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Effect of frying process on furan content in foods and assessment of furan exposure of Spanish population

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ABSTRACT

Furan content in eight bread-coated frozen foods (ham croquettes, squid rings, tuna pasties, churros, nuggets, fish fingers, onion rings and san jacobos) deep-fried in fresh and reheated olive oil, and in five cooked vegetables was evaluated. Deep fried foods showed the highest levels of furan between 12 μg kg^{-1} (tuna pasties) and 172 μg kg^{-1} (onion rings), with a furan increase tendency when reheated oil was used. In vegetables, furan was only found at low level in griddled onion (3.5 μg kg^{-1}). The lower temperature applied (<150 °C) in comparison to that of the deep-fried foods (190 °C), the furan volatilization during longer time cooking (15 min vs 6 min) together with the food composition differences might explain the low furan content in vegetables. As a preliminary approach for risk assessment, the margin of exposure (MOE) was calculated. The total daily intake of furan by Spanish population (239–4372 ng/kg bw/day) with MOEs below 10,000 indicates a human public health concern. However, MOEs for fried foods showed that furan could suppose a possible health risk only in people with a high consumption of these products. Nevertheless, further studies should be developed to provide furan exposure data of other fried foods.

1. Introduction

Frying is a culinary process applied to a great variety of foods. Innovation in the food industry with the development of new food products associated to social changes in Western countries have increased the consumption of a great variety of time-saving "ready-to-heat" frozen foods. In Spain, the consumption of pre-cooked frozen foods, most of them bread-coated, increased by 7.8% in the period 2001–2006 (MAGRAMA, 2006). When considering both household and catering and institutions consumption, recent data (2014) indicate that around 12.3 kg per capita per year of ready-to-serve foods (including pre-cooked frozen foods) were consumed in Spain, increasing every year (0.4% higher than in 2013) (MAGRAMA, 2014). Croquettes and pasties account for around 20% of precooked foods with high amount of cereals. Churros are also a typical Spanish fried food product with a high consumption among cereal products (1.32 g/capita/day) (AECOSAN, 2011). Other commonly consumed foods in the Mediterranean diet, and particularly in the Spanish cuisine, are vegetables, such as onions or peppers, which are often subjected to a frying process for further uses as base ingredients or garnish.

Fried food palatability is related to unique sensory characteristics, including brown colour, crunchy texture and other desired flavour and taste, mainly due to Maillard reactions (Rossell, 2001). Frying process induces significant changes in food such as water loss, melanoidins formation, increase of fat amount, and changes in the fatty acid profile due to the mass exchange between frying media and the fat of food (Miranda et al., 2010; Romero, Cuesta, & Sanchez-Muniz, 2000; Sanchez-Muniz, Viejo, & Medina, 1992). Maillard reaction also induces the formation of volatile compounds that provide the characteristic aroma and flavour of roasted and fried foods. Among them, furan and furanic compounds can significantly contribute to the sensory properties of heat treated foods (Anese & Suman, 2013; Maga, 1979). However, furan is a highly volatile compound, which has been classified as a possibly carcinogenic to humans (group 2B) by the International Agency for Research on Cancer (IARC, 1995). The Joint FAO/WHO Expert Committee on Food Additives estimates that furan exposure through diet is confirmed as a public health problem (JECFA, 2010). Therefore, Food Safety Agencies promote furan data collection in foods (EFSA, 2010; US FDA, 2008).

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Coffee (for adults) and commercial baby foods (for infants) have been proposed as the major contributors to furan exposure (Fromberg, Fagt, & Granby, 2009). Some authors have studied the risk assessment of furan in these products (Lachenmeier, Reusch, & Kuballa, 2009; Waizenegger et al., 2012), however other cooked foods could also contribute in a high extent to furan exposure due to the fact that furan formation can be influenced by the heat treatment conditions (Fromberg et al., 2009). Carbohydrate degradation, pyrolysis of sugars, decomposition of ascorbic acid and oxidation of polyunsaturated fatty acids during heat treatment can promote furan generation (Becalski & Seaman, 2005; Limacher, Kerler, Conde-Petit, & Blank, 2007; Limacher Kerler, Davidek, Schmalzried & Blank, 2008; Märk, Pollien, Lindinger, Blank, & Mark, 2006; Perez Locas & Yaylayan, 2004; Owczarek-Fendor et al., 2011). Some authors suggest that carbohydrate foods are more prone to the formation of furan, probably due to the Maillard reaction and that the retention of furan in foods is mainly caused by the lipid fraction, especially polyunsaturated fatty acids (Arisseto, Vicente, Ueno, Tfouni, & Toledo, 2011; Fromberg et al., 2009). So that, it may be expected that the content of furan in foods subjected to a frying process, especially those rich in carbohydrates, could be high. Nevertheless, an EFSA report highlights that only 8% of the furan data were reported after food preparation and it claims that future testing of furan should preferably analysed both as purchased and as consumed indicating the exact cooking preparation conditions (time, temperature and handling label information) (EFSA, 2010). Therefore, the main aim of this work was to evaluate the furan content in some of the most common Spanish fried foods (AECOSAN, 2011), both bread-coated frozen foods cooked by deep-frying and selected vegetables slowly fried in oil and commonly used as base ingredients for cooking some typical dishes in Spanish cuisine. The use of reheated oil for frying is also a common culinary practice. Thus, furan occurrence in foods fried with fresh or reheated oils was also tested. With the obtained results, a preliminary approach for risk assessment of furan in fried foods for Spanish population has been conducted.

2. Material and methods

2.1. Food samples and reagents

Three packages of different lots of frozen precooked foods (ham croquettes, squid rings, tuna pasties, churros, nuggets, fish fingers, onion rings and san jacobos) were obtained from a local supermarket. According to food labels of frozen precooked products, carbohydrates were the most abundant nutrient (19.4–30.0 g/100 g), followed by fat (0.7–16.3 g/100 g) and proteins (<10 g/100 g). Three different batches of vegetables (yellow onion, green pepper, cardoon, cabbage and chicory), as well as sunflower oil and olive oil (refined and virgin olive oil blend) were obtained from local stores. Furan and d4-furan, as well as sodium chloride were purchased from Sigma–Aldrich Chemical (Steinheim, Germany). The methanol (HPLC grade) was purchased from Panreac (Barcelona, Spain).

2.2. Standard solutions

Stock solution of d4-furan was prepared by adding 25 μL of d4-furan to 10 mL of methanol in a vial. A 2.5 μg mL^{-1} water working solution was prepared daily. Stock and working solutions of furan were prepared using the same procedure for d4-furan. Then, six calibration standard solutions at concentration ranging from 0.001 to 0.02 μg mL^{-1} were prepared by adding the appropriate amount of furan water working solution (2.5 μg mL^{-1}) into a 20 mL vial containing 3 g of NaCl and 5 mL of deionized water. In addition, 40 μL of the d4-furan water working solution (2.5 μg mL^{-1}) was added to each calibration solution as internal standard.

2.3. Food samples preparation

Eight frozen precooked foods (ham croquettes, squid rings, tuna pasties, churros, nuggets, fish fingers, onion rings and san jacobos (ham and cheese in breadcrumbs)) were deep fried in olive oil using a domestic deep fryer Princess 180710 (DOSEFES S.A., Barcelona, Spain) at 190 °C during 6 min. Temperature was checked with a digital thermometer type J/K Fluke 51 (Fluke, USA). In order to study the effect of the reuse of oil in the formation of furan, 20 times reheated olive oil was also employed. Oil polar compounds were measured with a quality-meter frying oil FOM-320 (Ebro Electronic, Ingolstadt, Germany).

Chopped vegetables (yellow onion, green pepper, cardoon, cabbage and chicory) (300 g) were fried with olive or sunflower oils (30 mL) at 115 °C for 10 min in a frying pan. Then, temperature was decreased to 108 °C for 5 min. Chopped vegetables were also submitted to heating at 150 °C for 10 min and then at 110 °C for 5 min in a griddle without oil addition.

2.4. Furan analysis

Furan content was analysed following the method described by Perez-Palacios, Petisca, Melo, and Ferreira (2012) with modifications. Samples were grinded with a fork until homogenization in an ice bath to avoid furan losses. Immediately after, 2 g of each sample were transferred to a 20 mL vial containing 3 g of NaCl and 5 mL of deionized water. For the oil samples, 2 mL of oil were transferred to the vial containing 3 g of NaCl. A volume of 40 μL of working solution of d4-furan (2.5 μg mL^{-1}) was added as internal standard to each vial which was immediately closed. Afterwards, the vial was sonicated for 15 min and stored at 4 °C until further analysis (<24 h). Each sample was prepared in triplicate.

A SPME fiber (Supelco Co., Canada) coated with carboxen/polydimethylxilosane (CAR/PDMS) 75 μm was used. The fiber was exposed to the headspace of the sample during 40 min at 37 °C. The SPME fiber was desorbed at 280 °C for 10 min in a HP6890 GC System gas chromatograph (Agilent Technologies, Palo Alto, CA) coupled to a mass selective detector (MS) (model 5973, Agilent Technologies). Volatiles were separated using a column HP PLOT/Q (30 m length × 0.32 mm internal diameter × 0.20 μm thickness). The carrier gas was helium at a flow of 1 mL min^{-1}. The temperature program was 40 °C for 5 min, then raised at 3 °C min^{-1} to 120 °C, and finally, raised at 10 °C min^{-1} up to 220 °C and held for 5 min. The GC–MS transfer line temperature was 270 °C. The MS operated in the electron impact mode with an electron impact energy of 70 eV and a multiplier voltage of 1247 V and collected data at a rate of 1 scan s^{-1} over a range of m/z 35–350. Ion source temperature was set at 230 °C. Furan and d4-furan were identified by comparing their retention time and their mass spectra with those of standard compounds and NIST 05L library. Selected ion monitoring (SIM) was used for the detection of furan and d4-furan, using m/z 68 and m/z 72, respectively. Furan was quantified using d4-furan as an internal standard by calibrate curve method.

The method was validated by obtaining a linear relationship between the concentrations of furan and the respective area ratio between m/z 68 and m/z 72 ($r = 0.999$). Results for repeatability showed a good precision of the method with coefficient of variation values below 5%. Taking into account that furan is a highly volatile compound, a narrow dispersion of values was also observed for intermediate precision, with coefficients of variation between 3.28 and 16.19%. The limit of detection (LOD) and limit of quantification (LOQ) were also calculated, obtaining 0.7 and 2.3 μg furan kg^{-1}