EXAMINATION OF AGRICULTURAL AREAS WHICH HAVE BEEN TREATED WITH PCDD/F CONTAINING SEWAGE SLUDGE, AND EMISSION MEASUREMENTS FROM THE FIRING INSTALLATIONS OF A WASTE WATER TREATMENT PLANT

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1. Introduction

The pollution of sewage sludge of a waste water treatment plant by polychlorinated dibenzodioxins (PCDD) and dibenzofurans (PCDF) in Bavaria, Germany, was measured in 1988 in high concentrations of up to 5780 ng TE/kg d.w. as defined by FHO, Berlin. The pattern of PCDD/Fs detected showed mainly HpCDD and OCDD congeners. The highly contaminated sewage sludge was disposed of in the incineration plant of the Corporation for the Disposal of Toxic Waste in Bavaria. The source of the PCDD/F contamination in the sewage sludge was pentachlorophenol (PCP), present in the waste water of a manufacturing company. It is known that PCP and PCP-sodium contain PCDD/F, mainly Hexa-to OctaCDD/F (1).

The PCDD/F homologue congener patterns in the sewage sludge and in PCP are similar.

Because of the high PCDD/F concentrations in the sewage sludge, all agricultural areas, on which the sludge was spread for fertilization, were examined in 1991/92. Furthermore, measurements of emissions from the firing installations of the waste water treatment plant, in particular from the biogas burner, were conducted. The investigations described in this paper were funded by the Bavarian government.

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2. Results

2.1 Soil investigation

Sampling was performed in 30 districts from 313 agricultural areas, which had been treated with sewage sludge. The analysis of the soil samples was carried out by the Bavarian State Agency of Water Research, Munich. All PCDD/F data of the sludge treated areas were compared with an investigation carried out by the Bavarian Geological Survey (2) on non contaminated rural areas, as shown in Tab. 1:

TE values for rural areas, farm land, sampling depth 30 cm							
	investigation of the Bavarian Geological Survey, 1991	investigation of sludge treated areas, 1991/92					
Minimum	< detection limit	< detection limit					
Maximum	3,7	23					
Arithmetic mea∩	0,41	3,7					
Median (50%)	0,12	1,7					
Percent-90	0,91	13					
<u> </u>	<u> </u>	<u> </u>					
	s, meadow, sampling dep	th 10 cm					
		th 10 cm investigation of sludge treated areas, 1991/92					
	s, meadow, sampling dep investigation of the Bavarian Geological	investigation of sludge					
TE values for rural areas	s, meadow, sampling dep investigation of the Bavarian Geological Survey, 1991	investigation of sludge treated areas, 1991/92					
TE values for rural areas	s, meadow, sampling dep investigation of the Bavarian Geological Survey, 1991 < detection limit	investigation of sludge treated areas, 1991/92 0,2					
TE values for rural areas Minimum Maximum	s, meadow, sampling dep investigation of the Bavarian Geological Survey, 1991 < detection limit 5,6	investigation of sludge treated areas, 1991/92 0,2 196					

Table 1 Statistical analysis of concentrations of PCDD/F in soil samples from the investigation of the sludge treated areas and non contaminated rural areas (Bavarian Geological Survey), expressed as ng TE(FHO)/kg d.w.

The comparison of both investigations shows that the mean and median (50-percent) values of the sludge treated areas are ten times higher than the respective values of the non contaminated areas of the investigation of the Bavarian Geological Survey.

The measured TE values from the investigation of sludge treated areas were compared with the limits proposed by the FHO: 5 ng TE/kg d.w., a limit which restricts the cultivation of certain vegetables and 40 ng TE/kg d.w., a limit which excludes the cultivation of any plants with the exception of those with very low transfer factors. The results of the soil investigation are shown by the following survey:

76 % of all areas (237 areas) < 5 ng TE(NATO/CCMS)/kg d.w.

76 % of all areas (237 areas) < 5 ng TE(NATO/CCMS)/kg d.w. 23 % of all areas (73 areas) 5-40 ng TE(NATO/CCMS)/kg d.w. 1 % of all areas (3 areas) > 40 ng TE(NATO/CCMS)/kg d.w.

Altogether in nearly a third of all districts TE values over 5 ng/kg d.w. were found. The most toxic 2,3,7,8-TCDD was not detected in any sample.

2.2 Investigation of the flue gas from a biogas burner and gas engine of the waste water treatment plant and soil investigation in the surroundings of the emission sources

Based on a maximum emission as toxic equivalents (FHO, Berlin) of 15 pg/Nm 3 from the biogas burner, a maximum emission of 460 μ g TE could be calculated for the period from 1966 till 1990. Even the maximum emission as toxic equivalents of 15 pg/Nm 3 is lower than 100 pg/Nm 3 , which is the EC standard for waste incineration. As shown in Fig. 1, the PCDD/Fs were determined in 3 different sections of the flame from the biogas burner. Fig. 2 shows the congener distribution of PCDD/Fs in different sections of the flame.

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plain	torch	temperature (°C)	oxygen (Vol%)	nitrogen- oxides (NOX) (mg/Nm ³)	carbon- monoxide (mg/Nm ³)	total- carbon (mgC/Nm3)	PCDD/F TE (BGA/Int.) (pg/Nm ³)
1: ca. 8 m		ca. 850	18,5 (14,5-20,7)	4 (0-14)	537 (13-2702)	4 (1-16)	2,3/3,3
2: ca. 5 m	flame	ca. 815	16,3 (0-20,9)	0 (0-10)	372 (0-4153)	24 (3-90)	14,9/13,1
3: ca. 3,5 m		— ca. 550	20,9	1 (0-5)	70 (0-151)	109 (3-625)	1,2/1,4

Figure 1 Operational parameters of combustion

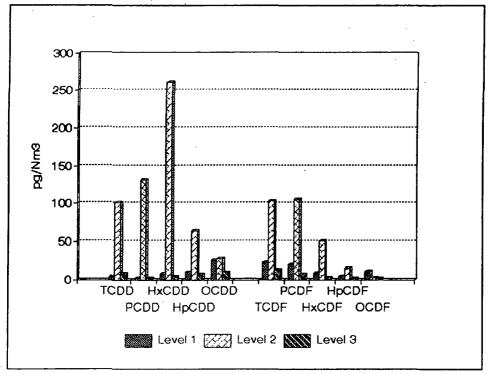


Figure 2 Congener distribution of PCDD/Fs in different sections of the flame

The concentrations of PCDD/Fs in the gas engine flue gas from the waste water treatment plant were lower than 0,009 ng TE/Nm³, as defined by NATO/CCMS. PCDD/Fs were also measured in 11 soil samples of non-sludge treated areas surrounding the emission sources of the waste water treatment plant. The 3 soil samples from the region where the flue gas descends in estimated highest deposition showed PCDD/F values lower than 1 ng TE/kg d.w. (NATO/CCMS). The congener distribution of PCDD/Fs in the 11 soil samples gave no indication of combustion residuals caused by the emission from the sewage gas burner.

2.3 Results of the analysis of hay, grass and milk samples

For this investigation the areas most highly contaminated with PCDD/F were compared with non contaminated areas. The analysis of the hay and grass samples were conducted by the Bavarian State Agency of Water Research and the milk samples by the Landesuntersuchungsamt für das Gesundheitswesen Südbayern. Compared with some of the very high PCDD/F levels in the soil samples (maximum value 173,6 ng TE/kg d.w., NATO/CCMS), the low PCDD/F content of the hay, grass and milk samples is remarkable. The PCDD/F content of the grass samples is on the whole lower than the content, measured in grass samples of rural areas in Nordrhein-Westfalen, Germany (3). Also the hay samples show low PCDD/F content (range 0,03-1,34 ng TE/kg d.w., NATO/CCMS). The PCDD/F contents of the cow's milk samples are with the exception of 3 samples below 0,9 ng TE/kg fat, NATO/CCMS, and at the lower end of background levels for non contaminated areas (0,7-1,5 ng TE/kg fat, NATO/CCMS). Even the maximum concentration in the cow's milk samples of 1,26 ng TE/kg fat (NATO/CCMS) is below the maximum values from non contaminated areas. A correlation between the PCDD/F values in soil and those in grass, hay and milk was not found.

The congener distribution of PCDD/Fs in the soil, grass and hay samples is similar (mainly HpCDD and OCDD congeners). The congener distribution of PCDD/Fs in the milk samples is different (mainly HxCDD, HpCDD, OCDD, PCDF and HxCDF congeners).

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