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# Potential impact of fertilization practices on human dietary intake of dioxins in Belgium

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#### ABSTRACT

Dioxins can enter the food chain at any stage, including crop fertilization. Therefore, we developed a simple method for estimating the introduction of dioxins in the food chain according to various fertilization practices. Using dioxin's contamination data taken from the literature, we estimated that fertilization accounts for approximately 20% of the dioxin inputs on agricultural soils at country scale. For the estimations at the field scale, 6 fertilization scenarios were considered: sludge, compost, digestate, manure, mineral fertilizers, and a common fertilization scenario that corresponds to an average situation in Belgium and combines mineral and organic fertilizers. According to our first estimations, mineral fertilizers, common fertilization practices or manure bring less than 1 ng TEQ/m<sup>2</sup> while atmospheric deposition or digestate bring between 1 and 3 ng TEQ/m<sup>2</sup> and sludge or compost bring more than 3 ng TEQ/m<sup>2</sup>. The use of solid fertilizers could potentially increase the dioxin levels in the 30 cm agricultural soil layer by 0 to  $\sim 1.5\%$  per year (up to  $\sim 9\%$  for the 5 cm thick surface layer). For animals, the increase in dioxin ingestion linked to the fertilization practices is lower than 1% for most scenarios with the exception of the compost scenario. Increases in human dietary intake of dioxin are estimated to be lower than 1% for conventional rearing methods (i.e. grazing animals are reared outdoor while pigs and poultry are reared indoor). Spraying liquid fertilizers on meadows and fodder crops, even if very limited in practice, deserves much more attention because this application method could theoretically lead to higher dioxin's intake by livestock (from 6 to ~300%). Considering an average half-life of dioxins in soils of 13 years, it appears that the risks of accumulation in soils and in the food chain are negligible for the various fertilization scenarios.

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#### 1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and dioxin-like polychlorinated biphenyls (DL-PCBs) belong to a class of organic compounds which are referred to as dioxins because they exhibit "dioxin-like toxicity", commonly expressed in toxicological equivalents (TEQ) (Van den Berg et al., 1998; 2006). Human exposure to dioxin is a matter of concern. These compounds cause impairment of the immune system, the nervous system, the endocrine system and the reproductive functions and are also suspected of causing cancer (European Commission, 2010). The World Health Organization recommends considering these compounds as priority contaminants for inclusion in monitoring programs (WHO, 2006).

Dioxins are classified as persistent organic pollutants (POPs) meaning, among others, that they bioaccumulate in the food chain (Stockholm Convention on Persistent Organic Pollutants, 2008). Nevertheless, limited attention is given to the possible incorporation of dioxins into the food chain through the use of agricultural fertilizers. At the same time, some organic fertilizers like sludge or compost are known to contain dioxins while the presence of dioxins in other fertilizers is little documented and cannot be excluded (Brändli et al., 2007; Kupper et al., 2007; Martinez et al., 2007; Van Gestel et al., 2007; Brändli et al., 2005; Zennegg et al., 2003; Groeneveld and Hébert, 2002; Stevens and Jones, 2002; Stevens et al., 2001; Eljarrat et al., 1999; Paulsrud et al., 1999; Rogowski et al., 1999).

In this work we use the term fertilizer to designate any kind of material that is added to the soil to increase the nutrient content or to improve the soil properties, including chemical fertilizers, sludge, compost and soil conditioners. Sludge is the biosolid produced during the purification process of urban waste water, industrial waste water or surface water. Although different in composition, these sludges will be grouped and simply referred to as sludge.

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Currently, no legal limit for dioxin concentration in fertilizers is set by the European legislation. National legislations are more restrictive, often imposing PCB limits and sometimes even PCDD/F limits for biowastes. In Belgium, organic fertilizers must contain less than 0.8 mg PCBs/kg dw for the sum of the seven marker PCBs in the Flemish region (OVAM, 2005) and less than 0.5 mg PCBs/kg dw for the sum of the seven marker PCBs in the Walloon region. No monitoring is done yet for the PCDD/Fs (FPS, 2010, unpublished document).

The present report has two objectives. Firstly, it proposes a simple method to assess the potential impact of fertilization practices on human intake of dioxins. Secondly, the developed method is applied to the Belgian situation as a proof of concept, using data gathered from the literature.

#### 2. Methods

#### 2.1. Concept

The impact of fertilization practices on the human dioxin intake has already been assessed for sludge (Wild et al., 1994; McLachlan et al., 1996; Rideout and Teschke, 2004) and for the combined use of biological wastes and chemical fertilizers (Fouchécourt and Beausoleil, 2001). Here, we describe a simple method which specifically aims at a comparison of fertilization practices at each step of the method.

In this study, two different scales are considered. At the territory scale, we assess the quantities of dioxin brought by fertilization practices throughout the territory in order to obtain information concerning the whole population. At field scale, we consider the impact of the worst case practices on the human dioxin intake. For each described fertilization practice, the effect on the soil dioxin levels is calculated. Further estimation of the impact on feed- and foodstuffs, as well as on the possible dioxin intake by humans, is carried out.

According to a literature review from the European Commission Joint Research Centre (Erhardt and Prüeß, 2001), four main pathways should be considered when assessing the transfer of POPs from sludge to humans. These are transfer (i) sludge-man by handling, (ii) soil-man (soil ingestion by human), (iii) soil (-plant)-animal and (iv) soil-water. However, here, we are only interested in transfer pathways that are relevant for the general population. Nonoccupational sludge or soil ingestion or inhalation by humans is very unlikely to occur at significant levels. Moreover, Wild et al. (1994) estimated that food accounts for more than 99% of the human dioxin intake, while the intakes from water and air are negligible. At the same time, if we exclude aquaculture, most of the fish eaten in Belgium is coming from the sea and not from rivers, lakes or ponds (Willemsen, 2003). The increase of the seawater dioxin level caused by fertilizers use, although uncalculated, can be reasonably considered as negligible. Indeed, dioxins are not mobile in soil (lipophilic) and direct water contamination by fertilizers is scarce (only through point contamination). Therefore, it is considered that merely the soil (-plant)-animal pathway is impacted by fertilization practices. The word "plant" is put in brackets because, most of the time, it only serves as a support for soil or fertilizers since dioxin uptake by plants is generally extremely low (Uegaki et al., 2006). In some cases (cucurbitaceae), soil-plant dioxin transfer is of importance but these species are exceptions and are of minor importance for human dioxin intake (Huelster et al., 1994).

Two kinds of fertilization practices are distinguished; solid fertilizers are believed to drop on soil after spreading, resulting in a soil contamination, while liquid fertilizers can be sprayed on plants, stick to leaves and lead to a plant contamination.

The developed method is valid for typical feeding conditions. Involuntary feed contamination by dioxins (e.g. by incorporation of contaminated fat into animal feeds) do not belong to the scope of this paper.

#### 2.2. Impact at territory scale

At territory scale, the total amount of dioxins brought to agricultural soils by fertilizing practices is assessed considering for each fertilizer the quantity applied to the entire territory and the estimated TEQ levels (Eq. (1)).

Amount of dioxins 
$$\left[\frac{\text{ng IEQ}}{\text{year}}\right]$$
  
= $\sum \left(\text{Fertilizer consumption}\left[\frac{\text{kg}}{\text{year}}\right] \times \text{Dioxins' concentaration}\left[\frac{\text{ng TEQ}}{\text{kg}}\right]\right)$ (1)

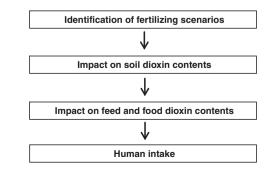
#### 2.3. Impact at field scale

The impact of fertilization practices at field scale is developed in 4 different steps that are described in this section and represented in Fig. 1.

#### 2.3.1. Identification of fertilizing scenarios

Six fertilizing scenarios are proposed for solid fertilizers use (manure, sludge, compost, digestate, NPK and common scenario) and four scenarios are proposed for liquid fertilizers sprayed on plants (manure, sludge, digestate and NPK). Each scenario tries to maximize the use of a specific fertilizer while the common scenario represents an average fertilization. The average fertilization is calculated by dividing, for each fertilizer, the known or estimated consumption throughout the territory by the agricultural surface. Liquid fertilizers are not considered in the common scenario because we have no data about the proportion sprayed on plants rather than on soils. By default, these fertilizers are considered to be spread on soils, like solid fertilizers, for the common scenario. Real practices are always combination between these scenarios and will always result in a dioxin transfer lower than the higher value for a specific fertilizer. Over fertilization is not considered in this report because the most common scenario does not imply over fertilization and because the proposed specific scenarios already represent worst case situations (organic fertilizers are scarcely applied at maximum rates in Belgium).

In Belgium, fertilization rates at field scale are limited by federal and regional regulatory constraints. For practical reasons, we regard Belgium as one single entity by considering for each fertilizer the maximum permitted application rate according to the most permissive legislation. The maximum fertilization rates are considered to be equal for solid and for liquid fertilizers. The maximum Nitrogen fertilization rate is 350 or 275 kg N/ha, on Belgian meadows or crops respectively with a maximum of 230 or 170 kg N/ha provided by organic fertilizers. For sludge, no more than 2 tons dw can be annually applied on meadows and 4 tons dw on crops. The chemical



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