

FLUXES OF Cu, Zn, Pb, Cd, Cr, AND Ni IN TEMPERATE FOREST ECOSYSTEMS

A Literature Review

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Abstract. The literature on the fluxes of six heavy metals in temperate forest ecosystems is reviewed. Special attention is given to wet and dry deposition and internal flux, to metal budgets for ecosystems and soils, to concentrations in aqueous compartments of the ecosystem and to speciation in soil solutions. Metal fluxes are discussed in relation to pollution load, soil type, tree species and land use. The mobility of Cu and Pb is strongly dependent on the solubility of organic matter. These metals are commonly accumulated in forest soils. Zinc, Cd and Ni are greatly influenced by soil acidity and are often lost in considerable amounts from acidified soils. Chromium is often at balance in forest ecosystems. Implications for metal solubility and budgets in forest soils are discussed in connection with an increase in soil acidification.

1. Introduction

Knowledge of the transfer and budgets of metals in forest ecosystems is important for the understanding of the function of ecosystems. The amounts of metals, especially those available in trace quantities, that are cycled through different compartments of the forest ecosystems are still poorly known. Such knowledge is necessary in estimating cycling rates and long-term effects on biological systems of metals from atmospheric deposition and in natural soil pools. The prevailing soil acidification (Butzke, 1981; Falkengren-Grerup, 1986; Tamm and Hallbäcken, 1986) will increase the release and leachability of many elements in soil (Norton, 1977; Bergkvist, 1986b), those of anthropogenic as well as those of natural origin. The degree to which the acidic deposition has increased the soil acidity varies according to, e.g., base cation reserves, weathering rate and the amount and duration of acidic deposition.

In the biologically most active part of the soil system – the organic top soil – the biological activity has been shown to be highly sensitive to heavy metal pollution (Tyler, 1972, 1976a; Rühling and Tyler, 1973). It is necessary to know the metal budgets in this horizon to be able to estimate the risk for adverse effects on nutrient mineralization, maybe also on primary productivity.

The loss of metals from the entire soil profile is also of great concern. Ulrich (1975) claimed that there is a risk of Mg deficiency in trees due to increased Mg loss from acidified soils. Raisch (1983) found Mg and Zn deficiency to prevail in

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five spruce-forest sites in the Black Forest, southwestern FRG. There is clear evidence of diminishing nutrient pools in northeastern USA (Norton *et al.*, 1980), central Europe (Raisch, 1983) and S Scandinavia (Nilsson, 1985; Falkengren-Grerup *et al.*, 1988).

Forest decline is becoming a widespread phenomenon in Europe (see Breloh and Dieterle, 1985, for the situation in FRG) and part of northeastern USA (Papke *et al.*, 1986). In parts of central Europe nutrient deficiency seems to be a main factor in forest decline (Zöttl, 1985; Zöttl and Hüttl, 1986) and fertilization with, e.g., Mg and Zn has been shown to improve growth and vigour rapidly.

Soil acidification is also considered to raise the soil-solution concentration of Al and heavy metals to levels that are toxic to tree roots (Ulrich, 1983; Matzner *et al.*, 1986).

The objectives of this literature review on Cu, Zn, Pb, Cd, Cr, and Ni are:

- (i) to compile and evaluate literature data on the deposition of these metals to temperate forest ecosystems;
- (ii) to quantify metal fluxes through the ecosystems;
- (iii) to calculate metal budgets for the ecosystems and for the soils;
- (iv) to compile data on metal concentrations in aqueous compartments of the ecosystems;
- (v) to discuss the speciation of metals in the soil solution; and
- (vi) to identify fields where more research is needed.

2. Materials

The search for data included only European and N American studies. Where a research group has published many reports from the same field sites, only the major reports have usually been included. In many cases, internal reports, etc., had to be consulted, however. Reports containing data only on deposition are gathered in Table I, others in Table II.

Metal budgets for forest ecosystems have been produced in different ways by different authors. In Table II two types of budgets have been calculated. An ecosystem budget is defined as the difference between the total deposition to the canopy and the amount that leaves the ecosystem, either with the soil percolate below the rooting zone or with the output from the catchment. Where the ecosystem budget is positive, the ecosystem thus accumulates the metal and acts as a net sink for the metal. A negative budget is correspondingly associated with metal release from the ecosystem which acts as a net source. Where possible, a soil budget has been calculated from the input to the forest floor minus the output from the soil (soil solution under the rooting zone, accumulation into above-ground biomass and root uptake) Data on all variables are seldom given in the same report. The values given in Tables I–III are usually mean values or the range of mean values.

TABLE I
Bulk and dry deposition (g ha⁻¹ yr⁻¹)

Site	Cu	Zn	Pb	Cd	Cr	Ni	Remarks
<i>F.R.G</i>							
Göttingen	104	470	229	3.8	91	24	Ruppert (1975). Bulk deposition. Urban
Solling	236	1377	285	15.9	14	27	Mayer (1981). Bulk deposition.
Lüneburger Heide	113	-	143	11.2	-	-	1969-1979
Solling	8.8	-	48	0.88	-	-	Schultz (1985). Bulk deposition
Harste	5.5	-	27	0.58	-	-	1983-1984
Spanbeck	6.9	-	27	0.66	-	-	
Teutoburger Wald	520	440	215	3.6	-	-	Godt et al. (1985). Bulk dep. 1982-1984
Göttinger Stadtwald	26	240	118	1.9	-	-	Meiwees (1985). Bulk dep. 1982-1983
Essen	-	-	620	15	-	-	Georgii et al. (1982). Bulk deposition.
Deuselbach	-	-	150	3.5	-	-	1979-1981
Schaumsland	-	-	150	16	-	-	
Bärhalde	18	210	110	4.5	-	-	Raisch and Zöttl (1983). Bulk dep. 1977-78
Obertheinebene	16	143	86	2.1	-	8-11	Trübny (1983). Bulk dep. 1978+79
Rural areas	32	140	110	2.7	-	-	Nürnberg (1983). Wet-only deposition.
Goslar	80	-	365-730	7.3	-	-	1980-1981
Urban/rural	34/38-89	170/220-5850	130/170-540	3.2/4.0-28	-	-	Nürnberg et al. (1984). Bulk deposition.
Bavaria	240-470	800-1410	260-420	5.8-13.6	-	-	Hantschel et al. (1985). Bulk deposition.
<i>Switzerland</i>							
Different pollut. load	-	-	18-60	0.2-1.4	-	-	Keller and Flückiger (1985). Bulk deposition.
<i>Czechoslovakia</i>							
Zvolen	40	-	-	-	-	-	Kabata-Pendias and Pendias (1984). Bulk deposition.
<i>Poland</i>							
Baltic coast	25-53	-	280-340	4.4-23	-	21-47	Szefer and Szefer (1986). Dissolved + suspended. Three stations. Yearly means. 1976, 1978-80.

Table I (continued)

Site	Cu	Zn	Pb	Cd	Cr	Ni	Remarks
<i>Hungary</i>							
Central H., rural	-	-	50-140	5.5-8.7	-	-	Mészáros <i>et al.</i> (1987). Wet-only deposition.
<i>United Kingdom</i>							
England	-	-	270	40	-	-	Cawse (1974). Bulk deposition. Urban.
	-	360	250	60	-	-	Bulk deposition. Rural.
<i>Europe</i>							
Urban	78-500; 320	520-1900; 1000	180-640; 400	-	-	33-530; 310	Jeffries and Snyder (1981). Bulk deposition.
Rural	14-320; 150	38-3900; 550	63-550; 220	2-13; 4	-	7-100; 32	Range; median. Large literature review
<i>N. America</i>							
Canada, Great Lakes	-	-	20-70	0.5-2.0	-	-	Reid and Lulis (1987). Wet-only dep. 1981-84.
Can., Ontario	11-14	38-73	21-61	0.8-1.0	-	4.0-5.0	Chan <i>et al.</i> (1986). Wet-only deposition.
	3.0-4.0	5.1-15	11-42	0.28-0.42	-	1.8-2.8	Dry deposition. 1982. 36 sites.
N. America	20-980; 160	80-4800; 3200	140-3500; 910	7-36; 18	-	-	Jeffries and Snyder (1981). Bulk deposition. Urban.
<i>U.S.A., Tennessee</i>							
	13-79; 37	93-970; 470	17-320; 170	1-8.8; 6.7	-	-	Rural. Range; median. Large literature review.
	-	76	72	4	-	-	Lindberg and Harriss (1981). Bulk deposition.
<i>U.S.A., Indiana</i>							
	-	17	84	1	-	-	Dry deposition. 1977.
	-	-	1400	15	-	-	Peyton <i>et al.</i> (1976). Bulk deposition. Urban.
	-	-	150	3	-	-	Rural.
<i>U.S.A., New Hampshire</i>							
	-	-	20	9	-	-	Schlesinger <i>et al.</i> (1974). Bulk deposition.
<i>U.S.A., New Hampshire</i>							
	-	-	317	-	-	-	Siccoma and Smith (1978). Bulk deposition.
<i>U.S.A., California</i>							
	>32	>106	-	-	-	-	McColl (1981). Wet + dry deposition. 1978-79.

TABLE IIa
Mean annual flux of Cu (g ha⁻¹ yr⁻¹)

Ecosystem/site	Bulk dep.		Dry dep.	Through-fall			Stem-flow	Litter-fall		Soil solution horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake		Ecosystem budget	Soil budget	Remarks
	A	B		C	D	E		F	G					H	I			
F.R.G.																		
1 Beech/Solling	236	234		142	75	175	20	240	106				310	77	364	-256		1. Mayer (1981, 1986), 11/1974-8/1979, Beech 125 yr, spruce 90 yr, standing crop, 311 and 324 ton/ha, resp; acid braunerde, Moder, pH 3.9 - 4.3, loess loam, clay 16 - 19%. F=zero-tension lysimeter, G=tension lysimeter, 80 cm.
Spruce/Solling	236	423		227		157			110				300	108	549	-51		2. Ulrich <i>et al.</i> (1979), Heinrichs and Mayer (1980), Mayer and Heinrichs (1980); 11/1974-10/1977, B=calc. from 1; G=tension lysimeter, 80 cm.
2 Beech/Solling	350	347		130	75		18	240	110				310	77	587	-274		3. Schultz <i>et al.</i> (1986), Schmidt (1987), 1 yr, 1983-1985, a=incl. in TF.
Spruce/Solling	350	627		230					110				300	108	867	-48		4. Schmidt and Schultz (1985), 1983, summer=May to Oct., winter=Nov. to Apr.
3 Beech/Solling	24	61		62	33		a							14				5. Asche (1985); 2 yr, 1982-1984, Alnus glutinosa; Quercus robur-Carpinus betulus.
Spruce/Solling	24	22		47	22									26				6. Schultz (1985); 3/1983-4/1984, a=incl. in TF.
4 Beech - summer/Solling	13	33		25	9		9											
Beech - winter/Solling	11	24		17	11		11											
summer + winter	24	57		42	20		4											
5 Alder/Riddagshausen	35			57	5.6		5.6							57				
Oak - Hornbeam/Riddagsh.	35			60	35		35							40				
Beech/Harste	15			37	13		a											
Spruce/Spaubeck	19			37	11													
Spruce/Wingst	20			47	10													
Spruce/Westerberg	24			37	17													
Beech/Solling	24			62	33		a											
Spruce/Solling	24			47	22													
Limburger Heide	30																	
7 Spruce/Solling	27.9	19.0		47.1	23.3	23.1		23.3	26.2				7.0		20.7	37.2		7. Schultz (1987).
Spruce/Spaubeck	20.6	18.0		43.2	15.1	30.3		15.1	6.2				64.0		32.4	-11.9		5/1983-4/1985.
Spruce/Wingst	27.8	38.0		58.0	40.5			40.5	3.3				3.0		62.5	92.2		F=lysimeter plates or funnel lysimeters.
Spruce/Westerberg	27.5	26.0		51.0	28.6			28.6	5.3				4.0		48.2	70.3		G=ceramic cups, tension lysimeters.
Pine/Heide	25.7	13.0		39.7	18.1	33.0		18.1	7.9				23.0		30.8	26.9		
Beech/Solling	27.9	57.0		40.4	45.2	32.9	9.6	45.2	6.0				24.0		78.9	65.2		
Beech/Harste	16.3	32.0		27.6	4.9	21.6	4.9	21.6	4.4				30.0		43.9	19.7		
Oak/Heide	23.7	56.0		46.8	44.8	75.9		44.8	6.1				17.0		75.6	68.5		
8 Spruce/Bairhalde (a)	18			16		10			9						9.0/11.0			8. Stahr <i>et al.</i> (1980), 5/1977-4/78, a=Braunerde, b=Podzol, Tension lysimeter: F=30 cm, G=100 and 80 cm, resp.
Spruce/Bairhalde (b)	18			16		30			25						-7.0/11.0			

Table 11a (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	K	L	
											(A+B) -G or H	(C+D+E)- (G+I+J)	
9 Spruce/Bärhalde (a)	27	-	-	-	-	-	12	-	-	-	15	-	9. Trüby and Zöttl (1984). 2 yr. G=tension lysimeter. a=Braunerde, b=Pararendzina.
Pine/Hartheim (b)	16	-	-	-	-	-	2	-	-	-	14	-	10. Zöttl (1985). 5/1977-4/79. a=Braunerde, b=Podzol. Tension lysimeter. F=30 cm, G=100 and 80 cm, resp.
10 Spruce/Bärhalde (a)	23	-	21	-	20	12	15	-	-	-	8	26	
Spruce/Bärhalde (b)	23	-	28	-	32	24	19	-	-	-	4	41	
11 S. Black Forest: Spruce/fir/beechn	40-50	-	30-50	-	10-26	-	-	-	-	-	-	-	11. Mies (1987). 1982-84.
12 Black Forest: Brown forest soil/podzol/podzol	-	-	-	-	-	-	-	11/8.0/10	-	-	-	-	12. Feger (1986). 1984-85
13 Spruce/Reinhardtswald	86	-	300	-	-	-	-	-	-	-	-	-	13. Brechtel <i>et al.</i> (1986). 1983.
14 NRW: Spruce/pine/beechn/oak	215-421	-	370/350/ 350/290	-	-/-/ 25/5	-	-	-	-	-	-	-	14. Block and Bartels (1985).
Austria 15 Beechn/Wienerwald	78	-	67	23	-	-	-	-	-	-	-	-	15. Kazda (1986). 5/1984-4/85.
Poland 16 Niepolomice Forest: Pine Oak-hornbeam	- 190 190	- - -	- - -	- - -	- - -	- - -	- - -	20 16	19 191	- -	170 174	151 -17	16. Grodzinski <i>et al.</i> (1984). Industrial region. B=free deposition, glass receptacle. I=trees above ground; L=B-(H+I).
N. America 17 Mixed forests/Can. Muskoka	-	-	-	-	-	-	-	1.3	-	-	-	-	17. Schut <i>et al.</i> (1986). 3/1982-3/83. 13 catchments.
18 Maple-birch/ Can., Turkey Lake	41	-	142	1	-	166	393	4	-	-	-352/37	-227	18. Foster and Nicolson (1986). 1983; remote site F=zero-tension lysimeter, F hor, G=tension lysimeter, 60 cm; L=F-G.
19 Pine-oak/ USA, NJ, Pine Barrons	53	-	-	-	-	-	19	7	-	-	34/46	-	19. Swanson and Johnson (1980). 5/1978-4/79. McDonalds Branch Basin; Pinus rigida and mixed oaks. Soil: pH-H2O 3.4-4.8; A: pH 3.8-4.1; H: pH 3.7-3.8. G=ground water, (conc. * estimated water flux).
Sweden 20 Spruce/Horröd	20	-	-	-	-	29	-	-	-	-	-9	-	20. Tyler (1981). 8/1977-12/79. Picea abies, 70 yr. podzol, mor. A=moss analysis. L=zero-tension lysimeter: 15 cm, A horizon. K=A-F.

Table 10a (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Litter-organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	K	L	
											(A+B) -G or H	(C+D+E) -(G+H+J)	
21 Spruce/Värsjö	8.1	1.6	21	-	17	16.7	8	-	13	>28.3	1.7	-11.3	21. Bergkvist (1987c). 2 yr. 1980+1981. Picea abies, 80 yr. Podzol, mor, pH-KCl: 2.8-4.4. F; G=Z+I lys: 15 cm. A hor/55 cm B hor. 7/1980-7/84. Picea abies, 80 yr. Podzol, mor, pH-KCl: 2.8-4.3.
22 Höröd:													
Spruce (a)	8.3	-	-	-	-	10.8	3.3	-	-	-	5	7.5	22. Bergkvist (1987a). 1980+1981. a=Picea abies, 60 yr. b=opening 30*30 m. c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970. Podzol, mor, pH-KCl: 2.9-4.5. F; G=zero-tension lysimeter: 15 cm, A hor; 35 or 55 cm. B hor
Spruce (b)	8.3	-	-	-	-	8.9	3.8	-	-	-	4.5	5.1	
Beech (c)	8.3	-	-	-	-	8.9	4.1	-	-	-	4.2	4.8	
Regeneration area (d)	8.3	-	-	-	-	15.3	7.4	-	-	-	0.9	7.9	L=Mineral soil budget: 15-35 or 15-55 cm.
Sjöbo:													
Spruce (e)	8	-	-	-	-	6.6	5.4	-	-	-	2.6	1.2	e=Picea abies, 50 yr. f=Fagus sylvatica, 100 yr.
Beech (f)	8	-	-	-	-	9.5	3.7	-	-	-	4.3	5.8	g=Spruce cleared 1967. Brown forest soil, pH-KCl: 3.1-4.3.
Regeneration area (g)	8	-	-	-	-	15.6	15.1	-	-	-	-7.1	0.5	23. Bergkvist <i>et al.</i> (1988). 6/1984-5/1987. Picea abies, Fagus sylvatica, Betula pendula.
23 Spruce:													
a	14	-	29	1.9	18	19	7.4	-	-	-	6.6	11.6	a-c: podzols; d-e: acidic brown forest soils.
b	11	-	32	0.97	12	22	5.9	-	-	-	5.1	16.1	F=Zero-tension lysimeter: 15 cm, A horizon.
c	11	-	34	0.85	19	27	4.8	-	-	-	6.2	22.2	G=Zero-tension lysimeter: 50 cm, B horizon.
d	7.6	-	25	1.2	12	5.9	5	-	-	-	2.6	0.9	L=Mineral soil budget: 15-50 cm.
e	7.6	-	16	1	15	7.8	5.3	-	-	-	2.3	2.5	
Beech:													
a	14	-	23	2	12	22	5.4	-	-	-	8.6	16.6	
b	11	-	17	1.8	12	24	6.1	-	-	-	4.9	17.9	
c	11	-	25	2.4	12	17	5.2	-	-	-	5.8	11.8	
d	7.6	-	24	2.2	24	7.7	4.3	-	-	-	3.3	3.4	
e	7.6	-	24	3.4	14	6.7	4.8	-	-	-	2.8	1.9	
Birch:													
a	14	-	34	3.7	15	9.3	4	-	-	-	10	5.3	24. Grahn and Rosen (1983). 10/80-9/81. Podzol; Pinus sylv., Picea abies; pH stream: 4.8.
b	11	-	25	2	9.7	6.3	4.2	-	-	-	6.8	2.1	Podzol; P.s., P.a.; (Betula), pH stream: 5.0.
c	11	-	18	1.2	12	11.5	9	-	-	-	2	2.5	Podzol; Pinus sylv., Picea abies; pH stream: 4.1.
d	7.6	-	31	0.85	16	9.6	1.9	-	-	-	5.7	7.7	pH stream: 4.2.
e	7.6	-	16	1.3	16	9.9	14.7	-	-	-	-7.1	-4.8	pH stream: 4.3.
24 Västerbotten, Svartberget	10.9	-	-	-	-	-	-	5.2	-	-	5.7	-	
Hälsingland, Kullarna	39.8	-	-	-	-	-	-	0.9	-	-	38.9	-	
Bohuslän, Gårdsjön II	7.4	1.6	13	-	-	-	-	5	-	-	4	-	
Bohuslän, Gårdsjön III	7.4	1.6	13	-	-	-	-	1.3	-	-	7.7	-	
Bohuslän, Gårdsjön IV	7.4	1.6	13	-	-	-	-	4.4	-	-	4.6	-	

TABLE IIb
Mean annual flux of Zn ($\text{g ha}^{-1} \text{ yr}^{-1}$)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	K	L	
											(A+B) -G or H	(C+D+E) -(G+H+J)	
<i>F.R.G.</i>													
1 Beech/Solling	1377	255	777	1392	260	2029	1125	-	110	907	507	287	1. Mayer (1981, 1986), 11/1974-8/1979. Beech 125 yr, spruce 90 yr, standing crop, 311 and 324 ton/ha, resp; acid braunerde, Moder, pH 3.9 - 4.3, loess loam, clay 16 - 19%. F=zero-tension lysimeter, G=tension lysimeter, 80 cm.
Spruce/Solling	1377	355	2121	-	250	3251	2364	-	246	885	-632	-1124	
2 Beech/Solling	1890	350	920	1800	260	-	1100	-	110	907	1140	863	2. Urrich <i>et al.</i> (1979), Heinrichs and Mayer (1980), Mayer and Heinrichs (1980); 11/1974-10/1977. B=calc. from 1., G=tension lysimeter, 80 cm.
Spruce/Solling	1890	487	2700	-	250	-	2400	-	250	885	-23	-585	3. Schmidt (1987). 1 yr, 1983-1985. a=incl. in TF.
3 Beech/Solling	410	350	680	a	170	-	-	-	-	90	-	-	4. Asche (1985); 2 yr, 1982-1984. Alnus glutinosa, Quercus robur-Carpinus betulus.
Spruce/Solling	410	280	570	-	110	-	-	-	-	80	-	-	5. Schultz (1985); 3/1983-4/1984. a=incl. in TF.
4 Alder/Riddagshausen	241	-	442	42	528	-	-	-	-	603	-	-	
Oak-Hornbeam/Riddagsh.	241	-	384	50	262	-	-	-	-	286	-	-	
5 Beech/Harste	90	-	233	a	78	-	-	-	-	-	-	-	
Spruce/Spanbeck	90	-	225	-	75	-	-	-	-	-	-	-	
Spruce/Wingst	180	-	353	-	118	-	-	-	-	-	-	-	
Spruce/Westerberg	200	-	326	-	114	-	-	-	-	-	-	-	
Beech/Solling	410	-	680	a	170	-	-	-	-	-	-	-	
Spruce/Solling	410	-	571	-	109	-	-	-	-	-	-	-	
Lüneburger Heide	220	-	-	-	-	-	-	-	-	-	-	-	
6 Spruce/Solling	316	333	531	-	112	1487	1573	-	153	-	-924	-1083	6. Schultz (1987). 5/1983-4/1985.
Spruce/Spanbeck	112	261	274	-	95	820	350	-	171	-	23	-152	F=lysimeter plates or funnel lysimeters. G=ceramic cups, tension lysimeters.
Spruce/Wingst	249	355	430	-	168	527	527	-	86	-	77	-15	
Spruce/Westerberg	235	343	429	-	143	557	557	-	173	-	21	-158	
Pine/Heide	184	297	369	-	107	467	599	-	25	-	-118	-148	
Beech/Solling	316	311	463	99	185	1363	664	-	110	-	129	27	
Beech/Harste	97	168	165	40	112	903	136	-	154	-	141	167	
Oak/Heide	184	168	288	-	131	299	211	-	41	-	92/93	177	7. Stahr <i>et al.</i> (1980). 5/1977-4/78. a=Braunerde, b=Podzol. Tension lysimeter: F=30 cm, G=100 and 80 cm, resp.
Spruce/Bärhalde (a)	174	-	259	-	96	242	82	81	-	-	-274/93	-103	
Spruce/Bärhalde (b)	174	-	345	-	182	400	448	81	-	-	-	-	
<i>8 S. Black Forest:</i>													
Spruce/fir/beech	140-230	-	200-360	-	50-160	-	-	-	-	-	-	-	8. Mies (1987), 1982-84.
9 Spruce/Rainhardswald	110	-	1800	-	-	-	-	-	-	-	-	-	9. Brechtel <i>et al.</i> (1986), 1983.

Table 11b (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	K	L	
											(A+B) -G or H	(C+D+E)- (G+I+J)	
10 Black Forest: Brown forest soil/podzol/podzol	-	-	-	-	-	-	-	40/130/ 160	-	-	-	-	10. Feger (1986), 1984-85.
<i>Poland</i>													
11 Niepolomice Forest:													
Pine	-	1200	-	-	-	-	-	530	242	-	670	428	11. Grodzinski <i>et al.</i> (1984), Industrial region.
Oak-hornbeam	-	1200	-	-	-	-	-	220	768	-	980	212	B=free deposition, glass receptacle. I=trees above ground, L=B-(H+I).
<i>Czechoslovakia</i>													
12 Moldava/polluted Spruce	-	-	9750	-	-	4680/2920	3050	-	-	-	-2420	1630	12. Lochman (1985), 1978-1979. Illimerized brown forest soil. pH-KCl: 2.3-4.2. F=zero-tension lysimeter, A0/20 cm. G=zero-tension lysimeter, 50 cm. Podzolized brown forest soil. pH-KCl: 3.8-4.4. F=zero-tension lysimeter, A0/30 cm. G=zero-tension lysimeter, 100 cm.
Zelivka/less polluted Spruce	-	-	4600	-	-	1770/240	120	-	-	-	490	1650	13. Lochman (1983), Materna (1985), 1973-79. Illimerized brown forest soil. pH-KCl: 2.3-4.2. F=zero-tension lysimeter, A0/20 cm. G=zero-tension lysimeter, 50 cm. Podzolized brown forest soil. pH-KCl: 3.8-4.4. F=zero-tension lysimeter, A0/30 cm. G=zero-tension lysimeter, 100 cm.
13 Moldava/polluted Spruce	630	-	2330	-	-	1200/730	770	-	-	-	-	-	14. Van Hook <i>et al.</i> (1977, 1980). L=(A+E)-(H+I+J).
Zelivka/less polluted Spruce	610	-	1220	-	-	780/78	25	-	-	-	-	-	15. Turner <i>et al.</i> (1985). 1981-82, wet-only.
<i>N. America</i>													
14 USA, TN, Walker Branch Mixed deciduous (four sites)	538	-	119	-	76	-	-	140	247	632	398	-405	16. Tyler (1981), 8/1977-12/79. Picea abies, 70 yr, podzol, mor. A=moss analysis. L=zero-tension lysimeter, 15 cm. A horizon. K=A-F.
15 USA, TN, NC (four sites)	110-180	5-83	-	-	-	-	-	11-41	-	-	252-74	-	17. Bergkvist (1987c), 2 yr, 1980+1981, Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4. F, G=z-t lys.: 15 cm, A hor/55 cm B hor. 7/1980-7/84. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.3.
<i>Sweden</i>													
16 Spruce/Horröd	180	-	-	-	-	210	-	-	-	-	-30	-	16. Tyler (1981), 8/1977-12/79. Picea abies, 70 yr, podzol, mor. A=moss analysis. L=zero-tension lysimeter, 15 cm. A horizon. K=A-F.
17 Spruce/Värsjö	100	13	320	-	210	140	560	-	270	>420	-447	-720	17. Bergkvist (1987c), 2 yr, 1980+1981, Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4. F, G=z-t lys.: 15 cm, A hor/55 cm B hor. 7/1980-7/84. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.3.
Pine-Spruce/Gårdsjön	318	13	283	-	230	210	456	-	270	>500	-125	-713	17. Bergkvist (1987c), 2 yr, 1980+1981, Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4. F, G=z-t lys.: 15 cm, A hor/55 cm B hor. 7/1980-7/84. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.3.

Table 11b (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	(A+B) -G or H	(C+D+E) -(G+I+J)	
18. Horröd:													
Spruce (a)	105	-	-	-	-	101	414	-	-	-	-309	-313	18. Bergkvist (1987a). 1980+1981. a=Picea abies, 60 yr, b=opening 30x30 m. c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970. Podzol, mor, pH-KCl: 2.9-4.5. F,G=zero-tension lysimeter: 15 cm, A hor: 35 or 55 cm, B hor
Spruce (opening) (b)	105	-	-	-	-	226	234	-	-	-	-129	-8	
Beech (c)	105	-	-	-	-	121	319	-	-	-	-214	-198	
Regeneration area (d)	105	-	-	-	-	132	85	-	-	-	20	47	
Sjöbo:													
Spruce (e)	100	-	-	-	-	344	485	-	-	-	-385	-141	L=Mineral soil budget: 15-35 or 15-55 cm.
Beech (f)	100	-	-	-	-	196	461	-	-	-	-361	-265	e=Picea abies, 50 yr. f=Fagus sylvatica, 100 yr.
Regeneration area (g)	100	-	-	-	-	219	195	-	-	-	-95	24	g=Spruce cleared 1967. Brown forest soil, pH-KCl: 3.1-4.3.
19. Spruce:													
a	220	-	250	35	160	470	650	-	-	-	-430	-180	19. Bergkvist <i>et al.</i> (1988). 6/1984-5/1987. Picea abies, Fagus sylvatica, Betula pendula.
b	200	-	440	20	110	700	790	-	-	-	-590	-90	a-c: podzols; d-e: acidic brown forest soils.
c	200	-	330	28	170	570	930	-	-	-	-730	-360	F=Zero-tension lysimeter: 15 cm, A horizon.
d	190	-	270	38	190	660	1200	-	-	-	-1010	-540	G=Zero-tension lysimeter: 50 cm, B horizon.
e	190	-	320	30	210	760	1300	-	-	-	-1110	-540	L=Mineral soil budget: 15-50 cm.
Beech:													
a	220	-	110	14	59	200	1200	-	-	-	-980	-1000	
b	200	-	180	10	77	310	1200	-	-	-	-1000	-890	
c	200	-	120	12	110	470	1400	-	-	-	-1200	-930	
d	190	-	120	11	200	650	1200	-	-	-	-1010	-550	
e	190	-	100	11	110	900	1800	-	-	-	-1610	-900	
Birch:													
a	220	-	190	58	330	290	200	-	-	-	20	90	20. Grahn and Rosén (1983). 10/80-9/81. Podzol; Pinus sylv., Picea abies; pH stream: 4.8.
b	200	-	280	26	230	86	180	-	-	-	20	-94	Podzol; P.s., P.ai.; (Betula); pH stream: 5.0.
c	200	-	240	31	330	220	170	-	-	-	30	50	Podzol; Pinus sylv., Picea abies; pH stream: 4.1.
d	190	-	230	21	480	180	58	-	-	-	132	122	pH stream: 4.2.
e	190	-	290	18	340	330	250	-	-	-	-60	80	pH stream: 4.3.
20. Västerbotten, Svartberget	263	-	-	-	-	-	-	30	-	-	233	-	
Hälsinglän, Kullarna	199	-	-	-	-	-	-	24	-	-	175	-	
Bohuslän, Gårdsjön II	318	13	283	-	-	-	-	170	-	-	161	-	
Bohuslän, Gårdsjön III	318	13	283	-	-	-	-	151	-	-	180	-	
Bohuslän, Gårdsjön IV	318	13	283	-	-	-	-	181	-	-	150	-	

Table 11b (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
A	B	C	D	E	F	G	H	I	J	K	(A+B)- (G+H)	(C+D+E)- (G+H+J)	
<i>Denmark</i>													
21 Klosterhede/Picea	350	-	a 410	-	-	b 240 a,c 770	b 50 a,c 380	-	-	-	d 300 c -30	d,f 190 c,f 390	21. Rasmussen (1988), 3-4 yr, 1983/84-87. Spruce, 68 yr, Podzol, pH-CaCl ₂ , 0-5 cm: 2.7. a=2 yr, 1985-87; b=zero-tension lysimeter; A hor. 0-25 cm, AB hor. 0-65 cm, resp.; c=tens. lys., depth as b.; d= z-t lys.; e=tension lysimeter, [=mineral soil budget; g=J yr, 1986-87. Spruce, 42 yr. Acid brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.1.
Strødam/Picea	190	-	310	-	-	b 360 a,c,g 1200	b 860 a,c,g 460	-	-	-	d -670 e -270	d,f -500 e,f 740	
Tange/Picea	150	-	a 230	-	-	b 150 a,c 150	b 90 a,c 120	-	-	-	d 60 c 30	d,f 60 e,f 30	Spruce, 47 yr; Brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.6.

TABLE IIc
Mean annual flux of Pb ($\text{g ha}^{-1} \text{yr}^{-1}$)

Ecosystem/site	Bulk dep.		Dry dep.		Through-fall		Stem-flow	Litter-fall	Soil organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	B	A	C	D	E	F	G	H	I	J	K	L	(C+D+E)- (G+H+I)	
<i>F.R.G.</i>																
1 Beech/Solling	285	152	229	73	120	314	24	-	49	34	413	315	1. Mayer (1981, 1986). 11/1974-8/1979. Beech 125 yr, spruce 90 yr, standing crop: 311 and 324 ton/ha, resp. acid braunerde, Moder, pH 3.9 - 4.3, loess loam, clay 16 - 19%, F=zero-tension lysimeter, G=tension lysimeter, 80 cm.			
Spruce/Solling	285	448	467	-	256	178	13	-	76	66	720	568	2. Ulrich <i>et al.</i> (1979), Heinrichs and Mayer (1980), Mayer and Heinrichs (1980); 11/1974-10/1977. B=calc. from 1.; G=tension lysimeter, 80 cm.			
2 Beech/Solling	310	165	255	85	120	-	30	-	39	34	445	357	3. Schultz <i>et al.</i> (1986), Schmidt (1987). 1 yr, 1983-1985. a=incl. in TF.			
Spruce/Solling	310	487	532	-	256	-	27	-	76	66	770	619	4. Schmidt and Schultz (1985). 1983, summer=May to Oct., winter=Nov. to Apr.			
3 Beech/Solling	130	189	195	a	62	-	-	-	-	2.7	-	-	5. Mayer (1983). 7/1979-12/1981 G=tension lysimeter, 50 cm			
Spruce/Solling	130	195	205	-	177	-	-	-	-	3	-	-	6. Asche (1985); 2 yr, 1982-1984. Alnus glutinosa; Quercus robur-Carpinus betulus.			
4 Beech - summer/Solling	60	73	61	15	9	-	-	-	-	-	-	-	7. Schultz (1985); 3/1983-4/1984. a=incl. in TF.			
Beech - winter/Solling	71	80	102	17	7	-	-	-	-	-	-	-				
summer + winter	131	153	163	32	16	-	-	-	-	-	-	-				
5 Oak/Lüneb. Heide	214	-	140	-	95	-	16	-	-	-	198	219				
Pine/Lüneb. Heide	214	-	175	-	30	-	260	-	-	-	-46.0	-55				
6 Alder/Riddagshausen	138	-	130	15	42	-	-	-	-	-	-	-				
Oak-Hornbeam/Riddagsh.	138	-	145	19	40	-	-	-	-	-	-	-				
7 Beech/Harste	74	-	97	a	34	-	-	-	-	-	-	-				
Spruce/Spaubeck	73	-	133	-	44	-	-	-	-	-	-	-				
Spruce/Wingst	100	-	154	-	57	-	-	-	-	-	-	-				
Spruce/Westerberg	100	-	160	-	72	-	-	-	-	-	-	-				
Beech/Solling	131	-	195	a	61	-	-	-	-	-	-	-				
Spruce/Solling	131	-	207	-	116	-	-	-	-	-	-	-				
Lüneburger Heide	104	-	-	-	-	-	-	-	-	-	-	-				
8 Spruce/Solling	158	250	242	-	147	198	4.7	-	46.0	-	403	338	8. Schultz (1987).			
Spruce/Spaubeck	84.0	105	130	-	50.0	13.8	3.0	-	17.0	-	186	160	5/1983-4/1985.			
Spruce/Wingst	133	196	197	-	118	-	1.2	-	6.0	-	328	308	F=lysimeter plates or funnel lysimeters.			
Spruce/Westerberg	119	222	207	-	118	-	1.2	-	11.0	-	340	313	G=ceramic cups, tension lysimeters.			
Pine/Heide	110	73.0	144	-	30.0	26.0	24.2	-	2.0	-	159	148				
Beech/Solling	158	177	167	22.0	81.0	254	0.8	-	40.0	-	334	229				
Beech/Harste	86.2	75.0	77.3	18.0	34.0	60.6	39.3	-	8.0	-	122	82				
Oak/Heide	110	89.0	121	-	40.0	40.0	9.9	-	13.0	-	189	138				

Table 11c (continued)

Ecosystem/site	A	B	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
				C	D	E	F	G	H	I	J	K	L	
												(A+B) -G or H	(C+D+E)- (G+H+J)	
9 Spruce/Bärhald (a)	110	-	-	70	-	-	40	12	5.9	-	-	>98/>104	-	9. Stehr <i>et al.</i> (1980), 5/1977-4/78. a=Braunerde, b=Podzol. Tension lysimeter: F=30 cm, G=100 and 80 cm, resp.
Spruce/Bärhald (b)	110	-	-	70	-	-	21	7	5.9	-	-	>103/>104	-	
10 Spruce/Bärhald (a)	132	-	-	-	-	-	-	29	-	-	-	103	-	10. Trüby and Zöttl (1984), 2 yr. G=tension lysimeter. a= Braunerde, b= Pararendzina.
Pine/Härthelm (b)	86	-	-	-	-	-	-	1	-	-	-	85	-	11. Zöttl (1985), 5/1977-4/79. a=Braunerde, b=Podzol. Tension lysimeter: F=30 cm, G=100 and 80 cm, resp.
11 Spruce/Bärhald (a)	128	-	-	74	-	110	29	18	7.5	-	-	110/120	166	
Spruce/Bärhald (b)	128	-	-	80	-	170	57	12	7.5	-	-	116/120	238	
12 S. Black Forest														
Spruce/fir/beech	70-120	-	-	50-130	-	16-34	-	-	-	-	-	-	-	12. Mies (1987), 1982-84.
13 Hessen														
Spruce/Reinhardswald	140	-	-	190	-	-	-	-	-	-	-	-	-	13. Brechtel <i>et al.</i> (1986), 1983.
Spruce/3 sites	390	-	-	430	-	-	-	-	-	-	-	-	-	
14 Black Forest: Brown forest soil/podzol/podzol	-	-	-	-	-	-	-	-	7.0/12/12	-	-	-	-	14. Feger (1986), 1984-85.
15 NRW: Spruce/pine/beech/oak	100-180	-	-	110/100/120/110	-/-/18/3	-	-	-	-	-	-	-	-	15. Block and Bartels (1985).
Austria														
16 Beech/Wienerwald	155	-	-	137	62	-	-	-	-	-	-	-	-	16. Kazda (1986), 5/1984-4/85.
Poland														
17 Niepolomice Forest:														
Pine	-	320	-	-	-	-	-	-	23	94	-	>297	203	17. Grodzinski <i>et al.</i> (1984). Industrial region. B=free deposition, glass receptacle.
Oak-hornbeam	-	320	-	-	-	-	-	-	18	250	-	>302	52	I=trees above ground; L=B=(H+I).
N. America														
18 Mixed forests/Can. Muskoka	-	-	-	-	-	-	-	-	2.8	-	-	-	-	18. Schut <i>et al.</i> (1986), 3/1982-3/83. 13 catchments.
19 Maple-birch/Can., Turkey Lake	25	-	-	23	1	-	79	6	2	-	-	19/23	73	19. Foster and Nicolson (1986), 1983; remote site F=zero-tension lysimeter, F hor, G=tension lysimeter, 60 cm; L=F-G.
20 Hardwood (a) Hardwood (b) USA, NH, Hubbard Brook	266 317	-	-	-	-	-	-	-	6.1 12	-	-	260 305	-	20. Siccama and Smith (1978), Smith and Siccama (1981), a=4 yr: 1975-1978. b=1 yr: 1975. Hardwood: 65 yr, podzol, Fagus grandifolia, Betula alleghaniensis, B. papyrifera, Picea rubens, Abies balsamea.

Table 1c (continued)

Ecosystem/site	Bulk dep.		Dry dep.	Through-fall		Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B		C	D										
21 USA, TN, Walker Branch Mixed deciduous	286	-	-	-	-	19	-	-	-	6	8	-	280	291	21. Van Hook <i>et al.</i> (1977). I=(A+E)-(H+I)
22 USA, TN, NC (four sites)	68-73	18-26	-	-	-	-	-	-	-	0.7-1.5	-	-	98-85	-	22. Turner <i>et al.</i> (1985). 1981-82, wet-only.
23 Pine-oak/ USA, NJ, Pine Barrens	254	-	-	-	-	-	-	14	17	-	-	-	240/237	-	23. Swanson and Johnson (1980). 5/1978-4/79. McDonalds Branch Basin; Pinus rigida and mixed oaks. Soil: pH-H2O 3.4-4.8; A: pH 3.8-4.1; H: pH 3.7-3.8. G=ground water, (conc. * estimated water flux).
24 Pine-oak/ USA, NJ, Pine Barrens	87	60	102	-	-	40	35	30/20/0	-	-	4.2	-	127	118	24. Turner <i>et al.</i> (1985). 1979-82. McDonalds Branch Basin; Pinus rigida and mixed oaks.
25 USA, Vermont Fir-spruce-birch Camels Hump	700	-	-	-	-	-	85/39	23/20	<12	-	-	-	>680/688	19	25. Friedland and Johnson (1985). 5/1983-10/1984. Abies balsamea- Picea rubens-Betula papyrifera. A=cloud dep. (400) incl. Zerotenston lysimeter: F=3 cm/12cm; G=25 cm/40 cm. L=Mineral-soil budget: 12 cm - 40 cm.
<i>Sweden</i>															
26 Spruce/Horröd	150	-	-	-	-	-	101	-	-	-	-	-	49	-	26. Tyler (1981). 8/1977-12/79. Picea abies, 70 yr, podzol, mor. A=moss analysis. L=zero-tension lysimeter: 15 cm, A horizon. K=A-F.
27 Spruce/Värsjö	87	13	77	-	-	74	81	6.4	-	-	34	>51	93.6	59.6	27. Bergkvist (1987c). 2 yr. 1980+1981. Picea abies, 80 yr, Podzol, mor, pH-KCl, 2.8-4.4. F;G=z-t lys.: 15 cm, A hor/55 cm B hor. 7/1980-7/84. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.3.
Pine Spruce/Gårdsjön	64	13	49	-	-	9	76	2.4	-	-	34	>43	74.6	-21.4	
28 Horröd:															
Spruce (a)	81.5	-	-	-	-	-	60	5.6	-	-	-	-	75.9	54.4	28. Bergkvist (1987a). 1980+1981. a=Picea abies, 60 yr, b=opening 30x30 m. c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970. Podzol, mor, pH-KCl: 2.9-4.5. F;G=zero-tension lysimeter: 15 cm, A hor: 35 or 55 cm, B hor
Spruce opening (b)	81.5	-	-	-	-	-	13.1	3.2	-	-	-	-	78.3	9.9	
Beech (c)	81.5	-	-	-	-	-	10.5	1.5	-	-	-	-	80	9	
Regeneration area (d)	81.5	-	-	-	-	-	14.4	3	-	-	-	-	78.5	11.4	
Sjöbo:															
Spruce (e)	77.9	-	-	-	-	-	17.1	3.5	-	-	-	-	74.4	13.6	L=Mineral soil budget: 15-35 or 15-55 cm. e=Picea abies, 50 yr. f=Fagus sylvatica, 100 yr.
Beech (f)	77.9	-	-	-	-	-	12.2	3	-	-	-	-	74.9	9.2	g=Spruce cleared 1967. Brown forest soil, pH-KCl: 3.1-4.3.
Regeneration area (g)	77.9	-	-	-	-	-	6.4	3.1	-	-	-	-	74.8	3.3	

Table 1c (continued)

Ecosystem/site	Bulk dep.		Dry dep.		Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	B	A											
29 Spruce:															
a	62	-	-	51	11	65	56	2.7	-	-	-	-	59.3	53.3	29. Bergkvist <i>et al.</i> (1988). 6/1984-5/1987. Picea abies, Fagus sylvatica, Betula pendula. a-c: podzols; d-e: acidic brown forest soils.
b	66	-	-	74	3	69	92	4.5	-	-	-	-	61.5	87.5	F=Zero-tension lysimeter: 15 cm, A horizon.
c	66	-	-	66	3.8	83	99	5.8	-	-	-	-	60.2	93.2	G=Zero-tension lysimeter: 50 cm, B horizon.
d	51	-	-	41	5.4	40	6.3	2.1	-	-	-	-	48.9	4.2	L=Mineral soil budget: 15-50 cm.
e	51	-	-	41	3.4	52	11	4.2	-	-	-	-	46.8	6.8	
Beech:															
a	62	-	-	28	3.4	20	110	2.4	-	-	-	-	59.6	108	
b	66	-	-	27	4.2	18	100	1.8	-	-	-	-	64.2	98.2	
c	66	-	-	24	4.1	22	66	2.8	-	-	-	-	63.2	63.2	
d	51	-	-	22	3.7	38	10	2.7	-	-	-	-	48.3	7.3	
e	51	-	-	13	2.2	21	1.6	1.3	-	-	-	-	49.7	0.3	
Birch:															
a	62	-	-	20	9	24	7.6	15	-	-	-	-	47	-7.4	
b	66	-	-	36	6.3	15	9.7	4.4	-	-	-	-	61.6	5.3	
c	66	-	-	28	3.6	23	30	1.7	-	-	-	-	64.3	28	
d	51	-	-	26	2.2	20	4	1.6	-	-	-	-	49.4	2.4	
e	51	-	-	28	2.2	30	2.9	2.5	-	-	-	-	48.5	0.4	
30 Västerbotten, Svartberget	26.6	-	-	-	-	-	-	-	-	2.8	-	-	23.8	-	30. Gråhn and Rosén (1983). 10/80-9/81. Podzol; Pinus sylv., Picea abies; pH stream: 4.8.
Hälsingland, Kullarna	71.3	-	-	-	-	-	-	-	-	1.5	-	-	69.8	-	Podzol; P.s., P.a.; (Betula); pH stream: 5.0.
Bohuslän, Gårdsjön II	64.2	13	-	49	-	-	-	-	-	3.7	-	-	73.5	-	Podzol; Pinus sylv., Picea abies; pH stream: 4.1.
Bohuslän, Gårdsjön III	64.2	13	-	49	-	-	-	-	-	4.5	-	-	72.7	-	pH stream: 4.2.
Bohuslän, Gårdsjön IV	64.2	13	-	49	-	-	-	-	-	2.8	-	-	74.4	-	pH stream: 4.3.

TABLE II
Mean annual flux of Cd ($\text{g ha}^{-1} \text{yr}^{-1}$)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget K	Soil budget L	Remarks
	A	B	C	D	E	F	G	H	I	J	(A+B)-G or H	(C+D+E)-L	
<i>F.R.G.</i>													
1 Beech/Solling	15.9	0.4	10.8	1.75	2.3	17.5	16.5	-	1.4	0.4	-0.2	-3.45	1. Mayer (1981, 1986), 11/1974-8/1979. Beech 125 yr, spruce 90 yr, standing crop: 311 and 324 ton/ha, resp; acid braunerde, Moder, pH 3.9 - 4.3, loess loam, clay 16 - 19%. F=zero-tension lysimeter, G=tension lysimeter, 80 cm.
Spruce/Solling	15.9	4.2	20.1	-	1.9	20	26.2	-	3.2	5.1	-6.1	-12.5	2. Ulrich <i>et al.</i> (1979), Heinrichs and Mayer (1980), Mayer and Heinrichs (1980); 11/1974-10/1977. B=calc. from I.; G=tension lysimeter, 80 cm.
2 Beech/Solling	35	0.9	14	2.1	2.6	-	17	-	1.4	0.4	18.9	-0.1	3. Schultz <i>et al.</i> (1986), Schmidt (1987). 1 yr, 1983-1985. a=incl. in TF.
Spruce/Solling	35	9.2	25	-	2.3	-	22	-	3.2	5.1	22.2	-3	4. Schmidt and Schultz (1985). 1983; summer=May to Oct., winter=Nov. to Apr.
3 Beech/Solling	2.3	2.9	4.1	a	1.3	-	-	-	-	0.4	-	-	5. Mayer (1983), 7/1979-12/1981 G=tension lysimeter, 50 cm
Spruce/Solling	2.3	2.7	3.9	-	1.1	-	-	-	-	0.6	-	-	6. Asche (1985); 2 yr, 1982-1984. Alnus glutinosa; Quercus robur-Carpinus betulus.
4 Beech - summer/Solling	1.1	1.2	1.3	0.5	0.1	-	-	-	-	-	-	-	7. Schultz (1985); 3/1983-4/1984. a=incl. in TF.
Beech - winter/Solling	1.2	1.2	1.8	0.5	0.1	-	-	-	-	-	-	-	8. Schultz (1987).
summer + winter	2.3	2.4	3.1	1	0.2	-	-	-	-	-	-	-	5/1983-4/1985.
5 Oak/Lüneb. Heide	5.6	-	5	-	0.8	-	4.9	-	-	-	0.7	0.9	F=lysometer plates or funnel lysimeters.
Pine/Lüneb. Heide	5.6	-	6.5	-	0.63	-	9.8	-	-	-	-4.2	-2.67	G=ceramic cups, tension lysimeters.
6 Alder/Riddagshausen	2.17	-	2.2	0.34	0.99	-	-	-	-	-	-	-	
Oak-Hornbeam/Riddagsh.	2.17	-	3.18	0.49	1.31	-	-	-	-	1.28	-	-	
7 Beech/Harste	1.6	-	2.7	a	0.8	-	-	-	-	-	-	-	
Spruce/Spanbeck	1.8	-	3.4	-	0.8	-	-	-	-	-	-	-	
Spruce/Wingst	2.2	-	3.5	-	0.5	-	-	-	-	-	-	-	
Spruce/Westerberg	2	-	3	-	0.5	-	-	-	-	-	-	-	
Beech/Solling	2.4	-	4.1	a	1.3	-	-	-	-	-	-	-	
Spruce/Solling	2.4	-	4	-	1.1	-	-	-	-	-	-	-	
Lineburger Heide	1.9	-	-	-	-	-	-	-	-	-	-	-	
8 Spruce/Solling	2.67	3.30	4.86	-	1.10	11.0	13.6	-	2.80	-	-7.62	-10.4	
Spruce/Spanbeck	1.67	2.50	3.29	-	0.89	8.41	19.8	-	3.50	-	-15.7	-19.2	
Spruce/Wingst	2.40	2.90	4.22	-	1.06	9.40	9.40	-	0.80	-	-4.10	-4.92	
Spruce/Westerberg	2.17	3.80	4.91	-	0.46	-	16.3	-	0.80	-	-10.3	-11.7	
Pine/Heide	1.77	2.90	3.35	-	1.32	3.49	7.63	-	1.00	-	-2.96	-3.96	
Beech/Solling	2.67	2.30	3.16	0.77	1.38	4.54	6.80	-	0.80	-	-1.83	-2.29	
Beech/Harste	1.57	1.40	2.08	0.25	0.84	12.6	3.73	-	1.10	-	-0.76	-1.16	
Oak/Heide	1.77	1.30	2.46	-	0.86	2.26	10.5	-	1.70	-	-7.41	-8.86	

Table III (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	(A+B) -G or H	(C+D+E) -L (G+I+J)	
9 Spruce/Bärhald (a)	4.5	-	12	-	-	3	3	1.4	-	-	1.5/3.1	9.00	9. Stahr <i>et al.</i> (1980), 5/1977-4/78. a=Braunerde, b=Podzol. Tension lysimeter: F=30 cm, G=100 and 80 cm, resp.
Spruce/Bärhald (b)	4.5	-	12	-	-	6	3	1.4	-	-	1.5/3.1	9.00	
10 Spruce/Bärhald (a)	4.5	-	-	-	-	-	4	-	-	-	0.5	-	10. Tribby and Zöttl (1984), 2 yr. G=tension lysimeter. a= Braunerde, b= Pararendzina.
Pine/Hartheim (b)	2.1	-	-	-	-	-	0.1	-	-	-	2	-	
11 Spruce/Bärhald (a)	4.3	-	10.5	-	0.6	4	3.7	-	-	-	0.6	7.4	11. Zöttl (1985), 5/1977-4/79. a=Braunerde, b=Podzol. Tension lysimeter: F=30 cm, G=100 and 80 cm, resp.
Spruce/Bärhald (b)	4.3	-	12.3	-	1.5	9	6	-	-	-	-1.7	7.8	
12 S. Black Forest: Spruce/fir/beech	1.9-2.5	-	3.1	-	0.5-1.2	-	-	-	-	-	-	-	12. Mies (1987), 1982-84.
13 Black Forest: Brown forest soil/podzol/podzol	-	-	-	-	-	-	-	0.5/1.3/2.0	-	-	-	-	13. Feger (1986), 1984-85.
14 Spruce/Reinhardswald	33	-	27	-	-	-	-	-	-	-	-	-	14. Brechtel <i>et al.</i> (1986), 1983.
15 NRW: Spruce/pine/beech/oak	2.3-8.2	-	7.9/8.8/ 4.9/5.9	-/- 1.1/0.3	-	-	-	-	-	-	-	-	15. Block and Bartels (1985).
Austria 16 Beech/Wienerwald	36	-	44	26	-	-	-	-	-	-	-	-	16. Glatzel <i>et al.</i> (1986), 5/1984-4/85.
Poland 17 Niepolomice Forest:	-	15	-	-	-	-	-	3	8.3	-	12	3.7	17. Grodzinski <i>et al.</i> (1984), Industrial region. B-free deposition, glass receptacle. I=trees above ground; L=B-(H+I).
Pine	-	15	-	-	-	-	-	2	22	-	13	-9	
Oak-hornbeam	-	-	-	-	-	-	-	-	-	-	-	-	
N. America 18 Mixed forests/Can.	-	-	-	-	-	-	-	<1.7	-	-	-	-	18. Schut <i>et al.</i> (1986), 3/1982-3/83, 13 catchments.
Muskoka	-	-	-	-	-	-	-	-	-	-	-	-	
19 USA, TN, Walker Branch	21	-	-	-	1.8	-	-	7	0.8	-	14	15	19. Van Hook <i>et al.</i> (1977).
Mixed deciduous	-	-	-	-	-	-	-	-	-	-	-	-	
20 USA, TN, NC (four sites)	1.0-2.2	0.21-0.49	-	-	-	-	-	0.1-0.52	-	-	0.69-2.59	-	20. Turner <i>et al.</i> (1985), 1981-82; wet-only. I=(A+E)-(H+I)
21 Pine-oak/ USA, NJ, Pine Barrens	11.3	-	-	-	-	-	<10	<10	-	-	1.3/1.3	-	21. Swanson and Johnson (1980), 5/1978-4/79. McDonalds Branch Basin; Pinus rigida and mixed oaks. Soil: pH-H2O 3.4-4.8; A: pH 3.8-4.1; H: pH 3.7-3.8. G=ground water, (conc. * estimated water flux).

Table IIa (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	K	L	
											(A+B) -G or H	(C+D+E) -L (G+I+J)	
<i>Sweden</i>													
22 Spruce/Horröd	2	-	-	-	-	3.2	-	-	-	-	-1.2	-	22. Tyler (1981). 8/1977-12/79. Picea abies, 70 yr, podzol, mor. A=moss analysis. L=zero-tension lysimeter: 15 cm, A horizon. K=A-F.
23 Spruce/Värsjö	1.2	1.3	2.3	-	0.7	2.5	5	-	1.5	>0.5	-2.5	-4	23. Bergkvist (1987c), 2 yr, 1980-1981. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4, F,G-z+lys.: 15 cm, A hor/55 cm B hor. 7/1980-7/84. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.3.
Pine Spruce/Gårdsjön	1.9	1.3	2.3	-	0.5	4.5	4.2	-	1.5	>2	-1	-5	24. Bergkvist (1987a), 1980+1981. a=Picea abies, 60 yr, b=opening 30*30 m, c=Fagus sylvatica, 90 yr, d=Spruce cleared 1970. Podzol, mor, pH-KCl: 2.9-4.5, F,G=zero-tension lysimeter: 15 cm, A hor; 35 or 55 cm, B hor L=Mineral soil budget: 15-35 or 15-55 cm. e=Picea abies, 50 yr. f=Fagus sylvatica, 100 yr. g=Spruce cleared 1967. Brown forest soil, pH-KCl: 3.1-4.3.
24 Horröd:													
Spruce (a)	1.2	-	-	-	-	1.2	4.1	-	-	-	-2.9	-2.9	
Spruce (opening) (b)	1.2	-	-	-	-	3.1	2.3	-	-	-	-1.1	0.8	
Beech (c)	1.2	-	-	-	-	3.5	3.8	-	-	-	-2.6	-0.3	
Regeneration area (d)	1.2	-	-	-	-	2.7	1.3	-	-	-	-0.1	1.4	
Sjöbo:													
Spruce (e)	1.1	-	-	-	-	4.7	7	-	-	-	-5.9	-2.3	
Beech (f)	1.1	-	-	-	-	3.4	6.9	-	-	-	-5.8	-3.5	
Regeneration area (g)	1.1	-	-	-	-	5	1.6	-	-	-	-0.5	3.4	
25 Spruce:													
a	1.2	-	1.8	0.28	0.72	5.6	6.5	-	-	-	-5.3	-0.9	25. Bergkvist <i>et al.</i> (1988). 6/1984-5/1987. Picea abies, Fagus sylvatica, Betula pendula. a-c: podzols; d-e: acidic brown forest soils. F=Zero-tension lysimeter: 15 cm, A horizon. G=Zero-tension lysimeter: 50 cm, B horizon. L=Mineral soil budget: 15-50 cm.
b	1.1	-	2.3	0.1	0.32	2.4	10	-	-	-	-8.9	-7.6	
c	1.1	-	2.5	0.12	1.2	2.4	12	-	-	-	-10.9	-9.6	
d	0.92	-	1.5	0.23	0.62	7.4	7.3	-	-	-	-6.38	0.1	
e	0.92	-	1.6	0.16	0.62	7.3	4.4	-	-	-	-3.48	2.9	
Beech:													
a	1.2	-	1	0.08	0.35	3.5	14	-	-	-	-12.8	-10.5	
b	1.1	-	0.98	0.12	0.39	3.4	15	-	-	-	-13.9	-11.6	
c	1.1	-	0.91	0.14	0.36	3.6	11	-	-	-	-9.9	-7.4	
d	0.92	-	0.91	0.12	1.2	7.3	12	-	-	-	-11.1	-4.7	
e	0.92	-	0.8	0.07	0.42	9.2	5.2	-	-	-	-4.28	4	
Birch:													
a	1.2	-	1	0.35	0.96	3.2	3.2	-	-	-	-2	0	
b	1.1	-	1.5	0.21	0.54	2	2.9	-	-	-	-1.8	-0.9	
c	1.1	-	1.2	0.17	0.55	1.6	2.4	-	-	-	-1.3	-0.8	
d	0.92	-	1.5	0.12	1.1	2.9	1.6	-	-	-	-0.68	1.3	
e	0.92	-	1.3	0.09	0.88	3.9	1.7	-	-	-	-0.78	2.2	

TABLE IIc
Mean annual flux of C_r (g ha⁻¹ yr⁻¹)

Ecosystem/site	Bulk dep.		Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B												
<i>F.R.G.</i>														
1 Beech/Solling	14.3	135	12.6	3.22	45	23.4	7.1	-	-	87	13	142	-46.3	1. Mayer (1981, 1986). 11/1974-8/1979. Beech 125 yr, spruce 90 yr, standing crop: 311 and 324 ton/ha, resp. acid braunerde, Moder, pH 3.9-4.3, loess loam, clay 16-19%. F=zero-tension lysimeter, G=tension lysimeter, 80 cm.
Spruce/Solling	14.3	151	23.3	-	77	18.5	5.5	-	-	65	14	160	15.8	
2 Beech/Solling	12.1	114	8.8	3	45	-	3.5	-	-	87	13	123	-46.7	2. Ulrich <i>et al.</i> (1979), Heinrichs and Mayer (1980), Mayer and Heinrichs (1980); 11/1974-10/1977. B=calc. from I.; G=tension lysimeter, 80 cm.
Spruce/Solling	12.1	128	21.9	-	77	-	3	-	-	65	14	137	16.9	
3 Beech/Solling	4.4	15.4	7.3	a	7.9	-	-	-	-	-	0.6	-	-	3. Schmidt (1987). 1 yr, 1983-1985. a=incl. in TF.
Spruce/Solling	4.4	11.7	6.5	-	7.3	-	-	-	-	-	1.1	-	-	
4 Beech - summer/Solling	1.8	6	3.6	0.5	0.6	-	-	-	-	-	-	-	-	4. Schmidt and Schultz (1985). 1983; summer=May to Oct., winter=Nov. to Apr.
Beech - winter/Solling	2.6	0.8	2.6	0.6	0.2	-	-	-	-	-	-	-	-	
5 Beech/Harste	4.4	6.8	6.2	1.1	0.8	-	-	-	-	-	-	-	-	5. Schultz (1985); 3/1983-4/1984. a=incl. in TF.
Spruce/Harste	2.2	-	4	a	15	-	-	-	-	-	-	-	-	
Spruce/Spangebek	2.3	-	4.3	-	12.8	-	-	-	-	-	-	-	-	6. Schultz (1987) 5/1983-4/1985. F=lysimeter plates or funnel lysimeters. G=ceramic cups, tension lysimeters.
Spruce/Wingst	3.2	-	4.1	-	2.9	-	-	-	-	-	-	-	-	
Spruce/Westerberg	3.3	-	4	-	3	-	-	-	-	-	-	-	-	7. Block and Bartels (1985).
Beech/Solling	4.4	-	6.2	a	8.9	-	-	-	-	-	-	-	-	
Lüneburger Heide	3.5	-	-	-	15.4	-	-	-	-	-	-	-	-	8. Tyler (1981). 8/1977-12/79. Picea abies, 70 yr, podzol, mor. A-moss analysis. I=zero-tension lysimeter; 15 cm. A horizon. K=A-F.
6 Spruce/Solling	5.74	13.6	6.89	-	19.2	6.79	1.60	-	-	5.80	-	17.7	18.7	
Spruce/Spangebek	2.39	7.10	4.68	-	8.20	7.84	1.51	-	-	2.10	-	7.98	9.27	
Spruce/Wingst	2.60	2.80	3.73	-	4.60	4.70	4.70	-	-	0.40	-	0.70	3.23	
Spruce/Westerberg	3.12	2.80	3.91	-	4.10	6.10	6.10	-	-	1.30	-	-0.18	0.61	
Pine/Heide	3.52	1.50	4.83	-	2.00	5.83	5.04	-	-	1.30	-	-0.02	0.49	
Beech/Solling	5.74	10.2	5.42	0.65	6.10	6.44	0.82	-	-	2.80	-	15.1	8.55	
Beech/Harste	2.11	44.6	2.63	0.71	31.7	11.6	1.37	-	-	4.00	-	45.3	29.7	
Oak/Heide	3.52	8.30	4.07	-	5.00	9.19	8.17	-	-	3.60	-	3.65	-2.70	
7 NRW: Spruce/pine/beech/oak	4.2-17	-	16/12/9/9	-/-/-	-	-	-	-	-	-	-	-	-	
<i>Sweden</i>														
8 Spruce/Horröd	8	-	-	-	-	12.5	-	-	-	-	-	-4.5	-	

Table IIe (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	K	L	
											(A+B) -G or H	(C+D+E)- (G+I+J)	
9 Spruce/Värsjö	2	0.5	4.2	-	5.1	5	3	-	8.1	>6.8	-0.5	-8.6	9. Bergkvist (1987c). 2 yr. 1980+1981. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4. F,G=zero-tension lysimeter: 15 cm, A hor/35 cm B hor. 7/1980-7/84. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.3.
Pine Spruce/Gårdsjön	2.5	0.5	6.5	-	1.9	11	8.3	-	8.1	>10	-5.3	-18	10. Bergkvist (1987a). 1980+1981. a=Picea abies, 60 yr, b=opening 30*30 m, c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970. Podzol, mor, pH-KCl: 2.9-4.5. F,C=zero-tension lysimeter: 15 cm, A hor: 35 or 55 cm, B hor
10 Horrdö:													
Spruce (a)	1.8	-	-	-	-	7.9	2	-	-	-	-0.2	5.9	L=Mineral soil budget: 15-35 or 15-55 cm.
Spruce (opening) (b)	1.8	-	-	-	-	1	1.2	-	-	-	0.6	-0.2	e=Picea abies, 50 yr. f=Fagus sylvatica, 100 yr.
Beech (c)	1.8	-	-	-	-	7.2	6.2	-	-	-	-4.4	1	g=Spruce cleared 1967. Brown forest soil, pH-KCl: 3.1-4.3.
Regeneration area (d)	1.8	-	-	-	-	3.6	2.5	-	-	-	-0.7	1.1	11. Grahn and Rosén (1983). 10/80-9/81.
Sjöbo:													
Spruce (e)	1.7	-	-	-	-	4.3	4.8	-	-	-	-3.1	-0.5	Podzol, Pinus sylv., Picea abies; pH stream: 4.8.
Beech (f)	1.7	-	-	-	-	1.2	2.3	-	-	-	-0.6	-1.1	Podzol, P.s., P.a.; (Betula), pH stream: 5.0.
Regeneration area (g)	1.7	-	-	-	-	4.3	4.6	-	-	-	-2.9	-0.3	Podzol, Pinus sylv., Picea abies; pH stream: 4.1.
11 Västerbotten/Svartberget	1.3	-	-	-	-	-	-	2.6	-	-	-1.3	-	pH stream: 4.2.
Hälsingland, Kullarna	1.3	-	-	-	-	-	-	0.6	-	-	0.7	-	pH stream: 4.3.
Bohuslän, Gårdsjön II	2.5	0.5	6.5	-	-	-	-	3.5	-	-	-0.5	-	
Bohuslän, Gårdsjön III	2.5	0.5	6.5	-	-	-	-	2.6	-	-	0.4	-	
Bohuslän, Gårdsjön IV	2.5	0.5	6.5	-	-	-	-	2.3	-	-	0.7	-	

TABLE III
Mean annual flux of Ni (g ha⁻¹ yr⁻¹)

Ecosystem/site	Bulk dep.		Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B												
<i>F.R.G.</i>														
1 Beech/Solling	27	96	29	41	4.1	41	29.9	21	-	88	39	102	-73.9	1. Mayer (1981, 1986), 11/1974-8/1979, Beech 125 yr, spruce 90 yr, standing crop: 311 and 324 ton/ha, resp; acid braunerde, Moder, pH 3.9 - 4.3, loess loam, clay 16 - 19%, F=zero-tension lysimeter, G=tension lysimeter, 80 cm.
Spruce/Solling	27	113	39	66	-	66	33.5	66	-	78	43	74	-82	
2 Beech/Solling	26	92.4	30	41	4	41	-	18	-	88	39	100	-70	2. Ulrich <i>et al.</i> (1979), Heinrichs and Mayer (1980), Mayer and Heinrichs (1980); 11/1974-10/1977, B=calc. from 1.; G=tension lysimeter, 80 cm.
Spruce/Solling	26	109	40	66	-	66	-	63	-	78	43	71.8	-78	3. Schultz (1985); 3/1983-4/1984, a=incl. in TF.
3 Beech/Harste	4.7	-	-	-	-	-	-	-	-	-	-	-	-	
Spruce/Spaubeck	4.7	-	-	-	-	-	-	-	-	-	-	-	-	
Spruce/Wingst	5.9	-	-	-	-	-	-	-	-	-	-	-	-	
Spruce/Westerberg	5.7	-	-	-	-	-	-	-	-	-	-	-	-	
Beech/Solling	8.6	-	-	-	-	-	-	-	-	-	-	-	-	
Spruce/Solling	8.6	-	-	-	-	-	-	-	-	-	-	-	-	
Lüneburger Heide	12.3	-	-	-	-	-	-	-	-	-	-	-	-	
4 Spruce/Solling	10.8	19.0	18.6	11.8	27.0	45.3	27.0	45.3	-	1.0	-	-15.5	-15.9	4. Schultz (1987).
Spruce/Spaubeck	4.5	16.0	12.5	8.0	36.9	158	36.9	158	-	2.0	-	-138	-140	5/1983-4/1985.
Spruce/Wingst	8.4	10.0	10.6	7.1	-	12.2	-	12.2	-	<1	-	-6.20	4.52	F=lysimeter plates or funnel lysimeters.
Spruce/Westerberg	5.8	12.0	12.1	5.7	-	27.7	-	27.7	-	<1	-	-9.88	-10.9	G=ceramic cups, tension lysimeters.
Pine/Heide	8.7	8.0	12.1	4.6	11.0	26.4	11.0	26.4	-	5.0	-	-9.62	-10.7	
Beech/Solling	10.8	8.0	11.1	2.4	19.8	16.5	19.8	16.5	-	5.0	-	2.34	-2.40	
Beech/Harste	4.5	25.0	10.6	1.8	18.3	60.6	10.7	10.7	-	5.0	-	18.8	15.0	
Oak/Heide	8.7	18.0	11.3	15.2	20.0	23.7	20.0	23.7	-	2.0	-	3.06	0.87	
5 Spruce/Bärhald (a)	42	-	-	-	-	35	-	35	-	-	-	7	-	5. Trüby and Zättl (1984), 2 yr. G=tension lysimeter. a= Braunerde, b= Pararendzina.
Pine/Harthelm (b)	8	-	-	-	-	2	-	2	-	-	-	6	-	6. Block and Bartels (1985).
6 NRW: Spruce/pine/beechnoak	11-18	-	-	-	-/-	-	-	-	-	-	-	-	-	
					12/1.2									
<i>Austria</i>														
7 Beech/Wienerwald	33	-	39	-	14	-	-	-	-	-	-	-	-	7. Kazda (1986), 5/1984-4/85.
<i>Poland</i>														
8 Niepolomice Forest:														
Pine	-	66	-	-	-	-	-	-	24	29	-	42	13	8. Grodzinski <i>et al.</i> (1984). Industrial region.
Oak-hornbeam	-	66	-	-	-	-	-	-	26	86	-	40	-46	B=free deposition, glass receptacle. I=trces above ground; L=B-(H+I).

Table III (continued)

Ecosystem/site	A	B	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks	
				C	D	E	F	G	H	I	J	K	L		
												(A+B) -G or H	(C+D+E) -(G+H+J)		
<i>N. America</i>															
9 Pine-oak/ USA, NJ, Pine Barrens	66	-	-	-	-	-	-	23	19	-	-	43/47	-	9. Swanson and Johnson (1980), 5/1978-4/79. McDonalds Branch Basin; Pinus rigida and mixed oaks. Soil: pH-H2O 3.4-4.8; A: pH 3.8-4.1; H: pH 3.7-3.8. G=ground water, (conc. * estimated water flux).	
<i>Sweden</i>															
10 Spruce/Horröd	10	-	-	-	-	-	7.4	-	-	-	-	-1.2	-	10. Tyler (1981), 8/1977-12/79. Picea abies, 70 yr, podzol, mor. A=moss analysis. L=zero-tension lysimeter: 15 cm, A horizon. K=A-F.	
11 Spruce/Värsjö	4.3	1.3	9.2	-	-	7.1	3.7	15.2	-	14	>10.7	-9.6	-23.6	11. Bergkvist (1987c), 2 yr, 1980+1981. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4. F,G=z-t lysimeter: 15 cm, A hor/55 cm B hor. 7/1980-7/84. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.3.	
Pine Spruce/Gårdsjön	4.4	1.3	8.5	-	-	8	10	21.8	-	14	>22	-16.1	-41.3	12. Bergkvist (1987a), 1980+1981. a=Picea abies, 60 yr, b=opening 30*30 m. c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970. Podzol, mor, pH-KCl: 2.9-4.5. F,G=zero-tension lysimeter: 15 cm, A hor; 35 or 55 cm, B hor. L=Mineral soil budget: 15-35 or 15-55 cm. c=Picea abies, 50 yr. e=Fagus sylvatica, 100 yr. g=Spruce cleared 1967. Brown forest soil, pH-KCl: 3.1-4.3.	
12 Horröd:															
Spruce (a)	4.3	-	-	-	-	-	2.9	14.7	-	-	-	-10.4	-11.8		
Spruce (opening) (b)	4.3	-	-	-	-	-	3.8	6.6	-	-	-	-2.3	-2.8		
Beech (c)	4.3	-	-	-	-	-	10.7	7.6	-	-	-	-3.3	3.1		
Regeneration area (d)	4.3	-	-	-	-	-	9.3	6.4	-	-	-	-2.1	2.9		
Sjöbo:															
Spruce (e)	4.1	-	-	-	-	-	15.4	31.9	-	-	-	-27.8	-16.5		
Beech (f)	4.1	-	-	-	-	-	11.7	26.1	-	-	-	-22	-14.4		
Regeneration area (g)	4.1	-	-	-	-	-	24.5	14.6	-	-	-	-10.5	9.9		
13 Västerbotten, Svarberget Hälsingland, Kullarna Bohuslän, Gårdsjön II Bohuslän, Gårdsjön III Bohuslän, Gårdsjön IV	1.8 1.6 4.4 4.4 4.4	- - 1.3 1.3 1.3	- - 8.5 8.5 8.5	- - - - -	- - - - -	- - - - -	- - - - -	- - - - -	- - - - -	1.2 0.6 8.3 4 5.7	- - - - -	- - - - -	0.6 1 -2.6 1.7 0	- - - - -	Podzol; Pinus sylv., Picea abies; pH stream: 4.8. Podzol; P.s., P.a.; (Betula), pH stream: 5.0. Podzol; Pinus sylv., Picea abies; pH stream: 4.1. pH stream: 4.2. pH stream: 4.3.

Table III (continued)

Ecosystem/site	Bulk dep.	Dry dep.	Through-fall	Stem-flow	Litter-fall	Soil solution organic horizon	Soil solution below rhizosphere	Output from catchment	Bio-mass increment	Internal root uptake	Ecosystem budget	Soil budget	Remarks
	A	B	C	D	E	F	G	H	I	J	K	L	
											(A+B) -G or H	(C+D+E) -(G+H+J)	
<i>Denmark</i>													
14 Klosterhede/Picea	12	-	a 14	-	-	b 12 a,c 9	b 14 a,c 14	-	-	-	d -2 e -2	d,f -2 e,f -5	14. Rasmussen (1988), 3-4 yr, 1983/84-87. Spruce, 68 yr, Podzol, pH-CaCl ₂ , 0-5 cm: 2,7 a=2 yr, 1985-87; b=zero-tension lysimeter; A hor. 0-25 cm, AB hor. 0-65 cm, resp.; e=lens, lys., dept as b.; d=g-z lys.; e=tension lysimeter; f=mineral soil budget; g=1 yr, 1968-87.
Strødam/Picea	10	-	13	-	-	b 17 a,c,g 20	b 35 a,c,g 40	-	-	-	d -25 e -30	d,f -18 e,f -20	Spruce, 42 yr; Acid brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.1.
Tange/Picea	11	-	a 7	-	-	b 17 a,c 8	b 7 a,c 12	-	-	-	d 4 e -1	d,f 10 e,f -4	Spruce, 47 yr; Brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.6.

TABLE IIIa
Concentrations of Cu ($\mu\text{g L}^{-1}$)

Ecosystem/site	Bulk dep.		Through-fall	Stem-flow	Soil organic horizon	Soil mineral horizon	Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
	A	B								
<i>F.R.G.</i>										
1 Beech/Solling	23	19	18	18	24 (60)	18	-	-	-	1. Mayer (1981). 11/1974-8/1979. D=zero-tension lysimeter, E=tension lysimeter, 80 cm (in brackets: max. conc. in the system).
Spruce/Solling	23	30	-	-	22	26	-	-	-	2. Heinrichs and Mayer (1980). 11/1974-10/1977. E=tension lysimeter, 80 cm.
2 Beech/Solling	33	17	18	18	-	18	-	-	-	3. Mayer <i>et al.</i> (1980). 11/1974-10/1977. summer=May to Oct, winter=Nov to Apr.
Spruce/Solling	33	30	-	-	-	27	-	-	-	D=zerotension lysimeter
3 Beech - summer/Solling	14	14	15	15	-	-	-	-	-	E=tension lysimeter, 80-100 cm.
Beech - winter/Solling	53	19	20	20	-	-	-	-	-	
summer + winter	33.5	16.5	17.5	17.5	19.5	18	-	-	-	
Spruce - summer/Solling	14	25	-	-	-	-	-	-	-	
Spruce - winter/Solling	53	35	-	-	-	-	-	-	-	
summer + winter	33.5	30	-	-	22	27	-	-	-	
4 Spruce/Solling	2.3-2.5	-	-	-	3.8	9	-	-	-	4. Schultz (1987). 5/1983-4/85.
Spruce/Spangebck	2.3-2.5	-	-	-	9	9.6	-	-	-	D=lysometer plates or funnel lysimeters.
Spruce/Wingst	2.3-2.5	-	-	-	-	5.1	-	-	-	E=ceramic cups.
Spruce/Westerberg	2.3-2.5	-	-	-	-	1.9	-	-	-	tension lysimeters
Pine/Heide	2.3-2.5	-	-	-	-	-	-	-	-	
Beech/Solling	2.3-2.5	6.1	8.4	8.4	-	-	-	-	-	
Beech/Harste	2.3-2.5	6.6	4.7	4.7	-	-	-	-	-	
Oak/Heide	2.3-2.5	-	-	-	-	-	-	-	-	
5 S. Black Forest:										
Spruce/fir/beech	2.5-3.3	3.0-4.7	5.3-5.5	5.3-5.5	-	-	-	-	-	5. Mies (1987). 1982-84.
6 Spruce/Reinhardswald	10	80	-	-	-	-	-	-	-	6. Brechtel <i>et al.</i> (1986). 1983.
<i>Austria</i>										
7 Beech/Wienerwald	11	14	17	17	-	-	-	-	-	7. Kazda (1986). 5/1984-4/85.

Table IIIa (continued)

Ecosystem/site	Bulk dep.		Through-fall	Stem-flow	Soil solution			Soil solution		Stream water	Remarks
	A	B			C	D	E	F	G		
14 Spruce											
a	2	6.1	16	5.5	2	-	-	-	-	-	14. Bergkvist <i>et al.</i> (1988). 6/1984-5/1987.
b	1.3	5.8	10	5.2	1.4	-	-	-	-	-	Picea abies, Fagus sylvatica, Betula pendula.
c	1.3	6.2	11	7.4	1.2	-	-	-	-	-	a-c: podzols; d-e: acidic brown forest soils.
d	1.3	6.5	10	1.8	1.9	-	-	-	-	-	D=Zero-tension lysimeter: 15 cm, A horizon.
e	1.3	5	14	2.1	1.6	-	-	-	-	-	E=Zero-tension lysimeter: 50 cm, B horizon.
Beech:											
a	2	4.2	3.5	5.4	1.1	-	-	-	-	-	
b	1.3	2.7	4	5	1.3	-	-	-	-	-	
c	1.3	3.3	3.1	3.2	0.86	-	-	-	-	-	
d	1.3	4.3	2.7	1.8	1	-	-	-	-	-	
e	1.3	3.7	6.5	2.7	1.1	-	-	-	-	-	
Birch:											
a	2	5.2	5.6	2.4	1.1	-	-	-	-	-	
b	1.3	3	5	1.6	0.79	-	-	-	-	-	
c	1.3	2.4	5	2.5	1.7	-	-	-	-	-	
d	1.3	5	5.3	4.4	1.1	-	-	-	-	-	
e	1.3	3	11	4.1	4.5	-	-	-	-	-	
15 Västerbotten/Svartberget	-	-	-	-	-	-	-	-	-	1.6	15. Gråhn and Rosén (1983), 10/1980-9/
Hälsingland/Kullarna	-	-	-	-	-	-	-	-	-	0.4	1981. Podzol; Pinus sylvestris, Picea abies; pH stream: 4.8.
Bohuslän/Gårdsjön II	-	-	-	-	-	-	-	-	-	0.7	Podzol; Pinus sylvestris, Picea abies, (Betula sp.); pH stream: 5.0.
Bohuslän/Gårdsjön III	-	-	-	-	-	-	-	-	-	0.5	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.1.
Bohuslän/Gårdsjön IV	-	-	-	-	-	-	-	-	-	0.6	pH stream: 4.2. pH stream: 4.3

TABLE IIIb
Concentrations of Zn ($\mu\text{g L}^{-1}$)

Ecosystem/site	Bulk dep.		Through-fall	Stem-flow	Soil organic horizon	Soil mineral horizon	Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
	A	B								
<i>F.R.G.</i>										
1 Beech/Solling Spruce/Solling	142	104		1322(3100)	278	191	-	-	-	1. Mayer (1981). 11/1974-8/1979. D=zerotension lysimeter, E=tension lysimeter, 80 cm (in brackets: max. conc. in the system).
	142	288		-	456	560	-	-	-	
2 Beech/Solling Spruce/Solling	180	120		1720	-	190	-	-	-	2. Heinrichs and Mayer (1980). 11/1974-10/1977. E=tension lysimeter, 80 cm.
	44	67	1500	-	-	570	-	-	-	
3 Beech - summer/Solling Beech - winter/Solling	320	170	1900	-	-	-	-	-	-	3. Mayer <i>et al.</i> (1980). 11/1974-10/1977. summer=May to Oct, winter=Nov to Apr.
	182	119	1700	-	360	190	-	-	-	
Spruce - summer/Solling Spruce - winter/Solling	44	217	-	-	-	-	-	-	-	D=zerotension lysimeter E=tension lysimeter, 80-100 cm.
	320	480	-	-	-	-	-	-	-	
summer + winter Spruce/Solling	182	349	-	-	470	570	-	-	-	4. Schultz (1987). 5/1983-4/85.
	19-24	-	-	-	247	520	-	-	-	
Spruce/Spaubeck Spruce/Wingst	19-24	-	-	-	234	546	-	-	-	D=lysometer plates or funnel lysimeters. E=ceramic cups.
	19-24	-	-	-	-	306	-	-	-	
Spruce/Westerberg Pine/Heide	19-24	-	-	-	-	195	-	-	-	tension lysimeters
	19-24	69	87	-	-	-	-	-	-	
Beech/Solling Beech/Harste	19-24	39	38	-	-	-	-	-	-	5. Zöttl (1985). 6/1977-5/1979. Braunerde. E=tension lysimeter, 100 cm.
	19-24	-	-	-	-	-	-	-	-	
5 Spruce/Bärhalde	-	-	-	-	-	6	-	-	-	
6 S. Black Forest:										
Spruce/fir/beechn	9-15	20-34	34-45	-	-	-	-	-	-	6. Mies (1987). 1982-84.
7 Spruce/Reinhardswald	15	440	-	-	-	-	-	-	-	7. Brechtel <i>et al.</i> (1986). 1983.

Table IIIb (continued)

Ecosystem/site	Bulk dep.	Through-fall			Stem-flow	Soil solution organic horizon	Soil solution mineral horizon	Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
		A	B	C							
12 Spruce											
a	34	50	340	120	160	-	-	-	-	-	12. Bergkvist <i>et al.</i> (1988). 6/1984-5/1987. Picea abies, Fagus sylvatica, Betula pendula. a-c: podzols; d-e: acidic brown forest soils. D=Zero-tension lysimeter: 15 cm, A horizon. E=Zero-tension lysimeter: 50 cm, B horizon.
b	25	85	190	120	180	-	-	-	-	-	
c	25	75	230	63	170	-	-	-	-	-	
d	37	73	300	170	430	-	-	-	-	-	
e	37	98	350	210	360	-	-	-	-	-	
Beech:											
a	34	20	20	48	230	-	-	-	-	-	
b	25	30	18	61	210	-	-	-	-	-	
c	25	14	16	50	230	-	-	-	-	-	
d	37	21	14	140	250	-	-	-	-	-	
e	37	21	21	260	470	-	-	-	-	-	
Birch:											
a	34	31	76	66	59	-	-	-	-	-	
b	25	38	64	22	32	-	-	-	-	-	
c	25	31	96	42	19	-	-	-	-	-	
d	37	39	110	65	20	-	-	-	-	-	
e	37	52	170	110	85	-	-	-	-	-	
13 Västerbotten/Svartberget	-	-	-	-	-	-	-	-	-	14	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.8.
Hälsingland/Kullarna	-	-	-	-	-	-	-	-	-	11	Podzol; Pinus sylvestris, Picea abies, (Betula sp.); pH stream: 5.0.
Bohuslän/Gårdsjön II	-	-	-	-	-	-	-	-	-	21	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.1.
Bohuslän/Gårdsjön III	-	-	-	-	-	-	-	-	-	21	pH stream: 4.2.
Bohuslän/Gårdsjön IV	-	-	-	-	-	-	-	-	-	38	pH stream: 4.3.

Table IIIb (continued)

Ecosystem/site	Bulk dep.	Through-fall	Stem-flow	Soil solution organic horizon	Soil solution mineral horizon	Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
	A	B	C	D	E	F	G	H	
<i>Denmark</i>									
14 Klosterhede/Picea	-	-	-	82	9.6	-	-	-	14. Rasmussen (1986), 2 yr: 1984-86. Picea abies, 68 yr. Podzol, pH-CaCl ₂ , 0-5 cm: 2.7.
Strødam/Picea	-	-	-	129	295	-	-	-	Picea abies, 42 yr. Acid brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.1.
Tange/Picea	-	-	-	31	28	-	-	-	Picea abies, 47 yr. Brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.6. Zerotension lysimeters: D=A horizon 0-25 cm, E=AB horizon 0-65 cm.

Table IIIc (continued)

Ecosystem/site	Bulk dep.	Through-fall			Stem-flow	Soil solution			Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
		A	B	C		D	E	F				
8 Black Forest: Brown forest soil/podzol/ podzol	-	-	-	-	-	-	-	7.0/12/12	-	-	-	8. Feger (1986), 1984-85.
9 beech/oak	-	110/110	-	-	-	-	-	-	-	-	-	9. Block and Bartels (1985).
<i>Austria</i>												
10 Beech/Wienerwald	21	29	46	-	-	-	-	-	-	-	-	10. Kazda (1986), 5/1984-4/85.
<i>N. America</i>												
11 Maple-birch/Can., Turkey Lake												
dormant season	nd	nd	1	6	nd	-	-	-	-	nd	-	11. Foster and Nicolson (1986), 1983; remote site; D=zerotension lysimeters, F hor, E=tension lysimeters, 60 cm; nd=not detectable.
growing season	nd	nd	1	19	nd	-	-	-	-	nd	-	12. Swanson and Johnson (1980), 5/1978-4/1979, McDonalds Branch Basin; Pinus rigida and mixed oaks, Soil: pH-H20: 3.4-4.8, E=groundwater.
12 Pine-oak USA, NJ, Pine Barrens	18	-	-	-	3.6	-	-	-	-	3.8	-	13. Turner <i>et al.</i> (1985), 1979-82, McDonalds Branch Basin; Pinus rigida and mixed oaks, Soil: pH-H20: 3.4-4.8, E=groundwater.
13 Pine-oak USA, NJ, Pine Barrens	9	10	-	5	5	4	4	nd	nd	-	-	14. Friedland and Johnson (1985) 5/1983-10/1984, Abies balsamea-Picea rubens-Betula papyrifera. A=conc. in cloud. Zerotionen lysimeters: D=3 cm, E=org. hor. 12 cm; F=25 cm, G=40 cm.
14 Fir-spruce-birch USA, Vermont, Camels Hump	17 (51)	-	-	3.7	1.8	1.1	1.1	1	1	<0.6	-	

Table IIIc (continued)

Ecosystem/site	Bulk dep.	Through-fall	Stem-flow	Soil solution organic horizon	Soil solution mineral horizon	Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
	A	B	C	D	E	F	G	H	
<i>Sweden</i>									
15 Spruce/Horröd	-	-	-	10-65	-	-	-	-	15. Tyler (1981). 8/1977-12/1979. Picea abies, 70 yr, podzol, mor. D=zerotension lysimeter: 15 cm, A horizon.
16 Spruce/Värsjö	9	14.7	-	9.96	19.5	2.47	1.02	1.19	16. Bergkvist (1987c). 2 yr, 1980+1981. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4. Zero tension lysimeters: D=5 cm, org. hor.; E=15 cm, A hor. F=35 cm, B1 hor; G=55 cm, B2 hor. 7/1980-7/84. Picea abies, 80 yr. Podzol, mor, pH-KCl: 2.8-4.3.
Pine-Spruce/Gårdsjön	-	-	-	11.2	17.1	1.16	0.95	0.7	17. Bergkvist (1987a). 1980+1981. a=Picea abies. 60 yr. b=opening 30*30 m. c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970. Podzol, mor, pH-KCl: 2.9-4.5. Zero-tension lysimeters: D=5 cm, org. hor.; E=15 cm, A hor. F=35 cm, B1 hor. G=55 cm, B2 hor.
17. Horröd:									
Spruce (a)	-	-	-	16.3	32	1.63	2.6	-	a=Picea abies. 60 yr. b=opening 30*30 m.
Spruce (opening) (b)	-	-	-	13.1	7.52	0.63	1.37	-	c=Fagus sylvatica, 90 yr. d=Spruce cleared
Beech (c)	-	-	-	11	3.72	0.5	-	-	1970. Podzol, mor, pH-KCl: 2.9-4.5.
Regeneration area (d)	-	-	-	8.76	2.9	0.57	-	-	Zero-tension lysimeters: D=5 cm, org. hor.; E=15 cm, A hor.
Sjöbo:									
Spruce (e)	-	-	-	18.7	6.28	0.95	1.19	-	F=35 cm, B1 hor; G=55 cm, B2 hor.
Beech (f)	-	-	-	7.09	5.57	0.69	1.75	-	e=Picea abies, 50 yr. f=Fagus sylvatica,
Regeneration area (g)	-	-	-	6.22	1.29	0.7	-	-	100 yr. g=Spruce cleared 1967. Brown forest soil, pH-KCl: 3.1-4.3
18 Spruce									18 Bergkvist et al. (1988). 6/1984-5/1987. Picea abies, Fagus sylvatica, Betula pendula.
a	8.2	9.3	76	17	1	-	-	-	a-c: podzols; d-e: acidic brown forest soils.
b	7.9	13	27	22	1	-	-	-	D=Zero-tension lysimeter: 15 cm, A horizon. E=Zero-tension lysimeter: 50 cm, B horizon.
c	7.9	12	38	26	1.4	-	-	-	
d	8.5	9.6	37	1.6	0.97	-	-	-	
e	8.5	11	34	2.4	0.8	-	-	-	

Table IIIc (continued)

Ecosystem/site	Bulk dep.		Through-fall		Stem-flow	Soil solution organic horizon	Soil solution mineral horizon	Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
	A	B	C	D							
Beech:											
a	8.2	4.5	5.7	27	0.57	-	-	-	-	-	
b	7.9	3.8	8.6	20	0.31	-	-	-	-	-	
c	7.9	3	4.5	12	0.51	-	-	-	-	-	
d	8.5	3.2	4.6	2.2	0.43	-	-	-	-	-	
e	8.5	2.1	4	0.39	0.36	-	-	-	-	-	
Birch:											
a	8.2	2.9	12	2.2	2.1	-	-	-	-	-	
b	7.9	4.4	14	2.1	0.73	-	-	-	-	-	
c	7.9	3.4	12	6.1	0.29	-	-	-	-	-	
d	8.5	3.6	12	1.4	0.54	-	-	-	-	-	
e	8.5	4.3	18	0.99	1.2	-	-	-	-	-	
19 Västerbotten/Svartberget	-	-	-	-	-	-	-	-	-	0.87	19. Grahn and Rosén 1983). 10/1980-9/1981. Podzol; Pinus sylvestris, Picea abies; pH stream: 4.8.
Hälsingland/Kullarna	-	-	-	-	-	-	-	-	-	0.68	Podzol; Pinus sylvestris, Picea abies, (Betula sp.); pH stream: 5.0.
Bohuslän/Gårdsjön II	-	-	-	-	-	-	-	-	-	0.66	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.1.
Bohuslän/Gårdsjön III	-	-	-	-	-	-	-	-	-	0.7	pH stream: 4.2.
Bohuslän/Gårdsjön IV	-	-	-	-	-	-	-	-	-	0.87	pH stream: 4.3.

TABLE IIIId
Concentrations of Cd ($\mu\text{g L}^{-1}$)

Ecosystem/site	Bulk dep.	Through-fall			Stem-flow	Soil solution organic horizon D	Soil mineral horizon E	Soil solution mineral horizon F	Soil mineral horizon G	Stream water	Remarks
		A	B	C							
F.R.G. 1 Beech/Solling	1.5	1.45	1.7		2.4	2.8	-	-	-	1. Mayer (1981). 11/1974-8/1979. D=zero-tension lysimeter, E=tension lysimeter, 80 cm (in brackets: max. conc. in the system).	
	1.5	2.7	-		2.8	6.2 (9.1)	-	-	-		
2 Beech/Solling	3.4	1.8	2		-	2.9	-	-	-	2. Heinrichs and Mayer (1980). 11/1974-10/1977. E=tension lysimeter, 80 cm.	
	3.4	3.2	-		-	5.1	-	-	-		
3 Beech - summer/Solling	1.5	0.6	1.4		-	-	-	-	-	3. Mayer <i>et al.</i> (1980). 11/1974-10/1977. summer=May to Oct, winter=Nov to Apr. D=zero-tension lysimeter	
	5.3	3	2.6		-	-	-	-	-		
4 Spruce - winter/Solling	3.4	1.8	2		2.6	2.9	-	-	-	E=tension lysimeter, 80-100 cm.	
	1.5	1.9	-		-	-	-	-	-		
5 S. Black Forest:	5.3	4.6	-		-	-	-	-	-	4. Schultz (1987). 5/1983-4/85. D=lysimeter plates or funnel lysimeters. E=ceramic cups. tension lysimeters	
	3.4	3.25	-		2.7	5.1	-	-	-		
6 Spruce/Reinhardswald	0.19-0.35	-	-		2	4	-	-	-	5. Mies (1987). 1982-84. 6. Brechtel <i>et al.</i> (1986). 1983.	
	0.19-0.35	-	-		2	3	-	-	-		
7 Beech/Wienerwald	0.19-0.35	-	-		-	4	-	-	-	7. Glatzel <i>et al.</i> (1986). 5/1984-4/1985.	
	0.19-0.35	-	-		-	4	-	-	-		
8 Beech/Harste	0.19-0.35	0.5	0.7		-	-	-	-	-	5. Mies (1987). 1982-84. 6. Brechtel <i>et al.</i> (1986). 1983.	
	0.19-0.35	0.5	0.2		-	-	-	-	-		
9 Beech/Wienerwald	0.19-0.35	-	-		-	-	-	-	-	7. Glatzel <i>et al.</i> (1986). 5/1984-4/1985.	
	0.12-0.16	0.18-0.37	0.55-0.75		-	-	-	-	-		
10 Beech/Wienerwald	0.4	1.1	-		-	-	-	-	-	7. Glatzel <i>et al.</i> (1986). 5/1984-4/1985.	
	4.3	6.8	-		-	-	-	-	-		
Austria											
7 Beech/Wienerwald	4.9	9.4	19		-	-	-	-	-	-	7. Glatzel <i>et al.</i> (1986). 5/1984-4/1985.

Table IIIId (continued)

Ecosystem/site	Bulk dep.	Through-fall	Stem-flow	Soil solution organic horizon	Soil solution mineral horizon	Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
	A	B	C	D	E	F	G	H	
<i>N. America</i>									
8 Pine-oak USA, NJ, Pine Barrens	0.8	-	-	0.55	-	-	-	<0.1	8. Swanson and Johnson (1980). 5/1978-4/1979. McDonalds Branch Basin; Pinus rigida and mixed oaks. Soil: pH-H2O: 3.4-4.8. E=groundwater.
<i>Sweden</i>									
9 Spruce/Horröd	-	-	-	0.2-1.7	-	-	-	-	9. Tyler (1981). 8/1977-12/1979. Picea abies, 70 yr, podzol, mor. D=zerotension lysimeter: 15 cm. A horizon.
10 Spruce/Vårsjö Pine-Spruce/Gårdsjön	0.13	0.46	-	0.63	0.61	0.97	1.73	0.2	10. Bergkvist (1987c). 2 yr, 1980+1981. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4. Zero tension lysimeters: D=5 cm, org. hor.; E=15 cm, A hor. F=35 cm, B1 hor; G=55 cm, B2 hor. 7/1980-7/84. Picea abies, 80 yr. Podzol, mor, pH-KCl: 2.8-4.3.
11 Horröd: Spruce (a) Spruce (opening) (b) Beech (c) Regeneration area (d)	-	-	-	0.78	0.88	1.88	1.8	-	11. Bergkvist (1987a). 1980+1981. a=Picea abies. 60 yr. b=opening 30*30 m. c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970. Podzol, mor, pH-KCl: 2.9-4.5. Zero-tension lysimeters: D=5 cm, org. hor.; E=15 cm, A hor. F=35 cm, B1 hor; G=55 cm, B2 hor. e=Picea abies, 50 yr. f=Fagus sylvatica, 100 yr. g=Spruce cleared 1967. Brown forest soil, pH-KCl: 3.1-4.3.
Sjöbo: Spruce (e) Beech (f) Regeneration area (g)	-	-	-	0.94	2.4	1.66	2.7	-	12. Bergkvist et al. (1988). 6/1984-5/1987. Picea abies, Fagus sylvatica, Betula pendula. a-c: podzols; d-e: acidic brown forest soils. D=Zero-tension lysimeter: 15 cm, A horizon. E=Zero-tension lysimeter: 50 cm, B horizon.
12 Spruce a b c d c	0.16	0.38	2.5	1.6	1.6	-	-	-	
	0.13	0.43	1	0.56	2.4	-	-	-	
	0.13	0.48	1.5	0.63	2.7	-	-	-	
	0.15	0.39	1.9	2	3.2	-	-	-	
	0.15	0.46	1.9	2.1	1.2	-	-	-	

Table IIIa (continued)

Ecosystem/site	Bulk dep.			Through-fall		Stem-flow		Soil solution organic horizon		Soil solution mineral horizon		Soil solution mineral horizon		Stream water		Stream Remarks	
	A	B	C	D	E	F	G	H									
Beech:																	
a	0.16	0.2	0.1	0.83	2.8	-	-	-	-	-	-	-	-	-	-	-	
b	0.13	0.15	0.23	0.68	3	-	-	-	-	-	-	-	-	-	-	-	
c	0.13	0.13	0.15	0.67	1.9	-	-	-	-	-	-	-	-	-	-	-	
d	0.15	0.16	0.15	1.7	2.3	-	-	-	-	-	-	-	-	-	-	-	
e	0.15	0.14	0.12	2.7	1.1	-	-	-	-	-	-	-	-	-	-	-	
Birch:																	
a	0.16	0.16	0.46	0.73	0.77	-	-	-	-	-	-	-	-	-	-	-	
b	0.13	0.19	0.46	0.49	0.55	-	-	-	-	-	-	-	-	-	-	-	
c	0.13	0.16	0.46	0.3	0.34	-	-	-	-	-	-	-	-	-	-	-	
d	0.15	0.25	0.6	0.96	0.51	-	-	-	-	-	-	-	-	-	-	-	
e	0.15	0.22	0.81	1.3	0.6	-	-	-	-	-	-	-	-	-	-	-	
13 Västerbotten/Svartberget	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.09	
Hälsingland/Kullarna	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.09	
Bohuslän/Gårdsjön II	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.15	
Bohuslän/Gårdsjön III	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.19	
Bohuslän/Gårdsjön IV	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.15	
Denmark																	
14 Klosterhede/Picea	-	-	-	2.9	7.4	-	-	-	-	-	-	-	-	-	-	-	14. Rasmussen (1986), 2 yr: 1984-86. Picea abies; 68 yr. Podzol, pH-CaCl ₂ , 0-5 cm: 2.7.
Strødam/Picea	-	-	-	3	8	-	-	-	-	-	-	-	-	-	-	-	Picea abies, 42 yr. Acid brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.1.
Tange/Picea	-	-	-	3	5	-	-	-	-	-	-	-	-	-	-	-	Picea abies, 47 yr. Brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.6. Zerotionen lysimeters: D=A horizon 0-25 cm, E=AB horizon 0-65 cm.

Table IIIe (continued)

Ecosystem/site	Bulk dep.	Through-fall	Stem-flow	Soil solution organic horizon	Soil solution mineral horizon	Soil solution mineral horizon	Soil solution mineral horizon	Stream water	Remarks
	A	B	C	D	E	F	G	H	
Sjöbo:									
Spruce (e)	-	-	-	1.16	2.04	1.54	1.71	-	e=Picea abies, 50 yr. f=Fagus sylvatica, 100 yr. g=Spruce cleared 1967.
Beech (f)	-	-	-	1.04	0.81	0.69	0.79	-	Brown forest soil, pH-KCl: 3.1-4.3.
Regeneration area (g)	-	-	-	0.6	0.75	0.99	-	-	
7 Västerbotten/Svartberget	-	-	-	-	-	-	-	0.81	7. Grahn and Rosén (1983). 10/1980-9/1981. Podzol; Pinus sylvestris, Picea abies; pH stream: 4.8.
Hälsingland/Kullarna	-	-	-	-	-	-	-	0.34	Podzol; Pinus sylvestris, Picea abies, (Betula sp.); pH stream: 5.0.
Bohuslän/Gårdsjön II	-	-	-	-	-	-	-	0.6	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.1.
Bohuslän/Gårdsjön III	-	-	-	-	-	-	-	0.5	pH stream: 4.2.
Bohuslän/Gårdsjön IV	-	-	-	-	-	-	-	0.36	pH stream: 4.3.

TABLE III
Concentrations of Ni ($\mu\text{g L}^{-1}$)

Ecosystem/site	Bulk dep.		Through-fall		Stem-flow	Soil solution organic horizon		Soil solution mineral horizon		Stream water	Remarks
	A	B	C	D		E	F	G	H		
<i>F.R.G.</i>											
1 Beech/Solling	2.6	3.9	3.9	4.1	3.9	3.6	-	-	-	-	1. Mayer (1981). 11/1974-8/1979. D=zero-tension lysimeter, E=tension lysimeter, 80 cm (in brackets: max. conc. in the system).
Spruce/Solling	2.6	5.2	-	4.7	-	15 (26)	-	-	-	-	2. Heinrichs and Mayer (1980). 11/1974-10/1977. E =tension lysimeter, 80 cm.
2 Beech/Solling	2.5	3.9	3.8	-	3.8	-	3.1	-	-	-	3. Mayer <i>et al.</i> (1980). 11/1974-10/1977.
Spruce/Solling	2.5	5.2	-	-	-	15	-	-	-	-	summer=May to Oct, winter=Nov to Apr.
3 Beech - summer/Solling	2.1	2	2.7	-	2.7	-	-	-	-	-	D=zero-tension lysimeter
Beech - winter/Solling	2.9	5.7	4.9	-	4.9	-	-	-	-	-	E=tension lysimeter, 80-100 cm.
summer + winter	2.5	3.85	3.8	3.2	3.8	3.1	-	-	-	-	
Spruce - summer/Solling	2.1	4.3	-	-	-	-	-	-	-	-	
Spruce - winter/Solling	2.9	6.1	-	-	-	-	-	-	-	-	
summer + winter	2.5	5.2	-	4.1	-	15	-	-	-	-	
4 Spruce/Solling	0.63-1.37	-	-	-	-	-	-	-	-	-	4. Schultz (1987).
Spruce/Spanbeck	0.63-1.37	-	-	-	-	-	-	-	-	-	5/1983-4/85.
Spruce/Wingst	0.63-1.37	-	-	-	-	-	-	-	-	-	D=lysimeter plates or funnel lysimeters.
Spruce/Westerberg	0.63-1.37	-	-	-	-	-	-	-	-	-	E=ceramic cups.
Pine/Heide	0.63-1.37	-	-	-	-	-	-	-	-	-	tension lysimeters
Beech/Solling	0.63-1.37	1.7	2.1	-	2.1	-	-	-	-	-	
Beech/Harste	0.63-1.37	2.5	0.7	-	0.7	-	-	-	-	-	
Oak/Heide	0.63-1.37	-	-	-	-	-	-	-	-	-	
<i>Austria</i>											
5 Beech/Wienerwald	4.5	8.3	10	-	-	-	-	-	-	-	5. Kazda (1986). 5/1984-4/85.
<i>N. America</i>											
6 Pine-Oak USA, Pine Barrens	4.6	-	-	-	-	5.8	-	-	-	4.2	6. Swanson and Johnson (1980). 5/1978-4/1979. McDonalds Branch Basin; Pinus rigida and mixed oaks. Soil: pH-H2O: 3.4-4.8. E=groundwater.

Table IIIf (continued)

Ecosystem/site	Bulk dep.	Through-fall	Stem-flow	Soil solution			Soil solution			Stream water	Remarks
				organic horizon	mineral horizon	E	mineral horizon	mineral horizon	G		
	A	B	C	D	F		F	G	H		
<i>Sweden</i>											
7 Spruce/Horröd	-	-	-	0.6-4.5	-	-	-	-	-	-	7. Tyler (1981), 8/1977-12/1979. Picea abies, 70 yr, podzol, mor. D=zerotension lysimeter: 15 cm, A horizon.
8 Spruce/Värsjö	0.47	1.68	-	1.22	2.15	0.89	2.47	4.14	0.49	-	8. Bergkvist (1987c), 2 yr, 1980+1981. Picea abies, 80 yr, Podzol, mor, pH-KCl: 2.8-4.4.
Pine-Spruce/Gårdsjön	-	-	-	2.16	2.47	1.8		3.6	1	-	Zero tension lysimeters: D=5 cm, org. hor.; E=15 cm, A hor. F=35 cm, B1 hor; G=55 cm, B2 hor. 7/1980-7/84. Picea abies, 80 yr. Podzol, mor, pH-KCl: 2.8-4.3.
9 Horröd:											
Spruce (a)	-	-	-	2.34	4.16	1.83		6.03	-	-	9. Bergkvist (1987a), 1980+1981. a=Picea abies. 60 yr. b=opening 30*30 m. c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970.
Spruce (opening) (b)	-	-	-	1.16	2.34	1.45		2.75	-	-	Podzol, mor, pH-KCl: 2.9-4.5. Zero-tension lysimeters: D=5 cm, org. hor.; E=15 cm, A hor. F=35 cm, B1 hor; G=55 cm, B2 hor. 7/1980-7/84. Picea abies, 80 yr. Podzol, mor, pH-KCl: 2.8-4.3.
Beech (c)	-	-	-	1.48	3.89	4.2			-	-	9. Bergkvist (1987a), 1980+1981. a=Picea abies. 60 yr. b=opening 30*30 m. c=Fagus sylvatica, 90 yr. d=Spruce cleared 1970.
Regeneration area (d)	-	-	-	1.35	1.08	1.8			-	-	Podzol, mor, pH-KCl: 2.9-4.5. Zero-tension lysimeters: D=5 cm, org. hor.; E=15 cm, A hor. F=35 cm, B1 hor; G=55 cm, B2 hor. 7/1980-7/84. Picea abies, 80 yr. Podzol, mor, pH-KCl: 2.8-4.3.
Sjöbo:											
Spruce (e)	-	-	-	3.79	9.37	7.04		12.9	-	-	10. Grahñ and Rosén (1983). 10/1980-9/1981. Podzol; Pinus sylvestris, Picea abies; pH stream: 4.8.
Beech (f)	-	-	-	3.35	6.81	7.16		10.7	-	-	Podzol; Pinus sylvestris, Picea abies, (Betula sp.); pH stream: 5.0.
Regeneration area (g)	-	-	-	2.65	3.05	4.38			-	-	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.1.
10 Västerbotten/Svartberget	-	-	-	-	-	-			0.47	-	Podzol; Pinus sylvestris, Picea abies, (Betula sp.); pH stream: 5.0.
Hälsingland/Kullarna	-	-	-	-	-	-			0.25	-	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.2.
Bohuslän/Gårdsjön II	-	-	-	-	-	-			1.26	-	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.1.
Bohuslän/Gårdsjön III	-	-	-	-	-	-			0.79	-	Podzol; Pinus sylvestris, Picea abies; pH stream: 4.3.
Bohuslän/Gårdsjön IV	-	-	-	-	-	-			1.18	-	

Table IIIf (continued)

Ecosystem/site	Bulk dep.	Through-fall	Stem-flow	Soil solution organic horizon	Soil solution mineral horizon E	Soil solution mineral horizon F	Soil solution mineral horizon G	Stream water	Remarks
	A	B	C	D	E	F	G	H	
<i>Denmark</i>									
11 Klosterhede/Picea	-	-	-	3.3	2.8	-	-	-	11. Rasmussen (1986), 2 yr: 1984-86. Picea abies, 68 yr. Podzol, pH-CaCl ₂ , 0-5 cm: 2.7.
Strødam/Picea	-	-	-	5.4	12.1	-	-	-	Picea abies, 42 yr. Acid brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.1.
Tange/Picea	-	-	-	3.6	2.3	-	-	-	Picea abies, 47 yr. Brown forest soil, pH-CaCl ₂ , 0-5 cm: 3.6. Zerotension lysimeters: D=A horizon 0-25 cm, E-AB horizon 0-65 cm.

3. Deposition to the Canopy

Metals are deposited to the tree canopy as wet and dry deposition. Wet deposition includes precipitation in the form of rain and snow as well as fog and cloud droplets. Dry deposition comprises particle deposition and gas sorption.

'Bulk deposition' is commonly defined as the total wet and dry fallout collected in a continuously open vessel placed in an open field. Bulk deposition includes an undefined portion of the true dry deposition. This portion varies with meteorology and the conditions of sampling, etc. The bulk deposition collected in continuously open vessels is sometimes called wet deposition, however, which contributes much confusion. 'Wet-only deposition' is collected in vessels that are open only during periods with precipitation. Standardized measuring devices for 'wet-only' and 'dry-only' sampling are commercially available. The sum of these two fractions need not be equal to bulk deposition as measured in a continuously open vessel, however.

The methods of collecting bulk or wet deposition vary greatly between studies. Design and spacing of vessels, duration of sampling and evaporation losses are examples of factors that greatly influence the results. Many reports lack a detailed description of the methods, however, which renders any comparison between studies difficult. For a careful and detailed evaluation or comparison of the data compiled in Tables I and II, the original reports should therefore be consulted.

Dry deposition to a forest is a process involving interaction with the vegetation surface. The surface of the unchanged receiving system (e.g. a tree canopy) would, strictly speaking, be the only acceptable sampler. Apart from all other processes involved in dry deposition, foliar uptake and excretion of elements as well as other results of biological activities in the canopy continuously interact with the processes of dry deposition. The true dry deposition can therefore hardly be quantified directly but may be estimated in several indirect ways requiring special measurement devices and/or the use of models (Bengtson *et al.*, 1977; Wiman, 1984; 1985; Wiman and Lannefors, 1984; Lindberg and Lovett, 1985; Schmidt and Mayer, 1986). Different techniques have been developed for the measurements of dry deposition of atmospheric compounds to underlying surfaces. Measurement capabilities are gradually improved, but discrepancies exist between measurement methods. Hicks *et al.*, (1986) discuss the state of the art and the causes of the differences that exist among measurement methods; these aspects have been reviewed recently by Nicholson (1988). Recent development in surface analysis methods such as analysis of foliage for natural radionuclides (Bondietti *et al.*, 1984), and throughfall measurements (Lovett and Lindberg, 1984) are promising both in the context of routine monitoring and intensive measurement. Natural surfaces, although difficult to simulate either mathematically or physically, integrate the net effects (emanating from deposited minus resuspended material) of dry deposition of various atmospheric constituents. Surface analysis methods can be used to quantify the amount of deposited material residing on such surfaces. From knowledge of the area of the surface in question and the duration of the period during which dry deposition occurred, the net dry

deposition flux to the ground can be computed.

The often great contribution of dry deposition to the total input of heavy metals to forest ecosystems makes calculations of ecosystem budgets using only the wet part of the total deposition basically unreliable.

According to a recent review, the reliability of many precipitation chemistry data for rural or remote sites has to be questioned because of possible contamination (Barrie *et al.*, 1987). Included in Table I are remote-site precipitation data that these authors consider reliable.

Despite the differences between methods and the frequent lack of a detailed method description, the data indicate that both wet (bulk) and dry deposition are greater in central Europe than in Scandinavia (Table I). The rather few cases where dry deposition has been measured in addition to wet (bulk) deposition indicate that dry deposition is often more important than bulk deposition in polluted regions like F.R.G. (Table II). In less polluted regions, like Sweden, bulk deposition is often more important than dry deposition. Strictly speaking, it should not be justified to compare bulk and dry deposition since bulk deposition includes an undefined contribution from the true dry deposition, as mentioned above.

The measurements at Solling, F.R.G. having been performed over an extended period of time, often reveal much higher concentration and bulk-deposition values for older sampling periods than currently. The reason is not explicitly stated in the later reports.

The dry deposition to a forest is much greater than to a grass-covered or otherwise open area. Due to aerodynamic properties and to the greater leaf area, conifers are much more efficient in trapping aerosols than are deciduous trees (Höfken and Gravenhorst 1982; Mayer and Ulrich 1982). The dry deposition to deciduous trees is greater when they are foliated than during the winter (Höfken *et al.*, 1983; Schmidt and Schultz, 1985). However, the higher pollution load of metals commonly occurring during the winter may well increase the deposition.

Dry deposition rates to spruce forests were estimated by the experiment and model approach by Wiman (1984; 1985). From these estimations the dry deposition to a mature Norway spruce forest was calculated by Bergkvist (1987c). The wet deposition of Cu, Zn, Pb, Cr and Ni dominated over dry deposition, but wet and dry deposition were of equal importance in the case of Cd (Table II).

In spruce forests in SW Sweden, bulk deposition contributes 10 to 30% of the total input (including litterfall) of Cu, Zn, Cr, and Ni to the forest floor (Bergkvist, 1987c). The corresponding value for Pb and Cd was about 50%.

It has also been shown that cloud and fog droplet deposition of certain elements to forest ecosystems at high altitude could be more important than bulk deposition (Lovett *et al.*, 1982; Grosch and Schmitt, 1985; Gietl and Rall, 1986; Glatzel *et al.*, 1986). In a wood of European beech close to Vienna, the proportion that fog contributed to the deposition to the forest floor was 43% for Cd, 24% for Pb, 24% for Ni and 15% for Cu (Kazda, 1986).

4. Tree-Layer Interactions

Passing the canopy, the precipitation is often enriched with heavy metals. The extent of the enrichment varies greatly between metals and tree species and with stand structure, soil fertility and pollution load (Table III; Nihlgård, 1970; Astrup and Bülow-Olsen, 1979; Heinrichs and Mayer, 1980).

Interception by the canopy reduces the water amount reaching the forest floor by about 25% (Parker, 1983). Interception is commonly greater in conifers than in deciduous trees (Block and Bartels, 1985; Bergkvist *et al.*, 1988). In spite of this, the metal amounts in throughfall are commonly greater than the amounts deposited by precipitation to the canopy, especially so in conifers. The metal concentrations in throughfall are much greater (often 2 to 4 times greater) under conifers than under deciduous trees (Table III). In some cases, e.g. Zn and Cd in deciduous stands in S Sweden, there is only little difference between the concentration in open-field precipitation and throughfall.

The annual flux of heavy metals in throughfall is thus commonly greater than in open-field precipitation (Table II). Lead is often an exception. Its internal cycling is small and aerial deposition and mobility of soil-organic matter determine the flux through the ecosystem. Lead is thus often trapped by canopies so that throughfall contributes a lower Pb flux than open-field precipitation. This has been reported from relatively unpolluted areas like Sweden and the Black Forest in F.R.G. In the more heavily polluted regions of F.R.G. throughfall in spruce forests can contribute more Pb than open-field precipitation, but in beech forests the flux of Pb is the same above and below the canopy (Godt, 1986; Mayer and Schultz, 1987). In other studies in F.R.G., lower as well as higher input of Pb via throughfall as compared with the open field are reported, as reviewed by Mies (1987). Lindberg

TABLE IV

Deposition to forest edge vs. forest interior, Teutoburger Wald, FRG, 1982/83; g ha⁻¹ yr⁻¹ Spruce, 86 yr, podzolic Braunerde, Moder, pH-KCl: 2.5-3.6 (Godt, 1986)

	Cu	Zn	Pb	Cd	Cr	Ni
<i>Edge</i>						
Wet deposition	558	548	292	4.3	8.3	80
Throughfall	482	2117	708	16.2	9.8	55
Dry deposition	660	650	350	5.1	9.9	100
Plant wash-off	740	920	70	6.7	-8.4	-120
Total deposition	1220	2670	640	9.4	18.2	180
<i>Interior</i>						
Wet deposition	558	548	292	4.3	8.3	80
Throughfall	361	1168	423	8.1	5.0	55
Dry deposition	128	130	66	1.0	1.9	18
Plant wash-off	325	490	66	2.8	-5.2	-44
Total deposition	686	680	358	5.3	10.2	98

and Harriss (1981) also found the internal cycling of Pb to be negligible. They also found that the input of Zn and Cd to the soil surface is mainly part of the internal cycling of these elements.

Zinc and Cd concentrations often show considerable augmentation from open-field precipitation to throughfall (Table III; Bergkvist, 1987c; Mies, 1987). These metal ions are readily taken up by the tree roots. A certain amount is incorporated into the biomass and temporarily removed from internal cycling. Well above half the amounts of these metals taken up from the soil have been reported to be recycled to the soil, either leached from the needles or returned as litterfall (Bergkvist, 1987c).

Very high throughfall input of Zn has been reported from Czechoslovakia, even in relatively little polluted areas (Lochman, 1985).

The local topography and canopy structure are important for the amount of atmospheric pollutants delivered to forest ecosystems as particles or gases by dry deposition. In general, wet deposition is known to be more evenly deposited over large areas, but in upland areas where orographic enhancement of rain is important there may also be large spatial variations in wet deposition of pollutants (Unsworth and Fowler, 1988). Hill tops and forest edges exposed to polluted air may receive a larger deposition than low elevation stands or the interior of a forest (Godt, 1986; Hasselrot and Grennfelt, 1987).

Spruce canopies, efficiently trapping acidic aerosols, are known to lower the pH of incident precipitation, whereas beech canopies increase it, at least in rural areas (Nihlgård, 1970; Mayer and Ulrich, 1977; Block and Bartels, 1985; Bergkvist *et al.*, 1987a). In studies reviewed by Mies (1987), the H⁺ input with the throughfall averaged 1.3 kg ha⁻¹ yr⁻¹ in coniferous stands and 0.5 in deciduous stands. This was 2.7 and 1.1 times the amount deposited by the bulk deposition to the open field, respectively. Also the internal production of acids differs between vegetation types. In a spruce stand in F.R.G. the total annual H⁺ load from deposition and internal sources was about twice the H⁺ load in a beech stand (Matzner and Ulrich, 1981). In a 3-yr study of five sets of adjoining stands of spruce, beech and birch in S Sweden, the average ratio of H⁺ flux between throughfall and bulk deposition was 2.4 for the spruce stands, 0.6 for the beech stands and 1.0 for the birch stands (Bergkvist *et al.*, 1988).

Stemflow usually contributes less than 10% of the water reaching the ground in a stand (Parker, 1983; Block and Bartels, 1985; Bergkvist *et al.*, 1988). Stemflow is characterized by high concentrations of metals, however, and especially so in conifers (Table III). The concentrations are highly dependent on the volume of stemflow which varies with the density and structure of the canopy, the twig insertion, the intensity and duration of rainfall, etc. Especially in conifers, stemflow can thus contribute a significant amount of metals to the ground. In the Swedish study mentioned above, the water flux in the stemflow of the birch trees was only 2 to 9% of that in the throughfall, but the stemflow flux of Zn was 6 to 31% of that in the throughfall (Table II; Bergkvist *et al.*, 1988). Very high Zn concentrations in stemflow water were reported in the older German reports (Table III).

Litterfall transports less Cu and Cd and usually Zn and Pb than does throughfall. However, litterfall is more important than throughfall for the flux of Zn in birch (Bergkvist *et al.*, 1988) and alder (Asche, 1985). In the relatively unpolluted Black Forest, more Pb is being transported in litter than in throughfall of spruce. In the S Swedish studies, throughfall and litterfall were found to be of equal importance for the transport of Pb in the tree species studied. Litterfall seems to be important for the transport of Cr and Ni.

5. Leaching Through the Soil

5.1. LEACHABILITY

The leachability in forest soils differs greatly among metals. Generally, metals demonstrate two distinct patterns of release. In the first pattern a crucial role is played by soluble organic acids, predominantly humic substances. These are released in great quantities when the organic matter in the top soil is mineralized. The organic acids are transported through the A horizon with the percolating soil water. Lead, Cu and Cr are known to form stable complexes with dissolved organic acids and are transported through the soil in a complexed form (Himes and Barber, 1957; Stevenson, 1972; 1975; 1976; Verloo *et al.*, 1973; Cheshire *et al.*, 1977; Kirkham, 1977; Keilen, 1978). The amounts of these metals show a close correlation with the dissolution of organic matter, as shown in a podzol by Tyler (1981) and in podzols and brown forest soils by Bergkvist (1986a, 1987a,c). A high biological activity in the mor layer favors the formation of dissolved organic acids and, hence, the release of these metals from the soil. Maximum release was found in late summer and autumn, at high soil temperature and soil moisture.

As the organic acids percolate through the A horizon they become saturated with Fe and Al and will finally precipitate in the upper part of the B horizon (Petersen, 1976). Most of the Cu, Pb and Cr released from the A horizon is accordingly accumulated in the upper part of the B horizon of podzols as well as in the upper (B) horizon of brown forest soils (Bergkvist, 1987a, c).

The second pattern is associated with soil acidity. A gradual release of metals from the mineral soil and an increase in soil solution concentration of metals through the B horizon are characteristic features of Zn, Cd, and Ni (Bergkvist, 1987a, c). These metals are very susceptible to changes in soil acidity, as demonstrated by experimental acidification of lysimeter-contained soils (Bergkvist, 1986a). The influence of accelerated soil acidification on the increased leachability of these metals is also shown in other studies (for references see Bergkvist, 1986a).

5.2. METAL SPECIATION

The chemical speciation of a metal may be defined as the distribution of the total concentration into various chemical forms (species). The most important factor controlling the speciation of metals in soil solutions is the concentrations of various

TABLE V

Inorganic speciation of Cu, Pb, Cd, and Zn at different pH and ligand concentrations. All data are from A horizons. Equilibrium calculations were performed using stability constants from Lindsay (1979). Only species contributing 1% or more of the total inorganic metal concentration are included

Ref.	pH	Ligand conc. (μM)				% of total inorganic metal concentration								
		SO_4^{2-}	Cl^-	NO_3^-	Cu^{2+}	CuSO_4^0	Pb^{2+}	PbSO_4^0	PbCl^+	Cd^{2+}	CdSO_4^0	CdCl^+	Zn^{2+}	ZnSO_4^0
1	4.04	68	16	8	99	1	98	2	<1	98	2	<1	99	1
2a	3.4	200	150	0	97	3	94	6	<1	95	4	1	97	3
2b	3.9	100	150	300	98	2	96	3	1	97	2	1	98	2
3	3.90	137	200	60	98	2	95	4	1	95	3	2	98	2

Ref. 1: Cronan (1980), balsam fir (*Abies balsamea*), podzol.

Ref. 2: Bourg and Védry (1986), (a) Scots pine (*Pinus sylvestris*), podzol. (b) European silver fir (*Abies alba*), acidic brown forest soil. 1 mM ionic strength was assumed.

Ref. 3: Nilsson and Bergkvist (1983), Norway spruce (*Picea abies*) and Scots pine, podzol. Cl^- and NO_3^- data not published. 1 mM ionic strength was assumed.

ligands present and the stability of the resulting complexes. The ligands present in forest soil solutions are either inorganic or organic in nature. Cl^- and SO_4^{2-} are the most important inorganic ligands. The organic ligands can be divided into two groups: i) biochemicals of the type known to occur in living organisms, such as simple aliphatic acids, amino acids, sugar acids etc., and ii) acidic, yellow to black colored polyelectrolytes referred to as humic substances (HS; fulvic + humic acids) (Stevenson and Fitch, 1986). In fresh water, the bulk of the dissolved organic carbon (DOC) consists of HS (Reuter and Perdue, 1977).

A recent study on soil solutions from a lysimeter-contained brown forest soil (0 to 15 cm) of a Silver birch forest in S Sweden showed that HS made up about 90% of the DOC (D. Berggren, unpublished data). The HS have also been shown to complex metals most effectively (Geering and Hodgson, 1969). The relative importance of common inorganic anions as ligands to Cu, Zn, Pb, and Cd is shown in Table V. The data originate from investigations in the north-east of USA (Cronan, 1980), in the north-east of France (Bourg and Védry, 1986) and on the Swedish west coast (Nilsson and Bergkvist, 1983). It is obvious that the inorganic ligands only insignificantly affect the speciation of the metals. The free hydrated metal makes up 94 to 99% of the total inorganic metal concentration, depending on metal and sampling site. The critical question is thus to what extent organic ligands affect the speciation. Since the bulk of the DOC in a soil solution is HS, which also form the strongest metal complexes, the interest must be focussed on HS.

The speciation of Cu, Pb, and Cd in HS-containing solutions from a brown forest soil is shown in Figures 1 and 2. The following conclusions can be drawn:

- (i) Under all the experimental conditions, the HS were the most important ligands for all metals studied;
- (ii) The strength of the complexes formed between the HS and the metals decreased in the following order: $\text{Cu} > \text{Pb} \gg \text{Cd}$;
- (iii) The relative importance of humic complexation on the speciation increased greatly with pH and concentration of HS; and
- (iv) The proportion of metal complexed to HS increased as the metal to HS ratio decreased.

At pH 5.0 and a HS concentration of 40 mg C L^{-1} , the proportion of Cu, Pb and Cd in complexes with HS was 43, 93 and 97%, respectively (Berggren, 1989). This C concentration is lower than the DOC concentrations of 100 to 150 mg L^{-1} found in soil solutions from the A2 horizon in Swedish podzol soils (Nilsson and Bergkvist, 1983; Bergkvist, 1987a, c), though in the same range as in soil solutions from the A horizon of brown forest soils of S Sweden (Bergkvist, 1987a). The conclusions above agree with previous findings in the field of metal-humic complexation (see, e.g., Stevenson and Fitch, 1986; Buffle and Altmann, 1987). From data of fulvic acid stability constants (Mantoura *et al.*, 1978) one can suggest that Zn would behave like Cd in the solutions of Figure 1.

Various analytical approaches have been used in order to determine the speciation of trace metals in natural waters and soil solutions:

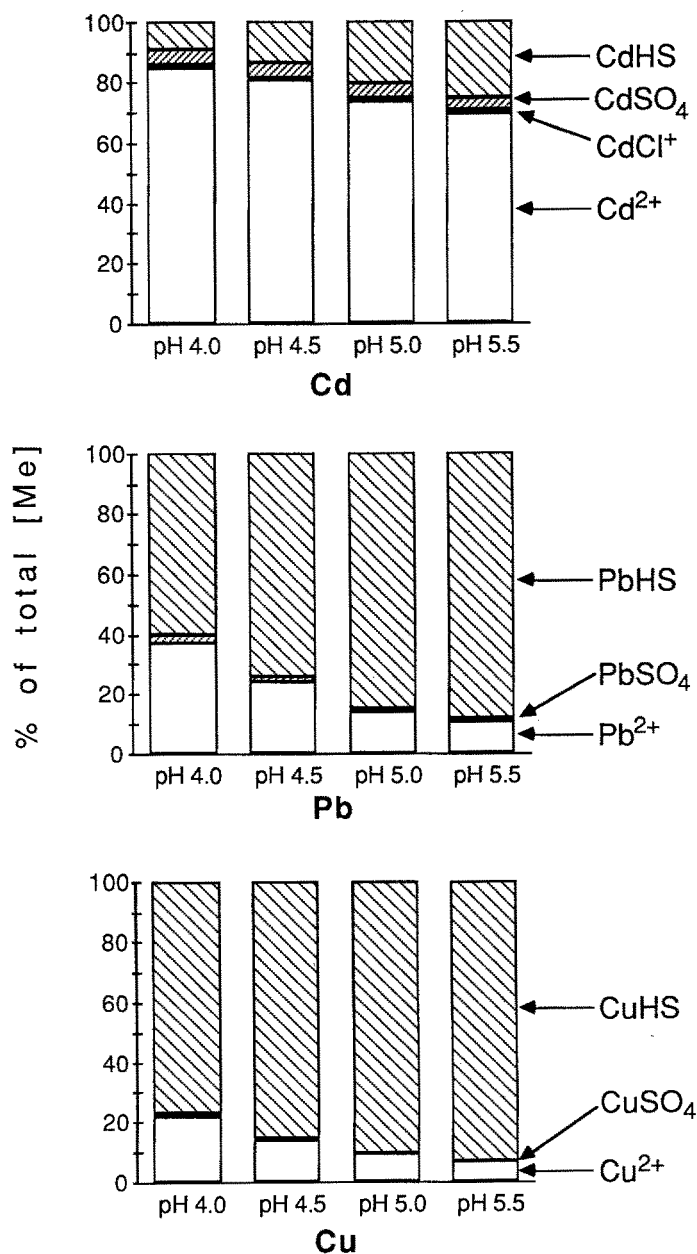


Fig. 1. The relative speciation of Cd, Pb and Cu in a soil solution (collected at 15 cm depth of a brown forest soil) dialyzed against a dilute inorganic solution having a composition of major cations and anions about the same as the soil solution (outer solution). The outer solution also contained 2.00, 10.0 and 10.0 $\mu\text{g L}^{-1}$ of Cd, Pb and Cu, respectively. The speciation of a metal, at equilibrium, was obtained by an analysis of total concentration in the soil solution and in the outer solution, followed by thermodynamic calculations of the inorganic speciation (stability constants from Lindsay, 1979). Total concentrations in the soil solutions, at equilibrium, increased with pH and were in the range 2.17 to 2.77, 21.0 to 70.3 and 28.9 to 88.1 $\mu\text{g L}^{-1}$ for Cd, Pb and Cu, respectively and the concentration of humic substances (HS) was 8.7 mg C L^{-1} at pH 4.0 and 9.8 mg C L^{-1} at pH 4.5 to 5.5. Data from Berggren (1989).

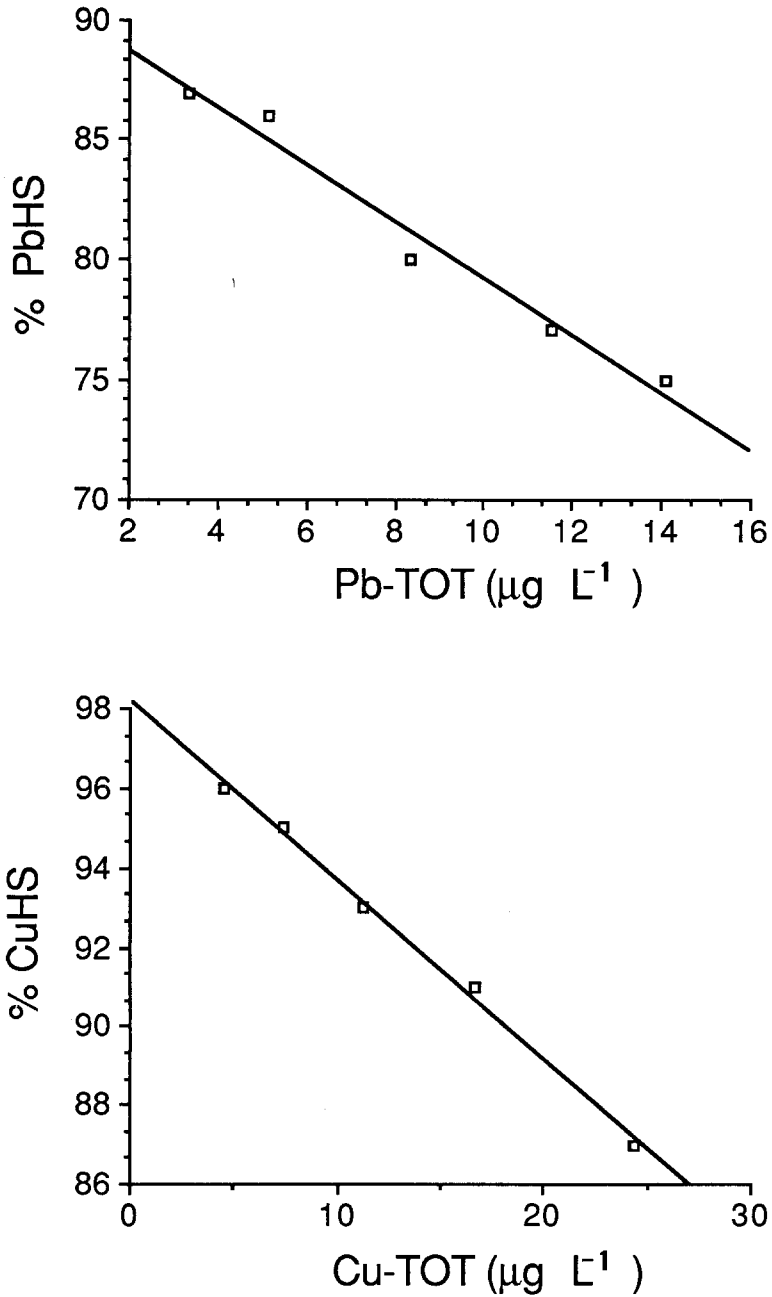


Fig. 2. The relationship between the total concentration of Pb and Cu and the fractions complexed to humic substances (HS) at constant pH (4.5) and HS concentration (\bar{x} =7.61 mg C L⁻¹, s.d.=0.18). For a brief explanation of the analytical method, see Figure 1. Data from Berggren (1989).

- (i) ion selective electrodes (Sanders, 1982 and Minnich and McBride, 1987 for Cu);
- (ii) Donnan dialysis (Cox *et al.*, 1984 for Cu, Zn, Pb, and Cd; Minnich and McBride, 1987 for Cu);
- (iii) an ion exchange equilibrium technique (Sanders, 1983 for Zn; Werner, 1987 for Zn and Cd);
- (iv) an ion exchange column technique (Cox *et al.*, 1984 for Cu, Zn, Pb, and Cd; König and Ulrich, 1986 for Cu, Zn, Pb, Cd and Cr; Berggren, 1989 for Cu, Pb, and Cd; and
- (v) an immiscible displacement method (Hodgson *et al.*, 1965 for Cu and Zn).

Generally, the analytical results agree very well with the results presented in Figure 1, i.e. Cu and Pb were predominantly present as organic complexes and Cd and Zn as free metals.

6. Metal Budgets

Metal cycling through forest ecosystems has been studied in central F.R.G. for a long period of time by Ulrich and co-workers. The research efforts have been concentrated to the Solling area (Heinrichs and Mayer, 1977, 1980; Seekamp, 1977; Mayer and Schultz, 1987). The soil type is an acidic brown forest soil on a residual loess loam. The mean annual precipitation is 1060 mm.

Positive ecosystem budgets as defined in the Materials section, were found for Cu, Pb, and Cr in both the spruce and the beech ecosystems at Solling (Table II; Figure 3). Zinc was likewise accumulated in the beech stand but was lost from the spruce stand. The ecosystem budgets were usually negative for Cd, most so for the spruce ecosystems (Table II; Figure 4). The loss of Zn, Cd and Ni via the soil solution under the rooting zone was remarkably high from the spruce stand. The more recent Solling data differ from the earlier data in revealing considerably lower fluxes of metals in the above-ground parts of the ecosystems.

From several studies on stands of different tree species in northern F.R.G. positive ecosystem budgets were calculated for Cu and Pb. Zinc displayed a positive budget in some cases, a negative in others, such as a pine stand on a podzol overlaying shifting sand. Chromium showed a net accumulation in a moderately acidic brown forest soil with beech but was almost at balance or was lost in spruce forests with very acidic podzols on sandy moraines. Net loss of Cd was reported from all sites. Nickel was considerably leached from an acidic brown forest soil supporting a heavily damaged spruce stand.

Budget studies are also available from a forested watershed in the Black Forest (Keilen, 1978; Zöttl *et al.*, 1979; Stahr *et al.*, 1980; Trüby and Zöttl, 1984). The soil parent materials are periglacial solifluction layers and moraines derived from and covering the extremely acidic granite in the Bärhalde watershed. This mountainous area is humid (mean annual precipitation: 1950 mm) and cool but less polluted than the Solling area further north. The ecosystems at Bärhalde showed

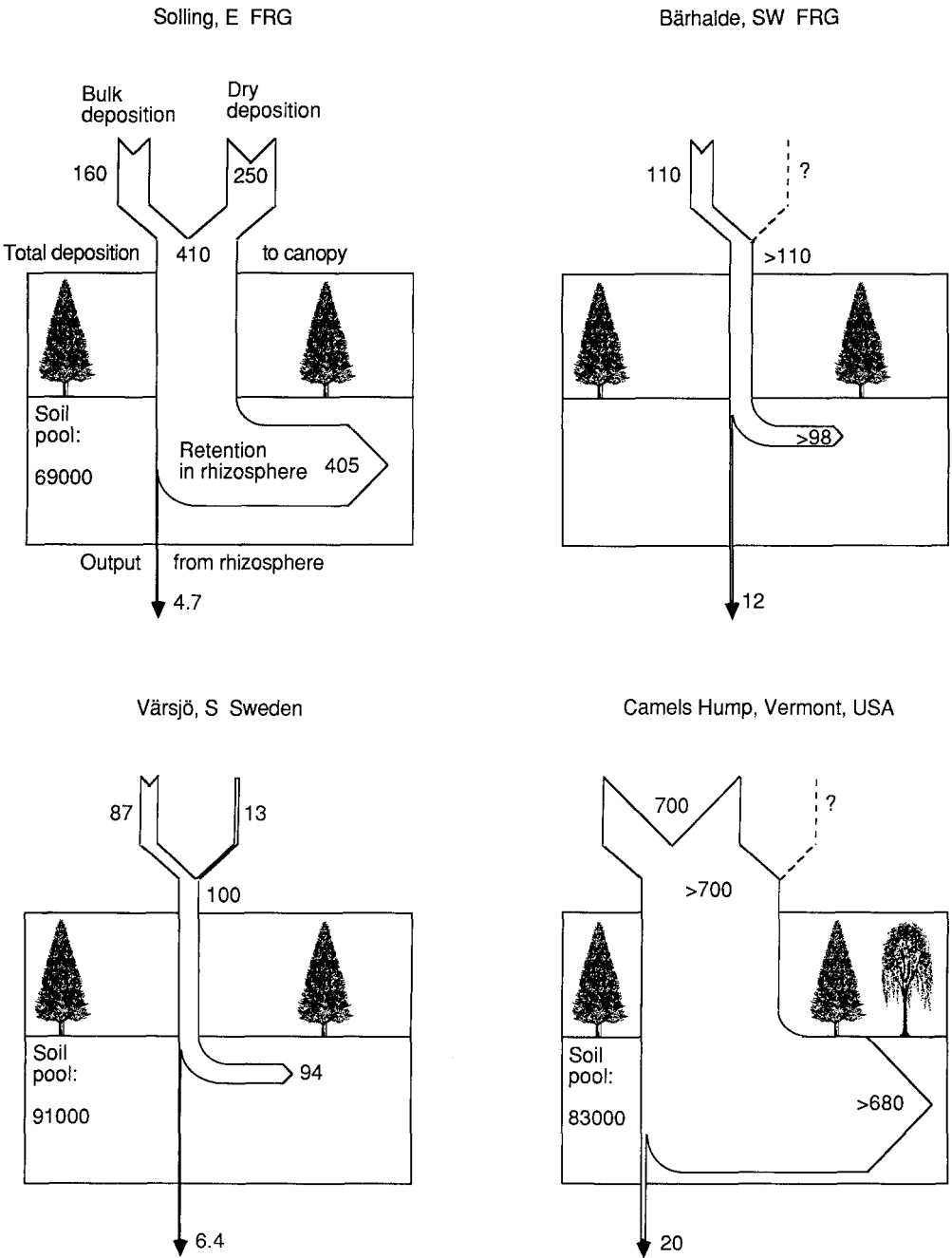
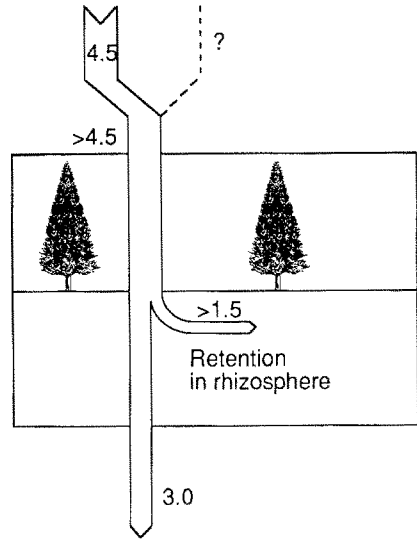
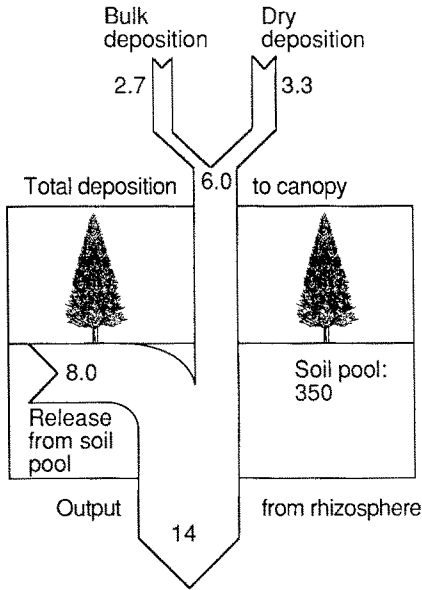


Fig. 3. Annual ecosystem budgets of Pb in four typical forest ecosystems. Flows in $\text{g ha}^{-1} \text{ yr}^{-1}$, soil pools in g ha^{-1} . Solling: *Picea abies*; acidic brown forest soil overlaying loess; soil pool (0–50 cm): forest floor HNO_3 digested, mineral soil EDTA extractable. From Schultz (1987). Bärhalde: *Picea abies*; acidic brown forest soil overlaying moraine, acidic granite; soil 0–100 cm. From Stahr *et al.* (1980). Vårsjö: *Picea abies*; podzol overlaying sandy glacial till, acidic siliceous rock; soil pool (0–55 cm): HNO_3 digested. From Bergkvist (1987c). Camels Hump: *Abies balsamea*, *Picea rubens*, *Betula papyrifera*; Haplorthods, Fragiorthods with O horizons > 10 cm; soil pool (0–52 cm): ashed + digested. From Friedland and Johnson (1985).

Solling, E FRG

Bärhalde, SW FRG



Strødam, E Denmark

Värsjö, S Sweden

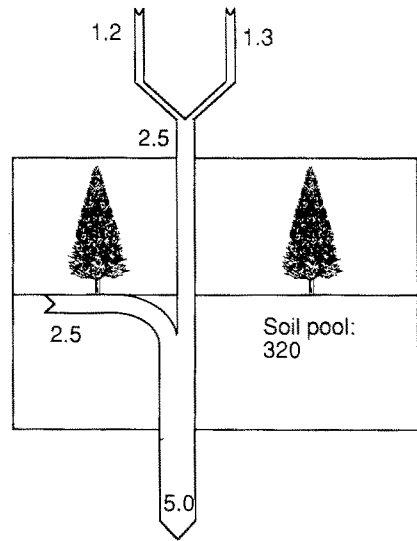
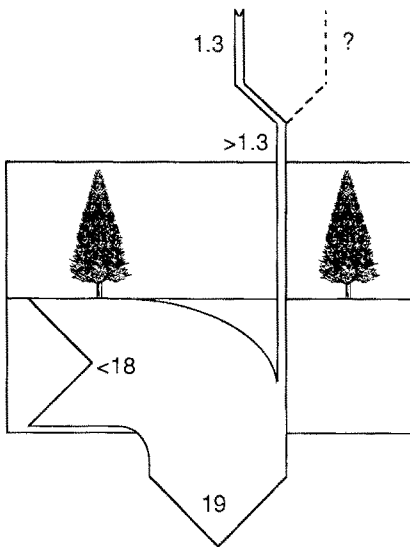


Fig. 4. Annual ecosystem budgets of Cd ($\text{g ha}^{-1} \text{yr}^{-1}$) in four typical coniferous-forest ecosystems. Strødam: *Picea abies*; acidic brown forest soil overlaying sandy glacial deposit; soil 0–65 cm. From Rasmussen (1986; 1988). See further Figure 3.

positive budgets of Cu, Pb, and Ni, usually also of Zn and Cd with the exception of a podzol (Table II; Figures 3 and 4). Percolating water leaving the subsoil had lower metal concentrations than at Solling (25 to 50% lower for Pb and Cd; 10 for Cu) but the deposition input is also lower at Bärhalde. Nevertheless, the differences between the two F.R.G. sites should in part be due to differences in humidity and soil properties.

Budgets for a spruce forest on the Swedish west coast (Gårdsjön) and one in southernmost Sweden (Värsjö) are given in Bergkvist (1987c). The soils are podzols on sandy-silty glacial tills originating from siliceous rocks. The mean annual precipitation amounts to 730 and 785 mm, respectively. The type of lysimeter used is an open zero-tension plexiglass lysimeter. Most metals are to some extent accumulated in the A horizon at Värsjö (Table II). The A horizon at Gårdsjön is losing Cd and Cr. There is a net release of several metals from the B horizon at the two sites. This is true for Zn, Cd, and Ni. These metals have a negative budget in the entire mineral soil, except Cd at Gårdsjön where input equals output.

The spruce forest ecosystem at Värsjö is losing Zn, Cd, and Ni – the ecosystem budget is negative (Figure 4). Almost at balance is Cr, whereas Cu and Pb accumulate in the ecosystem (Figure 3). Zinc, Cd, Cr, and Ni are being lost from the ecosystem at Gårdsjön, whereas Cu and Pb accumulate. Only insignificant amounts of Pb leave the rooting zone.

The exchangeable fractions of the soil stores of Zn and Ni are lost in minor quantities from the mineral soil. Evidently, there is a potential risk of a shortage to the plants of Zn which is more soluble and prone to leaching under acidic conditions. There is also a negative soil budget of the toxic cations Cd and Ni. However, the deposition load is high compared to the outflow from the soil and will probably balance the Cd output on a level where about half the current exchangeable soil store will remain. Therefore the problem with high Cd concentrations in acidic soil solutions will persist and even become more serious with increasing soil acidification.

There are indications that weathering does not keep pace with the documented losses of several macro-elements from the soil at Värsjö (Olsson and Melkerud, 1989). The long-term average annual loss rate by weathering was lower than the present metal fluxes monitored by the above-mentioned lysimeter studies (Bergkvist, 1987c). This is regarded as an indication of a recent increase in leaching caused by, e.g., acidic rain. It seems reasonable to suppose that the weathering rates of Zn, Cd, and Ni do not keep pace with the documented present leaching losses, and there is a potential risk that the exchangeable stores will decrease. The present-day extractable soil pool of Zn in deciduous and coniferous forests of S Sweden is only half the size 40 to 50 yr ago (Falkengren-Grerup *et al.*, 1988).

A net accumulation in the mineral soil is generally revealed by Cu and Pb. The first sink for these metals is the litter and mor layers. The organic topsoil is often regarded as an almost permanent sink for Pb (Benninger *et al.*, 1975; Bowen, 1975; Van Hook *et al.*, 1977; Siccama and Smith, 1978; Stahr *et al.*, 1980; Smith

and Siccama, 1981; Trüby and Zöttl, 1984; Turner *et al.*, 1985; Zöttl, 1985). In the acidic soils in SW Sweden (Bergkvist, 1987a, c) a close relationship was shown between vertical transport of organic matter and Pb. Therefore, the upper part of the B horizon seems to be the main sink for Pb in SW Swedish spruce podzols, though some Pb is transported even deeper.

In addition to the lysimeter studies at Gårdsjön, metal budgets have been calculated for entire microcatchments surrounding the Gårdsjön site (Grahn and Rosén, 1983). The general picture from the two different approaches is that the calculated budgets of certain elements are more negative in the lysimeter study – some elements also have different signs of the budget. Chromium and Ni are lost in greater amounts from the soil profile in the lysimeter study than from the entire catchment where Cr is almost at balance. Zinc and Cd give a negative budget in the lysimeter study but a positive budget in the catchment study. Both studies show positive budgets of Cu and Pb.

Differences between the two approaches may be attributable to several factors. The lysimeters include only the upper soil horizons and the flux is studied at an early stage of transport through the ground. Input to ground water is not considered in run-off studies, leading to an underestimation of ecosystem losses. Brook water has to a large extent passed through a freely drained soil profile, partly followed by an out-transport through soil layers rich in organic matter. The two budget approaches are thus not wholly comparable. Furthermore, soil acidification due to nitrification may increase the leaching in the lysimeters.

The concentrations of metals in different types of water from the Värnsjö and the Gårdsjön sites are listed in Table III. There was little change in the levels of Cu, Pb and Cr between the leachate from the 0 to 55 cm soil horizon (lysimeters) and the brook. In contrast, at Värnsjö the concentrations of Zn, Cd, and Ni in the brook were only 10% of the soil-water concentrations at the depth of 55 cm.

At the Gårdsjön site about 15 to 25% of the soil-solution concentration of Zn, Cd and Ni at 55 cm was found in the run-off water draining the microcatchments. In the lake water only Zn, Cd and Cr concentrations were significantly lowered compared to run-off. Lead concentrations were higher in the lake; the concentrations of Cu and Ni did not differ between the brook and the lake.

In the budget studies performed in Sweden and at Solling, the gross outflow of Cr and Cd from the rooting zone was at the same general level in the different studies. Copper, Zn and Pb were leached in larger amounts at Solling. Nickel was leached in much larger amounts from the Scandinavian soils. The great particulate deposition to the forest soils at Solling is combined with a considerable acidity of the precipitation. Spruce throughfall had a pH of 3.37 as an annual mean compared to 3.96 in that of beech whereas the wet deposition had a pH of 4.06 (Ulrich *et al.*, 1979). In S Sweden the corresponding pH values were 3.7, 4.7 and 4.1, respectively (Bergkvist *et al.*, 1987a). Great differences in the mineral soil properties may also be of importance for the differences in the leaching rates.

Element release below the rooting zone at the Swedish sites studied is usually

in the same range as those from Bärhalde. An exception is Zn showing a higher release rate from mineral soil in the Scandinavian studies.

The influence which different tree species exert on soil characteristics and nutrient cycling is often a subject of debate. Even though a considerable soil acidification has occurred in all forest types investigated in Europe, the dominating tree species is important for the metal cycling and acid-base properties of the soils. The acidifying potential of the spruce is definitely greater than that of deciduous trees, e.g. beech or birch (Nihlgård, 1971; Riha *et al.*, 1986). There are many reasons for this. Dry deposition is greater to a spruce stand, due to a larger aerosol trapping leaf area, particularly in winter (Höfken and Gravenhorst, 1982; Mayer and Ulrich, 1982). The spruce canopy increases the acidity of incident precipitation considerably, and metals and S are being enriched. The pH is usually considerably lowered when the precipitation passes the spruce canopy, at least in rural areas. Incident precipitation water passing through a beech canopy becomes less enriched in metals or even loses metals (Table III; Nihlgård, 1970). The internal production of acids is also greatest in a spruce stand (Matzner and Ulrich, 1981). When old farmland or beech-forest soil has been planted with spruce, podzolization and deterioration often follow rapidly (Nihlgård, 1971; Bråkenhielm, 1977; Brand *et al.*, 1986).

Further evidence of the great influence of the tree species on soil acidification and metal turnover is contributed by the above-mentioned study in S Sweden (Tables II and III; Bergkvist *et al.*, 1987a, b). Five sets of adjacent spruce, beech and birch stands on soils of the same origin were studied. Throughfall and soil solution were always most acidic in the spruce stand, and the concentrations and fluxes of metals were usually highest as well. The birch soil had the highest pH and lowest concentrations and fluxes of metals. Whereas the ecosystem budget of Zn was strongly negative in the spruce and beech stands, it was strongly positive in the birch stands (Figure 5). The ecosystem budget of Cd was clearly negative for spruce and beech but only weakly negative for birch. The three forest types were alike in having strongly positive ecosystem budgets of Cu and Pb.

The leachability of metals in the soil is considerably influenced not only by the vegetation type but also by the soil type. In the Swedish study the sites Skogslund and Rosenlund had an acidic brown forest soil in contrast to the other three podzol sites (Bergkvist *et al.*, 1987a, b). The three podzol soils clearly differed from the brown-forest soils in a much heavier displacement of Cu and Pb from the A horizon to the B horizon of the spruce and beech stands. This process went along with a heavy leaching of organic matter from the A horizon. The flux of Cd was usually greater from the B than from the A horizon at the podzol sites, whereas the opposite is true for the brown forest-soil sites.

Different soil types were also compared in another S Swedish study comprising spruce, beech and open grass-dominated regeneration areas (Bergkvist, 1987a). Metal concentrations in soil solutions and the leaching rate from a brown forest soil were generally higher than in a podzol. Soil solution concentrations of Zn, Cd, and Ni were higher in the brown forest soil, while metals with a leaching pattern

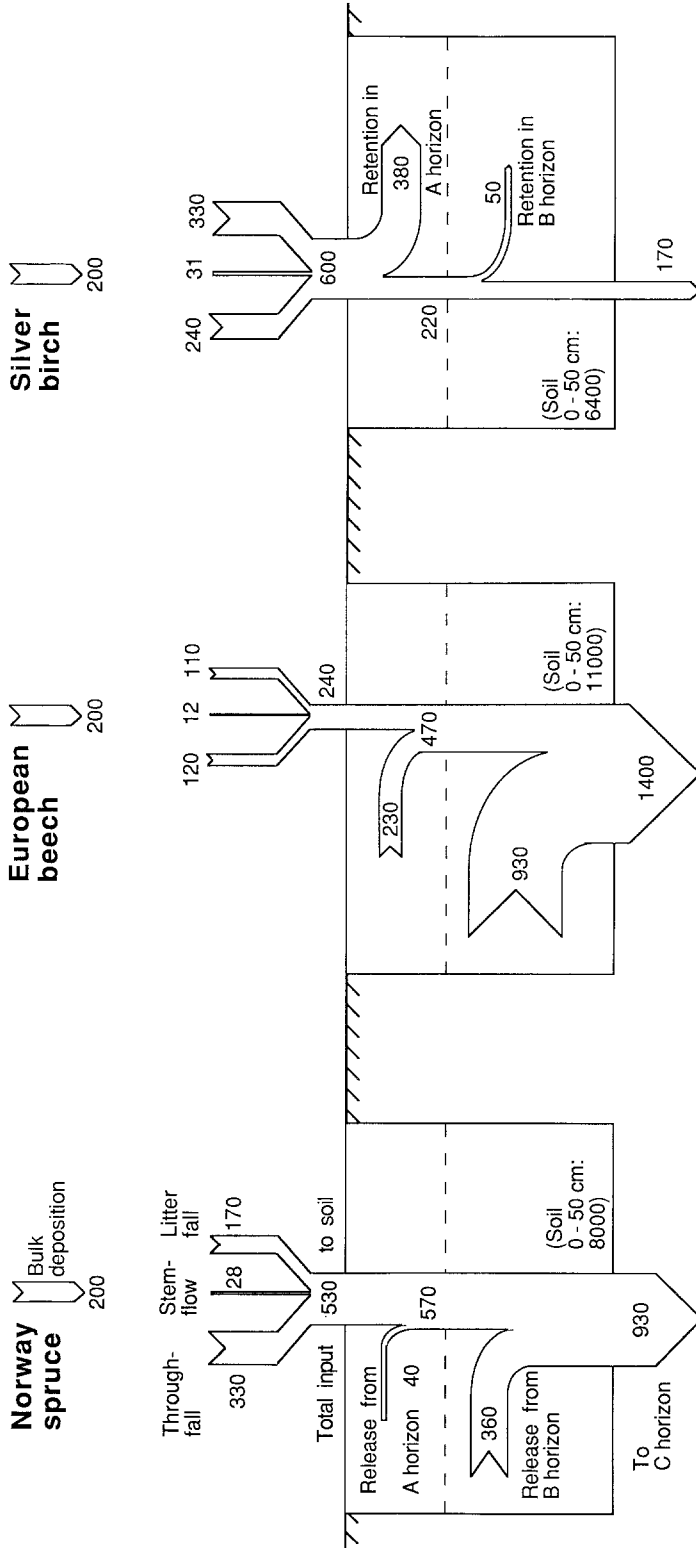


Fig. 5. Annual flow of Zn through *Picea abies*, *Fagus sylvatica* and *Betula pendula* forest ecosystems in S Sweden. Mean of five sites. June 1984 – May 1987. g ha⁻¹ yr⁻¹. Soil pools (EDTA-exchangeable): g ha⁻¹. Open zero-tension lysimeters. From Bergkvist *et al.* (1988).

associated with the transport of humus (Cu, Pb and Cr) were leached in larger amounts from the podzol, as was DOC. However, the most conspicuous metal pattern of the soil solutions was that in both soil types, the lowest concentrations of metals were found in the regeneration areas. In the regeneration areas, all metals showed a net accumulation or were at equilibrium in the mineral soil. In the forest stands, Zn and Cd were lost from the mineral soil, as was Ni in the spruce stand. Spruce seems to exert a more powerful influence than beech on the leachability of metals in soils, but also beech increases the release of some metals considerably. In all stands Cu, Pb, and Cr as well as organic matter accumulated in the mineral soil.

Results from a study of metal leaching rates from the upper 'C horizon' (110 cm soil depth) of the soil studied by Bergkvist (1987a) may be compared with the outflow recorded from the B horizon (55 cm depth; Bergkvist, 1987b). It was shown that the soil acidification penetrated far down into the morphological C horizon. The depth to which the pedological influence penetrated was governed by tree species and land management. In the C horizon, soil acidity and metal release were greater in afforested soils (spruce or beech) compared to open areas covered by grass swards.

In Denmark, Rasmussen (1986; 1988) has used the same lysimeter technique as in the Swedish studies. The precipitation is as acidic or somewhat more acidic at the Danish sites. When three planted spruce stands were compared, soil-solution concentrations of metals proved to be generally highest in the most acidic soil. Metal concentrations in soil solution from the lower B horizon in this acidic soil were also higher than in the Swedish soils; the concentration of Cd (c. 8 to 10 $\mu\text{g L}^{-1}$) was four times that reported from Vårsjö (c. 2 $\mu\text{g L}^{-1}$). Together with the Solling studies the Danish study differs from those at Bärhalde and Vårsjö in having a strongly negative ecosystem budget of Cd (Figure 4).

In a Canadian study of eleven watersheds, the amounts of Cu, Pb, and Cd retained within the watersheds were estimated to be 94, 97 and $> 75\%$, respectively, of the amounts deposited into the watersheds (Jeffries and Snyder, 1981; Schut *et al.*, 1986). Retention percentages of 72 to 100, 97 to 99 and 67 to 100 have been calculated for Cu, Pb and Cd, respectively, for Walker Branch Watershed, Tennessee (Schut *et al.*, 1986). Likewise, retention of Pb was 95 to 98% for a forested watershed at Hubbard Brook, New Hampshire (Siccama and Smith, 1978; Smith and Siccama, 1981). The doubling time of the Pb concentration in the humus was estimated at 50 yr (Siccama and Smith, 1978). Likewise, a strong retention of Pb in the soil was documented at Camels Hump by Friedland and Johnson (1985). The Pb deposition they reported was remarkably greater than in Europe (Figure 3).

Copper, Pb and Cd retention in the Bärhalde watershed in the Black Forest was estimated at 61, 95 and 69%, respectively (Stahr *et al.*, 1980, Schut *et al.*, 1986).

In the event of a total disintegration of the humus profile of a forest, there would be a great risk of considerable movement of Zn and Cd to the ground

water. In a future with a continuous accumulation of Cu, Zn, Pb and Cd in forest soils, the greatest threat to vegetation comes from Cd being a mobile element chemically resembling Zn. The largely immobile element Pb shows the least probability of accumulation into vegetation or leaching to the ground water (Keilen, 1978).

7. Interactions with Acidic Precipitation

The influence of acidification on the soil budgets of trace elements has not attracted as much concern as the macronutrients but some studies are reported. The leachability as affected by acidity of the leaching solutions differs greatly among metals. The mobility of Zn and Cd was shown to increase with soil acidity (Esser and el Bassam, 1981; Brümmer and Herms, 1983; Scokart *et al.*, 1983). From a field study where lysimeter soils were irrigated with acidified throughfall water (Bergkvist, 1986a) it is evident that Zn and Cd have higher concentrations in soil leachates throughout the soil profile and greater outflow from soil when more acid is added to the soil. This is in accordance with a laboratory study of the leaching of heavy metals from metal-polluted and unpolluted mor layers (Tyler, 1978). In an experimentally acidified mountain stream in the Hubbard Brook Experimental Forest, USA (mean pH 4.0), streamwater concentration of Cd increased with acidity (Hall and Likens, 1980).

The release of Cu and Pb does not seem to be enhanced at all by the acidity added to the soil. Bergkvist (1986a) reported that the release from the A horizon was in fact lowered by the addition of more acid to the soil. The release of these metals was closely related to the release of organic matter. The DOC content of soil leachates was significantly lowered in the A horizon when acid was added to the soil and so were the concentrations of Cu and Pb in the most acidic treatment.

Strong acidification seems to reduce the litter decomposition rate. It was demonstrated in an early study that the decomposition of humus and cellulose declined in direct proportion to the pH decrease caused by the addition of elemental S to a mineral soil (White *et al.*, 1934). Later studies have verified these findings. Tamm *et al.* (1977) obtained data suggesting that the addition of H₂SO₄ to soil reduced the C mineralization rate in humus. Irrigation with pH 2 water reduced birch litter decomposition, while pH 3 did not (Hågvar and Kjøndal, 1981). In an 'acid rain' experiment with a pine forest soil (Bååth *et al.*, 1979), a reduction in soil respiration was brought about by pH 2.0 water but not by pH 3.0 water.

The close relationships between Pb and humus release, and between Cd release and soil acidity were also shown by Tyler (1981) at ambient soil pH variations. In the above-mentioned laboratory experiment with mor layers, Tyler (1978) showed that Cu and Pb were also released when enough acid had been added to the soil.

Interestingly, Mn appears to be very important for the mobility of Cu and Zn. It is known that manganiferous nodules in soils contain varying amounts of other metals, e.g. Cu and Zn (Taylor and McKenzie, 1966; Sidhu *et al.*, 1976). Factors

affecting the solubility of these concretions also affect the mobility of the associated metals.

The solubility of many metals and the acidity of the soil solution are closely related, as demonstrated by Bergkvist (1987a) and Tyler *et al.* (1987). The relationship between the pH and the total concentration of Zn, Cd, and Ni in the soil solution is very close, though non-linear. There is usually a more or less distinct bend in the curves, indicating a rapidly increasing metal solubility below a critical soil solution pH. In all soils studied in the two reports, the critical acidity of the soil solution in the B horizon is within the pH range of 4.0 to 4.5. A drop in soil-solution pH by merely 0.2 units in this range results in a 3 to 5-fold increase of the Cd concentration.

There are indications that the root uptake of certain metals is increased by soil acidification. In rhizomes and leaves of *Anemone nemorosa* in S Swedish beech forests the concentrations of Zn and Cd were positively correlated with soil acidity (Tyler, 1976b). In the case of an increase in their availability, metals could be expected to accumulate in biologically active parts of the ecosystem, such as growing plant organs and the organic top soil (Tyler, 1972). There are also indications that acidification can disturb the function of tree roots (Puhe *et al.*, 1986). Subsequent changes in the metal uptake by roots may have significant nutritional and toxicological consequences to forest trees. These aspects are beyond the scope of this literature review, however.

8. Research Needs

During the last few decades a considerable amount of data on fluxes of heavy metals in forest ecosystems has accumulated. A more thorough understanding of metal cycling in the ecosystem is limited by the lack of knowledge in certain fields, however. This is especially true in the present situation where the current acidification greatly modifies the pattern of metal fluxes. Some fields where more research is needed are pointed out below.

The long-term influence of acidification on metal mobility is a widely neglected issue of great importance. This has special relevance not only in areas with a non-calcareous bedrock but also in certain areas where the soils have a great potential of metal leaching, e.g. clayey soils such as many soils of N and Central Europe. In these areas there are sufficient data on acidification trends and metal fluxes to support the development of models to predict long-term effects on metal mobility. Acidification influences not only the rate of leaching from soils but also the uptake by tree roots and the internal circulation of metals within the tree and the ecosystem.

Knowledge of these long-term effects is urgently needed not only from a scientific point of view but also since it has considerably practical implications in, e.g., forest nutrition. Even if data on weathering are scanty, there is clear evidence that the weathering rate does not keep pace with the loss of macronutrients from soils subject to acidification. There are indications that this applies also to heavy metals susceptible

to acidification, both the micronutrient Zn and toxic heavy metals like Cd and Ni. Since information on these elements cannot be extrapolated from data on base cations, special weathering research must be directed towards heavy metals.

The vertical extent of acidification in forest soils is widely unknown except for a small number of study sites. The liberation and downward movement of metals, especially heavy metals, have important practical implications in, e.g., fresh-water quality and community water supply.

Further, there is a great lack of knowledge as to the interactions between soil water, ground water and surface waters. Much research remains until a comprehensive model of the flux of water far down the soil profile in different types of soils can be established. This lack of knowledge hampers the calculation of accurate water and metal budgets for entire soil profiles. It also complicates the comparison of data from run-off studies with those gathered in studies using lysimeters.

Acidification experiments in the field have hitherto comprised plots of a rather limited area. Much information on the effects of acidification on metal fluxes and mobility would probably be gained from well-designed field experiments on a larger scale.

The role of anions as acidifying agents is another ignored field of great importance to the mobility of metals in acidified soils. This is of special concern in areas where aerially deposited sulphates have accumulated in soils.

As to metal speciation, more research is needed to elucidate to what extent metal-complexing properties of humic substances (HS) vary with the origin of the HS. Further, stability constants should be determined at metal/HS ratios normally present in forest-soil solutions. More attention should be paid to competitive binding between different metal ions for reactive sites on HS. Using analytical approaches, the interest must be focused on equilibrium techniques such as ion-selective electrodes, membrane separation techniques (e.g. dialysis and Donnan dialysis) and solvent extraction, i.e. methods capable of measuring ionic activities in the range of 10^{-9} to 10^{-8} M. Possible interferences must be critically studied.

The role of natural vs anthropogenic acidification in metal mobility is widely unknown. This question is connected with the influence of different land use, including the choice of tree species. European studies have usually included two or three tree species at the most, and American studies have often been performed in mixed stands.

Intercalibration of different lysimeter types is a technical question deserving more attention. The reliability of element flux calculations is dependent on the performance of the lysimeter equipment used and, as mentioned above, the choice of water-flux data to be used in the calculations.

There is also a demand for the development of more practicable methods for dry deposition measurements. This is important in the light of the great amount that dry deposition contributes to the total deposition. In this connection it can be mentioned that the methods of sampling of deposition and soil water are often inadequately described in the literature.

Further, many flux studies would be much more interpretable had environmental variables as well as site and soil characteristics been more precisely stated. Finally, more attention should be paid to the contamination risks in the collection and preparation of samples.

9. Concluding Remarks

Bearing in mind that every report reviewed here has its own assumptions and aims, that methods vary greatly and that budget calculations, where present, have been performed in different ways, it may not be considered justified to draw any general conclusions from the body of studies reviewed here. Furthermore, the descriptions of the methods used in many of the reports are not sufficient to make the basis of any straight-forward comparison between the budget figures arrived at in the different studies. Accordingly, any conclusions from a literature review like this would necessarily be subject to many caveats. There is, however, a body of information in support of the following implications pertaining to the mobility and budget of metals in forest soils subjected to continued and increased acidification:

(1) Release of Zn, Cd and Ni from the A horizon will increase with increasing soil acidity. These elements are released throughout the soil profile and, usually, the soil-solution concentration continues to increase at least through the B horizon.

(2) Copper and Pb are transported from the A horizon to the B horizon by soluble organic matter. Conditions favoring dissolution of organic compounds also favor this transport. The precipitation of Pb with organic matter in the upper B horizon is almost total.

(3) Forests are accumulating Cu and Pb. Chromium is accumulating or almost at balance at least in less acidic soils.

(4) Acidic forests lose significant quantities of Zn, Cd, and Ni.

(5) Increased soil acidification by acid rain may increase the concentrations of many metals in the soil solution, e.g. Zn and Cd.

(6) There are indications that weathering does not keep pace with the documented losses of Zn from forest soils, and there is a potential risk that exchangeable stores will decrease, with consequences for plant nutrition.

(7) Biological nutrient uptake and recycling may be impeded by acidic precipitation.

(8) With increased soil acidification the soil-solution concentration of, e.g., Cd is increased to such an extent that the risk of root damages cannot be disregarded.

(9) Both the soil type and the vegetation type seem to be of great importance for the soil acidification and the metal leaching rates.

(10) The soil acidity is greater and penetrates deeper into spruce-forest soils than into adjacent beech-forest soils or soils of other deciduous-tree stands of the same parent mineral origin.

(11) A birch soil or a soil in an open grass-dominated area shows much lower leaching rates of metals than soils of adjacent spruce or beech stands on soils with the same mineral origin.

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