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Case study of a highly dioxin contaminated sports field: Environmental risk assessment and human exposure

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Abstract

Copper slag, containing high levels of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/PCDF) was used as covering material of a community sports field. This led to a contamination up to 40 000 fold in comparison to background concentrations in soils. Levels and patterns of dioxins found in sediments of a nearby river proved a translocation of contaminated copper slag, while slightly elevated dioxin levels found in soil from nearby house gardens could not be attributed to the sports field as the source of dioxins.

The annual flux of dioxins and furans to the environment was estimated using worst-case assumptions and the potential exposure of humans was deduced. Calculations of possible PCDD/PCDF-fluxes to the environment showed that only one major process – wind erosion – may have contributed to more than 90% of the total dioxin flux from the contaminated site to the surroundings. Based on PCDD/PCDF-exposure pathways for humans it was concluded that the PCDD/PCDF-content of the copper slag of the sports field did not pose a health hazard to athletes nor to the residents living in the neighbourhood.

Keywords: Contaminated site; Environmental-impact-assessment; Human exposure; PCDD/PCDF; Risk-assessment; Transfer

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1. Introduction

At least 800 000 t of copper slag from ore mining has been brought onto the German market during the 1950 and 1960s [1]. Sold as "Kieselrot" this slag was used as covering material for sports fields and playgrounds as well as for road construction dibenzo-*p*-dioxins, dibenzofurans (PCDD/PCDF), and other chlorinated organic

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compounds such as polychlorinated biphenyls (PCB) and chlorophenols in mg/kg concentrations [2]. Consequently, a widespread contamination of sports fields and soils due to Kieselrot was assumed and German authorities were concerned about the potential risk of this contamination to humans as well as to the environment.

Communities and sports clubs as owners of the dioxin-contaminated sports fields asked for a risk assessment for existing sites although at that time only very little information was available about the environmental fate and impact of the copper slag. The study presented here was conducted on request of a community in the Frankfurt area, Germany.

2. Sampling and chemical analysis

A suitable sampling and analysis scheme was developed to investigate the site and the surroundings in order to locate the vertical and horizontal distribution of the PCDD/PCDF-contamination. In total, 45 composite samples were analysed with high resolution gas chromatography coupled with a high resolution mass spectrometer (HRGC/HRMS). A detailed description of the sampling and analysis and analysis scheme is given elsewhere [3].

The sports field itself consisted of two fields (Fields II and III, approx. 60 000 m^2 each) and one sprint-course (approx. 500 m^2). Due to the potential transfer of PCDD/PCDF to the vicinity, samples were also taken from nearby house gardens, river sediment, and the drainage system of the field (see Fig. 1).

The total concentration of PCDD/PCDF can be expressed as toxic equivalents (TEQ) which is a normalization of all toxic congeners (those with chlorine at least in the 2,3,7,8-positions of the dioxin or furan molecule) with respect to the reference substance 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-Cl_4DD). This compound gives the highest toxic response in in vivo and in vitro tests, therefore, an equivalency factor of 1 is attributed to the 2,3,7,8-Cl_4DD. All other congeners are characterised, due to their relative toxicity in comparison to 2,3,7,8-Cl_4DD with factors between 0.001 and 0.5 [4].

Results demonstrated that Field II and the sprint-course were heavily contaminated with dioxins and furans due to the use of Kieselrot. In contrast, results from Field III, from the surroundings, the drainage system and the river sediment indicated a secondary contamination with PCDD/PCDF due to transfer processes from Field II or the sprint course (Table 1).

After analysing the PCDD/PCDF, the homologue groups (tetra- through octachlorinated dioxins/furans) can be plotted vs. concentration. Environmental samples of different origin often show characteristic homologue profiles which can reflect the source of the PCDD/PCDF.

The top layer of the sports field showed the characteristic PCDD/PCDF-homologue profile of the Kieselrot-slag: In such samples levels of both PCDD and PCDF increase with increasing degree of the chlorination. The octachlorodibenzofuran (Cl_8DF) is the predominant compound so it is relatively easy to identify soil and sediment samples contaminated with Kieselrot-slag (see Figs. 2(a) and 3(a)).

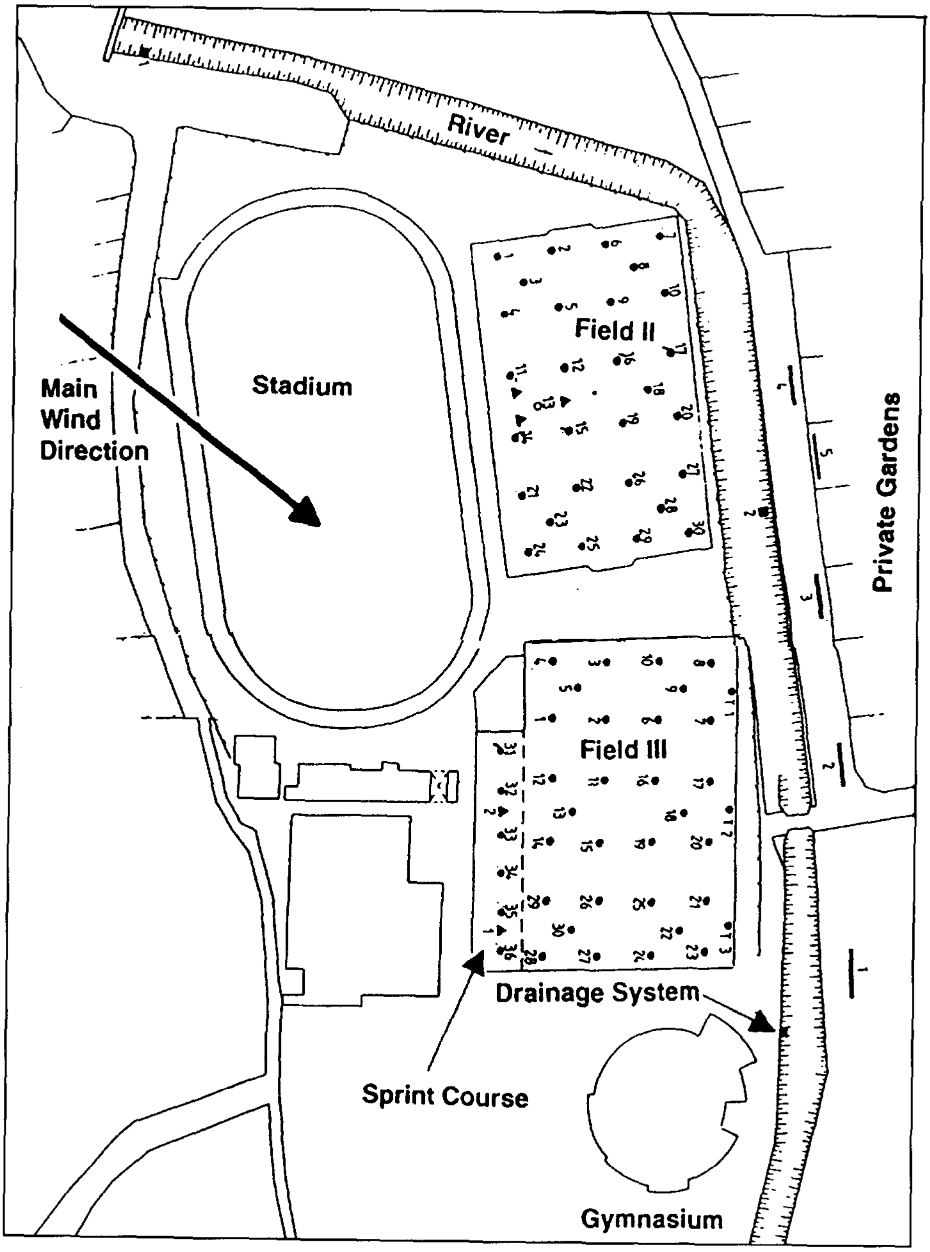

Fig. 1. Map of the contaminated sports fields and prevailing wind direction.

Table 1
PCDD/PCDF-concentration and TEQ (in ng/kg d.m.) of selected samples; samples from Field II, III and the sprint-course are composite samples

Location	Depth	Σ PCDD/PCDF	I-TEQ
Field II	0–5 cm	4 767 000	43 700
	5–12 cm	648 900	7450
	12–18 cm	170 100	1720
	18–23 cm	85 100	728
Field III	0–7 cm	6 808	85
	0–7 cm	27 960	245
	0–7 cm	22 580	181
Sprint-course	0–10 cm	289 600	2350
	10–17 cm	516 300	3950
Soil (five samples)	Surface	Range: 208–2880	2.2–58
Drainage system	Surface	605	7.7
River sediment, 100 m downstream	Surface	370	3.0
River sediment, 1 km downstream	Surface	143	1.3

3. Environmental risk assessment

3.1. Hazard identification

PCDD/PCDF were selected as indicator compounds of the risk assessment process. These substances have a high toxic potential to humans and are of special environmental concern. For exposure assessment possible pathways were identified and listed in a matrix (Table 2).

Some simplified assumptions were made to quantify the amount of PCDD/PCDF transferred to the environment:

- the average concentration to a depth of 0.1 m is 50 000 ng TEQ/kg, and ΣPCDD/PCDF 5 000 000 ng/kg for Field II as well as for the sprint-course;
- inhomogeneities of the horizontal and vertical PCDD/PCDF-concentration are neglected;
- mass fluxes due to the transfer processes are not time-dependent (steady state).

3.2. Hazard analysis

3.2.1. Transfer via sportswear of athletes

A merely mechanical transport of PCDD/PCDF with Kieselrot dust and particles, adhesively bound to sportwear (clothes and shoes) of athletes can lead to a contamination of the vicinity as well as causing a potential hazard to humans. An average uptake of 1–10 g of Kieselrot should reflect the correct order of magnitude.

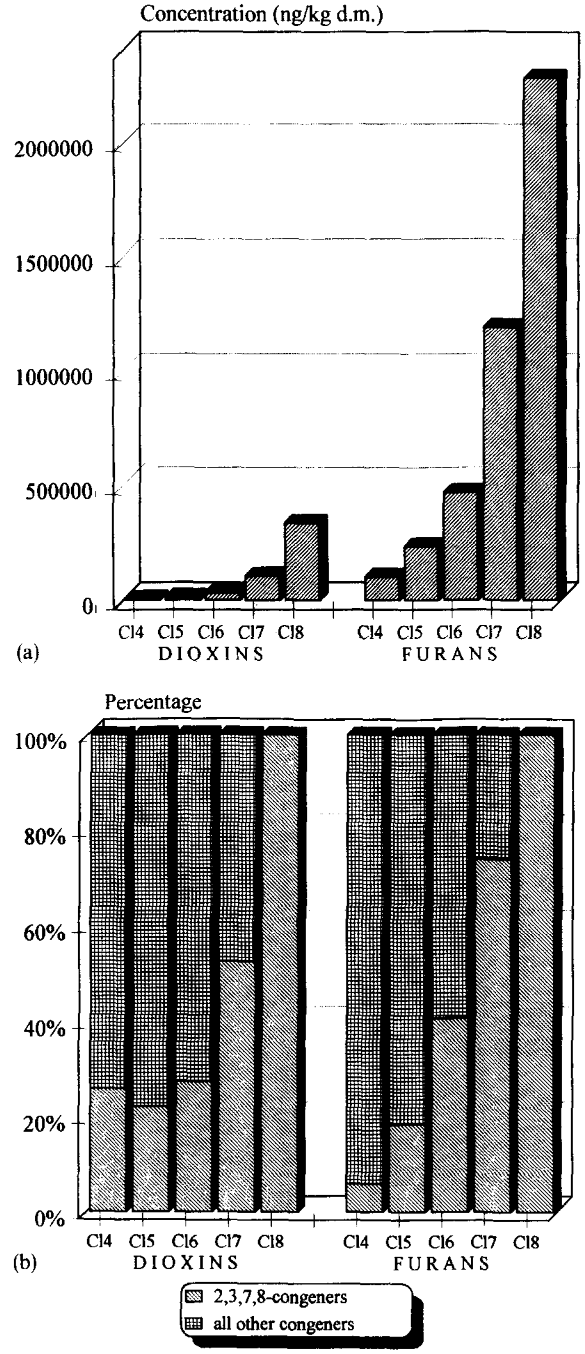

Fig. 2. Homologue profile (a) and percentage of 2,3,7,8-substituted congeners within the corresponding homologue groups (b) of Kieselrot-sample from Field II.

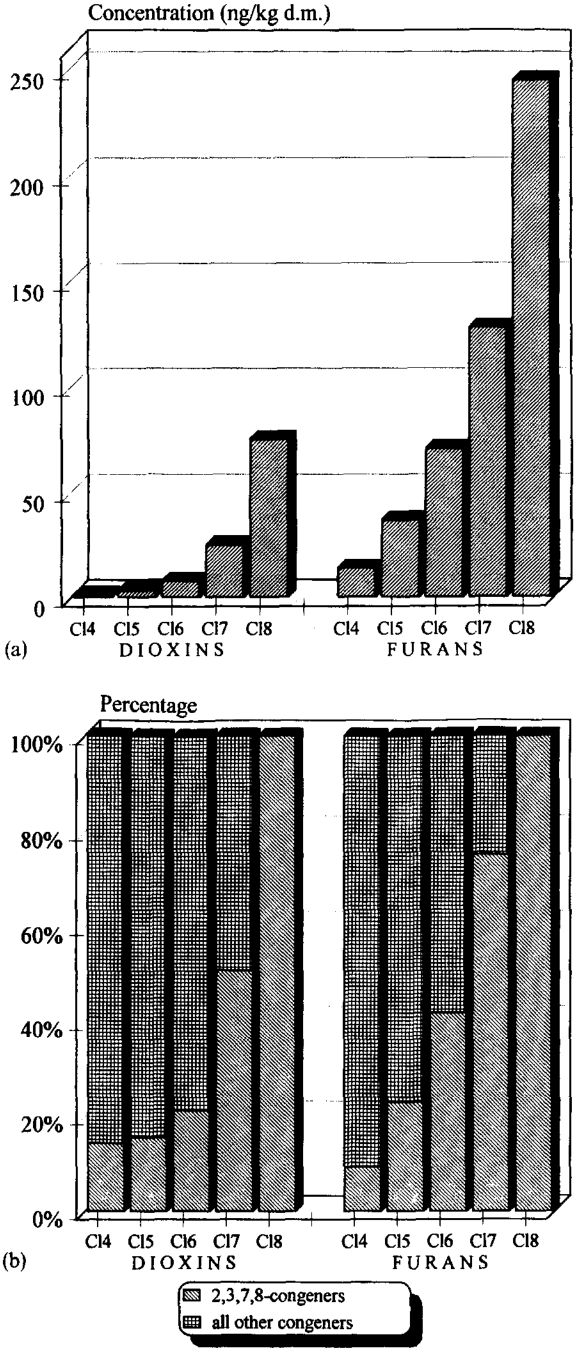

Fig. 3. Homologue profile (a) and percentage of 2,3,7,8-substituted congeners within the corresponding homologue groups (b) of a river sediment sample close to the drainage system.

Table 2
Environmentally relevant properties of PCDD/PCDF and copper slag

Type of process	Relevant properties of PCDD/PCDF	Relevant properties of Kieselrot-slag	Compartment(s) affected
Natural			
Wind erosion	—	Particle size	Air, soil, human
Abrasion	—	Particle size	
Evaporation	Vapour pressure	Heat transfer	Air, soil, human
Infiltration and surface run-off	Solubility and adsorption	Conductivity, particle size	Groundwater, water, sediment
Anthropogenic			
Maintenance, repair	—	Particle size	Air, soil, human
Adhesive transfer with sportswear of athletes	Adsorption	Particle size	Soil, human

Table 3
Transfer of Kieselrot-slag from Field II and the sprint-course via sportswear of athletes

Athletes per year	Uptake per training (g Kieselrot/athlete)	Annual transfer (ng TEQ/yr)
1000	1	50000
	10	500000
10000	1	500000
	10	5000000

An estimation of toxic equivalents being removed from the sports fields by 1000 or 10000 athletes (range of athletes per year on the site) is given in Table 3.

3.2.2. Transfer due to maintenance

Maintenance and repair of the sports field surface may lead to secondary emissions of dust due to the application of fresh material. Additionally, the change in particle size distribution to smaller particles by rolling the field might favour increasing wind erosion. On the other hand, the field was sprinkled regularly with water for dust control. The net transfer rate for PCDD/PCDF from these two effects cannot be quantified and had to be neglected.

3.2.3. Transfer via wind erosion

Uptake and transport via wind erosion depends on the particle size and compactness of the Kieselrot-slag. Lahl [5] assumed that the erosion rate of a sports field is in the same order of magnitude as from agricultural land although sports fields are sprinkled regularly (Table 4).

Table 4
Transfer of PCDD/PCDF from Field II and the sprint-course via wind erosion at two different erosion rates

Erosion rate (t/yr)	Concentration (ng TEQ/kg)	Annual transfer (g TEQ/yr)
1	50 000	0.05
10	50 000	0.5

3.2.4. *Transfer via evaporation*

A direct transfer of dioxins and furans into the ambient air can occur via evaporation due to the compound's vapour pressure at the actual on-site temperature. Assessing the real evaporation rates is very difficult because one has to take into account the specific properties of the Kieselrot-slag (e.g. humidity, organic carbon content) as well as environmental factors (e.g. rainfall, temperature).

Model calculations by Paustenbach et al. [6] showed that the evaporation flux of 2,3,7,8-Cl_4-Cl_4DD from a heavily contaminated soil (100 000 ng/kg of 2,3,7,8-Cl_4DD) is about 6.7×10^{-7} ng/s m^2. Higher chlorinated PCDD/PCDF doe not tend to evaporate significantly from soil and were therefore not considered in this calculation.

Total concentration of the 2,3,7,8-substitute3d tetra- and pentachlorinated dioxins and furans in the top layer of Field II was 53 000 ng/kg or expressed as toxic equivalents approx. 11 000 ng TEQ/kg, which means about 10% of the contamination used for the model calculation of Paustenbach et al. [6]. However, the transfer rate from the Field II to the air was not changed.

According to this model, evaporation took place at an annual rate of 15 mg/yr of tetra- and pentachlorinated dioxins and furans (approx. 3 mg TEQ/yr) from Field II.

3.2.5. *Transfer via water*

Due to the very low water solubility of PCDD/PCDF (e.g. 2,3,7,8-Cl_4DD < 2 μg/l), transport in the pure water phase can be almost excluded. Thus, a transfer of dioxins and furans in the water phase will only occur as transport of Kieselrot particulates. Surface run-off can be neglected for the sports fields because it is equipped with a drainage system. The sludge from the drainage system had been removed regularily for maintenance purposes which means that dioxin levels in drainage sediment reflected the actual transfer and not the accumulation over a long time.

Comparing the PCDD/PCDF concentrations from the sports field with those from the drainage system demonstrated that less than 0.05% of the dioxins were present in the sediments of the drainage system (Table 5). However, this is a very small transfer, the resulting sediment concentration of 7.7 ng TEQ/kg was clearly above background levels (approx. 1 ng TEQ/kg).

The amount of PCDD/PCDF translocated to the drainage system is very difficult to estimate. From the amount of sludge present in the system it was concluded that a total export of 1 μg TEQ/yr certainly represented an upper limit.

Table 5
Comparison of PCDD/PCDF concentrations and TEQ (in ng/kg d.m.) found on Field II and in the drainage system

Parameter	Field II	Drainage system	Concentration ratio
TEQ	43 700	7.7	5675:1
Σ PCDD/PCDF	4 767 000	605	7880:1

Table 6
Maximum transfer rates (in mg TEQ/yr) of PCDD/PCDF from Field II and the sprint-course to the environment

Compartment	Sediment	Air	Soil	Total
Kieselrot	0.001	3	500 + 5	508

3.2.6. *Overall transfer of PCDD/PCDF*

Summarizing the estimated transfer rates of PCDD/PCDF from the contaminated site to other environmental compartments (Table 6) via the paths described above leads to the following conclusions:

- transfer via drainage system to the river sediment does not exceed 1 µg TEQ/yr,
- transfer to air due to direct evaporation does not exceed 3 mg TEQ/yr,
- transfer to surrounding soils due to removal with sportswear of athletes is in the range of 0.05–5 mg TEQ/yr,
- transfer to surrounding soils due to wind erosion is the most important process with maximum transfer rates in the range of 50–500 mg TEQ/yr.

For an estimated total PCDD/PCDF content of 650 g TEQ (Σ PCDD/PCDF about 6.8 kg) of the whole contaminated site this means a maximum annual loss of about 500 mg TEQ or < 0.1% of the original amount.

4. Human exposure to dioxins from the sports field

The average daily intake of PCDD/PCDF of an adult person in Germany is approx. 120 pg TEQ. More than 95% of the uptake occurs with the consumption of food and only less than 5% can be attributed to other paths (dermal, oral, inhalative). Only these pathways have to be taken into consideration for the case study presented here.

Human exposure of dioxins from the sports field can take place via:

- dermal absorption from contaminated sports wear;
- oral ingestion and intestinal absorption via uptake of contaminated particles during activities on the sports field;
- inhalation of ambient air contaminated by evaporation of dioxins from the site.

4.1. Dermal uptake

Absorption of dioxins by skin contact is unlikely because of the strong binding of PCDD/PCDF to the copper slag [7]. Moreover, the penetration potential of PCDD/PCDF into human skin is very poor [8]. This path can therefore be neglected.

4.2. Oral uptake

Intestinal absorption of PCDD/PCDF from Kieselrot-slag after oral uptake is estimated to be about 1% with respect to the total TEQ of the slag. This value was determined with a synthetic digestive mixture [7]. Thus, the ingestion of 1 g copper slag (50 000 ng TEQ/kg) would result in a maximum uptake of 0.5 ng TEQ, which corresponds to about 1% of the annual background uptake of an adult person (approx. 40 ng TEQ/yr) in Germany via food.

Epidemiological studies on residents living close to a former copper slag production site clearly demonstrated that the bioavailability of PCDD/PCDF from Kieselrot is low [9]. Serum dioxin levels in this cohort which was directly exposed for years to Kieselrot particles and dust showed only a slight increase especially of higher chlorinated PCDD/PCDF. Although the serum dioxin concentrations are within the range of PCDD/PCDF background levels in Germany of 12–94 ng I-TEQ/kg, on fat basis [10], the dioxin pattern found in these blood samples supported an uptake of PCDD/PCDF from the copper slag. Toxicologically this additional intake was not relevant because it was mainly restricted to octachlorodibenzo-*p*-dioxin (Cl_8DD), a congener of low biological activity. Clinical laboratory tests of this cohort were not different from a control group without any Kieselrot contamination [8].

4.3. Inhalation

Generally, the inhalative uptake contributes to less than 1% of the total daily intake. This value was derived from air concentrations of 50 fg/m^3 which are typical for rural and semi-urban regions. Even with considerable higher dioxin levels in ambient air the contribution to the daily intake would be small in comparison to the uptake via food.

5. Conclusions

Based on a comprehensive risk assessment which included environmental and toxicological aspects, it was concluded that the dioxin burden of the contaminated sports fields did not represent a hazard to public health. The additional dioxin uptake due to the sports fields is so small that an impact on human health could not be deduced.

Nevertheless, the sports fields were closed to the public and the contaminated sites were covered completely with plastic foil to avoid further wind erosion and infiltration of rain water. So, the main transfer paths were cut and a contamination of the surroundings can now be excluded. For precautionary reasons the contaminated material will be removed and disposed of as hazardous waste.

Acknowledgements

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