A PROPOSAL FOR A DIET-BASED LOCAL PCDD/F DEPOSITION LIMIT

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SUMMARY: This study aims to propose a limit value for PCDD/F deposition to soil for an Alpine region located near a steel making plant. To derive the deposition limit value, some consolidated food chain models were applied, but they were run backwards, starting from the diet of people living in the region, from the guide value proposed by the World Health Organization for the Tolerable Daily Intake (1 pg WHO-TEQ kg^{-1} d^{-1}) and the consequent maximal concentration of PCDD/Fs in milk fat; running through the assimilation process of the cow, a deposition value on the ground that should not to be exceeded will be obtained. The study relates only to the intake of PCDD/Fs by the consumption of dairy products from cow's milk: indeed a comparison between the contribution of dairy products and vegetables to the daily PCDD/F intake will be carried out to demonstrate the most important role of the first ones. Afterwards, the obtained limit value for PCDD/F deposition will be compared with the average PCDD/F deposition measured in the same region during a monitoring campaign. The approach proposed in this paper allows assessing the overall deposition acceptable for an area.

1. INTRODUCTION

Exposure to Persistent Organic Pollutants (POPs) and their effects on human health have been object of important scientific and regulatory concern over the last years (European Commission, 1999). In particular, polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are considered the most toxic and diffuse family of POPs (Rada et al., 2011a). The primary health risk of PCDD/Fs is related to long-term exposure (Silbergeld and Gasiewicz, 1989; Sweeney and Mocarelli, 2000; Gascape, 2011). The toxicity of dioxin congeners is expressed in terms of a Toxic Equivalency Factor (TEF) in relation to the most toxic congener, the 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD). This methodology turns useful to study the environmental incidence of single industrial plants or the role of single industrial sectors (Rada et al., 2006; Rada et al., 2011b). The TEF concept was introduced in 1998 (Van den Berg et al., 1998) and in 2005 the Toxic Equivalency (TEQ) was adopted by the World Health Organization (WHO) for direct application to human risk assessment. Due to the lipophilic properties of POPs, bioaccumulation represents the most important way of contamination by PCDD/Fs, these compounds being capable to enter the food chain. Indeed, such compounds are accumulated in adipose tissues of animals and, consequently, of humans. The most important route for human exposure to PCDD/Fs is food consumption, primarily dairy products, followed by cereals and vegetables, meat and fish (Eduljee and Gair, 1996). This aspect was demonstrated in 1990 by a WHO survey which found out that more than 90% of the average daily intake (estimated in 2 pg I-TEQ kg^{-1} of body weight) derived from food consumption (Fürst and Wilmers, 1997). Contamination of food is primarily due to atmospheric deposition of semi-volatile compounds coming from various sectors (e.g. waste treatment, production of chemicals, metal industry, domestic heating) on farmland and the subsequent accumulation in the food chain (Liem et al., 2000; Fang et al., 2011). Grass is the primary food for the cattle (McLachlan, 1995) and, as a consequence, their main source of PCDD/Fs. On European scale, it was estimated that steel making plants would constitute the most important source of PCDD/Fs (Fang et al., 2011; Quaß et al., 2004). More in details the emission factors in this sector show a strong variability (European Commission, 2012), thus a generalization of their role in terms of local impact is not possible: specific analyses are needed case by case.

Since meat and dairy products have an important role in the diet of European and North American people, several studies have been conducted to reconstruct the accumulation process of PCDD/Fs in the grass-cattle-milk/beef food chain (McLachlan, 1995; Prinz et al., 1993; Slob and Van Jaarsveld, 1993; Slob et al., 1995; Van Lieshout et al., 2001) and to derive guide values for deposition on the ground and concentration in cow's milk, starting from the analysis of deposition samplers collected in specific regions (Van Lieshout et al., 2001; De Fré et al., 2000).

The aim of this study is to calculate a limit value for PCDD/F deposition, starting from the diet of people living in an Alpine region of Italy and from the WHO guide value for the Tolerable Daily Intake (TDI). The study relates only to the intake of PCDD/Fs by the consumption of dairy products from cow's milk: indeed a comparison between the contribution of dairy products and vegetables to the daily PCDD/F intake will verify the most important role of the first ones. The study will be carried out considering the diet entirely composed of local products. The food chain will be studied by applying some formulations presented in Slob and Van Jaarsveld (1993), Lorber et al. (2000) and McLachlan (1995), but the chain will be run backwards with respect to the classical approach, starting from the TDI and the consequent maximal concentration of PCDD/Fs in milk fat, running through the assimilation process of the cow and getting to a deposition value on the ground that should not to be exceeded. Afterwards, the obtained limit value will be compared with the average PCDD/F deposition measured in the same region during a monitoring campaign, in order to highlight possible exceedances. The main difference between this methodology and the work of Van Lieshout et al. (2001) consists in the purpose of calculating a deposition guide value directly on the basis of the diet and the TDI, instead of deriving the guide value iteratively starting from the deposition to get to a target TDI. The approach proposed in this paper allows assessing the overall deposition acceptable for an area. In order to complete the frame, the evaluation of the deposition related to a specific source can be based on a recent methodology (Rada et al., 2011b).

2. MATERIALS AND METHODS

In 1990 the WHO proposed an acceptable daily intake of 10 pg WHO-TEQ kg^{-1} d^{-1} for 2,3,7,8-TCDD based on information and studies available at that time (Kociba et al., 1978; Kimbrough et al., 1984). In 1998 the WHO revised this value and recommended a TDI of 1-4 pg WHO-TEQ kg^{-1} d^{-1} as maximal tolerable intake on a provisional basis but it stressed the need to reduce human intake to less than 1 pg WHO-TEQ kg^{-1} d^{-1} (Rada et al., 2011b). By applying the results of Buckley-Golder et al. (1999), who associated a one in a million cancer risk to an exposure of 0.006 pgTCDD kg^{-1} d^{-1}, the resulting cancer risk associated to the TDI of 1 pg WHO-TEQ kg^{-1} d^{-1} is $1.67 \cdot 10^{-4}$. Due to the absence of updated guide values for the TDI, this study focuses on the estimation of a limit value for PCDD/F deposition on the ground, starting from a maximal TDI of 1 pg WHO-TEQ kg^{-1} d^{-1}.

The region under investigation is an Alpine valley (E-W oriented) which has an important steel making plant located at the valley bottom. Near the plant, small villages, fields and some pastures are present. On annual average, the dominant wind direction follows the orientation of the valley, although no prevalent direction (E-W or W-E) is observed. PCDD/F deposition to soil has been kept under observation by means of bulk deposition samplers installed near sensitive receptors, such as kindergartens and primary schools. Each sample consists of a funnel coupled with a jar (both made of glass) collecting wet and dry deposition together, in accordance with the Bergerhoff method, which is widely used for PCDD/F samplings (Fang et al., 2011; Horstmann and McLachlan, 1997). The funnel/jar combination allows collecting settling particles and wet depositions, with contribution from dry gaseous depositions and impacting/diffusing particles (Horstmann and McLachlan, 1997). The here used deposition data refer to one year of measurements (September 2010 – September 2011) taken near a primary school, located eastward with respect to the plant, about 1.2 km far from its stacks. Since this study involves the transfer of PCDD/Fs through the food chain, the measured depositions are expressed in terms of WHO-TEQ (Table 1).

With the purpose of running the food chain backward, a characterization of the typical diet for seven age classes (0-11 months, 12-35 months, 36 months – 9 years, 10-17 years, 18-64 years, 65-74 years, over 75 years) was performed, based on the European Food Safety Authority data about food consumption in Italy (EFSA, 2012). Realistic body weight data for each age were assumed (Table 2), in order to calculate the daily amount of dairy products for every year of life. The average fat content of these products (expressed in g of fat per g of product), with particular regard to dairy products of the North of Italy, is 0.037 for milk, 0.039 for yoghurt, 0.305 for soft cheese, 0.290 for mature cheese, 0.285 for cream and 0.825 for butter (Gambelli et al., 1998; Mila, 2012; Trentingrana, 2012). The daily fat intake is obtained by multiplying each quantity of product ingested by its fat content. To indicate the TDI relating to each age, the term TDI_{bw} was introduced, calculated multiplying the TDI (1 pg WHO-TEQ kg^{-1} d^{-1}) by the body weight of each age class (Table 2).

The TDI_{bw} gives the maximal PCDD/F daily intake for every year of life. As showed in Table 2, the calculated TDI_{bw} is more restrictive for children, since it depends on the body weight. However, in view of the estimated long elimination half-life of PCDD/Fs, the proposed TDI should be regarded as a time-weighted average tolerable intake (COT, 2001). Considering a mean lifetime of 80 years for the local population (Italian Ministry of Health, 2004), the resulting average maximal PCDD/F daily intake is 59 pg WHO-TEQ d^{-1}. The average milk fat consumption for the lifetime is 29 g_{fat} d^{-1}. Dividing the average maximal intake by this value, a maximal tolerable PCDD/F concentration of 2.056 pg WHO-TEQ g^{-1} in cow's milk fat is obtained.

According to the formulation of Slob and Van Jaarsveld (1993), the PCDD/F concentration in milk fat (C_{mf}) can be expressed as follows:

$$C_{mf} = \frac{b \cdot I}{P_{mf}} \tag{1}$$

where b is the bioavailability of PCDD/F in the cow, I is the total daily intake of PCDD/F by the cow and P_{mf} is the average daily production of milk fat (Slob and Van Jaarsveld, 1993). The latter can be estimated starting from the average yearly milk production (P_m, expressed in g yr^{-1}), according to the relation:

Table 1. Mean daily PCDD/F depositions to soil measured at the monitoring site from the 31st of August 2010 to the 3rd of October 2011 and mean congener composition.

PCDD/F Congeners	Depositions to soil [pg WHO-TEQ m^{-2} d^{-1}]													mean composition
	2010	2010	2010	2010	2010-2011	2011	2011	2011	2011	2011	2011	2011	2011	
	31 aug 21 sep	22 sep 13 oct	14 oct 02 nov	03 nov 25 nov	24 dec 03 jan	04 jan 14 jan	01 feb 01 mar	02 mar 05 apr	08 apr 18 may	19 may 28 jun	29 jun 08 aug	09 aug 07 sep	08 sep 03 oct	[%]
2,3,7,8-TCDD	1.38E-02	8.48E-02	7.61E-03	2.60E-02	1.45E-02	1.45E-02	5.16E-03	4.15E-03	7.23E-03	3.54E-03	1.06E-02	6.49E-02	5.59E-03	1.49
1,2,3,7,8-PeCDD	4.12E-01	1.17E-01	7.61E-03	6.37E-01	1.45E-02	1.42E-01	3.99E-01	9.97E-03	3.20E-01	1.49E-02	7.08E-03	7.27E-02	2.07E-01	11.30
1,2,3,4,7,8-HxCDD	2.70E-03	6.95E-04	1.98E-03	6.75E-03	1.45E-03	6.47E-02	4.31E-02	3.90E-03	1.16E-03	2.26E-03	1.34E-03	1.45E-03	3.69E-03	0.72
1,2,3,6,7,8-HxCDD	1.24E-01	4.59E-03	1.67E-03	1.06E-01	4.35E-03	7.40E-02	1.09E-01	4.82E-02	7.15E-03	6.44E-03	3.61E-03	2.79E-02	4.58E-02	3.01
1,2,3,7,8,9-HxCDD	1.60E-03	6.95E-04	2.13E-03	1.06E-02	1.45E-03	5.49E-02	3.89E-02	7.97E-03	2.24E-03	3.33E-03	1.96E-01	1.03E-02	5.59E-03	1.30
1,2,3,4,6,7,8-HpCDD	9.25E-02	4.39E-02	4.24E-02	8.77E-02	1.57E-01	6.21E-02	3.62E-02	9.39E-03	3.42E-02	1.78E-02	1.08E-01	7.63E-02	2.17E-02	3.57
OCDD	3.80E-02	3.73E-02	2.37E-02	5.92E-02	1.92E-01	2.65E-02	2.13E-02	2.24E-02	3.07E-02	1.33E-02	1.32E-01	1.11E-01	3.71E-02	0.18
2,3,7,8-TCDF	2.45E-01	3.96E-01	5.93E-02	1.88E-01	3.45E-01	3.61E-01	1.14E-01	8.64E-02	7.98E-02	3.50E-02	1.54E-01	1.30E-01	8.17E-02	11.35
1,2,3,7,8-PeCDF	2.91E-02	7.92E-02	3.80E-04	1.41E-02	6.66E-02	8.24E-02	7.43E-02	1.48E-02	4.19E-03	5.73E-03	6.83E-02	8.23E-03	7.44E-02	2.03
2,3,4,7,8-PeCDF	5.09E-01	9.04E-01	1.25E+00	1.02E+00	1.49E+00	8.38E-01	6.30E-01	6.23E-03	5.02E-02	1.57E-01	7.29E-01	2.52E-01	2.38E+00	34.67
1,2,3,4,7,8-HxCDF	9.63E-02	1.54E-01	5.49E-02	1.93E-01	5.11E-01	1.88E-01	1.22E-01	1.86E-02	9.97E-03	7.15E-02	4.86E-01	4.09E-02	4.28E-01	8.64
1,2,3,6,7,8-HxCDF	2.12E-01	1.43E-01	1.16E-01	4.01E-01	1.23E-01	1.99E-01	1.36E-01	1.98E-02	2.02E-02	6.72E-02	1.34E-01	1.60E-02	2.60E-01	7.21
2,3,4,6,7,8-HxCDF	3.85E-03	1.89E-01	1.32E-01	1.71E-01	4.15E-01	3.35E-01	1.62E-01	9.05E-02	5.78E-04	8.42E-02	2.97E-01	4.94E-02	2.76E-01	9.79
1,2,3,7,8,9-HxCDF	1.69E-03	1.67E-03	1.52E-03	1.36E-02	1.45E-03	3.73E-02	1.87E-02	1.41E-03	1.16E-03	1.13E-03	1.78E-02	5.04E-03	1.44E-02	0.55
1,2,3,4,6,7,8-HpCDF	6.77E-02	3.81E-02	2.13E-02	5.68E-02	8.59E-02	6.82E-02	3.06E-02	3.17E-02	1.94E-02	3.23E-02	1.84E-01	2.52E-02	1.00E-01	3.70
1,2,3,4,7,8,9-HpCDF	4.31E-03	6.81E-04	3.06E-03	9.08E-03	7.20E-03	7.40E-03	1.05E-02	3.28E-03	3.18E-04	1.61E-03	1.42E-02	3.12E-03	1.80E-02	0.48
OCDF	4.76E-04	2.36E-03	6.08E-03	1.28E-02	1.48E-02	9.60E-03	4.19E-03	1.46E-03	8.96E-04	6.00E-03	3.51E-02	3.45E-03	2.30E-02	0.02
TOTAL	1.817	2.159	1.701	2.944	3.254	2.531	1.930	0.358	0.559	0.505	2.420	0.789	3.923	

Table 2. Estimated mean body weight (males and females), consumption of dairy products and consequent fat intake for a 80-year lifetime (EFSA, 2012).

age	**body weight**	**food consumption**				**total fat intake**	**TDI_{bw}**
		milk	fermented milk products	cream	cheese		
[yr]	[kg]	[g d^{-1}]	[g d^{-1}]	[g d^{-1}]	[g d^{-1}]	[g d^{-1}]	[pg WHO-TEQ d^{-1}]
<1	8	589	26	0	9	31	8
1	11	248	37	0	27	27	11
2	13	293	43	0	31	32	13
3	15	128	11	1	27	15	15
4	17	145	13	1	30	17	17
5	19	162	14	1	34	19	19
6	22	188	16	1	39	22	22
7	25	213	18	1	44	25	25
8	28	239	21	1	50	28	28
9	31	265	23	1	55	31	31
10	35	107	13	1	40	19	35
11	39	119	15	2	45	21	39
12	45	137	17	2	52	24	45
13	50	152	19	2	58	27	50
14	54	164	20	2	62	29	54
15	57	174	21	2	66	31	57
16	60	183	22	2	69	32	60
17	62	189	23	2	71	33	62
18	63	97	21	21	55	30	63
19	64	99	22	22	56	31	64
20	65	100	22	22	57	31	65
21	65	100	22	22	57	31	65
…	…	…	…	…	…	…	…
28	65	100	22	22	57	31	65
29	65	100	22	22	57	31	65
30	66	102	22	22	58	32	66
31	66	102	22	22	58	32	66
…	…	…	…	…	…	…	…
38	66	102	22	22	58	32	66
39	66	102	22	22	58	32	66
40	68	105	23	23	59	32	68
41	68	105	23	23	59	32	68
…	…	…	…	…	…	…	…
48	68	105	23	23	59	32	68
49	68	105	23	23	59	32	68
50	69	107	23	23	61	33	69
51	69	107	23	23	61	33	69
52	69	107	23	23	61	33	69
53	69	107	23	23	61	33	69
54	69	107	23	23	61	33	69
55	68	105	23	23	60	33	68
56	68	105	23	23	60	33	68
57	68	105	23	23	60	33	68
58	68	105	23	23	60	33	68
59	68	105	23	23	60	33	68
60	67	103	23	23	59	32	67
61	67	103	23	23	59	32	67
62	67	103	23	23	59	32	67
63	67	103	23	23	59	32	67
64	67	103	23	23	59	32	67
65	66	105	15	1	49	21	66
66	66	105	15	1	49	21	66
67	66	105	15	1	49	21	66
68	66	105	15	1	49	21	66
69	66	105	15	1	49	21	66
70	66	105	15	1	49	21	66
71	64	101	14	1	47	21	64
72	64	101	14	1	47	21	64
73	64	101	14	1	47	21	64
74	64	101	14	1	47	21	64
75	64	144	15	0	51	24	64
76	63	140	14	0	50	23	63
77	62	139	14	0	49	23	62
78	62	139	14	0	49	23	62
79	62	139	14	0	49	23	62
80	61	137	14	0	49	23	61

$$P_{mf} = \frac{f \cdot P_m}{t_m} \tag{2}$$

where f is the fraction of fat in cow's milk and t_m is the number of milk producing days per year (Slob and Van Jaarsveld, 1993). The maximal total daily intake of PCDD/F by the cow can be obtained combining (1) with (2):

$$I = \frac{f \cdot P_m \cdot C_{mf}}{t_m \cdot b} \tag{3}$$

where f could be assumed as 0.044 for raw milk, C_{mf} is the previously calculated value of 2.056 pg WHO-TEQ g^{-1} of fat and t_m is typically 300 d yr^{-1} (Slob and Van Jaarsveld, 1993). Assuming a milk production of about 7,000 l yr^{-1} per cow and a milk density of 1,030 g l^{-1}, the corresponding milk production in mass is 7.21 Mg yr^{-1}. Congener-specific values for the bioavailability b are listed in McLachlan (1995), on the basis of other previous studies (Stevens and Gerbec, 1988; Firestone et al., 1979; Olling et al., 1991; Heeschen et al., 1994; McLachlan, 1995; Slob et al., 1995). Since the current approach starts from the TDI for total PCDD/Fs proposed by the WHO and no TDI values are proposed for the single congeners, a value of 0.227 for b was adopted, which is the weighted average between the proposed congener-specific values of bioavailability on the basis of the TEF of each congener.

I can be seen as the sum of the PCDD/F daily intake from grass consumption (I_g) and that from soil ingestion (I_s). The first one depends on the PCDD/F deposition on the grass (seen as the total deposition D multiplied by the fraction c_g of surface covered by grass), the average area (A) of meadow grazed by one cow during one month and t_r is the average number of days per month (Slob and Van Jaarsveld, 1993), according to the following relation:

$$I_g = \frac{A \cdot c_g \cdot D}{t_r} \tag{4}$$

where A is approximately 3,000 m^2 $month^{-1}$, c_g is assumed to be 0.9 and t_r is 30.4 d $month^{-1}$.

The depositions on the grass should be corrected to account for runoff processes due to the rain. In the absence of information, no wash-off parameters were taken into account. Thus, such an approach leads to precautionary results. The PCDD/F daily intake from soil ingestion (I_s) depends on the amount of soil ingested (m_s) and the dioxin concentration in soil (C_s):

$$I_s = m_s \cdot C_s \tag{5}$$

According to Slob and Van Jaarsveld (1993), m_s can be assumed as 225 g d^{-1}. An expression for C_s is provided in Lorber et al. (2000):

$$C_s = D_{LT} \frac{1 - \exp(-k \cdot t_d)}{k \cdot M} \tag{6}$$

where D_{LT} represents the long-term deposition, intended as mean value of the total annual PCDD/F deposition (D_{LT}=12D), k is the first-order annual soil dissipation rate, t_d is the time (in years) since the annual deposition D_{LT} occurred and M is the soil mixing mass. This expression is included in a reservoir mixing model for predicting soil concentrations from available long-term deposition data (Lorber et al., 2000). A mid-range value for k is 0.02772 yr^{-1}, which corresponds to the PCDD/F half-life time of 25 yr used in Lorber et al. (2000). A value of 112.5 kg m^{-2} for M was chosen, which is the product between the soil bulk density (assumed to be 1,500 kg m^{-3}) and the maximal penetration depth of dioxins in soil (assumed to be 0.075 m) (Lorber et al., 2000). In order to obtain a deposition limit value for the future years, C_s will be calculated considering a temporal horizon of 30 yr, assuming D_{LT} equal to the average annual deposition measured during the monitoring campaign, which is 608 pg WHO-TEQ m^{-2} yr^{-1}. Using this value in (6) implies the assumption that a mean PCDD/F deposition of 608 pg WHO-TEQ m^{-2} will be encountered annually for the whole reference period of 30 yr.

I can now be expressed as a function of the total deposition D:

$$I = I_g + I_s = A \cdot t_g \cdot c_g \cdot D + m_s \cdot C_s \tag{7}$$

Thus, the deposition can be calculated as follows:

$$D = \frac{1}{A \cdot t_g \cdot c_g} \left(\frac{f \cdot P_m \cdot C_{mf}}{t_m \cdot b} - m_s \cdot C_s \right) \tag{8}$$

By attributing the maximal PCDD/F concentration value in milk fat to C_{mf}, D represents a limit value for the PCDD/F deposition on a monthly basis.

In addition to the consumption of dairy products, also cereals and vegetables represent an important route of dioxin intake, even though it is of secondary importance (Eduljee and Gair, 1996). The PCDD/F plant uptake mainly occurs through three pathways: interaction between soil and root system, particulate deposition and dry gaseous deposition to above-ground shoots, the first two playing a minor role with respect to the latter (Harrad and Smith, 1997; UK Environment Agency, 2006; UK Environment Agency, 2009a).

The contribution of root uptake ($C_{p,r}$) can be calculated by means of the soil-to-plant concentration factors (BCFs) used in the CLEA plant uptake models (UK Environment Agency, 2009b) and in Hülster et al. (1994) (Table 3). The BCFs refer to the following plant groups: green vegetables, root vegetables, tuber vegetables, herbaceous fruits and tree fruits. The PCDD/F concentration into the plant is calculated multiplying the BCFs by the PCDD/F concentration in soil. Since the aim of this part of the study is to evaluate the role of cereals and vegetables for the local contribution to the dioxin intake, in comparison with dairy products, C_s will be calculated from (6), starting from the long-term deposition value measured for the case study (608 pg WHO-TEQ m^{-2} yr^{-1}) and assuming the temporal horizon of 30 yr used for the calculation of the deposition limit value.

The dioxin concentration into the plant due to the contribution of dry gaseous deposition to above-ground shoots and leaves ($C_{p,dg}$) can be estimated through the following formulations presented in Harrad and Smith (1997), Lorber et al. (1994), Junge (1977) and Bidleman (1988):

$$C_{p,dg} = \frac{B_{vpa} \cdot f_v \cdot C_a}{\rho_a} \tag{9}$$

where B_{vpa} is the congener-specific air-to-leaves transfer factor (Table 3), ρ_a is the air density, assumed as 1.19 kg m^{-3} (Lorber et al., 1994), C_a is the dioxin concentration in air and f_v is the fraction of total air concentration in vapor phase at 293 K and is complementary to the fraction of total air concentration sorbed to particulates at the same temperature (f_p). The latter is obtained as follows:

Table 3. Congener-specific parameters adopted for calculating the contribution of root uptake, dry gaseous and particulate depositions to the PCDD/F concentration of vegetables (Menses et al., 2002; UK Environment Agency, 2009b; Hülster et al., 1994; Harrad and Smith, 1997).

PCDD/F Congeners	**WHO-TEF**	**v_d**	**C_s**	**C_a**	**BCF**					**B_{vpa}**	**P_l**	**f_p**	**f_v**
					green veg.	root veg.	tuber veg.	tree fruit	herb. fruit				
	[-]	[m s^{-1}]	[pg WHO-TEQ g^{-1}]		[-]					[-]	[atm]	[-]	[-]
2,3,7,8-TCDD	1	0.069	1.74E-03	2.80E-04	2.59E-05	2.38E-04	1.72E-04	9.33E-08	1.02E-02	1.43E+05	7.16E-10	4.54E-01	5.46E-01
1,2,3,7,8-PeCDD	1	0.091	1.32E-02	2.12E-04	4.68E-06	9.28E-05	6.60E-05	8.57E-09	1.02E-02	2.78E+05	2.13E-10	7.37E-01	2.63E-01
1,2,3,4,7,8-HxCDD	0.1	0.166	8.39E-04	1.16E-04	7.36E-07	3.62E-05	2.51E-05	6.51E-10	6.67E-02	1.06E+06	4.68E-11	9.27E-01	7.29E-02
1,2,3,6,7,8-HxCDD	0.1	0.166	3.52E-03	1.16E-04	7.36E-07	3.62E-05	2.51E-05	6.51E-10	6.67E-02	1.06E+06	2.18E-11	9.65E-01	3.54E-02
1,2,3,7,8,9-HxCDD	0.1	0.166	1.52E-03	1.16E-04	7.36E-07	3.62E-05	2.51E-05	6.51E-10	6.67E-02	1.06E+06	1.14E-11	9.81E-01	1.88E-02
1,2,3,4,6,7,8-HpCDD	0.01	0.291	4.17E-03	6.63E-05	1.01E-07	1.41E-05	9.43E-06	4.03E-11	1.60E-03	8.09E+05	1.23E-11	9.80E-01	2.02E-02
OCDD	0.0001	0.426	2.12E-04	4.53E-05	1.21E-08	5.47E-06	3.55E-06	2.00E-12	2.90E-04	2.01E+07	1.29E-12	9.98E-01	2.17E-03
2,3,7,8-TCDF	0.1	0.150	1.33E-02	1.29E-04	1.38E-04	6.54E-04	4.53E-04	9.73E-07	7.54E-03	1.90E+05	1.45E-09	2.90E-01	7.10E-01
1,2,3,7,8-PeCDF	0.05	0.178	2.37E-03	1.08E-04	3.11E-05	2.67E-04	1.87E-04	1.19E-07	8.27E-03	1.90E+05	4.26E-10	5.83E-01	4.17E-01
2,3,4,7,8-PeCDF	0.5	0.178	4.05E-02	1.08E-04	3.11E-05	2.67E-04	1.87E-04	1.19E-07	8.27E-03	1.90E+05	2.54E-10	7.01E-01	2.99E-01
1,2,3,4,7,8-HxCDF	0.1	0.178	1.01E-02	1.08E-04	6.20E-06	1.09E-04	7.54E-05	1.26E-08	5.51E-03	3.48E+05	3.79E-11	9.40E-01	5.99E-02
1,2,3,6,7,8-HxCDF	0.1	0.178	8.42E-03	1.08E-04	6.20E-06	1.09E-04	7.54E-05	1.26E-08	5.51E-03	3.48E+05	7.13E-11	8.93E-01	1.07E-01
2,3,4,6,7,8-HxCDF	0.1	0.178	1.14E-02	1.08E-04	6.20E-06	1.09E-04	7.54E-05	1.26E-08	5.51E-03	3.48E+05	4.04E-11	9.36E-01	6.35E-02
1,2,3,7,8,9-HxCDF	0.1	0.178	6.41E-04	1.08E-04	6.20E-06	1.09E-04	7.54E-05	1.26E-08	5.51E-03	3.48E+05	4.16E-11	9.35E-01	6.53E-02
1,2,3,4,6,7,8-HpCDF	0.01	0.107	4.33E-03	1.80E-04	1.12E-06	4.45E-05	3.00E-05	1.17E-09	1.74E-03	1.04E+06	2.69E-11	9.57E-01	4.32E-02
1,2,3,4,7,8,9-HpCDF	0.01	0.107	5.66E-04	1.80E-04	1.12E-06	4.45E-05	3.00E-05	1.17E-09	1.74E-03	1.04E+06	1.51E-11	9.75E-01	2.48E-02
OCDF	0.0001	0.065	2.60E-05	2.97E-04	1.74E-07	1.81E-05	1.19E-05	8.68E-11	8.70E-04	2.97E+06	1.25E-12	9.98E-01	2.09E-03

$$f_p = \frac{c \cdot S_t}{P_l + c \cdot S_t} \tag{10}$$

where c is the Junge constant ($1.7 \cdot 10^{-4}$ atm cm), S_t is the surface area of airborne particulates (assumed as $3.5 \cdot 10^{-6}$ cm^2 cm^{-3}) and P_l is the saturation vapor pressure of sub-cooled liquid. C_a can be calculated using congener-specific deposition velocities (v_d) and multiplying each of them by the respective measured deposition (Menses et al., 2002). Congener-specific parameters (v_d, BCFs, B_{vpa}) and the calculated congener-specific values for C_s, C_a, P_l and f_p are presented in Table 3. Since BCFs are provided for the single congeners, a C_s value for each congener was calculated considering the respective measured long-term total deposition value.

The PCDD/F concentration into the plant due to the contribution of particulate deposition ($C_{p,p}$) can be obtained as follows:

$$C_{p,p} = \frac{D_{LT} \cdot R_p}{Y_p \cdot k_p}\left[1 - \exp\left(-k_p \cdot T_p\right)\right] \tag{11}$$

where Y_p is the mean dry aerial biomass of grass (0.25 kg m^{-2}), k_p is the plant surface particle loss rate constant (126.6 yr^{-1}), T_p is the duration of vegetation exposure to deposition per harvest (0.12 yr) and R_p is the fraction of particles intercepted by the vegetation (0.51) (Harrad and Smith, 1997).

Finally, accumulation in fruit ($C_{p,f}$), due to the uptake from air and stem, can be estimated by means of the tree model developed by Trapp (2007), which was originally calibrated on an apple orchard.

3. RESULTS AND DISCUSSION

3.1 Verification of the main intake pathway

To confirm the main role of dairy products in the exposure to dioxin, the daily PCDD/F intake was calculated for cow's milk consumption, starting from the data about the Italian diet and from the deposition values measured from September 2010 to September 2011 for the case study and following the approaches presented in Slob and Van Jaarsveld (1993) and Lorber et al. (2000). The PCDD/F daily intake for the cow is the sum of the daily intake of dioxins due to soil ingestion and grass consumption. By applying (7), the total PCDD/F intake for the cow is 4.51 ng WHO-TEQ d^{-1}.This calculation was carried out assuming D as the mean daily deposition value measured during the period of study (1.67 pg WHO-TEQ m^{-2} d^{-1}) and adopting the previously presented values for A, t_g, c_g, m_s, D_{LT}, k, t_d and M. The PCDD/F concentration in milk fat can be calculated from (3), (2) and (1), resulting in 0.97 pg WHO-TEQ g^{-1}.

Similarly, the contribution of cereals and vegetables to the daily PCDD/F intake was estimated. Since the Italian and South European diet quite differs from the rest of Europe, being cereal products and vegetables generally more present, it is necessary to clarify the role of these products and verify if their consumption does not involve a greater PCDD/F intake. A total value of $C_{p,r}$ was then calculated for every plant group considered. By applying equations (9), (10) and (11), $C_{p,dg}$ and $C_{p,p}$ were also obtained. Finally, accumulation in fruit ($C_{p,f}$) was calculated (Table 4). For each congener, $C_{p,f}$ was two orders of magnitude lower than $C_{p,dg}$ and one order lower than $C_{p,p}$. In addition, $C_{p,r}$ was even lower than $C_{p,f}$ for green vegetables, tubers, root vegetables and tree fruit. $C_{p,r}$ was higher than $C_{p,f}$ (of one order of magnitude) only for herbaceous fruit (Table 4).

Table 4. Congener-specific concentrations related to different contributions: root uptake ($C_{p,r}$), dry gaseous deposition ($C_{p,dg}$), particulate deposition ($C_{p,p}$) and accumulation in tree fruit ($C_{p,f}$).

PCDD/F Congeners	$C_{p,r}$ green veg.	root veg.	tuber veg.	tree fruit	herb. fruit	$C_{p,dg}$	$C_{p,p}$	$C_{p,f}$
	[pg WHO-TEQ g^{-1}]							
2,3,7,8-TCDD	4.52E-08	4.15E-07	3.00E-07	1.63E-10	1.78E-05	1.84E-02	1.46E-04	6.34E-04
1,2,3,7,8-PeCDD	6.18E-08	1.23E-06	8.71E-07	1.13E-10	1.35E-04	1.31E-02	1.11E-03	4.95E-04
1,2,3,4,7,8-HxCDD	6.17E-10	3.04E-08	2.10E-08	5.46E-13	5.59E-05	7.54E-03	7.03E-05	3.43E-05
1,2,3,6,7,8-HxCDD	2.59E-09	1.27E-07	8.83E-08	2.29E-12	2.35E-04	3.66E-03	2.95E-04	3.43E-05
1,2,3,7,8,9-HxCDD	1.12E-09	5.51E-08	3.82E-08	9.91E-13	1.02E-04	1.95E-03	1.28E-04	1.37E-04
1,2,3,4,6,7,8-HpCDD	4.21E-10	5.88E-08	3.93E-08	1.68E-13	6.67E-06	9.10E-04	3.49E-04	2.17E-05
OCDD	2.56E-12	1.16E-09	7.52E-10	4.24E-16	6.14E-08	1.66E-03	1.78E-05	1.59E-05
2,3,7,8-TCDF	1.83E-06	8.67E-06	6.01E-06	1.29E-08	1.00E-04	1.46E-02	1.11E-03	2.33E-04
1,2,3,7,8-PeCDF	7.38E-08	6.34E-07	4.44E-07	2.82E-10	1.96E-05	7.22E-03	1.99E-04	1.58E-04
2,3,4,7,8-PeCDF	1.26E-06	1.08E-05	7.58E-06	4.82E-09	3.35E-04	5.17E-03	3.40E-03	9.67E-05
1,2,3,4,7,8-HxCDF	6.26E-08	1.10E-06	7.61E-07	1.27E-10	5.56E-05	1.90E-03	8.46E-04	6.07E-05
1,2,3,6,7,8-HxCDF	5.22E-08	9.18E-07	6.35E-07	1.06E-10	4.64E-05	3.39E-03	7.06E-04	6.07E-05
2,3,4,6,7,8-HxCDF	7.09E-08	1.25E-06	8.62E-07	1.44E-10	6.30E-05	2.01E-03	9.59E-04	1.15E-04
1,2,3,7,8,9-HxCDF	3.97E-09	6.99E-08	4.83E-08	8.08E-12	3.53E-06	2.07E-03	5.37E-05	6.46E-05
1,2,3,4,6,7,8-HpCDF	4.84E-09	1.92E-07	1.30E-07	5.06E-12	7.53E-06	6.80E-03	3.63E-04	4.39E-05
1,2,3,4,7,8,9-HpCDF	6.34E-10	2.52E-08	1.70E-08	6.62E-13	9.84E-07	3.90E-03	4.74E-05	6.03E-05
OCDF	4.52E-12	4.70E-10	3.09E-10	2.26E-15	2.26E-08	1.55E-03	2.18E-06	2.45E-05
TOTAL	3.47E-06	2.56E-05	1.78E-05	1.87E-08	1.18E-03	9.57E-02	9.80E-03	2.29E-03

The total PCDD/F concentrations in vegetables were then calculated as follows: the contribution of root uptake was considered for every group, accumulation in fruit was taken into account only for tree fruit, whilst the contribution of dry gaseous and particulate depositions were considered only for green vegetables and herbaceous fruit. Using the statistics on the Italian diet (EFSA, 2012), the PCDD/F daily intake was calculated multiplying the mass of vegetables ingested by the respective PCDD/F content. The resulting contribution of vegetables is comprised between 3% and 51% (mean value over the lifetime: 33%) with respect to the contribution of dairy products. This confirms the main role of the latter in the daily absorption of dioxins.

3.2 Deposition limit proposal and comparison with the measured deposition

To investigate the roles of the daily intake from grass consumption (I_g) and soil ingestion (I_s), these two terms were calculated separately through the equations (4), (5), (6), on the basis of the measured deposition: I_s turned out to be three orders of magnitude lower than I_g, thus the total intake I can be assumed equal to I_g. As a consequence, equation (8) can be simplified as follows:

$$D = \frac{f \cdot P_m \cdot C_{mf}}{A \cdot t_g \cdot c_g \cdot t_m \cdot b} \tag{12}$$

By applying this formulation, the resulting limit value for PCDD/F deposition is 3.55 pg WHO-TEQ m^{-2} d^{-1}. Dealing with carcinogenic pollutants, this value must be intended as average over a long period (for instance one year). This value is half of the deposition limit proposed by van Lieshout et al. (2001), which is 6.8 pg WHO-TEQ m^{-2} d^{-1}. For the case study, an average PCDD/F deposition of 1.67 pg WHO-TEQ m^{-2} d^{-1} was encountered during the one-year monitoring campaign. Thus, the present deposition is below the resulting limit value. The here calculated limit value should be intended as a threshold value below which the population can be considered subjected to a low PCDD/F exposure, since the present study started from a TDI of 1 pg WHO-TEQ m^{-2} kg^{-1} d^{-1}, that is the lower bound of the range suggested by the WHO (1-4 pg WHO-TEQ m^{-2} kg^{-1} d^{-1}). Therefore, a 4 times higher deposition limit can represent a threshold over which an investigation about the PCDD/F content of the locally produced food becomes opportune.

According to the approach presented here, the obtained deposition value is calculated only with respect to the consumption of dairy products, assuming that local products represent 100% of the diet for the local population.

4. CONCLUSIONS

An approach to derive a limit value for PCDD/F depositions to soil, starting from the diet of a region located in the North of Italy was presented. The calculation was carried out by applying existing reservoir models for predicting PCDD/F accumulation in cow's milk. The models were run backwards with respect to their original purpose, since the aim of this study was to calculate a PCDD/F deposition value assuming a dioxin daily intake of 1 pg WHO-TEQ m^{-2} kg^{-1} d^{-1} of body weight by from consumption of dairy products (which contribute most to the transfer of PCDD/F in humans) and for a lifetime of 80 yr. A deposition value of 3.55 pg WHO-TEQ m^{-2} d^{-1} was obtained, which could be considered a limit value, since the assumed daily intake of 1 pg WHO-TEQ m^{-2} kg^{-1} d^{-1} of body weight represents the TDI proposed by the WHO. The average PCDD/F deposition measured during the monitoring campaign in a selected site was 1.67 pg WHO-TEQ m^{-2} d^{-1}, thus lower than the resulting limit value.

The comparison between the contributions of dairy products and cereals, vegetables and fruits to the PCDD/F intake confirms that milk derived products play a major role in the exposure to dioxins. The calculated limit value overestimates the real PCDD/F daily intake because the consumption of local products was assumed to represent 100% of the diet.

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