VESIENTUTKIMUSLAITOKSEN JULKAISUJA
PUBLICATIONS OF THE WATER RESEARCH INSTITUTE 42

VESIHALLITUS — NATIONAL BOARD OF WATERS, FINLAND
Helsinki 1981

ISBN 951-46-6066-8
ISSN 0355-0982

Helsinki 1982. Valtion painatuskeskus

PARTICLE DEPOSITION IN URBAN AREAS

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MELANEN, M. & TÄHTELÄ, H. 1981. Particle deposition in urban areas. Publications of the Water Research Institute, National Board of Waters, Finland, No. 42.

In experiments carried out at seven urban test sites during 1977—1979, the average deposition rate compared to the one observed in regional background was roughly 100 % higher in respect to total organic carbon, 75 % higher in total phosphorus and the same in the cases of total nitrogen and chloride. In the case of calcium, the deposition rate was the same as the one in regional background in suburban catchments but 100 % higher in city centres. The corresponding comparative figures for sulphate were 50 % in suburban catchments and 100—200 % in city centres. In suburban catchments, the pH of precipitation was on the average 0.4 units lower than the pH of precipitation in regional background. The conductivity of precipitation was 25 % higher in suburban catchments and 100 % higher in city centres when compared to the average observed in regional background. On the order of one third to one half of the total variance of the deposition rates of total nitrogen, chloride, sulphate, vanadium, copper, lead and the variance of conductivity of precipitation could be explained by hydrometeorological factors, local emission levels and regional background deposition levels. In the 1970's, some increase has been observed in the deposition rate of sulphate. On the basis of deposition levels, the catchments in the study were classified to be satisfactory for residential living.

Index words: Particle deposition, emissions, urban hydrology.

1. INTRODUCTION

The field experiments of an extensive research programme, the Finnish Urban Storm Water Project, were carried out over the 1977—1979 period in seven urban test catchments.

The main objective of the research programme is to provide data on the quantity and quality of urban runoff as part of the hydrological cycle.

The field studies performed in the catchments included the recording of rainfall and runoff, sampling of the runoff, and sampling of the atmospheric particle deposition.

In the Urban Storm Water Project, one of the objectives is to study the variation in the urban runoff-water quality due to different catchment characteristics, hydrometeorological factors and airborne pollution.

Consequently, two objectives were put into the study dealing with atmospheric deposition: (1) to give the average deposition rate of various parameters in the test catchments, to be used in runoff quality modelling, and (2) to analyse and quantify the dependence of various deposition parameters on the hydrometeorological factors, local emissions and background pollution.

2. TEST CATCHMENTS

The main criterion in choice of the test catchments was that they should differ as much as possible in local activities, background pollution levels, and climatic and weather conditions.

When all the requirements derived from the various studies of the project and the budgetary constraints were taken into account, seven catchments were eligible for selection in the municipalities of Helsinki, Tampere, Oulu and Kajaani (Table 1, Fig. 1).

In Table 2 some characteristics of the test catchments are given which describe their degree of urbanization.

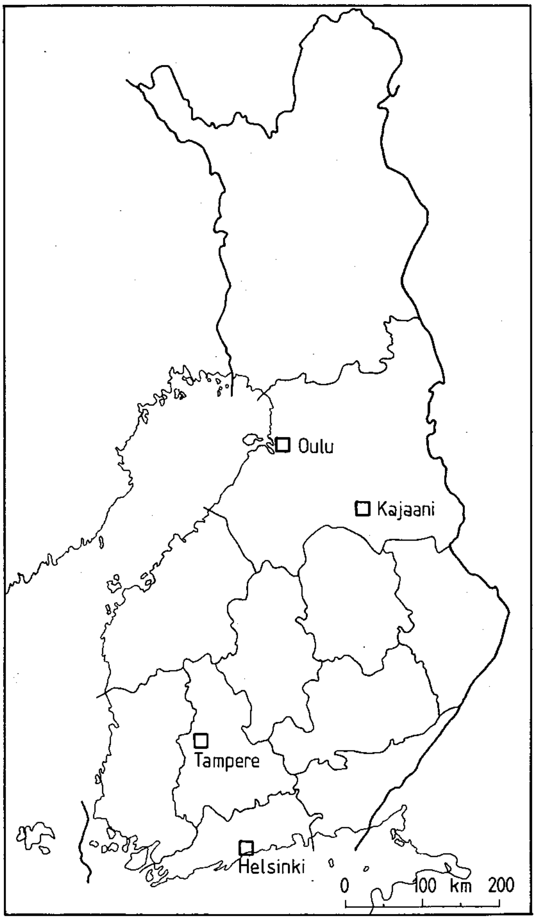

Fig. 1. Location of test municipalities.

Table 1. Location of test catchments.

Catchment	Municipality	Coordinates	
		latitude	longitude
Kontula	Helsinki	60°14'N	25°05'E
Pakila	Helsinki	60°15'N	24°57'E
Herttoniemi	Helsinki	60°12'N	25°02'E
Nekala	Tampere	61°29'N	23°48'E
Hämeenpuisto	Tampere	61°30'N	23°45'E
Kaukovainio	Oulu	65°00'N	25°32'E
Kajaani centre	Kajaani	64°13'N	27°44'E

2.1 Catchments in Helsinki

The three test catchments in the municipality of Helsinki are the high-rise residential area of Kontula, the low-rise residential area of Pakila, and the traffic area of Herttoniemi.

The Kontula catchment (Fig. 2) is a typical suburban residential area with houses of three to eight storeys, in which the activities are mainly related to dwelling. The percentage of paved surfaces in the catchment is 40 % and population density 160 persons per hectare. The topography of the catchment varies to some extent, and the area is bounded by an extensive green area in the west and south and by a similar residential area in the east and north. The area is connected to a district heating system. There is no industry in the catchment itself or in its immediate surroundings. A major point source, district heating plant, is situated roughly one kilometre to SSE of the catchment. Three traffic routes with a high traffic volume pass by the test catchment within a distance of one kilometre: E 3 and E 4 routes (26 000 motor vehicles a day) west and north, and Myllymestarintie (20 000 motor vehicles a day) south of the catchment.

The catchment of Pakila (Fig. 3) is an example of older single-family residential areas. The percentage of paved surfaces is 29 % and population density is 30 persons per hectare. Topographically, the area is slightly hilly, and it is bounded by the same type of low-rise developments in all directions. There is no industry in this test site either. The motor way of Tuusulantie (26 000 motor vehicles a day) passes by in close vicinity to the area on the eastern side. Within a distance of 100—300 metres to the south another traffic route, Muurimestarintie (19 000 motor vehicles a day) is located.

The Herttoniemi test catchment (Fig. 4) consists of a stretch of the motor way Itäväylä (45 000 motor vehicles a day), part of an underground railway, and a minor residential and

Table 2. Characteristics of test catchments.

Catchment	Altitude from sea level[a] m	Type	Catchment area ha	Population density persons ha^{-1}	Proportion of paved surfaces % of catchment area	Proportion of green area % of catchment area	Volume of growing stock solid m^3 ha^{-1}	Proportion of street area % of catchment area	Total length of street area km
Kontula	+35 to +50	suburban residential (high-rise houses)	22.9	160	40	53	12	10	1.5
Pakila	+23 to +41	suburban residential (low-rise houses)	20.2	30	29	67	24	15	3.1
Herttoniemi	+6 to +23	traffic (motor way, underground railway)	14.7	-[b]	35	45	12	23	3.0[c]
Nekala	+85 to +89	industrial (mixed industry)	14.2	-	40	32	11	14	1.6
Hämeenpuisto	+93 to +112	city centre-commercial	13.2	125	67	24	30	36	3.3
Kaukovainio	+13 to +19	suburban residential (high-rise houses)	40.5	85	30	61	52	10	4.2
Kajaani	+140 to +156	city centre-commercial	18.5	65	64	23	13	34	3.8

a: N_{60} elevation system; b: roughly 45 000 motor vehicles a day; c: length of motor way – carriage ways = 2.0 km (1.0 km to either direction)

industrial area in immediate surroundings of the route. The carriage ways of the motor way comprise roughly 22 % of the catchment area. On the northern side of the route the topography is in parts very steep. In the west and north the catchment is bounded by a green area and a high-rise residential area. To the south and east there is a significant industrial area with storage areas, metal and food product industries, and an oil delivery and storage harbour.

2.2 Catchments in Tampere

The two test catchments in the municipality of Tampere are the industrial area of Nekala and the city centre-commercial area of Hämeenpuisto.

The catchment of Nekala (Fig. 5) represents an industrial area with mixed small and medium-size industry. Ware houses and storage yards, and factories of food, metal and paper product industries are typical examples of the area. The percentage of paved surfaces in the catchment is 40 %. Topographically the area is flat. In the north the catchment is bounded by a low-rise residential area and in the east by an older high-rise area. In the other directions the catchment is surrounded by an industrial area;in the near surroundings there are factories for metal and food products, among other things. At a distance of roughly one kilometre in the west, the traffic route of Lempääläntie (25 000 motor vehicles a day) passes by and somewhat further there is a railway.

The Hämeenpuisto catchment (Fig. 6) is the most urbanized one among the test sites of the study; the percentage of paved surfaces in the catchment is 67 % and population density 125 persons per hectare. The terrain in the catchment descends towards the south. There is a substantial traffic volume in the test site because the main traffic routes of the city centre of Tampere go through the site. In the north and south the catchment is bounded by small green areas, elsewhere the area type is similar. The area is almost entirely connected to district heating. There is no industry in the test catchment itself. Yet, in the near surroundings there are, among other things, establishments of textile, metal, food and paper products. Significant point sources exist further away (1—5 km) from the catchment.

2.3 Catchment in Oulu

The test catchment of Kaukovainio (Fig. 7) in Oulu is a high-rise area with a low dwelling density. The percentage of paved surfaces in the catchment is 30 % and population density 85 persons per hectare. The area is flat and is bounded by a major green area in the south, and by low- and high-rise areas in the other directions. Practically all buildings in the cachment are connected to district heating. There is no industry in the catchment or in its immediate surroundings. Significant point sources exist, however, further from the catchment. The traffic route Nelostie (5 700 motor vehicles a day) passes the catchment in the west at a distance of roughly half a kilometre and the traffic route Kainuuntie (4 600 motor vehicles a day) in the southeast is equally distant.

2.4 Catchment in Kajaani

The test catchment of Kajaani (Fig. 8) is situated in the city centre. The percentage of paved surfaces in the catchment is 64 % and population density 65 persons per hectare. The traffic volume in the test site is high because the main traffic routes going via Kajaani city pass through it. The terrain descends towards northeast. In practically all directions the catchment is bounded by a similar area type of high-rise buildings and commercial blocks. There is no actual industry within the test catchment itself but a heat and power station is nearby. Some significant point sources exist further from the test catchment and the most prominent one is a cellulose factory utilizing calcium.

3. DEPOSITION PARAMETERS STUDIED

3.1 Particle deposition

A classification of air impurities, useful with regard to sampling and analysis, is as follows:

1) Gaseous substances
2) Vapor phase substances
3) Suspended particulate matter, which is generally divided into two subcategories:
   - nucleation, condensation and agglomeration mode, which is mostly a result of condensing vapors and atmospheric chemistry (S, N, C, Pb), range 0.001—1μm
   - dust mode, a result of wind erosion, fly ash emissions etc. (mostly typical soil minerals: Si, Fe, Al, Ca etc.), range 5—80μm

Particle deposition (particle fall, dustfall) refers to air contamination, consisting of the mass rate at which particles deposit from the atmosphere (Laamanen 1969). Deposition sampling includes techniques, which utilize open containers and man-made or natural surfaces to collect impurities deposited by natural forces.

In the samples gathered by various types of deposition gauges, the majority of particulate mass is in the range of 20—40 μm (Goettle 1978, Laamanen 1969, Laamanen and Jantunen 1979, Vornamo 1975).

The particle deposition caused by settling due to gravity during dry periods, is defined as dry deposition. The deposition of particles due to rainout and washout by precipitation is defined as wet deposition.

The deposition parameters analysed in this study are: total deposition, volatile deposition, total organic carbon, total phosphorus, total nitrogen, chloride, calcium, sulphate, vanadium, cadmium, titanium, zinc, copper, lead, and pH value and conductivity of the deposition samples (precipitation).

3.2 Total deposition

The total deposition equals the sum of the total insoluble and water soluble particles deposited. It contains inorganic, organic and biological particulate matter originating from both natural and anthropogenic (man-made) sources.

The particulate matter is defined as either primary or secondary. The primary particles are in the particulate form already as emitted, for example dust raised by wind and fly ash released from a stack. Secondary particles are being formed in the atmosphere as a consequence of chemical reactions of gaseous substances and by physical processes, e.g. condensation. In this manner the same amount of particles may be generated as are emitted from the primary sources (Seinfeld 1975).

On a global scale particulate emissions from natural sources have a dominant role covering roughly 90 % of the total emissions (Seinfeld 1975). On the global scale the major primary natural sources of particulate matter are wind-risen dust, sea spray, volcanoes, and forest and bush fires. Secondary natural sources are related to the carbon, sulphur and nitrogen cycles, and to the conversion of gases into particles. The particulate matter from the man-made sources consists mainly of solid particles formed in the combustion processes, and products of catalytic and photo-

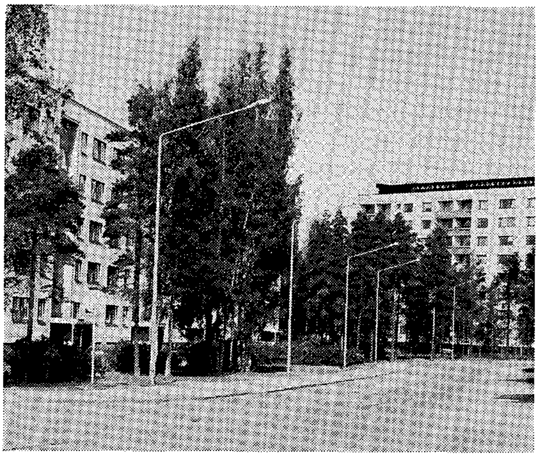

Fig. 2. View from test catchment Kontula in Helsinki.

Fig. 5. View to test catchment Nekala in Tampere.

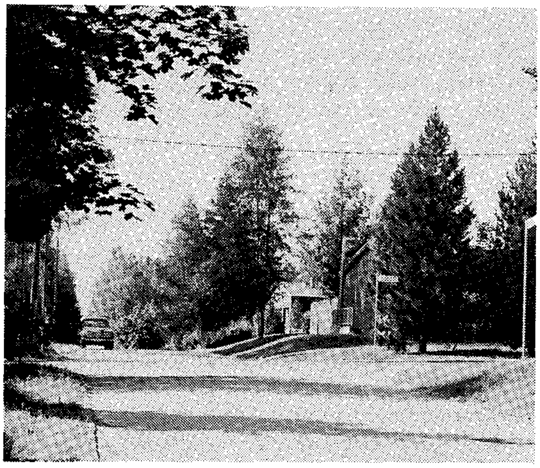

Fig. 3. View from test catchment Pakila in Helsinki.

Fig. 6. View to test catchment Hämeenpuisto in Tampere.

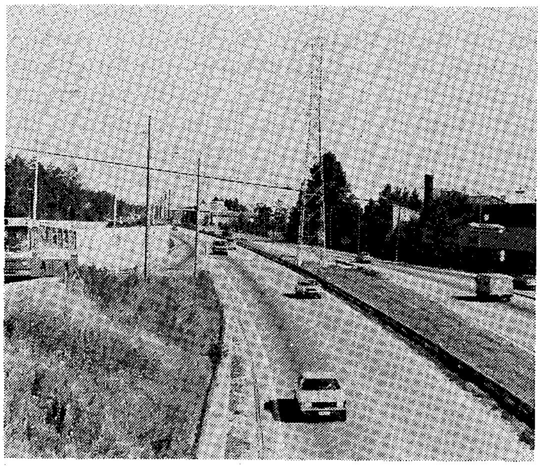

Fig. 4. View from test catchment Herttoniemi in Helsinki.

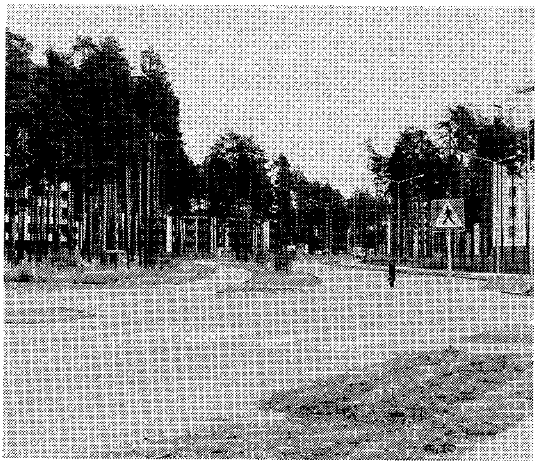

Fig. 7. View from test catchment Kaukovainio in Oulu.

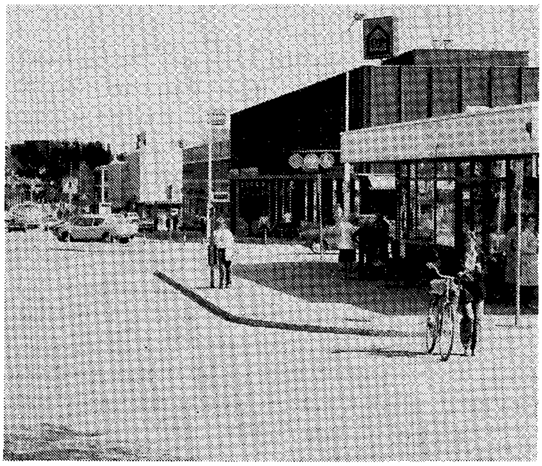

Fig. 8. View from test catchment Kajaani centre in Kajaani.

chemical reactions involving oxides of sulphur, unburned or partially burned hydrocarbons and nitrogen oxides (Seinfeld 1975). In a national scale emission inventory of 1970 in Finland, 40 % of the man-made particulate emissions were estimated to originate from heating, 30 % from industry, 10 % from traffic and 20 % from other sources (Laamanen and Jantunen 1979).

The proportion of water insoluble matter in the deposition generally grows with increasing industrial activity. In rural area samples the proportion of water soluble organic material is usually higher. During each season of the year the total deposition varies in both quantity and composition. One of the quantitative peaks occurs in the spring when the wind can raise loose dust prior to the sprouting of vegetation. In the summertime, the amount of various natural-source particulates, such as pollen grains and seeds, is at its maximum. In the autumn when the vegetation withers, the storms can raise mineral dust and organic material into the atmosphere. In the winter the snow prevents recirculation of the particulates deposited on the ground, but the emissions of particulates from heating, energy production and other man-made activities are at their maximum.

In the lower atmosphere the average residence time of particulate matter ranges from a couple of days to roughly a week (Seinfeld 1975). In the section closest to ground level the particles are removed by settling due to gravity or as a consequence of impaction against obstacles. At an altitude above about 100 metres, the principal removal mechanism of particles is rainout and washout.

3.3 Volatile deposition

The volatile deposition represents approximately the amount of organic matter in the deposition. Part of the volatile deposition originates from natural sources, i.e. directly from soil, vegetation and animal sources. In the summer this fraction contains for example pollen grains, seeds and other particulates originating from plants. In the winter the volatile deposition is mainly soot, tar compounds and other anthropogenic compounds originating from heating and traffic.

3.4 Total organic carbon

The amount of organic carbon in the deposition reflects the content of organic matter. The ratio of total organic matter to carbon is highest in complex compounds (for example proteins and fats) which contain a lot of oxygen, nitrogen and other "ingredients", and is smallest in pure hydrocarbons. The pollen grains and seeds for example contain plenty of proteins and fats.

Hydrocarbons appear in the air in gaseous, liquid and particulate form. A vast variety of hydrocarbons are emitted into the atmosphere from both natural and anthropogenic sources. Methane and terpenes (from vegetation) from the natural sources form a vast majority of total emissions on the global scale (Seinfeld 1975). Yet, the hydrocarbons from man-made sources are on the average more active and are consequently of crucial importance in air pollution chemistry.

The combustion of gasoline is the single most important contribution to anthropogenic hydrocarbon emissions, being approximately 40 % of the total man-made emissions on the global scale (Seinfeld 1975). Roughly 30 % originate from refuse incineration. The rest of hydrocarbon emissions are mainly due to refinement and evaporation of petroleum and the use of solvent. In the national inventory of 1970 in Finland, 65 % of the man-made emissions of hydrocarbons were estimated to be due to traffic activities (Laamanen and Jantunen 1979).

Accurate lifetimes of hydrocarbons in the air cannot be presented. In the urban areas, conversion of hydrocarbons to other organic compounds takes place rapidly in the presence of oxides of nitrogen. In general, the residence times of higher-molecular-weight hydrocarbons have been estimated to range from days to months (Seinfeld 1975).

3.5 Total phosphorus

Phosphorus is emitted into the air with mineral dust, for example as phosphates from soils with high apatite content. Phosphorus is also emitted into the atmosphere with organogenic substances, mostly as organically fixed and as calcium phosphate. In the summer the organic particulates originating from vegetation, increase the amount of phosphorus compounds in the air. As to the anthropogenic sources, phosphorus is emitted from some industrial processes, and from manufacturing and spreading of fertilizers, for example.

3.6 Total nitrogen

The important nitrogen containing compounds in the atmosphere are: nitrogen oxides, nitrates and ammonium salts, nitric acid and ammonia.

Of the oxides, nitric oxide (NO) and nitrogen dioxide (NO_2) are considered air pollutants. They are emitted by both natural and anthropogenic sources. The principal man-made source of nitric oxide is the burning of fuels at high temperatures, for example in heat and power stations, refuse incineration plants, motor vehicles and some chemical plants. On the global scale a vast majority of nitrogen oxides are emitted from natural sources as N_2O, roughly 90 % (Seinfeld 1975). Yet, the oxides originating from man-made activities are more significant as to air pollution phenomena. On the global scale roughly 50 % of the anthropogenic nitrogen oxides come from coal combustion and 40 % from petroleum processing and combustion (Seinfeld 1975). According to the national inventory of 1970 in Finland, of the man-made nitrogen oxides roughly 50 % are emitted by heating, 40 % by traffic and 10 % by industry and other sources (Laamanen and Jantunen 1979).

Atmospheric ammonia is primarily due to biological decay on the earth's surface. Some ammonia is emitted by the chemical industry. Nitrates and ammonium salts are not emitted in significant quantities but are formed from conversion of nitric oxide, nitrogen dioxide and ammonia (Seinfeld 1975).

The oxides of nitrogen are usually removed from the atmosphere by particle formation. Ultimately most of the oxides are converted into nitrates which in turn are removed by rainout, washout and dry deposition. Most of the ammonia is converted into ammonium compounds and roughly one fourth is oxidized to nitrates. The average lifetimes of nitrogen compounds range from two days to two weeks (Seinfeld 1975).

3.7 Chloride

Highly reactive gaseous chlorine appears only locally in the air. The most common chlorine compounds are hydrochloric acid and its various salts, chlorides. Chlorine is a commonly used industrial raw material and several compounds which contain chlorine, such as chlorinated hydrocarbons and phenols, are almost indecomposable in the environment.

Chlorides are emitted into the atmosphere by heating and industrial processes. The emissions from traffic are not significant. The salted sand used for de-icing on the traffic routes contains chlorides, which are consequently emitted to the air with dust to some extent. On the sea coasts chlorides are emitted into the atmosphere with sea spray. Chlorine compounds occur also in the surroundings of pulp and paper factories, refuse incineration plants and chlorine alkali factories, for example.

3.8 Calcium

Dust raised by vehicles from the traffic routes, fly ash released with flue gases and dust originating from the decay and corrosion of the plaster in buildings emit calcium into the air. The industry emits calcium into the atmosphere through fly ash from heating and through waste compounds of various industrial processes, such as a cellulose factory utilizing calcium.

3.9 Sulphate

The most important sulphur containing compounds in the atmosphere are: sulphur dioxide (SO_2), sulphur trioxide (SO_3) and hydrogen sulfide (H_2S) in gaseous form, liquid sulphuric acid (H_2SO_4) and sulphate (SO_4^{-2}) salts in particulate form.

On the global scale approximately 25—40 % of the total emissions of sulphur were estimated to originate from anthropogenic sources in the late 1960's (Seinfeld 1975). The main sources are combustion of fossil fuels, decomposition and combustion of organic matter, sea spray over oceans and volcanoes. Since the 1960's, the proportion of the anthropogenic sulphur emissions has been continually growing.

In an urban environment the sulphur compounds are emitted into the atmosphere in flue gases. Some amount of sulphur dioxide is absorbed into the particulates, the rest is generally spread over extensive areas. Other sulphur compounds are also fixed on the particles (sulfides, sulphites) due to absorption and adsorption and these are ultimately oxidized to sulphates.

The most abundant source of sulphur dioxide in urban areas is the burning of coal, with substantial contributions from petroleum combustion and smelting (copper, lead, zinc) (Seinfeld 1975). Emissions of sulphur dioxide from traffic are minor. In the year 1970 in Finland, 48 % of the anthropogenic emissions of sulphur oxides were estimated to originate from heating, 45 % from industry, 2 % from traffic and 5 % from other sources (Laamanen and Jantunen 1979).

Ultimately, sulphur dioxide is converted to sulphate salts, the lifetime of the sulphur dioxide before conversion being of the order of days (Seinfeld 1975). In the first stage, the sulphur dioxide is oxidized to sulphur trioxide either catalytically or photochemically. The presence of a number of foreign substances, such as metal salts and ammonia, promotes reactions of sulphur dioxide and oxygen dissolved in water drops in the catalytic process, leading to the formation of sulphate. Normally, several reaction products occur in the air simultaneously.

Hydrogen sulfide is formed in biological decomposition processes in the soil and sea and in some industrial processes (the manufacture of sulphate cellulose, for example). In the atmosphere, hydrogen sulfide is rapidly oxidized to sulphur dioxide, the lifetime of hydrogen sulfide being of the order of hours (Seinfeld 1975). Part of the sulphate salts in the atmosphere originate from seas, the rest is the oxidation products of sulphur dioxide and hydrogen sulfide.

The removal of sulphur from the atmosphere occurs by several processes (Seinfeld 1975): (1) rainout and washout, (2) diffusion into soil and vegetation, and (3) dry deposition of sulphate particles.

3.10 Vanadium

The main source of vanadium in the air of urban areas is heating by fuel oil which produces approximately half of the vanadium emissions in the urban environment (Statens Naturvårdsverk 1976). The fuel oils, mainly heavy fuel oil, contain varying amounts of vanadium compounds which are emitted as oxides. The second important source of vanadium emissions is the combustion of anthracite coal and coke. Some amounts of vanadium are emitted by industrial processes, for example in the manufacture of ferrous vanadium.

3.11 Cadmium

Cadmium is used in industry for example as a colour pigment, as a stabilizer for plastics and in various alloys. The cadmium used is emitted into the environment to a great extent. The main sources of emissions are cadmium mining and processing, and refuse incineration. At the end of the 1960's, mining and processing, and refuse incineration, each produced emissions equalling one sixth of the total cadmium consumption in the United States (Statens Naturvårdsverk 1976). In the combustion of fossil fuels, cadmium is also emitted into the atmosphere.

3.12 Titanium

Titanium is mainly used as a colour pigment (TiO_2) in paints and as a metal in the aircraft and space industry. It is also used in the manufacture of paper and in the rubber industry. Under Finnish conditions the majority of atmospheric titanium originates from the combustion of fossil fuels, and from the incineration. Some titanium may be emitted in the surroundings of traffic routes as a result of decaying paints used for road markings (Statens Naturvårdsverk 1976).

3.13 Zinc

Zinc is used principally to deter corrosion, in alloys and as a colour pigment (ZnO). In Sweden (Statens Naturvårdsverk 1976) roughly 70 % of total zinc emitted into the air originates from industrial processes; of the total emissions the manufacture of zinc and copper emit approximately 25 and 20 % respectively, and iron and steel industry about 25 %; roughly 16 % of the emissions are due to combustion of anthracite coal and coke, and 8 % is from incineration. In the United States an estimated 5 % of the zinc emissions into the air were a result of wearing out of automobile tires in the late 1960's (Statens Naturvårdsverk 1976).

3.14 Copper

Copper compounds are used in industry for electrical components, building construction and various industrial equipment. In Sweden (Statens Naturvårdsverk 1976) roughly 75 % of copper emitted into the air is estimated to be due to the mining and manufacture of copper. Refuse incineration (actually the burning of cables) is responsible for approximately 16 % of the copper emissions. Emissions due to the combustion of anthracite coal and coke total roughly 7 % of the emissions. Small amounts of copper are emitted by iron and steel industries and by the combustion of fuel oils.

3.15 Lead

Lead is mainly used in batteries, as a gasoline additive, in various pipes and sheets and in cables.

In 1973 in Sweden, approximately 55 % of the emissions of lead were estimated to be due to gasoline-powered traffic, 40 % to industry (concentration of ore, battery industry), roughly 5 % to refuse incineration and less than one per cent to combustion of fossil fuels (Statens Naturvårdsverk 1976).

In the United States roughly 99 % of the emissions of lead into the air were estimated to originate from the gasoline-powered traffic in the late 1960's (Statens Naturvårdsverk 1976).

3.16 pH value

The pH value of the deposition samples gives a view of the content of acid or alkaline impurities deposited as wet and dry deposition.

3.17 Conductivity

The conductivity of the deposition samples is proportional to the amount of dissolved ions in water thus giving a view of the occurrence of water soluble salts in the deposition.

4. METHODOLOGY AND DATA

4.1 Emission inventory

To provide basic data for the analysis of the observed depositions, emission inventories were performed (Hokkanen et al. 1979). In each test site, emissions originating from traffic, heating, refuse incineration and industrial processes were estimated both in the catchments and in their near surroundings. Moreover, the major point sources located further away from the test sites were analysed.

The emission parameters chosen to indicate the overall emission levels were: particulates, hydrocarbons, oxides of nitrogen, oxides of sulphur, vanadium, cadmium, titanium and lead. The principal urban sources of these compounds are listed in Table 3.

4.11 Principles of inventory

For estimation of the emissions from motor-vehicle traffic, the yearly mileages were computed, from which the emission rates were derived by emission factors given in Table 4. The calculations were based on traffic statistics available in the municipalities, supplemented by estimated traffic densities derived from the street classifications and the area's population. In conversion to the yearly mileages, seasonal, daily and hourly variation coefficients derived in the Helsinki region (Helsingin kaupunkiseudun liikennesuunnittelun koordinointitoimisto 1974) were applied in all test sites. The emission factors used are based on studies performed in Sweden in city traffic (Slutbetän-

Table 3. Sources of emission parameters included in emission inventory performed in catchments. Order of importance of sources depends on local circumstances.

Parameter	Sources
Particulates	heating and energy production industry refuse incineration and solid waste management traffic
Hydrocarbons	traffic heating and energy production refuse incineration
Oxides of nitrogen	heating and energy production traffic industry refuse incineration
Oxides of sulphur	heating and energy production industry refuse incineration traffic (diesel oil-powered)
Vanadium	heating and energy production industry
Cadmium	industry refuse incineration heating and energy production
Titanium	heating and energy production refuse incineration industry
Lead	traffic (gasoline-powered) industry refuse incineration

Table 4. Emission parameters and factors used in emission inventory (Hokkanen et al. 1979).

Parameter	Emission factors									
	gasoline-powered traffic $g\ km^{-1}$	diesel oil-powered traffic $g\ km^{-1}$	heavy fuel oil $kg\ m^{-3}$	light fuel oil $kg\ m^{-3}$	wood $kg\ m^{-3}$	ground gas $10^{-6} kg\ m^{-3}$	anthracite coal $kg\ t^{-1}$	peat $kg\ t^{-1}$	refuse incineration in oven $kg\ t^{-1}$	refuse incineration, open burning $kg\ t^{-1}$
Particulates	0.078	0.8	1.0	1.2	15	300	5	2	18	8
Hydrocarbons	2.3	3.9	0.25	0.35	1	50	1.25	-	50	15
Oxides of nitrogen	4.2	2.3	12.6	1.5	3	2 000	1.5	10	0.5	3
Oxides of sulphur	0.6	0.8	50.1	12.1	-	9.6	27.6	8	0.25	0.5
Vanadium	-	-	0.05	0.05	-	-	0.01	-	-	-
Cadmium	-	-	$2 \cdot 10^{-6}$	$2 \cdot 10^{-6}$	-	-	0.01	-	0.01	0.01
Titanium	-	-	-	-	-	-	0.1	-	0.1	0.1
Lead	0.035	-	0.001	0.001	-	-	0.01	-	0.01	0.01

Table 5. Average rates of emission parameters in test catchments (Hokkanen et al. 1979).

Catchment	Emission ($kg\ ha^{-1}\ a^{-1}$) of parameter							
	particulates $E_{part,c}$	hydrocarbons $E_{H_xC_y,c}$	oxides of nitrogen $E_{NO_x,c}$	oxides of sulphur $E_{SO_x,c}$	vanadium $E_{V,c}$	cadmium $E_{Cd,c}$	titanium $E_{Ti,c}$	lead $E_{Pb,c}$
Kontula	5.8	86	160	24	0.00	0.00000	0.000	0.86
Pakila	220	92	190	300	1.0	0.016	0.160	0.58
Herttoniemi	180	2 500	3 800	620	0.00	0.00000	0.000	18
Nekala	170	450	610	1 100	1.5	0.0087	0.087	1.2
Hämeenpuisto	120	1 600	2 500	400	0.00	0.00000	0.000	11
Kaukovainio	53	220	170	70	0.18	0.025	0.25	0.84
Kajaani	200	670	1 200	1 000	3.5	0.0055	0.005	5.0

kande av kommunikationsdepartementets ledningsgrupp rörande utvecklingsarbete på bilavgasområdet 1971).

The basic data on emissions from heating, refuse incineration and industrial processes were acquired by questionnaire techniques. The percentage of the return of questionnaires was 65 %; the percentages were 55 % in Tampere, 68 % in Helsinki, 71 % in Kajaani and 75 % in Oulu. When basic data were missing the emissions were directly estimated based on building volumes and type of activity. In the computation of emissions the newest emission factors available were used (Table 4) which are mainly based on the American circumstances (U.S. Environmental Protection Agency 1973). These factors were supplemented by more recent American (Campbell 1976) and Swedish (Statens Naturvårdsverk 1976) results. In computations the sulphur content (per cent by weight) was taken as 1.5 % for anthracite coal, 2.6 % for heavy fuel oil and 0.7 % for light fuel oil (Aspo Oy 1977).

There is no reliable quantitative method for estimation of the effects of the distant point sources. In the study the significant point sources at a distance of roughly 1—5 km from the test sites were analysed. The effects of the distant sources were computed as sum indices, "sum effects", which give a basis for mutual comparison of the test sites. The index of an emission parameter was computed as follows. The annual emission from a point source was multiplied by the size of the sector in which the source is seen from a test site. The amount derived was weighted by the permanence of wind from the direction of the point source. These "sum effects", summed over the analysed point sources, describe the yearly "matter flows" towards the test sites.

4.12 Emission data

The computed emission rates in the test catchments and in their surroundings, in local background, are given in Tables 5 and 6. The rough "sum effects" of the distant point sources are given in Table 7. The emission rates are graphically illustrated in Fig. 9. The local background emissions cover an area with a radius of one kilometre. Due to inaccuracies in both the basic data and emission factors, the emission rates are mainly given by two significant numbers.

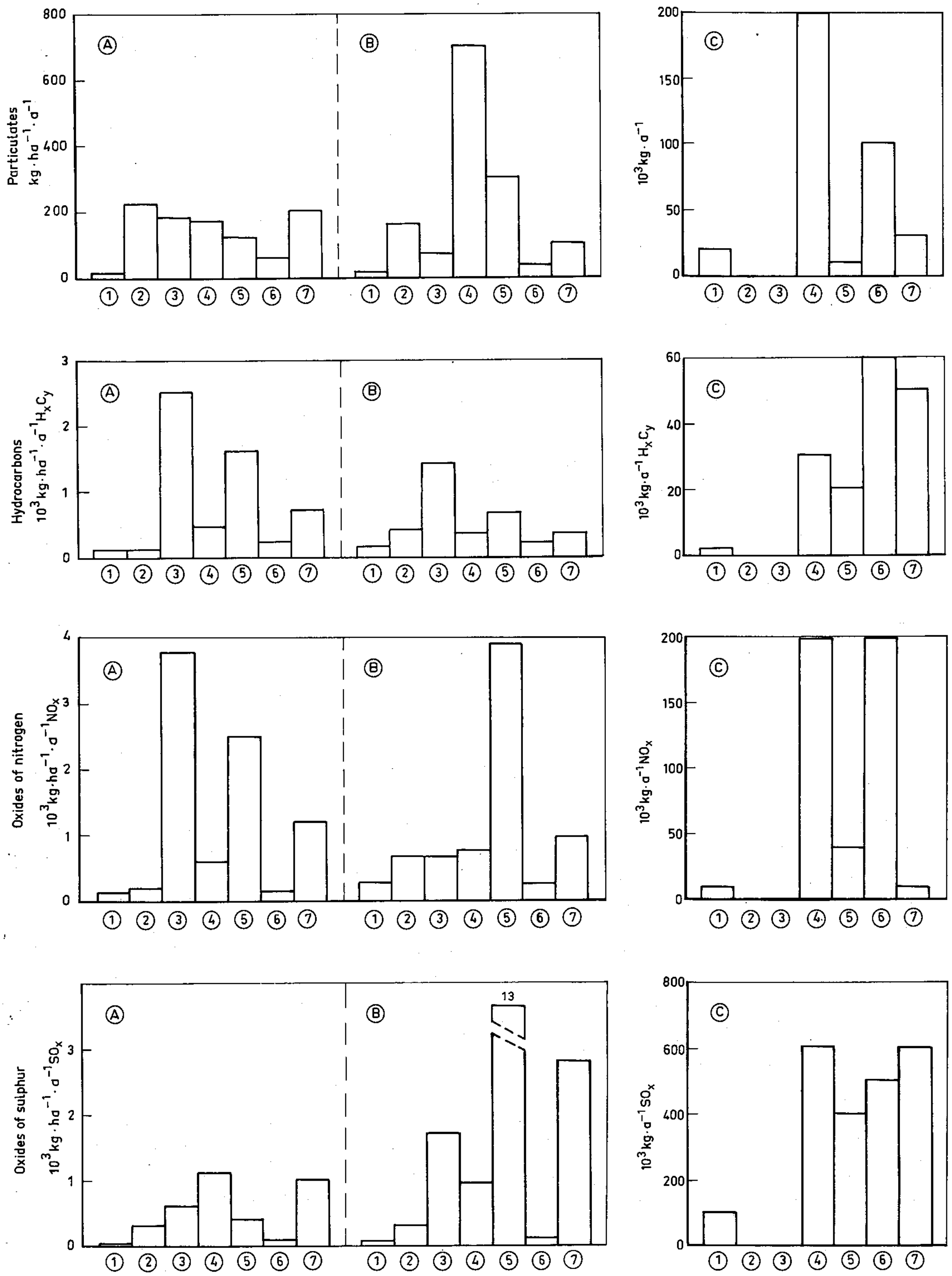

Fig. 9. Estimated emissions. A = emission rates in test catchments, B = emission rates in local background, C = "sum effects" of distant point sources. 1 = Kontula, 2 = Pakila, 3 = Herttoniemi, 4 = Nekala, 5 = Hämeenpuisto, 6 = Kaukovainio, 7 = Kajaani centre.

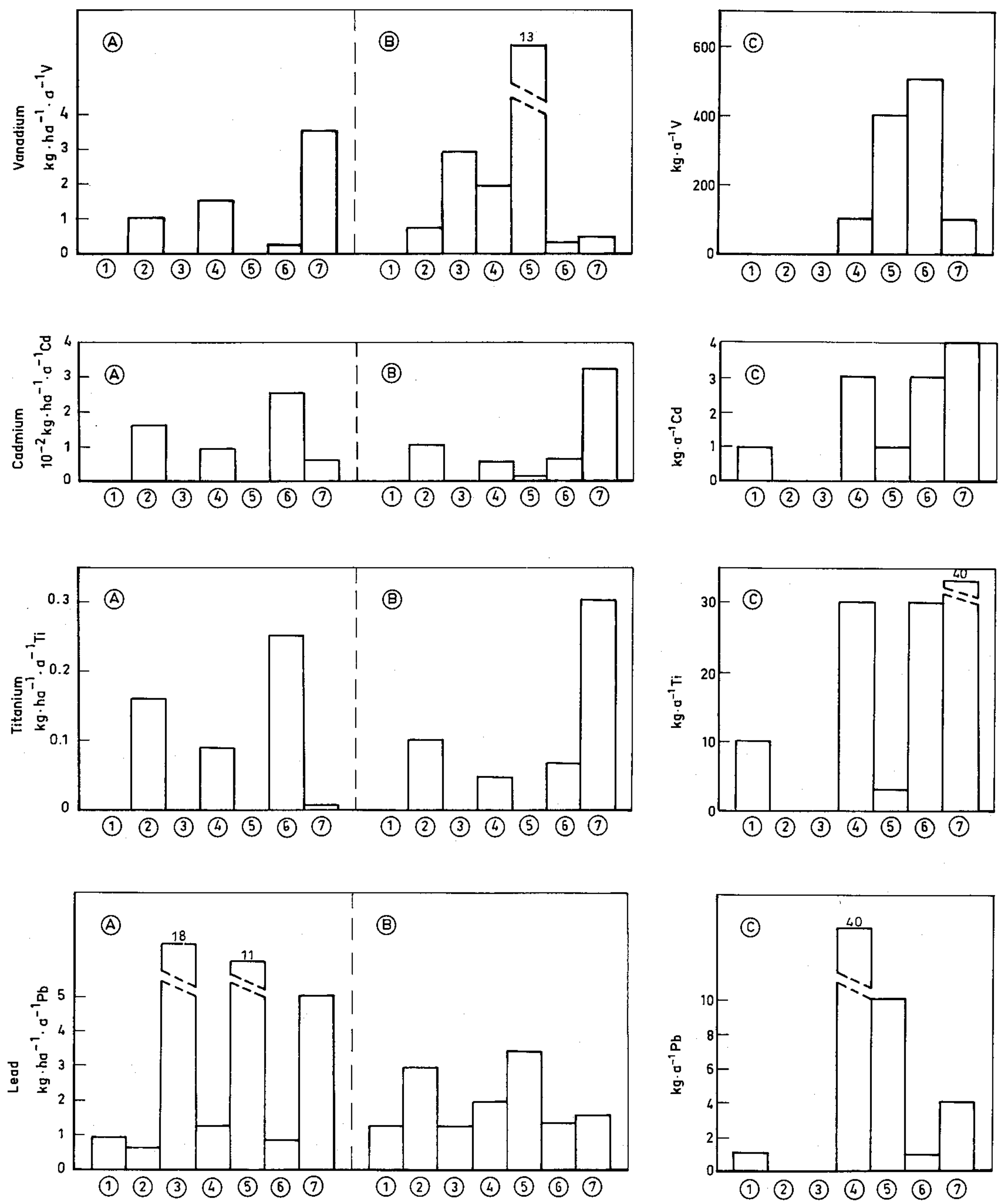

Fig. 9. Continued.

Table 6. Average rates of emission parameters in local backgrounds (Hokkanen et al. 1979).

Catchment	Emission (kg ha^{-1} a^{-1}) of parameter							
	particulates $E_{part,lb}$	hydrocarbons $E_{H_xC_y,lb}$	oxides of nitrogen $E_{NO_x,lb}$	oxides of sulphur $E_{SO_x,lb}$	vanadium $E_{V,lb}$	cadmium $E_{Cd,lb}$	titanium $E_{Ti,lb}$	lead $E_{Pb,lb}$
Kontula	9.4	150	250	39	0.00	0.00000	0.000	1.2
Pakila	160	400	660	280	0.66	0.010	0.100	2.9
Herttoniemi	70	1 400	670	1 700	2.9	0.00011	0.000	1.2
Nekala	700	350	730	940	1.9	0.0046	0.045	1.9
Hämeenpuisto	300	630	3 900	13 000	13	0.00051	0.000	3.4
Kaukovainio	26	180	270	120	0.32	0.0064	0.064	1.3
Kajaani	100	360	960	2 800	0.40	0.032	0.32	1.5

Table 7. "Sum effects" of distant point sources (Hokkanen et al. 1979).

Catchment	Sum effect (kg a^{-1}) of parameter							
	particulates $E_{part,ds}$	hydrocarbons $E_{H_xC_y,ds}$	oxides of nitrogen $E_{NO_x,ds}$	oxides of sulphur $E_{SO_x,ds}$	vanadium $E_{V,ds}$	cadmium $E_{Cd,ds}$	titanium $E_{Ti,ds}$	lead $E_{Pb,ds}$
Kontula	20 000	2 000	10 000	100 000	1	1	10	1
Pakila	-	-	-	-	-	-	-	-
Herttoniemi	-	-	-	-	-	-	-	-
Nekala	200 000	30 000	200 000	600 000	100	3	30	40
Hämeenpuisto	10 000	20 000	40 000	400 000	400	1	3	10
Kaukovainio	100 000	60 000	200 000	500 000	500	3	30	1
Kajaani	30 000	50 000	10 000	600 000	100	4	40	4

In further analysis, emissions from three main sources — traffic, heating and others — were studied. The emission rates of compounds from these sources in the catchments and local backgrounds are graphically illustrated in Fig. 10. The percentage distributions are given in Table 8.

According to the inventory performed, the emissions are due to the following sources in the test catchments themselves:

Kontula	practically all from traffic
Pakila	from heating of buildings, from traffic and from combustion of domestic refuse
Herttoniemi	practically all from traffic
Nekala	from heating of buildings, from industry and from traffic
Hämeenpuisto	practically all from traffic
Kaukovainio	from heating of some buildings which are not connected to district heating, from refuse incineration facilities used in some houses, and from traffic
Kajaani centre	from heating of buildings and from traffic

The catchment types based on indices derived for the housing, industrial and traffic activities with a method suggested by Laamanen (1969) are shown in Table 9.

The emission rates in the test catchments of Kontula, Nekala and Hämeenpuisto are in general lower than the emission rates in the local background. Thus the background can be expected to affect the deposition in these catchments especially. The emission rates in the catchments Herttoniemi and Kajaani centre are in general higher than those of the local background. The emissions in the catchments Pakila and Kaukovainio are roughly at the same level as in the local background. According to the inventory, the effects of the distant point sources are expected to be largest in the test catchments of Kaukovainio, Nekala and Kajaani centre, and negligible in the catchments of Pakila and Herttoniemi.

When the estimated emission rates are compared to the rough average emission rates computed for whole areas of Helsinki, Tampere and Oulu in the late 1960's (Laamanen 1973, Laamanen and Rautanen 1971, Laamanen et al. 1973) it is found that they are distinctly lower than the overall averages as to other catchments but Herttoniemi, Nekala and Hämeenpuisto. Although the anti-pollution measures probably have lowered the background levels in the cities since the late 1960's, the

Table 8. Source distribution of main emission parameters in catchments and in local backgrounds (per cent of total emission).

Parameter	Source	Percentage of emission from source in													
		catchment							local background						
		Kontula	Pakila	Herttoniemi	Nekala	Hämeenpuisto	Kaukovainio	Kajaani	Kontula	Pakila	Herttoniemi	Nekala	Hämeenpuisto	Kaukovainio	Kajaani
Particulates	traffic	100	1	100	30	100	9	20	100	14	30	4	8	28	9
	heating	0	98	0	15	0	8	80	0	85	54	13	25	28	90
	others	0	1	0	55	0	83	0	0	1	16	83	67	44	1
Hydrocarbons	traffic	100	73	100	83	100	44	95	100	96	15	82	64	82	49
	heating	0	23	0	2	0	0	4	0	3	1	5	3	1	7
	others	0	4	0	15	0	56	1	0	1	84	13	33	17	44
Oxides of nitrogen	traffic	100	64	100	63	100	96	89	100	93	43	57	17	96	31
	heating	0	36	0	32	0	4	11	0	7	8	26	21	4	69
	others	0	0	0	5	0	0	0	0	0	49	17	62	0	0
Oxides of sulphur	traffic	100	6	100	7	100	34	16	100	35	3	8	1	33	2
	heating	0	94	0	81	0	64	84	0	65	22	87	25	67	98
	others	0	0	0	12	0	2	0	0	0	75	5	74	0	0
Vanadium	traffic	-	0	-	0	-	0	0	-	0	0	0	0	0	0
	heating	-	100	-	63	-	100	100	-	100	55	95	26	100	100
	others	-	0	-	37	-	0	0	-	0	45	5	74	0	0
Lead	traffic	100	93	100	100	100	98	98	100	99	100	100	91	100	100
	heating	0	7	0	0	0	0	2	0	1	0	0	3	0	0
	others	0	0	0	0	0	2	0	0	0	0	0	6	0	0

Table 9. Indices for housing, industrial and traffic activities of test catchments. 1 = low activity, 2 = medium activity, 3 = high activity (Hokkanen et al. 1979).

Catchment	Index for			Catchment type based on emissions
	housing	industry	traffic	
Kontula	1	1	2	traffic
Pakila	3	1	1	housing
Herttoniemi	1	1	3	traffic
Nekala	1	2	2	industrial
Hämeenpuisto	1	1	3	traffic
Kaukovainio	2	1	2	housing
Kajaani	3	1	3	housing

conclusion drawn in the emission inventory was that at least the catchments Kontula, Pakila and Kaukovainio represent areas cleaner than the average in the cities under study.

4.2 Sampling of particle deposition

4.21 Sampling techniques

Deposition sampling was performed by applying the Finnish standard SFS 3865 (Suomen Standardisoimisliitto 1978) which is based on the proposal DIS 4222 outlined by the technical committee TC 146 of ISO.

The standard sampler used is a polyethene gauge (Norwegian-made NILU cylinder) with an inner diameter of 200 ± 5 mm and a height of 400 ± 10 mm (Fig. 11). The gauge is supported on a stand at height of 1.8 ± 0.2 m from the base as specified by the standard.

The sampling period was 30 ± 2 days and the deposition rates have been computed in milligrams per square metre over 30 days.

In each test catchment two deposition gauges were located. The monthly deposition was computed as an arithmetic mean of the depositions measured by the two samplers. When one of the measurements was missing, the other was used as the monthly deposition in the catchment.

The altitude and location of the two gauges in the catchments are given in Table 10. In the test sites Pakila and Herttoniemi the samplers were located on ground level. In the others the gauges were installed on roofs. In the Nekala test site a reference sampler was located in the test catchment itself. Reference measurements were performed for the analysis of deposition observed in the Hämeenpuisto catchment, too. The reference point was situated in Hatanpää, 1.5 km south of the test catchment (Table 10).

In the Herttoniemi test site, wet and dry deposition were sampled separately by a Finnish-made AAPS sampler (Fig. 12). This equipment contains two NILU gauges and automatically

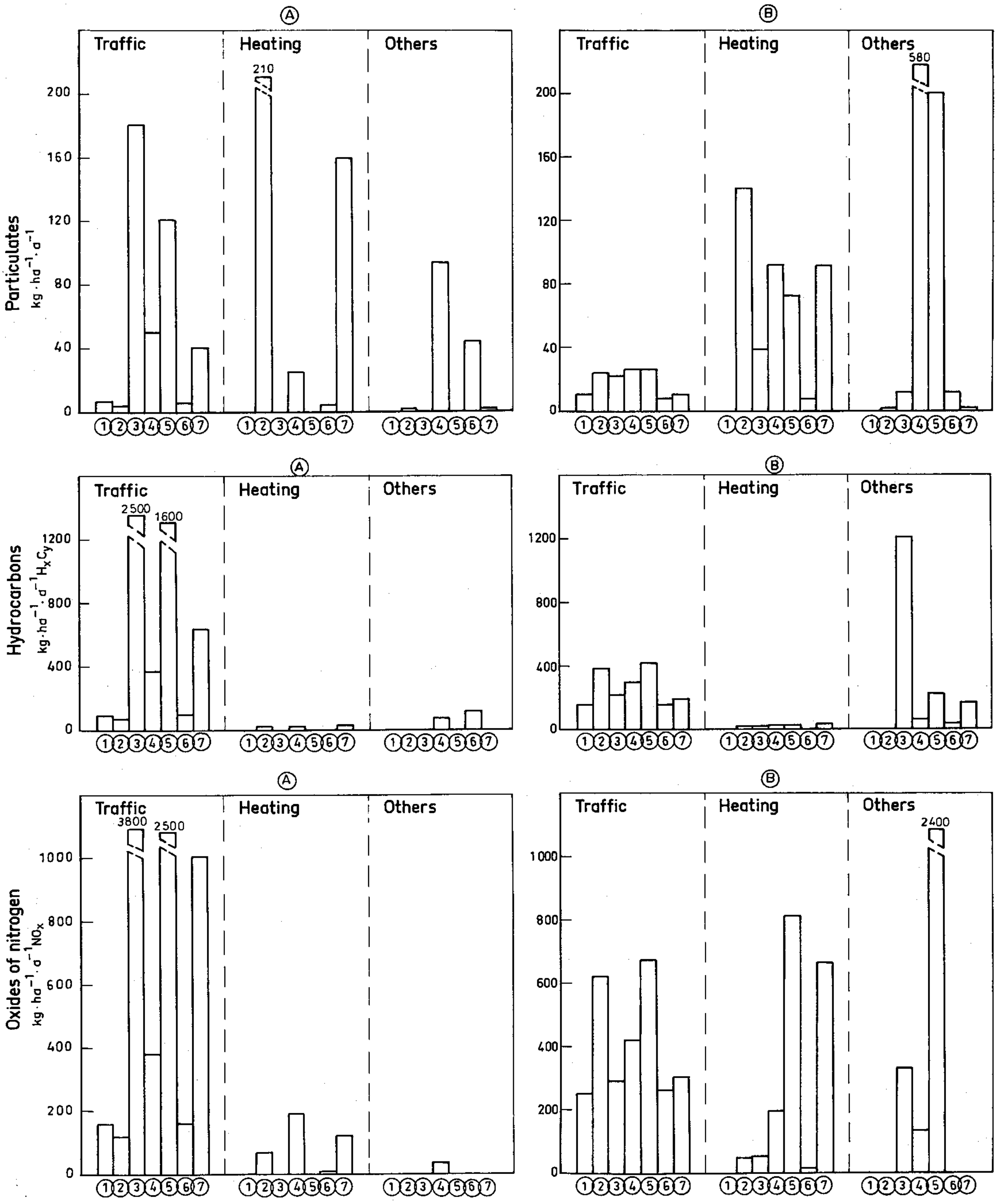

Fig. 10. Estimated emissions of main parameters from various sources. A = emission rates in test catchments, B = emission rates in local background. 1 = Kontula, 2 = Pakila, 3 = Herttoniemi, 4 = Nekala, 5 = Hämeenpuisto, 6 = Kaukovainio, 7 = Kajaani centre.

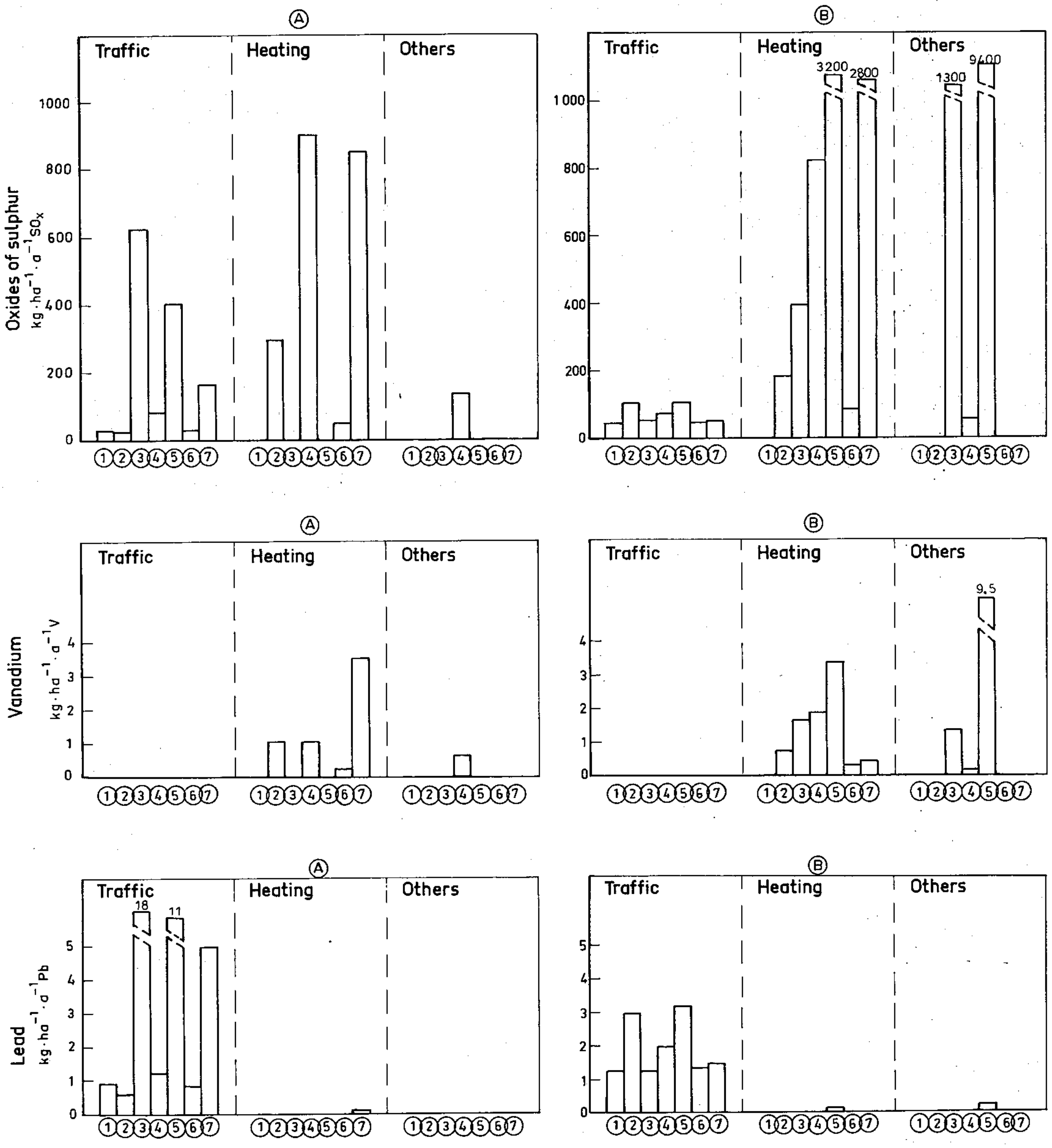

Fig. 10. Continued.

collects the wet deposition during precipitation in one of the gauges and the dry deposition during dry periods in the other gauge.

4.22 Treatment of samples and methods of analysis

To prevent the samples from drying out during the measurement period, 0.5 to 2 litres of distilled water was added to the gauges in the beginning of the sampling period.

In the laboratory the matter which does not belong to deposition (leaves, insects etc.) was removed by passing the samples through a strainer with mesh of 1 mm.

The analyses were performed directly of homogenized samples by standard methods of analysis (Appendix 1). An intercalibration study has been made between the three laboratories performing the analyses.

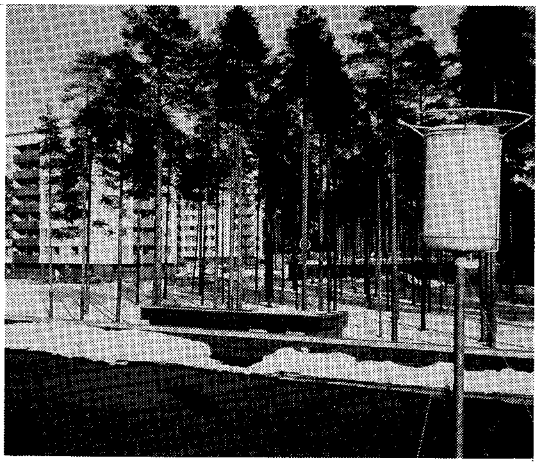

Fig. 11. Deposition sampler used in study (NILU gauge).

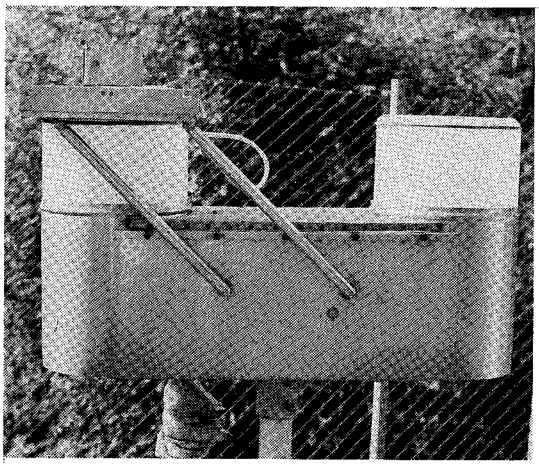

Fig. 12. Wet and dry deposition sampler used in study (AAPS sampler).

Table 10. Altitude and location of deposition samplers in test catchments. Sampler no 3 refers to reference sampling points of Nekala and Hämeenpuisto.

Catchment	Sampler no	Altitude of sampler m	Altitude of ground level in sampling point m	Distance between samplers 1 and 2 m
Kontula	1	+47	+41	400
	2	+45	+41	
Pakila	1	+27	+25	200
	2	+27	+25	
Herttoniemi[a]	1	+15	+13	3
	2	+15	+13	
Nekala	1	+92	+87	400
	2	+95	+88	
	3[b]	+88	+85	
Hämeenpuisto	1	+105	+99	500
	2	+98	+93	
	3[c]	+86	+80	
Kaukovainio	1	+22	+15	400
	2	+21	+16	
Kajaani	1	+150	+142	350
	2	+153	+146	

a: wet and dry deposition sampler has same location as standard samplers

b: located in test catchment

c: located in Hatanpää, 1.5 km to south of Hämeenpuisto catchment and 2.0 km to west of Nekala catchment

The following precision is estimated for the various laboratory analyses:

total solids (total deposition)	± 5 mg l^{-1}
volatile solids (volatile deposition)	± 6 mg l^{-1}
total organic carbon	± 1 mg l^{-1} C
total phosphorus	± 0.005 mg l^{-1} P
total nitrogen	± 0.1 mg l^{-1} N
chloride	± 0.1 mg l^{-1} Cl
calcium	± 0.1 mg l^{-1} Ca
sulphate	± 0.3 mg l^{-1} SO_4
vanadium	± 0.005 mg l^{-1} V
titanium	± 0.005 mg l^{-1} Ti
cadmium	± 0.001 mg l^{-1} Cd
zinc	± 0.005 mg l^{-1} Zn
copper	± 0.001 mg l^{-1} Cu
lead	± 0.005 mg l^{-1} Pb
pH	± 0.1
conductivity	± 0.1 mS m^{-1}

4.23 Accuracy of deposition measurement

The Finnish standard SFS 3865 (Suomen Standardisoimisliitto 1978) does not give any measure for accuracy of the sampling method because of several factors influencing the measurement.

In the study two gauges were used to measure the deposition in a catchment. Consequently, the measured monthly deposition deviates from the real deposition concerning the whole area, but the results are expected to give a reasonable basis for mutual comparisons between the test sites, i.e. the deposition levels have been derived.

Table 11. Climatological and synoptic stations of Finnish Meteorological Institute (1980) referred in study.

Station	Coordinates		Altitude from sea level	Distance
	latitude	longitude	m	km
Helsinki, Kaisaniemi	60°10'N	24°57'E	+4	5 from Herttoniemi 8 from Pakila
Helsinki, Malmi, Airport	60°15'N	25°03'E	+17	6 from Herttoniemi 2 from Kontula
Helsinki, Airport/Vantaa	60°19'N	24°58'E	+53	8 from Pakila
Tampere, Airport	61°28'N	23°44'E	+85	3 from Nekala 3 from Hämeenpuisto
Kajaani, Airport/Kajaanin mlk	64°17'N	27°41'E	+134	8 from Kajaani centre
Oulu, Airport/Oulunsalo	64°56'N	25°22'E	+12	10 from Kaukovainio
Oulu, Koskikeskus	65°01'N	25°29'E	+5	3 from Kaukovainio

The variation between deposition rates measured by the two samplers, gives a view of the magnitude of the measurement accuracy. For most deposition parameters, the difference between the monthly rates measured by the two gauges was found to be wide, being on the average of the order of ± 20 to ± 100 %. Thus a very rough average of ± 50 % might be presented to illustrate the magnitude of the deposition measurement accuracy.

4.3 Hydrometeorological data

The monthly precipitation, mean air temperature and mean wind speed used in the study, were taken from the climatological stations of the Finnish Meteorological Institute (1980), given in Table 11.

In the various catchments, the following data of precipitation and air temperature were used in computations:

Kontula	data in Helsinki, Malmi, Airport
Pakila	mean of data in Helsinki, Kaisaniemi and in Helsinki, Airport/Vantaa
Herttoniemi	mean of data in Helsinki, Kaisaniemi and in Helsinki, Malmi, Airport
Nekala and Hämeenpuisto	data in Tampere, Airport
Kaukovainio	data in Oulu, Koskikeskus
Kajaani centre	data in Kajaani, Airport/Kajaanin mlk

The monthly mean wind speeds were taken as follows:

Kontula and Herttoniemi	data in Helsinki, Malmi, Airport
Pakila	data in Helsinki, Airport/Vantaa
Nekala and Hämeenpuisto	data in Tampere, Airport
Kaukovainio	data in Oulu, Airport/Oulunsalo
Kajaani centre	data in Kajaani, Airport/Kajaanin mlk

The measured monthly precipitation is estimated to be on the average 5 % smaller than the precipitation reaching the ground in the case of rainfall, and 20—30 % smaller in the case of snowfall. The average accuracy of temperature measurement is estimated to be ± 0.1 °C. The measured monthly precipitation, air temperature and wind speed have been used in the computations.

4.4 Regional background deposition data

The deposition data describing the regional background levels were taken from the nearest non-urban precipitation stations of the National Board of Waters, given in Table 12 (Järvinen and Haapala 1980).

Statistics on the monthly deposition rate of total organic carbon, total phosphorus, total nitrogen, chloride, calcium and sulphate, plus statistics on the pH and conductivity of the deposition samples gathered in these stations over the 1971—1977 period are given in Table 13. The sampler used for gathering consisted of two polyethene cylinders placed one on top of the other. The analyses were made of homogenized samples. This enables direct comparison of the results to the results achieved in this study.

Of the figures given in Table 13, the regional background deposition rates were computed for the test catchments of this study as arithmetic means of the medians in the three nearest non-urban stations, as shown in Table 14.

Table 12. Location of nonurban precipitation (deposition) stations of National Board of Waters referred (Järvinen and Haapala 1980).

Station	Coordinates	
	latitude	longitude
Espoo	60°13'N	24°43'E
Sipoo	60°24'N	25°14'E
Vihti	60°25'N	24°24'E
Lammi	61°03'N	25°03'E
Orivesi	61°40'N	24°21'E
Jämijärvi	61°44'N	22°43'E
Pyhäntä	63°56'N	26°25'E
Kuhmo	64°16'N	29°50'E
Hailuoto	65°02'N	24°48'E
Pudasjärvi	65°22'N	27°01'E

4.5 Statistical and computational methods

The hypothesis on normal distribution of the variables was tested by the Kolmogorov-Smirnov test. Two-sided testing was used at the 95 % significance level.

Variance ratio test, t test and Welch's approximation method were used in testing the equality of variances and means of two populations. Conclusions were drawn by two-sided testing at the 95 % significance level.

Stepwise multiple regression was applied in modelling the dependent deposition parameters by the explanatory variables. The derived models fulfill the following requirements: (1) F statistic of the regression equation equals at least the 95 % point of the F distribution, and (2) t statistics of the individual regression coefficients are significant at least at the 95 % level in two-sided testing. The residuals were plotted graphically against observed values of the dependent variables in order to examine the adequacy of the regression equations derived. For the correlation and regression analysis, the accepted level of missing observations was approximately 10 %.

The Kolmogorov-Smirnov test was programmed on the UNIVAC 1108 computer by utilizing the IMSL (International Mathematical & Statistical Libraries, Inc.) subroutines. The statistical program package HYLPS was used in the correlation and regression analysis.

5. HYDROMETEOROLOGICAL CONDITIONS DURING TEST PERIOD

Observations in the climatological stations of the Finnish Meteorological Institute (1980), given in Table 11, were used to analyse the hydrometeorological conditions during the test period 1977—1979. The annual precipitation, air temperature, wind speed and wind distribution were studied and compared to the long-term averages of the 1931—1960, 1961—1975 and 1966—1975 periods available (Helimäki 1967, Kolkki 1966, Heino 1976).

According to Table 15 the annual precipitation and mean air temperature observed on the southern coast — in the Helsinki region (test catchments Kontula, Pakila and Herttoniemi) — are higher than those observed in the regions of Tampere, Oulu and Kajaani.

In 1977 the precipitation was generally higher than on the average (roughly 21—25 % in the stations of Helsinki, 20 % in Tampere, and 15 % in Kajaani; in the Oulu region the precipitation of 1977 was normal). The mean air temperature in 1977 was generally lower than the long-term average.

The precipitation in 1978 was in general lower than the long-term average (rouhly 6—24 % in the Helsinki region, 6 % in Tampere, 27 % in Oulu and 29 % in Kajaani). The mean air temperatures for 1978 were also lower than the long-term values, and even more distinctly than in 1977.

The precipitation in 1979 was slightly lower (4—6 %) than the long-term average in the stations of Helsinki, Kaisaniemi and Helsinki, Airport/Vantaa. In the other stations the 1979 precipitation was slightly (7—11 %) higher than on the average. The annual mean temperatures of 1979 were nearly normal.

The total precipitation during the 1977—1979 period has been close to the long-term average, the difference between the 1977—1979 total and the long-term average over three years is of the order of − 3 to + 12 % in the regions of Helsinki and Tampere, and − 6 to − 1 % in the regions of Oulu and Kajaani.

On an annual basis, the wind speeds during the 1977—1979 period differ little from the long-term averages (Table 16). The average wind speed in the inland stations (Tampere, Airport and Kajaani, Airport/Kajaanin mlk) is somewhat lower than that in the stations of the coastal zone.

The winds blowing from the south-southwest direction are prevailing in the Helsinki and Tampere regions, and the winds from the southeast-

Table 13. Deposition of parameters observed over 1971–1977 period in stations of National Board of Waters. min = smallest observed value, $\bar{x}_{a,min}$ = smallest annual arithmetic mean, $\bar{x}_{a,max}$ = highest annual arithmetic mean, m_{71-77} = median value over 1971–1977 period, max = highest observed value (Järvinen and Haapala 1980).

Station	Values of parameter																			
	total organic carbon mg m^{-2} C in 30 days					total phosphorus mg m^{-2} P in 30 days					total nitrogen mg m^{-2} N in 30 days					chloride mg m^{-2} Cl in 30 days				
	min	$\bar{x}_{a,min}$	$\bar{x}_{a,max}$	m_{71-77}	max	min	$\bar{x}_{a,min}$	$\bar{x}_{a,max}$	m_{71-77}	max	min	$\bar{x}_{a,min}$	$\bar{x}_{a,max}$	m_{71-77}	max	min	$\bar{x}_{a,min}$	$\bar{x}_{a,max}$	m_{71-77}	max
Espoo	19	84	232	75	340	<0.1	1.4	4.8	1.5	29.0	18	53	113	75	356	9	38	119	50	271
Sipoo	18	99	208	124	556	0.3	2.7	5.9	2.4	37.8	9	41	108	59	246	<4	37	91	45	235
Vihti	12	61	477	62	1 237	0.1	1.1	3.5	1.1	23.2	11	48	93	60	191	6	26	67	37	122
Lammi	11	103	391	108	1 511	<0.1	2.5	8.9	2.1	49.6	11	40	72	46	166	7	31	97	48	244
Orivesi[a]	17	81	426	93	1 160	0.1	1.3	2.7	1.2	11.0	11	39	49	40	103	6	18	56	30	240
Jämijärvi	25	85	320	105	1 115	0.1	1.6	3.4	1.9	10.7	25	59	120	82	226	5	37	92	55	241
Pyhäntä	10	54	245	71	384	0.1	1.0	2.0	0.9	7.6	7	35	68	45	161	3	17	75	24	224
Kuhmo	8	58	434	80	2 122	<0.1	1.1	2.5	0.9	14.1	10	30	53	40	123	4	16	81	24	268
Hailuoto	14	44	97	51	221	0.1	1.4	4.3	0.8	21.4	10	33	60	42	141	5	20	70	30	301
Pudasjärvi	26	78	354	99	618	0.2	0.9	3.3	1.4	20.6	<4	28	59	35	172	5	15	162	47	378

Table 13. Continued.

Station	Values of parameter																			
	calcium mg m^{-2} Ca in 30 days					sulphate mg m^{-2} SO_4 in 30 days					pH[b]					conductivity[c] ($\varkappa_{25}$) mS m^{-1}				
	min	$\bar{x}_{a,min}$	$\bar{x}_{a,max}$	m_{71-77}	max	min	$\bar{x}_{a,min}$	$\bar{x}_{a,max}$	m_{71-77}	max	min	$\bar{x}^{d}_{a,min}$	$\bar{x}^{d}_{a,max}$	m_{71-77}	max	min	$\bar{x}_{a,min}$	$\bar{x}_{a,max}$	m_{71-77}	max
Espoo	<7	48	100	63	178	101	256	424	313	853	3.9	4.4	5.0	4.6	7.8	1.4	3.3	4.5	3.7	9.5
Sipoo	2	29	65	47	171	41	201	346	237	870	3.6	4.5	4.8	4.7	7.6	1.0	3.3	4.0	3.5	9.8
Vihti	6	27	80	36	353	32	192	279	221	600	3.7	4.4	5.4	4.4	7.0	1.1	3.5	4.5	3.4	14.5
Lammi	5	24	74	42	260	43	156	405	209	703	3.9	4.4	5.0	4.8	6.8	1.2	2.4	5.0	3.2	11.0
Orivesi[a]	4	32	58	36	158	28	139	191	167	381	3.9	4.5	4.9	4.8	7.2	0.7	2.8	4.3	3.1	8.6
Jämijärvi	3	18	67	33	143	22	195	272	264	438	4.0	4.5	4.9	4.9	6.5	1.3	2.8	4.4	3.4	8.7
Pyhäntä	1	11	66	26	256	27	123	224	145	503	4.2	4.6	5.0	4.9	6.7	0.8	2.5	3.9	2.4	12.4
Kuhmo	4	16	48	23	182	31	100	142	117	275	3.6	4.4	5.1	4.8	7.0	0.8	2.3	3.9	2.6	12.0
Hailuoto	<1	14	40	18	131	25	111	182	151	432	4.0	4.3	4.6	4.5	6.9	1.2	2.9	3.6	3.1	7.7
Pudasjärvi	<2	15	70	37	141	26	109	198	153	509	3.9	4.4	4.9	4.7	6.4	0.9	2.1	4.3	2.8	7.5

a: period 1971–1975; b: pH value of deposition samples (precipitation); c: conductivity of deposition samples (precipitation); d: median values

Table 14. Regional background deposition rates used in regression analysis. In test catchments Kontula, Pakila and Herttoniemi means of medians over 1971–1977 period in stations Espoo, Sipoo and Vihti. In test catchments Nekala and Hämeenpuisto means of medians over 1971–1977 period in stations Lammi, Orivesi and Jämijärvi. In test catchment Kaukovainio means of medians over 1971–1977 period in stations Pyhäntä, Hailuoto and Pudasjärvi. In test catchment Kajaani centre means of medians over 1971–1977 period in stations Pyhäntä, Kuhmo and Pudasjärvi.

Test catchment	Background deposition of parameter							
	total organic carbon TOC_{rb} $mg\,m^{-2}\,C$ in 30 days	total phosphorus $tot\,P_{rb}$ $mg\,m^{-2}\,P$ in 30 days	total nitrogen $tot\,N_{rb}$ $mg\,m^{-2}\,N$ in 30 days	chloride Cl_{rb} $mg\,m^{-2}\,Cl$ in 30 days	calcium Ca_{rb} $mg\,m^{-2}\,Ca$ in 30 days	sulphate $SO_{4,rb}$ $mg\,m^{-2}\,SO_4$ in 30 days	pH[a] pH_{rb}	conductivity[b] $\gamma_{25,rb}$ $mS\,m^{-1}$
Kontula, Pakila & Herttoniemi	87	1.7	65	44	49	257	4.6	3.5
Nekala & Hämeenpuisto	102	1.7	56	44	37	213	4.8	3.2
Kaukovainio	74	1.0	41	34	27	150	4.7	2.8
Kajaani	83	1.1	40	32	29	138	4.8	2.6

a: pH value of precipitation; b: conductivity of precipitation

Table 15. Measured annual precipitation and mean air temperature in studied climatological and synoptic stations in years 1977–1979 and long-term averages (Finnish Meteorological Institute 1980, Helimäki 1967, Kolkki 1966, Heino 1976).

Station	Precipitation (mm) and mean air temperature (°C) in year									
	1977		1978		1979		1931–1960 average		1961–1975 average	
	mm	°C	mm	°C	mm	°C	mm	°C	mm	°C
Helsinki, Kaisaniemi	785	4.6	605	3.9	618	4.7	647	5.4[a]	548	5.4
Helsinki, Malmi, Airport	798	4.0	575	3.3	689	4.3	-[b]	4.4[c]	637	4.7
Helsinki, Airport/Vantaa	758	4.0	475	3.2	586	4.2	-	-	624	4.7
Tampere, Airport	612	3.5	476	3.0	542	3.8	-	3.8[d]	508	4.0
Kajaani, Airport/Kajaanin mlk	614	1.2	376	−0.2	591	1.5	-	-	533	1.6
Oulu, Koskikeskus	517	1.6	375	1.1	557	2.5	514[e]	-	516	2.2

a: Helsinki, Vuorikatu; b: not available; c: 24 observation years; d: 13 observation years; e: Oulu, Kaupunki

south direction in the Oulu and Kajaani regions. As to the wind distribution, minor variation occurs over the 1977—1979 period. The year 1978 differs most from the average distribution observed in the period 1966—1975 (Table 16).

6. RESULTS AND DISCUSSION

6.1 Wet and dry deposition observed in Herttoniemi catchment

The monthly wet and dry deposition measured by the AAPS sampler over the 1978—1979 period, and the deposition collected by the standard samplers (containing both the wet and dry depositions) over the same sampling period in the Herttoniemi catchment are graphically illustrated in Appendix 2.

6.11 Variable distributions and magnitude of variance

The number of observations, and the values of the key statistics of the hydrometeorological variables and wet and dry deposition parameters are given in Tables 17 and 18.

At the 95 % significance level the hypothesis on normal distribution cannot be rejected for any of the hydrometeorological variables or wet and dry deposition parameters.

The variance of the deposition rates is generally wide, the standard deviations are on the average of the order of 50—100 % of the arithmetic means.

6.12 Deposition parameters correlating with hydrometeorological variables and with each other

The deposition parameters that correlate with the

Table 16. Measured annual mean wind speed and wind distribution in five of studied synoptic stations in years 1977–1979 and long-term averages (Finnish Meteorological Institute 1980, Heino 1976).

Station	Year	Mean wind speed m s^{-1}	Percentage of wind blowing from direction (%)								
			N	NE	E	SE	S	SW	W	NW	calm
Helsinki, Malmi, Airport	1977	3.9	11	8	12	13	18	14	8	10	6
	1978	3.7	14	12	10	9	15	15	9	9	7
	1979	3.9	9	7	12	11	20	17	10	7	7
	1966–1975 average	3.9	12	6	11	10	19	16	10	9	7
Helsinki, Airport/Vantaa	1977	3.9	12	11	9	14	11	19	10	11	3
	1978	3.8	10	20	5	12	7	20	10	12	4
	1979	4.1	9	13	8	12	16	21	11	9	1
	1966–1975 average	4.0	11	9	10	10	18	15	13	11	3
Tampere, Airport	1977	3.4	12	8	11	9	20	15	11	7	7
	1978	3.2	11	12	10	7	14	14	12	8	12
	1979	3.3	8	5	12	10	21	17	12	5	10
	1966–1975 average	3.3	10	7	11	9	20	14	12	7	10
Kajaani, Airport/Kajaanin mlk	1977	3.4	9	7	11	14	21	12	12	9	5
	1978	3.4	8	9	14	10	16	12	15	10	6
	1979	3.5	5	9	13	16	18	19	10	8	2
	1966–1975 average	3.5	6	6	11	14	22	12	15	9	5
Oulu, Airport/Oulunsalo	1977	3.7	13	9	10	19	15	9	10	12	3
	1978	3.7	13	10	9	17	13	9	11	13	5
	1979	3.8	8	7	12	22	19	11	10	7	4
	1966–1975 average	4.1	10	7	9	20	16	12	12	11	3

Table 17. Properties of distributions of hydrometeorological variables in connection with wet and dry deposition sampling in Herttoniemi catchment. n = number of observations, min = minimum value, m = median value, max = maximum value, $\bar{x}$ = arithmetic mean, s = standard deviation, D = Kolmogorov-Smirnov test statistic. Hypothesis on normal distribution of variables cannot be rejected at 95 % significance level in two-sided testing (critical value of D is 0.349).

Variable and unit	Values of statistics						
	n	min	m	max	$\bar{x}$	s	D
Monthly precipitation P mm	14	23.1	49.6	136.3	62.4	38.5	0.229
Mean monthly air temperature T °C	14	−3.2	7.0	16.6	7.3	6.5	0.159
Mean monthly wind speed W m s^{-1}	14	3.2	4.0	4.9	4.0	0.5	0.158

hydrometeorological variables, and those correlating with each other at the significance level ≥ 95 % in the wet and dry deposition, and in the deposition collected by the standard samplers (including both the wet and dry depositions) are given in Table 19.

Precipitation has an increasing effect on the deposition of total nitrogen, chloride and sulphate. In total nitrogen, this effect can be found both in the wet deposition and in the deposition collected by the standard samplers. In dry deposition the correlation between precipitation and sulphate deposition is negative, thus reflecting strong washout effect of sulphur compounds caused by precipitation.

In general, the correlations of air temperature with the wet and dry deposition parameters are non-significant. In wet deposition conductivity decreases with increasing air temperature, indicating a higher amount of water soluble salts in wet deposition during the winter. In the deposition collected by the standard samplers, the deposition of total organic carbon increases during the summer period. The deposition of vanadium increases during wintertime due to the emissions from combustion of fossil fuels.

Table 18. Properties of distributions of wet and dry deposition parameters in Herttoniemi catchment. w refers to wet deposition, d refers to dry deposition, parameter notation without footnote refers to deposition collected by standard samplers over same period. n = number of observations, min = minimum value, m = median value, max = maximum value, $\bar{x}$ = arithmetic mean, s = standard deviation, D = Kolmogorov-Smirnov test statistic. Hypothesis on normal distribution of wet and dry deposition parameters cannot be rejected at 95 % significance level in two-sided testing[a].

Parameter and unit	Values of statistics						
	n	min	m	max	$\bar{x}$	s	D
Total deposition mg m^{-2} in 30 days							
tot D_w	13	500	1 200	5 590	1 660	1 340	0.318
tot D_d	14	120	2 290	13 200	2 870	3 300	0.258
tot D	13	1 010	3 150	20 100	4 440	4 850	
Volatile deposition mg m^{-2} in 30 days							
vol D_w	12	270	870	1 730	970	440	0.170
vol D_d	13	30	1 100	5 660	1 510	1 580	0.281
vol D	12	270	1 620	11 700	2 520	3 010	
Total organic carbon mg m^{-2} C in 30 days							
TOC_w	11	72	146	505	193	138	0.206
TOC_d	10	38	181	724	222	190	0.309
TOC	11	153	422	802	471	233	
Total phosphorus mg m^{-2} P in 30 days							
tot P_w	14	0.1	1.6	5.4	1.8	1.5	0.145
tot P_d	14	0.3	2.0	12.5	3.2	3.3	0.206
tot P	13	0.8	7.0	28.6	8.1	7.9	
Total nitrogen mg m^{-2} N in 30 days							
tot N_w	14	15	54	170	65	42	0.179
tot N_d	14	4	25	54	25	15	0.124
tot N	13	26	97	132	84	35	
Chloride mg m^{-2} Cl in 30 days							
Cl_w	14	15	44	166	55	40	0.174
Cl_d	14	7	26	134	37	39	0.328
Cl	13	36	85	150	81	39	
Calcium mg m^{-2} Ca in 30 days							
Ca_w	14	10	41	95	43	23	0.178
Ca_d	13	8	41	118	47	35	0.174
Ca	13	42	87	152	87	32	
Sulphate mg m^{-2} SO_4 in 30 days							
$SO_{4,w}$	14	113	355	1 043	410	231	0.157
$SO_{4,d}$	14	46	269	570	276	189	0.149
SO_4	13	480	651	1 077	717	195	
Vanadium[b] mg m^{-2} V in 30 days							
V_w	12	0.05	0.22	0.59	0.27	0.16	0.170
V_d	12	0.04	0.24	0.89	0.28	0.22	0.236
V	12	0.16	0.44	1.14	0.48	0.30	
Cadmium[c] mg m^{-2} Cd in 30 days							
Cd_w	14	<0.01	-	<0.06	-	-	-
Cd_d	14	<0.01	-	<0.02	-	-	-
Cd	13	<0.01	-	<0.06	-	-	-
Zinc mg m^{-2} Zn in 30 days							
Zn_w	14	0.58	1.56	2.89	1.56	0.66	0.107
Zn_d	14	0.13	2.57	7.86	3.13	2.37	0.273
Zn	13	0.80	4.88	10.39	5.52	2.95	

Table 18. Continued.

Parameter and unit	Values of statistics						
	n	min	m	max	$\bar{x}$	s	D
Copper mg m^{-2} Cu in 30 days							
Cu_w	14	0.15	0.30	1.10	0.36	0.24	0.265
Cu_d	14	0.04	0.28	0.56	0.29	0.14	0.137
Cu	13	0.26	0.41	0.54	0.40	0.09	
Lead mg m^{-2} Pb in 30 days							
Pb_w	14	0.18	0.88	2.96	1.02	0.71	0.178
Pb_d	14	0.15	1.13	2.72	1.15	0.78	0.152
Pb	13	0.77	2.17	3.96	2.17	0.84	
pH value							
pH_w	14	3.5	4.2	5.4	-	-	0.258
pH	13	3.4	3.9	4.8	-	-	
Conductivity mS m^{-1}							
$\gamma_{25,w}$	14	1.3	3.8	7.1	4.2	1.8	0.138
γ_{25}	13	5.3	6.7	15.6	7.8	2.7	

a: critical values of Kolmogorov-Smirnov test statistic:

n	= 10	11	12	13	14
$D_{critical}$	= 0.409	0.391	0.375	0.361	0.349

b: concentration of vanadium is at detection limit (< 5 $\mu g\ l^{-1}$ V) in 1 (V), 2 (V_d) and 3 (V_w) samples

c: concentration of cadmium is at detection limit (<1 $\mu g\ l^{-1}$ Cd) in all samples

Wind has an effect on deposition collected by the techniques used. In the wet and dry deposition, the total and volatile deposition and the deposition of phosphorus are found to decrease with increasing wind speed.

As to mutual correlations between the deposition parameters, positive correlation exists between total and volatile depositions, total deposition and zinc deposition, and depositions of calcium and sulphate in both the wet and dry deposition, and in the deposition collected by the standard samplers. The pH value decreases with a growing deposition of sulphate and lead both in the wet deposition and in the deposition collected by the standard samplers. The value of conductivity increases with a growing deposition of lead both in the wet deposition and in the deposition containing the wet and dry fractions.

The correlation analysis shows that in this catchment sulphate, nitrogen and chloride are especially related to the wet deposition, and volatile matter (organic matter), zinc, phosphorus and organic carbon to the dry deposition.

The pH value of the deposition samples collected by the standard samplers (containing both wet and dry deposition) is on the average 0.3 units lower than pH of the wet deposition samples. The two pH values correlate positively at the 99.9 % significance level. The conductivity of samples containing both the wet and dry fractions is noticeably higher than that of wet deposition samples. The correlation between these two conductivities was yet statistically non-significant.

6.13 Fraction of variance explained by hydrometeorological variables

Regression models were formed for the wet and dry deposition parameters with stepwise regression technique by using monthly precipitation, air temperature and wind speed as explanatory variables. To allow square-root and logarithmic transformations of the temperature, a modified variable was formed by adding 20 °C to each monthly temperature ($T_{20} = T + 20$).

The variables in the final models, the fraction of variance explained by them, and the significance level of coefficients in the regression equations are shown in Table 20.

Roughly one third of the variance of total deposition in both the wet and dry deposition is explained by the monthly wind speed, which has a negative regression coefficient, thus reflecting the sensitivity of the sampling method to aerodynamic effects. The same holds true as to the deposition of organic matter in dry deposition, and of phosphorus in wet deposition.

Table 19. Parameters having positive (+) or negative (–) correlation in wet deposition (w), dry deposition (d) and deposition collected by standard samplers (s) over same period in Herttoniemi catchment. Significance levels of correlation ≥95 %.

	Monthly precipitation	Mean monthly air temperature	Mean monthly wind speed	Total deposition	Volatile deposition	Total organic carbon	Total phosphorus	Total nitrogen	Chloride	Calcium	Sulphate	Vanadium	Zinc	Copper	Lead	pH	Conductivity
Monthly precipitation																	
Mean monthly air temperature																	
Mean monthly wind speed																	
Total deposition			– w														
Volatile deposition			– d	+ + + w d s													
Total organic carbon		+ s															
Total phosphorus			– w	+ s	+ s	+ + d s											
Total nitrogen	+ + w s				+ d	+ w											
Chloride	+ w					+ d	+ d	+ d									
Calcium						+ d	+ d	+ + w d									
Sulphate	+ – w d							+ + w s	+ + w s	+ + + w d s							
Vanadium		– s				– s	– s	+ d	+ w		+ + w d						
Zinc		+ d		+ + + w d s	+ + d s		+ s										
Copper	+ d			+ w	+ w	– d	– s		– s				+ w				
Lead			+ d			– s		+ w	+ w	+ + w d	+ + w d	+ + w d					
pH			– s					– s			– – w s	– w			– – w s		
Conductivity		– w									+ w	+ w			+ + w s	– w	

Precipitation explains approximately half of the variance of total nitrogen in the wet deposition indicating the washout mechanism of nitrogen compounds from the atmosphere.

Precipitation and temperature explain nearly three fourths of the variance of wet deposition of chloride. The negative regression coefficient of the air temperature reflects the increase of chloride emissions due to heating in the winter.

More than half of the variance of the sulphate's wet deposition is explained by precipitation and air temperature. The negative coefficient of temperature reflects the growth of the emissions of sulphur oxides due to heating in the wintertime.

Temperature and precipitation explain more than half of the variance of the vanadium's wet deposition. The negative regression coefficient of temperature refers to the principal source of vanadium emissions — combustion of the fossil fuels (principally fuel oil).

Half of the variance of the wet deposition of lead can be explained by precipitation and temperature. The negative coefficient of temperature refers to the increased amount of cold starts of the

Table 20. Hydrometeorological variables explaining wet and dry deposition in Herttoniemi catchment and fraction of variance explained. w refers to wet deposition, d refers to dry deposition, wpl refers to percentage wet deposition of sum of wet and dry deposition, wp2 refers to percentage wet deposition of deposition collected by standard samplers. (+) indicates increasing effect of explanatory variable, (−) indicates decreasing effect of explanatory variable. Significance level shows level at which each or weakest of regression coefficients of explanatory variables in model is significant in two-sided testing by t test. N.S. indicates statistically non-significant model (significance level < 95 %).

Parameter	Variables in model	Fraction of variance explained 100 R^2 %	Significance level %
Total deposition			
tot D_w	(−) ln W	36.9	95
tot D_d	(−) ln W	29.5	95
tot D_{wp1}	N.S.	-	-
tot D_{wp2}	N.S.	-	-
Volatile deposition			
vol D_w	N.S.	-	-
vol D_d	(−) ln W	34.2	95
vol D_{wp1}	N.S.	-	-
vol D_{wp2}	(−)T	50.9	95
Total organic carbon			
TOC_w	N.S.	-	-
TOC_d	N.S.	-	-
TOC_{wpl}	(+) P	50.7	95
TOC_{wp2}	(+) P (−) T	77.7	99
Total phosphorus			
tot P_w	(−) ln W	32.7	95
tot P_d	N.S.	-	-
tot P_{wp1}	(+) ln P	28.9	95
tot P_{wp2}	N.S.	-	-
Total nitrogen			
tot N_w	(+) P	52.2	99
tot N_d	N.S.	-	-
tot N_{wp1}	N.S.	-	-
tot N_{wp2}	(+) P	37.6	95
Chloride			
Cl_w	(+) P (−) T	70.4	99
Cl_d	N.S.	-	-
Cl_{wp1}	N.S.	-	-
Cl_{wp2}	(+) P	56.0	99
Calcium			
Ca_w	N.S.	-	-
Ca_d	N.S.	-	-
Ca_{wp1}	N.S.	-	-
Ca_{wp2}	N.S.	-	-
Sulphate			
$SO_{4,w}$	(+)P(−)lnT_{20}	62.2	95
$SO_{4,d}$	(−) P	32.8	95
$SO_{4,wp1}$	(+) P	41.4	95
$SO_{4,wp2}$	N.S.	-	-
Vanadium			
V_w	(−)lnT_{20}(+)lnP	62.4	95
V_d	N.S.	-	-
V_{wp1}	N.S.	-	-
V_{wp2}	N.S.	-	-
Zinc			
Zn_w	N.S.	-	-
Zn_d	(+) T	31.3	95
Zn_{wp1}	(−) ln T_{20}(+) P	61.7	95
Zn_{wp2}	(−) T (+) P	66.0	99
Copper			
Cu_w	N.S.	-	-
Cu_d	N.S.[a]	-	-
Cu_{wp1}	N.S.	-	-
Cu_{wp2}	N.S.	-	-
Lead			
Pb_w	(+) P (−) lnT_{20}	50.3	95
Pb_d	N.S.[b]	-	-
Pb_{wp1}	N.S.	-	-
Pb_{wp2}	(+) P (−) T	55.8	95
pH_w	N.S.	-	-
$\gamma_{25,w}$	(−) ln T_{20}	38.8	95

a: precipitation excluded as explanatory variable
b: wind excluded as explanatory variable

automobiles and the less efficient performance of the internal combustion engine at low speeds in the winter.

The models of organic carbon, calcium and copper proved to be non-significant both in the wet and dry depositions. The model of pH value of the wet deposition samples was not significant either.

6.14 Proportion of wet deposition

The proportions of the wet deposition of various parameters were studied by computing the ratio of wet deposition to the sum of wet and dry deposition and to the deposition collected by the standard samplers (containing both the wet and dry fractions) for each sampling month.

Regression models were formed for the proportions by using the hydrometeorological variables as explanatory variables (Table 20).

The models derived for the wet — deposition proportions of total deposition, calcium, vanadium and copper proved to be non-significant.

As can be seen in Table 20, the significances of the models derived for the proportions, computed

Table 21. Proportion of wet deposition in Herttoniemi catchment. Averages of percentages computed by arithmetic means of parameter depositions.

Parameter	Proportion of wet deposition of sum of wet and dry deposition %			Proportion of wet deposition of deposition collected by standard samplers %		
	minimum	average	maximum	minimum	average	maximum
Total deposition	22	37	95	25	37	117
Volatile deposition	23	39	96	15	38	113
Total organic carbon	25	47	86	14	41	89
Total phosphorus	8	36	88	3	22	100
Total nitrogen	29	72	97	46	77	147
Chloride	27	60	90	36	68	104
Calcium	16	48	92	14	49	102
Sulphate	17	60	96	18	57	97
Vanadium	15	49	78	21	56	83
Zinc	15	33	95	17	28	100
Copper	34	55	82	38	90	423
Lead	21	47	95	20	47	100

in the two ways, differ from each other, which is due to inaccuracies of the sampling method, and to the relatively short measurement period. Yet, the following conclusions may be drawn in this catchment. The dry proportion of volatile deposition increases in the summer period. The wet proportion of organic carbon deposition increases with precipitation, but decreases during summer period. The wet proportion of phosphorus deposition slightly increases with precipitation. The wet proportions of nitrogen, chloride and sulphate increase with a growing amount of precipitation. The dry proportion of zinc deposition increases during the summer period but decreases with precipitation. The wet proportion of lead increases with precipitation and decreases during the summertime.

The minimum, average and maximum of the wet proportions of various parameters observed over the 1978—1979 period are shown in Table 21. Inaccuracies in the data are reflected by wide variation of the wet proportions. Yet, the following wet proportions may be concluded:

proportion of wet deposition

< 50 %	> 50 %
total deposition	nitrogen
volatile deposition	chloride
organic carbon	sulphate
phosphorus	
zinc	

On the basis of observations, the wet and dry proportions of calcium, vanadium and lead are roughly equal. Some obviously erroneous copper values in the analysis prevent the making of conclusions about the wet proportion of this parameter.

Based on the observations of wet deposition, the average composition of rainwater in the Herttoniemi area has been computed as shown in Table 22.

6.15 Reliability of observations

A comparison of the computed sum of measured wet and dry deposition to the measured deposition containing both the wet and dry fractions (by standard samplers) reveals the wide range in which the ratio of these two values may vary on a monthly basis (Table 23).

On the average, over a long period of time, the sum of the wet and dry depositions equals the combined deposition surprisingly well. Some anomalous values in the observations of phosphorus and copper have their effects even on the averages in the data analysed.

6.2 Monthly deposition observed in test catchments

The monthly rates of deposition of the various parameters measured during 1977—1979 in the seven test catchments are graphically illustrated in Appendix 3.

To emphasize the importance of seasons on the deposition levels, the hydrological year was divided into two parts: vegetation period and non-vegetation period. The vegetation period was defined as May-October in the case of the test catchments in Helsinki (Kontula, Pakila, Herttoniemi) and Tampere (Nekala, Hämeenpuisto), and

Table 22. Average composition of rainwater (wet deposition) in Herttoniemi catchment.

Parameter	Unit	Concentration
Total solids (total deposition)	mg l^{-1}	25
Volatile solids (volatile deposition)	mg l^{-1}	15
Total organic carbon	mg l^{-1} C	2.8
Total phosphorus	mg l^{-1} P	0.029
Total nitrogen	mg l^{-1} N	1.0
Chloride	mg l^{-1} Cl	0.88
Calcium	mg l^{-1} Ca	0.69
Sulphate	mg l^{-1} SO_4	6.6
Vanadium	mg l^{-1} V	0.004
Cadmium	mg l^{-1} Cd	<0.001
Zinc	mg l^{-1} Zn	0.025
Copper	mg l^{-1} Cu	0.006
Lead	mg l^{-1} Pb	0.016
pH		4.2
Conductivity	mS m^{-1}	3.8

Table 23. Comparison of sum of wet and dry deposition to deposition collected by standard samplers in Herttoniemi catchment. Averages of percentages computed by arithmetic means of parameter depositions.

Parameter	Percentage of wet + dry deposition of deposition collected by standard samplers %			
	minimum	median	maximum	average
Total deposition	67	98	172	102
Volatile deposition	63	111	211	98
Total organic carbon	39	81	141	88
Total phosphorus	10	89	131	62
Total nitrogen	74	93	196	107
Chloride	67	116	286	114
Calcium	54	107	131	103
Sulphate	57	101	132	96
Vanadium	74	124	189	115
Zinc	37	103	148	85
Copper	73	143	531	163
Lead	63	99	123	100

May-September in the case of the catchments in Oulu (Kaukovainio) and Kajaani (Kajaani centre). The main purpose of this classification was to separate the season requiring heating from the rest of the hydrological year.

6.21 Variable distributions and magnitude of variance

The number of observations and the values of the key statistics illustrating distributions of the hydrometeorological variables and deposition parameters are given in Tables 24 and 25. The averages are graphically presented in Figs. 13 and 14. The average depositions per millimetre of precipitation, or composition of "precipitation" in milligrams per litre, are graphically illustrated in Fig. 15.

Fig. 13 shows that both the average precipitation and air temperature decrease when moving from the catchments on the southern coast to the catchments in central and northern Finland.

At the significance level of 95 % the hypothesis on normal distribution of the hydrometeorological variables cannot be rejected in any of the catchments. In the case of combined observations (Nekala excluded) the hypothesis on normally distributed monthly precipitation has to be rejected in two-sided testing.

As to the deposition parameters, the hypothesis on normal distribution cannot in general be rejected in the individual catchments. The exceptions are the distributions of total deposition in the Kontula, Pakila and Herttoniemi catchments, the distribution of chloride in the Nekala catchment, and the distribution of zinc in the Kontula catchment. Yet, in the case of the combined observations, the hypothesis on normal distribution has to be rejected for all other parameters but sulphate and conductivity.

Only a few observations on titanium deposition were gathered. The concentrations of cadmium have been within the detection limit ($< 1\ \mu g\ l^{-1}$ Cd) in 77 to 100 % of the samples in the individual catchments, thus making these results unreliable. Consequently, the deposition of cadmium was not included in further analyses. As to vanadium, the concentrations have been at the detection limit ($< 5\ \mu g\ l^{-1}$ V) in 0 to 17 % of the samples.

A typical characteristic of the measured deposition rates is the wide variation which is found in every catchment for all parameters studied. On the average, the standard deviations are of the order of 50—100 % of the arithmetic means.

The statistics given in Table 26 show that the deposition measured over 1977—1979 period in the Hämeenpuisto catchment has been higher than the reference deposition measured in the reference point of Hatanpää (1.5 km to the south of Hämeenpuisto catchment) concerning the following parameters: total deposition, organic carbon, calcium, sulphate, vanadium, copper and lead. A big difference is found to occur especially in the deposition of lead.

The deposition measured during 1979 over a seven-month period in the reference point located in the test catchment of Nekala is shown in Table

Table 24. Properties of distributions of hydrometeorological variables. Season: I = hydrological year, II = vegetation period, III = non-vegetation period. n = number of observations, min = minimum value, m = median value, max = maximum value, $\bar{x}$ = arithmetic mean, s = standard deviation, D = Kolmogorov-Smirnov test statistic. Value of D underlined shows that hypothesis on normal distribution of variable has to be rejected at 95 % significance level in two-sided testing[a].

Catchment	Season	Values of statistics																				
		monthly precipitation P mm							mean monthly air temperature T °C								mean monthly wind speed W m s^{-1}					
		n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D
Kontula	I	19	9.4	51.3	142.3	65.4	40.8	0.161	19	−11.4	4.2	16.3	5.6	8.4	0.159	19	2.8	3.9	4.6	3.8	0.5	0.184
	II	11	26.0	58.7	127.6	64.8	31.1		11	4.1	14.0	16.3	11.1	4.8		11	3.1	3.9	4.6	3.8	0.5	
	III	8	9.4	37.4	142.3	66.1	53.8		8	−11.4	1.6	2.0	−2.0	5.7		8	2.8	3.9	4.1	3.7	0.5	
Pakila	I	19	8.9	43.7	132.1	60.7	38.7	0.196	19	−10.8	4.5	16.4	5.8	8.2	0.160	19	3.1	3.9	4.5	3.8	0.4	0.204
	II	11	19.9	55.2	132.1	63.4	35.5		11	4.5	14.1	16.4	11.3	4.7		11	3.1	3.8	3.9	3.6	0.3	
	III	8	8.9	33.4	118.7	56.9	44.9		8	−10.8	1.7	2.3	−1.7	5.5		8	3.5	4.1	4.5	4.1	0.3	
Herttoniemi	I	21	10.2	46.4	136.3	62.6	38.6	0.207	21	−10.8	4.5	16.6	5.5	8.0	0.147	21	2.8	3.9	4.9	3.8	0.5	0.118
	II	11	23.4	54.0	136.3	66.4	35.3		11	4.5	14.2	16.6	11.6	4.4		11	3.1	3.9	4.6	3.8	0.5	
	III	10	10.2	39.3	125.0	58.5	43.5		10	−10.8	1.7	2.8	−1.2	5.1		10	2.8	4.0	4.9	3.9	0.6	
Nekala	I	22	5.5	53.6	130.2	51.1	34.6	0.157	22	−13.7	2.6	20.1	3.2	9.5	0.123	22	2.2	3.2	4.0	3.2	0.5	0.163
	II	11	12.0	67.7	130.2	66.2	34.5		11	3.5	9.1	20.1	10.6	5.6		11	2.2	3.1	4.0	3.2	0.6	
	III	11	5.5	24.8	90.2	36.0	28.8		11	−13.7	−3.4	1.7	−4.2	6.0		11	2.3	3.3	3.9	3.2	0.5	
Hämeenpuisto	I	22	5.5	53.6	130.2	51.1	34.6	0.157	22	−13.7	2.6	20.1	3.2	9.5	0.123	22	2.2	3.2	4.0	3.2	0.5	0.163
	II	11	12.0	67.7	130.2	66.2	34.5		11	3.5	9.1	20.1	10.6	5.6		11	2.2	3.1	4.0	3.2	0.6	
	III	11	5.5	24.8	90.2	36.0	28.8		11	−13.7	−3.4	1.7	−4.2	6.0		11	2.3	3.3	3.9	3.2	0.5	
Kaukovainio	I	29	0.8	35.1	78.7	40.6	22.3	0.113	29	−16.2	0.0	17.7	1.5	9.6	0.111	29	2.4	3.8	4.9	3.8	0.6	0.079
	II	11	0.8	51.1	78.7	53.0	25.5		11	7.4	12.4	17.7	11.7	3.6		11	3.1	3.8	4.8	3.8	0.5	
	III	18	9.2	32.5	70.0	33.0	16.6		18	−16.2	−2.3	2.1	−4.8	5.9		18	2.4	3.8	4.9	3.8	0.6	
Kajaani	I	26	0.1	39.3	121.4	41.0	28.4	0.144	26	−19.3	−0.5	15.3	1.0	10.2	0.114	26	2.2	3.5	5.7	3.5	0.9	0.126
	II	10	0.1	48.5	121.4	56.2	35.4		10	6.7	12.3	15.3	11.6	3.2		10	2.4	3.2	5.5	3.4	0.9	
	III	16	3.5	28.1	72.5	31.4	18.5		16	−19.3	−2.5	1.0	−5.7	6.7		16	2.2	3.5	5.7	3.5	0.9	
All catchments (Nekala excluded)	I	136	0.1	43.5	142.3	52.0	34.4	0.13	136	−19.3	2.2	20.1	3.5	9.2	0.10	136	2.2	3.7	5.7	3.7	0.6	0.09

a: critical values of Kolmogorov-Smirnov test statistic:

n	=	19	21	22	26	29	136
$D_{critical}$	=	0.301	0.287	0.281	0.259	0.246	0.12

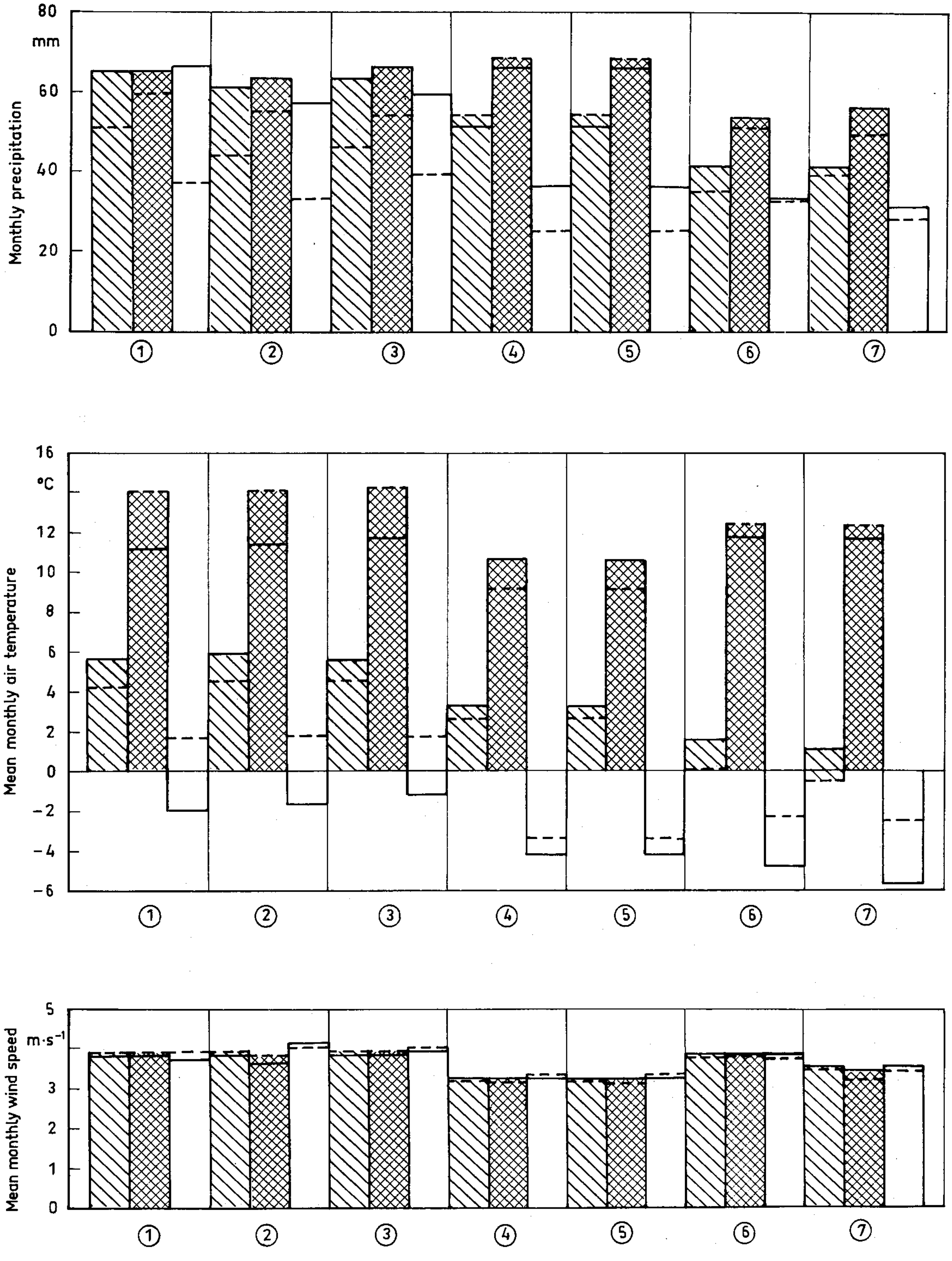

Fig. 13. Averages of hydrometeorological variables studied. Bar on left represents hydrological year, bar in middle represents vegetation period, bar on right represents non-vegetation period. Solid line shows arithmetic mean, dotted line shows median value. 1 = Kontula, 2 = Pakila, 3 = Herttoniemi, 4 = Nekala, 5 = Hämeenpuisto, 6 = Kaukovainio, 7 = Kajaani centre.

Table 25. Properties of distributions of deposition parameters. Season: I = hydrological year, II = vegetation period, III = non-vegetation period. n = number of observations, min = minimum value, m = median value, max = maximum value, $\bar{x}$ = arithmetic mean, s = standard deviation, D = Kolmogorov-Smirnov test statistic. Value of D underlined shows that hypothesis on normal distribution of variable has to be rejected at 95 % significance level in two-sided testing[a].

Catchment	Season	Values of statistics																				
		total deposition tot D mg m^{-2} in 30 days							volatile deposition vol D mg m^{-2} in 30 days							total organic carbon TOC mg m^{-2} C in 30 days						
		n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D
Kontula	I	19	710	2 340	13 300	3 350	3 400	0.315	16	220	800	4 800	1 350	1 190	0.237	16	41	200	579	246	166	0.232
	II	11	1 310	1 970	13 300	4 010	4 300		9	220	1 630	4 800	1 750	1 430		9	112	227	579	319	182	
	III	8	710	2 590	4 940	2 430	1 290		7	390	730	2 020	840	540		7	41	143	274	151	81	
Pakila	I	19	1 010	2 090	16 000	2 920	3 300	0.317	16	150	930	6 040	1 320	1 390	0.248	16	41	213	899	258	205	0.221
	II	11	1 010	2 090	16 000	3 330	4 290		9	150	1 380	6 040	1 700	1 730		9	134	268	899	341	230	
	III	8	1 100	2 390	4 060	2 350	1 040		7	180	700	2 000	820	560		7	41	138	344	152	105	
Herttoniemi	I	19	1 010	2 860	20 100	3 730	4 130	0.314	18	270	1 360	11 700	2 020	2 550	0.270	17	42	267	802	351	252	0.160
	II	10	1 490	3 180	20 100	4 710	5 520		10	620	1 760	11 700	2 820	3 230		9	221	505	802	522	244	
	III	9	1 010	2 430	5 140	2 640	1 220		8	270	900	2 070	1 030	560		8	42	143	308	158	88	
Nekala	I	16	210	1 790	2 580	1 660	670	0.154	12	90	720	1 520	690	340	0.256	17	23	80	218	91	45	0.186
	II	9	210	1 940	2 580	1 720	690		8	90	720	1 520	730	400		11	23	87	218	100	52	
	III	7	760	1 540	2 450	1 570	680		4	290	670	810	610	230		6	47	74	107	75	19	
Hämeenpuisto	I	20	1 320	2 800	12 300	3 860	2 580	0.198	14	400	1 210	2 380	1 250	570	0.160	17	71	150	464	184	113	0.196
	II	11	1 920	2 960	6 880	3 570	1 640		9	750	1 400	2 380	1 440	560		11	87	174	464	222	121	
	III	9	1 320	2 630	12 300	4 220	3 490		5	400	900	1 580	890	460		6	71	100	204	115	47	
Kaukovainio	I	27	500	1 180	7 580	1 620	1 410	0.214	25	70	690	1 900	760	460	0.124	24	47	96	267	124	68	0.267
	II	10	650	2 080	3 000	1 810	770		10	370	990	1 530	1 000	380		10	71	108	266	144	76	
	III	17	500	870	7 580	1 510	1 700		15	70	570	1 900	600	440		14	47	87	267	110	61	
Kajaani	I	26	1 320	2 850	7 060	3 370	1 810	0.183	24	370	880	1 890	990	410	0.150	22	52	136	414	161	88	0.161
	II	10	1 610	3 000	5 590	3 290	1 500		10	450	1 150	1 890	1 140	470		9	90	145	414	185	105	
	III	16	1 320	2 850	7 060	3 420	2 030		14	370	780	1 520	880	330		13	52	122	302	143	73	
All catchments (Nekala excluded)	I	130	500	2 330	20 100	3 060	2 860	0.22	113	70	920	11 700	1 230	1 310	0.20	112	41	155	899	211	169	0.18

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a: critical values of Kolmogorov-Smirnov test statistic:

n =	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	90	91	112	113	127-136
$D_{critical}$ =	0.375	0.361	0.349	0.338	0.327	0.318	0.309	0.301	0.294	0.287	0.281	0.275	0.269	0.264	0.259	0.254	0.250	0.246	0.14	0.14	0.13	0.13	0.12

Table 25. Continued.

Catchment	Season	Values of statistics																				
		total phosphorus tot P mg m^{-2} P in 30 days								total nitrogen tot N mg m^{-2} N in 30 days							chloride Cl mg m^{-2} Cl in 30 days					
		n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D
Kontula	I	19	0.3	2.4	11.2	3.3	3.0	0.253	19	37	69	346	91	74	0.267	19	17	43	195	59	45	0.193
	II	11	0.9	2.8	11.2	4.3	3.5		11	39	67	114	66	23		11	17	43	104	48	27	
	III	8	0.3	2.3	3.6	2.0	1.1		8	37	100	346	126	105		8	22	54	195	75	61	
Pakila	I	19	0.2	3.0	12.5	3.6	2.7	0.211	19	34	62	219	86	57	0.272	19	12	45	191	57	45	0.202
	II	11	1.4	4.6	12.5	4.8	3.0		11	38	54	219	69	52		11	12	34	83	40	21	
	III	8	0.2	2.1	3.4	2.0	1.1		8	34	86	189	108	60		8	17	69	191	81	60	
Herttoniemi	I	20	0.8	2.3	28.6	5.9	7.0	0.278	21	22	56	132	70	37	0.252	21	16	45	160	65	40	0.218
	II	11	1.7	8.8	28.6	9.0	8.1		11	26	56	116	67	36		11	16	45	121	63	37	
	III	9	0.8	1.5	7.0	2.2	1.9		10	22	56	132	73	41		10	20	52	160	68	45	
Nekala	I	19	0.6	1.8	4.6	2.1	1.2	0.185	18	12	45	85	48	21	0.175	22	2	16	100	20	19	0.341
	II	9	0.6	2.1	4.6	2.2	1.2		9	12	44	85	50	25		11	2	16	100	24	26	
	III	10	0.7	1.5	4.5	2.0	1.3		9	25	45	74	46	18		11	7	17	21	16	5	
Hämeenpuisto	I	22	0.4	2.6	14.3	3.7	2.9	0.215	20	29	63	99	64	23	0.145	22	16	35	116	45	28	0.223
	II	11	1.6	3.4	14.3	4.6	3.6		11	35	69	98	69	23		11	16	31	116	50	37	
	III	11	0.4	2.4	6.6	2.8	1.8		9	29	63	99	59	24		11	21	37	61	39	12	
Kaukovainio	I	29	0.6	2.1	9.9	2.5	2.1	0.215	28	8	47	86	46	18	0.193	26	2	12	31	14	7	0.170
	II	11	0.9	2.4	9.9	3.5	2.9		11	8	48	86	54	23		11	2	12	31	13	8	
	III	18	0.6	1.8	3.5	1.8	0.9		17	15	42	61	41	12		15	8	15	27	15	6	
Kajaani	I	26	1.0	2.6	9.4	3.3	2.2	0.178	26	19	38	60	39	12	0.103	22	6	18	27	18	6	0.147
	II	10	1.2	2.5	5.7	2.7	1.4		10	19	39	56	38	12		9	6	17	27	18	7	
	III	16	1.0	2.9	9.4	3.7	2.6		16	24	36	60	39	12		13	12	18	27	19	5	
All catchments (Nekala excluded)	I	135	0.2	2.4	28.6	3.6	3.7	0.24	133	8	50	346	63	44	0.19	129	2	28	195	41	37	0.19

Table 25. Continued.

Catchment	Season	Values of statistics																				
		calcium Ca mg m^{-2} Ca in 30 days							sulphate SO_4 mg m^{-2} SO_4 in 30 days							vanadium[b] V mg m^{-2} V in 30 days						
		n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D
Kontula	I	19	33	61	141	70	33	0.205	19	175	475	1 090	475	208	0.190	18	0.10	0.27	0.94	0.36	0.26	0.177
	II	11	33	53	108	57	22		11	259	480	613	448	110		11	0.10	0.16	0.86	0.26	0.23	
	III	8	40	87	141	88	40		8	175	441	1 090	513	303		7	0.23	0.42	0.94	0.51	0.25	
Pakila	I	19	20	47	132	62	34	0.251	19	180	364	1 010	424	201	0.150	18	0.05	0.21	1.02	0.30	0.24	0.203
	II	11	20	39	90	44	19		11	229	364	514	371	96		11	0.05	0.16	0.54	0.18	0.14	
	III	8	44	93	132	87	36		8	180	436	1 010	497	282		7	0.21	0.46	1.02	0.49	0.26	
Herttoniemi	I	21	42	87	152	86	32	0.140	21	187	588	1 077	591	241	0.138	20	0.12	0.43	1.61	0.57	0.40	0.179
	II	11	48	72	120	76	24		11	343	588	973	599	198		11	0.12	0.35	0.55	0.32	0.15	
	III	10	42	105	152	98	36		10	187	569	1 077	582	292		9	0.41	0.78	1.61	0.88	0.39	
Nekala	I	22	7	52	109	50	22	0.115	21	55	330	582	322	140	0.132	22	0.03	0.43	1.27	0.46	0.26	0.147
	II	11	7	55	74	50	18		11	55	411	582	381	137		11	0.03	0.38	0.61	0.34	0.17	
	III	11	18	51	109	50	27		10	70	284	413	258	117		11	0.19	0.57	1.27	0.59	0.28	
Hämeenpuisto	I	22	24	58	123	63	26	0.129	21	128	460	945	459	212	0.069	22	0.12	0.96	2.68	1.12	0.65	0.136
	II	11	24	57	81	55	17		11	335	522	945	558	172		11	0.12	0.66	1.64	0.74	0.45	
	III	11	26	66	123	70	33		10	128	333	747	350	205		11	0.44	1.63	2.68	1.50	0.60	
Kaukovainio	I	25	8	21	50	23	11	0.152	27	89	218	995	252	170	0.239	26	0.04	0.19	0.46	0.22	0.13	0.167
	II	10	8	17	50	22	14		11	178	313	995	358	219		11	0.04	0.13	0.42	0.16	0.11	
	III	15	11	22	40	25	9		16	89	169	337	180	68		15	0.11	0.23	0.46	0.26	0.13	
Kajaani	I	23	27	97	704	148	147	0.238	23	157	516	1 230	586	286	0.173	23	0.10	0.51	1.15	0.55	0.30	0.111
	II	8	27	99	338	114	98		9	350	553	1 230	599	302		9	0.10	0.18	0.79	0.33	0.24	
	III	15	44	97	704	166	168		14	157	500	1 220	577	287		14	0.31	0.69	1.15	0.70	0.24	
All catchments (Nekala excluded)	I	129	8	56	704	75	77	0.19	130	89	410	1 230	457	250	0.10	127	0.04	0.41	2.68	0.52	0.47	0.17

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b: concentration of vanadium is at detection limit (<5 $\mu g\ l^{-1}$ V) in 0 to 17 % of samples in individual catchments

Table 25. Continued.

Catchment	Season	Values of statistics																				
		cadmium^c Cd mg m^{-2} Cd in 30 days							titanium Ti mg m^{-2} Ti in 30 days							zinc Zn mg m^{-2} Zn in 30 days						
		n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D
Kontula	I	15	0.00	-	0.08	-	-	-	4	0.46	0.88	2.72	1.24	1.01	-	13	0.87	1.67	14.27	2.64	3.53	0.412
	II	8	0.01	-	0.05	-	-		2	0.46	-	0.88	0.67	-		8	0.92	1.80	14.27	3.22	4.49	
	III	7	0.00	-	0.08	-	-		2	0.88	-	2.72	1.80	-		5	0.87	1.67	2.74	1.72	0.68	
Pakila	I	15	0.00	-	0.08	-	-	-	4	0.27	1.45	1.83	1.16	0.67	-	13	0.06	1.12	2.71	1.34	0.74	0.157
	II	8	0.02	-	0.07	-	-		2	0.27	-	1.07	0.67	-		8	0.06	1.21	2.71	1.34	0.77	
	III	7	0.00	-	0.08	-	-		2	1.45	-	1.83	1.64	-		5	0.78	0.98	2.61	1.34	0.77	
Herttoniemi	I	18	0.00	-	0.06	-	-	-	3	0.38	1.62	1.70	1.23	0.74	-	16	0.80	4.15	10.39	5.12	2.79	0.197
	II	9	0.01	-	0.06	-	-		1	-	-	-	0.38	-		9	2.46	4.88	10.39	5.67	2.83	
	III	9	0.00	-	0.06	-	-		2	1.62	-	1.70	1.66	-		7	0.80	3.90	9.32	4.40	2.77	
Nekala	I	19	0.01	-	0.15	-	-	-	3	0.94	1.35	3.29	1.86	1.26	-	16	0.20	1.17	4.39	1.54	1.15	0.185
	II	9	0.01	-	0.14	-	-		2	0.94	-	3.29	2.12	-		9	0.20	1.16	4.39	1.74	1.47	
	III	10	0.01	-	0.15	-	-		1	-	-	-	1.35	-		7	0.69	1.18	2.13	1.29	0.54	
Hämeenpuisto	I	19	0.01	-	0.15	-	-	-	4	1.51	5.63	8.08	4.22	3.20	-	16	1.09	2.28	3.48	2.21	0.88	0.223
	II	9	0.01	-	0.04	-	-		2	1.51	-	5.63	3.57	-		9	1.09	2.94	3.48	2.46	0.93	
	III	10	0.01	-	0.15	-	-		2	1.66	-	8.08	4.87	-		7	1.10	1.56	2.95	1.89	0.76	
Kaukovainio	I	26	0.00	-	3.13	-	-	-	4	0.23	0.57	0.79	0.51	0.24	-	17	0.21	1.11	2.50	1.09	0.50	0.184
	II	11	0.00	-	3.13	-	-		1	-	-	-	0.23	-		7	0.69	1.11	1.39	1.04	0.24	
	III	15	0.00	-	0.21	-	-		3	0.44	0.57	0.79	0.60	0.18		10	0.21	1.09	2.50	1.13	0.63	
Kajaani	I	23	0.00	-	0.14	-	-	-	3	1.08	1.25	4.55	2.29	1.96	-	16	0.63	1.88	8.86	2.30	1.99	0.308
	II	9	0.01	-	0.05	-	-		0	-	-	-	-	-		6	0.63	2.13	8.86	3.21	3.11	
	III	14	0.00	-	0.14	-	-		3	1.08	1.25	4.55	2.29	1.96		10	0.95	1.66	2.78	1.76	0.60	
All catchments (Nekala excluded)	I	116	0.00	-	3.13	-	-	-	22	0.23	-	8.08	1.78	1.94	-	91	0.06	1.57	14.27	2.47	2.38	0.21

c: concentration of cadmium is at detection limit (<1 μg l^{-1} Cd) in 77 to 100 % of samples in individual catchments

Table 25. Continued.

Catchment	Season	Values of statistics																				
		copper Cu mg m^{-2} Cu in 30 days							lead Pb mg m^{-2} Pb in 30 days							pH						
		n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D	n	min	m	max	$\bar{x}$	s	D
Kontula	I	13	0.26	0.39	0.67	0.42	0.12	0.195	19	0.27	1.43	2.28	1.37	0.56	0.162	19	3.7	4.1	7.9	-	-	0.300
	II	8	0.26	0.39	0.67	0.43	0.13		11	0.65	0.98	1.89	1.14	0.37		11	4.0	4.1	7.9	-	-	
	III	5	0.28	0.40	0.55	0.42	0.10		8	0.27	1.88	2.28	1.70	0.63		8	3.7	4.0	4.7	-	-	
Pakila	I	13	0.15	0.39	0.93	0.42	0.22	0.220	19	0.24	1.03	2.53	1.31	0.70	0.180	19	3.9	4.3	6.6	-	-	0.251
	II	8	0.15	0.30	0.49	0.32	0.12		11	0.46	0.90	1.39	0.92	0.30		11	4.0	4.3	6.6	-	-	
	III	5	0.32	0.46	0.93	0.57	0.26		8	0.24	2.04	2.53	1.84	0.76		8	3.9	4.0	4.6	-	-	
Herttoniemi	I	16	0.26	0.45	0.69	0.43	0.11	0.109	21	0.77	2.07	4.67	2.22	0.89	0.170	21	3.4	4.0	5.2	-	-	0.190
	II	9	0.26	0.40	0.69	0.41	0.14		11	0.77	1.97	2.65	1.93	0.63		11	3.8	4.1	5.2	-	-	
	III	7	0.35	0.45	0.54	0.46	0.07		10	1.29	2.12	4.67	2.53	1.06		10	3.4	3.9	4.9	-	-	
Nekala	I	16	0.04	0.45	2.19	0.53	0.48	0.294	22	0.15	1.28	2.24	1.28	0.44	0.098	22	3.6	4.1	5.6	-	-	0.206
	II	9	0.04	0.46	2.19	0.61	0.63		11	0.15	1.38	1.73	1.26	0.46		11	3.6	4.1	4.9	-	-	
	III	7	0.20	0.44	0.66	0.43	0.14		11	0.63	1.15	2.24	1.30	0.45		11	3.7	4.3	5.6	-	-	
Hämeenpuisto	I	16	0.18	0.57	1.13	0.62	0.25	0.158	22	0.75	4.46	6.98	4.28	1.75	0.098	22	3.6	4.1	5.1	-	-	0.236
	II	9	0.38	0.55	1.13	0.68	0.26		11	0.75	4.51	6.98	4.47	2.07		11	3.7	4.0	4.8	-	-	
	III	7	0.18	0.58	0.88	0.55	0.24		11	1.01	4.05	6.17	4.09	1.44		11	3.6	4.2	5.1	-	-	
Kaukovainio	I	17	0.10	0.85	1.77	0.84	0.51	0.133	27	0.37	0.79	1.55	0.81	0.24	0.141	29	3.3	4.4	6.9	-	-	0.191
	II	7	0.30	0.79	1.38	0.76	0.35		11	0.37	0.74	0.99	0.72	0.18		11	3.3	4.3	5.8	-	-	
	III	10	0.10	0.89	1.77	0.89	0.61		16	0.56	0.83	1.55	0.87	0.26		18	3.3	4.4	6.9	-	-	
Kajaani	I	15	0.42	1.16	3.14	1.37	0.76	0.202	24	0.96	1.92	3.01	1.96	0.55	0.112	26	2.9	4.6	7.5	-	-	0.148
	II	6	0.49	1.26	2.67	1.40	0.78		9	0.96	1.91	2.45	1.84	0.45		10	3.8	4.2	5.9	-	-	
	III	9	0.42	1.16	3.14	1.35	0.80		15	1.24	2.02	3.01	2.03	0.62		16	2.9	5.2	7.5	-	-	
All catchments (Nekala excluded)	I	90	0.10	0.51	3.14	0.69	0.52	<u>0.20</u>	132	0.24	1.64	6.98	1.97	1.44	<u>0.15</u>	136	2.9	4.2	7.9	-	-	<u>0.19</u>

Table 25. Continued.

Catchment	Season	Values of statistics						
			conductivity γ_{25} mS m^{-1}					
		n	min	m	max	$\bar{x}$	s	D
Kontula	I	19	2.4	4.5	10.3	5.5	2.2	0.205
	II	11	3.3	5.4	10.3	5.8	2.2	
	III	8	2.4	4.1	8.6	5.1	2.3	
Pakila	I	19	2.5	3.9	9.2	4.4	1.6	0.273
	II	11	3.0	3.6	6.0	4.0	0.9	
	III	8	2.5	4.0	9.2	4.9	2.3	
Herttoniemi	I	21	2.7	6.7	15.6	7.6	3.2	0.208
	II	11	5.2	6.4	9.2	6.8	1.4	
	III	10	2.7	7.1	15.6	8.5	4.3	
Nekala	I	22	1.9	5.6	19.0	6.8	4.2	0.194
	II	11	4.1	6.8	19.0	7.7	4.3	
	III	11	1.9	4.4	14.9	5.9	4.1	
Hämeenpuisto	I	22	2.8	6.1	16.0	6.9	3.4	0.189
	II	11	4.9	6.8	16.0	7.8	3.2	
	III	11	2.8	5.3	13.8	6.0	3.5	
Kaukovainio	I	29	1.2	3.8	9.8	4.2	2.5	0.180
	II	11	3.0	4.5	9.8	5.8	2.5	
	III	18	1.2	2.5	8.2	3.2	1.9	
Kajaani	I	25	1.8	6.4	12.0	6.5	2.4	0.098
	II	10	5.1	7.4	12.0	7.4	2.0	
	III	15	1.8	5.4	11.2	5.9	2.6	
All catchments (Nekala excluded)	I	135	1.2	5.4	16.0	5.8	2.9	0.08

27. In the same Table, also the deposition measured by the actual gauges of the catchment over the same period is given. As can be seen from the figures, only conductivity is higher in the samples measured on the actual gauges, their deposition rates in general are distinctly lower than those of the reference point. These results and the emission data yield the conclusion that the deposition gauges in the Nekala catchment — located on roofs — were exposed to wind to such an extent that the deposition data of the catchment are not representative (the gauges were prohibited from being located at the points originally chosen because of the objection of the real estate owners). Therefore, the Nekala data were excluded from the analyses of combined deposition observations.

6.22 Deposition parameters correlating with hydrometeorological variables and with each other

A summary on the correlation of the deposition parameters with the hydrometeorological variables, and on their mutual correlation is given in Table 28 in which the statistically significant correlations (significance level ≥ 95 %) are shown (Nekala excluded).

Precipitation has a positive correlation with the deposition of total nitrogen and sulphate in more than half of the six test catchments, thus confirming the dependence of these depositions in particular on the amount of precipitation. In half of the catchments the deposition of organic carbon correlates positively with air temperature showing increasing deposition due to vegetation in the summertime. In five of the six catchments, the deposition of vanadium correlates negatively with the air temperature, thus showing a growth in the vanadium deposition during the winter period.

As to the intercorrelation between the deposition parameters, the following parameters have a positive correlation in at least half of the catchments: total and volatile depositions (including the non-normally distributed total deposition of Kontula, Pakila and Herttoniemi), total deposition and total phosphorus (as above), total organic carbon and total phosphorus, total nitrogen and sulphate, total nitrogen and copper, total

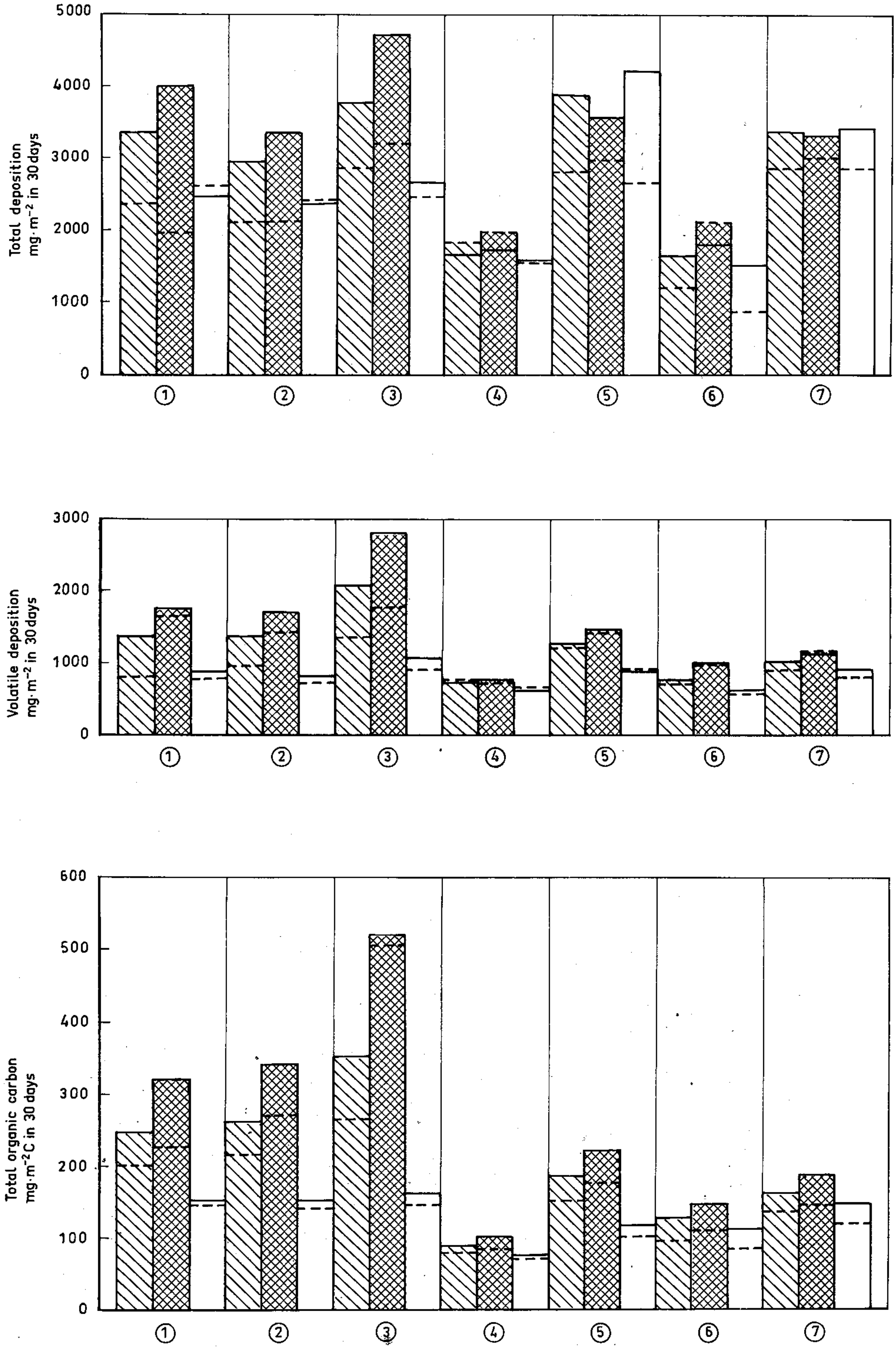


Fig. 14. Averages of deposition parameters studied. Bar on left represents hydrological year, bar in middle represents vegetation period, bar on right represents non-vegetation period. Solid line shows arithmetic mean (in case of pH top of bar shows highest value observed and bottom of bar lowest value observed), dotted line shows median value. 1 = Kontula, 2 = Pakila, 3 = Herttoniemi, 4 = Nekala, 5 = Hämeenpuisto, 6 = Kaukovainio, 7 = Kajaani centre.

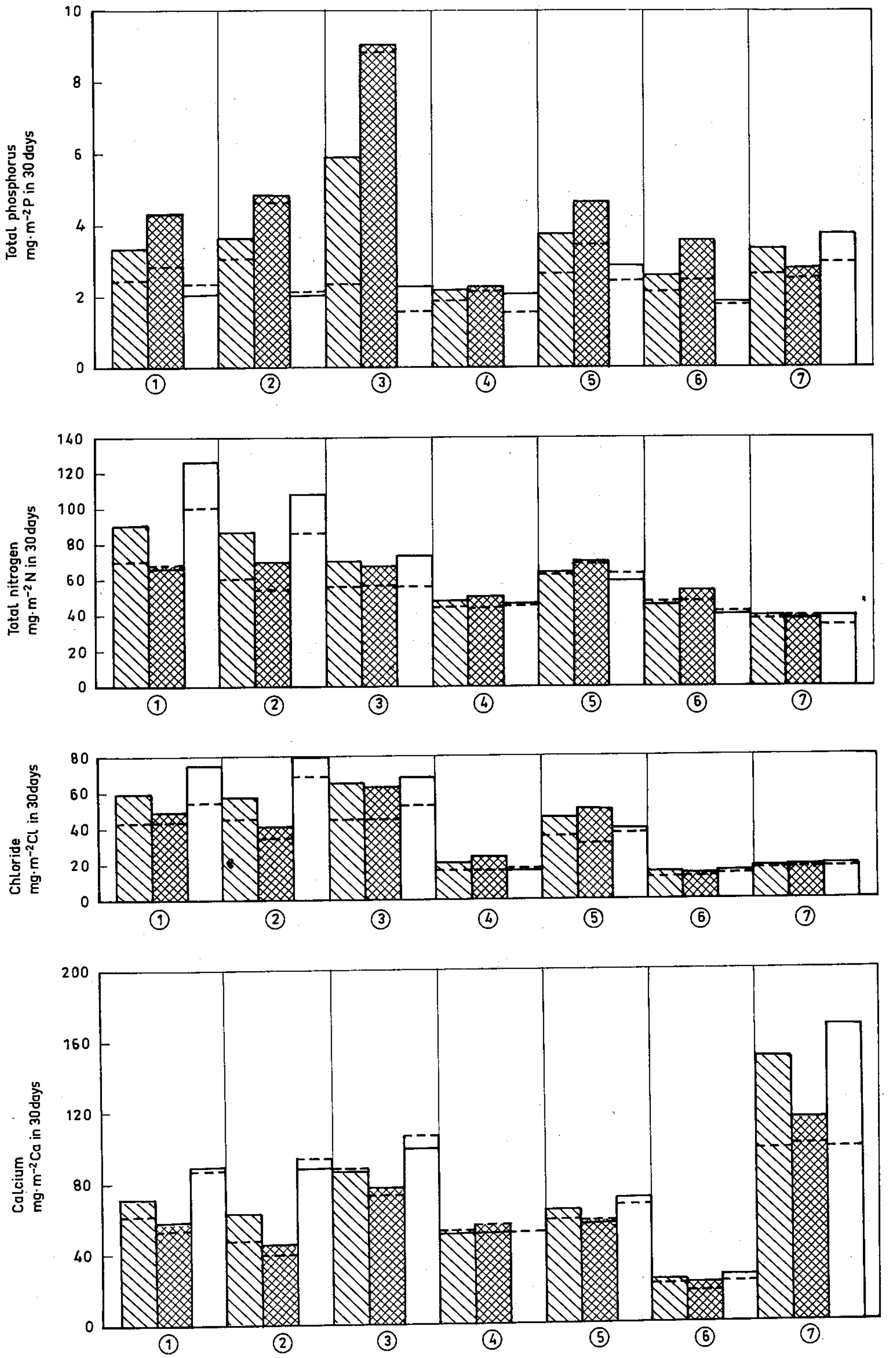
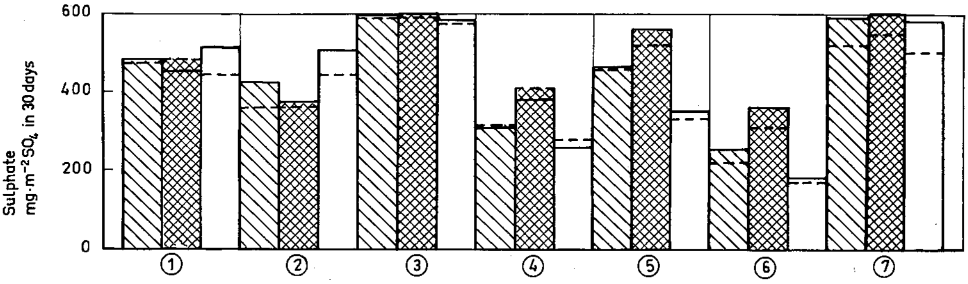
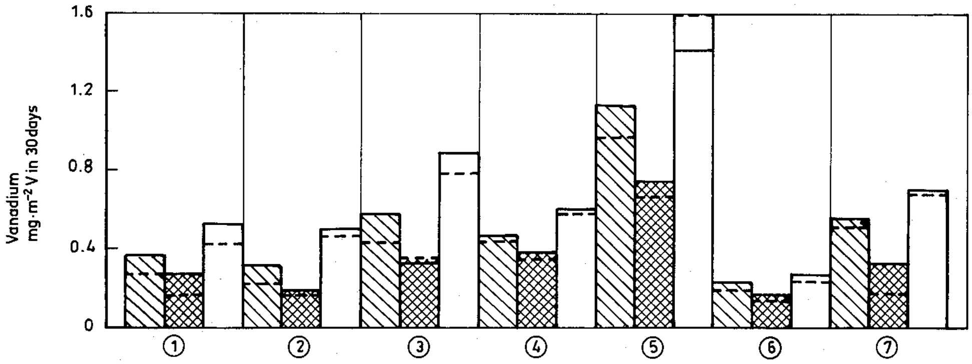
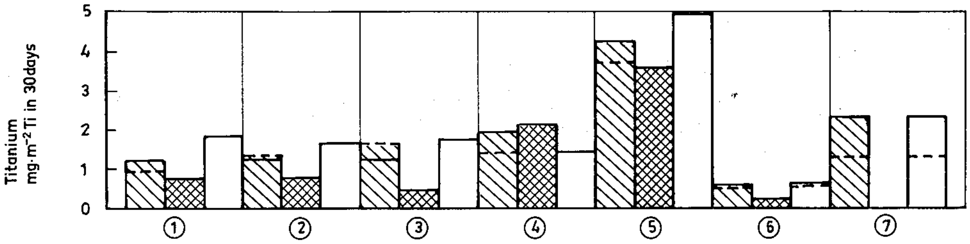


Fig. 14. Continued.







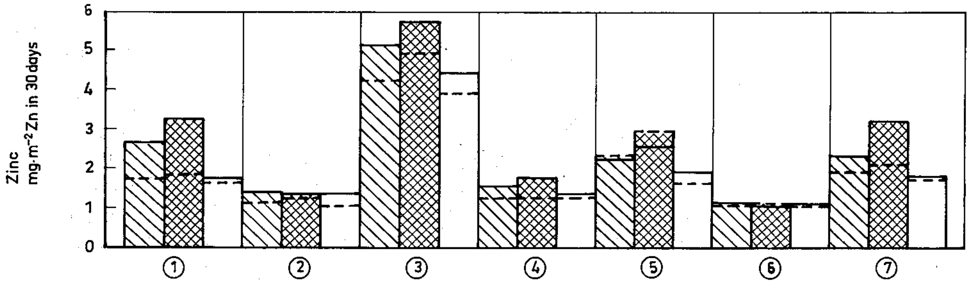


Fig. 14. Continued.

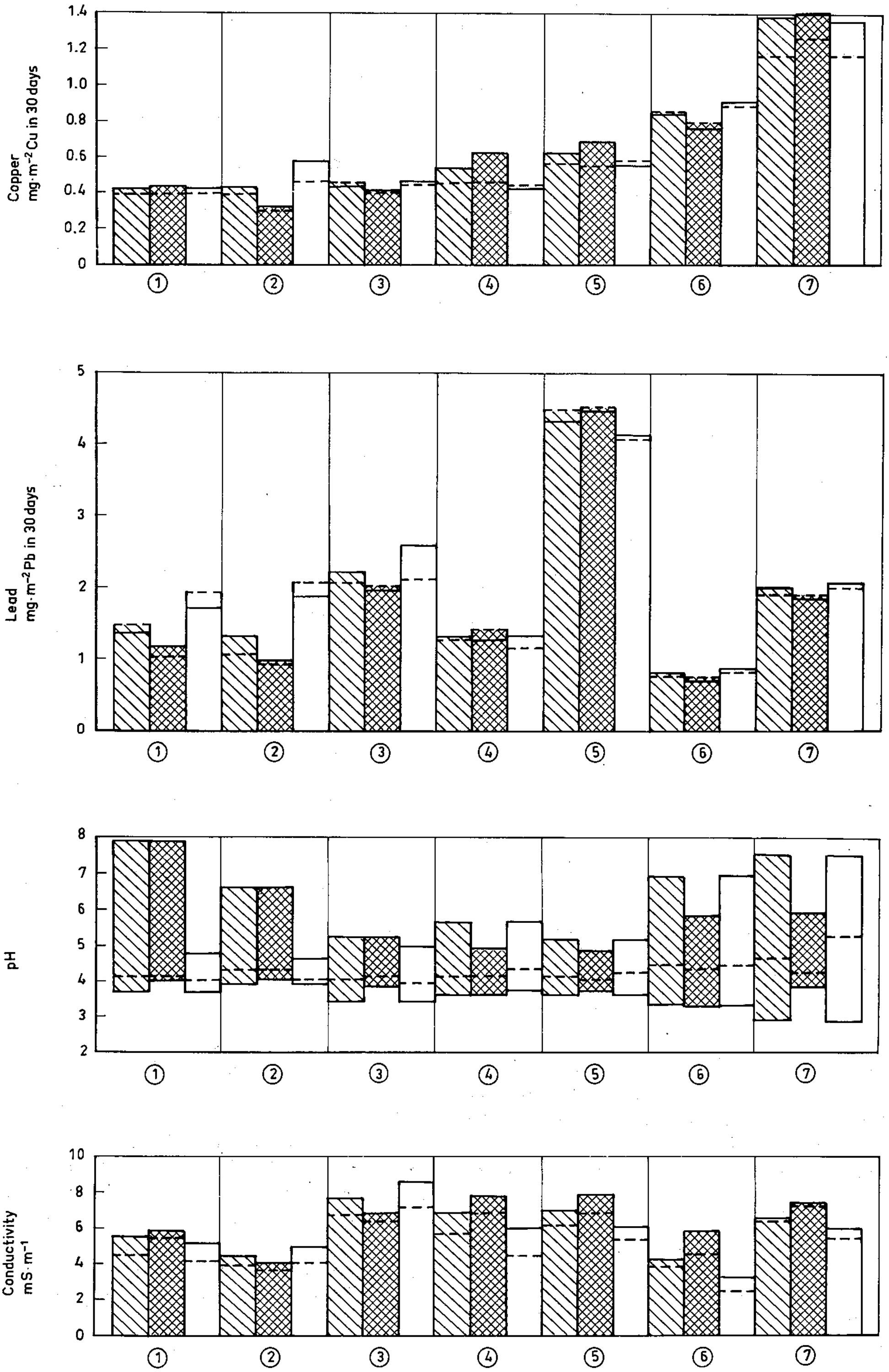


Fig. 14. Continued.

Table 26. Comparison of deposition measured in test catchment of Hämeenpuisto (upper row) and reference point of Hatanpää (lower row) over same period. n = number of observations, min = minimum value, max = maximum value, $\bar{x}$ = arithmetic mean, s = standard deviation.

Parameter and unit	Values of statistics				
	n	min	max	$\bar{x}$	s
Total deposition	20	1 320	12 300	3 860	2 580
mg m^{-2} in 30 days	17	880	5 840	2 120	1 490
Volatile deposition	14	400	2 380	1 250	570
mg m^{-2} in 30 days	12	390	2 320	1 030	570
Total organic carbon	17	71	464	184	113
mg m^{-2} C in 30 days	16	43	318	151	76
Total phosphorus	22	0.4	14.3	3.7	2.9
mg m^{-2} P in 30 days	15	1.0	13.0	3.2	2.9
Total nitrogen	20	29	99	64	23
mg m^{-2} N in 30 days	15	34	105	63	21
Chloride	22	16	116	45	28
mg m^{-2} Cl in 30 days	22	11	153	53	28
Calcium	22	24	123	63	26
mg m^{-2} Ca in 30 days	22	23	95	52	21
Sulphate	21	128	945	459	212
mg m^{-2} SO_4 in 30 days	21	85	615	380	154
Vanadium[a]	22	0.12	2.68	1.12	0.65
mg m^{-2} V in 30 days	22	0.13	2.40	0.81	0.56
Cadmium[b]	19	0.01	0.15	-	-
mg m^{-2} Cd in 30 days	20	0.01	0.16	-	-
Zinc	16	1.09	3.48	2.21	0.88
mg m^{-2} Zn in 30 days	16	0.63	5.25	2.41	1.11
Copper	16	0.18	1.13	0.62	0.25
mg m^{-2} Cu in 30 days	16	0.18	0.92	0.46	0.21
Lead	22	0.75	6.98	4.28	1.75
mg m^{-2} Pb in 30 days	22	0.37	6.15	1.46	1.19
pH value	22	3.6	5.1	-	-
	21	3.4	5.0	-	-
Conductivity	22	2.8	16.0	6.9	3.4
mS m^{-1}	21	2.7	24.0	8.0	5.2

a: concentration of vanadium is at detection limit (<5 $\mu g\ l^{-1}$ V) in one sample
b: concentration of cadmium is at detection limit (<1 $\mu g\ l^{-1}$ Cd) in 80 % of samples

nitrogen and lead, calcium and sulphate, calcium and vanadium, sulphate and lead, and sulphate and conductivity. These correlations especially reveal a growth in the deposition of nitrogen, calcium, vanadium and lead during the non-vegetation period.

6.23 Fraction of variance explained by hydrometeorological variables in individual catchments

Regression models were formed for the deposition parameters by using the hydrometeorological variables as explanatory variables, as was done for the wet and dry deposition in the Herttoniemi catchment.

The explanatory variables in the final models of the various catchments, the fraction of variance explained, and the significance level of coefficients in the regression equations are given in Table 29.

In general, none of the variance of total deposition can be explained in the individual catchments by the hydrometeorological variables used. This is partly due to the inaccuracies in the data that hampers modelling. The main reason is that, though the composition of total deposition varies depending on the season, the magnitude of the deposition rate need not change, which may be seen by comparing the median values of total

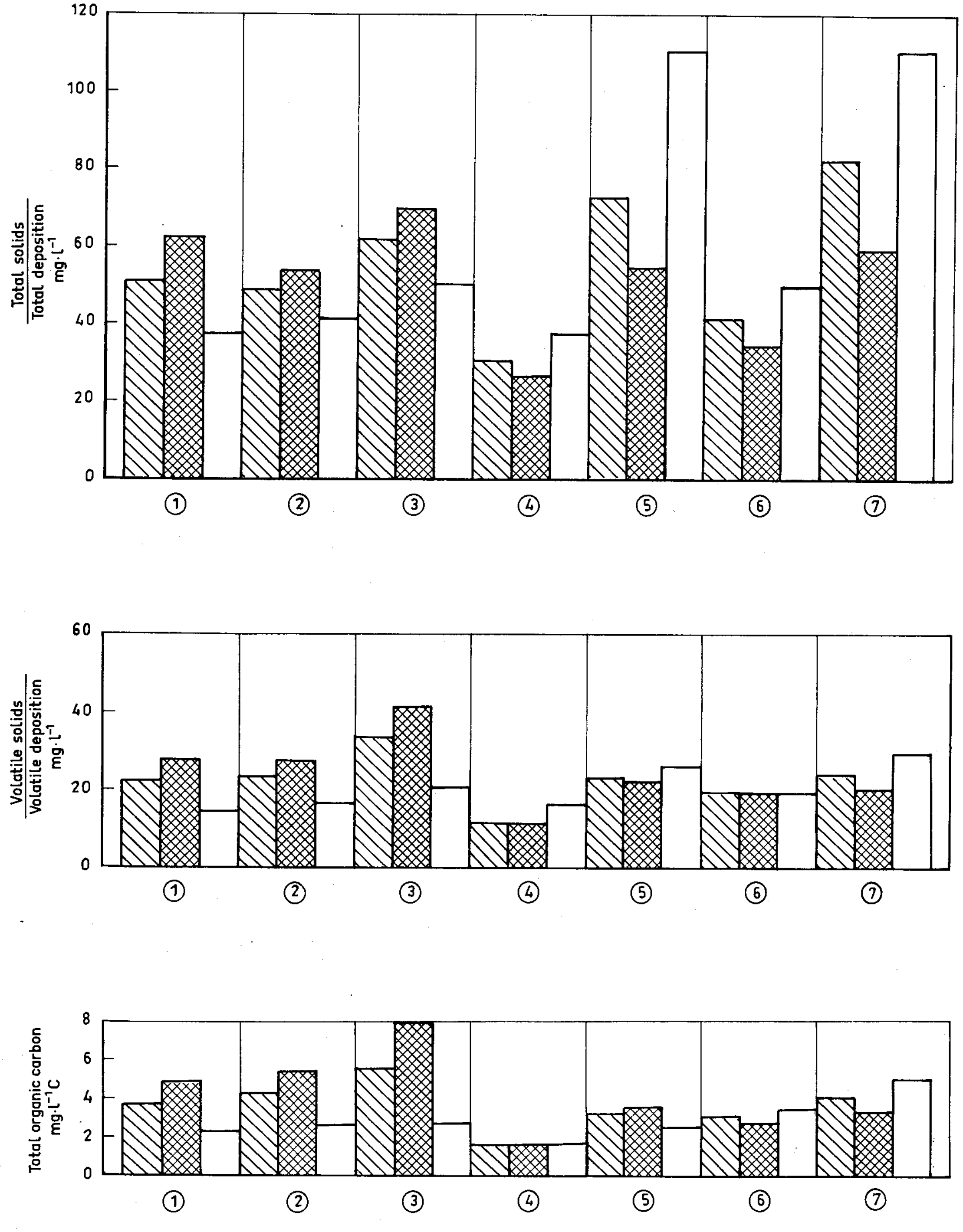
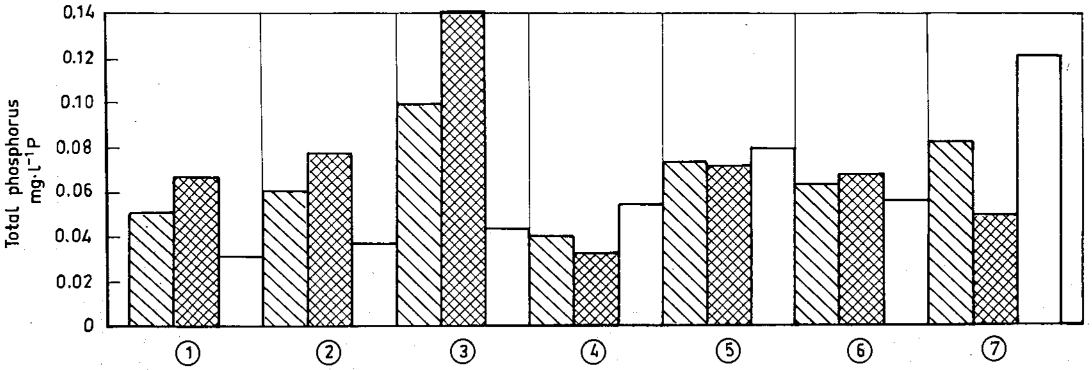
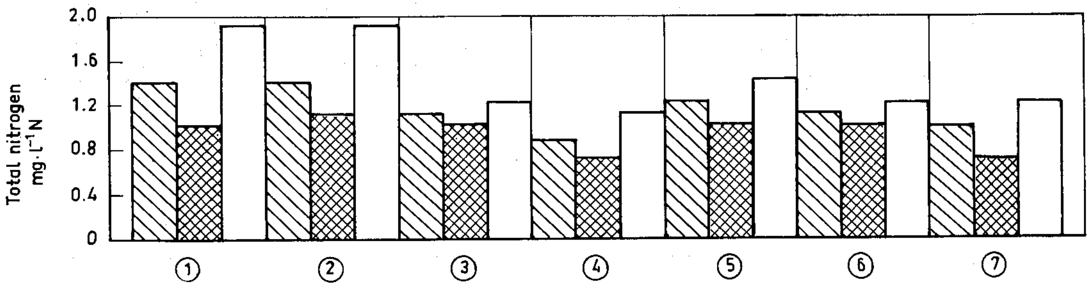
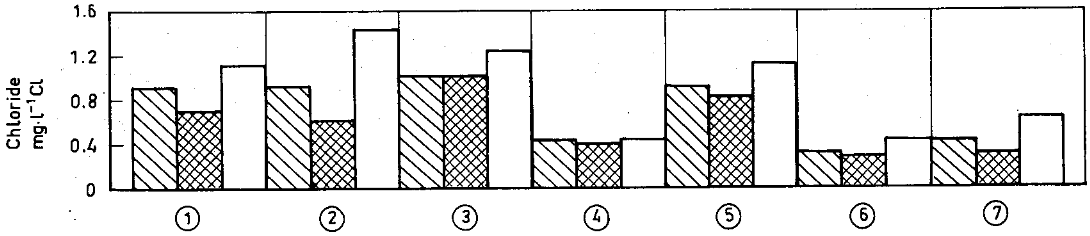
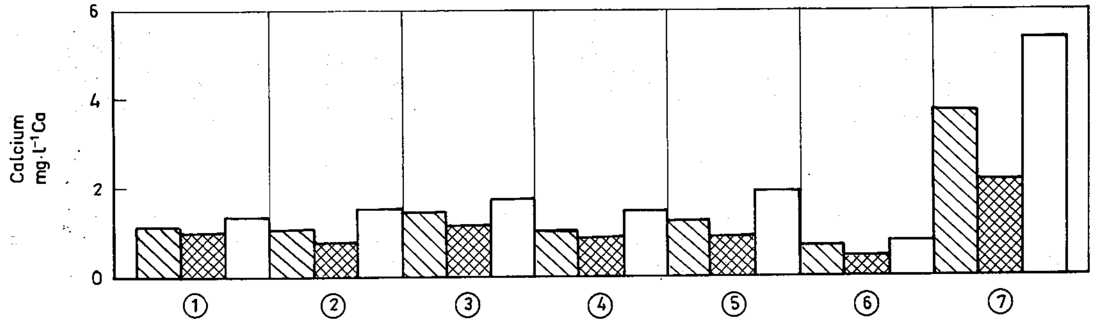


Fig. 15. Average composition of precipitation. Bar on left represents hydrological year, bar in middle represents vegetation period, bar on right represents non-vegetation period. 1 = Kontula, 2 = Pakila, 3 = Herttoniemi, 4 = Nekala, 5 = Hämeenpuisto, 6 = Kaukovainio, 7 = Kajaani centre (pH and conductivity in Fig. 14).









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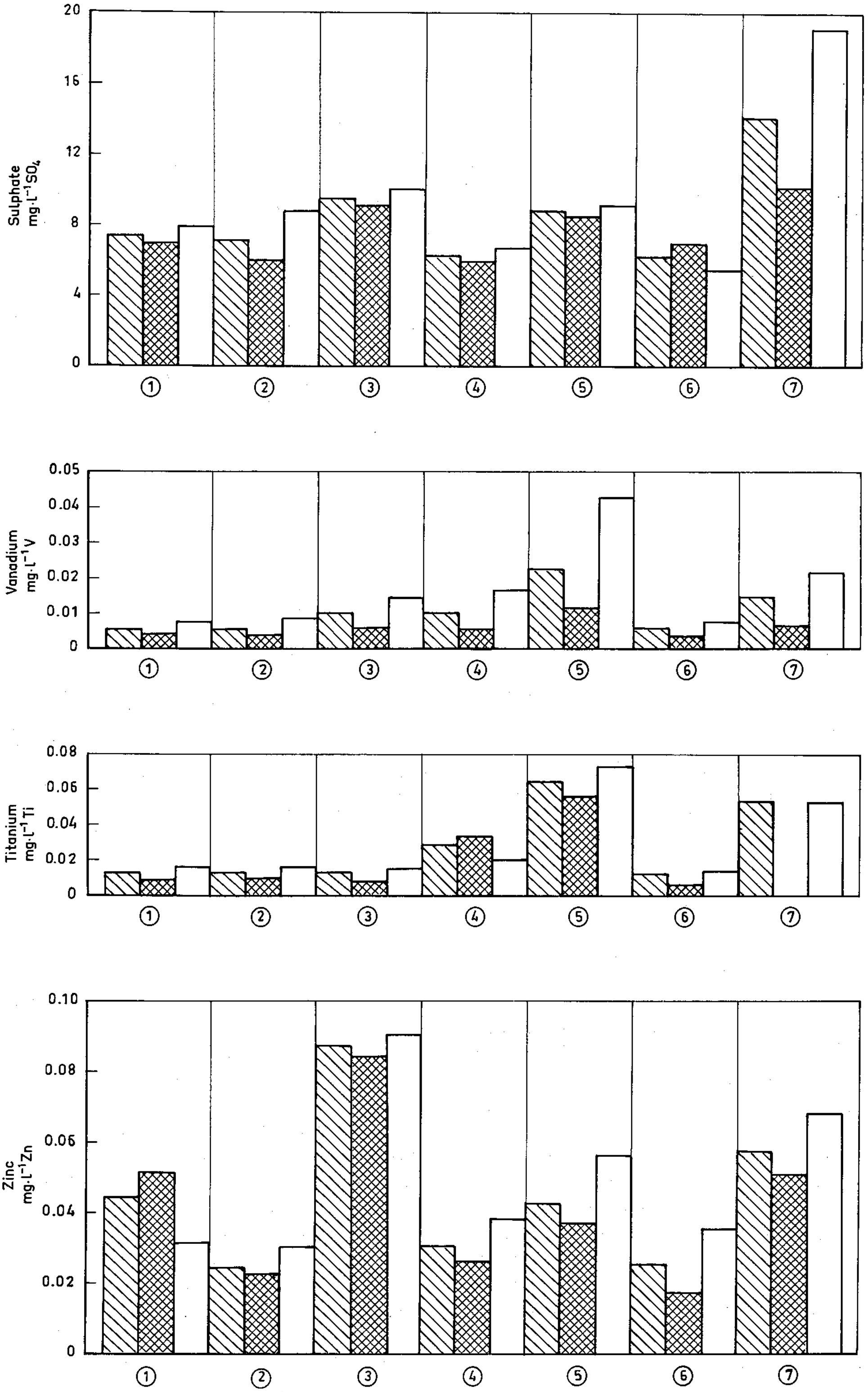

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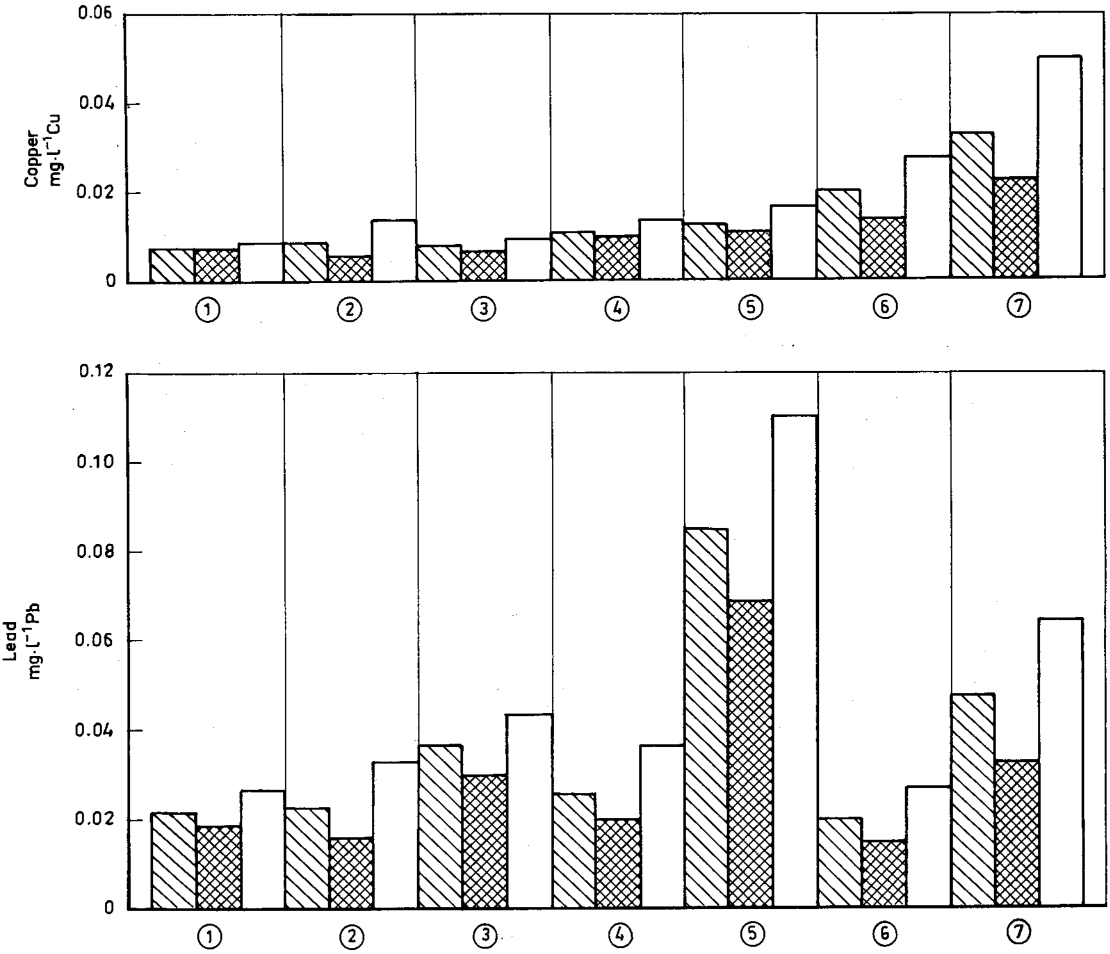

Fig. 15. Continued.

deposition during the vegetation and non-vegetation periods (Table 25, Fig. 14). A statistically significant model is derived for the Kontula catchment where 21 % of the variance of total deposition is explained by temperature. The model derived for the Kajaani centre has no physical explanation and is thus the creation of statistics only.

Air temperature with a positive regression coefficient appears as a significant explanatory variable in the models of volatile deposition in the catchments of Kontula, Herttoniemi and Kaukovainio and explains roughly one fourth of the variance. None of the variance of the volatile deposition can be explained in the catchments of Nekala, Hämeenpuisto and Kajaani centre. The sensitivity of the sampling to the aerodynamic effects may be seen in the catchments of Helsinki where wind speed with a negative regression coefficient appears in the models of volatile deposition.

Roughly one third of the variance of the organic carbon deposition can be explained by air temperature in the test catchments of Helsinki. In the other catchments the models remain non-significant.

The models derived for the deposition of total phosphorus are somewhat contradictory in the individual catchments; in general, the phosphorus deposition cannot be explained by the hydrometeorological variables used.

In five of the catchments of the order of 20—60 % of the variance of the nitrogen deposition can be explained by precipitation. In the city-centre catchments of Hämeenpuisto and Kajaani centre the models of nitrogen remain non-significant.

The deposition of chloride is not well modelled

Table 27. Comparison of deposition measured in test catchment (upper row) and reference point (lower row) of Nekala over same period. n = number of observations, min = minimum value, max = maximum value, $\bar{x}$ = arithmetic mean, s = standard deviation.

Parameter and unit	Values of statistics				
	n	min	max	$\bar{x}$	s
Total deposition	4	1 660	2 050	1 890	170
mg m^{-2} in 30 days	6	2 190	10 600	5 660	3 620
Volatile deposition	4	720	810	750	40
mg m^{-2} in 30 days	6	760	1 340	1 130	220
Total organic carbon	5	59	218	107	64
mg m^{-2} C in 30 days	6	88	299	147	77
Total phosphorus	6	0.8	4.6	2.6	1.7
mg m^{-2} P in 30 days	7	1.4	7.6	4.1	2.3
Total nitrogen	6	30	78	61	20
mg m^{-2} N in 30 days	7	35	106	68	23
Chloride	7	7	100	27	32
mg m^{-2} Cl in 30 days	7	14	51	26	12
Calcium	7	18	74	46	19
mg m^{-2} Ca in 30 days	7	23	140	80	48
Sulphate	7	181	517	347	103
mg m^{-2} SO_4 in 30 days	7	203	697	429	161
Vanadium[a]	7	0.12	0.76	0.50	0.23
mg m^{-2} V in 30 days	7	0.39	1.95	1.06	0.63
Cadmium[b]	7	<0.01	<0.05	-	-
mg m^{-2} Cd in 30 days	7	<0.01	<0.04	-	-
Zinc	7	0.69	3.94	1.43	1.16
mg m^{-2} Zn in 30 days	7	0.83	5.74	2.75	1.51
Copper	7	0.20	0.68	0.36	0.16
mg m^{-2} Cu in 30 days	7	0.32	1.34	0.73	0.34
Lead	7	0.83	1.81	1.31	0.33
mg m^{-2} Pb in 30 days	7	0.91	5.25	2.90	1.59
pH value	7	3.6	4.9	-	-
	7	3.8	4.8	-	-
Conductivity	7	2.9	19.0	8.0	5.8
mS m^{-1}	7	3.1	11.0	6.2	2.8

a: concentration of vanadium is at detection limit (< 5 μg l^{-1} V) in one sample
b: concentration of cadmium is at detection limit (<1 μg l^{-1} Cd) in 80 % of samples

in the individual catchments. Only in Kontula precipitation and air temperature explain more than half of the variance. The models of the chloride deposition remain non-significant in four of the catchments.

A significant model for the calcium deposition was derived only in the Kajaani centre where the calcium emitted from the cellulose factory raises the calcium deposition to a significantly higher level than in the other catchments (Figs. 14 and 15).

Precipitation explains roughly one third of the variance of sulphate deposition in the catchments of Kontula, Pakila and Kaukovainio. The models in Herttoniemi and Kajaani centre are non-significant. In the catchments of Tampere, air temperature with a positive regression coefficient explains roughly one third of the variance of the sulphate deposition.

Air temperature with a negative regression coefficient appears in the models of vanadium deposition in five of the catchments explaining 14—64 % of the variance. A low fraction of variance explained is achieved in the catchments with low emissions of vanadium (Kontula: non-significant model, Kaukovainio: 14 % of the variance explained).

In general, the deposition of zinc and copper cannot be modelled by the hydrometeorological factors in the individual catchments. Yet, in

Table 28. Levels of correlation between hydrometeorological variables and deposition parameters. Nekala catchment excluded. + or − indicates positive or negative correlation at 95 % significance level, ++ or −− indicates correlation at 99 % level, +++ or −−− indicates correlation at 99.9 % level. Number of catchments is given in brackets.

	Monthly precipitation	Mean monthly air temperature	Mean monthly wind speed	Total deposition	Volatile deposition	Total organic carbon	Total phosphorus	Total nitrogen	Chloride	Calcium	Sulphate	Vanadium	Zinc	Copper	Lead	pH	Conductivity
Monthly precipitation																	
Mean monthly air temperature	++... +++ (3)																
Mean monthly wind speed																	
Total deposition		+ (1)															
Volatile deposition		++ (2)	− (1)	+++ (4)													
Total organic carbon		+... ++ (3)		++ (1)	++ (1)												
Total phosphorus		+ (1)		+... +++ (3)	+++ (1)	+... +++ (3)											
Total nitrogen	+... +++ (6)	+ (1)		+ (1)		+ (1)	++ (1)										
Chloride	+... ++ (2)		+ (2)		++ (1)		+ (1)	+++ (1)									
Calcium		− (2)	+ (1)	++ (1)				++ (2)	+... ++ (2)								
Sulphate	+... ++ (4)	++ (2)	++ (1)	+ (1)	++ (1)		+ (1)	++... +++ (5)	+ (2)	+... +++ (4)							
Vanadium	− (1)	−... −−− (5)	+ (1)			− (1)	− (1)	+ (2)		+ (3)	+... ++ (2)						
Zinc	++ (1)	+ (1)		+ (1)	+ (1)	+ (1)	+ (1)	++ (1)			+ (1)						
Copper							− (1)	+ (3)	++ − (1) (1)	+ (2)		++ (1)	++ (2)				
Lead	+ (1)		+ − (1) (1)	+... +++ (2)		++ − (1) (1)	+ (1)	++... +++ (3)	++ − (1)(1)	++ (1)	+... +++ (4)	++ (2)	++ (1)	++ (2)			
pH	− (1)	−−... −−− (2)	− (1)	− (1)				−− (1)		+++ (1)	−−... −−− (2)				− (2)		
Conductivity		+... ++ (2)		+++ (2)	+ (2)	+ (1)		+ (2)			+... +++ (3)				+... ++ (2)	−−... −−− (2)	

Kajaani centre, precipitation with a positive regression coefficient explains 47 % of the variance of zinc, and in Hämeepuisto air temperature with a positive coefficient 26 % of the zinc's variance.

Deposition of lead cannot be modelled by the hydrometeorological variables in the individual catchments except in Kontula. There, 26 % of the variance of lead's deposition is explained by precipitation.

The models derived for the pH value are nonsignificant in the catchments of Kontula, Pakila, Herttoniemi and Kaukovainio. In the city-centre catchments of Hämeenpuisto and Kajaani, and in the industrial catchment of Nekala, air temperature with a negative regression coefficient explains roughly half of the variance of pH. In the catchments of Tampere an obvious explanation for the decrease of pH value in the summertime, is the increase of the sulphate deposition found in the data analysed. In the Kajaani centre the increase of the calcium deposition in the winter raises pH during that period.

Table 29. Hydrometeorological variables explaining deposition in various test catchments and fraction of variance explained. (+) indicates increasing effect of explanatory variable, (−) indicates decreasing effect of explanatory variable. Significance level shows level at which each or weakest of regression coefficients of explanatory variables in model is significant in two-sided testing by t test. N.S. indicates statistically non-significant model (significance level <95 %).

Catchment	Model of parameter								
	total deposition tot D			volatile deposition vol D			total organic carbon TOC		
	variables in model	fraction of variance explained $100 R^2$ %	significance level %	variables in model	fraction of variance explained $100 R^2$ %	significance level %	variables in model	fraction of variance explained $100 R^2$ %	significance level %
Kontula	(+)T	21.1	95	(+)T (−)ln W	61.3	95	(+)T	46.2	99
Pakila	N.S.	-	-	(−)ln W (+)ln P	50.2	95	(+)T	29.8	95
Herttoniemi	N.S.	-	-	(+)T (−)ln W	44.0	95	(+)$T_{20}^{0.5}$	28.3	95
Nekala	N.S.	-	-	N.S.	-	-	N.S.	-	-
Hämeenpuisto	N.S.	-	-	N.S.	-	-	N.S.	-	-
Kaukovainio	N.S.	-	-	(+)T	24.6	99	N.S.	-	-
Kajaani	(−)ln P	18.8	95	N.S.	-	-	N.S.	-	-

Table 29. Continued.

Catchment	Model of parameter								
	total phosphorus tot P			total nitrogen tot N			chloride Cl		
	variables in model	fraction of variance explained $100 R^2$ %	significance level %	variables in model	fraction of variance explained $100 R^2$ %	significance level %	variables in model	fraction of variance explained $100 R^2$ %	significance level %
Kontula	(+)T	25.9	95	(+)P	37.5	99	(+)P (−)T	56.4	95
Pakila	N.S.	-	-	(+)P	55.0	99.9	(+)P^a	28.4	95
Herttoniemi	N.S.	-	-	(+)P	27.3	95	N.S.	-	-
Nekala	N.S.	-	-	(+)P	21.1	95	N.S.	-	-
Hämeenpuisto	N.S.	-	-	N.S.	-	-	N.S.^{a}	-	-
Kaukovainio	(−)ln P (+)T	50.9	99.9	(+)ln P	40.2	99.9	N.S.	-	-
Kajaani	(−)$P^{0.5}$	17.1	95	N.S.	-	-	(−)ln T_{20}	18.3	95

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a: W excluded as explanatory variable

Table 29. Continued.

Catchment	Model of parameter								
	calcium Ca			sulphate SO_4			vanadium V		
	variables in model	fraction of variance explained 100 R^2 %	significance level %	variables in model	fraction of variance explained 100 R^2 %	significance level %	variables in model	fraction of variance explained 100 R^2 %	significance level %
Kontula	N.S.	-	-	(+)P	39.5	99	N.S.	-	-
Pakila	N.S.^{a}	-	-	(+)P^a	28.0	95	(−)T (+)ln P	60.7	99
Herttoniemi	N.S.	-	-	N.S.	-	-	(−)ln T_{20}	64.4	99.9
Nekala	N.S.	-	-	(+)T	33.0	99	N.S.	-	-
Hämeenpuisto	N.S.	-	-	(+)T	40.7	99	(−)T	35.2	99
Kaukovainio	N.S.	-	-	(+)P	32.0	99	(−)T	14.2	95
Kajaani	(−)ln T_{20}	27.8	99	N.S.	-	-	(−)T	27.2	99

Table 29. Continued.

Catchment	Model of parameter								
	zinc Zn			copper Cu			lead Pb		
	variables in model	fraction of variance explained 100 R^2 %	significance level %	variables in model	fraction of variance explained 100 R^2 %	significance level %	variables in model	fraction of variance explained 100 R^2 %	significance level %
Kontula	N.S.	-	-	N.S.	-	-	(+)P	25.8	95
Pakila	N.S.	-	-	N.S.	-	-	N.S.^{a}	-	-
Herttoniemi	N.S.	-	-	N.S.	-	-	N.S.	-	-
Nekala	N.S.	-	-	N.S.	-	-	N.S.	-	-
Hämeenpuisto	(+)T	25.8	95	N.S.	-	-	N.S.^{a}	-	-
Kaukovainio	N.S.	-	-	N.S.	-	-	N.S.	-	-
Kajaani	(+)P	47.0	99	N.S.	-	-	N.S.	-	-

a: W excluded as explanatory variable

Table 29. Continued.

Catchment	Model of parameter					
	pH			conductivity γ_{25}		
	variables in model	fraction of variance explained 100 R^2 %	significance level %	variables in model	fraction of variance explained 100 R^2 %	significance level %
Kontula	N.S.	-	-	$(+)T_{20}^{0.5}\ (-)P^{0.5}$	44.1	95
Pakila	N.S.	-	-	N.S.	-	-
Herttoniemi	N.S.	-	-	N.S.	-	-
Nekala	$(-)\ln T_{20}$	46.2	99.9	$(+)\ln T_{20}\ (-)P$	35.9	95
Hämeenpuisto	$(-)\ln T_{20}$	55.7	99.9	$(+)\ln T_{20}\ (-)P$	38.4	95
Kaukovainio	N.S.	-	-	$(+)T_{20}^{0.5}$	32.7	99
Kajaani	$(-)\ln T_{20}$	49.1	99.9	N.S.	-	-

Air temperature with a positive regression coefficient appears as an explanatory variable in the model of conductivity in four of the catchments, accompanied by precipitation with a negative coefficient in three of them. The fraction of variance explained is slightly over one third.

6.24 Fraction of total variance explained by hydrometeorological factors, local emissions and background depositions

The deposition observations of the six catchments — Kontula, Pakila, Herttoniemi, Hämeenpuisto, Kaukovainio and Kajaani centre — were combined in order to derive regression models for the various deposition parameters by using as explanatory variables the hydrometeorological variables, emissions in the test catchments (Table 5) and in local backgrounds (Table 6), "sum effects" of the distant point sources (Table 7), and the regional background deposition rates (Table 14).

The following dichotomous variables were also included as explanatory variables:

d_1 $d_1 = 0$ in the case of suburban residential catchments (Kontula, Pakila, Kaukovainio)
$d_1 = 1$ in the case of other type of catchments (Herttoniemi, Hämeenpuisto, Kajaani centre)
d_2 $d_2 = 0$ in the case of catchments in northern Finland (Kaukovainio, Kajaani centre)
$d_2 = 1$ in the case of catchments in southern Finland (Kontula, Pakila, Herttoniemi, Hämeenpuisto)
d_3 $d_3 = 0$ in the case of inland catchments (Hämeenpuisto, Kajaani centre)
$d_3 = 1$ in the case of catchments on the coastal zone (Kontula, Pakila, Herttoniemi, Kaukovainio)

The final models of the combined observations are given in Table 30.

Only less than 10 % of the total variance of the total deposition can be explained by the explanatory variables used in the analysis. On the average, the total deposition increases slightly with increasing air temperature and is higher in the more urbanized areas (city centres and traffic catchments) in comparison to the suburban areas.

The volatile deposition is somewhat better explained by the explanatory variables. Air temperature and the rate of emission of hydrocarbons in the local background explain roughly one fifth of the total variance of volatile deposition. Thus, the emissions due to vegetation in the summer period affect the deposition of organic matter. It is of interest to note that the emission rate of hydrocarbons (primarily originating from the traffic) appears as a significant explanatory variable in the model, indicating conversion of the hydrocarbons to depositing organic matter.

Air temperature and the emission rate of hydrocarbons in the local background explain even a larger fraction of the variance of the organic carbon deposition, slightly less than one third. The regional background does not have an influence on the deposition of the organic carbon.

Only 10 % of the variance of the deposition of total phosphorus can be explained. According to the model derived, the deposition of phosphorus increases during the vegetation period and with the increase of urbanization. The regional background deposition rate of phosphorus proves to be non-significant as an explanatory variable.

Roughly 40 % of the variance of the deposition of total nitrogen is explained by precipitation, regional background deposition rate and air temperature. Thus, nitrogen is one of the compounds for which the deposition level is determined by conditions over extensive areas. The

Table 30. Final models of deposition for combined observations. Nekala catchment excluded from analysis. Significance level indicates level at which each or weakest of regression coefficients of explanatory variables in model is significant in two-sided testing by t test. Explanatory variables: P = monthly precipitation (mm), T = monthly air temperature (°C), T_{20} = T+20.0 (°C), d_1 = 0 for residential catchments, d_1 = 1 for other catchments, d_2 = 0 for catchments of northern Finland, d_2 = 1 for other catchments, $E_{H_xC_y,lb}$ = emission rate of hydrocarbons in local background (kg ha^{-1} a^{-1} H_x C_y), $E_{SO_x,c}$ = emission rate of oxides of sulphur in catchment (kg ha^{-1} $a^{-1}SO_x$), $E_{V,lb}$ = emission rate of vanadium in local background (kg ha^{-1} a^{-1} V), $E_{Pb,c}$ = emission rate of lead in catchment (kg ha^{-1} a^{-1} Pb), $E_{Pb,ds}$ = "sum effect" of lead from distant point sources (kg a^{-1} Pb), tot N_{rb} = deposition of total nitrogen in regional background (mg m^{-2} N in 30 days), $SO_{4,rb}$ = deposition of sulphate in regional background (mg m^{-2} SO_4 in 30 days).

Deposition parameter	Number of observations in regression n	Model	Fraction of variance explained 100 R^2 %	Significance level %
Total deposition tot D mg m^{-2} in 30 days	136	tot D = 2240+73T+1130d_1	9.4	95
Volatile deposition vol D mg m^{-2} in 30 days	117	vol D = 700+49T+0.72$E_{H_xC_y,lb}$	19.2	99
Total organic carbon TOC mg m^{-2} C in 30 days	117	TOC = 129+7.1T+0.125$E_{H_xC_y,lb}$	28.6	99.9
Total phosphorus tot P mg m^{-2} P in 30 days	136	tot P = 2.6+0.11T+1.3d_1	10.3	95
Total nitrogen tot N mg m^{-2} N in 30 days	136	tot N = −30+0.73P+1.1 tot N_{rb}−0.8T	41.5	95
Chloride Cl mg m^{-2} Cl in 30 days	117	Cl = −1+37d_2+0.52P−1.4T	48.3	99.9
Calcium Ca mg m^{-2}Ca in 30 days	117	Ca = 208 − 52ln T_{20}+54d_1	28.2	99.9
Sulphate SO_4 mg $m^{-2}SO_4$ in 30 days	136	SO_4 = 34+2.71P+0.309$E_{SO_x,c}$+0.76$SO_{4,rb}$	33.3	95
Vanadium V mg m^{-2} V in 30 days	136	V = 0.36+0.049$E_{V,lb}$−0.019T+0.18d_1	54.0	99
Zinc Zn mg m^{-2} Zn in 30 days	90	Zn = −1.11+1.68d_1+0.56$T_{20}^{0.5}$	18.0	95
Copper Cu mg m^{-2} Cu in 30 days	90	Cu = 0.42−0.68d_2+0.27d_1+0.15 ln P	40.8	95
Lead Pb mg m^{-2} Pb in 30 days	136	Pb = 0.86+0.24$E_{Pb,ds}$+0.076$E_{Pb,c}$	58.2	99.9
pH	136	pH = 6.4 − 0.6 ln T_{20} − 0.3d_2	22.6	95
Conductivity γ_{25} mS m^{-1}	136	γ_{25} = 2.4d_1+1.2$T_{20}^{0.5}$ −0.020P	31.8	99

negative coefficient of temperature refers to heating emissions in the wintertime.

For the chloride deposition, roughly half of the variance can be explained by the explanatory variables. According to the model the deposition of chloride is higher in the catchments located in southern Finland. The deposition increases with the increasing amount of monthly precipitation and with the decreasing air temperature. The latter reflects emissions due to heating in the non-vegetation season. The dichotomous variable (d_3) distinguishing the catchments located on the coastal zone proves to be non-significant as an explanatory variable.

Temperature and catchment category explain 28 % of the total variance of the calcium deposition. The negative coefficient of temperature indicates emissions due to heating in the wintertime. Modelling efficiency is somewhat hampered by the deviation of the calcium deposition in the Kajaani centre catchment (Figs. 14 and 15).

One third of the total variance of the deposition of sulphate can be explained by precipitation, emission rate of sulphur oxides in the catchments and deposition rate of sulphate in the regional background. Sulphate is therefore another parameter on the deposition rate of which the regional background has a significant influence.

The average emission rate of vanadium in the local background plus the temperature and the catchment type explain slightly more than half of the total variance of the vanadium deposition. The negative regression coefficient of air temperature reflects the main source of vanadium emissions, i.e. combustion of fuel oil during the non-vegetation period.

Less than one fifth of the total variance of the zinc deposition rate can be explained by the variables used. According to the regression model, the deposition of zinc increases with a growing degree of urbanization and during the vegetation period. The latter refers to a growing corrosion rate of zinc-covered surfaces due to increased temperature.

The model derived for copper shows that the deposition rate of copper is significantly higher in the test catchments of northern Finland (Kaukovainio and Kajaani centre). Morever, the copper depositions are higher in the more urbanized catchments and increase with a growing amount of monthly precipitation. Of the total variance roughly 40 % can be explained by the explanatory variables used.

The emission levels are found to be the dominant factor determining the level of lead's deposition rate. The rough "sum effect" of the distant point sources and the average emission rate in the catchments explain 58 % of the total variance of the lead deposition. Though the order of significance of the explanatory variables in this case may be more due to statistics, the model anyway suggests that the emissions of lead from sources other than traffic also have a significant impact on the lead deposition.

Only roughly one fifth of the total variance of pH of the deposition samples can be explained. According to the model, pH decreases in the vegetation period contrary to the behaviour expected. In the model this is due to the fact that pH decreases in the vegetation period in the catchments of Hämeenpuisto and Kajaani centre. Moreover, the model indicates that pH is lower in the test catchments of southern Finland.

Slightly less than one third of the total variance of conductivity is explained. According to the model, the values of conductivity increase with the increasing degree of urbanization and with air temperature, and decrease with a growing amount of monthly precipitation. The latter reflects the diluting effect of precipitation.

6.25 Adequacy of models

In general, due to the degree of inaccuracy in the data as dealt with in this study, the regression models derived should be considered descriptive in the first place, showing the most significant explanatory factors and direction of their effects. When the residuals of the models derived were examined, they were considered adequate. However, a tendency was found for the models to underestimate the highest observed deposition rates. As examples of the residuals, the residual plots of the total nitrogen and vanadium deposition models (combined observations) are shown graphically in Appendix 4.

6.3 Discussion

The purposes of this study are specific. The primary objective was to find out the deposition-rate level of various parameters for use in modelling of the runoff quality in the test catchments. Based on the observations, the average composition of precipitation is shown in Table 31 and in Fig. 15. The average concentration of a parameter was computed by dividing the cumulative deposition by the cumulative precipitation. The computational concentration is in general given by two significant numbers.

Table 31. Average composition of precipitation (deposition) in test catchments.

Parameter	Unit	Season	Concentration in catchment						
			Kontula	Pakila	Hertto-niemi	Nekala	Hämeen-puisto	Kauko-vainio	Kajaani
Total solids (total deposition)	mg l^{-1}	hydrological year	51	48	61	30	72	41	82
		vegetation period	62	53	69	26	54	34	59
		non-vegetation period	37	41	50	37	110	49	110
Volatile solids (volatile deposition)	mg l^{-1}	hydrological year	22	23	33	11	23	19	24
		vegetation period	27	27	41	11	22	19	20
		non-vegetation period	14	16	20	16	26	19	29
Total organic carbon	mg l^{-1} C	hydrological year	3.7	4.1	5.4	1.5	3.1	2.9	3.9
		vegetation period	4.8	5.3	7.8	1.5	3.4	2.6	3.2
		non-vegetation period	2.2	2.5	2.6	1.5	2.4	3.3	4.8
Total phosphorus	mg l^{-1} P	hydrological year	0.050	0.059	0.098	0.039	0.072	0.062	0.081
		vegetation period	0.066	0.076	0.14	0.031	0.070	0.066	0.048
		non-vegetation period	0.030	0.035	0.042	0.053	0.078	0.055	0.12
Total nitrogen	mg l^{-1} N	hydrological year	1.4	1.4	1.1	0.86	1.2	1.1	0.95
		vegetation period	1.0	1.1	1.0	0.71	1.0	1.0	0.68
		non-vegetation period	1.9	1.9	1.2	1.1	1.4	1.2	1.2
Chloride	mg l^{-1} Cl	hydrological year	0.90	0.94	1.0	0.39	0.88	0.33	0.43
		vegetation period	0.74	0.63	0.95	0.36	0.76	0.25	0.31
		non-vegetation period	1.1	1.4	1.2	0.44	1.1	0.44	0.63
Calcium	mg l^{-1} Ca	hydrological year	1.1	1.0	1.4	0.98	1.2	0.57	3.7
		vegetation period	0.88	0.69	1.1	0.76	0.83	0.44	2.1
		non-vegetation period	1.3	1.5	1.7	1.4	1.9	0.73	5.3
Sulphate	mg $l^{-1}SO_4$	hydrological year	7.3	7.0	9.4	6.1	8.6	6.0	14
		vegetation period	6.9	5.9	9.0	5.8	8.4	6.8	10
		non-vegetation period	7.8	8.7	9.9	6.6	9.0	5.3	19
Vanadium	mg l^{-1} V	hydrological year	0.005	0.005	0.009	0.009	0.022	0.005	0.014
		vegetation period	0.004	0.003	0.005	0.005	0.011	0.003	0.006
		non-vegetation period	0.007	0.008	0.014	0.016	0.042	0.007	0.021
Cadmium	mg l^{-1} Cd	hydrological year	<0.001	<0.001	<0.001	<0.001	<0.001	~0.001	<0.001
		vegetation period	<0.001	<0.001	<0.001	<0.001	<0.001	~0.001	<0.001
		non-vegetation period	<0.001	<0.001	<0.001	<0.001	<0.001	~0.001	<0.001
Titanium	mg l^{-1} Ti	hydrological year	0.012	0.012	0.012	0.028	0.064	0.011	0.052
		vegetation period	0.008	0.008	0.005	0.033	0.055	0.005	-
		non-vegetation period	0.015	0.015	0.014	0.019	0.072	0.013	0.052
Zinc	mg l^{-1} Zn	hydrological year	0.044	0.024	0.087	0.030	0.042	0.025	0.057
		vegetation period	0.051	0.022	0.084	0.026	0.037	0.017	0.050
		non-vegetation period	0.031	0.030	0.090	0.038	0.056	0.035	0.068
Copper	mg l^{-1} Cu	hydrological year	0.007	0.008	0.007	0.010	0.012	0.019	0.032
		vegetation period	0.007	0.005	0.006	0.009	0.010	0.013	0.022
		non-vegetation period	0.008	0.013	0.009	0.013	0.016	0.027	0.049
Lead	mg l^{-1} Pb	hydrological year	0.021	0.022	0.036	0.025	0.084	0.019	0.047
		vegetation period	0.018	0.015	0.029	0.019	0.068	0.014	0.032
		non-vegetation period	0.026	0.032	0.043	0.036	0.11	0.026	0.064
pH		hydrological year	4.1	4.3	4.0	4.1	4.1	4.4	4.6
		vegetation period	4.1	4.3	4.1	4.1	4.0	4.3	4.2
		non-vegetation period	4.0	4.0	3.9	4.3	4.2	4.4	5.2
Conductivity	mS m^{-1}	hydrological year	4.5	3.9	6.7	5.6	6.1	3.8	6.4
		vegetation period	5.4	3.6	6.4	6.8	6.8	4.5	7.4
		non-vegetation period	4.1	4.0	7.1	4.4	5.3	2.5	5.4

In the coming discussion some comparisons are made between this study and the following: the deposition studies performed in some Finnish cities in the late 1960's (Table 32), a proposed classification of the total deposition and some of its components from the early 1970's (Table 33), and some foreign investigations in which the interaction between air and runoff-water quality in the urban areas has been studied.

Direct comparison of the deposition rates reported from various studies is hampered (and is virtually impossible) due to differences in the representativeness of results (number of gauges and length of monitoring periods), and in the methods of sampling and analysis. In the Finnish studies referred to in Table 32, the sampler used was the B.S.1747 gauge (polyethene funnel with a bottle); the treatment of samples differed from the one adopted in this study (the water soluble and insoluble parts were separated). Due to similar treatment and analysis of the samples in this study and in the investigation concerning the regional background (Järvinen and Haapala 1980), the results in these two investigations are in practice directly comparable; yet, the different duration of the measurement periods has to be emphasized (Tables 13 and 14).

6.31 Total deposition

It is concluded world-wide that since the 1960's the amount of particle fall in the urban areas has been decreasing due to improved pollution-control strategies and practices (Goettle 1978).

In the suburban residential catchments of this study (Kontula, Pakila, and Kaukovainio) the average total depositions are found to be close to the levels measured in the background points of the cities a decade ago (Table 32). In the other catchments (Herttoniemi, Hämeenpuisto and Kajaani centre) the average total depositions rise to levels measured in the late 1960's; yet, they are closer to the lower averages observed at that time.

In the classification of Table 33, five of the six test catchments belong to the class with satisfactory air for residential purposes; the Kaukovainio catchment would be classified as good for residential living. On a world-wide scale the test catchments belong to the category rural-urban areas; Goettle (1978) suggests following average dustfall rates (mg m^{-2} month^{-1}) for the various land-use categories on a world-wide scale:

rural areas	2 500— 3 300
urban areas	3 300— 6 300
large urban areas	5 500—30 000
industrial areas	10 000—60 000

The Finnish National Board of Health presents in its recommendations (Lääkintöhallitus 1978) a total deposition rate of 10 000 mg m^{-2} month^{-1} (annual mean) as a limit which is considered clearly polluting. During some months in the test period this limit has been exceeded in all three test catchments of Helsinki and in the Hämeenpuisto catchment in Tampere (Table 25).

The seasonal variations of the total deposition rate and composition are rather different in the results published in literature. In some investigations the dustfall loads have a maximum in the non-vegetation period, while others show maxima in the vegetation period. According to the results of this study, on the average, higher total deposition rates are found in the summer period; yet, in city centres the total deposition rates are roughly equal during summer and winter periods, or even slightly higher in the wintertime (Figs. 14 and 15).

6.32 Volatile deposition

On the average, the rates of volatile deposition grow during the vegetation period (Table 30). Yet, in the city-centre areas this increase is not statistically significant (Table 29). The lowest average proportion of volatile matter of the total deposition is found in the Kajaani centre where it is about 30 % and the highest one in the suburban residential catchment of Kaukovainio in which it is roughly 50 %.

6.33 Total organic carbon

The rates of organic carbon grow on the average with increasing air temperature (Table 30). Yet, as to the individual catchments, this increase is statistically significant only in the three test sites of Helsinki (Table 29). In the catchments of northern Finland (Kaukovainio and Kajaani centre) the average deposition rates are roughly equal during the vegetation and non-vegetation periods (Figs. 14 and 15).

The observed deposition rates are roughly double compared to the rates in the regional background (Tables 13 and 14). The increase is mainly due to hydrocarbons emitted in the urban areas.

In the foreign studies referred to, Randall et al. (1978) report on average contents of total organic carbon in the precipitation at nine stations in the Washington D.C. metropolitan area, ranging from 2.5 to 18 mg l^{-1} C. As shown in Table 31, the average concentrations found in this study are of

Table 32. Deposition rates of some parameters observed in Helsinki, Tampere, Oulu, Turku, Lappeenranta and Kokkola in late 1960's (Laamanen and Rautanen 1969, Laamanen and Rautanen 1971, Laamanen et al. 1973, Laamanen 1969).

Parameter and unit	Average deposition rate in municipality															
	Helsinki (1967–1968)				Tampere (1969)				Oulu (1970)				Turku (1966), Lappeenranta (1966–1967), Kokkola (1966–1967)	
	measurement points[a]		background[b]		measurement points		background		measurement points		background		measurement points	
	min	max	min	max	min	max	min	max	min	max	min	max	min	max
Total deposition mg m^{-2} month^{-1}	3 930	14 600	1 270	3 080	2 220	7 720	380	3 120	2 450	7 710	280	3 670	2 460	7 270
Sulphate (water soluble) mg m^{-2} month^{-1} SO_4	290	610	80	350	110	570	30	340	140	510	50	290	250	450
Vanadium (ash) mg m^{-2} month^{-1} V	1.10	5.80	0.09	0.50	0.50	2.90	0.10	0.30	0.60	1.30	0.02	0.22	0.15	1.50
Copper (ash) mg m^{-2} month^{-1} Cu	-	-	-	-	0.03[c]	9.15[d]	-	-	0.03[c]	17.90[d]	-	-	1.40	7.20
Lead (ash) mg m^{-2} month^{-1} Pb	2.30	10.70	0.13	1.44	1.10	6.50	0.10	0.60	0.70	4.20	0.01	0.60	0.35	3.80
pH value (water soluble)	4.4[c]	7.1[d]	-	-	3.9[c]	6.8[d]	-	-	4.1[c]	7.9[d]	-	-	5.0	7.1

a: lowest (min) and highest (max) average deposition rate in actual measurement points; b: lowest and highest deposition rate observed in reference points; c: lowest value observed; d: highest value observed

Table 33. Proposal for classification of total deposition and some of its components (Laamanen 1972a and 1972b).

Level	Evaluation	Deposition (as annual mean) of parameter				
		total deposition mg m^{-2}month^{-1}	sulphate mg m^{-2} month^{-1}SO$_4$	vanadium mg m^{-2} month^{-1} V	copper mg m^{-2} month^{-1}Cu	lead mg m^{-2} month^{-1}Pb
I	clean air and relatively clean air, good for residential or base level area	$\leq$2 000	$\leq$200	0–2.4	0–4	0–4
II	slightly unclean, satisfactory air for residential area	2 010–5 000	201–400	2.5–4.9	5–9	5–9
III	medium pollution, tolerable for residential area	5 010–10 000	401–600	5.0–7.4	10–14	10–14
IV	rather polluted, unsatisfactory for residential area	10 010–15 000	601–800	7.5–9.9	15–19	15–19
V	highly polluted, too dirty and unacceptable for residential area	>15 000	>800	$\geq$10	$\geq$20	$\geq$20

the same order as the lowest ones in the Washington study.

6.34 Total phosphorus

The combined model in Table 30 shows that only a small fraction of the variance of the phosphorus deposition can be explained. On the average, the deposition grows during the summer period and is higher in the more urbanized catchments. As shown in Figs. 14 and 15, the increase of phosphorus deposition in the vegetation period is questionable in the test catchments outside Helsinki.

The observed deposition rates of total phosphorus are roughly 75 % higher than those of the regional backgrounds (Tables 13 and 14).

In the foreign studies, the following average phosphorus contents in the precipitation are reported: 0.3 mg l^{-1} P in the Munich area (Goettle 1978), 0.04—0.62 mg l^{-1} P in the Washington D.C. metropolitan area (Randall et al. 1978), 0.04—0.12 mg l^{-1} P in Gothenburg (Malmquist 1978, Malmquist and Svensson 1977). The average concentrations found in this study (Table 31) are of the same order of magnitude as those reported in Gothenburg.

6.35 Total nitrogen

As shown by the combined model (Table 30), the amount of precipitation, plus the regional background and the season, determine the level of nitrogen deposition in the test catchments. In the regional background the deposition of total nitrogen clearly decreases from southern to northern Finland (Järvinen and Haapala 1980). As to nitrogen, the median values of the deposition observed in the test catchments differ little from the ones observed in the regional backgrounds (Tables 13 and 14).

In general, the deposition rates of nitrogen in Finland seem lower than those observed in more urbanized and industrialized countries: for example, an average deposition of 96 mg m^{-2} month^{-1} N is reported in Gothenburg (Malmquist 1978, Malmquist and Svensson 1977), and average concentrations in the precipitation ranging from 0.79 to 9.9 mg l^{-1} N in the metropolitan area of Washington D.C. (Randall et al. 1978).

6.36 Chloride

The average deposition of chloride is lower in the northern test catchments and increases with a growing amount of precipitation and during the non-vegetation period (Table 30, Figs. 14 and 15).

In all catchments the medians of the chloride deposition are of the same order of magnitude as those observed in the regional backgrounds (Tables 13 and 14).

The average chloride contents in precipitation are in general reported to be higher in the foreign studies; for example 2.0—2.7 mg l^{-1} Cl in Gothenburg (Malmquist 1978, Malmquist and Svensson 1977). Probably this is mainly due to wider sea areas but also to the higher level of urban and industrial activities.

6.37 Calcium

The deposition of calcium increases in the winter in all test catchments (Figs. 14 and 15). The higher level of the calcium deposition in the Kajaani centre is due to the emissions from the cellulose factory (calcium line) located further away from the test site.

In the suburban catchments (Kontula, Pakila and Kaukovainio) the observed calcium depositions are of the same order as those in the regional backgrounds. In the other catchments (Herttoniemi, Hämeenpuisto and Kajaani centre) the observed deposition is roughly double compared to the one measured in the regional background (Tables 13 and 14).

6.38 Sulphate

The amount of precipitation, the level of emission of sulphur oxides and the regional background deposition level explain one third of the total variance of the sulphate deposition in the test catchments (Table 30). In general, the average deposition rates during the vegetation and non-vegetation periods differ little (Figs. 14 and 15).

In the suburban catchments the sulphate's deposition rates are on the average 50 % higher than those of the regional background. In the city-centre catchments the deposition is roughly two- to threefold compared to the average of the regional background (Tables 13 and 14).

When analysing the observations gathered from the nonurban stations over 1971—1977 period, Järvinen and Haapala (1980) concluded that the average deposition rates of sulphate are distinctly higher than the reference values in the period 1955—1958, which they used in the comparison. Some growth in the sulphate deposition can be concluded on the basis of the results of this study, too. In all test catchments (suburban residential catchments included) the average rates are roughly of the same order as those measured at the actual measurement points in the studies of the late 1960's (Table 32). This implies that the pollution-control measures taken in the 1970's may not have been able to decrease the sulphur dioxide emissions as effectively as they obviously did with the other emissions. Secondly, some of the increase is probably due to the long-range transport of sulphur. According to inventories performed by OECD, the proportion of sulphur deposition in Finland coming from abroad is of the order of 50—75 % (Laamanen and Jantunen 1979).

Following the proposed classification shown in Table 33, the depositions of sulphate in the test catchments are on the levels, which are satisfactory or tolerable for the residential purposes.

As an example of the foreign studies referred to, the average content of sulphate in the precipitation in the Munich area is 40 mg l^{-1} SO_4. This is roughly four times higher than the averages found in this study. In Gothenburg the average values determined on single rain events ranged from 4.0 to 4.8 mg l^{-1} SO_4 (Malmquist 1978, Malmquist and Svensson 1977). These are of the same order as the content found in the rainwater (wet deposition) of the Herttoniemi catchment (Table 22).

6.39 Vanadium

In all test catchments the deposition rate of vanadium increases during the winter period due to the emissions from heating (Figs. 14 and 15). In the catchments of Helsinki and Oulu the average deposition is closer to the levels measured in the background points of the studies of the late 1960's (Table 32). The deposition rate in the Hämeenpuisto catchment rises to the level observed in Tampere a decade ago. According to the classification of Table 33, the vanadium depositions in all catchments are at a level which is defined as good for residential purposes.

6.310 Cadmium and titanium

There are not enough observations on the deposition of titanium in the test catchments so that any conclusions could be drawn.

The concentrations of cadmium within the detection limit of analysis, observed in the deposition samples of this study, hamper making conclusions on this element. The average concentration of cadmium in the precipitation is less than 0.001 mg l^{-1} Cd. Goettle (1978) gives an average of 0.001 mg l^{-1} Cd for the Munich area. Randall et al. (1978) report on averages ranging from 0 to 0.013 mg l^{-1} Cd in the Washington D.C. metropolitan area.

The deposition of cadmium in the Kaukovainio catchment is obviously higher than the depositions in the other catchments (Table 25). Consequently, according to the emission inventory, the emission of cadmium is the highest in the Kaukovainio catchment where probably the distant point sources also have an influence on the cadmium deposition of the area (Fig. 9).

6.311 Zinc

In the foreign studies referred to, the following average contents of zinc in the precipitation are given: 0.08 mg l^{-1} Zn in the Munich area (Goettle 1978), 0.020—1.9 mg l^{-1} Zn in the Washington D.C. metropolitan area (Randall et al. 1978), 0.05—0.15 mg l^{-1} Zn in Gothenburg (Malmquist 1978, Malmquist and Svensson 1977). These averages are slightly higher than the ones found in this study (Table 31). The deposition rates in the Gothenburg study were on the average 1.7—4.6 mg m^{-2} $month^{-1}$ Zn which are of the same order as the averages in this study. Mance and Harman (1978) give an average of 4.8 mg m^{-2} $month^{-1}$ Zn from Stevenage New Town, U.K.

6.312 Copper

The average deposition of copper in the test catchments of Kaukovainio and Kajaani centre is significantly higher than the deposition in the other catchments (Table 30). The average rate in the Kajaani centre is equal to the one measured in Kokkola in the late 1960's (Table 32).

As to the levels of the copper deposition, the test catchments would be defined as good for residential purposes (Table 33).

Depositions of 0.3—1.0 mg m^{-2} $month^{-1}$ Cu are reported from Gothenburg (Malmquist 1978, Malmquist and Svensson 1977), and a deposition of 1.5 mg m^{-2} $month^{-1}$ Cu in Stevenage New Town, U.K. (Mance and Harman 1978), which are of the same order of magnitude as the ones in this study. The following average concentrations in the precipitation have been reported: 0.012 mg l^{-1} Cu in the Munich area (Goettle 1978), 0—0.060 mg l^{-1} Cu in the metropolitan area of Washington D.C. (Randall et al. 1978).

6.313 Lead

In the test catchments of Helsinki, the deposition of lead in Herttoniemi is roughly equal to the lowest averages measured in the late 1960's (Table 32). The lead deposition in Hämeenpuisto is equal to the ones measured in Tampere a decade ago. The deposition in Kaukovainio is approximately equal to the lowest averages measured in Oulu ten years ago.

Following the proposed classification of Table 33, the catchments would be defined as good for residential purposes. Yet, the Hämeenpuisto catchment should be classified as satisfactory.

In Gothenburg (Malmquist 1978, Malmquist and Svensson 1977) average depositions of 1.6—2.7 mg m^{-2} $month^{-1}$ Pb are reported, and an average of 3.8 mg m^{-2} $month^{-1}$ Pb in Stevenage New Town, U.K. (Mance and Harman 1978) which are of the same order as the ones in this study. In the Munich area, the average content of lead in precipitation is reported as 0.11 mg l^{-1} Pb (Goettle 1978). In the Washington D.C. metropolitan area (Randall et al. 1978), the averages range up to 0.24 mg l^{-1} Pb and show significantly higher contents than the ones in this study.

6.314 pH value

The medians of pH in the monthly samples in the test catchments range from 4.0 to 4.6. On the average they are 0.4 units lower than the pH values measured in the regional backgrounds (Tables 13 and 14). An obvious decrease has taken place in pH since the late 1960's (Table 32). This is mainly due to the increased deposition of strong acids.

6.315 Conductivity

According to the model in Table 30, the values of conductivity grow with the increase of urbanization. In the suburban catchments the average conductivity is roughly 25 % higher than the one of the regional background. In the city centres the values of conductivity are on the average double compared to those of the regional backgrounds (Tables 13 and 14).

6.316 Regional comparisons

Deposition rates observed in the test catchments of equal regional location (Kontula, Pakila, Herttoniemi in Helsinki, and Kaukovainio in Oulu and Kajaani centre) were compared statistically as catchment pairs. The hypothesis on equal variances was tested by the variance ratio test and the hypothesis on equal means by either the t test or Welch's approximation method (Tables 34 and 35).

Between the catchments of Kontula and Pakila, none of the differences in the means of deposition parameters is statistically significant.

Between Pakila and Herttoniemi, a statistically significant difference is found in the case of parameters reflecting the volumes of mainly heating and traffic activities; i.e. the averages of calcium, sulphate, vanadium, zinc, lead and conductivity are higher in Herttoniemi.

Between Kontula and Herttoniemi, only the deposition of lead and the conductivity of precipitation differ statistically, the averages being higher in Herttoniemi.

Between the suburban residential catchment of Kaukovainio and the city centre of Kajaani the deposition rates of most of the parameters differ statistically, being higher in Kajaani centre. The difference is statistically non-significant for volatile deposition, organic carbon, phosphorus and nitrogen. This is understandable considering the sources and behaviour of these components as discussed above.

7. CONCLUSIONS

The following main conclusions were drawn from the observations of this study:

1) The deposition parameters for which the proportion of wet deposition is distinctly over 50 % are: total nitrogen, chloride and sulphate. The proportion of wet deposition is distinctly less than 50 % in the case of total deposition, volatile deposition, and depositions of total organic carbon and total phosphorus.
2) The regional background deposition level has an effect on the deposition level of total nitrogen and sulphate in the urban catchments.
3) Compared to the average deposition rates observed in the regional backgrounds, the deposition rates in the urban catchments are:
    a) roughly 100 % higher in the case of total organic carbon
    b) roughly 75 % higher in the case of total phosphorus
    c) of the same order of magnitude in the case of total nitrogen and chloride
    d) of the same order of magnitude in suburban catchments and roughly 100 % higher in city centres in the case of calcium
    e) roughly 50 % higher in suburban catchments and 100 to 200 % higher in city centres in the case of sulphate

   Moreover, the pH value of precipitation is on the average 0.4 units lower, and conductivity 25 % higher in suburban catchments and 100 % higher in city centres when compared to the average observed in the regional background.
4) Of the order of one third to one half of the total variance can be explained by the hydrometeorological factors, local emission levels and regional background deposition levels in the case of the following deposition parameters: total nitrogen, chloride, sulphate, vanadium, copper, lead and conductivity.
5) The parameters which are especially weakly modelled are: total deposition and total phosphorus.
6) In the 1970's some increase can be observed in the deposition rate of sulphate, while the pH of precipitation has decreased.
7) The studied test catchments are classified as satisfactory for residential purposes.

Table 34. Test of hypothesis on equal variances of deposition parameters in test catchments of Helsinki and in test catchments of Oulu and Kajaani. Variance ratio test. Two-sided testing at 95 % significance level.

Catchment pair	Statistic	Values of statistics for parameter												
		tot D	vol D	TOC	tot P	tot N	Cl	Ca	SO_4	V	Zn	Cu	Pb	γ_{25}
Kontula/Pakila	variance ratio	-[a]	1.36	1.53	1.23	1.69	1.00	1.06	1.07	1.17	-	3.36	1.56	1.89
	ν_1, ν_2		15,15	15,15	18,18	18,18	18,18	18,18	18,18	17,17		12,12	18,18	18,18
	$F_{0.975}(\nu_1, \nu_2)$		2.86	2.86	2.60	2.60	2.60	2.60	2.60	2.68		3.28	2.60	2.60
Pakila/ Herttoniemi	variance ratio	-	3.37	1.51	6.72	2.37	1.27	1.13	1.44	2.78	14.21	4.00	1.62	4.00
	ν_1, ν_2		17,15	16,15	19,18	18,20	18,20	18,20	20,18	19,17	15,12	12,15	20,18	20,18
	$F_{0.975}(\nu_1, \nu_2)$		2.82	2.84	2.58	2.50	2.50	2.50	2.56	2.64	3.18	2.96	2.56	2.56
Herttoniemi/ Kontula	variance ratio	-	4.59	2.30	5.44	4.00	1.27	1.06	1.34	2.37	-	1.19	2.53	2.12
	ν_1, ν_2		17,15	16,15	19,18	18,20	18,20	18,20	20,18	19,17		12,15	20,18	20,18
	$F_{0.975}(\nu_1, \nu_2)$		2.82	2.84	2.58	2.50	2.50	2.50	2.56	2.64		2.96	2.56	2.56
Kaukovainio/ Kajaani centre	variance ratio	1.65	1.26	1.67	1.10	2.25	1.36	178.59	2.83	5.33	3.98	2.22	5.25	1.09
	ν_1, ν_2	25,26	24,23	21,23	25,28	27,25	25,21	22,24	22,26	22,25	15,16	14,16	23,26	28,24
	$F_{0.975}(\nu_1, \nu_2)$	2.22	2.31	2.35	2.17	2.22	2.37	2.31	2.26	2.28	2.79	2.82	2.24	2.23

a: not studied due to non-normal distribution

Table 35. Test of hypothesis on equal means of deposition parameters in test catchments of Helsinki and in test catchments of Oulu and Kajaani. Welch's approximation method and t test. Two-sided testing at 95 % significance level.

Catchment pair	Parameter	Values of statistics								Test used	$\bar{x}_1$ and $\bar{x}_2$ differ from each other
		$\bar{x}_1$	$\bar{x}_2$	s_1	s_2	n_1	n_2	t^a	ν		
Kontula/ Pakila	vol D	1 350	1 320	1 190	1 390	16	16	0.07	30	t test	no
	TOC	258	246	205	166	16	16	0.18	30	t test	no
	tot P	3.6	3.3	2.7	3.0	19	19	0.32	36	t test	no
	tot N	91	86	74	57	19	19	0.23	36	t test	no
	Cl	59	57	45	45	19	19	0.14	36	t test	no
	Ca	70	62	33	34	19	19	0.74	36	t test	no
	SO_4	475	424	208	201	19	19	0.77	36	t test	no
	V	0.36	0.30	0.26	0.24	18	18	0.72	34	t test	no
	Cu	0.42	0.42	0.22	0.12	13	13	0.00	19.7	Welch	no
	Pb	1.37	1.31	0.56	0.70	19	19	0.29	36	t test	no
	γ_{25}	5.5	4.4	2.2	1.6	19	19	1.76	36	t test	no
Pakila/ Herttoniemi	vol D	2 020	1 320	2 550	1 390	18	16	1.01	28.1	Welch	no
	TOC	351	258	252	205	17	16	1.16	31	t test	no
	tot P	5.9	3.6	7.0	2.7	20	19	1.37	25.4	Welch	no
	tot N	86	70	57	37	19	21	1.06	38	t test	no
	Cl	65	57	40	45	21	19	0.60	38	t test	no
	Ca	86	62	32	34	21	19	2.30	38	t test	yes
	SO_4	591	424	241	201	21	19	2.37	38	t test	yes
	V	0.57	0.30	0.40	0.24	20	18	2.55	33.0	Welch	yes
	Zn	5.12	1.34	2.79	0.74	16	13	5.20	17.9	Welch	yes
	Cu	0.43	0.42	0.11	0.22	16	13	0.15	17.6	Welch	no
	Pb	2.22	1.31	0.89	0.70	21	19	3.57	38	t test	yes
	γ_{25}	7.6	4.4	3.2	1.6	21	19	4.06	31.1	Welch	yes
Herttoniemi/ Kontula	vol D	2 020	1 350	2 550	1 190	18	16	1.00	25.6	Welch	no
	TOC	351	246	252	166	17	16	1.40	31	t test	no
	tot P	5.9	3.3	7.0	3.0	20	19	1.52	26.8	Welch	no
	tot N	91	70	74	37	19	21	1.12	26.7	Welch	no
	Cl	65	59	40	45	21	19	0.45	38	t test	no
	Ca	86	70	32	33	21	19	1.56	38	t test	no
	SO_4	591	475	241	208	21	19	1.62	38	t test	no
	V	0.57	0.36	0.40	0.26	20	18	1.89	36	t test	no
	Cu	0.43	0.42	0.11	0.12	16	13	0.23	27	t test	no
	Pb	2.22	1.37	0.89	0.56	21	19	3.57	38	t test	yes
	γ_{25}	7.6	5.5	3.2	2.2	21	19	2.39	38	t test	yes
Kaukovainio/ Kajaani centre	tot D	3 370	1 620	1 810	1 410	26	27	3.94	51	t test	yes
	vol D	990	760	410	460	24	25	1.84	47	t test	no
	TOC	161	124	88	68	22	24	1.60	44	t test	no
	tot P	3.3	2.5	2.2	2.1	26	29	1.38	53	t test	no
	tot N	46	39	18	12	28	26	1.69	48.9	Welch	no
	Cl	18	14	6	7	22	26	2.10	46	t test	yes
	Ca	148	23	147	11	23	25	4.07	22.3	Welch	yes
	SO_4	586	252	286	170	23	27	4.91	35.7	Welch	yes
	V	0.55	0.22	0.30	0.13	23	26	4.89	29.9	Welch	yes
	Zn	2.30	1.09	1.99	0.50	16	17	2.36	17.0	Welch	yes
	Cu	1.37	0.84	0.76	0.51	15	17	2.34	30	t test	yes
	Pb	1.96	0.81	0.55	0.24	24	27	9.47	31.3	Welch	yes
	γ_{25}	6.5	4.2	2.4	2.5	25	29	3.43	52	t test	yes

a: percentiles of Student's t distribution:

$t_{0.975}(15) = 2.13$, $t_{0.975}(20) = 2.09$, $t_{0.975}(30) = 2.04$, $t_{0.975}(40) = 2.02$, $t_{0.975}(50) = 2.01$, $t_{0.975}(60) = 2.00$

ACKNOWLEDGEMENTS

The Finnish Urban Storm Water Project (Valtakunnallinen hulevesitutkimus 1977—1979) is mainly financed by the Maj and Tor Nessling Foundation. The project is performed by the Technical Universities of Helsinki and Tampere, and the University of Oulu. The Water Administration — the National Board of Waters, and the Offices of the Water Districts of Helsinki, Tampere, Oulu and Kainuu — and the municipalities of Helsinki, Tampere, Oulu and Kajaani took part in the financing and accomplishment of the field studies. Reporting of the results is mainly financed by the National Board of Waters, Finland.

The authors express their sincere gratitude to the Financiers and Governing Board of the project for permission to publish this report. We also wish to thank M.Sc. Satu Vuolas and Dr. Risto Laukkanen for their advice during the work, and professors Eero Kajosaari, Arvo Laamanen and Seppo Mustonen, and Lic. Kirsti Haapala and Dr. Matti Jantunen for their comments concerning the manuscript. Mr. Bernard Siegel has revised the language.

Of the authors, Mr. Tähtelä has been in charge of preliminary data preparation. Mr. Melanen has planned the study, been in charge of data collection, performed the statistical analysis, drawn the conclusions on results and written the report.

Helsinki, April 1981

Matti Melanen, Heikki Tähtelä

LOPPUTIIVISTELMÄ

Valtakunnallisen hulevesitutkimuksen kenttäkokeet suoritettiin vuosina 1977—1979 Helsingissä, Tampereella, Oulussa ja Kajaanissa sijainneilla seitsemällä taajamakoealueella.

Tutkimuksen yhtenä osana suoritettiin alueilla ilmaperäisen laskeuman mittaus. Laskeuman havainnoinnin tavoitteena on ollut selvittää ilman kautta tulevan kuormituksen suuruus koealueilla ja tarkastella sen riippuvuutta hydrometeorologisista tekijöistä, paikallisista emissiotasoista ja alueellisesta taustakuormituksesta.

Havainnoista on tehty mm. seuraavat johtopäätökset:

1) Laskeuman komponentit, joiden laskeumasta selvästi yli 50 % tulee märkälaskeuman muodossa, ovat kokonaistyppi, kloridi ja sulfaatti. Komponentit, joiden laskeumasta selvästi yli 50 % tulee kuivalaskeumana, ovat kokonaislaskeuma, orgaaninen aine, orgaaninen hiili ja kokonaisfosfori.
2) Alueellisella taustakuormituksella on selvä vaikutus kokonaistypen ja sulfaatin laskeumien tasoon taajama-alueilla.
3) Verrattuna alueellisessa taustassa havaittuihin laskeumiin ovat taajama-alueilla
   a) orgaanisen hiilen laskeumat noin 100 % korkeampia
   b) kokonaisfosforin laskeumat noin 75 % korkeampia
   c) kokonaistypen ja kloridin laskeumat samaa suuruusluokkaa
   d) kalsiumin laskeumat samaa suuruusluokkaa esikaupunkialueilla ja noin 100 % korkeampia keskusta-alueilla
   e) sulfaatin laskeumat noin 50 % korkeampia esikaupunkialueilla ja 100—200 % korkeampia keskusta-alueilla

   Sadannan pH-arvo on taajama-alueilla keskimäärin 0,4 yksikköä alempi kuin alueellisen taustan sadannan pH-arvo. Sadeveden sähkönjohtavuus on esikaupunkialueilla noin 25 % korkeampi ja keskusta-alueilla noin 100 % korkeampi kuin alueellisessa taustassa.
4) Suuruusluokkaa kolmasosa-puolet kokonaistypen, kloridin, sulfaatin, vanadiinin, kuparin ja lyijyn laskeumien sekä sadeveden sähkönjohtavuuden kokonaisvarianssista voidaan selittää hydrometeorologisia tekijöitä, paikallisia emissiotasoja ja alueellista taustakuormitusta kuvaavilla muuttujilla.
5) Kokonaislaskeuman ja fosforin laskeuman varianssista kyetään selittämään vain noin kymmenesosa.
6) Sulfaatin laskeumissa havaitaan tapahtuneen kasvua 1970-luvun aikana. Samalla ovat sadeveden pH-arvot laskeneet.
7) Tutkimuksen koealueet voidaan havaittujen laskeumatasojen perusteella luokitella tyydyttäviksi asumistarkoituksiin.

LIST OF SYMBOLS

Deposition parameters

tot D	total deposition (mg m^{-2} in 30 days)
tot D_w	wet total deposition (mg m^{-2} in 30 days)[a]
tot D_d	dry total deposition (mg m^{-2} in 30 days)[a]

---

a: analogically for other deposition parameters

tot D_{wp1}	percentage wet deposition of sum of wet and dry depositions in case of total deposition (%)[a]
tot D_{wp2}	percentage wet deposition of deposition collected by standard samplers in case of total deposition (%)[a]
vol D	volatile deposition (mg m^{-2} in 30 days)
TOC	deposition of total organic carbon (mg m^{-2} C in 30 days)
tot P	deposition of total phosphorus (mg m^{-2} P in 30 days)
tot N	deposition of total nitrogen (mg m^{-2} N in 30 days)
Cl	deposition of chloride (mg m^{-2} Cl in 30 days)
Ca	deposition of calcium (mg m^{-2} Ca in 30 days)
SO_4	deposition of sulphate (mg m^{-2} SO_4 in 30 days)
V	deposition of vanadium (mg m^{-2} V in 30 days)
Cd	deposition of cadmium (mg m^{-2} Cd in 30 days)
Ti	deposition of titanium (mg m^{-2} Ti in 30 days)
Zn	deposition of zinc (mg m^{-2} Zn in 30 days)
Cu	deposition of copper (mg m^{-2} Cu in 30 days)
Pb	deposition of lead (mg m^{-2} Pb in 30 days)
pH	pH value of monthly deposition samples
γ_{25}	conductivity of monthly deposition samples (mS m^{-1})

Emission parameters

$E_{part,\ c}$	emission rate of particulates in catchment (kg ha^{-1} a^{-1})
$E_{part,\ lb}$	emission rate of particulates in local background (kg ha^{-1} a^{-1})
$E_{part,\ ds}$	"sum effect" of particulates from distant point sources (kg a^{-1})
$E_{H_xC_y,c}$	emission rate of hydrocarbons in catchment (kg ha^{-1} a^{-1} H_xC_y)
$E_{H_xC_y,lb}$	emission rate of hydrocarbons in local background (kg ha^{-1} a^{-1} H_xC_y)
$E_{H_xC_y,ds}$	"sum effect" of hydrocarbons from distant point sources (kg a^{-1} H_xC_y)
$E_{NO_x,c}$	emission rate of oxides of nitrogen in catchment (kg ha^{-1} a^{-1} NO_x)
$E_{NO_x,lb}$	emission rate of oxides of nitrogen in local background (kg ha^{-1} a^{-1} NO_x)
$E_{NO_x,ds}$	"sum effect" of oxides of nitrogen from distant point sources (kg a^{-1} NO_x)
$E_{SO_x,c}$	emission rate of oxides of sulphur in catchment (kg ha^{-1} $a^{-1}SO_x$)
$E_{SO_x,lb}$	emission rate of oxides of sulphur in local background (kg ha^{-1} a^{-1} SO_x)
$E_{SO_x,ds}$	"sum effect" of oxides of sulphur from distant point sources (kg a^{-1} SO_x)
$E_{V,c}$	emission rate of vanadium in catchment (kg ha^{-1} a^{-1} V)
$E_{V,lb}$	emission rate of vanadium in local background (kg ha^{-1} a^{-1} V)
$E_{V,ds}$	"sum effect" of vanadium from distant point sources (kg a^{-1} V)
$E_{Cd,c}$	emission rate of cadmium in catchment (kg ha^{-1} a^{-1} Cd)
$E_{Cd,lb}$	emission rate of cadmium in local background (kg ha^{-1} a^{-1} Cd)
$E_{Cd,ds}$	"sum effect" of cadmium from distant point sources (kg a^{-1} Cd)
$E_{Ti,c}$	emission rate of titanium in catchment (kg ha^{-1} a^{-1} Ti)
$E_{Ti,lb}$	emission rate of titanium in local background (kg ha^{-1} a^{-1} Ti)
$E_{Ti,ds}$	"sum effect" of titanium from distant point sources (kg a^{-1} Ti)
$E_{Pb,c}$	emission rate of lead in catchment (kg ha^{-1} a^{-1} Pb)
$E_{Pb,lb}$	emission rate of lead in local background (kg ha^{-1} a^{-1} Pb)
$E_{Pb,ds}$	"sum effect" of lead from distant point sources (kg a^{-1} Pb)

Background deposition parameters

TOC_{rb}	average deposition of total organic carbon in regional background (mg m^{-2} C in 30 days)
tot P_{rb}	average deposition of total phosphorus in regional background (mg m^{-2} P in 30 days)
tot N_{rb}	average deposition of total nitrogen in regional background (mg m^{-2} N in 30 days)

Cl_{rb}	average deposition of chloride in regional background (mg m^{-2} Cl in 30 days)
Ca_{rb}	average deposition of calcium in regional background (mg m^{-2} Ca in 30 days)
$SO_{4,rb}$	average deposition of sulphate in regional background (mg m^{-2} SO_4 in 30 days)
pH_{rb}	average pH value of precipitation in regional background
$\gamma_{25,rb}$	average conductivity of precipitation in regional background (mS m^{-1})

Hydrometeorological variables

P	monthly precipitation (mm)
T	mean monthly air temperature (°C)
T_{20}	$= T + 20.0$ (°C)
W	mean monthly wind speed (m s^{-1})

Catchment category

d_1	dichotomous variable: $d_1 = 0$ for suburban residential catchments, $d_1 = 1$ for other catchments
d_2	dichotomous variable: $d_2 = 0$ for catchments of northern Finland, $d_2 = 1$ for other catchments
d_3	dichotomous variable: $d_3 = 1$ for catchments in coastal zone, $d_3 = 0$ for other catchments

Statistical notations

n	number of observations
min	minimum value
m	median value
m_{71-77}	median value over 1971—1977 period
max	maximum value
$\bar{x}$	arithmetic mean
$\bar{x}_{a,min}$	smallest annual arithmetic mean
$\bar{x}_{a,max}$	highest annual arithmetic mean
s	standard deviation
D	Kolmogorov-Smirnov test statistic
100 R^2	fraction of variance explained (multiple correlation squared) (%)
v	degrees of freedom of t distribution
v_1, v_2	degrees of freedom of F distribution
t	test statistic in Student's t test and Welch's approximation method
$t_{0.975}(v)$	critical value of t distribution with v degrees of freedom in two-sided testing at 95 % significance level
$F_{0.975}(v_1, v_2)$	critical value of F distribution with v_1, v_2 degrees of freedom in two-sided testing at 95 % significance level

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Appendix 1. Methods of analysis.

Total solids
Method: Evaporation and drying at 103–105 °C (total residue).
Reference: Vesianalyysitoimikunnan mietintö 1968. Komiteanmietintö 1968: B19.

Volatile solids
Method: Ignition of total residue at 600 °C.
Reference: Vesianalyysitoimikunnan mietintö 1968. Komiteanmietintö 1968: B19.

Total organic carbon
Method: Conversion of organic carbon to carbon dioxide at 950 °C, measurement of amount of carbon dioxide by infra-red analyzer.
Reference: APHA, AWWA & WPCF 1976. Standard methods for the examination of water and wastewater. 14th ed.

Total phosphorus
Method: Oxidation of phosphorus compounds to orthophosphates in acid solution with $K_2S_2O_8$, formation of a complex as result of reactions with ammonium molybdate and ascorbic acid, measurement of absorbance of the complex compound with spectrophotometer at wave length 880 nm.
Reference: Standardiehdotus INSTA–VH 23. 1975.

Total nitrogen
Method: Oxidation of nitrogen compounds to nitrates with $K_2S_2O_8$, reduction of nitrates to nitrites, formation of an atso colour compound as result of reactions with sulfanilamide and N-(1-naphthyl) – ethylenediamine, measurement of absorbance of the complex compound with spectrophotometer at wave length 545 nm.
Reference: Henriksen, A. 1969. Analyseforskrifter for auto-analyzer. NIVA. & Standardiehdotus INSTA–VH 27. 1975.

Chloride
Method: Determination with chemical auto-analyzer.
Reference: Henriksen, A. 1969. Analyseforskrifter for auto-analyzer. NIVA.

Calcium
Method: Determination with atomic absorption spectrophotometer with lantanide addition.
Reference: Manual of the instrument.

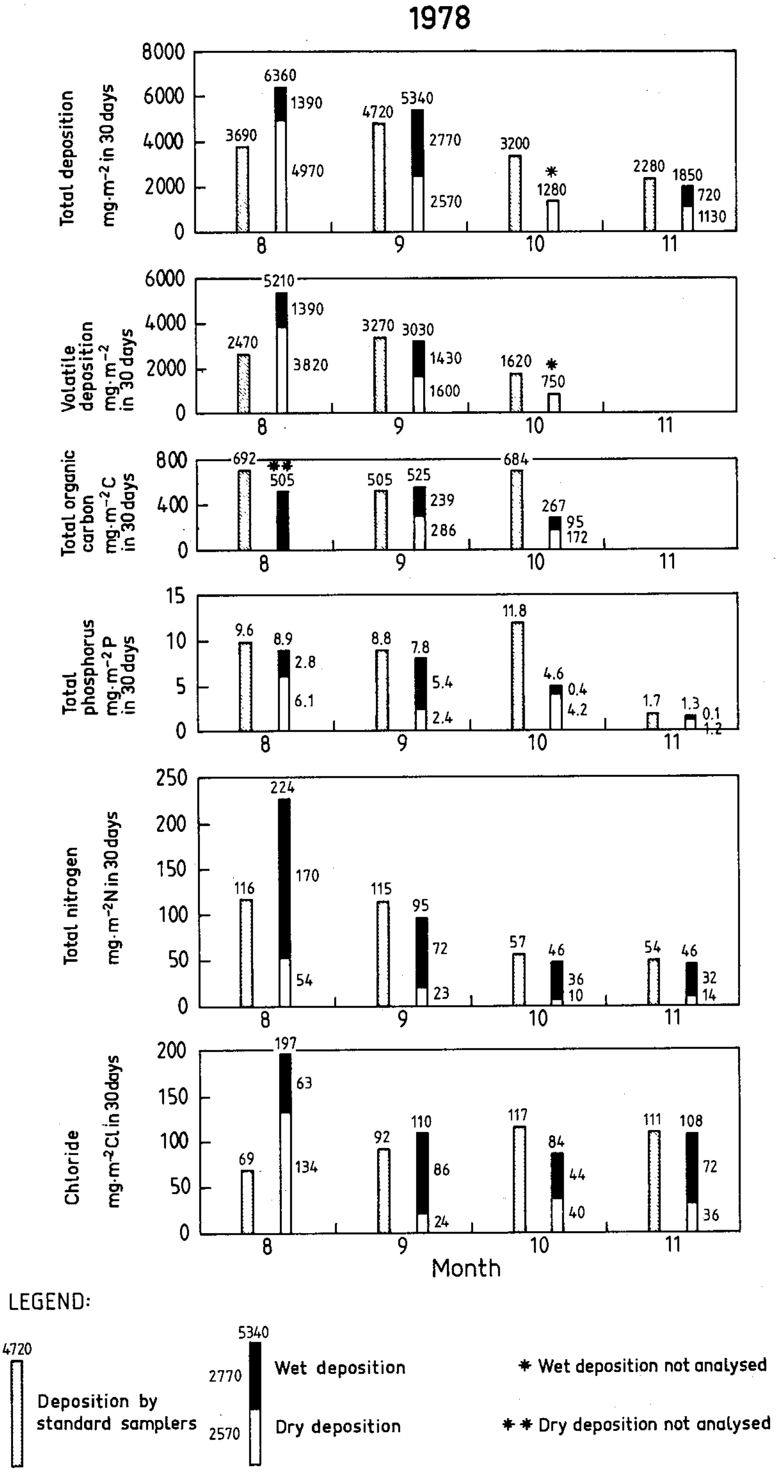
Sulphate
Method: Determination with chemical auto-analyzer based on thorin method.
Reference: Henriksen, A. & Bergmann-Paulsen, I-M. 1974. An automatic method for determining sulfate in natural soft water and precipitation. Vatten 74, 2.

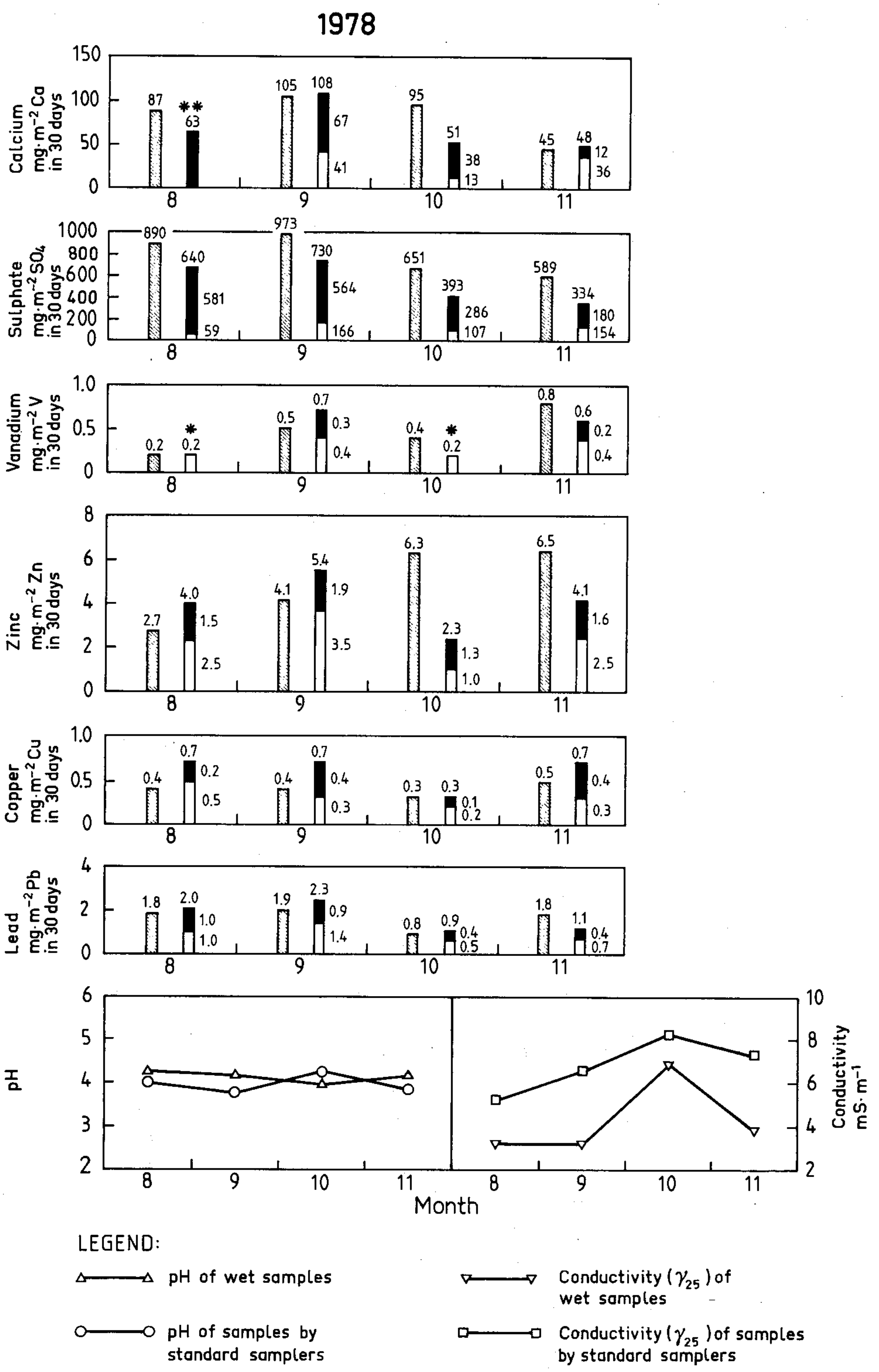
Vanadium, cadmium, titanium, zinc, copper, lead
Method: Determination with atomic absorption spectrophotometer with graphite furnace.
Reference: Manuals of the instruments.

pH
Method: Potentiometric determination at 25 ± 1 °C with glass electrode.
Reference: Suomen Standardisoimisliitto 1979. Standardi SFS 3021.

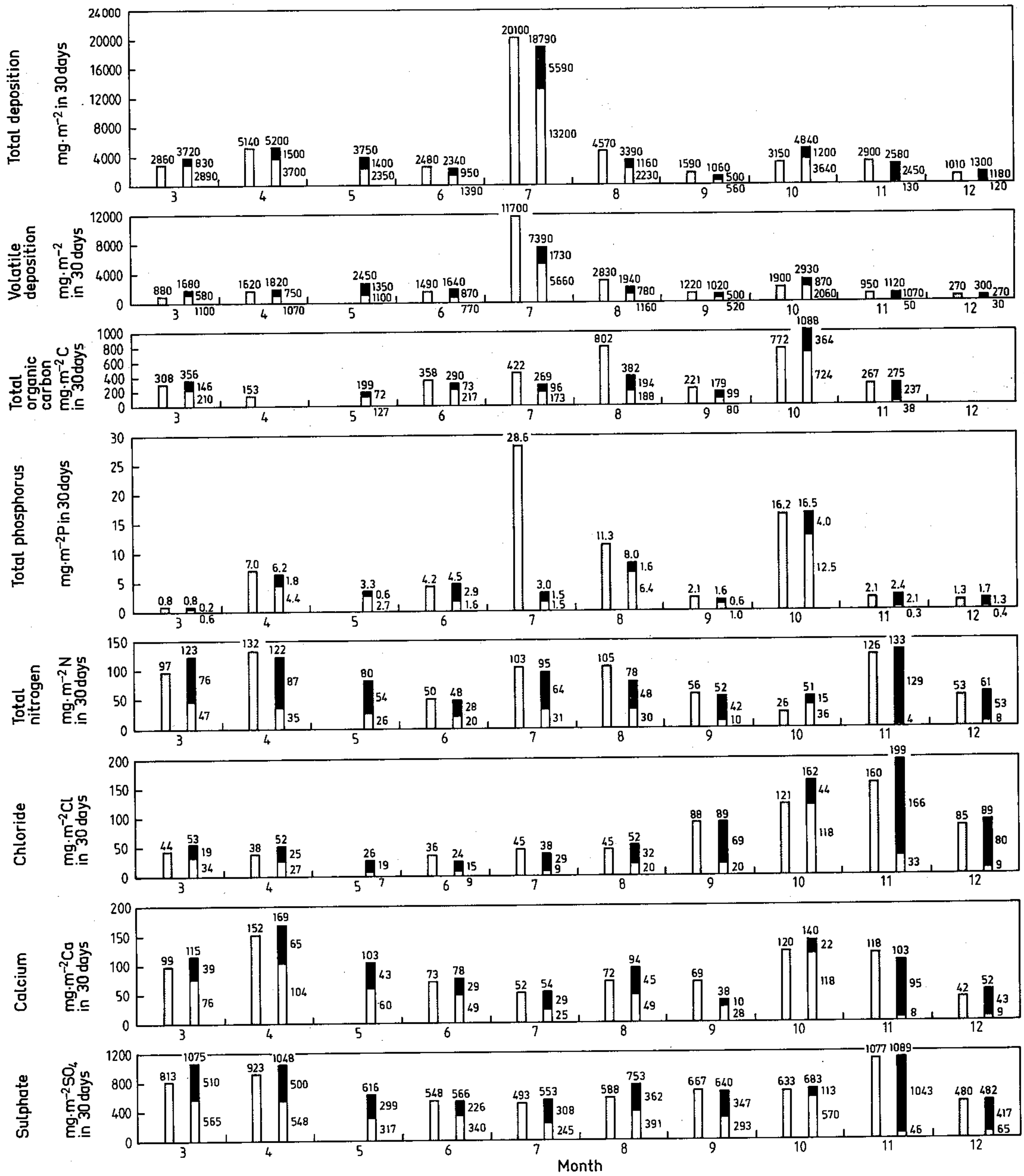
Conductivity
Method: Determination at 25 °C in conductivity cells supplied with platinum electrodes.
Reference: Suomen Standardisoimisliitto 1974. Standardi SFS 3022.

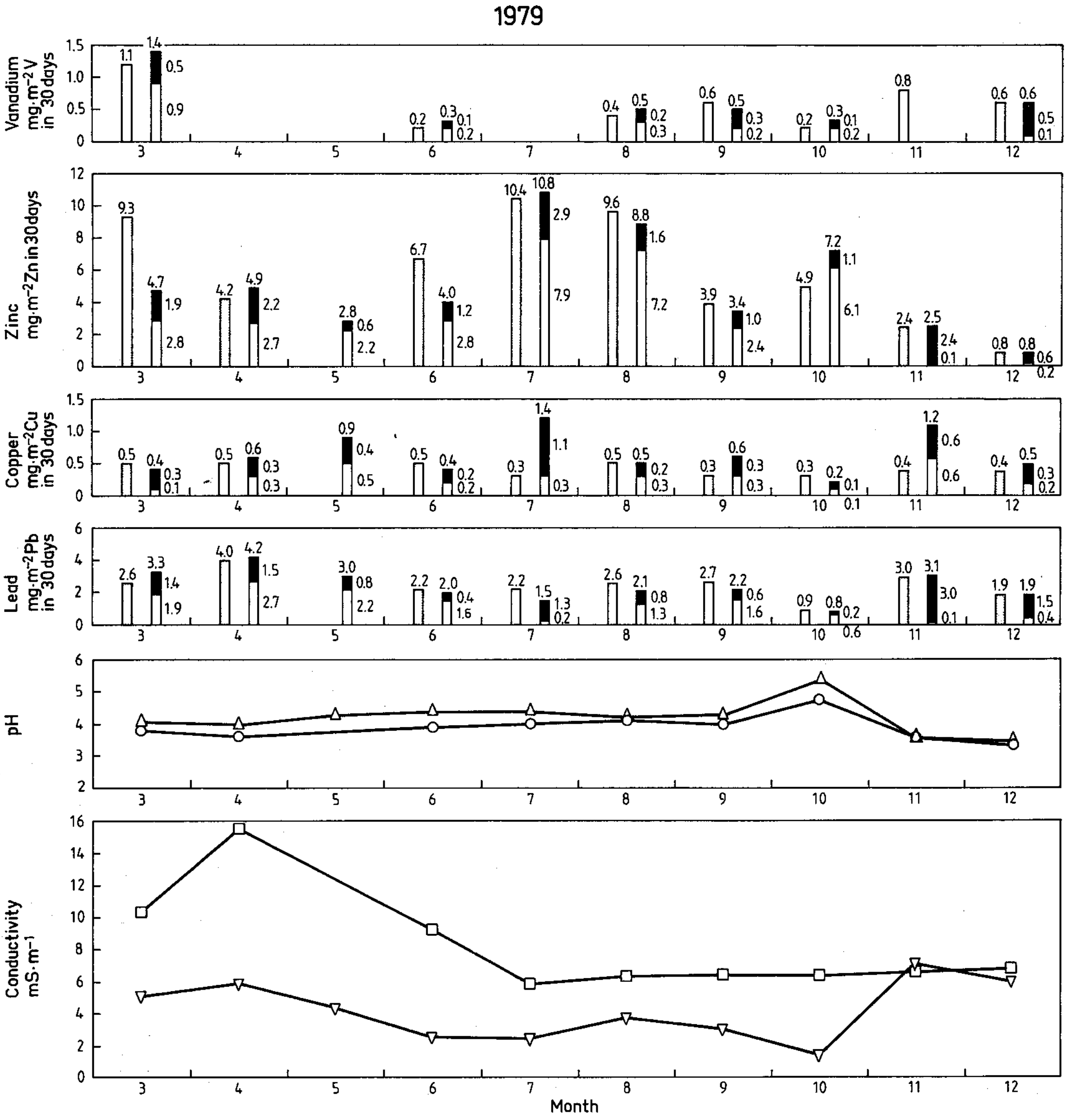
Appendix 2. Wet and dry deposition observed in Herttoniemi catchment.



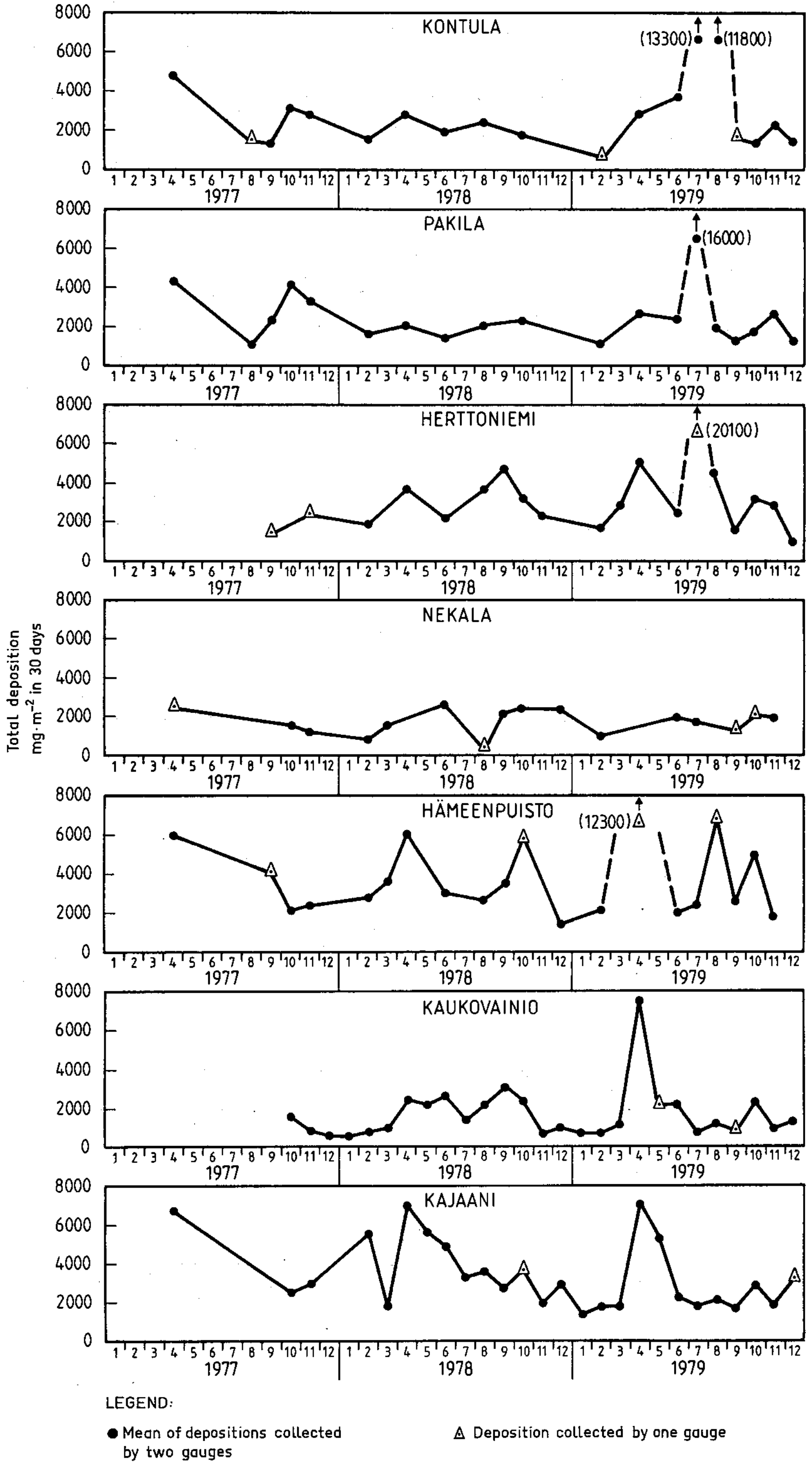

1978
Calcium mg·m⁻² Ca in 30 days
150
100
50
0
87
**
63
105
108
67
41
95
51
38
13
45
48
12
36
8
9
10
11
Sulphate mg·m⁻² SO₄ in 30 days
1000
800
600
400
200
0
890
640
581
59
973
730
564
166
651
393
286
107
589
334
180
154
Vanadium mg·m⁻² V in 30 days
1.0
0.5
0
0.2
*
0.2
0.5
0.7
0.3
0.4
0.4
*
0.2
0.8
0.6
0.2
0.4
Zinc mg·m⁻² Zn in 30 days
8
6
4
2
0
2.7
4.0
1.5
2.5
4.1
5.4
1.9
3.5
6.3
2.3
1.3
1.0
6.5
4.1
1.6
2.5
Copper mg·m⁻² Cu in 30 days
1.0
0.5
0
0.4
0.7
0.2
0.5
0.4
0.7
0.4
0.3
0.3
0.3
0.1
0.2
0.5
0.7
0.4
0.3
Lead mg·m⁻² Pb in 30 days
4
2
0
1.8
2.0
1.0
1.0
1.9
2.3
0.9
1.4
0.8
0.9
0.4
0.5
1.8
1.1
0.4
0.7
pH
6
5
4
3
2
Conductivity mS·m⁻¹
10
8
6
4
2
Month
LEGEND:
pH of wet samples
pH of samples by standard samplers
Conductivity (γ_{25}) of wet samples
Conductivity (γ_{25}) of samples by standard samplers

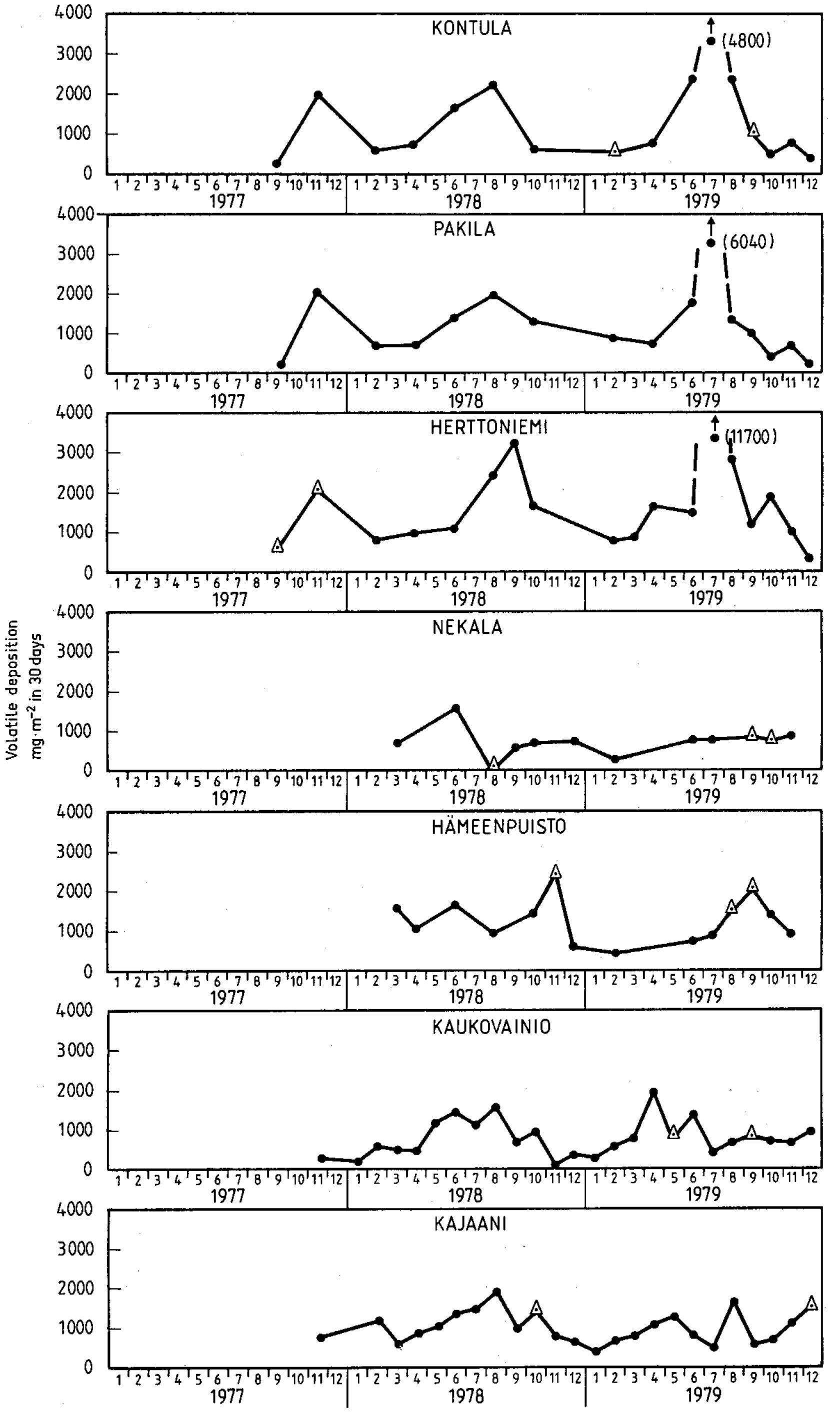
1979

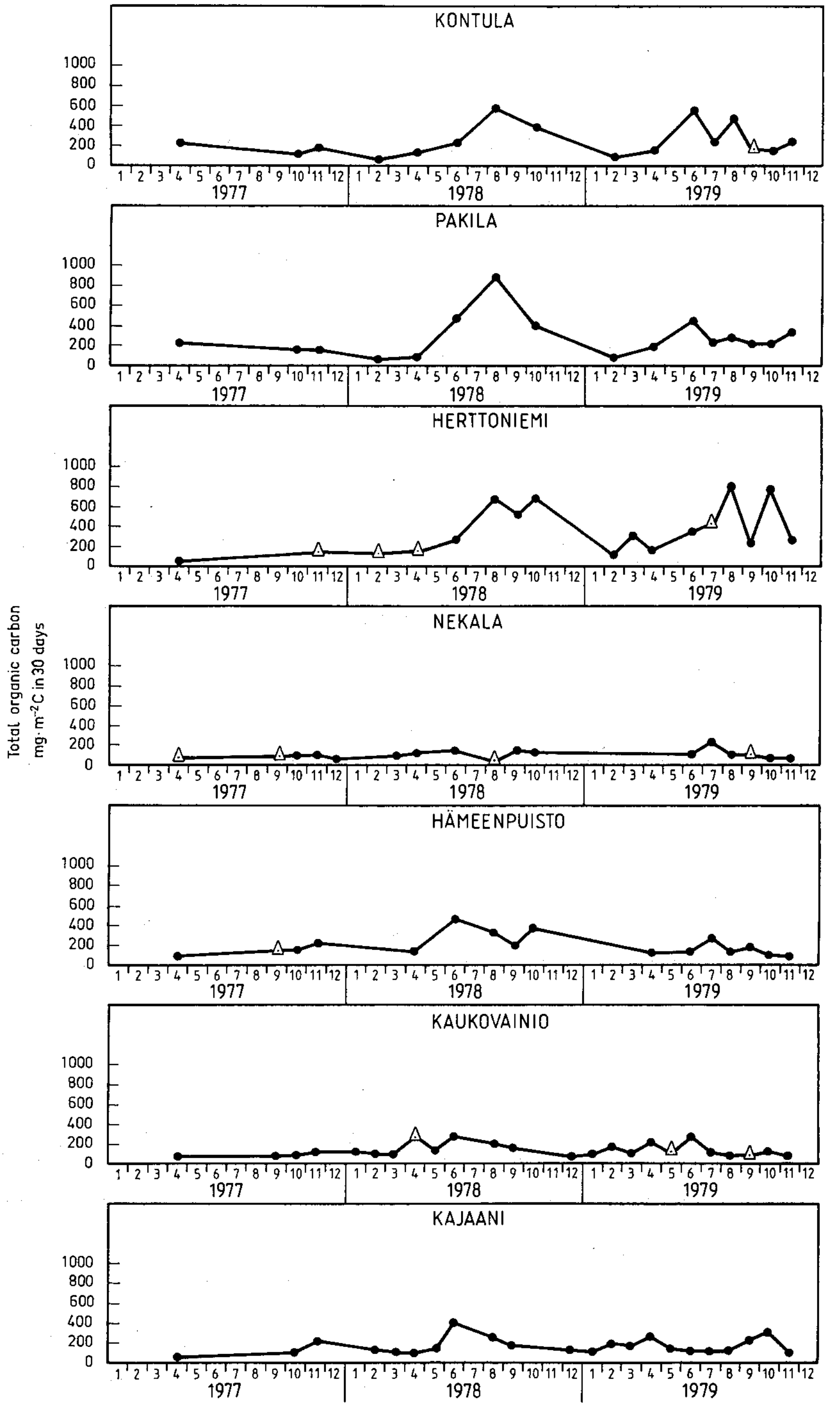

Total deposition
mg·m⁻² in 30 days
24000
20000
16000
12000
8000
4000
0
2860
3720
830
2890
5140
5200
1500
3700
3750
1400
2350
2480
2340
950
1390
20100
18790
5590
13200
4570
3390
1160
2230
1590
1060
500
560
3150
4840
1200
3640
2900
2580
2450
130
1010
1300
1180
120
Volatile deposition
mg·m⁻² in 30 days
12000
8000
4000
0
880
1680
580
1100
1620
1820
750
1070
2450
1350
1100
1490
1640
870
770
11700
7390
1730
5660
2830
1940
780
1160
1220
1020
500
520
1900
2930
870
2060
950
1120
1070
50
270
300
270
30
Total organic carbon
mg·m⁻² C in 30 days
1000
800
600
400
200
0
308
356
146
210
153
199
72
127
358
290
73
217
422
269
96
173
802
382
194
188
221
179
99
80
772
1088
364
724
267
275
237
38
Total phosphorus
mg·m⁻² P in 30 days
30
25
20
15
10
5
0
0.8
0.8
0.2
0.6
7.0
6.2
1.8
4.4
3.3
0.6
2.7
4.2
4.5
2.9
1.6
28.6
3.0
1.5
1.5
11.3
8.0
1.6
6.4
2.1
1.6
0.6
1.0
16.2
16.5
4.0
12.5
2.1
2.4
2.1
0.3
1.3
1.7
1.3
0.4
Total nitrogen
mg·m⁻² N in 30 days
150
100
50
0
97
123
76
47
132
122
87
35
80
54
26
50
48
28
20
103
95
64
31
105
78
48
30
56
52
42
10
26
51
15
36
126
133
129
4
53
61
53
8
Chloride
mg·m⁻² Cl in 30 days
200
150
100
50
0
44
53
19
34
38
52
25
27
26
19
7
36
24
15
9
45
38
29
9
45
52
32
20
88
89
69
20
121
162
44
118
160
199
166
33
85
89
80
9
Calcium
mg·m⁻² Ca in 30 days
200
150
100
50
0
99
115
39
76
152
169
65
104
103
43
60
73
78
29
49
52
54
29
25
72
94
45
49
69
38
10
28
120
140
22
118
118
103
95
8
42
52
43
9
Sulphate
mg·m⁻² SO₄ in 30 days
1200
800
400
0
813
1075
510
565
923
1048
500
548
616
299
317
548
566
226
340
493
553
308
245
588
753
362
391
667
640
347
293
633
683
113
570
1077
1089
1043
46
480
482
417
65
3
4
5
6
7
8
9
10
11
12
Month

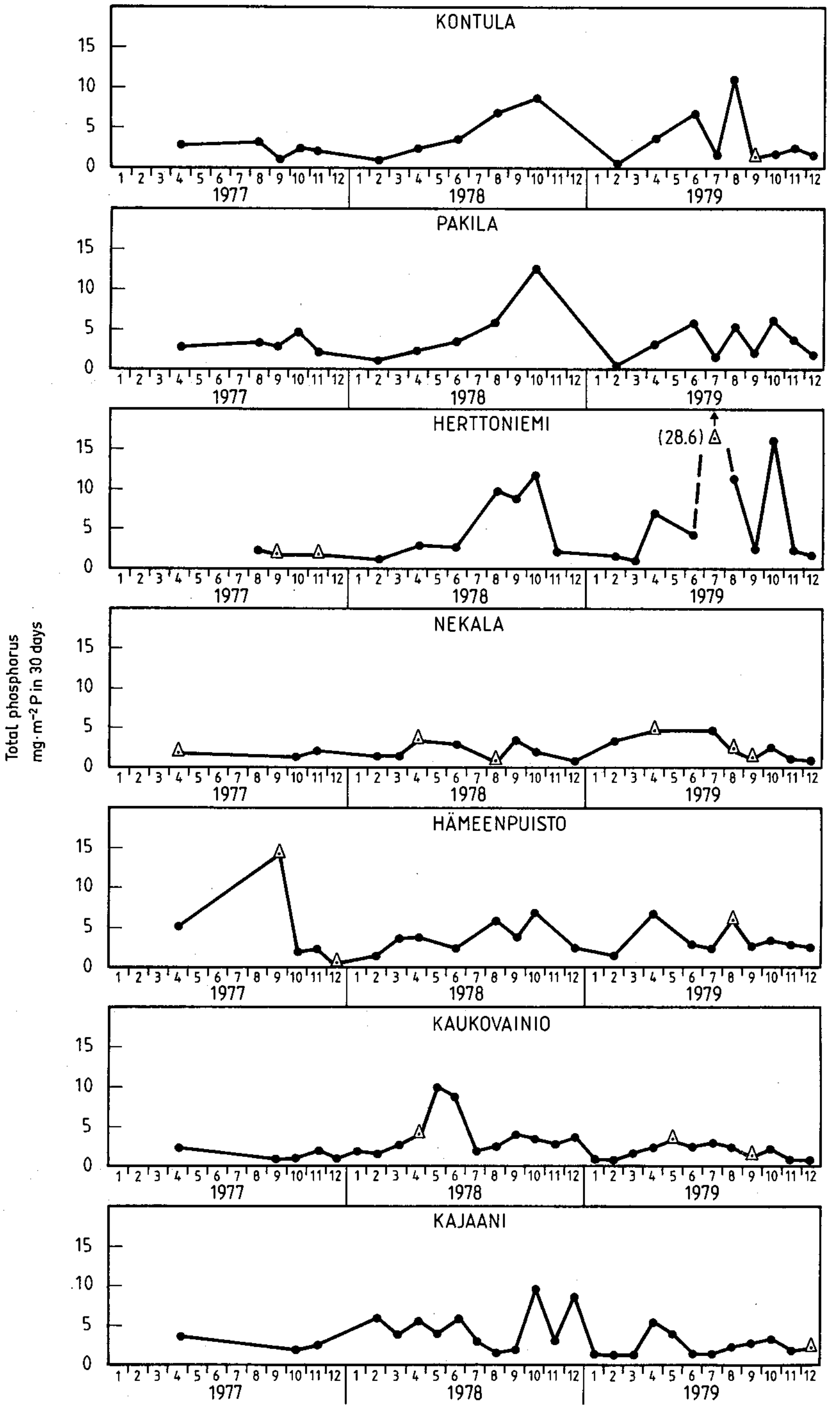

1979
Vanadium mg·m⁻²V in 30 days
1.5
1.0
0.5
0
1.1
1.4
0.5
0.9
0.2
0.3
0.1
0.2
0.4
0.5
0.2
0.3
0.6
0.5
0.3
0.2
0.2
0.3
0.1
0.2
0.8
0.6
0.6
0.5
0.1
Zinc mg·m⁻²Zn in 30 days
12
10
8
6
4
2
0
9.3
4.7
1.9
2.8
4.2
4.9
2.2
2.7
2.8
0.6
2.2
6.7
4.0
1.2
2.8
10.4
10.8
2.9
7.9
9.6
8.8
1.6
7.2
3.9
3.4
1.0
2.4
4.9
7.2
1.1
6.1
2.4
2.5
2.4
0.1
0.8
0.8
0.6
0.2
Copper mg·m⁻²Cu in 30 days
1.5
1.0
0.5
0
0.5
0.4
0.3
0.1
0.5
0.6
0.3
0.3
0.9
0.4
0.5
0.5
0.4
0.2
0.2
0.3
1.4
1.1
0.3
0.5
0.5
0.2
0.3
0.3
0.6
0.3
0.3
0.3
0.2
0.1
0.1
0.4
1.2
0.6
0.6
0.4
0.5
0.3
0.2
Lead mg·m⁻²Pb in 30 days
6
4
2
0
2.6
3.3
1.4
1.9
4.0
4.2
1.5
2.7
3.0
0.8
2.2
2.2
2.0
0.4
1.6
2.2
1.5
1.3
0.2
2.6
2.1
0.8
1.3
2.7
2.2
0.6
1.6
0.9
0.8
0.2
0.6
3.0
3.1
3.0
0.1
1.9
1.9
1.5
0.4
pH
6
5
4
3
2
Conductivity mS·m⁻¹
16
14
12
10
8
6
4
2
0
3
4
5
6
7
8
9
10
11
12
Month

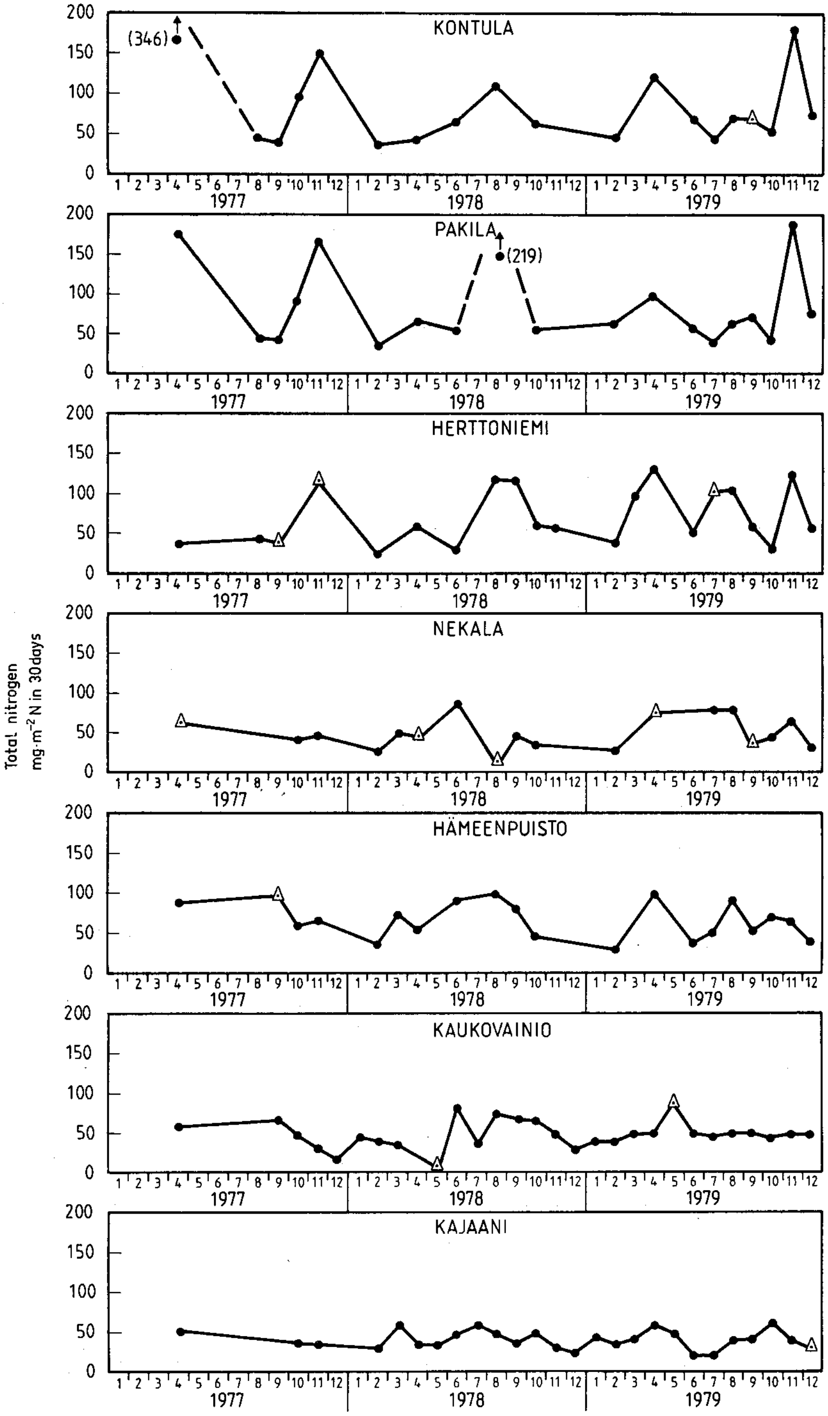
Appendix 3. Measured monthly rates of deposition in test catchments during 1977—1979.

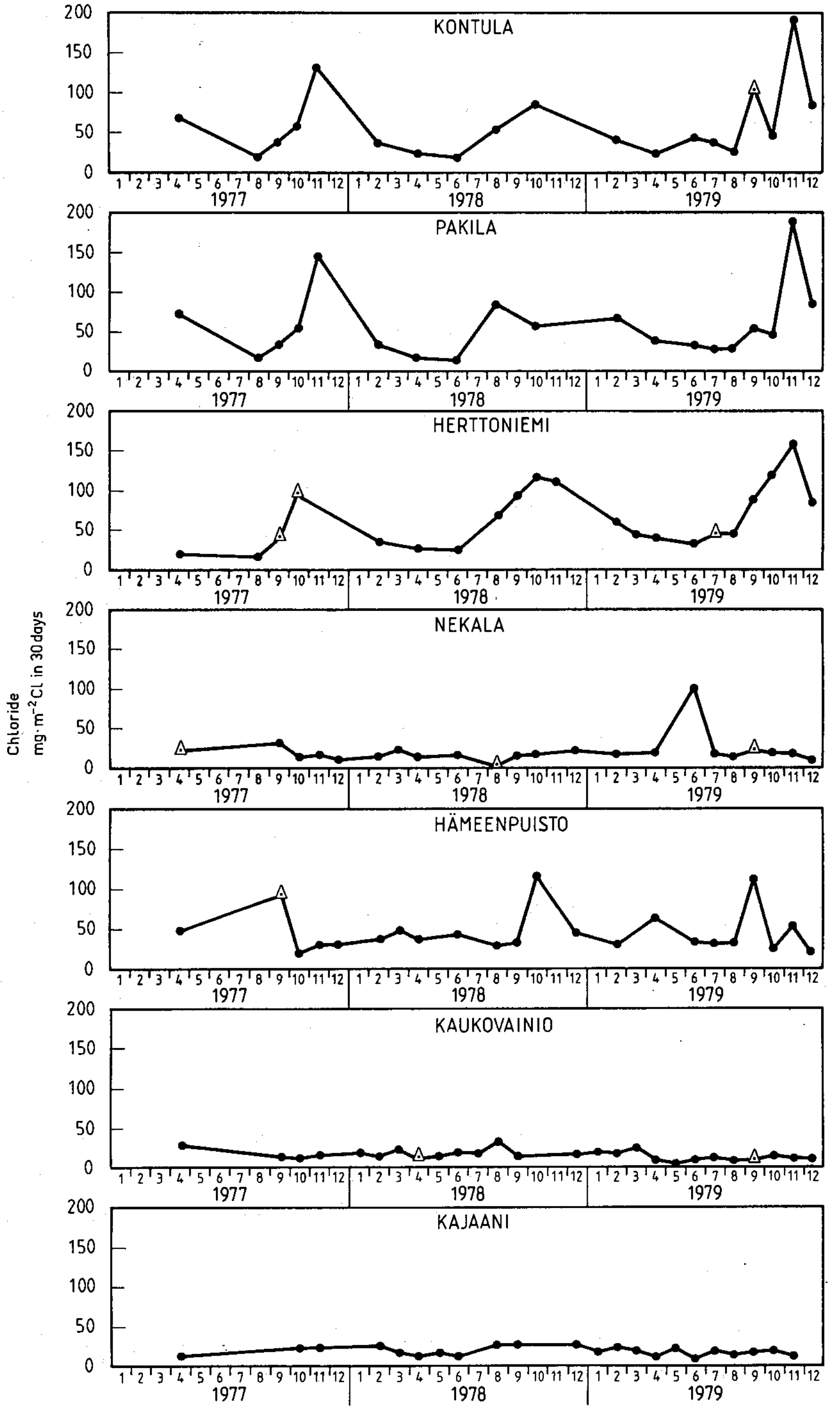


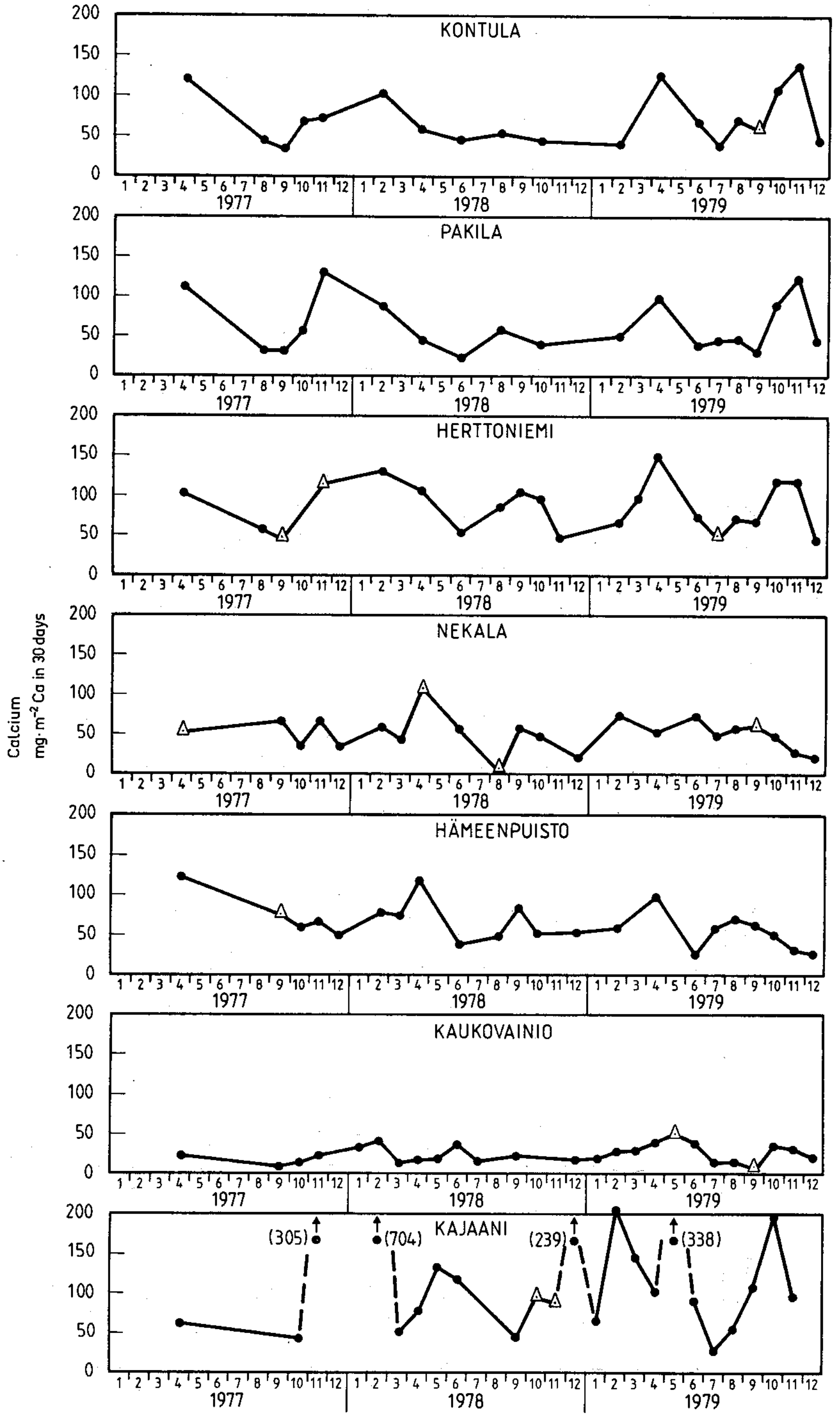

KONTULA
(4800)
PAKILA
(6040)
HERTTONIEMI
(11700)
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
Volatile deposition
mg·m⁻² in 30 days
4000
3000
2000
1000
0
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979

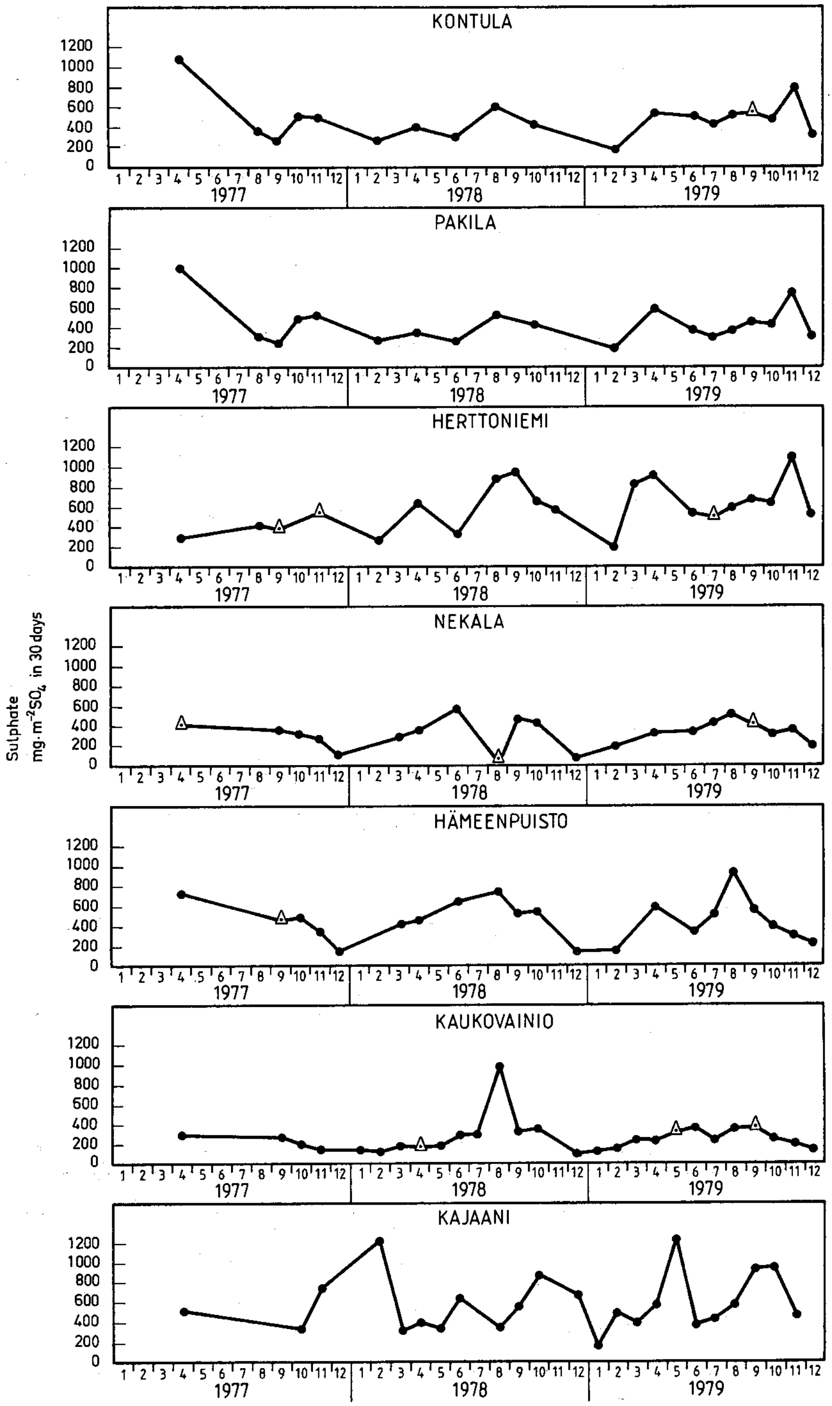

KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
Total organic carbon
mg·m⁻²C in 30 days
1000
800
600
400
200
0
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979

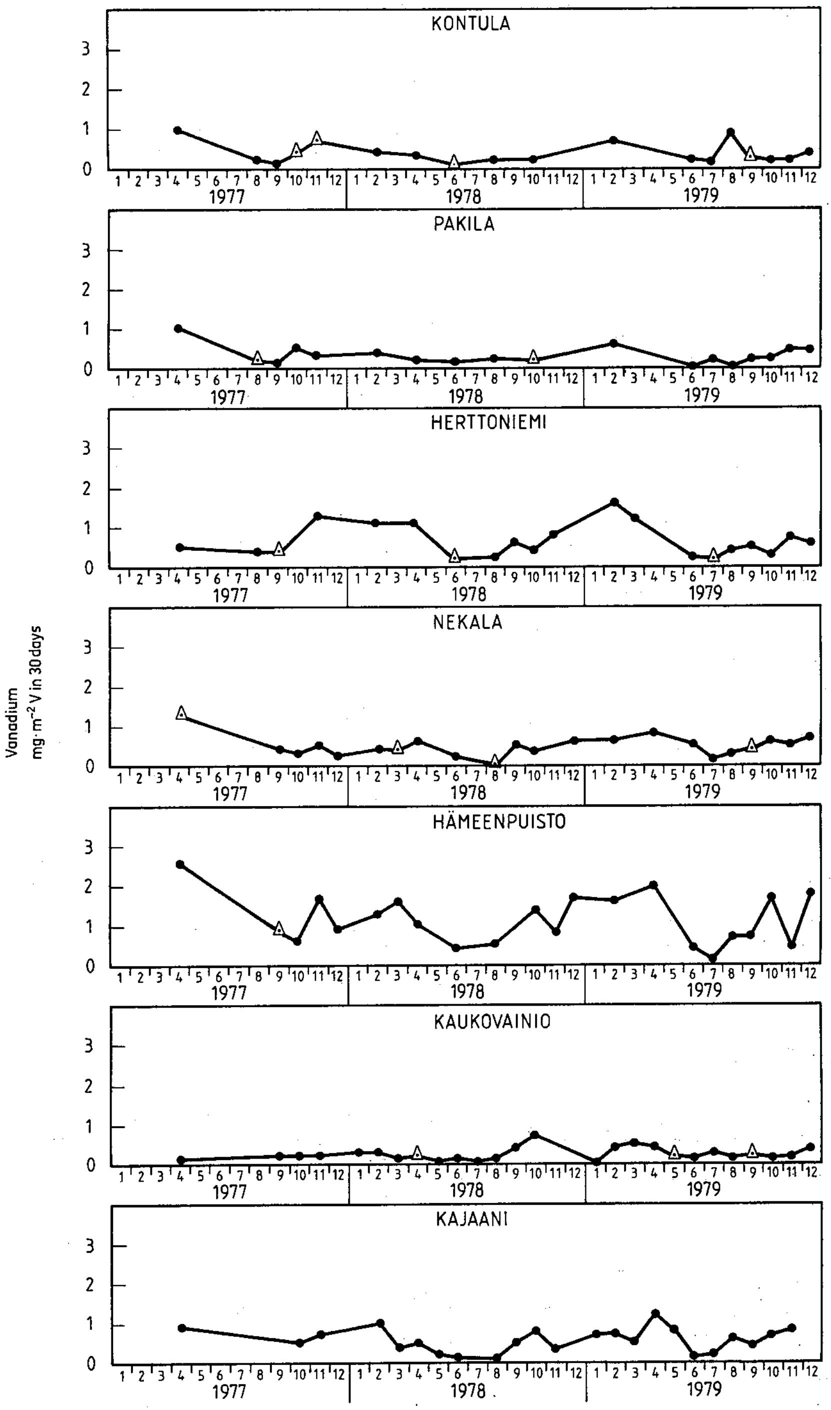

KONTULA
PAKILA
HERTTONIEMI
(28.6)
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
Total phosphorus
mg·m^{-2} P in 30 days
0
5
10
15
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979

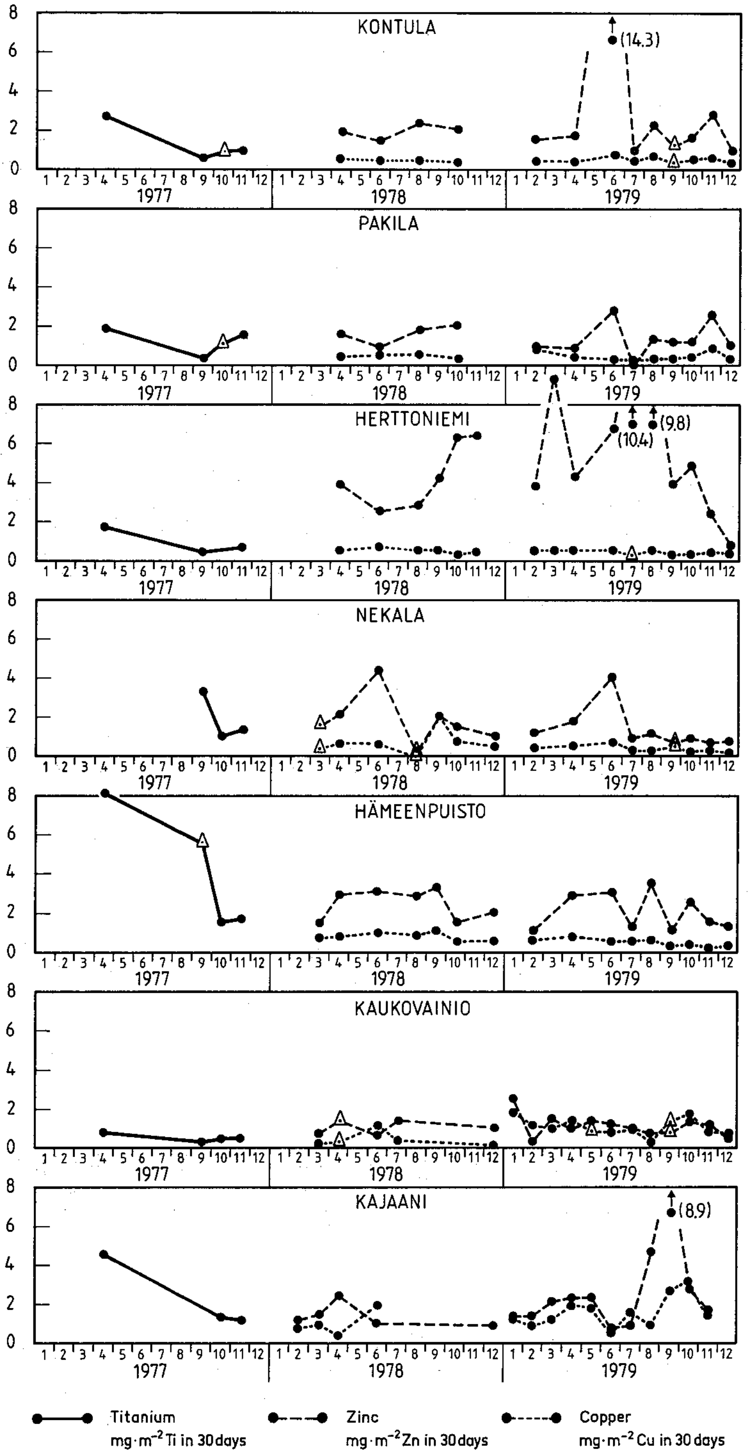

KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
(346)
(219)
Total nitrogen
mg·m⁻² N in 30days
0
50
100
150
200
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979

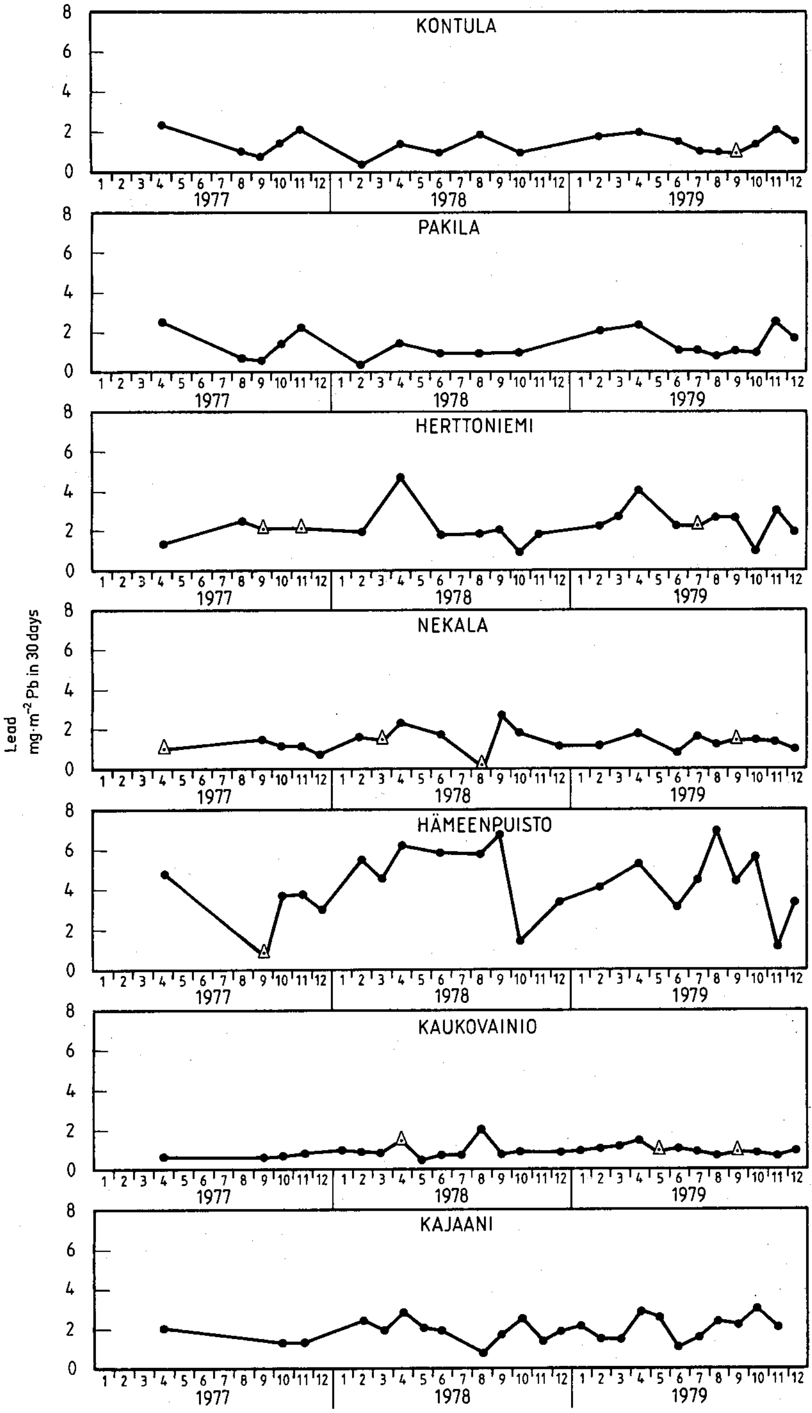

KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
Chloride
mg·m⁻²Cl in 30days
0
50
100
150
200
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979

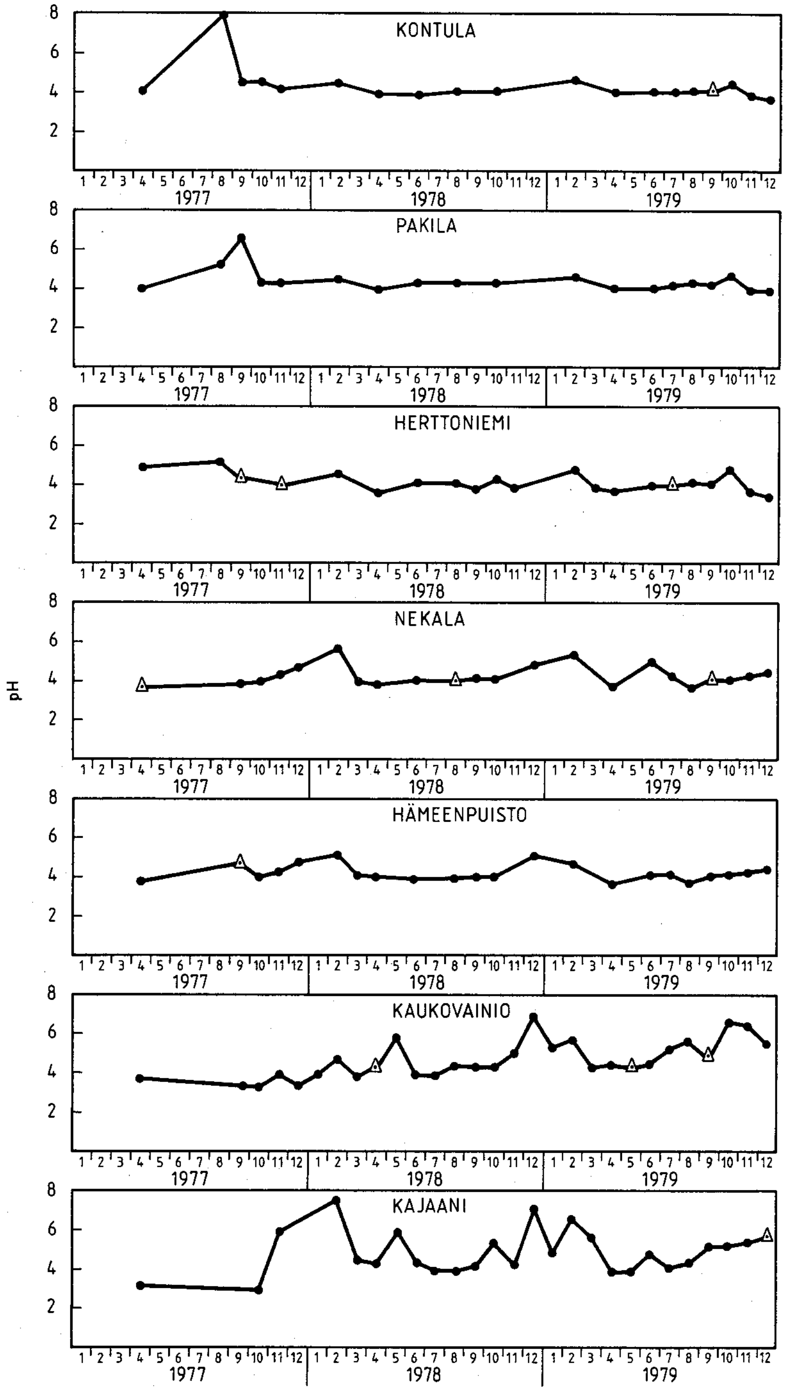

Calcium
mg·m⁻² Ca in 30 days
200
150
100
50
0
KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
(305)
(704)
(239)
(338)
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979

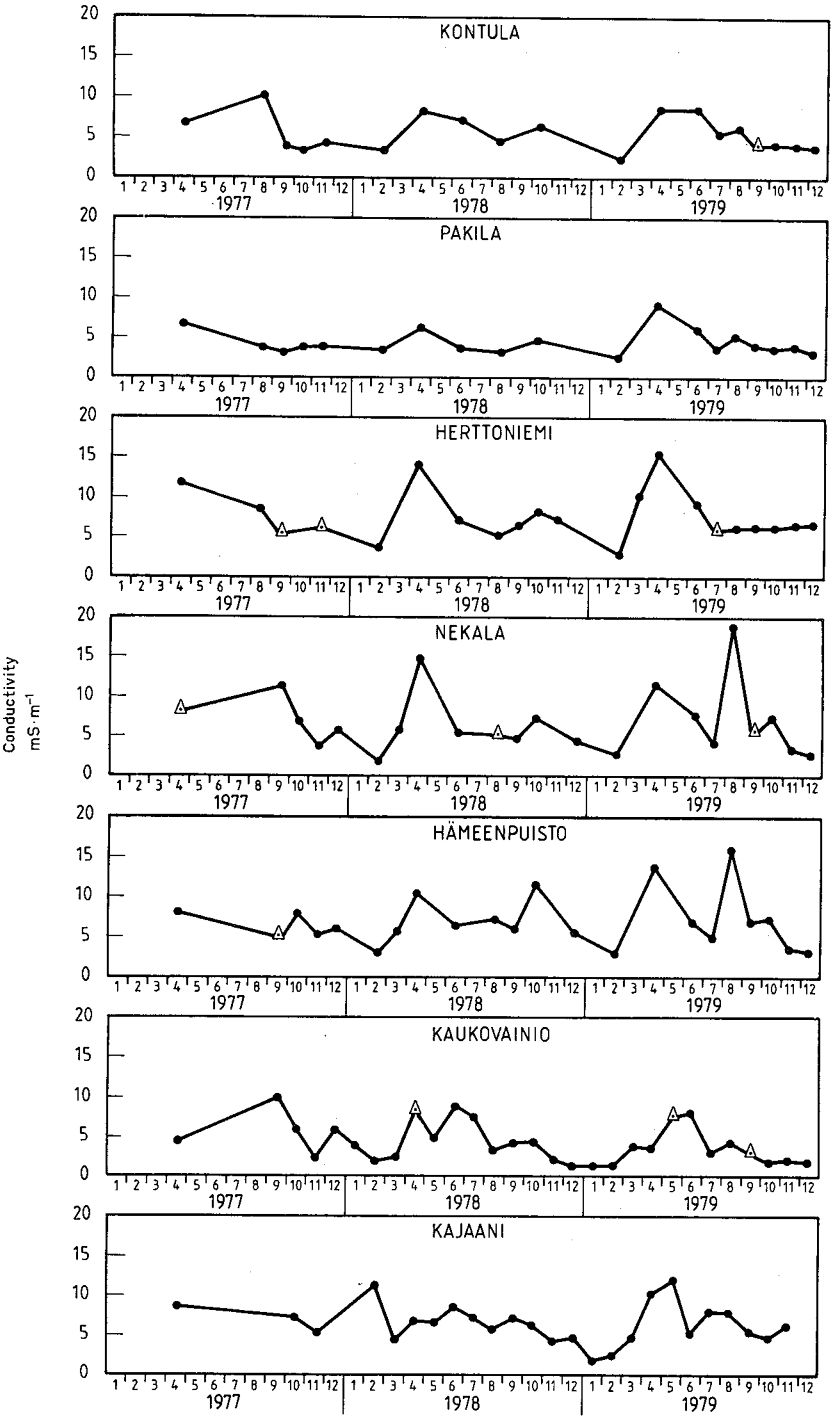

KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
Sulphate
mg·m^{-2}SO$_4$ in 30 days
0
200
400
600
800
1000
1200
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979


KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
Vanadium
mg·m⁻² V in 30 days
0
1
2
3
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979


KONTULA
(14.3)
PAKILA
HERTTONIEMI
(10.4)
(9.8)
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
(8.9)
0
2
4
6
8
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979
Titanium
mg·m⁻² Ti in 30 days
Zinc
mg·m⁻² Zn in 30 days
Copper
mg·m⁻² Cu in 30 days


KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
Lead
mg·m⁻² Pb in 30 days
0
2
4
6
8
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979


pH
8
6
4
2
KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979


Conductivity
mS·m⁻¹
KONTULA
PAKILA
HERTTONIEMI
NEKALA
HÄMEENPUISTO
KAUKOVAINIO
KAJAANI
0
5
10
15
20
1 2 3 4 5 6 7 8 9 10 11 12
1977
1978
1979

Appendix 4. Residual plots of models of total nitrogen (n = 136, 100 R^2 = 41.5 %) and vanadium (n = 136, 100 R^2 = 54.0 %) in case of combined observations.