min by applying jets of superheated steam mixed with air. Fluorescence measurements were performed by fixing the excitation wavelength of 250–550 nm using an offset of 0–160 nm with an interval of 10 nm between excitation and emission wavelength. The results showed that the best PARAFAC model had two components which were attributed to Maillard reaction products and tryptophan residues generated from the effect of cooking. More recently, Sahar et al. (2016) reported the effectiveness of SFS in tandem with chemometrics for detection of thermal changes in meat during cooking process (Sahar et al., 2016). Purposely, bovine meat samples were cooked at 66, 90 and 237 °C for various time intervals (0, 1, 2, 5, 7, and 10 min) using the same cooking method (jets of superheated steam mixed with air). The fluorescence spectra were taken in the excitation wavelength range of 250–550 nm using offsets ($\Delta\lambda$) of 20, 30, 40, 50, 60, 70, 80, 90, 100, 110, 120, 130, 140, 150, and 160 nm between excitation and emission wavelengths. The study confirmed that fluorescence spectroscopy can be used as a rapid and non-invasive technique for detection of thermal changes in meat (Sahar et al., 2016).

However, cooking of meat products at high temperature and for long period may generate some levels of mutagenic/carcinogenic heterocyclic aromatic amines (HAA). Traditional methods used to measure these compounds are time consuming and labour intensive. Fluorescence spectroscopy has shown its efficiency to detect the production of HAA when meat is exposed to high cooking temperatures. For example, SFS was applied to detect the concentration of HAA in grilled meat at temperature of 237 °C for 5, 7 and 10 min, using PARAFAC and N-PLS regression (Sahar, Portanguen, & Dufour, 2010, pp. 803–812). Four HAA and their concentrations were successfully predicted from SFS data using two components of PARAFAC. In a similar approach, derivative-SFS technique was found to be successful for a rapid determination of benzo(*a*)pyrene, as one of the most carcinogenic polycyclic aromatic hydrocarbons in processed meat and fish samples (Li, Luo, Jia, Zhou, & Li, 2011).

5.2.3. Use of fluorescence spectroscopy for some authenticity issues

It was suggested that fluorescence spectroscopy can be used as a valuable tool for detecting some authenticity issues, such as differentiation between meat muscles, discrimination between fresh and frozen thawed meat, etc.

Fluorescence spectroscopy was successfully applied to differentiate between beef muscles on the basis of different chemical and rheological parameters (Sahar et al., 2009). In this study, seven bovine muscles were differentiated by using FFFS by taking emission spectra in the ranges of 305–400 nm, 340–540 nm, and 410–700 nm obtained by fixing the excitation wavelength at 290, 322, and 382 nm, respectively. It was shown that fluorescent spectra could classify various bovine muscles based on their protein, fat, collagen, and dry matter contents. Later, Sahar and Dufour (2015) reported similar potential of FFFS for classification and characterization of beef muscles. Three different beef muscles namely, *Semitendinosus*, *Rectus abdominis,* and *Infraspinatus* were analysed through fluorescent spectroscopy followed by treatment of data through partial least square discriminant analysis (PLS-DA) and PLS regression. The outcomes of the study revealed that fluorescent spectroscopy coupled with multivariate analysis offer considerable potential to characterize different beef muscles (Sahar & Dufour, 2015). Similar results were obtained recently (Aït-Kaddour et al., 2018), confirming the ability of fluorescence spectroscopy in performing discrimination between muscle samples.

Studies reported use of fluorescence spectroscopy for discriminating between fresh and frozen thawed meat are scarce. One of these studies investigated the potential of porphyrin fluorescence (excitation at 420 nm) as indicator of changes occurring during freezing storage and other storage conditions of pork slices (Durek, Bolling, Knorr, Schwägele, & Schlüter, 2012). One of the results obtained from this study was a decrease in fluorescence intensity of meat samples after freezing and thawing process.

5.2.4. Use of fluorescence spectroscopy for monitoring microbial growth

Fluorescence spectroscopy has also been widely used as a rapid and non-destructive detection of microbial spoilage of meat. In this context, Oto et al. (2013) used fluorescence spectroscopy in EEM mode to evaluate the microbial growth on aerobically stored (15 °C for 3 days) pork meat surface (Oto et al., 2013). The fluorescent intensity was measured in the range of 200–900 nm wavelength and two highest peaks were obtained, the first peak was observed at excitation = 295 nm and emission = 335 nm which is attributed to the tryptophan and the second peak was observed at excitation = 335 nm and emission = 450 nm that represents NADH (Oto et al., 2013). Recently, Durek et al. (2016) also investigated the microbial contamination on pork and lamb meat surface stored at 5 °C for up to 20 days by using FFFS. The results showed that fluorescence of NADH (excitation = 340 nm, emission = 370–600 nm) and porphyrin (excitation = 420 nm, emission = 550–750 nm) were correlated to the growth of diverse bacteria. Besides, a rapid increase (from the 2nd day of storage) in porphyrin fluorescence for lamb meat was observed and attributed to the high activity of microorganisms on the meat surface compared with the pork meat (Durek et al., 2016). Another study conducted by Aït-Kaddour et al. (2011) explored the potential of portable spectrofluorimeter (Fig. 5) to detect microbial spoilage of minced beef stored at 5 and 15 °C under two packaging conditions (vacuum and aerobic) and concluded that spectrofluorimeter is a promising device to predict microbial spoilage ($R^2 = 0.50–0.99$) of minced meat (Aït-Kaddour et al., 2011). More recently, FFFS was applied in EEM mode with excitation wavelengths in the range of 200–500 nm and emission wavelengths ranging from 200 to 900 nm for prediction of aerobic plate count on sliced beef samples (Mita Mala et al., 2016). Several fluorescence peaks were observed corresponding to five intrinsic fluorophores, including tryptophan, NADH, vitamin A, porphyrins, and flavins. The results showed that the coefficient of the PLSR model was relatively high ($R^2 = 0.83$), suggesting that this method has the potential to predict microbial spoilage of meat in a rapid and non-destructive method. The above-mentioned studies have shown that fluorescence spectroscopy has a significant potential to detect microbial spoilage of meat and this technique can be efficiently used at commercial level for

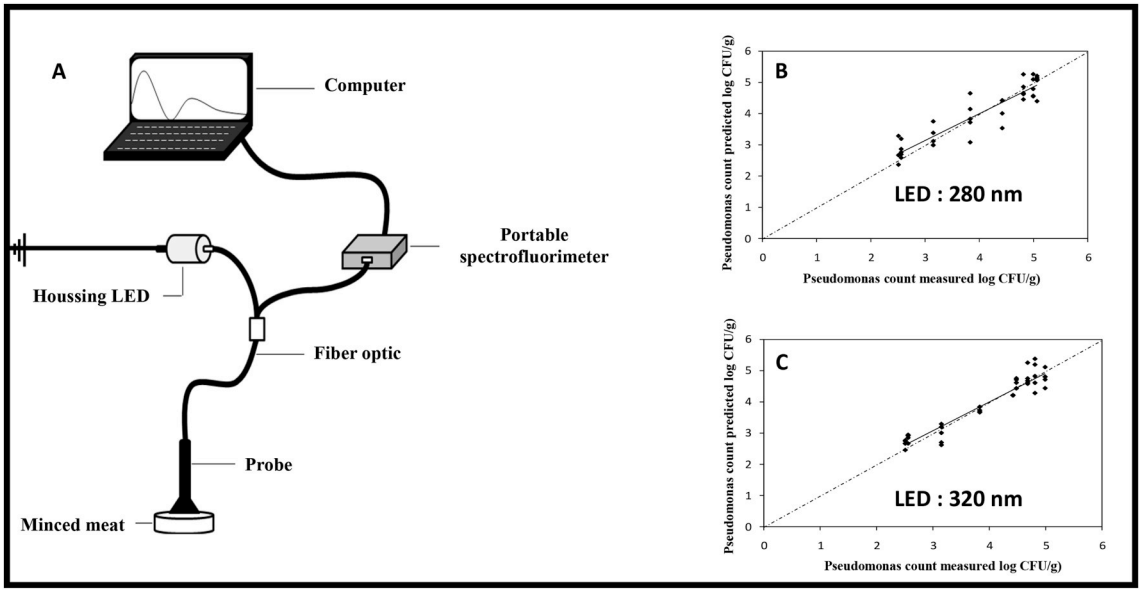

Fig. 5. Portable spectrofluorimeter (A) and PLS regression models (B and C) of *Pseudomonas* count obtained after vacuum packaging of minced meat stored at 5 °C.

online prediction of microbial safety of meat products under varied storage conditions.

5.2.5. Evaluation lipid and protein oxidation by fluorescence spectroscopy

Oxidative deterioration of lipid and protein is one of the prime non-microbial factors involved in quality degradation and spoilage of meat (Guyon et al., 2016). Several studies confirmed the ability of fluorescent spectroscopy to detect oxidation of meat products (Pouzo, Zaritzky, Pavan, Rossetti, & Descalzo, 2016; Veberg et al., 2006; Wold, Mielnik, Pettersen, Aaby, & Baardseth, 2002). Most of these studies used correlation between thiobarbituric acid reactive substances (TBARS) value and fluorescent spectral data to predict the oxidative stability of meat products during different processing and storage conditions. For instance, Veberg et al. (2006) explored the potential of fluorescence spectroscopy for rapid detection of oxidative damage of pork and turkey meat samples stored for 7 and 12 days under various conditions of light exposure and oxygen availability (Veberg et al., 2006). In more details, high oxygen modified atmosphere and vacuum-packed meat samples were subjected to spectroscopic analysis (excitation = 382 nm, emission = 410–750 nm) and some traditional measurements (sensory and lipid oxidation analysis). From a technological point of view, it was reported that the turkey samples packed in high oxygen were oxidised only after 7 days of storage, while from an analytical point of view, the results revealed higher correlations of fluorescence spectra and porphyrin contents during the storage (Veberg et al., 2006). The obtained results were confirmed later by Gatellier and other (2007) who reported the possibility of evaluating lipid oxidation in three chicken genotypes by FFFS (Gatellier et al., 2007). The findings showed correlations between TBARS, as a reference lipid oxidation index, and fluorescence mesurements, as a rapid and valuable method. In a similar investigation (Gatellier et al., 2009), the relationship between fluorescence and lipid oxidation was studied during cooking of bovine meat (*Longissimus thoracis*). In this study, the authors reported a high correlation between TBARS and fluorescent pigments formed because of interaction between proteins and aldehyde products.

In addition to lipid oxidation, fluorescence spectroscopy can also be used for the detection of protein degradation and oxidation in meat samples. For instance, Armenteros, Heinonen, Ollilainen, Toldra, and Estévez (2009) reported the use of fluorescence spectroscopy to investigate the production of protein carbonyl during processing. The excitation wavelength was set at 350 nm and emission spectra were recorded in the range of 400–500 nm wavelength. The study demonstrated that fluorescent spectra can be employed as a useful tool to detect protein oxidation in meat samples (Armenteros et al., 2009). In another study, fluorescence emission spectra obtain after excitation set at 295 nm were used as indicator of oxidation of sarcoplasmic proteins during processing of Cantonese sausage (Sun, Zhao, Yang, Zhao, & Cui, 2011). The results showed a gradual decrease in fluorescence intensity with increasing processing time, which was attributed to structural unfolding of the sarcoplasmic proteins. In a recent study, the authors used tryptophan fluorescence of porcine myofibrillar protein to obtain information about protein oxidation and modifications in secondary and tertiary structure of the proteins induced by different thawing methods (F. Li et al., 2019). Increased fluorescence intensity was observed for thawed samples, especially for microwave-thawed and water-thawed ones, indicating more exposure of tyrosine and tryptophan in proteins of these samples to a hydrophilic environment, compared to other thawing methods.

6. Modelling fluorescence data of intact foods

As can be noticed from our literature review, several research groups applied multivariate chemometrics methods for calibration and modelling fluorescence data from fish and meat and other intact foods without referring to the circumstances in which fluorescence measurements were obtained. Hence, one must be careful when using the chemometrics methods to fluorescence data measured from intact food samples, such as fish or meat. Indeed, fluorescence spectra measured from such biological samples are very often distorted due to absorption and scattering of the exciting light known as primary inner filter effect, as well as reabsorption of the fluorescent light known as secondary inner filter effect (Lakowicz, 2006). Additional dips or shoulders could occur in the measured emission and excitation spectra at the position of absorption bands of natural chromophores, such as hemoglobin. The inner-filter effect is immensely strong in the spectra from biological media because multiple scattering increases the path lengths of photons inside the sample (Zhadin & Alfano, 1998). In this case, fluorescence emission intensity is not proportional neither to the amplitude of the fluorescence excitation wavelength or to the concentration of the fluorophores, whereas all multivariate chemometrics models are multilinear. Lakhal, Acha, and Aussenac (2012) used Monte Carlo method to simulate fluorescence data originating from mixtures of three known fluorophores embedded in different absorbing and scattering environments (Lakhal et al., 2012). It has been shown that the fluorescence

spectral information is severely altered by multiple scattering and re-absorption events in the surrounding medium, thereby preventing the consistent resolution and quantitative determination of the mixture by using multivariate data analysis methods such as PARAFAC and multivariate curve resolution–alternating least squares (MCR-ALS). Therefore, fluorescence studies in undiluted biological samples like fish and meat require that inner filter effects be minimized or compensated for before a qualitative and quantitative analysis can be made. Contrary to what is often stated in the literature (Møller Andersen & Mortensen, 2008), Lakhal et al. (2012) have proved that the acquisition of spectra from turbid media with front-face fluorescence detection mode does not necessarily guarantee the conditions of linearity.

The inner filter effects were studied for a long time now and several models for correcting the fluorescence emission spectra for inner filter effects were proposed (Credi & Prodi, 1998; Gu & Kenny, 2009; Larsson, Wedborg, & Turner, 2007; Luciani, Mounier, Redon, & Bois, 2009; T.; Wang et al., 2017). However, the formulation of these models assumes that the fluorescence spectra are measured in non-scattering media and the light intensity decreases exponentially along the axis of propagation. Unfortunately, these assumptions do not hold in the context of fluorescence spectroscopy of biological samples. In 2014, Lakhal and co-authors proposed Monte Carlo based approach to remove the effects from EEM of biological tissue-equivalent phantoms and recover the ideal EEM. This model is suitable for deconvolving absorption and scattering from fish and meat fluorescence emission spectra (Lakhal et al., 2014).

7. Limitations and future trends

Based on the above studies, it was fully demonstrated that fluorescence spectroscopy, as a rapid and non-invasive technique, would be a promising tool for practical applications in quality evaluation of fish and meat products in the future. Moreover, the technique enables valuable technological information to be obtained during storage of these products under different preservation conditions. However, some limitations and obstacles are still observed and need to be overcome before the implementation of fluorescence spectroscopy in the fish and meat industrial sectors.

One of the main drawbacks of fluorescence spectroscopy is that not all materials can be excited to fluorescence due to the lack of intrinsic fluorophores. On the other hand, presence of several fluorophores in the examined samples may lead to overlapping peaks, which makes identification of specific fluorophores more complicated (Xiaobo et al., 2016). As fish and meat products are heterogeneous matrices, variations in fluorescence analysis may be occurred, depending on the location of the measurements. To overcome this limitation, multispectral and hyperspectral imaging techniques have been developed to obtain both spectral and spatial information on a large part of the analysed samples, which are more representative of the initial fish or meat product. Although this emerging technology has been widely used in VIS/NIR spectroscopy (reflectance/transmittance) (Cheng & Sun, 2014; Feng et al., 2018) and Raman spectroscopy (Lohumi, Kim, Qin, & Cho, 2017; Yaseen et al., 2017), very limited applications are available concerning the fluorescence hyperspectral imaging. Therefore, this could be a focal area of research in the coming years to obtain detailed chemical images along with the spectral data.

Response time is an important parameter to be considered when choosing the fluorescence acquisition mode; while a spectrum can be measured in terms of seconds with fixed excitation or emission wavelength (classical mode), response times are clearly longer in the case of the synchronous or EEM modes. Generally speaking, most of the studies on fish and meat products have been conducted using the classical mode, whiles only a few investigations were performed using the synchronous and EEM modes, especially for seafood products. Additionally, in comparison with traditional techniques, this technique seems to be a very selective and sensitive analytical method, and it is even far more sensitive than the other spectroscopic techniques. Nevertheless, the extreme sensitivity of this technique could be considered as a double-edged sword since the fluorescence measurements can be influenced by several parameters due to the product composition complexity (e.g. energy transfer between fluorophores …) and to the industrial environment (e.g. temperature, vibration …). This is probably the reason why the applications of fluorescence spectroscopy for quality and authenticity analysis of fish and meat products are still limited compared with other spectroscopic techniques, particularly NIR spectroscopy.

Besides, apart from some applications for discrimination between fresh and frozen products, no studies have been found in the literature about the use of fluorescence spectroscopy for other authenticity issues such as determination of geographical origin or discrimination between the species. Therefore, more research is needed, and application fields should be enlarged in this respect to make the use of fluorescence spectroscopy available optimally. Technological advancements in fluorescence spectroscopy technique, as well as development in chemometric multivariate techniques and their rapid implementation are expected for the near future, allowing further progress of such fingerprint technique towards the development of an online or in-line system for real-time applications in the field of fish and meat products.

8. Conclusion

The high perishability of food muscle requires both adequate methods for food preservation and rapid methods for quality determination. This review has covered some of the recent applications of fluorescence spectroscopy for quality evaluation and authenticity of fish and meat products. Depending on the study's objective and equipment availability, fluorescence spectroscopy can be used in different acquisition modes in fish and meat products, even though more efforts are still needed in order to choose the most suitable acquisition method. In addition, more research is needed to identify the exact fluorophores responsible for fluorescence properties and the factors affecting fluorescence phenomenon in such complex systems. Thus, it is expected that fluorescence spectroscopy will continue to be a dynamic research field over the coming years to overcome the few limitations of this technique and making it an interesting prospect for future industrial applications.

From the information available in the literature, it could be concluded that fluorescence spectroscopy can be used in many applications of quality as well as some authenticity issues of fish and meat products, in a rapid and non-destructive manner. The results of studies reviewed during this work are encouraging enough to promote and expect the future applications of fluorescence spectroscopy in the food industry.

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