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Field investigation to determine the environmental source of PCBs in a pig farm

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ABSTRACT

During a national monitoring plan, a pork fat sample was declared non-compliant for the sum of dioxins and PCB-DL (EU regulation). The National Reference Laboratory together with competent authorities conducted extended investigations to determine rapidly the contamination source at the farm level. A range of samples (n = 129), representative of potential contamination sources, was selected for further characterization (fat, feed, materials, dust, soil) and was analyzed for PCDD/Fs and DL-PCBs by GC-HRMS. A hot spot was localized in the farm, which corresponded to a pre-feed storage tank, the paints of which presented huge DL-PCB concentrations ($> 1 \times 10^6$ pg g^{-1}), responsible for the contamination. The present case report describes a new source of PCB contamination, previously undescribed.

1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDD), polychlorinated dibenzofurans (PCDF) and polychlorinated biphenyls (PCBs) have been listed in the Stockholm convention for both their persistence and toxicity (Amiard, Meunier, & Babut, 2016; UNEP – United Nations Environment Programme, 2001). PCDD/Fs appear during thermal and chemical processes, whereas PCBs are industrial mixtures produced and used in agriculture and industry from the 1930s for their insulating properties in electrical transformers (Breivik, Sweetman, Pacyna, & Jones, 2002) and their chemical and physical stability (cutting oils, inks, paint) (Anezaki & Nakano, 2014; Ling, Han, & Xu, 2008). Their presence in the environment led to their restriction of use, firstly in 1970 in closed systems (transformers, capacitors) (Breivik et al., 2002). The production and use of PCBs were banned in many European countries at the end of 1980's. Despite such prohibition, PCDD/Fs and PCBs are still present in the food and feed chain (Traoré, Echaux, Sirot, & Crépet, 2016; Zhang et al., 2013) and food from animal origin, such as meat, eggs and dairy products are considered most important contributors to human exposure (Arnich et al., 2009; EFSA, 2014). Therefore, in a public health perspective, regulatory limits have been defined in foodstuffs (EC, 2011) and feed (EC, 2012). Product conformity (from the domestic or imported markets) is checked in the EU through the implementation of official PCDD/Fs and PCBs controls at an appropriate frequency, based on risk (EC, 2004).

Several food contamination episodes and crises have occurred worldwide during the last decades (Hoogenboom, Traag, Fernandes, & Rose, 2015; Malisch & Kotz, 2014), e.g., the Yusho and Yucheng rice oil poisonings in Taiwan in 1979 (Soong & Ling, 1997), the Belgian dioxins/PCBs poultry crisis (Bernard et al., 1999; Covaci et al., 2008; Hens, Elskens, De Bont, Baeyens, & van Larebeke, 2004; Van Larebeke et al., 2001), and more recently the Irish dioxin incident in pork and pork products (Casey, Lawless, & Wall, 2010; Daff, 2008). The detection of the original source of a contamination is most of the time a challenging task, which requires specific knowledge on production processes and changes of patterns during bioaccumulation (Malisch & Kotz, 2014). Consequently, thorough investigations are conducted on the polluted area to determine the source and implement mitigation solutions. Reporting such crises or case reports strongly contributes to enhanced risk management by competent authorities for appropriate management options to be decided and applied.

In 2016 in France a monitoring plan, commissioned by the French ministry of agriculture, highlighted PCB contamination in a subcutaneous pork fat sample. The concentration of 3.0 pg g^{-1} lw WHO-TEQ 2005, exceeded the maximum level fixed at 1.25 pg g^{-1} lw for the considered animal species (EC, 2011). The sample was declared non-compliant and it was then possible to identify the farm where the pig was raised. In order to explain this contamination, disclose its source and ensure that livestock were no longer exposed, further investigations were led by the relevant French National Reference Laboratory

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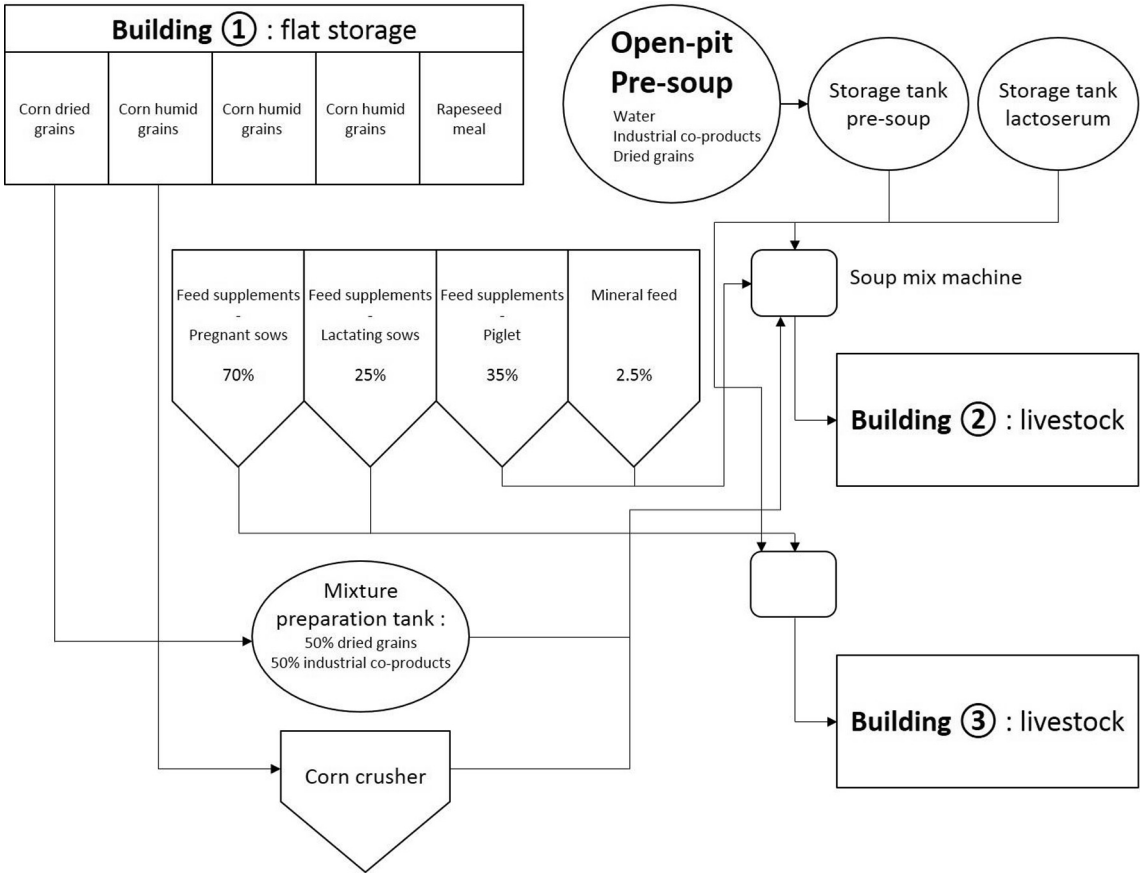

Fig. 1. Schematic representation of the pig farm and the feeding process.

(Laboratoire d'Étude des Résidus et Contaminants dans les Aliments (LABERCA)), in collaboration with other national veterinary services.

The aim of our study was to develop a thorough and efficient methodology to determine the origin of the PCBs contamination. This article presents the sample collection methodology, the analytical strategy and associated results, to explain and prevent further PCB contamination in this particular farm.

2. Materials and methods

2.1. Site presentation

A schematic drawing of the farm and the feeding process is presented in Fig. 1. The pig farm of interest was composed of three main buildings, one for feed storage (Building 1) and two for livestock (Buildings 2 and 3). Two main steps composed the established feed process. The first one consisted of the "pre-soup" preparation, which was a mix of water, industrial co-products and dried grains, in a 250-m^3 cement open pit. Part of the prepared pre-soup was stored in a 50-m^3 dedicated tank. The second one involved the "soup" elaboration in a mix machine as follows: lactoserum, feed supplements, corn, minerals and water were added to the pre-soup. The resulting soup was then used to feed the pigs. Building 1 was built in 2012, Building 2 in the 1980s and Building 3 in the 1990s. The roofs were made of asbestos-cement fiber plates for the 1980s and 1990s buildings and fiber cement for the 2012 building.

2.2. Collected samples

More than 120 samples were collected at different locations in the pig farm throughout the complete investigation period. The sampling scheme included pork samples (peritoneal fat, colostrum, milk, $n = 16$), feed (n = 32), materials in contact with feed like silos and open-pit cement ($n = 17$), building materials and dust roofs ($n = 50$), soils ($n = 10$) and paint samples ($n = 4$). Milk and colostrum were collected from three different lactating sows and pooled. Peritoneal fat samples were collected at the slaughterhouse from pork samples from the five different weight groups (5 kg, 8 kg, 20 kg, 45 kg and 60 kg) representative of the farming production. Building materials, dusts, soils and paint samples were collected in 50 and 250-mL polypropylene flasks by using either a small shovel, a cement grinder or wipes wetted with ethanol (32 cm × 17 cm). Whatever the collection mode, all samples underwent subsequently the same extraction/purification protocol complying quantification requirements (see Sections 2.3 and 2.4).

2.3. Chemicals and standards

The chemicals used were of high quality grade for trace analysis; acetone, *n*-hexane, and toluene were purchased from Promochem (Molsheim, France), and anhydrous sodium sulfate from Merck (Darmstadt, Germany). Ultrapure water was obtained using a Nanopure system from Barnstead (Waltham, MA). The four GO-HT columns used to purify samples were acquired from the Miura Institute of Environmental Science (Miura Co., Ltd., Matsuyama, Japan) and kept at room temperature. These columns can be used up to one year after production. Wet wipes were purchased from Sodibox.

Standard solutions of native PCDD/Fs, DLPCBs and NDL-PCBs, together with their ^{13}C-labeled internal and ^{13}C-labeled external standards, were provided by BCP Instruments (Lyon, France). External standard (^{13}C PCB111) was spiked into all samples after the purification step, to determine the recoveries. Purity of all standards was higher than 98%. Seven calibration standard solutions containing all the native PCDD/Fs and PCBs in different concentrations and ^{13}C-labeled