Probabilistic health risk assessment of PCDD/F emissions from MSW incineration

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Introduction

Potential public health effects associated with the emissions of toxic trace contaminants have significant implications in current regulatory practice for municipal solid waste incineration plants (MSWI). Human risk assessment is actually involved in decision processes for siting of new facilities and for the evaluation of design retrofit options of existing plants, as well as in addressing emission limits and standards included in the regulations. Quantitative risk assessment has to be conducted through a rather complex multipathway approach, by considering the environmental distribution of the emitted contaminant for evaluating the intake of the human subject arising from direct and indirect exposure and assessing the resulting final risk level through dose-response relationships. The evaluation tools utilised in the field during the last years[1,2] are essentially based on several simulation models, each requiring a relatively broad set of input parameters affected by a certain degree of uncertainty, arising from lack of knowledge and from intrinsic variations of the particular data value, and influencing with more or less significance the final risk calculated. More recently, a significant emphasis in the analysis of risk has focused on the possibilities of incorporating uncertainties into final risk estimates by the integration of the conventional procedure with proper statistical techniques. This probabilistic approach is essentially based on the utilisation of distribution functions for describing the variability of the input parameters and to propagate this variability throughout the calculation with Monte Carlo or modified Monte Carlo methods[3,4]: final risk results are consequently obtained in terms of probability distributions of the expected values, instead of single point estimates normally derived from the conventional deterministic applications.

The approach outlined is utilised for evaluating carcinogenic risk distributions associated to the uncertainty of the main emission parameters of stack gas

PCDD/Fs from a waste to energy plant. Results are compared with values derived from conventional deterministic applications and the effect of the uncertainty of every emission parameter is analysed.

Materials and Method

Methodological approach: **The main objective of the analysis is the identification and quantification of predicted health risk uncertainty arising from variability in PCDD/F emission characteristics and essentially associated to design options adopted for combustion chamber and flue gas treatment and to normal fluctuations during plant operation. Consequently, PCDD/F risk evaluation is conducted through a combined probabilistic/deterministic approach: probability density functions are thus utilised for describing the variability of the parameters associated with the estimation of emission rates, dry and wet deposition fluxes and concentrations of PCDD/F in soil, whilst constant point values are adopted for input data required for the evaluation of exposure and toxicity. Distribution functions were derived from best data fitting through standard statistical techniques (Kolmogorov-Smirnov's test) and the resulting variability propagated throughout the risk calculation utilising a Monte-Carlo simulation, with a sample size of 10000 randomly drawn parameter values: all the evaluations were performed with the commercial package Crystal Ball[5].**

Distribution functions for PCDD/F emitted concentrations and gas-particle partitioning were derived from a data set consisting of several stack measurements performed on full scale MSWI during continuous operating regime at design burning capacity located in Northern Italy[6] and of similar available literature data[7,8,9,10]. The data series includes 198 concentration values, considered as a whole and also in terms of distributions arising from different air pollution control equipment (APCD): prior to distribution fitting, the series were analysed for statistical detection and elimination of outliers through Huber's test. The same approach was utilised for evaluating probability distribution of PCDD/F gas-particle partitioning, in terms of the fraction V of total toxicity equivalents emitted in vapour phase available in the data set.

The variability of dry deposition flux (D_D), due to both variability and uncertainty of the granulometric distribution of the total suspended particulate (TSP) emitted, was analysed through the probability distribution of simulated D_D values obtained by running ISC3 model (described later in this section) with seven lognormal TSP granulometric distributions as input, available from literature.

Since direct measurements of size fractionated PCDD/Fs concentrations on emitted particulates were not available, the corresponding distributions were calculated by assuming a partitioning proportional to the surface to volume ratio of every representative fraction: under this assumption, PCDD/Fs mass distribution on emitted particulates corresponds thus to the TSP granulometric surface distribution. Wet deposition flux D_W was separately calculated for the gas phase and for the particle phase by the following equation:

$$D_w = C \cdot W \cdot H \qquad (1)$$

where C is the atmospheric concentration of PCDD/F (gas or particle phase) at ground level, W the washout coefficient and H the annual mean rainfall depth. Probability distributions for C, W and H were utilised as inputs in equation (1) to obtain a probability distribution for D_W. Distributions of washout coefficients for gas phase W_V and particle phase W_P were derived from literature data[11,12].

The concentration of PCDD/F in soil (C_S), deriving from dry and wet deposition processes was calculated with the following equation:

$$C_S = \frac{(D_D + D_W) \cdot \left[1 - \exp(-K_S \cdot T)\right]}{z \cdot K_S \cdot BD} \qquad (2)$$

where K_S is the PCDD/F soil loss constant, T is the period of exposure, z the soil mixing depth and BD the soil bulk density. Probability distributions for K_S, z and BD were derived from available literature data[13], while a period of exposure of 30 years was assumed, as usual for similar risk assessment evaluations. According to the results of a preliminary sensitivity analysis performed on equation (2), the soil mixing depth z has a largely predominant role in determining the variability of C_S, whereas a negligible effect is due to the soil loss constant K_S and bulk density BD.

Case study definition: The approach outlined was applied for evaluating carcinogenic risk distributions associated to stack gas PCDD/Fs emission of a MSWI incineration plant in its present (Scenario 1) and past configuration (Scenario 2). The actual plant has a design burning capacity of 1000 $t_{MSW}d^{-1}$ and is equipped with flue gas treatment configured with selective non catalytic reduction (SNCR), electrostatic precipitation, dry system absorption with injection of activated carbon (AC) and final fabric filtration for respecting the 0.1 $ng_{I\text{-}TEQ}\ m_n^{-3}$ emission limit established for PCDD/Fs by current regulation. The plant previously operating in the same area had a capacity of about 300 $t_{MSW}\ d^{-1}$ and was equipped with a flue gas treatment consisting of an electrostatic precipitator and a wet scrubber, not specifically designed for removing organic trace pollutants. Former regulations set an emission limit of 4 µg m_n^{-3} as total PCDD/F mass, roughly corresponding to 80 $ng_{I\text{-}TEQ}\ m_n^{-3}$.

Atmospheric transport and diffusion of emitted PCDD/Fs were simulated with ISC3 model[14] runs in the long term version for calculating annual average concentration and total (dry + wet) deposition fluxes in an area of 20x20 km around the facility. At each receptor point of the area, Monte Carlo simulation techniques were applied for the determination of the distribution of gas and solid phase ground-level concentration of PCDD/F, as well as total annual deposition and soil concentrations. Dry deposition phenomena was considered not to be active during rain events; wet deposition D_W was evaluated in terms of the probability distribution obtained as described previously coupled with a proper distribution for the time-fraction of the year during which rain events occur in the area. Impact pathways considered for human health risk include direct inhalation of gas and particle, soil dermal absorption, soil ingestion and food ingestion: this latter was considered only for residents in non urban receptors of the area by considering the exposure deriving from dietary intake of vegetables grown locally, assumed to constitute 10% of the total consumption.

Results and discussion

The probability functions and their corresponding parameters utilised throughout the evaluation are reported in Table 1. PCDD/Fs stack concentrations in terms of equivalent toxicity (I-TEQ according to NATO weighting scheme) result generally well described by lognormal probability models. The distribution parameters evaluated for the data series derived from full scale measurements on modern and older Italian plants, applied for risk assessment in Scenario 1 and 2 respectively, are reported in Table 1. Modern plants, equipped with AC flue gas treatment and, in some instances, with final SCR units, result in stack emissions with mean values more than two orders of magnitude lower than current regulatory limits (geometric mean of 4.7 $pg_{I\text{-}TEQ}\ m_n^{-3}$), thus confirming the highest potential of these techniques in controlling toxic trace organics emissions to the atmosphere. As expected, stack concentrations from older plants not specifically designed for PCDD/F removal from flue gas release emissions significantly exceeding current regulations, with a geometric mean value around 2 $ng_{I\text{-}TEQ}\ m_n^{-3}$. Vapour fraction V of the emitted PCDD/F in terms of I-TEQ result adequately fitted by a Beta probability distribution, with an arithmetic mean value of 0.76 as a whole, indicating a predominant partition of the emission in the gas rather than on particulates, regardless of the flue gas cleaning device. The resulting gas phase PCDD/F stack concentrations are still well described by lognormal distributions, with geometric means and geometric standard deviations of 3.2 $pg_{I\text{-}TEQ}$ m_n^{-3} and 2.57 for modern plants and 1.34 $ng_{I\text{-}TEQ}$ m_n^{-3} and 2.83 for older installations.

Dry deposition fluxes evaluated at all the receptor points of the area for both Scenarios through model simulation were also found to be well described by a lognormal probability distribution model. All the single receptor point values arising form the different particulate size distributions result further in rather smooth differences, with variation coefficients CV included within the relatively restricted range between 0.9 and 1.3 and mean values of the same statistical parameter equal to 1 for modern plants (Scenario 1) and 1.03 for older installations (Scenario 2). Consequently, the variability of dry deposition was described with a lognormal distribution, with mean equal to the arithmetic mean of the corresponding series of seven values and CV equal to the mean CV value on the area. Regarding washout coefficients for wet deposition calculations, vapour phase W_V results well fitted by a Beta probability distribution, while particle bound W_P is adequately described through Weibull probability model (Table 1). Based on hourly rainfall depth data in Milan, a logistic probability distribution was found for the time-fraction of the year during which rain events take place, whilst an extreme value distribution was derived for the annual mean rainfall depth H, still based on local meteorological data.

For soil concentration evaluations, probability distributions drawn from literature were used for the soil mixing depth in urbanised (z_U) and non-urbanised soil (z_{NU}): a uniform probability distribution and a lognormal distribution were respectively adopted for z_U and z_{NU}. For the soil bulk density BD a lognormal distribution was considered regardless to the nature of soil. Even though a probability distribution for K_S is available from literature (lognormal distribution with M_g = 0.07 yr^{-1} and S_g = 1.09) a value of 0.06 yr^{-1} was used, since this parameter has a negligible role for soil concentration variability, as already explained.

Table 1 - Probability functions and corresponding parameters

Variable	Distribution	Parameters value
PCDD/F stack concentration ($ng_{I\text{-}TEQ}$ m_n^{-3})	Lognormal	Scenario 1: M_g= 0.0047, S_g = 2.15 Scenario 2: M_g= 1.95, S_g = 2.37
PCDD/F vapour fraction	Beta	α = 1.24, β = 0.36, Sc = 100
PCDD/F dry deposition flux ($g_{I\text{-}TEQ}$ m^{-2} yr^{-1})	Lognormal	See text
Washout coefficient vapour phase	Beta	α = 0.64, β = 6.36, Sc = 266426
Washout coefficient solid phase	Weibull	L = 17660, Sc = 14096, Sh = 1.19
Annual mean rainfall (mm yr^{-1})	Extreme value	Mean = 861, Sc = 210
Annual fraction of rain periods	Logistic	Mean = 0.0606, Sc = 0.0119
Mixing depth in urbanised soil (cm)	Uniform	End points 1 - 5
Mixing depth in non-urbanised soil (cm)	Lognormal	M_g = 20, S_g = 1.28, end points 15- 25
Soil bulk density (g cm^{-3})	Lognormal	M_g = 1.4, S_g = 1.1, end points 0.93-1.84

M_g = geometric mean; S_g = geometric standard deviation; L = location; Sh = Shape; Sc = Scale

The calculation of direct and indirect exposure resulting from the distribution functions of the main parameters depending from emissions variability was performed following a conventional deterministic approach, utilising standard models applied in MSW incineration risk assessment and with fixed points values of the required parameters normally used and reported elsewhere[3].

The resulting distributions of maximum individual risk for the area, in terms of the excess probability of cancer development following the exposure to stack PCDD/F emissions, are reported in Figure 1 and 2, for Scenario 1 and 2 respectively, with the most significant statistical parameters of the distributions summarised in Table 2. For present plant configuration (Scenario 1), the mean value obtained is $6.7 \cdot 10^{-10}$, with an estimated maximum of $1.1 \cdot 10^{-8}$: even the most extreme percentiles of the calculated risk appear thus largely insignificant with respect to the reference value of 10^{-6} most commonly considered as acceptable in actual risk regulatory practice. For Scenario 2, representative of the exposure situation existing before the installation of the new plant, higher values are obtained, in accordance with measured stack PCDD/F concentrations: mean individual maximum risk results $1.1 \cdot 10^{-7}$, with maximum value fairly in excess of reference limit ($2.1 \cdot 10^{-6}$). By comparing the Scenarios evaluated in terms of individual risk, the beneficial of the optimum control of trace organics emissions obtained with the new plant is thus further confirmed: even with a significant three-fold increase in burning capacity, expected risks result almost three orders of magnitude lower with respect to the former plant configuration.

Figure 1: Total maximum individual risk for Scenario 1

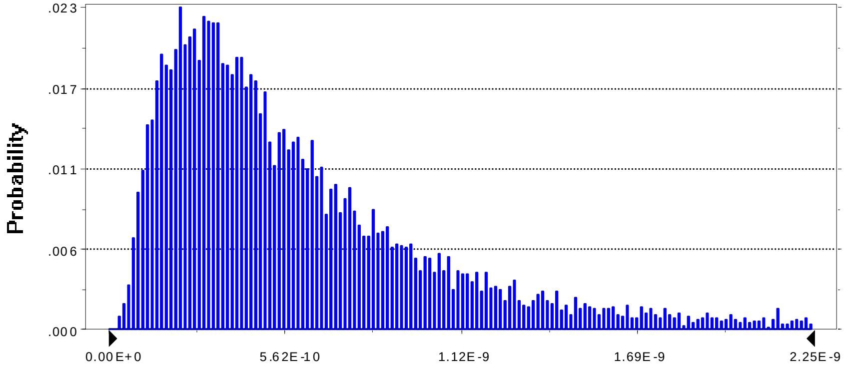

Figure 2: Total maximum individual risk for Scenario 2

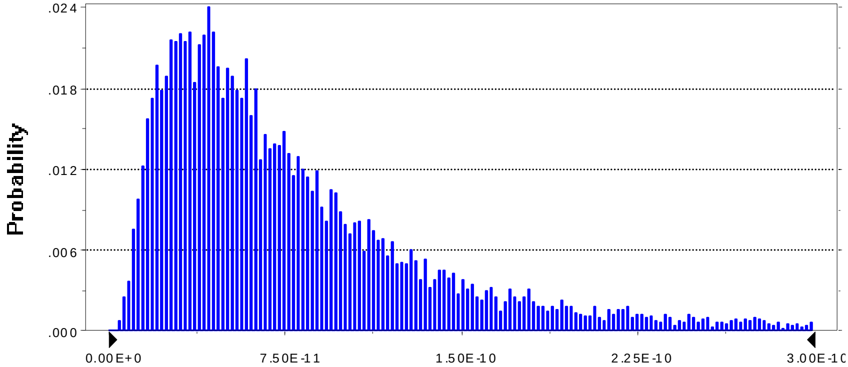

Table 2 - Distribution of maximum individual risk for the area

Scenario	Mean	Standard deviation	Minimum	Maximum	Percentile		
					10^{th}	50^{th}	90^{th}
Scenario 1	$6.7 \cdot 10^{-10}$	$6.1 \cdot 10^{-10}$	$2.9 \cdot 10^{-11}$	$1.1 \cdot 10^{-8}$	$1.8 \cdot 10^{-10}$	$4.8 \cdot 10^{-10}$	$1.3 \cdot 10^{-9}$
Scenario 2	$1.1 \cdot 10^{-7}$	$1.2 \cdot 10^{-7}$	$3.1 \cdot 10^{-9}$	$2.1 \cdot 10^{-6}$	$2.5 \cdot 10^{-8}$	$7.6 \cdot 10^{-8}$	$2.3 \cdot 10^{-7}$

Within the probabilistic approach, the significance of the variability of every emission parameter considered on the resulting variability of the final predicted risk values might be derived, as already mention, through sensitivity analysis. For both evaluated Scenarios the figures obtained in terms of the total variance of risk explained by input parameters indicate an almost exclusive contribution of PCCD/F stack concentration (over 99%). This is mainly a consequence of the large predominance, on total exposure, of direct inhalation and vegetables dietary intake pathways, accounting for roughly 99% of the final risk values predicted (Table 3) and regulated almost exclusively by atmospheric PCDD/F concentrations. Exposures from soil ingestion and dermal absorption are substantially negligible, thus leading to a contribution without any practical significance of particle deposition and related emissions.

Table 3 – Apportionment of maximum individual risk for the area (percentage contribution)

Pathway	Scenario 1	Scenario 2
Inhalation	$1.5 \cdot 10^{-10}$ (22.8%)	$2.6 \cdot 10^{-8}$ (22.7%)
Dermal absorption	$2.1 \cdot 10^{-12}$ (0.2%)	$3.7 \cdot 10^{-10}$ (0.3%)
Soil ingestion	$1.2 \cdot 10^{-12}$ (0.3%)	$2.1 \cdot 10^{-10}$ (0.2%)
Diet	$5.1 \cdot 10^{-10}$ (76.7%)	$8.7 \cdot 10^{-8}$ (76.8%)
Total	$6.7 \cdot 10^{-10}$ (100%)	$1.1 \cdot 10^{-7}$ (100%)

Individual risk distributions evaluated with the probabilistic approach were finally compared with the results obtained through conventional deterministic methods. The evaluation was performed, following the usual criteria adopted for MSWI plants, through the adoption of single conservative data point values of all the input parameters considered as distributed variables in the probabilistic analysis. In particular, PCDD/F stack concentrations were assumed equal to the regulatory limit of 0.1 $ng_{I\text{-}TEQ}$ m^{-3} both in gas than in particle phase and with the particulate size distribution resulting in the highest deposition. Further conservative hypothesis were also adopted for deposition phenomena, by considering the simultaneous occurrence of dry and wet processes and the more restrictive values available for W_V and W_P. With respect to the probabilistic approach, individual risks estimated for the area are essentially comparable with the maximum values of the distributions, and result more than two orders of magnitude higher than mean values for Scenario 1 ($1.1 \cdot 10^{-8}$) and about one order of magnitude higher for Scenario 2 ($3.3 \cdot 10^{-6}$). The largely conservative assumptions, adopted in conventional deterministic approaches for compensating, at least partially, the lack of knowledge about uncertainty and variability, lead thus to very high estimates of risk, corresponding to situations characterised by extreme and very low probability of occurrence.

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