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Packaging contaminants in former food products: Using Fourier Transform Infrared Spectroscopy to identify the remnants and the associated risks

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HIGHLIGHTS

- The presence of packaging contaminants was investigated in former food products.
- The Fourier Transform Infrared Spectroscopy coupled with a microscope was used.
- Plastic, cellulose and aluminum particles were detected in former food products.

GRAPHICAL ABSTRACT

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ABSTRACT

Food waste and feed-food competition can be reduced by replacing traditional feed ingredients such as cereals, with former food products (FFPs) in livestock diets. These foodstuffs, initially intended for human consumption, are recovered, mechanically unpacked, and then ground. Despite this simple and inexpensive treatment, packaging contaminants (remnants) are often unavoidable in the final product. To maximize the exploitation of FFPs and to minimize the associated risks, packaging remnants need to be quantified and characterized. This study tested the efficacy of the Fourier Transform Infrared Spectroscopy coupled with an optical microscope (μFT-IR) in identifying packaging remnants in 17 FFP samples collected in different geographical areas. After a visual sorting procedure, presumed packaging remnants were analyzed by μFT-IR. The results showed significant differences ($p < 0.05$) between the FFPs in terms of the total number of foreign particles found (plastics, cellulose and aluminum remnants, ranging from 4 to 19 particles *per* 20 g fresh matter), and also regarding the number of cellulose and aluminum particles. These data clearly demonstrate the need for sensitive instruments that can characterize the potential contaminants in the FFPs. This would then help to reduce the overestimation of undesirable contaminants typical of simple visual sorting, which is currently the most common method.

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1. Introduction

Despite the huge need for natural resources in our modern society, there is still significant food waste [1]. An effective animal nutrition strategy could be a useful means to reduce food losses. In fact, food industry leftovers re-used as alternative ingredients in animal diets are increasingly being used by animal-feed producers [2-4]. Surplus food is thus not considered as waste but as an existing resource rich in nutrients which can be recovered and enhanced by becoming part of the food chain again [5,6].

Food leftovers, also called former food products (FFPs), include all those foods produced by the baking industries (e.g. bread, pasta, salty cakes) and confectionery industries (e.g. biscuits, chocolate, snacks, cakes, breakfast cereals [2]). Due to logistical or production errors, surplus problems in the food supply chain, or problems with packaging, these products are no longer suitable for sale and human consumption [7].

According to the feed legislation and the European Commission, the legal status of FFPs is different from food waste [8,9]. Food waste cannot be returned to the food chain, while FFPs are still microbiologically safe and rich in valuable nutrients [5,6,10–12].

FFPs are mechanically unpacked during their processing, before being included in the animal feed. There are different types of mechanical packaging removal such as sieving, magnetic attraction, air flows and density methods, as described by Van Raamsdonk et al. [13]. The mechanical process removes large pieces of packaging, while smaller residues inevitably remain in the feed [10,14].

Packaging materials are often characterized by complex compositions [15] because each type of food requires a specific type of protection, which plays an important role in food safety. The packaging layer acts as a physical barrier to ensure product preservation from any chemical, physical or biological hazard, and to extend its shelf-life as long as possible [16]. Among all the food packaging materials, plastics are the most commonly used materials, followed by paper and pressure paperboard, regenerated cellulose, resin and aluminum foil [5,6,17]. The packaging is essential to guarantee the quality and the shelf-life of products during transport and storage; however these materials need to comply with specific laws. European Regulation (EC) 1935/2004 states that the packaging materials must not release their constituents in order to safeguard human health [13,18].

Assuming that food is safe for humans, it should also be safe for animals, therefore packaging remnants are not accepted as feed ingredients as stated by article 6 of Regulation (EC) 767/2009 of the European Parliament and of the Council of 13 July 2009 [5,6,19,20]. Although animals should be protected from consuming potentially harmful materials, it is difficult to apply zero tolerance concerning the presence of packaging in FFPs. However, a sufficiently low-level risk is achievable [13]. To minimize the risk, some control authorities such as the German Federal Ministry of food, agriculture and consumer protection, have established the maximum tolerance limit for the presence of packaging remnants in animal feed at about 0.125% (wet weight; *w/w*). Although a level of 0.15% (*w/w*) is often unavoidable, it is negligible regarding safety issues [11,13].

To ensure the sustainability and the safety of FFPs in feed, the possible presence packaging remnants always needs to be monitored [10,14]. Several studies have applied different methods to detect packaging remnants in FFPs [10–13,20,21]. The RIKILT institute (Institute of Food Safety, Wageningen University and Research Center, Netherlands) validated a method for establishing the level of packaging contamination in FPPs. Although it was not possible to formally identify the original packaging materials, 90% of the analyzed FFP samples showed a level of ''presumed residuals'' of below 0.15% *w/w* [13,20].

Stereomicroscopes have also been used to detect remnants. However, operator inspection can be time-consuming, unpredictable, and inconsistent, because the results depend on the ability of the operator to correctly visually recognize and quantify the different types of remnants [11]. As reported by van Raamsdonk et al. [13], the difficulty of the operator depends on the particle size. Particles smaller than one mm are difficult to see and results can be affected by the level of training of the operator.

In a previous study [11] we used Computer Vision (CV) coupled with a stereomicroscope for image acquisition. This led to a fast-qualitative screening to estimate the presence of foreign materials in feed. Although CV seemed to be able to differentiate between contaminated and uncontaminated samples, it could only be applied when images of FFP samples were obtained using a stereomicroscope. Moreover, the CV method did not detect all kinds of packaging remnants [10,11]. We also studied the application of the electronic nose to recognize the presence of presumed foreign materials in FFP samples [14]. The e-Nose was able to differentiate between clean FFP samples and contaminated FFP samples when the feed matrix was characterized by a low variability (e.g. same producer company, same odor background). However, when different packaging materials were involved, the e-Nose was less effective [14].

A feasibility study [10] proposed the Multivariate Image Analysis method coupled with imaging methods (analysis of red, green, and blue (RGB) images of FFP samples), to rapidly detect presumed packaging remnants. This method was also useful when the color of the foreign material could be distinguished from the color of the FFP matrix. All these results, however highlight that the different methods have some limitations when identification/characterization and especially quantification of the presumed contaminants of the packaging remnants is required.

The first aim of this study was thus to shed light on another potential source of environmental contamination due to specific pollutants that should normally be recycled or disposed of separately and which instead enter the environment through a particular food supply chain. This was achieved through the evaluation of the effectiveness of Fourier Transform Infrared Spectroscopy coupled with an optical microscope (μFT-IR) to characterize the nature of the remnants and investigate their origin, composition and quantity, at the same time guarantying the traceability, quality and safety of the FFPs.

Many studies have shown that μFT-IR spectroscopy is a reliable method to characterize and quantify plastics in aquatic ecosystems [22–24], additives and chemical contaminants in food [25], food chemical components [26], plastic packaging additives [27] and organic molecules for food packaging [28]. However, none of these studies have considered animal feed. This is thus, to the best of our knowledge, the first study in which the potential of μFT-IR spectroscopy to also characterize packaging remnants in FFPs has been tested.

2. Materials and methods

The procedure followed in this study for the recognition and characterization of packaging remnants in FFPs is divided into two steps: i) extraction of packaging remnants from the different FFP samples; ii) analysis of each extracted contaminant using μFT-IR.

2.1. FFP sample preparation

Seventeen FFP samples obtained from both European and *non*-European former food processors were analyzed. The samples, did not represent the entire range of FFPs potentially processed in different areas. However, to ensure as wide a range as possible, samples were obtained from five different countries and from different processing plants. All of the samples were collected in cooperation with FFP processing plants, and feeding stuff manufacturers in 2021. Due to rights issues, the authors agreed not to identify the sample sources (in terms of both the region/country and processing plant).

After transportation to our laboratory, samples were stored at a refrigerated temperature (+ 4 °C) in glass containers to prevent contamination. The final lab samples had different weights depending

on the quantities made available by the companies. To perform the analysis, each lab sample was stirred/mixed and picked up to obtain random tests samples. In total, 60 g *per* type of FFP were analyzed. Each aliquot was placed in a Petri dish and analyzed separately by visual sorting using a stereomicroscope (OLYMPUS SZX9) and tweezers, avoiding any external contamination. Sampling was carried out in three technical replicates for each type of FFP. Each replicate was composed of 20 g of feed. The identification of an undesirable substance or of a legally applied ingredient relies heavily on the expertise of the laboratory staff [29]. In this study, the laboratory staff were trained specifically to improve their skills in feed visual examination. All foreign materials suspected of being cellulose, plastics and aluminum were extracted, and transferred into closed Petri dishes and labeled. Throughout the visual sorting procedure, in order to monitor any environmental contaminants and to prevent any accidental contamination of the samples, a cellulose nitrate membrane filter was placed in the workstation as a blank for each group of technical FFP replicates.

2.1.1. Extracted remnant preparation

Depending on their origin, FFPs can be very rich in lipids, oil starch and sugars [3]. Packaging materials can be soiled by oils and sugars, and thus a layer of fat and oil, forming a sort of patina, can cover the remnants. Before proceeding with their analysis and characterization, the extracted remnants were washed to prevent organic matter from being identified by the instrument instead of their actual chemical nature.

Each foreign remnant was defatted with a specific detergent (Triton X-100, 1:4 dilution *v/v*) and then rinsed in several steps with ultrapure water [30]. The cleaned remnants were then placed on clean cellulose nitrate membrane filters (Sartorius™ 50 mm) inside closed Petri dishes, which were labeled with the corresponding feed name. To count the number of remnants for each 20 g-aliquot, each particle was numbered on the filter (Fig. 1A).

2.2. μFT-IR analysis: quantification and characterization of foreign remnants

After extraction, washing, drying and placing the particles on filters, their chemical composition and size were analysed using the μFT-IR (Spotlight 200i equipped with a Spectrum Two microscope by Perkin Elmer; Fig. 1B).

Infrared spectra were obtained in attenuated total reflectance (ATR) mode with 32 scans and wavelengths between 600 and 4000 cm^{-1} and analyzed using the Spectrum 10 software (Perkin Elmer). The spectra obtained were matched with standard spectra using the Perkin Elmer libraries. Only the spectra with a matching score of ≥ 0.70 were accepted.

In addition, the degree of similarity between the measured samples and the reference spectra was considered reliable only after visual analysis of the spectrum peaks by the operator to prevent identification errors [22–24]. At the end of the procedure, the environmental particles found on the cellulose nitrate membrane filter used as blanks (one for each replicate) were also counted and characterized by μFT-IR. According to the spectra obtained (Figs. S1−S4), particles were classified as: (i) cellulose and (ii) plastics. Plastics were also specifically sub-classified *per* type of polymer (e.g. polypropylene-PP, polyethylene-PE) and their colors were registered.

Given that the instrument cannot obtain spectra from aluminum, due to its reflectance property, identification and counting of aluminum particles were based only on visual inspection.

Using ImageJ, each remnant was also characterized based on its size, measuring only the length (mm) of the largest dimension. In addition, the mean particle size of the total remnants and the mean size *per* category (cellulose, plastic, aluminum) were calculated. The definition of plastics (micro-, meso-, macro plastic) based on size was evaluated in accordance with the size classification proposed by Hartmann et al. [31].

2.3. Statistical approach

The results were analyzed by one-way ANOVA with R software (v 4.0.5). For pairwise comparisons, the Sidak function was performed, which is a modified post-hoc Tukey test for multiple comparisons of means. Statistical means and their standard deviation (SD) were calculated with the lsmeans function from the package “emmeans”. Residuals of Lmer models were checked for normality and homoscedasticity.

3. Results and discussion

3.1. Detection of packaging remnants

The box plots of the mean and median number of total remnant particles for each FFP sample are reported in Fig. 2. Samples from FFP10 to FFP14 showed the highest number of foreign remnants obtained by visual sorting. An intermediate number was identified by samples FFP1, FFP2, FPP4, FFP6, FFP8, FFP15. By contrast the remaining samples were characterized by a low level of contamination (i.e. FFP3, FFP5, FFP7, FFP9, FFP16, FFP17). The results thus suggest that approximately two-thirds of the samples were contaminated, whereas the remaining

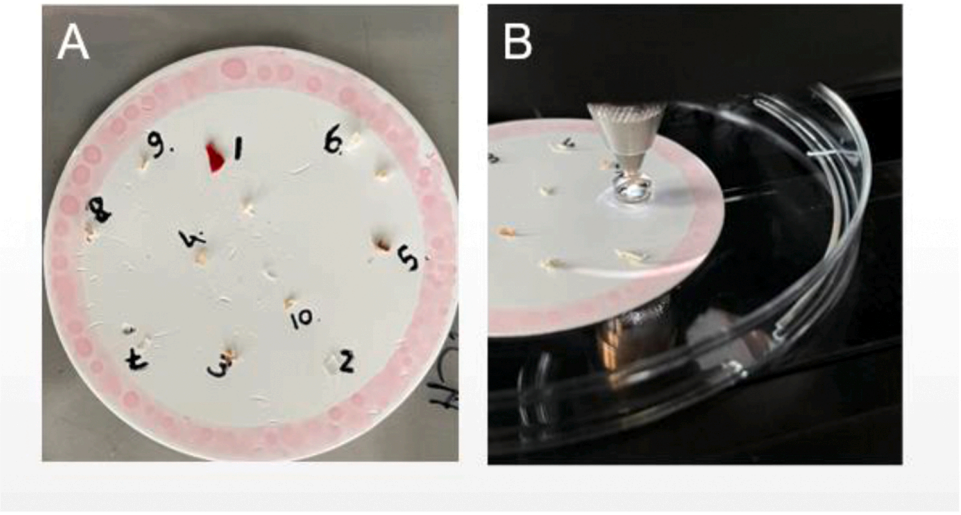

Fig. 1. Numbered materials, after the visual sorting procedure, on cellulose nitrate membrane filter (A) and μFT-IR characterization of particle chemical nature (B).

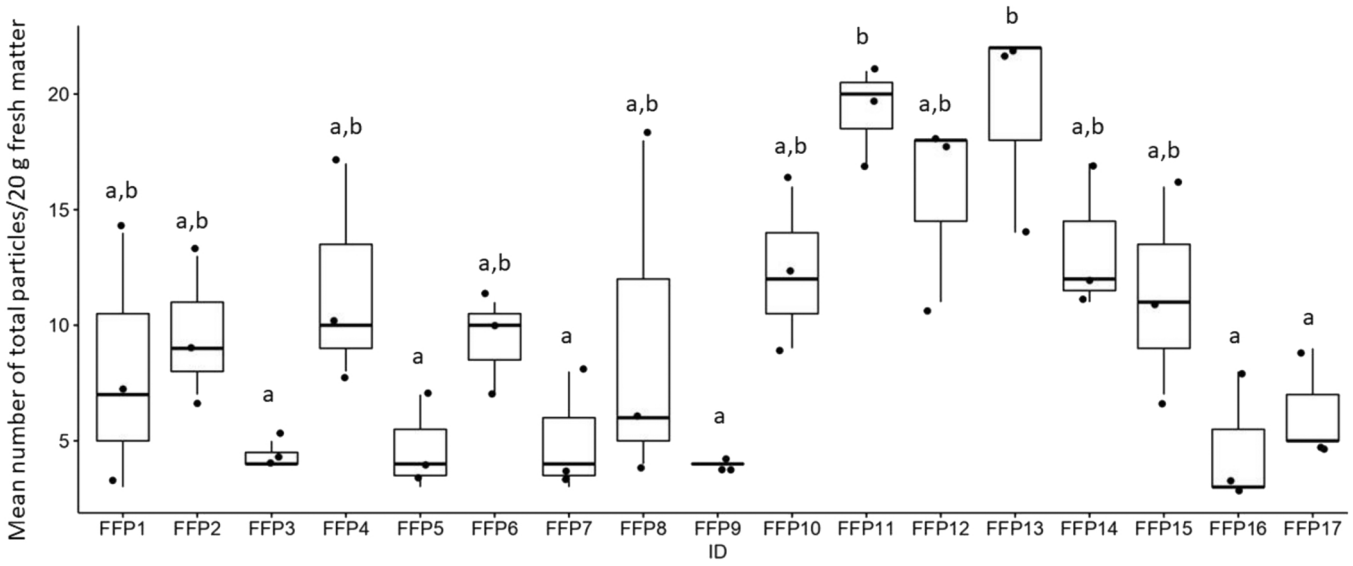

Fig. 2. The mean number of total remnant particles (cellulose, plastics, aluminum) for each FFP sample. Bold lines show the medians, box borders indicate the 25th and 75th percentiles as determined by R software. Different letters indicate a statistically significant difference ($p < 0.05$) among the FFP groups.

samples showed a low level of contamination (Fig. 2).

This distribution could be due to the inherent variability of the cleaning and packaging remnant removal procedures adopted in each plant. Another source of variability is the initial food material used since single-serving packages have increased in recent years. There are various reasons for the popularity of smaller products, such as an overall increase in small family units, the prevalence of snacking, and concern over portion control. Smaller portions require smaller packages which ultimately can increase the amount of packaging and wasted space on a pallet when shipping the product. This means that the balance between single-serve and bulk packaging can vary according to the type of original material used in the former food plant. Multipacks can also be categorized as single-serve packaging because they consist of multiple smaller packages within a larger container, such as a multipack of various bagged snacks [32]. All these aspects can affect the presence of packaging remnants in former foods since their removal is mechanical [5,6]. However, based on our results, it is difficult to speculate whether some FFP samples (group 10–14) were produced from single-serve or bulk packaging.

3.2. *Nature, classification and different size distribution of remnant particles*

The analysis of blanks by μFT-IR showed no external sample

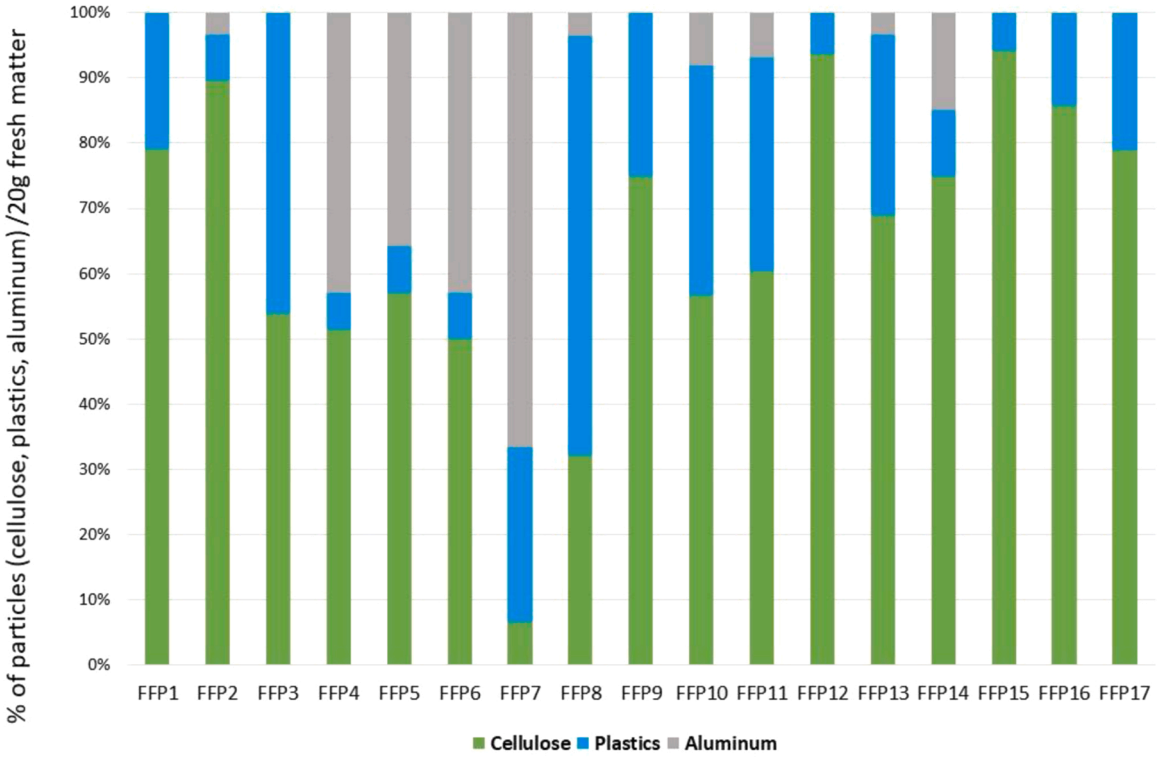

Fig. 3. The relative abundance (percentage of particles) of cellulose, plastics and aluminum remnants found in each FFP sample.

contamination during processing. Generally, the results showed a prevalence of cellulose, followed by plastic and aluminum particles (Fig. 3). This finding is in accordance with van Raamsdonk et al. [13] who found that the fibers from paper and paperboard were the most prevalent in the FFPs analyzed. Considerable variability between samples was observed: cellulose particles were present in all the FFPs with values ranging from 6% to 93% (of the total remnants) in FFP7 and FFP12, respectively. Plastic particles were observed in all the samples, with values ranging from 5% to 64% in FFP5 and FFP8, respectively, and aluminum particles were revealed with a percentage up to 66% in FFP7, but were absent in seven of the samples analyzed (FFP1, FFP3, FFP9, FFP12, FFP15, FFP16, FFP17).

In terms of the abundance of packaging remnants in the FFPs analyzed, many aspects need to be taken into account. First, the types of ingredients used in the different products by the different FFP producers can vary and may have different kinds of packaging materials. FFP samples were not collected in the same period of the year, and the seasonality of the ingredients used for the FFP production could also affect the packaging materials found in the final products. Chocolate products for instance, which are usually packed in aluminum, may be more abundant in specific countries and/or in specific periods of the year. Other FFP processors may also prefer unpacked ingredients to packed ones.

Different countries may also have different food leftovers, when considering the confectionary industry or bakery products, with different types of packaging. For example, in FFPs from confectionary industries, the packaging material may be aluminum for chocolate bars, plastic for candies and sweet snacks. On the other hand, in FFPs derived from bakery industry ingredients, paperboard may be used for packaging pasta, although most biscuit products and bread (except for processed bread), are processed unpacked [13].

Other aspects to consider are the way these products are ground, the technological process they undergo (e.g. pelleting, extrusion), the mechanical technologies used to unpack them, as well as the efficiency of the equipment used for unpackaging and removing the packaging remnants. All these factors could explain the great variability in terms of presumed packaging material among the FFP samples analyzed. However, the companies did not disclose to us the ingredient list of the FFPs, thus limiting the interpretation of our results.

The final particle number of the remnants also greatly depends on the particle size limit. As reported by van Raamsdonk et al. [29], macroscopic detection of remnants larger than approximately 1 mm permits the physical extraction from the sample and counting of the particles. On the other hand, microscopic detection at a magnification of between 100x and 400x only determines the presence of the potential packaging remnants. In the present study, the overall mean remnant particle size was 2.74 ± 0.56 mm, while the particle sizes of the three FFP categories (cellulose, plastics, aluminum) are reported in Fig. 4, which shows the lack of significant differences in the three different materials. The mean size of the cellulose particles was 2.57 ± 0.37 mm, 3.79 ± 1.85 mm for plastics and 1.46 ± 0.25 mm for aluminum, respectively.

This variability in size distribution, although not significant, may depend on the former food pre-treatment methods adopted during the production, such as in the packaging remnant removal [33] and grinding process [29]. As reported by Luciano et al. [33], the processes used in packaging removal of bakery products are: air, drying, sieving and the use of blown air to remove the remaining packaging materials such as plastic and paper, magnets to remove ferrous metals, and eddy current separation (ECS) to remove nonferrous metals. Depending on the different processing plants, FFPs are then ground, with extra-fine grinding to increase the homogeneity of the final product [29]. Grinding can result in a higher fragmentation of packaging remnants which influences both their size and number in the sample.

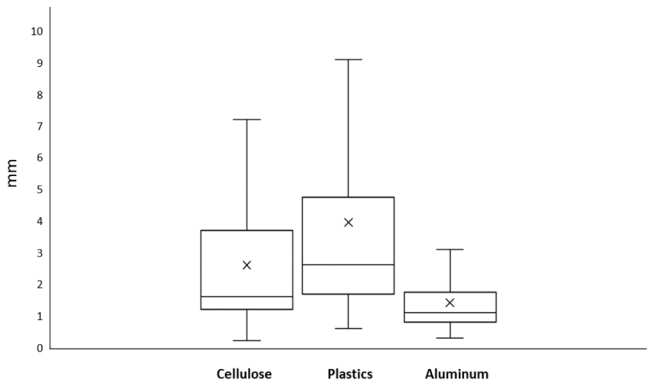

Fig. 4. The sizes (mm) of the three categories (cellulose, plastics, aluminium). Central lines show the medians, the X represents the mean, box limits indicate the 25th and 75th percentiles as determined by R software. There is no statistically significant difference ($p > 0.05$) among the FFP samples.

3.2.1. *Cellulose particles*

The pattern of cellulose particles resembled the total number of remnants. In fact, the same samples that were characterized by the highest number of impurities had approximately the highest number of cellulose particles, namely FFPs 1–2 and FFPs 10–14. Some differences were observed for samples FFP2 and FFP15 which showed a high number of cellulose particles, even though, as shown in Fig. 2, they were allocated to the middle level group. This indicates that cellulose was the main contributor to the sample contamination.

The results showed that sample FFP12 had the highest number of cellulose particles, which were significantly more abundant than sample FFP7 (Fig. 5).

The cellulose category includes paper, paperboard and natural fibers such as vegetable (e.g. cotton [34]). Paper was the second-most commonly used packaging material together with pressed paperboard, considering that about 37% of all food packaging materials are made from paper [35], followed by regenerated cellulose (not identifies by the IR as being different from cellulose [3,5,6,13]).

Paper, paperboard and carton as food packaging are principally used to protect milk and milk-based products, dry powders, confectionary and bakery products and some kinds of beverages [36]. Since these materials are in direct contact with both dry and fatty or wet foodstuffs, they need to have good barrier properties, high heat salability and strength [36]. For this reason, paper, paperboard and carton are treated with specific additives, such as sizing and retention agents, biocides, surface refining and coating agents or are reinforced with aluminum or plastic layers [13,36]. As paper and paperboard are made from natural fibers, the only issue when livestock ingest these materials are these additives that are intended to remain in the paper. Although these substances become more available after the degradation of paper in the gastrointestinal tract, no health concerns have been identified in view of the small amount of paper found in animal feed [13].

3.2.2. *Plastic particles*

After a general analysis of remnant particles and their classification into the three main groups (cellulose, plastics and aluminum), the mean number of plastics in each type of FFP sample was calculated. The results showed no significant differences between the FFP samples regarding the amount of plastic particles (Fig. 6). Numerically, the first group showed the lowest number of plastic particles (i.e., FFP1, FFP2, FFP3, FFP4, FFP5, FFP6, FFP7, FFP9, FFP12, FFP14, FFP15, FFP16, FFP17), indicating that more than 70% of the FFP samples had a low level of plastic contamination. The second group (i.e., FFP8, FFP10, FFP11, FFP13) with the highest number of plastic particles resembled the total number of remnants. In fact, the same samples that were characterized by the highest number of remnants were approximately the same as

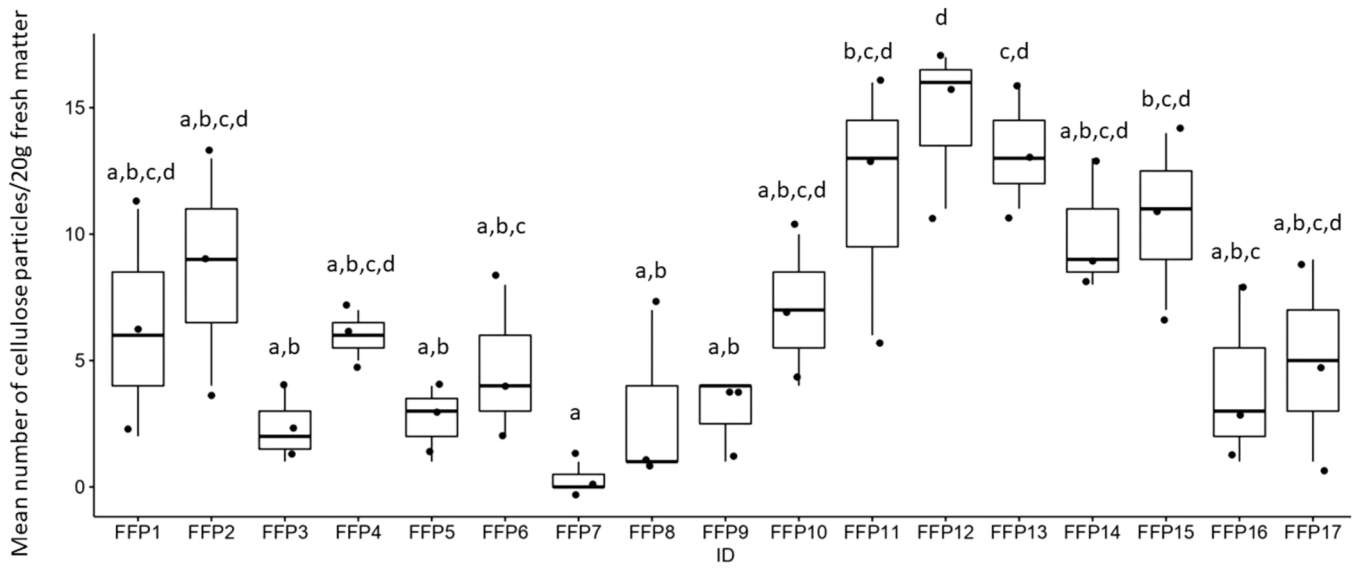

Fig. 5. The mean number of cellulose particles for each FFP sample. Bold lines show the medians, box limits indicate the 25th and 75th percentiles as determined by R software. Different letters indicate a statistically significant difference ($p < 0.05$) among the FFP groups.

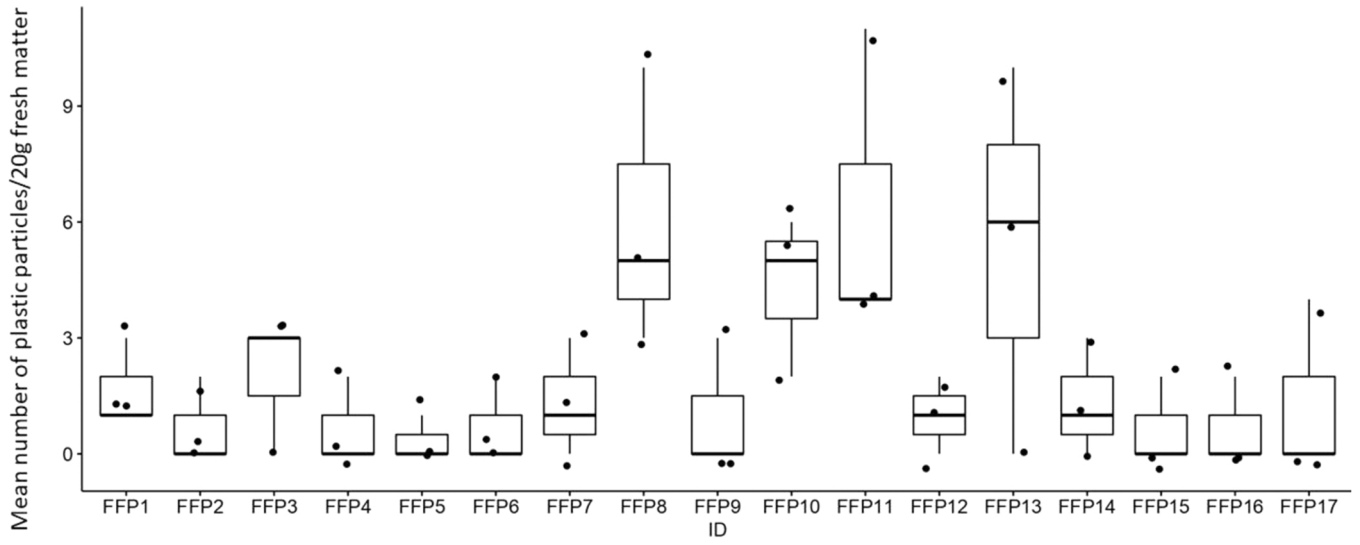

Fig. 6. The mean number of plastic particles for each FFP sample. Bold lines show the medians, box limits indicate the 25th and 75th percentiles as determined by R software. There is no statistically significant difference ($p > 0.05$) among the FFP samples.

those with the highest number of plastics, except for FFP8.

Of all food-packaging materials, plastic was the most frequently used (in Europe 38% of plastics use is for packaging; [37]). Durability, flexibility, lightness and low cost are some of the features that make plastic suitable for protecting food and contributing to food quality [38]. Since FFPs are intended for livestock including both ruminants and monogastrics, all the potential effects on the gastrointestinal tract and on the animal physiology related to the ingestion of plastic packaging need to evaluated.

Much is known about the effects of plastics on marine organisms and also on plastics entering the food chain in aquatic environments, while few studies regarding terrestrial animals refer to the transfer of plastics in the food chain [39]. With regard to monogastrics, remnant particles, such as fragments of polystyrene packaging, can cause gastric lesions in newborn birds kept in the poultry house litter. There is also evidence of inappetence, lethargy and ulcerative lesions with the formation of granulomas in broiler chickens [40].

Basini et al. [41] exposed swine granulosa cells to different concentrations of polystyrene nanoplastics. The results showed that the highest concentration tested stimulated cell proliferation and induced the disruption of the cell redox status, confirming the potential of plastic to promote an oxidative stress status [42]. Studies on bovines, sheep and goats have reported potential effects that the ingestion of macroplastics could have on the rumen [39,43]. After the systematic ingestion of plastics, inappetence and suspended rumination, indigestion, inflammation, ruminal bloat, rumen microflora dysbiosis, and poor production have been reported [44]. Plastic is not digested in the rumen and it tends to accumulate causing atrophy of ruminal papillae and abnormalities during the fermentation process [44]. Moreover, Mekuanint et al. [43] showed how large amounts of plastic residues in the rumen and reticulum prevent the normal absorption of volatile fatty acids (VFA) leading to a reduction in milk yield and in the rate of animal fattening [43,44].

Many studies have assessed the potential consequences of plastic ingestion and exposure on animal health [39,42,45-47], however further research is needed to explain the mechanisms behind their effects.

Our use of μFT-IR led not only to the characterization of cellulose and plastic particles, but also differentiated between different types of polymers. The most abundant polymers in this study were polypropylene (PP) and polyethylene (PE). Regarding PP the maximum percentage value (100%) was found in FFP1, FFP5, FFP9, FFP14, FFP17, while the minimum percentage content (16%) was found in FFP1 and was absent in FFP2, FFP4, FFP6, FFP8, FFP10 and FFP15. Regarding PE, the maximum percentage (100%) was found in FFP4 and FFP6, the minimum percentage content (15%) was found in FFP10, while it was absent in FFP1, FFP5, FFP9, FFP14 and FFP17. Although their content in the FFP samples was very low, also polyesters (PEST), such as the particular class of polyethylene terephthalate (PET), polyurethane (PU), polylactic acid (PLA), polyacrylic rubber (ACM), ethylene propylene rubber (EPR), polyammide (PA) and ethylene vinyl acetate (EVA) were found in the different samples. This thus indicates a large amount of different packaging materials intended for different types of recovered products (Fig. 7).

These polymers cover 84% of plastic production and are mainly thermoplastics, which offer great mechanical performance together with valuable barrier properties [48]. Polymers include PET, which has different colors, and is temperature resistant up to 140 °C. PP is usually made from transparent rigid material and is resistant to temperatures between 220 and 240 °C [5,6]. In all the samples only irregular shaped fragments were detected, except for FFP13 in which one fiber of PEST was found in the second replicate, and transparent, white, grey and blue were the main colors of the detected fragments in the majority of FFPs analyzed.

Polymers are used in many plastic containers. PET, for example, is found in food jars, soft drink bottles and plastic films, while PP is used to produce bottles for milk and food containers [48]. These polymers, which are known to have a carbon-carbon backbone, a high molecular weight and few functional groups, are very resistant to degradation [49]. Wu et al. [47] showed how PP and PE are the most frequently used polymers in plastic products. Depending on the type of polymer produced, plain plastic also requires treatment with specific additives to obtain a physical or chemical effect during the polymerization of monomers [13] and to improve the polymer performance [37]. The different types of additives include antioxidants, fillers, polymeric additives, stabilizers, optical brighteners and antistatics [13]. These additives can migrate from plastics to the external environment, such as plasticizers, which are not stable additives because they are not bound to the polymer matrix [42].

These particles can also fragment into smaller debris, both in the environment and in the feed matrix. This fragmentation can be caused by exposure to light (photo-degradation), oxidation, hydrolysis, temperature changes or mechanical abrasion [39,46,49]. Since the plastic remnants in the FFP samples undergo grinding together with physical and microbial processes in the stomach and gastrointestinal tract, the residual plastic chemicals migrating from the plastic remnants may be more available when the animal ingests the plastic packaging.

Plastic remnants have several toxic effects on animal health [39,42,44]. For example, when the chemicals migrate from the packaging to the rumen fluid, they can reach the circulation causing immunosuppression [44]. One possible toxicological effect is also due to the additives in the plastics. For instance, di (2-ethylhexyl) phthalate (DEHP) has a strong impact on the reproductive organs, heart, liver, kidney and lungs, while bisphenol A, which is often used in beverage and food packaging, is an endocrine disrupter [42].

During the manufacturing of plastic, heavy metals such as cobalt, lead, mercury, chromium and theirs salts, can also be incorporated. These heavy metals can accumulate in the blood, liver, kidney and in the rumen fluid when the animal ingests plastic [39]. Heavy metals, as well as chemicals such as polychlorinated biphenyl, can enter the blood circulation, also reaching the food chain.

The plethora of effects that plastics may have on the quality of the animal product therefore needs to be considered, since there are safety issues related to the release of these materials when consumers eat meat and milk products [44].

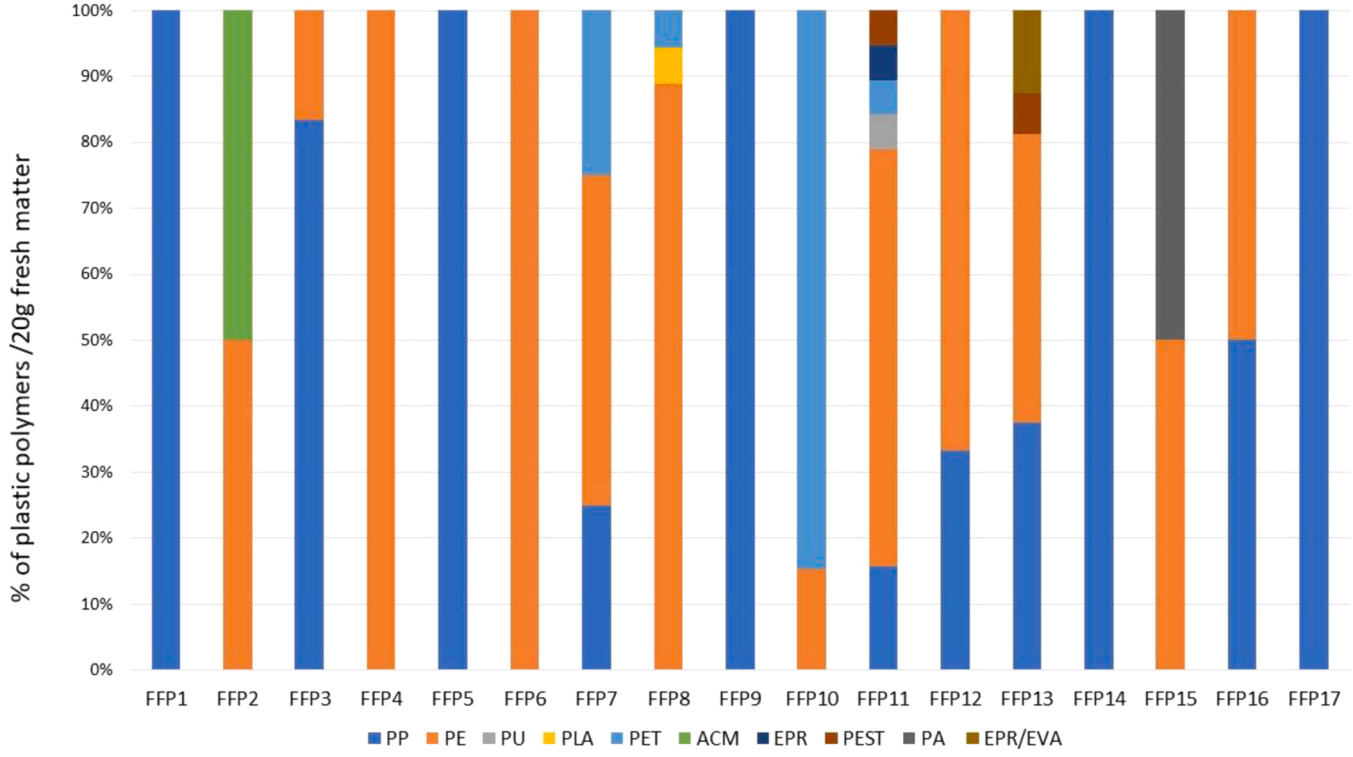

Fig. 7. The relative abundance (in percentages) of plastic particles classified by the different types of polymers; polypropylene (PP), polyethylene (PE), polyurethane (PU), polylactic acid (PLA), polyethylene terephthalate (PET), polyacrylic rubber (ACM), ethylene propylene rubber (EPR), polyester (PEST), polyammide (PA), ethylene vinyl acetate (EVA).

One of the main concerns about plastic is the size. In the present study, the plastic particle size was found to have an overall mean of 3.8 mm (Fig. 4). The size measurements showed that mesoplastics were the main category found in the FFP samples (Fig. 8). Only 23% of the samples were slightly contaminated (4–6 particles/20 g of fresh matter) which is reassuring. In addition, more that 75% of samples were contaminated by one particle.

In addition to the risk derived from the migration of plastic additives, the size of the polymer particles also needs to be considered. Depending on the size of the plastic particles, the potential hazard on organisms and ecosystem can increase. The first definition of microplastics was given by Thompson et al. [50] who defined them as all plastic particles or debris smaller than 5 mm in diameter including nanoplastics (nano--scale particles) according to the criteria of the US National Oceanic and Atmospheric Administration (NOAA) [51-53]. Currently, however, the classification of plastics according to their size, is more complex. In fact, the smaller debris from the fragmentation of larger plastic pieces can be classified in four main groups: macro-plastics (≥1 cm), meso-plastics (from 1 mm to < 10 mm), microplastics (from 1 μm to < 1 mm) and nano-plastics (from 1 nm to < 1 μm; [31]). The main focus is currently on micro-plastics, defined as emerging and persistent pollutants, since their presence is ubiquitous in many environmental systems, thus representing a health concern for animals, humans and environment [46].

The main consequences linked to the ingestion of plastic particles tend to be alterations in feeding activity, and decrease in food assimilation efficiency with a consequent reduced body weight and slower growth [42]. Depending on their size, microplastics can cross the intestinal barrier of mammals, enter in the systemic circulation and reach the tissues, establishing a general status of inflammation and necrosis in the worst case [54]. The consequences for the digestive tract following the ingestion of microplastics concerns both mechanical damage to the intestinal mucosa and alteration in the gut microbiota that activate a mucosal immune response. The inflammatory process leads to several changes in the bacterial gut community, promoting the growing of actinobacteria, proteobacteria, and enterobacteriaceae (which are known to increase during inflammation) and a condition of dysbiosis which exposes the animal to a weak immune system and diseases [42, 45]. However, the risk assessment conducted by European Food Safety Authority (EFSA) showed that the intestinal absorption of microplastics is very low (up to 0.3% with particles of 2–3 μm on human and rodent models [54]. In contrast, nanoplastics (NPs) are more easily absorbed by the gut mucosa than microplastics [41].

Lastly, microplastics can accumulate in the food chain [49,55] although there is a lack of information on the dietary exposure, kinetics and biodistribution in food [54]. Microplastics have been found principally in the food from marine environments, however no data are available on microplastics residues in meat products. Since the absorption of microplastics in the gut is very low, there should be no high exposure via food for humans, as products from livestock are consumed after the removal of the intestinal packet. Regarding the terrestrial environment, studies have investigated the presence of microplastics in soil, soil earthworms, fruit and vegetables [56-59].

3.2.3. *Aluminum particles*

The pattern of the aluminum particles presented three distinct groups. Sample FFP4 showed the highest mean number of aluminum particles, significantly more abundant compared with FFP1, FFP2, FFP3, FFP8, FFP9, FFP12, FFP15, FFP16 and FFP17. An intermediate group was represented by FFP5, FFP6, FFP7, FFP10, FFP11, FFP13, FFP14 (Fig. 9). These results thus indicate, that more than 50% of the FFP samples were very slightly contaminated, while the rest was moderately contaminated.

Aluminum foil and aluminum-coated paper have a wide range of applications in many products, such as chocolate, candy bars and many kinds of sweets. It can also be combined with other materials such as plastic especially when the food is acid or very salty. All these features make aluminum suitable for oxygen, light, water vapor-sensitive food [60]. Most foods have a pH of between 4 and 7, and thus aluminum packaging is not affected by corrosion and consequently the migration of aluminum in food is not a concern, as reported by Lamberti et al. [60]. As with paper and plastics, aluminum fragments also break down into smaller particles and some components can be absorbed by the gut [13, 61]. Although the absorption is very low, there is some evidence of aluminum bioaccumulation in the spleen, liver, kidneys, heart, lungs and in particular in bones in experimental animals [62,63]. Esquerre et al. [64] showed how an oral ingestion of a low dose aluminum can cause bowel irritation, visceral pain and hypersensitivity in experimental rodents. Little was found regarding the effect of aluminum ingestion by livestock, however it is assumed that the impact of this material could be relatively high because of its long half-life in tissues

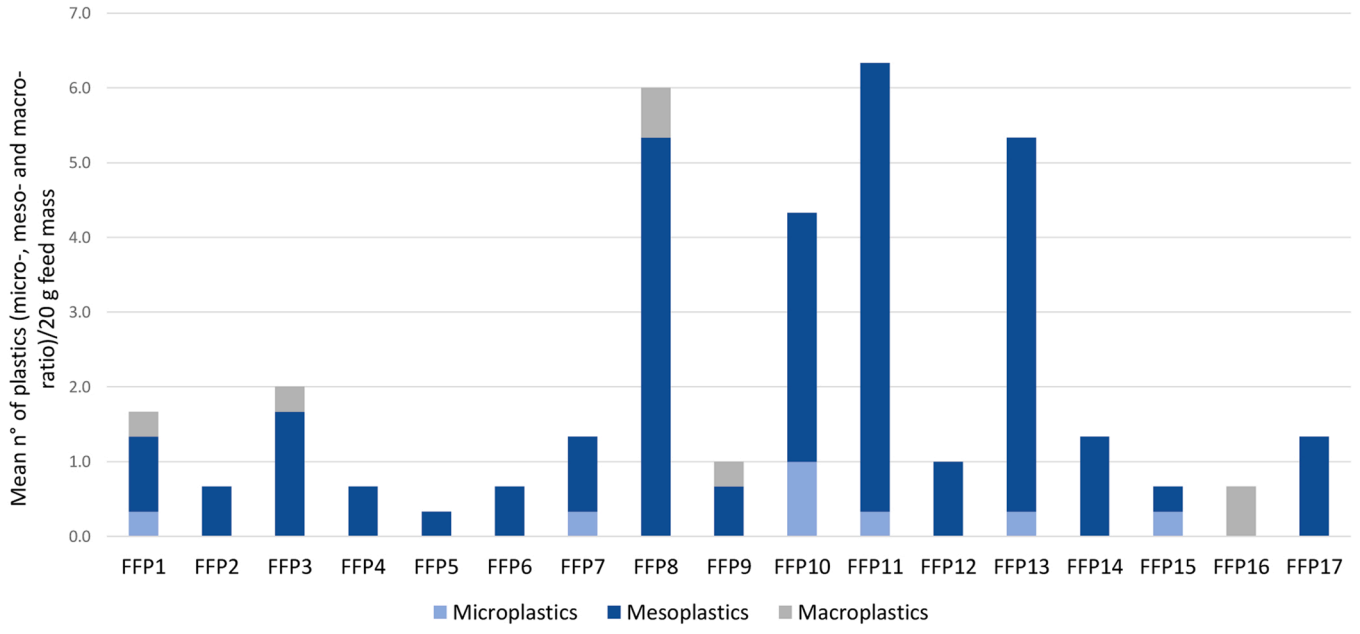

Fig. 8. The mean number of microplastics, mesoplastics and macroplastics for each type of FFP sample. Mesoplastics were the main particle size found.

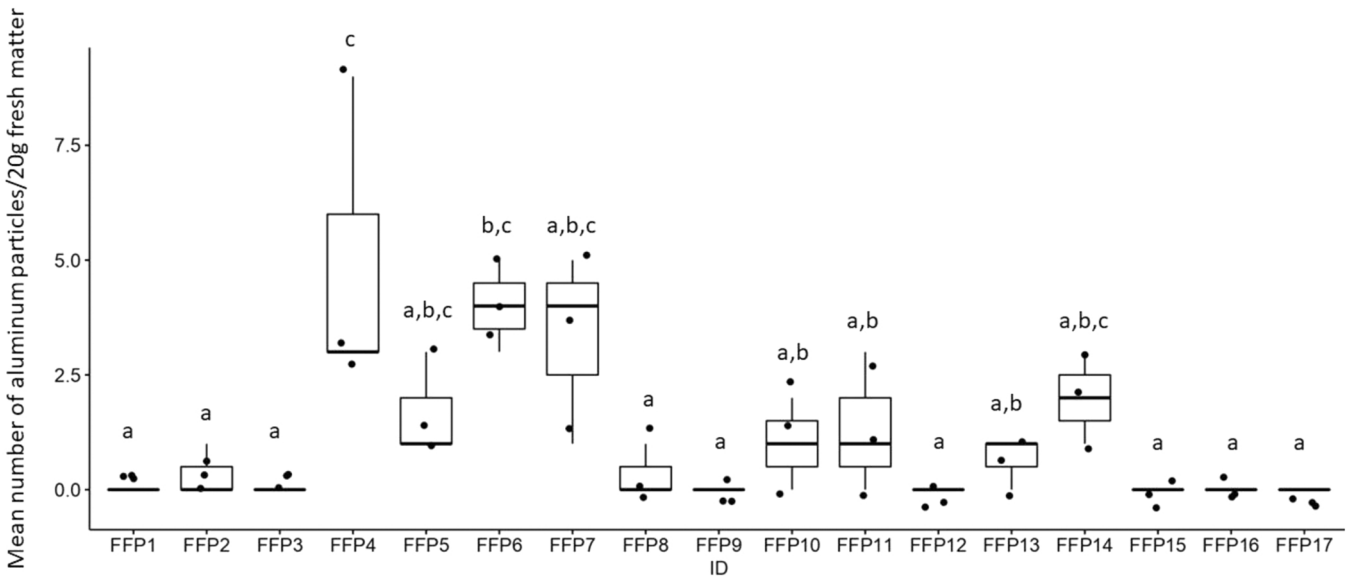

Fig. 9. The mean number of aluminum particles for each FFP sample. Bold lines show the medians, box limits indicate the 25th and 75th percentiles as determined by R software. Different letters indicate a statistically significant difference ($p < 0.05$) among the FFP groups.

[13].

According to the European regulation (EC) 1935/2004 [18], packaging materials must not release additives substances in a way they could damage human health. For this reason, EFSA established some Specific Migration Limits. In addition, FFPs comply with the accepted tolerable level of packaging remnants in feed, according to the German Federal Ministry of food, agriculture and consumer protection [11]. The inclusion of 30% of FFPs in animal feed (level of inclusion suggested by [5,6]) would result in a level of 0.005% (wet weight; *w/w*) of packaging material in the final feed. In the current study, the residual amount of paper, plastic and aluminum particles in the analyzed FFPs samples was negligible, as also reported in other studies [3,11–14] and thus can be considered safe for animal feeding.

4. Conclusions

The development of validated monitoring methods that control hazardous contaminants in feed and food has been strongly recommended [54], especially in a circular economy where reducing food losses must be combined with minimizing potential risks and ensuring food and feed safety [16]. However, little is known about using a μFT-IR for the analysis of feed contaminants. Several methods have been used to detect presumed packaging remnants in FFPs, but without characterizing the real chemical nature of these materials.

We demonstrated that a μFT-IR enables a semi-quantitative and qualitative evaluation of packaging remnants and prevents the overestimation of undesirable contaminants. It can thus be considered as an innovative method to better investigate presumed packaging remnants in FFPs.

Our findings showed significant differences in the number of particles found in the different samples, irrespectively of their chemical nature. However, cellulose particles were the most abundant compared to plastic and aluminum remnants. Moreover, there was no significant difference in the abundance of plastic particles between the different FFP samples.

Despite all the dangers linked to plastic ingestion, the risk for animal health is sufficiently low considering the low quantity of remnants found and considering the low percentage of FFPs (5–30%) usually included in the animal feed. From these results, we can conclude that the amount of packaging remnants found in the analyzed FFPs is low. The quantification of the packaging remnants in FFPs using a μFT-IR was reliable and confirmed that the issue of packaging remnants is limited. A μFT-IR could thus be considered for use in the routine monitoring of presumed packaging remnants in FFPs.

CRediT authorship contribution statement

Sharon Mazzoleni: Methodology, Investigation, Formal analysis, Validation, Data curation, Writing – original draft**. Stefano Magni:** Conceptualization, Methodology, Investigation, Data curation, Writing – review & editing, Supervision. **Marco Tretola:** Formal analysis, Data curation, Writing – review & editing. **Alice Luciano:** Investigation, Writing – review & editing**. Luca Ferrari:** Writing – review & editing**. Cristian Edoardo Maria Bernardi:** Writing – review & editing**. Peng Lin:** Software**. Matteo Ottoboni:** Software**. Andrea Binelli:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition. **Luciano Pinotti:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Statemnts of environmental implications

Plastic characterization techniques have been applied in various environmental research fields, as plastic has a significant ability to enter in the trophic chain. However very little information on the presence of plastic in feed and food is available. It is therefore essential to investigate the fate of plastics in the environment starting with their presence in former food products (FFPs, i.e. food leftovers originally intended for human consumption) used in livestock diets. FFPs are unpacked mechanically and then ground down, however particles from packaging may potentially remain in the feed, and thus enter the food chain.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2023.130888.

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