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## Abstract

This study was part of a three-year (2004-2007) project entitled “Assessment and reduction of heavy metal inputs into Finnish agro-ecosystems” that was funded by the Ministry of Agriculture and Forestry in Finland. The aims of the project were to clarify: 1) *aqua regia* extractable trace elements in Finnish cultivated soils with the international standard method at a national level; 2) *aqua regia* and AAAC-EDTA extractable trace elements in the top- and subsoil of Finnish arable land at selected crop and dairy farms; and 3) field mass balances of trace elements on the same selected crop and dairy farms at the farm level.

The main aim of this study was to estimate field balances of trace elements at the farm level on typical crop and dairy farms in Finland in 2004. The trace elements studied were arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), nickel (Ni), selenium (Se), vanadium (V) and zinc (Zn). Five crop farms were selected from southwestern Finland that is typical of arable farming and five dairy farms from Ostobothnia that is typical of milk production. For balance estimations data on the amounts of agricultural production resources imported to the farm and products exported from the farms were collected. Samples from fertiliser products, commercial feeds, crop plants, milk, meat and manure and also from top- and subsoil were collected and analysed for the ten trace elements. The soil type on the crop farms was clay and dairy farms finesand.

Balance calculations were made with two models. The conventional field balance model called RAKAS-model was used for all the elements studied here and the European AROMIS-model for Cd, Pb, Cu, Cr, Ni and Zn. The main

differences between the models were in the leaching and erosion values. The RAKAS-model used the leaching figures measured in Nordic countries and the AROMIS-model used the leaching figures measured in other parts of Europe. Erosion was taken into account in the RAKAS-model, but not in the AROMIS-model. The AROMIS-model was able to estimate internal flows of the trace elements in homegrown feeds and manure as well as the inputs of the trace elements from unidentified sources on the dairy farms.

Field balances estimated at the farm level showed that the balances of the harmful heavy metals, Cd and Hg, were often slightly positive leading in accumulation of these metals into the soil on the crop and dairy farms. Selenium indicated a highly positive balance resulting in enrichment of this element in the soil in both farming systems. Also, the Cu and Zn balances were positive on the dairy farms but on the crop farms, clearly negative showing a depletion trend in the soil. In general, inputs and outputs of As, Cr, Ni, Pb and V were rather well balanced. However, if steel slag was used on the farm, accumulation of V into the soil was even 400 times that on the farm not used steel slag.

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*Index words: arsenic, cadmium, chromium, copper, lead, mercury, nickel, selenium, vanadium, zinc, arable land, soil, inputs, outputs, mass balance, fertiliser products, feeds, plants, crops, milk, meat, leaching*

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# Hivenalkuaineiden tilakohtaisia peltotaseita suomalaisilla kasvin- ja maidontuotantotiloilla vuonna 2004

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## Tiivistelmä

Tutkimuksen tavoitteena oli selvittää hivenalkuaineiden tilakohtaisia peltotaseita kasvin- ja maidontuotantotiloilla vuonna 2004. Tutkittavat hivenalkuaineet olivat arseeni, kadmium, kromi, kupari, lyijy, elohopea, nikkeli, seleeni, vanadiini ja sinkki. Tutkimukseen valitut viisi kasvinviljelytilaa sijaitsivat lounaisessa Suomessa tyypillisellä viljanviljelyalueella ja viisi maidontuotantotilaa Pohjois-Pohjanmaalla tyypillisellä karjatalousalueella. Taselaskelmia varten viljelijöiltä saatiin tiedot tilojen tuotantopanosten hankintamääristä ja tuotteiden myyntimääristä. Tiloilta kerättiin näytteitä lannoitevalmisteista, kaupallisista rehuista, lannasta, kasvisadoista, maidosta ja lihasta sekä peltomailta muokkauskerroksesta ja jankosta. Näytteistä analysoitiin kaikki edellä mainitut kymmenen hivenalkuainetta. Kasvintuotantotiloilla peltomaiden vallitseva maalaji oli savi ja maidontuotantotiloilla hieta.

Maatilaikohtaiset peltotaseet selvitettiin kahdella tavalla. Tavanomaisella peltotase-mallilla eli RAKAS-mallilla taseet voitiin laskea kaikille kymmenelle hivenalkuaineelle. Eurooppalaisella AROMIS-mallilla taseiden laskeminen onnistui vain kadmiumille, kromille, kuparille, lyijylle, nikkelille ja sinkille. Mallit erosivat toisistaan siinä, että RAKAS-malli huomioi eroosion yhtenä hivenalkuaineiden poistumisreittinä maasta ja AROMIS-malli taas ei huomionut eroosiota lainkaan. Toiseksi RAKAS-mallissa käytettiin pääasiassa pohjoismaisia hivenalkuaineiden huuhtoutumistietoja, kun AROMIS-malli perustui eurooppalaisiin huuhtoutumismääriin, jotka vaikuttivat Suomen olosuhteisiin suurilta.

RAKAS- ja AROMIS-malleilla saadut tilakohtaiset peltotaseet erosivat jossain määrin toisistaan. Taselaskelmat osoittivat, että maidontuotantotiloilla hivenalkuaineiden lisäykset maahan olivat keskimäärin suuremmat ja taas poistumat maasta pienemmät kuin kasvinviljelytiloilla. Siten alkuaineiden kertyminen

maahan on todennäköisempää maidontuotannossa kuin kasvinviljelyssä. Euroopassa maatiloille tulevien ja tiloilta poistuvien hivenalkuaineiden kokonaismäärät ovat keskimäärin suurempia kuin Suomessa. Lisäksi taselaskelmien vertailu osoitti, että peltojen raskasmetallipitoisuuksien kasvu on muualla Euroopassa selvästi nopeampaa kuin Suomessa.

Suomen tiukoista päästörajoituksista huolimatta haitallisimpien raskasmetallien, kadmiumin ja elohopean, lisäykset viljelymaahan olivat vuonna 2004 edelleen lievästi suurempia kuin niiden poistumat. Näiden metallien pääasiallinen lähde oli ilmasta tuleva laskeuma. Arseenin, kromin, lyijyn, nikkelin ja vanadiinin kokonaispoistumat saattoivat olla jopa hiukan suuremmat kuin niiden kokonaislisäykset. Kuitenkin silloin kun peltojen kalkitukseen käytettiin teräskuonaa, vanadiinilisäykset olivat jopa 400-kertaiset poistumiin verrattuna. Jo yhdestä teräskuonan kerta-annoksesta tuleva vanadiinimäärä saattaa kaksinkertaistaa viljelymaan vanadiinipitoisuuden. Myös suurimmat kromilisäykset tulivat teräskuonasta.

Kuparin ja sinkin lisäykset maahan olivat maidontuotantotiloilla suuremmat ja kasvinviljelytiloilla pienemmät kuin näiden hivenravinteiden poistumat maasta. Taseiden perusteella kuparin ja sinkin pitoisuudet tulevaisuudessa pienevät kasvinviljelytiloilla ja kasvavat maidontuotantotiloilla. Kasvinviljelytiloilla kuparia poistui maasta keskimäärin hiukan enemmän kuin maahan eri lähteistä tuli. Kasvinviljelytiloilla suurimmat kuparilisäykset tulivat ilmasta laskeumana ja maidontuotantotiloilla kaupallisista rehuista. Sinkki kulkeutui maatilalle molemmassa tuotantosuunnissa ennen kaikkea lannoitevalmisteissa. Seleenilisäykset maahan olivat kasvintuotantotiloilla noin viisinkertaisia ja maidontuotantotiloilla noin kymmenkertaisia seleenipoistumiin verrattuna. Mikäli tilanne jatkuu samanlaisena, viljelymaiden seleenipitoisuudet keskimäärin kaksinkertaistuvat seuraavan sadan vuoden aikana. Tuotantosuunnasta riippumatta tärkein seleenilähde olivat kivennäislannoitteet.

Tutkimus oli osa vuosina 2004–2007 toteutetusta, Maa- ja metsätalousministeriön rahoittamasta yhteistutkimushankkeesta ”Raskasmetallikuormitusten selvittäminen ja vähentäminen Suomen maatalousekosysteemeissä”. Hankkeen tavoitteena oli selvittää hivenalkuaineiden kokonaispitoisuudet viljelymaassa valtakunnallisesti käyttäen kuningasvesiuuttoa. Toiseksi mitattiin kasvin- ja maidontuotantotilojen hivenalkuaineiden kokonaispitoisuuksia peltojen muokkauskerroksesta ja jankosta. Muokkauskerroksesta tutkittiin lisäksi alkuaineiden liukoiset osuudet käyttäen hapan (pH 4,65) ammoniumasetaatti-EDTA -uuttoa. Kolmanneksi samoille maatiloille laskettiin hivenalkuaineiden tilakohtaiset peltotaseet.

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*Avainsanat: arseeni, kadmium, kromi, kupari, elohopea, lyijy, nikkeli, seleeni, vanadiini, sinkki, viljelymaa, maaperä, massatase, lannoitteet, rehut, laskeuma, kasvintuotanto, maidontuotanto, huuhtoutuminen, eroosio*

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## Foreword

This study was part of a three-year (2004-2007) project “Assessment and reduction of heavy metal inputs into Finnish agro-ecosystems” (acronym RAKAS, Project number 310925) which was jointly funded by the Ministry of Agriculture and Forestry in Finland (MMM) and participating organisations. The project was coordinated by MTT Agrifood Research Finland, Plant Production Research (Ritva Mäkelä-Kurtto). Also other scientific staff (Annukka Laitonen) from the same Department was participated in the project. Other participating organisations were MTT Laboratories (Merja Eurola), Geological Survey of Finland, GTK (Timo Tarvainen, Tarja Hatakka), Evira Finnish Food Safety Authority (Arja Vuorinen, Kimmo Suominen, Riitta Rankanen), Viljavuuspalvelu Ltd (Pirkko Laakso) and Suomen Rehu Ltd (Juha Salopelto). The project was monitored by a steering committee consisting of Senior Officer Pirjo Salminen (MMM), Senior Officer Elina Nikkola (MMM), Dr. Liisa Rajakoski (Ministry of Trade and Industry in Finland), Senior Officer Titta Pasanen (Evira Finnish Food Safety Authority), Dr. Matti Verta (Finnish Environment Institute) and Dr. Kari Kiltilä (Suomen Rehu Ltd). Aims of the project were to study: 1) total contents of trace element in Finnish cultivated soils with the international standard method at a national level; 2) total contents of trace elements in top- and subsoil of Finnish arable land on crop and dairy farms, and effects of the production sector on the possible enrichment or depletion of the trace elements; and 3) field mass balances of trace elements at the farm level on crop and dairy farms.

An aim of this study was to clarify field mass balances of trace elements on ten farms, five crop farms in south-western Finland and five dairy farms in Ostrobothnia. MTT Plant Production Research was responsible for collecting data on material flows from the farms and for top- and subsoil sampling and pretreatment. Farmers took crop, manure, and milk samples according to the guidelines given by the project. Meat samples were officially taken in slaughter houses. Soil samples were analysed for *aqua regia* extractable trace elements by GTK, for particle size distribution, humus, fertility, easily soluble trace elements by Viljavuuspalvelu Ltd (Soil Analysis Service Ltd) which analysed also manure samples for trace elements. Trace element contents in agricultural products, plants, milk and meat, were analysed by MTT Laboratories. Sampling and analysis of fertiliser products and feeding stuffs were organised by Evira.

## List of Abbreviations

Al	Aluminium
As	Arsenic
B	Boron
Cd	Cadmium
Ca	Calcium
Co	Cobalt
Cr	Chromium
Cu	Copper
Fe	Iron
Hg	Mercury
K	Potassium
Mg	Magnesium
Mn	Manganese
Ni	Nickel
P	Phosphorus
Pb	Lead
S	Sulphur
Se	Selenium
V	Vanadium
Zn	Zinc
AR	<i>Aqua regia</i>
AAAc	Acid (pH 4.65) ammonium acetate
AAAc-EDTA	Acid (pH 4.65) ammonium acetate –EDTA
Bulk dens.	Bulk density
EDTA	Na <sub>2</sub> -ethylenediaminetetra acetic acid
El. cond.	Electrical conductivity
dm	Dry matter
fm	Fresh matter
Max	Maximum
Med	Median
Min	Minimum
n	Number
r	Correlation coefficient
SD	Standard deviation
Org. C	Organic carbon
OM	Organic matter
Topsoil	Plough layer
Subsoil	Horizon under the topsoil

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# 1 Introduction

The natural sources of trace elements in arable land are soil parent rock material and volcanic eruptions. Major anthropogenic sources in the cultivated soils are fertiliser products (mineral fertilisers, liming agents, municipal and industrial sewage sludge, composts, ashes, slag, etc.), feeds and pesticides, particularly in the past and atmospheric depositions as a result of the emissions from industry, traffic, energy production and waste incineration. Heavy metals in farm animal manure used on a farm are in the internal flow at that farm. Anthropogenic inputs slowly increase element contents in the soil. This has been found in Finland, too. Concentrations of soluble cadmium in arable soil increased by 30% from 1974 to 1989 (Erviö et al. 1990). During that period, phosphorus fertilisers were manufactured in Finland from African high-Cd raw material.

Recently, much attention has been paid to soil pollution and soil protection. According to Finnish Act Nr 86 on the protection of the environment (2000), it is not allowed to leave or put into the soil waste or substances that may cause harm or risk to health or the environment. In addition, the person(s) or organisation(s) that have caused soil pollution are responsible for paying for soil restoration. In 2007, a Government Decree 2007/214 was given on the Finnish criteria for the evaluation of soil contamination and soil restoration. The decree presents soil background and threshold values as well as low and high guideline values for total contents of As, Cd, Co, Cr, Cu, Hg, Ni, Pb, V and Zn among other substances. In addition, the European Commission (EC) gave a final communication “Towards a Thematic Strategy for Soil Protection” (CEC 2006). Its purpose is to build on the political commitment to soil protection in coming years. The soil thematic strategy contains a proposal for a European act (a framework directive) that could set out common principles and a common methodology. The other thematic strategies of the European Commission cover air pollution, waste prevention and recycling, natural resources, the urban environment, pesticides and the marine environment. All these strategies will give support to sustainable development in agriculture and in the environment.

Healthy soil is one of the most valuable resources for nature and for human society. Globally, about 95% of the protein and most of the energy of the human population are obtained from traditional land-based agriculture of crops and livestock (Botkin & Keller 1995). Keeping soil clean and healthy is the best way to ensure qualitatively and quantitatively sufficient food production for future generations and for the natural flow of substances and energy on the globe. Soil quality affects not only the quality and quantity of crops, but also the quality of ground and surface waters, atmosphere and climate. However, cultivation practices with nutrient and heavy metal loads and atmospheric depositions may gradually pollute soils which may lead to soil, water and air contamination.

Many trace elements (e.g. Cr, Co, Cu, Se, Mn, Mo, Zn) are essential to metabolic functions of human and animals in small amounts but toxic in excessive amounts. Some of the trace elements are used as feed additives. Others, such as Cd, Hg, Pb and As are mainly known as harmful elements to animals. Carry over of heavy metals into agricultural products may cause a risk to human. Heavy metals are widely present in environment and can enter the food chain via animal feed. Feed plants can take some heavy metals and inorganic elements (e.g. Cd, As) from soil. For some elements (e.g. Hg), airborne deposition is the major route for pollution. Exposure to lead is mainly a result of consuming soil while grazing or foraging in contaminated soil or consumption of contaminated feed.

Arsenic, mercury, cadmium, and lead are considered as undesirable substances in animal feed, and these elements have limit values in legislation (Directive 2002/32/EC of the European Parliament and of the Council). Chromium, nickel and aluminium are not considered to be very harmful substances in animal nutrition, and they do not have any limit values. Copper, zinc, and selenium are trace elements. These compounds can be used as additives in animal feed and their limit values are defined in feed additive legislation. These statutory limits vary depending on the type of feed and on the animal for which the feed is intended.

There exist various tools to identify impacts on or trends in the trace element contents in the soil. Monitoring of cultivated soils for the trace elements reveals afterwards how much the trace elements have increased or decreased in the topsoil of arable land between the sampling years. Also, analysing both top- and subsoil for trace elements provides the possibility to see afterwards how much the trace elements have been enriched or depleted in the topsoil. By calculating the trace element balances in the agro-ecosystems, we are able to evaluate the future state of the system and how the current land use affects soil quality. The balances will show the coming trends in the soil trace element contents with the current trace element inputs and outputs. If the balance is negative, it means that inputs are lower than the outputs and in future years, trace element contents in the soil will decrease. However, if the balance is positive, it means that trace element accumulation will occur.

General principles of the heavy metal balances in agro-ecosystems have been described well by Moolenaar and Lexmond (1999). Estimations on the trace element balance in the agro-ecosystem take into account not only the agricultural actions and measures but also external actions. Agricultural actions lead to trace element inputs from the production resources imported to the farm and to the trace element outputs in the products exported. Inputs from the external pressures are atmospheric deposition and soil weathering and outputs from the external pressures are leaching and soil erosion (Wilcke & Döhler 1995, Ref.

Römkens et al. 2004). Homegrown feeds and manure used on the farm are part of internal circling and are not taken into account in the balance calculations. However, if the manure or homegrown feeds have been imported or exported, then they have to be included in the estimations. To approach sustainability, inputs and outputs should be balanced. In practice, there are two options for no accumulation of trace elements: 1) contents in applied fertilisers and soil amendments are at the same background level (“same to same” or “similar to similar” or loads are at the level of tolerable exports from soil via harvested crops, leaching or erosion (“import = export”)) (Römkens et al. 2004). Also, Moolenaar (1999) reported systems for managing heavy metals in the agro-ecosystems.

The main aim of this study was to investigate trace element inputs, outputs and field mass balances of arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), lead (Pb), nickel (Ni), selenium (Se), vanadium (V) and zinc (Zn) on the typical crop and dairy farms in Finland. The more detailed aims at the farm scale were to: 1) collect data on the amounts of agricultural production resources imported to the test farms and products exported from the test farms; 2) sample fertiliser products, crop plants, milk and meat from the test farms and analyse them for the trace elements; 3) analyse top- and subsoil of the fields on the test farms for trace elements; 4) estimate field balances of trace elements at the farm level in 2004 by using the RAKAS-model and the European AROMIS-model (AROMIS 2005, Eckel et al. 2005).

## **2 Material and methods**

### **2.1 Test farms**

The influences of the production sector on the field mass balances of trace elements were studied on five crop farms (Numbers 1-5) in southwestern Finland (Appendix 1: Fig. 1), where crop farming has been a typical production sector. Trace element investigations were also conducted on five dairy farms (Numbers 6-10) in Ostrobothnia (Appendix 1: Fig. 1) where milk production has been a typical production sector. These production sectors have traditionally been the main production sectors in Finland. In 2004, 58% of the 72 054 Finnish farms specialized in crop farming and 24% in dairy farming (Tike 2005). In 2004, 1.22 million ha of 2.22 million ha of the total cultivated land area was used for cereal production and 0.62 million ha for grassland (Tike 2005)

The total number of cattle in Finland has been about one million but this number is gradually decreasing along with the number of dairy cows, which was 324 000 in 2004 (Tike 2005). All the farms selected to this study practiced conventional Finnish farming and followed the EU's agro-environmen-

tal programme. Areas of the fields in the crop farms varied between approximately 30 and 250 ha and in the dairy farms between 30 and 70 ha. In 2004, the mean arable land per Finnish farm was 31.4 ha (Tike 2005). Dairy farms in this study had approximately 30 cows, 10 heifers, 20 calves and possibly one bull, on the average.

In 2004, the effective temperature during the growing season was at an average level in Finland (Table 1), but precipitation was locally exceptionally high, particularly in the summer and autumn (Kuusisto & Mäkinen 2005). Regional precipitation, evaporation and leaching in southwestern Finland and Ostrobothnia in 2004 are presented in the chapter 3.5.3. Mean precipitation, evaporation and leaching were about 100, 63 and 37 mm higher in southwestern Finland than in Ostrobothnia in 2004, respectively.

Annual material flows in 2004 on each test farm were clarified using a questionnaire completed by the farmers. The questionnaires also gave information on additional agricultural and environmental circumstances on the farms needed for the balance calculations. Detailed descriptions and exact geographical information for the target farms or sampling sites could not be published here due to privacy protection. According to the Commission Directive on private ownership (Directive 2003/4/EC), the farms, farmers, sampling sites and other personal or individual data must be protected and will be published in a form that the identity of the farmers or farms cannot be recognized. The RAKAS-project had an agreement with the farmers to keep all the information and data collected from the farm or farmers anonymous. Annual crop yields and milk and meat production figures in Finland in 2004 are presented in Table 2 and 3, respectively.

Table 1. Weather conditions during the growing season in Finland (Finnish Meteorological Institute 2005, Ref. Finfood 2005).

	Effective temperature °C			Precipitation mm		
	1971-2000	2003	2004	1971-2000	2003	2004
Jokioinen	1 225	1 347	1 253	346	349	478
Kauhava	1 102	1 283	1 188	285	335	414
Joensuu	1 150	1 323	1 220	336	463	399
Oulu	1 079	1 245	1 146	241	227	460
Sodankylä	759	955	790	229	246	243

Table 2. Annual crop yields in 2004 and earlier in Finland (Tike 2005).

Annual yield (exl. straw and tops)	1995	2001	2002	2003	2004
million feed units	..	6 434	6 613	6 291	6 388
feed units per ha	..	3 472	3 558	3 404	3 592
<b>Wheat</b> area (in 1 000 ha)	101	143	174	191	225
Wheat yield kg/ha	3 770	3 420	3 270	3 550	3 470
<b>Rye</b> area (in 1 000 ha)	21	29	30	31	27
Rye yield kg/ha	2 770	2 210	2 400	2 390	2 320
<b>Barley</b> area (in 1 000 ha)	516	543	522	530	532
Barley yield kg/ha	3 420	3 290	3 330	3 210	3 240
<b>Oats</b> area (in 1 000 ha)	329	416	451	425	326
Oats yield kg/ha	3 330	3 090	3 350	3 050	3 080
<b>Sugar-beets</b> area (in 1 000 ha)	35	31	31	29	30
Sugar-beets yield kg/ha	31 900	35 520	34 960	30 950	35 090
<b>Oil plants</b> area (in 1 000 ha)	85	72	66	75	68
Oil plants yield kg/ha	1 500	1 400	1 550	1 260	1 100
<b>Peas area</b> (in 1 000 ha)	5	5	5	4	3
Peas first grade product %	96	95	95	91	70
<b>Potatoes area</b> (in 1 000 ha)	36	30	30	29	27
Potatoes total yield (M kg)	798	733	780	617	619
Potatoes first grade product %	88	90	89	88	90

Table 3. Annual cattle number and milk and meat (beef and veal) production in 2004 and earlier in Finland (Tike 2005).

	1995	2001	2002	2003	2004
<b>Livestock</b>					
Cattle (in thousands)	1 148	1028	1019	1000	969
of which dairy cows	399	353	345	334	324
<b>Milk</b> (M litres)					
production	2 397	2 456	2 458	2 400	2 378
- yield litres/cow	5 981	6 932	7 117	7 251	7 404
received at dairies	2 296	2 378	2 376	2 323	2 304
<b>Meat</b> (M kg)					
Beef and veal					
total production	95	90	91	96	93

## 2.2 Soils

Soil sampling on the crop farms in southwestern Finland was carried out between 27.09.-01.10.2004 and on the dairy farms in Ostrobothnia during 04.-06.10.2004. Top- and subsoil samples were collected from 23 fields on the crop farms and from 21 fields on the dairy farms. In total 44 fields were sampled for top- and subsoil. A number of the fields studied per farm varied from three to six. A sampling site was on the same field that was sampled for the crop plant, as well. The distance of the sampling site from the railway or highway/

motorway was at least 400 m, 100 m from the road 100 m and 50 m from the electric or telephone cables.

The soil sampling system was principally the same as that used for arable land in the national soil testing (Agricultural Research Centre of Finland 1986) and in the national soil monitoring study (Sippola & Tares 1978, Erviö et al. 1990, Mäkelä-Kurtto & Sippola 2002). Soil samples from both the topsoil (plough layer) and subsoil that were collected at each sampling site were conducted on the same day as four subsamples from the four corners of the 10 x 10 m sampling area. First, the plough layer was carefully opened down to the subsoil with a spade and the depth of the plough layer was measured. After removing all plant material from the soil surface, a small slice from the whole depth was taken for a topsoil subsample with a spade. The volume of the slice was about one litre. Subsamples from the topsoil were collected into a colourless plastic vessel of five litres. After carefully removing all the topsoil material, a subsample from the subsoil was taken with an auger (a diameter 22 mm) or a spade 20 cm below the topsoil and the volume of subsoil subsample was 0.15 litre. The subsoil subsamples were collected directly to the soil boxes (600-700 ml) made of cardboard. The geographical coordinates (X, Y, Z) were taken with a GPS receiver (Trimble Geo XT or Trimble ProXR) in the middle of the sampling site. A layout of each site with its surroundings was drawn in a field book.

In the laboratory, fresh top- and subsoil samples were crushed and homogenized. Fresh top- and subsoil samples were air-dried at 35°C in an oven with air circulation and homogenized. Air-dried soils were ground, avoiding disintegration of primary particles by pressing the soil with a rotating wooden disc through a 2-mm sieve of hardened steel. The sieved soils were homogenized again and stored at room temperature in cardboard boxes for analyses.

Detailed descriptions of all the soil analysis methods used in this study were given by Hatakka et al. (2007). In this report, soil trace elements determined by *aqua regia* extraction with a modification of the international standard method (ISO 11466) are discussed.

## **2.3 Crop plants, milk and meat**

Sampling time depended on the plant species. Each crop plant was collected at the time of its normal harvest. Farmers themselves with or without the MTT staff took samples from the crop plants according to the MTT guidelines. Thus, the grass samples from the second cut were taken around midsummer and the cereal samples were taken in the summer or in early autumn.

Grass samples were collected from the test field as four subsamples, 150-200 g of grass, that were cut with scissors at least five cm above the soil surface to avoid

dead and dirty plant parts and soil contamination. The subsamples were combined to form one grass sample. In the laboratory, the samples were allowed to dry at room temperature for a couple of days and then in an oven at 60°C with air circulation.

After harvesting, the farmers took cereal samples of 1-2 kg of grains, representing the same field that was sampled for soil. After drying at the farm, the cereal samples were sent to the MTT Laboratory for analysis. All the plant samples were homogenized and ground in a hammer mill of pure carbon steel to pass a 2-mm sieve. The dried samples were stored in plastic bags at a room temperature until analysed.

From the crop farms, a total of 23 plant samples were taken from the fields to be sampled for top- and subsoil and 8 additional plant samples. From the dairy farms, 21 plant samples were taken from the fields to be sampled for the top- and subsoil along with 6 additional plant samples.

Milk samples were collected twice a year; in April before the outdoor feeding season started (winter milk) and in September before the indoor feeding season (summer milk). The samples were taken directly from the farm tank to the clean plastic container and frozen. Meat samples were taken in slaughterhouses from four points on the carcass (chuck, brisket, steak, outer fillet) that were pooled together. Meat was cut into small pieces with titanium knife to avoid contamination, freeze dried and homogenized with a blender. The silage samples were taken at the same time as the milk from several places in the silo or container. Silage was dried at 60°C and milled. All samples were stored in -25°C until analysed.

Heavy metals (As, Cd, Pb, Cu, Ni, V, Zn) in plants, milk and meat were determined by inductively coupled plasma mass spectrometry (ICP-MS). A freeze dried 0.5-2 g sample was digested in concentrated nitric acid, diluted with MILLI-Q purified water and filtrated (digestion: Kumpulainen & Paakki 1987). Elemental concentrations were measured by ICP-MS (Perkin Elmer Elan 6000). Calibration were performed by external standards, except for As, where the method of standard additions was used.

Se was analysed by an electrothermal atomic absorption spectrometry (ETAAS) method (Kumpulainen et al. 1983). The dried samples were digested in a mixture of concentrated HNO<sub>3</sub>, HClO<sub>4</sub> and H<sub>2</sub>SO<sub>4</sub>. Se was reduced to Se(IV) with hydrochloric acid, chelated with ammonium pyrrolidine dithiocarbamate (APDC) and extracted into methyl isobutyl ketone (MIBK) for AAS measurement ( $\lambda$  196.0 nm).

For Hg determinations, <1 g of sample was digested (60 ± 2°C) in a mixture of concentrated sulfuric acid and nitric acid and for further oxidation and prevention of Hg loss, potassium permanganate solution was added. Hg was measured

with the Varian M-6000A Cetac mercury analyser ( $\lambda$  253.6 nm) with the cold vapour technique. Tin chloride  $\text{SnCl}_2 \times 2 \text{H}_2\text{O}$  was used as reductant.

Cr was determined by the electrothermal atomic absorption spectrometry (ETAAS) method. The sample was digested in microwave oven.

The accuracy of the analytical methods was tested by determining certified or in-house reference materials of suitable matrix in every batch of samples. The samples were analysed as duplicates and all laboratory vessels were acid washed and rinsed with MILLI-Q purified water. The Chemistry Laboratory of Agri-food Research Finland follows the requirements of the Standard SFS-EN ISO/IEC 17025 and is the testing laboratory accredited by the Centre of Metrology and Accreditation. The following methods used in this study were accredited: As, Se, Cd, Pb, Cu, and Zn. The detection limits were: As 0.010, Se, 0.010, Cd 0.0007, Pb 0.006, Cu 0.4, Zn 0.3  $\text{mg kg}^{-1} \text{ dm}$ .

## 2.4 Fertiliser products

All the fertiliser products sampled from the test farms were inorganic field or grass fertilisers produced by Kemira GrowHow Ltd, because fertilisers from other producers were not used on these farms. Fertiliser sampling on the farms were carried out by the Evira, Finnish Food Safety Authority (in 2004 KTTK) according to the standard method SFS-EN 1482:1996. Sampling time was spring 2004 before sowing time.

At the crop farms, fertiliser samples were taken between 27.04.-29.04.2004. Each farm used 2-3 different NPK-fertilisers. The total number of fertilisers collected from the crop farms was 12. Due to the homogeneity of the fertiliser material, only seven fertiliser samples were analysed for trace elements (Cd, Hg, Zn, Pb, Ni, Cu, V, Cr, As and Se) at the Evira (in 2004 KTTK) (Table 4).

Table 4. Fertiliser samples collected from the five crop farms in 2004 and analysed for trace elements at the Evira (in 2004 KTTK).

Sample Nr.	Date of sampling	Trade mark of fertiliser
1442 RAKAS	28.04.04	Kevätviljan Y1
1443 RAKAS		Kevätviljan Y2
1428 RAKAS + NPK	27.04.04	Kevätviljan Y3
1430 RAKAS		Kevätviljan Y6
1426 RAKAS + NK	27.04.04	Suomensalpietari
1444 RAKAS + NPK	29.04.04	Kevätviljan Y4
1441 RAKAS	28.04.04	Syysviljan Y1

At the dairy farms, fertiliser samples were collected between 06.05.-11.05.2004. Each farm used 3-4 different NPK-fertilisers. Since the farms used the same fertilisers, there was a farm where the fertiliser sample was not taken at all. The total number of fertilisers collected from the crop farms was 15. Due to the homogeneity of the fertiliser material, only eight fertiliser samples were analysed for trace elements (Cd, Hg, Zn, Pb, Ni, Cu, V, Cr, As and Se) at the Evira (in 2004 KTTK) (Table 5).

Table 5. Fertiliser samples collected from the four dairy farms in 2004 and analysed for trace elements at the Evira (earlier KTTK).

Sample Nr.	Date of sampling	Trade mark of fertiliser
1642 RAKAS	06.05.04	Syysviljan Y1
1645 RAKAS		Nurmen Y2
1652 RAKAS	11.05.04	Suomensalpietari
1651 RAKAS		Kevätviljan Y4
1641 RAKAS	06.05.04	Nurmen Y1
1636 RAKAS		Nurmen NK2
1649 RAKAS	07.05.04	Nurmen NK 1
1650 RAKAS		Kevätviljan Y5

For the trace element determinations the samples of inorganic fertilisers were digested by nitric acid. Concentrations of Cu, Cr, Ni, Zn and V were measured by ICP-OES (KTTK's/Evira's method code P03356) and of Cu, Cu, Zn (Evira's method code) and Cd and Se by AAS (KTTK's/Evira's method code P10715). In addition, the Evira analysed some slag samples for the trace elements.

## 2.5 Cattle manure

For collecting samples from cattle manure on the dairy farms, special boxes made of colourless thick plastic were sent from Viljavuuspalvelu (Soil Analysis Service in Finland) to the farmers in spring 2004. The boxes (about one litre) also contained guides for sampling. Sampling was carried out using the same method recommended by the Viljavuuspalvelu (Soil Analysis Service in Finland) and demanded by the national agri-environmental programme. The guides were also available on the home pages of the Viljavuuspalvelu ([www.viljavuuspalvelu.fi](http://www.viljavuuspalvelu.fi)). The farmer took one manure sample in early summer before grazing time to indicate indoor feeding and one manure sample in late autumn at the end of grazing time to indicate outdoor feeding. The samples were sent to Viljavuuspalvelu for analysis.

The dry matter and volume weight of manure were analysed on the wet samples. For the other analyses, the samples were dried at 105°C, ground and homogenized. Standard methods SFS 3044:1980 and SFS 3047:1980 were used for the heavy metals As, Cd, Cr, Cu, Hg, Ni, Pb, Zn and V. Bulk density was

determined by weighing a 200 ml wet, moderately compressed sample. All the results were calculated based on dry matter, wet weight and on volume bases. All the manure analysis was done by the Viljavuuspalvelu and presented in Table 6. Analytical results on manure samples excluding the results on trace elements are presented in Table 7.

Table 6. Analysis methods used in this study to determine trace elements in manure samples.

Element, fraction, concentration unit	Method
Nitrogen (N), soluble, %	Kjeldahl-method. SFS 5505: 1988
Nitrogen (N), total, g/kg dm	
Phosphorus (P) total, g/kg dm	Dry ashing 550°C, HCl-extraction, measurements by ICP-AES. ISO 5516: 1978
Potassium (K), total, g/kg dm	
Selenium (Se), total, mg/kg dm	
Copper (Cu) total, mg/kg dm	HNO <sub>3</sub> -extraction, measurements by ICP-AES. SFS 3044: 1980 and SFS 3047: 1980
Zinc (Zn), total, mg/kg dm	
Nickel (Ni), total, mg/kg dm	
Arsenic (As), total, mg/kg dm	
Cadmium (Cd), total, mg/kg dm	HNO <sub>3</sub> -extraction, measurements by GFAAS. SFS 3044: 1980 and SFS 3047: 1980
Chromium (Cr), total, mg/kg dm	
Lead (Pb), total, mg/kg dm	
Vanadium (V), total, mg/kg dm	
Mercury (Hg), total, mg/kg dm	
	HNO <sub>3</sub> -extraction, measurements by FIAS. SFS 3044: 1980 and SFS 3047: 1980

Table 7. Manure samples collected from the five dairy farms (6-10) in Ostrobothnia in 2004 and total contents of nitrogen (N), phosphorus (P) and potassium (K) and concentrations of soluble N (g kg<sup>-1</sup> dm) of the samples.

Sample Nr.	Sampling	Manure type 1)	Farm Nr.	Dm %	Volume weight kg/m <sup>3</sup>	N sol.	N tot.	P tot.	K tot.
1	07.06.2004	1	6	6.3	920	34	55	8.8	54
2	22.10.2004	1	6	7.8	1000	30	55	8.8	63
3	03.06.2004	1	7	3.7	1000	46	83	11	72
4	11.10.2004	1	7	2.1	1000	45	61	12	84
5	11.06.2004*	1	8	1.8	1000	73	96	8.6	92
6	05.10.2004	1	8	6.5	950	24	45	11	29
7	16.05.2004**	1	9	2.4	1000	61	120	11	96
8	No sample		9						
	<b>Mean</b>	<b>1</b>		<b>4.4</b>	<b>981</b>	<b>45</b>	<b>74</b>	<b>10</b>	<b>70</b>
9	20.05.2004	2	10	13.4	880	15	39	11	11
12	22.10.2004	2	10	16.1	940	15	36	4.6	39
	<b>Mean</b>	<b>2</b>		<b>14.8</b>	<b>910</b>	<b>15</b>	<b>37.5</b>	<b>7.8</b>	<b>25</b>
10	20.05.2004	3	10	17.5	930	14	33	5.1	30
11	20.05.2004	4	10						

1) Manure types: 1 = dairy slurry; 2 = faeces and urine in peat; 3 = faeces; 4 = peat

## 2.6 Commercial feeds

Feed samples (Table 8) from the test farms were collected in spring 2004 according to the guidelines of the Plant Production Inspection Centre that are based on the Commission Directive 76/371/EEC.

Table 8. Feeds (n = 13) sampled from the five dairy farms in 2004 and analysed for Cd, Pb, Hg, Cu, Cr, Ni, Zn, V, As and Se by Evira (in 2004 KTTK).

Sample Nr.	Time of sampling	Producer / Date imported to farm	Trade mark	Moisture content %
2004-01786-001	06.05.	Suomen Rehu Oy/ 15.12.03	Huippu Krossi 23	11.9
2004-01790-001	06.05.	Rehu Raisio/ 05.04	Aseto-Melli	9.8
2004-01812-001	06.05.	Rehu Raisio/ 08.04.04	Amino-Maituri	10.4
2004-01814-001	06.05.	Rehu Raisio/ 11.03	Mulli-Melli	5.3
2004-01816-001	06.05.	Suomen Rehu/ 16.04.04	Kesä-Namino	4.6
2004-01818-001	07.05.	Kinnusen Mylly/ 23.03.04	Tähti-145	12.7
2004-01820-001	07.05.	Rehumelica Oy/ 04.10.03	Onni-Kivennäinen	0.9
2004-01823-001	07.05.	Rehumelica Oy/ 23.12.03	Milkkeri melassoitu rypsi	11.2
2004-01830-001	07.05.	Rehu Raisio Oy/ 02.05.04	Maituri 10000	9.9
2004-01831-001	07.05.	Rehu Raisio Oy/ 02.05.04	Rouhe-Tiiviste	11.2
2004-01845-001	11.05.	Rehumelica Oy/ 30.12.03	Melassileike	11.9
2004-01846-001	11.05.	Tilan raaka-aineet lis. öljy/ 21.02.04	Tilaseos väkevämpi <sup>1)</sup>	11.8
2004-01849-001	11.05.	Suomen Rehu Oy/ 23.03.04	Viher Hertta-Minera Muro	1.9

<sup>1)</sup> Feed manufactured by a mobile mixer

For cadmium and lead analysis, 0.5 g of homogenised feed was digested in 5 ml of 10% HNO<sub>3</sub> in a Teflon-sealed vessel under high pressure (55 bar) for 30 min. The cadmium and lead content of the feed samples were analysed with atomic adsorption spectrometer (AAS) at wavelengths of  $\lambda_{Cd} = 228.8$  nm and  $\lambda_{Pb} = 283.3$  nm. Nickel was analysed by flame atomic adsorption spectrometer (Unicam 939) at 232.0 nm

For arsenic, selenium, nickel, copper, zinc, chrome and vanadium analysis, 5 g homogenised sample was allowed to stand in a 100 ml glass beaker in 65% HNO<sub>3</sub> for 16 h at room temperature. The mixture was heated to 60°C for 1 h,

to 100°C for 1h and to 120°C for 1 h. After cooling to 50°C, 50 ml of ion exchanged H<sub>2</sub>O and 3 ml of H<sub>2</sub>O<sub>2</sub> were added, and the temperature was raised again to 100°C for 40 min. The mixture was filtered (589/2 ash less filter paper Ø 125 mm) and the volume was adjusted to 100 ml in a volumetric flask at room temperature.

The acid liquid was injected through a Perkin Elmer Flow Injection Hydride Analysis 100 (FIAS) unit, which reduced the As(V) to As(III) and Se(VI) to Se(IV). The gas was transported through FIAS unit to atomic-absorption spectrometry (AAS, Perkin-Elmer 4100) where the elements of arsenic (As) and selenium (Se) were analysed at wavelengths of  $\lambda_{As} = 193.7$  nm and  $\lambda_{Se} = 196.0$  nm. Concentrations of copper (Cu), zinc (Zn), chromium (Cr) and vanadium (V) were analysed by an inductively coupled plasma equipment (ICP-Optima 3300DV) at wavelengths of  $\lambda_{Cu} = 327.4$  nm,  $\lambda_{Zn} = 213.8$  nm,  $\lambda_{Cr} = 267.7$  nm and  $\lambda_{V} = 290.8$  nm.

The mercury (Hg) concentration of a homogenised 5 g sample was analysed using Mercury Analyser atomic absorption spectrometry at a dry condition at wavelength of  $\lambda_{Hg}$ : 253.6nm.

## 2.7 Mass balance calculations

Balance calculations were made by two different models. The RAKAS-model was used for all the ten trace elements studied here. The AROMIS-model developed for EU's AROMIS-project was applicable only for Cd, Cr, Cu, Ni, Pb and Zn (AROMIS 2005).

### 2.7.1 RAKAS-model

In the simple RAKAS-model, based on the data on the annual amounts of production resources and products obtained from the farmers and on the analytical results on trace element contents in the resources and products obtained in this study, the trace element inputs and outputs were calculated, respectively. In this way, the trace element outputs in manure were also estimated. Trace element inputs from the annual atmospheric depositions were based on the measurements from the Meteorological Institute of Finland, as described in the Chapter 3.4.3. The trace element inputs from various sources and the outputs in crop plants, milk, meat and manure were the same in the RAKAS- and AROMIS-model. National or Swedish research results were used for leaching values in the RAKAS-model, as presented in the Chapter 3.5.3. The trace element outputs via eroded soil material were based on the mean trace element content of each study farm. Erosion calculations are described in more detailed in Chapter 3.5.4. Also, weathering (Chapter 3.4.4) of the soil parent material is

an input of trace elements, but it was not taken into account in the RAKAS- nor AROMIS-models.

### **2.7.2 AROMIS-model**

The European AROMIS-model (Version rev04, Appendix 2) was used to calculate the total heavy metal balances for Cd, Cr, Cu, Zn, Ni and Pb. The AROMIS-model is an Excel-based tool used to calculate the heavy metal balances for crop ([www.ktbl.de/english/aromis/forum/blance/plant.xls](http://www.ktbl.de/english/aromis/forum/blance/plant.xls)) and dairy farms ([www.ktbl.de/english/aromis/forum/blance/animal.xls](http://www.ktbl.de/english/aromis/forum/blance/animal.xls)). This version of the model was obtained directly from Dr. Henning Eckel, Coordinator of the EU concerted action AROMIS (Assessment and reduction heavy metal inputs into agro-ecosystems). Most of the member states of the EU participated in the AROMIS-project (AROMIS 2005).

There were mainly two differences between the AROMIS- and RAKAS-model: leaching and erosion. In the AROMIS-model, leaching values were based on the European measurements and were clearly higher than those used in the RAKAS-model. Trace element outputs via erosion were not taken into account in the AROMIS-model. The AROMIS-model was also able to calculate the internal flows of trace elements in the homegrown feeds and manure used on the dairy farms themselves (Chapter 3.7). In addition, the AROMIS-model automatically estimated unidentified inputs of trace elements on the dairy farms (Chapter 3.7).

## **2.8 Statistical methods**

For statistical processing, Microsoft Office Excel 2003 software was used. If the results were below the detection limit, half of the detection limit value was used in calculations.

# **3 Results and discussion**

## **3.1 Soils**

The soils between the farming types clearly differed in the particle size distribution (Table 9 and 10). On the crop farms, the mean clay (<0.002 mm) content was 45% and in the dairy farms 10%, while the mean finesand (0.02-0.2 mm) content were 15% and 60%, respectively. Also, according to Kurki (1972), the clay soil was the dominating soil type in both the top- and subsoil in southwestern Finland and in the finesand soil in Ostrobothnia (Appendix 1: Fig. 1 and 2).

The clay soils in southwestern Finland contained much more Al and Fe than the finesand soils in Ostrobothnia (Hatakka et al. 2007). On the crop farms, the soil humus content and electrical conductivity were slightly lower than those on the dairy farms, on average (Hatakka et al. 2007).

Table 9. Mean particle size distributions (%) from 23 fields (plough layer) on the five crop farms (1-5) in southwestern Finland in 2004.

Crop farms									
Particle size distribution, %									
Fraction	Clay	Silt		Finesand		Sand		Gravel	
Particle size, mm	<0.002	0.002 - 0.006	0.006 - 0.02	0.02 - 0.06	0.06 - 0.2	0.2 - 0.6	0.6 - 2.0	2.0 - 6.0	6.0 - 20.0
Symbol	S	Fine HHS	Coarse KHS	Fine HHT	Coarse KHT	Fine HHK	Coarse KHK	Fine HSR	Coarse KSR
Farm									
1	46	20	19	9	4	2	2	0	0
2	51	20	14	9	3	2	1	0	0
3	23	10	7	7	13	17	12	8	2
4	36	29	21	6	5	3	1	0	0
5	56	9	7	11	13	2	2	0	0
Mean	45	17	13	9	7	4	3	1	0

Table 10. Mean particle size distributions (%) from 21 fields (plough layer) on the five dairy farms (6-10) in Ostrobothnia in 2004.

Dairy farms									
Particle size distribution, %									
Fraction	Clay	Silt		Finesand		Sand		Gravel	
Particle size, mm	<0.002	0.002 - 0.006	0.006 - 0.02	0.02 - 0.06	0.06 - 0.2	0.2 - 0.6	0.6 - 2.0	2.0 - 6.0	6.0 - 20.0
Symbol	S	Fine HHS	Coarse KHS	Fine HHT	Coarse KHT	Fine HHK	Coarse KHK	Fine HSR	Coarse KSR
Farm									
6	14	20	29	21	10	4	3	0	0
7	5	8	21	29	35	2	1	0	0
8	24	22	19	14	11	6	4	1	0
9	5	3	6	21	61	4	1	0	0
10	2	0	3	15	77	3	1	0	0
Mean	10	11	16	19	38	4	2	0	0

Means of *aqua regia* extractable trace elements in the top- and subsoil of the fields on the five crop and five dairy farms are presented as statistical indicators in Appendix 3 (Table 11). Clearly higher contents of all the elements occurred

in both the soil horizons of the clay soils on the crop farms than in the finesand soils on the dairy farms. Trace element contents in the clay soils were two to three times the respective contents in the finesand soils, on average. Thus, the difference in the trace element contents between the farm types seemed to be due more to the difference in the geology of the regions than to the difference in the land use or farming. All the crop farms were located in the clay region in southwestern Finland and dairy farms were in the finesand region in Ostrobothnia. However, the dominant soil type on the dairy farm (Nr. 8) was organic soil.

Based on the enrichment factors estimated by Hatakka et al. (2007), enrichment of trace elements in the topsoil was clearly more pronounced on the dairy farms than on the crop farms, on average (Appendix 3: Table 3). On the dairy farms, Cd, Cr, Cu, Pb, Hg, Se, V and Zn were enriched, and no trace element was depleted. On the crop farms, Cd, Pb, Hg, Se and Zn were also enriched, while As, Cr, Cu and Ni were depleted. When the amounts enriched in the topsoil per ha were compared between the farm types, more Cd, Pb and Hg was enriched in the clay soils in southwestern Finland than in the finesand soils in Ostrobothnia. Enrichments of Cd, Pb, Hg and Zn were 0.239, 4, 0.042 and 17 kg per ha on the crop farms and on the dairy farms 0.124, 3, 0.039 and 22 kg per ha, respectively (Hatakka et al. 2007). On the crop farms As, Cr, Cu and Ni were depleted by 3, 37, 11, 13 kg per ha, respectively, and in the dairy farms, Cr, Cu, Se and V were enriched by 3, 7, 0.121 and 26 kg per ha, respectively.

According to Swedish studies (Eriksson et al. 1997), trace element concentrations were often somewhat higher in the subsoil than topsoil. Important excep-

Table 11. Means of *aqua regia* extractable (ISO 11466) trace element contents (mg kg<sup>-1</sup>) in the top- and subsoil of 23 fields on five crop farms in southwestern Finland and 21 fields on five dairy farms in Ostrobothnia in 2004.

	Crop farms		Dairy farms	
	Topsoil	Subsoil	Topsoil	Subsoil
As	5.9	7.2	3.1	3.1
Cd	0.227	0.116	0.112	0.061
Cr	68	85	25	24
Cu	32	37	14	11
Pb	20	18	6.7	5.3
Hg	0.047	0.028	0.033	0.017
Ni	33	39	11	11
Se	- *	0.25	-*	0.13
V	90	90	43	32
Zn	114	106	42	33

\* Over 50% of the values were below the detection limit, 0.2 mg kg<sup>-1</sup> and the mean value was not computed.

tions were Cd, Hg, and Pb, for which there has been a significant input to Swedish agricultural soils in the form of deposition from long-range-transport air pollution. The higher concentrations of many elements in the subsoil than in topsoil was observed for As, Cu, Cr and Ni based on the analyses of samples from environmental monitoring of agricultural soils. Several possible explanations were presented in that report. One hypothesis was that the net supply of agricultural soils has been low. During 1900s, the net supply of As, Cu, Cr and Ni represented only a few per cent of topsoil concentrations (Andersson 1992). A contributory reason to why concentrations tend to be higher in subsoil than in topsoil may be that Swedish soils, for geological and / or colloid-chemical reasons, have in many cases higher clay content in the subsoil than the topsoil, as in Finland as well. The higher clay content usually means a higher metal content. Another factor that influences the metal content is the weathering the topsoil has been exposed to since the ice age or since the land was raised above sea level. Weathering releases metals in a soluble form, which means these metals can leach down to the subsoil or out of the soil profile. In contrast, many soils have topsoil that can be described as organic, while the subsoil is a mineral soil type. There are often quite large differences in element concentrations in the topsoil between the different soil types. The highest concentrations of most of the trace elements were found in the heavy clay soils and the lowest in the sandy soils.

## **3.2 Trace elements in production resources**

### **3.2.1 Fertiliser products**

In 2004, the test farms studied used mineral fertilisers, lime, calcites and slag as fertiliser product, but not ashes or municipal sewage sludge. However, all these fertiliser products are discussed below.

#### **3.2.1.1 *Mineral fertilisers***

Continuous application of fertilisers that contain trace levels of harmful heavy metals may result in the contamination of cultivated soil, thereby reaching the food chain through plant uptake. Fertiliser products are known to be one major source of cadmium into cultivated soils. Heavy metals find their way into mineral fertilisers from parent rock materials, particularly from rock phosphate depending on its geologic characteristics and also from equipment corrosion, catalysts and reagents, fillers, coaters, and conditioners. Thus, P containing mineral fertilisers can be rich in heavy metals. In the 1970's and 1980's, rock phosphates from North Africa were imported into Finland. In Finland, a rock phosphate mine was opened in the beginning of the 1980's in Siilinjärvi. This mine produces rock phosphate of igneous origin. Finnish rock phosphate is known to contain very little harmful heavy metals.

The use of fertilisers has decreased markedly since the 1980's and is now little more than 700 000 tonnes annually (Table 12). For instance, the use of P has been halved until today (Table 12). Implementation of the EU agro-environmental programme into the Finnish agriculture system in the 1990's has continuously diminished the application of all N, P and K macronutrients as well as Ca (liming). The use of lime has decreased more drastically than the use of fertilisers (Table 12). Since 1994, the use of lime has decreased nearly to one third. This means that today, on average, 240 kg of lime is applied annually per hectare.

Table 12. Use of fertilisers and lime in Finland (Tike 2005).

Fertilisers		1994	2001	2002	2003	2004
total use <sup>1)</sup>	Million kg	861	770	753	743	721
nutrients	kg ha <sup>-1</sup>	153	114	115	114	108
nitrogen (N)	kg ha <sup>-1</sup>	94	78	78	78	74
phosphorus (P)	kg ha <sup>-1</sup>	19	10	10	10	9
potassium (K)	kg ha <sup>-1</sup>	40	26	27	27	26
Fertilisation costs	MEUR	249	163	180	177	175
Lime, total use	Million kg	1 217	1 105	1 197	555	478 2)

<sup>1)</sup> During the fertilising period 1.7.-30.6; <sup>2)</sup> Pure lime

All the mineral fertilisers used by the test farms and studied here were manufactured by Kemira GrowHow Ltd. Trace element contents measured here are presented in Table 13. The contents of the most harmful elements, Cd, Pb, Hg and As in the mineral fertilisers were very low and under their analytical detection limits of 0.2, 1, 0.01 and 0.5 mg kg<sup>-1</sup>, respectively. Nickel, Cr, Cu and V contents were mostly <5 mg kg<sup>-1</sup>. Selenium contents were around 10 mg kg<sup>-1</sup>.

In Finland, a Cd content in mineral fertilises has been regulated since 1986 and the limit value has varied between 30 and 100 mg kg<sup>-1</sup> P. Currently, the Cd content of the fertiliser products is regulated in Finland by the Fertiliser Act 2006/539 and the Decree of the Ministry of Agriculture and Forestry 2007/12 on fertiliser products. In agriculture, the Cd limit for all the fertiliser products is 1.5 mg kg<sup>-1</sup> dm and 30 mg kg<sup>-1</sup> P for mineral fertilisers containing P at least 2.2%. In addition, the highest Cd load from fertiliser products must not exceed 1.5 g ha<sup>-1</sup> for one year or 6 g ha<sup>-1</sup> for four years.

In Finland, mineral fertilisers have been supplemented with Se to increase Se intake by humans and animals. According to the Decree of the Ministry of Agriculture and Forestry in Finland 2007/12, currently, the maximum allowable Se content in the fertilisers is 20 mg kg<sup>-1</sup> but 30 mg kg<sup>-1</sup> for the fertilisers to be used on animal farms or farms receiving manure from another farm.

The Decree of the Ministry of Agriculture and Forestry 2007/12 sets limit values not only for Cd, but also for As, Hg, Cr, Cu, Pb, Ni, and Zn in all the fertiliser products entering the Finnish markets and to be used in agriculture, forestry or green building (Table 14). In general, the trace element contents measured here were clearly lower than the respective limit values for the fertiliser products to be accepted for the Finnish markets. In Finland, exceptionally low trace element contents occur in the fertilisers manufactured by the Kemira GrowHow (Table 14). In addition, the Kemira GrowHow gives a certificate for the quality of the mineral fertilisers produced by the company (Table 14). Medians and ranges of the trace element contents in the mineral fertilisers used in other European countries are presented in Table 15. A variation of trace element contents in mineral fertilisers is large in Europe. European fertilisers may result in much higher heavy metal loads to the arable land than the Finnish fertilisers do.

Table 13. Trace element contents (mg kg<sup>-1</sup>) of fertiliser products used by the ten test farms and nutrient (N/P/K/Zn) contents (%) in fertiliser products according to Kemira GrowHow (no data for Cu content, Se content 0.001 % in each fertiliser product).

Trade mark (N/P/K/Zn) <sup>1)</sup>	Cu	Se	Zn	Cr	V	As	Hg	Cd	Pb	Ni
Kevätviljan Y1 (26/2/3)	6	10	10	4.4	4.6	<0.5	<0.01	<0.2	<1	2
Kevätviljan Y2 (23/3/5)	8	10	1100	6.8	6.5	0.5	<0.01	<0.2	1	5
Kevätviljan Y3 (20/3/8)	12	10	150	2.2	8.7	<1	<0.01	<0.2	<1	2
Kevätviljan Y4 (20/2/12)	<5	8	82	3.2	2.3	<0.5	<0.01	<0.2	<1	2
Kevätviljan Y4 (20/2/12)	11	10	21	4.1	4.6	<0.5	<0.01	<0.1	<1	2
Kevätviljan Y5 (22/5/5)	<5	9	41	3.6	3.1	0.6	<0.01	0.1	<1	2
Kevätviljan Y6 (17/4/13)	3	11	56	3.5	2.8	<1	<0.05	<0.2	<1	2
Syysviljan Y1 (13/7/13)	<5	11	33	3.2	2.4	0.7	<0.01	<0.2	<1	2
Syysviljan Y1 (13/7/13)	<5	10	39	3.3	2.6	0.7	<0.01	<0.2	<1	2
Nurmen Y1 (18/3/5/0.1)	<5	11	630	3.7	3.5	<0.5	<0.01	<0.2	<1	1
Nurmen Y2 (18/6/8/0.1)	<5	10	630	3.4	2.7	<0.3	<0.01	<0.2	<1	2
Nurmen NK1 20/0/7/0.15)	<5	9	720	5.0	5.5	<0.5	<0.01	<0.2	<1	2
Nurmen NK2(20/0/15/0.1)	<5	10	750	3.6	4.2	<0.5	<0.01	<0.2	<1	1
Suomensalpietari (26/0/1)	6	10	6	3.6	4.6	<1	<0.05	<0.2	<1	1
Suomensalpietari (20/0/1)	<5	12	15	1.1	4.4	<0.5	<0.01	<0.2	<1	<1
Blast-furnace slag, Koverhar	<5		9	<10	78	<1	<0.01	<0.2	<2	<2
Steel slag, Koverhar	18	<1	75	600	10000	2	<0.05	<0.2	<2	10
Detection limit	5			10	10	0.5	0.01	0.2	1	

<sup>1)</sup> Kemira GrowHow Ltd 2004.

Table 14. Current limit values for trace elements in fertiliser products on the market in Finland (MMMa 12/2007) and certified limit values for trace elements in the mineral fertilisers manufactured by the Kemira GrowHow (2007a).

	MMMa 12/2007	Kemira GrowHow
Element	mg kg <sup>-1</sup> dm	mg kg <sup>-1</sup>
Cadmium	1.5	<0.5
Mercury	1.0	<0.2
Lead	100	<4
Arsenic	25	<5
Nickel	100	<20
Chromium	300	<20
Vanadium		<20
Zinc	1 500*	
Copper	600*	

\* Value can be exceeded, if the soil is deficient

Table 15. Mean heavy metal contents (mg kg<sup>-1</sup> dm) in mineral fertilisers in Europe (KTBL 2005).

Fertilisers	Cd	Cr	Cu	Ni	Pb	Zn
<b>N</b>						
Range	0.01-6.9	0.2-23	0.3-14	0.2-49	0.1-14	0.6-39
Median <sup>1</sup>	0.9	3.4	2.0	6.0	1.9	5.0
<b>P</b>						
Range	0.11-47	2.5-990	2.9-109	1.0-60	0.9-79	24-557
Median <sup>1</sup>	13	60	26	22	13	236
<b>K</b>						
Range	0.1-2.6	1.8-14	1.0-14	0.1-223	0.4-11	0.7-11
Median <sup>1</sup>	0.5	3.4	2.0	1.4	4.2	6.6
<b>NPK</b>						
Range	0.1-12	7.0-68	1.5-70	3.8-23	0.5-3.2	15-240
Median <sup>1</sup>	2.5	32	13	10	0.5	166
<b>Lime</b>						
Range	0.03-1.0	1.3-1893	0.9-79	1.7-25	0.6-37	2.5-166
Median <sup>1</sup>	0.2	6.5	5.6	6.3	8.2	22

<sup>1</sup> = Median of reported values

In 1997, the quantity of imported inorganic fertilisers was over 105 166 tonnes, of which 71 180 tonnes was N-fertilisers (29 455 tonnes as N), 7 055 tonnes P-fertilisers (1 305 tonnes as P<sub>2</sub>O<sub>5</sub>), 444 tonnes K-fertilisers (70 tonnes as K<sub>2</sub>O) and 26 487 tonnes other mineral fertilisers (National Board of Customs 1998 and 1999). In the same year, Finnish fertiliser exports totalled 41 951 tonnes, of

which 40 080 tonnes was N-fertilisers (9 351 tonnes as N), 14 tonnes P-fertilisers (0.5 tonnes as  $P_2O_5$ ), 231 tonnes K-fertilisers (41.7 tonnes as  $K_2O$ ) and 1 626 tonnes other mineral fertilisers. The balance between import and export was as follows: all fertilisers together +63 215 tonnes, N-fertilisers +31 100 tonnes (+20 104 tonnes as N), P-fertilisers +7 041 tonnes (+1 304.5 tonnes as  $P_2O_5$ ) and K-fertilisers +213 tonnes (+28.3 tonnes as  $K_2O$ ). The quality of the fertiliser products and fertiliser raw materials imported and for sale in Finland is controlled by the Finnish Food Safety Authority, Evira.

According to statistics from the National Board of Customs, Finland's (2005) total import and total export of fertilisers manufactured (Table 15) were clearly higher in 2005 than in 1997. The total balance between the imported and exported fertilisers was +63 215 tonnes in 1997 and -332 154 tonnes in 2005. The balances indicate that in 1997, more fertilisers came into Finland than was exported, but in 2005, much larger amounts of fertilisers were sold to other countries than were bought into Finland. The total fertiliser import was about four times larger in 2005 than in 1997, but the total export was about 20 times greater than in 1997. The biggest change in imports occurred for K-fertilisers, which were imported into Finland about 700 times more in 2005 than in 1997. For exports, there seems to be an increasing trend, especially for K fertilisers (Table 15). Fertilisers that may contain relatively high amounts of harmful elements are mainly P-containing mineral fertilisers. In both 1997 and 2005, P-containing mineral fertilizers were exported very little. In 2005, 2 739 tonnes of P-fertilisers were imported, which is about one third of that in 1997 and less than 0.4% of all the fertilisers sold for agriculture in Finland in 2004. Thus, it seems to be very probable that P-fertilisers imported into Finland do not pose a great risk to Finnish agro-ecosystems.

Table 16. Finnish imports and exports (in metric tonnes) by fertiliser groups in 2005 and the change from 2004 (National Board of Customs, Finland 2005).

Fertiliser group	January-December 2005			
	Import		Export	
	Quantity	Change	Quantity	Change
Fertilisers, manufactured	450 641	+3.0	782 795	+15.0
- Mineral or chemical fertilisers, nitrogenous	94 734	+13.0	69 641	-47.0
- Mineral or chemical fertilisers, phosphatic	2 739	+7.0	18.0	+50.0
- Mineral or chemical fertilisers, potassic	304 375	-1.0	72 477	+99.0
Total balance = Import - Export			-332 154	

### 3.2.1.2 Liming materials

Since 1994, the use of lime has radically decreased in Finnish agriculture (Table 12). In 2004, lime or calcite was used in five test farms, mainly on the dairy farms. No lime samples could not be taken from the farms because lime is not stored on the farms, but was immediately applied on the soil. Trace element contents of various types of the lime used by the test farms (Table 17) were obtained from Nordkalk, Kanerva Timo (personal communication 28.4.2005) and used for the balance calculations. In Europe (KTBL 2005), trace element contents in the lime may vary (Table 15). Limes are classified to the fertiliser products so the limit values for the trace elements (MMMa 12/2007) presented in Table 14 are valid to limes as well.

Table 17. Trace element contents of lime, calcites and Dolomite lime used on the test farms (Nordkalk, Timo Kanerva, personal communication 28.4.2005).

	mg kg <sup>-1</sup> fm	
	Lime and calcites	Dolomite lime
Cd	0.06	0.15
Pb	1	3
Cu	4	3
Zn	23	23
Ni	2.5	14
Cr	2.5	4
Hg	0.01	0.01
V	17	8
As	3	2.5
Se	0.1	0.17

### 3.2.1.3 Slags

EUROSLAG (2007) describes slags as follows:

“Metallurgical slags are co-products of the iron and steel industry. They are non-metallic materials that are produced together with the metallic products of these processes. Slags are rock-like or glassy materials composed chiefly of lime and silica. Minor components are magnesia and alumina. Metallurgical slags can be subdivided into blast furnace- and steel slags.

Blast furnace slags: When iron ore is reduced at 1500°C in a blast furnace, it yields molten iron while the other components (from the ore, fluxes and coke ash) form a slag. These two liquids flow to the bottom of the furnace, where the less dense slag forms a layer above the molten iron. When the slag is tapped from the furnace it, can be treated in a number of ways but most commonly, it

is air-cooled, granulated or pelletised. Air-cooling produces a crystalline structured mass that after crushing and screening provides an eminently suitable material for use as a construction aggregate in a bound or unbound form, like any natural rock. Granulated and pelletised slags are rapidly cooled by the addition of water. Granulation gives a glass with all the components in non-crystalline solution and is inherently hydraulic, while pelletising provides an expanded material. Both can be used as construction materials and, if ground to a fine powder, become an excellent constituent of cement material for concretes, mortars and grouts.

Steel slags: Steel slag is produced from the further refining iron in a Basic Oxygen Furnace or from melting recycled scrap in an Electric Arc Furnace. Both processes produce a slag that is hard and durable, suitable for use in bound or unbound form and is recommended particularly for use in asphalt. Steel slag can also be used for clinker manufacture and as a fertiliser and soil improvement agent.”

In Europe, 0.2% of 27.2 million tonnes of blast furnace slag and 3% of 15.0 million tonnes of steel slag was used as fertiliser in 2004 (European Slag Association 2006). In Finland, the steel industry annually generates about 2 million tonnes of slags that are used for soil and road construction, soil improvement and raw material in industry. The use of slag in agriculture started in the 1970's and has increased noticeably. At the moment, totally 150 000 tonnes of slag of which 70% is steel slag and 30% blast furnace slag, is used annually in agriculture (Hiltunen et al. 2007). Only the steel slag analysed in this study was very rich in V and contained relatively high amounts of Cr (Table 13).

Currently, it is assumed that slags could also protect plants against some diseases (Hiltunen et al. 2007). In a field experiment on potato (Hiltunen et al. 2007), one application of 10 tonnes of steel slag (Table 18) increased the V content in test soil from 16 to 32 mg kg<sup>-1</sup> which meant that the V content doubled. The vanadium contents of the potatoes were not analysed in this field experiment. In general, the V content in plants is low. However, potato tubers contained more V than wheat grains or timothy grass (Mäkelä-Kurto et al. 2006). However, peeling reduced V content in potatoes by about 75%. Thus, vanadium is mainly in the peels of the potatoes.

Table 18. Trace element contents (mg kg<sup>-1</sup>) of slag (Ruukki 2007) and current limit values for trace elements in fertiliser products to be used in agriculture (MMM<sub>a</sub> 12/2007).

	Blast-furnace slag		Steel slag		Limit
	Raahe	Koverhar	Raahe	Koverhar	12/2007
Cadmium	<0.1	<0.1	<0.1	<0.1	1.5
Mercury	<0.05	<0.05	<0.05	<0.05	1.0
Lead	<1	<1	<1	<1	100
Nickel	<10	<10	<10	<20	100
Arsenic <sup>1)</sup>	nd	nd	nd		25
Chromium <sup>1)</sup>	0.012	0.005	1.48		300
Copper <sup>1)</sup>	nd	nd	0.010		600*
Nickel <sup>1)</sup>	0.002	0.001	0.009		100
Vanadium <sup>1)</sup>	250	88	9700		no limit
Zinc <sup>1)</sup>	0.019	0.030	0.010		1500*

<sup>1)</sup> Hiltunen et al. 2006.

\* Value can be exceeded, if the soil is deficient

An especially high V content, 10000 mg kg<sup>-1</sup>, and also a relatively high Cr content, 600 mg kg<sup>-1</sup>, occurred in the steel slag of Koverhar (Table 13). The limit values for the trace elements in the fertiliser products to be used in agriculture are presented in the Decree of the Ministry of Agriculture and Forestry 2007/12 (Table 14). However, there is no limit value for V. Mobility and bioavailability of V added in slag to the soil are the main factors determining the possible health and ecological risks of V. According to Hatakka et al. (2007), 7% from the *aqua regia* extractable V was soluble in AAAC-EDTA from clay soils on crop farms in southwestern Finland and 17% from finesand soils on dairy farms in Ostrobothnia in 2004, on average, but only 1.5% from silt soils in the Pirkanmaa region, on average (Mäkelä-Kurtto et al. 2006). Thus, depending on the soil type of the arable land, even 20% of the total vanadium may be in a soluble form in cultivated soil.

Steel slag applied to the soil increased the soluble concentration of vanadium as indicated by a monitoring study conducted by Sippola et al. (2001) at the MTT research station, Ruukki. The Finnish soil monitoring study showed that from 1992 to 1997, the soil AAAC-EDTA extractable V increased from 7 mg l<sup>-1</sup> to 18 mg l<sup>-1</sup> and from 3 mg l<sup>-1</sup> to 28 mg l<sup>-1</sup> in the topsoil of the fields after the application of steel slag. The steel slag used contained 1.15% vanadium. In addition, the vanadium concentrations in the topsoil that received slag were many times greater than the concentrations in the respective subsoil (about 1 mg l<sup>-1</sup>). This indicates that the concentration of soluble V may increase notably in the soil after the application of steel slag.

In Sweden, the converter slag (LD-slag) has been banned since 1992 as a fertiliser for Swedish agriculture and forestry (Lindvall 2006). Vanadium in converter slag is a growing problem in Sweden, mainly because there is no general use for it since it was banned as a lime source in agriculture Johnsson 2006. In northern Sweden, 23 heifers out of 98 cattle died of acute vanadium toxicity in a 10-day period (Frank et al. 1996). Eight months earlier, a pasture had been fertilised with basic slag, containing 3% vanadium. The fertiliser was laid on the surface without being ploughed under. Mainly heifers, and some cows, were fed with basic slag contaminated fresh hay. Vanadium-containing basic slag used as fertiliser poisoned the herd of cattle and its regional use was detectable in the livers of cattle at regular slaughter.

Leaching tests on the reuse of steel slags in road construction showed that only low amounts of Cr but high amounts of V may be released (Mroueh et al. 2000, Chaurand et al. 2006). In addition, Chaurand et al. (2006) showed that Cr is present in the less mobile and less toxic trivalent form and that its speciation does not evolve during leaching. However, V that is predominantly present in the 4+ oxidation state seems to become oxidized to the pentavalent form (the most toxic form) during leaching. In Sweden, certain types of slag may contain high amounts ( $> 10000 \text{ mg kg}^{-1}$ ,  $>1\%$ ) of Cr as well (Walterson 2007). In contrast, vanadium may be relatively soluble and bioavailable only under strongly alkaline conditions, when vanadate predominates. Leaching tests with slag from the steel industry sometimes show relatively large vanadium concentrations. The probable reason is the relatively large pH value of these materials. The chemical behaviour of V in the environment depends on many factors and is not yet sufficiently understood (Gustafsson & Johnsson 2004).

According to Jones (2004), most slag contains a significant amount of heavy metals, the release of which can cause environmental problems. However, recovery of these metals may make it economic to clean the slag to the point where they are not only safe for disposal, but where they can be used as products in their own right. This is sometimes a viable alternative to the conventional method of slag disposal by dumping, which occupies land unproductively, can cause water pollution, and can be seen as a waste of resources. Vanadium-bearing slag is used as an intermediate feed material in the V industry. Over two-thirds of worldwide V production is derived from by-product or co-product sources, and mining capacity has only a small role (Jones 2004). The production of V is mainly based on secondary materials, such as steelmaking slag. The world V production in 2004 was about 41 000 tonnes and the annual increase is expected to be 5% in the coming years. In Sweden, the content of V in the slag (in the SSAB LD-slugs) is about 4000 tonnes per year (Mefos 2006).

### 3.2.1.4 Ashes

The farms studied here did not use ashes for soil improvement. Therefore, no ash samples were analysed for trace elements. The limit values for trace elements are different for ashes to be used in agriculture and forestry (MMMa 12/2007). If the ash will be used in agriculture the lower values are valid and if the ash will be used in forestry then the higher values are valid (Table 19). In addition, a Cd loading rate is 6 g in agriculture for four years, 15 g in green building for 10 years and 60 g in forestry for 40 years (MMMa 12/2007). The Finnish control authority, Evira (former KTTK) has monitored the quality of ashes used for agriculture, green building and forestry and the results are presented in reports that are available from the Evira homepages ([www.evira.fi](http://www.evira.fi)). Finnish research results (Saarela 2007 data from Aalto 1991) from a pot study carried out on silty clay (clay content >50%) indicated that soluble, AAAC-EDTA extractable Cd clearly increased in the soil along with increasing Cd amounts added via ashes into the soil (Fig. 1). Chemical characteristics of the test soil were pH(H<sub>2</sub>O) 5.7, electrical conductivity 1.15 x 10<sup>-4</sup> S cm<sup>-1</sup>, Ca 1639, K 338, Mg 334, P 7.3, S 23, B (water soluble) 0.47, Cu 5.2, Zn 0.88, Mn 30.5 Fe 688, Cd 0.09 and Mo 0.019 mg l<sup>-1</sup> of air-dry soil (Aalto 1991).

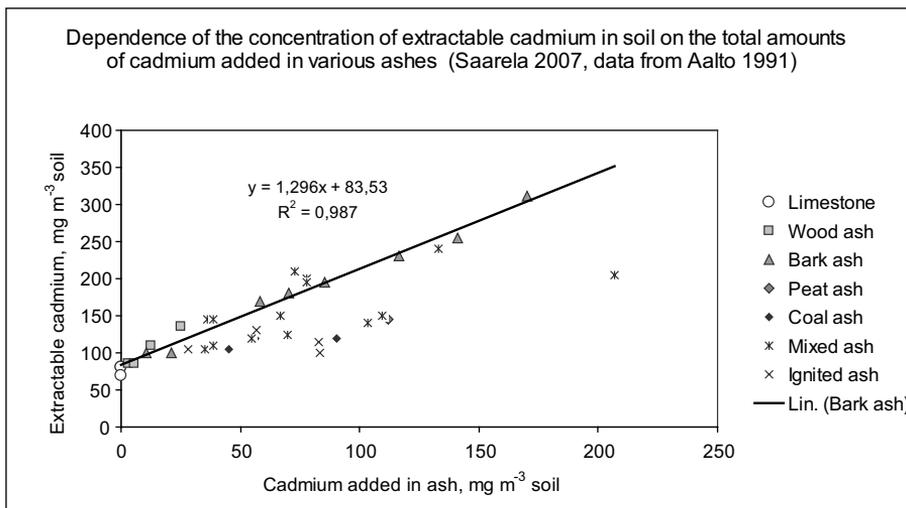


Figure 1. Dependence of the concentration of AAAC-EDTA extractable cadmium in soil on the total amounts of cadmium added in various ashes (Saarela 2007, data from Aalto 1991).

### 3.2.1.5 Municipal sewage sludge

In Finland, municipal wastewater treatment plants have generated sewage sludge about 1 000 000 tonnes of sewage sludge annually as fresh matter and

160 000 tonnes as dry matter (Ministry of Environment in Finland 1995, 2003). The amounts will probably remain the same in the upcoming years. Since 2000, about 80% of municipal sewage sludge has been used as a growth medium in green buildings and as cover medium in landfills, 12% in agriculture after various treatments, 6% in landfills and 2% has been stored (Ministry of Environment in Finland 2003). By 2005, the target for sludge use was 90% and that has been reached. However, the use of sewage sludge as a cover medium in landfills will be reduced. Thus, additional routes for recycling of sewage sludge should be found out.

Use of municipal sewage sludge on the cultivated soils may have beneficial fertilising, liming and humus increasing effects on the soil. However, sewage sludge may cause potential environmental and agricultural problems related to an excessive and/or unbalanced supply of nutrients, the introduction of pollutants, such as heavy metals and organic compounds, and the spreading of human, animal or plant pathogens (EC 2003). Sewage sludge production in the EU is more than 8 million tonnes of dry matter. According to Sauerbeck (1986), sewage sludge has physical influences on soil texture, structure, pore volume, pore size distribution and soil colour, chemical influences on pH value, redox potential, carbon content, exchange capacity (cations and anions), colloidal and ionic precipitation and bonding reactions (like complexation), nutrient and pollutant content and biological influences on soil biological activity, microflora (bacteria, fungi and algae) and fauna (microfauna, mesofauna and macrofauna)

In the EU member states, agricultural use of sewage sludge has been regulated by the Sewage Sludge Directive of EEC (278/1986), which is now under revision. Ranges of heavy metal contents in municipal sewage sludges in EU member states are presented in Table 19. Due to the small population, low population density and low application rates of sludge in Finland, the importance of municipal sewage sludge as a trace element source is small, but can be locally great, particularly if sludge is continuously applied to the same fields. The fields situated in the vicinity of the waste water treatment plant are at the highest risk to frequently receive sewage sludge. The Government Decree on the use of sewage sludge in agriculture in Finland 1994/282 gives the limit values for the heavy metal contents in the sludge to be used in agriculture (Table 19) and also the limit values for the mean annual loading rates of heavy metals per hectare and for the heavy metal contents of the soil to which the sewage is to be applied. The Decree of the Ministry of Agriculture and Forestry in Finland on fertiliser products 2007/12 sets limit values for heavy metals when the sludge is recycled in agriculture as fertiliser product (Table 19).

A high P content of the municipal sewage sludge efficiently reduces the rate of application and heavy metal loads into the soil as well. In the future, use of sludge in agriculture may be enhanced especially in non-food production. A tar-

get of the EU is to double the use of renewable energy sources from the current 6% to 12% by 2010 (ProAgria Group 2006). One renewable energy source is the production of bioenergy plants on arable land. This would be a sustainable way to increase recycling of nutrients and organic matter in side-products generated by municipalities and food or feed industry for bioenergy production, if the side-products have been proven to be safe to the agro-ecosystems.

Table 19. Ranges of heavy metal contents (mg kg<sup>-1</sup> dm) in municipal sewage sludge in EU member states (EC 2001), maximum heavy metal contents in sewage sludge according to EEC (Directive 86/278/EEC), to the Council of the State Decision on the use of the sewage sludge in agriculture in Finland 1994/282 (VNp 1994/282) and to the Decree of the Ministry of Agriculture and Forestry in Finland 2007/12 on fertiliser products (including inorganic and organic fertilisers, limes, ashes, slags, composts, etc.) to be used in agriculture (Agric) or forestry (Forest) (MMMa 2007/12).

	Ranges in the Member States	Directive 86/278/EEC	VNp 1994/282 Agriculture	MMMa 2007/12 Agric/Forest
Cadmium	0.4 - 3.8	20 – 40	1.5	1.5/15**
Chromium	16 - 275	1 000 – 1 750	300	300/300
Copper	39 - 641	1 000 – 1 750	600*	600*/700
Mercury	0.3 - 3	16 – 25	1.0	1.0/1.0
Nickel	9-90	300 – 400	100	100/150
Lead	13 - 221	750 – 1 200	100	100/150
Zinc	142 - 2000	2 500 – 4 000	1500*	1 500*/4500
Arsenic	-	-	-	25/30

\* Value can be exceeded, if the soil is deficient.

\*\* 17.5 mg dm in wood, peat or field biomass ashes to be used in forestry.

### 3.2.2 Commercial feeds

Maximum concentrations of As, Cd, Hg and Pb and other undesirable substances in animal feed are described in the Directive of the European Parliament and of the Council 2002/32/EEC. Limit values for micronutrients (Co, Cu, Fe, Mn, Se and Zn) is given in feed additives legislation. In Finland, the competent authority for control of quality of feeds is Evira ([www.evira.fi](http://www.evira.fi)). The Evira publishes the results of the control of undesirable substances in feed twice a year. If exceeding contents come out, use and marketing of the feed in question has to be stopped immediately.

Feeds are one source for the intake of trace elements by domestic animals and one source for soils via farm animal manure. The higher the trace element content in the feeds, the greater the arsenic input into the soil through manure, in general. Heavy metals may also accumulate into food. For instance, liver is known to be

a source of heavy metals (especially Cd) for human. Contents of heavy metals in agricultural products are described in Chapters 3.3.1, 3.3.2 and 3.3.3.

Total annual amounts of feeds manufactured for the farms in Finland are presented in Table 20. In 2004, the amount was 1 252 million kg, which was more than the total annual amount of mineral fertilisers and lime together (1 199 million kg) sold to the Finnish markets (Table 14). About half of the feeds were used for cattle and about a fourth for pigs. Use of commercial feeds has increased while the use of mineral fertilisers and lime has decreased over the last few years.

Table 20. Amounts (million kg) of feeds manufactured annually for cattle, pig, poultry and broiler farms in Finland (Salopelto 2006).

Feeds for	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004
Cattle	444	493	458	458	524	616	640	642	655	645	661
Pig	252	279	270	270	263	254	246	246	268	288	285
Poultry	85	92	84	76	69	67	70	67	67	72	71
Broiler	110	121	138	145	169	185	178	211	228	235	235
Totally	892	985	951	949	1024	1121	1133	1165	1218	1240	1252

Concentrations of As, Cr, Cu, Ni, Zn, V, Hg, Cd, Pb and Se in bovine feeds are reported in Table 21 and 22. Concentrations of undesirable substances (Hg, Pb, Cd and As) in all studied samples were below limit values set in the legislation (Directive 2002/32/EC of the European Parliament and of the Council and the Decree of the Ministry of Agriculture and Forestry in Finland 2007/1. The limit values presented in the Decree for each element depends on feed type (mineral feed, compound feed or complementary feed etc.), feed origin (plant, animal, sea, mineral, etc.), target animal, and age and purpose of the animal (e.g. dairy cow, meat cow). The limit values for As varied between 2 and 40 mg kg<sup>-1</sup> (88% dm), Pb between 5 and 400 mg kg<sup>-1</sup> (88% dm), Hg between 0.1 and 0.5 mg kg<sup>-1</sup> (88% dm) and Cd between 0.5 and 30 mg kg<sup>-1</sup> (88% dm).

The concentrations of Cd in all complete or complementary feeds except minerals reported in this study were 0.2 mg kg<sup>-1</sup> (88% dm) or less. This is similar or less than in feed materials of plant origin reported by EFSA (2004a) (0.05 to 0.6 mg kg<sup>-1</sup> (88% dm) or in commercial compound feeds (complete feeding stuff and complementary feed) for ruminants in the EU (0.03 to 0.8 mg kg<sup>-1</sup> (88% dm). The concentrations of cadmium in minerals and premixtures in EU member states were similar or higher (<0.01 to 2.0 mg kg<sup>-1</sup> (88% dm) (EFSA, 2004a) than concentrations reported in this study, <0.1 to 0.3 mg kg<sup>-1</sup>

(88% dm). The significance of the concentration of heavy metals in minerals to exposure to livestock is difficult to establish since these products are included in diets at varying inclusion rates.

Concentrations of Pb in compound feed (complete feeding stuff and complementary feeds) for cattle in EU member states were less than  $4.7 \text{ mg kg}^{-1}$  (88% dm)<sup>-1</sup> with averages of 0.9 and  $1.0 \text{ mg kg}^{-1}$  (88% dm) for dairy and beef cattle, respectively (EFSA 2004b), which is somewhat higher than concentrations in our study,  $< 1 \text{ mg kg}^{-1}$  (88% dm). Mean concentrations of lead in grass/herbage, hay and grass silage in the EU were 4.0, 3.4 and  $1.8 \text{ mg kg}^{-1}$  (88% dm), respectively (EFSA 2004b).

Concentrations of lead in mineral feeds in this study were lower, 1 to  $2.1 \text{ mg kg}^{-1}$  (88% dm), than the mean concentration in mineral supplements reported by EFSA (2004b) in the EU member states,  $3.0 \text{ mg kg}^{-1}$  (88% dm).

Concentration of arsenic in compound feed in EU member states ranged from 0.1 to  $0.5 \text{ mg kg}^{-1}$  (88% dm) with averages of 0.4 and  $0.2 \text{ mg kg}^{-1}$  (88% dm) for beef and dairy cattle, respectively (EFSA 2005). In our study, concentrations of arsenic in complete feeds were below detection limit,  $0.5 \text{ mg kg}^{-1}$  (88% dm). The concentration of arsenic in mineral feeds in our study ranged from 0.8 to  $3.1 \text{ mg kg}^{-1}$  (88% dm) with an average of  $2.0 \text{ mg kg}^{-1}$  (88% dm). This is lower than reported in the EU, 0.01 to  $14 \text{ mg kg}^{-1}$  (88% dm) with an average of  $6.0 \text{ mg kg}^{-1}$  (88% dm) (EFSA, 2005). Plants take arsenic from soil and concentration of arsenic in soil influences the concentration of arsenic in plants.

Concentrations of As, Cd and Pb were equal or below the detection limits:  $0.5 \text{ mg kg}^{-1}$  (88% dm),  $0.1 \text{ mg kg}^{-1}$  (88% dm) and  $1 \text{ mg kg}^{-1}$  (88% dm) in all feeds except in mineral feeds, respectively. Mineral feeds are added into feed in minor amounts, and thus, the concentrations of heavy metals will be diluted in complete feed.

Most pastures and crops contain Hg less than  $0.1 \text{ mg kg}^{-1}$  dm (European Commission, 2003a). Typically, feed material of plant origin contains mercury between 0.001 and  $0.03 \text{ mg kg}^{-1}$  dm (Dudka & Miller 1999). In our study, concentration of Hg in all samples was below the detection limit,  $0.01 \text{ mg kg}^{-1}$  (88% dm).

Copper, cobalt (Co), iron (Fe), manganese (Mn), molybdenum (Mo), Se and Zn are essential elements in animal nutrition. They are used as trace element and feed additive in animal nutrition (European Commission, 2003a, b). Concentrations of these elements are regulated in feed additive legislation. Recommendations for concentrations of copper, zinc and selenium in complete feed for ruminants are 10, 50 and  $0.1 \text{ mg kg}^{-1}$  dm feed, respectively (MTT 2004). Farm animal copper and zinc intake are almost entirely excreted and the slurry is spread onto arable lands. Thus, the soil will be exposed to copper and zinc.

Transfer of chromium into plants is low (European Commission, 2003a) and the chromium content of minerals is variable. Adsorption of Cr(III) is poor, and consequently, the tissue residues are very low (European Commission 2003).

There are no guidelines or statutory limits for concentration of Ni, Cr or V in animal feed. Some animals may be sensitive to excess nickel. However, evidence exists that nickel can also be a useful trace element for domestic animals (Spears 1984).

Table 21. Concentrations, mg kg<sup>-1</sup> (88% dm), of trace elements in 13 different individual feeds sampled from the five dairy farms (6-10) in Ostrobothnia in 2004.

Trade mark	Feed type*	Cr	Cu	Ni	Se	Zn	V	As	Hg	Cd	Pb
Huippu Krossi 23	1	0.6	17	<1	0.79	190	0.5	<0.5	<0.01	<0.1	<1
Aseto-Melli	1	0.8	7.5	<1	0.15	54	0.5	<0.5	<0.01	<0.1	<1
Amino-Maituri	2	1	21	<1	0.68	120	0.9	<0.5	<0.01	0.1	<1
Mulli-Melli	3	5.5	410	26	17	1600	1	0.75	<0.01	<0.1	1,1
Kesä-Namino	3	10	400	19	18.6	2500	18	3.1	<0.01	<0.1	1
Tähti-145	1	0.6	8.7	<1	0.13	60	0.5	<0.5	<0.01	<0.1	<1
Molassed rade seed meal	1	1	6.6	<1	0.34	56	0.3	<0.5	<0.01	0.2	<1
Onni-Kivennäinen	3	14	280	79	20	1260	2.9	1.4	<0.01	<0.1	1.2
Maituri 10000	2	1	21	<1	0.45	96	0.6	<0.5	<0.01	<0.1	<1
Rouhe-Tiiviste	1	0.6	10	<1	0.17	67	0.2	<0.5	<0.01	<0.1	<1
Sugar beet pulp, molassed	4	1.3	<5	1	0.08	11	1	<0.5	<0.01	0.1	<1
Home mixture for cattle	5	1.6	23	<1	0.6	120	0.8	<0.5	<0.01	<0.1	<1
Viher Hertta-Minera Muro	3	24	530	36	10	3600	29	2.7	<0.01	0.3	2.1

\* Feed types: 1 = Complementary feed for cattle; 2 = Complete feed for cattle; 3 = Mineral feed for cattle; 4 = Feed for cattle; 5 = Home mixture for cattle

Table 22. Mean trace element concentrations, mg kg<sup>-1</sup> (88% dm), in different feed types. Samples (n = number of samples) were collected from the five dairy farms (6-10) in Ostrobothnia in 2004.

Feed type*	Cr	Cu	Ni	Se	Zn	V	As	Hg	Cd	Pb	n
1	0.7	10	<1	0.3	85	0.4	<0.5	<0.01	<0.1	<1	5
2	1	21	<1	0.6	110	0.8	<0.5	<0.01	<0.1	<1	2
3	14	400	40	16	2200	13	2.0	<0.01	0.2	1.4	4
4	1.3	<5	1	0.1	11	1	<0.5	<0.01	0.1	<1	1
5	1.6	23	<1	0.6	120	0.8	0.5	<0.01	<0.1	<1	1

\* Feed types: 1 = Complementary feed for cattle; 2 = Complete feed for cattle; 3 = Mineral feed for cattle; 4 = Feed for cattle; 5 = Home mixture for cattle

### 3.3 Trace elements in agricultural products

#### 3.3.1 Crop plants

At the study farms, various plant species were cultivated: oats (*Avena sativa* L.), rye (*Secale cereale* L.), wheat (*Triticum aestivum* L.), barley (*Hordeum vulgare* L.), Rapeseed (*Brassica rapa ssp oleifera*), pea (*Pisum sativum*) and timothy (*Phleum pratense*).

The Commission regulation (EC) No 1881/2006 establishes maximum levels of lead and cadmium in certain foodstuffs and mercury in fish products. The regulation has been in force since 1 March 2007. The maximum levels of lead and cadmium in cereal based foods are presented in Table 23. For other elements, no maximum limit for cereals has yet been established.

Table 23. Maximum levels of cadmium (Cd) and lead (Pb) in cereal based foods according to the Commission regulation (EC) No 1881/2006.

Product	Maximum Cd level mg kg <sup>-1</sup> fm	Maximum Pb level mg kg <sup>-1</sup> fm
Cereals, excluding bran, germ wheat grain and rice	0.1	
Bran, germ, wheat grain and rice	0.2	
Cereals, legumes and pulses		0.2

Table 24. Mean trace element contents (mg kg<sup>-1</sup> dm) of crop plants on five crop and five dairy farms in Finland in 2004.

	Oats		Barley		Spring wheat		Winter wheat	Rape-seed	Rye	Pea
	Dairy farm	Crop farm	Dairy farm	Crop farm	Dairy farm	Crop farm	Crop farm	Crop farm	Crop farm	Crop farm
n	4	2	5	10	3	2	4	4	3	1
As	0.031	0.010	0.016	<0.010	<0.010	<0.010	<0.010	0.040	<0.010	0.019
Cd	0.016	0.022	0.019	0.017	0.022	0.017	0.057	0.190	0.024	0.015
Cu	5.0	3.8	6.7	5.3	5.4	4.8	4.7	4.2	5.6	7.8
Hg	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
Ni	1.44	0.758	0.055	0.063	0.039	0.130	0.191	0.920	0.110	1.66
Pb	0.041	0.012	0.050	0.130*	0.028	0.035	0.011	0.120*	0.013	0.058
Se	0.047	0.210	0.038	0.160	0.079	0.180	0.170	0.150	0.120	0.031
V	0.170	0.007	0.062	0.013	0.006	<0.005	<0.005	0.490	0.007	0.23
Zn	35	26	31	22	33	26	26	34	27	28

\* In one crop farm, the Pb concentration was very high which increased the mean value.

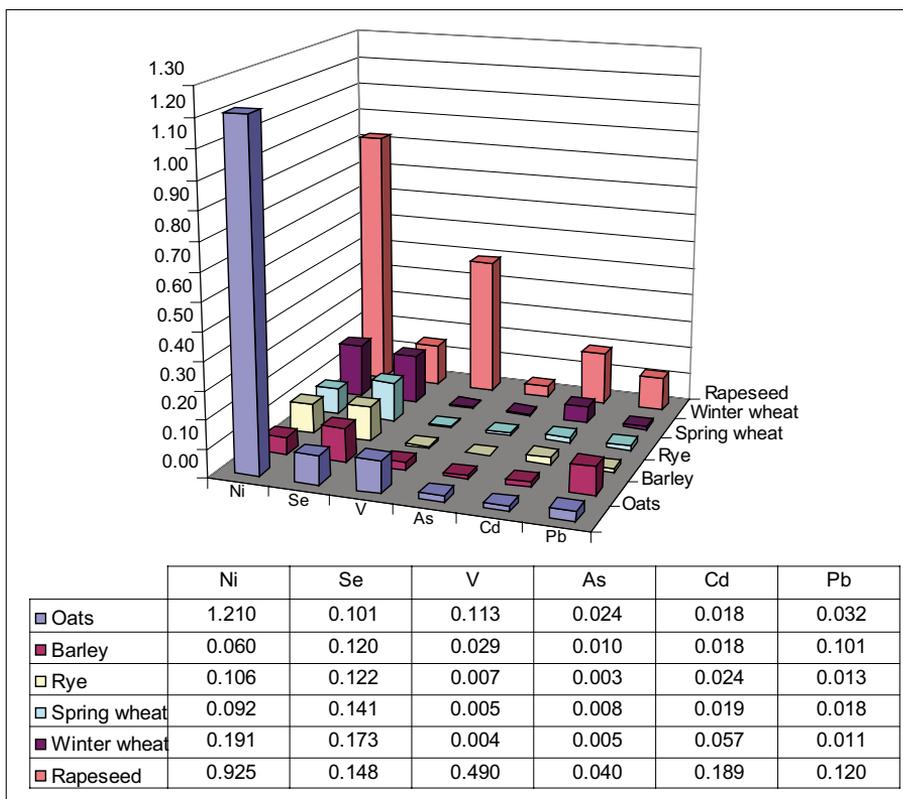


Figure 2. Mean nickel, selenium, vanadium, arsenic, cadmium and lead contents ( $\text{mg kg}^{-1} \text{ dm}$ ) in the crops of oats, barley, rye, spring wheat, winter wheat and rape seed collected from the study farms in Finland in 2004.

While a direct comparison of the elemental concentrations between the crop and dairy farms was not possible because of regional and climatic differences, there were some clear differences. Selenium concentrations in crop plants were clearly lower in dairy farms. Vanadium, zinc and copper were slightly lower in the crop plants on the cereal farms.

Elemental concentrations varied between the plant species. Nickel and vanadium contents were highest in oats, rapeseed and pea samples, whereas cadmium contents were highest in rapeseed and winter wheat, lead and arsenic in rapeseed. Lead concentrations were systematically higher in one cereal farm, which increased the mean value of lead in barley over the maximum permitted level.

Elemental concentrations of 16 grass feed samples represented typical national levels (Table 25). Some variation between the study farms and harvest years were observed. Largest differences were in Se, V, Ni and Cr contents in silage

samples and Ni, Pb and Se contents in pasture grass samples. This may be due to the different fertilisation (Se) and growing conditions or contamination from soil (Pb, Cr, V, Ni).

Toxic heavy metal contents in grass feeds were low. According to the DMA-FF 1/2007, the maximum Pb content for grass feed is 30 mg kg<sup>-1</sup> 88% dm. The maximum lead concentration in grass feed was found from pasture grass, 0.55 mg kg<sup>-1</sup> dm. This was less than 2% of the maximum level.

Mäkelä-Kurtto et al. (2006) determined the trace element contents of wheat, potato and timothy in the Pirkanmaa region in 2005. This study showed that there were clear differences in the trace element contents between the plant species. In general, the highest As, Cu, Pb, Ni and Zn contents were found in the timothy grass; the highest Cr and V contents in the unpeeled potato tubers; and the highest Cd and Se contents in the wheat grains (Appendix 4, 5 and 6: Table 1). Peeling the potatoes remarkably reduced the V, Al and Fe contents and to a lesser extent Cr and Ni contents in potatoes (Appendix 5). Often, trace element contents are high in the roots and decrease from the roots to the shoots and fruits. In general, leafy plants are more susceptible to atmospheric depositions and soil dust than seeds and grains. Soil-to-plant uptake factors (element content in plant tissue, mg kg<sup>-1</sup> dm / *aqua regia* extractable element content in soil, mg kg<sup>-1</sup> dm) were the highest for Se, Zn, Cu, and Cd and the lowest for V, Cr, As, Ni and Pb (Appendix 6: Table 2). Element contents of timothy samples collected from the first cut all over Finland during the national soil monitoring programme in 1998 are reported in Table 26 (Mäkelä-Kurtto & Sippola 2002, unpublished research results of the MTT).

Table 25. Trace element contents of grass feed samples collected from the dairy farms in Ostrobothnia in 2004.

Element	Grass silage 2003 Mean mg kg <sup>-1</sup> dm	Grass silage 2004 Mean mg kg <sup>-1</sup> dm	2003-2004 Mean ± SD mg kg <sup>-1</sup> dm	Pasture grass Mean ± SD mg kg <sup>-1</sup> dm	Hay Mean mg kg <sup>-1</sup> dm
n	5	5	10	4	2
As	0.050	0.050	0.050 ± 0.030	0.037 ± 0.009	0.021
Cd	0.021	0.013	0.017 ± 0.008	0.020 ± 0.011	0.007
Cr	0.130	0.100	0.120 ± 0.090	0.110 ± 0.076	0.090
Cu	6.4	7.9	7.2 ± 1.6	7.6 ± 2.3	5.1
Hg	<0.005	<0.005	<0.005	<0.005	<0.005
Ni	0.330	0.370	0.350 ± 0.210	0.460 ± 0.270	0.280
Pb	0.100	0.130	0.120 ± 0.036	0.240 ± 0.210	0.095
Se	0.340	0.260	0.300 ± 0.210	0.210 ± 0.210	0.140
V	0.290	0.063	0.180 ± 0.280	0.064 ± 0.019	0.028
Zn	27	25	26 ± 3.6	31 ± 7.0	25

Table 26. Statistical indicators on element contents in timothy samples (n = 200) collected from the first cut evenly from the whole Finland during the national soil monitoring programme in 1998 (Mäkelä-Kurto & Sippola 2002, unpublished research results of the MTT).

Element	Unit	Min	Median	Mean	Max
Ca	g kg <sup>-1</sup> dm	0.99	2.06	2.15	4.43
K	g kg <sup>-1</sup> dm	9.23	20.6	20.4	39.1
Mg	g kg <sup>-1</sup> dm	0.54	1.04	1.06	2.48
P	g kg <sup>-1</sup> dm	1.40	2.54	2.53	3.75
S	g kg <sup>-1</sup> dm	0.72	1.28	1.35	2.45
Al	mg kg <sup>-1</sup> dm	3.97	7.62	10.5	126
B	mg kg <sup>-1</sup> dm	2.97	4.90	5.15	11.4
Cd	mg kg <sup>-1</sup> dm	0.002	0.014	0.015	0.051
Co	mg kg <sup>-1</sup> dm	0.007	0.030	0.043	0.418
Cr	mg kg <sup>-1</sup> dm	0.020	0.260	0.302	1.310
Cu	mg kg <sup>-1</sup> dm	1.78	4.09	4.13	6.60
Fe	mg kg <sup>-1</sup> dm	19.0	33.2	43.4	170
Mn	mg kg <sup>-1</sup> dm	10.3	46.7	49.7	192
Ni	mg kg <sup>-1</sup> dm	0.11	0.48	0.57	2.62
Pb	mg kg <sup>-1</sup> dm	0.009	0.056	0.067	0.492
Zn	mg kg <sup>-1</sup> dm	10.9	24.0	24.9	53.5

### 3.3.2 Milk

Factors influencing the composition of milk are feeding, seasonal changes, milking frequency, milking systems and breed of cow. In this study, the toxic heavy metal (Cd, Pb, Hg) and arsenic contents in milk were low, generally below the detection limit (Table 27). The maximum level of lead in milk is 0.020 mg kg<sup>-1</sup> fm. Other maximum levels of toxic elements for milk have not yet been established. In this study, the lead content of milk was <0.006 mg kg<sup>-1</sup> fm and thus, well below the maximum permitted level.

Table 27. Heavy metal, arsenic and selenium contents in milk samples collected on five dairy farms in Ostrobothnia in 2004.

MILK	Winter milk	Summer milk	Mean ± SD	Maximum level*	Mean**
Element	mg kg <sup>-1</sup> fm	mg kg <sup>-1</sup> dm			
n	5	5	10		10
As	<0.010	<0.010	<0.010		0.009
Cd	<0.007	<0.007	<0.007		0.0008
Cr	<0.020	<0.020	<0.020		<0.020
Cu	<0.4	<0.4	<0.4		0.46
Hg	<0.005	<0.005	<0.005		<0.005
Ni	0.0028	0.0023	0.0025 ± 0.001		0.019
Pb	<0.006	<0.006	<0.006	0.020	0.015
Se	0.025	0.024	0.025 ± 0.005		0.190
V	<0.005	<0.005	<0.005		0.012
Zn	4.0	4.0	4.0 ± 0.3		0.031

\* Commission regulation (EC) No 1881/2006

\*\* calculated with the moisture content of 87%

Table 28. Arsenic, cadmium and lead in milk and dairy products excluding cheese in some European countries (SCOOP 2004).

MILK Country	As mg kg <sup>-1</sup> fm	Cd mg kg <sup>-1</sup> fm	Pb mg kg <sup>-1</sup> fm
Belgium		0.0004	0.0040
Denmark		0.0010	0.0017
Finland	<0.005	0.001	<0.010
France		0.000327	0.002
Germany	0.003	0.006	0.017
Greece		0.001	0.0123
Ireland		0.005	0.05
Italy		<0.004	0.007
The Netherlands		0.001	
Norway			0.0024
Sweden		<0.001	<0.002
United Kingdom	0.0004	0.0002	0.001

Trace element concentrations represented typical national levels in conventionally produced milk. Varo *et al.* (1980) reported zinc concentration in untreated whole milk that varied between 4.2-4.7, copper 0.065-0.12, nickel 0.004-0.01 and chromium 10-20 mg kg<sup>-1</sup> fm. In this study, zinc and copper concentrations were now slightly lower, zinc 3.6-4.5, copper 0.05-0.08 mg kg<sup>-1</sup>. However, the samples in this study represent the situation in the five farms studied, whereas in the previous study samples were taken from large dairies. As well, analytical methods differed between the studies.

In Finland selenium concentrations in milk are affected by the selenium supplemented fertilisation that has been used for over 20 years. During this study, the selenium supplementation level was 10 mg kg<sup>-1</sup> fertiliser. Milking cows also received complete, complementary and/or mineral feeds of variable selenium concentration (Table 21). The maximum permitted amount of selenium in compound feeds or daily rations is 0.5 mg kg<sup>-1</sup> dw. In the present study selenium concentrations in milk varied between 0.017-0.036 mg kg<sup>-1</sup> fw. The mean selenium concentration of 0.025 mg kg<sup>-1</sup> fw represents the typical national selenium level (Eurola *et al.* 2003) and is among the highest in Europe. The mean selenium content of milk in many European studies varies between 0.007-0.019 mg kg<sup>-1</sup> fm (Lindmark-Månsson *et al.* 2003, Simonoff *et al.* 1988, Smrkolj *et al.* 2005, Bratakos *et al.* 1987, Kadrabova *et al.* 1997). However, in some areas like the Sava basin, Croatia, the mean selenium concentrations of 0.0409 mg kg<sup>-1</sup> fm in milk have been found (Klapec 2004). Selenium contents were slightly lower in summer milk (cows feeding in the pasture). Such a seasonal variation in selenium contents of milk has been detected previously. Selenium is mainly bound to the protein constituents of milk. According to Muñiz-Naveiro *et al.* (2005), 53.6% of selenium is distributed in whey, 42.6% in casein and 9.3% in fat.

### 3.3.3 Meat

During this study, only 4 meat samples were received. Toxic heavy metal and arsenic concentrations were low. Lead and cadmium contents were well below the maximum permitted levels (Table 29) and generally lower than most European countries (Table 30).

Table 29. Heavy metal, arsenic and selenium contents in meat samples collected on the dairy farms in Ostrobothnia in 2004.

MEAT Element	Mean $\pm$ SD mg kg <sup>-1</sup> dm	Mean mg kg <sup>-1</sup> fm	Maximum level* mg kg <sup>-1</sup> fm
n	4	4	
As	0.007 $\pm$ 0.001	0.002	
Cd	0.0005 $\pm$ 0.0004	0.0001	0.050
Cr	<0.030	<0.020	
Cu	3.3 $\pm$ 0.9	0.9	
Hg	<0.005	<0.005	
Ni	0.025 $\pm$ 0.017	0.006	
Pb	0.017 $\pm$ 0.005	0.015	0.10
Se	0.530 $\pm$ 0.070	0.140	
V	0.002 $\pm$ 0.0004	0.0006	
Zn	0.160 $\pm$ 0.030	0.040	

\* Commission regulation (EC) No 1881/2006

Table 30. Arsenic, cadmium and lead in meat and meat products including poultry and game in some European countries (SCOOP 2004).

MEAT Country	As mg kg <sup>-1</sup> fm	Cd mg kg <sup>-1</sup> fm	Pb mg kg <sup>-1</sup> fm
Belgium		0.024	0.0040
Denmark	0.024	0.0022	0.073
Finland		0.001-0.004	0.01-0.03
France		0.004	0.012
Germany	0.009-0.033	0.007-0.016	0.054-1.32
Greece		0.0074	0.1137
Ireland	0.0037-0.01	0.001-0.01	0.02
Italy			0.038
The Netherlands		0.051	
Norway	0.023	0.046	0.034
Sweden		0.007	<0.00-0.035
United Kingdom	0.0033	0.0008-0.0097	0.0073

### 3.3.4 Cattle manure

Over the past few years, the total amount of biodegradable waste in Finland has annually been about 34 million tonnes as fresh matter (fm) of which about 20 million tonnes are animal manure (Ministry of Environment 1995, 2003). This is about 20 times the amount of sewage sludge generated annually in the municipal waste water treatment plants in Finland. Numbers of animals in Finland are presented in Table 3. The numbers of cattle and sheep have decreased, while the number of horses has increased. The amount of manure generated in Finland is based on the numbers of farm animal species. Finnish agricultural production is mainly based on livestock and milk is the most important product of Finnish agriculture. Therefore, a major part of animal manure, 18 million tonnes, was generated by cattle and consisted of about 85% of all the manure generated annually. If fresh animal manure contains about 20% dry matter, a total amount of animal manure as dry weight is about 4 million tonnes.

Analytical results obtained for the manure samples collected from the five dairy farms in 2004 are presented in Table 29. Micro-nutrients like Zn and Cu, were highest contents in manure. Manure contained >1 mg of Ni, Cr and V, about 1 mg of Se, As and Pb, and clearly less than 1 mg of Cd and Hg in kg dry matter. Peat seemed to have relatively high Pb and V contents and faeces a high Pb content, but low Cu and Zn contents. Due to the low number of manure samples, it was difficult to see any systematic difference in the trace element contents between the spring and autumn. The Cu and Zn contents of manure measured in this study were lower than the long-term mean in Finland (Table 32). However, the results were still well within the standard deviation. The variability on the contents of micronutrients in manure is extensive. Due to mineral additives in feeding stuffs, pig manure and slurry is especially known for its significant elevated concentrations of Cu and Zn (Table 33, 34 and 35).

Trace element contents in manure generated by different animal species may vary much both inside and between the species shown by Table 32 and 33. In general, pig and poultry manures are particularly rich in Zn and Cu. Compared to other animal manure, cattle slurry seemed often to have lower contents of trace elements and harmful elements, than the manure of other animal species or other manure types.

Table 31. Trace element concentrations (mg kg<sup>-1</sup> dm) in cattle manure samples collected on five dairy farms in Ostrobothnia in 2004. For calculation of the means, values below the analytical detection limit (DL) were replaced by the value DL/2.

Sample Nro.	Type <sup>1)</sup>	Cd	Pb	Cu	Zn	Ni	Cr	Hg	V	As	Se
1	1	0.10	<1.5	31	150	<10	1.8	<0.07	<5.0	0.96	0.04
2	1	<0.10	<1.5	27	130	<10	1.7	<0.07	<5.0	<0.45	0.19
3	1	0.17	<1.5	41	270	<10	1.7	<0.07	<5.0	0.73	1.40
4	1	<0.10	<1.5	46	240	<10	2.0	<0.07	<5.0	0.49	1.80
5	1	0.15	<1.5	46	320	<10	1.8	<0.07	<5.0	1.7	1.30
6	1	<0.10	<1.5	52	370	<10	1.9	<0.07	5.4	1.3	1.10
7**	1	0.17	<1.5	55	290	<10	3.0	<0.07	6.2	<0.45	1.50
<b>Mean</b>		<b>0.11</b>	<b>0.75</b>	<b>43</b>	<b>253</b>	<b>5</b>	<b>2.0</b>	<b>0.035</b>	<b>3.4</b>	<b>0.80</b>	<b>1.05</b>
9	2	0.14	<1.5	53	250	<10	2.4	<0.07	5.2	0.61	1.30
12	2	0.11	8.1	18	87	<10	2.4	<0.07	<5.0	0.60	0.22
<b>Mean</b>		<b>0.13</b>	<b>4.4</b>	<b>36</b>	<b>169</b>	<b>5</b>	<b>2.4</b>	<b>0.035</b>	<b>3.9</b>	<b>0.61</b>	<b>0.76</b>
10	3	0.13	4.5	27	140	<10	4.5	<0.07	15.0	<0.45	1.90
11	4	0.30	23	3.2	24	<10	1.6	<0.07	<5.0	1.2	0.24

1 = dairy slurry; 2 = faeces and urine in peat; 3 = faeces; 4 = peat; \*\* Sample collected in spring, but analysed in autumn

Table 32. Mean copper (Cu) and zinc (Zn) contents of liquid manure in Finland between 2000-2004 (Unpublished data of Viljavuuspalvelu 2005) and in the five dairy farms in Ostrobothnia in 2004.

	Cu, mg kg <sup>-1</sup> fm	Zn, mg kg <sup>-1</sup> fm
Mean	2.7	17.1
Standard deviation	1.6	10.5
Minimum	0.2	1.3
Maximum	17	190
Number of samples	638	638
Mean of dairy farms	1.7	10.1

Table 33. Ranges of mean values (mg kg<sup>-1</sup> dm) found for different types of liquid and solid manure (Amlinger et al. 2004).

	Cd	Cr	Cu	Hg	Ni	Pb	Zn	As
Cattle slurry	0.21-0.61	4.6-17	42-71	0.04-0.33	3-21	5-29	166-237	0.35-0.62
Cattle manure	0.22-0.76	5-56	33-60	0.04-0.38	4-47	5-18	109-347	0.67-1.77
Sheep & goat manure	0.28-41	6.3-60	26-41	0.17	6.8-16	3.4-28	107-204	0.99-2.65
Pig slurry	0.31-0.82	8.4-20	294-499	0.03-0.2	11-26	3.5-29	619-1270	0.52-0.83
Pig manure	0.43-1	10.5-14	276-740	0.04	8.7-24	8.7-13	733-1450	nd
Poultry manure/slurry	0.18-2.86	4.4-13	61-137	0.02-0.15	7.1-31	3.4-29	302-636	0.49-0.89

nd. = no data

Table 34. Trace element contents (mg kg<sup>-1</sup> dm) in dairy slurry (n = 4), pig liquid and solid manure (n = 4 per type of manure) (Eriksson 2001).

	Dairy slurry			Pig liquid manure			Pig solid manure		
	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
As	0.62	0.20	1.1	1.0	0.71	1.2	1.3	0.52	1.8
Cd	0.12	0.09	0.18	0.28	0.27	0.32	0.25	0.19	0.36
Cr	2.3	1.2	2.9	6.8	5.7	7.8	11	7.1	17
Cu	24	23	25	149	136	161	113	50	161
Hg	0.014	0.012	0.018	0.009	<0.01	0.016	0.019	0.013	0.024
Ni	4.6	2.1	7.3	4.5	3.3	5.8	6.1	3.3	9.4
Pb	0.98	0.81	1.2	1.19	0.92	1.15	2.0	1.3	2.7
Se	0.56	0.40	0.77	1.4	1.4	1.5	0.89	0.19	1.3
V	2.5	1.5	3.3	6.6	5.2	9.2	6.1	4.1	7.7
Zn	154	128	170	582	394	680	680	347	821

Table 35. Heavy metal contents (mg kg<sup>-1</sup> dm) in livestock manures (KTBL 2005).

Livestock manures	Cd	Cr	Cu	Ni	Pb	Zn
<b>Cattle FYM</b>						
Weighted mean	0.3	7.5	23	4.4	3.8	119
Range	0.04-3.1	0.1-170	0.3-191	0.2-28	0.1-92	9.6-691
n	348	200	345	175	331	345
<b>Cattle slurry</b>						
Weighted mean	0.4	6.9	42	6.2	5.6	207
Range	0.04-5.5	0.2-170	0.1-741	0.1-57	0.1-75	2.0-1908
n	680	431	720	475	679	721
<b>Pig FYM</b>						
Weighted mean	0.5	14	237	8.3	3.6	926
Range	0.1-1.3	0.7-257	38-1227	0.9-24	0.6-9.9	116-5513
n	151	109	150	109	150	150
<b>Pig slurry</b>						
Weighted mean	0.3	0.4	193	12	3.0	934
Range	0.02-4.0	0.2-37	12-1802	0.1-50	0.3-112	5.0-5832
n	1128	235	1143	180	1126	1143
<b>Broiler/turkey FYM</b>						
Weighted mean	0.4	20	89	6.2	3.7	353
Range	0.1-1.2	1.3-1980	8.4-760	2.2-21	1.0-24	52-790
n	102	49	105	46	102	105
<b>Laying hen FYM</b>						
Weighted mean	0.4	6.5	62	7.0	3.2	471
Range	0.1-2.0	2.1-13	15-486	2.5-11	1.4-15	120-789
n	60	25	66	22	60	66

### 3.4 Trace element inputs

Total trace element inputs from fertiliser products and atmospheric depositions on the crop farms and additionally from feeds on the dairy farms estimated by the RAKAS-model are presented in Table 36. Total inputs were the same for Cd, Cr, Cu, Ni, Pb and Zn in the AROMIS-model. Compared to the respective European values, the trace element inputs were clearly lower in Finland than in Europe, on average. Due to lower crop yields and farm animal density, smaller amounts of fertiliser products and feeds are imported the Finnish farms. Furthermore, imported materials may have lower trace element contents in Finland than in other parts of Europe. In addition, atmospheric depositions of the trace elements in Finland were nearly the lowest ones in Europe. Contrary to Finland, the total trace elements inputs were higher on the crop farms than on the dairy farms in Europe (Table 36).

Table 36. Total trace element inputs on the five crop farms (1-5) in southwestern Finland and on the five dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model and their means by farm types in Finland and Europe (EUR) according to the AROMIS-project (Eckel et al. 2005).

Farm	Total trace element inputs, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
1*	0.393	(364)	18	8.3	9.4	124	0.067	(6039)	2.3	5.1
2	0.330	2.3	7.6	2.3	8.8	31	0.052	4.5	1.1	4.3
3	0.320	2.2	8.0	2.5	8.8	158	0.050	4.4	1.1	3.6
4	0.401	4.9	13	5.3	10	134	0.063	25	4.5	4.8
5*	0.427	(515)	24	12	9.8	281	0.074	(8543)	2.9	5.7
<b>Mean</b>	<b>0.375</b>	<b>3.1</b>	<b>14</b>	<b>6.1</b>	<b>9.4</b>	<b>146</b>	<b>0.061</b>	<b>11</b>	<b>2.4</b>	<b>4.7</b>
EUR	4.4	107	426	98	86	1014	-	-	-	-
6*	0.392	(726)	43	18	6.9	283	0.089	(12027)	4.9	5.5
7	0.437	13	49	5.4	6.8	719	0.070	28	3.9	9.0
8	0.347	7.9	17	12	7.5	369	0.062	12	2.6	4.9
9	0.317	13	45	2.6	6.1	383	0.063	14	1.1	5.4
10	0.313	6.9	25	3.6	5.9	449	0.061	16	2.3	6.2
<b>Mean</b>	<b>0.361</b>	<b>10</b>	<b>36</b>	<b>8.3</b>	<b>6.6</b>	<b>441</b>	<b>0.069</b>	<b>18</b>	<b>3.0</b>	<b>6.2</b>
EUR	2.9	63	273	40	44	923	-	-	-	-

\*Slag was used for soil improvement. () not included in the mean.

#### 3.4.1 Fertiliser products

In 2004, the test farms used the following fertiliser products: mineral fertilisers, limes, calcites and slag. Total inputs from the various fertiliser products by the farms are presented in Table 37. In general, mean trace element inputs seem to be somewhat higher on the dairy farms than on the crop farms. To produce

high grass yields for cattle, strong fertilisation is needed. To neutralize the field after manure applications sufficient liming is also needed. However, the biggest inputs of many trace elements, especially V, came from the use of steel slag that contained about 1% of V. The steel slag was also a remarkable source of Cr. Trace element inputs from all the fertiliser products excluding steel slag were clearly lower in Finland than in Europe, on average (Table 37).

Table 37. Trace element inputs from fertiliser products on the five crop farms (1-5) in southwestern Finland and on the five dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model and their means by farm types in Finland and in Europe (EUR) according to the AROMIS-project (Eckel et al. 2005).

Farm	Inputs from fertiliser products, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
1*	0.100	(364)	13	(6.7)	0.80	97	0.017	(6036)	1.3	4.4
2	0.036	1.5	2.1	0.66	0.18	4.2	0.002	1.6	0.10	3.6
3	0.026	1.5	2.5	0.90	0.20	131	0.002	1.6	0.11	2.9
4	0.108	4.2	7.6	3.7	1.4	108	0.013	22	3.5	4.1
5*	0.133	(515)	(18)	(10)	1.2	255	0.024	(8540)	1.9	5.1
<b>Mean</b>	<b>0.081</b>	<b>2.4</b>	<b>4.1</b>	<b>1.8</b>	<b>0.8</b>	<b>119</b>	<b>0.006</b>	<b>8.4</b>	<b>1.4</b>	<b>4.0</b>
EUR**	2.5	26	12	6.6	6.1	59	-	-	-	-
6*	0.190	(723)	(25)	(14)	2.0	199	0.038	(12024)	4.3	4.2
7	0.125	4.8	5.5	3.1	1.3	480	0.013	19	3.1	7.1
8	0.149	4.3	3.2	11	2.4	237	0.009	7.2	2.0	4.0
9	0.024	1.5	1.5	0.49	0.20	173	0.002	1.7	0.13	4.1
10	0.079	2.9	3.2	1.7	0.70	321	0.008	10	1.6	5.2
<b>Mean</b>	<b>0.113</b>	<b>3.4</b>	<b>3.4</b>	<b>4.1</b>	<b>1.3</b>	<b>282</b>	<b>0.008</b>	<b>9.5</b>	<b>2.2</b>	<b>4.9</b>
EUR**	1.9	22	7.0	6.8	4.2	40	-	-	-	-

\*Slag was used for soil improvement. \*\* Inputs only from mineral fertilisers. () = not included in the mean.

### 3.4.2 Commercial feeds

The five dairy farms in Ostrobothnia used more than 20 different commercial feeds for animal feeding. Feeds were among others: Tähti-145, Onni-Kivennäinen, Milkkeri melassoitu rypsi, Amino-Maituri, Mulli-Melli, Kesä-Namino, Mullin herkku, Lypsymelli, Startti, Huippu Krossi 23, Aseto-Melli, Hertta-kivennäinen, Primo vasikanrehu, Start instant, Maituri 10000, Rouhetiiviste, Melassileike, Tilaseos nautakarjalle, Viher Hertta-Minera Muro, Amino maituri, Pihatto-Melli and Krono 135. Only about ten of them were possible to analyse here for the trace elements. The commercial feeds selected for the analysis were those that were most often used on the test farms. In the balance calculations, we also had to apply the best available analytical results for those feeds not analysed. Thus, reliability of the trace element inputs calculated for those feeds cannot be the highest.

In 2004, mean total inputs from the commercial feeds varied between 0.005 and 141 g ha<sup>-1</sup> a<sup>-1</sup> on the test farms. Trace element inputs in the commercial feeds decreased in the following order: Zn>Cu>Cr>V>Ni>Se>Pb>As>Cd>Hg, on average. Compared to the inputs from the fertiliser preparations, commercial feeds were a major source for only Cr and Cu (Table 37), but a minor source for the other trace elements studied on the dairy farms (Table 37). In general, the trace element inputs from commercial feeds were lower in Finland than in Europe (Table 38). Chromium was an exception.

Table 38. Trace element inputs from commercial feeds on the five dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model and their means in Finland and in Europe (EUR) according to the AROMIS-project (Eckel et al. 2005).

Farm	Inputs from commercial feeds, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
6	0.034	1.8	14	3.4	0.17	67	0.001	1.4	0.10	0.90
7	0.145	7.4	40	1.3	0.75	222	0.007	7.5	0.40	1.4
8	0.031	3.1	10	0.30	0.31	115	0.003	3.1	0.15	0.48
9	0.108	11	40	1.1	1.1	193	0.011	11	0.54	0.80
10	0.067	3.5	18	0.89	0.30	110	0.003	3.5	0.20	0.46
<b>Mean</b>	<b>0.077</b>	<b>5.4</b>	<b>24</b>	<b>1.4</b>	<b>0.53</b>	<b>141</b>	<b>0.005</b>	<b>5.3</b>	<b>0.28</b>	<b>0.81</b>
EUR	1.17	3.0	75	7.5	11	432	-	-	-	-

### 3.4.3 Atmospheric depositions

In Finland, the main national anthropogenic source of trace elements into the air is the burning of fossil fuels for energy production and for work machines (Table 39). However, agriculture is not a source of any of the monitored trace elements (SYKE 2005).

Table 39. Domestic micro-element emissions (tonnes) by sectors into air in Finland in 2003 (SYKE 2005).

Sectors	Domestic micro-element emissions (tonnes) into air in Finland in 2003									
	Pb	Cd	Hg	As	Cr	Cu	Ni	Zn	V	
Energy	28.5	0.9	0.5	2.2	12.4	14.1	23.9	37.3	67.3	
Traffic	0	0	0	0	0	0	0	0	0	
Industrial processes	5.0	0.3	0.2	1.0	15	7.4	9.8	25.3	1.0	
Use of solvents and other chemicals	0	0	0	0	0	0	0	0	0	
Agriculture	0	0	0	0	0	0	0	0	0	
Waste	0.02	0	0	0.03	0.02	0.01	0.02	0.2	0.05	
Alltogether	33.5	1.2	0.8	3.2	27.4	21.5	33.8	62.8	68.3	

Trace element depositions onto the fields of the crop farms in southwestern Finland were estimated from the measurements made at the three research stations of the Meteorological Institute (Utö, Virolahti, Kotinen) (Table 40). As well, trace element depositions onto the dairy farm fields in Ostrobothnia were estimated from the measurements of two Meteorological Institute research stations (Hailuoto and Hietajärvi) (Table 41). Regional bulk deposition values were collected from the annual reports (years 1997-2002) of Meteorological Institute (Leinonen 1999a, Leinonen 1999b, Leinonen 2000, Leinonen 2001, Salmi 2001, Salmi 2002). For selenium, the bulk deposition value was 0.67 g ha<sup>-1</sup> a<sup>-1</sup> in southern Finland and 0.47 g ha<sup>-1</sup> a<sup>-1</sup> in northern Finland (Alfthan et al. 1995). For mercury, a value of 0.05 g ha<sup>-1</sup> a<sup>-1</sup> was used for both farm types. There seems to be a clear decreasing trend in the trace element atmospheric depositions from 1997 to 2002 for trace elements monitored in Finland. Atmospheric depositions of most elements decreased towards the north. Thus, the annual trace element depositions per hectare on the dairy farms were about half of those on the crop farms (Table 42). Mostly, the trace elements depositions were lower in Finland than in Denmark or Sweden (Table 42).

Table 40. Annual trace element depositions ( $\mu\text{g m}^{-2}$ ) measured in three research stations (Utö, Virolahti and Kotinen) between 1997-2002 by the Finnish Meteorological Institute (1998-2003). NL = northern latitude; EL = Eastern longitude; Prec. = Precipitation of water.

Station	NL	EL	Year	Prec. mm	Trace element deposition, $\mu\text{g m}^{-2}$							
					Zn	Pb	Cu	Cd	Cr	Ni	V	As
Utö	59° 47'	21° 23'	1997	430	3080	1160	940	28	90	198	357	149
Utö	59° 47'	21° 23'	1998	390	3170	1130	810	28	74	234	331	108
Utö	59° 47'	21° 23'	1999	329	2580	980	890	27	132	168	335	137
Utö	59° 47'	21° 23'	2000	335	2320	1010	650	32	84	151	252	119
Utö	59° 47'	21° 23'	2001	363	2300	890	520	24	40	109	254	82
Utö	59° 47'	21° 23'	2002	269	2040	600	400	24	86	118	226	68
Virolahti	60° 32'	27° 41'	1997	438	2490	970	640	27	74	241	390	143
Virolahti	60° 32'	27° 41'	1998	616	5270	1030	670	41	80	240	380	143
Virolahti	60° 32'	27° 41'	1999	326	2260	880	390	38	127	153	340	109
Virolahti	60° 32'	27° 41'	2000	514	3020	1470	500	53	87	180	380	144
Virolahti	60° 32'	27° 41'	2001	582	2420	860	480	43	52	116	300	94
Virolahti	60° 32'	27° 41'	2002	412	2580	660	470	27	95	165	280	52
Kotinen	61° 14'	25° 04'	1997	526	1900	590	520	17	63	221	221	63
Kotinen	61° 14'	25° 04'	1998	756	3440	800	580	27	45	249	280	80
Kotinen	61° 14'	25° 04'	1999	528	3230	690	380	22	53	127	206	86
Kotinen	61° 14'	25° 04'	2000	662	2140	760	420	30	40	113	219	74
Kotinen	61° 14'	25° 04'	2001	662	1930	600	400	24	46	86	199	62
Kotinen	61° 14'	25° 04'	2002	515	1620	400	310	16	57	67	175	22
<b>Mean</b>	<b>(g/ha)</b>				<b>26.6</b>	<b>8.60</b>	<b>5.54</b>	<b>0.29</b>	<b>0.74</b>	<b>1.63</b>	<b>2.85</b>	<b>0.96</b>

Table 41. Annual trace element depositions ( $\mu\text{g m}^{-2}$ ) measured in two research stations (Hailuoto, Hietajärvi) between 1997-2002 by the Finnish Meteorological Institute (1998-2003). NL = Northern latitude; EL = Eastern longitude; Prec. = Precipitation.

Station	NL	EL	Year	Prec. mm	Trace element deposition, $\mu\text{g m}^{-2}$							
					Zn	Pb	Cu	Cd	Cr	Ni	V	As
Hailuoto	63° 10'	30° 43'	1997	268	1790	310	490	9	35	121	153	35
Hailuoto	63° 10'	30° 43'	1998	459	1320	370	330	11	23	124	161	39
Hailuoto	63° 10'	30° 43'	1999	335	3170	600	380	26	47	97	201	47
Hailuoto	63° 10'	30° 43'	2000	418	1720	630	410	19	50	96	255	59
Hailuoto	63° 10'	30° 43'	2001	364	1270	440	330	14	22	62	149	40
Hailuoto	63° 10'	30° 43'	2002	311	1010	260	320	8	71	50	149	13
Hietajärvi	65° 00'	24° 41'	1997	571	1190	470	550	13	51	171	160	50
Hietajärvi	65° 00'	24° 41'	1998	727	3980	710	460	23	51	174	204	64
Hietajärvi	65° 00'	24° 41'	1999	510	1430	580	300	20	46	107	204	58
Hietajärvi	65° 00'	24° 41'	2000	640	1670	610	430	24	90	102	173	61
Hietajärvi	65° 00'	24° 41'	2001	511	1140	480	280	19	26	61	153	49
Hietajärvi	65° 00'	24° 41'	2002	476	1040	280	290	14	76	57	124	11
<b>Mean</b>	<b>(g/ha)</b>				<b>17.3</b>	<b>4.78</b>	<b>3.81</b>	<b>0.17</b>	<b>0.49</b>	<b>1.02</b>	<b>1.74</b>	<b>0.44</b>

Table 42. Trace element inputs from atmospheric deposition on the five crop farms (1-5) in southwestern Finland and on the five dairy farms (6-10) in Ostrobothnia in 2004 used in the RAKAS-model and means of atmospheric depositions in Sweden (SWE) and Denmark (DE) (Eckel et al. 2005).

Farm	Atmospheric deposition of trace elements, $\text{g ha}^{-1} \text{a}^{-1}$									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
1	0.293	0.74	5.5	1.6	8.6	27	0.050	2.85	1.0	0.67
2	0.293	0.74	5.5	1.6	8.6	27	0.050	2.85	1.0	0.67
3	0.293	0.74	5.5	1.6	8.6	27	0.050	2.85	1.0	0.67
4	0.293	0.74	5.5	1.6	8.6	27	0.050	2.85	1.0	0.67
5	0.293	0.74	5.5	1.6	8.6	27	0.050	2.85	1.0	0.67
<b>Mean</b>	<b>0.293</b>	<b>0.74</b>	<b>5.5</b>	<b>1.6</b>	<b>8.6</b>	<b>27</b>	<b>0.050</b>	<b>2.85</b>	<b>1.0</b>	<b>0.67</b>
6	0.167	0.49	3.8	1.0	4.8	17	0.050	1.74	0.44	0.47
7	0.167	0.49	3.8	1.0	4.8	17	0.050	1.74	0.44	0.47
8	0.167	0.49	3.8	1.0	4.8	17	0.050	1.74	0.44	0.47
9	0.167	0.49	3.8	1.0	4.8	17	0.050	1.74	0.44	0.47
10	0.167	0.49	3.8	1.0	4.8	17	0.050	1.74	0.44	0.47
<b>Mean</b>	<b>0.167</b>	<b>0.49</b>	<b>3.8</b>	<b>1.0</b>	<b>4.8</b>	<b>17</b>	<b>0.050</b>	<b>1.74</b>	<b>0.44</b>	<b>0.47</b>
SWE	0.8	5.0	15	0.5	6.3	118	-	-	-	-
DE	0.5	1.5	17	4.2	14	94	-	-	-	-

### 3.4.4 Weathering

The annual inputs of heavy metals from the weathering of soil parent material were estimated according to Mäkelä-Kurtto and Sippola (2001) and are presented in Table 43. Similar method has been earlier applied for the long-term average weathering release rates of calcium and magnesium by Olsson et al. (1993) and Johansson and Tarvainen (1996). The trace element inputs seem to be minor as compared to the inputs from other sources. Hence, the annual weathering of the trace elements was not taken into account in the RAKAS-model. However, Starr and others (2003) reported that the magnitude of the annual weathering release rates were found to be similar to current annual deposition, litterfall and leaching fluxes of heavy metals in a remote, boreal coniferous forest catchment in eastern Finland. Thus, in areas where other input of heavy metals is low, weathering can be a significant input source.

Table 43. Estimated average annual heavy metal inputs from the litho- into the pedosphere (Johansson & Tarvainen 1996).

	g ha <sup>-1</sup> a <sup>-1</sup>					
Element	Cd	Cr	Cu	Ni	Pb	Zn
Input	0.004	0.7	0.4	0.5	0.8	0.7

### 3.5 Trace element outputs

Total trace element outputs through various routes on the crop and dairy farms in Finland in 2004 estimated by the RAKAS-model are presented in Table 44. On the crop farms, the total outputs of Cr, Cu, Ni, Pb, V and As were higher than the respective total inputs (Table 36). A total output for Cd was lower than the total input for Cd. A total Se output was much lower than the total Se input. In the case of Zn, a total input and total output were quite equal. On the dairy farms, total outputs were clearly lower than the total inputs (Table 36). An exception was Cr. In general, the trace element outputs were smaller in Finland than in Europe, but the total outputs of Cr, Ni and Pb on the crop farms and Cr on the dairy farms were larger than the respective figures, on average (Table 44). The main reason might be outflow of these trace elements via erosion in Finland.

Table 44. Total trace element outputs on the five crop farms (1-5) in southwestern Finland and on the five dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model and their means by farm types in Finland and in Europe (EUR) according to the AROMIS-project (Eckel et al. 2005).

Farm	Total trace element outputs, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
1*	0.390	34	34	20	11	156	0.030	52	4.2	0.73
2	0.250	40	31	25	12	113	0.026	54	3.1	0.25
3	0.300	16	36	11	7.2	163	0.028	27	2.0	0.80
4	0.323	34	33	20	7.8	150	0.045	65	3.1	0.76
5*	0.265	41	37	24	14	123	0.040	63	4.3	0.72
<b>Mean</b>	<b>0.306</b>	<b>33</b>	<b>34</b>	<b>20</b>	<b>10</b>	<b>141</b>	<b>0.034</b>	<b>52</b>	<b>3.3</b>	<b>0.65</b>
EUR	1.0	9.0	54	18	5.3	265	-	-	-	-
6*	0.180	18	17	12	4.8	65	0.036	29	3.0	0.26
7	0.172	12	14	8.3	8.2	78	0.043	14	2.3	0.44
8	0.240	22	45	15	5.7	259	0.067	53	3.6	1.0
9	0.127	7.1	8	6.7	2.4	47	0.029	10	1.3	0.19
10	0.168	6.7	18	7.6	3.3	83	0.034	13	1.3	0.39
<b>Mean</b>	<b>0.177</b>	<b>13</b>	<b>20</b>	<b>10</b>	<b>4.9</b>	<b>106</b>	<b>0.042</b>	<b>24</b>	<b>2.3</b>	<b>0.46</b>
EUR	0.6	6.4	42	20	6.1	154	-	-	-	-

\*Slag was used for soil improvement.

### 3.5.1 Crop plants

The magnitude of the trace element outputs depends on the yields, their trace element contents and the amount of the yield exported. On the crop farms, the outputs in crop plants were clearly higher than on the dairy farms, because most of the crop yields were exported by the crop farms (Table 45). Instead, a great deal of the crop plants were grown for own use, for cattle feeding, on the dairy farms. On average, 78 g of Zn and 15 g of Cu were annually transported in the crop plants out of the crop farms from one hectare. The figures for dairy farms were only 3.6 g and 0.84 g, respectively. Also, the outputs of other elements in the crops studied were much lower on the dairy farms than on the crop farms.

In general, only a small part of the trace elements totally added into the soil flowed out of the farms in crop plants (Table 45). However, on the crop farms, the total Cu outputs were slightly higher than the Cu inputs, on average (Table 36). Trace element outputs in the crop plants were remarkably higher in Europe (Table 45) than in Finland. The main reason might be the higher crop yields and their higher trace element contents in Europe than in Finland.

Table 45. Trace element outputs in crop plants on the five crop farms (1-5) in southwestern Finland and on the five dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model and their means by farm types in Finland and in Europe (EUR) according to the AROMIS-project (Eckel et al. 2005).

Farm	Trace element outputs in crop plants, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
1*	0.195	0.17	18	1.5	0.08	94	0.000	0.14	0.03	0.64
2	0.050	0.13	7.6	0.54	1.1	37	0.001	0.13	0.02	0.19
3	0.050	0.20	24	0.74	0.15	120	0.028	0.10	0.02	0.70
4	0.078	0.06	15	0.58	0.06	77	0.010	0.03	0.02	0.65
5*	0.055	0.08	12	0.42	0.03	61	0.000	0.02	0.03	0.55
<b>Mean</b>	<b>0.086</b>	<b>0.13</b>	<b>15</b>	<b>0.76</b>	<b>0.28</b>	<b>78</b>	<b>0.008</b>	<b>0.08</b>	<b>0.024</b>	<b>0.55</b>
EUR	0.7	2.7	34	6.7	1.6	153	-	-	-	-
6*	0.005	0.01	2.6	0.02	0.10	10	0.001	0.01	0.00	0.02
7	0.000	0.00	0.00	0.00	0.00	0.00	0.000	0.00	0.00	0.00
8	0.001	0.01	0.60	0.02	0.02	2.3	0.000	0.00	0.00	0.00
9	0.000	0.00	0.00	0.00	0.00	0.00	0.000	0.00	0.00	0.00
10	0.001	0.01	1.0	0.01	0.02	5.5	0.000	0.05	0.00	0.01
<b>Mean</b>	<b>0.0001</b>	<b>0.006</b>	<b>0.84</b>	<b>0.01</b>	<b>0.028</b>	<b>3.6</b>	<b>0.000</b>	<b>0.01</b>	<b>0.00</b>	<b>0.006</b>
EUR	0.7	1.4	17	5.6	1.2	96	-	-	-	-

\*Slag was used for soil improvement.

The ability of plants to take up elements varies between plant species (Table 46). Elements in plants are taken up partly from the soil by the roots and partly from the air after the trace elements have been deposited on the plant surfaces. Thus, the roots may have the highest trace element content. The leafy plants are sensitive to atmospheric depositions and can contain elevated levels of trace elements. Cereal grains are well protected from the atmospheric depositions by the husks and the trace element contents in the plant usually decrease towards the fruits and seeds. According to Mäkelä-Kurtto (1996) mean uptakes of Cd, Pb, Cu and Zn varied between 0.02-1.47, 0.22-1.72, 6.8-27.3 and 58-213 g ha<sup>-1</sup> a<sup>-1</sup>, respectively, and depended on the metal content of the plants and annual crop yields.

Table 46. Mean annual uptakes ( $\text{g ha}^{-1} \text{a}^{-1}$ ) of cadmium (Cd), lead (Pb), copper (Cu) and zinc (Zn) by various crop plants in Finland (Mäkelä-Kurtto 1996).

Crop plant	Cd	Pb	Cu	Zn
Wheat	0.19	0.22	20.1	156
Rye	0.07	0.48	17.4	119
Barley	0.09	0.31	21.7	115
Oat	0.07	0.29	11.2	120
Sugar beet roots	1.47	1.10	33.0	213
Oil rape seeds	0.11	0.37	6.8	58
Pea seeds	0.02	0.27	19.4	94
Potato	0.22	0.24	27.3	78
Hay	0.11	1.32	15.7	90
Silage 1. cut	0.15	1.72	25.8	133
Silage 2. cut	0.08	1.22	11.6	60
Mean, g/ha	0.14	0.75	18.7	115

### 3.5.2 Milk and meat

In general, total trace element outputs in milk and meat were very low in the dairy farms studied due to their low trace element contents (Table 47). The annual Zn output was the highest,  $21 \text{ g ha}^{-1}$ , on average. The mean Cu and Se outputs were  $0.34$  and  $0.12 \text{ g ha}^{-1} \text{a}^{-1}$ , respectively. Only Zn, Se and Cr were exported more into the milk and meat (Table 47) than in the crop plants (Table 45) on the dairy farms. Compared to the European level, the trace element outputs in milk and meat were very low in Finland (Table 47).

Table 47. Trace element outputs in milk and meat on the five dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model and their means in Finland and in Europe (EUR) according to the AROMIS-project (Eckel et al. 2005).

Farm	Trace element outputs in milk and meat, $\text{g ha}^{-1} \text{a}^{-1}$									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
6*	0.000	0.05	0.20	0.00	0.01	16	0.008	0.00	0.00	0.10
7	0.001	0.08	0.40	0.02	0.01	22	0.014	0.01	0.01	0.13
8	0.001	0.07	0.39	0.01	0.01	21	0.011	0.01	0.01	0.10
9	0.000	0.08	0.43	0.02	0.01	28	0.013	0.00	0.01	0.14
10	0.000	0.06	0.30	0.01	0.01	16	0.010	0.01	0.01	0.12
<b>Mean</b>	<b>0.0002</b>	<b>0.07</b>	<b>0.34</b>	<b>0.012</b>	<b>0.01</b>	<b>21</b>	<b>0.011</b>	<b>0.006</b>	<b>0.008</b>	<b>0.12</b>
EUR	0.2	0.9	22	1.8	0.7	113	-	-	-	-

\*Slag was used for soil improvement.

### 3.5.3 Leaching

Leaching values used in the RAKAS-model were mainly based on the Swedish measurements (Andersson 1992). Leaching values for cadmium and selenium were based on Finnish measurements (Mäkelä-Kurtto et al. 2003 and Yläran-ta 1982). Leaching of selenium was considered to be zero (Vääriskoski 1987). According to Holodov (1968), the amount of vanadium leached from the soil is about 15% of the amount of vanadium transported from the soil with erod-ed soil particles. In this study, vanadium outputs via leaching were calculated through erosion (Chapter 3.5.4). Trace element leaching values used in the RA-KAS-model on the crop and dairy farms are presented in Table 46.

In the RAKAS-model, the trace element leaching figures were the same for the crop and dairy farms (Table 48). However, they were rather reasonable, because the clay soils of crop farms contained much more of all the trace elements than the finesand soils of the dairy farms (Table 11). As well, leaching is known to be lower from clay soils than from coarse mineral soils.

Table 48. Trace element leaching values used in the RAKAS-model on the five crop farms (1-5) in southwestern Finland and on the five dairy farms (6-10) in Ostrobothnia in 2004.

Farm	Leaching, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
1	0.100	0.70	4.3	3.9	0.50	7.5	0.010	6.8	0.48	0.00
2	0.100	0.70	4.3	3.9	0.50	7.5	0.010	7.0	0.48	0.00
3	0.100	0.70	4.3	3.9	0.50	7.5	0.010	3.5	0.48	0.00
4	0.100	0.70	4.3	3.9	0.50	7.5	0.010	6.5	0.48	0.00
5	0.100	0.70	4.3	3.9	0.50	7.5	0.010	8.3	0.48	0.00
<b>Mean</b>	<b>0.100</b>	<b>0.70</b>	<b>4.3</b>	<b>3.9</b>	<b>0.50</b>	<b>7.5</b>	<b>0.010</b>	<b>6.4</b>	<b>0.48</b>	<b>0.00</b>
6	0.100	0.70	4.3	3.9	0.50	7.5	0.010	3.7	0.48	0.00
7	0.100	0.70	4.3	3.9	0.50	7.5	0.010	1.9	0.48	0.00
8	0.100	0.70	4.3	3.9	0.50	7.5	0.010	6.4	0.48	0.00
9	0.100	0.70	4.3	3.9	0.50	7.5	0.010	1.2	0.48	0.00
10	0.100	0.70	4.3	3.9	0.50	7.5	0.010	1.5	0.48	0.00
<b>Mean</b>	<b>0.100</b>	<b>0.70</b>	<b>4.3</b>	<b>3.9</b>	<b>0.50</b>	<b>7.5</b>	<b>0.010</b>	<b>2.9</b>	<b>0.48</b>	<b>0.00</b>

The AROMIS-model defaulted trace element concentrations in the leachates that were based on the European measurements and which depended on the soil type (Table 49). In this model, the precipitation and evaporation of water on the farms were needed and fed in the model. The annual precipitation of water used in the model was based on the measurements made by Finnish Environmental Institute in the station, closest (10-70 km) to the farm. Evaporation values for water were estimated according to Järvinen and Kuusisto (1995). The model calculated leaching of water (Table 50). The dominant soil type on all the five

crop farms (1-5) was clay which in the model was classified as river clay (RC). The dominant soil type on the four dairy farms (6, 7, 9 and 10) was finesand and on one farm (8) organic soil. In the AROMIS-model, the soils were classified as sand (S) and peat (P), respectively.

Table 49. Default values for trace element concentrations ( $\mu\text{g l}^{-1}$ ) in leachate used in the AROMIS-model.

Soil type	Concentration in leachate, $\mu\text{g l}^{-1}$					
	Cd	Cr	Cu	Ni	Pb	Zn
Sand (S)	0.34	2.1	11	1.6	8.5	57
Sea clay (SC)	0.03	0.35	2	0.8	4.0	3.5
River clay (RC)	0.03	0.35	5	0.7	2.6	11
Peat (P)	0.03	1.0	6	5.4	4.6	33

According to estimations from the AROMIS-model, the mean trace element outputs via leaching were much higher from the finesand soils on the dairy farms than from the clay soils on the crop farms (Table 50). Also the leaching estimations were much higher than the respective figures used in the RAKAS-model (Table 48).

Table 50. Trace element leaching ( $\text{g ha}^{-1} \text{a}^{-1}$ ) from the soil estimated by the AROMIS-model for the five crop farms (1-5) in southwestern Finland and for the five dairy farms in Ostrobothnia in 2004 as estimated with the AROMIS-model and their means by farm types in Finland and Europe (EUR) according to the AROMIS-project (Eckel et al. 2005). In addition, precipitation (Prec.), evaporation (Evap.) and leaching (Leach.) of water in Finland in 2004.

Farm	Soil type	AROMIS-Leaching, $\text{g ha}^{-1} \text{a}^{-1}$						In 2004, $\text{mm year}^{-1}$		
		Cd	Cr	Cu	Ni	Pb	Zn	Prec.	Evap.	Leach.
1	RC	0.112	1.30	18.6	2.60	9.67	40.9	787	415	372
2	RC	0.122	1.42	20.3	2.84	10.5	44.6	813	408	405
3	RC	0.050	0.59	8.4	1.18	4.37	18.5	631	463	168
4	RC	0.105	1.23	17.5	2.45	9.10	38.5	779	429	350
5	RC	0.118	1.38	19.7	2.76	10.2	43.3	779	385	394
<b>Mean</b>		<b>0.101</b>	<b>1.2</b>	<b>17</b>	<b>2.4</b>	<b>8.8</b>	<b>37</b>	<b>758</b>	<b>420</b>	<b>338</b>
EUR		0.2	1.8	14	7.9	2.1	41			
6	S	1.044	6.44	33.8	4.91	26.10	175	636	329	307
7	S	0.904	5.59	29.3	4.26	22.61	152	642	376	266
8	P	0.095	3.17	19.0	17.1	14.58	105	646	329	317
9	S	1.044	6.45	33.8	4.91	26.10	175	683	376	307
10	S	1.044	6.45	33.8	4.91	26.10	175	683	376	307
<b>Mean</b>		<b>1.009*</b>	<b>6.2*</b>	<b>33*</b>	<b>4.8*</b>	<b>25*</b>	<b>169*</b>	<b>658</b>	<b>357</b>	<b>301</b>
EUR		0.3	3.6	27	15	6.6	80			

\* = Mean does not include the Farm 8 with peat; RC = River clay, S = Sand; P = Peat

### 3.5.4 Erosion

Measurements of trace elements eroded with soil particles from cultivated soils were not available for Finland. The mean slope (m/100 m) of arable land is 1.6%, on average, and only 10% of the fields have a mean slope more than 5% (Puustinen et al. 1994). An annual loss of total solids from Finnish fields may vary from 50 kg to 7 000 kg/ha depending on many factors such as soil type, slope of the fields, and precipitation, as reported by Uusi-Kämpä (1989). In the RAKAS-model, the annual loss of total solids from cultivated soil was assumed to be 500 kg ha<sup>-1</sup>. The trace element contents of the eroded solids were assumed to be the same as the initial soil. Thus, the trace element figures for erosion depended on the soil trace element content, respectively. The higher the trace element contents in the soil, the higher the trace element outputs via eroded soil material. Hence, the outputs via erosion were about two to three times higher in the crop farms than in the dairy farms (Table 51). Erosion was not taken into account in the AROMIS-model.

Table 51. Trace element outputs via eroded soil material on the five crop farms in southwestern Finland and on the five dairy farms in Ostrobothnia in 2004 as estimated with the RAKAS-model and their means by farm types.

Farm	Trace element outputs via eroded soil material, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
1*	0.095	33	12	15	10	54	0.020	45	3.7	0.09
2	0.100	39	19	21	10	68	0.015	46	2.9	0.08
3	0.140	15	7.9	6.2	6.5	34	0.020	23	1.5	0.20
4	0.145	33	14	15	7.2	66	0.035	43	2.5	0.11
5*	0.110	40	21	20	13	55	0.030	55	3.8	0.18
<b>Mean</b>	<b>0.118</b>	<b>32</b>	<b>15</b>	<b>15</b>	<b>9</b>	<b>55</b>	<b>0.024</b>	<b>42</b>	<b>2.9</b>	<b>0.13</b>
6*	0.075	17	10	7.9	4.3	32	0.019	25	2.5	0.15
7	0.050	11	4.1	3.7	3.8	16	0.015	12	1.1	0.11
8	0.080	20	11	8.1	4.8	28	0.025	43	2.3	0.20
9	0.027	6.3	2.9	2.8	1.9	11	0.006	8.3	0.84	0.05
10	0.035	5.4	3.9	2.4	1.7	12	0.005	10	0.67	0.07
<b>Mean</b>	<b>0.053</b>	<b>12</b>	<b>6.4</b>	<b>5.0</b>	<b>3.3</b>	<b>20</b>	<b>0.014</b>	<b>20</b>	<b>1.5</b>	<b>0.12</b>

\*Slag was used for soil improvement.

### 3.5.5 Cattle manure

Farm animal manure used on the farm itself is of internal flow and was not taken into account in the balance calculations. If the manure was imported or exported then it was considered as an input or output, respectively. In this study, no farm imported the manure, but three of the dairy farms exported 60 to 700 tonnes of manure to another farm as fresh matter. In these farms, manure is a route for the trace elements out of the farm. As

indicated by Table 52, remarkable amounts of the trace elements could be transported to another farm and thus, reduce the trace element inputs into own fields. In the cases of some trace elements, the mean outputs via exported manure (Table 52) might be even higher than the trace element inputs in the commercial feeds (Table 38), respectively. Thus, manure exporting is a good way to reduce the trace element loads into the soil on the farm itself. Depending on the trace element content of manure and the amount of manure exported, the mean trace element outputs increased in the following order: Hg<Cd<As=Se<Pb<Cr<Ni<V<Cu<Zn.

Table 52. Trace element outputs in manure exported from the three dairy farms in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Farm	Outputs in cattle manure, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
7	0.021	0.20	5.4	0.63	3.9	32	0.004	0.30	0.08	0.20
8	0.058	1.1	28	2.9	0.43	200	0.020	4.60	0.89	0.70
10	0.032	0.61	9.0	1.3	1.1	43	0.009	0.97	0.15	0.19
<b>Mean</b>	<b>0.037</b>	<b>0.64</b>	<b>14.1</b>	<b>1.6</b>	<b>1.8</b>	<b>92</b>	<b>0.011</b>	<b>1.96</b>	<b>0.37</b>	<b>0.36</b>

### 3.6 Internal flow and unidentified input of trace elements on dairy farms

The AROMIS-model automatically calculated the internal flows of the trace elements in the home-grown feeds (Table 53) and manure (Table 54) used on the dairy farm itself. The mean internal flows for Cd, Cu and Zn were higher and for Cr, Ni and Pb lower in the home-grown feeds than in manure. Annually, 0.35 g of Cd, 4 g of Cr, 132 g Cu, 10 g Ni, 4 g of Pb and 573 g of Zn was cycled in the home-grown feeds and manure together, on average.

Table 53. Internal flow (g ha<sup>-1</sup> a<sup>-1</sup>) of trace elements in total home-grown feeds used on the five dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the AROMIS-model.

Farm	Cd	Cr	Cu	Ni	Pb	Zn
6	0.23	0.79	87	5.4	1.2	285
7	0.20	0.55	56	1.8	1.1	256
8	0.21	1.93	92	4.5	1.4	368
9	0.16	0.93	100	5.5	1.4	367
10	0.17	2.28	80	3.6	1.5	296
<b>Mean</b>	<b>0.19</b>	<b>1.30</b>	<b>83</b>	<b>4.2</b>	<b>1.3</b>	<b>314</b>

Table 54. Internal flow ( $\text{g ha}^{-1} \text{a}^{-1}$ ) of trace elements in manure used on the dairy farms (6-10) themselves in Ostrobothnia in 2004 as estimated with the AROMIS-model.

Farm	Cd	Cr	Cu	Ni	Pb	Zn
6	0.12	2.13	35	6.1	0.91	170
7*	0.06	0.93	22	2.5	0.38	128
8*	0.06	1.07	28	2.9	0.43	200
9	0.24	4.30	79	7.2	1.07	416
10*	0.28	5.45	81	11	10.0	382
<b>Mean</b>	<b>0.16</b>	<b>2.76</b>	<b>49</b>	<b>6.0</b>	<b>2.57</b>	<b>259</b>

\* Part of total manure generated on the farm was exported.

An unidentified input of trace elements on the dairy farms was estimated in the model as follows: Unidentified input in livestock farming = (trace element flow via animal manure + trace element flow via animal products) minus (trace element flows via home-grown feeds, imported livestock feeds and feed additives, and, if necessary, other inputs, which end up in the manure and the animal products and would thus be counted twice). No unidentified input of trace elements occurred on the farms 6-8 (Table 55, Appendix 8: Fig. 12, 14 and 16), and only for Ni on the farm 9 (Appendix 8: Fig. 18). Instead, a major part of the total inputs of Ni and Pb and a minor part of the total inputs of Cd and Cr originated from unidentified sources on farm 10 (Appendix 8: Fig. 20). This study could not determine the unidentified sources. We assumed that the metal structures in the cow house might be one source. Also, a reason could be the inaccuracy in the data for the materials flown in and out of the farms.

Table 55. Unidentified trace element input (%) from the total inputs on the five dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the AROMIS-model.

Farm	Cd	Cr	Cu	Ni	Pb	Zn
6	0	0	0	0	0	0
7	0	0	0	0	0	0
8	0	0	0	0	0	0
9	0	0	0	20	0	0
10	18	3	0	60	60	0

### 3.7 Trace element balances

Detailed figures on the trace element inputs and outputs and balances of the RAKAS-model can be seen by elements in Appendix 7: Tables 1-10. The respective figures estimated by the AROMIS-model are described in Appendix 8: Figs. 1-20. Field balances of the five crop farms and five dairy farms estimated by the RAKAS-model are summarized in Table 56 and AROMIS-model in Table 57. Balances obtained by the RAKAS-model for the crop farms on the clay soils seemed to be quite similar to those obtained by the AROMIS-model. However, there were clear differences in the balances estimated for the dairy farms on the finesand soils between the models. The most probable reason for the differences might be the lack of real values for the trace element leaching and erosion in the Finnish agro-ecosystems. In most European countries, the balances of Cd, Cr, Cu, Ni, Pb and Zn estimated with the AROMIS-model were positive regardless of the farm type (Eckel et al. 2005). Both models showed that in most cases, the trace element inputs and accumulation in the soil are higher on the dairy farms than on the crop farms. That was observed also in earlier studies by Mäkelä-Kurtto and Sipola in the AROMIS-project (AROMIS 2005).

Table 56. Trace element field balances for the five crop farms (1-5) on clay soil in southwestern Finland and for the five dairy farms (6-10) on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Farm	RAKAS-balances, g ha <sup>-1</sup> a <sup>-1</sup>									
	Cd	Cr	Cu	Ni	Pb	Zn	Hg	V	As	Se
1*	0.003	(330)	-16	-12	-1.3	-32	0.037	(5986)	-1.2	4.4
2	0.079	-38	-23	-23	-3.2	-82	0.026	-49	-2.0	4.0
3	0.023	-14	-28	-8.5	1.6	-5	0.020	-23	-1.0	2.7
4	0.078	-29	-20	-15	2.2	-16	0.018	-41	1.5	4.0
5*	0.161	(474)	-13	-12	-3.7	(158)	0.034	(8479)	-1.4	5.0
<b>Mean</b>	<b>0.069</b>	<b>-27</b>	<b>-20</b>	<b>-14</b>	<b>-0.88</b>	<b>-34</b>	<b>0.027</b>	<b>-38</b>	<b>-0.82</b>	<b>4.0</b>
6*	0.212	(708)	26	6.5	2.1	219	0.053	(11998)	1.9	5.3
7	0.265	0.7	35	-2.9	-1.4	641	0.027	14	2.3	8.6
8	0.108	-14	-27	-2.6	1.8	110	-0.004	-41	-1.0	3.9
9	0.190	5.8	37	-4.1	3.7	335	0.034	4.8	-0.22	5.2
10	0.145	0.13	6.6	-4.0	2.6	365	0.027	3.1	1.0	5.8
<b>Mean</b>	<b>0.184</b>	<b>-1.9</b>	<b>16</b>	<b>-1.4</b>	<b>1.8</b>	<b>334</b>	<b>0.027</b>	<b>-4.8</b>	<b>0.80</b>	<b>5.8</b>

\* Steel slag was used on the farm; ( ) = not included in the mean

Table 57. Trace element field balances for the five crop farms (1-5) on clay soil in southwestern Finland and for the five dairy farms (6-10) on finesand soil in Ostrobothnia in 2004 as estimated with the AROMIS-model and their means by farm types in Finland and in Europe according to the AROMIS-project (Eckel et al. 2005).

Farm	Soil type	AROMIS-balances, g ha <sup>-1</sup> a <sup>-1</sup>					
		Cd	Cr	Cu	Ni	Pb	Zn
1*	RC	0.090	(363)	-18	4.0	-0.30	-11
2	RC	0.160	0.70	-20	-1.1	-2.8	-51
3	RC	0.210	1.5	-25	0.58	4.3	19
4	RC	0.220	3.6	-20	2.3	0.80	19
5*	RC	0.250	(514)	-7.6	8.5	-0.50	(178)
<b>Mean</b>	<b>RC</b>	<b>0.186</b>	<b>1.9</b>	<b>-18</b>	<b>2.9</b>	<b>0.3</b>	<b>-24</b>
EUR		3.3	98	372	78	81	745
6*	S	-0.660	(719)	6.1	13	-19	83
7	S	-0.490	6.8	14	0.6	-16	514
8	P	(0.190)	(3.6)	(-31)	(-6.8)	(-8)	(40)
9	S	-0.720	6.3	11	-1.7	-20	179
10	S	-0.690	0.10	-19	5.6	-12	245
<b>Mean</b>	<b>S</b>	<b>-0.640</b>	<b>4.4</b>	<b>3.0</b>	<b>4.3</b>	<b>-17</b>	<b>255</b>
EUR		2.2	54	232	12	35	769

\* Steel slag was used on the farm; ( ) = not included in the mean

The RAKAS-model also revealed the main in- and outflow routes for the trace elements studied (Table 58 and 59). On the crop farms in southwestern Finland, the major source of Cd, Pb, Hg, Cu, V and As was atmospheric deposition. The other elements, Zn, Ni, Cr and Se, were imported into the crop farms through the fertiliser products. On the dairy farms in Ostrobothnia, Cd, Pb and Hg mainly came into the farm from the atmosphere, Zn, Ni, V, As and Se from the fertiliser products and Cu and Cr from the commercial feeds. If steel slag was used on the farm, huge amounts of V and smaller amounts of Cr flowed into the farm via the slag (Table 37).

In the crop farms, the major outflow of Zn and Se occurred in the crop plants and the other elements, Cd, Pb, Cu, Ni, Cr, Hg, V and As, via erosion (Table 58). On the dairy farms, crop plants did not play an important role in the outflow of any element (Table 59). Instead, Zn and Se were flowing out from the dairy farms in the milk and meat. For Cd, leaching was the main route out of the dairy farms and for Pb, Cu, Ni, Cr, Hg, V and As the main route was erosion.

Field balances were mostly positive for Cd, Hg, Se and negative for Cr, Cu, Ni, Pb, Zn, V and As on the crop farms (Table 58). On the dairy farms, the balances were more or less positive for Cd, Cu, Pb, Zn, Hg, As and Se and negative for Cr, Ni, and V (Table 59). Regardless of farm type, the field balance for V

was highly positive, if steel slag was used on the farm (Table 58 and 59). Manure was a remarkable outflow route for Cu, Zn and Se, if manure was exported from the dairy farm.

Table 58. Main in- and outflow routes for trace elements on the five crop farms (1-5) on clay soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Crop farms	Trace elements
<b>Inflow route</b>	
Atmospheric deposition	Cd, Pb, Hg, Cu, V, As
Fertiliser products	Zn, Ni, Cr, Se
<b>Outflow route</b>	
Crops	Zn, Se
Leaching	-
Erosion	Cd, Pb, Cu, Ni, Cr, Hg, V, As
<b>Balance</b>	
Negative	Cr, Cu, Ni, Pb, Zn, V, As
Positive	Cd, Hg, Se

Table 59. Main in- and outflow routes for trace elements on the five dairy farms (6-10) on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Dairy farms	Trace elements
<b>Inflow route</b>	
Atmospheric deposition	Cd, Pb, Hg
Fertiliser products	Zn, Ni, V, As, Se
Commercial feeds	Cu, Cr
<b>Outflow route</b>	
Crops	-
Leaching	Cd
Erosion	Pb, Cu, Ni, Cr, Hg, V, As
Milk and meat	Zn, Se
Manure, if exported	Cu, Zn, Se
<b>Balance</b>	
Negative	Cr, Ni, V
Positive	Cd, Cu, Pb, Zn, Hg, As, Se

### 3.7.1 Cadmium

According to the balance calculations the major input of Cd to both farm types seemed to be atmospheric deposition (Table 60) although Cd emissions and bulk

depositions have decreased during the last decade. Cadmium inputs from fertiliser products in Finland were less than a tenth of the respective figures in Europe (Table 37). The phosphorus mine in Siilinjärvi, Finland, produces low-Cd raw phosphate for fertiliser manufacturing.

On the dairy farms, the mean input of Cd was to some extent higher from the commercial feeds than from fertiliser products (Table 60). The Decree of the Ministry of Agriculture and Forestry in Finland 2007/1 on harmful elements in feeds set a limit value for Cd in animal feeds that varied from 0.5 to 10 mg kg<sup>-1</sup> in 12% water content depending on the feed type. The limit value is the highest for feed phosphates. In these feeds, the maximum Cd content must not exceed 0.5 mg per 1% of P.

On the crop farms, located on the clay soil in southwestern Finland, the main output route of Cd was erosion and on the dairy farms located on finesand soil in Ostrobothnia was leaching (Table 60). As a whole, Cd was the only element studied here whose biggest route out of the farm was leaching. Due to the low Cd contents in the crop plants, Cd output was less important than erosion or leaching. The field mass balance of Cd was positive in both farm types (Table 60). Also, comparisons of the Cd contents in top- and subsoil of the same crop and dairy farms indicated that Cd has clearly enriched in the topsoil regardless of the farm type (Hatakka et al. 2007). However, enrichment was higher in the crop farms in southwestern Finland due to the higher Cd deposition.

Table 60. Mean input and output routes of cadmium (Cd) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Cd	Crop farms
Input	Atmospheric deposition > fertiliser products
Output	Erosion > leaching > crop plants
Balance	Positive
Cd	Dairy farms
Input	Atmospheric deposition > fertiliser products > commercial feeds
Output	Leaching > erosion > manure > crop plants
Balance	Positive

The results obtained by the AROMIS-model showed that the main input of Cd was atmospheric deposition (Appendix 7, Table 1, and Appendix 8), but the main output occurred via leaching (Appendix 8) because this model did not take into account the erosion, on average. Also, this model showed a positive Cd balance for both the crop and dairy farms.

### 3.7.2 Chromium

Chromium mainly came into the crop farms in the fertiliser products and into the dairy farms in the commercial feeds (Table 61). In Finland, there is no limit value for Cr in the animal feeds, but there is a limit value for Cr in the fertiliser products of 300 mg kg<sup>-1</sup> dm (the Decree of the Ministry of Agriculture and Forestry 2007/12 on the fertiliser products). In both farm types, erosion was the most important way for Cr to move out of the farms. The field balance of Cr was negative on the crop farms, on average, but slightly positive on the dairy farms. These results are well in line with the observations made by Hatakka et al (2007). They showed that Cr was depleted in the topsoil of the crop farms but enriched in the topsoil of the dairy farms.

According to the AROMIS-model, the field balances of Cr were positive for all the farms studied. However, both the RAKAS-model and AROMIS-model indicated that Cr inputs and outputs were rather well balanced meaning that the balance values were only a little lower or higher than zero (Appendix 7: Table 2). However, if steel slag was used for liming, the Cr balance was highly positive regardless of the farm type (Appendix 7: Table 2, Appendix 8: Figs. 1-12). Steel slag contained Cr 600 mg kg<sup>-1</sup> dm (Table 13).

Table 61. Mean input and output routes of chromium (Cr) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Cr	Crop farms
Input	Fertiliser products > atmospheric deposition
Output	Erosion > leaching > crop plants
Balance	Negative
Cr	Dairy farms
Input	Commercial feeds > fertiliser products > atmospheric deposition
Output	Erosion > leaching > milk and meat > crop plants
Balance	Mostly positive

### 3.7.3 Copper

Atmospheric deposition was the main route for Cu on those crop farms that did not use steel slag (Table 62). On the dairy farms, the commercial feeds were the most important source of Cu into the farm (Table 62). Copper flowed out of the farms, including the dairy farms, through erosion if manure was not exported (Table 62). Over the past twenty years, the total sale of Cu in field fertilisers in Finland has diminished from 693 tonnes in the fertiliser year 1983/1984 to 43 tonnes in the fertiliser year 2003/2004 (Table 63). On the crop farms, total Cu inputs were lower (Table 36) than the Cu outputs (Table 45). That was a reason

for a negative Cu balance. Also, according to the AROMIS-model, the Cu balance was clearly negative in all the crop farms and mostly positive on the dairy farms (Appendix 7: Table 3, Appendix 8: Figs. 1-12). Also, highly negative Cu balances occurred on the crop farms on clay soil in Norway, France and Hungary and on sandy soil in Denmark (AROMIS 2005). The copper balance was mostly positive on the dairy farms (Table 60, Appendix 7: Table 3). However, the mean fertility status of Cu in the fields on both farm types was of the Class 4 (Satisfactory) (Hatakka et al. 2007). According to Hatakka et al. (2007), Cu was depleted in the soils of the crop farms and enriched in the soils of dairy farms.

Table 62. Mean input and output routes of copper (Cu) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Cu	Crop farms
Input	Atmospheric deposition > fertiliser products
Output	Erosion > crop plants > leaching
Balance	Negative
Cu	Dairy farms
Input	Commercial feeds > fertiliser products > atmospheric deposition
Output	Manure > erosion > leaching > milk and meat > crop plants
Balance	Positive

Table 63. Total annual sales of copper (Cu) and zinc (Zn) in field fertilisers in Finland (Kemira Agro 1999). \* Kemira GrowHow (2007b).

Fertiliser year	Total sale of Cu and Zn in field fertilisers, in tonnes		
	Copper	Zinc	
1983/1984	660	263	
1984/1985	693	322	
1985/1986	657	382	
1986/1987	595	381	
1987/1988	524	417	
1988/1989	589	414	
1989/1990	579	354	
1990/1991	323	288	
1991/1992	221	238	
1992/1993	185	268	
1993/1994	180	163	
1994/1995	183	148	
1995/1996	121	168	
1996/1997	79	142	
1997/1998	64	201	
1998/1999	57	194	
1999/2000*	36	204	continues

2000/2001*	59	192
2001/2002*	44	215
2002/2003*	50	252
2003/2004*	43	249

### 3.7.4 Nickel

In the case of Ni, a major route into the farms was fertiliser products and a major route out of the farms was erosion on both farms, on average (Table 64). The Ni balance was remarkably more negative on the crop farms than on the dairy farms (Table 64). Comparisons of the Ni contents in various soil layers (Hatakka et al. 2007) indicated a clear Ni depletion in the topsoil of crop farms. Instead, on the dairy farms, the Ni contents were at the same level in these horizons. The Ni balances shown by the AROMIS-model were most often slightly positive, but near zero (Appendix 7: Table 4). According to the AROMIS-model, the main Ni flow out of the farms was leaching (Appendix 8: Figs. 1-12).

Table 64. Mean input and output routes of nickel (Ni) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Ni	Crop farms
Input	Fertiliser products > atmospheric deposition
Output	Erosion > leaching > crop plants
Balance	Negative
Ni	Dairy farms
Input	Fertiliser products > commercial feeds = atmospheric deposition
Output	Erosion > leaching > manure > crop plants > milk and meat
Balance	Negative

### 3.7.5 Lead

Although lead emissions from traffic into the atmosphere have virtually ceased since 1994 (Mäkelä 1996), when unleaded gasoline became available for all motor vehicles, atmospheric deposition was further the main source for Pb on both farms (Table 65). Erosion was the major route for Pb out of the crop and dairy farms (Table 65). The field balances for Pb were mostly negative on the crop farms and positive on the dairy farms, but close to zero on both farms (Table 65). Differences in the Pb balance were not large between the RAKAS- and AROMIS-model (Appendix 7: Table 5) on crop farms, but clearly greater on the dairy farms. Leaching was the most distinct route out of the dairy farms in the AROMIS-model (Appendix 8: Figs. 1-12). In a long-term, Pb has noticeably enriched in the topsoil of both farm types (Hatakka et al. 2007).

Table 65. Mean input and output routes of lead (Pb) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Pb	Crop farms
Input	Atmospheric deposition > fertiliser products
Output	Erosion > leaching > crop plants
Balance	Close to zero
Pb	Dairy farms
Input	Atmospheric deposition > fertiliser products > commercial feeds
Output	Erosion > manure > leaching > crop plants > milk and meat
Balance	Close to zero

### 3.7.6 Zinc

On both farm types, fertiliser products were the biggest source of Zn (Table 64). The main route out from the farms was crop plants on the crop farms and milk and meat (or erosion) on the dairy farms (Table 66). On the crop farms, the Zn balance was mostly negative but clearly positive on each of the dairy farms. As to the Zn status, the fertility class of Zn on the crop farms was 2 (Rather poor) and dairy farms 4 (Satisfactory) (Hatakka et al. 2007). Hatakka et al. (2007) reported that Zn had more enriched in the topsoil of the crop farms than in the topsoil of dairy farms. In Finland, the annual sale of Zn in the field fertilisers (Table 63) has varied over the last 20 years and was highest (about 400 tonnes) at the end of 1980's and lowest (about 150 tonnes) in the middle of 1990's. In the 2000's, the annual sale of Zn has been between 200-250 tonnes. The Zn balance indicated by the AROMIS-model was not as negative on the crop farms and not as positive on the dairy farms as those indicated by the RAKAS-model (Appendix 7: Table 6, Appendix 8: Figs. 1-12). Clearly negative Zn balances were estimated for crop farms on clay soil in Norway and France and on sandy soil in Denmark (AROMIS 2005).

Table 66. Mean input and output routes of zinc (Zn) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Zn	Crop farms
Input	Fertiliser products > atmospheric deposition
Output	Crop plants > erosion > leaching
Balance	Negative
Zn	Dairy farms
Input	Fertiliser products > commercial feeds > atmospheric deposition
Output	Milk and meat = erosion > leaching > crop plants
Balance	Positive

### 3.7.7 Mercury

The primary source of Hg on all the farms was atmospheric deposition and the primary route out of the farms was erosion (Table 67). The field balances of Hg were positive on both farm types, on average (Table 67). Since 1992, the use of Hg containing pesticides in agriculture has been banned, and Hg inputs into the cultivated soil have been relatively low in Finland. Clear enrichment of Hg in the topsoil of both crop and dairy farms was observed by Hatakka et al. (2007).

Table 67. Mean input and output routes of Hg on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Hg	Crop farms
Input	Atmospheric deposition > fertiliser products
Output	Erosion > leaching > crop plants
Balance	Positive
Hg	Dairy farms
Input	Atmospheric deposition > fertiliser products > commercial feeds
Output	Erosion > leaching > milk and meat > crop plants
Balance	Positive

### 3.7.8 Vanadium

In southwestern Finland, the main source of V into the crop farms was mostly atmospheric deposition (Table 68). Instead, In Ostrobothnia, however, the fertiliser products and commercial feeds contributed about equal amounts of V to the dairy farms (Table 68). On both farm types, erosion was the major route for V out of the farms. Also, leaching of V was a pronounced output route. The crop plants, milk and meat had almost no importance to the V outflow from the farms (Table 68). Generally, the V balance was negative on the crop farms and slightly positive on the dairy farms (Appendix 7: Table 8). Hatakka et al. (2007) found clear enrichment of V in the topsoil of the dairy farms but not in the crop farms. If steel slag was used on the farm, the V balance was highly positive and the annual accumulation or enrichment of V in the soil was at least 1000 times that on the farm that did not use the steel slag (Appendix 7: Table 8).

Table 68. Mean input and output routes of vanadium (V) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

V	Crop farms
Input	Steel slag > atmospheric deposition = fertiliser products
Output	Erosion > leaching > crop plants
Balance	Negative
V	Dairy farms
Input	Steel slag > fertiliser products = commercial feeds > atmospheric deposition
Output	Erosion > leaching > manure > crop plants > milk and meat
Balance	Positive

### 3.7.9 Arsenic

In southwestern Finland, atmospheric deposition and fertiliser products were equal sources of As. The main source in Ostrobothnia was fertiliser products (Table 69, Appendix 7: Table 9). In all cases, As was mainly flowing out of the farm via erosion (Table 69, Appendix 7: Table 9). The balance was negative on the crop farms and slightly positive on the dairy farms, on average. Arsenic showed depletion in the topsoil on the crop farms but no difference in the contents between the top- and subsoil on the dairy farms (Hatakka et al. 2007).

Table 69. Mean input and output routes of arsenic (As) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

As	Crop farms
Input	Atmospheric deposition = fertiliser products
Output	Erosion > leaching > crop plants
Balance	Negative
As	Dairy farms
Input	Fertiliser products > atmospheric deposition > commercial feeds
Output	Erosion > leaching > manure > milk and meat > crop plants
Balance	Positive

### 3.7.10 Selenium

A major source of selenium on both farm types was the fertiliser products (Table 68). Mineral fertilisers to be used on animal farms or farms receiving manure from another farm are allowed to contain more Se than the fertilisers to be used in other types of farms (MMMa 12/2007). This might be one reason for the higher Se in-

puts on the dairy farms than on the crop farms. The main output of Se from the crop farms was through crop plants and on the dairy farms, through milk and meat and erosion. If manure was exported, Se was noticeably flowing out of the farm. In both farm types, the field balance for Se was highly positive (Table 70). The annual enrichment of Se in the soil was 4 g on the crop farms and 5.8 g ha<sup>-1</sup> on the dairy farms, on average. If Finnish cultivated soil contains Se 0.2 mg kg<sup>-1</sup> that means about 400 g per ha in the plough layer (20 cm) and if 4 g of Se will be annually accumulated per ha, it will take no more than one hundred years to double the initial Se content in the soil.

According to the calculations made by Yli-Halla (2005), the cumulative Se application was 37 g ha<sup>-1</sup> from 1992 to 2004, on average, and the highest application took place in intensive grassland cultivation. According to soil analyses, the Se content of a 23-cm deep plough layer was on average 23 g ha<sup>-1</sup> higher in 2004 than in 1992, but the difference was not statistically significant obviously due to the heterogeneity of the sampled fields. However, these results give support to the findings made in the present study. In addition, Se was distinctly enriched in the topsoil of both farm types, but clearly more on the dairy farms than on the crop farms (Hatakka et al. 2007).

To improve the quality of Finnish foods and to increase the Se intake of the Finnish population, an official decision was made in 1984 to supplement mineral fertilisers with Se as sodium selenate. In 1983, the Ministry of Agriculture and Forestry appointed a selenium working group and gave it the task to organize a national Se monitoring programme. The follow-up was initiated in 1984 and the effects of the Se content of fertilisers on basic foods, human serum and Se intake have been monitored, but less on soils. Since 1984, the supplementation level has been varied depending on the trends measured in the Se content of foods, feeds and human serum, and Se intake and the use of fertilisers. A large amount of data has been collected and published (Euroala et al. 2003). In the future, the soil Se content in arable land should be regularly monitored either in the national Se monitoring programme or in the national soil monitoring programme.

Table 70. Mean input and output routes of selenium (Se) on the five crop farms on clay soil in southwestern Finland and on the five dairy farms on finesand soil in Ostrobothnia in 2004 as estimated with the RAKAS-model.

Se	Crop farms
Input	Fertiliser products > atmospheric deposition
Output	Crop plants > erosion > leaching
Balance	Positive
Se	Dairy farms
Input	Fertiliser products > commercial feeds > atmospheric deposition
Output	Manure > milk and meat = erosion > crop plants > leaching
Balance	Positive

## 4 Conclusions

Although efficient measures to reduce the emissions of Cd and Hg into the air have been made in Finland over the long term, the major part of these heavy metals flowing into the farms still originates from the atmosphere.

The main inflow route of Se is fertiliser products. Compared to the low Se content in the soil in Finland, the accumulation rate of Se shown by the balance calculations seems to be so high that the initial Se content in the soil will be doubled in one hundred years.

Since the balance calculations showed a clear depletion trend for Cu and Zn in the soil on the crop farms, attention should be paid to the sufficiency of these micronutrients in the plant production. Instead, excessive amounts of these elements are flowing into the dairy farms. Hence, transport of manure generated on the dairy farms to the crop farms seems to be a good way to reduce excessive Cu and Zn loading on the dairy farms and at the same time, to balance the loadings of these micro-nutrients between the farms.

In general, inputs and outputs of As, Ni, Cr, Pb and V are rather well balanced on both the crop and dairy farms. However, if steel slag was used on the farm, accumulation of V into the soil was 400 times greater than on the farm that did not use steel slag. Currently, there is no limit value for V in the fertiliser products. Since mobility and bioavailability of V in the Finnish agro-ecosystems is not sufficiently known, the biogeochemical behaviour of V originating from steel slag in the soil should be studied.

In addition, to improve the reliability of the balance calculations, national measurements will be needed to determine leaching and erosion values for the trace elements studied, particularly for V, if steel slags are acceptable for use in agriculture.

## 5 Summary

This study was part of a three-year (2004-2007) project entitled “Assessment and reduction of heavy metal inputs into Finnish agro-ecosystems”, which was funded by the Ministry of Agriculture and Forestry in Finland. The aims of the project were to clarify: 1) *aqua regia* extractable trace elements in Finnish cultivated soils with the international standard method at a national level; 2) *aqua regia* and AAAC-EDTA extractable trace elements in the top- and subsoil of Finnish arable land on selected crop and dairy farms; and 3) field mass balances of trace elements on the same selected crop and dairy farms at the farm level.

The main aim of this study was to estimate field balances of trace elements at the farm level on selected crop and dairy farms in Finland in 2004. The trace elements studied were arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), nickel (Ni), selenium (Se), vanadium (V) and zinc (Zn). Five crop farms were selected from southwestern Finland typical to arable farming and five dairy farms from Ostrobothnia typical to milk production. For balance estimations, data on the amounts of agricultural production resources imported to and products exported from the farms were collected. Samples from fertiliser products, commercial feeds, crop plants, milk, meat and manure and also from top- and subsoil were collected and analysed for the ten trace elements. The dominant soil type on the crop farms was clay and on the dairy farms finesand.

Balance calculations were made with two models: the RAKAS-model for all the elements to be studied and the European AROMIS-model for Cd, Pb, Cu, Cr, Ni and Zn. The main differences between the models were in the leaching and erosion values. The RAKAS-model used the leaching figures measured in the Nordic countries and in the AROMIS-model, leaching figures measured in other parts of Europe. Erosion was taken into account in the RAKAS-model, but not in the AROMIS-model. The AROMIS-model was able to estimate internal flows of the trace elements in home-grown feeds and manure and also the inputs of trace elements from unidentified sources on the dairy farms.

Field balance estimated at the farm level showed that on the crop farms on clay soil in southwestern Finland in 2004, the main sources for Cd, Pb, Hg, Cu, V and As was atmospheric deposition and for Zn, Ni, Cr and Se fertiliser products. Based on the RAKAS-model, the main outputs of Zn and Se were via crop plants and other trace elements via erosion. Leaching was of negligible importance on the crop farms. Mean field balances were slightly positive for Cd, Hg and highly positive for Se resulting in enrichment of these elements in the soil. Instead, balances were mainly negative for the other elements, even for Cu and Zn, resulting in a depletion of these elements in the soil.

On the dairy farms on finesand soil in Ostrobothnia in 2004, the main source for Cd, Pb and Hg was atmospheric deposition; Zn, Ni, V, As and Se fertiliser products; and Cu and Cr commercial feeds. Based on the RAKAS-model, the main output for Cd was via leaching; Pb, Cu, Ni, Cr, Hg, V and As via erosion; and Zn and Se via milk and meat. Crop plants had almost no importance in the trace element outputs on the dairy farms. Mean field balances were negative for Cr, Ni, Pb and V resulting in depletion and positive for the other trace elements resulting in enrichment of these elements in the soil. Field balances obtained by the AROMIS-model indicated that manure export clearly increased the trace element output from the dairy farm and reduced the trace element load to the soil on the farm itself.

Trace element outputs in milk and meat were low because the trace element contents in these products were also very low and clearly lower than the respective contents in the crop plants. Thus, the trace element balances were mostly more positive on the dairy farms than on the crop farms. Field balances of the harmful heavy metals, Cd and Hg, at the farm level were mostly slightly positive. This means that enrichment of these metals into the soil will slowly continue in the coming years in both production sectors. The balances of As, Cr, Ni, Pb and V were quite well balanced for both farming types. However, if steel slag was used for liming, huge amounts of V and lesser amounts of Cr were added into the soil, leading to very positive balances of V and Cr. Selenium balances in the crop and dairy farms were positive as well. If the loading rate of Se into the soil will be the same as measured here, it will take about one hundred years to double the current selenium content in soil. The Cu and Zn balances were positive on the dairy farms, but negative on the crop farms leading gradually to the depletion of these elements in the crop farm's soil.

Research results obtained with the RAKAS-model were compared to those obtained with the European AROMIS-model. In the AROMIS-model, leaching seemed to be the major route for many trace elements out of the farms. National measurements on the trace element outputs via leaching and erosion are needed to improve the accuracy of the balance calculations.

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# 8 Appendixes

## Appendix 1

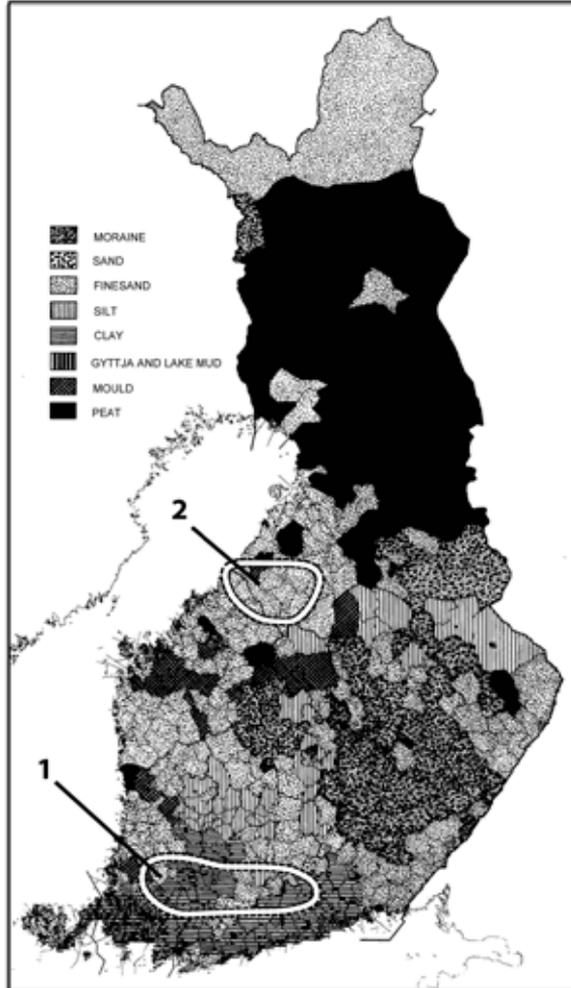


Figure 1. Dominant soil types in topsoil (plough layer) of arable land in Finland after Kurki (1972). A circle (1) in southwestern Finland indicates the region where the crop farms are located and a circle (2) in Ostrobothnia indicates the region where the dairy farms are located.

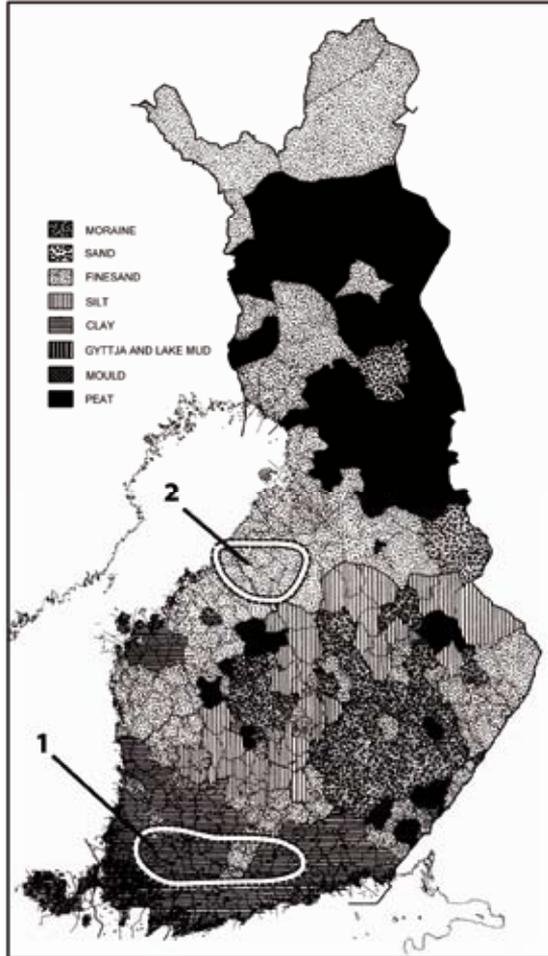


Figure 2. Dominant soil types in subsoil of arable land in Finland after Kurki (1972). A circle (1) in southwestern Finland indicates the region where the crop farms are located and a circle (2) in Ostrobothnia indicates the region where the dairy farms are located.

## Appendix 2

AROMIS - Excel calculation tools for the calculation of heavy metal balances for model farms

**Version rev04 (end of Sept. 02):** [Another correction in the formula for the diffuse input calculation was made. Additionally, a checking routine was set up for the leaching and irrigation routine.]

These Excel-folders provide a calculation tool to generate a heavy metal balance for model farms. Separate folders were developed for animal and for plant production farms. The revised sheets are called **Bal\_animprod\_rev04.xls** and **Bal\_plantprod\_rev04.xls**. (The previous sheets were called Balance\_animprod.xls and Balance\_plantprod.xls, respectively). To facilitate the distinction between the two models the sheet titles within the folders are always provided with the suffix “\_ap” or “\_pp”.

Only the fields highlighted in blue can be filled. The fields with fixed contents are blocked and cannot be changed by the user. Yet, if for any reason changes in blocked fields are necessary, please contact Paul Römken or KTBL. The contents of those fields, which enter into subsequent calculations, are automatically transmitted to the consequent fields in the following sheets.

Sheet “assumpt”

The basic assumptions for the model farm have to be defined in the first sheet “assumpt\_ap” and “assumpt\_pp”, respectively.

### 1. General information

- Country (automatic transmission to the following sheets)
- Type of farm: animal or plant production farm
- Real/average: existing farm or an average farm based on literature values
- Surface area: filled automatically by summing up the numbers of the crop information section

### 2. Crop information

- Metals in crops measured: yes/no/in part (This can later be indicated separately for each product in the “in”- and “out”-sheets; see in these chapters)
- Home-grown feed and commercial crops

For animal production a differentiation is made between the area used for the cultivation of home-grown feeds and that used for commercial

crops. The yearly produced amount of the different crops is automatically calculated from the area, the fresh matter yield per ha and the water content, i.e. these fields have to be filled to enable correct calculation. The result is then - as well as the culture names - transferred to the corresponding fields in the sheets “out” (for commercial crops) and “intern” (for home-grown feeds in animal farms).

### **3. Animal farm model: Animal manure and other organic fertilisers information**

- Type and number of animals
- Manure applied (on the farm itself): yes/no and type (e.g. dairy slurry)  
The total amount of manure produced and the percentage used on the farm itself have to be indicated. The percentage exported and the corresponding amounts are automatically calculated and transferred to the sheets “internal” (used on the farm itself) and “out” (exported manure) as well as the manure type.
- Other organic fertilisers applied: yes/no and indication of amounts of sewage sludge, compost or other organic fertiliser used (automatic transmission to “in”-sheet)

#### **Plant farm: Organic fertilisers information**

- Animal manure applied and other organic fertilisers, respectively: the type and amount of fertiliser are transferred to the “in”-sheet

### **4. Irrigation**

- Fields irrigated: Y/N  
If yes, the irrigated area and the amount of water per ha have to be indicated, because an automatic calculation of the irrigation input is made in the “in”-sheet assuming irrigation with local groundwater (i.e. the metal concentration equals that of the leaching, which is either calculated or indicated in the leaching section). Because of possible errors during filling in the leaching section a checking routine was set up, which also affects irrigation (see below).

### **5. Leaching**

- Precipitation and Evapo(transpi)ration have to be indicated; the leaching is then automatically calculated; if precipitation minus evapo(transpi)ration is negative (e.g. in southern Europe), leaching is automatically set to zero. This does not affect irrigation calculations - in this case default values are used.
- Measured data: Y/N  
If yes, the values have to be indicated as they are then used for the cal-

ulation of the metal output with leaching in sheet “leachingcalc” (and for the input with irrigation, see above).

If no, the leaching metal concentration is replaced by default values depending on the soil texture indicated in the general soil properties section below. For the default values see sheet “leachingcalc” lines 27-30.

Automatic checking routine: If the user indicates, that measured values are available without filling them in below, the model automatically displays a warning in the input (column irrigation), output (column leaching) and balance (lines irrigation and leaching) sheets (**CHECK!!**). If instead of y/n nothing or something else is typed in, there is a warning only in the leaching cells, irrigation is then calculated based on the default values.

## 6. General soil properties

- Soil type: For the calculation of irrigation and leaching concentration the indication of the soil texture is essential, if no measured values are available for leaching (see sheet “leachingcalc”). If nothing or others than the indicated abbreviations are used here, “FALSE” is displayed in the leaching cells in the other sections. Irrigation is then again based on default values.
- Other information requested in this section is not used for further calculation. The soil metal contents might be useful in future, if we decide to calculate the time left until threshold values for soil are reached.

## 7. Other relevant information

This information helps to further characterise the site.

### Sheet “in”

On this sheet all heavy metal inputs into the farm are listed and grouped together in the below listed categories. Column titles not highlighted in blue are fixed and in the case of organic fertilisers automatically filled with the indications in the “assumpt”-sheet.

The yearly heavy metal load is calculated from the yearly used amount (in tons of dry matter per year) and the heavy metal content (in mg per kg dry matter). These two figures have to be entered by the user, the calculation routine is already set. Within the different categories the flow is in a first step calculated separately for each of the listed media. The overall load (e.g. the different mineral fertilisers) is then automatically summed up and the result is transmitted to the table in the “balance”-sheet.

If you are using own values for the heavy metal concentrations please indicate

in the tick boxes below the tables (each medium and metal can be chosen separately). You'll find these boxes in the sheets "out" and "intern" as well.

### Categories

- atmospheric deposition (here the heavy metal flow has to be filled in directly, as no calculation possible)
- irrigation (if no irrigation takes place, values are automatically zero; otherwise the amount is calculated from the indications in the "assumpt"-sheet and the metal concentration is taken from sheet "leachingcalc"; see also comment on the "assumpt"-sheet); see also the leaching section in the "assumpt"-sheet for the automatic checking routine (**CHECK!!** is displayed in case of errors there)
- mineral fertilisers
- organic fertilisers - including imported animal manure in the plant production model. In the animal production model the manure used on the farm itself is seen as an internal flow (see "intern"-sheet for more detailed comment).
- imported livestock feeds (only in the animal production model)
- feed additives (only in the animal production model)
- diffuse input by animal farming

Blank columns ("others") have been inserted within the different categories and for additional inputs, so that the sheet can be adopted to the particularities of each country. For imported livestock feeds and feed additives the column titles are not fixed by the authors, as the media of these groups change depending on the animal.

### Unidentified input animal farming

To calculate the diffuse heavy metal input by animal farming the following calculation routine was set up: **diffuse input by animal farming = (heavy metal flow via animal manure + heavy metal flow via animal products) minus (heavy metal flows via homegrown feeds, imported livestock feeds and feed additives - and, if necessary, other inputs, which end up in the manure and the animal products and would unless be counted twice).**

As the additional inputs vary depending on the animal etc. this calculation routine is not blocked, so that the user can add, if necessary, manually a further medium to the formulae in the column "diffuse input by animal farming" (highlighted in bright green). In the example sheet this is the case for hoof disinfection. The remaining diffuse input from animal farming might include sources such as abrasion of stable interior, for which very little data exist.

### Appendix 3

Table 1. Means of *aqua regia* extractable trace elements in top- (1) and subsoil (2) on the five crop farms (1-5) in southwestern Finland in 2004.

Farm Nr.	Soil		Mean concentration of the element, mg kg <sup>-1</sup> air dried soil									
	depth	n	As	Cd	Cr	Cu	Pb	Hg	Ni	Se	V	Zn
1*	1	4	7.4	0.19	66	24	20	0.039	29	-*	91	108
	2		11	0.12	79	28	19	0.031	34	0.26	86	104
2	1	6	5.2	0.20	79	38	21	0.025	41	-*	93	136
	2		6.3	0.11	98	41	18	0.021	46	0.22	98	118
3	1	3	3.1	0.28	29	16	13	0.044	12	0.35	47	67
	2		4.8	0.15	45	21	14	0.032	21	0.25	55	70
4	1	4	5.1	0.29	65	27	14	0.075	31	0.22	87	131
	2		6.0	0.16	84	34	13	0.026	36	0.33	91	118
5*	1	6	7.6	0.22	80	42	25	0.058	39	0.35	110	110
	2		8.0	0.07	96	50	23	0.032	46	0.21	101	107

\* Over 50% of the values were below the detection limit, 0.2 mg kg<sup>-1</sup> and the mean value was not computed.

Table 2. Means of *aqua regia* extractable trace elements in top- (1) and subsoil (2) on the five dairy farms (6-10) in Ostrobothnia in 2004.

Farm Nr.	Soil		Mean concentration of the element, mg kg <sup>-1</sup> air dried soil									
	depth	n	As	Cd	Cr	Cu	Pb	Hg	Ni	Se	V	Zn
6*	1	5	5.0	0.15	34	19	8.6	0.035	16	0.29	50	63
	2		4.1	0.06	37	17	7.5	0.007	17	0.12	47	49
7	1	3	2.2	0.10	21	8.3	7.5	0.034	7.4	0.21	25	33
	2		2.2	0.03	24	8.2	5.0	0.008	10	0.10	29	28
8	1	5	4.6	0.16	40	22	10	0.049	16	0.39	85	56
	2		5.4	0.11	33	16	7.0	0.039	16	0.27	47	43
9	1	3	1.7	0.05	13	5.9	3.8	0.015	5.5	-*	17	23
	2		1.8	0.03	14	5.4	3.7	0.011	5.4	0.07	17	18
10	1	5	1.3	0.07	11	7.8	3.3	0.012	4.8	-*	20	24
	2		1.3	0.05	8.6	4.5	2.6	0.008	3.4	0.03	12	18

\* Over 50% of the values were below the detection limit, 0.2 mg kg<sup>-1</sup> and the mean value was not computed.

Table 3. Mean ratios of *aqua regia* extractable trace elements in top- and subsoil on the five crop farms (1-5) in southwestern Finland and on the five dairy farms (6-10) in Ostrobothnia in 2004. (n = number of fields studied).

Farm Nr.	n	Ratios (topsoil/subsoil)									
		As	Cd	Cr	Cu	Pb	Hg	Ni	Se	V	Zn
1*	4	0.70	1.56	0.84	0.86	1.09	1.26	0.88	0.70	1.06	1.03
2	6	0.83	1.80	0.80	0.90	1.13	1.38	0.90	0.72	0.95	1.16
3	3	0.65	1.87	0.64	0.74	0.92	1.38	0.60	1.41	0.85	0.95
4	4	0.85	1.78	0.78	0.80	1.13	2.88	0.87	0.67	0.95	1.12
5*	6	0.95	3.10	0.84	0.85	1.11	1.81	0.84	1.68	1.10	1.03
<b>Mean</b>	<b>23</b>	<b>0.80</b>	<b>2.02</b>	<b>0.78</b>	<b>0.83</b>	<b>1.08</b>	<b>1.74</b>	<b>0.82</b>	<b>1.04</b>	<b>0.98</b>	<b>1.06</b>
6*	5	1.24	2.50	0.91	1.15	1.16	5.00	0.91	2.42	1.06	1.29
7	3	1.00	3.33	0.88	1.02	1.51	4.25	0.72	2.10	0.87	1.15
8	5	0.85	1.45	1.19	1.36	1.37	1.26	1.03	1.44	1.81	1.29
9	3	0.94	1.67	0.92	1.10	1.02	1.36	1.03	1.43	0.97	1.25
10	5	1.03	1.40	1.24	1.71	1.27	1.50	1.42	4.53	1.71	1.33
<b>Mean</b>	<b>21</b>	<b>1.01</b>	<b>2.07</b>	<b>1.03</b>	<b>1.27</b>	<b>1.27</b>	<b>2.67</b>	<b>1.02</b>	<b>2.38</b>	<b>1.28</b>	<b>1.26</b>

## Appendix 4

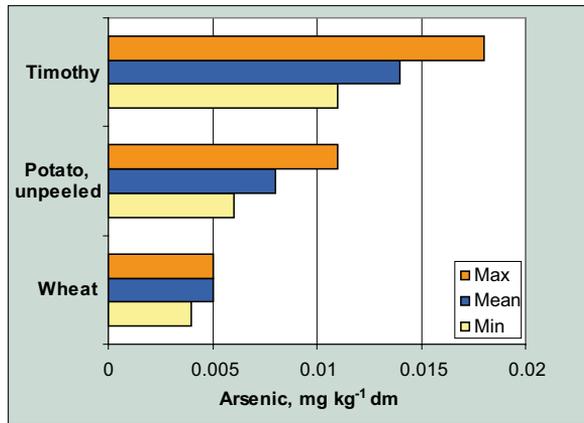


Figure 1. Arsenic contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurto et al. 2006).

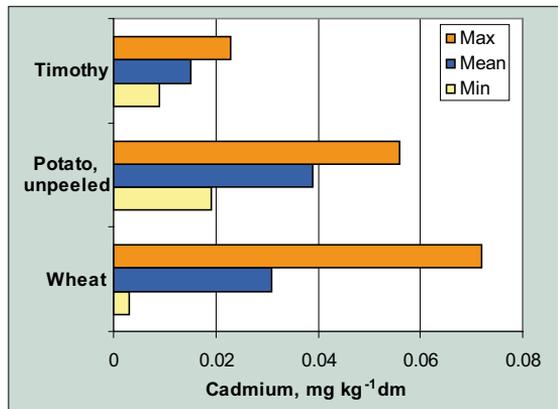


Figure 2. Cadmium contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurto et al. 2006).

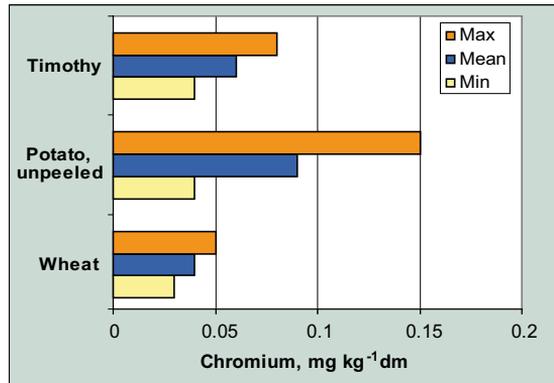


Figure 3. Chromium contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurtto et al. 2006).

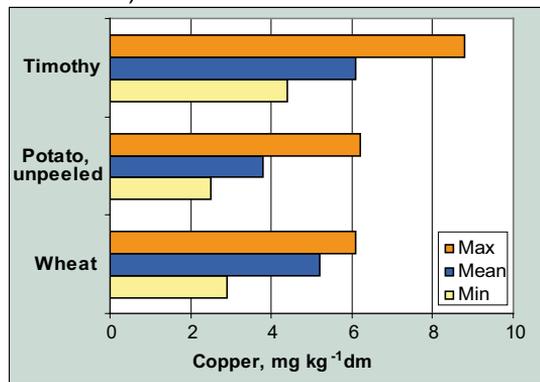


Figure 4. Copper contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurtto et al. 2006).

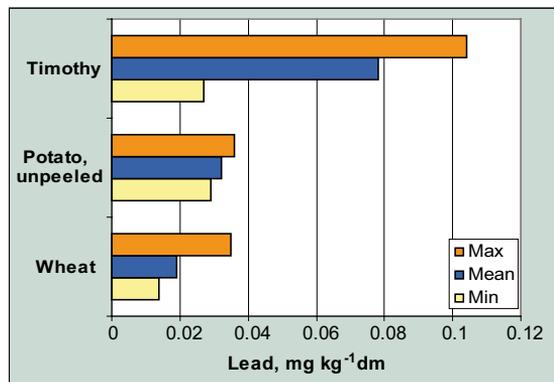


Figure 5. Lead contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurtto et al. 2006).

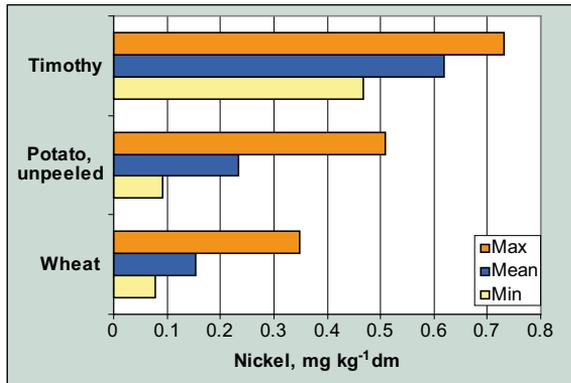


Figure 6. Nickel contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurto et al. 2006).

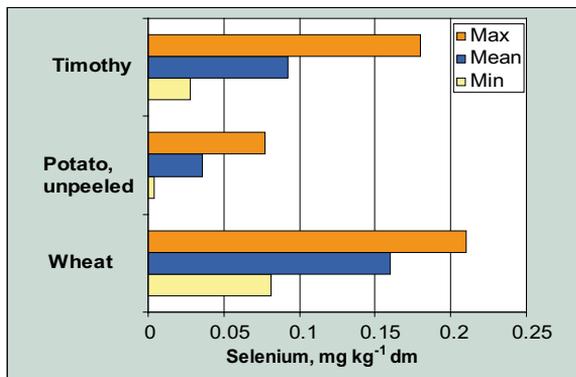


Figure 7. Selenium contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurto et al. 2006).

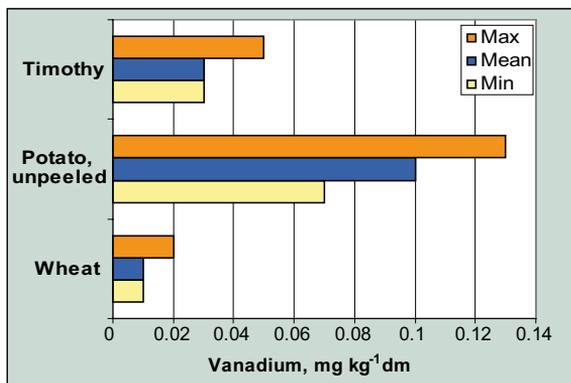


Figure 8. Vanadium contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurto et al. 2006).

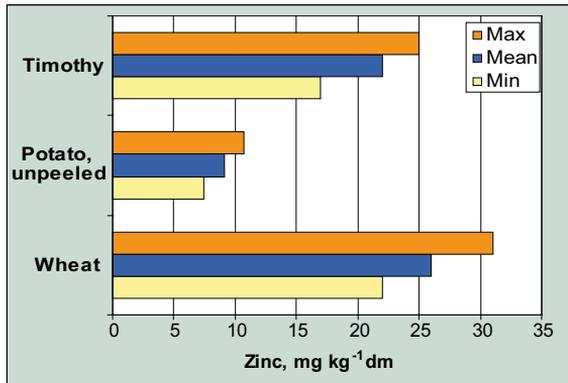


Figure 9. Zinc contents (mg kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurtto et al. 2006).

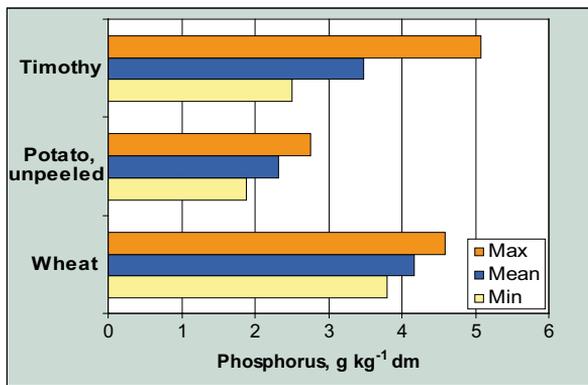


Figure 10. Phosphorus contents (g kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurtto et al. 2006).

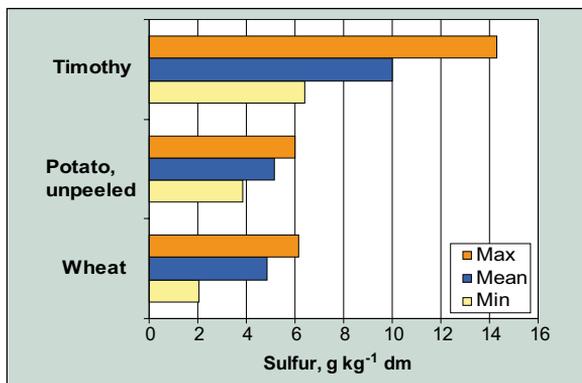


Figure 11. Sulphur contents (g kg<sup>-1</sup> dm) in timothy grass, the 2<sup>nd</sup> cut, potato tubers, unpeeled, and wheat grains on the farms in the Tampere region in 2005 (Mäkelä-Kurtto et al. 2006).

## Appendix 5

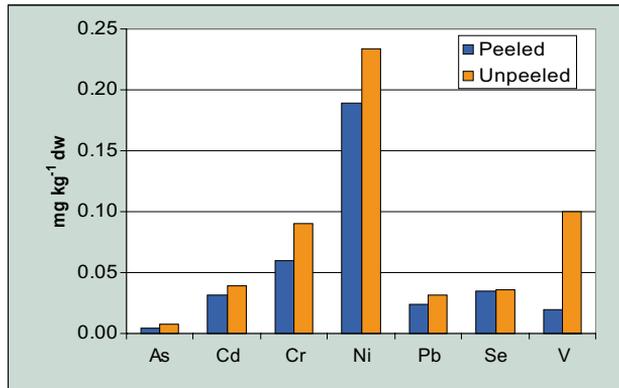


Figure 1. Arsenic (As), cadmium (Cd), chromium (Cr), nickel (Ni), lead (Pb), selenium (Se) and vanadium (V) contents (mg kg<sup>-1</sup> dw) in potato tubers peeled (column left) and unpeeled (column right). Samples collected from the farms in the Tampere region in 2005 (Mäkelä-Kurto et. al 2006).

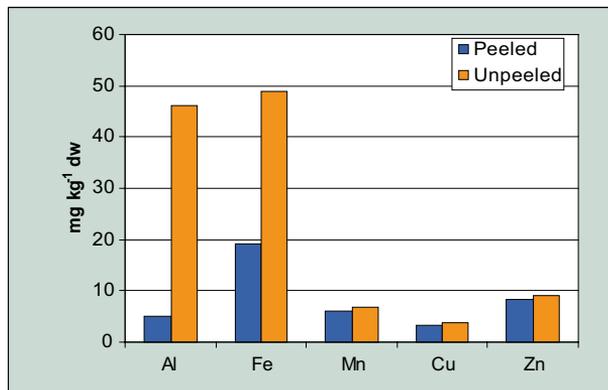


Figure 2. Aluminium (Al), iron (Fe), manganese (Mn), copper (Cu) and zinc (Zn) contents (mg kg<sup>-1</sup> dw) in potato tubers peeled (column left) and unpeeled (column right). Samples collected from the farms in the Tampere region in 2005 (Mäkelä-Kurto et. al 2006).

## Appendix 6

Table 1. Trace element, phosphorus (P) and sulphur (S) contents (mg kg<sup>-1</sup> dm) in wheat grains, potato tubers (unpeeled), and timothy grass, the second cut, by crop plants grown in the Pirkanmaa region and sampled in 2005 (Mäkelä-Kurtto et al. 2006).

	Wheat (n=5)				Potato tubers unpeeled (n=5)				Timothy grass 2nd cut (n = 5)			
	Min	Med	Mean	Max	Min	Med	Mean	Max	Min	Med	Mean	Max
P	3800	4160	4170	4590	1880	2260	2320	2750	2510	3040	3480	5070
S	1180	1320	1320	1430	1280	1390	1360	1420	1560	1890	2470	4350
Al	2.3	2.5	2.5	2.6	22.2	51.7	46.2	72.7	12.1	15.6	16.3	22.9
As	0.004	0.005	0.005	0.005	0.006	0.008	0.008	0.011	0.011	0.014	0.014	0.018
Cd	0.003	0.025	0.031	0.072	0.019	0.041	0.039	0.056	0.009	0.016	0.015	0.023
Cr	0.03	0.04	0.04	0.05	0.04	0.07	0.09	0.15	0.04	0.06	0.06	0.08
Cu	2.9	5.7	5.2	6.1	2.5	3.6	3.8	6.2	4.4	5.5	6.1	8.8
Fe	29.0	26.9	34.9	41.1	35.9	48.5	49.0	61.9	79.3	87.3	85.0	88.2
Mn	20.3	31.2	33.9	47.7	9.0	6.2	6.8	7.1	29.6	44.5	43.5	51.7
Ni	0.078	0.098	0.153	0.349	0.0910	0.137	0.234	0.510	0.468	0.646	0.620	0.732
Pb	0.014	0.015	0.019	0.035	0.029	0.031	0.032	0.036	0.027	0.09	0.078	0.104
Se	0.081	0.190	0.160	0.210	0.004	0.043	0.036	0.077	0.028	0.089	0.092	0.180
V	0.01	0.02	0.01	0.02	0.07	0.09	0.10	0.13	0.03	0.03	0.03	0.05
Zn	21.9	23.9	26.1	30.9	7.4	9.5	9.1	10.7	17.0	22.1	21.6	25.5

Table 2. Soil-to-plant uptake factors (element content in plant tissue, mg kg<sup>-1</sup> dm / *aqua regia* extractable element in soil, mg/kg dm) of phosphorus (P), sulphur (S) and trace elements by crop plants grown in the Pirkanmaa region and sampled in 2005 (Mäkelä-Kurtto et al. 2006).

	Soil-to-plant uptake factor											
	Wheat grains (n = 5)				Potato tubers, unpeeled (n = 5)				Timothy grass 2nd cut (n = 5)			
	Min	Med	Mean	Max	Min	Med	Mean	Max	Min	Med	Mean	Max
P	2.980	4.380	3.940	4.840	1.840	2.060	2.290	3.180	3.430	4.300	4.240	4.970
S	4.050	4.620	4.860	6.130	3.860	5.520	5.150	6.020	6.400	10.07	9.990	14.25
Al	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.001	0.001
Fe	0.001	0.001	0.001	0.001	0.000	0.001	0.001	0.001	0.002	0.003	0.003	0.003
Mn	0.020	0.040	0.040	0.080	0.000	0.010	0.010	0.010	0.030	0.040	0.040	0.050
As	0.001	0.001	0.001	0.002	0.001	0.001	0.001	0.001	0.003	0.003	0.004	0.006
Cd	0.010	0.130	0.150	0.370	0.110	0.150	0.180	0.280	0.040	0.070	0.070	0.100
Cr	0.001	0.001	0.001	0.001	0.000	0.001	0.002	0.004	0.001	0.001	0.001	0.002
Cu	0.120	0.230	0.220	0.300	0.070	0.130	0.170	0.370	0.210	0.350	0.350	0.420
Ni	0.003	0.008	0.007	0.01	0.003	0.009	0.011	0.025	0.019	0.031	0.030	0.038
Pb	0.001	0.001	0.002	0.003	0.001	0.002	0.002	0.003	0.002	0.006	0.006	0.009
Se	0.340	0.930	0.830	1.340	0.010	0.300	0.230	0.510	0.140	0.430	0.440	0.870
V	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.001	0.001
Zn	0.140	0.240	0.250	0.360	0.060	0.100	0.090	0.110	0.150	0.200	0.220	0.360

## Appendix 7

Table 1. Field mass balances of cadmium at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance) and with the European AROMIS-model (AROMIS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable Cd in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Cadmium (Cd)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>0.393</b>	<b>0.330</b>	<b>0.323</b>	<b>0.401</b>	<b>0.427</b>	(g/ha/a)
Fertiliser products	0.100	0.036	0.029	0.108	0.133	(g/ha/a)
Atmospheric deposition	0.293	0.293	0.293	0.293	0.293	(g/ha/a)
<b>Outputs</b>	<b>0.390</b>	<b>0.250</b>	<b>0.300</b>	<b>0.323</b>	<b>0.265</b>	(g/ha/a)
Leaching	0.100	0.100	0.100	0.100	0.100	(g/ha/a)
Crop plants	0.195	0.050	0.060	0.078	0.055	(g/ha/a)
Erosion	0.095	0.100	0.140	0.145	0.110	(g/ha/a)
<b>RAKAS-balance</b>	<b>0.003</b>	<b>0.079</b>	<b>0.023</b>	<b>0.078</b>	<b>0.161</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>0.090</b>	<b>0.160</b>	<b>0.220</b>	<b>0.220</b>	<b>0.250</b>	(g/ha/a)
Cd in topsoil in 2004	0.190	0.200	0.280	0.290	0.220	(mg/kg)
Cd in subsoil in 2004	0.120	0.110	0.150	0.160	0.070	(mg/kg)
Cd ratio: Top-/subsoil	1.56	1.80	1.87	1.78	3.10	
Soil type	RC	RC	RC	RC	RC	

Cadmium (Cd)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>0.392</b>	<b>0.437</b>	<b>0.347</b>	<b>0.317</b>	<b>0.313</b>	(g/ha/a)
Fertiliser products	0.190	0.125	0.149	0.042	0.079	(g/ha/a)
Commercial feeds	0.034	0.145	0.031	0.108	0.067	(g/ha/a)
Atmospheric deposition	0.167	0.167	0.167	0.167	0.167	(g/ha/a)
<b>Outputs</b>	<b>0.180</b>	<b>0.172</b>	<b>0.240</b>	<b>0.127</b>	<b>0.168</b>	(g/ha/a)
Leaching	0.100	0.100	0.100	0.100	0.100	(g/ha/a)
Crop plants	0.005	0.000	0.001	0.000	0.001	(g/ha/a)
Erosion	0.075	0.050	0.080	0.027	0.035	(g/ha/a)
Milk and meat	0.000	0.001	0.001	0.000	0.000	(g/ha/a)
Manure	0.000	0.021	0.058	0.000	0.032	(g/ha/a)
<b>RAKAS-balance</b>	<b>0.212</b>	<b>0.265</b>	<b>0.108</b>	<b>0.190</b>	<b>0.145</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>-0.660</b>	<b>-0.490</b>	<b>0.190</b>	<b>-0.720</b>	<b>-0.690</b>	(g/ha/a)
Cd in topsoil in 2004	0.150	0.100	0.160	0.053	0.070	(mg/kg)
Cd in subsoil in 2004	0.060	0.030	0.110	0.030	0.050	(mg/kg)
Cd ratio: Top-/subsoil	2.50	3.30	1.45	1.67	1.40	
Soil type	S	S	P	S	S	

Table 2. Field mass balances of chromium at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance) and with the European AROMIS-model (AROMIS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable Cr in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Chromium (Cr)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>364</b>	<b>2.3</b>	<b>2.2</b>	<b>4.9</b>	<b>515</b>	(g/ha/a)
Fertiliser products	364	1.5	1.5	4.2	515	(g/ha/a)
Atmospheric deposition	0.74	0.74	0.74	0.74	0.74	(g/ha/a)
<b>Outputs</b>	<b>34</b>	<b>40</b>	<b>16</b>	<b>34</b>	<b>41</b>	(g/ha/a)
Leaching	0.70	0.70	0.70	0.70	0.70	(g/ha/a)
Crop plants	0.17	0.13	0.19	0.06	0.08	(g/ha/a)
Erosion	33	39	15	33	40	(g/ha/a)
<b>RAKAS-balance</b>	<b>330</b>	<b>-38</b>	<b>-14</b>	<b>-29</b>	<b>474</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>363</b>	<b>0.70</b>	<b>1.5</b>	<b>3.6</b>	<b>514</b>	(g/ha/a)
Cr in topsoil in 2004	66	79	29	65	80	(mg/kg)
Cr in subsoil in 2004	79	98	45	84	96	(mg/kg)
Cr ratio: Top-/subsoil	0.84	0.80	0.64	0.78	0.84	
Soil type	RC	RC	RC	RC	RC	

Chromium (Cr)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>726</b>	<b>13</b>	<b>7.9</b>	<b>13</b>	<b>6.9</b>	(g/ha/a)
Fertiliser products	723	4.8	4.3	1.5	2.9	(g/ha/a)
Commercial feeds	1.8	7.4	3.1	11	3.5	(g/ha/a)
Atmospheric deposition	0.49	0.49	0.49	0.49	0.49	(g/ha/a)
<b>Outputs</b>	<b>18</b>	<b>12</b>	<b>22</b>	<b>7.1</b>	<b>6.7</b>	(g/ha/a)
Leaching	0.70	0.70	0.70	0.70	0.70	(g/ha/a)
Crop plants	0.01	0.00	0.01	0.00	0.01	(g/ha/a)
Erosion	17	11	20	6.3	5.4	(g/ha/a)
Milk and meat	0.05	0.08	0.07	0.08	0.06	(g/ha/a)
Manure	0.00	0.20	1.1	0.00	0.61	(g/ha/a)
<b>RAKAS-balance</b>	<b>708</b>	<b>0.7</b>	<b>-14</b>	<b>5.8</b>	<b>0.13</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>719</b>	<b>6.8</b>	<b>3.6</b>	<b>6.3</b>	<b>0.10</b>	(g/ha/a)
Cr in topsoil in 2004	34	21	40	13	11	(mg/kg)
Cr in subsoil in 2004	37	24	33	14	8.6	(mg/kg)
Cr ratio: Top-/subsoil	0.91	0.88	1.19	0.92	1.24	
Soil type	S	S	P	S	S	

Table 3. Field mass balances of copper at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance) and with the European AROMIS-model (AROMIS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable Cu in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Copper (Cu)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>18</b>	<b>7.6</b>	<b>8.0</b>	<b>13</b>	<b>24</b>	(g/ha/a)
Fertiliser products	13	2.1	2.5	7.6	18	(g/ha/a)
Atmospheric deposition	5.5	5.5	5.5	5.5	5.5	(g/ha/a)
<b>Outputs</b>	<b>34</b>	<b>31</b>	<b>36</b>	<b>33</b>	<b>37</b>	(g/ha/a)
Leaching	4.3	4.3	4.3	4.3	4.3	(g/ha/a)
Crop plants	18	7.6	24	15	12	(g/ha/a)
Erosion	12	19	7.9	14	21	(g/ha/a)
<b>RAKAS-balance</b>	<b>-16</b>	<b>-23</b>	<b>-28</b>	<b>-20</b>	<b>-13</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>-18</b>	<b>-20</b>	<b>-25</b>	<b>-20</b>	<b>-7.6</b>	(g/ha/a)
Cu in topsoil in 2004	24	38	16	27	42	(mg/kg)
Cu in subsoil in 2004	28	41	21	34	50	(mg/kg)
Cu ratio: Top-/subsoil	0.86	0.90	0.74	0.80	0.85	
Soil type	RC	RC	RC	RC	RC	

Copper (Cu)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>43</b>	<b>49</b>	<b>17</b>	<b>45</b>	<b>25</b>	(g/ha/a)
Fertiliser products	25	5.5	3.2	1.5	3.2	(g/ha/a)
Commercial feeds	14	40	10	40	18	(g/ha/a)
Atmospheric deposition	3.8	3.8	3.8	3.8	3.8	(g/ha/a)
<b>Outputs</b>	<b>17</b>	<b>14</b>	<b>45</b>	<b>8</b>	<b>18</b>	(g/ha/a)
Leaching	4.3	4.3	4.3	4.3	4.3	(g/ha/a)
Crop plants	2.6	0.00	0.60	0.00	1.0	(g/ha/a)
Erosion	10	4.1	11	2.9	3.9	(g/ha/a)
Milk and meat	0.20	0.40	0.39	0.43	0.30	(g/ha/a)
Manure	0.00	5.4	28	0.00	9.0	(g/ha/a)
<b>RAKAS-balance</b>	<b>26</b>	<b>35</b>	<b>-27</b>	<b>37</b>	<b>6.6</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>6.1</b>	<b>14</b>	<b>-31</b>	<b>11</b>	<b>-19</b>	(g/ha/a)
Cu in topsoil in 2004	19	8.3	22	5.9	7.8	(mg/kg)
Cu in subsoil in 2004	17	8.2	16	5.4	4.5	(mg/kg)
Cu ratio: Top-/subsoil	1.15	1.02	1.36	1.10	1.71	
Soil type	S	S	P	S	S	

Table 4. Field mass balances of nickel at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance) and with the European AROMIS-model (AROMIS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable Ni in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Nickel (Ni)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>8.3</b>	<b>2.3</b>	<b>2.5</b>	<b>5.3</b>	<b>12</b>	(g/ha/a)
Fertiliser products	6.7	0.66	0.90	3.7	10	(g/ha/a)
Atmospheric deposition	1.6	1.6	1.6	1.6	1.6	(g/ha/a)
<b>Outputs</b>	<b>20</b>	<b>25</b>	<b>11</b>	<b>20</b>	<b>24</b>	(g/ha/a)
Leaching	3.9	3.9	3.9	3.9	3.9	(g/ha/a)
Crop plants	1.5	0.54	0.74	0.58	0.42	(g/ha/a)
Erosion	15	21	6.2	15	20	(g/ha/a)
<b>RAKAS-balance</b>	<b>-12</b>	<b>-23</b>	<b>-8.5</b>	<b>-15</b>	<b>-12</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>4.0</b>	<b>-1.1</b>	<b>0.58</b>	<b>2.3</b>	<b>8.5</b>	(g/ha/a)
Ni in topsoil in 2004	29	41	12	31	39	(mg/kg)
Ni in subsoil in 2004	34	46	21	36	46	(mg/kg)
Ni ratio: Top-/subsoil	0.88	0.90	0.60	0.87	0.84	
Soil type	RC	RC	RC	RC	RC	

Nickel (Ni)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>18</b>	<b>5.4</b>	<b>12</b>	<b>2.6</b>	<b>3.6</b>	(g/ha/a)
Fertiliser products	14	3.1	11	0.49	1.7	(g/ha/a)
Commercial feeds	3.4	1.3	0.30	1.1	0.89	(g/ha/a)
Atmospheric deposition	1.0	1.0	1.0	1.0	1.0	(g/ha/a)
<b>Outputs</b>	<b>12</b>	<b>8.3</b>	<b>15</b>	<b>6.7</b>	<b>7.6</b>	(g/ha/a)
Leaching	3.9	3.9	3.9	3.9	3.9	(g/ha/a)
Crop plants	0.02	0.00	0.02	0.00	0.01	(g/ha/a)
Erosion	7.9	3.7	8.1	2.8	2.4	(g/ha/a)
Milk and meat	0.00	0.02	0.01	0.02	0.01	(g/ha/a)
Manure	0.00	0.63	2.9	0.00	1.3	(g/ha/a)
<b>RAKAS-balance</b>	<b>6.5</b>	<b>-2.9</b>	<b>-2.6</b>	<b>-4.1</b>	<b>-4.0</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>13</b>	<b>0.6</b>	<b>-6.8</b>	<b>-1.7</b>	<b>5.6</b>	(g/ha/a)
Ni in topsoil in 2004	16	7.4	16	5.5	4.8	(mg/kg)
Ni in subsoil in 2004	17	10	16	5.4	3.4	(mg/kg)
Ni ratio: Top-/subsoil	0.91	0.72	1.03	1.03	1.42	
Soil type	S	S	P	S	S	

Table 5. Field mass balances of lead at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance) and with the European AROMIS-model (AROMIS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable Pb in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Lead (Pb)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>9.4</b>	<b>8.8</b>	<b>8.8</b>	<b>10</b>	<b>9.8</b>	(g/ha/a)
Fertiliser products	0.80	0.18	0.20	1.4	1.2	(g/ha/a)
Atmospheric deposition	8.6	8.6	8.6	8.6	8.6	(g/ha/a)
<b>Outputs</b>	<b>11</b>	<b>12</b>	<b>7.2</b>	<b>7.8</b>	<b>13.5</b>	(g/ha/a)
Leaching	0.50	0.50	0.50	0.50	0.50	(g/ha/a)
Crop plants	0.08	1.1	0.15	0.06	0.03	(g/ha/a)
Erosion	10	10	6.5	7.2	13	(g/ha/a)
<b>RAKAS-balance</b>	<b>-1.3</b>	<b>-3.2</b>	<b>1.6</b>	<b>2.2</b>	<b>-3.7</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>-0.30</b>	<b>-2.8</b>	<b>4.3</b>	<b>0.80</b>	<b>-0.50</b>	(g/ha/a)
Pb in topsoil in 2004	20	21	13	14	25	(mg/kg)
Pb in subsoil in 2004	19	18	14	13	23	(mg/kg)
Pb ratio: Top-/subsoil	1.09	1.13	0.92	1.13	1.11	
Soil type	RC	RC	RC	RC	RC	

Lead (Pb)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>6.9</b>	<b>6.8</b>	<b>7.5</b>	<b>6.1</b>	<b>5.9</b>	(g/ha/a)
Fertiliser products	2.0	1.3	2.4	0.20	0.70	(g/ha/a)
Commercial feeds	0.17	0.75	0.31	1.1	0.30	(g/ha/a)
Atmospheric deposition	4.8	4.8	4.8	4.8	4.8	(g/ha/a)
<b>Outputs</b>	<b>4.8</b>	<b>8.2</b>	<b>5.7</b>	<b>2.4</b>	<b>3.3</b>	(g/ha/a)
Leaching	0.50	0.50	0.50	0.50	0.50	(g/ha/a)
Crop plants	0.10	0.00	0.02	0.00	0.02	(g/ha/a)
Erosion	4.3	3.8	4.8	1.9	1.7	(g/ha/a)
Milk and meat	0.01	0.01	0.01	0.01	0.01	(g/ha/a)
Manure	0.00	3.9	0.43	0.00	1.1	(g/ha/a)
<b>RAKAS-balance</b>	<b>2.1</b>	<b>-1.4</b>	<b>1.8</b>	<b>3.7</b>	<b>2.6</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>-19</b>	<b>-16</b>	<b>-8</b>	<b>-20</b>	<b>-12</b>	(g/ha/a)
Pb in topsoil in 2004	8.6	7.5	9.6	3.8	3.3	(mg/kg)
Pb in subsoil in 2004	7.5	5.0	7.0	3.7	2.6	(mg/kg)
Pb ratio: Top-/subsoil	1.16	1.51	1.37	1.02	1.27	
Soil type	S	S	P	S	S	

Table 6. Field mass balances of zinc at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance) and with the European AROMIS-model (AROMIS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable Zn in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Zinc (Zn)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>124</b>	<b>31</b>	<b>158</b>	<b>134</b>	<b>281</b>	(g/ha/a)
Fertiliser products	97	4,2	131	108	255	(g/ha/a)
Atmospheric deposition	27	27	27	27	27	(g/ha/a)
<b>Outputs</b>	<b>156</b>	<b>113</b>	<b>163</b>	<b>150</b>	<b>123</b>	(g/ha/a)
Leaching	7.5	7.5	7.5	7.5	7.5	(g/ha/a)
Crop plants	94	37	121	77	61	(g/ha/a)
Erosion	54	68	34	66	55	(g/ha/a)
<b>RAKAS-balance</b>	<b>-32</b>	<b>-82</b>	<b>-5</b>	<b>-16</b>	<b>158</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>-11</b>	<b>-51</b>	<b>19</b>	<b>19</b>	<b>178</b>	(g/ha/a)
Zn in topsoil in 2004	108	136	67	131	110	(mg/kg)
Zn in subsoil in 2004	104	118	70	118	107	(mg/kg)
Zn ratio: Top-/subsoil	1.03	1.16	0.95	1.12	1.03	
Soil type	RC	RC	RC	RC	RC	

Zinc (Zn)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>283</b>	<b>719</b>	<b>369</b>	<b>383</b>	<b>449</b>	(g/ha/a)
Fertiliser products	199	480	237	173	321	(g/ha/a)
Commercial feeds	67	222	115	193	110	(g/ha/a)
Atmospheric deposition	17	17	17	17	17	(g/ha/a)
<b>Outputs</b>	<b>65</b>	<b>78</b>	<b>259</b>	<b>47</b>	<b>83</b>	(g/ha/a)
Leaching	7.5	7.5	7.5	7.5	7.5	(g/ha/a)
Crop plants	10	0.00	2.3	0.00	5.5	(g/ha/a)
Erosion	32	16	28	11	12	(g/ha/a)
Milk and meat	16	22	21	28	16	(g/ha/a)
Manure	0.00	32	200	0,00	43	(g/ha/a)
<b>RAKAS-balance</b>	<b>219</b>	<b>641</b>	<b>110</b>	<b>335</b>	<b>365</b>	(g/ha/a)
<b>AROMIS-balance</b>	<b>83</b>	<b>514</b>	<b>40</b>	<b>179</b>	<b>245</b>	(g/ha/a)
Zn in topsoil in 2004	63	33	56	23	24	(mg/kg)
Zn in subsoil in 2004	49	28	43	18	18	(mg/kg)
Zn ratio: Top-/subsoil	1.29	1.15	1.29	1.25	1.33	
Soil type	S	S	P	S	S	

Table 7. Field mass balances of mercury at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable Hg in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Mercury (Hg)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>0.067</b>	<b>0.052</b>	<b>0.050</b>	<b>0.063</b>	<b>0.074</b>	(g/ha/a)
Fertiliser products	0.017	0.002	0.002	0.013	0.024	(g/ha/a)
Atmospheric deposition	0.050	0.050	0.050	0.050	0.050	(g/ha/a)
<b>Outputs</b>	<b>0.030</b>	<b>0.026</b>	<b>0.028</b>	<b>0.045</b>	<b>0.040</b>	(g/ha/a)
Leaching	0.010	0.010	0.010	0.000	0.010	(g/ha/a)
Crop plants	0.000	0.001	0.028	0.010	0.000	(g/ha/a)
Erosion	0.020	0.015	0.020	0.035	0.030	(g/ha/a)
<b>RAKAS-balance</b>	<b>0.037</b>	<b>0.026</b>	<b>0.020</b>	<b>0.018</b>	<b>0.034</b>	(g/ha/a)
Hg in topsoil in 2004	0.039	0.025	0.044	0.075	0.058	(mg/kg)
Hg in subsoil in 2004	0.031	0.021	0.032	0.026	0.032	(mg/kg)
Hg ratio: Top-/subsoil	1.26	1.38	1.38	2.88	1.81	
Soil type	RC	RC	RC	RC	RC	

Mercury (Hg)	Crop farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>0.089</b>	<b>0.070</b>	<b>0.062</b>	<b>0.063</b>	<b>0.061</b>	(g/ha/a)
Fertiliser products	0.038	0.013	0.009	0.002	0.008	(g/ha/a)
Commercial feeds	0.001	0.007	0.003	0.011	0.003	(g/ha/a)
Atmospheric deposition	0.050	0.050	0.050	0.050	0.050	(g/ha/a)
<b>Outputs</b>	<b>0.036</b>	<b>0.043</b>	<b>0.067</b>	<b>0.029</b>	<b>0.034</b>	(g/ha/a)
Leaching	0.010	0.010	0.010	0.010	0.010	(g/ha/a)
Crop plants	0.001	0.000	0.000	0.000	0.000	(g/ha/a)
Erosion	0.019	0.015	0.025	0.006	0.005	(g/ha/a)
Milk and meat	0.008	0.014	0.011	0.013	0.010	(g/ha/a)
Manure	0.000	0.004	0.020	0.000	0.009	(g/ha/a)
<b>RAKAS-balance</b>	<b>0.053</b>	<b>0.027</b>	<b>-0.004</b>	<b>0.034</b>	<b>0.027</b>	(g/ha/a)
Hg in topsoil in 2004	0.035	0.034	0.049	0.015	0.012	(mg/kg)
Hg in subsoil in 2004	0.007	0.008	0.039	0.011	0.008	(mg/kg)
Hg ratio: Top-/subsoil	5.00	4.25	1.26	1.36	1.50	
Soil type	S	S	P	S	S	

Table 8. Field mass balances of vanadium at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable V in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Vanadium (V)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>6039</b>	<b>4.5</b>	<b>4.4</b>	<b>25</b>	<b>8543</b>	(g/ha/a)
Fertiliser products	6036	1.6	1.6	22	8540	(g/ha/a)
Atmospheric deposition	2.9	2.9	2.9	2.9	2.9	(g/ha/a)
<b>Outputs</b>	<b>52</b>	<b>54</b>	<b>27</b>	<b>65</b>	<b>63</b>	(g/ha/a)
Leaching	6.8	7.0	3.5	22	0.02	(g/ha/a)
Crop plants	0.14	0.13	0.10	0.03	8.3	(g/ha/a)
Erosion	45	46	23	43	55	(g/ha/a)
<b>RAKAS-balance</b>	<b>5986</b>	<b>-49</b>	<b>-23</b>	<b>-41</b>	<b>8479</b>	(g/ha/a)
V in topsoil in 2004	91	93	47	87	110	(mg/kg)
V in subsoil in 2004	86	98	55	91	101	(mg/kg)
V ratio: Top-/subsoil	1.06	0.95	0.85	0.95	1.10	
Soil type	RC	RC	RC	RC	RC	

Vanadium (V)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>12027</b>	<b>28</b>	<b>12</b>	<b>14</b>	<b>16</b>	(g/ha/a)
Fertiliser products	12024	19	7.2	1.7	10	(g/ha/a)
Commercial feeds	1.4	7.5	3.1	11	3.5	(g/ha/a)
Atmospheric deposition	1.7	1.7	1.7	1.7	1.7	(g/ha/a)
<b>Outputs</b>	<b>29</b>	<b>14</b>	<b>53</b>	<b>10</b>	<b>13</b>	(g/ha/a)
Leaching	3.7	1.9	6.4	1.2	1.5	(g/ha/a)
Crop plants	0.01	0.00	0.00	0.00	0.05	(g/ha/a)
Erosion	25	12	43	8.3	10	(g/ha/a)
Milk and meat	0.00	0.01	0.01	0.00	0.01	(g/ha/a)
Manure	0.00	0.30	4.60	0.00	0.97	(g/ha/a)
<b>RAKAS-balance</b>	<b>11998</b>	<b>14</b>	<b>-41</b>	<b>4.8</b>	<b>3.1</b>	(g/ha/a)
V in topsoil in 2004	50	25	85	17	20	(mg/kg)
V in subsoil in 2004	47	29	47	17	12	(mg/kg)
V ratio: Top-/subsoil	1.06	0.87	1.81	0.97	1.71	
Soil type	S	S	P	S	S	

Table 9. Field mass balances of arsenic at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable As in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Arsenic (As)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>2.3</b>	<b>1.1</b>	<b>1.1</b>	<b>4.5</b>	<b>2.9</b>	(g/ha/a)
Fertiliser products	1.3	0.10	0.11	3.5	1.9	(g/ha/a)
Atmospheric deposition	1.0	1.0	1.0	1.0	1.0	(g/ha/a)
<b>Outputs</b>	<b>4.2</b>	<b>3.1</b>	<b>2.0</b>	<b>3.1</b>	<b>4.3</b>	(g/ha/a)
Leaching	0.48	0.48	0.48	0.48	0.48	(g/ha/a)
Crop plants	0.03	0.02	0.02	0.02	0.03	(g/ha/a)
Erosion	3.7	2.9	1.5	2.5	3.8	(g/ha/a)
<b>RAKAS-balance</b>	<b>-1.2</b>	<b>-2.0</b>	<b>-1.0</b>	<b>1.5</b>	<b>-1.4</b>	(g/ha/a)
As in topsoil in 2004	7.4	5.2	3.1	5.1	7.6	(mg/kg)
As in subsoil in 2004	11	6.3	4.8	6.0	8.0	(mg/kg)
As ratio: Top-/subsoil	0.70	0.83	0.65	0.85	0.95	
Soil type	RC	RC	RC	RC	RC	

Arsenic (As)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>4.9</b>	<b>3.9</b>	<b>2.6</b>	<b>1.1</b>	<b>2.3</b>	(g/ha/a)
Fertiliser products	4.3	3.1	2.0	0.13	1.6	(g/ha/a)
Commercial feeds	0.10	0.41	0.15	0.54	0.20	(g/ha/a)
Atmospheric deposition	0.4	0.4	0.4	0.4	0.4	(g/ha/a)
<b>Outputs</b>	<b>3.0</b>	<b>2.3</b>	<b>3.6</b>	<b>1.3</b>	<b>1.3</b>	(g/ha/a)
Leaching	0.48	0.48	0.48	0.48	0.48	(g/ha/a)
Crop plants	0.00	0.00	0.00	0.00	0.00	(g/ha/a)
Erosion	2.5	1.1	2.3	0.84	0.67	(g/ha/a)
Milk and meat	0.00	0.01	0.01	0.01	0.01	(g/ha/a)
Manure	0.00	0.08	0.89	0.00	0.15	(g/ha/a)
<b>RAKAS-balance</b>	<b>1.9</b>	<b>2.3</b>	<b>-1.0</b>	<b>-0.22</b>	<b>1.0</b>	(g/ha/a)
As in topsoil in 2004	5.0	2.2	4.6	1.7	1.3	(mg/kg)
As in subsoil in 2004	4.1	2.2	5.4	1.8	1.3	(mg/kg)
As ratio: Top-/subsoil	1.24	1.00	0.85	0.94	1.03	
Soil type	S	S	P	S	S	

Table 10. Field mass balances of selenium at the farm level in the soil of the crop farms (1-5) in southwestern Finland and of the dairy farms (6-10) in Ostrobothnia in 2004 as estimated with the RAKAS-model (RAKAS-balance). Contents (mg kg<sup>-1</sup>) of *aqua regia* extractable Se in the top- and subsoil in 2004 and content ratio: top-/subsoil. Soil types: RC = river clay; S = sand; and P = peat. \*Steel slag was used for soil improvement on the farm in 2004.

Selenium (Se)	Crop farms					Unit
	1*	2	3	4	5*	
<b>Inputs</b>	<b>5.1</b>	<b>4.3</b>	<b>3.6</b>	<b>4.8</b>	<b>5.7</b>	(g/ha/a)
Fertiliser products	4.4	3.6	2.9	4.1	5.1	(g/ha/a)
Atmospheric deposition	0.67	0.67	0.67	0.67	0.67	(g/ha/a)
<b>Outputs</b>	<b>0.73</b>	<b>0.25</b>	<b>0.80</b>	<b>0.76</b>	<b>0.72</b>	(g/ha/a)
Leaching	0.00	0.00	0.00	0.00	0.00	(g/ha/a)
Crop plants	0.64	0.19	0.70	0.65	0.55	(g/ha/a)
Erosion	0.09	0.08	0.20	0.11	0.18	(g/ha/a)
<b>RAKAS-balance</b>	<b>4.4</b>	<b>4.0</b>	<b>2.7</b>	<b>4.0</b>	<b>5.0</b>	(g/ha/a)
Se in topsoil in 2004	0.18	0.16	0.35	0.22	0.35	(mg/kg)
Se in subsoil in 2004	0.26	0.22	0.25	0.33	0.21	(mg/kg)
Se ratio: Top-/subsoil	0.70	0.72	1.41	0.67	1.68	
Soil type	RC	RC	RC	RC	RC	

Selenium (Se)	Dairy farms					Unit
	6*	7	8	9	10	
<b>Inputs</b>	<b>5.5</b>	<b>9.0</b>	<b>4.9</b>	<b>5.4</b>	<b>6.2</b>	(g/ha/a)
Fertiliser products	4.2	7.1	4.0	4.1	5.2	(g/ha/a)
Commercial feeds	0.90	1.4	0.48	0.80	0.46	(g/ha/a)
Atmospheric deposition	0.47	0.47	0.47	0.47	0.47	(g/ha/a)
<b>Outputs</b>	<b>0.26</b>	<b>0.44</b>	<b>1.0</b>	<b>0.19</b>	<b>0.39</b>	(g/ha/a)
Leaching	0.00	0.00	0.00	0.00	0.00	(g/ha/a)
Crop plants	0.02	0.00	0.00	0.00	0.01	(g/ha/a)
Erosion	0.15	0.11	0.20	0.05	0.07	(g/ha/a)
Milk and meat	0.10	0.13	0.10	0.14	0.12	(g/ha/a)
Manure	0.00	0.20	0.70	0.00	0.19	(g/ha/a)
<b>RAKAS-balance</b>	<b>5.3</b>	<b>8.6</b>	<b>3.9</b>	<b>5.2</b>	<b>5.8</b>	(g/ha/a)
Se in topsoil in 2004	0.29	0.21	0.39	0.10	0.14	(mg/kg)
Se in subsoil in 2004	0.12	0.10	0.27	0.07	0.03	(mg/kg)
Se ratio: Top-/subsoil	2.42	2.10	1.44	1.43	4.53	
Soil type	S	S	P	S	S	

## Appendix 8

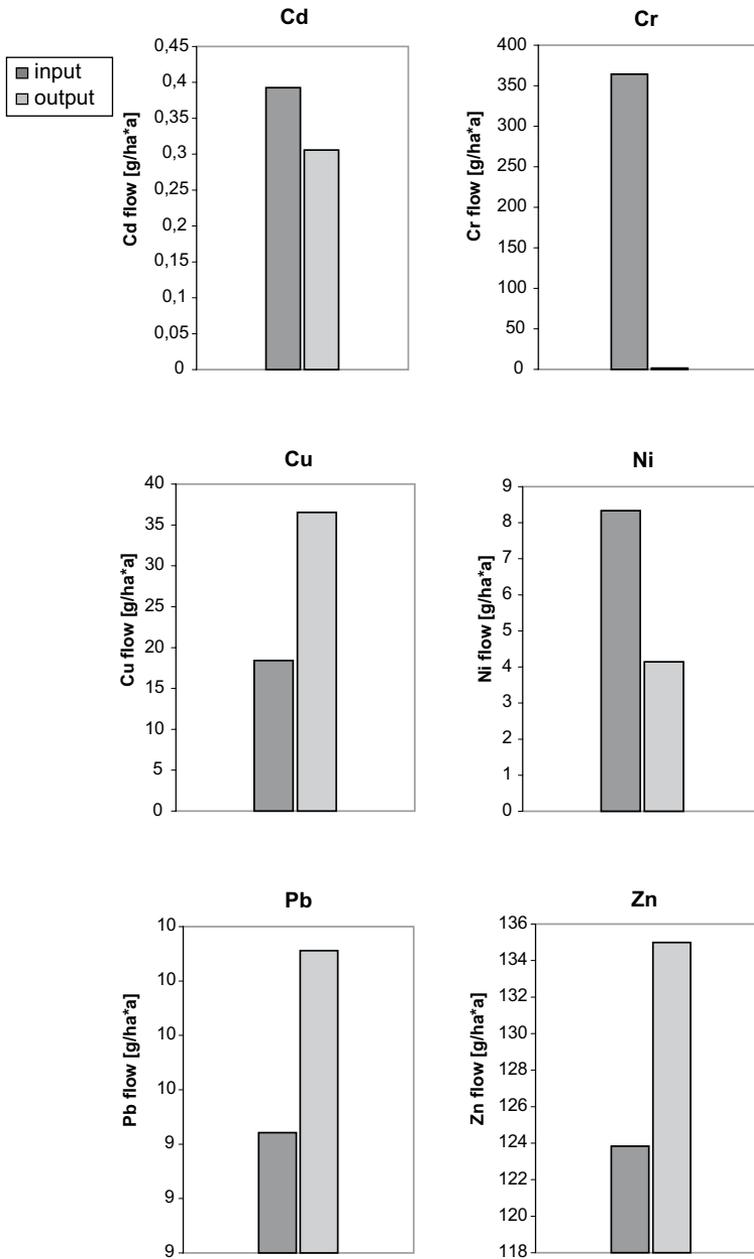
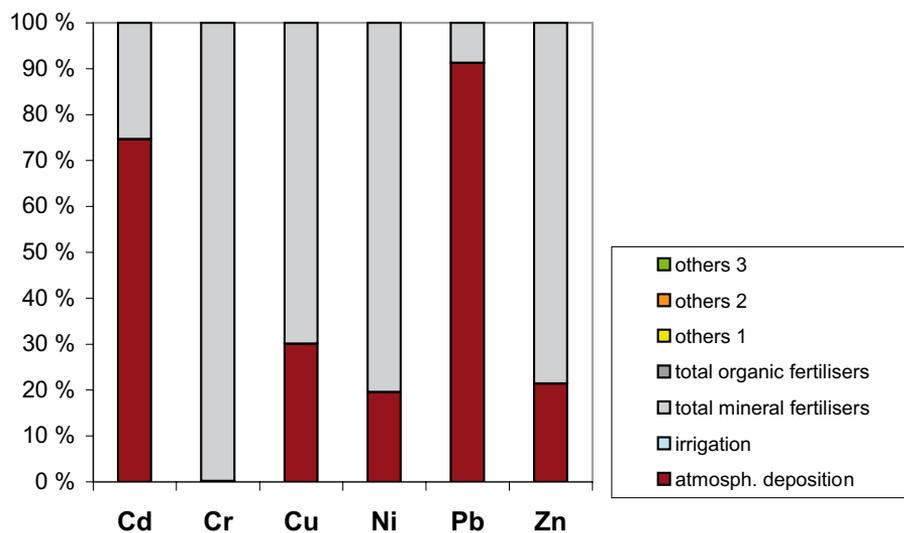


Figure 1. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 1 in southwestern Finland in 2004. Soil type river clay = RC. Steel slag was used on the farm in 2004.

### Heavy metal inputs



### Heavy metal outputs

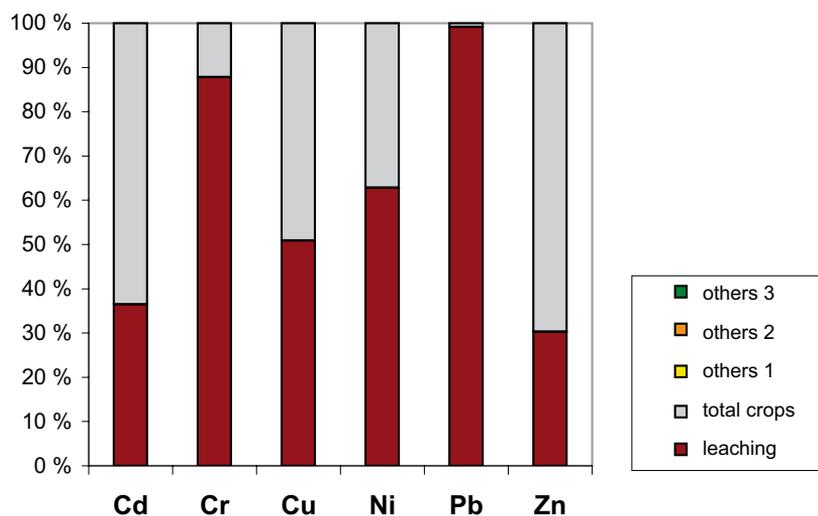


Figure 2. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various source to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 1 in southwestern Finland in 2004. Soil type river clay = RC. Steel slag was used on the farm in 2004.

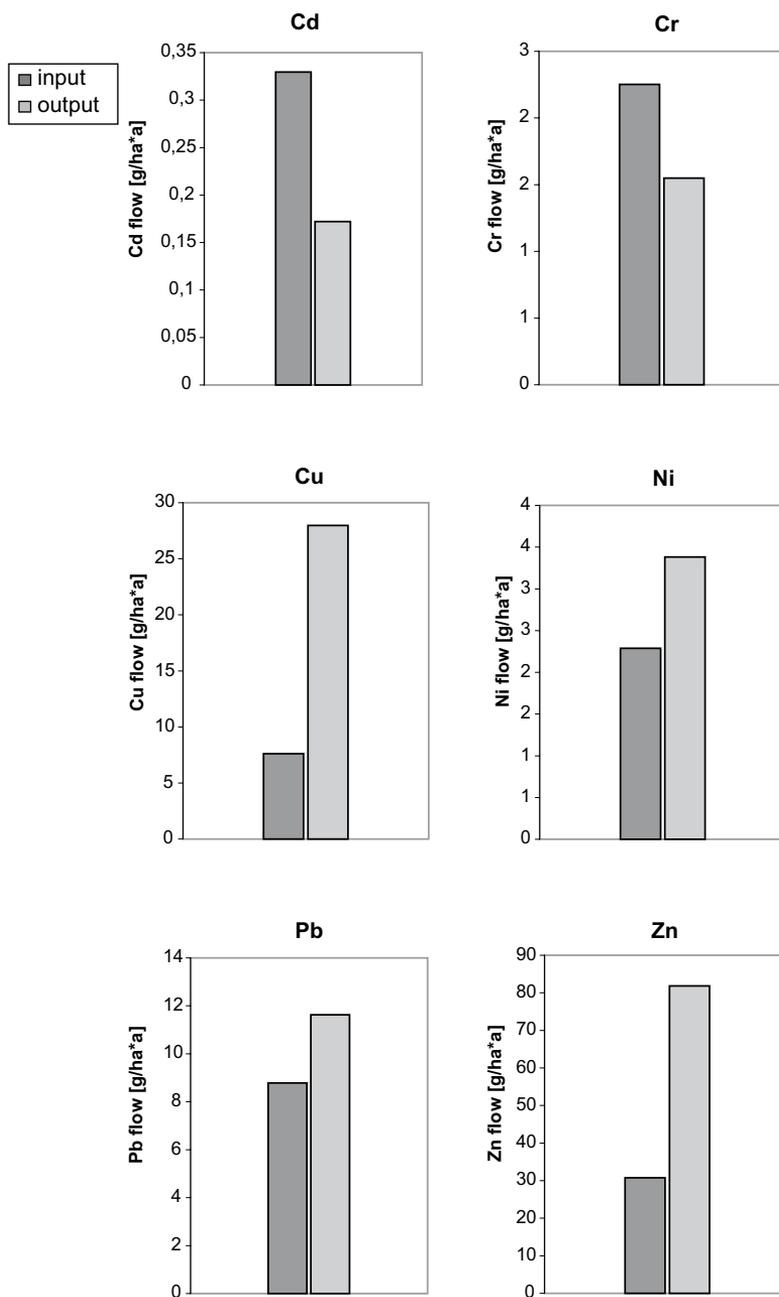
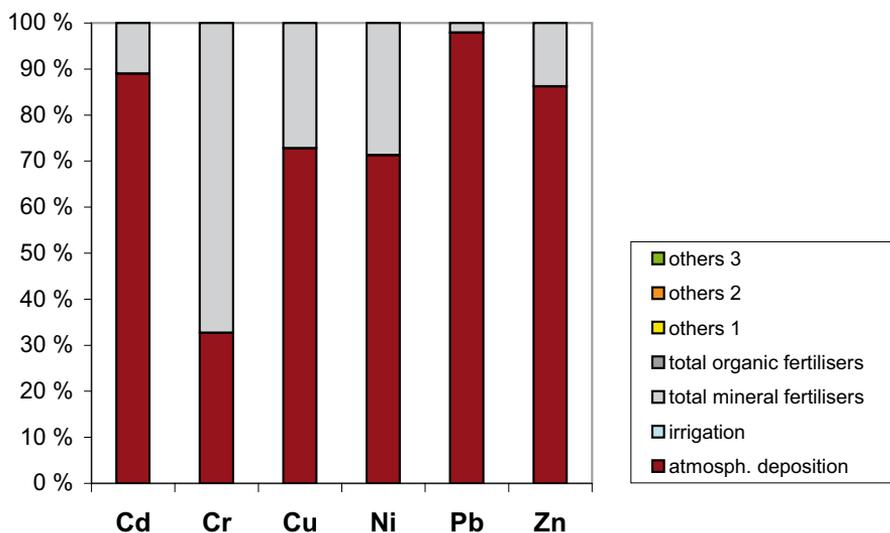


Figure 3. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 2 in southwestern Finland in 2004. Soil type river clay = RC.

### Heavy metal inputs



### Heavy metal outputs

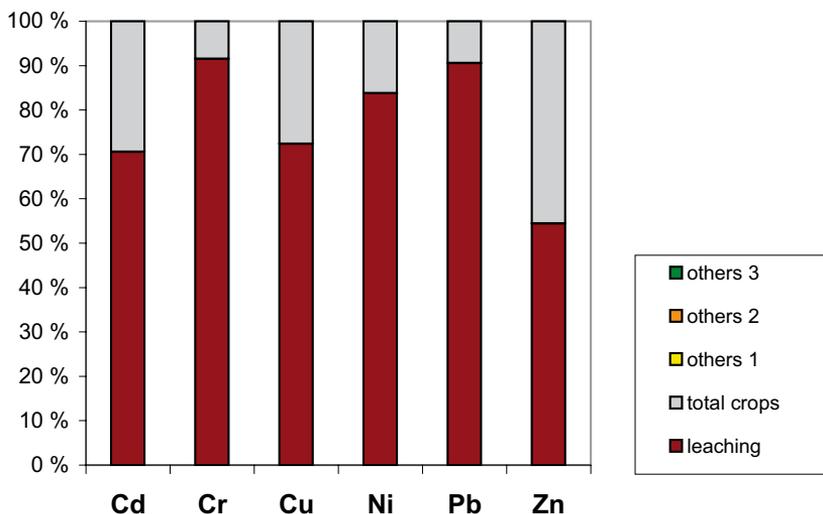


Figure 4. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 2 in southwestern Finland in 2004. Soil type river clay = RC.

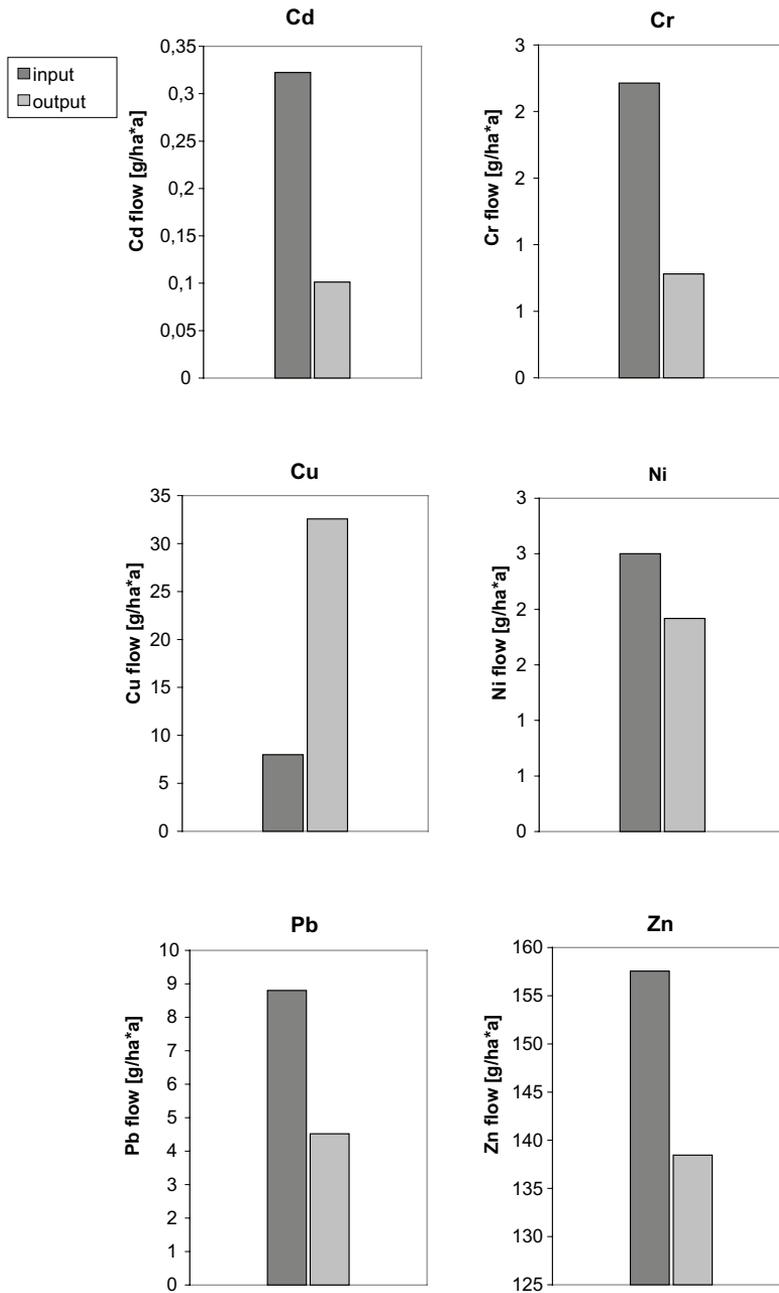


Figure 5. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 3 in southwestern Finland in 2004. Soil type river clay = RC.

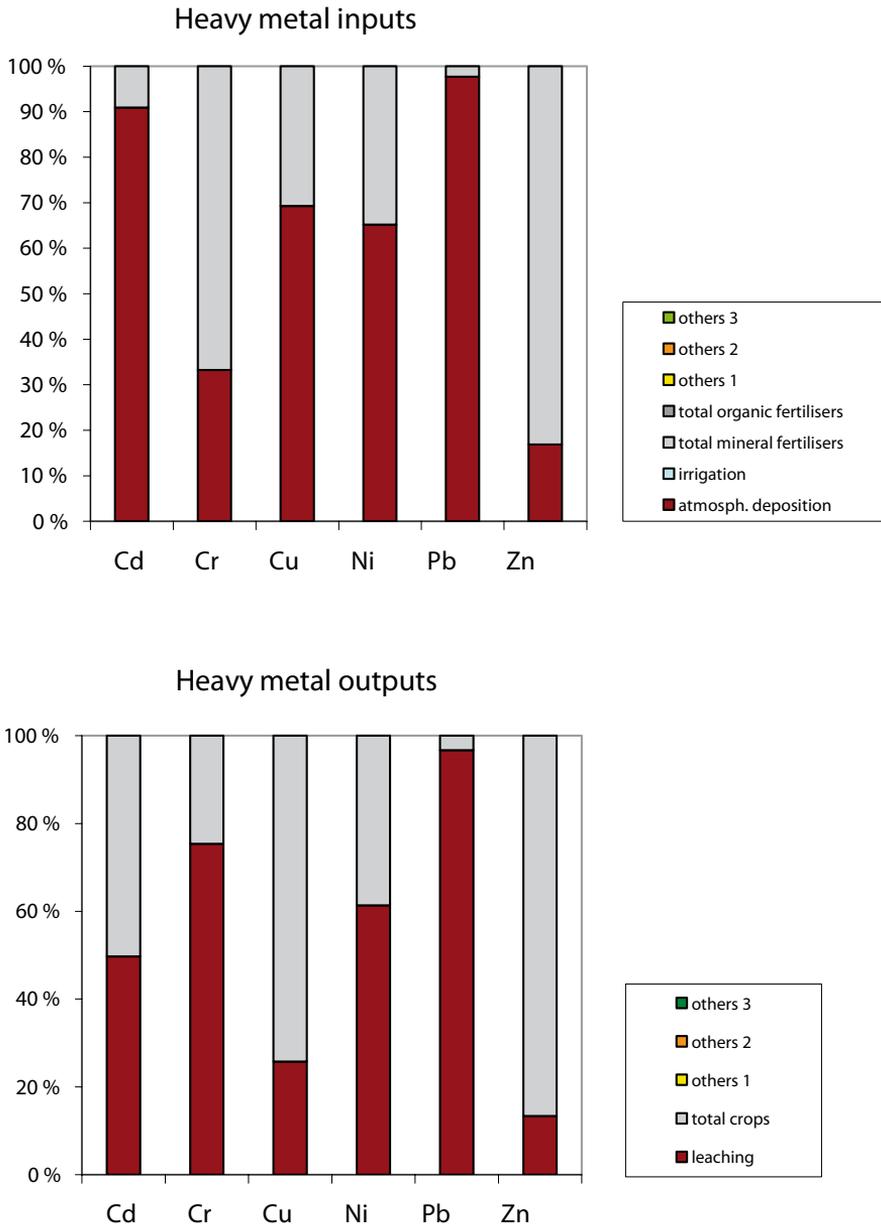


Figure 6. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 3 in southwestern Finland in 2004. Soil type river clay = RC.

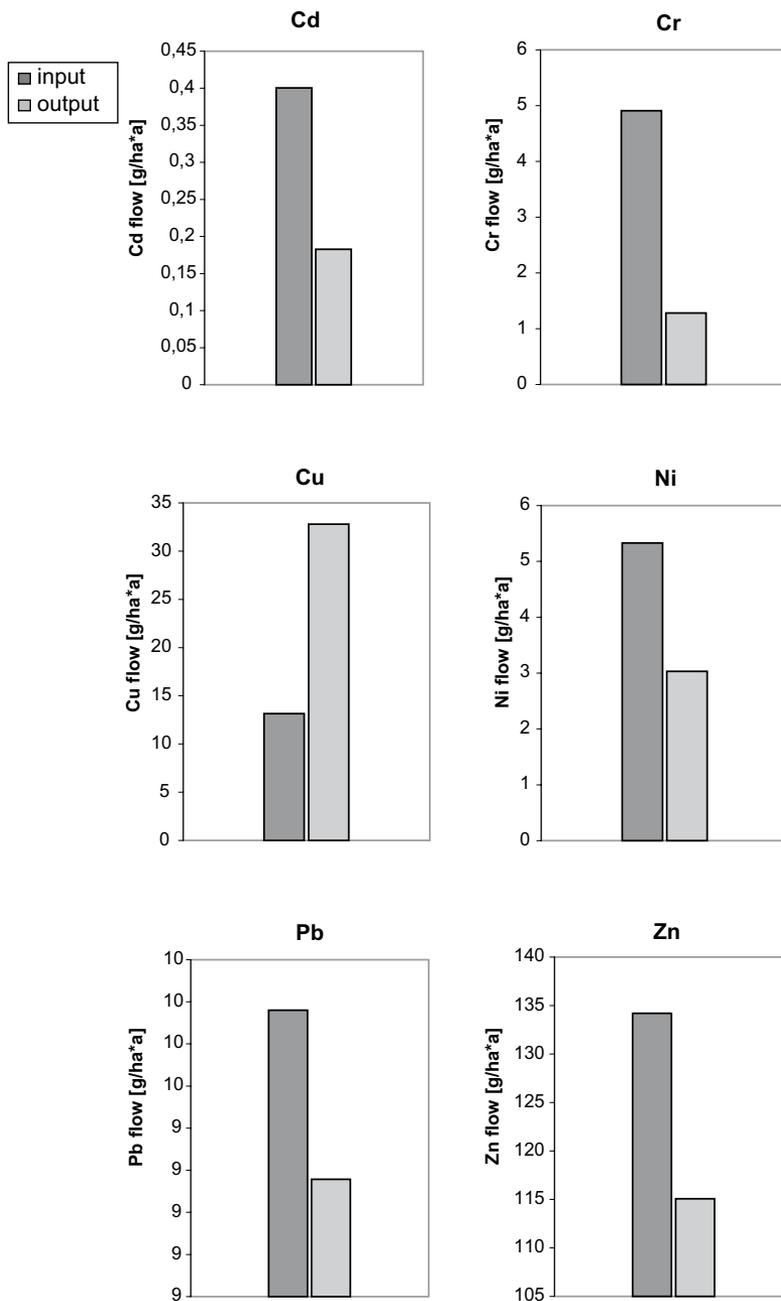
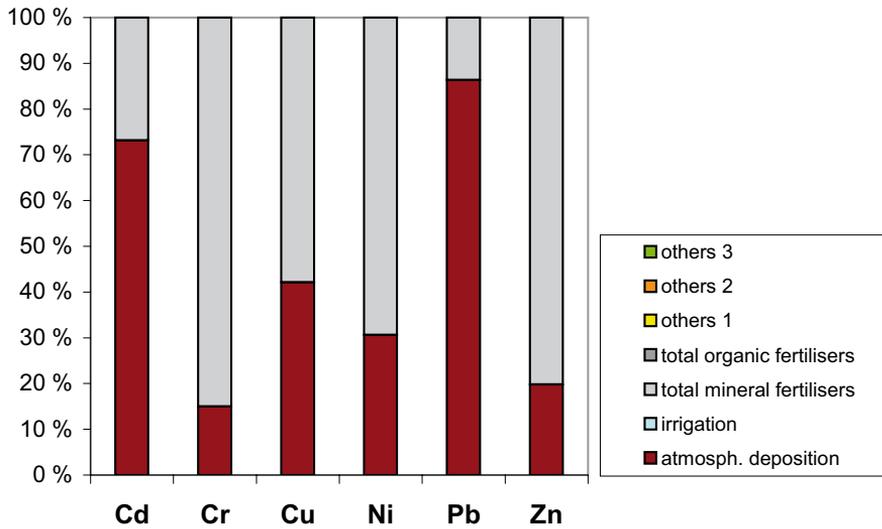


Figure 7. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 4 in southwestern Finland in 2004. Soil type river clay = RC.

### Heavy metal inputs



### Heavy metal outputs

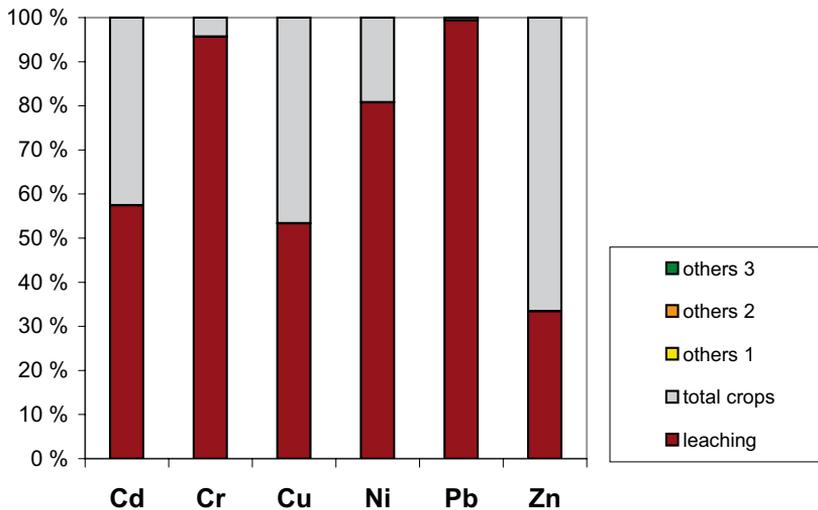


Figure 8. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 4 in southwestern Finland in 2004. Soil type river clay = RC.

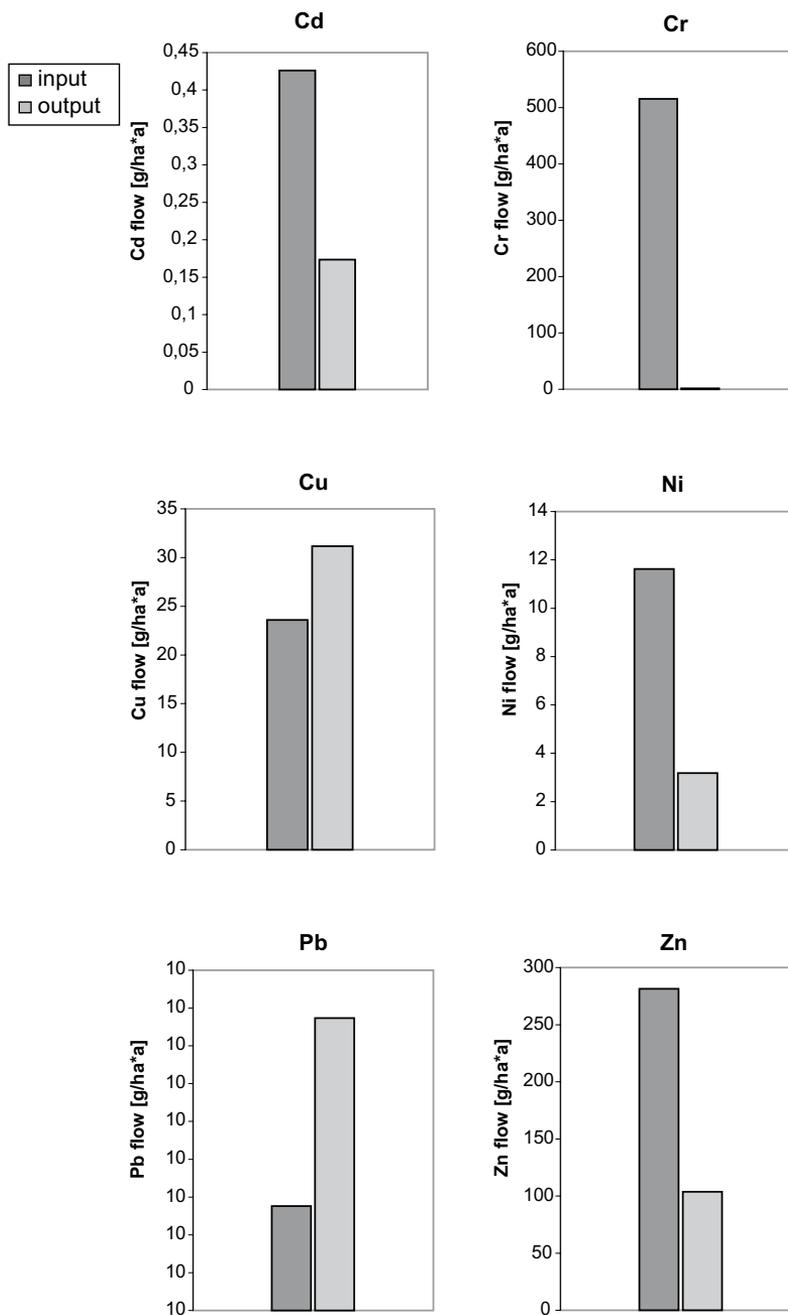
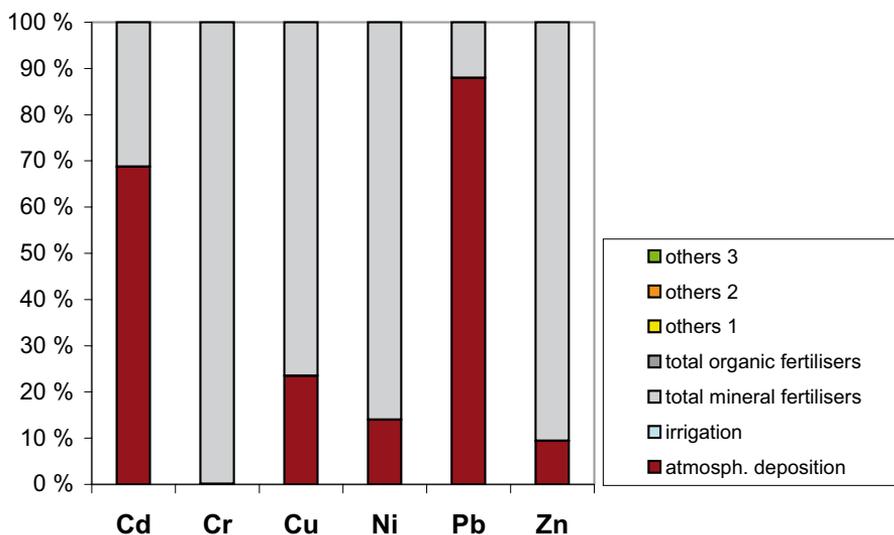


Figure 9. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 5 in southwestern Finland in 2004. Soil type river clay = RC. Steel slag was used on the farm in 2004.

### Heavy metal inputs



### Heavy metal outputs

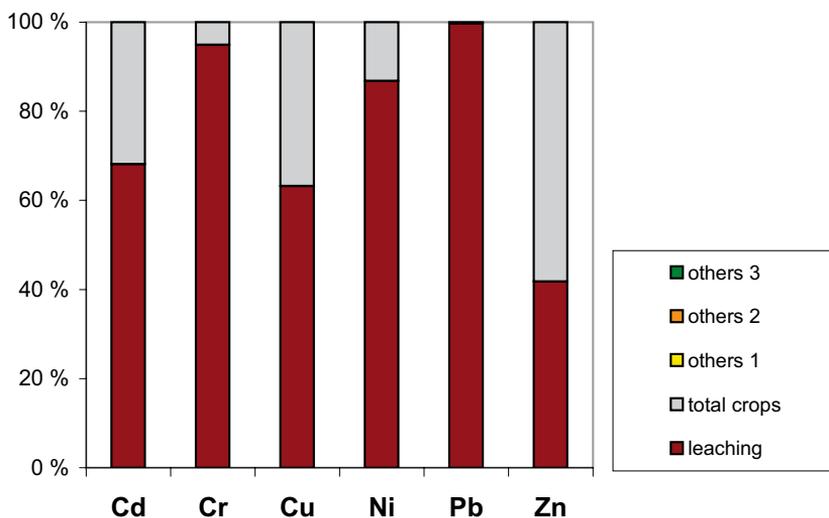


Figure 10. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for crop farm Nr. 5 in southwestern Finland in 2004. Soil type river clay = RC. Steel slag was used on the farm in 2004.

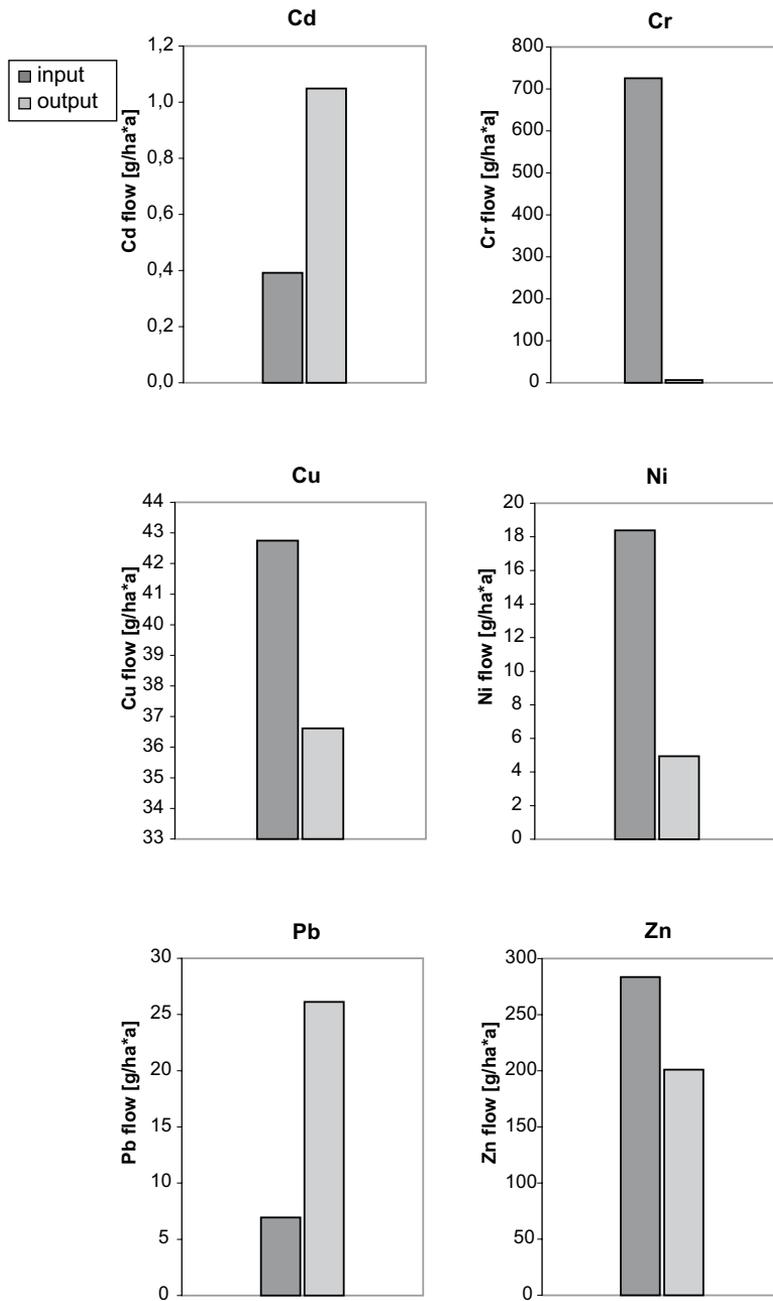
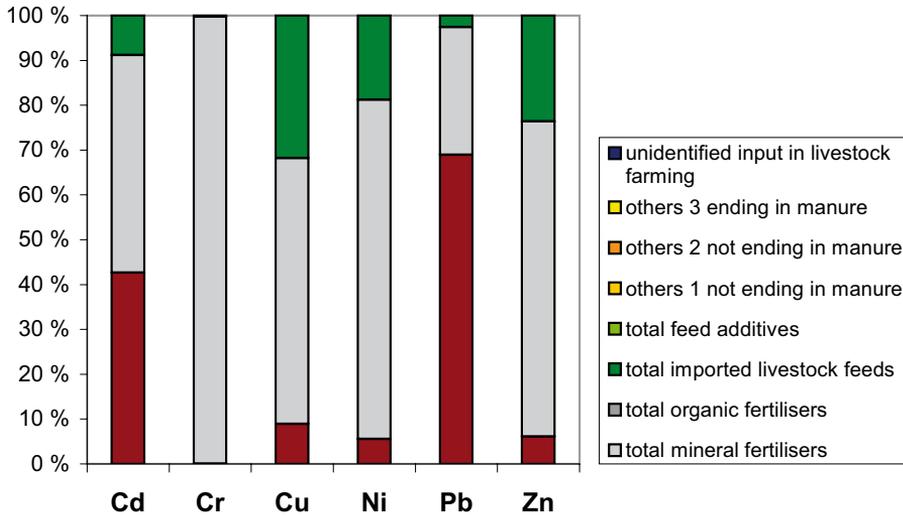


Figure 11. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 6 in Ostrobothnia in 2004. Soil type sand = S. Steel slag was used on the farm in 2004.

### Heavy metal inputs



### Heavy metal outputs

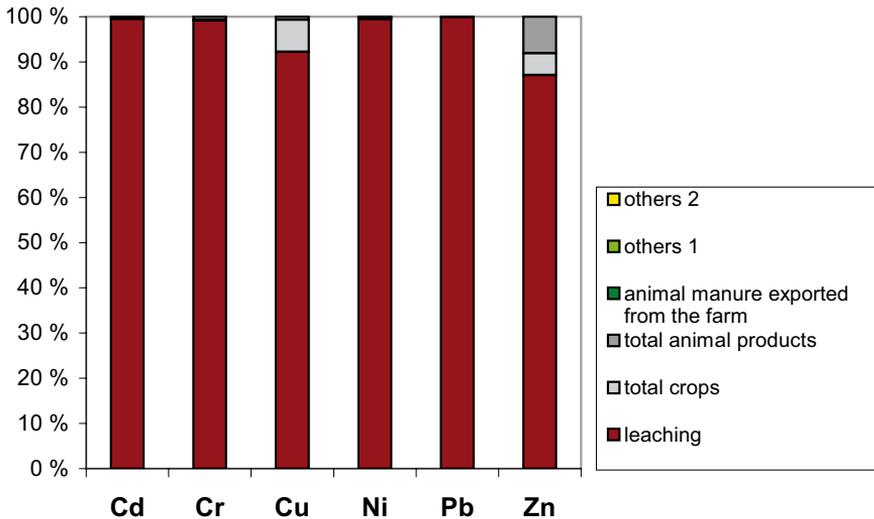


Figure 12. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 6 in Ostrobothnia in 2004. Soil type sand = S. Steel slag was used on the farm in 2004. In the graph of heavy metal inputs: ■ = atmospheric deposition.

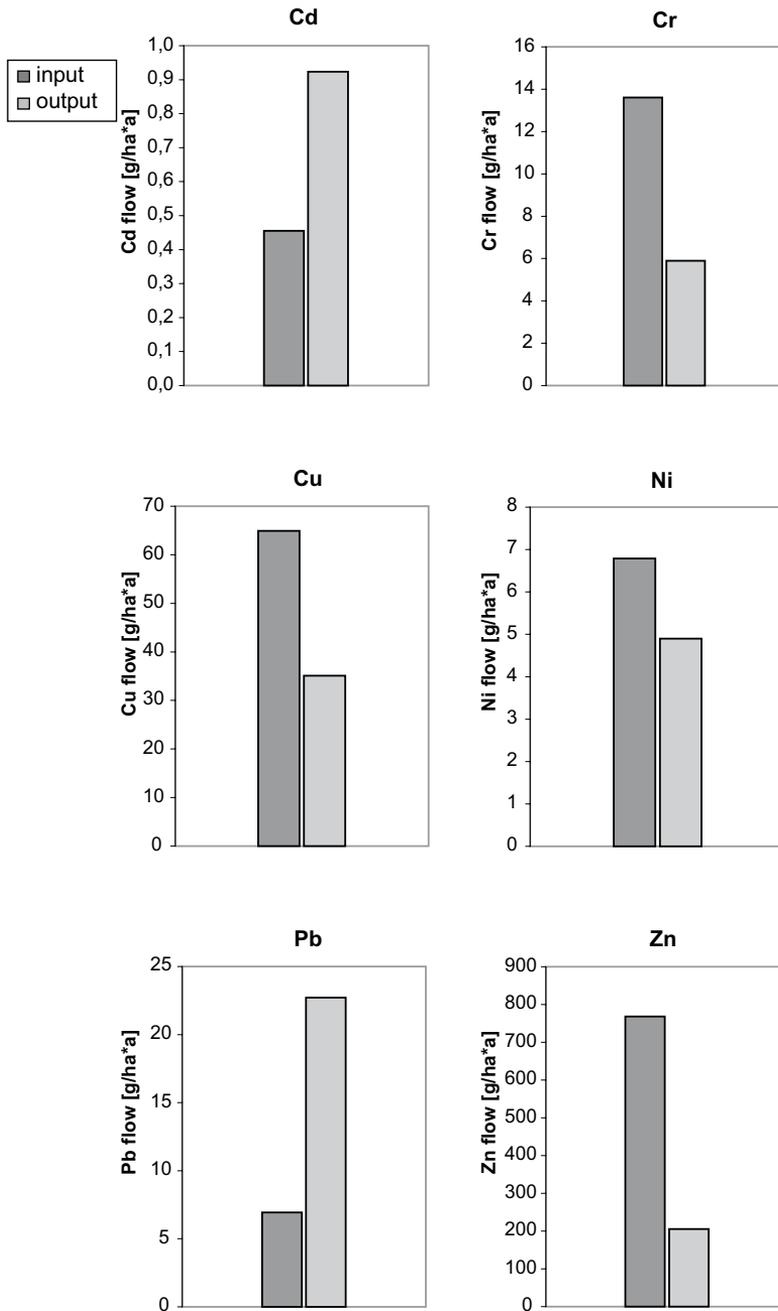
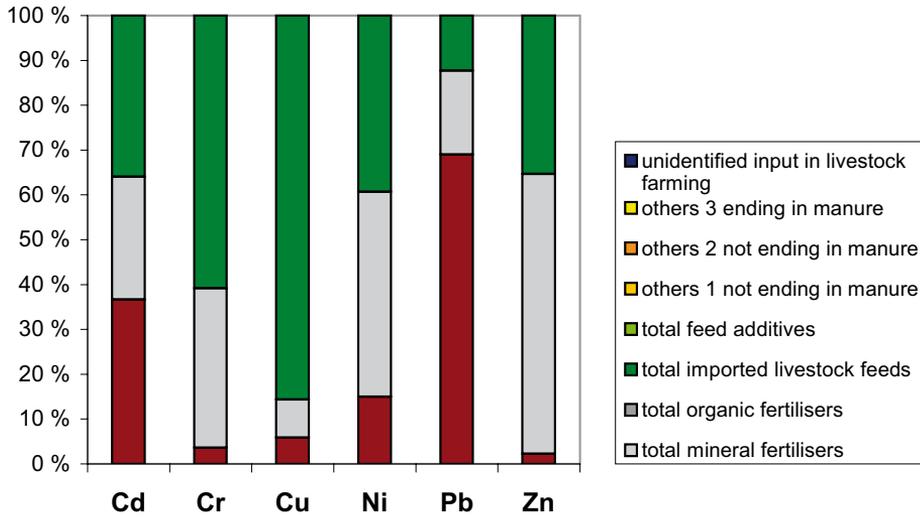


Figure 13. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 7 in Ostrobothnia in 2004. Soil type sand = S.

### Heavy metal inputs



### Heavy metal outputs

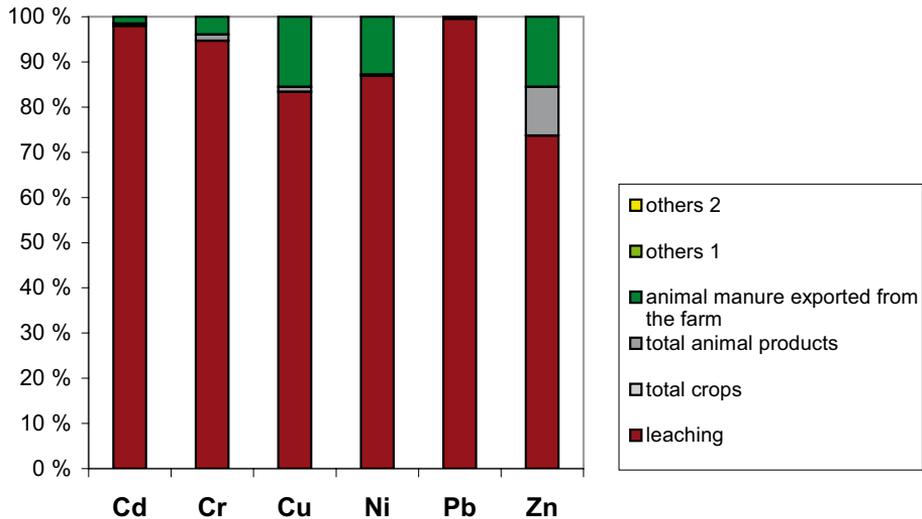


Figure 14. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 7 in Ostrobothnia in 2004. Soil type sand = S. In the graph of heavy metal inputs: ■ = atmospheric deposition.

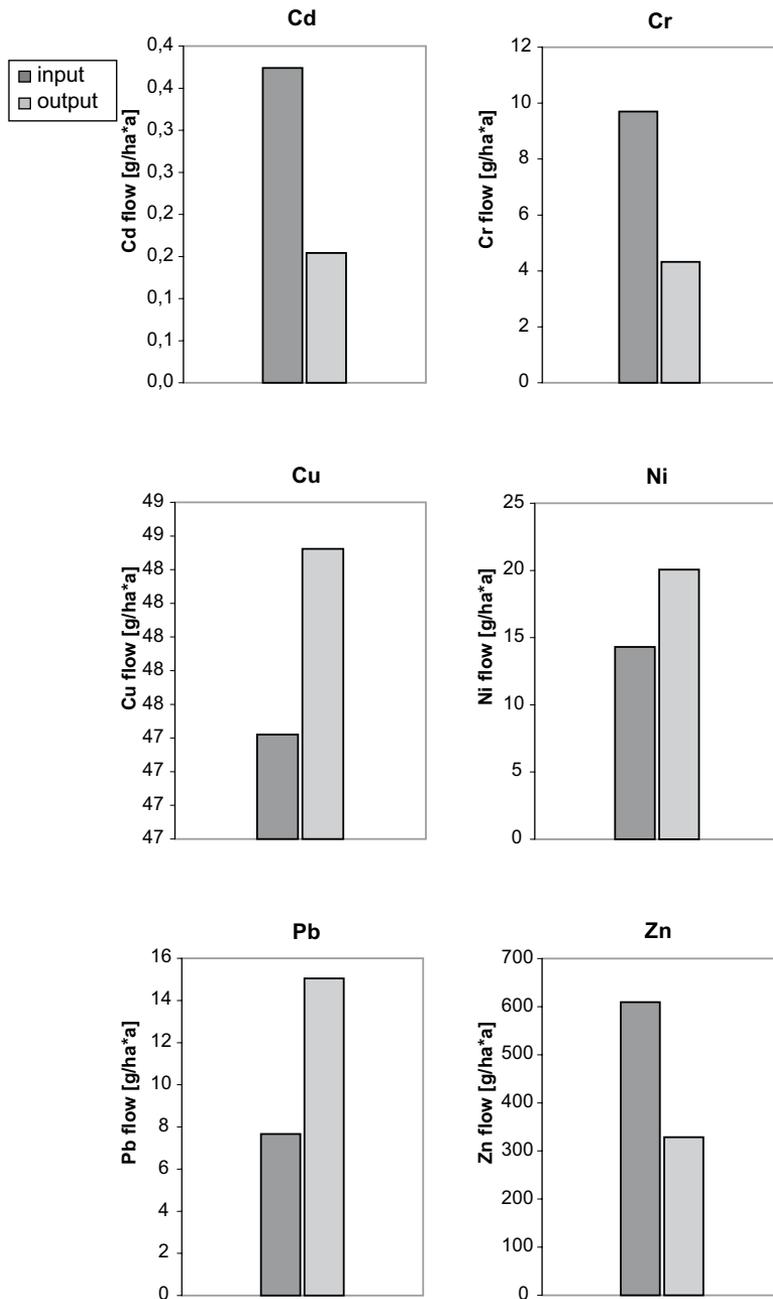
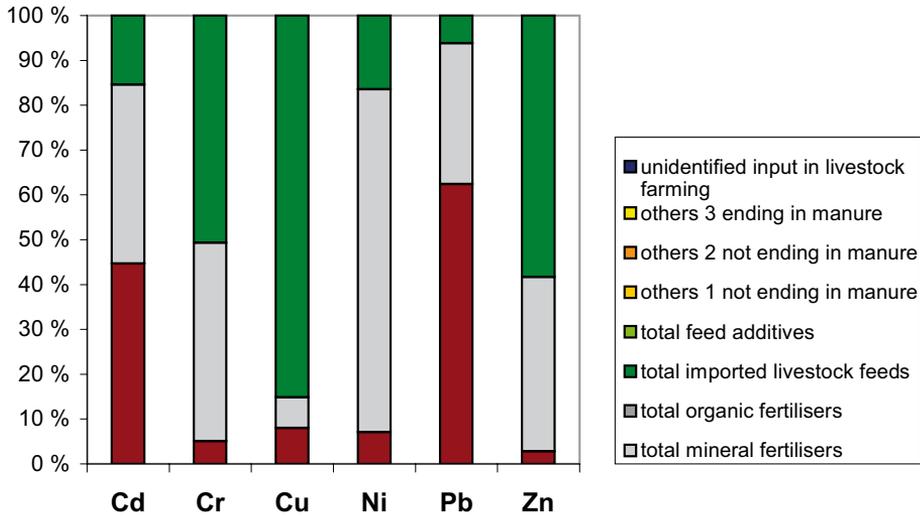


Figure 15. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 8 in Ostrobothnia in 2004. Soil type peat = P.

### Heavy metal inputs



### Heavy metal outputs

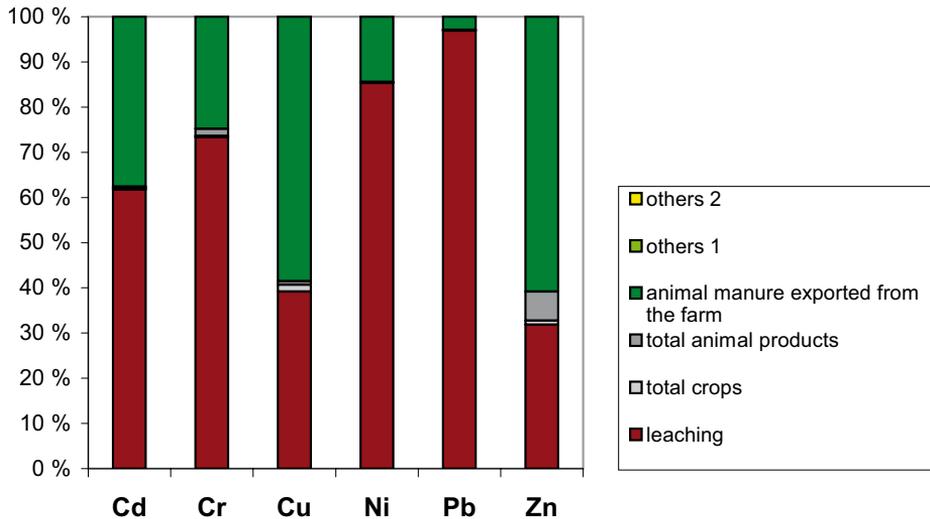


Figure 16. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 8 in Ostrobothnia in 2004. Soil type peat = P. In the graph of heavy metal inputs: ■ = atmospheric deposition.

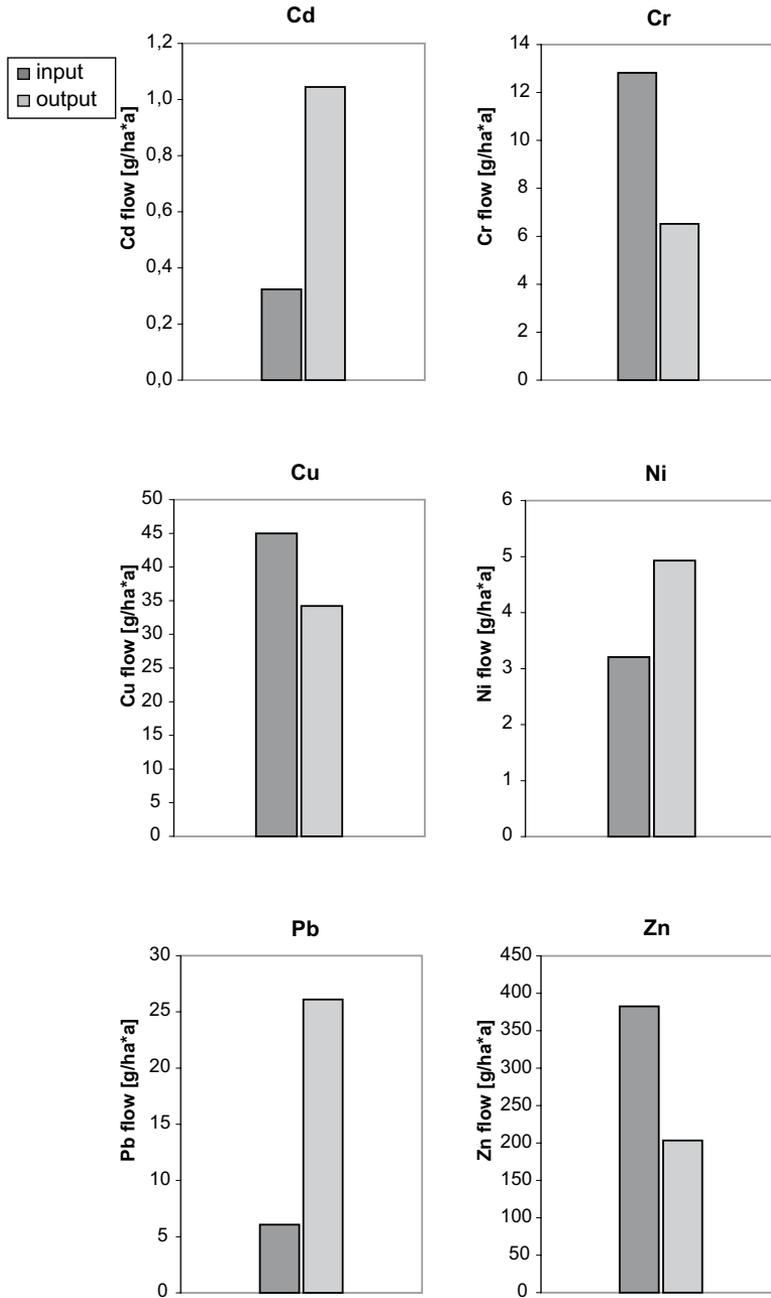


Figure 17. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 9 in Ostrobothnia in 2004. Soil type sand = S.

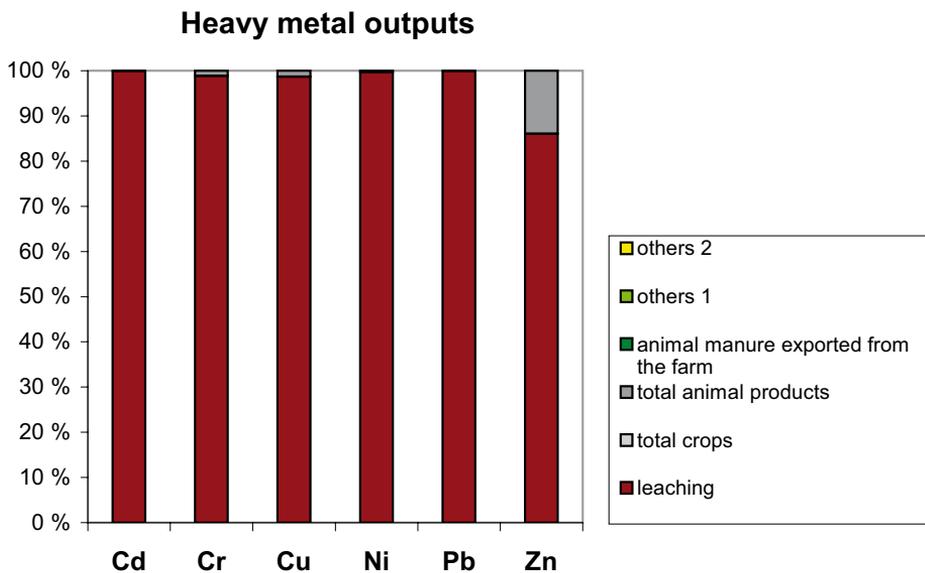
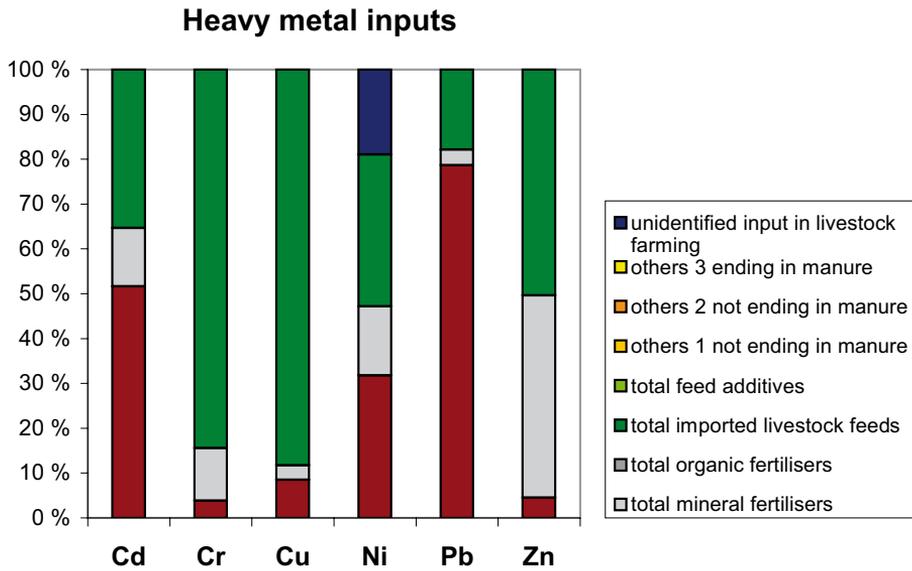


Figure 18. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 9 in Ostrobothnia in 2004. Soil type sand = S. In the graph of heavy metal inputs: ■ = atmospheric deposition.

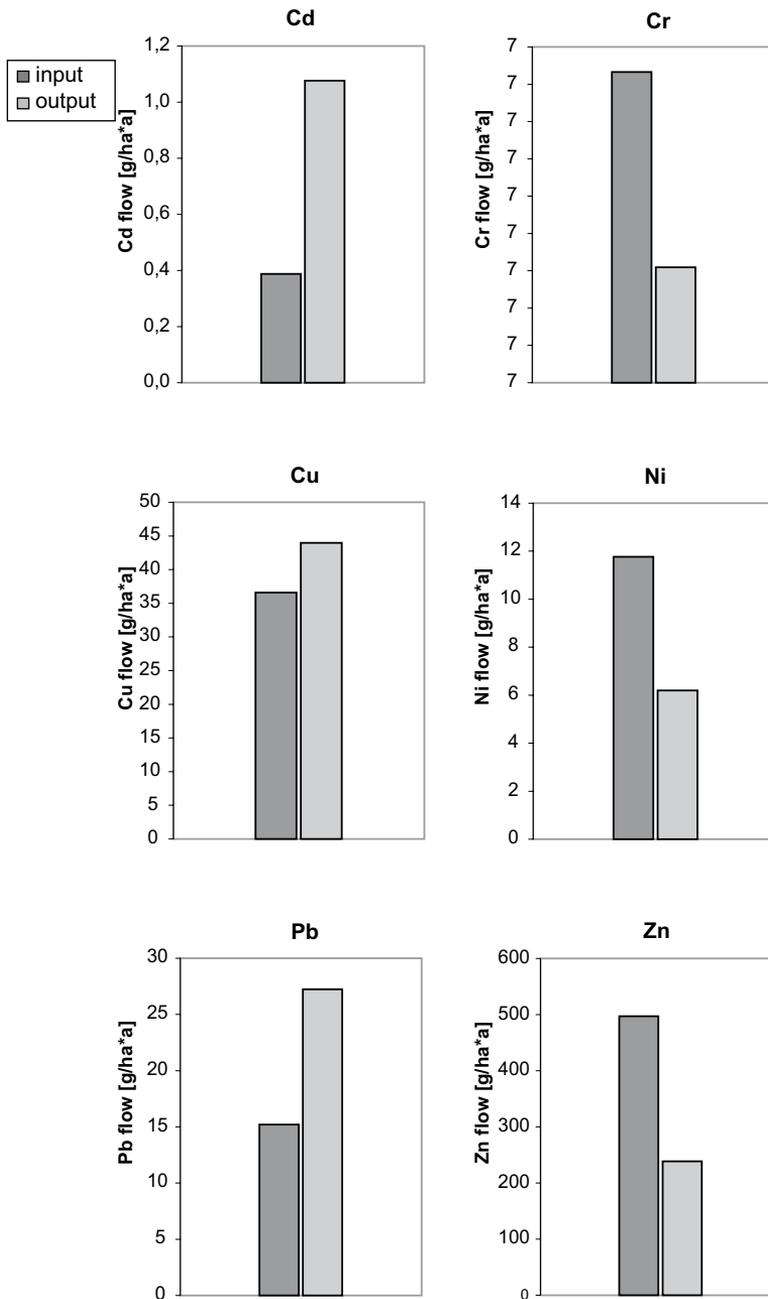
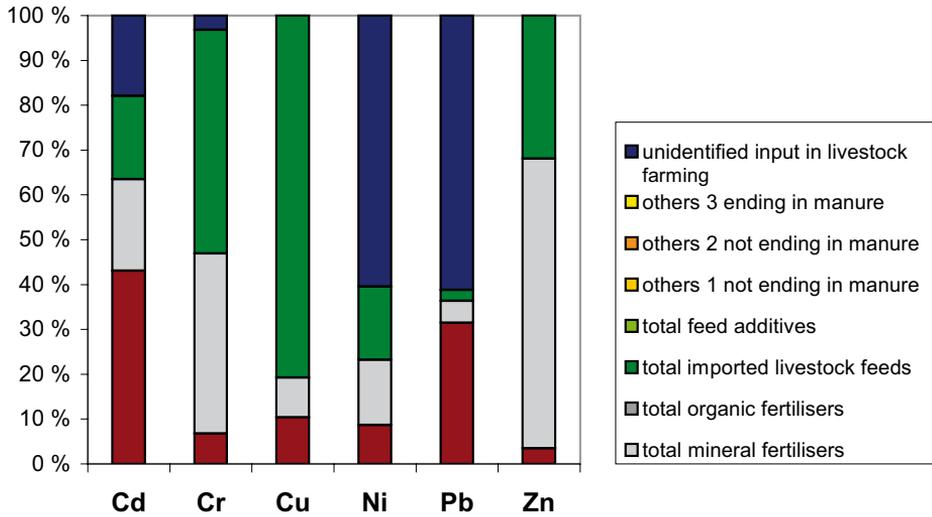


Figure 19. Total cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs to the soil and outputs from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 10 in Ostrobothnia in 2004. Soil type sand = S.

### Heavy metal inputs



### Heavy metal outputs

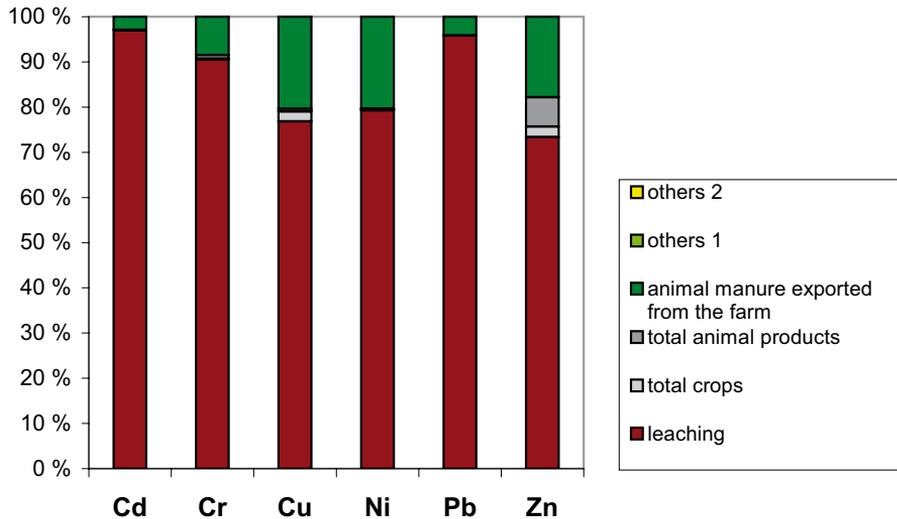


Figure 20. Percentages of cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) inputs from various sources to the soil and outputs through various routes from the soil at the farm level as calculated by the AROMIS-model for dairy farm Nr. 10 in Ostrobothnia in 2004. Soil type sand = S. In the graph of heavy metal inputs: ■ = atmospheric deposition.

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- 111 Field balances of trace elements at the farm level on crop and dairy farms in Finland in 2004. *Mäkelä-Kurtto, R.* ym. 131 s. Hinta 25 euroa.
- 108 Trace elements in top- and subsoil on selected crop and dairy farms in Finland in 2004. *Hatakka, T.* ym. 80 s. Hinta 20 euroa.
- 104 Monitoring programme of Finnish arable land. *Agua regia* extractable trace elements in cultivated soils in 1998. *Mäkelä-Kurtto, R.* ym. 61 s. Hinta 20 euroa.
- 96 Maatalous Itämeren rehevöittäjänä. *Uusitalo, R.* ym. 34 s. Hinta 15 euroa.

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- 66 Lyijy ja kadmium rohdos- ja yrttikasveissa. Kirjallisuuskatsaus. *Roitto, M. & Galambosi, B.* 103 s. Hinta 25 euroa.
- 65 Recept ur marknadsförarens kokbok: ingredienser och tillredningsanvisningar för en inbjudande lägerskola. Miljölägerskola Eco Learn. *Miemois, A.* 53 s. (webbpublikation: [www.mtt.fi/met/pdf/met65.pdf](http://www.mtt.fi/met/pdf/met65.pdf))

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