Assessment of the PCDD/Fs Emissions From Coal Fired Residential Heating Appliances By Air Dispersion Modelling

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Introduction

Coal fired stoves for residential heating could generate significant PCDD/Fs emissions to the air. This is now reflected also in the new version of the Standardised Toolkit for Identification and Quantification of Dioxin and Furan Releases which in addition to given emission factor of 3 µg TEQ/ t provides also the very high value of 400 µg TEQ/ t for high chlorine coal combusted in the stoves.[1] With such a high emission factor residential heating could be the dominant source of PCDD/Fs where coal with high chlorine content is used in stoves. High levels of the PCDD/Fs in December 2002 were measured by Christoph et al[2] in air particulate matter in centre of Krakow, Poland. These levels were attributed to the residential heating by congener profile comparison. This work presents dispersion modelling of residential heating emissions to assess whether high or low emission factors give better match to the measured PCDD/F ambient air levels.

Materials and methods

Ambient air particulate samples, taken with High Volume PM 10 samplers, were obtained at two permanent air monitoring sites in Krakow (Figure 1). The site Aleje is situated in the centre of the city, where the density of coal-heated apartments is still substantial. The distance from Nowa Huta industrial complex to Aleje station is about 8 km. The Nowa Huta air monitoring station is located in the suburb of Krakow, less than one km from the Nowa Huta industrial complex. Buildings in the vicinity of Nowa Huta are connected to the district heating network, and coal fired residential heating appliances are used there very rarely.

The PM10 was collected on the filters for 24 hours for each sample. The samples were taken on 10th, 20th and 25th of December 2002. The average daily ambient temperature on these days was: –12, -7 and -15 °C. The PCDD/Fs analysis of the PM10 samples was performed as described by Christoph et al.[2] The PCDD/Fs emission inventory for the residential heating was prepared on the basis of the emission inventory for regulated pollutants that was compiled with 1x 1 km^2 resolution for Krakow area of 30 x 20 km^2. In the 1998/99 heating season, for which it was prepared, the quantity of combusted hard coal was estimated to be 80,000 t for stoves and 10,000 t for small residential boilers. The daily coal consumption was determined taking into account the average daily temperature, which influences the heating demand. Assuming that half of the combusted coal was of high chlorine content the emission factor for PCDD/Fs was averaged to 200 µg TEQ/ t for coal combusted in the residential stoves.[1] For small residential boilers the emission factor was assumed to be 10 times lower than the one for stoves. This assumption is based on Moche et al., who found very high emissions from Polish coal fired stoves and more than ten times lower emissions from small residential boilers, fuelled with the same coal.[3]

The sinter plant and the coke production are deemed to be the most important sources of PCDD/Fs emissions in the Nowa Huta industrial complex. Yearly emissions of PCDD/Fs from the sinter plant stack were estimated to be 40 g TEQ. In addition to the stack emissions, sinter plants exhibit fugitive emissions due to hot sieving and crushing as well as to the leakage from the sinter belt.[4] These emissions were assumed to be 8 g TEQ per year. A rough estimation for the fugitive coke plant emission was 4 g TEQ per year, again at rough estimate. All emissions from the Nowa Huta industrial complex were assumed constant throughout the entire year. No data on the measurements of PCDD/Fs emissions were available for the year 2002 and exact activity data are confidential, that is why the rough estimation of PCDD/Fs was done. Other combustion sources were not taken into account in Krakow PCDD/Fs emission inventory due to the lack of activity data. PCDD/Fs emission factors for liquid and gases fuels combustion as well as for coal combustion in boilers in the industry and commercial-institutional sectors are two orders of magnitude lower than the ones selected for coal- fired stoves.[1] For this reason omission of other combustion

sources does not affect significantly the accuracy of the emission inventory.

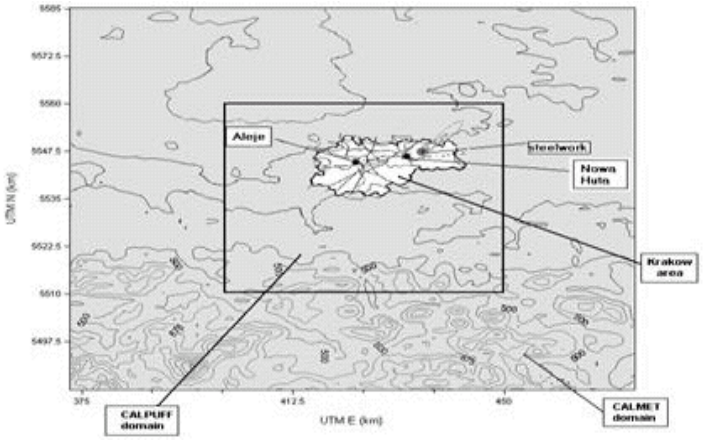
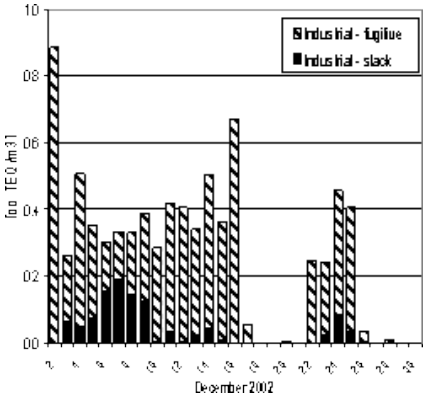


Fig.1.CALMET and CALPUFF modeling domains

Dispersion simulations were performed with the use of CALMET/ CALPUFF meteorological and air quality modelling system developed by Earth Tech, Inc.[5] CALMET/ CALPUFF have been used at the Malopolska Inspectorate for Environment Protection for analysis of emission abatement measures. In this study CALMET domain was 100 x 100 km^2 (Figure 1). The input surface and upper air meteorological data were obtained from the ALADIN model (9 sites on 9 vertical layers). The horizontal resolution of the grid was set to 1x 1 km^2.The domain of CALPUFF dispersion simulations was 50 x 50 km^2 with the grid cell of 1x 1 km^2 centred in the CALMET domain. PCDD/Fs were modelled as primary PM10 emissions from pre-selected steelwork point sources and home heating gridded sources as described earlier. It was assumed that in the plume the majority of the PCDD/Fs emitted in the gas phase are adsorbed on the particulate matter in the low ambient temperature conditions. The advection of PCDD/Fs from outside of the modelling domain was not taken into account. The degradation of the PCDD/Fs in the atmosphere was also neglected since: (i) the reaction timescale is longer than the advection one thus leading to the transport of the species outside the modelling domain prior to chemical transformation, (ii) the considered case relates to the weather conditions in December at 50˚ North latitude.

Results and discussion

Air dispersion modelling shows an influence of the industrial emissions on both monitoring sites (Figure 2, Figure 3). At the Nowa Huta monitoring site the fugitive emissions released close to the ground have the greatest share due to the vicinity of the industrial complex while the height of the stack produces long distance dispersion and a reduced effect on the nearby monitoring site. The effect of the stack is in fact more evident at the Aleje site in the Krakow downtown. On average the simulations performed for December 2002 show that levels of PCDD/Fs are 4 times higher at Nowa Huta monitoring site than at Aleje site due to Nowa Huta industrial complex activities.



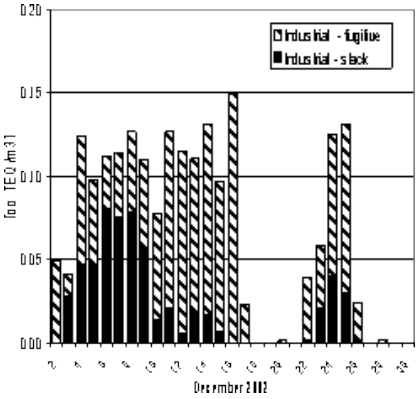


Fig.2. Modelled average daily PCDD/F concentration at Nowa Huta resulting from emissions from Nowa Huta industrial complex

Fig.3. Modelled average daily PCDD/F concentrations at Aleje resulting from emissions from Nowa Huta industrial complex

Not surprisingly the residential heating emissions give always higher levels at the Aleje than at Nowa Huta site. The modelled concentrations at the Aleje site were less influenced by the wind direction, since residential areas, where coal stoves are used, encircle this site.

Table 1. Comparison of the of the measured and modelled PCDD/F levels (pg TEQ/m^3)

	Nowa Huta site				**Aleje site**			
	measured	modelled			measured	modelled		
		residential	industrial fugitive	industrial stack		residential	industrial fugitive	industrial stack
10.12.	1.6	0.07	0.28	0.00	-	2.77	0.06	0.01
20.12.	2.2	0.67	0.00	0.00	1.6	3.46	0.00	0.00
25.12.	0.8	0.09	0.37	0.03	0.71	3.70	0.10	0.03

At the Nowa Huta site significant discrepancies are found between the modelled and measured PCDD/F levels (Table 1). The residential heating dominates for instance on the 20th Dec. This contradicts the results of congener profile analysis performed by Christoph et al.[2] The reasons of these results can be attributed to: (i) the wind field for Dec. 20th didn't reflect the actual wind fields thus leading to the transport of the industrial emissions away and the residential emissions toward the sampling site; (ii) industrial emissions used in the dispersion modelling are roughly estimated, fugitive in particular.

At Aleje site the influence of residential heating clearly dominates (Table 1). This agrees with the conclusions based on the congener profile by the Christoph et al.[2] Modelling results for residential heating emission for Aleje site surpass the measured PCDD/F levels. Two reasons for the model to measurement results discrepancies could be inadequate modelling of the atmospheric conditions and the 4- year time lag of the activity data for residential heating emission inventory, especially taking into consideration that the fuel switch program from coal to gas in Krakow is going on.

Our results indicate that the PCDD/Fs emissions factor for coal combustion in the stoves in the Krakow area is approximately in the order of 100 µg TEQ per ton of coal. The use of an emission factor of 3 µg TEQ/ t of coal would significantly underestimate measured air ambient levels.

This study has outlined the necessity for further investigation. Namely: once more accurate simulation of atmospheric circulation will be obtained and the emission inventory will be updated the sensitivity analysis should be performed in order to more precisely assess the PCDD/Fs emission factors.

Acknowledgement

We would like to thank Malopolska Inspectorate for Environment Protection for providing us with residential heating emission inventory. We also express our thanks to Mr. Jerzy Burzynski for the support at the preparation of meteorological data, Mrs. Helle Skejo for the assistance in the PCDD/Fs analysis and Dr Stefano Galmarini for his comments on the manuscript and on the interpretation of the modelling results.

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