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Application of the ISCST3 model for predicting PCDD/F concentrations in agricultural soil in the vicinity of a MSWI plant in China*

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Abstract: Based on the Industrial Source Complex Short-Term Version 3 (ISCST3) model, a simplified modeling approach was developed to predict concentrations of congeners of polychlorinated-*p*-dioxins and dibenzofurans (PCDD/Fs) of agricultural soil, within a radius of 3 km from a municipal solid waste incinerator (MSWI) plant after its 4-year operation in Hangzhou, China. Comparisons were made between the measured and estimated congener-specific concentrations and the international-toxic equivalent (I-TEQ) values of soil samples with respect to distance from the stack. The results indicate that the predictions of soil PCDD/F concentrations and I-TEQ values were generally lower than their observations, and that the higher the degree of underestimation seems, the greater the further downwind one gets. Nevertheless, most of the predictions were in good agreement with the trend of measured ones and were within a factor of ten for samples located within 1 km of the plant. Besides, analysis of contributions of various deposition pathways confirms that in addition to wet particle deposition, the dry gaseous deposition is essential for realistic prediction of PCDD/F depositions to soil, especially for tetra- and penta-chlorinated dioxins.

Key words: Agricultural soil, Dispersion model, ISCST3, Municipal solid waste incinerator (MSWI), Polychlorinated-*p*-dioxins and dibenzofurans (PCDD/Fs)
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INTRODUCTION

Since the first detection of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in the flue gas of municipal solid waste incinerator (MSWI), the emissions of PCDD/Fs have become one of the most controversial issues worldwide (Olie *et al*., 1977). Consequently, comprehensive researches have been conducted to investigate the air and soil PCDD/F levels in the vicinity of the MSWI plant since the end of 20th century (Lorber *et al*., 1998; Ohta *et al*., 2000; Chao *et al*., 2003; Cheng *et al*., 2003; Caserini *et al*., 2004; Chang *et al*., 2004; Oh *et al*., 2006; Schumacher and Domingo, 2006; Coutinho *et al*., 2007).

Due to the characteristics of less time/cost consuming, consistent data integration and quantitative data extrapolation, model simulations have been widely used for the assessment of soil PCDD/F concentrations near the MSWI plant in addition to the conventional environmental monitoring (Yamamoto and Fukushima, 1993; Basham and Whitwell, 1999; Lober *et al*., 2000; Yoshida *et al*., 2001; Meneses *et al*., 2002; 2004; Ogura *et al*., 2003; Choi and Chang, 2005; Lee *et al*., 2007). Most of the simulations were based on the USEPA regulatory dispersion model: Industrial Source Complex Short-Term Version 3 (ISCST3). The ISCST3 model is derived from the Gaussian-plume formula and is generally applicable for near-field (within 10 km) impact assessment of chemically inert pollutant in meteorologically and topographically uncomplex conditions (NIWA, 2004).

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Being the largest developing country in the world, China generates approximately 150 million metric tons of municipal solid waste (MSW) per year (Peng and Gu, 2006). The lack of landfill sites for the wastes has forced local governments (especially for developed regions) to choose the incineration as a substitute option. As a result, more than 20 large scale fluidized bed incinerators (FBIs) (unit capacity between 150 and 400 metric tons of MSW per day) have been commercially operated in China since 1998 (Yan *et al*., 2006). However, the PCDD/F emissions from the MSWI plants have stirred public concerns for their potential adverse effects on the surrounding environment. It was estimated that 3.20~924 g international-toxic equivalent (I-TEQ) of PCDD/Fs was released into the atmosphere by the MSWI plants during the year of 2002 in China (Jun *et al*., 2004).

Until now, however, in China there are limited studies focused on the investigation and modeling of PCDD/F concentrations in environmental media in the neighbourhood of a MSWI plant. In the previous study in September 2006, we initiated an investigation of PCDD/F concentrations of agricultural soil in the vicinity of a MSWI plant in Hangzhou, China (Yan *et al*., 2007). Comparisons of homologue and congener patterns and multivariate analyses of soil and flue gas samples strongly indicate that most of the soil samples were influenced by the MSWI plant. Accordingly, the objectives of this study were to test whether a simplified modeling approach based on the ISCST3 model could serve as a screen means to predict the incremental PCDD/F concentrations of agricultural soil near a MSWI plant in China.

EXPERIMENTAL DESIGN

Study area

The study area is within a satellite town in the northeast of Hangzhou, China, where industrial and residential areas coexist (Fig.1). The MSWI plant investigated in this study is located in an industrial zone in the center of the town, adjacent to two motorways with heavy traffics, in its west and north sides, respectively. It was equipped with three FBIs, began operations of its first two lines in 2002, and has been in full operation with a total daily capacity of 800 t since 2003. In addition, a small-scale hazardous waste incinerator (HWI) plant, about 800 m northward to the MSWI plant, had once been occasionally in operation during 2002 and 2004. However, the capacity and PCDD/F emission data from this HWI plant were not available due to plant confidentiality. Moreover, agricultural activities with a possible use of agrochemicals including sodium pentachlorophenate (PCP-Na) and 1,3,5-trichloro-2-(4-nitrophenoxy) benzene (CNP) were unclear, although suspicious.

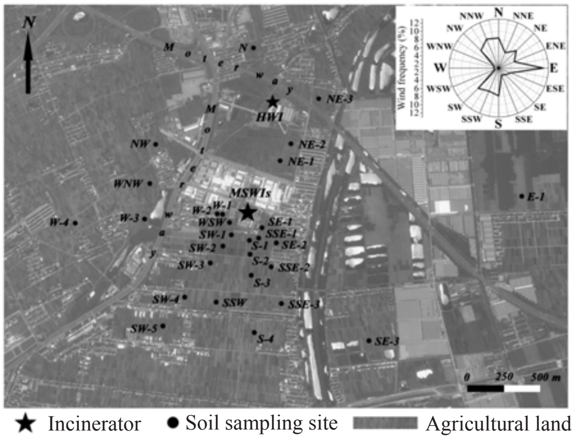

Fig.1 Wind frequency diagram and the distribution of soil samples around the MSWIs

Soil sampling

A total of 30 soil samples were collected from agricultural fields within a radius of 3 km from the MSWI plant in September, 2006. These sampling sites were distributed mainly in the historically prevailing downwind directions (W, NE, SSE, SE, S, SW) (Fig.1) and the exact sampling points were determined and recorded within ~10 m in accuracy by a handheld GPS device (Meridian Color, Thales Navigation, USA). Soil samples were collected by mixing five different aliquots within a 25 m^2 area. The soil was subsequently dried in a ventilated room until constant weight. Then, they were ground and filtered through a 2-mm sieve. About 500 g soils of each sample were finally homogenized through a 60-mesh sieve, and refrigerated until analysis.

PCDD/F analysis

The Method of USEPA 1613 (USEPA, 1994) was followed to analyze PCDD/F in soil samples. Briefly, 10 g (dry matter) of each sample were extracted by using a fully automated accelerated solvent extraction (ASE) 300 system (Dionex, Sunnyvale, CA, USA), subsequently followed by rotary evaporation, multi-layer silica gel column clean-up procedure, and finally subjected to a high-resolution gas chromatography/high-resolution mass spectrometer (HRGC/HRMS). The 17 toxic 2,3,7,8-substituted PCDD/Fs (referred to as congeners) were quantified based on isotope ratios within ±15% of the theoretical values and signal-to-noise ratios of equal to or greater than 2.5. The recoveries of internal standards as well as the limits of detection (LOD) all fulfilled the requirements set by the Method of USEPA 1613. All the results were expressed on a dry weight basis in this study, and the I-TEQ values were estimated using the NATO/CCMS factors. In the case of values below the detection limit, I-TEQ calculations were carried out by using the half of the LOD.

A detailed description of the studied area, soil sampling as well as the soil PCDD/F analysis method were previously reported (Yan *et al*., 2007).

MODELING PROCEDURE

Model framework

The model framework to predict incremental soil congener concentrations is shown in Fig.2.

Data pertaining to source characteristics, meteorological parameters, terrain receptor network and nominal emission were used as input to the ISCST3 model to estimate the nominal gaseous concentrations and particle depositions of three representative diameter classes (i.e., <2 μm≡1 μm, 2~10 μm≡6.78 μm, >10 μm≡20 μm) (Basham and Whitwell, 1999). The nominal gaseous concentrations were subsequently used as the input to estimate the dry and/or wet gaseous depositions with the combination of the dry gaseous deposition rate and/or the washout ratios and rainfall parameters of local areas. The calculation algorithms of dry/wet gaseous depositions were referred to Yoshida *et al*.(2001) and Ogura *et al*.(2003). On the assumption that the partitioning of particle-phase PCDD/F congeners between different size particles is dependent on the surface area to volume ratio of the particles present (Basham and Whitwell, 1999), the following percentage values for distribution of particle-bound congeners by particle diameter are used: <2 μm=57%, 2~10 μm=39%, >10 μm=4%. The actual four categories of congener-specific depositions (dry gaseous, wet gaseous, dry particle-bound and wet particle-bound) could then be estimated

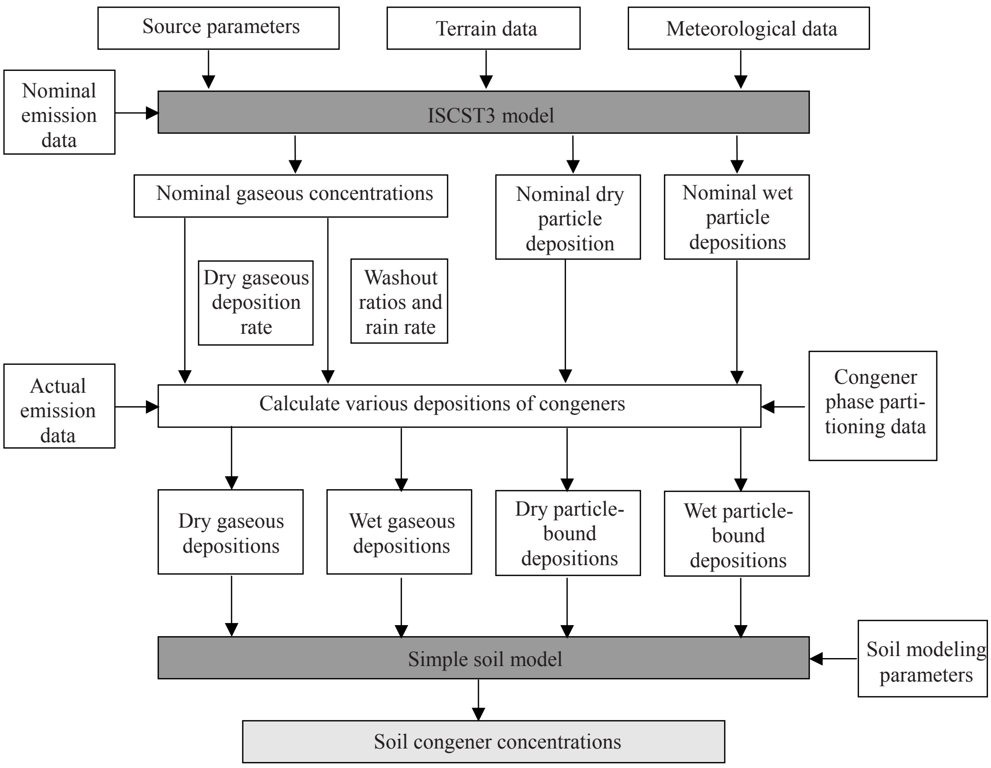

Fig.2 Model framework to predict incremental soil congener concentrations

by incorporating the real emission data as well as the congener phase partitioning data. Finally, the total depositions of the above four categories were subsequently served as the source term for a simple reservoir mixing model to predict the incremental soil congener concentrations (Lorber and Pinsky, 2000).

Source characterization

The stack height, diameter, exit gas temperature, and exit gas velocity of the MSWI plant were 60 m, 3.2 m, 125 °C, and 12 m/s, respectively. An evaluation of PCDD/F emission data of three FBIs obtained during their fully operations in 2003 showed that although the emission levels changed significantly, the congener profiles of flue gas were quite similar (Yan *et al*., 2007). The maximum PCDD/F emission level (0.1961 ng I-TEQ/N m^3) detected in the FBI-B was used in this model exercise as the representative of conservative scenario.

Meteorological data

Sequential hourly surface meteorological data were obtained from the Meteorological Bureau of Hangzhou from year 2002~2005 inclusive. The raw surface meteorological data was then transformed into the SCRAM (MET144) format via FORTRAN 77 and was subsequently modified for use in ISCST3 using the USEPA meteorological pre-processor program, PCRAMMET (USEPA, 1995). Parameterization of fixed quantities was as follows: anemometer height, 10 m; surface roughness length, 1 m at Meteorological Bureau of Hangzhou and 0.073 m at the MSWI plant; noontime albedo, 0.28; Bowen ratio, 0.75; anthropogenic heat flux, 0; and fraction of net radiation absorbed at the ground, 0.15.

Topographical data

Topographic data was obtained from the website, http://www.webgis.com/, supplied in the GTOPO30 DEM Format and was processed by the USEPA terrain processor AERMAP to obtain the information of a height scale and a base elevation for each receptor. Terrain elevation (between 9 m and 13 m) and land use within a 3 km-radius of the MSWI plant indicated that the ISCST3 "flat" and "rural" modeling algorithms, respectively, were suitable for our study.

Model parameters

The ISCST3 modeling procedure used the regulatory default options in this study. Particle parameters for deposition algorithms (including diameters, density, and scavenging coefficients), homologue-specific V/P partitioning coefficients, as well as the dissipation rate of PCDD/Fs were all referred to Lorber and Pinsky (2000). Dry gaseous deposition velocity (2 m/h) and gaseous congener-specific washout ratios were referred to Yoshida *et al*.(2001) and Ogura *et al*.(2003), respectively. Parameters for the simple reservoir mixing model were as follows: a time of operation, *t*, of 4 years was used, corresponding to the time of operation of the MSWI plant; the soil mixing mass, *M*, equaled 320000 g/m^2, which assumes a mid-range of soil bulk density of 1.6 g/cm^3 and the soil sampling depth of 20 cm.

Model assumptions

To predict the incremental PCDD/F concentrations of agricultural soil caused by the emissions of the FBIs, the following assumptions were made prior to the modeling: (1) In order to simplify the calculation of total mass of gas-phase congeners in air above the receptor coordinate (x, y), the mixing height was assumed to be 700 m (the average value of the local area), and was divided into 15 layers (with an interval of 50 m); besides, the annual maximum concentrations were used for conservative purposes; (2) As discussed in (Yan *et al*., 2007), the unusually high concentrations found in Samples WSW, NW, and N were speculated to have mainly resulted from uncontrolled dispersion of fly ash and the emissions of the HWI plant/motor vehicles, respectively, whereas Sample E was suspected to be polluted with the impurities of CNP. Therefore, 17 congener concentrations as well as I-TEQ values of the above 4 soil samples were not to be considered in the context of model testing; (3) As the soil data before the operation of the MSWI plant was not available, Sample SE-3 with the least I-TEQ value (0.39 pg/g) was served as the background level and subsequently subtracted from each of the 25 observed soil measurements to estimate the incremental congener concentrations; when this subtraction resulted in a concentration less than 0, the concentration was set to 0; (4) Soil samples were clustered into 6 groups according to their distances from the stack: 0~250, 250~500, 500~750, 750~1000, 1000~1500, and 1500~3000 m, respectively.

RESULTS AND DISCUSSIONS

The measured and estimated congener-specific concentrations as well as I-TEQ values of clustered soil samples are shown in Fig.3, with error bars indicating the standard deviations.

It appears that the predicted values demonstrate similar profiles to those of the measured ones, indicating agricultural soil in the vicinity of the MSWI plant was strongly influenced by the FBIs. Of both predicted and measured values, OCDD exhibits the highest level, followed by the 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF and OCDF, whereas the 2,3,7,8-TCDD, 1,2,3,7,8,9-HxCDF and 1,2,3,4,7,8,9-HpCDF indicate relatively low concentrations.

As shown in Fig.3, the mean values of estimated congener-specific concentrations in soil at each group were generally lower than those of the observations, and the higher the degree of underestimation seems, the greater the further downwind one gets. The underestimations of predictions for most high-chlorinated congeners might be partly attributed to the application of some pesticides such as PCP-Na. The abundant congeners in impurities of Chinese commercial PCP-Na consist of OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,4,6,7,8-HpCDF and 1,2,3,4,7,8-HxCDF (Bao *et al*., 1995). Therefore, the scattered usage of the pesticides might result in the unusual elevation of high-chlorinated congeners in some clustered groups. For instance, the mean levels of OCDD, OCDF, 1,2,3,4,6,7,8-HpCDD of Group V (Fig.3e) are even higher than those of Group

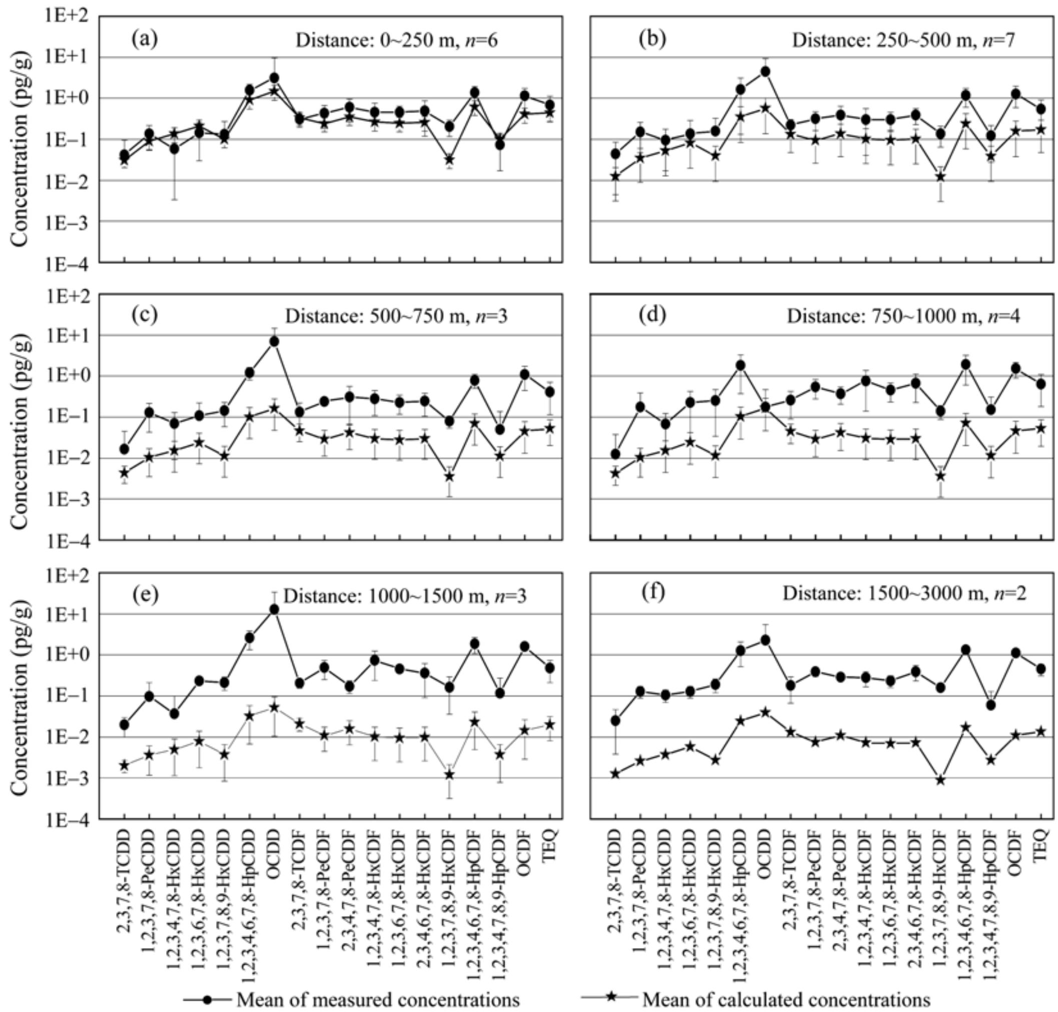

Fig.3 (a)~(f) showing comparison between measured and estimated congener-specific concentrations as well as I-TEQ values of clustered soil samples (a, b, c, d, e, and f), with error bars indicating the standard deviations

I (Fig.3a), which is believed to be mostly affected by the emissions of the FBIs. Moreover, historical emissions of the HWI and motor vehicles might also contribute to the discrepancies between the predicted and measured congener concentrations of soil. Besides, the background level used in our model exercise may not be ideal since the congener-specific concentrations of each soil sampling site could not be identical prior to the operation of the MSWI plant.

Nevertheless, most of the estimated congener-specific concentrations are in good agreement with measured ones within a factor of 2, 5, 10, and 20 for soil samples obtained 0~200, 250~500, 500~750 and 750~1000 m from the stack, respectively. As for I-TEQ value, the corresponding ratios of measured to predicted ones are within a factor of 2, 4, 8, and 10, respectively.

Fig.4 shows the estimated contributions of 4 deposition categories to the total atmospheric depositions of 17 PCDD/F congeners with respect to the distances from the plant. As indicated by Fig.4, dry gaseous deposition contributes considerably to the transfer of tetra-chlorinated (16.8%~51.9%) and penta-chlorinated (3.0%~16.7%) congeners from atmosphere to soil. Besides, the contribution of dry gaseous depositions increases with the increasing distance from the stack and reaches the maximum value within 1000~1500 m from the stack (Fig.4e). Wet gaseous deposition also plays an important role in the transfer of tetra- and penta-chlorinated congeners to soil, but to a lesser extent. By contrast, wet particle depositions contribute markedly to the transfer of hepta- and octa-chlorinated congeners to soil. Although the contribution of wet particle depositions decreases with the increasing distance from the stack, wet particle deposition contributes more than 97.6% and 96.2% to the total deposition of hepta- and octa-chlorinated congeners, respectively. Dry particle deposition seems to play a negligible role in the transfer of congeners from the atmosphere to soil, but

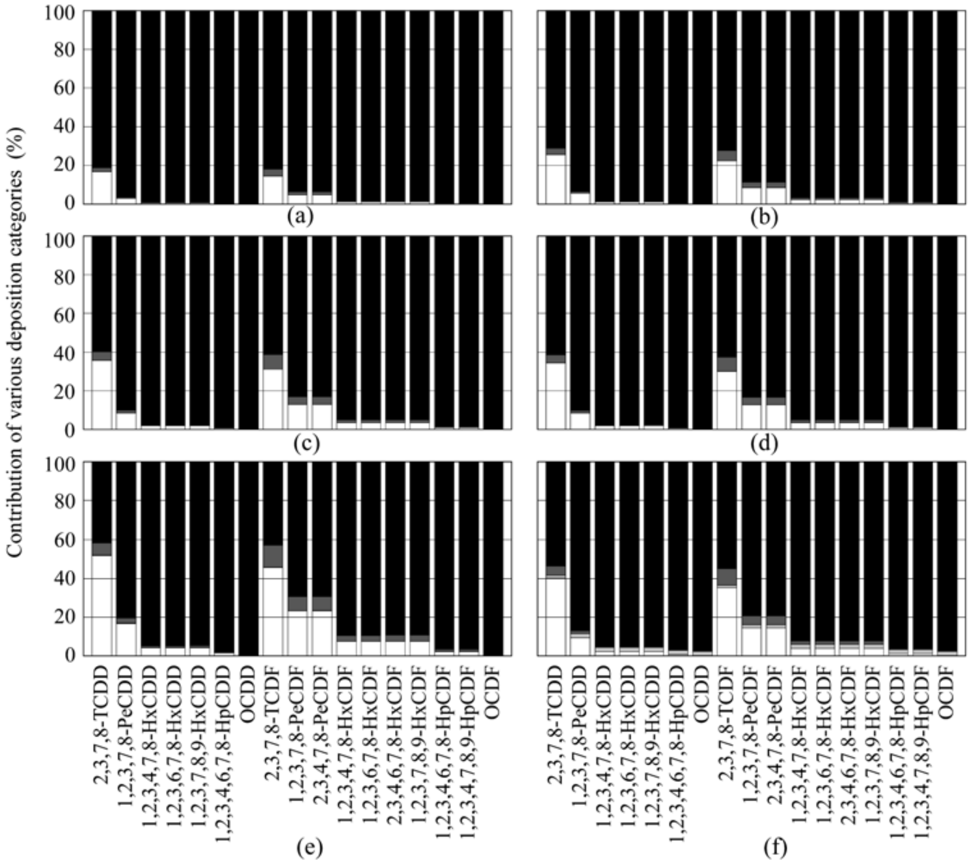

Fig.4 Estimated contributions of dry gaseous, dry particle-bound, wet gaseous and wet particle-bound to the total atmospheric depositions of congeners with respect to distance from the stack
(a) 0~250 m; (b) 250~500 m; (c) 500~750 m; (d) 750~1000 m; (e) 1000~1500 m; (f) 1500~3000 m

its contribution enhances with the increasing distance from the stack and accounts for 1.2%~2.3% for soils located 1500~3000 m from the plant (Fig.4f). A few previous studies (Yoshida *et al*., 2001; Ogura *et al*., 2003) also revealed that the dry gaseous deposition and wet particle deposition were an important pathway for lower chlorinated and higher chlorinated PCDD/Fs, respectively, indicating that these two pathways are essential for predicting PCDD/F depositions to soil.

CONCLUSION

Measured and estimated congener-specific concentrations as well as I-TEQ values of agricultural soil samples with increasing distance from the stack are compared. Most of the estimated concentrations are in good agreement with the trend of measured ones and are within a factor of 10 for soils located within 1 km from the stack. Analysis of contributions of various deposition pathways confirms that in addition to wet particle deposition, the dry gaseous deposition is essential for realistic prediction of PCDD/F depositions to soil, especially for tetra- and penta-chlorinated dioxins.

Our approach based on the ISCST3 model could be used as a screen means for predicting incremental congener-specific and I-TEQ values of soil caused by the operation of combustion sources such as the MSWI plant. Further studies may, however, be needed to validate the assumptions and simplifications made in the modeling exercise.

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