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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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Abstract: The on-soil depositions of dioxins and furans estimates using different atmospheric dispersion models (ISC3ST, AERMOD, ADMS) are illustrated. This comparison allows to highlight the variability among the results, due to the large number of parameters needed and to the intrinsic differences between the algorithms used in the models. Nevertheless, for the case-study analyzed, the obtained results are in agreement to delimit the relevant impacts only to the extreme proximity to the source.

Key words: on-soil deposition, PCDD/Fs.

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. These estimates are also necessary for population's health evaluations, such as risk analysis or epidemiological investigations.

According to the World Health Organization, the largest part of human background exposure to PCCD/Fs occurs through the diet, with food from animal origin being the predominant source, while daily intake through inhalation constitutes less than 5% of the total intake from food. PCDD/Fs contamination of food is mainly caused by deposition of emissions on farmland and water bodies, followed by bioaccumulation up terrestrial and acquatic food-chains. Therefore it is very important to estimate the PCDD/Fs on-soils depositions.

Reference levels for evaluating and comparing estimated values of the PCDD/Fs air concentrations and on-soil depositions have not yet been established by the European Union and Italy regulations. Usually literature values or threshold values adopted in other Countries are used as benchmarks in estimates.

In the present study we consider the relevant level to be 14 pg TEQ/m² as daily average (TEQ: equivalent toxicity), evaluated as long term estimate; this value is the one suggested as corresponding to a daily intake of 4 pg TEQ/kg(Bw) day^{-1} (Van Lieshout et al. 2001).

A reliable estimation of the PCDD/Fs air concentrations and on-soil depositions is complicated from a methodological point of view, because the pollutant is a mixture of up to 17 toxic distinct congeners,with distinct physical-chemical characteristics. Moreover, the congeners emitted may occur both in particulate and vapour forms, making more challenging the correct estimate of environmental impacts.

The US-EPA document "Human Health Risk Assessment Protocol for Hazardous Waste Combustion Facilities" (HHRAP, US-EPA 2005) suggests a methodology to be used for this kind of estimates. However these indications are sometimes specific for the ISC3ST model, replaced since 2005 by others included in the "preferred air quality models list" recommended by US-EPA for regulatory applications. So, it is often necessary to interpret and adapt the HHRAP guidelines when using a different model.

ARPAT has applied the HHRAP methodology to estimate the impact of emissions associated to some specific installations on the environment and population's health in Tuscany. The results obtained with different dispersion models show variability due to the input data, the optional choices and the algorithms used in the models.

In the following section, we summarize some relevant aspects in the HHRAP's methodology. Then we describe a case-study, related to a copper foundry in the industrial area of Pisa, and we compare the estimated PCCD/Fs impacts obtained by using ISC3ST, AERMOD and ADMS models, applying some of the HHRAP's suggestions. In particular, we discuss the connection of the depositions results with the choices of the input data.

SYNTESIS OF HHRAP'S METHODOLOGY AND SOME REMARKS

The main aspect 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 mass of congeners with a vapour fraction of less than 5% is attributed to particulate matter PM, while that of congeners with vapour fraction greater than 5% is subdivided into the vapour and PMB components.

Therefore, it is generally necessary to run the atmospheric dispersion model considering a gaseous pollutant and two different pollutants such as PM and PMB particles. The latter two will have different dimensional distributions because the first one follows the actual mass distribution, while the other follows the distribution of the surface area of the particles (for simplicity in HHRAP a spherical shape is assumed).

The on-soil deposition estimate with HHRAP therefore also requires a definition of a dimensional distribution of the particulate in the emission.

This methodology suggests to simulate the dispersion of each congener separately and to obtain the total value by adding the contributions of the individual congeners, with the equivalent toxicity as the weight factor. However, this is not necessary when both the dry deposition velocity and the washout coefficient of the vapour-phase component are fixed. Likewise, it is not necessary for the particulate component since the dry deposition velocity and the scavenging or washout coefficients depend mainly on the physical characteristics of the particles (size, density etc.).

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 (having size 0.1 μm according to ISC3 technical documentation, US-EPA 1995). These simple recipes cannot be implemented in AERMOD (US-EPA, 2004), but can be used by setting the dry deposition velocity and the washout coefficients in ADMS appropriately (CERC, 2012).

THE CASE STUDY

In the industrial area of Ospedaletto, on the south-east of Pisa town, there are some industrial 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 standardized flow rate of around 50000 Nm^3/h, and a high PCDD/Fs concentration in emission, measured during control as approximately 0.6 ng I-TEQ/Nm^3, thus the mass flow is about 8.5 ng I-TEQ/s. This relatively low stack is placed on a very large building which certainly produces an interference effect on wind flow and pollutants dispersion.

In the modeling assessment of these emissions, ARPAT has tried to apply the HHRAP methodology indications, using AERMOD model and ISC3ST model for checking. The differences in the results obtained are the reason of the analysis presented in this study, which has been carried out also by using ADMS model.

The area surrounding the plant is substantially flat, without significant relief for distances of several kilometers. 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.

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.

First, the AERMET pre-processor has been used to produce the needed files for AERMOD, so with these data the input meteorological files for ISC3ST and ADMS have been generated. In this way, the meteorological data are basically the same for the models or in any case come from the same source and can be considered equivalent.

From analytical 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. Instead of performing 17 simulations for the congeners vapour components, we calculate the weighted value (in TEQ) for the parameters Da (air diffusivity, 0.040 cm^2/s), Dw (water diffusivity, 0.0000073 cm^2/s), rcl (cuticular resistance for leaves, 6.22 s/m) and Henry constant law H (1.09 Pa m^3/mol) requests by AERMOD model for gas deposition estimation (US-EPA, 2004).

Finally, 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 upstream of the abatement plant, and on which it has been hypothesized an efficiency of about 99.9% of the control system (US-EPA, 1990). In this distribution there are three classes of particles with HHRAP's equivalent 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. In AERMOD for the first distribution is has been used the "method 1" and for the second the "method 2" option for deposition calculation (US-EPA, 2004). The version 00101 of ISCST, the version 15181 of AERMOD and the version 5.1 for ADMS have been used. The air concentrations and 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 orographic data derive from the SRTM data (CGIAR-CSI). BPIP (ISC3ST) and BPIPPRM (AERMOD) utilities have been used to take in account building downwash calculation; the ADMS building specific option has been used.

AERMOD VERSUS ISC3ST RESULTS

The results obtained using the AERMOD and ISC3ST models are compared in Figure 1. The estimates obtained with ISC3ST following the HHRAP indications for the vapour component and the particulate distribution indicated by "01" (ISC3ST-01) are considered as reference. The AERMOD results (AERMOD-01 and AERMOD-02) have the same vapour component and differ for the distribution of the particulate matter employed.

Figure 1 shows that with ISC3ST no significant difference in total deposition is estimated by changing particulate distribution (ISC3ST-02).

The scatterplots in Figure 1 show that 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. It is possible to observe that the receptors where the highest estimates are obtained do not coincide between AERMOD and ISC3ST: this is essentially due to the different algorithms used by the two models to evaluate the effects of building downwash. In fact, the ISC3ST model used here does not implement the PRIME model as AERMOD does.

Observing the apportionment of the total deposition between dry and wet depositions (not showed here), we note that for ISC3ST most of the contribution is due to the wet component; for AERMOD the wet deposition effect is negligible, so only dry deposition seems to be relevant. This result seems to be in agreement with those reported in test cases (see https://www.epa.gov/scram/air-quality-dispersion-modeling-preferred-and-recommended-models#aermod).

ADMS VERSUS ISC3ST RESULTS

Taking advantage of the wide flexibility of ADMS model, a series of simulations has been carried out trying to reproduce those already done with ISC3ST and AERMOD models, and also introducing some variations.

Figure 2 shows the results of the total on-soil deposition obtained by using ADMS compared to the reference estimates (ISC3ST-01). In this case, the following three different schemas for the vapour-phase have been used: dry deposition velocity of 0.005 m/s as in ISC3ST-01 (case A), non-reactive gas option according to ADMS (case B, approximately a dry deposition velocity of 0.001 m/s), and hourly variable deposition velocity corresponding to values calculated by AERMOD, obtained using the debug option (case C). For the scavenging coefficients in the vapour-phase, in case A we adopt washout coefficients that allow to reproduce the linear behavior of ISC3ST-01 or for which $\Lambda = aP^b \sim sP$ (with Λ washout coefficient, P hourly rain rate in mm/h, s scavenging coefficient used in ISC3ST), ie a=0.0018 and b=0.96. In the B and C case we adopt the default values of ADMS , ie a=0.0001 and b=0.64.

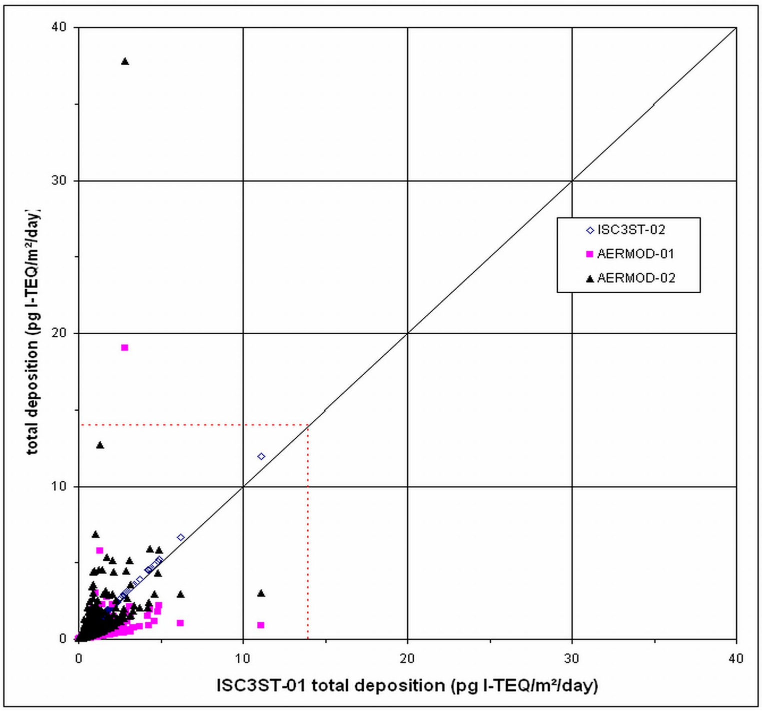

Figure 1. Scatterplots of total deposition results obtained with AERMOD and ISC3ST simulations. The dashed line indicates the reference value adopted (14 pg I-TEQ/m²/day).

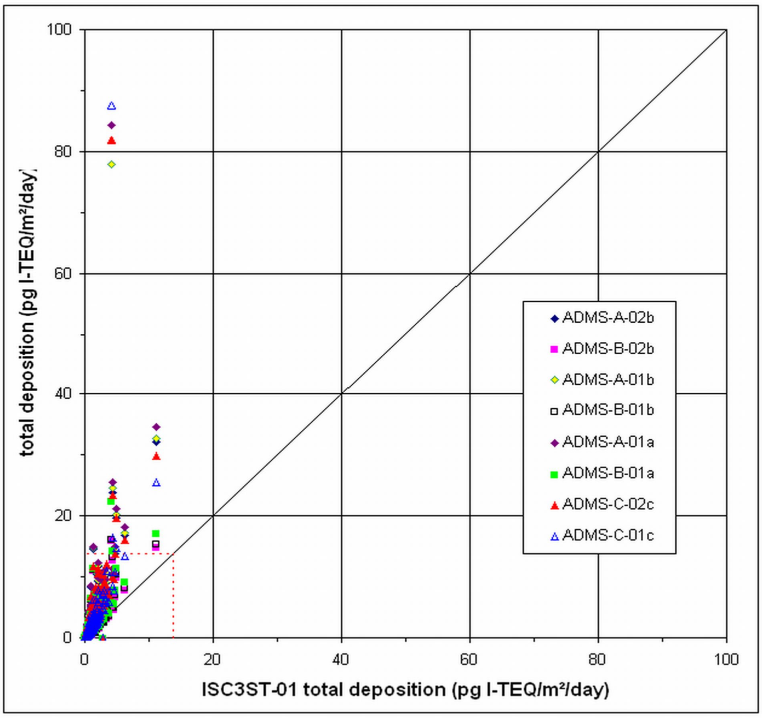

Figure 2. Scatterplots of total deposition results obtained with ADMS simulations. The dashed line indicates the reference value adopted (14 pg I-TEQ/m²/day).

For the particulate matter, the pollutants with the characteristics previously used with ISC3ST and AERMOD (01 and 02) have been reproduced: the case indicated with 01a adopts washout coefficients adapted to be equivalent to those of ISC3ST, while the cases indicated with 01b and 02b adopt the ADMS default values. We also have simulated pollutants as particulate using the dry deposition velocity produced by AERMOD (01c and 02c), but with washout coefficients ADMS default values.
Combining the various choices of the vapour and particulate components together we get the eight cases of total deposition whose the results are shown in the scatterplots of Figure 2. In particular, the case ADMS-A-01a is equivalent to ISC3ST-01. For the dry component ADMS-C-01c and ADMS-C-02c are equivalent to AERMOD-01 and AERMOD-02.
It is possible to observe that ADMS estimates are generally higher than ISC3ST-01 ones.
This is also true when considering the cases in which the ADMS's default values are used (ADMS-B-01b and ADMS-B-02b). For the receptors associated with the highest values, there is a large variability (in some cases, the maximum is about 80 pg I-TEQ/m²/day; for other cases, values are also 4 times lower). The highest values are obtained from the cases in which we use the values of the dry deposition velocity calculated from AERMOD (ADMS-C-01c and ADMS-C-02c) and those with the suggested values in HHRAP for the vapour phase (ADMS-A-01a, ADMS-A-01b, ADMS-A-02b).
The apportionment between dry and wet deposition (not showed here) indicates that the contribution of the wet component is generally relevant, although its importance is different depending on the case considered and the receptors.

CONCLUSIONS

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 estimated by all models and in all cases simulated are lower than the reference level adopted. This allows to conclude that for sources such the ones examined in the present study, the possible critical situations for the environment are limited to areas closest to the plant.

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