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Soil deposition estimates of PCDD/F: comparison between the results obtained with different atmospheric dispersion models (ISC3ST, AERMOD, ADMS).

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Introduction

The environmental impact assessment (Directive 2011/92/EU) for some installations (such as waste incinerators, smelters and power stations) requires to estimate the dioxins and furans (PCDD/Fs) released in the atmosphere and their environmental fate, by the application of dispersion models. In order to evaluate human exposure, it is important to estimate the deposition of these pollutants on the soil. Below we show the results obtained for PCDD/Fs deposition estimates on the ground with some applications of the ISC3ST, AERMOD and ADMS models. In the absence of reference values in European and Italian legislation, to evaluate the environmental significance of the obtained values we consider the relevant level to be 14 pg TEQ/m² as daily average (TEQ: equivalent toxicity), evaluated as long term estimate (Van Lieshout et al. 2001). In modeling applications we try to adopt some of the methodological guidelines in "Human Health Risk Assessment Protocol for Hazardous Waste Combustion Facilities" (HHRAP, US-EPA 2005), actually specific to ISC3ST.

1 - Syntesis of HHRAP's Methodology

The PCDD/Fs is a mixture of up to 17 toxic different congeners, with distinct physical-chemical characteristics. Moreover, the congeners emitted may occur both in particulate and vapour forms. The main aspect in HHRAP methodology is the partition of PCDD/Fs emissions into the vapour and particle phase: the latter is in turn subdivided in mass particulate (PM, weighted as mass) and surface's particulate, named "particle-bound" (PMB, weighted as surface area), that is the portion of the vapour condensed onto the surface of particles. The subdivision is performed taking into account the value of the vapour fraction of the various compounds. The on-soil deposition estimate requires the definition of a dimensional distribution of the particulate in the emission. For PCDD/Fs vapour-phase pollutants, HHRAP suggests to use a fixed deposition velocity of 0.005 m/s for dry deposition (that is attributed to 2,3,7,8-tetrachlorodibenzo(p)dioxin), while for wet deposition it is recommended to use a scavenging coefficient associated to very fine particles.

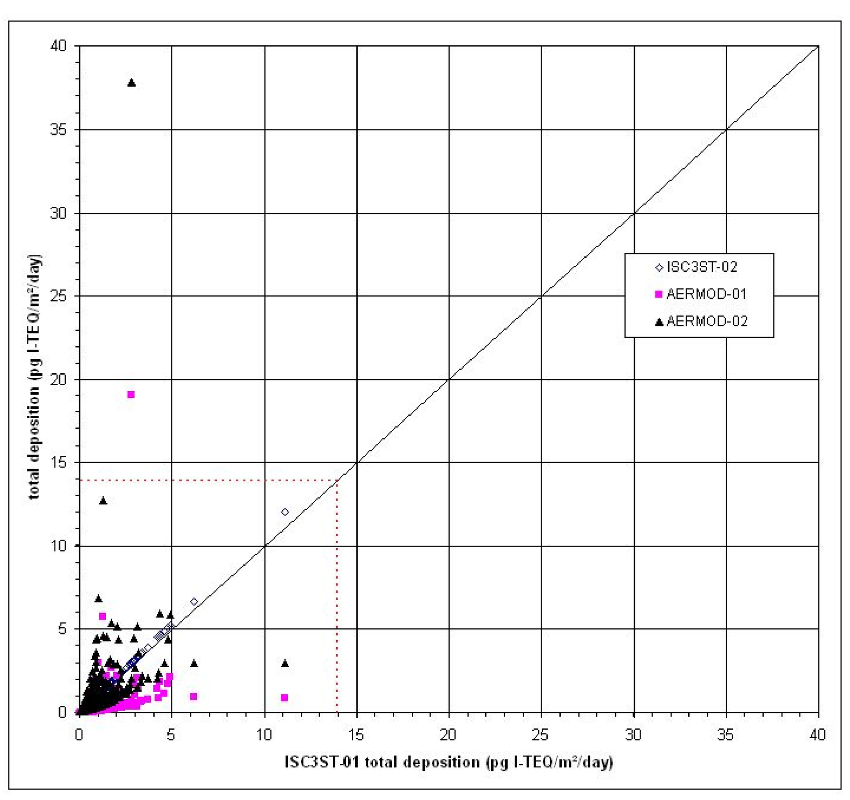

Figure 2: scatterplots of total deposition results obtained with AERMOD and ISC3ST simulations. The dashed line indicates the reference value adopted (14 pg I-TEQ/m²/day).

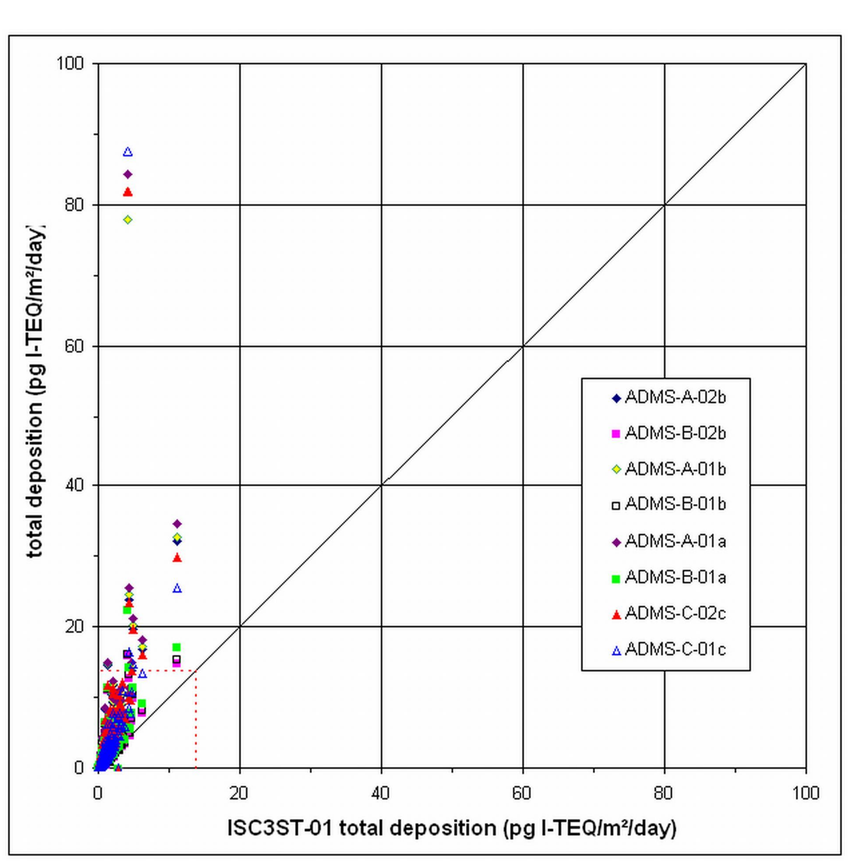

Figure 3: scatterplots of total deposition results obtained with ADMS simulations respect those of ISC3ST-01. The dashed line indicates the reference value adopted (14 pg I-TEQ/m²/day).

Table 1: List of the simulations

simulation	Vapour-phase characteristics		Particle-phase characteristics	
	Dry deposition	Wet deposition	Dry deposition	Wet deposition
ISC3ST-01 (reference)	V_d=0.005 m/s	Λ=sP, φ=0.1 μm, s=1.6E-4	Classes of PM $φ_1$=1.57 μm, $φ_2$=6.92 μm $φ_3$=21.54 μm	Λ=sP, $φ_1$=1.57 μm, s=7E-5 $φ_2$=6.92 μm, s=4.8E-4 $φ_3$=21.54 μm, s=6.7E-4
ISC3ST-02	V_d=0.005 m/s	Λ=sP, φ=0.1 μm, s=1.6E-4	Classes of PM $φ_1$=0.1 μm $φ_2$=6.92 μm	Λ=sP, $φ_1$=0.1 μm, s=1.6E-4 $φ_2$=6.92 μm, s=4.8E-4
AERMOD-01	Da=0.040 (cm²/s), Dw=7.3E-6 (cm²/s) rcl=6.22 (s/m), H=1.09 (Pa m³/mol)		Classes of PM (Method1) $φ_1$=1.57 μm, $φ_2$=6.92 μm $φ_3$=21.54 μm	
AERMOD-02	Da=0.040 (cm²/s), Dw=7.3E-6 (cm²/s) rcl=6.22 (s/m), H=1.09 (Pa m³/mol)		Method2 ($φ_1$=0.1 μm)	
ADMS-A-01a	V_d=0.005 m/s	Λ=aP^b, with a=0.0018, b=0.96 (about equivalent to ISC3ST)	Classes of PM $φ_1$=1.57 μm, $φ_2$=6.92 μm $φ_3$=21.54 μm	Λ=aP^b, with a=0.00012, b=0.85 for $φ_1$ a=0.0005, b=0.99 for $φ_2$ a=0.0008, b=0.95 for $φ_3$ (about equivalent to ISC3ST)
ADMS-A-01b	V_d=0.005 m/s	Λ=aP^b, with a=0.0018, b=0.96 (about equivalent to ISC3ST)	Classes of PM $φ_1$=1.57 μm, $φ_2$=6.92 μm $φ_3$=21.54 μm	Λ=aP^b, with a=0.001, b=0.64 (ADMS default values)
ADMS-A-02b	V_d=0.005 m/s	Λ=aP^b, with a=0.0018, b=0.96 (about equivalent to ISC3ST)	Classes of PM $φ_1$=0.1 μm $φ_2$=6.92 μm	Λ=aP^b, with a=0.001, b=0.64 (ADMS default values)
ADMS-B-01a	Non reactive gas (V_d≈0.001 m/s)	Λ=aP^b, with a=0.001, b=0.64 (ADMS default values)	Classes of PM $φ_1$=1.57 μm, $φ_2$=6.92 μm $φ_3$=21.54 μm	Λ=aP^b, with a=0.00012, b=0.85 for $φ_1$ a=0.0005, b=0.99 for $φ_2$ a=0.0008, b=0.95 for $φ_3$ (about equivalent to ISC3ST)
ADMS-B-01b	Non reactive gas (V_d≈0.001 m/s)	Λ=aP^b, with a=0.001, b=0.64 (default values)	Classes of PM $φ_1$=1.57 μm, $φ_2$=6.92 μm $φ_3$=21.54 μm	Λ=aP^b, with a=0.001, b=0.64 (ADMS default values)
ADMS-B-02b	Non reactive gas (V_d≈0.001 m/s)	Λ=aP^b, with a=0.001, b=0.64 (default values)	Classes of PM $φ_1$=0.1 μm $φ_2$=6.92 μm	Λ=aP^b, with a=0.001, b=0.64 (ADMS default values)
ADMS-C-01c	Hourly values of V_d from AERMOD-01	Λ=aP^b, with a=0.001, b=0.64 (default values)	Hourly values of V_d from AERMOD-01	Λ=aP^b, with a=0.001, b=0.64 (ADMS default values)
ADMS-C-02c	Hourly values of V_d from AERMOD-02	Λ=aP^b, with a=0.001, b=0.64 (default values)	Hourly values of V_d from AERMOD-02	Λ=aP^b, with a=0.001, b=0.64 (ADMS default values)

with: V_d deposition velocity, Λ washout coefficient (s^{-1}), s scavenging coefficient ($h\ s^{-1} mm^{-1}$)
P rate of rain precipitation (mm/h), $φ_i$ average diameter of i-class of particles,
Da air diffusivity, Dw water diffusivity, rcl cuticular resistance for leaves,
H Henry constant law

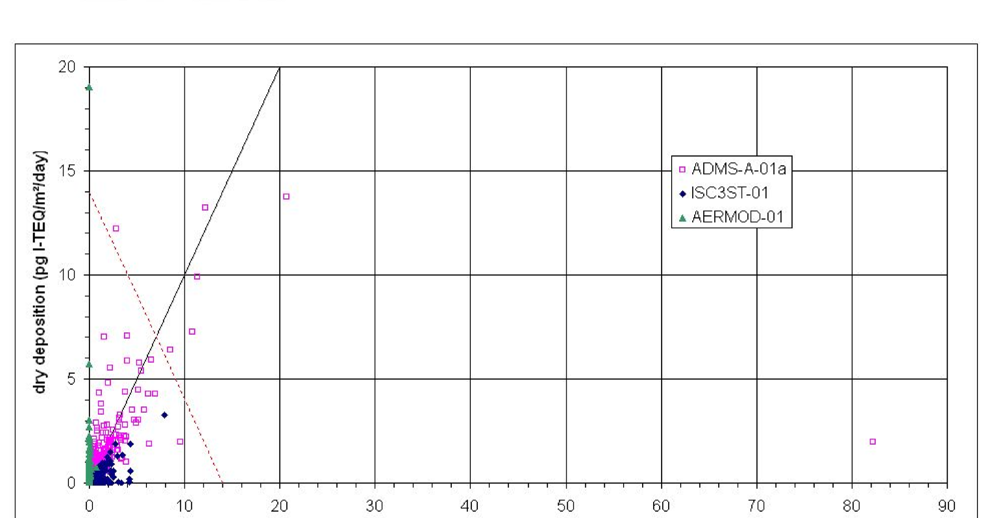

Figure 4: comparison between dry deposition and wet deposition values obtained in the simulation ISC3ST-01, AERMOD-01 and ADMS-A-01a. The dashed line indicates the reference value adopted (14 pg I-TEQ/m²/day).

4 - Comment on the results -A-

The results obtained using the AERMOD and ISC3ST models are compared in Figure 2. It shows that with ISC3ST no significant difference is estimated by changing particulate distribution. The values of AERMOD-02 are higher than those of AERMOD-01: this implies that the finer particulate distribution used results in higher depositions with AERMOD.

The receptors where the highest estimates are obtained do not coincide between AERMOD and ISC3ST: this is due to the different algorithms used to evaluate the effects of building downwash.

Figure 3 shows the results of the total on-soil deposition obtained by using ADMS compared to ISC3ST-01. We observe that ADMS estimates are generally higher than ISC3ST-01 ones. The highest values obtained in the different simulations vary by a factor of about 4 (from 15 pg I-TEQ/m²/day to over 80).

5 - Comment on the results -B-

Observing the apportionment of the total deposition between dry and wet depositions (showed in Figure 4 for ISC3ST-01, AERMOD-01 and ADMS-A-01a simulation), we note that for ISC3ST most of the contribution is due to the wet component, while for AERMOD the wet deposition effect is negligible and only dry deposition seems to be relevant. For ADMS the contribution of the wet component is generally relevant, although its importance depending on the receptor.

2 - The case study

In the industrial area of Ospedaletto, on the south-east of Pisa town, there are some plants (urban solid wastes incinerator, aluminum and copper foundries) that generate PCDD/Fs emissions. These emissions have been the subject of a modeling assessment and of a survey soil monitoring by ARPAT in the year 2016. The most significant emission is associated to a copper foundry and consists of a 20 m height chimney with a flow rate of around 50000 Nm³/h, and a PCDD/Fs concentration in emission, measured during control, as approximately 0.6 ng I-TEQ/Nm³: thus the mass flow is about 8.5 ng I-TEQ/s. This low stack is placed on a very large building which certainly produces an interference effect on wind flow and pollutants dispersion. The area surrounding the plant is substantially flat. The territory is predominantly made up of farmlands cultivated soils and only the northern sector is covered by buildings in the industrial area and of the outskirts of the city.

Figure 1: photographic images showing the copper foundry that represents the case-study. The stack and the extensive adjacent building are well visible.

3 - Simulation features

To obtain the total deposition values, we must estimate the dry deposition and wet deposition of the vapor-phase and particulate phase components, providing the necessary parameter values.

With ISC3ST and ADMS for vapor phase we can directly assign the dry deposition velocity and the scavenging or washout coefficient. For the particulate phase, we assign the distribution of the particles to classes identified by the characteristic diameter (according to HHRAP) and the relative scavenging or washout coefficients.

From measurement at the stack and from fraction vapour data in HHRAP, we found that the vapour-phase component is about 21% of mass in TEQ, while the particulate component results the 79% of mass in TEQ, subdivided in 86% as PMB and the remaining 14% as PM. We calculate the weighted value (in TEQ) for the parameters Da (air diffusivity, 0.040 cm²/s), Dw (water diffusivity, 0.0000073 cm²/s), rcl (cuticular resistance for leaves, 6.22 s/m) and Henry constant law H (1.09 Pa m³/mol) requests by AERMOD model for gas deposition estimation (US-EPA, 2004).

Having no information about particles size distribution, two different distributions have been adopted: the first (hereinafter referred to as 01) derives from the distribution indicated by US-EPA for foundries emissions. In this distribution there are three classes of particles with diameters of 1.57 µm, 6.92 µm and 21.54 µm, with PM (PMB) ratios of 93.4% (97.2%), 5.7% (2.7%) and 0.9% (0.1 %) respectively. The second (referred to as 02) assumes that 90% of particulate matter belongs to the PM2.5 class with an average particle diameter of 0.1 µm; the remaining 10% is coarse particulate matter. The version 00101 of ISC3ST, the version 15181 of AERMOD and the version 5.1 for ADMS have been used. BPIP (ISC3ST) and BPIPPRM (AERMOD) utilities have been used to take in account building downwash calculation; the ADMS building specific option has been used. In Table 1 we summarize the main features used in the simulations.

The on-soil depositions (total, dry and wet) have been estimated on a regular receptors grid wide 8 km x 8 km, centered on the source, with a pitch of 100 m. The meteorological data used for the simulations consists of observations collected at the nearby "Galileo Galilei" airport (LIRP, about 3 km from plant) and at a private weather station located at a distance of about 5 km. The meteorological profile data are supplied by LaMMA (Tuscany Regional Meteorologic Service) and derives from the application of the WRF model. The data used refer to year 2014, when the annual cumulative value of rainfall has been 1207 mm.

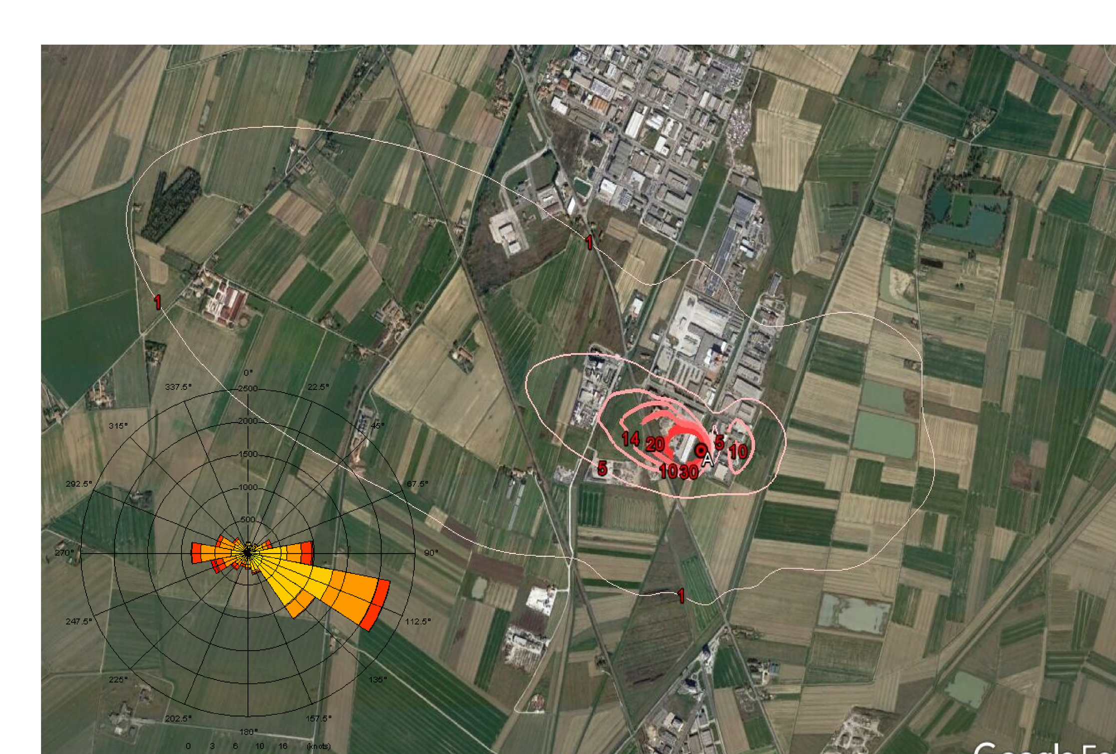

Figure 5: qualitative representation with isolines of the on-soil total deposition estimates (pg I-TEQ/m²/day) obtained by the ADMS-A-01a simulation. In the left corner it is shown the surface winds rose.

References

US-EPA 2005: Human Health Risk Assessment Protocol for Hazardous Waste Combustion Facilities. EPA-530-R-05-006, US-EPA.

US-EPA 2004: User's guide for the AMS/EPA regulatory model – AERMOD. EPA-454/B-03-001 (Addendum 2006, Addendum 2011, Addendum 2015). US-EPA.

Van Lieshout, L, M. Desmedt, E. Roekens, R. De Fré, R.Van Cleuvenbergen and M. Wevers, 2001: Deposition of dioxins in Flanders (Belgium) and a proposition for guide values. *Atmospheric Environment*, **35** Suppl. no. 1, s83-s90.

6 - Syntesis of the results

The previous analysis highlights the variability of PCDD/Fs on-soil deposition estimates due both to the different dispersion models used and the variation of the parameters required by these models for the dry and wet deposition estimates. However, except for some receptors very close to the source, on-soil depositions of PCDD/Fs estimated by all models and in all cases simulated are lower than the reference level adopted (14 pg I-TEQ/m²/day). Each of the simulations carried out can be considered acceptable in the context of an environmental impact assessment process. This allows to conclude also that for sources such the ones examined in the present study, the possible critical situations for the environment are limited to the areas closest to the plant.