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Potential impact on food safety and food security from persistent organic pollutants in top soil improvers on Mediterranean pasture

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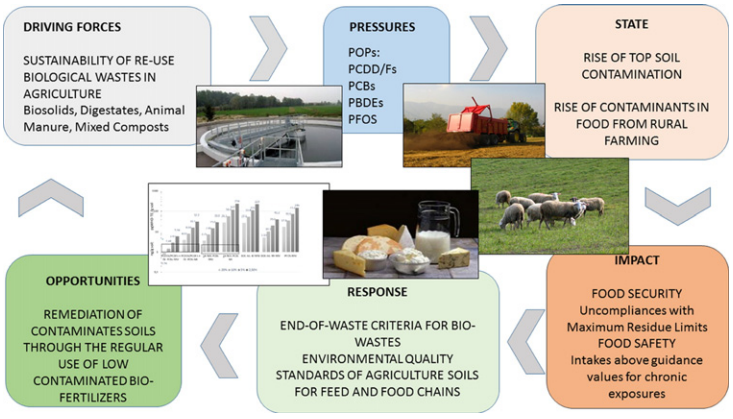
HIGHLIGHTS

- Top soil improvers were characterized for selected POPs content, in Italy.
- Grazing behaviour makes sheep sensitive to top soil contamination.
- Environmental quality standards for grazing areas were modelled
- The impact on Mediterranean sheep milk safety/security was evaluated.
- Low contaminated TSIs support safe intake and compliance of dairy products.

GRAPHICAL ABSTRACT



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ABSTRACT

The organic carbon of biosolids from civil wastewater treatment plants binds persistent organic pollutants (POPs), such as polychlorodibenzo -dioxins and -furans (PCDD/Fs), dioxin and non-dioxin -like polychlorobiphenyls (DL and NDL-PCBs), polybrominated diphenyl ethers (PBDEs), and perfluorooctane sulfonic acid (PFOS). The use of such biosolids, derived digestates and composts as top soil improvers (TSIs) may transfer POPs into the food chain. We evaluated the potential carry-over of main bioavailable congeners from amended soil-to-milk of extensive farmed sheep. Such estimates were compared with regulatory limits (food security) and human intakes (food safety). The prediction model was based on farming practices, flocks soil intake, POPs toxicokinetics, and dairy products intake in children, of the Mediterranean area. TSI contamination ranged between 0.20–113 ng WHO-TEQ/kg dry matter for PCDD/Fs and DL-PCBs (N = 56), 3.40–616 μg/kg for $\sum_6$ NDL-PCBs (N = 38), 0.06–17.2 and 0.12–22.3 μg/kg for BDE no. 47 and no. 99, 0.872–89.50 μg/kg for PFOS (N = 27). For a 360 g/head/day soil intake of a sheep with an average milk yield of 2.0 kg at 6.5% of fat percentage, estimated soil quality standards supporting milk safety and security were 0.75 and 4.0 ng WHO-TEQ/kg for PCDD/Fs and DL-PCBs, and 3.75 and 29.2 μg/kg for

Abbreviations: BMDL, Bench Mark Dose Level; COR, carry over rate; DL, dioxin-like; dm, dry matter; EEA, European Environmental Agency; EFSA, European Food Safety Authority; fw, fresh weight; lb, lipid base; ML, maximum Limit; NDL, non-dioxin like; OC, organic carbon; PBDEs, polybrominated diphenyl ethers; PCB, polychlorobyphenyls; PCDDs/PCDFs, polychlorodibenzo -dioxins and -furans; PFAAs, perfluoro alkyl acids; PFOS, perfluoroctane sulfonic acid; POPs, persistent organic pollutants; TDI, Tolerable Daily Intake; TEQ, toxic equivalency; TSI, top soil improvers; UNEP, United Nations Environmental Program; WHO, World Health Organization; WWTP, waste water treatment plant.

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Sheep
Dairy
Mediterranean

$\sum_6$ NDL-PCBs, respectively. The possibility to use low-contaminated TSIs to maximize agriculture benefits and if the case, to progressively mitigate highly contaminated soils is discussed.

1. Introduction

The contamination of bioaccumulative persistent organic pollutants (POPs) in food of animal origin represents a priority issue for the society both in terms of food security (the possibility to place on the market and to give access to sufficient and nutritious food), and of food safety (the possibility to prevent foodborne diseases in humans) (WHO, 2013). Polychlorodibenzo -dioxins and -furans (PCDD/Fs) polychlorobiphenyls with and without dioxin-like activity (DL and NDL-PCBs), polybrominated diphenyl ethers, (PBDEs) and perfluoro -octan sulfonic acid (PFOS) are framed among POPs in the Stockholm Convention (UNEP, 2013). These compounds are targeted towards controls and a progressive reduction of their environmental release. POPs intake through the consumption of food of animal origin is acknowledged as the main route of exposure in the general population (EFSA, 2010, 2011a, b, 2012a, 2012b). Within the European Union, a particular attention has been placed on the reduction of alimentary intakes of the aforesaid contaminants through the implementation of regulatory levels in food and feeds, as in the case of PCDD/Fs and PCBs (EU Commission Regulation, 2011; EU Recommendation 2014/663/EU). For PBDEs and PFOS, a specific recommendation to develop monitoring plans able to estimate their occurrence in foodstuffs on national basis has been issued (EU Commission Recommendation 2010/161/EU; 2014/118/EU). The final goal is to progressively reduce the human intake of such priority contaminants, thus bringing the exposure of most of the population below the pertinent guidance values for chronic alimentary toxicity, such as the Tolerable Daily Intake (TDI) and the Bench Mark Dose Level (BMDL). Recent papers are consistent to indicate the environmental quality of agricultural soil as a key factor to guarantee the safety of food from extensively farmed animals. A particular attention is paid to sheep that, for their grazing behaviour, are considered among the most sensitive animals (EFSA, 2011a; Lake et al., 2015, Perugini et al., 2012). Some countries have already proposed PCDD/Fs quality standards for agriculture soils for a healthy food accounting for human intakes: Canada indicated 4 ng WHO_{1998}-TEQ/kg dry matter (dm) (Canadian Council of Ministers of the Environment, 2007), The Netherlands 2 ng I-TEQ/kg dm (Health Council of The Netherlands, 1996), Switzerland 5 ng I-TEQ/kg dm, and Germany 5 ng WHO_{1998}-TEQ/kg dm (OSPAR, 2007), respectively. More recently, the Italian National Institute of Health suggested a 4 ng WHO_{1998}-TEQ/kg dm as environmental quality standard for soils devoted to dairy farming in the Campania Region (ISS, 2010). Nowadays, apart from regular emissions by thermic sources of industrial and civil plants, the use of top soil improvers (TSIs) originating from biosolids from the civil wastewater treatment plants (WWTPs) may be acknowledged as a potential emerging risk for the progressive POPs entry in the food webs. Biosolids-derived TSIs represent a cheap and sustainable source of organic carbon (OC) and a water-saving tool for agriculture soils (Saveyn and Eder, 2014). Top soils (30 cm) of Southern Italy possess favourable conditions for an effective use of BSOs directly on soil, or indirectly after their anaerobic digestion and/or their mix with household and green wastes, as digestates and mixed composts. In particular, they have a low organic carbon content (on average 50 t/ha) (ISPRA, 2013a) (Fig. 1), and a geographical vulnerability to extended summer drought periods, also as consequence of climate changes (EEA, 2015). Biosolids could be conveniently used in small family farms where sheep management is mainly based on the production of local forages (Molle et al., 2001).

However, there is the evidence that biosolids and derived TSIs can result 10–100 fold more contaminated than the classical animal manure with PCDD/Fs and DL-PCBs (5.6 vs 0.2 pg WHO-TE/g dry matter) (Elskens et al., 2013). The use of such TSIs (up to 5 ton/ha/year for stabilized biosolids BSOs, up to 35 ton/ha/year for derived digestates and mixed composts with an averaged 30% content of organic carbon, on dry matter basis, according to national provisions of law) brings to the amended top soil a contamination in the range of the hundreds of ng/g dry matter for NDL-PCBs and/or PFOS, and in the range of the thousands for PBDEs (Zenneg et al., 2013; Suominen et al., 2014). The TSIs use in Mediterranean rural areas, where there is a poor POPs bulk depositions from regular emissions (Brambilla et al., 2015a), may progressively increase POPs top soil contamination to levels able to compromise the food security and safety of dairy products (Dumortier et al., 2012).

The POPs contamination derived from soil presence in grass and derived forages may be affected by cut height (Pecoraro and Camera, 2008). During the late summer the probability of direct soil ingestion increases because sheep use to spend up to 8 h/day in the grazing activity due to the reduced grass availability (Molle et al., 2001). On the basis of the above considerations, this paper aims to investigate the potential impact of the proposed agriculture soil dressing with biosolids and derived digestates and mixed composts on sheep farming. This could allow identification of the end-of-waste criteria of TSI able to support both food safety and food security issues related to dairy productions in the Mediterranean area.

2. Materials and methods

2.1. Sampling design

As far as in some Countries biosolids from civil WWTPs and derived composts and digestates are not allowed for agriculture use due to the potential risk determined by biological, pharmacological and chemical hazards burden (Saveyn and Eder, 2014), the main focus was addressed to such category of wastes already checked for their regulatory compliance with heavy metals content by Regional Environmental Agencies in

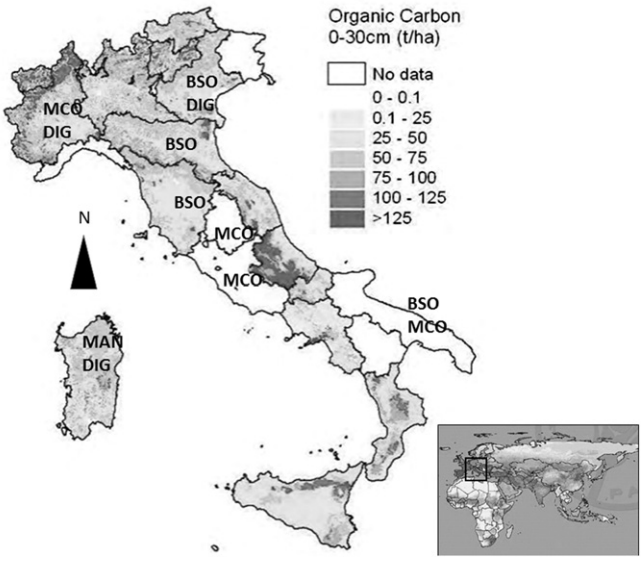

Fig. 1. The organic carbon content in Italian top soil along with the location of the sites of Biosolids (BSO), Digestates (DIG), Mixed Composts (MCO), and pig Manure (MAN) sampling. In the bottom right corner, the worldwide sheep flock density (darker areas) and boxed the Mediterranean region.