APPLICATION OF SEWAGE SLUDGE ON AGRICULTURAL SOILS: A SAFE PRACTICE?

Passuello A[1], Nadal M[2], Schuhmacher M[1], Domingo JL[2]

[1]Environmental Engineering Laboratory, Departament d'Enginyeria Quimica, Universitat Rovira i Virgili, Av. Països Catalans 26, 43007 Tarragona, Catalonia, Spain; [2]Laboratory of Toxicology and Environmental Health, School of Medicine, IISPV, Universitat Rovira i Virgili, Sant Llorenç 21, 43201 Reus, Catalonia, Spain

Introduction

Sewage sludge (SS) is the main residue of wastewater treatment plants. Due to the improvement of wastewater collection and treatment systems, SS production has increased in recent years. Its recycling for agricultural purposes is a suitable option to handle the increasing quantities of sludge annually produced. Sewage sludge contains contaminants of concern, which could be potentially transferred to the food chain (i.e., polychlorinated dibenzo-*p*-dioxins and dibenzofurans, PCDD/Fs)[1]. These persistent pollutants are highly toxic, characterized by being subject to bioaccumulation potential and long-range transport capacity.

Recently, we developed an integrated model to assess the human health risks due to long-term SS application on agricultural soils[2]. The model, designed for persistent organic pollutants (POPs), takes into consideration diverse parameters such as application dose, soil characteristics, transfer parameters, and dietary habits. Ultimately, the execution of the model allows modeling environmental concentrations of a specific organic chemical, and characterizing the long-term human exposure and the associated health risks. The application of this tool may be useful to guide future legislative efforts regarding this practice.

In this study, we have assessed the potential increase on human health risks as a consequence of amending agricultural soils with SS. For that purpose, 2,3,7,8-TCDD fate from sludge to the human food chain was studied, while the trends in its levels in soil, plants, meat and milk were determined. The previously referred model was applied considering current data from Catalonia (NE Spain). Finally, the effect of the uncertainty present in the model on the predicted values was studied within a probabilistic framework.

Materials and methods

Details of the model here applied were recently reported[2]. Sewage sludge was supposed to be applied once per year. The mean concentration of 2,3,7,8-TCDD ($7.51 \cdot 10^{-1}$ ng kg dry weight $(dw)^{-1}$) was obtained from a previous survey where the levels of PCDD/Fs were determined in 200 sludge samples from wastewater plants across Spain. A high application rate was considered (30 ton SS $\cdot$ $ha^{-1} \cdot yr^{-1}$), with the objective of simulating a worst-case scenario. As the present study was aimed at defining the risk increase related to SS amendment, the initial concentration of 2,3,7,8-TCDD in soil was set to zero, and no external sources were considered. The model was applied for a time span of 30 years with a time step of one day. Transfer processes considered in the soil model were volatilization, diffusion and chemical degradation.

The plant model was calculated considering 3 transfer processes (root uptake, particle deposition, and diffusion plant-air) for 2 types of vegetables (lettuce and grass). While the concentration in lettuce was used to calculate human exposure, the concentration in grass was related to cattle exposure. The food chain model considered the ingestion of soil and grass by cattle, and evaluated the concentration of 2,3,7,8-TCDD in cattle milk and beef. Finally, two scenarios were selected for human exposure: occupational (farmers) and non-occupational (general population). The selected human exposure routes were air inhalation, food and soil ingestion for the occupational scenario, and food ingestion for the non-occupational scenario.

The interrelation between the submodels is depicted in Figure 1. Uncertainty analysis was performed through the generation of 5,000 scenarios by means of Monte Carlo analysis.

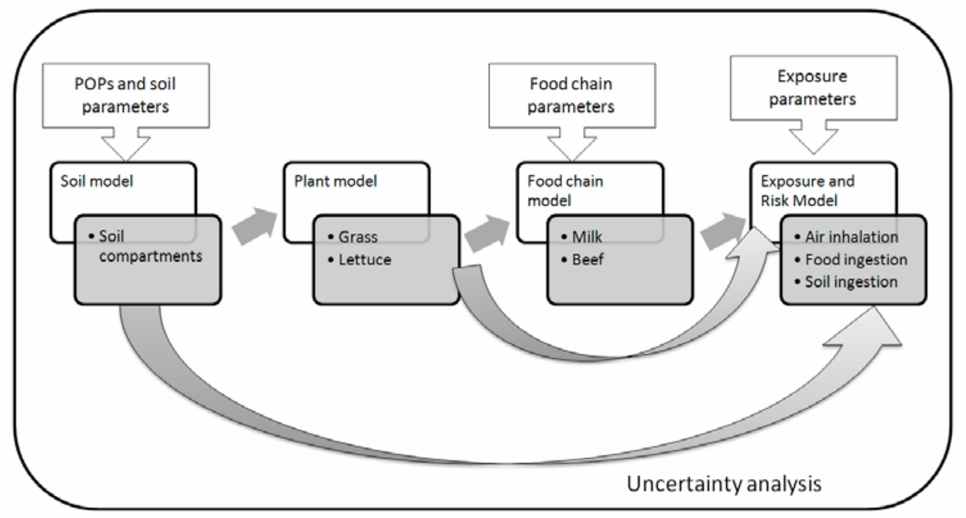

Figure 1: Schematic representation of the model development (adapted from Passuello et al.[2])

Results and discussion:

The probabilistic trends in the soil concentration of 2,3,7,8-TCDD, for a period of 30 years are shown in Figure 2. Increased mean levels were observed through the evaluated period, indicating that this congener tends to accumulate in soils. As expected, the uncertainty study showed higher ranges of concentrations in relation to the period of time assessed, with a notable long-term uncertainty. However, by the end of the evaluation period (30 years), a tendency of reaching a steady state was observed. Moreover, when compared with experimental data, the final concentration obtained from the model was similar to that found in a sampling study performed in Catalonia[3], with soils presenting the same physical-chemical properties. Nevertheless, it must be noted that this comparison is only illustrative, as pollutant inputs from external sources were not considered here.

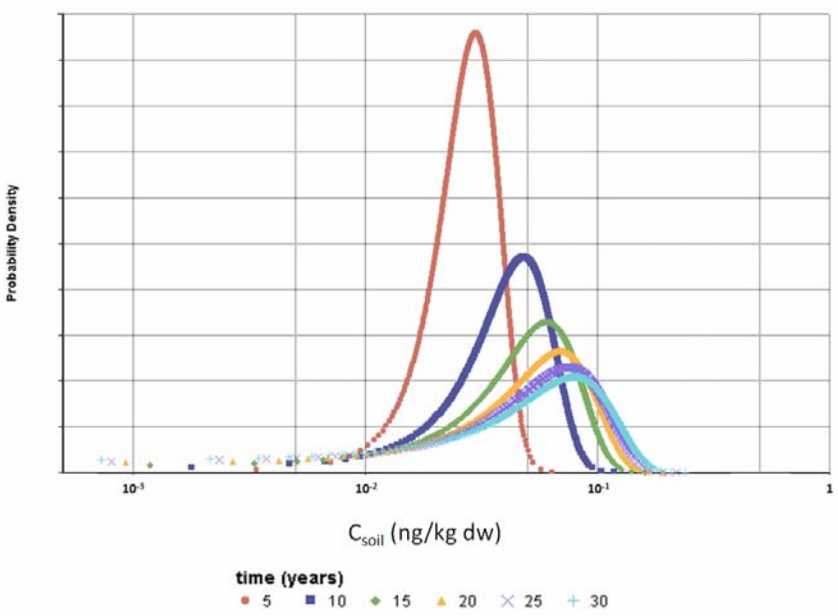

Figure 2: Concentration of 2,3,7,8-TCDD in soil (ng kg^{-1} dw) along the simulation time. Colors indicate the distribution of the calculated levels for the evaluated time spans.

Table 1 summarizes the probabilistic results of 2,3,7,8-TCDD levels in grass and lettuce. The concentrations after 30 years of application in both types of plants were similar (mean: 0.67 and 0.70 pg · kg dw^{-1}, respectively). Among the evaluated transfer processes, diffusion plant-air was the most contributive pathway, with 90% of the input fluxes, as a consequence of the volatilization levels obtained. In contrast, root transfer was found to be the minor process (less than 1%).

Table 1: Total concentration of 2,3,7,8-TCDD in grass and lettuce (mg kg^{-1}) for a simulation time of 30 years.

Grass				**Lettuce**			
		Percentiles				Percentiles	
Mean	St. Dev.	50th	90th	Mean	St. Dev.	50th	90th
6.69E-10	3.00E-10	6.63E-10	1.06E-09	7.00E-10	2.91E-10	6.97E-10	1.07E-09

The calculated values of 2,3,7,8-TCDD levels in both meat and milk are shown in Table 2. Soil ingestion was found to be the most important route for cattle, representing more than 99% of 2,3,7,8-TCDD intake.

Table 2: Total concentration of 2,3,7,8-TCDD in meat and milk (mg kg^{-1}) for a simulation time of 30 years.

Meat				**Milk**			
		Percentiles				Percentiles	
Mean	SD	50th	90th	Mean	SD	50th	90th
1.08E-08	7.20E-09	8.77E-09	2.16E-08	7.05E-09	5.05E-09	5.27E-09	1.38E-08

The human exposure to 2,3,7,8-TCDD through different pathways is presented in Table 3, while the associated cancer and non-cancer risks are shown in Table 4. The ingestion of food cultivated in the SS-amended agricultural soils was found to be the most contributive (72%), contrasting with the soil ingestion (0.1%). However, those values were not of concern in comparison to environmental exposure levels reported in the scientific literature, and also considering the dietary intake of dioxins[4].

Table 3: Environmental exposure to 2,3,7,8-TCDD (mg kg^{-1} day^{-1}) through different routes for the two scenarios. Simulation time: 30 years.

	Occupational				**Non-occupational**			
			Percentiles				Percentiles	
	Mean	SD	50th	90th	Mean	SD	50th	90th
Air inhalation	6.67E-12	2.72E-12	6.82E-12	1.07E-11				
Soil ingestion	1.65E-14	1.12E-14	1.32E-14	3.41E-14				
Food ingestion	1.72E-11	1.68E-11	1.21E-11	3.74E-11	5.82E-12	5.23E-12	3.57E-12	1.22E-11

In the evaluated scenarios, the hazard quotient (HQ) for 2,3,7,8-TCDD was below the threshold value, set at 1.0, being the highest mean HQ value found for ingestion in the occupational scenario ($1.72 \cdot 10^{-2}$). The highest cancer risk values were associated to the ingestion pathway, mainly food. The mean oral cancer risk for ingestion was $6.86 \cdot 10^{-7}$ for the occupational scenario, and $2.26 \cdot 10^{-7}$ for the non-occupational scenario. The 90th percentiles for the oral cancer risk were $1.31 \cdot 10^{-6}$ and $4.14 \cdot 10^{-7}$ for farmers and local residents, respectively. This difference would be related to the consumption rate of foods produced in the area, which is higher for the farmers than for the general population.

Cancer risk due to inhalation was lower than that derived from the ingestion of pollutants, presenting a mean value of $1.14 \cdot 10^{-9}$ (occupational scenario only). However, none of the exposure pathways showed risk values above the threshold (range: 10^{-6}–10^{-4}). Therefore, the current concentrations of 2,3,78-TCDD in SS used for agricultural soil-amending did not mean an important increase on human health risks for both occupationally and non-occupationally exposed populations, if the described conditions are maintained.

Table 4: Carcinogenic and non-carcinogenic risks due to environmental exposure to 2,3,7,8-TCDD. Simulation time: 30 years.

	Occupational				**Non-occupational**			
			Percentiles				Percentiles	
	Mean	SD	50th	90th	Mean	SD	50th	90th
Oral cancer risk	6.86E-07	4.98E-07	5.57E-07	1.31E-06	2.26E-07	1.46E-07	1.8E-07	4.14E-07
Oral HQ	1.72E-02	1.68E-02	1.21E-02	3.74E-02	5.82E-03	5.23E-03	3.57E-03	1.22E-02
Inhalation cancer risk	1.14E-09	2.33E-10	1.12E-09	1.45E-09				
Inhalation HQ	1.67E-04	6.81E-05	1.71E-04	2.67E-04				

HQ: Hazard Quotient

It is important to remark that POP concentrations in SS may vary in several orders of magnitude. The results of the current study are related to the concentration of 2,3,7,8-TCDD, a PCDD/F congener with a high toxicity and environmental persistence. However, depending on the physical-chemical properties and the toxicological characteristics of each congener, its accumulation in soil may differ. Therefore, legislation efforts should consider the evaluation of the different congeners to define the safe levels of organic contaminants on SS matrix intended to be used as an organic amendment. In this context, the applicability of risk models, such as that here presented, may be of high usefulness for stakeholders participating in decision-making processes, as they usually need powerful tools of easy comprehension for the general public.

Acknowledgements:

Financial support was provided by the Ministry of Science and Innovation, Spain, through grant No. CTM2007-64490, and the SOSTAQUA project, founded by CDTI in the framework of the Ingenio 2010 Program under the CENIT call. Ana Passuello received a PhD studentship from AGAUR (Commissioner for Universities and Research of the Department of Innovation, Universities and Enterprises of the “Generalitat de Catalunya” and the European Social Fund).

References:

1. Eljarrat E, Caixach J, Rivera J. (2003); *Chemosphere* 51: 595-601
2. Passuello A, Mari M, Nadal M, Schuhmacher M, Domingo JL. (2010); *Environ Int.* 36: 577-83
3. Nadal M, Mari M, Schuhmacher M, Domingo JL. (2009); *Environ Int.* 35: 227-35
4. Nadal M, Schuhmacher M, Domingo JL. (2004); *J Environ Monit.* 6: 926-31