

Timo Walter Assmuth

**Toxicant distributions and impact models in
environmental risk analysis of waste sites**

Yhteenveto: Myrkyllisten aineiden jakaumat ja vaikutusmallit
jätealueiden ympäristöriskien analyysissä

20

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ISBN 952-11-0017-6
ISSN 0783-9472

Yliopistopaino 1995

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"You must also also know that the Law ... was not given with a view to things that are rare ... but it has with all the knowledge ... it teaches the most prevailing conditions in mind ..."

- Moses Maimonides (1135 - 1206): *The Guide of the Perplexed*. S. Pines (ed.), University of Chicago Press 1963, part 3, ch. 34, p. 534-535, and A. Weiss (ed.), Hamburg 1973, Vol. 2, p. 211.

"So many circumstances of a small and accidental nature are relevant that no broad and simple uniformities are possible"

- Bertrand Russell (1872 - 1970): *Human Knowledge, Its Scope and Limits, Part V*. Simon and Schuster, New York 1948.

TOXICANT DISTRIBUTIONS AND IMPACT MODELS IN ENVIRONMENTAL RISK ANALYSIS OF WASTE SITES

Timo Walter Assmuth

Assmuth, T. W. Toxicant distributions and impact models in environmental risk analysis of waste sites. Helsinki, Finnish Environment Agency. 79 p. Publications of the Water and Environment Research Institute. No. 20.

The occurrence and distributions of toxicants at various Finnish waste sites, the toxicity of their leachates and the associated environmental risks were investigated. The studies involved chemical, biological or physical measurements of solid, liquid, gaseous and biological samples from 43 sites, including municipal mixed-waste landfills, industrial waste deposits and contaminated soil sites. Toxic elements and organic compounds were often identified in the sites and in their emissions. The probability distributions of the concentrations of most toxicants at the study sites were skewed to small values. Many distributions were lognormal and truncated due to non-detects especially in leachate runoff; this affected the estimation of the statistics of the variables. In solid wastes, the ratio of mobile to total heavy metals mainly correlated with organic matter content and pH. Estimates of toxicant fluxes in surface waters varied strongly; the greatest average specific fluxes (per waste volume) were those of Zn, Cr and Ni. Water quality goals were exceeded particularly by peak concentrations of metals and chloroaromatic compounds in nearfield ground water and surface water, and air quality guidelines by chloromethanes and monoaromatic hydrocarbons in gas from wastes of municipal landfills. 2,3,7,8-chlorinated PCDD/PCDFs in high concentrations and characteristic patterns (mainly hexa- and heptachlorinated dibenzofurans) were found in soils and sediments at waste sites contaminated by chlorophenolic wood preservatives. Acute lethality of *Daphnia magna* in a 24-h screening test was noted in ca. 1/3 and mutagenicity to *Salmonella typhimurium* strains in all of the assayed leachate samples. Concentration-response relationships in log-probit scales showed variable forms and temporal development of toxicity to *Daphnia*. 48h-EC(I)50 estimates by power function correlated with concentrations of chlorides (serving as leachate indicator) and of some toxic elements in linear regression models. Yet, toxicity was not always noted despite concentrations exceeding reportedly toxic values.

Models of environmental risks of waste sites were devised and applied to toxicant prioritization in the aggregated leachate and landfill gas data, and to specific analyses of substances and impacts. Using linear models and estimates of present emissions, durations of the releases of toxicants from old municipal landfills spanned tens to hundreds of thousands of years. Cd, Ni and several chlorinated hydrocarbons were important in leachate runoff regarding observed distributions, persistence or bioaccumulation, tumorigenicity and health norms; ecotoxicological risks were associated mainly with

pesticides and other biomagnifying chemicals. The infrequently detected compounds were assigned high ranks only by upper fractiles of risk indices. In landfill gas the greatest overall health risks on the basis of air quality norms were caused by chloroform whereas ethylene dibromide was the most prominent carcinogen. By extrapolation from LC50 values of fish to NOAEC of hypothetical most sensitive species in loglinear sensitivity distributions, and to no-effect levels by assessment factors, margins of safety between observed and toxic values of pentachlorophenol concentrations decreased by orders of magnitude. Observed lethality of *Daphnia* correlated with impacts predicted by additive models which were based on published substance-specific LC50 data, only when flow-weighted concentrations and toxicity fluxes were used, displaying inconsistencies of chemical and biological based analyses. Estimates of the toxicity of PCDD/PCDFs were sensitive to the toxic equivalency system, e.g. differing between mammal and fish based systems. Estimates of carcinogenic risks of PCDD/PCDFs at waste sites associated with chlorophenolic wood preservatives ranged from > 1 (hypothetical worst case of sawmill soil ingestion) to background levels of $5 \cdot 10^{-6}$ using more plausible exposure and impact model assumptions; acceptable doses based on safety factors were exceeded by a factor of 0.4 to 32 000 for soil ingestion and 0.06 - 2 for fish consumption. The results were discussed with particular emphasis on the resolution, attributability and significance of toxicant and impact distributions in a space-time continuum under measurement and model uncertainty. Important research needs include gas emissions and leachate pulses due to attenuation capacity breakdown e.g. in soils, exposure conditions, variations in receptor vulnerability, chronic effects e.g. from mixtures of biomagnifying substances and future risks due e.g. to land use changes. Combined chemical and biological measurements coupled with risk models in a staged procedure are advisable in waste site monitoring and control for efficient resolution of such emissions, exposures, impacts and risks.

Key words: Waste, landfills, hazardous waste sites, chemicals, polychlorinated dibenzo-p-dioxins and furans, contamination, risks, toxicity, *Daphnia magna*, assessment, models, field data, uncertainties, probability distributions.

1 INTRODUCTION AND LITERATURE EVALUATION

1.1 General concepts and definitions

'Distribution' as a statistical term refers to the distribution function (DF) or cumulative distribution function (CDF) of a random variable, defined by the probability of values below a certain value (Dudewicz and Mishra 1988), and by the corresponding probability density function (PDF). Generally, distribution is the spatial, temporal or structural spread of properties.

'Risk' is "exposure to the chance of injury or loss, or the probability of such an injury" (The Random House Dictionary of the English Lan-

guage 1987). Risks are divided e.g. in natural and anthropogenic, chronic and catastrophic, individual and collective, and voluntary and involuntary risks (Cox and Ricci 1989).

'Environmental risks' encompass those natural or man-made risks which are directed to the environment of organisms. The concept is often used in a restricted sense of risks in the ambient environment. 'Toxicological risk' is associated with toxic effects of chemicals on organisms. The concept of 'health risk' is usually restricted to humans, and includes also other kinds of health impairment than toxic effects (Gaylor 1988). 'Ecotoxicological risks' involve adverse effects on other species, on communities and on species interactions (Suter 1993).

'Environmental impact' is defined broadly as an alteration in the quality of the environment by natural or artificial process. A stricter definition includes man-made impacts harmful to man, or to other organisms and ecological systems. Alternatively, one may include non-harmful impacts but limit the scope to human values.

'Risk analysis' is the systematic intellectual activity striving to understand risks, and is often used as a synonym for risk assessment. In environmental health, 'risk assessment' has been divided in risk identification, exposure and dose-response assessment and risk characterization (U.S. National Academy of Science 1983).

'Waste' is a substance or object which is discarded, or intended or required to be discarded (Council of the European Communities 1991a). 'Hazardous wastes' have been defined by reference to their properties, source and composition (Council of the European Communities 1991b). In Finland, the roughly equivalent concept 'problem waste' has been used in Waste Act of 1993. Only non-radioactive wastes are included in these statutory definitions. 'Chemical' is a substance produced or used in a chemical process (The Random House Dictionary of the English Language 1987). According to Finnish statutes, the term includes organic and inorganic substances. The emphasis is on synthetic or artificially

processed non-radioactive substances. 'Contaminant' is a substance which contaminates materials to render them harmful to organisms. 'Toxicant' is a toxic chemical substance.

'Waste disposal site' is a limited area used for prolonged disposal of wastes. 'Landfill' has been defined as a site used for the controlled deposit of waste onto or into land (Commission of the European Communities 1991). Waste disposal sites may be classified by their main waste qualities and properties (Fig. 1). 'Uncontrolled hazardous waste site' is a common but inexact concept for sites containing hazardous wastes disposed in an uncontrolled manner, often in old and abandoned disposal sites (cf. Franzius 1992). 'Contaminated soil site' is a term used for local chemical contamination of soil (Franzius 1989). Natural contamination and radionuclides or pathogenic organisms are usually excluded. 'Waste site' is a broad term for spatially limited concentrations of wastes, and includes uncontrolled hazardous waste sites and landfills as well as other kinds of chemically contaminated sites (Fig. 1). The justification for such a broad definition is that wastes cause soil contamination and vice versa, and chemically contaminated soil sites and disposal sites for wastes containing toxicants are conceptually similar phenomena from the point of view of risk analysis (1.3.2).

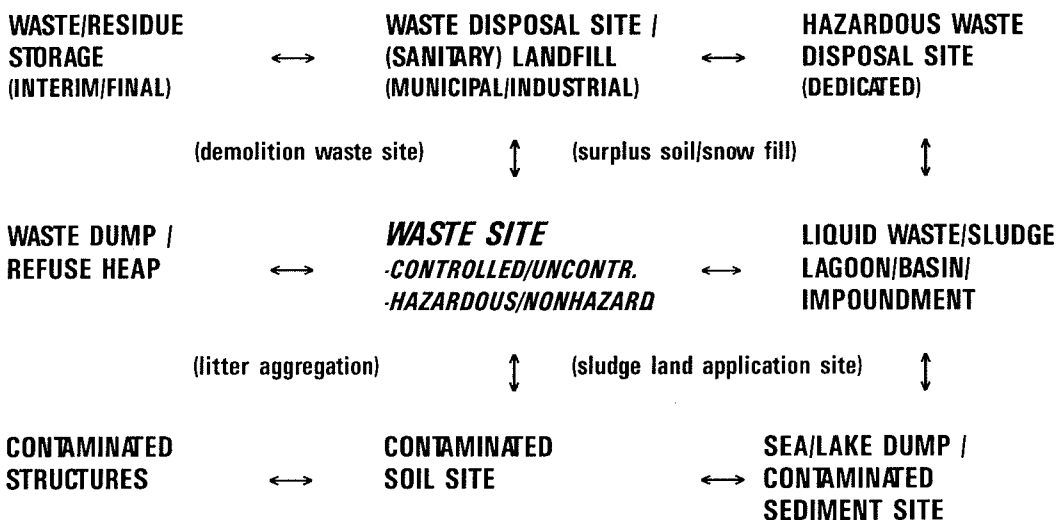


Fig. 1. Definitions and delimitations of the concept of waste site.

1.2 Character and scope of the problem field

1.2.1 Waste sites

Development and importance

Waste sites such as dumps of household and industrial wastes (Fig. 1) have existed throughout history. They were developed to sanitary landfills due to problems of pests, pathogens and nuisance caused by open dumps. More recently, waste sites have become a large-scale problem worldwide due to concerns of impacts caused by toxic chemicals in such sites. Extensive and increasing work with inventory, investigation, assessment and control of waste sites and contaminated sites has taken place during the last decade in industrialized countries (Table 1).

Research and development related to waste sites has grown to be one of the main areas of environmental science and technology. Considerable resources have been directed to the problem, e.g. special legislation and financing mechanisms such as the Comprehensive Environmental Responsibility, Compensation and Liability Act (CERCLA, known as the Superfund) and the Superfund Amendments and Reauthorization Act (SARA) in the USA, the Soil Cleanup Act in the Netherlands, and the Chemical Waste Dump Act and the 1990 Contaminated Site Act in Denmark (cf. references in Table 1).

Such developments have largely been responses to cases of local contamination leading to publicity and emergency action, such as Love Canal in USA and Lekkerkerk in the Netherlands. In many countries the focus has gradually extended from waste dumps to include other kinds of contaminated sites and more widespread soil and environmental contamination (Franzius 1989, Hojsholt 1993, Wolf 1993). In Finland, the case of groundwater contamination at a sawmill in Kärkölä has been an important case of local contamination (Lampi et al. 1992); thousands more sites have been identified in national inventories as potentially contaminated (Table 2).

Origins

Seen generally as a physical system, waste sites are the consequence of man-made and natural processes which emit chemicals into the environment, while still keeping them relatively concentrated. The contaminant source may be due to old or recent activities or accidents. The distinction between such toxicant aggregations and diffuse contamination is not sharp.

Waste sites are caused by many activities in the production, handling and disposal of materials and chemicals. Typical categories of waste sites and other kinds of potentially contaminated soil sites in Finland include old landfills, industrial waste areas, and scrap yards (Table 2).

Table 1. Evaluations of the magnitude of the problems and activities associated with uncontrolled hazardous waste sites (HWS) and other kinds of contaminated soil sites (CSS) in some countries.

Country	Potential/suspected contaminated sites		Prioritized sites/ contamination observed	Sites with implemented remedial action	Cleanup costs, estimated total (10 ⁹ ECU ¹)
	HWS	CSS			
Finland	400 ²	10 000 (20 000) ^{3,4}	700 (3 500) ^{3,5}	~50 ^{3,4}	0.4 - 0.8 ³
Denmark ⁶	3 000 ⁷	3 500	500 ⁸ (2 600) ⁹	several	> 0.8
Germany ¹⁰	75 000	> 8 000 (75 000) ¹¹	12 500	several	> 9 (20 - 200) ¹¹
Austria ¹²	2 200	5 400	900	> 10	> 0.7
Holland ¹³	3 700	140 000	25 000	≈ 1 000	11 (300) ¹⁴
USA ¹⁵	HWS + CCS ≈ 40 000		1 300 ¹⁶	> 400 ¹⁶	50 (500) ¹⁷

¹ECU = European Currency Unit; ²Seppänen 1986a; ³Puolanne et al. 1994; ⁴Assmuth et al. 1990a; ⁵In parentheses, sites on vulnerable areas; ⁶Welinder 1993, Hojsholt 1993; ⁷Including all dumps; ⁸Prioritized waste dumps; ⁹Registered sites; ¹⁰Franzius 1989, Ruppe 1991; ¹¹Higher estimates by Wolf 1993; ¹²Zirm 1990; ¹³van Veen and van der Galien 1988, Holtham and Gravesteyn 1993; ¹⁴Higher figure based on cleanup to target levels, de Vries (personal communication); ¹⁵Johnson 1995, ¹⁶On National Priorities List; ¹⁷Higher figure including Federal sites.

Disposal sites and technologies

Disposal still is the main treatment method for Finnish municipal and industrial solid wastes. The amount of registered Finnish waste disposal sites exceeds 1 700 (Table 2). Most of them are small municipal landfills; the total area covered by registered municipal or industrial landfills is roughly 40 km² (Seppänen 1986b). However, the bulk of the wastes are disposed in industrial landfills, e.g. in metal and forest industries.

The main waste categories in municipal landfills are mixed municipal solid wastes, construction wastes and sewage sludge. Sludges,

soil and energy production wastes are also disposed in dedicated sites (Seppänen 1986b).

Hazardous substances are included in both municipal and industrial wastes. Such substances have been disposed in wastes presently defined as hazardous particularly before the development of hazardous waste regulations and treatment (Wahlström et al. 1985). Most of Finland's hazardous wastes are now treated in a central incineration plant (Isaksson and Vahvelainen 1991), and so-called codisposal of hazardous wastes in municipal solid waste landfills is no more officially approved or commonly practiced.

Table 2. Types, toxicants, features and minimum amounts of Finnish waste sites and other kinds of potential chemically contaminated sites, on the basis of surveys (Assmuth et al. 1990a, Puolanne et al. 1994), and of information on branch-specific chemicals (Wahlström et al. 1985).

Contaminating activity or site type	Typical contaminants or contaminant groups ¹	Common site characteristics	Number of sites		
			total	poll. ²	vulner. ³
Sawmills	CPs/other chloroaromatics	shores	} 1 032	154	449
Wood impregnation sites	As/Cr/Cu/creosote (PAHs)				
Cardboard industries	phenols/pesticides				
Pulp and paper industries	(chlorinated) organics/Hg	large, shores	51	16	11
Mining waste areas ⁴	heavy metals/organics	piles, lagoons	37		
Metal industries ⁵	metals/solvents/cyanide		607	34	200
Graphic industries	solvents/heavy metals		177	2	39
Electronics industries	heavy metals/solvents		+		
Chemical industries ⁶	various branch-specific		346	26	117
Oil refineries	hydrocarbons/Pb/As		2		
Textile and leather industries	Cr/solvents/pesticides		166	11	51
Construction industries ⁷	oil/metals/phenols/asbestos		526	7	232
Food industries	solvents/pesticides		239	14	69
Gasoline stations	hydrocarbons/solvents	subsurface tanks	> 816	16	530
Mobile asphalt stations ⁸	hydrocarbons/solvents	gravel eskers	270	6	86
Waste sites	various	dumps/lagoons	1 682	170	427
Scrapyards and repair shops	oil/solvents/metals (Pb)	scrap piles	1 701	46	541
Wastewater purification plants	various	lagoons/sludge	560	27	116
Chemical laundries	chlorinated solvents		+		
Oil and chemical spill sites	various (oils/solvents)	transport areas	115	48	40
Chemical storage sites	various branch-specific	tanks/containers	68	3	38
Power plants	hydrocarbons/metals/PCBs	fuel/ash piles	419	29	109
Plant schools and gardens ⁹	biocides/oil		251	4	55
Domestic animal shelters	nutrients/organic matter	feed/residues	> 661	2	70
Complex industrial areas	various		146	9	88
Total			> 10 396	653	3 685

¹See Table 4 and list of symbols and abbreviations; ²Ascertained to be contaminated; ³On important ground water areas or near housing; ⁴Ore and mineral mines; ⁵Incl. smelters and foundries; ⁶Incl. plastics, pharmaceuticals, rubber and fertilizer; ⁷Concrete and cement mills and building industries; ⁸Incl. oil gravel production; ⁹Large trade gardens.

Wastes in disposal sites are transformed depending on waste properties, disposal techniques and environmental conditions (Department of the Environment 1978, Ehrig 1980, Baccini et al. 1987). In Finland, important factors affecting disposed wastes include e.g. frost and snow (Ettala 1988). Inert wastes may remain unchanged for long periods, while wastes containing organic matter change biologically and chemically. A waste disposal site is thus a semi-closed reactor, affected by and affecting the environment (Collins and Spillmann 1986).

The emissions from waste disposal sites to the environment may include

- runoff of leachate and surface water (solutes, colloids and suspended solids) (Fig. 2)
- leachate discharge to ground water (Fig. 2)
- transport of gases (including water vapor)
- transport of aerosols (dust)
- large particles carried by the wind, by vehicles, humans and animals (or by gravitation)
- organisms migrating from wastes
- physical energy in dispersing temperature, noise, pressure (in explosions) and radiation.

Landfill technology in Finland has been rather low level especially at old and small landfills. Usually only natural soils have been used as bottom layers. Waste pretreatment and compaction have been practiced mainly at large sites. Covering frequency and intensity have been modest particularly at small sites. Leachates have been mainly discharged without treatment in dikes or directly to recipients or soil (Seppänen 1986b). In recent years, leachate diversion to treatment has increased. Landfill gas collection and treatment systems are still rare.

1.2.2 Description of the risks caused by waste sites

Toxicological risks

Toxicological risks of waste sites may be divided by their receptors, rapidity, action sites and mechanisms, severity, and spatial extension (Table 3), as well as by their source (e.g., synthetic and natural) and the affected compartment (e.g., aquatic and terrestrial).

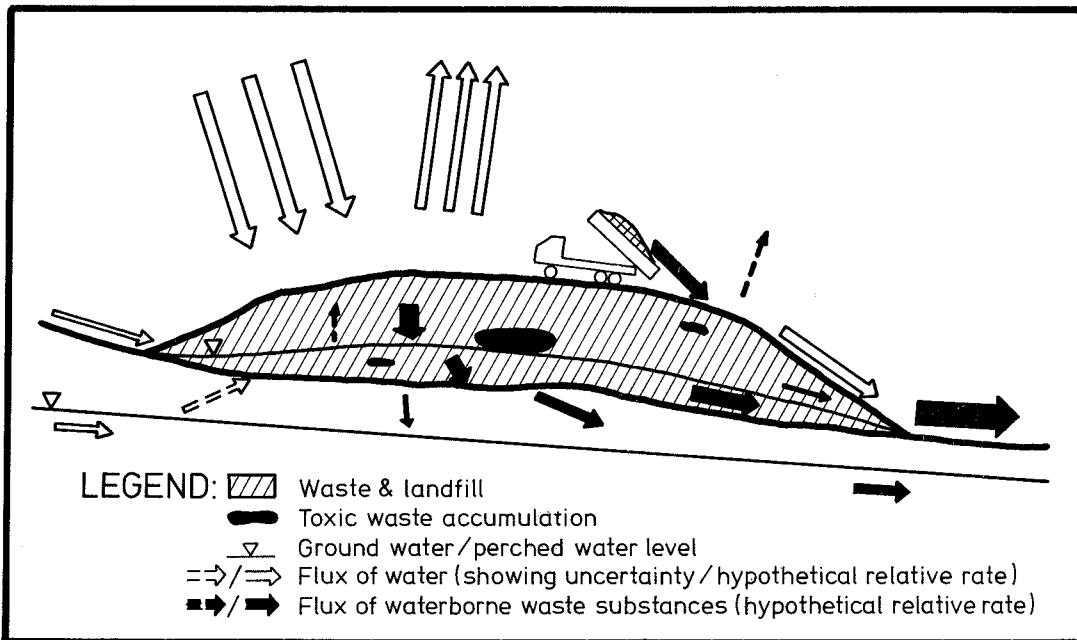


Fig. 2. Schematic presentation of the processes and fluxes of waterborne hazardous substances in waste disposal sites (Assmuth et al. 1991).

Table 3. Classifications of the toxic effects of waste sites and chemically contaminated sites.

Ecological level	Target class	Rapidity	Mechanism or target system	Action route	Severity	Areal extension
molecular	(virus)	acute	irritative	oral	lethal	<i>in situ</i>
cellular	bacteria	subacute	respiratory	dermal	sublethal,	on site
tissue	fungi	subchronic	hepatotoxic	inhalation	irreparable	local
organ	algae	chronic	nephrotoxic		repairable	(regional)
organism	higher plants	hereditary	neurotoxic			
population	animals		behavioral			
community			immunotoxic			
(ecosystem)			hormonal			
			reproductive/developmental			
			genotoxic/mutagenic			
			teratogenic/carcinogenic			

Risks may have existed in the past or may exist in the future, also after actual disposal operations. Indirect risks are caused e.g. as waste leachates disturb wastewater purification plants (cf. Ettala and Rossi 1994).

The environmental risks of toxicants in waste sites are formed in stages through dispersal along various routes. People potentially exposed to toxicants around a waste site include visitors, nearby residents, and consumers of water, fish or wildlife contaminated by site emissions (e.g., Kruse and Wieben 1989, Paustenbach 1989).

Ecotoxicological risks are posed to organisms residing in or visiting wastes, or to organisms and communities in site surroundings (U.S. EPA 1989b, Suter 1993). There is a variety of potential effects on these targets (Table 3).

Particular risks for human health may be caused by waste sites located on ground water and housing areas. About 200 Finnish landfills are sited on permeable soils or in ground water protection areas (Seppänen 1986a). Many of these sites are small and closed municipal landfills. Manyfold more other kinds of contaminated sites are located on aquifers (Table 2). Yet, other exposure routes may be more commonly dominant than ground water (Puolanne et al. 1994). In housing areas, air pollution, direct contact and bioaccumulation mediate risks from waste sites (Kimbrough et al. 1984).

Toxicants in waste sites also cause occupational health risks. Landfill workers may be exposed to both pathogens and toxicants, e.g. metals, in normal operations (Rahkonen 1987) and in landfill fires (Rahkonen et al. 1994).

Other occupational groups at risk include industrial workers in facilities located at contaminated sites; construction, maintenance or demolishing crews at such sites; emergency response teams (e.g., firemen); and site remediation personnel (Brett et al. 1989).

Toxicological risks from with waste sites may be caused by catastrophic events such as fires and floods, or by more continuous releases of toxicants. The cumulative impacts on the environment are due to both kinds of mechanisms and can often not be separated.

Toxicological risks may be caused by substances in municipal wastes and industrial wastes not designated as hazardous. Apart from synthetic chemicals and hazardous elements, organic compounds occurring naturally in wastes may cause environmental risks; e.g., methane from anaerobic decomposition of organic wastes may cause suffocation, explosions and atmospheric impairment (see references in II).

Other risks

Environmental risks are also caused e.g. by pathogens and pests in wastes and through physical factors such as accidental injuries, explosions (of landfill gas and explosives in wastes), fires, radiation, and corrosion and structural failures (e.g., landslides) (Fig. 3). Some of these risks, e.g. those caused by fires, involve chemical releases and toxicological risks as well (Persson and Bergström 1991, Rahkonen et al. 1994).

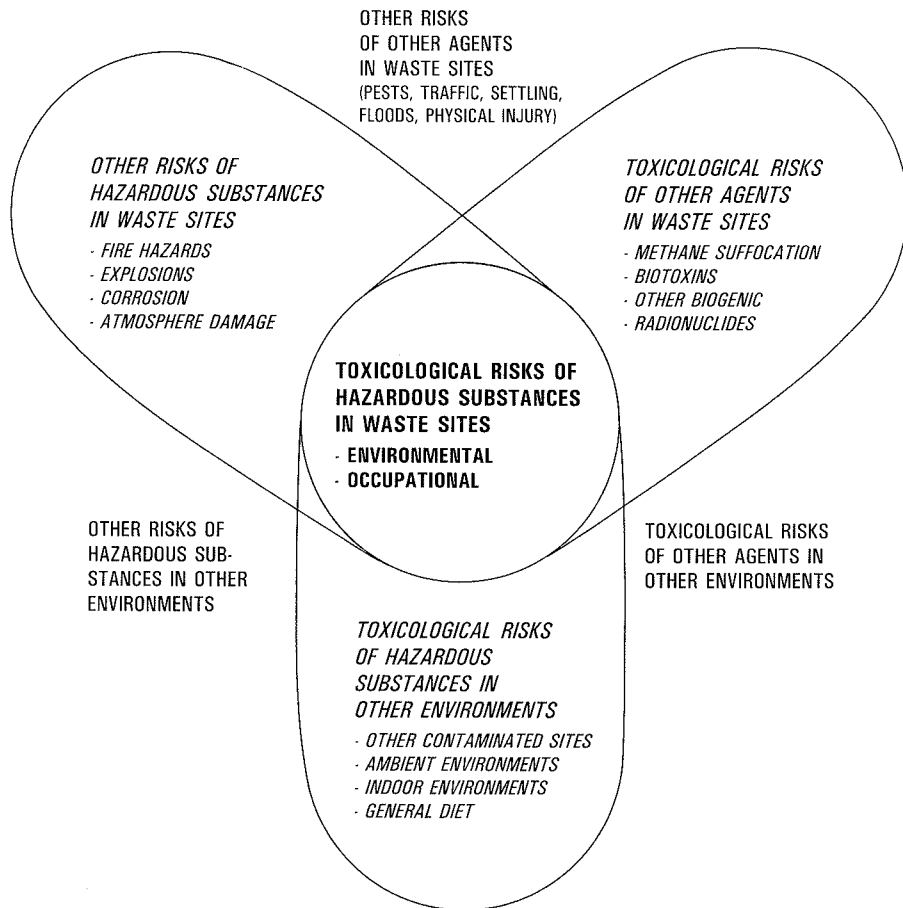


Fig. 3. Conceptual presentation of the toxicological risks associated with hazardous substances in waste sites and of related risks, and scoping of the studies (shown by delineation).

1.3 Previous studies of toxicological environmental risks of waste sites

1.3.1 Quantification of toxicant occurrence and exposures

Many measurements of the occurrence of toxic substances at chemically contaminated sites have been made in recent years. These studies have addressed municipal and industrial landfills, hazardous waste dumps and industrial areas. Laboratory and field analyses have been made of the quality of wastes and soil, surface and

ground water, soil gas (Table 4) and of sediments, sludges, biota, ambient air and structures.

The results of studies of toxicant concentrations in and around waste disposal sites have been reviewed in articles (I - IV). These data are in part still very incomplete. They are also heterogeneous, and their representativity and reliability are not always good or clear. Even the more comprehensive and consistent investigations have produced widely varying data (Table 4). Most importantly, the significance of chemical measurements in terms of toxicological risks has seldom been explicitly assessed in studies and monitoring of waste sites.

Table 4. Results from measurements of toxicants at municipal mixed-waste landfills, hazardous waste disposal sites and chemically contaminated soil sites in USA and Germany (mainly from references in I - IV, Kruse and Wieben (1989), Kerndorff et al. (1992), Plumb (1992), and Chilton and Chilton (1992)). Previous appropriate Finnish data (Ettala et al. 1988, Pärjälä 1989) are in parentheses.

Substance or group (chemical symbol or abbreviation)	Landfill leachate		Impacted groundwater		Impacted soil conc. ($\mu\text{g g}^{-1}$), Max ³	Landfill gas conc. (mg m^{-3}), Max
	Conc. ($\mu\text{g l}^{-1}$), f ₅₀ ¹	f _{Detn} ² (%)	Conc. ($\mu\text{g l}^{-1}$), f ₅₀	f _{Detn} (%)		
<u>Elements</u>						
Arsenic (As) ⁴	14 - 34	15	1.3		394	
Cadmium (Cd)	12 - 20 (0.7)	15	nd		7.3	
Chromium, total (Cr)	97 - 780 (25)	15	3		119	
Copper (Cu)	50 - 58 (15)		7.0		1 200	
Mercury (Hg)			nd		62	
Lead (Pb)	68 - 100 (18)	30	nd		870	
Nickel (Ni)	260 - 390 (37)		13		75	
Zinc (Zn)	510 - 890 (220)	14	65			
<u>Unhalogenated aromatic hydrocarbons</u>						
Benzene	300	26	14	2.4 - 34	144	180
Toluene (Tol)	360	28	3.5	7.7 - 33	19	460
1,3/1,4-Xylene ⁵ (13/14-Xyl)	73	13	2.8		22	
Ethylbenzene	44	13	4.4	2.6 - 18		330
Naphthalene			1.1	0 - 21	4 000	
Acenaphthene			0.3	0 - 3.8		
Phenanthrene			0.6	0 - 3.7		
Phenol		15	1.5		≤ 27	1 440
4-Cresol (4-Cres)			86			
<u>Chlorinated aliphatic hydrocarbons</u>						
Dichloromethane (DCM)	440 - 2 700	12	440	4.9 - 31		190
Chloroform	7.1 - 130 (0.04)	20	1.4	1.5 - 44	21	0.8
Carbon tetrachloride (CCl ₄)	202 (0.04)		0.2	0 - 10		
1,2-Dichloroethane (12-DCE _a)	220		nd	2.3 - 28		8
1,1,1-Trichloroethane (111-TCE _a)	(0.2)	14	1.0	1.8 - 24	0.74	177
cis-1,2-Dichloroethene (c12-TCE _a)			170			
trans-1,2-Dichloroethene (t12-TCE _a)		11	50	4.9 - 40		302
Trichloroethene (TCE _a)	(0.09)	33	2.3	7.3 - 76	1.9	170
Tetrachloroethene (TeCE _a)	16 (0.2)	16	1.4	4.3 - 52	85	350
Vinyl chloride	40		100	0 - 17		
<u>Chlorinated aromatic hydrocarbons</u>						
1,2-Dichlorobenzene (12-DCBz)			0.9	0 - 7.1		
1,3-Dichlorobenzene (13-DCBz)			1.1	0 - 2.7		
1,4-Dichlorobenzene (14-DCBz)			2.2	0 - 3.9		
Hexachlorobenzene (HCBz)				0 - 1.5	0.22	
2,4,5-Trichlorophenol (245-TCP)	(nd)		1.3 (0.3)			
2,4,6-Trichlorophenol (246-TCP)	(0.2)		(0.02)	0 - 15		
2,3,4,6-Tetrachlorophenol (2346-TeCP)	(4)		(0.12)			
Pentachlorophenol (PeCP)	25 (0.8)	22	(0.1)	0 - 18		
Dieldrin					≤ 12	
Endrin					≤ 2.6	
4,4'-DDT					≤ 7.6	
Lindane					≤ 18	
PCB compounds (PCBs)					≤ 18 ⁶	12
I-TEQ ⁷ (water soluble)	0.00156					
I-TEQ (oil soluble)	0.06				varies	

¹Median value; for Finnish data, medians of site averages; ²Detection frequency; ³Maximum value; ⁴Arsenates; ⁵The two isomers unseparated; ⁶Aroclor 1254; ⁷International 2378-TCDD (2,3,7,8-tetrachlorodibenzo-p-dioxin) toxic equivalents (V, VII).

The applicability of foreign results to Finland is limited. The previous sparse Finnish studies of toxicants at waste sites (Table 4) have generally found lower concentrations than those measured abroad. Among other kinds of contaminated sites, chlorophenols and related chloroaromatic hydrocarbons have been commonly observed in high concentrations at Finnish sawmills (e.g., Kitunen and Salkinoja-Salonen 1990, Valo et al. 1984).

Controlled experiments of releases of toxicants and other materials from wastes have been conducted in field lysimeters (e.g., Collins and Spillmann 1986) and in a few full-scale field investigations (e.g., Baccini et al. 1987). In addition, several studies have been made of the transport and behavior of waste leachates in soil, both in the field (e.g., Borden and Bedient 1987) and in laboratory tests (e.g., Kjeldsen 1986). These researchers have stressed the role of the properties and processes of wastes for the retention, decomposition and release of toxicants. Transport of toxicants from wastes into their environment is governed by substance properties and environmental setting. Transport has been investigated particularly in relation to solutes in surface water (I) and in porous soils (III) by empirical and modeling methods. Previous research in chemicals at contaminated sites in Scandinavian conditions are sparse, including mainly Swedish studies of leachate migration in glacial till and ground water (Semán 1986, Gustafsson and Holm 1989, Lindmark 1989).

1.3.2 Assessments of responses to toxicants

1.3.2.1 Studies of health effects

Epidemiologic and clinical studies

Few studies of waste sites have given conclusive evidence of their adverse effects and of causes (Table 5, cf. Grisham 1986), although associations between health disorders and exposures to site emissions are often reported. This is largely due to difficulties in ascertaining statistically significant effects based on interviews and other cross-sectional surveys of small and heterogeneous exposed groups, and in attributing such effects to the local and often poorly defined sources of exposure (Talbot et al. 1987, Layard and Silvers 1989). The role of psychosomatic

factors in sensitization has been stressed (Neutra et al. 1991).

Analyses based on exposure assessment

Quantitative assessments of health risks of waste sites and contaminated sites on the basis of exposure estimation have become common. They vary in scope and complexity, and often rely on simple linear models (Table 6). A common approach is that proposed by U.S. EPA (1986), involving selection of indicator substances, estimation of exposures and calculations of intake, derivation of toxicity from published data, comparisons with reference values, and risk characterization (Zamuda 1989b).

Modified procedures have been presented and used for the analysis of waste site risks (Table 6). Mauskopf and Curtis-Powell (1985) estimated carcinogenic risks caused by toxicants in an unregulated landfill incorporating a life table analysis in the basic impact assessment. McKone and Daniels (1991) analyzed health risks from soil toxicants by linear models linking concentrations to exposure by pathway-specific factors. Risk models are used also inversely to derive safe levels of chemical concentrations (Hertzman et al. 1989, Table 6). The above exposure-oriented health risk assessment procedure for waste sites has been applied e.g. to remedial measures (Stephanathos and Sotsky 1989).

Also site-aggregating risk analyses have been made using similar basic approaches, e.g. by Brown and Donnelly (1988, cf. Table 6). Kerndorff et al. (1991) proposed a model for the standardized semiquantitative assessment of ground water contamination by waste sites using an index which is based on normalized ranks of toxicant occurrence, concentrations and toxicity.

Substance-specific quantitative analyses of risks from local soil contamination have been made (Table 6) by detailed but basically simple exposure models. More extensive health risk assessments have been made e.g. for chromium (Golden and Karch 1989) and for PCDD/PCDF compounds (Paustenbach 1989, cf. IV). Exposure analyses have utilized empirical data and models of soil ingestion (Stanek and Calabrese 1995), dermal absorption (McKone 1990), dust inhalation (Steele et al. 1990) and household water use (McKone and Bogen 1992).

Table 5. Empirical health studies of waste sites and of other kinds of contaminated sites (main sources: Andelman and Underhill 1987, Marsh and Caplan 1987, National Research Council 1991).

Site/state, problems and risk factors ¹	Study methodologies, data and exposure inference	Findings and conclusions of health effects and of potential causes
industrial waste site/Alabama, DDT runoff	clinical examination, chemical analysis	serum DDT level correlated with ambient DDT level
Hyde Park fill/NY (New York), organochloride pollution	employee questionnaire, clinical tests, exposure surrogate	significant hiatal hernia increase in site vicinity compared to control
Jackson landfill/NJ (New Jersey), ground water pollution	questionnaire, chemical analysis	complaints of skin symptoms, causes not ascertained
Gems (Camden) landfill/NJ, ground water pollution	questionnaire, respiration tests	complaints of respiratory damage, causes not ascertained
Price (Atlantic) landfill/NJ, ground water pollution	questionnaire, chemical analysis	reports of various symptoms, reliability and causes unclear
Love Canal chemical dump/NY, mixed air, water & soil pollution	cancer register examination; child length & weight measurement	weight and length decrease correlate with exposure estimates
Lorain chemical dump/Ohio, air pollution	questionnaire, census register	no apparent mortality increase
Hardeman landfill/Tennessee, CCl ₄ , ground water	questionnaire, clinical and chemical analyses, exposure estimation	no certainty of effects and causes; liver damage suspected
Woburn dump/Massachusetts, ground water pollution	questionnaire, cancer/pregnancy registers, chemical analyses, exposure surrogate	leukemia incidence related to estimated exposure
Drake Superfund site/Pennsylvania	cancer register analysis, questionnaire, exposure surrogate (area of residence)	bladder cancer mortality increase, reported skin and sleep problems
Mixed-waste landfill, Hamilton/Canada	retrospective case-control self-reports, exposure estimate by residence & work	correlation between exposure and various reported symptoms
Lowell/Massachusetts contaminated site	cross-sectional questionnaire of symptoms, exposure by residence	increased prevalence of minor symptoms
Stringfellow/California, industrial waste dump	questionnaire, medical records, demographic surveys	rate and prevalence increase in several (reported) disorders
Dauphin county/Pennsylvania TCE _c contamination of wells	case-control questionnaire, exposure surrogate by residence	increased reported prevalence of eye, gut and sleep problems
contaminated aquifer/Arizona, chlorinated solvents	retrospective case-control study, exposure estimates by residence	association between parent exposure and congenital heart disease
various contaminated sites ² , New York	congenital malformation register, exposure by residence proximity	association between mother's proximity to site and defect incidence
hazardous waste site/Germany, various pollutants ³	blood analysis of case and control groups	decrease in blood quality, increase in Hg and creatinine
4 waste sites/California, various (volatile) toxicants ⁴	questionnaire, clinical examination, exposure estimation by proximity	elevated level of symptom reports in typical profiles (neurol.) near sites
12 disposal sites/USA ⁵ , emissions of PCBs	census data evaluation, serum exposure measurements	at 2 sites higher serum levels in high-risk persons, causes unclear
Kärkölä sawmill/Finland chlorophenols, ground water ⁶	questionnaire, chemical analysis, clinical tests, registers	RR ⁷ =1.1-74 for non-Hodgkins dis. in those exposed to contamination
1281 hazardous waste sites on National Priorities List/USA ⁸	survey data on low birth weight (LBW) and maternal proximity to site	no association between LBW and site proximity (RR ⁶ = 0.99)
2 Superfund sites/Texas, various volatile contaminants ⁹	questionnaire and clinical examination (cross-sectional) of case and control area	clusters of symptom reports in nearby area; causes uncertain
Industrial waste site/France, volatile organic toxicants ¹⁰	case-control study with questionnaires and clinical examinations	elevated prevalence of symptoms

¹For abbreviations, see Table 4; ²Stehr-Green et al. 1988; ³Kruse and Wieben 1989; ⁴Neutra et al. 1991; ⁵Geschwind et al. 1992; ⁶Lampi et al. 1992; ⁷RR = Risk Ratio; ⁸Sosniak et al. 1994; ⁹Dayal et al. 1995; ¹⁰Deloraine et al. 1995.

Table 6. Quantitative assessments of environmental exposures and health risks of waste site toxicants.

Study or information source	Site type(s) ¹	Substance(s) assessed ²	Risks addressed	General methodology ³
Mauskopf & Curtis-Powell 1985	unregulated landfills (solute emissions)	model chemicals	environmental health (carcinogenic & acute)	release estimation & EA, life table & LMS D-RA
Fingleton & al. 1986	hazardous waste land disposal	various (unspecified)	environmental health	uncertainty analysis of transport & D-R model
Hwang 1986	aquifer contamination by waste sites	various	environmental health (carcinog. & acute)	3-D groundwater model, simple EA, LMS D-RA
Bogen & Spear 1987	aquifer contamination (drinking water)	various	carcinogenic	variation & uncertainty analysis (Bayesian)
Whelan & al. 1987	inactive waste sites (chemical & radioactive)	various (As, Sr-90)	environmental health	detailed EA model for remedy prioritization
Brown & Donnelly 1988	waste disposal sites	various	environmental health (carcinog. & acute)	exposure approximation, LMS & RfD based D-RA
Brett & al. 1989	hazardous waste site soil contamination	various volatile org.	environmental health, of excavation options	indicator selection, LMS & ADI based D-RA
Burmester & Stackelberg 1989	waste disposal sites	PAHs etc.	environmental health (carcinog. & acute)	PDF generation for linear functions in EA & D-RA
Byard 1989	aquifer contamination	111-TCE _a	environmental health (carcinog. & chronic)	multipath EA, NOAEL based D-RA (determin.)
Heida et al. 1989	waste disposal site	TCDD	environmental health (carcinogenic)	multipath EA, LMS estim. of potency (determinist.)
Hertzman & al. 1989	contaminated soil site/ groundwater contam.	B(a)P/ benzene	environmental health	simple determinist. EA in reverse mode
LaGoy & al. 1989	mining waste site soil & indoor dust	Pb (As, Cd)	environmental health chronic (& carcinog.)	soil/blood Pb model, regular EA, LMS D-RA
Marshall & al. 1989	contaminated industrial area	various organic, As	environmental health (carcinog. & acute)	indicator selection, EA, LMS based D-RA
Maxim 1989	contaminated soil sites	PCBs	environmental health (carcinog. & chronic)	exposure & potency scenarios (detailed)
Murphy & al. 1989	metal ore smelter site soil contamination	As	carcinogenic	multipath EA, LMS estim. of potency (determinist.)
Reichard & Evans 1989	aquifer contamination	As	environmental health (carcinog. & chronic)	detailed EA, D-RA for monitoring optimization
Sheehan & al. 1991	soil contamination (residential)	Cr ^{III} , Cr ^{VI}	environmental health (carcinog. & acute)	multipath EA, standard D-R models (determin.)
Chilton and Chilton 1992	Municipal mixed-waste landfills	various	carcinogenic (mainly ground water)	RCRA simple determin. EA & LMS D-RA
McKone & Bogen 1992	aquifer contamination	TeCE _c	environmental health (ambient & home)	multipath EA, detailed D-R & uncertainty anal.
Copeland & al. 1993	soil contamination at wood preservation site	PCDD/Fs	environmental health (carcinogenic)	multipath EA & D-RA with uncertainty anal.
Finley & al. 1993	contamination of potable groundwater	chloroform, TeCE _c etc.	carcinogenic	multipath EA with PDF based uncertainty anal.
Donnelly & al. 1995	soil contamination at wood impregnation site	PAHs, PCDD/Fs	ecotoxicol. (mutagen.) and human cancer	biomarker-based and EA for ingestion route

¹Assessments of risks from aquatic and solid media are mainly included; ²For abbreviations, see Table 4 (additional symbols: Sr-90 = strontium-90, PAHs = polycyclic aromatic hydrocarbons, B(a)P = benzo(a)pyrene); ³Abbreviations: LMS = linear multistage (model of carcinogenesis), RfD = reference dose, D-R(A) = dose-response (analysis), EA = Exposure analysis, ADI = Acceptable daily intake, NOAEL = No observed adverse effect level, 3-D = 3-dimensional, PDF = probability density function; ⁴In some cases, modified equations or examples. Symbols or abbreviations: R = (individual) risk, c₀ = concentration (in route i), b = potency, R_c = lifetime excess cancer risk, D = dose, HI = hazard index, CDI = chronic daily intake, I₀ = intake

Table 6. (continued).

Mathematical formul. of dose/risk ¹	Cofactors included	Treatment of uncertainties ⁵	Empirical data base	Risk estimates ⁶ (range/Max)	PDFs ⁷ used in analysis
$I_{EC}=I_{CR} \cdot c \cdot b_1 \cdot f_T$	dilution, mobility, persistence (semiquant.)	modular uncert. anal. system	some (leachate initial conc.)	$R_c \leq 6 \cdot 10^{-3}$ (bladder tumor)	assumed (simple)
$p(D)=1-\exp(-(b_0+b_1 D+...+b_{k-1} D^{k-1}))$	dispersivities etc. transport variables	Monte Carlo anal., use of 90 % UCL	fair	- (only b_1 and VSD computed)	assumed LN,N,Uni
$R_c=D \cdot b_1$ $R \sim c \cdot A \cdot D^{-1}$	3-D transport in plume (no attenuation)	-	fair (selected case data)	$R_c=0.028-1$	-
$R=p(U,v)$	interindividual variation	by analysis of Bayesian p's	some (ground-water)	$R_{50}=10^{-6}-10^{-5}$ $R_{Max}=10^{-3}-10^{-3}$	Poisson
$R_c=1-e^{-Db}$ $HI=CDI \cdot R \cdot fD^{-1}$	transport and exposure variables & parameters	-	-	- (relative)	-
unexplicated (R=c·b implied)	-	(in potency estimation)	extensive (heterog.)	' R_c ' < 10^{-3} (toxicant-spec.)	-
$CDI=f_C \cdot f_A \cdot f_E \cdot t_L^{-1} \cdot m_B^{-1}$ $R=CDI \cdot b$	emissions & absorpt. exposure durat., age	(mean & max. dose estimates)	some (not stated)	$R_c=2.8 \cdot 10^{-3}$, ADD ADI ⁻¹ =20	-
$R_c=c \cdot CR \cdot f_A \cdot b_1 \cdot m_B^{-1}$ $HI=c \cdot CR \cdot f_A \cdot m_B^{-1} \cdot R \cdot fD^{-1}$	-	by Monte Carlo simulation	some (soil)	$R_c < 10^{-4}$	LN,N etc. (assumed)
$D=c_i \cdot CR_i \cdot m_B^{-1} \cdot f_A$	absorption	safety margins	limited	not applicable (NOEL based)	-
$R_j=\Sigma(D_{ij} \cdot b_1(i))$ (for route j)	bioaccumulation, intake rates	-	rather extensive	$R_c=1.6 \cdot 10^{-5}$	-
unexplicated (NOAEL based linear model)	exposure scenarios, bioconc., gas diffusion	range estimation	some	not applicable (preset safe c)	-
unexplicated (comparison of CDI and Rfd)	intake, absorption (soil, dust)	(range estimation)	fair (for Cd)	$R_c=3 \cdot 10^{-6}$	-
unexplicated (routine LMS model)	dilution, retardation etc. transport factors, popul.	-	fair (soil, groundwater)	$R_c=8 \cdot 10^{-3}$	-
unexplicated	degradation, bioavail., bioassay interpretation	conserv./plausible scenarios	limited (reference)	$R_c=3.7 \cdot 10^{-9}$ (plausible)	-
$D=\Sigma c_j \cdot CR_j \cdot b$ $\approx \Sigma c_j \cdot CR_j \cdot m_B^{-1} \cdot b_1$	ingested fraction, ventilation etc.	-	modest	$R_c=3.6 \cdot 10^{-6}$ (all j indoors)	-
$R_p=n \cdot R=n \cdot b \cdot d$ $\approx n \cdot b_1 \cdot c \cdot CR \cdot t \cdot m_B^{-1}$	potency extrapolation, population size	model/param., σ_{inX}	limited	$3.5 \cdot 10^{-1} \cdot a^{-1}$ excess cancers	mainly LN
$R=CDI \cdot b$ $=\Sigma c_i \cdot CR_i \cdot f_E \cdot f_C \cdot t \cdot m_B^{-1} \cdot b$	age groups, exposure scenarios, bioavail.	MLEI&MEI risks, MOS=Rfd/ADD	limited (topsoil)	$R_c \approx 10^{-8}-10^{-9}$ MOS $\approx 10-1000$	-
various (unspecified)	leachate c, dist. to well, site size, aquifer, infiltr.	MEI risk estim. (within 1 mile)	variable (leachate)	$R_{c60}=10^{-10}$ $R_{c95}=10^{-4}-10^{-5}$	-
various linear (standard form)	transfer coefficients, several cofactors	PDFs generated by Monte Carlo	fair		various LN,N,Uni,Tri
various linear (standard form)	path-specific exposure factors, LMS versions	PDFs generated by Monte Carlo	fairly good (for soil)	$R_{c50}=2 \cdot 10^{-8}$ $R_{c95}=1 \cdot 10^{-7}$	various LN,Uni,Tri
various linear (standard form)	transfer and exposure factors	PDFs generated by Monte Carlo	fair	$R_c=10^{-4}-10^{-6}$	various LN,N,Uni,Tri
unexplicated (mutag.) standard for R_c (Db)	bioavail. & toxicokinet. partly incl. in markers	-	limited (2 samples)	$R_c \approx 10^{-2}-10^{-3}$ (direct ingest.)	-

of carrier (in route i), f_C = fraction of contaminated mass, f_A = absorbed fraction, f_E = fraction of exposed time, t_L = life length, m_B = body weight, b_1 = relative potency by LMS model, $CR_{(j)}$ = contact rate (of medium in exposure route j), R_p = population risk, N = exposed population, t = exposure duration, p = probability, U = vector of uncertain variables, v = vector of particular values of known variables, I_{EC} = excess incidence of cancer, I_{CR} = baseline incidence of cancer, f_T = time factor, NOAEL = No Observed Adverse Effect Level; ³UCL = upper confidence limit, σ = standard deviation, MLEI/MEI = most likely/maximally exposed individual, MOS = margin of safety, Rfd = reference dose; ⁶VSD = virtually safe dose; ⁷PDF = probability density function; LN = lognormal, N = normal, Uni = uniform, Tri = triangular.

Health risks from lead in waste sites and other contaminated sites have been assessed using relationships between environmental concentrations and body burdens and between body burdens and neurological disorders in children observed in epidemiological studies (Theelen and Nijhof 1995).

Quantitative uncertainty analysis has increased in health risk assessment of waste sites (Table 6). The U.S. EPA used the transport model EPASMOD to back-calculate a distribution of leachate concentrations, choosing the upper 90 % value as the basis for accept values (Fingleton et al. 1986). However, the estimate of potency was shown to introduce large errors. Many investigators have used and further developed such approaches, generating PDFs for exposure variables (Table 6, see also Broshears 1986, McKone and Ryan 1989, Medina et al. 1989, Smith and Charbeneau 1989). These models are dependant on definitions of the operative factors and of their functional relationships and on assumptions of distributions. Uncertainties in impacts have been analysed more seldom than those in exposure factors, and attempts to validate estimates of ranges or distributions of exposures and risks have rarely been made.

1.3.2.2 Ecotoxicological risk analyses

The application of ecological risk analysis to chemically contaminated sites is still relatively undeveloped (e.g., U.S. EPA 1989b, Suter 1993). However, investigations of the ecotoxicological effects of waste sites have increased (Table 7, see also Menzie et al. 1992 and 1993, Scarano and Woltering 1993). Ecotoxicological risks are usually expressed as the probability in a population of exceeding a certain level of a specified end point; individual risks are thus not as relevant as in human risk analysis (Kylä-Harukka-Ruonala 1989, Suter 1993).

The responses of organisms to emissions from contaminated sites have been measured mainly in the laboratory by acute lethality tests of single aquatic animal species, such as the common water flea *Daphnia magna* (V, Table 7). Increasingly, other effects and selections of species have been studied. The comparability and interpretation of these data are limited due e.g. to variation in site and test factors.

Recently, laboratory toxicity tests have been complemented by studies of the occurrence and function of species present at the site (Table 7). Also bioaccumulation of waste site toxicants in food webs has been studied (e.g., Fordham and Reagan 1990, Menzie et al. 1992).

Ecological risks of waste site toxicants are additionally evaluated by approaches similar to those in human health risk analysis on the basis of observed or simulated toxicant concentrations (Zamuda 1989a).

The distributions of exposure factors and of response variables have seldom been evaluated in these site investigations (e.g., Cardwell et al. 1993). Specification of end point and formulation of models is often insufficient (Suter 1993). The multi-invertebrate test of Perrodin (1988) used a lognormally distributed PDF linking exposure to waste site leachates and response.

Acceptable concentrations of toxicants have been based also on ecotoxicological effects, assuming that the sensitivity of organisms is distributed loglogistically; e.g., Van Straalen and Denneman (1989) used the relationship between the share of protected species and concentrations to estimate safe soil concentrations.

Comparisons of ecotoxicological and health risks have seldom been attempted in studies of waste sites. Suter (1993) noted that for some sites contaminated heavily by PCDD/PCDFs, no human health effects were demonstrated, while severe effects on animals were observed. Combined ecotoxicological and health risk analysis has recently been attempted by use of biochemical markers (Donnelly et al. 1995).

1.3.3 Summarizing evaluation

In Finland, few studies of the occurrence, impacts and risks caused by toxic substances in waste sites have been made. The properties and implications of toxicant distributions have rarely been analysed.

In many other countries, locally significant contamination has been simulated or verified around many waste sites, sometimes also in exposure media. Indications of impacts and of environmental conditions conducive to toxicant dispersal have also been identified.

Table 7. Empirical studies of ecotoxicological effects of waste sites (in part from V).

Studied sites/materials	Studied effects	Studied taxa	Results/max. toxicity ^{1,2}
Waste disposal sites/ leachates ³	acute lethality, tissues, enzymes	<i>Salmo gairdneri</i>	96h-LC ₅₀ = 6.5 %; interrenal nuclei damage in 0.5 %/7 d
Landfill and lysimeter/ leachates ⁴	acute lethality, residual O ₂ TLV ⁵	<i>Salmo gairdneri</i>	96h-LC ₅₀ = 6 % (fill), 0.5 % (wood), 0.06 % (lysimeter)
Solid waste landfills/ leachates ⁶	acute lethality	<i>Daphnia pulex</i> , <i>Oncorhynchus sp.</i>	48h-LC ₅₀ ^D = 4.1 % 96h-LC ₅₀ ^O = 3.2 %
Municipal mixed-waste landfills/runoff ⁷	acute lethality	<i>Daphnia magna</i>	48h-EC ₅₀ = 11 %
Solid waste landfills/ leachates ⁸	acute toxicity, chlorophyll-a content	<i>Daphnia m.</i> , <i>Selenastrum</i> <i>c.</i> , <i>Photobacterium sp.</i> ,	48h-EC ₅₀ ^D = 62 % EC ₅₀ ^S = 10 %, EC ₅₀ ^P = 14 %
Hazardous waste site/ soil, leachate ⁹	acute toxicity, germination tests	<i>Daphnia m.</i> , <i>Photobacter</i> , algae, lettuce, earthworms	EC ₅₀ ^{algae} = 0.4 %, EC ₅₀ ^D = 3.3 %
Contaminated soil site/ soil, leachate ¹⁰	acute toxicity, root elongation	<i>Daphnia m.</i> , <i>Photobacter</i> , algae, cultivated plants	EC ₅₀ ^{algae} = 0.002 %, EC ₅₀ ^D = 0.003 %
Landfill, ash & slag/ leachates ¹¹	acute lethality, Ames mutagenicity	<i>Daphnia m.</i> , <i>Salmonella</i> <i>typhimurium</i>	24h-EC ₅₀ = 2.3 % (landfill), 0.022 % (ash leachate)
Industrial waste/ leachates ¹²	acute toxicity	<i>Daphnia m.</i> , <i>Scenedesmus</i> <i>sp.</i> , <i>Photobacterium sp.</i>	EC ₅₀ ^P = 1.5 %, EC ₅₀ ^S = 2 %, EC ₅₀ ^D = 9 %
Solid waste landfill/ leachates ¹³	acute toxicity	aquatic invertebrates (<i>Lymnaeidae</i> , <i>Lumbricidae</i>)	LC ₅₀ = 17 %, LC ₁₀ = 2.9 %
Creosote site/ sediments ¹⁴	acute toxicity, germination & rooting	<i>Daphnia</i> , <i>Photobacter</i> , let- tuce, <i>Selenastrum</i> , <i>Eisenia</i>	EC ₅₀ ^D = 0.2 %, EC ₅₀ ^S = 0.6 %
Municipal landfills/ leachates ¹⁵	acute toxicity	<i>Photobacter</i> , <i>Ceriodaphnia</i> , <i>Lemna minor</i>	14-56 % light ext., LC ₅₀ ^C = 11-54 %, LC ₅₀ ^C = 34-100 %
Hazardous waste and slag/extracts ¹⁶	mutagenicity	<i>Salmonella t.</i> strain TA98, <i>Aspergillus nidulans</i>	mutag. ^{TA98} = 9 600 net rev. l ⁻¹ , mutation freq. ^A = 3.4 10 ⁻⁵
Mixed waste site/ leachate ¹⁷	acute & chronic toxic., genotox., bioaccumul.	<i>Salmo gairdneri</i> , <i>Daphnia</i> <i>p.</i> , algae, <i>Photobacter</i> etc.	96h-LC ₅₀ ^S = 2 %, ^D = 4 % 8d-EC ₅₀ ^{algae} = 0.4 %
Waste disposal sites/ leachates ¹⁸	acute toxicity	<i>Daphnia m.</i> , algae, fish	EC ₂₀ ^D = 0.05 %
Pesticide disposal site/ soil extracts ¹⁹	acute (developmental) toxicity, mutagenic.	<i>Oryzias latipes</i> , <i>Salmonella typhimurium</i>	72h-LC ₅₀ = 2.2 % (larvae), embryo inhibition at 10 %
Landfill, creosote sites/ soil, leachate ²⁰	acute toxicity	<i>Daphnia m.</i> , <i>Eisenia sp.</i> <i>Photobacterium sp.</i>	EC ₅₀ ^P = 5 %, LC ₅₀ ^E < 10 %, EC ₅₀ ^D = 62 %
Waste disposal sites/ soil ²¹	acute & chronic toxic., bioaccum. (lab, field)	<i>Eisenia sp.</i> , <i>Lumbricus t.</i> , other (popul., bioaccum.)	14d-LC ₅₀ ^E = 10 % 0 % survival in 7 d
Hazardous waste site/ leachates ²²	acute & chronic toxic.	<i>Daphnia</i> , <i>Photobacter</i> , <i>Scen-</i> <i>enedesmus</i> , <i>Brachydanio</i> etc.	EC ₅₀ ^P = 1.6 %, EC ₅₀ ^S = 2.4 %, EC ₅₀ ^B = 17 %, EC ₅₀ ^D = 15 %
Industrial (metal, paint) waste leachates ²³	acute & chronic toxic., genotoxicity	<i>Photobacter</i> , <i>Daphnia m.</i> , <i>Raphidocelis</i> , <i>Salmonella</i>	24h-EC ₅₀ ^D = 0.6 - > 100 %, 28d-EC ₅₀ ^D = 0.2 - > 100 %
Wood impregnation site/ soil leachates ²⁴	genotoxicity/ mutagenicity	<i>Salmonella t.</i> TA98, <i>Escherichia coli</i>	<470 net rev. of <i>Salmonella</i> with 20 mg ml ⁻¹ dose (+S9) ²⁵

¹Superscripts denote toxicities to the taxa with the same letter in family name; ²EC₅₀ and LC₅₀ = effective and lethal concentration to 50 % of test animals, respectively; ³McBride et al. 1979; ⁴Cameron and Koch 1980; ⁵O₂ TLV = oxygen threshold limit value; ⁶Atwater et al. 1983; ⁷Nuorteva et al. 1983; ⁸Plotkin and Ram 1984; ⁹Miller et al. 1985; ¹⁰Thomas et al. 1986; ¹¹Mezzanotte et al. 1988; ¹²Deneuvy et al. 1988 (ref. Perrodin 1988), ¹³Perrodin 1988; ¹⁴Athey et al. 1989; ¹⁵Björklund 1989; ¹⁶Davol et al. 1989; ¹⁷Van Coillie et al. 1989; ¹⁸Hagendorf and Börner 1991; ¹⁹Marty et al. 1991; ²⁰Deschamps et al. 1993; ²¹Menzie et al. 1992 and 1993; ²²Hund and Traunspurger 1994; ²³Lambolez et al. 1994; ²⁴Donnelly et al. 1995; ²⁵With enzymatic activation.

The toxicological risks caused by waste sites and other contaminated sites have been quantitatively estimated mainly in connection with human carcinogens; the estimates have varied from levels near natural baseline risks to high levels which have usually been obtained by using several conservative assumptions. Toxic effects of emissions from waste sites on test organisms and also on local biota have been observed. The attributes and significance of these impacts have not been often analysed. Epidemiological studies have yielded little conclusive evidence of human health effects attributable to toxicants in waste sites, but have usually not excluded the possibility of such effects.

Methodologically, most of the quantitative assessments of health risks caused by waste sites have addressed carcinogenicity and been based on exposure analyses using simple deterministic (mainly linear) models. Uncertainties have been considered by the use of safety factors, scenarios and spread statistics, and increasingly also by analysis of probability distributions, usually using Monte Carlo methods for exposure estimation. Predictions made by such models have rarely been verified. Toxic effects of waste sites have been rarely investigated together with exposure measurements or formal risk analyses. Ecotoxicological risk analyses have mostly been limited to appraisals of laboratory biotests.

Despite increasing investigations, previous assessments have thus given insufficient information of the risks of waste sites. There is a pronounced need for improved risk assessment, considering the great resources spent on other parts of risk management. This requires the production and better use of empirical data, coupled with increased theoretical scrutiny of the processes operative in risk formation and with improved risk analysis methodologies.

2 PURPOSE, OBJECTIVES AND SCOPE OF THE PRESENT STUDIES

The general purpose of the summarized studies has been to investigate toxicants in waste sites and in their emissions, to measure their impacts and to assess the resultant toxicological risks to

the environment. To accomplish this, the empirical data and descriptive and comparative analyses in I - VI are completed by a synthesizing quantitative and qualitative assessment of their significance based on a structured risk analytical framework emphasizing uncertainties.

The specific objectives of the work contained in this thesis may be defined as follows:

1. to identify organic and inorganic toxicants in solid, liquid and gaseous media in waste sites, and to analyze the distribution and leachability of toxicants in wastes
2. to measure concentrations of toxicants in leachate runoff from waste sites, to examine the statistical properties of the distributions of the concentrations, and to estimate fluxes of toxicants in runoff
3. to measure the toxicity of leachates from waste sites, to characterize the concentration-response relationships in acute laboratory biotests with *Daphnia magna*, and to study the connections between leachate quality and toxicity
4. to identify chemicals and properties contributing to toxicological risks in ground water and soils impacted by waste sites
5. to quantify the spatial and isomer distributions of PCDD/PCDF compounds at chlorophenolic wood preservative waste sites
6. to devise and apply risk analytical models to prioritization of toxicants in waste site emissions, to site-aggregating analyses of toxicological risks, and to quantitative assessment of the risks of selected sites and substances, with particular reference to uncertainties.

This thesis deals with toxicological risks caused by chemicals in waste sites through emissions to the environment. Local soil contamination associated with waste sites is also treated (cf. 1.1).

Occupational health risks are dealt with at a general level only in connection with toxic substances in landfill gas. Physical and technical properties of waste sites which are related to toxicological risks have been studied to a limited extent. Catastrophic risks caused by fires etc. are not analysed explicitly and directly. Risks associated with methane or other main decomposition products of organic wastes, with pathogens or with radionuclides have not been analysed.

3 EMPIRICAL METHODS

3.1 Study sites

Waste sites for field studies were selected from all districts of Finland (Fig. 4). Candidate sites were compiled from landfill inventories, surveys of suspected hazardous waste sites and other background information (Assmuth et al. 1990b).

After field visits and evaluation of written, graphical (map) and oral information on site characteristics, 37 sites were selected for screening investigations. Six waste disposal sites were added at a later stage.

The selection criteria included risk-related factors such as estimated hazardous waste disposal and environmental conditions, factors related to the representativity of the sample (e.g. in terms of site owner and waste age and qualities), as well as practical and organizational considerations.

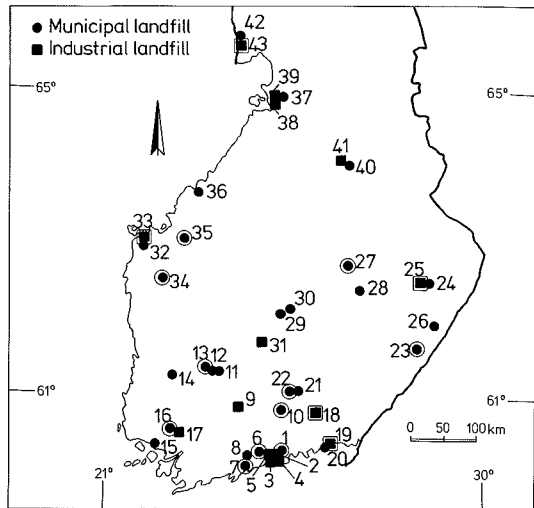


Fig. 4. Location of study sites (from I and III). Sites subject to investigation of ground water quality have been indicated.

Table 8. Basic characteristics of the 43 studied waste disposal sites¹ (modified after Assmuth 1992).

Characteristic (unit)	Min	\bar{x}^2	Max	Finnish average ³
Operated by municipality (% of sites)		65		84
Presently used for disposal (% of sites)		35		74
Use period (a)	7	23	80	16
Control of incoming wastes (at % of sites)		28		31
Leachate treatment system present ⁴ (at % of sites)		21		11
Fill area (10^4 m ²)	0.1	9.3	40	4.2
Readily permeable bottom soils ⁵ (at % of sites)		19 (37)		26
Distance to habitation (m)	50	400	1 300	1 000
Distance to extracted aquifer (m)	0	700	1 400	5 500

¹Evaluation of site properties mostly refer to the situation during the study period; ²Arithmetic mean; ³Statistics on municipal and industrial landfills from a national register of data from municipalities in 1983 (Seppänen 1986a); ⁴Other than leachate collection basins, dikes or uncontrolled soil infiltration; ⁵Laboratory-measured $K_c > 10^{-5}$ m s⁻¹ (in parentheses, percentage of sites with significant occurrence (> 50 % of site area) of gravel or sand, or gravelly or sandy silt, cf. Assmuth et al. 1990b).

The field study sites include municipal and industrial landfills of varying properties and environment in southern and central Finland (Table 8, I - III). Also other kinds of contaminated sites and their wastes were examined (IV, VI).

Most of the sites may be regarded as uncontrolled waste sites. They are estimated to contain a large proportion of the hazardous wastes accumulated in the respective area. Due to their com-

paratively great size, the standard of landfill technology is not below the (internationally seen low) average for the country (see I and Assmuth et al. 1990b).

After screening investigations, sites were selected for in-depth studies of toxicants and other properties in wastes (Assmuth 1992), leachate (I, VI), landfill gas (II), ground water (III) and sediment or soil (IV, VI).

3.2 Sampling and field methods

3.2.1 General principles

Site properties and emissions were investigated with a screening methodology. Simple measurements were initially used, emphasizing coverage instead of sensitivity. During in-depth studies, spatially or temporally more extensive sampling and measurements were used. The frequency of analysis varied between substances.

Sampling was focused by background information, maps, field surveys and screening measurements to obtain samples representative of toxicant distributions. Samples were also taken from background points unlikely to be impacted by the sites, especially of surface waters. Attention was paid to the quality of equipment and methods to diminish the unrepresentativity of samples (I - VI, Assmuth 1992).

3.2.2 Leachate runoff and surface water

Runoff from waste deposits was sampled in the screening investigations approximately two times per year from peripheral ditches and creeks which represent the dominant waterborne emission route. The average dilution factor, f_{dil} (the ratio of catchment area uncovered by waste to that covered by waste) was 3.8. At several sites more extensive sampling was done.

Sampling methods for leachate and surface waters are detailed in Table 9 and (I). The guidelines of National Board of Waters (1973) were followed. Flows were measured whenever possible by recommended methods, mostly 90° V-notch weirs (National Board of Waters 1984).

3.2.3 Ground water and perched water

Ground water was sampled at 16 sites mainly within 200 m in the flow direction from the wastes. Samples were collected from piezometers or wells which had been equipped with sieves extending vertically through a large part of the depth horizon of typically 5 m, usually to impermeable layers. The wells were designed on the basis of hydrogeologic information to ensure samples representative of the saturated zone (III). Whenever possible, the wells and piezometers were flushed after installation.

Samples were taken by bailers or by suction or electric pumps. Prior to sampling, several well volumes were removed except in soils of very low permeability. Silty samples were usually filtered for heavy metal analysis, and otherwise treated as surface water samples (Table 9). Samples of perched water in landfills were taken mainly from piezometers by similar equipment and procedures as with ground water samples.

3.2.4 Solid wastes, soils and sediments

The quality of disposed wastes was investigated at 8 sites. Sampling was focussed by information on site properties, by visual examination and by grids. Samples were taken from excavated pits or by drills using inert and cleaned equipment. Large particles were avoided when sampling. Combination and splitting of samples were frequently used to increase representativity (Assmuth 1992).

Topsoil samples were taken from ca. 20 sites for physical analysis by manual core samplers. For chemical analyses, soil and peat samples were taken by drilling or from trial pits with similar methods as in waste sampling. At one contaminated soil site, auger drilling was used.

Sediments in dikes and streams were sampled by hand shovels. The samples were contained in clean plastic or aluminium foil, and preserved deep-frozen or refrigerated, depending on the analyses (Table 9, IV).

3.2.5 Landfill gas

Gas samples were taken from four landfills by gas wells or piezometers which were sealed with lids and surrounded by compacted soil (Table 9). Samples for laboratory analysis were taken by suction pumps. Samples for field measurements were taken by direct reading instruments (II). The samples were collected in Tenax™-filled steel tubes and stored cold.

The integrity of samples in +4 °C and -20 °C was checked by subsequent analyses. Duplicate samples and field blanks were collected. Before sampling the equipment was flushed with sample gas. Field measurements were made of main and trace components using gas analyzers and Dräger™ tubes, mainly to ensure and improve the representativity of laboratory samples (II).

Table 9. Summary of sampling methods used for the principal laboratory analyses.

Analysis	Sample type	Sampling equipment and procedures	Sample amounts and containment	Storage and pretreatment
Main constituents	liquid solid	SS ¹ /TEF ² /HDPE ³ bailers/pumps, direct drilling, excavation or SS ¹ /PE ³ shovel	1 l HDPE ³ bottle ~5 l plastic bag/cylinder	+4 °C, dark +4 - +20 °C
Heavy metals	liquid solid	non-metal equipment, flushed (filter) non-metal equipment	0.12 l PPN ⁴ bottle 0.5 l PE ³ bag	+4 °C, pH 2 +4 °C
AOX/EOX ⁵	liquid solid	solvent-rinsed, flushed equipment drilling, excavation	1 l BSi ⁶ flask pure Al ⁷ foil	+4 °C, dark -20 °C
Mineral oils	liquid	solvent-rinsed, flushed flasks	1 l NaSi ⁸ flask, TEF ² septa	+4 °C, dark
Volatile organic compounds	liquid gaseous	solvent-rinsed, filled completely (or gastight injection vials; Al lid) suction pumps (~ 0.01 l s ⁻¹)	1 l gastight BSi ⁶ flask (20 ml NaSi ⁸ vial, TEF ²) Tenax TM in SS ¹ tube	+4 °C, dark (+4 °C, dark) +4 °C, dark
Non-volatile organics	liquid solid	solvent-rinsed, flushed equipment drilling, excavation	1 l BSi ⁶ flask, Al ⁷ +TEF ² pure Al ⁷ foil	+4 °C, dark -20 °C
<i>Daphnia</i> tests	liquid	samplers/direct closure	1 l HDPE ³ bottle	-20 °C
Ames tests	liquid	samplers/direct closure	2.5 l BSi ⁶ flask	+4 °C, dark

¹Stainless steel; ²TeflonTM or other related fluorohydrocarbons; ³(High Density) Polyethylene; ⁴Polypropylene; ⁵Adsorbable/extractable organohalides; ⁶Borosilicate (laboratory) glass; ⁷Aluminium; ⁸Sodium silicate (industrial) glass.

3.3 Laboratory treatment and analyses

3.3.1 Physical and chemical measurements

Basic physico-chemical analyses

Solid samples were analyzed for basic physical properties (grain size, moisture, ignition loss, K_c) using standard methods or established procedures at our soil laboratory (Fig. 5, III).

Basic physico-chemical analyses of liquid samples were made at the Water and Environment District laboratories by standard procedures with minor modifications (Table 10, Fig. 5).

Metals and other trace elements

Solid samples were dried at 105 °C, homogenized, sieved to < 0.5 mm with metal-free meshes, extracted generally with both 1M NH₄Ac (solid: liquid ratio 1:20, pH 4.5, 3 h) and strong HNO₃ (1:10, 1 h), and filtered (0.45 µm) (Fig. 5).

The samples or extracts were analyzed after filtration mainly by AAS using standard methods and quality assurance (Table 10, Fig. 5, I, III).

Organic compounds

The analyses of specific organic compounds were selected on the basis of AOX and GC screening, site information, previous research and pollutant rankings (I, III, Table 10). Most compounds were analysed after extraction with solvents, chosen to suit the sample type and analysis; hexane was mainly used. Gases were analysed after thermodesorption (II). Cleanup was made particularly for extracts of solid samples. Separation, detection and quantification were mainly made by GC using capillary columns and EC detection (Table 10).

The internal standard for chlorinated aliphatics and benzenes was regularly 1,2-dibromoethane, for unchlorinated hydrocarbons 1,2-dimethoxybenzene, for pesticides PeCBz and for chlorophenols 3,5-dichlorophenol. For PCBs, a mixture of Aroclor 1242 and 1268 and later a mixture of 9 - 19 pure isomers were used (Table 10).

PCDD/PCDF compounds were analyzed by sensitive isomer-specific methods after extensive cleanup and separation (Table 10). Identification and quantification was made using rigorous quality assurance procedures (IV, VI).

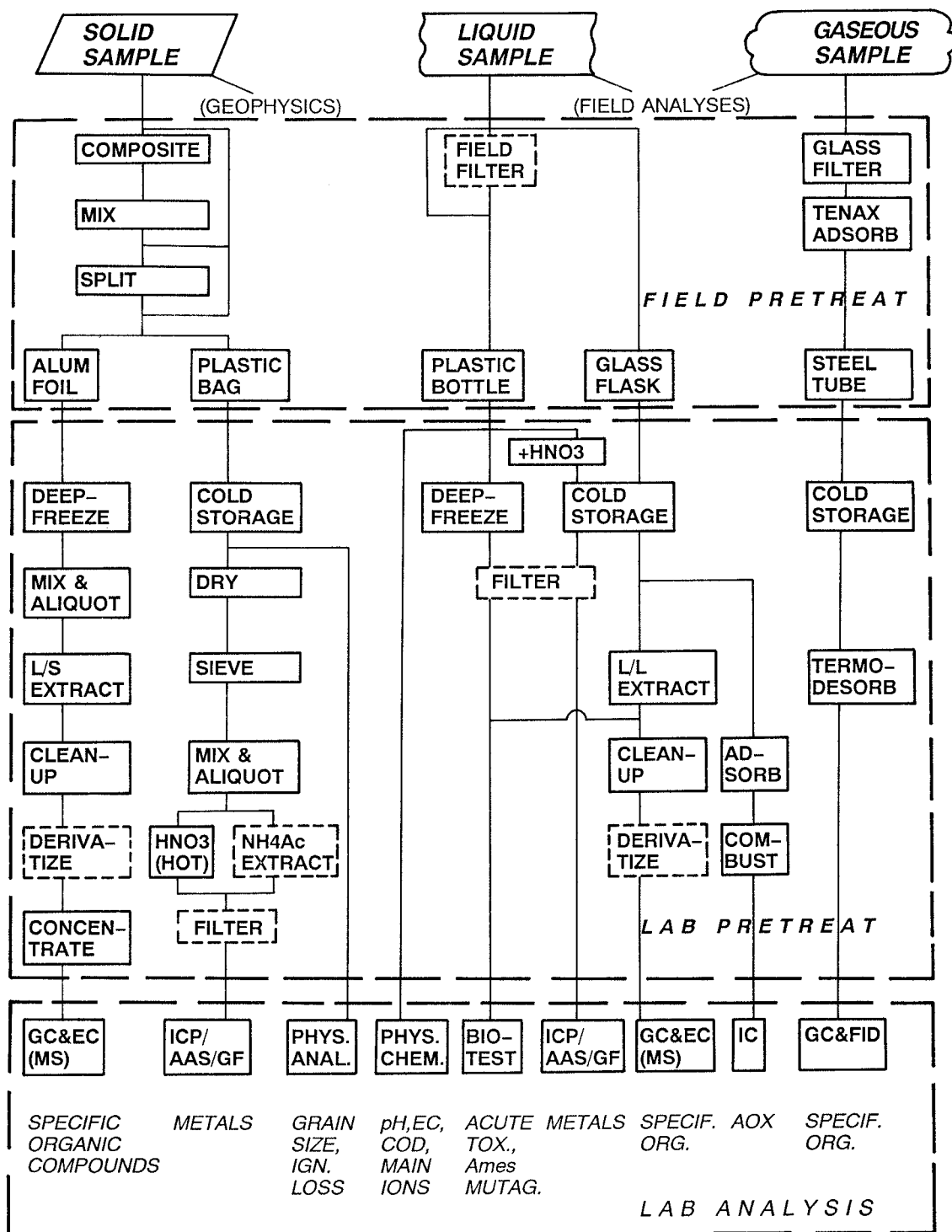


Fig. 5. Generalized scheme of the treatment and analysis of the various types of samples.

Table 10. Overview of main laboratory analysis methods. (For chemical abbreviations, see Table 4.)

Analysis	Separation, concentration and cleanup	Detection and quantitation	Detection limits (DL)			Reference methods
			Liquid ($\mu\text{g l}^{-1}$)	Solid ($\mu\text{g kg}^{-1}$)	Gas ($\mu\text{g l}^{-1}$)	
Main constituents	various	spectrometry etc.	varies			SFS ¹
Light metals	filtration, acid extraction	AAS ² (ICP ³)	30-100	1000		GSF ⁴ , FRI ⁵
Fe, Mn	filtration, acid extraction	AAS (ICP)	100	1000-10000		SFS, GSF
Zn	filtration, extractions	AAS (ICP)	20-45	1000		SFS, GSF
Pb	filtration, extractions	AAS, GF ⁶	0.01-6	1000		SFS, GSF
Cd	filtration, extractions	AAS, GF	0.01-6	10		SFS, GSF
Cr, Cu	filtration, extractions	AAS, GF (ICP)	1-50	100-1000		SFS, GSF
As	filtration, extractions	AAS, GF	0.1-1	100		SFS, GSF
Co	filtration, extractions	GF	6			SFS, GSF
Ni	filtration, extractions	GF	0.1-1	1000		SFS, GSF
Hg	wet digestion	cold vapor	0.003-0.05	20		SFS, WERI ⁷
Volatile chlorinated aliphatics	solvent extraction (headspace)	capillary GC ⁸ , ECD ¹⁰	0.05-10		0.1-0.5	NIPH ⁹ , WERI
Chlorobenzenes	hexane extraction	capillary GC, ECD	0.01-0.1	0.1	0.01	NIPH, WERI
Volatile aromatic compounds	hexane extraction	capillary GC, FID ¹¹	0.05-10	1	0.1	NIPH, WERI
Chlorophenols	ether extraction, asylation	capillary GC, ECD (GC/MS ¹²)	0.005-0.2	5		NIPH, WERI
Cresols	acidification, ether extraction, asylation	capillary GC, ECD	0.5		0.01	NIPH
Chlorinated pesticides	hexane (+EtAc ¹³ for solids) extraction	capillary GC, ECD (GC/MS)	0.01-0.1	~0.1		NIPH, WERI
PCBs ¹⁴	hexane (+EtAc) extract. (H ₂ SO ₄ purification)	capill. GC, ECD (GC/HRMS ¹⁵)	0.05 (0.005-0.01)	0.01-5		NIPH, WERI
PCDD/Fs	toluene/EtOH extract., SiO ₂ /H ₂ SO ₄ /AC ¹⁶ /Al	GC/HRMS, 16 ¹⁶ C standards	0.0000005	0.00005		NIPH, Lampi & al. 1992
Phthalates	Liquid/liquid extraction	capillary GC, FID	0.2-1			NBWE ¹⁷
AOX/EOX compounds	(toluene elution,) AC ¹⁹ & XAD-8 adsorpt.	950 °C, IC ¹⁸ (coulometry)	0.5-5	100		Vartiainen 1988
Acute toxicity to <i>D. magna</i>	(filtering, neutralization)	24 h & 48 h immobilization	6 % immob.			SFS 1984, ISO 1982
Mutagenicity to <i>S. typhimurium</i>	acidific., EtAc elution, DMSO ²⁰ concentration	gene disorder (with/without S9)				Vartiainen 1988

¹Finnish Standardization Association; ²Atomic Absorption Spectrometry; ³Inductively Coupled Plasma (atomic emission spectrometry); ⁴Geological Survey of Finland; ⁵Forestry Research Institute; ⁶Graphite Furnace; ⁷Water and Environmental Research Institute; ⁸Gas Chromatography; ⁹National Institute of Public Health; ¹⁰Electron Capture Detection; ¹¹Flame Ionization Detection; ¹²Gas chromatography-Mass spectrometry; ¹³Ethylene acetate; ¹⁴Initially Aroclors 1242 and 1268, then a selection of pure isomers (usually IUPAC numbers 8, 18, 28, 31, 52, 77, 80, 101, 105, 118, 126, 137, 138, 153, 156, 159, 169, 180, 181); ¹⁵High Resolution Mass Spectrometry; ¹⁶Activated Carbon; ¹⁷National Board of Waters and the Environment; ¹⁸Ion Chromatography; ¹⁹Activated Carbon; ²⁰Dimethylsulphoxide.

The internal standard for chlorinated aliphatics and benzenes was regularly 1,2-dibromoethane, for unchlorinated hydrocarbons 1,2-dimethoxybenzene, for pesticides PeCBz and for chlorophenols 3,5-dichlorophenol. For PCBs, a mixture of Aroclor 1242 and 1268 and later a mixture of 9 - 19 pure isomers were used (Table 10).

PCDD/PCDF compounds were analyzed by sensitive isomer-specific methods after extensive cleanup and separation (Table 10). Identification and quantification was made using rigorous quality assurance procedures (IV, VI).

3.3.2 Toxicity tests

Acute toxicity to *Daphnia* water flea

D. magna and their feed, *Selenastrum capricornutum* green alga, were cultivated by standard methods (mainly SFS 1984, cf. V). The day before testing, 40 females were prepared for breeding by placing them in dilution water prepared according to ISO (1982). The sensitivity of the *D. magna* was checked (V).

Twenty-four hour screening tests were made with neonate daphnids without dilutions. A control was made of dilution water only. Samples with oxygen deficit were discarded. The samples exhibiting a lethality > 10 % were tested by the standard 48 h procedure (SFS 1984) in triplicate 10 ml tubes with 5 neonates in each.

Mutagenicity to *Salmonella* bacterial strains

Bacterial mutagenicity and toxicity of acidified leachate samples to *S. typhimurium* strains TA97 and TA100 were assayed by the Ames test, with and without enzymatic activation by rat liver extract (Vartiainen 1988, Assmuth et al. 1990b).

3.4 Characteristics and treatment of empirical data

3.4.1 Representativity of measurements

Spatial representativity

The spatial representativity of sampling varied between sites and types of samples and measurements (Table 11).

The study sites include waste sites in various areas and environmental conditions, which are generally rather typical of Finland.

The surface water samples generally represent main leachate runoff route (depending on the hydrological characteristics of the sampling points and schemes). All subsurface fluxes were not covered, due to the heterogeneity of hydrogeological properties. Spatial representativity is particularly limited for wastes and other solid samples. Thus, risk analyses were based mainly on measurements of site emissions.

Temporal representativity

Temporal representativity is affected by the frequency and duration of measurements and by the temporal variation of the properties measured.

The frequency of measurements of toxicants in emissions was generally rather limited (Table 11). More frequent or aggregative measurements of leachate constituents were used to elucidate the temporal dimension of emissions (I).

Measurements of accumulated toxicants e.g. in soil and sediments increase the temporal coverage of the data. More frequent measurements of main constituents of leachate also serve as indicators of toxicants.

Although the studies did not cover the long time development of single sites, the selection of sites with different average age of waste, including many old sites, gives information on the temporal extension of site properties and risks.

Structural representativity

Emphasis was put on sites close to large towns. Thus, the median size of the sites were above Finnish average (Table 8). Still, most sites are small by international standards.

Various industrial waste sites were investigated, especially disposal sites of forest industries, including sawmills. The industrial sites do not represent all important kinds of industrial waste deposits, e.g. those of mining or metal industries.

Several study sites represent a common type of contamination, that caused at sawmills. They were included as cases which are principally comparable with waste sites. Some of the waste sites also include other types of contamination, e.g. from adjacent industry.

Table 11. Sources, assessment and treatment of unrepresentativity of the empirical methods.

Types of variation and unrepresentativity at the various levels 1.-4.				Evaluation of representativity	
1.	2.	3.	4.	Dgr ¹	Characteristics/improvement
Spatial	Intersite	Environmental setting	Regional Local	++ +/-	for special types modest (e.g., sites on aquifers)
Spatial	Intrasite	Leachate emission routes	Surface water Ground water Waste and soil	++ +/- +/-	questionable in some sites questionable; simultaneous leachate sampling improves
Spatial	Intrasite	Gas emission routes		-	
Spatial	Intrasite	Bioaccumulation routes		-	
Tempo- ral	Intersite	Timing/duration of emissions/cofactors	Waste age Long-time impacts	+ +	site selection improves site selection improves
Tempo- ral	Intrasite	Fluctuation of emissions	Leachate quality Gas quality	- -	frequent/solid samples improve
Struc- tural	Intersite	Waste and general site properties	Site category Size of sites	+ +	for special types not good ² bias to large/medium-sized
Struc- tural	Intersite/ intrasite	Measurements of chemical analytes	All media	-/+	some organics lacking; selection, screening and indicator measures improve
Struc- tural	Intersite/ intrasite	Measurements of other variables	Transport factors Exposure factors Sensitivity factors Response factors	+/- - -- +/-	estimable in water; by K_c ³ qual./semiquant. only for test population only for ca. one species only

¹Ordinal-scale estimate of the degree of representativity: ++ = good, + = fair, +/- = variable, - = modest, -- = poor;

²Industrial waste sites in particular; ³Saturated hydraulic conductivity.

The measurements of toxic responses included mainly assays of acute lethality to an aquatic invertebrate (Table 11). Assessments of other responses were made on the basis of extrapolation and of known effects of analyzed toxicants.

The focusing of measurements according to indications from previous measurements of the same properties or of related properties (by indicator or group analyses) increases the representativity of the empirical data.

3.4.2 Reliability and error

Measurement reliability involves accuracy and precision of the measurement systems, including field and laboratory operations. In addition to random error, systematic errors may be caused e.g. for volatile pollutants by losses.

The reliability of measurements was increased and controlled by several methods in the various parts and phases of study, including the following quality assurance procedures:

- inert and pure materials and equipment, and recommended methods in sample treatment chains

- replicate samples (especially for organic compounds in water samples)
- composite samples (especially for measurements of toxicants in solid samples)
- field blanks (especially for analyses of organic compounds)
- calibration of instruments and of measurement systems, and method tests
- high-quality and pure internal and external standards and reagents (cf. 3.3.1)
- laboratory blanks (for analyses of toxicants and toxicity, usually 1 blank per batch (day))
- replicate analyses within one measurement system (especially for GC analyses)
- confirmatory and parallel analyses with many measurement systems (especially for organics, e.g. by several GC columns and programs)
- standard laboratory methods and practices, and, with lacking standards, established quality assurance procedures of the analytical laboratories (cf. Table 10)
- checks of calculation and data management methods (independently by many persons at various stages).

Errors are introduced in the field particularly when sampling from ground water, due e.g. to sorption, volatilization, contamination and coprecipitation (cf. III). Such error sources could not be extensively quantified. However, analytical quality control was extended to field and trip blanks (of water samples). If results exceeding detection limits were obtained with these blanks, the data on the substances, sites and dates in question were omitted. Such cases occurred mainly with volatile chlorinated aliphatic compounds, notably DCM and 12-DCE_a.

For most trace elements, analytical error was typically within 20 %, based on replicate measurements, field blanks and intralaboratory comparisons during the present work and previous analyses at publicly supervised laboratories (Mäkinen 1991 and 1994). Accuracy and precision depended on the element, matrix, concentration, laboratory, and method. Errors are higher at low concentrations in heterogeneous mixtures and matrices such as waters containing abundant suspended solids and organic matter. The applicability of AAS analyses to leachates has been shown to be better for Cr, Ni and Zn than for other elements (Gallorini et al. 1993).

Errors in measurements of organic compounds may be higher, due e.g. to losses, reactions and

other disturbances. On the basis of quality control, measurement variation and error was estimated to occur particularly with some volatile chlorinated aliphatic compounds in low concentration ranges; these results may thus be largely considered semiquantitative. For unchlorinated hydrocarbons and for chloroaromatic compounds measurement error was less. For PCDD/PCDF compounds the within-laboratory analytical variation was remarkably small (VI).

Variation between replicate samples as a result of measurement system variability was usually smaller than the variation caused by the representativity of sampling. In measurements of soil concentrations of chlorophenols, the combined intralaboratory variation in the sample treatment and analysis stages, as characterized by replicates split in the field, was small in comparison with the interlaboratory variation particularly in the low concentration ranges (Fig. 6). This may have been largely due to different pretreatment (e.g., extraction) methods. The sampling variation was appreciable for CPs in soil particularly in high concentrations, as illustrated by comparative measurements of single and composite samples with the same treatment system (Table 12).

Pretreatment of the samples for toxicity tests (freezing, aeration and filtration for *Daphnia* tests, extraction for *Salmonella* tests) introduces analytical variation and errors which may not be extensively evaluated (V). Variation was present also in responses between the replicate *Daphnia* tests (V). Generally the lethality in the three replicates was very similar.

The overall maximum frequency of flow measurements of one week may introduce errors of over 100 % in fluxes due to the low representativity of top flows. Load estimation methods may cause systematic errors of 30 % and differences by a factor of two even in more frequent sampling (Godfrey et al. 1995). Additional errors in estimates of fluxes from small catchments are caused by temporal variation in the concentrations of sorbed substances in particular (Ekholm et al. 1995). Also the variations in flow regimes affect fluxes (Droppo and Jaskot 1995). In small flows the low measurement precision produces relative errors of ca. 100 % (Ettala 1988). However, small flows are not as crucial for flux estimates. At one site the flows were recorded continuously, allowing estimates of cumulative flows.

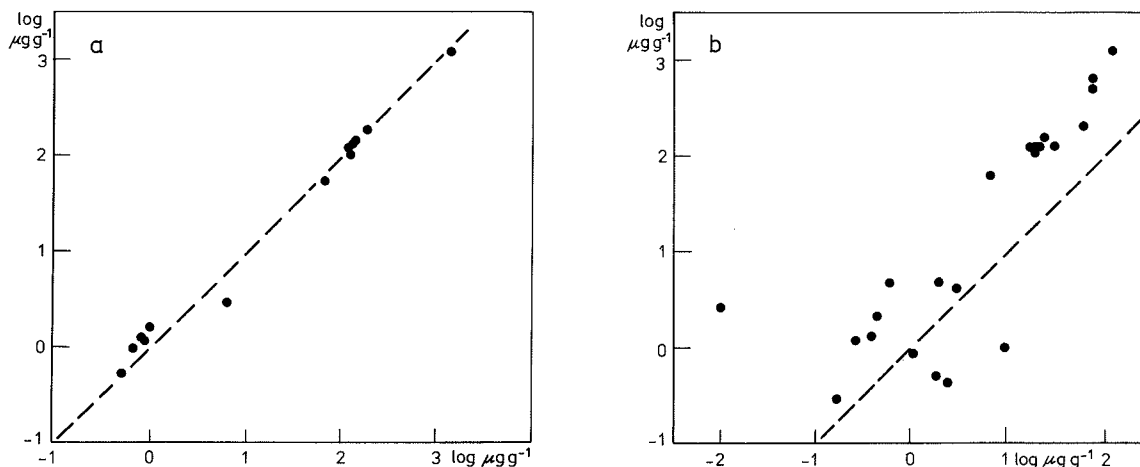


Fig. 6. Variation in pentachlorophenol concentrations in soil between replicate samples analysed in the same laboratory (a) and in two laboratories (b). 1:1 correlation is shown by hatched lines.

Table 12. Comparisons of the concentrations of some chlorophenols in single and composite soil samples¹.

Sampling transect and points	Concentration ($\mu\text{g g}^{-1}$ d.w. ²) of composite sample			Average concentration ($\mu\text{g g}^{-1}$ d.w. ²) in single samples		
	PeCP	2346-TeCP	246-TCP	PeCP	2346-TeCP	246-TCP
A1-6	2.1	4.1	0.59	1.9	3.7	0.38
B4-6	40	530	72	130	2 600	160

¹For abbreviations, see Table 4; ²In dry weight.

3.4.3 Statistical treatment

The computerized data were analyzed mainly by the SAS package (SAS Institute, Inc. 1991) after common statistical data management procedures.

In descriptive statistics, mainly linear univariate and multivariate methods were used.

In estimation and testing and in covariation analysis methods suited to the particular distributions of the variables or of their appropriate transformations were used, such as the non-parametric χ^2 test and rank correlation statistics.

The normality of distributions was generally tested by the Shapiro-Wilk test which is the appro-

priate default method for the sample sizes at hand (SAS Institute, Inc. 1991).

Regression analysis was done by the ordinary least squares method and by stepwise regression analysis using several alternative methods.

In order to derive a measure of toxicity on the basis of the response to increasing concentrations (of waste leachates), a procedure which is based on fitting a power function of test media concentrations and linear estimates of median survival time (MST) was used, as specified by the Finnish standard (V). Mutagenicity was calculated on the basis of the slope of the linear portion of the D-R function.

4 KEY EMPIRICAL RESULTS

4.1 Distributions of toxicants in wastes

4.1.1 Solid phase

Cr, Cu, Pb and Zn were the most abundant heavy metals in wastes (Table 13). The easily leached fractions of Pb and Zn were consistently largest, whereas Cr, Cu, As and Ni were more strongly bound in waste matrix (Fig. 7).

Several toxic organic compounds were measured at elevated concentrations, even if their median values were generally low. The variation in waste concentrations was consistently larger between sites than within sites (Table 13).

The spatial variation of toxicant concentrations in a municipal waste landfill was pronounced and irregular (see Fig. 3 in Assmuth 1992).

4.1.2 Liquid phase

In perched water in wastes several toxicants were also present, in variable median concentrations (Table 13, cf. Assmuth 1992).

The decline in average toxicant concentrations from the fractions in solid wastes to perched water and to leachate varied between substances and sites (Fig. 4 in Assmuth 1992, cf. I). The relative decline was greatest for metals.

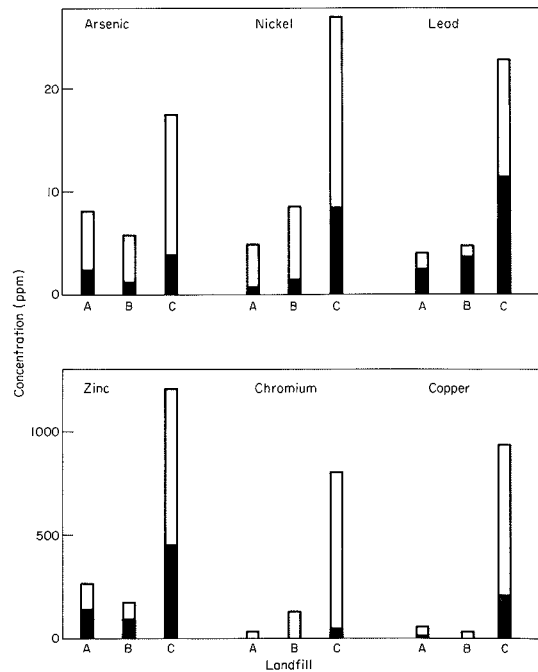


Fig. 7. The relationships between easily leached (NH_4Ac -extracted) concentrations (black bars) and total (HNO_3 -extracted) concentrations (whole bars) of some toxic elements in wastes from three municipal waste disposal sites (A - C) (from Assmuth 1992). All values of bar lengths are arithmetic means. (Cf. leachability indices in 4.5.1.)

Table 13. Statistics of toxicant concentrations in wastes and in perched water of municipal solid waste landfills (compiled from Assmuth 1992).

Substance ¹	Concentrations in solid wastes ($\mu\text{g g}^{-1}$ d.w. ² , HNO_3 -extractable)						Concentrations in perched (interstitial) water in waste ($\mu\text{g l}^{-1}$)		
	Range of site medians	Range of site means	Max	CV ³ (%)		n	Range of site medians	Max	n
				Total	Intra- site				
As	5 - 15	6 - 17	55	635	80	61	< 1 - 20	60	13
Cd	0.18 - 1.6	0.3 - 4.3	45	295	135	61	0.3 - 3.0	5	12
Cr	27 - 220	36 - 810	13 000	380	106	61	4.6 - 7.3	39	13
Cu	24 - 360	41 - 950	6 400	268	107	61	1.4 - 20	90	13
Pb	27 - 290	80 - 450	2 400	194	125	61	1.5 - 1.9	800	13
Zn	89 - 910	180 - 1 200	3 600	150	95	61	23 - 60	90	13
2346-TeCP	<0.005 - 0.6	0.007 - 0.95	3	232	79	24	0.11 - 5	47	8
PCBs	0.002 - 0.16	0.002 - 0.18	0.47			24			

¹For abbreviations, see Tables 4 and 12; ²In dry weight; ³CV = coefficient of variation = $\sigma \cdot \bar{x}^{-1} \cdot 100$ %, where σ = standard deviation and \bar{x} = arithmetic mean.

4.1.3 Gas phase

Skewness in the concentration distributions of gaseous compounds was not so pronounced as in solid or liquid phases (II). Truncation as a result of concentrations below detection level occurred with some relatively non-volatile analytes only.

Chlorinated methanes, ethanes and ethenes were the dominant organic trace compounds analyzed (II). Several aromatic hydrocarbons were commonly found in concentrations above their background levels as well.

Variation in concentrations between the sampling points and, particularly, between study sites was notable, whereas variation between duplicate samples was smaller (II). Significant covariation was observed between the dominant chlorinated trace gases (Fig. 3 in II).

4.2 Distributions of toxicants in the soil environment

4.2.1 Contaminated soil

High concentrations of As, Cu and Cr were found in the surface peat layers around the waste site of a plant where wood impregnation mixtures containing these elements have been produced (Fig. 36 in Assmuth et al. 1990a). The areal variation in peat, e.g. in relation to leachate dispersal routes, was noticeable.

The spatial distribution of chlorophenols in soil and debris at a sawmill contaminated by wood preservatives revealed heavily contaminated 'hot spots' in the vicinity of the wood preservation treatment area (Fig. 8). They extended to the relatively impermeable silt layers.

High concentrations of PCDD/PCDF compounds could be found in both surface soil and subsoil at this site, with considerable spatial variation. Highly chlorinated dibenzofurans (123478-HxCDF, 1234678-HpCDF and OCDF) were the dominant isomers (VI). The latter two were the most abundant isomers also in sediments at the waste site of the wood preservative plant (IV). No simple relationships were observed between the concentrations of PCDD/PCDFs and chlorophenols in sawmill soil (VI).

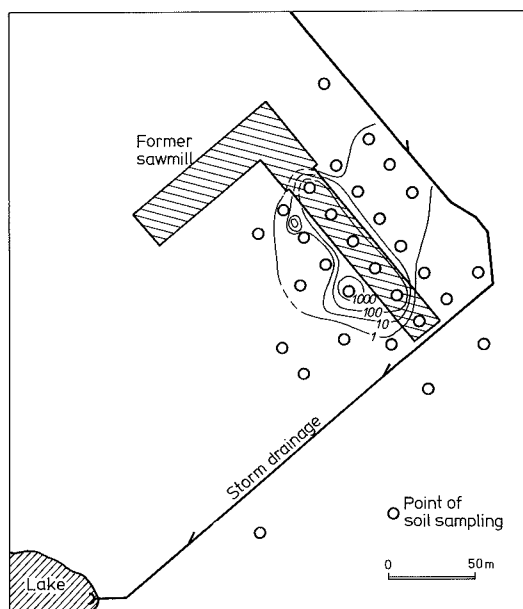


Fig. 8. Isophleths of pentachlorophenol concentrations (in $\mu\text{g g}^{-1}$ dry weight) in surface soil and debris at an abandoned sawmill.

4.2.2 Ground water

In nearfield ground water down gradient of the study sites, the concentrations of heavy metals and some organic toxicants, e.g. chlorophenols, frequently exceeded detection limits. Also some organochloropesticides were found in ground water samples. The dominance of small values was typical for most toxicants, causing skewed and truncated distributions and medians below detection limits (see Table III in III, toluene and As in Fig. 4 in III).

The median and maximum concentrations for ca. $\frac{1}{4}$ of the analytes were greater than those in leachate runoff; Pb and PCBs had both greater medians and maxima in ground water (III, VII). For Pb the concentrations may be affected by silt and by sampling structures, for PCBs by samples from a single well next to waste. Generally, quantitative comparisons are difficult due to methods of sampling and analysis.

The probability distributions of saturated hydraulic conductivity (K_s) in surface soils were asymmetric and displayed considerable within-site and between-site variation (III).

4.3 Chemical quality of leachate runoff

4.3.1 Main constituents

The most abundant of the measured leachate constituents were ammonium nitrogen, chlorides and alkali and earth alkali metals (Table 14, I). Their medians and means were generally higher at municipal landfills than at industrial sites (χ^2 , $p > 95\%$) (Assmuth et al. 1990b).

4.3.2 Toxic trace substances

The concentrations of many toxicants in leachate runoff were dominated by low values, causing skewed distributions (cf. Sokal and Rohlf 1981). Values below detection limits were common particularly with organic compounds even in leachate discharge points.

The infrequent detection produced truncated concentration distributions (Table 14, I, VII). Some of the statistics in Table 14 differ from those in (I) and (VII) due to the inclusion of additional data and due to the exclusion of unimpacted sampling points, of points without surface runoff and of data (for metals) below the higher DLs in initial screening analyses.

Variation in toxicant concentrations was regularly large, as reflected in high values of coefficient of variation, a tentative measure of variation because of the non-normality of concentration distributions (Table 14).

Normality in the distributions of the logarithmic transformations of concentrations was encountered with a few toxicants and data subsets (Table 14). In addition, the distribution of the concentrations of many toxicants on a logarithmic scale appeared to be composed of two linear sections of differing steepness (Fig. 9).

Table 14. Distribution properties of concentrations of selected toxicants in leachate runoff data (in part from VII, cf. I). Non-detects have been substituted by 0.5 detection limits (DL) or, with low-sensitivity measurements (of elements), by missing values. In parentheses, numbers of observations and maximum values of data including low-resolution plasma emission measurements (for elements).

Substance ¹	General properties		Statistics of location of concentration distribution ($\mu\text{g l}^{-1}$)			Attributes of distribution spread and form		
	n	f_{Detn} (%)	f_{50}	f_{90}	Max	CV (%)	SK ²	Fit to theor. ³
Cl ⁻	266	100	67 000	330 000	1 800 000	140	3.1	
NH-N ₄	279	100	2 800	110 000	340 000	190	2.8	
As	103 (221)	72	4.0	16	760 (760)	380	7.1	
Cd	101 (278)	59	0.25	0.80	12 (70)	510	11	
Cr	128 (275)	98	8	24	65 (1 400)	590	7.5	
Ni	98 (158)	100	12	43	83 (90)	93	2.3	(LogN)
Pb	102 (278)	85	1.0	7.2	15 (125)	190	4.6	
Zn	217 (293)	94	90	1 600	34 000 (110 000)	310	7.4	(LogN)
DCM	68	57	3.6	140	4 800	500	6.9	
12-DCE _s	78	47	< 1.5	110	670	290	5.0	
Toluene	81	72	0.80	70	1 500	380	5.3	
12-DCBz	77	29	< 0.05	2.0	58	560	8.5	LogN
HCBz	71	13	< 0.025	0.10	2.7	330	4.6	LogN
2346-TeCP	52	77	0.074	1.5	5.6	220	3.0	(LogN)
PeCP	52	88	0.057	0.50	3.0	240	5.4	
Dieldrin	54	9.3	< 0.05	0.061	1.1	250	6.9	(LogN)
Lindane	52	29	< 0.025	0.40	15	550	7.1	(LogN)
44'-DDD	52	27	< 0.025	0.27	2.2	270	4.5	(LogN)
PCBs	50	44	< 0.025	1.5	7.6	220	3.5	LogN

¹Cf. Table 4; ²Skewness; ³Probability of significance of the test statistic $p(r < W) > 0.25$, under the null hypothesis that the variable (its logarithm) is distributed normally (only data > DLs are included, decreasing n values from those given elsewhere). (In parentheses, variables which fit to theoretical distributions only depending on data selection and non-detection substitution.)

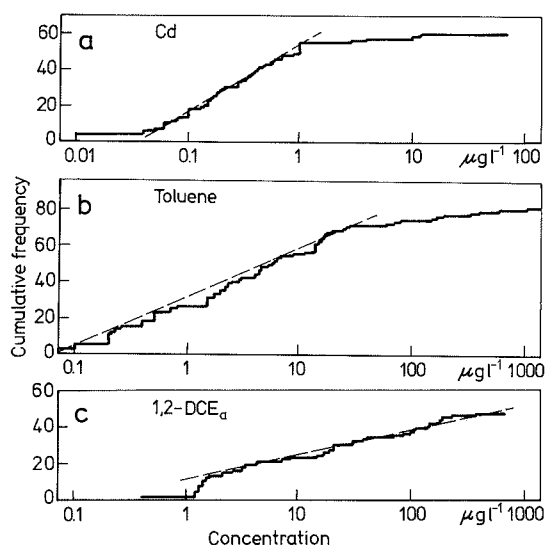


Fig. 9. Cumulative distribution functions of the concentrations of some toxicants in waste site leachate runoff. Note the logarithmic scales for concentrations. Frequencies are in %. Linear portions of the curves according to graphical evaluation have been shown by hatched lines.

There were statistically significant covariations between the concentrations of many toxicants and especially between main constituents, as measured by linear and, particularly, rank correlations (I).

Due to the variation introduced by sampling focus, the concentration distributions in aggregated data are strongly affected by those of individual sites. Site-averaged statistics were therefore generally used in the analysis of distributions (I).

Relatively few statistically significant differences in trace substance concentration statistics were found between municipal and industrial landfills (I, Assmuth et al. 1990b).

Due to the skewness of the distributions of most trace elements, their mean values often exceeded the median values. Of analysed heavy metals, Zn was the most abundant, while Cd, As and Pb were most often undetected (Table 14). The infrequently detected elements exhibited particularly pronounced differences in the rank order of concentration statistics between subsets of data which resulted from different methods of truncation and between various methods of non-detect treatment (Table 15, cf. Table 14 and I).

Of the analysed toxic organic trace substances, those most frequently detected in leachate runoff included chlorinated aliphatic hydrocarbons, simple unchlorinated monoaromatic hydrocarbons (toluene, xylenes, ethylbenzene) as well as chlorophenols and cresols (Table 14, I, VII). The measurements of chlorinated volatile aliphatic compounds in particular are semiquantitative. Due to the common truncation and skewness in distributions, median values better illustrate their locations than arithmetic means do. Medians were for many compounds at background levels (below detection limit).

Table 15. Distribution statistics of the concentrations of highly toxic elements in leachate runoff data subsets with different forms of substitution of non-detects (values below detection limits). (Cf. I.)

Element ¹	Median concentrations ($\mu\text{g l}^{-1}$)								Mean concentrations ($\mu\text{g l}^{-1}$)							
	All data, non-detects substituted by				Data with low DL ² , non-detect substit. by				All data, non-detects substituted by				Data with low DL ² , non-detect substit. by			
	0	½DL	DL	miss.	0	½DL	DL	miss.	0	½DL	DL	miss.	0	½DL	DL	miss.
As	4.0	4.0	9.5	15	4.0	4.0	4.0	7.0	14	15	20	24	15	15	16	21
Cd	0.06	0.25	0.50	0.24	0	0.28	0.50	0.28	0.51	2.1	1.5	2.2	0.46	0.56	0.66	0.80
Cr	8.0	8.0	8.0	8.1	3.3	11	11	12	76	120	120	120	11	11	11	11
Hg	0	0.03	0.05	0.03	0	0.20	0.20	0.20	0.07	0.16	0.16	0.16	0.02	0.04	0.06	0.07
Pb	0.25	2.5	2.5	4.0	1.0	1.0	1.0	1.3	5.2	9.1	9.1	10	2.2	2.3	2.3	2.6

¹For abbreviations, see Table 4; ²Data subsets which result in median values exceeding DLs (detection limits) by a factor of approximately 5.

4.4 Measures of toxicity and bioaccumulation

4.4.1 Acute lethality to *Daphnia magna*

The lethality in 24 h screening tests without dilutions was used as a screening measure of toxicity. Its distribution was two-peaked, concentrated in high or no toxicity (Fig. 1 in V).

The standard calculated EC(I)50 values sometimes exceeded 100 %. Apart from such high values, the frequency distribution of 48h-EC(I)50 was approximately normal, as reflected in the central statistics (Table 1 in V). Medians were close to means, and 48 h toxicity values were consistently higher than 24 h values.

The appreciable aggregate variation between samples (Table 1 in V) includes interindividual and population variation (cf. Forbes 1993).

Covariation was observed between screening and standard test results. The observations were concentrated in the high/high or low/low toxicity regions (V, cf. 4.5.2).

Additional information on toxicity was obtained by the methods of ISO (1982) and ASTM (1984), where EC(I)50 is determined by linear regression fit to final lethality, presented on a probit scale, and to test concentration, measured on a logarithmic scale (cf. Stephan 1977). Variations in the location and form (e.g., steepness) of the D-R curve and in the spread of the effect of exposure duration (i.e., temporal development of toxicity) were evident (Fig. 10). In many cases a lognormal response was implied by D-R curves approaching linearity at least in a (central) part of the curve, while in other cases sub-linear curves were observed (sustained toxicity with increasing dilution).

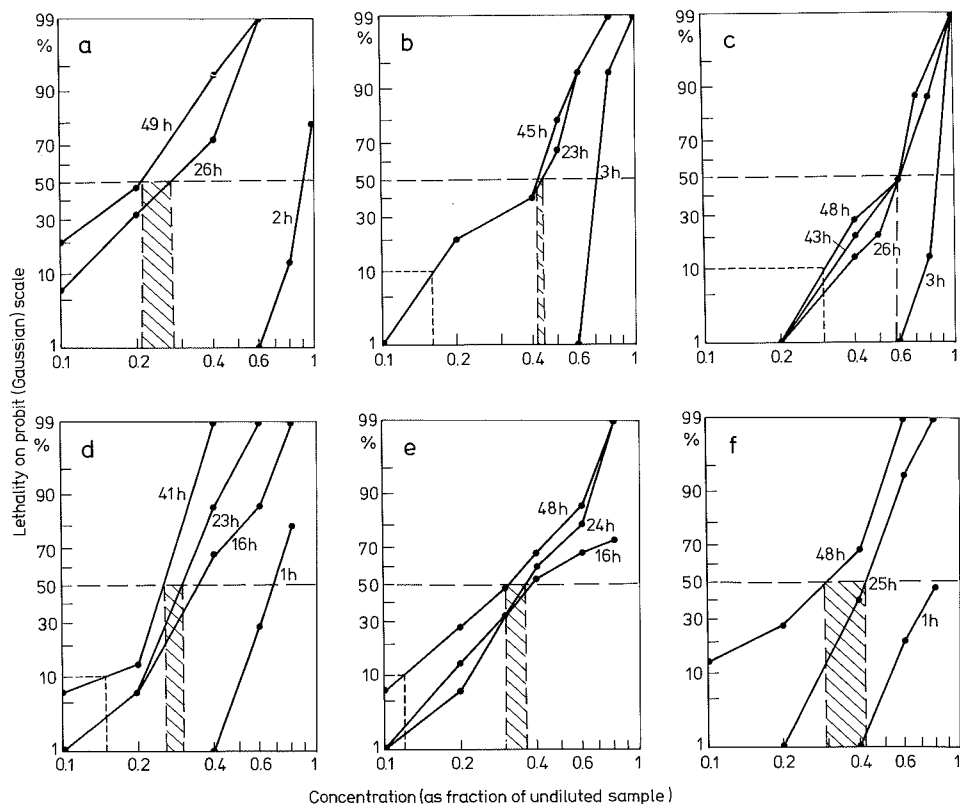


Fig. 10. Logprobit dose-response curves observed after variable exposure times in toxicity tests of leachates with *Daphnia*. a) and b) are from the same sampling point at two different dates; b) and c) are from the discharge point and from a downstream point, respectively; c) is impacted by a new waste disposal site, b) is impacted also by an old waste fill containing e.g. chlorophenols. Note the logarithmic scales of concentration and the normal (Gaussian) distribution based scales of lethality.

4.4.2 Genotoxicity to *Salmonella* strains

The mutagenicity of leachate to *Salmonella typhimurium* (after enzymatic activation) was 140 - 2 900 net revertants l⁻¹ for strain TA97 and 20 - 450 for TA100 (Assmuth et al. 1990b). The responses of the two strains were not correlated; their ratio varied from 1.5 to over 60.

The distribution of the mutagenicity of TA97 seemed more symmetric (median 930, arithmetic mean 1 000 net rev. l⁻¹) than that of TA100 (median 60, mean 120 net rev. l⁻¹).

4.4.3 Bioaccumulation of PCDD/PCDFs in fish downstream a chlorophenolic waste site

Accumulation of PCDD/PCDFs in roach (*Rutilus rutilus*) and pike (*Esox lucius*) from the recipient of the waste site of chlorophenolic wood preservative production was investigated on the basis of samples ca. 0.5 km downstream from the site.

The concentrations of PCDD/PCDFs in fish were slightly above those in the upstream lake and in sediment (Table 16).

Table 16. Mean concentrations of PCDD/PCDFs in fish and sediment downstream of the waste site of a chlorophenolic wood preservative plant (IV, VI, unpublished data by Assmuth and Vartiainen).

Species	Downstream samples			Upstream (background) samples		
	n ¹	conc. (pg I-TE g ⁻¹ l.w. ²)		n	conc. (pg I-TE g ⁻¹ l.w.)	
		PCDD/PCDFs	PCBs ⁴		PCDD/PCDFs	PCBs ⁴
Roach	2·3	20	19	2·3	13	10
Pike	2·3	52		2·3	49	
Sediment	3	17 000 ³				

¹Amount of samples from different points or age classes; ²Lipid weight; ³Dry weight; ⁴TEFs by Safe (e.g., 1992).

4.5 Relationships between measures of toxicants, site properties and toxicity

4.5.1 Leachability and waste properties

The leachability of element *i* in wastes was defined as the relation of the easily leached fraction to total concentrations (cf. Fig. 7):

$$LI_i = c_i(i) \cdot c_{tot}(i)^{-1} \quad (1)$$

LI_{Cu} in particular was explained by the pH and organic content of wastes (Table 17).

No statistically significant relationships were discerned between general landfill characteristics and site-averaged toxicant concentrations in waste (χ^2 tests, $p < 0.01$, Assmuth et al. 1990b). In single cases the origins and general quality of wastes were reflected in concentrations of toxicants e.g. in sites containing wood preservatives, solvents and PCDD/PCDFs (Assmuth 1992, IV).

Table 17. Measures and models of leachability indices of metals in a closed municipal waste site.

Element	Leachability, LI (%)		Best determinants of the natural logarithm of LI ¹					
	\bar{x}	CV	1st	r ²	2 first ones	r ²	3 first ones	r ²
Cd	41	48	ln(OM)	0.44	ln(OM), (H ⁺) ²	0.54		
Cu	12	116	ln(OM)	0.67	ln(OM), (H ⁺)	0.73	ln(OM), (H ⁺), (H ⁺) ²	0.77
Ni	32	51	(H ⁺)	0.31				
Pb	47	38	(H ⁺)	0.58				
Zn	35	34	(H ⁺)	0.38				

¹Significant at a level of ≤ 0.15 in stepwise regression model; The organic matter content (OM) is approximated by ignition loss; hydrogen ion concentration (H⁺) is calculated from the pH, measured in water extract after elution with distilled water (1:10).

4.5.2 Emissions and general site conditions

General landfill characteristics explained little of the variation in toxicant concentrations in pooled runoff data, but in individual sites relationships could be observed between toxicant concentrations and site features (Assmuth et al. 1991).

The fluxes of toxicants from study sites in leachate runoff (F_i) were estimated by various methods of non-detect substitution, flow estimation and background subtraction. F_i of a site was calculated as the sum of fluxes in the main discharge routes. These fluxes were flow-weighted mean concentrations in a sampling point multiplied by the catchment area of the point (Assmuth et al. 1990b) and the average yearly runoff in reference areas (Leppäjärvi 1990, 1991, 1992, 1993, cf. Ekholm et al. 1995).

The resulting tentative estimates are site-attributable fluxes, being dominated by near-field discharge points.

Volume-specific fluxes of a toxicant, $F_v(i)$, were calculated by dividing F_i by the waste volume of the site, V_w :

$$F_v(i) = F_i \cdot V_w^{-1} \quad (2)$$

The F_v estimates were averaged over points in time, sampling points, and sites (Table 18).

Variation in estimates of F_v was considerable. Most specific risk fluxes were greater at municipal than at industrial sites, with the exception of e.g. As which is affected by fluxes from a pesticide waste site. Some specific fluxes were also greater from landfills of high technical status, which are on the average large sites (Table 18).

Table 18. Estimates of fluxes and specific fluxes of some toxicants in leachate runoff from study sites¹.

Substance ²	Volume-specific F_v ($\text{mg m}^{-3} \text{a}^{-1}$) ³		Correlation of site characteristics with F_v ⁴			
	\bar{x} range	Max	technical status	size	age	industrial waste input
As	0.055 - 0.2	0.97				
Cr	0.79 - 0.92	20	+			-
Ni	0.76 - 0.86	15	-	-	-	-
Pb	0.089 - 0.68	1.7		-	-	
Zn	2.9 - 4.3	67				
PeCP	0.0084 - 0.0011	0.15	+			-
PCBs	0.022 - 0.077	0.52	+			-

¹Based on arithmetic means for downstream sampling points and for sites, and on substitution of non-detects by 0, 0.5 DL or DL; ²For abbreviations, see Table 4; ³Per total waste volume; ⁴Sign of correlation significant at 95 % level; technical status (on scale 0 - 2) defined on the basis of main structural and functional properties (Table 8, Assmuth et al. 1990b), size by area and volume of waste; proportion of industrial waste (dummy variable) by owner class = private.

4.5.3 Chemical quality and toxicity of leachates

There were significant correlations between toxicity and chemical indicators of leachate (I). Screening test lethality correlated with more chemical variables than did 48h-EC(I)50 (V).

Although 24h-lethality explained a relatively large share of the variation in standard test response, the model could be improved by inclusion of chemical variables, especially chlorides

(Fig. 3 in V). The best fit ($r = 0.77^{***}$) with these variables in stepwise regression was (V):

$$48\text{h-EC(I)50} = -0.005 (24\text{h-leth})^2 - 0.46 (\ln \text{Cl})^2 + 100 \quad (3)$$

The best ($r = 0.98^{***}$) stepwise linear regression with toxicant concentrations (in $\mu\text{g l}^{-1}$) was:

$$48\text{h-EC(I)50} = 170 (\text{Zn}) + 3200 (\text{Hg}) - 52 \quad (4)$$

5 RISK ANALYSIS

5.1 Model formulation

5.1.1 Conceptual framework

General

The standard definition for the risk (R) of an adverse event (E) is a function of its probability (p) and severity or magnitude (M) (e.g., Merkhofer 1987). It may be defined for individuals and populations; the latter particularly introduce magnitude dimensions (cf. Cox and Ricci 1989).

Often risk is viewed as the outcome of a sequence of events, with associated (conditional) probabilities (Marin et al. 1989). For environmental risks of waste site toxicants, the sequence involves source releases, transport and transformation, and exposures and responses of living receptors (Fig. 11).

The transport routes of toxicants are divided to surface and ground water runoff, air, solids and biota. The exposure routes include ingestion, inhalation and dermal uptake (Fig. 12).

Risks are further expressed relative to background risks (R_0) in a system (cf. Fig. 11). The risk due to a specified cause is the attributable risk (cf. Seiler and Scott 1987).

Based on this general conceptualization of the risk formation and of toxicant transport between the physical compartments of the process (Fig. 12), route-specific process models of risk formation are devised (e.g., Fig. 13). The processes are affected by environmental factors, properties of toxicants and receptors, and technology. Instead of a compartmentalized source and target, they can be black boxes.

Applying these concepts to the classic definition by Paracelsus, the toxicological risk (R_i) of

an adverse (toxic) impact (I) of substance i is a probability of impact conditional on dose, and a function of its effective dose (D_i^E) and potency relative to the impact (b_i):

$$R_i = p(I | D) = f(D_i^E, b_i) \quad (5)$$

Probability is implicit also in the function, since b_i denotes the probability of toxic impact or effect (e.g., tumors) in a population.

Omitting toxicokinetics, D_i^E may be expressed as a function (e.g., linear product) of administered dose D_i and a bioavailability factor f^{BA} .

The total attributable risk of an impact I to a receptor from exposure to toxicants is then a function of the sum of the risk estimates for each toxicant (i), route of transport (j) and exposure (k) minus corresponding background risk (cf. integral by Göttinger 1984):

$$R_{ijk} = f(\sum \sum \sum D_{ijk}^E, f_{ijk}^{BA}, b_{ik}) - R_{ijk}^0 \quad (6)$$

The toxicological risk of a site to its environment is correspondingly the probability that its emissions cause I , and is also a function of the sums over the doses it causes and of their potencies, minus background risk for I at the site.

The risk of a waste site is further expressed by a function of the fluxes and toxicities of emitted substances, assuming the former are proportional to long-term exposures:

$$R_n = f(F_{in}, b_{ik}) \quad (7)$$

Ecotoxicological risks of waste site emissions are analysed as functions of observed toxic impacts, which are approximated by effective doses (ED values). When laboratory organisms are used as a surrogate model of real world, they are extrapolated to other organisms, impacts and exposure situations.

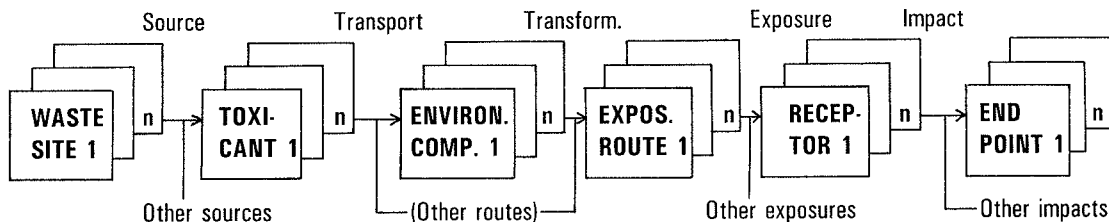


Fig. 11. General model of the main stages and entities in the formation of risks of waste site toxicants.

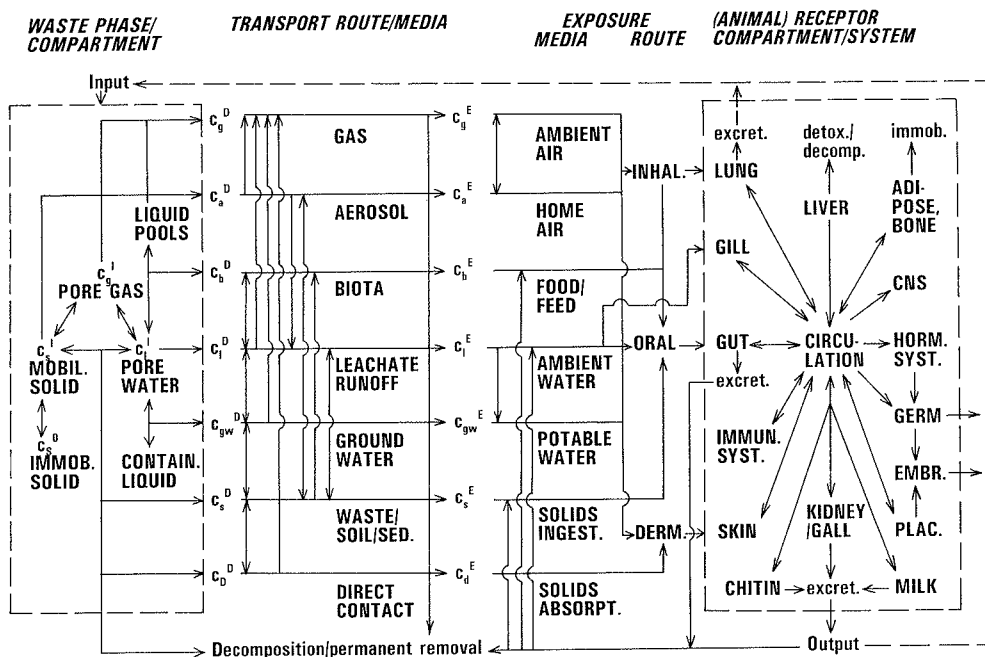


Fig. 12. Conceptual model of the distribution and transport routes of waste site toxicants between compartments of waste, environment and receptor.

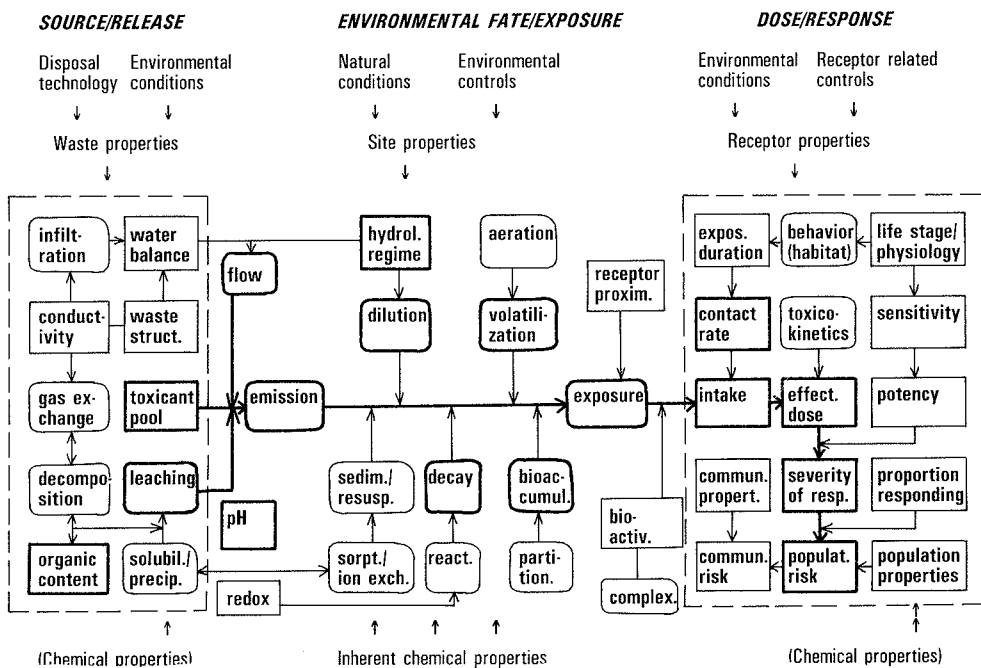


Fig. 13. Sequential model of risk formation for the transport route surface water runoff. Processes are marked with circles and properties with rectangles, and uni- and bidirectional influences with arcs. The entities and influences subject to measurement or estimation are indicated with bold lines.

Impact based models are implemented by biotests of toxicant mixtures (V). As a first approximation, the risk to aquatic animals is modeled by acute lethality to *Daphnia magna*.

Substance based analyses are combined with impact based analyses by examining the relationships between predicted and observed acute toxicity of leachate runoff to *Daphnia*.

The conceptual models are realized by a staged methodology proceeding from risk identification based on indices to quantitative toxicological risk analysis and to risk characterization. In these realizations, deterministic models are used as approximations. Model uncertainties are analysed by examining the influence of model assumptions and parameters. Measurement uncertainty in risk variables is accounted for by analysing the variation in distributions.

5.1.2 Toxicant sources and mobility

The pool of toxicants in a waste site is the product of their mean total concentration (c_{tot}) and waste volume (V_w). The leachable pool is the NH_4Ac -extractable fraction (Eq. 1, Fig. 13).

The time until the toxicant pool has been released (t^R) is estimated from assuming constant yearly flux (F) and no decay:

$$t^R = c_{tot} \cdot V_w \cdot F^{-1} = c \cdot F_v^{-1} \quad (8)$$

The t^R of the mobilizable pool is estimated from easily leachable fractions:

$$t_{mob}^R = c_l \cdot V_w \cdot F^{-1} = c_l \cdot F_v^{-1} \quad (9)$$

5.1.3 Environmental fate and exposure

Volatilization is considered an irreversible sink, modeled by coefficients of Henry's law (H) (VIII, cf. Thomas 1990).

Decomposition is treated as a sink by first-order aerobic decay constants (k_D , Schnoor et al. 1987) or aerobic aquatic half-lives ($t_{1/2}$, Scow 1990), assumed to be related as (cf. VII):

$$K_D = \ln 2 \cdot (t_{1/2})^{-1} \quad (10)$$

Bioconcentration of organic compounds is modeled by bioconcentration factors (BCF) assuming linear uptake and no depuration (cf. Farrington 1991). With lacking data, BCF is obtained from K_{ow} by regression models for groups of (non-ionic) compounds (Bysshe 1990, VII):

$$\log BCF_{fish/wat} = 0.76 \cdot \log K_{ow} - 0.23 \quad (11)$$

Assuming equilibrium partitioning (Di Toro et al. 1991), $BCF_{fish/sed}$ is expressed as the ratio of fillet to sediment concentration (VI).

Exposure to a toxicant (i) is quantified as the sum (D_i) of the doses (D_{ij}) through each route of exposure j (U.S. EPA 1988, 1989a, 1991):

$$D_i = \sum D_{ij} = \sum (c_{ij} \cdot CR_j \cdot m_B^{-1} \cdot f_T) \quad (12)$$

where

c_{ij} = ambient concentration of i in path j

CR_j = contact rate for medium in path j

m_B = body weight of receptor

f_T = time factor = (exposure frequency · exposure duration · averaging time).

Assuming constant exposure and no concentration-modifying factors, c_{ij} may be substituted by mean exposure concentration, c_{ij}^E , which includes f_T ; a duration factor f_D may be added. A bioavailability factor f^{BA} is also included. Chronic daily intake (CDI) then becomes:

$$CDI_i = \sum c_{ij}^E \cdot CR_j \cdot f^{BA}_{ij} \cdot m_B^{-1} \quad (13)$$

Due to variation in CDI, it is estimated in various exposure scenarios, e.g. for likely and maximally exposed individuals.

In ecotoxicological risk analyses, exposure is assumed to take place readily near the waste site. Sinks in volatilization and decay are included in indices of ecotoxic effects (VII).

5.1.4 Toxic impacts

Human health effects

For carcinogenic risk analysis the multistage model is used. Assuming D is small, as usual in environmental applications, and background incidence of the tumor is also small, the linearized form is used as a default (Crump 1984):

$$R_c \approx b_1 \cdot D \quad (14)$$

where

R_c = lifetime excess cancer risk (for a particular tumor type and tissue)

b_1 = tumorigenic potency (1st order term of the multistage model, or slope of D-R curve, inherently including a probability, Eq. (5).

The following assumptions of models and choices of data are made in the estimation of b_1 :

- Carcinogens acting orally or by inhalation exposure are treated separately (Pease 1990)
- of b_1 values based on animal tests, 95 % UCL estimates by the LMS model (b_1^*) are used
- otherwise, values for the most sensitive species, strain, sex and tumor site are used.
- interspecies dose conversion is based on body surface areas as by Anderson et al. (1983).

The use of b_1 values implies a non-threshold D-R model. As additional conservative assumptions, all tumors are taken to be malignant and incurable. Background incidence rates are included in potency. Assuming dose additivity, carcinogenic risks R_c are expressed as the sum:

$$R_c = \sum (D_i \cdot b_1^*(i)) \quad (15)$$

Estimates of genotoxicity are based on the data of IARC (1987) on lowest genotoxic doses, originally normalized as $5 + \log D^{-1}$ ($\text{ppm}^{-1} \text{d}^{-1}$). For comparability $\log D^{-1}$ values are used (Lewis 1991b). Reproductive and developmental toxicity is assessed by the lowest toxic dose reported by Lewis (1991b). These and other health risks (R^h_i) are modeled by the probability that CDI exceeds ADI (U.S. EPA 1989a, Lewis 1992, Lu 1995):

$$R^h_i = p(\text{CDI} > \text{ADI}) \sim a \cdot (\text{CDI} \cdot \text{ADI}^{-1}) \quad (16a)$$

$$\text{ADI} = (\text{LOAEL or NOAEL}) \sum \text{SF} \quad (16b)$$

where, in addition to symbols defined earlier,

ADI = acceptable daily intake

LOAEL = lowest observed adverse effect level

NOAEL = no observed adverse effects level

SF = safety factor for extrapolation between species, individuals, test durations and LOAEL and NOAEL.

Ecotoxicological effects

A basic measure of the ecotoxicological risk of substance i (R^e_i) is obtained by the probability that its observed concentration (or statistic of concentration distribution) exceeds the LOAEC for the exposure route j :

$$R^e_i \sim p(c_{ij} > \text{LOAEC}_{ij}) \quad (17)$$

Lowest LOAEC for all species (relevant to the route) is used. NOAECs of a hypothetical most sensitive species are estimated by extrapolation from the distributions of LC50s and LOAECs. Observed concentrations are also compared with no-effect concentrations (PNEC) derived from acute and chronic LC and LOAEC data by assessment (safety) factors (OECD 1995).

Substance-based risk estimates are summed to measures of all analytes assuming additivity of impacts (cf. Logan and Wilson 1995).

For structurally related substances, equivalent quantities are calculated (Safe 1992, VI). For mixture effects, concentration additivity based on simple similar action is assumed.

In impact based analysis, risks are approximated by statistics of effective concentrations (EC) of waste site emissions observed in biotests, and by the additional dilution required to diminish EC values under observable effects. Ecotoxicity fluxes (TF^{obs}) and lethality fluxes (LF^{obs}) are also defined on the basis of flows and observed toxicity (cf. Nikunen and Miettinen 1985):

$$\text{TF}^{\text{obs}} = \text{EC}^{-1} \cdot Q \quad (18)$$

$$\text{LF}^{\text{obs}} = 24\text{h-leth} \cdot Q \quad (19)$$

Combining the substance based and impact based analysis, values of TF and EC obtained from biotests are compared with the respective simulated toxicities, defined as follows:

$$\text{T}^{\text{pred}} = \sum (c_i \cdot \text{LOAEC}_i^{-1}) \quad (20)$$

$$\text{TF}^{\text{pred}} = \sum c_i \cdot (\text{LOAEC}_i^{-1} \cdot Q) \quad (21)$$

where, in addition to symbols defined earlier,

T^{pred} , TF^{pred} = toxicity and toxic flux simulated by published LOAECs.

5.2 Applications of the models

5.2.1 Analysis of toxicant sources and releases

Using the simple model described in 5.1.2, the temporal distribution of the releases of some substances in emissions from old municipal solid waste landfills were estimated (Table 19).

Table 19. Estimates of the duration of releases of toxic substances in leachate emissions from old municipal landfills on the basis of concentration fractions in wastes and of flux estimates.

Substance (group) ¹	Minimum t ^R (1 000 a)	
	Based on easily leachable pool	Based on total (extractable) pool
As	6.8 - 8.3	37 - 39
Cd	24 - 140	69 - 340
Cr	11 - 45	160 - 710
Pb	48 - 1 900	240 - 3 800
Zn	13 - 39	25 - 110
PeCP		13 - 29
PCBs		2.6 - 4.1

¹Cf. Table 4; ²Duration of release assuming constant yearly emission and no background baseline.

Based on estimated present leachate emissions and on assumption of linear decrease of toxicants in these sites, the duration of releases for metals in particular is around 10⁵ - 10⁶ a. The easily leachable pool will be released slightly faster.

5.2.2 Comparative analysis of toxicants in waste site emissions by risk indices

Prioritization of toxicants in leachate runoff

A comparative analysis of the toxicological risks of analytes in site-aggregated data was made by substance-specific risk indices (VII). They are based on concentration statistics or flux estimates, environmental quality guidelines (LQGs), toxicity, and environmental sinks:

$$EHI = c_D \cdot (LQG)^{-1} \cdot H^{-1} \cdot t_{1/2} \quad (22)$$

$$ECI = c_D \cdot b_1^* \cdot H^{-1} \cdot t_{1/2} \quad (23)$$

$$ETI = c_D \cdot (LOAEC)^{-1} \cdot H^{-1} \cdot t_{1/2} \quad (24)$$

where, in addition to symbols defined earlier, EHI, ECI, ETI = effective environmental health, cancer and ecotoxic risk index, respectively.

Indices accounting for bioconcentration were also defined as $c \cdot BCF \cdot (LOAEC^{-1} \text{ or } LQG^{-1})$.

The most important analytes on the basis of concentration distributions and toxicities included chlorinated aliphatic compounds, toluene and some chloroaromatic compounds (Table 20). When environmental fate factors were included, non-volatile persistent and biomagnifying organic compounds emerged as the most significant ones. Using flux based indices, heavy metals due to ecotoxicity and organic compounds due to health criteria ranked high (Table 20, VII).

The estimation of non-detects and distribution statistics affected index values. The ratio of 50 % to 90 % fractiles and the ratio of statistics in truncated distributions to those without non-detects was > 100 for some toxicants in leachate.

Table 20. The rank order of risks associated with toxicants in waste site runoff as analysed by 90th percentiles of various risk indices (based mainly on VII)¹.

Health risk rank by conc. and norms (HI)	Health risk rank accounting for fate		Ecotoxic risk rank by conc. and toxicity (TI)	Ecotoxic risk rank accounting for fate		Rank order by specific fluxes ⁴	
	sinks (EHI) ²	BCF ²		sinks (ETI) ³	BCF ²	SHRF	STRF
1 12-DCE ₃	dieldrin	I-TEQ	PCBs	dieldrin	PCBs	HCBz	Cu
2 246-TCP	lindane	PCBs	toluene	lindane	44'-DDD	Cd	Cd
3 TCE ₆	246-TCP	HCBz	endrin	aldrin	toluene	PCBs	PCBs
4 HCBz	44'-DDT	245-TCP	Cu	44'-DDT	PeCP	246-TCP	Pb

¹For abbreviations of chemicals, see Table 4; ²Based on indices of the form $ETI(\text{sec}) = c \cdot BCF$, the latter taken from literature (cf. VII) or derived from K_{ow} (Eq. 11); ³Elements excluded from comparison due to zero decay and volatility (cf. VII); ⁴SHRF = specific health risk flux, STRF = specific toxic risk flux (cf. VII).

Risk indices were also used to analyse the extension of leachate concentrations exceeding effects criteria. Such concentrations were observed within maximally 900 m down stream from the sites.

Prioritization of toxicants in landfill gas

Comparative risk analysis of compounds in landfill gas was done as with leachate, but omitting environmental fate factors and using means due to distribution properties (cf. II).

Prioritized constituents included chlorinated aliphatic compounds, whose average HIs (prior to dilution) exceeded health guidelines (Table 21).

Ethylene dibromide was the most prominent carcinogen, and styrene scored high on TI due to its high inhalation toxicity to animals.

Prioritization of toxicants in ground water

The relative health risk of toxicants in ground water in nearfield points was analysed by comparison with drinking water guidelines (III).

Concentrations in ground water maximally exceeded guideline values by a factor of 400. Strong violations were mainly caused by Fe and were not ascertained in water used as potable water. Risks were tentatively identified by occasional high values of health risk indices for also organic compounds.

Table 21. Comparative risk analysis of toxicants in landfill gas based on statistics of observed concentration distributions and on published toxicity values and air quality guidelines (cf. II).

Toxicant	Health risk index HI_g		Cancer risk index CI_g		Ecotoxic risk index TI_g	
	\bar{x}	Rank order	\bar{x}	Rank order	\bar{x}	Rank order
CCl_4	17	2.	3.8	3.	13	2.
Chloroform	27	1.	4.4	2.	0.67	
1122-TeCE _e	0.1		0.14		5.0	3.
EDB	0.054		320	1.	0.02	
Benzene	9.0	3.	0.26		0.49	
Styrene	0.022		0.004		35	1.

5.2.3 Site-aggregating risk analysis by substance and impact based approaches

Health risks

The health risks from exposure to ground water and leachate runoff were compared by total health risk indices, computed as sums of substance-specific indices omitting exposure:

$$HI_{tot} = \sum HI_i \quad (25)$$

A criterion for relative health risk was defined as the probability of exceeding drinking water guideline values by a specified magnitude (cf. hazard indices, e.g. U.S. EPA 1989a). More frequent violations seemed to be present in ground water than in surface water (Table 22).

The difference was significant (t test, 95 % level), particularly for p ($HI_{tot} > 10$), with all methods of non-detect substitution. Sampling and analysis methods affect these relationships.

Table 22. Comparative analysis of initial health risks from leachate emissions to surface and ground water on the basis of probabilities (p) of exceeding drinking water guideline levels.

Risk criterion	Ground water	Leachate runoff
	p range ¹	p range ¹
$(HI_{tot}) > 1$	0.46 - 0.53	0.34 - 0.43
$(HI_{tot}) > 10$	0.14 - 0.20	0.07 - 0.14
$(HI_{tot}) > 100$	0.03	0.02

¹Based on various methods of non-detect substitution.

Ecotoxicological risk analysis by biotests and substance concentrations

Aquatic ecotoxicological risks were analysed using acute toxicity to *Daphnia magna* as a model. Substance-specific ecotoxic risk indices were devised as described earlier (Eq. 24, VII). They were combined to total ecotoxic risk index TI_{tot}^{pred} , a measure of predicted toxicity to *Daphnia*, by summing the TIs of analysed substances (cf. Eq. 25). Non-detects were substituted by various methods to account for effects of empirical

distributions. Flow-weighted indices of toxic flux were also defined (Eqs. 18 - 19).

There was significant correlation between predicted toxic risk and observed impacts only when flow-weighted measures were used, which is in part an artefact caused by weighting with the same variable (Table 23). The lack of association with predicted and observed toxicity corresponds with that observed earlier (Table 4 in V), and contrasts with the significant covariation between EC50 values and original concentrations for some substances (Eq. 5).

Table 23. Statistical relationships between aggregate measures of the acute toxicity of leachates to *Daphnia magna* based on biotest responses and on toxicant concentrations exceeding reportedly toxic levels (cf. Table 4 in V and text). Ranges of p correspond to values of substance-based measures obtained with various methods of non-detect substitution.

Measure of impact based on biotest	Probability ¹ of uncorrelated risk measures based on c^{obs} and published EC50		
	TI_{tot}^{pred}	TI_{tot}^{pred} , Q-weighted	TRF_{tot}^{pred}
48h-EC50	0.73 - 0.88	n.a. ²	n.a.
48h-EC50, Q-weighted	n.a.	0.0001	0.0001
TF (toxic flux)	n.a.	n.a.	0.95
24h-leth	0.20 - 0.21	n.a.	n.a.
24h-leth, Q-weighted	n.a.	0.0001	n.a.
LF (lethal flux)	n.a.	n.a.	0.0001

¹p = (H0: r = 0); ²n.a. = not applicable or relevant.

Pentachlorophenol concentration distributions in relation to toxicity estimates from extrapolation between species and impacts

The distributions of toxicants and of sensitivity were studied more closely with pentachlorophenol, being prioritized by risk indices and common in leachates. Data was mainly from Nikunen et al. (1990) including original data by Oikari (1987), and Expert Panel (1994).

Differences were large between PeCP concentrations and the cumulative distribution function of acute LC_{50} values in fish and other aquatic species, especially considering medians (Fig. 14). Based on the log-linear distribution of fish LC_{50} , extrapolation was made to a hypothetical most sensitive species (cf. Van Straalen and Denne-

man 1989). This resulted in lower estimates of minimum LC_{50} levels. Margins of safety were still smaller when LC_{50} values were extrapolated to chronic NOAEC values using species with appropriate data, and when upper fractiles of concentrations were used as reference.

The 'risk window' was even more wide when comparing the distribution of observed concentrations with water quality goals assumed to protect aquatic life (Expert Panel 1994) and with no-effect concentrations (PNEC) predicted from short-term and long-term LOAEC data using assessment factors according to the end point and to the phyla represented (Fig. 14).

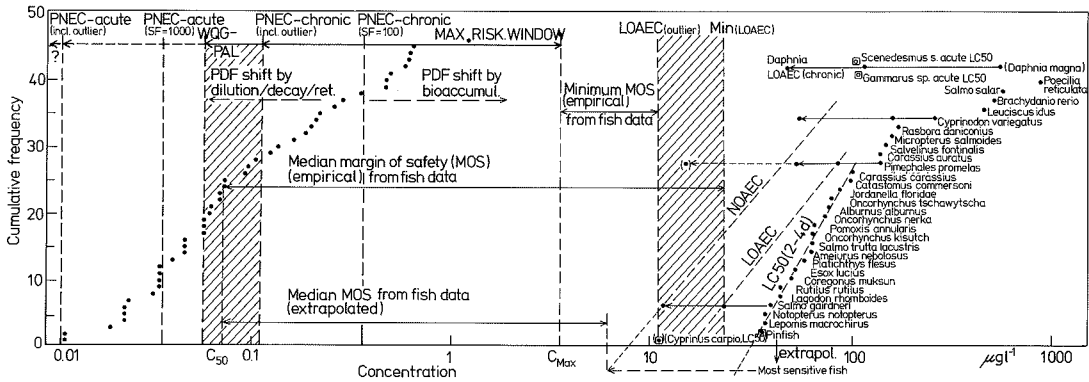


Fig. 14. Relationships between cumulative distribution functions of pentachlorophenol concentrations in leachate runoff and of published toxicity values for fish and other aquatic species, and extrapolation of lowest observed adverse effect concentrations (LOAECs) and NOAECs to predicted no-effect concentrations (PNECs).

5.2.4 Quantitative risk and uncertainty analysis of PCDD/PCDF compounds

Distributions and impact models based on toxicity equivalents

The influence of the distribution of PCDD/PCDF isomers and of their toxicities for relative estimates of toxicological risks was examined on the basis of data from waste sites with and without predominant disposal of chlorophenols (IV).

The relative toxicity and isomer distributions strongly affect estimates of the risks of such compounds. Closer analysis of the toxicity of PCDD/PCDFs quantitated in some samples was made by published data on their relative toxicity (or activity) to various species or biological model systems. Generally, toxic equivalent quantities were higher when considering enzymatic biomarkers and salmonids as compared with standard and rodent-based I-TEQ estimates (Table 24).

Table 24. Evaluation of the toxicity of PCDD/PCDFs in several samples on the basis of mean concentrations and alternative toxicity equivalent systems (in part from VII).

Site or material	Sample	I-TEQs (ppb, d.w./solids, l.w./fish) by TEF system (impact, receptor)					
		I-TEF, rodent ¹	TA, rat ²	EROD, mink ³	LC ₅₀ , trout ⁴⁻⁶	AHH, trout ⁷	EROD, trout ⁸⁻⁹
Sawmill site (A)	soil	20	50	110	46	74	120
CP waste site (C)	sediment ¹⁰	15	39	100	15	25	97
Site (C) recipient	pike	0.05	0.10	0.055	0.07	0.19	0.24
Site (C) recipient	roach	0.02	0.015	0.02	0.02	0.06	0.10
Wood preservative ¹¹	solid (salt)	860	3 300	8 400	1 500	3 000	9 700

¹Based on various effects (Safe 1993); ²Thymic atrophy, Safe 1987, ref. Niimi and Oliver (1989); ³Tillitt et al. (submitted); ⁴Walker and Peterson (1991); ⁵Zabel et al. (1995); ⁶Bol et al. (1989); ⁷Aryl hydrocarbon hydroxylase activity in liver, Parrott 1991 and 1992, ref. Zabel et al. (1995); ⁸Ethoxyresorufin-O-deethylase activity in liver cells, Clemons et al. 1994, ref. Zabel et al. (1995); ⁹Parrott et al. (1995); ¹⁰Based on new samples (cf. IV); ¹¹From IV and Vartiainen et al. (1995).

Site-specific analysis of health risks of wastes related with wood preservatives

The importance of various factors for quantitative estimates of health risks from PCDD/PCDFs were analysed with data from two chlorophenol-related sites. Soil ingestion and fish consumption were chosen as exposure routes (cf. Travis and Hattemer-Frey 1991). Concentrations in sawmill soil prior to removal for treatment were used for model application in the ingestion case.

In the worst-case scenario, constant exposure of a pica child (with excessive soil ingestion) was assumed; in the more plausible case, chronic exposure is in residential land use by child, and median values are used.

The worst-case estimates of cancer risks reached high values (Table 25). More realistic estimates were much lower. Uncertainties were due to both exposure and response models and parameters. A similar range of measures of health risk based on benchmark doses was noted.

Table 25. Quantitative estimates of the magnitudes and uncertainties of factors in the formation of toxicological risks from exposure to PCDD/PCDF compounds in sawmill soil and in the recipient of leachates from chlorophenolic wood preservative production wastes (in part from IV and VI)¹.

Exposure scenario, impact model and variable or factor ¹ (unit)	Worst case		Plausible case	
	Estimate	basis	Estimate	basis
Ingestion of soil in sawmill area				
<u>LMS model of carcinogenicity</u>				
Conc. <i>c</i> (pg I-TE g ⁻¹)	20 000	mean obs. (d.w.)	6 000	<i>f</i> ₅₀ , obs. (w.w.)
Contact rate CR (g d ⁻¹)	10	pica child ²	0.10	<i>f</i> ₅₀ , child ²
Body weight <i>m</i> _B (kg)	10	infant	20	\bar{x} , child
Fraction of time exposed <i>f</i> _T	0.8	daily ³	0.4	bidaily ³
Exposure duration fraction <i>f</i> _D	1	same CR all life	0.1	\bar{x} , child ⁴
CDI ⁵ (pg kg ⁻¹ d ⁻¹)	16 000		1.2	
Absorbed fraction	1.0		0.4	max. obs. ⁶
Effective dose D ^E (pg kg ⁻¹ d ⁻¹)	16 000		0.6	
Potency <i>b</i> ₁ (pg kg ⁻¹ d ⁻¹) ⁻¹	1.6·10 ⁻⁴	UCR by LMS ⁷	0.97·10 ⁻⁵	revised UCR ⁸
Excess cancer risk R _c	2.5		6·10 ⁻⁶	
<u>Threshold model of toxicity</u>				
ADI (pg kg ⁻¹ d ⁻¹)	0.5	LOAEL/300 ⁹	1.5	LOAEL/100 ¹⁰
D ^E ADI ⁻¹	32 000		0.4	
Consumption of contaminated fish				
<u>LMS model of carcinogenicity</u>				
<i>c</i> (fish) (pg I-TE g ⁻¹ w.w.)	0.27	Max, pike fillet	0.23	Mean, pike
CR (g d ⁻¹)	100	staple food ¹¹	15	sport fishing ¹²
<i>m</i> _B (kg)	20	child	30	\bar{x} for age classes
Cooking loss (%)	0		25	published data ¹³
CDI = D ^E (pg kg ⁻¹ d ⁻¹)	1.4		0.086	
R _c	2·10 ⁻⁴		8·10 ⁻⁷	
<u>Threshold model of toxicity</u>				
D ^E ADI ⁻¹	2.4		0.06	

¹For fish consumption, only variables deviating from those in the soil ingestion case have been shown; ²From data by Stanek and Calabrese (1995) and van Wijnen et al. (1990) for play; ³During melt period; ⁴From average duration of residence of a child, cf. Pohl et al. (1995); ⁵Chronic daily intake; ⁶Gut absorption for soil-bound TCDD (Expert Panel 1994); ⁷U.S. EPA's 95 % UCL estimate of unit cancer risk (corresponding to a R_c of 10⁻⁶) by the LMS model from the data of Kociba et al., cf. Pease (1990); ⁸95 % UCL estimate of UCR from reanalysis of the Kociba et al. data, Copeland et al. (1993); ⁹From LOAEL of 0.15 ng I-TE g⁻¹ d⁻¹ for learning impairment and endometriosis in monkeys by Schantz and Bowman 1979 and Reier et al. 1993, respectively, ref. De Vito et al. (1995), interspecies scaling factor of 10 and safety factor of 30; ¹⁰From a LOAEL of 0.2 ng I-TE g⁻¹ d⁻¹ for behavioral toxicity in monkeys observed by Bowman et al. 1993, ref. Pohl et al. (1995), interspecies scaling factor of 10 and safety factor of 10; ¹¹Fish as main diet, Ahlborg et al. (1988); ¹²Average consumption among recreation fishers (Travis and Hattemer-Frey 1991, Law and Gudaitis 1994); ¹³From observed loss for TCDD, Law and Gudaitis (1994).

6 DISCUSSION

6.1 General features and importance of toxicant distributions at waste sites

General considerations

Spatial, temporal and structural distributions of toxicants, impacts and other risk variables and associated probability distributions play an important role in the formation and assessment of toxicological risks caused by waste sites.

The significance of probability distributions of risks is related to the role of variation and uncertainty in risk factors, both physical and societal or technological ones (Finkel 1990b). Risks inherently carry a notion of probability, but point estimates of these probabilities are insufficient. Associated variation and uncertainties have to be identified and characterized more explicitly (Copeland et al. 1993, Dakins et al. 1994). Distributions of risks are also important for comparisons of risks and thus for decisions of risk management options (Goodman 1986).

Empirical and theoretical probability distributions

The empirical distributions of risk variables are approximations of theoretical distributions. They may be statistically indistinguishable, but this does not prove the generalizable existence of the theoretical distribution in the sampled environment, as was suggested by Assmuth (I). Still, uncertain distributions may be inferred by analogues and by information on the processes producing them. For instance, the lognormal distribution observed also with many toxicants in the present studies (I, II) has been implied in other studies of environmental contamination (Sielken 1987, Birmingham 1990) and frequently arises where the variables are products of multiplication of factors (Dudewicz and Mishra 1988).

The high frequency of non-detects with many toxicants, particularly in liquid media, has been noted also by other researchers of waste sites (Gibbons 1992, Kerndorff et al. 1992). The existence of several (varying) detection limits (DLs) complicates the elucidation of the form and parameters of the distribution (Haas and

Scheff 1990, Haas and Jacangelo 1993). It is common to substitute $\frac{1}{2}$ DL for non-detects or prudently by DL (cf. Helsel 1990). A more consistent approach is estimation based on the properties of the distribution above DL. Even though these estimation methods are in part arbitrary and include uncertainty, they are preferable to the omission of non-detects. In that case, much higher statistics are obtained; however, these are unrealistic estimates of general conditions and do not serve e.g. as the sole basis for loading and risk estimates due to their rare occurrence. On the other hand, also the few values exceeding high DLs are important for identification of peak contamination and risks.

Long tails of probability distributions of risk variables imply the possibility of low-probability but high-consequence risks. Long tails may be caused by tails in the distribution of toxicant occurrence in emissions, of exposure factors, or of receptor susceptibility (e.g., Finkel 1990a).

The implications of risk variable distributions were explored mainly on the basis of single variables in subsequent stages of risk formation. Their codistributions are important in hypothesizing relationships between variables, in identifying indicator variables, and in modeling risks. Joint distributions of both exposure and response variables were examined in the combined analysis of substance-based risk estimates and observed impact measures (5.2.2, 6.6).

The question of individual and population risks is closely related to the probability distribution of risks.

Temporal and spatial distributions

The distribution of risk variables in space and in time are important characteristics. The spatial dimension is inherent in the concept of waste sites, being (to some degree) local phenomena (1.1, 1.2). Their impacts and risks in the environment extend beyond the local scale (6.7). Spatial distributions are also related to partitioning between various phases in wastes and their environment (Fig. 13, 6.2).

The temporal distribution relates to risk variables in all stages of risk formation (5.1.1) and involves trends, rapid alterations, fluctuations and lags. Consequently, present and future risks may be distinguished (e.g., Zamuda 1989b).

6.2 Phase distribution, mobility and attenuation of toxicants

Leachability from wastes

The leachabilities of heavy metals were found to correlate with organic matter and pH. Organic matter is known to retain metals by sorption (Christensen et al. 1994). Organic matter also contributes to anaerobic decomposition and thus to metal precipitation in sulfides, which has been found important especially for Cd, Zn and Pb in wastes (Förstner 1988). The correlation of pH with the retention of Cd and Cu in wastes has been reported by others (Bartlett 1991, Peiffer et al. 1990), and can be expected based on the geochemical behavior of these metals. For many metals, greatest mobility occurs in the acid fermentation stage. Complexation and ion exchange is pH and redox dependant (Förstner 1988). The present study is among the first where relationships between the mobility of toxic metals and waste properties were quantified in samples from old disposal sites. The acetate extraction catches sorbed species, HNO₃ extraction also sulfides (Förstner 1988, Ton et al. 1993). Sorbed species may be easily leached, while metals in precipitates are retained to a higher degree. Speciation particularly governs the attenuation of Cr (Bartlett 1991) and of As (Blakey 1984, Holm 1990).

Organic toxicants are retained in waste by sorption. Many of them are also volatilized and decomposed or transformed biologically. For instance, pentachlorophenol and lindane degrade relatively rapidly in methanogenic landfills (Fischer and Pecher 1993). They were found also in old mixed municipal wastes; initial concentrations may have been much higher. However, decomposition could not be directly observed. Attenuation of organic toxicants has been noted by concentration gradients between various phases (Assmuth 1992).

Attenuation capacity

It is clear from this and other research (see references above and in Assmuth 1992) that wastes possess attenuation and retention capacity for toxicants as a result of several reversible and irreversible processes (decomposition). The use

of this capacity of wastes has been considered for management of toxicants in codisposal (e.g., Department of the Environment 1978, Young et al. 1984, Collins and Spillman 1986). Attenuation is important also in remediation of waste sites.

However, the present studies have also produced evidence of variable and lacking attenuation. A mixed-waste landfill is a storage and a reactor with great heterogeneity and limitations. A limitation for the reliance on decomposition is the possible formation of more mobile and potent substances, such as vinyl chloride from chlorinated ethenes under reducing conditions.

The present studies have produced estimates of long durations of leachate emissions with present levels of release, e.g. in comparison with the estimated time to reach final storage quality of controlled municipal landfill leachates (Belevi and Baccini 1991). For all toxicants retained in wastes (or in other media), the release rates and duration of retention may also change due e.g. to the pH and redox of the wastes (Fig. 13). In the very long run, all persisted (and not permanently sedimentated) toxicants will be dispersed to the environment, while organic compounds may degrade already in wastes, thus reducing risks.

Climatic and other environmental conditions in combination with disposal technology influence the behavior and attenuation of toxicants in wastes (Fig. 13). In Finland, freezing and thawing and flooding e.g. at snowmelt are important for leaching, and low temperatures for gas generation (Ettala 1988). Waste compaction and cover affect these and other aspects of water and material fluxes (Collins and Spillmann 1986). The effects of such factors on toxicant attenuation were not explicitly studied. Qualitative and indirect indications was obtained e.g. of the effect of climate on waste properties and emissions by observations of structural failures of some waste sites (cf. Assmuth et al. 1990b).

Attenuation and distribution outside wastes

Attenuation of toxicants continues after releases from wastes. For instance, with increasing redox potential from anaerobic wastes toward the environment, some metals are precipitated or sorbed and some organics are decomposed readily, while others are more mobile and persistent (see review by Christensen et al. 1994). These

processes operate in both surface waters (see discussion in I) and ground water and soil (III, VI) and in other natural environments. The purification and retardation capacity of soils for leachate toxicants such as monoaromatic and chlorinated aliphatic hydrocarbons has been found to be dependant on site conditions (e.g., Borden and Bedient 1987). This capacity of soils and other media and systems is limited e.g. by their heterogeneity allowing mobilization and transport of toxicants, particularly persistent ones. The stress on soil organisms hampers self-purification. Attenuation also depends on technological systems designed for external emission control (e.g., landfill leachate and gas collection and treatment) and other artificial structures and processes, e.g. sewer lines at uncontrolled waste sites in industrial areas. Attenuation capacity is variable and limited also in these external systems (Diependaal et al. 1993).

At wetland landfills, the leachate toxicants are affected by peat, e.g. through retention of light and heavy metals by sorption (Assmuth et al. 1990b, Ton et al. 1993, Warth 1993). The leachate constituents migrate in the hydrologically active surface peat layers, e.g. in the low peripheral parts of moors (Assmuth et al. 1983).

6.3 Toxicant concentrations and fluxes in relation to reference values

General considerations

The toxic substances which were most frequently detected in runoff, ground water and gas at study sites have been among the dominant toxicants in leachate runoff from other waste sites (Tables 4 and 14, I - III, see also Assmuth 1992). This suggests that these substances are ubiquitous in wastes in industrialized countries.

The differences between the concentrations of toxicants measured in this study and in other studies may be due to both measurements and models of processes. Sampling focus, analytical methods and data analysis influence the estimates of concentrations and fluxes for many toxicants (3.4.2). Variations in modeled processes exist in all stages of toxicant transport and transformation, from waste generation and disposal to subsequent environmental fate (6.2).

Thus, generalization and comparison of concentrations between studies is difficult. Tracers such as chlorides, and fluxes of toxicants, give additional information, but the latter are constrained by the paucity of flow data in this and in most reference studies (I, II). Comparison of observed concentrations is more meaningful within sites, e.g. in relation to background quality of site, and between sites after consideration of their differences e.g. in terms of dilution.

Toxicants in leachate runoff

The detection frequency of most analytes exceeded that in foreign surveys (Table 4, I). This difference is affected by analytical sensitivity. The lower concentrations of many toxicants in leachate runoff from Finnish than foreign waste sites do not necessarily indicate better qualities or smaller emissions of the former. In foreign studies samples may have often been taken from primary leachates. Comparisons of leachate runoff data within study sites showed that the median concentrations of many toxicants were in the background range, whereas average concentrations were significantly elevated due to high although in many cases infrequent peak values.

According to related work by Assmuth et al. (1990b), the estimates of toxicant fluxes in leachates from study sites were minor in comparison with fluxes from e.g. industry for the few substances allowing comparisons. On the other hand, the flux estimates were greater than those for a hazardous waste treatment plant, which is the principal alternative for disposal of hazardous wastes in landfills (Assmuth et al. 1991).

Toxicants in landfill gas

The principal chloroaliphatic compounds in landfill gas have been found in other studies (II, Scott et al. 1988). Their concentrations were above background levels. Skewed probability distributions of trace compounds in landfill gas have been observed earlier (Ettala et al. 1988). Such distributions were not so pronounced as in solid or liquid phases. Variation in gas concentrations between sites may be due to sampling, to the age of the waste (Rettenberger and Stegmann 1991) and to the original presence of toxicants.

Toxicants in ground water, soil and other media

Concentrations of toxic substances in ground water were in the same range as those near other Finnish landfills (Pärjälä 1989) and in some Scandinavian studies (for references see III), but were often below those found in Germany, USA and Canada (Kerndorff et al. 1992, Lesage et al. 1990). Measurement related variation and error is involved especially with volatile compounds and reactive species (of metals) in subsurface environments (see discussion in III and West et al. 1995). Ground water quality is also affected by general hydrogeochemical conditions. Many of the high concentrations were measured in perched water or stagnant water in aquitards and not in aquifers (which were present near few sites). However, it is evident also from the present work that waste sites alter the quality of subsurface waters and cause toxicant concentrations exceeding background levels.

In addition to ground water contamination, soil contamination is present in the areas exhibiting elevated ground water contaminants. Soil contamination in itself causes risks at waste sites, e.g. to ecological soil functions (see data in Table 7 and Van Straalen and Denneman 1989).

The concentrations of PCDD/PCDFs in solids at sites associated with chlorophenols were relatively high in comparison with data from studies of similar sites (see Table 1 in IV). Still, the observed concentrations were below values measured e.g. at sites associated with production of 245-T (IV) and chlorine (Wilken et al. 1994). The concentrations in biota were not higher than in other contaminated environments (VI).

6.4 Comparative analysis and prioritization of toxicants

The substances assigned greatest risks varied according to the effect used as criteria for ranking. The substances displaying greatest carcinogenic risks differed from those prioritized on the basis of health norms and ecotoxicity (VII). Health norms in principle integrate all types of adverse effects on humans and emphasize critical ones (IPCS 1994). However, norms may be biased to established (and more easily observed

and proven) effects. In any case, the selection of receptors (species) affects the comparative evaluation of toxicants; this was illustrated by the marked differences between values of risk indices e.g. for landfill gas (VII).

Considering environmental fate factors, the highly toxic persistent nonvolatile and biomagnifying substances, e.g. PCBs, PCDD/PCDFs and neutral chlorinated biocides and their metabolites (dieldrin, aldrin, endrin, 44'-DDD, lindane, HCBz), emerged as the most important organic toxicants in leachate emissions (Table 20). Yet, most of these substances rank high only when considering peak values (distribution tails). Among the more common toxicants, toluene and Cu were important also when environmental fate and fluxes were considered. However, toluene as a volatile compound is difficult to quantify and it may actually decompose rapidly in aerobic conditions (e.g., Berwanger and Barker 1988). These rankings differ also for carcinogens from those based on carcinogenicity and leachate concentrations only (Brown and Donnelly 1988). The present analysis also differs from the scaled assessment of ground water contamination at waste sites by Kerndorff et al. (1991), who emphasize mobility in soil. Their values of groundwater transport potential obtained e.g. from K_{ow} and solubility may be more appropriate for that exposure route. They may also correlate with the exposure factors in the present indices. Fiedler et al. (1990) have also produced toxicant assessment methods based on toxicity and volatility which are principally similar to the indices in the present work.

Prioritization of toxicants on site-aggregating basis does not account for intersite differences. While the differences in source terms are to some extent covered by the use of flux-based indices which imply weighing by (site-specific) flow data, intersite differences e.g. in exposure conditions are not included in the indices (6.9).

The probability distributions of concentrations and of other risk factors in the models underlying indices are crucial in comparative risk analysis. Due to the common skewness of the distributions of toxicants and to low detection frequencies, many substances which were important on the basis of high isolated maxima did not have high (or even estimable) median values. The rankings are also highly sensitive to the non-detect substitution methodology (6.9).

6.5 Health risk assessment

Estimation and characterization of risks of substances and effects of concern

Quantitative estimation of present and still more future risks to human health from toxicants at study sites is possible to a limited extent only. Health risk assessment is constrained by both lack of clinical or epidemiological investigations among human populations at study sites and by exposure data. Concentration distributions at point of exposure were in most cases not estimable, and neither were other exposure factors such as duration and frequency, which are governed by the behavior of the exposed population (cf. 6.6). This situation is a common problem in the assessment of health risks from waste sites (Talbot et al. 1987). Moreover, concentrations in exposure media are not equivalent to actual exposures in terms of body or tissue levels of toxicants (e.g., Travis et al. 1989). However, semiquantitative comparative analysis of health risks was possible on the basis of substance-based risk indices (6.4), and some analysis of risks associated with prioritized toxicants may be made qualitatively and, as with PCDD/PCDFs, quantitatively by exposure scenarios. These estimates are approximate illustrations of the risk analysis methodology.

The high estimates of excess carcinogenic risks from PCDD/PCDFs in worst-case exposure situations correspond to some other estimates of cancer risks from waste sites (Tables 26 and 6). However, more plausible scenarios of exposure and potency, still based on prudent assumptions of longtime residential and fish consumption activities (and on the LMS model), produced much lower estimates. They approached background levels (10^{-6} - 10^{-5} excess cancers in a lifetime), being of the same order of magnitude as some foreign estimates of cancer risks from comparable contamination of soil or fish (Pausenbach 1989, Parsons et al. 1994). Nisbet et al. (1988) obtained higher estimates for heavy consumption of fish contaminated by PCDD/PCDFs from chlorophenolic production.

A risk for non-Hodgkins lymphomas of ca. $2 \cdot 10^{-3}$ (based on reported excess cases and population size of 2 500) was observed in a population in Kärkölä with prolonged exposure to chlorophenols in ground water at concentrations of up to

100 $\mu\text{g l}^{-1}$, including the tumorigen 246-TCP and PeCP (cf. Lampi et al. 1990, 1992).

Maximum estimates of human carcinogenic risks from exposure to gases emissions from waste sites have reached high values (Minott 1989). However, assessments accounting for dilution and exposure factors have resulted in estimates in the background range (I, Crouch et al. 1990, cf. Wallace 1990). Also other effects may occur (Scott et al. 1988). Occupational exposures may be greater for airborne substances (gases, aerosols). PCDD/PCDFs have been recently measured in landfill air as a result of fires (Rahkonen et al. 1994), and may contribute to health risks from many waste sites.

Reproductive and developmental effects have been suggested to be caused by waste sites, but evidence is inconclusive (Table 5). Among the toxicants in this study, PCDD/PCDFs and PCBs and some other organochlorine compounds (e.g., neutral pesticides) impair reproduction and hormonal, neurological and behavioral development (Hemminki and Vineis 1985, DeVito et al. 1995), and cause immunotoxicity (McGrath et al. 1995) and chronic neurotoxicity (Peper et al. 1993). For the sawmill case, ADI values based on such effects were exceeded also with more plausible assumptions in residential exposure scenario based on LOAECs for behavioral toxicity. However, this risk level does not represent actual conditions at the site, which is used for industrial purposes only and contains a large part of the contamination in compost strings causing less exposure than residential soils.

Guideline values for health protection were exceeded near study sites by peak concentrations of many toxicants. The high concentrations of Fe in ground water may be caused by mobilized natural Fe (Christensen et al. 1994). Also other toxicants in water and gas samples clearly exceeded guideline values (I, III, Table 21), but such concentrations are subject to attenuation in surface waters and in soil prior to exposure. However, elevated values of health risk index also near wastes point to a potential for health risks. The occurrence and causes of, exposures to and health risks from leachate toxicants particularly in ground water have not yet been analysed sufficiently (cf. Pärjälä 1989). Increased local health risks may be caused e.g. by future releases and uses of water or other contaminated media near waste sites.

Models and cofactors of health risks

Health risks were modeled assuming simple similar action and concentration additivity of individual substances, as usual (U.S. EPA 1989a and b, Table 6). However, Chen et al. (1990) found higher upper-bound estimates of total risks than by simply adding the risks of individual contaminants. Even if additive, the relative toxicities vary. The present work showed notable differences in the toxicity of samples contaminated by PCDD/PCDFs depending on the TEF system (6.6, Table 24, IV). The inclusion of PCBs in equivalency calculations may further increase the level and variation of risk estimates.

In carcinogenic risk assessment, the linear multistage model was applied as the default considered plausible and prudent (Anderson et al. 1983, Reichard et al. 1990). Yet, there is no consensus on models of carcinogenesis (Cothorn and Van Ryzin 1985, Thorslund and Charnley 1988), and the choice of model e.g. for TCDD alters low-dose risk estimates by orders of magnitude (Bonvalot et al. 1989). Carcinogenic potency in humans correlates with that in other animals (Allen et al. 1988, Goodman and Wilson 1991), but interspecies extrapolation has been criticized due to differences in background incidence of tumors (Sielken and Stevenson 1994) and to test artifacts (Rieth and Starr 1989). Also the body weight scaling method for conversion of animal data affects risk estimates (Travis and White 1988). Other factors include the definition of tumors (Copeland et al. 1993), the level of their aggregation (Gold et al. 1993), and the time to tumor (Sielken 1989). In this work as in most analyses of carcinogenic risks of waste sites, such factors were not explicitly considered. Conservative assumptions are used but may result in high risk estimates (Table 6, 26). On the other hand, individual variation in susceptibility due to age, genome, nutrition and illness may be large (Finkel 1990a).

For other forms of chronic toxicity, Weibull models (Kodell et al. 1995) and power function models (McGrath et al. 1995) are used for quantitative dose-response assessment. Yet, analyses based on LOAELs and safety factors (threshold models) are most common (Barnes and Dourson 1988, IPCS 1994). This approach was also used in the present work implicitly with health risk indices and explicitly with CDI/ADI ratios (for

PCDD/PCDFs). Most variation in these ratios was due to exposure assumptions; however, also safety factors differ depending e.g. on the end point and data used (Pohl et al. 1995). In both safety factor based and dose-response function (and distribution) based extrapolation from animal data to humans, the default assumption used also in the present work is greater human susceptibility. This is justified because of prudence, but may contradict evidence.

Toxicokinetics may alter health risk estimates (Travis et al. 1989). Lower estimates of cancer risks have been obtained by consideration of metabolites and other kinetic factors in exposure to vinyl chloride and trichloroethene (Clewell et al. 1995) and to PCDD/PCDFs (Rao and Unger 1995). However, toxicokinetics may also involve potentiation and formation of toxic metabolites.

6.6 Ecotoxicological risks of prioritized analytes

Exposure factors

Decay in both biological and other processes influences the risks of many organic toxicants (5.2.1., VII). Decay was accounted for only in the semiquantitative risk identification by simple first-order linear models (cf. 6.4). The data on biodegradability and their interpretations (Cowan et al. 1995) depend heavily on the environmental compartments and processes (e.g., creek, wastewater treatment plant). The assumption of zero decay is justified and prudent for substances such as PCDD/PCDFs with $t_{1/2}$ values of ca. 10 a in soil (e.g., Paustenbach 1989) and for conditions not favoring transformation. Transformation products of degradable compounds may cause additional risks.

Sedimentation is a sink for many substances. However, there is transport from sediment e.g. in resuspension, bioturbation, gas convection and solubilization. Partitioning between sediment and pore water is involved in these fluxes (Di Toro et al. 1994). In a long-time equilibrium much of the toxicants in sediment will be available for exposure through water or food chains.

Bioavailability may significantly affect the toxicity of contaminants in aquatic (Dobbs et al. 1994) and terrestrial systems (Umbreit et al.

1987, van den Weiden et al. 1989). Generally lower risks are caused than those predicted from readily available test substances. Bioavailability to animals was considered indirectly in connection with PCDD/PCDFs accumulated in fish.

The importance of biomagnification of persistent lipophilic substances such as PCBs and non-polar organochlorine pesticides was suggested by risk indexes (Table 20, cf. Romijn et al. 1993).

However, biomagnification of PCDD/PCDFs to fish from highly contaminated sediments was slight. PCDD/PCDFs in these sediments may be immobile, or physical transport may dominate. Sampling effects may also be involved. Endicott and Cook (1994) found low sediment-to-fish bioconcentration of PCDDs and discussed the role of partitioning. Organic carbon content is central in partitioning but was not measured. Highest biomagnification of PCDD/PCDFs is to filter-feeders (Muir et al. 1989). Biomagnification of PCDD/PCDFs is constrained e.g. by molecule size (Loonen et al. 1994), reducing the uptake of higher chlorinated PCDD/PCDFs also in pike (Parkerton et al. 1993). Such factors may explain the low biomagnification (by I-TEQs) observed in the present study site where highly chlorinated PCDFs dominate. Overprediction of biomagnification may also be associated with differences in lipid content between trophic levels (Leblanc 1995).

Impact assessment

Other species than humans are likely to include organisms both more exposed and more vulnerable to waste site toxicants than humans (Suter 1993). However, comprehensive characterization or quantitative estimation of ecotoxicological risks and comparisons of human or non-human risks could not be made explicitly.

Interspecies variation in sensitivity is large e.g. for pentachlorophenol (Hedtke et al. 1986) and even larger for PCDD/PCDFs (Vanden Heuvel and Lucier 1993). Interspecies differences are observed also in acute effects especially between remotely related species (Oikari 1987). Extrapolation based on sensitivity distributions is used to account for this (Okkerman et al. 1993, Aldenberg and Slob 1993). In a simple prudent approach, lowest LOAEC or NOAEC values aggregate responses of several species. In

this work, extrapolation from distributions of LC_{50} of fish to PeCP yielded estimates of the LC_{30} of a hypothetical most sensitive species. The margin of safety was thereby reduced from that based on medians of observed and acutely toxic concentrations, and also from that implied by ecotoxic risk indices based on lowest LOAEC and upper concentration fractiles. Yet, the dominating difference between observed and critical concentrations (omitting exposure) stemmed from the extrapolation to PNECs (OECD 1995). In addition to the selection of phyla, test duration and respective assessment factor, the PNEC predictions and risk estimates were dependant on a single outlier (low LC_{50} value for carp).

Toxic equivalency factors are used particularly for PCDD/PCDFs which act through same receptor mechanisms (Safe 1992); the use has extended to other dioxinlike compounds (Niimi et al. 1989, Heaton et al. 1995) and to chlorophenols (Kovacs et al. 1993). Combination of the toxicities of several compounds to risk indices represents a still more integrative assessment. This work has pointed out the variation and limits of toxic equivalent factors when extrapolating from one species to another and between end points (Table 24). PCBs also play a role (Table 16). Toxic equivalent concepts for dioxin risk assessment has been criticized (Neubert et al. 1992). The method and level of compound integration is dictated by the purposes of the analysis.

Reproductive and developmental effects are important for populations and communities (Barnthouse et al. 1990). Of the analysed toxicants, PCDD/PCDFs and other dioxinlike compounds (e.g., PCBs) and some pesticides at low doses impair reproduction and development of e.g. birds (Giesy et al. 1994b) and minks (Heaton et al. 1995). The observations of deformations of sediment-living insect stages in the recipient of the study site exhibiting high sediment concentrations of PCDD/PCDFs (Kukkonen, personal communication) may be partly associated with emissions from the site.

Behavioral effects especially of persistent neurotoxicants have significance also at higher ecological levels. Swimming disorders are caused e.g. by low TCDD concentrations (Little and Finger 1990, cf. Bowman et al. 1989).

Already before such effects, subtle but widespread effects are likely to be caused. For instance, Van der Weiden et al. (1989) measured

significantly elevated EROD activities in carps after semichronic exposure to PCDD/PCDFs in sediments at concentrations of 0.5 ppb I-TE (d.w.), which is below the concentrations observed in this study (IV).

The toxicity of complex effluents may differ from that anticipated on the basis of some components (Oikari and Lindström-Seppä 1990). The assumption of simple similar action and concentration additivity holds reasonably for (narcotic) chemicals lacking specific mechanisms of action, including most analytes in the present work (De Wolf et al. 1988, Deneer et al. 1988, Broderius et al. 1995, Logan and Wilson 1995). However, complex mixture effects also occur e.g. for metals in aquatic systems (Calabrese 1991) and for some PCDD/PCDFs and PCBs (Bol et al. 1989). PeCP is a metabolic inhibitor and not a narcotic, and does not behave strictly additively (McCarthy et al. 1992); its environmental and biological fate is also affected by acid hydrolysis (Lyman 1990).

Toxicokinetics modify ecotoxicological risks. Biologically based toxicokinetic models have the principal advantage of avoiding errors in toxicity assessment associated with multiple sources, non-steady state exposures or pulsed exposures, which are common at waste sites (Landrum et al. 1992). In the present work, toxicokinetics were not analysed (cf. Fig. 12), but tissue contaminants reflect part of their compound effect.

6.7 Assessment of leachate impacts by biotests

Comparison and extrapolation of test results

Leachate runoff samples from study sites were acutely lethal to *Daphnia magna* particularly when their original dilution was small (V). The median and mean lethality in standard 48 h tests was lower than that measured in many other studies of waste leachate toxicity to *Daphnia magna* (Table 7, V). However, the comparison of these results is difficult for several reasons.

Biotest comparability is inherently limited. Test conditions and populations vary (Baird et al. 1991, Grothe and Kimerle 1985). Lack of dilution data is also an obstacle for comparisons, as with chemical analysis.

Short-term tests are limited in their ability to predict effects, although their results correlate for many species with chronic responses (Holcombe et al. 1989). Chronic effects are difficult to estimate especially with mixtures (McBride et al. 1979), even when the test duration is 10 % of life length, as in 48 hour *Daphnia* tests. The age of test population may introduce large variation (White and Champ 1983). The development of *Daphnia* populations is also governed by toxicokinetics (Hallam et al. 1990).

Daphnia magna is a sensitive species also to waste site eluates (Table 7, see also Miller et al. 1985, Thomas et al. 1986), and suitable for standard tests. Yet, it represents a low taxonomic level and some environments only (Suter 1993). Interspecies extrapolation is complicated by exposure to many substances (Hermens et al. 1984a). Tests with several species are thus advocated (Perrodin 1988, Van Coillie et al. 1989).

Bacterial mutagenicity of leachate samples was observed as in other foreign studies of waste sites (Table 7, Davol et al. 1989). Mutagenicity tests give indications also of carcinogenicity (Gold et al. 1993). Of the substances frequently abundant in leachates, Cr, DCM, chloroform and 12-DCE_a have caused mutagenicity to *Salmonella* strains TA97 or TA100 (Norpoth 1982). However, relationships between mutagenicity and concentrations of substances are difficult to infer from the present data of complex effluents, given the above uncertainties in biotest interpretation.

The reproducibility of laboratory zooplankton tests in the field has sometimes been relatively good (Cowgill 1987). Yet, variations of environmental conditions between laboratory and nature and between different environments, e.g. in population and community dynamics, hinder the prediction of real effects from biotests.

Impact models and risk inference based on biotests, chemical data and cofactors

Many models have been used to analyse toxicity data, e.g. the classic probit model of Bliss and Finney and its modifications (Stephan 1977), Weibull model (Christensen 1984) and the power function model used mainly in this work and with immunotoxicity (McGrath et al. 1995). All of these involve uncertainty and may be suboptimal for some cases of estimation.

Statistically significant covariation was found between observed toxicity and predicted effects (Table 23) only with flow weighting which may induce artificial correlation. The lack of a more general association reflects the insufficiency of empirical and literature data and of models in explaining effects of complex leachates (cf. Lambolez et al. 1994). Toxicity was also not observed in some cases when concentrations exceeded levels reportedly toxic to *D. magna*. This may partly be due to differing availability in different sample types (see discussion in V). However, the regression model connecting toxicity to Zn and Hg gains credibility also from other results (e.g., Khangarot and Ray 1989).

There is empirical support for the assumption of simple similar action and concentration addition, used in the present work also for *Daphnia* (Hermens et al. 1984a,b). Still, mixture effects may have contributed to lacking correlation in the impact based and substance based measures.

Causal relationships between the occurrence of chemicals and toxicity are difficult to establish (Suter 1993) due e.g. to other adverse agents, variation, and factors modifying toxicity. Toxicity suppression may occur e.g. by sorption of metals on organic matter in leachates (Biesinger and Christensen 1972). Humus has been found to decrease the bioavailability of organic contaminants to *Daphnia* (Oikari and Kukkonen 1990), but it also increases the toxicity of organic pollutants to *Daphnia* (Virtanen et al. 1989). Some of the general features of waste leachates may make exposed organisms more vulnerable (e.g., oxygen deficit) while others (e.g., nutrients) may protect them.

Thus, samples not causing acute lethality may be otherwise toxic (false negatives), and observed toxic effects may be due to other factors than toxicants (false positives) (Chapman et al. 1987). In screening for risks, the former cases are of most concern (Johnson 1988).

6.8 Characterization and uncertainties of risks

The role of uncertainties of risk variables

Uncertainty of measurements and models exists in all stages of risk formation (Finkel 1990b,

Suter 1993). Uncertainty is inherent in the identification and resolution of impacts or causes. False negative and positive results are obtained with both chemical and biological measurements and associated models, and quantitative risk estimates carry uncertainty. Analysis of uncertainty is thus important for risk evaluation, comparison and management, and for directing research both at single sites (Dakins et al. 1994) and generally. It reveals a more nuanced picture of risks and of associated unknowns. Although varied and great, their elucidation serves to guide risk analysis and management. It can aid e.g. in defining and applying precautionary principles.

Information on distributions of risk variables is needed for characterization of uncertainties (Finkel 1990a). Consideration of distributions is particularly important when many risk formation stages contain large uncertainties. The significance of this cascading uncertainty when proceeding from toxicant source to response has been stressed by many researchers of local contamination (Paustenbach 1989, Maxim 1989, Copeland et al. 1993). It was demonstrated in the present work for quantitative health risk estimates of PCDD/P-CDFs, and illustrates the tension between repeated default precaution and best estimates.

It must be stressed that not all risks and uncertainties are tractable by such quantitative analyses. Qualitative aspects are also involved. Risks from other sites, substances, transport routes and effects than those addressed may be important.

Measurement uncertainty along the analytical chain was large in the present work for many analytes particularly in heterogeneous matrices. This is reflected in the large coefficients of variation. Uncertainty of flux estimates is caused by flow measurements (3.4.2); Droppo and Jaskot (1995) pointed out the importance of the total hydrological regime of rivers for sampling waterborne soluble and sorbed toxics. Measurement uncertainty is also inherent in background data. For instance, the reported toxicity of PeCP may have been affected by its toxic impurities (Hamilton et al. 1986). In bioassays there is variation of responses, requiring a dose-response statistic such as LC50 or LC25. Secondly, variation occurs in these compound statistics, requiring the specification of e.g. 50 % fractile of LC50 values (see in particular Suter 1993). Great uncertainty is also produced by many bioassay factors (Cothem and van Ryzin 1985).

Still, models may be the dominating source of uncertainty in risk estimates, as demonstrated by the present work (6.5, 6.6, Table 25). The estimates differ by orders of magnitude e.g. with many dose-response models.

Model and measurement uncertainty is involved in the incomplete coverage of substances, routes, receptors, impacts and associated processes (Table 11). Many important toxicants were not analysed. Laboratory measurements of biological responses only partly compensate for this. The exclusion of some types of waste sites increases uncertainty of the general risks of waste sites. For instance, a large part of the Cd produced in Finland is disposed of in one industrial deposit (Mukherjee 1989), but the impacts and risks of such dominating toxicant pools may not be evaluated on the basis of the present data.

Temporal and areal distribution of risks in a space-time continuum

The toxicological risks of waste sites, being largely due to toxicant disposal long ago, change due to alterations in wastes and in their environments as a result of natural and artificial processes (Fig. 13). Also models and decision rules of risks (e.g., acceptable concentrations) change with accruing information.

The temporal development of exposures and risks could not be extensively analysed in the present work. Constant exposure concentrations and contact rates were assumed, as common (U.S. EPA 1986, 1989a, Table 6). The present estimates of release durations convey the order of magnitude. True duration may be greater due to accelerated leaching, or smaller due e.g. to its exponential level-off.

In a long-time perspective, risks may increase due to higher release rates of product-bound toxicants, to loss of attenuation capacity resulting e.g. from declining pH and organic content, as well as due to increased exposures following loss of societal recognition of the site (Okrent and Xing 1993). Long-time risk distributions are associated with the selection of containment and dispersal options (Diependaal et al. 1993). The estimated release at present emission level from a site during hundreds of thousands of years to deplete the pool implies some risk during extensive periods. Regardless of the decisions of risk

management strategies, analyses of the (temporal and spatial) distribution of toxicants and risks are valuable.

The results of older waste sites give indications of the temporal development of risk factors but may not be generalized. Unpredictable risks may arise e.g. through catastrophic emissions (Vandergrift and Ambrose 1988, Stuart et al. 1984). The accumulation of sporadic emissions was examined retrospectively on the basis of concentrations in solid media. Long-time impacts were analysed e.g. by conversion from acute to chronic toxicity (6.5, 6.6). However, there is much uncertainty of lagged risks.

Areal distributions of toxicants, cofactors, receptors and thus of risks are important with waste sites which by definition are local phenomena. These distributions are conceivable as representations (surfaces) of space-time continuums. At the source, hot spots may be present (Assmuth 1992). The environmental impact range depends on transport and fate, e.g. accumulation and persistence (cf. VII). Some impacts are global, e.g. those of CFCs (chlorofluorohydrocarbons) in landfill gas. Other persistent substances such as metals will disperse and contribute to global risks according to their relative abundance, dispersivity and potency. Further, the spatial range of exposed organisms is important for human and, particularly, for non-human effects (Clifford et al. 1995).

The areal distribution of toxicants and risks at many study sites were not quantified in detail. Violations of critical effect levels were observed up to ca. 1 km down stream, but may extend further. Toxicants in surface waters are distributed though natural or artificial routes (e.g., sewers), and concentrated e.g. in sediments and sludges. Longtime emissions of PCDD/PCDFs from a study site may have contributed to high levels in river sediments far downstream.

Extension of landfill impacts to ca. 1 km has been found abroad in ground water (Christensen et al. 1994). In Finland, impacts on ground water are not likely to be as extensive as in many other countries due to prevailing hydrogeological conditions (see discussion in III). However, they also imply little protective cover and dilution, and comparatively short lags before impact. Long-range subsurface migration may occur e.g. in bedrock, and toxicants may migrate undetected in dense non-aqueous phase liquids.

Toxicants in landfill gas migrate easier and in more uniform routes than those in water, but are also relatively rapidly diluted to background levels. Soils, sediments and other solids may be distributed by active transport e.g. in construction operations. Landfill animals transport toxicants over long distances (Assmuth 1983).

The estimated emissions from study sites seemed smaller than those from other sources (II, Assmuth et al. 1990b). However, the relative magnitude may change e.g. as controls are implemented on other fluxes and those from waste sites continue (for CFCs, see Laugwitz et al. 1990).

Even if risks of waste sites are local and of less absolute magnitude than risks in other areas, they may not be commensurable. For instance, a high local (health) risk is important for the individuals concerned, even if not as a population risk. The resolution of this political and ethical issue requires analysis of risk distributions and associated uncertainties (Finkel 1990a).

Attributability of risks

The attributability of impacts and risks particularly with mixtures of toxicants (Seiler and Scott 1987) adds to the difficulty of establishing that impacts and risks exist.

The attributability of risks was accounted for by subtraction of measured background values (of toxicant concentrations or impacts). Attributability was indirectly analysed by indicator variables, e.g. site-specific 'fingerprint' measurements (cf. Kerndorff et al. 1985).

At many study sites, there are several other sources of toxicants than the wastes. Industries producing the wastes have emitted waste constituents also in water, air, soil and other media (cf. Small et al. 1995). Emissions and impacts may also be due to more remote other (local) factors, e.g. other facilities. Thus, it is often difficult to ascertain the particular risks of a waste site. The issue is connected to the resolution of differences in distributions (Goodman 1986).

Models of toxicological risks of waste sites can not easily be verified as risks of well-defined events in extensive statistics (Suter 1993, see also the critique of verification by Oreskes et al. 1994). Traditional etiologic criteria for ascer-

taining cause and effect in Koch's postulates and its modifications (Suter 1993, Giesy et al. 1994a) are seldom met in ecological investigations (6.6).

Some general-level comparison and attribution of health risks is provided by apportionment of sources in applying ADI values (IPCS 1994). For toxicants like PCDD/PCDFs the attribution of exposures and effects to waste sites is particularly difficult due to their widespread background exposure e.g. in diet (Environ Dioxin Risk Characterization Panel 1995). From a risk management perspective however, even small excess emissions may be regarded unacceptable with such potent toxicants (Ahlborg et al. 1998).

6.9 Research, risk analysis and monitoring methodologies and needs

Sampling and measurement strategies

The spatial variation of toxicant concentrations in solid media and the temporal variation in more mobile media emphasize the difficulty in sampling and in estimating parameters of their distributions (VI, II, Assmuth 1992). For instance, the samples of interstitial pore water give a better reflection of the quality of larger waste volumes than solid samples do (Assmuth 1992), but have limited vertical representativity (Ettala 1990). A still more important consideration is the influence on risk estimates of toxicant distributions in site emissions and in the environment.

The screening strategy of chemical analysis is natural in investigation of waste sites, due to the lack of *a priori* information on toxicant occurrence and on risk factors. Specific chemical analyses are evolving rapidly. It seems feasible to accompany and direct them by indicator and group analyses and by initial measurements, also in field, of low sensitivity but good coverage.

Use of biotests

The characterization and assessment of leachates was aided by acute *Daphnia* lethality tests. Their value is illustrated by the inexplicable variation in relationships between impacts and chemical quality. The complexity of leachates limits bioassays but also justifies their use to supp-

lement chemical analyses. Biological measurements of waste site emissions are important since it is difficult to assess toxic responses to a mixture of (partly unknown) toxicants with chemical analyses only (Lambolez et al. 1994). Also during remedial actions, bioassays and biomarkers may give evidence of impacts and risks (or lack of them) not available with chemical analyses (Donnelly et al. 1992 and 1995).

However, there are important problems and development needs in applying biotests to the assessment of impacts also from waste sites (V). Problems are caused by both the technical conduct and interpretation of results. Particularly single-species acute lethality tests are a screening tool with considerable limitations. Suggestions for the use of such biotests have been given (V).

Risk analytical approaches and methods

The risk analytical approach in this work is partly based on earlier methodologies in toxicological risk analysis and on their application to waste sites (2). Some of the features of the present approach are original, such as the conceptual models of risk formation, the indices devised for comparative analysis, and applications of ecotoxicological risk assessment to chemical and biological data. The distributions of risk variables and associated uncertainties proved important for quantitative risk estimation and characterization. The emphasis on mixture effects and on arrays of end points was another crucial feature of the present analysis.

Methodologically, the present work emphasized descriptive measurement and theoretical modeling of risks. Risk estimates were mainly based on chemical occurrence and chemical-related data on one hand, and on biotests on the other. Instead of controlled experiments, inference was largely based on statistical and conceptual relationships. A more profound restriction is the reliance on measurements and models of the physical state of the environment; opinions about risks were not studied directly. These may give important information, but involve substantial inconsistency with nature (e.g., Assmuth 1983).

The approaches in the present work will be applicable to risk and uncertainty analysis of also other types of (local) chemical contamination.

However, the methodology of toxicological risk analysis for waste sites needs further development in connection with future empirical and theoretical research a.g. in the following topics:

- improved characterization of exposures together with measurements of biological responses in both humans and animals
- quantitative analysis of chronic toxicological risks from mixture effects
- uncertainty analysis of risks, e.g. by probabilistic methods, to complement point estimates
- theoretical concepts and models of risk formation acknowledging its multidimensionality
- risk analysis of ground water contamination and airborne emissions from waste sites.
- analyses of the spatial and temporal distributions of waste site impacts for risk management decision making and implementation

Comparative risk indices of toxicants

The indices developed serve as general descriptors of toxicological risks from waste sites, and as screening measures e.g. for the selection of toxicants in site-specific risk analyses.

A key methodological conclusion is that more attention should be paid to the distributions of toxicant concentrations, to fluxes, to environmental fate and to end points (VII). No previous report of index-based analysis methodology of waste site toxicants systematically and explicitly accounts for all of these classes of factors.

Substance-specific risk indices are affected by the choice, combination and weighing of component factors. Although based on relational scales, the final indices are interval-scale and relative measures of risk. The underlying models are somewhat arbitrary. The environmental fate factors may not be independent or expressible as mechanistic models. The indices also strongly depend on models of toxicity.

Availability and quality of data significantly affect risk indices. The lack of data may also exclude classes of toxicants from consideration, e.g. as the persistence of metals can not be assigned values. It emphasizes the need for several indices and judgment in interpreting them. This is related to the general need to characterize different kinds of risks by different measures. No single risk index, or model, is universally applicable. Otherwise e.g. very different

types of violation could give the same estimate for the same magnitude of the risk.

The proposed indices resemble the hazard indices (HI) of U.S. EPA (1989a). However, they do not address distributions and environmental fate. The relationship between estimated and acceptable doses (e.g., ADI values) is more comparable to the present indices.

The aggregate health risk indices (HI_{tot}) used in overall comparison of exposure routes (Table 22) represent a logical extension of the standard substance-specific indices. Comparable aggregate risk indices have been devised in other contexts (e.g., U.S. EPA 1989a). They may be used also to identify sites possessing particular risks, and further expanded e.g. to ecotoxicological effects.

Values of $HI > 1$ imply violation of health norms, and the absolute values of these indices display the magnitude of violation. The interpretation of CI is more complex. CI may be converted to estimates of cancer risks as implied by Brown and Donnelly (1988). Confidence in such estimates varies with data (Gold et al. 1993) and models. The procedure of Kerndorff et al. (1991) includes carcinogenicity only qualitatively.

Indicators and monitoring of waste site impacts

The suitability of substances as indicators of waste site impacts depends on the relationship between concentrations in emissions and those in the environment (Christensen and Kjeldsen 1984). The resolution of impacts depends on the deviation of the PDF of the variable in an impacted point from that in an unimpacted point (Goodman 1986). With different distribution statistics, different realizations of indicator indices are obtained (Kerndorff et al. 1992).

The suitable indicators in the present studies varied between sites. One may hypothesize a matrix of variables jointly defining the anomalies in environmental quality as compared with background quality. The main constituents chloride, ammonium, alkali metals and Zn often served as leachate indicators (I). Individual trace organic compounds are not general indicators due to their infrequent occurrence (Sabel and Clark 1984). AOCl measurements have been considered to indicate anthropogenic emissions (Kerndorff et al. 1985), but they may include naturally

formed compounds (Grimvall et al. 1989), and do not necessarily indicate the presence of single organochlorides significant at low concentrations (Levine and Kroemer 1989).

The covariations between toxicant concentrations in phases and between toxicants and main constituents aid in defining indicators of impacts. The distribution of substances within groups served as a tracer of contamination sources, e.g. PCDD/PCDF compounds associated with chlorophenols (IV, Wenning et al. 1993).

The use of several complementary approaches and analytical methods (including chemical and biological studies at species, population and community level in laboratory and in the field), and of staged procedures has been emphasized in investigations of hazardous waste sites (e.g., Burmaster et al. 1991). Such approaches are needed in view of the complex site characteristics. The present work has produced a framework for systematic monitoring and investigation of waste sites and their emission for the purpose of risk assessment to allow more informed risk management decisions and implementation.

7 SUMMARIZING CONCLUSIONS

1. The most abundant toxicants in waste sites varied between and within sites, but such toxicants generally included Zn, Cu, Cr and Pb, and chlorinated aliphatic and unchlorinated monoaromatic compounds. Other toxicants, including biomagnifying elements and chloroaromatic compounds, were identified more infrequently.

Several toxic organic compounds, especially chlorinated ethanes and toluene, were measured in landfill gas at concentration levels exceeding background levels by orders of magnitude. The central statistics varied between the sites, although they were approximately symmetrically distributed within a site.

The attenuation of many metals in wastes, as approximated by leachability indices based on acetate- and HNO_3 - extractable fractions, was associated with the pH and organic content of the matrix.

The rough estimates of the duration of the releases of toxic substances (heavy metals) in leachates before the easily leachable or total pool

will be mobilized, based on waste analyses and loading estimates, were tens to hundreds of thousands of years. This emphasizes longtime perspectives in risk assessment and management.

2. In leachate runoff from study sites many toxic substances were measured. Those commonly found included the heavy metals Zn, Cu, Cr and Ni, several chlorinated aliphatic and aromatic compounds (e.g., chlorophenols) and unchlorinated monoaromatic compounds.

The skewed distributions of toxicant concentrations and the frequency of non-detects affected the estimation and use of distribution statistics and imply the possibility of isolated significant emissions.

The estimates of toxicant fluxes in leachates are sensitive to variation in concentrations and flows, and to estimation methods. Specific fluxes (per unit waste volume) were greatest for abundant elements such as Zn. Some of the specific fluxes were statistically associated with general site properties such as technical status and industrial waste input. High specific fluxes at many municipal landfills reflect the impact of wastes containing hazardous substances.

3. Leachate runoff samples from waste sites were commonly acutely toxic to neonate *Daphnia magna* in acute tests.

Some concentration-response functions on a log-probit scale were linear, suggesting lognormality, but sublinear functions were also found, implying greater toxicity in low concentrations than expected from that in higher concentrations. The temporal development of toxicity varied between samples.

The frequent bacterial mutagenicity of leachates presents the possibility of genotoxic effects also to other taxa.

Toxicity to *Daphnia* correlated with the concentrations of chlorides which indicate the presence of other leachate constituents, and with concentrations of some heavy metals, which was plausible considering their reported toxicity. However, the absence of toxic responses in samples which contained substances at levels known to be toxic to *Daphnia* suggests immobilizing or inhibiting mechanisms, and illustrates the limits of both substance-based and biotest-based risk analysis.

4. Several toxic substances were measured in nearfield ground water down gradient of study sites. Those most consistently exceeding detection limits and background levels included Zn, Pb and chlorophenols.

The laboratory-measured saturated hydraulic conductivity of site soils exhibited considerable variation both within and between sites. High values reached levels where permeation of water is efficient and may enable transport of soluble toxicants in ground water.

Aquifer properties, toxicant occurrence in ground water and their implications for toxicological risks at waste sites could not be extensively elucidated on the basis of the present data.

5. 2378-chlorinated PCDD/PCDFs were identified particularly in solid samples from waste sites associated with chlorophenolic wood preservatives in spatially varying concentrations which are high in relation to many other contaminated sites. Substantial bioaccumulation in recipient fish was not observed in the limited analyses.

The isomer patterns at these sites, dominated by specific hexa- to octachlorinated PCDFs, suggested the sources of PCDD/PCDFs were impurities and transformation products in chlorophenolic wood preservative residues.

6. The compounds in leachate prioritized by risk indices included carcinogenic or otherwise highly toxic chlorinated compounds such as dieldrin, lindane, PCDD/PCDFs and PCBs. Considering leachate fluxes, Cu, Zn, Cd, PCBs, hexachlorobenzene and 2,4,6-trichlorophenol were important. Ranking depended on the distributions of observed concentrations, on environmental fate properties, and on effect criteria. Prioritization of compounds in landfill gas revealed differences between rankings by various criteria, e.g. tumorigenicity vs. other toxicity or air quality norms.

Ecotoxicological risks of pentachlorophenol were analysed on the basis of relationships between observed distributions of concentrations and published toxicity data. By extrapolation from the loglinear distribution of LC₅₀ for fish and from acute toxicities to NOAEC levels, critical concentrations for hypothetical most sensitive species were estimated. The major source of uncertainty was due to extrapolation using safety factors to no-effect levels, reaching to values below typical observed concentrations.

The observed toxicity to *Daphnia* and predicted toxicity based on measured concentrations and published toxic values correlated only when flow weighting or toxic fluxes were used. This illustrates the insufficiency of biological or chemical methods as the sole assessment base.

The environmental health risks of studied toxicants and waste sites varies depending on measurements and models. Risk estimates include uncertainties due e.g. to the lack of coverage of potentially important substances and routes, to exposure and effect data, and to attributing toxicant occurrences to waste sites. Occasionally toxicant concentrations in surface and ground water near study sites exceeded guideline values by several hundred fold, even if not in exposure media. Local health risks may be caused by prolonged exposure to such concentrations.

Risks from PCDD/PCDFs at sites related to chlorophenolic wood preservatives depended on the definition of the spatial distribution of the isomers e.g. in soil and of their relative toxicities, as analysed by various toxic equivalent systems for mammal and fish effects. Worst-case estimates of excess human cancer risks from PCDD/PCDFs reached levels of unity, while more plausible but prudent estimates approached background levels, due to consideration of other exposure and response factors. Dioxin-like compounds also cause risks of ecotoxicological risks, e.g. adverse developmental effects to sediment-feeding organisms in waste site recipients.

The importance of toxicant distributions and of multiple impact models for the assessment of waste sites was demonstrated. The temporal, spatial and structural coverage of measurements in combination with risk analysis is to be increased for better resolution of the time-space variation of risks attributable to waste sites.

Important uncertainties and research needs include ground water mediated and airborne risks of waste sites, e.g. longtime impacts of toxicant pulses and mixtures on humans and ecosystems.

The measures serving as toxicant indicators at waste sites depended on relationships between waste qualities and environmental conditions. Combinations of physicochemical variables commonly associated with leachates, group and specific analyses of toxic substances, and biotests are to be selected for monitoring on a site-specific basis and applied in a staged manner.

ACKNOWLEDGEMENTS

These studies have been carried out at the Water and Environment Research Institute of National Board of Waters and the Environment (presently Finnish Environment Agency) in cooperation with other units of the National Board and with the Water and Environment District Offices. During the core work, liaisons were established with the National Institute of Public Health and Geological Survey of Finland. Many site investigations were carried out in collaboration with consulting firms, site owners or authorities.

My fellow researchers Tapio Strandberg, Kirsti Kalevi and Sirpa Penttilä at the Water and Environment Research Institute and Terttu Vartiainen at the National Public Health Institute participated in the conduct of the empirical studies and in parts of some of the original articles; Terttu Vartiainen also offered valuable criticism of a draft of the summary. Several assistant researchers and technicians in our waste research group contributed as well, including Pertti Järvelä, Mirja Castren, Leena Laitinen and, particularly, Helena Dahlbo who also took part in the management of empirical data. I am indebted to all of them.

Among other employees of the Water and Environment Research Institute, valuable assistance was willingly offered by Markku Liponkoski in computer operations, Terttu Halme in kindly and efficiently drawing figures and in the overall layout of this publication, Heikki Jyllilä in field work, Päivi Laaksonen and Meri Kostian in many office duties, the staffs of our Laboratories in physico-chemical analysis, and the staff of our library in efficiently and patiently retrieving information. Hannu Laikari and Seppo Mustonen as my chiefs during most of the work provided essential general support; Seppo Mustonen also commented on a draft of this summary. Pertti Heinonen checked a draft very precisely and found many errors in references. Heikki Pitkänen gave advice for the dissertation procedures. In the last stages of the work, colleagues in the Chemicals Division of Finnish Environment Agency, particularly Esa Nikunen and Jukka Malm, provided fruitful input and support. Mikael Hilden gave valuable comments on the manuscript to clarify risk concepts, along with overall inspiration. I wish to thank these and other helpful colleagues. My special thanks go to Matti Melanen for his continuous encouragement and advice.

The contact persons and many other staff members at all Water and Environment District Offices, too numerous to mention individually, gave valuable assistance in the field, in laboratories and in offices. I am indebted to all of them.

Many authorities at several provincial governments and municipalities are also to be acknowledged for their contributions.

The laboratory analyses were mainly accomplished by the staffs at the Research Laboratory and the Soil and Waste Laboratory of our Institute, at the Water and Environment District Offices, at the National Institute of Public Health in Kuopio, and at the Geological Survey of Finland. The personnel of several other laboratories in many organizations, including municipalities and firms, also provided valuable analytical services.

Matti Vehkalahti and Klaus Pfister at the Ministry of the Environment are to be credited for support in their capacity of supervisors for much of the empirical study projects. Also Rainer Lahti provided support during most stages of the work. In particular, I am indebted to Ari Seppänen for his involvement and support, initially as a fellow researcher, later as a supervisor and as a partner in work related to risk management of waste sites.

I wish to thank my professor Pekka Nuorteva for his lasting inspiration and support and Martin Lodenius for promoting the final stages of the work. I also wish to acknowledge the input from the referees of the original articles and of the manuscript of this summary, particularly Matti Ettala and Helena Mussalo-Rauhamaa. I am indebted to many other persons as well, both in Finland and abroad, who have influenced and helped this work. The English language of a draft of the summary was swiftly checked by Donald Smart.

These studies have been funded primarily by the Ministry of the Environment and the National Board of Waters and the Environment. For the final part of the work financial support was offered by the Jenny and Antti Wihuri Foundation. I wish to thank warmly all these organizations for their donations.

Finally, I want to express my most sincere gratitude to my wife Veronica and to my children Kimmo, Lari and Sarah for support, patience, inspiration and love that was essential in making this work possible and worthwhile.

I dedicate this work to the memory of my father.

Helsinki, December 1995
Timo Walter Assmuth

SELOSTUS

Työssä tutkittiin haitallisten aineiden esiintymistä, pitoisuuksia ja jakaumia suomalaisilla jätealueilla, niiden suotovesien myrkyllisyyttä ja niiden ympäristössä aiheuttamia toksikologisia riskejä.

Tutkimuksissa tehtiin kemiallisia, fysikaalisia tai biologisia mittauksia vesistä, maaperästä, jätteistä, sedimenteistä, kaasuista ja eliöistä. Näytteitä otettiin 43 kohteesta, jotka sisältävät erilaisia kunnallisia ja teollisuuden kaatopaikkoja, muita jätealueita ja kemiallisesti saastuneita alueita eri puolilta maata.

Useimpien analysoitujen haitallisten aineiden pitoisuudet painottuivat pieniin arvoihin erityisesti vesinäytteissä. Monet suotovesinäytteiden pitoisuusjakaumat olivat lognormaalisia, ja katkaistuja johtuen määritysherkkyuden alittavista mittauksista. Jakaumien ominaisuudet vaikuttivat huomattavasti havaintoaineistojen tilastolliseen käsittelyyn ja estimointiin.

Jätenäytteissä todettiin useiden raskasmetallien liukoisten pitoisuuksien ja kokonaispitoisuuksien suhteen korreloivan organisen aineen pitoisuuteen ja pH-arvoon. Pidäytyminen ja pitoisuuksien vaimeneminen jätteestä ympäristöön vaihteli aineen ja kohteen mukaan. Jätetilavuutta kohti lasketun ominaiskuormituksen laskeva suuruusjärjestys oli sinkki > kromi > nikkeli. Eräiden aineiden ominaiskuormitus oli yhteydessä jätealueiden yleisiä ominaisuuksia kuvaaviin muutujiin, mutta nämä yhteydet eivät olleet yksiselitteisiä. Haitallisten aineiden päästöjen kestoksi yhdyskuntajätteen kaatopaikoilta arvioitiin jätteen liukoisen fraktion ja kokonaispitoisuuden sekä nykyisen päästötason perusteella 10^4 - 10^6 vuotta aineesta (metallista) riippuen.

Vedenlaadun terveydelliset pitoisuusrajasuositukset ylittyivät yleisesti pinta- ja pohjavesinäytteissä tutkimuskohteiden lähellä; voimakkaita ylityksiä aiheuttivat erityisesti eräät metallit ja torjunta-aineet. Kaatopaikoilla kehittyvässä kaassussa kloorimetaanien ja yksinkertaisten aromaattisten yhdisteiden korkeat pitoisuudet ylittivät hengitysilman terveydelliset pitoisuusrajat.

2,3,7,8-kloorattuja PCDD/PCDF-yhdisteitä (dibentso-p-dioksiineja ja furaaneja) mitattiin suhteellisen korkeina pitoisuuksina ja tunnusomaisissa suhteissa maaperässä ja sedimenteissä jätealueilla, joilla on käsitelty ja sijoitettu kloorifenolipitoista puunsuojausainetta.

Useimpien tutkimuskohteiden suotovedet olivat myrkyllisiä *Daphnia magna* -vesikirpulle 24 tunnin testissä, ja mutageenisia *Salmonella typhimurium* -bakteerikannoille varsinkin entsyymaattisella aktivoinnilla. Myös monet lähemmin tutkituista näytteistä olivat myrkyllisiä 48 tunnin *Daphnia*-standarditestissä; pitoisuus-vastesuhteiden muoto ja ajallinen kehitys vaihtelivat näytteiden välillä, noudattaen usein suurin piirtein lognormaalia jakaumaa. 48h-EC(I)50 -arvot korreloivat suotovesien voimakkuutta indikoivaan kloridipitoisuuteen ja eräiden myrkyllisten aineiden (metallien) pitoisuuksiin lineaarisissa regressiomalleissa. Aina ei kuitenkaan ollut havaittavissa myrkyllisyyttä, vaikka näytteiden haitallisten aineiden pitoisuudet ylittivät puhasainekokeissa myrkylliset pitoisuudet.

Käsitteellisiä, tilastollisia ja mekanistisia riskianalyysimalleja hahmoteltiin kirjallisuudessa esitettyjen metodologisten lähestymistapojen pohjalta. Malleja sovellettiin analysoitujen haitallisten aineiden priorisointiin suotovesi- ja kaasuaineistoissa käyttäen lineaarisia indeksifunktioita, sekä eräiden aineiden ja vaikutusten yksityiskohtaiseen arviointiin.

Kadmium, nikkeli ja monet klooratut hiilivedyt olivat tärkeitä haitallisia aineita suotovesissä ottaen huomioon pitoisuusjakaumat, pysyvyyden tai bioakkumuloituvuuden, syöpävaarallisuuden ja terveydelliset ohjearvot; ekotoksikologisia riskejä liittyi erityisesti ravintoketjuissa rikastuviin aineisiin. Kokonaisriski-indeksien arvot, jotka saatiin laskemalla yhteen eri aineiden indeksit, olivat korkeampia pohjavedessä (jätealueiden lähellä) kuin pintavedessä. Kaatopaikkakaasussa suurimmat terveysriskit liittyivät ilmanlaatonormien perusteella arvioiden kloroformiin; etyleenidibromidi oli huomattavin kaasufaasista mitattu karsinogeeni.

Arvio pentakloorifenolin pitoisuusjakauman ekotoksikologista riskeistä muuttui huomattavasti, kun haitattomat pitoisuudet ekstrapoloitiin sekä lajien herkkyysvaihtelun perusteella että altistuskestojen välillä, erityisesti kun käytettiin arvioinnissa suositeltuja turvakertoimia.

Havaittu myrkyllisyys *Daphnia*lle korreloi ennustettuun myrkyllisyyteen, joka määritettiin mitattujen pitoisuuksien ja kirjallisuudessa esitettyjen toksisuustietojen perusteella, vain kun käytettiin virtaamalla painotettuja pitoisuusarvoja ja toksisen kuormituksen suureita. Muiden säännömukaisten yhteyksien puute viittaa tunte-

mattomiin myrkyllisyyttä vähentäviin tai lisääviin tekijöihin, ja osoittaa sekä kemiallisten että biologisten mittausten merkityksen jätealueiden vaikutusten ja riskien tutkimuksessa.

PCDD/PCDF-yhdisteiden myrkyllisyysarvio riippui toksisuusevivalenttijärjestelmästä ja sen pohjana olevasta eliöstä ja vaikutuksesta; kalavaikutuksiin ja entsyymiaktiivisuuksiin perustuvat ekvivalenttipitoisuudet olivat keskimäärin korkeampia kuin nisäkävaikutuksiin perustuvat.

Arviot syövän lisääntymisen todennäköisyydestä PCDD/PCDF-yhdisteiden saastuttamilla puunsuojausainejätteitä sisältävillä alueilla laskivat pahimman skenaarion hyvin korkeista arvoista taustatasolle käytettäessä uskottavampia oletuksia altistustekijöistä (saastuneen maan ja kalan syönnissä) ja vaikutuksen voimakkuudesta. Myös arviot kroonisen annoksen ja sallitun annoksen suhteesta vaihtelivat paljon.

Tulosten tarkastelussa kiinnitettiin erityistä huomiota vaikutusten ja riskien havaitsemiseen ja syiden erittelyyn sekä tässä esiintyvään epävarmuuteen. Sen merkitystä korostaa riskimuuttujien monilukuisuus ja monitahoisuus sekä vaikuttavien tekijöitä koskevien tietojen puute. Haitallisten aineiden, niiden päästöjen ja vaikutusten sekä muiden riskimuuttujien jakaumat ovat siksi tärkeä osa jätealueiden toksikologisten riskien arviointia.

Tärkeitä jatkotutkimusaiheita ovat jätealueiden aiheuttama pohjavesien ja ilman pilaantuminen, eliöiden altistusastot, pitkän aikavälin yhteisvaikutukset terveyteen ja ekosysteemeihin, sekä vaikutusten ja riskien ja niiden ajallisten ja alueellisten jakaumien yhteydet riskinhallinnan päätöksentekoon ja toteutukseen mm. jätealueiden kunnostuksessa. Näissä ja muissa yhteyksissä voidaan hyödyntää tutkimuksessa kuvattuja lähestymistapoja, menetelmiä ja malleja. Tutkimuksen ja seurannan menetelmissä painotettiin vaiheittaista toteutusta, jossa yhdistetään biologisia ja kemiallisia mittauksia teoreettiseen tarkasteluun.

LIST OF SYMBOLS AND ABBREVIATIONS

used repeatedly or without explanation in the text (outside tables)

Symbol/ abbreviation	Unit(s) of measure	Meaning
AAS		Atomic absorption spectrometry
ADI	mg kg ⁻¹ d ⁻¹	Acceptable daily intake
AOCl		Adsorbable organochlorides
AOX		Adsorbable organohalides
As		Arsenic
b, b _i	varies	Potency (carcinogenic) (of substance i or tumorigenesis stage i)
b ₁	ppm ⁻¹ d	Potency of carcinogen estimated by LMS model (1st order term)
b ₁ [*]	ppm ⁻¹ d	95 % UCL estimate of carcinogenic potency by the LMS model
BCF	dimensionless	Bioconcentration factor
c, c _i	varies	Concentration (of toxicant i)
c _D , c _i ^D	(µg l ⁻¹)	Concentration (of toxicant i) in discharge route
Cd		Cadmium
CDI	g kg ⁻¹ d ⁻¹	Chronic daily intake
CDF		Cumulative distribution function
c _E , c _{ij} ^E	(µg l ⁻¹)	Concentration of toxicant (i) at point of exposure (along route j)
CI	(kg l ⁻¹ d)	Carcinogenic risk index
c _i (i)	µg g ⁻¹	NH ₄ Ac-extractable concentration (of metal i in solid sample)
CPs		Chlorophenols
Cr		Chromium
CR _(j)	m ³ d ⁻¹	Contact rate of medium (in path j)
c _{tot} (i)	µg g ⁻¹	HNO ₃ -extractable concentration (of metal i in solid sample)
Cu		Copper
D, D _{ij}	varies	Dose (of toxicant i along exposure route j)
12-DCE _a		1,2-Dichloroethane
DCM		Dichloromethane
D _(i) ^E	mg kg ⁻¹ d ⁻¹	Effective dose (of toxicant i in target tissue(s))
DF		Distribution function
DL	varies	Detection limit
D-R		Dose-response (model or function)
EC	varies	Effective concentration
EC(I)50	varies	Effective inhibitory concentration to 50 % of test animals
ECI	dimensionless	Effective (environmental) cancer risk index
ED ₅₀	varies	Effective dose to 50 % of exposed organisms
EHI	dimensionless	Effective (environmental) health risk index
ETI	dimensionless	Effective (environmental) ecotoxic risk index
F, F _i	g d ⁻¹ (etc.)	Flux (of substance i)
f ^{BA}	dimensionless	Bioavailability factor
f _{Dil}	dimensionless	Dilution factor
f _T	dimensionless	time factor (in exposure analysis)
F _V	g a ⁻¹ m ⁻³	Specific flux (per unit waste volume)

Symbol/ abbreviation	Unit(s) of measure	Meaning
GC		Gas chromatography
H	Pa m ⁻³ mol (/ none)	Coefficient of Henry's law
Hg		Mercury
HI	dimensionless	Hazard index or (in the present work) health risk index
HI _{tot}	dimensionless	Total health risk index
24h-leth	% (of sample)	Lethality (of <i>Daphnia magna</i>) in 24 h
48h-EC50	varies (% of sample)	48 hour effective concentration to 50 % of test animals
HNO ₃		Nitric acid
1234678-HpCDF		1,2,3,4,6,7,8-Heptachlorodibenzofuran
123478-HxCDF		1,2,3,4,7,8-Hexachlorodibenzofuran
I		Impact
I-TE, I-TEQ	varies	International TCDD toxicity equivalent (concentration or quantity)
I-TEF	none (fraction)	International TCDD toxicity equivalent factor
K _c	m s ⁻¹	Saturated hydraulic conductivity
K _D	d ⁻¹	(First-order) decay constant
K _{ow}	dimensionless	Octanol-water partition coefficient
LC50, LC ₅₀	varies (conc.)	Lethal concentration to 50 % of test animals
LEC, LEC	varies (conc.)	Lowest effective concentration
LF ^{obs}	(% l s ⁻¹)	Lethal flux (to <i>Daphnia magna</i>)
LI _i	%	Leachability index of substance i
LMS		Linearized multistage (model of carcinogenesis)
LOAEC	varies (conc.)	Lowest observed adverse effect concentration
LOAEL	varies	Lowest observed adverse effect level
LQG	varies	Lowest relevant environmental quality guideline
M	varies	magnitude (severity) of adverse event or impact
m _B	kg	Body weight
MOS	varies	Margin-of-safety
MST	h	Median survival time
NH ₄ Ac		Ammonium acetate
Ni		Nickel
NOAEC	(concentration)	No observed adverse effect concentration
NOAEL	varies	No observed adverse effect level
OCDF		Octachlorodibenzofuran
p	none (fraction)	Probability
PAHs		Polyaromatic hydrocarbons
Pb		Lead
PCBs		Polychlorinated biphenyls
PCDD/PCDF(s)		Polychlorinated dibenzo-p-dioxin/polychlorinated dibenzofuran(s)
PDF		Probability density function
PeCBz		Pentachlorobenzene
PeCP		Pentachlorophenol
Q	l s ⁻¹	Flow (of water) (instantaneous or average)

Symbol/ abbreviation	Unit(s) of measure	Meaning
r	none (fraction)	Pearson's linear coefficient of correlation
$R_{(ijk)}$	dimensionless	Risk (of substance i in transport route j and exposure route k)
R_c	dimensionless	Carcinogenic risk (lifetime excess cancer risk)
R_e, R_i^e	dimensionless	Ecotoxicological risk (of toxicant i)
R_h	dimensionless	Health risk (excluding carcinogenicity)
R_0	dimensionless	Background risk
SF	dimensionless	Safety factor
$t_{1/2}$	d	Half-life
t_A	a	Averaging time (in exposure analysis)
TA97, TA100		<i>Salmonella typhimurium</i> strains TA97, TA100
TCDD		2,3,7,8-Tetrachlorodibenzo-p-dioxin
246-TCP		2,4,6-Trichlorophenol
t_E	a	Exposure duration
TeCE _e		Tetrachloroethene (Perchloroethylene)
2346-TeCP		2,3,4,6-Tetrachlorophenol
TF ^{obs}	(% ⁻¹ l s ⁻¹)	Observed toxic flux (to <i>Daphnia magna</i>) based on EC50 values
TF ^{pred}	(l s ⁻¹)	Predicted toxic flux (to <i>Daphnia magna</i>) (cf. T ^{pred})
TI	dimensionless	Toxic risk index (of substance)
TI _{tot}	dimensionless	Total toxic risk index (sum of substance-specific indices)
TM		Trade mark
T ^{pred}	dimensionless	Predicted toxicity based on addition of the quotients of observed toxicant concentrations and substance-specific literature values
t ^R	a	Estimated time of emission duration
TRF	(m ³ d ⁻¹)	Toxic risk flux
UCL	varies	Upper confidence limit
V _w	m ³	Volume (of waste)
χ^2	dimensionless	Chi ² (distribution statistic)
Zn		Zinc

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Summarized publications

This publication is a synthesis and an extension of the following articles, referred to in the text by their Roman numerals:

- I Assmuth, T. 1992. Toxic chemical concentrations in waste deposit runoff. *Aqua Fenn.* 21(2):183-194. ISSN 0356-7133.
- I Assmuth, T. & Kalevi, K. 1992. Concentrations and toxicological significance of trace organic compounds in municipal solid waste landfill gas. *Chemosphere* 24(9):1207-1216. ISSN 0045-6535.
- III Assmuth, T.W. & Strandberg, T. 1993. Ground water contamination at Finnish landfills. *Water, Air, and Soil Pollut.* 69:179-199. ISSN 0049-6979.
- IV Assmuth, T. & Vartiainen, T. 1994. Concentration patterns of 2,3,7,8-chlorinated dibenzo-p-dioxins and dibenzofurans at landfills and disposal sites for chlorophenolic wood preservative wastes. *Chemosphere* 28(5):971-979. ISSN 0045-6535.
- V Assmuth, T. & Penttilä, S. 1995. Characteristics, determinants and interpretations of acute lethality of daphnids exposed to complex waste leachates *Aquat. Toxicol.* 31:125-141.
- VI Assmuth, T.W. & Vartiainen, T. 1995. Analysis of toxicological risks of local contamination by PCDD/PCDFs - importance of isomer distributions and toxic equivalents. *Chemosphere* 31(3):2853-2861. ISSN 0045-6535.
- VII Assmuth, T.W. Comparative risk analysis of waste site toxicants by indices based on concentration distributions, environmental fate, fluxes and effect modes (accepted for publication in *Journal of Hazardous Materials*).

ERRATA AND ADDENDA

Assmuth, T. 1995. Toxicant distributions and impact models in environmental risk analysis of waste sites. Publ. Water Environ. Res. Inst. 20, p. 1-79.

- p. 22, column 1, chapter 1 in Leachate runoff and surface water, row 7: '3.8' should read '2.8'
- p. 26, column 2, chapter 1 in Structural representativity, row 2: 'median size of the sites were' should read 'mean size of the sites was'
- p. 31, column 2, chapter 2 in Ground water, row 1: 'median and maximum' should read 'median or maximum'
- p. 40, column 1, chapter 3 (excluding equations), row 5 and 7: 'Lewis 1991b' should read 'Lewis 1991'
- p. 44, Table 24, column heading: 'I-TEQs' should read 'TEQs'
- p. 44, Table 24, footnotes, row 1: '(Safe 1993)' should read '(Safe 1992)'
- p. 46, column 2, chapter 1, row 3: 'by' should be deleted
- p. 46, column 2, chapter 1 in Temporal and spatial distributions, row 1: 'distribution' should read 'distributions'
- p. 58, column 2, chapter 1 in Summarizing conclusions, row 6: 'biomagnifying elements' should read 'other elements'
- p. 60, column 1, chapter 3, row 12: 'risks of ecotoxicological risks should read 'risks of ecotoxicological impacts'
- p. 63, 3rd row from bottom: 'Fe <TAB> <TAB> iron' should be added
- p. 64, 2nd row from the bottom:
'PNEC <TAB>varies <TAB>predicted no-effect concentration' should be added
- p. 66, column 1, reference 6, row 1: 'Assmuth, T. 1993' should read 'Assmuth, T. 1983'
- p. 68, column 2, reference 4, row 3: 'Thomas, N.A. & Paquin, P.R. 1991' should read 'Thomas, N.A. & Paquin, P.R. 1994'
- p. 70, column 2, reference 7, row 1: 'Hemminki, K. & Vineis, P.' should read 'Hemminki, K. & Vineis, P. 1985.'
- p. 72, column 1, reference 13 (Lewis, R.J. Sr. 1991a), 3 rows: should be deleted
- p. 72, column 1, reference 14, row 1: 'Lewis, R.J. Sr. 1991b' should read 'Lewis, R.J. Sr. 1991'
- p. 75, column 1, reference 5 (Reichard, E.G. & Evans, J. 1989), 4 rows: should be deleted
- p. 77, column 1, reference 10 (Wolfinger, T.F. 1989), 3 rows: should be deleted
- p. 79, 2nd of summarized publications: 'I' should read 'II'