ELSEVIER

Ecological Economics 30 (1999) 47–65

ECOLOGICAL
ECONOMICS

ANALYSIS

Evaluation of risks of metal flows and accumulation in economy and environment

J.B. Guinée [a,*], J.C.J.M. van den Bergh [b], J. Boelens [c], P.J. Fraanje [d], G. Huppes [a], P.P.A.A.H. Kandelaars [b], Th.M. Lexmond [e], S.W. Moolenaar [e], A.A. Olsthoorn [c], H.A. Udo de Haes [a], E. Verkuijlen [d], E. van der Voet [a]

[a] *Centre of Environmental Science, Leiden University, P.O. Box 9518, 2300 RA Leiden, The Netherlands*
[b] *Department of Spatial Economics, Free University, De Boelelaan 1105, 1081 HV Amsterdam, The Netherlands*
[c] *Institute for Environmental Studies, Free University, De Boelelaan 1115, 1081 HV Amsterdam, The Netherlands*
[d] *Interfaculty Department of Environmental Science, University of Amsterdam, Nieuwe Prinsengracht 130, 1018 VZ Amsterdam, The Netherlands*
[e] *Department of Environmental Sciences, Sub-department of Soil Science and Plant Nutrition, Wageningen Agricultural University, P.O. Box 8005, 6700 EC Wageningen, The Netherlands*

Received 29 January 1998; received in revised form 30 June 1998; accepted 2 July 1998

Abstract

The contrast between, on the one hand, decreasing emissions of the metals cadmium, copper, lead and zinc and, on the other, their continuously increasing input into the economy is analysed for three case studies: The Netherlands as a whole, the Dutch housing sector and the Dutch agricultural sector. Flows of these metals through and their accumulation within the economy and the environment have been quantified for 1990 and for a constructed steady-state situation. To this end, the substance flow analysis method has been applied. The case studies show that there is a strong increase to be expected in the emissions from the 1990 to the steady-state situation. This increase is mainly due to the shift from landfill accumulation to emission to non-agricultural soil. At the same time, however, there is also an increase in the emissions to other media: air, water and agricultural soil. Emissions along these critical routes with respect to human and ecotoxicity show an approximately 30% increase for cadmium, lead and zinc and more than a doubling for copper. It is shown that this increase may lead to the surpassing of critical levels for human toxicity and terrestrial and aquatic ecotoxicity. Some possible measures are suggested to prevent critical levels being exceeded.

Keywords: Economy; Environment; Metals; Risks; Steady-state

* Corresponding author. Tel.: +31 71 5277432; fax: +31 71 5277434; e-mail: guinee@rulcml.leidenuniv.nl

PII: S0921-8009(98)00069-X

1. Introduction

Heavy metals are key issues in environmental policy and management. They pose risks to human health and ecosystems and may become scarce. In The Netherlands and several other industrialised countries, policy measures have been implemented in order to reduce these risks (OECD, 1993, 1994). In The Netherlands such measures successfully reduced emissions to air and water between 1985 and 1990 by 10–80% depending on the metal concerned (VROM, 1993). This downward trend, combined with a number of more recent policy measures like the phasing-out of leaded petrol, might indicate that heavy metals no longer pose an environmental problem.

However, primary production of most metals is still increasing (USBM, 1985, 1992, 1993), which will result in either higher emissions to water, air and soil or accumulation in the economy, i.e. in capital goods, intermediate products, consumer goods and wastes. In part, this accumulation is a consequence of closed-loop recycling (i.e. closed-loop accumulation[1]). If metals are accumulating in the economy, emissions may be only temporarily decreasing and may well increase in the future—e.g. through corrosion, inadequately controlled incineration, and landfill of municipal and industrial solid waste (cf. Ayres and Rod, 1986, Stigliani and Anderberg, 1994, Bergbäck and Lohm, 1997). Individual countries may also export these metals to foreign countries and thus shift the problem abroad.

Thus, the emission reductions mentioned above might be a temporary trend rather than a sustainable solution. The potential risks to human and ecological health due to high inputs of metals in the economy are still unclear: what will be the risks for human health and ecosystem health now or in the longer term if the present management regime is maintained? For this purpose, a scenario maintaining the present management regime is elaborated in this article for The Netherlands, and two sector cases are elaborated in more detail: the housing and agricultural sectors. Four metals have been selected on the basis of their inherent toxicity in various environmental media and their extensive use: cadmium, copper, lead and zinc. Steady-state modelling is used to estimate the magnitude of flows, stocks and risks in the long run.

In Section 2 a description of methods is given. In this article we take an industrial ecology approach (Ayres and Simonis, 1994, Socolow et al., 1994, Allenby, 1999), stressing the analogy between the mobilising, use and excretion of materials in the biosphere and the technosphere. Within the framework of this concept various methods and analytic tools have been developed (see, e.g., Bringezu et al., 1997). These comprise substance flow analysis (SFA), which is used as a framework for the models needed for the various calculations and for indicators to assess the current and future state of flows and accumulations in the economy and environment. Section 3 gives the results of the analysis of flows and accumulation in the economy and environment for The Netherlands as a whole. Section 4 presents the results of a more detailed analysis of copper flows and accumulation in the economy related to the housing sector, and Section 5 presents the results of a more detailed analysis of cadmium flows and accumulation in the environment due to the agricultural sector. Section 6 presents the main conclusions.

2. Methods: framework, models and indicators

The framework for the analysis is SFA, which deals with the analysis of flows and stocks of specific substances (cf. Baccini and Brunner, 1991, Baccini and Bader, 1996, Van der Voet, 1996). Within this framework a number of models were developed to calculate the magnitude of flows, accumulation and stocks in 1990 and at steady-state. Finally, the results of the model computations were aggregated into a number of policy-relevant indicators.

[1] An illustrative example of closed loop accumulation of metals concerns agriculture: reuse of organic (e.g. vegetable) refuse as fertiliser causes a flow of heavy metals back to agricultural soils, on top of the fresh metal input from industrial fertilisers, thus leading to rising metal concentrations in agricultural soil and crops.

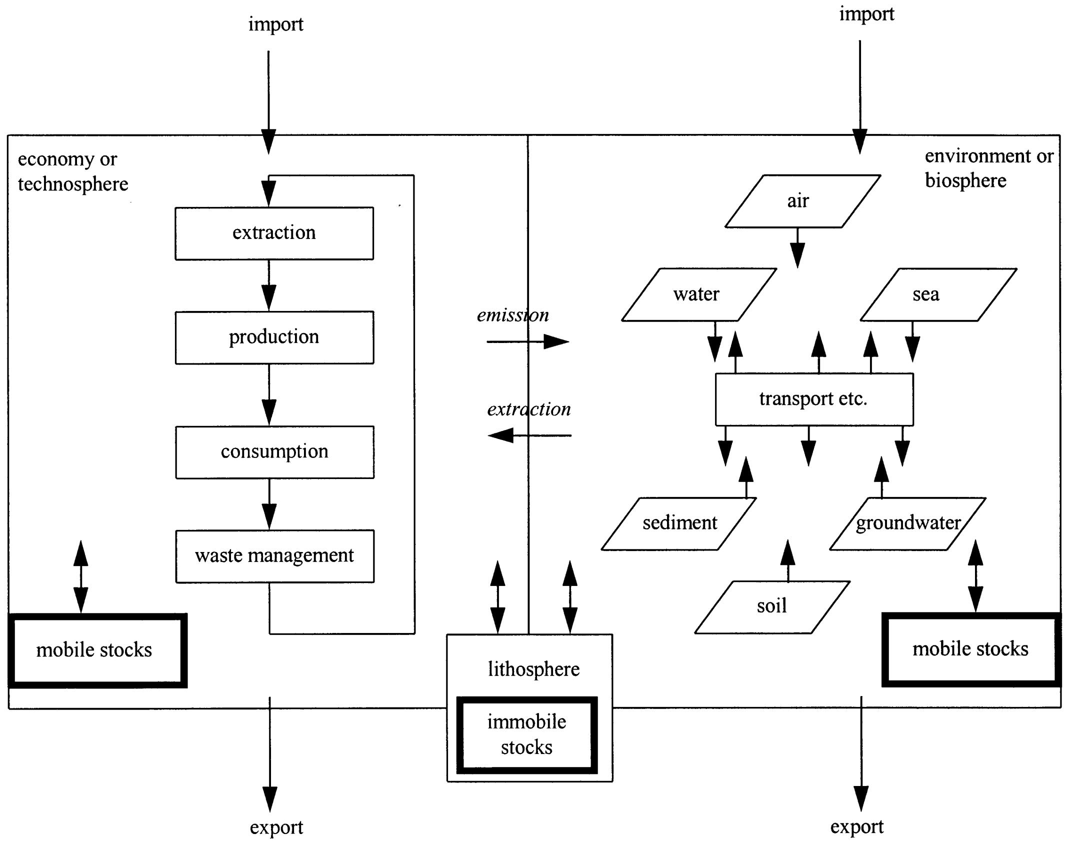

Fig. 1. Substance flows and stocks in economy, environment and lithosphere. Stocks of substances are depicted as bold squares, flows as arrows, environmental media as parallelograms and all stages in the economy as squares.

2.1. Framework

SFA describes the material or substance stocks within and flows within and between the economy and the environment in a certain time period and for a certain region. SFA is based on physical input–output analysis in which the materials-balance principle holds for each economic or environmental sector. Fig. 1 illustrates the framework. There are three subsystems: the economy or technosphere, the environment or biosphere, and the lithosphere, i.e. the ultimate source and sink of the metals. Stocks in the lithosphere are assumed to be immobile, while all stocks in the economy and environment are mobile.[2]

To analyse the structure of the economy the processes have been grouped into five stages: extraction and refining, covering both functional[3] and non-functional mining and refining of metal, as well as secondary recovery of metal scrap; production, which covers the production of both

[2] Mobile stocks are stocks that under the given conditions, on a time-scale of a few centuries (as opposed to geological time scales), can circulate within the system studied. Immobile stocks are just the opposite: they do not circulate within the (lithosphere) system, but there can be interactions with other systems (e.g. extraction).

[3] Functional flows include metals added to products or materials to fulfil a specific function in that product or material. Non-functional flows are the opposite of functional flows and include some inelastic inputs of metals (such as cadmium in zinc ore), contamination in materials (such as copper in bulk building materials) and also immobilised waste flows in materials (vitrification of metals in, e.g., slags).

functional and non-functional applications of metals (e.g. copper wire and cadmium in fertiliser, respectively); use, which includes, for example, storing soft drinks in aluminium cans and applying fertiliser in agriculture; waste management, which comprises collection, sorting, treatment and storage of metals in municipal and industrial waste flows.

2.2. Models, steady-state and dynamics

Three models have been used in the analysis: FLUX, DYNABOX and the dynamic balance (DB) model. FLUX describes the flows and accumulations within the economy. FLUX consists of a database of flows and accumulations of metals in The Netherlands, allowing the user to make data selection at different scales and with different system boundaries. FLUX does not model environmental processes such as actual intermedia transport, but it supports different output formats, enabling links to other models doing so, such as the generic environmental multi-media model DYNABOX (Heijungs et al., 1998). For further details, see Boelens and Olsthoorn (1998) and Olsthoorn and Boelens (1998).

DYNABOX builds on USES (RIVM et al., 1994), but has been adapted for use with metals and extended with a more detailed environmental metabolism for the soil medium. It models the flows and stocks (i.e. concentrations) of metals in the environment on the basis of the emissions calculated by FLUX. Thus, DYNABOX calculates, for example, concentrations in different environmental media such as air, water, agricultural soil, non-agricultural soil, sediment and groundwater, taking into account the relevant environmental processes such as partitioning, immobilisation, etc. For further details, see Heijungs et al. (1998).

For modelling cadmium in agricultural soils (see Section 5) a technically similar, but more refined, DB model was used; the emissions to agricultural soil as calculated by FLUX can also be used as input data for this model. The DB model calculates crop uptake, leaching from the topsoil and accumulation in the topsoil in relation to the metal content of the soil. For further details, see Moolenaar et al. (1997a).

Both DYNABOX and DB, which are conceptually very similar, can describe dynamic developments as well as steady-state situations.[4] The dynamic module of DYNABOX and DB is used to calculate the time that elapses, given constant emission inputs, before an average concentration of a substance in some environmental medium reaches a critical level (cf. Moolenaar et al., 1997b). This time is called the transition period.

Steady-state is an important concept in this article. The steady-state situation of, for example, the 1990 flows and stocks of a substance indicates the eventual magnitude of these flows required to maintain the 1990 regime indefinitely. The outcome of this type of modelling provides an assessment of the long-term sustainability of a certain substance regime compared to the present situation. It is not a prediction, since it obviously includes many uncertainties (see also Hansen and Lassen, 1997).

In FLUX the steady-state is calculated as follows. The first step is bookkeeping: gathering all written information on the flows and stocks of a given substance for a given year. To an extent, this information will be inconsistent; choices then have to be made as to which information is the most reliable. Based on this bookkeeping and the reliability information, a static model is drafted by applying a balancing procedure (Boelens and Olsthoorn, 1998). Subsequently, coefficients are deduced from the balancing results in order to derive a static model by determining how outputs depend on inputs or stocks for a specific process. At steady-state the accumulation is zero and the magnitudes of the flows and stocks considered are calculated for which, given a set of linear equations, OUT equals IN. The assumptions concerning the relationships between the flows are the same as those used in Leontief-type input–output modelling (e.g. the fixed-technology assumption). An extra assumption is that the demand for metal(-containing) flows remains constant

[4] The assumptions for dynamic modelling of the environment are less strong than for modelling the economy. In contrast to the residence times of economic goods, environmental residence times (or flow-rate constants) are physical parameters that are by definition constant over time.

(e.g. the production of apartments or the use of phosphate fertiliser); the resource stocks are thus considered unlimited.

In mathematical terms, the steady-state calculation can be described as follows. Consider a single node with an input flow I, a stock $N(t)$, and an output flow $O(t)$. $O(t)$ is proportional to N ($O = kN(t)$), where k is a rate constant that can be considered as the reciprocal lifetime (of the substance in the node). As $O(t)$ is not necessarily equal to I, $N(t)$ will change over time. After writing the mass balance as a first-order differential equation:

$$\frac{dN}{dt} = I - kN(t)$$

and integrating, it follows, assuming I is constant, that:

$$N(t) = e^{-kt}\left(N(0) - \frac{I}{k}\right) + \frac{I}{k}$$

where $N(0)$ is the initial stock.

After a long or even infinite time, N will no longer change: an equilibrium is reached whereby $N(\infty) = I/k$. This equilibrium is called the steady-state. For complex systems it is similarly possible to calculate the behaviour of stocks and flows over time given a set of constant inputs. For more details, see Van der Voet (1996) and Olsthoorn and Boelens (1998).

2.3. Indicators

For interpreting the results of the substance flow analyses, three types of policy-relevant indicators are used (Van der Voet, 1996 see also Ayres, 1996, Azar et al., 1996, Wernick and Ausubel, 1995):

- Environment indicators, which interpret the state of, or processes within, the environment.
- Indicators on emissions and export, which summarise the burdens on the environment and on other countries.
- Economy indicators, which show the backgrounds of emissions and can help find solutions.

The definition, use and simplified equations of these indicators are described briefly below; for a more elaborate description and for detailed equations, see Van der Voet et al. (1997).

2.3.1. Environment indicators

2.3.1.1. Relative environmental accumulation (1). The relative environmental accumulation (Q') is the increase of the environmental stocks in a given year (Q) divided by the total flows into the environment in that year (I), i.e. environmental import, emissions and mobilisation, expressed in %:

$$Q'\,(\%) = \frac{Q\ \text{(mass/year)}}{I\ \text{(mass/year)}} \times 100$$

It indicates to what degree the environment is off-balance under the current regime, and, apart from current risks, a growing risk because of rising concentrations in the future.

2.3.1.2. Risk ratio (2a) and transition period (2b). The risk ratio (dimensionless) is calculated for human toxicity and aquatic and terrestrial ecotoxicity, being the daily intake or the environmental concentration in a medium divided by the acceptable/tolerable daily intake or concentration standard, respectively, for that medium. It indicates the potential risk (values > 1) emissions pose to human and ecosystem health. This indicator can be calculated for the 1990 situation by using empirically measured concentrations, and for the steady-state situation by using DYNABOX (Section 3) and DB (Section 5).

The transition period is the time it takes for the ratio to equal 1. This indicator is only applicable for the steady-state situation.

2.3.2. Indicators on emissions and export

2.3.2.1. Total emissions (3). The indicator gives the aggregate emissions (mass/year) from the economy to the different environmental media (air, water, agricultural and non-agricultural soil). It indicates environmental pressure, and it is an early warning for the steady-state risk ratios. It may be compared with emission targets for each medium.

2.3.2.2. Pollution export (4). Pollution export (P) stands for the total emissions (E_W) occurring both inside and outside the region on behalf of demand within the region, minus the total emissions within the region (E_R):

$$P \text{ (mass/year)} = E_W \text{ (mass/year)} - E_R \text{ (mass/year)}$$

Production of metals and metal-containing products for foreign demand is thus a positive contributor and extraction of metals taking place in foreign countries for Dutch demand is a negative one. It indicates the amount of environmental pressure due to regional demand that occurs elsewhere in the world, and is therefore a measure of problem-shifting to other regions. If the indicator is positive, the region exports pollution; if it is negative, it imports pollution.

2.3.3. Economy indicators

Technical efficiency and recycling rate are calculated per economic stage and economic accumulation for the economy as a whole; the technical efficiency and recycling rate are calculated for both 1990 and steady-state, while the accumulation in the economy is calculated only for 1990, since accumulation at steady-state is by definition equal to 0.

2.3.3.1. Technical efficiency (5). The technical efficiency of an economic stage (T_s) is the useful outflow (thus excluding flows to landfill and emissions) from stage s (O_s) to a following economic stage divided by the total flows into stage s (I_s), expressed in %:

$$T_s\ (\%) = \frac{O_s \text{ (mass/year)}}{I_s \text{ (mass/year)}} \times 100$$

The higher the technical efficiency the better. An efficiency of 100% is environmentally the most preferable and may serve as a reference value to strive for. Low rates indicate options for improvement.

2.3.3.2. Functional recycling rate (6a) and non-functional recycling rate (6b). The functional recycling rate of an economic stage (F_s) is the amount of metal that is functionally recycled from stage s (O_s) to an earlier life-cycle stage within The Netherlands or in foreign countries, divided by the total output of stage s (O_s):

$$F_s\ (\%) = \frac{F_s \text{ (mass/year)}}{O_s \text{ (mass/year)}} \times 100$$

Recycling is not a goal in itself and it is difficult to determine an optimal recycling rate (cf. Van der Voet et al., 1994, Instituut voor Europees Milieubeleid and Environmental Resources Management, 1996, Guinée et al., 1997), but for substances with an elastic supply it generally holds that the higher the recycling rate, the better it is for the environment.

The non-functional recycling rate of an economic stage s (N_s) is the amount of metal that is non-functionally recycled (i.e. without any intention to recycle the metal in itself) to an earlier life-cycle stage (within or outside The Netherlands), divided by the total output of stage s (O_s):

$$N_s\ (\%) = \frac{N_s \text{ (mass/year)}}{O_s \text{ (mass/year)}} \times 100$$

2.3.3.3. Relative accumulation in the economy (7). The relative economic accumulation (A') is the mass increase of stocks in the economy in a given year (A) divided by the mass total inflow into the economy in a given year (I), i.e. economic import, extraction and mobilisation, expressed in %:

$$A'\ (\%) = \frac{A \text{ (mass/year)}}{I \text{ (mass/year)}} \times 100$$

It indicates the risk of a future increase in the generation of waste and emissions and may thus function as a warning signal for future environmental problems.

The indicators can be applied flexibly with respect to the definition of landfill and emissions. Flows to landfill can be seen either as emissions to soil or as flows to (mobile) stocks in the economy, with emissions as leakage from the landfill. Here, flows to landfill are regarded as flows to stocks in the economy; in the economy indicators these flows are treated, ultimately, as unavoidable leakage to the environment without any useful economic function. Similarly, flows from top soil to deeper soil may be treated as immobilisation when assessing risks from agricultural soils, or as

Table 1
Values of environment indicators for cadmium, copper, lead and zinc in The Netherlands, for 1990 and the steady-state (SS)

Indicators	Cadmium		Copper		Lead		Zinc	
	1990	SS	1990	SS	1990	SS	1990	SS
1. Relative environmental accumulation (%)	22	0	64	0	27	0	57	0
2a. Risk ratio								
Human toxicity	0.3	0.7	0.2	2.0	3.2	41	0.8	2.2
Aquatic ecotoxicity								
Concentration ÷ reference value	8	20	0.4	157	0.8	74	1.4	55
Concentration ÷ limit value	1.3	3.3	0.3	12		0.1	11	0.4
Concentration ÷ MPC	0.2	0.5	0.3	105	0.0	1.4	0.3	11
Terrestrial ecotoxicity								
Concentration ÷ reference value	0.7	1.6	0.7	7.9	0.5	6.9	0.7	3.1
Concentration ÷ limit value	0.0	0.1	0.1	1.7	0.1	1.2	0.1	0.6
Concentration ÷ MPC	0.3	0.7	0.6	7.1	0.3	4.2	0.6	2.7
2b. Transition period (years)								
Human toxicity		∞		460		0		130
Aquatic ecotoxicity								
Concentration ÷ reference value		0		2		13		0
Concentration ÷ limit value		0		3		90		12
Concentration ÷ MPC		∞		3		1000		16
Terrestrial ecotoxicity								
Concentration ÷ reference value		200		19		200		90
Concentration ÷ limit value		∞		720		80000		∞
Concentration ÷ MPC		∞		30		550		120

flows within the environment when analysing the ultimate sinks of the system. Here, part of the soil and the sediment—the deep soil (below 0.20 m) and the deep sediment (below 0.03 m)—are regarded as immobile sinks for metals.

The indicators as defined above form the kernel of the following presentation of results.

3. Metals in The Netherlands

In this section the results of a case study on the environmental risks relating to the metabolism of cadmium, copper, lead and zinc for the total Dutch economy are presented. The situation for 1990 is compared with the steady-state situation calculated with FLUX. Input data for 1990 such as data on flows in the economy, accumulations, emissions and transboundary pollution have been taken mainly from Annema et al. (1995). The economy indicators and the environmental accumulation for 1990 have been calculated by simple spreadsheet manipulations of FLUX results. The risk indicators have been calculated using the multi-media model DYNABOX (Heijungs et al., 1998). All the indicators discussed in Section 2 are applied in this case study. The results for 1990 and for the steady-state situation are presented below per group of indicators.

3.1. Environment indicators (indicators 1, 2a and 2b)

The results for the relative environmental accumulation (Table 1) show that about 50% of the environmental inflow of copper and zinc and about 25% of the inflow of cadmium and lead accumulated in the environment in 1990.

Table 1 shows the risk ratios for human toxicity and aquatic and terrestrial ecotoxicity. Acceptable daily intake (ADI) values defined by the World Health Organization and tolerable daily intake

Table 2
Concentrations in water and soil as used in the calculation of the risk ratios

	Cadmium		Copper		Lead		Zinc	
	1990	SS	1990	SS	1990	SS	1990	SS
Water concentration (μg/l)	0.08	0.2	0.4	157	0.2	15	2.8	101
Soil concentration (mg/kg dry weight)	0.6	1.3	26	328	38	624	88	442

(TDI) values similarly defined by Vermeire et al. (1991) and Cleven et al. (1992) have been applied in calculating the risk ratio for human toxicity. Three different standards have been applied in calculating the risk ratio for aquatic and terrestrial ecotoxicity: the Dutch reference value, the Dutch limit value and the Dutch maximum permissible concentration (MPC). The reference value is the concentration at which the risk of adverse ecotoxic effects is considered negligible; it indicates the final quality level to be reached and it does not take into account the natural background concentration. The limit value is the concentration of a chemical in a medium which should not be exceeded; it indicates the short-term ecotoxic quality level to be reached. Reference and limit values are policy standards. The MPC is defined as the sum of the maximum permissible addition (MPA) and the existing background concentration in The Netherlands, with the MPA defined as the amount of a metal originating from antropogenic sources that is allowed on top of the natural background concentration. The MPC is an ecotoxicological value.[5]

Reference and limit values are taken from Janus et al. (1994); MPC values are taken from Crommentuijn et al. (1997); background concentrations are taken from Crommentuijn et al. (1997) and Van Drecht et al. (1996). All standards are used as risk indicators; it has not been analysed which exposure levels and effects are actually found at the concentrations calculated.

The concentrations in water and soil used for calculation of the risk ratios are shown in Table 2. It appears that several risk ratios are expected to be above 1, depending on the type of environmental standard applied. For human toxicity the risk ratio of lead is already above 1 for 1990 and the risk ratio for the steady-state situation is above 1 for lead, zinc and copper, in decreasing order of magnitude. For aquatic and terrestrial ecotoxicity copper gives the highest risk ratios, then lead and zinc, and then cadmium. These results mean that the current metabolism of these metals is generally not sustainable.

The transition periods for the various metals are also shown in Table 1. In calculating the transition periods, current background levels in the various environmental media have been taken into due account. The transition periods vary from 0 years for cadmium in water to reach the reference and the limit value, to 8000 years for lead in soil to reach the limit value.[6] The results for soil have been compared with the results of the more sophisticated DB model for cadmium (see Section 5) and appear to be fairly similar. The results for human toxicity and aquatic ecotoxicity could not be compared with any other model. For aquatic ecotoxicity the risk ratios for the steady-state situation (note: based on the steady-state emissions as calculated by FLUX and not on the 1990 emissions!) seem quite high. This may be due to uncertainties attached to the partitioning coefficients used and the solubility value used, which is different for different metal species.

[5] In The Netherlands a discussion is being held on the derivation of ecotoxicological values for zinc, also taking into account the essential meaning of zinc for human and other life. The discussion may result in new ecotoxicological values, but these have not yet been proposed (Gezondheidsraad, 1998).

[6] The transition periods are calculated by DYNABOX and based on the steady-state emissions presented in Table 1. Note, however, that FLUX and DYNABOX are separate and not integrated models, which makes consistent analysis of the transition periods in relation to the flows in the economy unfeasible at present.

Table 3
Values of economy–environment indicators for cadmium, copper, lead and zinc in The Netherlands, for 1990 and the steady-state (SS)

Indicators	Cadmium		Copper		Lead		Zinc	
	1990	SS	1990	SS	1990	SS	1990	SS
3. Total emissions	17.1	148.9	1203	69044	1136	52716	5192	73060
Air	4.2	5.5	52	93	152	156	183	160
Water	4.8	5.0	152	477	185	191	617	1221
Agricultural soil	6.2	9.4	987	1914	266	510	1992	3124
Non-agricultural soil	1.9	129.1	12	66560	533	51859	2400	68555
Landfill	0.04	127.1	0.04	66521	0.12	51230	2	61276
Other sources	1.9	2.0	12	39	533	559	2398	7278
4. Pollution export	−10.0	−14.3	−5086	−7665	n.a.	n.a.	−6644	−7434

Values are given in tonnes/year. n.a. = not applicable.

3.2. Indicators on emissions and exports (indicator 3 and 4)

The results for the emission indicators (Table 3) show, for almost all media and all metals, an increase of emissions in the steady-state situation compared to the 1990 situation. The increase of air emissions in the steady-state situation compared to the 1990 situation is generally moderate. The increase for cadmium is caused by the incineration of spent NiCad batteries; the increase for copper is due to overhead railway wires. For zinc, air emissions for the steady-state compared to 1990 decrease, since the amount of zinc in galvanised iron is decreasing. Besides emissions, transboundary pollution via air from foreign countries is an important source for the total input to air for all four metals; this source is not included in the emissions indicator, however.

For all four metals, the increase of water emissions in the steady-state situation compared to the 1990 situation is due mainly to the corrosion of metals in building materials (e.g. zinc gutters, galvanised steel, tapwater heating equipment and bulk materials such as concrete). However, with respect to the total input to water, it is not emissions within The Netherlands but inflow of metals from outside The Netherlands via rivers like the Rhine and Meuse that constitute the dominant source for all four metals (up to over 70%).

The increase of steady-state emissions to agricultural soils compared to 1990 emissions is significant for all metals and is due to increasing flows of organic manure and of source-separated vegetable, fruit and garden waste (the latter being less relevant for lead). The ultimate source behind these increasing flows of copper and zinc is animal fodder. It appears that in the steady-state situation the agricultural soil emissions of copper and zinc are due overwhelmingly (about 80–90%) to the addition to fodder of copper and zinc, respectively. This is an example of closed-loop accumulation: copper and zinc are added to fodder, which is imported from abroad and fed to Dutch cattle. The manure produced by the cattle, including its copper and zinc content, is spread on agricultural land as an organic fertiliser. Soil concentrations of copper and zinc consequently rise and, with them, the copper and zinc concentrations in maize, pit grass, fresh grass and hay. The livestock are additionally fed with maize, pit grass, fresh grass and hay, and the metals are thus returned to the economy. The eventual steady-state soil concentration due to this cycling of copper and zinc leads to several risk ratios above 1.

The increase of steady-state emissions compared to 1990 emissions is most eye-catching for non-agricultural soil. For all metals, the increase of emissions at steady-state is completely dominated by emissions from landfill sites, which are

Table 4
Values of economy indicators for cadmium, copper, lead and zinc in The Netherlands, for 1990 and the steady-state (SS)

Indicators	Cadmium		Copper		Lead		Zinc	
	1990	SS	1990	SS	1990	SS	1990	SS
5. Technical efficiency (%)								
Extraction	98	98	99	99	3	3	98	98
Production	97	93	99	99	100	100	97	96
Use	98	83	100	96	99	91	95	79
Waste management	87	83	88	85	95	95	91	94
6a. Functional recycling rate (%)								
Extraction	0	0	0	0	0	0	0	0
Production	0	0	0	0	0	0	7	7
Use	0	0	0	0	0	0	0	0
Waste management	47	66	85	80	94	93	88	92
6b. Non-functional recycling rate (%)								
Extraction	0	0	0	0	0	0	0	0
Production	0	0	0	0	0	0	0	0
Use	0	0	0	0	0	0	0	0
Waste management	34	17	3	5	1	1	3	2
7. Economy accumulation ÷ total economy inflow (%)	12	0	14	0	7	0	11	0

assumed to be quite low in 1990. In a steady-state the outflow equals the inflow. Since it is assumed that emissions to non-agricultural soil are the only outflow from a landfill, the emission to non-agricultural soil at steady-state will equal the inflow at steady-state. In the end any leakage to the environment from a waste storage site will lead to a non-sustainable situation, but the time this will take may be very long (up to thousands of years). For the risk ratios, however, emissions to non-agricultural soil make a contribution of only about 10% and are thus not a major source, even in the steady-state situation.

The results for the pollution export indicator in Table 3 indicate that The Netherlands is, and will remain, a net importer of pollution for cadmium, copper and zinc. This means there is no net shifting of problems to other countries. For lead this indicator is not useful, since it only gives useful information if the economic processes in the region are more or less representative of the average economic processes in the world. The latter is not the case for lead extraction and refining processes in The Netherlands (see below).

3.3. Economy indicators (indicators 5, 6a, 6b and 7)

The results on technical efficiency (Table 4) show that the efficiency of the extraction and production stages is generally high. This indicates that, in order to prevent emissions, not much can be gained by a further boost of industrial efficiency. An exception is the extraction stage of lead (3%).[7] Comparing the steady-state efficiencies to the 1990 efficiencies, the decrease in use and waste-management efficiencies—due, for example, to corrosion of asphalt, cement and concrete in (utility) buildings, overhead rail wires, cement

[7] This is due to the definition of extraction, which includes non-functional extraction and refining, and the fact that no primary extraction of metals takes place in The Netherlands. The non-functional lead flow, which is separated from the iron and disposed of in a landfill during the process of iron refining, is also included in this indicator. Thus, the low extraction efficiency value indicates the potential for useful application of this lead flow.

and landfill emissions—is the most eye-catching result for all metals.

Table 4 shows that functional recycling takes place mainly in the waste-management stage; to a large extent, this is due to the definition of this stage, which is taken to include collection and storage of waste flows. For copper, lead and zinc the recycling rate is determined largely by the recycling of building materials, and for cadmium by the recycling of various types of NiCad batteries. Furthermore, Table 4 shows that non-functional recycling is highest for cadmium and lowest for zinc. Note that the efficiency of the waste-management stage is determined by the functional and non-functional recycling rate and by the accumulation rate (which, by definition, is 0 for the steady-state situation).

The results for the relative accumulation in the economy show that it ranges between 7 and 14%, being highest for copper and lowest for lead. An indication of the time it will take to reach the steady-state situation in the economy can be obtained from the lifespans of the products and applications involved. For example, the average lifespan of functional applications as building materials lies somewhere between 30 and 50 years, while for non-functional flows of metals in bulk building materials such as concrete this may be over 100 years. Thus, 100 years seems a reasonable estimate of the time it will take to reach steady-state for the metal flows in the economy.

It should be noted, however, that all steady-state indicators presented in this section are based on 1990 data and do not take into account any effects of policy measures taken since. For example, the decrease of lead in fuel and the decrease of cadmium in zinc gutters have not been taken into account in the current steady-state results. The use of copper and zinc in fodder has also been reduced since 1990, but this has been neutralised by an increase of the Dutch pig stock, resulting in a higher flow of copper and an equal flow of zinc in fodder in 1994 (Westhoek et al., 1997) compared with 1990. The closed-loop accumulation example of copper and zinc in fodder is thus still valid.

3.4. Conclusions

From this case study the following conclusions can be drawn:

- The 1990 flows and accumulations of cadmium, copper, lead and zinc pose significant long-term risks to human health and ecosystem health.
- For all metals, the built environment, agriculture and landfills are the most important sources of the increase in emissions for the steady-state situation based on the 1990 regime.

In contrast to the apparent general view that these metal flows are well under control, the conclusions of this case study points in a different direction. The problem is all the more pressing since the recycling rates of the metals are already quite high.

4. Copper in the Dutch housing sector

In this section, a case study is presented on copper flows and accumulation in the Dutch housing sector. The study focuses on flows and accumulation in the economy as related to the housing sector. An extensive inventory has been made of inputs, outputs, stocks, accumulations and emissions of copper in the Dutch housing sector (Fraanje and Verkuijlen, 1996). Applications and use, both functional and non-functional, have been mapped and analysed. Activities and processes are included with regard to houses and residential blocks: the handling of building materials in the context of construction, maintenance, renovation and demolition as well as the (natural) degradation processes occurring during the lifetime of buildings. The analysis has focused on all flows and stocks of building materials as far as these belong to real estate, such as floors, walls, roofs and technical installations for space heating and drinking water. Flows and stocks of copper due to the use of houses, such as washing machines, domestic electronics, etc., were left out of consideration. Stocks and flows from non-functional use in bulk building materials such as cement, concrete, clay and applied waste materials

Table 5
Indicator values for copper for functional and non-functional applications in the Dutch housing sector in 1990 and at steady-state (SS)

	Functional		Non-functional	
	1990	SS	1990	SS
1. Technical efficiency (%)				
Extraction	n.a.	n.a.	n.a.	n.a.
Production	100	98.4	100	100
Use	100	100	99.9	99.9
Waste management	99.9	99.8	71	71
2a. Functional recycling rate (%)				
Extraction	n.a.	n.a.	n.a.	n.a.
Production	0	0	n.a.	n.a.
Use	0	0	n.a.	n.a.
Waste management	98.4	98	n.a.	n.a.
2b. Non-functional recycling rate (%)				
Extraction	n.a.	n.a.	n.a.	n.a.
Production	0	0	0	0
Use	0	0	0	0
Waste management	1.4	1.8	71	71
3. Economy accumulation ÷ total economy inflow (%)	21	0	84	0
4. Pollution export (tonnes/year)	−46	−491		
5. Total emissions (tonnes/year)	52	868	18	112
Air	0.0	0.0	0	0
Water	29	45	0	0
Agricultural soil	23	36	0	0
Non-agricultural soil	0.0	787	18	112
Landfill	0.0	748	18	112
Other sources	0.0	39	0	0

The steady-state calculations proceeded from a fixed inflow. This assumption leads to a steady-state housing stock of 12 million, which seems very large. If the housing stock is instead fixed at the 1990 level (5.7 million), the total emissions from non-functional applications, which are directly related to the number of houses, drops to 51 tonnes/year.

have been estimated separately. Most data were collected or estimated at a detailed level; the results were aggregated for the Dutch housing sector as a whole, which in 1990 comprised over 5.7 million dwellings. Flows associated with the import and export, production, waste management and recycling of building materials as well as flows of resulting emissions have been estimated at a more aggregate level.

Similarly to the case study presented in Section 3, the steady-state situation has been calculated with the FLUX model. Based on these results, the indicator values for copper for 1990 and for the steady-state were determined.

4.1. Results

Table 5 presents the indicator values. Since functional and non-functional applications of copper in the housing sector have highly separated cycles, they are presented separately. Both the functional and non-functional results reflect the main characteristics of the respective applications of copper in the Dutch housing sector. With regard to the functional applications, copper is most commonly used as the metal itself (copper piping, electrical wiring) or as an alloy (brass, tombac and solder). Therefore, copper applications are easy to identify, and as a consequence also relatively easily to withdraw from a waste stream, allowing high recycling and technical efficiency rates. Corrosion also occurs, but since copper is used mainly in indoor applications this results in a flow to the sewer system. Such flows may lead to emissions during the waste management stage and therefore do not count as losses from the use stage, which explains the high efficiency values.

The amount of copper in non-functional applications (i.e. as a contaminant in building materials such as concrete and cement) is much smaller (in 1990, 360 vs 22000 tonnes of copper). The difference in emissions is not nearly as large (18 vs 52 tonnes of copper), which implies that the functional chain is much more efficient. Another difference is seen in the accumulation rate: of all non-intentional inflow, 84% accumulates; for the intentional applications this is only 21%. This is due to the longer lifespan of the housing shells.

The increase of copper emissions at steady-state compared to 1990 is due to the build-up of stocks. This results in an increase of stock-related emissions during all stages. As a consequence, the technical efficiency rate of the functional applications decreases slightly.

4.2. Conclusion

Current emissions of copper from the housing sector contribute about 6% to the Dutch total (see Table 1). Since the inflow to the housing sector is 17% of the Dutch total, the housing sector can be regarded as a relatively efficient sector. Due to the high accumulation in 1990 the steady-state emissions will be much higher, but compared to the total Dutch steady-state emissions the share of the housing sector will decline to 1%. Another conclusion is that the functional chain is more efficiently managed than the non-functional chain of copper in the Dutch housing sector.

5. Cadmium in Dutch agricultural soil

In this section a case study is presented on cadmium flows and accumulation in the agricultural soils of The Netherlands. The study focuses on the modelling of environmental flows and accumulation related to the agricultural sector—i.e. crop uptake, leaching and accumulation in the topsoil. The inputs considered originate from fertilisers and atmospheric deposition.

To quantify the flows and accumulation of cadmium for the Dutch agricultural sector, two diverging approaches are possible:

- Top-down, i.e. based on national average data on farming systems and fertiliser use.
- Bottom-up, i.e. based on an extrapolation of data on, for example, crop rotation, fertiliser use, cadmium concentrations in fertilisers and produce, and soil characteristics collected on specific farms.

Here, the top-down approach is followed because it allows for an aggregated analysis at the national level. For a discussion of possible bottom-up approaches, see Moolenaar and Lexmond (1998).

Here, flows and accumulations in the Dutch agricultural sector have been quantified using Dutch statistical data on current manuring and fertilising practices. Three types of agricultural production system were distinguished: dairy farming, arable farming and mixed farming. These are characterised by specific crops (grass for dairy farming, arable crops like cereal, grains, potatoes and sugar beet for arable farming and silage maize and grass for mixed farming). These crops or groups of crops were combined with three soil types: sandy, clay and peat soil. This results in seven main crop–soil combinations that are representative of Dutch agriculture:

1. Grass on sandy soil (6% organic matter): 30% of total Dutch agricultural area (600000 ha).
2. Grass on clay soil: 20% (400000 ha).
3. Grass on peat soil: 10% (200000 ha).
4. Maize on sandy soil (6% organic matter): 5% (100000 ha).
5. Maize on sandy soil (1% organic matter): 5% (100000 ha).

Table 6
Risk ratios and transition periods for cadmium in soil and groundwater, and soil accumulation of cadmium related to the Dutch agricultural sector (for an explanation of crop–soil combinations, see text)

Indicator	Crop–soil combination							Area weighted mean
	1	2	3	4	5	6	7	
1. Soil accumulation 1990								
1990 (g/ha/year)	2.6	2.7	2.7	2.1	1.2	6.6	6.7	3.8
1990 (ton/year)	1.56	1.08	0.53	0.21	0.12	1.32	2.68	
2a. Risk ratio								
Soil								
1990	0.46	0.60	0.52	0.46	0.58	0.46	0.60	0.53
Steady-state	1.04	1.04	1.17	0.63	0.68	1.89	1.67	1.22
Groundwater								
1990	0.43	0.33	0.38	0.43	1.20	0.43	0.33	0.41
Steady-state	0.98	0.78	0.88	0.60	1.43	1.83	1.45	1.11
2b. Transition period (years)								
Soil	862	966	514	∞	∞	212	260	
Groundwater	∞	∞	∞	∞	0	240	500	

Soil concentrations have been compared with the Dutch reference value for total cadmium content in soil, which depends on the clay (L) and organic matter (H) content: $REF_{Cd} = 0.4 + 0.007[3H + L]$, with the reference value expressed in mg/kg and both L and H as mass percentages of the dry soil. Groundwater concentrations have been compared to the Dutch reference value for the cadmium concentration in groundwater (0.4 mg/m^3). Source: Janus et al. (1994).

6. Arable farming on sandy soil: 10% (200000 ha).
7. Arable farming on clay soil: 20% (400000 ha).

As mentioned in Section 2, a dynamic balance (DB) model was developed in this case study to model the soil processes for these combinations[8] (Moolenaar et al., 1997a). Input values for soil characteristics in the dynamic balance model (e.g. soil bulk density, soil composition and metal distribution coefficients, i.e. K_d values)) were based on (empirical) data gathered by Bakker and van den Hout (1993).

5.1. Results

The results of the calculations are shown in Table 6. They are presented as the soil accumulation rate (indicator 1) in 1990. Also presented are the risk ratios (indicator 2a) for soil and groundwater, for both 1990 and the steady-state situation, as well as the corresponding transition periods (indicator 2b).

Table 6 shows that the risk ratio for soil was below 1 in 1990, but will exceed 1 for most crop–soil combinations in the steady-state in the event of unchanged inputs and agricultural management. The transition periods, however, are over 200 years. For groundwater[9] the risk ratio already exceeds 1 for maize grown on sandy soil with low organic matter (crop–soil combination 5). In the steady-state it will also exceed 1 for arable farming on sandy soil and clay soil (crop–soil combination 6 and 7). Again, the transition period here exceeds 200 years.

[8] The DB model used can be coupled to FLUX in the same way as DYNABOX. However, coupling of soil processes with prior processes in the economy, such as the industrial production of fertilisers, has not yet been realised.

[9] In balance studies, it is generally very difficult to obtain proper leaching data. Leaching is therefore often neglected or estimations based on literature data are used. Using the minimum or maximum leaching rates instead of the average rate affects the balance results significantly. This shows that figures on leaching rates are both unreliable and important with regard to determining the resulting heavy-metal balance at farm and field scales.

Accumulation rates are lowest for silage maize (crop–soil combinations 4 and 5), which have a low input (animal manure with a low cadmium content) and a relatively high offtake. Accumulation rates are highest in the arable farming systems (crop–soil combinations 6 and 7), which have the highest net input because of a high input (superphosphates with a high cadmium content) and a relatively low offtake (50% cereal grains in the crop rotation). The difference between crop–soil combinations 4 and 5 is due mainly to the much lower K_d value of the soil of combination 5.

A rough assessment of possible contents in crops (i.e. grains and potatoes) indicates that the risk ratio[10] for crops is likely to rise at steady-state to a value between 1 and 5, depending on the specific crop.

5.2. Further explorations

The above results concern average farming practice per crop–soil combination with respect to fertiliser use and crop rotation, with the concentration in the crops themselves not being systematically taken into account. Therefore, the importance of fertiliser use and crop rotation were further explored: for arable farming on clay (crop–soil combination 7). This exploration is based on a study of different farming systems as practised on an experimental farm in The Netherlands in 1995 (Moolenaar et al., 1996). The following systems were investigated:

- Conventional arable farming systems (CAFS).
- Integrated arable farming system (IAFS).
- Ecological arable farming system (EAFS).

For the conventional system, two subsystems were distinguished: a system with use of mineral and organic fertilisers (CAFS OF), and a system with exclusive use of mineral fertilisers (CAFS MF).

In this case the risk ratio (indicator 2a) was calculated not only for soil and groundwater, but also for different crops. Table 6 shows that the risk ratio for soil is exceeded in all farming systems, and that the transition periods may become significantly shorter compared with the results in Table 5. The risk ratio for groundwater exceeds 1 only for the conventional system using mineral fertilisers, and the transition periods are longer compared with the results in Table 5. Furthermore, for some crops the risk ratio exceeds 1. The transition periods differ significantly (from about 150 to 700 years) among the different farming systems. One surprising result is that it appears from Table 7 that, in contrast to the most obvious notion, not ecological (EAFS) but integrated (IAFS) and conventional arable farming with use of mineral and organic fertilisers (CAFS OF) appears to be the best farming type with respect to environmental quality.

Finally, another qualitative exploration should be made. If input and management characteristics do not change, at steady-state cadmium concentrations will exceed the Dutch reference value for soil in most farming systems studied and for crop and groundwater quality in some. There exists an important interconnection between cadmium and phosphorus flows in agricultural systems. Both mineral phosphorus fertilisers and feed phosphates (ending up in manure) contain cadmium occurring as a contaminant in phosphate ores. Sustainable cadmium management implies, among other things, that cadmium additions to the soil through fertiliser applications should not exceed the acceptable output by harvest and leaching, and that the total cadmium content in soil should not exceed the relevant standards. In order to safeguard these criteria, farmers can be more careful in selecting fertilisers with the lowest cadmium and the highest phosphorus content, select the most appropriate farming system and cultivate crops with pronounced cadmium removal (within critical levels). In this context, due care should be taken to avoid problem-shifting and trade-offs. For example, the present Dutch phosphorus fertiliser industry has too high cadmium emissions to the North Sea. If Dutch production were to be phased out for this reason, the present phosphorus fertiliser might be substituted by fertilisers with even an higher cadmium content. Consequently, cadmium input to agricultural soils would increase substantially.

[10] The standard used in calculating the risk ratio for crops is based on the legal limit for food crops as defined in the Dutch Commodities Act.

5.3. Conclusion

The results show that present agricultural fertiliser schemes are not sustainable in the long term. Efforts are required to substantially reduce cadmium input through use of mineral fertilisers and manure, in order to reach a sustainable steady-state situation (i.e. to reduce all concentrations to below critical levels). In contrast, there is a risk that current inputs may further increase in practice.

6. General discussion and conclusion

Whilst emissions of metals are decreasing, their input into the economy is still increasing. What happens to this input, now and in the long run? These questions were addressed through integrated modelling of metal flows within both the economy and the environment. In this section a summary is given of the main findings and of their implications for policy and future research.

First the models FLUX (flows in the economy) and DYNABOX (flows in the environment) were developed and linked. Both models have the capacity to make steady-state calculations, and in relation to these models a system of policy-relevant indicators dealing with processes within and between the economy and the environment was developed.

Taking the demand structure and technological processes of 1990 as a starting point, the metal emissions from the economy to the environment appeared to increase markedly from 1990 to the constructed steady-state. This increase is due

Table 7
Steady-state concentrations of cadmium in soil, groundwater and crops for different farming systems

Indicator	Farming system			
	EAFS	IAFS	CAFS OF	CAFS MF
2a. Risk ratio				
Soil				
1990	0.8	0.8	0.8	0.8
Steady-state	2.2	1.3	1.5	2.9
Groundwater				
1990	0.0	0.0	0.0	0.0
Steady-state	0.7	0.1	0.2	1.0
Crops, steady-state				
Seed potato	0.3	0.1		
Ware potato	0.3	0.1	0.2	0.3
Spring barley		0.2	0.2	0.5
Winter barley	0.1		0.0	0.1
Spring wheat	1.1			
Winter wheat	0.9	0.5	0.6	1.3
Bean	1.0			
Oats	0.5			
Onion	0.4	0.1	0.1	0.3
Celeriac	1.3			
Sugar beet		0.3	0.4	0.9
Chicory			0.3	0.7
2b. Transition period (years)				
Soil	145	306	245	70
Groundwater	∞	∞	∞	3366
Crops	620	360	700	150

Sources: Moolenaar et al. (1996, 1997b).

mainly to balancing inputs to landfills and non-functional applications (e.g. fly ash in roads) with emissions thereof to non-agricultural soil. In addition, however, emissions along sensitive routes to air, water and agricultural soil showed an increase of about 30% for cadmium, lead and zinc and over 100% for copper. The latter is caused by a shift from the present accumulation in products and materials to emissions to the environment.

In the constructed steady-state lead and to a lesser degree zinc and copper appear to pose significant risks to human health; all four metals pose serious risks to aquatic and to terrestrial ecosystems, indicating that the present economy is not sustainable with respect to metal metabolism. In addition, the agricultural case study on cadmium indicated an exceeding of crop standards, in dependence on such factors as crop rotation and fertiliser scheme.

Analysis of the economy indicators allowed a closer look at the underlying causes of the increased emissions. This increase takes place despite the quite substantial functional recycling rates; apart from cadmium, these were at least 80% in 1990. The non-functional recycling flows are the major cause of diffuse emissions to the important media (air, water, agricultural soil) for human and ecotoxicity. So even with a further increase of functional recycling, there will be leakage due to the non-functional squandering of the metals in the economy. In fact, relatively small concentrations in specific flows in the economy cause a marked increase of risks through a closed-loop accumulation process, as in the example of copper and zinc in fodder.

Three main approaches for enhanced management appear feasible: the input into the economy can be lowered, the output can be delayed and the output can be controlled. The first main approach, lowering of the input, can be achieved by substitution of metals in functional applications (e.g. zinc gutters; cf. Kandelaars and van den Bergh, 1997), by recycling or increasing the lifespan of metals with an elastic supply (e.g. copper, lead or zinc), and by reducing non-functional inflows (e.g. along with fossil fuels). The second main approach, delaying the output, can be achieved by keeping non-functional metals in the economy (e.g. fly ash in roads). This option offers time for further development of the third main approach: control of the output, which can be achieved by physicochemical immobilisation of the waste flow (e.g. vitrification), by waste disposal outside the biosphere, and by bypassing the sensitive environmental routes (e.g. redirection to mines). It is beyond the scope of the present article to explore these possibilities, which are to some extent already being applied in current policies.

Although the models used include the full spectrum of flows and accumulations in the economy and environment, the results are merely indicative. Besides the uncertainties in economy–environment modelling, a further limitation is that resource availability has not yet been taken into account; this is in fact assumed to be infinite. So perhaps the high risk ratios will not be reached because of enforced declines in resource extraction. However, this is by no means certain, given the continually rising estimates of resource availability. Consequently, the results at least imply a warning signal as to the sustainability of current metal metabolism.

Further study is advocated along the following three lines. Firstly, one line encompassing dynamic modelling should be developed to consistently assess resource depletion as well as emissions. Secondly, the system of indicators should be extended, to include a 'dissipation indicator', describing the squandering within the economy; an 'immobilisation indicator', addressing different metal species; and an 'environmental pathway indicator', describing the environmental routes of metal species to the final sinks. Thirdly, the above management options should be systematically evaluated as to their economic and environmental costs and benefits.

Acknowledgements

This research was supported by a grant from The Netherlands Organization for Scientific Research (NWO). The authors wish to thank Reinout Heijungs for his work on the DYNABOX model, and several anonymous referees for their helpful comments.

References

Allenby, B.R., 1999. Industrial Ecology—Policy Framework and Implementation. Prentice Hall, Upper Saddle River, NJ.

Annema, J.A., Paardekoper, E.M., Booij, H., van oers L.F.C.M., van der Voet, E., Mulder, P.A.A., 1995. Stofstroomanalyse van zes zware metalen—Gevolgen van autonome ontwikkelingen en maatregelen. RIVM report no. 601014010, Bilthoven.

Ayres, R.U., 1996. Statistical measures of unsustainability. Ecol. Econ. 16, 239–255.

Ayres, R.U., Rod, S.R., 1986. Patterns and pollution in the Hudson-Raritan Basin. Environment 28, 14–20 and 39–43.

Ayres, R.U., Simonis, U.E. (Eds.), 1994. Industrial Metabolism: Restructuring for Sustainable Development. United Nations University Press, Tokyo.

Azar, C., Holmberg, J., Lindgren, K., 1996. Socio-ecological indicators for sustainability. Ecol. Econ. 18, 89–112.

Baccini, P., Bader, H.-P., 1996. Regionaler Stoffhaushalt, Erfassung, Bewertung und Steuerung. Spektrum Akademischer Verlag, Heidelberg.

Baccini, P., Brunner, P.H., 1991. Metabolism of the Antroposphere. Springer-Verlag, Berlin.

Bakker, D.J., van den Hout, K.D., 1993. De invloed van atmosferische depositie op de kwaliteit van bodem en oppervlaktewater in Nederland. Beschrijving rekenmethode en berekeningsresultaten. TNO—Milieu en Energie, IMW-R 93/200, Apeldoorn.

Bergbäck, B., Lohm, U., 1997. Metals in society. In: Brune, D., Chapman, V. (Eds.), The Global Environment—Science, Technology and Management. Scandinavian Scientific Press, Oslo, pp 276–289.

Boelens, J., Olsthoorn, A.A., 1998. Software for material flow analysis. In: Vellinga, P., Berkhout, F., Gupta, J., 1998. Sustainable Sustainability. Kluwer, Dordrecht, pp. 115–130.

Bringezu S., Fischer-Kowalski, M., Kleijn, R., Palm, V. (Eds.), 1997. Regional and National Material Flow Accounting: From Paradigm to Practice of Sustainability. Institute for Climate, Environment and Energy, Wuppertal.

Cleven, R.F.M.J., Janus, J.A., Annema en W. Slooff, J.A., 1992. Basisdocument zink. Report no. 710401019, RIVM, Bilthoven.

Crommentuijn, T., Polder, M.D., van de Plassche, E.J., 1997. Maximum permissible concentrations and negligible concentrations for metals, taking background concentrations into account. Report no. 601501001, RIVM, Bilthoven.

Fraanje, P.J., Verkuijlen, E., 1996. Balansen van non-ferro metalen in de Nederlandse woningbouw in 1990. IVAM onderzoeksreeks nr. 76, Amsterdam.

Gezondheidsraad (Commissie Risico-evaluatie van Stoffen), 1998. Zink. Gezondheidsraad publicatie nr. 1997/34, Rijswijk.

Guinée, J.B., Kandelaars, P.P.A.A.H., Huppes, G., 1997. Linking economic models to physical substance flow models: possibilities and difficulties. In: Bringezu, S., Fischer-Kowalski, M., Kleijn, R., Palm, V. (Eds.), Regional and National Material Flow Accounting: From Paradigm to Practice of Sustainability. Institute for Climate, Environment and Energy, Wuppertal.

Hansen, E., Lassen, C., 1997. Sustainability of the current environmental load of persistent chemical compounds—focus on lead. In: Bringezu, S., Fischer-Kowalski, M., Kleijn, R., Palm, V. (Eds.), Regional and National Material Flow Accounting: From Paradigm to Practice of Sustainability. Institute for Climate, Environment and Energy, Wuppertal.

Heijungs, R., Guinée, J.B., Wegener Sleeswijk, A., 1998. DYNABOX: a generic dynamic multi-media fate model. In preparation.

Instituut voor Europees Milieubeleid (IEM), Environmental Resources Management (ERM), 1996. Mercury stock management in The Netherlands. Draft background document prepared for the workshop Kwik, uitbannen of beheersen? IEM, Brussel.

Janus, J.A., Hesse, J.M., Rikken, M.G.J., 1994. Aaandachtstoffen in het Nederlandse milieubeleid—overzicht 1994. Report no. 601014006, RIVM, Bilthoven.

Kandelaars, P.P.A.A.H., van den Bergh, J.C.J.M., 1997. Dynamic analysis of materials–product chains: an application to window frames. Ecol. Econ. 22, 41–61.

Moolenaar, S.W., Lexmond, Th.M., 1998. Balancing heavy metals: III. Heavy-metal balances of agro-ecosystems at field scale. Submitted for publication.

Moolenaar, S.W., Hatziotis, P., Lexmond, Th.M., 1996. Aspects of cadmium accumulation in agriculture. In: Fertilizers as a source of cadmium. Proceedings of OECD Inter-Organization Programme for the Sound Management of Chemicals Workshop, held in Saltsjöbaden, Sweden, 16–20 October 1995, pp. 119–132.

Moolenaar, S.W., Lexmond, Th.M., van der Zee, S.E.A.T.M., 1997a. Calculating heavy metal accumulation in soil: a comparison of methods illustrated by a case-study on compost application. Agric. Ecosyst. Environ. 66, 71–82.

Moolenaar, S.W., van der Zee, S.E.A.T.M., Lexmond, Th.M., 1997b. Indicators of the sustainability of heavy metal management in agro-ecosystems. Sci. Total Environ. 201, 155–169.

OECD, 1993. Lead. Risk Reduction Monograph 1. OECD, Environment Directorate, Paris.

OECD, 1994. Cadmium. Risk Reduction Monograph 5. OECD, Environment Directorate, Paris.

Olsthoorn, A.A., Boelens, J., 1998. Substance flow analysis with FLUX. Report no. W98\15. Institute for Environmental Studies, Free University, Amsterdam.

RIVM, VROM, WVC, 1994. Uniform system for the evaluation of substances (USES), version 1.0. National Institute of Public Health and Environmental Protection (RIVM), Ministry of Housing, Spatial Planning and Environment (VROM), Ministry of Welfare, Health and Cultural Affairs (WVC). Ministry of Housing, Spatial Planning and Environment. Distribution no. 11144/150, The Hague.

Socolow, R., Andrews, C., Berkhout, F., Thomas, V., 1994. Industrial Ecology and Global Change. Cambridge University Press, Cambridge.

Stigliani, W.M., Anderberg, S., 1994. Industrial metabolism at the regional level: the Rhine Basin. In: Ayres, R.U., Simonis, U.E. (Eds.), Industrial Metabolism: Restructuring for Sustainable Development. United Nations University Press, Tokyo, pp. 119–162.

United States Department of the Interior–Bureau of Mines (USBM), 1985. Mineral Facts and Problems. US Government Printing Office, Washington, DC.

United States Department of the Interior–Bureau of Mines (USBM), 1992. Minerals in the World Economy—1989 International Review. US Government Printing Office, Washington, DC.

United States Department of the Interior–Bureau of Mines (USBM), 1993. Mineral Commodity Summaries. US Government Printing Office, Washington, DC.

Van der Voet, E., 1996. Substances from cradle to grave: development of a methodology for the analysis of substance flows through the economy and the environment of a region—with case studies on cadmium and nitrogen compounds. Ph.D. Dissertation, Leiden.

Van der Voet, E., van Egmond, L., Kleijn, R., Huppes, G., 1994. Cadmium in the European Community: a policy oriented analysis. Waste Manage. Res. 12, 507–526.

Van der Voet, E., Guinée, J.B., Udo de Haes, H.A., 1997. Indicators for integrated substance chain management as a measure for environmental quality and sustainable development. CML report no. 139. Section Substances and Products, Leiden.

Van Drecht, G., Boumans, L.J.M., Fraters, D., Reijnders H.F.R., van Duijvenbooden, W., 1996. Landelijke beelden van de diffuse metaalbelasting van de bodem en de metaalgehalten in de bovengrond, alsmede de relatie tussen gehalten en belasting. Report no. 714801006, RIVM, Bilthoven.

Vermeire, T.G., van Apeldoorn, M.E., de Fouw, J.C., Janssen, P.J.C.M., 1991. Voorstel voor de humaan-toxicologische onderbouwing van C-(toestings)waarden. Report no. 725201005, RIVM, Bilthoven.

VROM, 1993. Emissies in Nederland: Trends—vierde inventarisatieronde basisjaar 1988. Publikatiereeks Emissieregistratie Supplement nr. 5A, VROM, The Hague.

Wernick, I.K., Ausubel, J.H., 1995. National materials flows and the environment. Annu. Rev. Energy Environ. 20, 463–492.

Westhoek, H.J., Beijer, L., Bruins, W.J., Hotsma, P.H., Janssen, J.W.M., Maathuis, E.J.R., 1997. Aan- en Afvoerbalansen van Zware Metalen van Nederlandse Landbouwgronden. Informatie- en KennisCentrum Landbouw, Ede.