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Doctoral Thesis

**A new indicator for the assessment of
anthropogenic substance flows to regional sinks.**

submitted in satisfaction of the requirements for the degree of
Doctor of Science in Civil Engineering
of the Vienna University of Technology, Faculty of Civil Engineering

Dissertation

**Ein neuer Indikator zur Bewertung von
anthropogenen Stoffflüssen in regionale Senken.**

ausgeführt zum Zwecke der Erlangung des akademischen Grades eines
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Abstract

Satisfying human needs requires an anthropogenic material turnover. After utilization, materials either remain in the anthroposphere in terms of recycling products, or they leave the anthroposphere in terms of waste and emission flows. The last two enter downstream sinks, man-made and natural ones. The problem is that material flows to natural sinks may cause risks for human and environmental health. To avoid overloading, several assessment frameworks have been put forward. In an economy-wide perspective, a single score indicator focusing on substances that leave the anthroposphere to regional sinks is missing. To overcome this gap, the thesis aims to develop a new indicator and to compute the score for selected case studies.

To achieve these goals, four steps are needed. First, the indicator is defined as the environmentally acceptable mass share of a substance in material flows that leave the anthroposphere to downstream sinks. The resulting score ranges between 0% as worst case and 100% as best case. Second, a methodology to determine the indicator components is presented, including (i) inventories based on substance flow analysis, and (ii) impact assessment based on a distance-to-target approach. Third, the framework developed is applied in three case studies including copper (Cu) and lead (Pb) on an urban scale (City of Vienna) and Perfluorooctane Sulfonate (PFOS) on a national scale (Switzerland). Fourth, recommendations are given for increasing the indicator score by means of sink load reduction or enhancement of sink capacities.

The following results are obtained: In Vienna, 99% of Cu mass flows to regional sinks are acceptable. However, the 0.7% of Cu entering urban soils and the 0.3% entering receiving waters surpass acceptable levels. In the case of Pb, 92% of all mass flows to sinks prove to be acceptable, and 8% are disposed of in local landfills with limited capacity. For PFOS, 96% of all flows to sinks are acceptable. 4% cannot be evaluated due to a lack of quality criteria, despite posing a risk for human health and the environment. The examples demonstrate the need for: (i) enhanced regional landfill capacities or increased recycling rates, (ii) regional standards for assessing substance flows to urban soils and receiving waters, (iii) appropriate data of good quality, and (iv) the extension of the methodology to include exports to sinks in the hinterland.

The new indicator is of relevance for managing wastes and emissions because it identifies substance flows to sinks that observe or neglect quality criteria, or that cannot be assessed due to missing knowledge. Moreover, it serves for monitoring the performance of waste and environmental management within a region, and for comparing the performance with other regions. For strategic decisions such as design and evaluation of policies, the indicator allows an examination of the effectiveness of directing substance flows to appropriate sinks. Finally, the indicator aggregates complex information into an easy to understand score and is therefore highly instrumental for communicating scientific research to decision makers and the public.

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I dedicate my special thanks and admiration to Claudia. She looked after our two sons Jakob and Adrian with loving care while I was traveling to scientific conferences around the world to present my research. Beyond that she is going to enrich our lives by giving birth to our third son, Lorin, in the near future.

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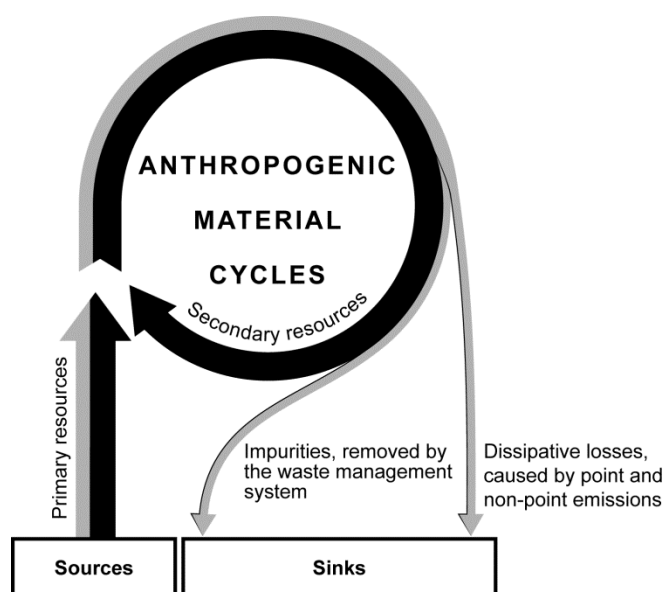
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1 INTRODUCTION

1.1 Background and problem definition

Satisfying human needs requires an anthropogenic material turnover. The turnover encompasses material cycles and stocks that grow with the input of primary and secondary materials and that decrease with the removal and loss of materials (Figure 1).

Figure 1: Anthropogenic material cycles are inevitably open, thereby including removals by the waste management system and dissipative losses from point and non-point emission sources (adopted from Stumm et al. 1974). Clean material cycles encompass materials without detrimental substances (black colored flows). Grey colored flows include substances that pose threats to human and environmental health.



Globally, the input of primary resources satisfies an increasing demand for materials (Dittrich et al. 2013) and compensates for the (i) removal and (ii) loss of materials from anthropogenic material cycles:

- (i) The removal of materials has three key drivers: First, the limiting factors for recovering valuable secondary resources, for instance, arise from waste collection schemes that might not be oriented towards recycling, technologies that separate recyclable and non-recyclable mass fractions, and economic conditions including the costs and benefits of secondary material production (UNEP 2013). Second, to establish clean material cycles encompassing secondary materials without impurities, hazardous substances contained in waste flows must be removed by, before or during recycling (Brunner 2010, Kral et al. 2013). Despite regulatory frameworks including the obligation to remove hazardous substances from waste flows (e.g. Republik Österreich 2008, Republik Deutschland 2012), waste management partly fails to fulfill this requirement. Examples are mineral oil in waste paper (ESFA 2012),

brominated flame retardants in recycled plastic (Chen et al. 2009, Samsonek et al. 2013) and recycled carcinogenic substances in construction waste (Rubli 2013, Deutscher Bundesrechnungshof 2014). Third, strategies to minimize the environmental impacts of material production contrast impacts from primary and secondary material production. Therefore, the minimization of environmental impacts is rather based on optimal than on high recycling rates. For copper, Stumm and Davis (1974) demonstrated that one ton has to be composed of 60% secondary and 40% primary copper in order to require a minimum of production energy. For treating cooling appliances, Laner et al. (2007) discussed the tradeoff between environmental protection and resource conservation.

- (ii) Material losses in terms of emissions occur throughout the entire process chain, including the upstream, use, and downstream phases. Depending on the spatial release pattern, losses originate from point or non-point sources. Hence, losses either result from the deliberate intention to dissipate substances in the environment, such as with pesticides applied on land, or might but cannot be avoided, such as with wear from brake pads (Lifset et al. 2012).

To conclude, materials leave the anthropogenic material cycle because the recovery rate of secondary materials inevitably falls below 100% and because emissions occur along the life cycle of materials. If materials leave the material cycle, sinks are required to accommodate these materials.

The problem is that substance flows to natural sinks cause risks for human and environmental health. To avoid burdens, natural sinks are available to a certain extent yet man-made sinks have to be provided where natural sinks are lacking. Policy makers in the field of waste and environmental management should consider to what extent the sinks can be loaded with substance flows and, if needed, should provide suitable strategies to undershoot the limits. The effectiveness of directing waste and emission flows to appropriate sinks is a measure of the environmental dimension of sustainability.

1.2 Goal, scope and research questions

The thesis investigates the regional anthropogenic material turnover with respect to limits for the discharge of substances to sinks. The goal is the development of an indicator that quantifies the relative magnitude between acceptable and entire flows to regional sinks. The framework to calculate the indicator score combines inventory analysis based on substance flow analysis and impact assessment based on a distance-to-target weighting approach. Selected case studies demonstrate both the applicability of the methodology and the benefits of the indicator score for managing waste and emission flows.

The following research questions ensue from the goal of the thesis:

- What are substance flows to sinks?
- How can the indicator be defined and quantified?
- What is the indicator score for selected case studies?
- What's the benefit of the new indicator?

The investigation into the regional metabolism focuses on substance flows (a) removed and lost from the anthroposphere and entering regional sinks downstream, (b) discarded to natural and man-made sinks, and (c) limited because of quality standards. Economic and social issues are excluded from the assessment, despite their relevance for managing regional waste and emission flows.

1.3 Structure of the thesis

The thesis is based on three research articles (see appendix III), which are framed from chapter 1 to chapter 6. Chapter 1 points to the need for sinks that accommodate substance flows, defines the problem, goal, scope and research questions which are tackled in the thesis. Chapter 2 describes state-of-the-art policy instruments and science-based indicators, and, finally, proposes a new indicator for assessing mass flows to regional sinks. Chapter 3 defines the proposed indicator, including the indicator components, and presents a framework to calculate the indicator score. Chapter 4 presents the application of the indicator framework to three case studies for Copper and Lead on a city level (Vienna) and PFOS on a national level (Switzerland). Chapter 5 presents the results, thereby providing answers to the research questions. Chapter 6 gives an outlook for future research.

The appendices include (I) background information regarding the definition of the term “sink”, (II) the findings from a literature study regarding inventories based on substance flow analysis, and (III) the articles as an integrative part of the thesis and puts them in the context of the thesis.

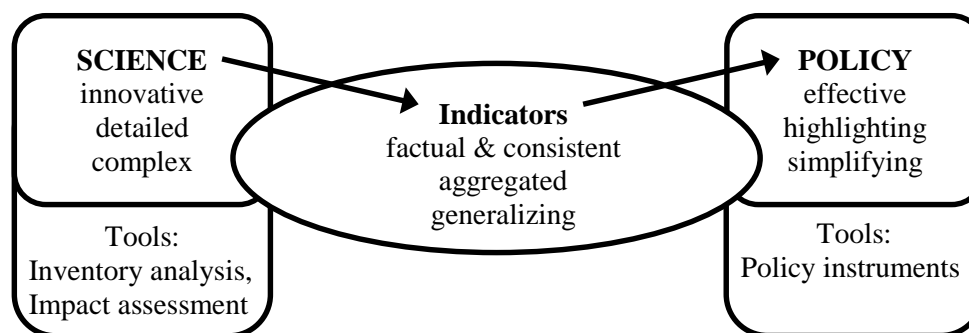
2 STATE OF THE ART

This chapter (1) outlines the chapters' framework, (2) describes policy instruments to manage waste and emission flows, (3) presents science-based indicators and, finally, (4) highlights the need for a new indicator.

2.1 Introduction

To avoid burden on human and environmental health, policy instruments are needed to manage substance flows to natural sinks. Each instrument draws upon an informative decision base. Therefore, science-based indicators quantify phenomena in the anthroposphere and in the environment. In particular, indicators serve as a communication tool between science and non-scientists, the public, and policy-oriented decision makers (Figure 2).

Figure 2: Science, indicators and policy (adopted from Jesinghaus 1999)



2.2 Policy

Indicators play a key role for communicating “if a development is on the right track or if adaptations or changes in policy directions need to take place in order to fulfill decided objectives or reaching targets” (Frederiksen et al. 2013). Hence, the authors list three important drivers for using indicators. First, the wish to “compare environmental and other performances across countries and sectors”. Second, the need of “governments to satisfy transparency and accountability requirements in policy performance evaluations”. Third, a “call for general information and communication with the public on sustainable development and the state of the environment”. The selection of indicators depends on regional circumstances, for instance, on the respective policy instrument used to manage flows into sinks. Apart from moral suasion, the creation of property rights, taxation and environmental performance bonds, Common et al. (2005) list two policy instruments in direct relationship with the scope of the thesis: (i) tradable permits, and (ii) command and control instruments.

- (i) Tradable permits are used to control emission flows by giving economic incentives. Therefore, a critical flow in mass per time is determined for a specific substance, region or sector, and

period. With respect to the critical flow, tradable permits are issued by regulatory authorities and a market-based mechanism allocates the permits to participating agents. For instance, the Kyoto Protocol includes three options if a participating agent exceeds the allowed emissions (United Nations 1998): (1) To buy permits from other participating agents. (2) To reduce emissions in external regions that either participate or do not participate as agents. (3) To enhance the removal of greenhouse gases by sinks on the basis of land use, land use change and forestry.

- (ii) Command and control instruments or “direct regulations” are widely used for managing substance flows into sinks. Examples of direct regulations in Austria include quality standards for surface waters (Republik Österreich 2006) and standards for waste disposal (Republik Österreich 2008). Defining quality standards is a consensus-based process influenced, for example, by scientific knowledge, available data, and technical and economic feasibility. The points of command are identical with the points of control, supported by indicators along the cause-effect chain of substances. For instance, pressure indicators include the level of substance concentration in flows or annual flow rates. State indicators refer to the level of substance concentration in environmental media. Damage indicators might refer to the acceptable risk levels, despite the difficulty in defining them (Hunter et al. 2001).

2.3 Science

With respect to science, researches have devoted much effort to identify relevant indicators to support policy instruments. From a methodological point of view, the indicators result either from inventory analysis or from impact assessment (Table 1). Therefore, the focus is on the cause-effect chain of substances, from the stage of "primary source" to the stage of "damage". Depending on the available knowledge, the indicators either refer to “known damage due to known causalities”, or “known damage due to unknown causalities”, or “unknown damage due to unknown causalities” (adopted from Hofstetter 1998). If damage and causalities are known, *damage indicators* can be provided. For the majority of substances placed on the market, the damage and causalities are partly or totally unknown (Berg et al. 1994, Grandjean 2013). In this case, *proxy indicators* with more or less predictive power are used to approximate potential damage. With respect to the cause-effect-chain, the *proxy indicators* can be further categorized into "*pressure indicators*", "*state indicators*", "*exposure indicators*", and "*effect indicators*".

Table 1: Stages on the cause-effect chain of substances and corresponding indicator examples.

Method	Stage	Stage description	Type of indicator	Indicator examples
Inventory analysis	Source	Material stocks as part of biogeochemical cycles	Proxy	Material stock in terms of resources and reserves
	Utilization	Man-made material cycle, potential of waste and emissions		Material intensity per service, Hibernating stock, stock-in-use, Recycling rates
	Pressure State	Flow into sinks Fate in man-made sinks		Emissions and waste flows Substance concentration, accumulation or transformation rate
Impact assessment	State	Fate in natural sinks	Damage	Substance concentration, accumulation or transformation rate
	Exposure	Standard characteristics of exposed organism		Exposed dose, collective effective dose
	Effect	Dose-Response-Relationship		Number of human diseases, or of vanishing plant species
	Damage	Damage to human health or ecosystem quality		Disability adjusted life years, Share of vanishing plant species per area and time unit.

(1) Inventory analysis

Inventory analysis focuses on the anthropogenic material turnover, including the fate and behavior of substances throughout the life-cycle-chain, from its geogenic or synthetic origin to the whereabouts in the anthroposphere, and the flow to natural sinks. To create inventories, including anthropogenic waste and emission flows to sinks, several tools have been developed and put forward, such as substance flow analysis (SFA) (Baccini et al. 1991, Baccini et al. 1996, Brunner et al. 2004), physical-input-output-analysis (PIO) and corresponding methods like Environmental Extended PIO and Full PIO (Hoekstra et al. 2006), and Economy-wide Material Flow Analysis (Adriaanse et al. 1997, European Communities 2001). With respect to the scope of the thesis (substance flows to regional sinks), two examples highlight region-wide inventories:

First, an international standardized inventory is available for carbon. Member states under the Kyoto protocol have to “establish and maintain a national system for the estimation of anthropogenic emissions by sources and removals by sinks” (UNFCCC 2008). When it comes to the regional and city level, international standardized inventories are lacking. Mohareb et al. (2012) proposed a framework on city level and initiated a discussion about the definition and classification of carbon flows to sinks (Wiedmann 2012).

Second, multiple SFA studies have been conducted by authorities or have been established by research groups (e.g. Brunner et al. 1993, Palm et al. 1996, van der Voet et al. 1999, Döberl et al. 2002, Laner et al. 2009, Månsson et al. 2009, Lifset, Eckelman et al. 2012, Burger Chakraborty et al. 2013). Due to the varying scope and methodology for each study, inventory data differ in terms of selected substances, flows, sinks, regions and periods of time (see appendix II). Despite their relevance in the respective context, the comparison and validation of inventory data across regions is lacking because of the absence of a standardized framework or data.

(2) Impact assessment

Inventory data are of a descriptive nature. Because of their physical accounting unit “mass”, they rely on the “equivalence of different emissions with unequal environmental impacts” (Jungbluth et al. 2012). To assess the impacts, the substance flows have to be characterized, normalized and weighted by normative methods (Brunner 2002). Therefore, inventory data are combined with impact assessment methods such as Life Cycle Impact Assessment (e.g. Boesch et al. 2009, Venkatesh et al. 2009, Rochat et al. 2013, Vadenbo et al. 2013, Boesch et al. 2014), Exposure and Risk Assessment (e.g. Guinée et al. 1999, Gottschalk et al. 2010, Jang et al. 2012, Chen et al. 2013, Lesmes-Fabian et al. 2013), cost-effective assessment in the field of waste management (Döberl, Huber et al. 2002), and critical load concepts for specific flows into environmental media (Nagl 1996, Spranger et al. 2004). With respect to the cause-effect chain of substances, each method provides indicators at a specific stage (Table 2).

Table 2: Impact assessment methods provide proxy and damage indicators.

Method	Type of indicator		Reference
	Proxy	Damage	
Material intensity per service	X		Ritthoff et al. (2002)
Ecological Footprint	X		Wackernagel et al. (1996)
Sustainable Process Index	X		Krotscheck et al. (1996)
Entropy index	X		Rechberger et al. (2002), Sobańtka (2013)
Ecological Scarcity 2006			Frischknecht et al. (2009)
ReCiPe 2008		X	Goedkoop et al. (2012)
Risk assessment		X	McKone et al. (2002)
Cost-Effective Method	X		Döberl, Huber et al. (2002)
Critical load method	X		van het Bolcher et al. (2000), Hellweg et al. (2005)

The Material Intensity per Service aggregates the amount of materials along the life cycle chain of products and services. Ecological Footprint and the Sustainable Process Index provide a single indicator on an area base. The entropy index is used to quantify the dilution or concentration performance of processes for elements and compounds. Life cycle impact methods provide indicators related to impact categories with an option to aggregate them into a single score. For example, Ecological Scarcity 2006 provides a *pressure indicator* in terms of a single score with Eco-Points for each intervention. ReCiPe 2008 provides three *damage indicators* (damage to resources, ecosystems, and human health) and weighting factors to aggregate the three indicators into a single score. Risk assessment methods provide *damage indicators* of absolute risk in view of human and environmental health. The cost-effective method is based on a distance-to-target approach and yields a single score. Critical load concepts provide *pressure indicators* such as the critical deposition on land, and the critical flow from landfills to subsurface layers.

2.4 Need for a new indicator

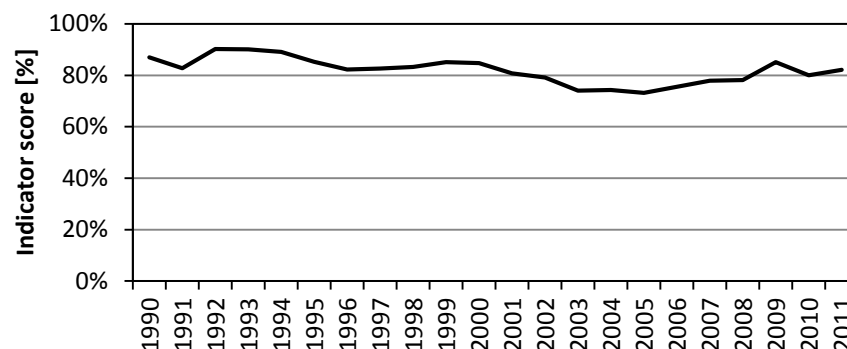
According to chapter 1.1, the problem is that substance flows into natural sinks pose risks for human and environmental health. To manage these risks, decision makers should consider to what extent the sinks can be loaded with substance flows and, if needed, should provide suitable strategies to undershoot the limits. Therefore, policy instruments encompass tradable permits and command and control strategies (chapter 2.2), including relevant indicators (chapter 2.3). If substance flows to regional sinks are managed, the evaluation of success depends on the policy instrument and therefore on the number of managed flows:

- Tradable permits regulate a specific substance flow to a specific sink. The success control can be monitored by a simple indicator: The relative magnitude of accepted and entire flows to the sink:

$$\text{Indicator score} = \frac{\text{Accepted flow to sink}}{\text{Entire flow to sink}} * 100$$

For example, the greenhouse gas emissions in the Austrian economy are reported on an annual base, and the reduction target has been agreed on under the Kyoto Protocol at minus 13% compared to the emissions in the year 1990 (Anderl et al. 2013). Based on this information, Figure 3 plots the indicator score for greenhouse gas emissions in Austria from 1990 – 2011. As long as the score stays below 100%, adjustments need to be made or measures need to be taken in order to reach the emission target. Based on Figure 3, it is obvious that the Austrian economy emitted more than has been allowed. To conclude, tradable permits from external regions have to be allocated to the Austrian economy.

Figure 3: Indicator score for greenhouse gases (CO₂-equivalents) in the Austrian economy, from the year 1990 to 2011.



Up to now, tradable permits have been available for less than 10 substances and for selected regions only (Common and Stagl 2005). For most substances and regions, cross-regional assessment and management of substance flows with tradable permits is lacking. This lack therefore amounts to the absence of an easily understandable indicator for most substances.

- In contrast to tradable permits, command and control instruments evaluate a specific substance in several flows to multiple sinks by various indicators. For instance, carbon to air has to meet an international agreed emission standard, whereas carbon to landfills has to meet national standards. Consequently, success control in view of reaching targets is based on flow by flow assessment. To provide overall region-wide information about the disposal of substances in sinks, Döberl et al. (2004) proposed a new indicator for monitoring the environmental dimension of sustainability:

$$\text{Indicator score} = \frac{\text{Amount of substances a region or process directs in appropriate final sinks}}{\text{Total amount of substances emitted by a region or process}}$$

Apart from the proposed indicator definition, a framework for calculating the indicator components has not been presented.

To conclude, with respect to policy instruments and relevant indicators today, the focus is rather on single flows than on entire flows to sinks. An overall assessment of substances is lacking since substances that have been removed or lost from the anthroposphere to regional sinks are not evaluated by a common framework today. To overcome the gap in favor of a substance-oriented policy, a new indicator is needed. Consequently, the thesis proposes a new indicator for quantifying the relative magnitude of acceptable and entire flows to regional sinks. The regional assessment of flows is (a) needed to manage material stocks and flows within the region itself, and is (b) required to identify the need for sinks in external regions and to develop suitable strategies to monitor and control external sink loads.

3 PROPOSAL FOR A NEW INDICATOR

This chapter proposes a new indicator, including (1) its definition and interpretation, and (2) the framework to calculate the indicator score.

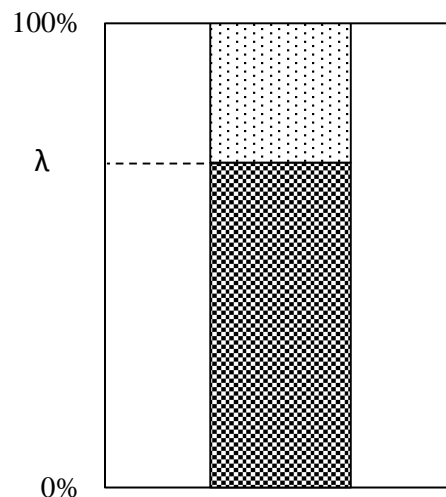
3.1 Indicator definition and interpretation

A new indicator is proposed for assessing substance flows to regional sinks caused by the removal and loss of substances from the anthroposphere. With respect to a specific substance, region and period, the indicator λ quantifies “the acceptable mass share in flows to regional sinks”. Therefore, it determines the relative magnitude between acceptable and entire sink loads:

$$\lambda = \frac{\text{Acceptable sink load}}{\text{Entire sink load}} * 100 \quad \text{Equation 1}$$

Both the acceptable and entire sink loads are given in mass per time. Consequently, the indicator score is dimensionless, ranging from 0% to 100% (Figure 4). Achieving 100% means that the total mass of a substance within sink loads is acceptable. Less than 100% means that there is a certain mass fraction within sink loads that overshoot accepted quality standards. Consequently, the increase of the indicator score up to 100% is seen as good and worth striving for.

Figure 4: The entire sink load represents 100%, whereas the indicator score λ is equal to the acceptable mass share in flows to regional sinks and the complementary amount (100% minus λ) is equal to the unacceptable mass share in flows to regional sinks.



- ☐ Unacceptable mass share in flows to regional sinks
- ▣ Acceptable mass share in flows to regional sinks

To operationalize the qualitative indicator definition from Equation 1, the following definitions are given:

$$\lambda = \frac{\sum F_{a,i}^{Region}}{\sum F_i^{Region}} * 100 \quad \text{Equation 2}$$

$$F_{a,i}^{Region} = \begin{cases} F_{c,i}^{Region} & \text{for } \alpha_i^{Region} \geq 0 \\ F_i^{Region} & \text{for } \alpha_i^{Region} < 0 \end{cases} \quad \text{with} \quad \text{Equation 3}$$

$$\alpha_i^{Region} = F_i^{Region} - F_{c,i}^{Region} \quad \text{Equation 4}$$

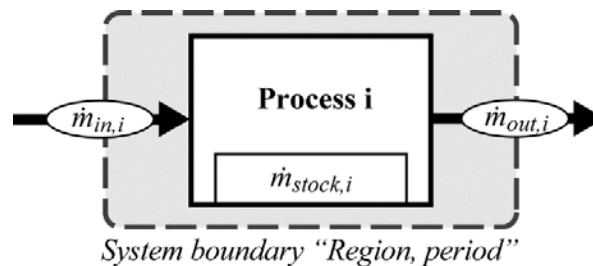
$$F_i^{Region} = \begin{cases} \beta_i & \text{for } \beta_i > 0 \\ 0 & \text{for } \beta_i \leq 0 \end{cases} \quad \text{with} \quad \text{Equation 5}$$

$$\beta_i = \dot{m}_{in,i} - \dot{m}_{out,i} \quad \text{Equation 6}$$

where the index i stands for a process i , $\sum F_{a,i}^{Region}$ is the acceptable sink load, $\sum F_i^{Region}$ is the entire sink load. $F_{a,i}^{Region}$ is an acceptable flow in a region, F_i^{Region} is an actual flow in a region, $F_{c,i}^{Region}$ is a critical flow in a region, α_i^{Region} is the distance-to-target value, β_i is the net flow of process i , $\dot{m}_{in,i}$ is the sum of flows into process i , and $\dot{m}_{out,i}$ is the sum of flows out of process i .

A process i stands for the transportation, transformation and/or storage of goods and substances within spatial and temporal system boundaries (ÖWAV 2003). Figure 5 plots a generic (sub-) process, including the inflows $\dot{m}_{in,i}$, stock changes $\dot{m}_{stock,i}$ and outflows $\dot{m}_{out,i}$.

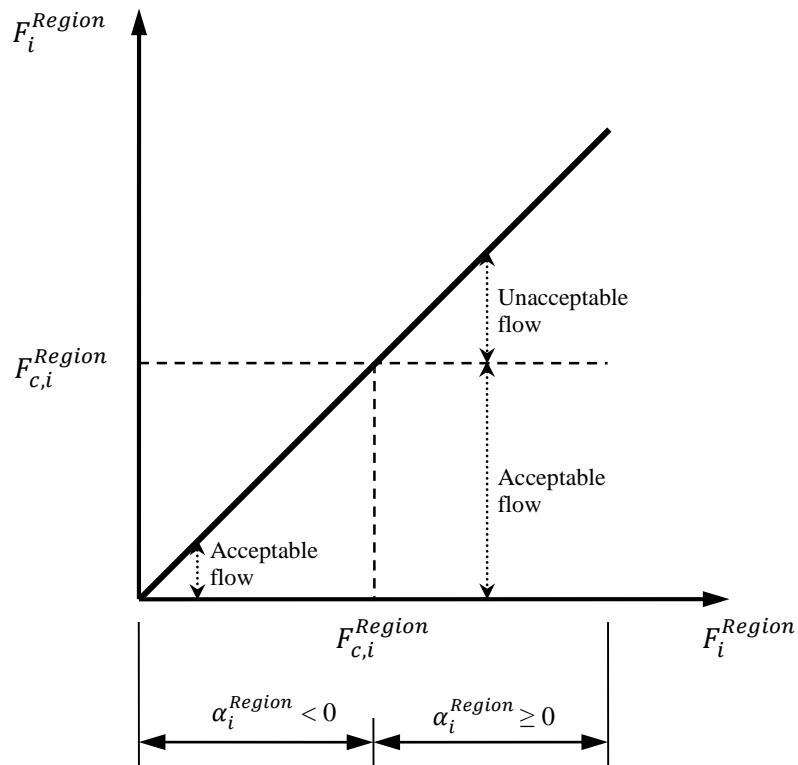
Figure 5: A process i balances inflows $\dot{m}_{in,i}$, stock changes $\dot{m}_{stock,i}$ and outflows $\dot{m}_{out,i}$.



The definition of the net flow β_i is of relevance because the positive net flow or actual flow F_i^{Region} represents mass fractions per time that enter a sink process. This definition avoids double accounting because each mass fraction per time is accounted for once a time. With respect to the proposed indicator, a sink is "a process that accommodates materials that have been removed or lost from the anthroposphere". The definition of the term "sink" has been derived from existing definitions in the field of waste management and environmental chemistry. Based on the findings of a literature survey (see appendix I), the common denominator of various definitions is a set of three features: substance specific, process, removal function.

The determination of the acceptable flow $F_{a,i}^{Region}$ is based on the distance-to-target value α_i^{Region} (Figure 6). If the distance-to-target value $\alpha_i^{Region} < 0$, the acceptable flow is equal to the actual flow. If the distant-to-target value $\alpha_i^{Region} \geq 0$, the acceptable flow is equal to the critical flow.

Figure 6: The classification of the actual flow F_i^{Region} is based on the distance-to-target value α_i^{Region} . If $\alpha_i^{Region} < 0$ then the entire actual flow F_i^{Region} is acceptable. If $\alpha_i^{Region} \geq 0$ then the actual flow F_i^{Region} contains an acceptable and an unacceptable flow.

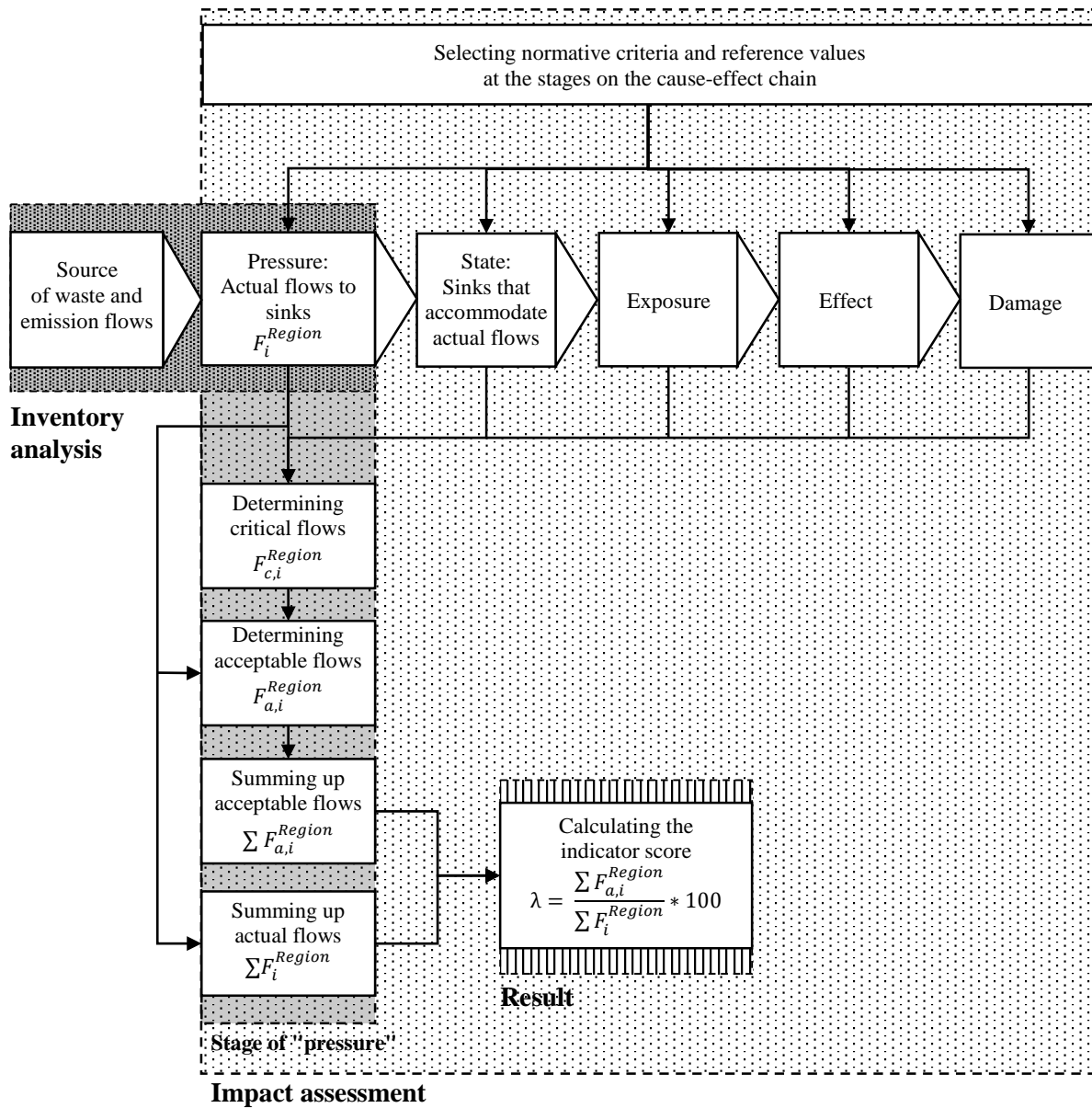


The framework to calculate the indicator score λ is outlined in chapter 3.2, the method for calculating the actual flow F_i^{Region} is described in chapter 3.3, the method for calculating the critical flow $F_{c,i}^{Region}$ can be found in chapter 3.4.

3.2 Framework

The framework results in the score for the proposed indicator λ (Figure 7). Inventory analysis applies the substance flows analysis tool and selects the actual flows F_i^{Region} (chapter 3.3, chapter 4.2). Impact assessment associates normative criteria and reference values to each actual flow, and calculates the critical and acceptable flows $F_{a,i}^{Region}$ (chapter 3.4, chapter 4.3). Finally, the outcomes from inventory analysis and impact assessment are merged in order to compute the indicator score (chapter 5.3).

Figure 7: Framework to calculate the indicator score λ . With respect to a specific substance, region and period, the indicator λ quantifies “the acceptable mass share of flows to regional sinks”.



3.3 Inventory analysis

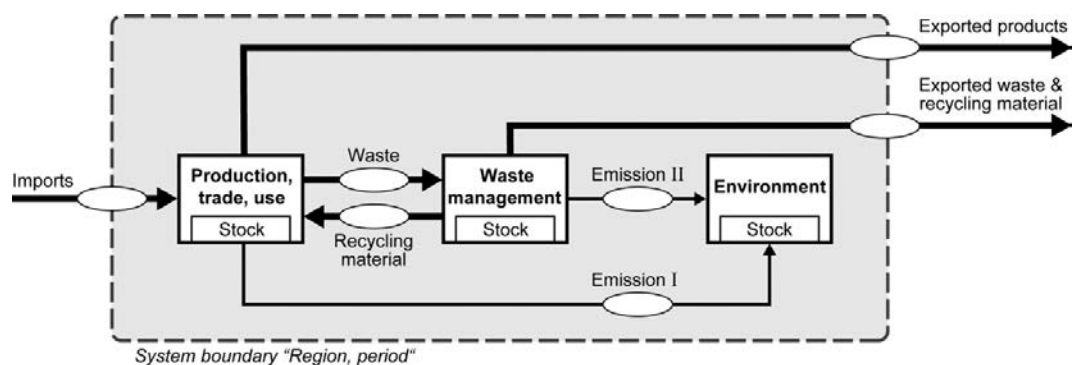
The inventory analysis results in the entire sink load $\sum F_i^{Region}$. To establish the inventory, two major steps are needed: First, inventory analysis is based on the substance flow analysis (SFA) tool (chapter 3.3.1). Second, SFA results are used to quantify the actual flows F_i^{Region} (chapter 3.3.2).

3.3.1 Applying substance flow analysis tool

To investigate the anthropogenic metabolism, SFA has been proven to be a practical tool. It tracks the flow of selected substances through systems such as households, enterprises, cities or regions. The applied methodology is in accordance with Brunner and Rechberger (2004) and includes four steps:

- (i) Setting the scope of the assessment includes the selection of a substance, a reference region and a period of interest. First, the notion “substance” is defined as “matter of constant composition best characterized by the entities (atoms, molecules, formula units) it is composed of” (Nic et al. 2012). This is in line with the SFA framework used in this study, which defines the notion as “any (chemical) element or compound composed of uniform units” (Brunner and Rechberger 2004). Second, the assessment deliberately focuses on region-wide flows instead of flows along a life cycle chain. A region is bounded by administrative limits. Accordingly, communities, cities, federal states, nations or continents are subjects of interest. Third, setting the system boundary in time includes the selection of a reference period (day, week, month, year, decade, and so on) and a reference point in time (e.g. the year 2013). Temporal variations of flows within the reference period are often neglected because of lack of data.
- (ii) Model development focuses on the identification of relevant processes and their links in terms of flows. The qualitative SFA model (Figure 8) is used as a basis to develop the case- study specific, quantitative SFA models (Figure 12, Figure 14 and Figure 16). The critical flows are computed for the actual flows “Waste”, “Emission I” and “Emission II” (see chapter 3.4). The critical flows for “Exported waste & recycling material” are not computed and constitute a part of future research (see chapter 6).

Figure 8: Qualitative stock and flow model as a basis for inventory analysis.



(iii) Model equations and data acquisition: The following model equations are used to calculate each flow and stock:

$$\dot{m}_{flow} = f(p_1, p_2, p_3, \dots, p_j) \quad \text{Equation 7}$$

$$m_{stock} = f(q_1, q_2, q_3, \dots, q_j) \quad \text{Equation 8}$$

where \dot{m}_{flow} is either the inflow $\dot{m}_{in,i}$ to a process i or the outflow $\dot{m}_{out,i}$ from a process i in mass per time, m_{stock} is the stock in terms of mass, p_j are the input parameters for \dot{m}_{flow} , and q_j are the input parameter for m_{stock} , whereas j is an index for each input parameter. The input parameter p_j and q_j are assumed to be normally distributed with $N(m_{p_j}, s_{p_j})$ and $N(m_{q_j}, s_{q_j})$. The mean values m can either be determined by non-process based models (as carried out by Månsson et al. 2009, Ott et al. 2012) or by specific process-based models such as landfill models (e.g. Laner 2011), water quality models (e.g. Mitchell et al. 2010), or traffic emission models (e.g. Hausberger 1997). The standard deviation s is derived from the uncertainty factor uf according to the data vagueness concept from Hedbrant et al. (2001):

$$s_p = m_p * \frac{uf - 1}{2} \quad \text{Equation 9}$$

$$s_q = m_q * \frac{uf - 1}{2} \quad \text{Equation 10}$$

with

$$uf = 1 + 0,0036 * e^{1,105 * l} \quad \text{Equation 11}$$

The uncertainty level l ranges from “1” to “5” and depends on the classification of the data sources.

(iv) Balance equations are applied for each (sub-) process i (see Figure 5):

$$0 = \dot{m}_{in,i} - \dot{m}_{out,i} + \dot{m}_{stock,i} \quad \text{Equation 12}$$

where $\dot{m}_{in,i}$ is the input flow, $\dot{m}_{out,i}$ is the output flow, $\dot{m}_{stock,i}$ is the alteration of stock. Since multiple data sources are used, data quality and quantity is heterogeneous. Consequently, contradictions in fulfilling the mass balance criteria occur. To overcome this gap, the freeware STAN (Cencic 2012) is used, including data reconciliation based on the error propagation law.

3.3.2 Quantifying actual flows

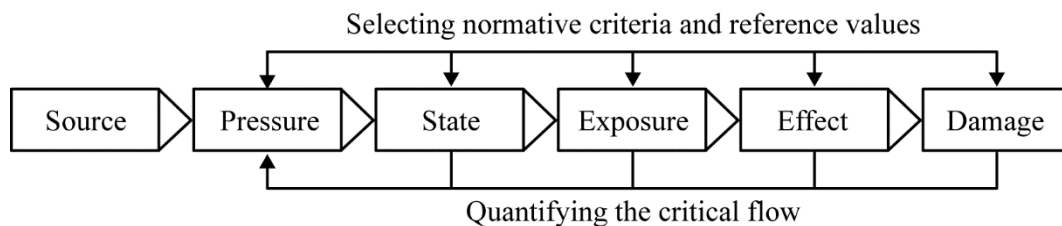
The actual flows F_i^{Region} are selected based on SFA results. Therefore, each sub-process is either related to the process “Production, trade & use”, to the process “Waste management”, or to the process “Environment”. Hence, the net flow β_i of each sub-process i is calculated and plotted. This kind of plot allows the comparisons of various stock and flow diagrams in a comparable manner. A positive net flow ($\beta_i > 0$) indicates a sink process, and a negative net flow ($\beta_i < 0$) indicates a source process.

In view of the impact assessment (chapter 3.4), the distance-to-target weighting refers to the stage of "pressure". The stage of "pressure" is identified where a positive net flow enters a sub-process in the processes “Waste management” and “Environment” (see Figure 8). The positive net flow β_i is defined as the actual flow F_i^{Region} .

3.4 Impact assessment

The impact assessment results in the acceptable flows $\sum F_{a,i}^{Region}$. Therefore, the framework to determine a critical flow $F_{c,i}^{Region}$ is presented in Figure 9, including (1) the selection of normative criteria and reference values, referring to a specific stage in the cause-effect chain (chapter 3.4.1) and (2) the quantification of the critical flow, by varying the corresponding actual flow as long as the reference value is achieved (chapter 3.4.2).

Figure 9: Framework to quantify a critical flow referring to an actual flow. The stages throughout the cause-effect chain of substances are in the boxes, the models that link the stages are represented by arrows.



3.4.1 Selecting normative criteria and reference values

Normative criteria and references values are derived from goal-oriented frameworks such as legal framework, standards, political agreements, or agreements among stakeholders. Their selection depends on circumstances in the case study region. The circumstances might differ from region to region and might change over time, for example, as a consequence of new scientific knowledge, by improved data availability, by changes in the ethical value-sphere, by different political agreements for accepted emission rates, and by environmental quality standards. To select normative criteria and reference values in a specific region, the outcome of stakeholders’ involvement might be

considered, without being further discussed in this thesis. Examples for normative criteria and reference values are given in Table 3.

Table 3: Examples for normative criteria and reference values in the context of the cause-effect chain, from the source of waste and emissions towards the damage (adopted from Hofstetter 1998, Frischknecht 2009).

Stage	Stage description	Normative criteria	Reference values
Source	Potential of waste and emissions	n.r.	n.r.
Pressure	Flow to sink	Emission rates Waste into landfill [mass per time]	Legal limits, political agreements
State	Substance fate in a) natural and b) man-made sinks.	Substance concentration, accumulation or transformation rate in a) environmental media, and b) recycling goods, landfills, underground storage facilities.	Geogenic reference values in soil; Approved landfill capacity and service time
Exposure	Standard characteristics of exposed organism	Exposed dose, collective effective dose	MAK values, BAT values
Effect	Dose-Response-Relationship	Number and type of human diseases, Number of vanishing plant species	Accepted number of cases regarding a human disease
Damage	Damage to human health or ecosystem quality	DALYs, Share of vanishing plant species per area and time unit.	Value weighted DALYs, Accepted share of vanishing plant species per area and time unit

Notes: n.r. = not relevant; MAK values = Maximum Concentrations at the Workplace, BAT values = Biological Tolerance Values; DALY = disability-adjusted life years or disease-adjusted life years;

Normative criteria determine the acceptance of flows into sinks in a qualitative manner. A criterion can be located at any stage throughout the cause-effect chain of a substance, ranging from the pressure stage to the damage stage. At the stage of “pressure”, the actual flow enters the sink. Normative criteria can be, for instance, substance concentration in waste flows and emission rates. The stage of “state” includes the fate of substances within the sink process. With respect to natural sinks, the normative criteria can be, for example, the substance concentrations and the accumulation rate in environmental compartments. With respect to man-made sinks, normative criteria for landfills can be the waste accumulation rate. For recycling goods it can be the substance concentration, and for incinerators it can be the efficiency to mineralize organic substances. The stage of “exposure” refers to the adsorption of a substance by a receptor. In terms of human health, the normative criteria can be, for example, the exposed dose. The stage of “effect” includes a dose-response relationship and yields effects on humans and environment like the number of cancer cases in human population. The stage of “damage” combines the effects with a weighting scale. In terms of human health, this might be the DALY (disability-adjusted life years or disease-adjusted life years) concept. It summarizes the time in which people are disabled or summarizes the time people have lost due to death.

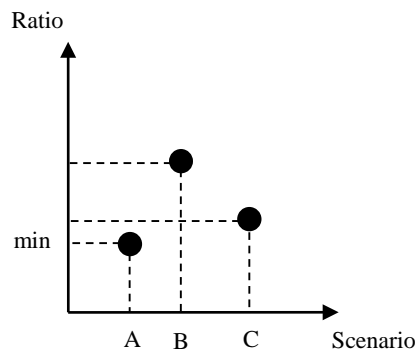
Reference values quantify the acceptance of flows into sinks. It relates to a specific normative criterion. For a single normative criterion, multiple reference values are possible. For instance, the substance concentration in environmental media can refer either to legal limits or to the geogenic background concentration.

3.4.2 Quantifying critical flows

The critical flow refers to the stage of “pressure” as the actual flow does. To determine the critical flow rate in mass per time, the actual flow is varied until the associated reference value is achieved. The causality between the actual flows and the reference values is determined by specific models. In general the type of model depends on the type and characteristic of the normative criteria. If the normative criteria are at the stage of “pressure”, the model is rather simple. The critical flow is either directly given by e.g. law in terms of a flow in mass per time, or it can be calculated based on legal substance concentration and the actual flow on the level of goods.

The quantification of the critical flow might become more complicated if the normative criteria are applied at the stage of “state”, “exposure”, “effect” or “damage”. The location determines to what extent the cause-effectchain has to be analysed. *State criteria* require fate analysis in the respective sink. Natural sinks have to be analysed with single-media or multi-media models (Hertwich et al. 2002). Man-made sinks have to be analysed with SFA models that pose an extension of the SFA model for inventory analysis. For instance, landfills can be investigated (e.g. Laner 2011) in order to quantify the accumulation rate of substances in the landfill body. *Exposure criteria* need a pathway and exposure analysis in view of a certain receptor. The receptor-based approach quantifies substance concentrations that reach, for instance, people. Exposure scenarios include the intermediate transfer ratio between the environmental media and the receptor, the uptake factor, the exposure frequency, duration and an average time for the exposed receptor. *Effect criteria* require the definition of a dose-response relationship and yield the effect, for instance, on human health in terms of number of people affected by cancer. *Damage and risk criteria* cause the most effort because analysis refers to the entire cause-effect chain, from the origin source towards the risk for human and environmental health. In the past, several tools have been developed that quantify *risk criteria* including multi-media, exposure and risk analysis (e.g. for human health, see McKone and Enoch 2002). In this case, an infinite number of critical flows is possible, for instance, as a consequence of flows into three environmental compartments. To reduce the number of possibilities, each actual flow is varied while the two others remain constant as long as the reference values is fulfilled. Accordingly, three actual flows result in three scenarios (A, B, C). The most stringent scenario is determined by the minimum ratio between the critical and the actual flow, and becomes selected (Figure 10).

Figure 10: Each scenario yields a ratio between the critical flow and the actual flow. To select the critical flow for indicator computation, the scenario with the minimum (min) ratio is determinant.



Two general rules are defined for calculating the critical flow:

First, the effort to analyse the cause-effect chain might be reduced if the reference value is achieved at an earlier stage in the cause-effect chain. For instance, if the legal limits for heavy metal concentrations in the receiving water (stage of “state”) are already detected in the effluent from the waste water treatment plant to the receiving water (stage of “pressure”). In this case, the fate analysis in the environment can be skipped.

Second, the most stringent normative criteria and reference value leads to the least critical flow. This specification determines the critical flow.

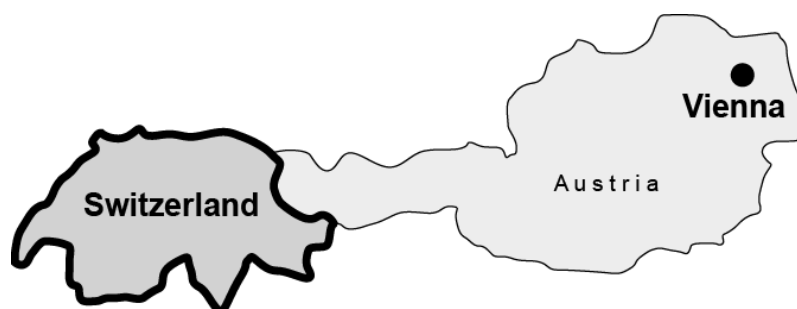
4 CASE STUDIES

This chapter presents three case studies, including (1) the reasons for selecting these case studies, (2) the results from inventory analyses, and (3) details about impact assessment. The ultimate result in terms of the indicator score can be found in chapter 5.

4.1 Selection of case studies

The indicator score is calculated in three case studies, including Copper (Cu) and Lead (Pb) on the city level and Perfluorooctane Sulfonate (PFOS) on the national level (Figure 11). For the metals, the city level is of relevance because the anthropogenic metal density (mass per spatial unit) in urban areas is larger than in rural areas (e.g. van Beers et al. 2007). Consequently, urban waste and emission flows are likely to cause higher metal contents and thus higher sink loads than those in rural areas. For PFOS, the national level is of relevance because the European Union urges member states to implement strategies for careful PFOS management. The two metals (Cu and Pb) and the organic substance (PFOS) have been selected in order to demonstrate the need for different man-made sinks in dependence on geogenic and man-made substances.

Figure 11: The case studies focus on Cu and Pb on the city level (Vienna) and on PFOS on the national level (Switzerland)



In detail,

- Cu is relevant from both a resource use and environmental impact viewpoint. On the one hand, Cu is essential for modern lifestyles, resulting in Cu waste fractions that are disposed of in man-made sinks. From 1900-2000, about 0.7% of the Swiss Cu stock have been annually discarded to landfills (Wittmer 2006), which pose a future resource deposit. On the other hand, Cu is emitted from point and non-point sources to natural sinks and poses a risk for aquatic life. Apart from the rural areas, urban areas are of major relevance. For instance, in Germany about 30% of total Cu loadings in receiving waters originate from urban areas (Böhm et al. 2001). In Vienna, Cu concentrations in sewage sludge are significantly larger than in rural areas (Kroiss et al. 2008), and Cu concentrations in urban soils are higher than in surrounding rural areas (Pfleiderer 2011).

- Pb and Pb compound emissions directly affect human health. In Austria, Pb emissions to air decreased from 218 tons per year (t/yr) in the year 1990 to 13 t/yr in 2009 (Anderl et al. 2011). In 1993 lead was banned from the Austrian petrol market. It is still used in accumulators, building coatings, tires and paints. Due to former Pb depositions and present diffusive losses, anthropogenic Pb is found in urban soils (Kreiner 2004). Hence, it can be transferred to fodder and food, and may affect human health (WHO 2007).
- Perfluorooctane sulfuric acid and its derivatives, collectively named PFOS, are persistent bio-accumulative and toxic substances. They are regulated under the Persistent Organic Pollutants Regulation 850/2004 (European Parliament 2004) and Regulation 2006/122/EG (European Parliament 2006). In 2010, PFOS has been added to the convention with some exemptions for specific applications (European Commission 2010). The European Union (EU) urges member states to implement strategies for careful PFOS management. Even though Switzerland is a non-EU member state, comprehensive data about anthropogenic stocks and flows are readily available.

The temporal system boundaries are defined on an annual basis. The reference years include the years 2007 and 2008 due to data availability.

4.2 Inventory analysis

To determine the inventory entries, two steps are needed. (1) The substance flow analysis tool is applied. (2) The actual flows are selected. This section highlights the results of both steps. Further details about the calculation including explanations and data are given in article II (Kral et al. 2014) and article III (Kral et al. 2014).

For Cu in Vienna, Figure 12 represents the stocks and annual flows for the year 2008. The actual flows are the positive net flows to sub-processes within the process “Waste management” and “Environment” (Figure 13). Calculating the sum of actual flows yields 1,129 t Cu/yr. Thereof, 97.3% are disposed of in a local landfill, 1.5% are shipped to a foreign underground storage facility, 0.8% are deposited on urban soil, and 0.4% enter receiving waters.

Figure 12: Copper flows and stocks in the city of Vienna for the year 2008. Annual flow rates and changes in stocks are given in tons per year (t/yr), for stocks in tons (t). The flows are represented as Sankey arrows proportional to the flow rate; figures for stocks are given within the process boxes. Deviations from mass balance are due to rounding.

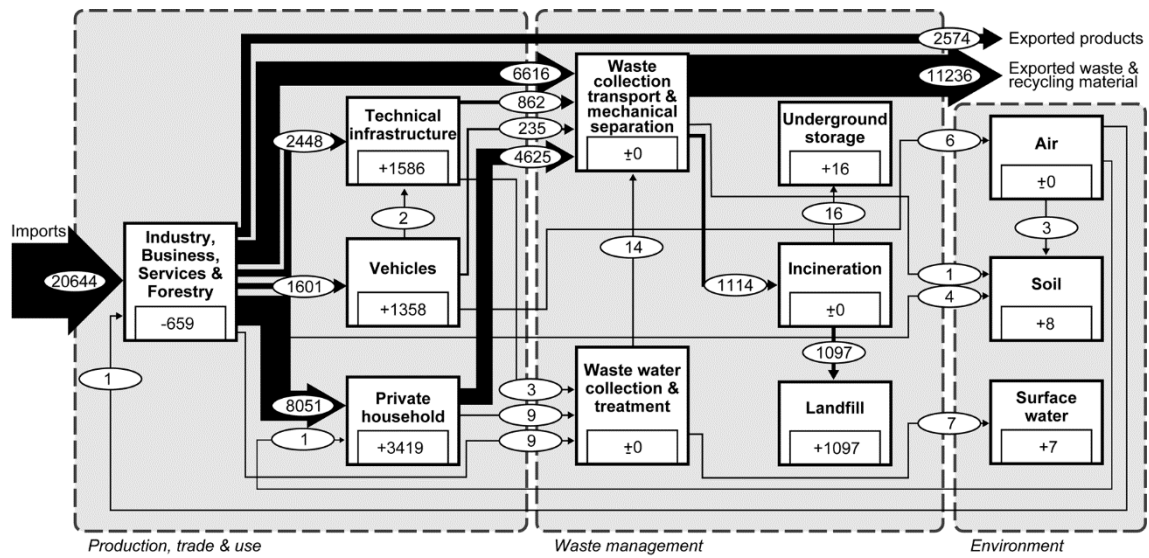
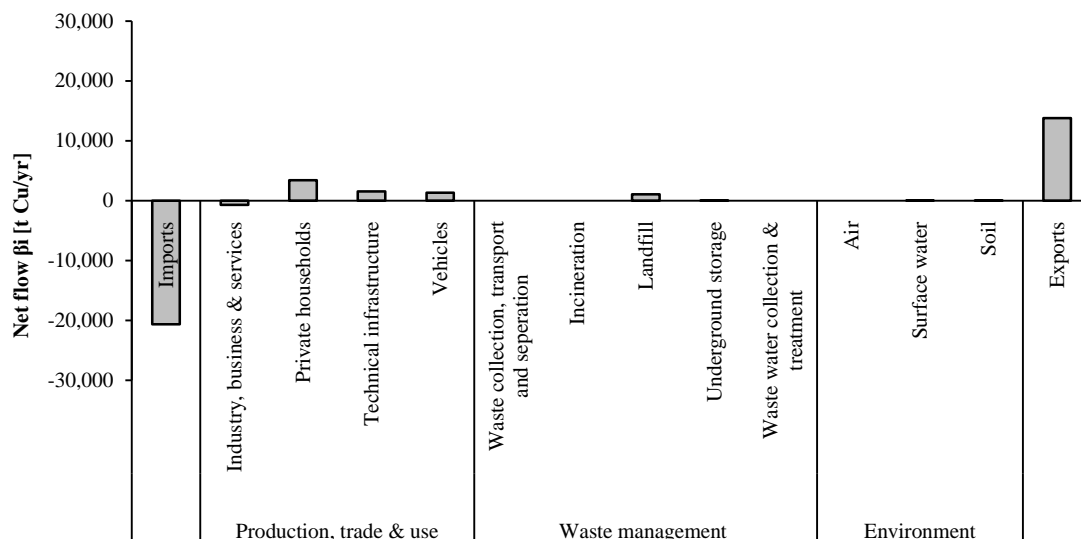


Figure 13: Copper net flow β_i for each sub-process, categorized into the processes “Production, trade & use”, “Waste management” and “Environment”. The sum of net flows β_i results in zero.



For Pb in Vienna, Figure 14 represents the stocks and annual flows for the year 2008. The actual flows are the positive net flows to sub-processes within the process “Waste management” and “Environment” (Figure 15). Calculating the sum of actual flows yields the sum of actual flows with 191 t Pb/yr, of which 75.4% entered a local landfill, 22.9% entered a foreign underground storage facility, 0.8% entered ambient air, 0.5% entered urban soil, and 0.3% entered receiving waters.

Figure 14: Lead flows and stocks in the city of Vienna for the year 2008. Annual flow rates and changes in stocks are given in tons per year (t/yr), for stocks in tons (t). The flows are represented as Sankey arrows proportional to the flow rate; figures for stocks are given within the process boxes. Deviations from mass balance are due to rounding.

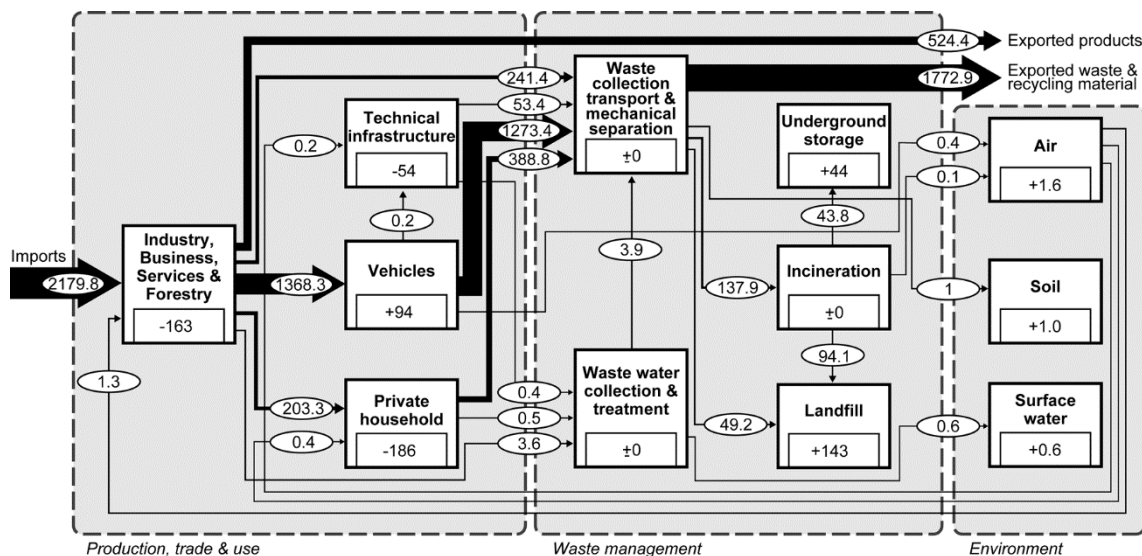
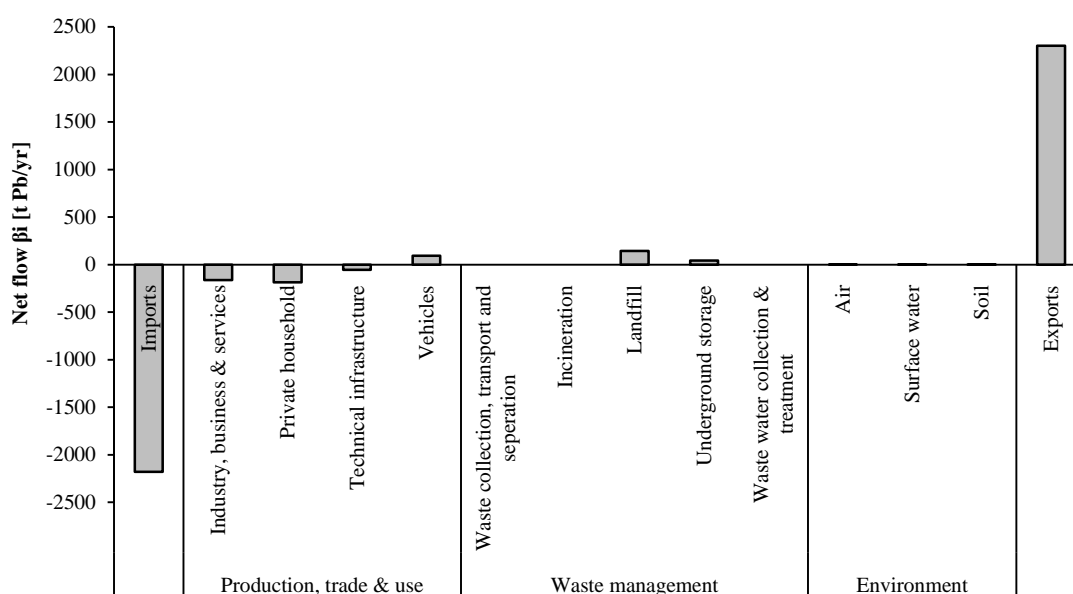


Figure 15: Lead net flow β_i for each sub-process, categorized into the processes “Production, trade & use”, “Waste management” and “Environment”. The sum of net flows β_i results in zero.



For PFOS in Switzerland, Figure 16 represents the stocks and annual flows for the year 2007. The actual flows are the positive net flows to sub-processes within the process “Waste management” and “Environment” (Figure 16). Calculating the sum of actual flows yields 2,260 kg PFOS/yr, of which 77.1% entered incineration, 0.4% entered landfills, and 22.5% entered geogenic sinks within the hydrosphere, atmosphere and soil.

Figure 16: PFOS flows and stocks in Switzerland for the year 2007. Annual flow rates and changes in stocks are given in kilograms per year (kg/yr), for stocks in kilograms (kg). The flows are represented as Sankey arrows proportional to the flow rate; figures for stocks are given within the process boxes. Deviations from mass balance are due to rounding.

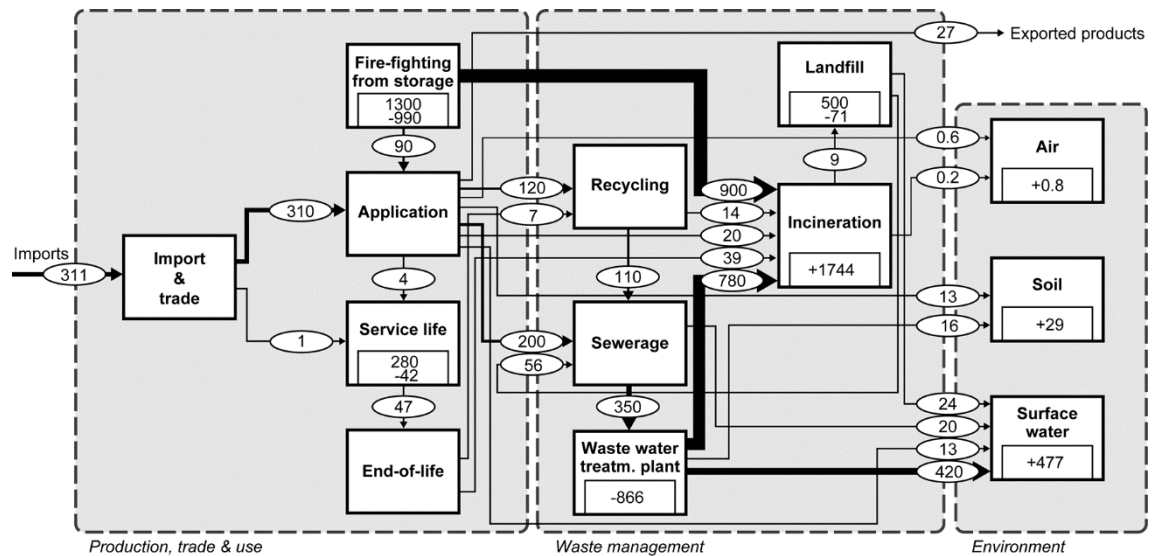
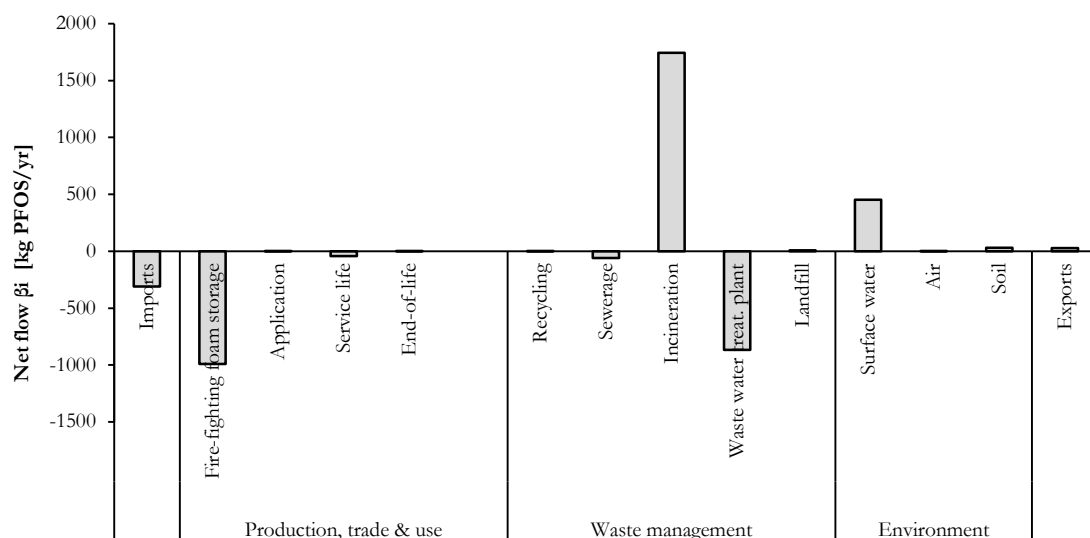


Figure 17: PFOS net flow β_i for each sub-process, categorized into the processes “Production, trade & use”, “Waste management” and “Environment”. The sum of net flows β_i results in zero.



4.3 Impact assessment

The chapter includes (1) the selection of normative criteria and reference values, and (2) the calculation of the critical flows. Further details about the calculation including explanations and data are given in article III (Kral, Brunner et al. 2014).

4.3.1 Selecting normative criteria and reference values

Table 4 gives an overview of the normative criteria and corresponding reference values, based on each case study. The normative criteria refer to three different stages on the cause-effect chain.

Table 4: Normative criteria and reference values associated to actual flows.

Scope	Actual flow	Sink	Normative criteria and reference values at the stage of		
			Pressure	State	Damage
Cu Vienna 2008	Bottom-ash from MSW incineration	Landfill	-	Accumulation rate 2,075.5 t/yr	-
	APC residues from MSW incineration (exported filter cake)	Underground storage facility	Mass flow 16.4 t/yr	-	-
	Stormwater overflow from mixed sewer system	Surface water	Legal standard 9.3 µg Cu / l	-	-
	Effluent from WWTP	Surface water	Legal standard 9.3 µg Cu / l	-	-
	Deposition, Fertilizer, Pesticides, Compost	Soil	-	Accumulation rate 0 t/yr	-
Pb Vienna 2008	Bottom-ash from MSW incineration	Landfill I	-	Accumulation rate 198.5 t/yr	-
	Excavated soil	Landfill II	-	Accumulation rate 4.3 t/yr	-
	Demolition waste	Landfill III	-	Accumulation rate 29.8 t/yr	-
	APC residues from MSW incineration (exported filter cake)	Underground storage facility	Mass flow 43.9 t/yr	-	-
	Emissions	Air	-	-	-
	Stormwater overflow from mixed sewer system, Runoff from separated sewer system, Effluent from WWTP	Surface water	-	-	Human health risk Risk level: 10 ⁻⁶ Hazardous index: 1
PFOS Switzerland 2007	Solid waste & sewage sludge	Incineration	-	Mineralization rate 99.475245%	-
	Bottom-ash from MSW incineration	Landfill	Mass flow 0.009 t/yr	-	-
	Effluent from WWTP	Surface water	Provisional health advisory 200 ng PFOS / l	-	-
	Emissions from other sources	Surface water	n.a.	n.a.	n.a.
	Emissions	Soil	n.a.	n.a.	n.a.
	Emissions	Air	n.a.	n.a.	n.a.

Note: Cu = copper, Pb = lead; PFOS = Perfluorooctane Sulfonate; - = not relevant; n.a. = not available due to a lack of normative criteria; APC = air pollution control; WWTP = waste water treatment plant; µg Cu / l = 10⁻⁶ gram Cu per liter; ng PFOS / l = 10⁻⁹ gram PFOS per liter;

Pressure criteria are associated with actual flows into

- underground storage facilities. Air pollution control (APC) residues from municipal solid waste (MSW) incineration are classified as hazardous waste. Because adequate landfill sites are missing in the case study regions, the waste is exported to Germany, filled into big-packs and stored in salt mines underground (Prognos 2012). In Germany, the salt mines are classified as long-term storage facilities. The acceptance criteria for disposal waste are defined by exclusions criteria according to §7 of the Federal Waste Act (Bundesrepublik

Deutschland 2009). Assuming that the disposal practice complies with German law, it is just a question of societal and political acceptance in the exporting region if the APC residues should be disposed of in underground salt mines. This acceptance is given in the case study regions. Both Switzerland and Vienna agree to the waste export. So, the entire flow into the man-made sink is classified as accepted, which is in accordance with the Ecological Scarcity method (Frischknecht, Steiner et al. 2009).

- surface waters. Cu in Vienna enters the River Danube via storm water overflow from the mixed sewer system and effluents from WWTP. The normative criterion is the Cu concentration in the water flow, and the reference value is determined with $9.3 \cdot 10^{-6}$ gram Cu per liter water in accordance with environmental standards (Republik Österreich 2006). PFOS in Switzerland is detected in effluents from WWTP. Swiss limits regarding PFOS concentrations are actually missing but under development (Götz et al. 2011). Consequently, the reference value of $200 \cdot 10^{-9}$ gram PFOS per liter water is based on the provisional health advisory for drinking water, published by U.S. EPA (2009).

State criteria are associated with actual flows into

- landfills. The acceptance for waste flows into sanitary landfills is defined in national waste acts. According to the Swiss (Schweizer Bundesrat 2011) and Austrian (Republik Österreich 2008) regulation, the acceptance includes waste quality criteria such as the organic carbon content, calorific value, the share of soluble salts, and heavy metal concentrations. However, these regulations determine possible *pressure criteria*. They are not used in the case studies because data quantity and quality regarding waste characteristics were not available to its' full extent. For Cu and Pb in Vienna, multiple *proxy criteria* are replaced by a single *state criteria*, which is determined by the constraints imposed by official permits for each landfill. The normative criteria is the waste accumulation rate in the landfill. The reference value is quantified by relating the approved and remaining landfill volume to the remaining service time.
- soil. The Swiss Regulation on the Impact on Soils (Schweizer Bundesrat 1998) aims to ensure long-term soil fertility. Accumulation of heavy metals in soil is not accepted. Consequently, the criterion is the accumulation rate in soil, and the reference value is set at zero. This approach is in accordance with the Ecological Scarcity Method (Frischknecht, Steiner et al. 2009) and is selected for the case studies due to a lack of more precise data.
- incinerators. The criterion is determined by the efficiency of the thermal treatment process to mineralize PFOS. The reference value is the mineralization rate, which stands for the mineralized PFOS mass fraction in relation to the entire PFOS flow into the incinerator.

Damage criteria are associated with actual flows into air, surface water and soil. For Pb in Vienna, the flows into natural sinks are assessed in view of human health risks. In accordance with McKone and Enoch (2002), the Risk Level (RL) and the Hazardous Index (HI) are selected as normative criteria. The RL considers carcinogenic risk, and the HI non-carcinogenic risk on human health. To demonstrate the method, widely-used acceptable risks of 10^{-6} for RL and 1 for HI are selected as reference values, without discussing further acceptable risks (Kelly 1991).

No criteria are associated with actual flows without available criteria, for instance, as a consequence of missing data or legal standards. Due to the lack of regulations and standards, the PFOS flows into soil, air and partly into water are excluded from impact assessment. Nevertheless, the inventory includes these flows for calculating the sum of actual flows.

4.3.2 Quantifying critical flows

To determine the critical flows in mass per time, the actual flow is varied as long as the associated reference value is achieved. The link between the actual flows and the reference values is presented for each criterion:

Pressure criteria refer to flows into

- underground storage facilities. The critical flow is equal to the actual flow [mass per time] (see chapter 4.3.1).
- surface waters. To calculate the critical flow, the reference value [mass per volume] is multiplied by the amount of water [volume] to surface waters.

State criteria refer to flows into

- landfills. The critical flow is equal to the accepted waste accumulation rate in the respective landfill. The accepted rate is defined as the available landfill volume [volume], multiplied with a specific waste density [mass per volume] and divided by the remaining service time for disposal [time].
- soil. The accumulation rate of Pb and Cu in soil is set at zero. To achieve this reference value, the inflow balances the outflow [mass per time]. According to the Ecological Scarcity method, the critical flow into soil is equal to the uptake of heavy metals by plants. The calculation of the uptake by plants is based on land use and plant uptake data. This simplified approach, which neglects leaching from the soil, is justified due to a lack of more precise data.
- incinerators. To calculate the critical flow, the PFOS flow [mass per time] into the incinerator is multiplied by the mineralization rate [percent].

Damage criteria refer to Pb flows into air, soil and water. The risk assessment model CalTOX 4.0 beta (McKone and Enoch 2002) has been developed to assess human exposures from continuous emissions to multiple environmental media. The model is used to quantify both the *damage criteria* RL and the HI. To calculate the critical flows, the actual flows are varied as long as the *damage criteria* result in the predefined reference values (see chapter 4.3.1). Each scenario is based on the variation of a one actual flow out of three, while the two remaining flows are kept constant. Each variation results in two critical flows. One meets the acceptable RL, another meets the acceptable HI. To conclude, three actual flows result in six scenarios. The minimum ratio between the critical flow and the actual flow represents the most stringent scenario, which determines the selection of the critical flow.

5 RESULTS AND DISCUSSION

The chapter gives answers to the research question raised in chapter 1.2.

5.1 What are substance flows to sinks?

The physical movement of substances between single processes is called substance flow, whereas the unit is mass per time (Brunner and Rechberger 2004). A process stands for the accumulation, transport and transformation of substances, thereby located in the anthroposphere or environment. A “sink” is a process too. The term is widely used in environmental chemistry and waste management (see appendix I). With respect to the definitions already given, a new definition is given for the term “sink” with "a process that accommodates materials that have been removed or lost from the anthroposphere". Natural sinks are part of biogeochemical cycles (e.g. Abeles et al. 1971, Molina et al. 1974, ICSU 1989, Paterson et al. 1996, Fong et al. 2000, Berg et al. 2004, Feichter 2008, Yanai et al. 2013). Man-made sinks include technologies such as incinerators, sanitary landfills, and sewage treatment plants (e.g. Brunner 1999, Vogg 2004, Zeschmar-Lahl 2004, Morf et al. 2005, Brunner et al. 2012, Heiskanen 2013). In a time perspective, the material can be temporarily or permanently excluded from the anthropogenic material cycle. Temporarily refers, for instance, to waste stored in landfills (Bertram 2012) that might become a valuable resource in the future (e.g. Johansson 2013) or to substances stored in environmental media that might re-enter the anthropogenic material cycle (e.g. Salomons 1998, Bogdal et al. 2009). Permanently refers, for instance, to organic substances that are mineralized in incinerators (e.g. Van Caneghem et al. 2010).

5.2 How can the indicator be defined and quantified?

The indicator assesses substance flows to regional sinks, caused by the removal and loss of substances from the anthroposphere. With respect to a specific substance, region and period, the indicator quantifies the relative magnitude between acceptable and entire flows to regional sinks. The indicator score is dimensionless, ranging from 0% to 100%. Achieving 100% means that the total mass of a substance within flows to regional sinks is acceptable. Less than 100% means that there is a certain mass fraction within flows to regional sinks that overshoot accepted quality standards. Consequently, the increase of the indicator score up to 100% is seen as good and worth striving for.

The calculation of the indicator score includes inventory analysis based on substance flow analysis, and impact assessment based on a distance-to-target approach. The inventory provides actual flows to sinks, whereas the impact assessment provides acceptable flows to sinks. The aggregation of single flows results in the proposed indicator.

5.3 What is the indicator score for selected case studies?

This chapter presents the case study results by giving (1) a brief overview across all case studies, and (2) details on an individual case study basis.

5.3.1 Overview

The major results refer to the indicator score λ , which quantifies acceptable mass share in flows to regional sinks (Figure 18). The indicator score is 99% for Cu in Vienna, 92% for lead in Vienna, and 96% for PFOS in Switzerland.

Figure 18: Indicator score λ , given in percent (%), for each case study.

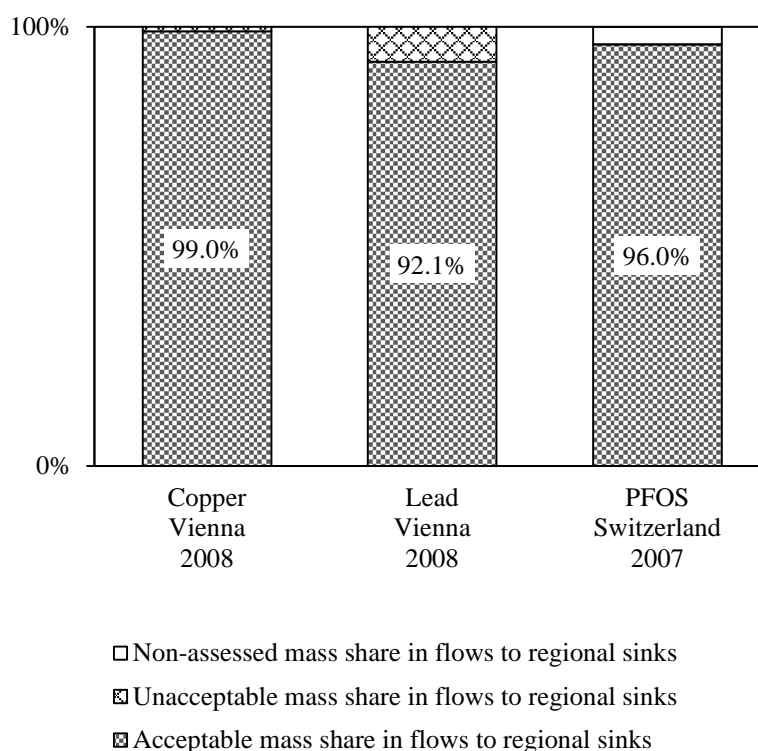


Table 5 includes the results from inventory analysis (actual flows), impact assessment (critical and acceptable flows), and lists the ratio between acceptable and actual flows. Therefore, the Cu metabolism in Vienna is limited by the storm water overflow from the mixed sewer system to receiving waters and by flows to urban soil. The Pb metabolism in Vienna is mainly constrained by excavated soil flows into landfills. The PFOS metabolism in Switzerland partly lacks normative assessment due to lack of data.

Table 5: Results from inventory analysis and impact assessment, on a case study base.

Case study	Sink	Actual flow name	Actual flow	Critical flow	Acceptable flow	Ratio $F_{a,i}$ to F_i []	
			F_i [t/yr]	$F_{c,i}$ [t/yr]	$F_{a,i}$ [t/yr]		
Cu	Landfill	Bottom-ash from MSW incineration	1,097.2	2,075.5	1,097.2	1.00	
	Underground storage facility	APC residues from MSW incineration (exported filter cake)	16.4	16.4	16.4	1.00	
	Surface water	Stormwater overflow from mixed sewer system	3.9	0.3	0.3	0.08	
	Surface water	Effluent from WWTP	1.6	1.7	1.6	1.00	
	Soil	Deposition, Fertilizer, Pesticides, Compost	8.8	1.1	1.1	0.13	
		Sum		1,127.9	-	1,116.6	0.99
	Pb	Landfill I	Bottom-ash from MSW incineration	94.7	198.5	94.7	1.00
Landfill II		Excavated soil	17.3	4.3	4.3	0.25	
Landfill III		Demolition waste	31.9	29.8	29.8	0.93	
Underground storage facility		APC residues from MSW incineration (exported filter cake)	43.9	43.9	43.9	1.00	
Air		Emissions	1.6	6.1	1.6	1.00	
Surface water		Stormwater overflow from mixed sewer system	0.6	0.6	0.6	1.00	
		Runoff from separated sewer system					
Soil		Effluent from WWTP					
		Compost		1.0	1.0	1.0	1.00
		Sum		190.9	-	175.8	0.92
PFOS	Incineration	Solid waste and sewage sludge	1.744	1.744	1.744	1.00	
	Landfill	Bottom-ash from MSW incineration	0.009	0.009	0.009	1.00	
	Surface water	Effluent from WWTP	0.420	0.737	0.420	1.00	
	Surface water	Emissions from other sources	0.060	n.a.	n.a.	n.a.	
	Soil	Emissions	0.029	n.a.	n.a.	n.a.	
	Air	Emissions	0.001	n.a.	n.a.	n.a.	
		Sum		2.263	-	2.173	0.96

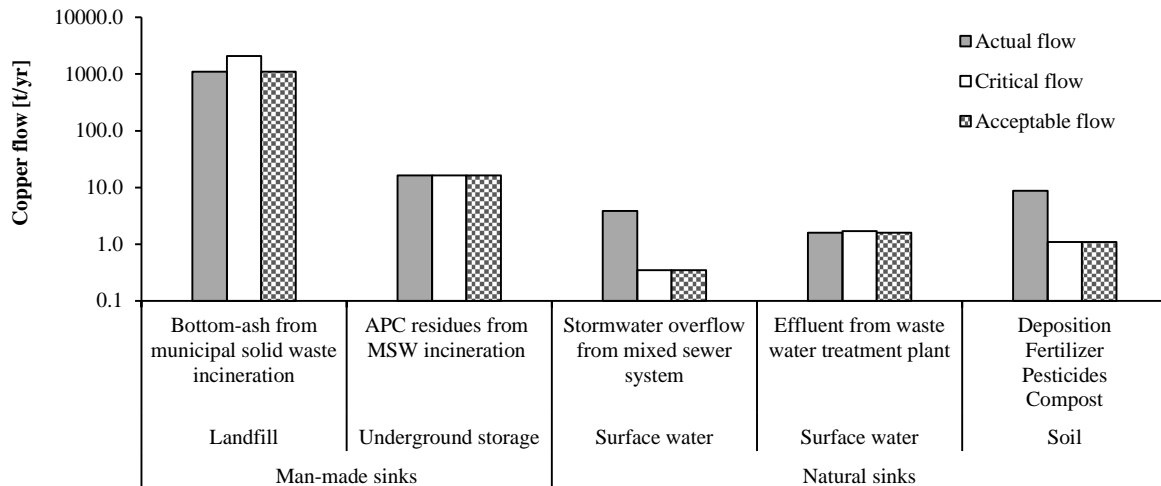
Note: - = not relevant, APC = air pollution control; WWTP = Waste water treatment plant; MSW = municipal solid waste

In the following chapters, the results are discussed on a case study basis, for Cu in Vienna in 2008, for Pb in Vienna for 2008, and for PFOS in Switzerland in 2007.

5.3.2 Cu in Vienna

In Vienna, 99.0% of Cu flows to regional sinks are acceptable. The sink “soil” poses a constraint for 0.7% of all actual Cu flows only. The sink “surface water” poses a constraint for 0.3% of all actual Cu flows.

Figure 19: Case study results from inventory analysis (actual flows) and impact assessment (acceptable and critical flows) regarding Cu in Vienna for the year 2008.



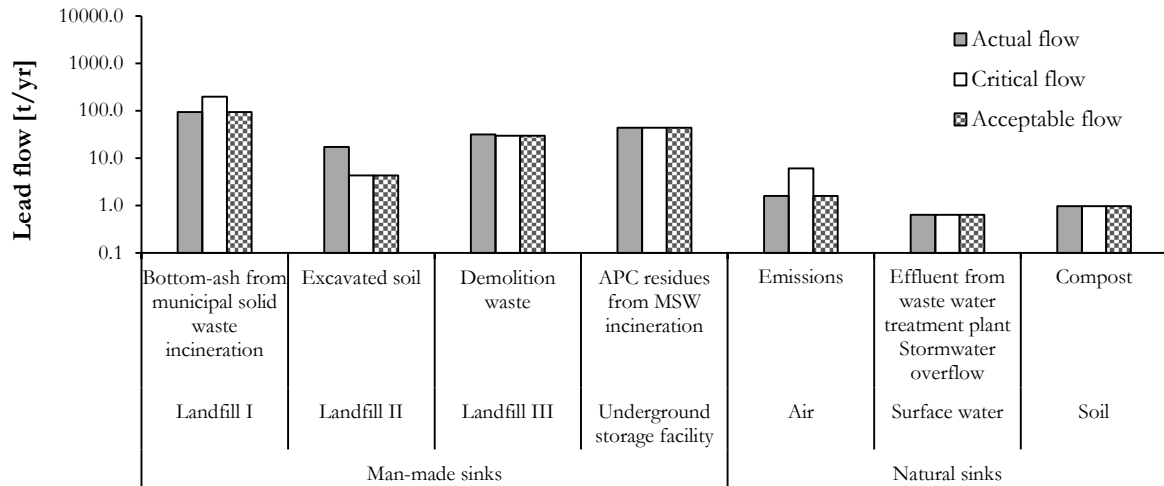
The following results have been obtained (Figure 19), whereas the percentage numbers refer to the sum of actual flows ($100\% \cong 1.128$ t/yr):

- Landfill: 97.3% of flows are due to bottom-ash from MSW incineration. This is in compliance with landfill regulation. Calculated by means of the approved volume and service time of the landfill, the critical flow is 189% higher than the actual flow. Consequently, there is no constraint for the disposal of bottom-ash until the end of the approved service time.
- Underground storage facility: 1.5% of flows are due to APC residues from MSW incineration. These residues are exported and disposed of in approved underground storage facilities.
- Water: 0.1% of flows are from effluents from the waste water treatment plant (WWTP), fulfilling quality standards for surface waters. However, the actual flow is only 6% below the critical flow. Besides WWTP effluents, 0.3% of flows are within storm water overflow from mixed sewer systems to receiving waters. Applying the same standards as for effluents shows that the actual flow is 11 times higher than the critical flow.
- Soil: 0.1% of flows are acceptable for urban soil. 0.7% of flows are unacceptable because they exceed the critical level by a factor of eight. Cu flows into the soil are higher than those caused by removal by plants. If no Cu leaching from soil is assumed, Cu will accumulate in specific soil compartments. The determination of the critical flow was rather crude due to a lack of accurate local data.

5.3.3 Pb in Vienna

In Vienna, 92.1% of Pb flows to regional sinks are acceptable. The sink “landfill” poses a constraint for 7.9% of all actual Pb flows.

Figure 20: Case study results from inventory analysis (actual flows) and impact assessment (acceptable and critical flows) regarding Pb in Vienna for the year 2008.



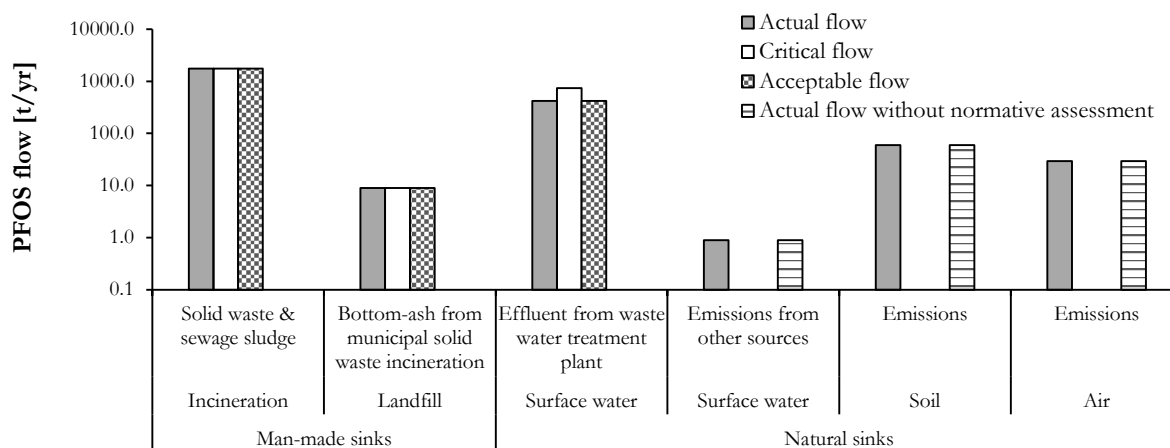
The following results have been obtained (Figure 20), whereas the percentage numbers refer to the sum of actual flows ($100\% \triangleq 191$ t/yr):

- Landfills: 49.6% of flows are due to bottom-ash from MSW incineration. Due to the permit of the landfill, the critical flow is twice as large as the actual flow. 9.1% of all actual Pb flows originates from excavated soil to local landfills. If the landfill capacity should be fully utilized at the end of the approved disposal time, only 2.3% of all actual Pb flows (instead of 9.1%) can be disposed of. The same pattern was found for Pb in demolition waste. 16.7% of Pb originates from demolition waste to landfills. If the landfill capacity should be utilized at the end of the approved service time, only 15.6% of all actual Pb flows (instead of 16.7%) can be disposed of. If the disposal practice continues, landfills for excavated soil and for demolition waste will exceed their approved landfill volume within the approved time for disposal. Based on legal permits, the landfill volumes will be exhausted before the approved time for disposal ends. In other words, the disposal of actual flows is in accordance with legal limits, but the disposal practice faces constraints.
- Underground storage: 22.9% of flows originate from APC residues from MSW incineration. The fractions are acceptable and exported into foreign underground storage facilities.
- Air, soil, water: 0.8% of flows enter ambient air, 0.5% enters urban soil, and 0.3% enters the water. The actual flows yield acceptable risks (RL: $7 \cdot 10^{-6}$, HI: 0.26). Increasing the actual flow into air by a factor of three results in a critical flow (HI: 1).

5.3.4 PFOS in Switzerland

In Switzerland, 96.0% of PFOS flows to regional sinks are acceptable. 4% of all actual PFOS flows lack normative assessment due to insufficient knowledge.

Figure 21: Case study results from inventory analysis (actual flows) and impact assessment (acceptable and critical flows) regarding PFOS in Switzerland for the year 2007.



The following results have been obtained (Figure 21), whereas the percentage numbers refer to the sum of actual flows ($100\% \triangleq 2.260$ t/yr):

- Incineration: 77.1% of PFOS flows originate from solid waste and sewage sludge and are treated by MSW incineration. This flow is mineralized and is classified as acceptable flow, which is in accordance with the EU Regulation (European Commission 2010).
- Landfill: 0.4% of flows derive from residues of waste incineration and enter landfills in an acceptable manner. Today's PFOS emissions from landfills result from former rather than from present waste disposal. They have been assessed as well.
- Water: Two flows enter the aquatic sphere. 18.6% of PFOS are within WWTP effluents and fulfill provisional drinking water standards (U.S. EPA 2009). Up to now in Switzerland, legal limits have been lacking but are under development (Götz, Kase et al. 2011). 2.7% of PFOS originate from additional sources. This flow has not been assessed because data and standards were missing.
- Soil and air: 1.3% of PFOS enter the soil, and 0.04% enter air. These flows, together with the 2.7% of flows into water, have not been assessed due to a lack of data.

5.4 What's the benefit of the new indicator?

The benefits of the new indicator refer (i) to implications of the case study findings on waste and environmental management, (ii) to the relevance of the indicator score, and (iii) to implications of the proposed indicator on resource use in general.

First, the three substances taken into account are widely present in the respective regions, but appear to include fractions that exceed quality standards. For each substance, proactive strategies and measures are needed to increase the indicator score up to 100% in the future.

- For Cu in Vienna, the deposition on urban soil exceeds the acceptance level by a factor of eight. Due to a lack of local data and non-site-specific assessment criteria, the assessment is rather crude. Consequently, the quantity and quality of data has to be improved. This finding is in agreement with the recommendation of the Austrian Environmental Agency for a nationwide monitoring program for metals in soil (Umweltbundesamt 2010). Based on the case study findings, two measures are recommended: On the one hand, improved sampling with respect to horizontal and vertical sample collection, and including the identification of potential hot spots, would allow a decrease in the uncertainty when determining the amount of critical flows to the soil. On the other hand, the development and definition of site-specific reference values in terms of substance concentrations for urban soil are needed. The stormwater overflow into receiving waters exceeds the quality standards by a factor of 11. The stormwater overflow was estimated with a combined water and copper balance model for Vienna, calibrated on the inflow to the waste water treatment plant. Enhanced data quality will improve the quantification of the storm water overflow. For the future, the indicator score might increase due to ongoing measures for reducing water bypassing waste water treatment via storm water overflow (e.g. Stadt Wien 2013).
- For Pb in Vienna, the flow into landfill exceeds the approved landfill capacity before the service time runs out. To overcome these constraints, landfill permits might be extended in time. Alternatively, wastes are directed to remote landfills beyond the system limits, which increases transport distances and costs, or waste fractions can be recycled, complying with advanced quality standards (BRV 2009). Even though the flows into natural sinks yield acceptable human health risks, it has to be noted that the method is based on a uniform approach without spatial resolution. Thus, accidental hot spots representing possible risks for the local population are not included in this approach.
- For PFOS in Switzerland, some flows into air, soil and water have not been assessed due to a lack of data. These flows still pose risks for human and environmental health. For most applications PFOS has been banned, but PFOS still hibernates in present stocks due to former imports. So, PFOS will continue to enter waste flows, whereas the monitoring of PFOS is

rather challenging and expensive (e.g. Schultz et al. 2006, Becker et al. 2008). Due to the obligation to destroy or irreversibly transform the POP content (European Parliament 2004) to PFOS for waste, the concentrations might exceed regulated concentration limits. This effects recycling systems, which “may challenge another environmental priority of ensuring the sustainable use of resources” (European Commission 2010).

Second, the indicator score is of relevance because it fulfills three needs according to Frederiksen and Gudmundsson (2013): First, a need of governments, institutions and companies to satisfy transparency and accountability requirements in policy and measure performance evaluation. The temporal alignment of the indicator score appears to represent the effects of measures undertaken. Second, a need to compare the performance of environmental and waste management across regions. Third, a need to communicate scientific research to the public and decision makers on environmental quality. The indicator processes complex information about the quantity and quality of anthropogenic material flows into an easily understandable score.

Third, materials are removed or lost from the anthroposphere. To accommodate these material without burdens on human and environmental health, natural sinks are available to a certain extent, and man-made sinks have to be provided where natural sinks are missing or lacking. If appropriate sinks are not available, for instance, as a consequence of technological and economic feasibility or ineffective policy instruments, there is a need for strategies that change the anthropogenic material turnover in terms of quantity or quality. The proposed indicator diagnoses to which extent sinks are loaded in an acceptable manner. This information is of major relevance for strategies managing materials that leave the anthroposphere to regional sinks.

6 FUTURE RESEARCH AND OUTLOOK

Investigating the regional material turnover is (a) needed to manage material stocks and flows with respect to quality criteria, and is (b) required to identify the need for sinks in external regions and to develop suitable strategies to monitor and control external sink loads. Future research is needed to evaluate the flow “exported waste & recycling material” (see figure 4). It crosses the spatial system boundary and enters external regions. It might be that the flow is not acceptable due to the same constraints as in the exporting region, for instance, because it surpasses standards for pollution control or overloads the sink “sanitary landfill”. The relevance of waste exports fraught with risk is well documented, for example, for electrical and electronic equipment waste (e.g. Widmer et al. 2005, Sthiannopkao et al. 2013). With respect to the case studies, the flow “exported waste & recycling material” to external sinks is not taken into account, except those to underground storage facilities. To allocate export flows to the exporting region, critical flows for flows within “export waste & recycling material” must be defined. In other words, the removal and loss of substances from exported flows to external sinks must be considered. Therefore, the spatial system boundary has to be extended by including the hinterland. In this case, the proposed method for assessing substance flows to regional sinks is applied for the external regions too. After the critical flow has been determined, the exporting region might increase the critical flow by a market based trading scheme. The proposed scheme allocates external sink loads to the exporting region. This management option is inspired by the mechanism under the Kyoto Protocol (see chapter 2.3). Therefore, allocation procedures are based on tradable permits, including trading units for reducing emissions and enhancing appropriate sinks capacities in external regions. In the context of the flow “exported waste & recycling material”, the critical flows might be increased by reducing emissions or enhancing appropriate sink capacities in the external region. Of course, the proposed management option is visionary, but it respects a globally integrated economy that exchanges wastes and recyclables across regions.

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APPENDICES

Appendix I: Literature survey “The definition of sinks”

The literature survey was carried out between January 2010 and May 2014. It was a web based survey, supported by the search engine “google”. The keywords included “sink”, “final sink”, “substance sink”, and “man-made sink”.

The relevance of defining technical terms is widely recognized in science. A drawback when defining technical terms consists of two fundamental problems: “First, theories that are based on a specific term without defining it cannot be readily tested. Second, communication is difficult; agreements based on certain terms are illusory if different stakeholders have a different comprehension of the term’s meaning” (Heink et al. 2010).

The motivation to define the term “sink” is driven by the scope of the thesis. The framework strives for the assessment of anthropogenic substance flows into natural and man-made sinks. It is essential that different stakeholders in science and policy are addressed because natural sinks are provided by the environment and man-made sinks are provided by the waste management sector. To ensure a common understanding of the term “sink”, state-of-the-art definitions in various fields are investigated (Table I-1) in order to give a definition for the thesis’ framework.

Summarizing the findings, several institutions and scientific disciplines clarify the term “sink” and operationalized it with respect to a theory or framework. It categorizes each definition based on the following aspects: descriptive versus normative, natural versus man-made, place versus process, four different sink functions, and pollution versus resource. Maybe the most prominent one is given by the Intergovernmental Panel on Climate Change: A sink is “any process, activity or mechanism that removes a greenhouse gas, an aerosol, or a precursor of a greenhouse gas or aerosol from the atmosphere” (IPCC 2011). Independent of the substance of interest, environmental chemistry defines the term sink as “an area or part of the environment in which, or a process by which, one or more pollutants is removed from the medium in which it is dispersed” (Duffus et al. 2007). Hence, a pollutant is coined as “any undesirable solid, liquid or gaseous matter in a solid, liquid or gaseous environmental medium” (Duffus, Nordberg et al. 2007). The two definitions of “sink” are very close to each other because they include a substance, a process and the removal function. But, they differ in a descriptive (greenhouse gas) and normative (pollutant) context. However, the definitions in the field of chemistry are rather precise compared to those in the field of Ecological Economics.

In Ecological Economics, the environment is treated as a service unit with different functions in order to support economy, whereas the “sink function” of environment is part of every functional classification (e.g. OECD 2007, Rogall 2008). Among others, Harries (2002) states the following:

“the sink function describes an environment's ability to absorb and render harmless waste and pollution: when waste output exceeds the limit of the sink function, long-term damage occurs”. Hence, the sink function influences the production functions twofold. On the one hand, polluted natural resources decrease the productive function. On the other hand, the productive function falls back on sinks as a valuable resource. The resource aspect is highlighted, for instance, by Daly et al. (2003) who defines a sink as “that part of the environment that receives the waste flow of the throughput and may, if not overwhelmed, be able to regenerate the waste through biogeochemical cycles back to usable sources”. Apart from the environment as natural sink, man-made sinks are of major interest when it comes to the investigation of the anthropogenic metabolism in general, and waste management in particular.

Man-made sinks are integrative elements of the anthropogenic metabolism (Baccini et al. 2012). Waste management is a key process within the anthropogenic metabolism and coins the term “sink” for specific waste treatment and disposal processes as well as for specific waste and recycling flows, respectively. Among others, Zeschmar-Lahl (2004) explains the disposal of various substances in man-made sinks, which either accumulate or transform specific substances. The German Federal Environmental Agency distinguishes between sinks for inorganic and organic substances. On the one hand, incinerators are classified as “typical sinks for pollutants” which are organic (Umweltbundesamt 2008). On the other hand, hazardous substances within the products/waste shouldn't be enriched in the economic cycle, they should be phased out. Exactly for these substances, landfills are “mostly the only economic acceptable sink for pollutants” (Umweltbundesamt 2011). This strategy is also figured out by Brunner (2010), who asks for clean material cycles and safe final sinks. Because hazardous substances might enter recycling products, several authors argue to classify recycling products as “sinks” too (e.g. Chen et al. 2010, Wittmer et al. 2011, Brunner et al. 2012).

To assess material flows into natural sinks and man-made sinks alike, as intended by the thesis, an effective definition of the term “sink” is needed. Concluding the literature survey, the common denominator of definitions in environmental chemistry and waste management includes the following constituents: substance specific, process, removal functions.

Table I-1: Definitions of the term “sink”.

Definition	Reference
Sink: any process, activity or mechanism which removes a greenhouse gas, an aerosol or a precursor of a greenhouse gas from the atmosphere. Note: The UNFCCC distinguishes between "sink" (= process, activity or mechanism) and "reservoir" (= component(s) of the climate system where a greenhouse gas or a precursor of a greenhouse gas is stored). I.e.: A forest can be both a sink (photosynthesis) and a reservoir.	United Nations (1992)

Ein Umweltkompartiment, in dem Stoffe angereichert werden und aus diesem ggf. durch Abbauvorgänge eliminiert werden können.	Deutscher Bundestag (1994)
A special kind of compartment type that accounts for chemical mass no longer available for transport or uptake within a scenario. There are three types of sinks: advection sinks, flush rate sinks, and degradation/reaction sinks.	U.S. EPA (2002)
Endpunkt von Stoffströmen. Im Kontext natürlicher Ressourcen wird unter Senken die Aufnahmefunktion der Natur, z.B. für Schadstoffe, verstanden.	Umweltbundesamt (2012)
Environmental Sink: The redistribution, storage, processing, and absorption of human made waste by the environment. This does not include the waste handled and stored and managed in human-made facilities (see Waste endowment under Human-Made Capital).	SDI Group (1996)
This is a removal process for chemicals in the atmosphere such as dissolution in bodies of water, removal via rain, photolysis, or by reactions with other atmospheric components.	Sam Houston State University (2013)
Place in the environment where a compound or material collects.	U.S. EPA (2009)
Pollution Sink: Vehicle for removal of a chemical or gas from the atmosphere-biosphere-ocean system, in which the substance is absorbed into a permanent or semi-permanent repository, or else transformed into another substance. A carbon sink, for example, might be the ocean (which absorbs and holds carbon from other parts of carbon cycle) or photosynthesis (which converts atmospheric carbon into plant material). Sinks are a fundamental factor in the ongoing balance which determines the concentration of every greenhouse gas in the atmosphere. If the sink is greater than the sources of a gas, its concentration in the atmosphere will decrease; if the source is greater than the sink, the concentration will increase.	EEA (2012)
Sink is a reservoir that takes up a pollutant from another part of its cycle. Soil and trees act as natural "sinks" for carbon.	EPAW (2002)
In environmental chemistry, an area or part of the environment in which, or a process by which, one or more pollutants is removed from the medium in which it is dispersed. Note: For example - moist ground acts as a sink for sulfur dioxide in the air.	Duffus, Nordberg et al. (2007)
<i>noun</i> (technology) a process that acts to absorb or remove energy or a substance from a system; `the ocean is a sink for carbon dioxide`	Princeton University (2013)
Sink is a reservoir that takes up a pollutant from another part of its cycle. Soil and trees act as natural "sinks" for carbon.	EP@W (2002)
A process or mechanism by which water or chemicals are removed from the subsurface system. A Karst Channel is sometimes a sink for groundwater flow. Biodegradation is a sink for a biodegradable chemical.	GOWen Environmental Limited (2014)
The process of providing storage for a substance. For example, plants--through photosynthesis--transform carbon dioxide in the air into organic matter, which either stays in the plants or is stored in the soils. The plants are a sink for carbon dioxide.	NASA (2014)
Nitrogen sink: A subsystem with more nitrogen input than output, as opposed to a source.	NYSERDA (2013)
Carbon sink Both natural and man-made. Accumulates carbon-containing products. Examples include the oceans, plants and landfills.	Green in Seven (2012)
Sink: That part of the environment that receives the waste flow of the throughput and may, if not overwhelmed, be able to regenerate the waste through biogeochemical cycles back to usable sources.	Daly and Farley (2003)
An environmental sink is a repository for potentially damaging by-products of human activity. As with a kitchen sink, we can send a certain amount of "gunk" into an environmental sink before problems occur. Limited amounts of some pollutants can be transformed, diluted, disperse, or absorbed into the environment without causing any damage. Animals can store small traces of DDT pesticide in their fat before harm in	Anderson (2013)

done. And by a process called carbon sequestration, the oceans and vegetation absorb carbon dioxide (CO₂), release oxygen (O), and store carbon (C).

- Sink* is the antonym for the term source, which stands for the origin of an import of a substance into the anthroposphere. A sink can be one of the following: (1) A place on planet Earth where anthropogenic substances (emissions, products of corrosion and weathering, wastes) are disposed of in the environment such as a lake, agricultural soil, or a landfill. (2) A conveyor belt that transports anthropogenic materials from the anthroposphere to certain environmental compartments, such as a river, urban air, or soil erosion. (3) A transformation process such as incineration or microbial degradation that transforms a substance A into a different substance B. Sinks can be located within the anthroposphere (e.g., substance transformation in an incinerator), at the border between anthroposphere and environment (e.g., a river receiving treated wastewater), or in the environment (e.g., the stratosphere for halogenated hydrocarbons). Baccini and Brunner (2012)
- Sink: A body or process that acts as a storage device or disposal mechanism; e.g., plants and the oceans act as sinks absorbing atmospheric carbon dioxide. Also, a location in a plant where sugar is being consumed, either in metabolism or by conversion to starch. Super Glossary (2013)
- Sink: It is the medium, which interacts and retains the long lived pollutant. The oceans are the sinks for atmospheric carbon dioxide. Ground water and subsoil water act as sinks for pesticides employed in agriculture. Subramanian (2011)
- A *sink* is a reservoir that takes up a chemical element or compound from another part of its natural cycle. ELC (2002)
- The "medium" (e.g. soil) or organism (e.g. fish) affected by the pollutant or contaminant is called a *receptor*, whilst a *sink* is a chemical medium or species that retains and interacts with the pollutant. Wikipedia contributors (2011)
- Kompartiment, aus dem eine Substanz durch Abfangvorgänge entfernt wird (Absorption, Ablagerung, Abbau) Stich (2011)
- A carbon sink is a natural or artificial reservoir that accumulates and stores some carbon-containing chemical compound for an indefinite period. The process by which carbon sinks remove carbon dioxide (CO₂) from the atmosphere is known as carbon sequestration. Public awareness of the significance of CO₂ sinks has grown since passage of the Kyoto Protocol, which promotes their use as a form of carbon offset. The main natural sinks are: Absorption of carbon dioxide by the oceans via physicochemical and biological processes, Photosynthesis by terrestrial plants. Natural sinks are typically much larger than artificial sinks. The main artificial sinks are: Landfills, Carbon capture and storage proposals. Wikipedia contributors (2014)
- Sink: Dissipation of degraded (low exergy and negentropy) energy as heat to the environment (and, from earth, to deep space), involving entropy generation and return towards thermodynamic equilibrium. Jørgensen (1997)
- A sink is a net consumer of carbon over a 24h period. The author uses this definition in a plant framework. Farrar (1996)
- Carbon Sinks: sites that soak up carbon (forests) Chalquist (2004-2009)
- Sink: a depression or hole opened by sand or soil erosion caused by poor drainage. Common after a lot of rain, especially in heavily paved areas. Also: a place where unrecycled compounds accumulate.
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Appendix II: Literature survey “SFA studies”

The literature survey was carried out in May 2013, including various sources such as the scientific literature database provided by Elsevier (ScienceDirect) and Thomson Reuters (web of science), and the web search engines “google”. The keywords included “material flows analysis” and “substance flow analysis”. The survey screened individual SFA studies in terms of consideration of waste and emission flows, and natural and man-made sink processes (Table II-1). The list of SFA studies doesn’t claim to cover all SFA studies that have been carried out.

Table II-1: SFA studies

Spatial Level	Year	Substance	Flow type		Sink type		Reference
			waste	emission	Man-made	Natural	
Global Level							
World	1997	Ag	X		X	X	Johnson et al. (2005)
World	1927-2005	Sn	X		X		Izard et al. (2010)
World	2008	Al	X		X		Liu et al. (2013)
World	1997	Ag	X	X	X	X	Eckelman et al. (2007)
World	2000	Ni, Cr	X		X	X	Reck et al. (2008)
World	1994	Cu	X		X	X	Graedel et al. (2004)
World	2000	Pb	X	X	X	X	Mao et al. (2008)
World	2000	Pb	X	X	X	X	Mao et al. (2009)
World	n.a.	Cd		X		X	ICdA (2013)
World Regions							
EU	~2010	p	X	X	X	X	Ott and Rechberger (2012)
EU	2000	Hg, Cd		X		X	Pacyna (2009)
EU	2000	HCB		X		X	Pacyna (2009)
EU	2000	PentaBDE		X		X	Pacyna (2009)
EU	2000	DecaBDE		X		X	Pacyna (2009)
EU	2000	TBT		X		X	Pacyna (2009)
EU	1997	DEHP		X		X	Pacyna (2009)
EU	2000	Atrazine		X		X	Pacyna (2009)
EU	2000	Isoproturon		X		X	Pacyna (2009)
EU	2000	Nonylphenol		X		X	Pacyna (2009)
EU	2000	PAHs		X		X	Pacyna (2009)
EU	2005	Hg	X	X	X	X	Sundseth et al. (2012)
EU		Al	X		X		Bertram et al. (2006)
EU	n.a.	Cu		X			Odnevall Wallinder et al. (2007)
EU	2000	As, Cd, Cr, Ni, Pb		X		X	Pacyna et al. (2007)
EU	1970-2020	DecaBDE	X	X	X	X	Graedel, Van Beers et al. (2004)
Afrika	1994	Cu	X		X	X	Graedel, Van Beers et al. (2004)
Europe	1994	Cu	X		X	X	Graedel, Van Beers et al. (2004)
Latin America and the Caribbean	1994	Cu	X		X	X	Graedel, Van Beers et al. (2004)

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Asia			X	X	X		Graedel, Van Beers et al. (2004)
Middle East	1994	Cu	X	X	X		Graedel, Van Beers et al. (2004)
Commonwealth of Independent States	1994	Cu	X	X	X		Graedel, Van Beers et al. (2004)
North America	1994	Cu	X	X	X		Graedel, Van Beers et al. (2004)
Oceania	1994	Cu	X	X	X		Graedel, Van Beers et al. (2004)
Afrika	1997	Ag	X	X	X		Johnson, Jirikowic et al. (2005)
Europe	1997	Ag	X	X	X		Johnson, Jirikowic et al. (2005)
Latin America and the Caribbean	1997	Ag	X	X	X		Johnson, Jirikowic et al. (2005)
Asia	1997	Ag	X	X	X		Johnson, Jirikowic et al. (2005)
Middle East	1997	Ag	X	X	X		Johnson, Jirikowic et al. (2005)
Commonwealth of Independent States	1997	Ag	X	X	X		Johnson, Jirikowic et al. (2005)
North America	1997	Ag	X	X	X		Johnson, Jirikowic et al. (2005)
Oceania	1997	Ag	X	X	X		Johnson, Jirikowic et al. (2005)
Afrika	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Europe	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Latin America and the Caribbean	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Asia	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Middle East	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Commonwealth of Independent States	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
North America	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Oceania	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
<hr/>							
National level							
<hr/>							
Austria			X	X	X	X	Brunner et al. (2001)
Switzerland		PFOS, PFOA	X	X	X	X	Buser and Morf (2009)
China	2001, 2004, 2007	Al	X	X	X		(2010)
Switzerland	1990	Cd	X	X	X	X	Kaufmann et al. (1997)
Germany		Cu, Zn, Pb		X	X	X	Hillenbrand et al. (2005)
Switzerland	2005	N	X	X	X	X	Federal Office for the Environment (2010)
Australia	1998/1999	Cd	X	X	X	X	Kwonpongsagoon et al. (2007)
Japan		Cu	X	X	X	X	Daigo et al. (2009)
Taiwan		Cd	X	X	X	X	Chang et al. (2007)
U.S.	1975-2000	Cu		X		X	Lifset, Eckelman et al. (2012)
Switzerland	1190, 1925, 1950, 1975, 2000	Cu					Wittmer (2006)
			X	X	X	X	

Austria	2003	Fe, Cu, Zn, Al	X		X		Döberl et al. (2004)
China	1994, 200, 2004	Cu, Zn, Ni	X	X	X	X	Wang et al. (2008)
Austria		FCKW	X	X	X	X	Obernosterer et al. (2005)
Austria	2001	N	X	X	X	X	Obernosterer et al. (2003)
Austria	1994	Zn	X	X	X	X	Daxbeck et al. (1998)
Norway	n.a.	Cd	X				Mykeelbost et al. (1997)
Norway	n.a.	DEHP	X				Mykeelbost and Rypdal (1997)
Switzerland		Sb	X	X	X	X	Mathys et al. (2007)
Taiwan		Zn	X				Ma et al. (2011)
Switzerland	1980-2020	DecaBDE, HBCD, BDE-27	X	X	X	X	Morf et al. (2007)
Austria	n.a.	Estron, 17- β -Estradiol, Estriol, Ethinylestradiol (EE2), Bisphe-nol A und Nonylphenol	X	X	X	X	Skutan et al. (2003)
Brazil	2005	Cu	X		X	X	Tanimoto et al. (2010)
Germany		Ga, In, Mn, Ni, Pa, Ag, Ti, Zn, Sn	X	X	X	X	Wittmer, Erren et al. (2011), Wittmer et al. (2011)
Austria	2001	Al	X		X		Pilz et al. (2003)
Austria	2003	Cu	X	X	X	X	Daxbeck et al. (2006)
India	2010	Ag	X	X	X	X	Burger Chakraborty, Qureshi et al. (2013)
Netherland	1990	Cd, Cu, Pb, Zn	X	X	X	X	Guinée, van den Bergh et al. (1999)
Peru	1997	Ag	X		X	X	Johnson, Jirikowic et al. (2005)
United States	1997	Ag	X		X	X	Johnson, Jirikowic et al. (2005)
Mexico	1997	Ag	X		X	X	Johnson, Jirikowic et al. (2005)
France	1997	Ag	X		X	X	Johnson, Jirikowic et al. (2005)
India	1997	Ag	X		X	X	Johnson, Jirikowic et al. (2005)
Germany	1997	Ag	X		X	X	Johnson, Jirikowic et al. (2005)
Zambia	1994	Cu	X		X	X	Graedel, Van Beers et al. (2004)
Germany	1994	Cu	X		X	X	Graedel, Van Beers et al. (2004)
Saudi Arabia	1994	Cu	X		X	X	Graedel, Van Beers et al. (2004)
China	1994	Cu	X		X	X	Graedel, Van Beers et al. (2004)
United States	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
France	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
United Kingdom	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Italy	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Japan	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
India	2000	Pb	X	X	X	X	Mao, Dong et al. (2008)
Regional level							

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Minnesota	1990, 2000, 2005	Hg	X	X	X	X	Barr Engineering Company (2001)
Unteres Bünztal	1986/1987	N, P, Cl, Cu, Zn, Cd, Pb (waterborne)	X			X	Henseler et al. (1992)
Sundgau region (CH)	n.a.	P, Cd, Zn,	X	X		X	Keller et al. (2001)
Upper Parramata river catchment (Australia)	2004-2005	Cu (waterborne)	X	X		X	Seelsaen (2007)
Rural region in Lower Austria	n.a.	N, P (waterborne)	X				Zessner et al. (2002)
Rural region in Lower Austria	n.a.	Zn, Cu, Pb, Cd (waterborne)	X				Zessner and Lampert (2002)
Swedish municipalities	1995, 1998	Cd	X		X	X	Lindqvist et al. (2004)
Rhine River	Mid 1960, 1988	Cd, Zn, Pb		X		X	Stigliani et al. (1993)
Hudson River basin		Heavy metal, pesticides and herbicides, other critical pollutants		X		X	Ayres et al. (1986)
Los Angeles Basin	1972	Pb		X		X	Huntzicker et al. (1975)
Generic region	n.a.	DEHP (waterborne)	X	X	X	X	De Keyser et al. (2010)
City level							
Stockholm	1995	Cd, Cr, Cu, Pb, Hg, Ni, Zn	X	X	X	X	Bergbäck et al. (2001)
Vienna	1991	N, C, Pb	X	X	X	X	Daxbeck et al. (1996)
Bangkok	1996	N, P	X	X		X	Færge et al. (2001)
Stockholm	1900-1995	Ce, Cr, Cu, Pb, Hg, Zn	X	X	X	X	Hedbrant (2001)
Stockholm	1995	Cu, Zn, Cd, Pb (waterborne)	X	X	X	X	Jonsson (2000)
Stockholm	2002, 2004, 2005	DEHP, APEO, PBDE, CP		X		X	Jonsson et al. (2008)
Vienna	n.a.	Al, Cu, Pb,		X		X	Obernosterer et al. (2003)
Stockholm		Pb, Zn		X		X	Palm and Ostlund (1996)
Villach		Pb, Cd, Zn, Cu (waterborne)	X	X		X	Rebernick (2007)
Stockholm		Cd, Cr, Cu, Hg, Ni, Pb, Zn	X				Sörme et al. (2001)
Sydney	2000	P	X	X	X	X	Tangsubkul et al. (2005)
Chaohu City	2008	P	X	X	X	X	Yuan et al. (2011)
Vienna	2010	c-PentaBDE, c-OctaBDE	X	X	X	X	Vyzinkarova et al. (2013)
Greater Toronto Area	2005	C	X	X	X	X	Mohareb and Kennedy (2012)
Gothenburg	2009	P	X	X	X	X	Kalmykova et al. (2012)
Paris	1801-1914	N	X			X	Barles (2007)
Paris	2006	N	X	X	X	X	Svirejeva-Hopkins et al. (2011)
Nhat Tan, Hoang Tay (Vietnam)	2008	N, P	X	X	X	X	Do-Thu et al. (2011)

Note: n.a. = not available

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Appendix III: Articles

The appendix III includes (1) the list of articles and their link to the thesis, (2) the statement from co-authors regarding the permit to include the articles, (3) my contributions to each article, and (4) the articles itself.

Included articles and link to the thesis

This thesis includes three original research articles:

- I. Kral, U., Kellner, K., Brunner, P.H., 2013. Sustainable resource use requires “clean cycles” and safe “final sinks”. Online: <http://dx.doi.org/10.1016/j.scitotenv.2012.08.094>. Science of the Total Environment 461–462, 819-822.
- II. Kral, U., Lin, C.Y., Kellner, K., Ma, H.-W., Brunner, P.H., 2014. The copper balance of cities: Exploratory insights into a European and an Asian city. Journal of Industrial Ecology. Online: <http://dx.doi.org/10.1111/jiec.12088>. In press.
- III. Kral, U., Brunner, P.H, Chen, P.C., Chen, S.R.. Sinks as limited resources? - A new indicator for evaluating anthropogenic material flows. Journal of Ecological Indicators. Submitted on 18th January 2014, 1st revisions submitted on 16th May 2014.

Two (article I, article II) out of three articles are published in peer-reviewed, international journals. They are available online under open access license and are included in the thesis. They are reprinted according to the authors’ rights of the Publishers Elsevier and Wiley-Blackwell, respectively. One (article III) out of three articles was submitted to the Journal of Ecological Indicators on 18 January 2014, the 1st revision was submitted on 16 May, and is at present with the editor. The article is included in appendix III in accordance with the author’s rights of the Publisher Elsevier.

Table III-1: Included articles and link to individual chapters of the thesis

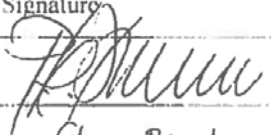

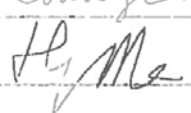
	Background and problem definition chapter 1.1	Indicator definition and interpretation chapter 3.1	Inventory analysis chapter 3.3, 4.2	Impact assessment chapter 3.4, 4.3	Results and discussion chapter 5
Article I	X				X
Article II			X		
Article III		X	X	X	X

Statement from Co-authors

**Statement from co-authors
confirming the authorship contribution of the PhD candidate**

As co-authors we authorize the inclusion of the articles in the candidate's thesis and certify that

- the declaration made by the candidate (see annex) correctly reflects the extent of the candidate's contribution to this work;
- the candidate contributed greater than 80% of the content of the articles and is the "primary author". The candidate was primarily responsible for the planning, execution and preparation of the work for publication.

Co-Author (alphabetical order)	Signature	Date
Brunner, Paul H.		24/4/2014
Chen, Pi-Cheng	Chen Pi-cheng	24/4/2014
Chen, Sih-rong	Chen Sih-Rong	25/4/2014
Kellner, Katharina		15/5/2014
Lin, Chih-Yi	Chihyi Lin	30/4/2014
Ma, Hwong-Wen		25/4/2014

Included articles:

- I. Kral, U., Kellner, K., Brunner, P.H., 2013. Sustainable resource use requires "clean cycles" and safe "final sinks". Online available: [dx.doi.org/10.1016/j.scitotenv.2012.08.094](https://doi.org/10.1016/j.scitotenv.2012.08.094). Science of the Total Environment 461-462, 819-822.
- II. Kral, U., Lin, C.Y., Kellner, K., Ma, H.-W., Brunner, P.H., 2014. The copper balance of cities: Exploratory insights into a European and an Asian city. Journal of Industrial Ecology, Online available: [dx.doi.org/10.1111/jiec.12088](https://doi.org/10.1111/jiec.12088), in press.
- III. Kral, U., Brunner, P.H., Chen, P.C., Chen, S.R. . Sinks as limited resources? - A new indicator for evaluating anthropogenic material flows. Journal of Ecological Indicators, submitted on 16th January 2014.

Annex: Declaration by the candidate

Note: The Annex "Declaration by the candidate" can be found in the next chapter "Authors contribution".

Authors contribution

For all three articles I have (a) reviewed the literature for contextualizing the research done, (b) suggested article content and structure, (c) took the lead in text writing and figure creation, and (d) handled the submission, review and editorial phase until publication. All articles are the result of collaborative work with theoretical and empirical contributions from my side:

Table III-2: Authors contribution to article I

Task	Kral, U.	Kellner, K.	Brunner, P.H.
Initial idea about the research topic	o	o	++
Conception and design of the article	++	+	o
Supervision	n.r.	n.r.	++

Notes: ++ = major contribution; + = minor contribution; o = no contribution; n.r. = not relevant

Table III-3: Authors contribution to article II

Task	Kral, U.	Lin, C.Y.	Kellner, K.	Ma, H.W.	Brunner, P.H.
Model development	+	o	+	+	+
Data acquisition for Vienna	+	o	++	o	o
Data acquisition for Taipei	o	++	o	o	o
Data treatment in view of city comparison	++	+	o	o	o
SFA indicator development	+	+	o	o	+
SFA indicator quantification	+	+	o	o	o
Supervision	n.r.	n.r.	n.r.	+	++

Notes: ++ = major contribution; + = minor contribution; o = no contribution; n.r. = not relevant

Table III-4: Authors contribution to article III

Task	Kral, U.	Brunner, P.H.	Chen, P.C.	Chen, S.R.
Sink indicator development	++	+	o	o
Case study “Cu Vienna”	++	o	o	o
Case study “Pb Vienna”	++	o	+	+
Case study “PFOS Switzerland”	++	o	o	o
Supervision	n.r.	++	n.r.	n.r.

Notes: ++ = major contribution; + = minor contribution; o = no contribution; n.r. = not relevant

Appended articles

Article I – Main article	AIII-1
Article II – Main article	AIII-7
Article II – Supporting information	AIII-21
Article III – Main article	AIII-77
Article III – Supporting information	AIII-107

Article I

Kral, U., Kellner, K., Brunner, P.H., 2013. Sustainable resource use requires “clean cycles” and safe “final sinks”. Online: <http://dx.doi.org/10.1016/j.scitotenv.2012.08.094>. Science of the Total Environment 461–462, 819-822.



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Sustainable resource use requires “clean cycles” and safe “final sinks”

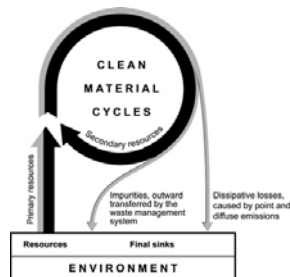
Ulrich Kral*, Katharina Kellner, Paul H. Brunner

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HIGHLIGHTS

- ▶ Present recycling policies focus too much on maximizing recycling rates.
- ▶ Hazardous materials are kept in cycles instead of eliminating them from cycles.
- ▶ New priorities must be set to establish clean cycles.
- ▶ A clean cycle strategy results in residues that must be disposed of in sinks.
- ▶ Waste management must supply suitable sinks for substances eliminated from cycles.

GRAPHICAL ABSTRACT



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ABSTRACT

In order to fulfill the objectives of environmental protection, today's focus on quantitative recycling rates must be amended by a more qualitative approach. Because modern products represent a mix of numerous and sometimes hazardous substances, ways must be explored to remove detrimental substances during recycling and to establish “clean cycles”. On the one hand, such a “clean cycle” strategy will result in better recycling qualities of secondary products and less dissipation of hazardous substances during further product use. On the other hand, the elimination of hazardous substances during recycling requires sinks for the disposal of the eliminated materials. These topics are presented in general as well as by case studies. In particular, the sink issue is addressed, differentiating between sinks and final sinks and discussing the challenge to supply appropriate final sinks for all materials that cannot be recycled.

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1. Introduction

On a global scale, there's a continuous increase of material extraction from the earth's crust (Weber et al., 2011), resulting in large accumulations of materials particularly in urban areas. While the anthropogenic material stocks are growing, the amount of waste flows and emission rates increase with a certain delay. To satisfy contemporary resource

needs and to decrease environmental loadings, an obvious solution is to recycle as much as possible in order to substitute primary resources. Therefore, European legislation aims to increase recycling rates continuously. This quantitative approach does not take into account the presence of unwanted substances ending up in the second generation products. To avoid this, a so called “clean cycle” strategy is proposed.

2. “Clean cycle” strategy

Sustainable resource use is characterized by an ecologically acceptable impact on nature. Recycling strategies play a key role by

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extending materials' lifespan and reducing environmental impacts. Until now, the focus of environmental evaluation methodologies has been laid on emission rates, the composition of second life products has only rarely been considered in view of hazardous constituents, multiple recycling loops, and long term effects. There is no focus yet on the fact that today's recycling loops contain valuable as well as harmful substances. Even though cycling of hazardous substances is prohibited by waste management directives (Republik Österreich, 2008), the recycling quota regulation as well as economic constraints result in hazardous substances to remain in cycles. Hence, "tramp elements" accumulate in the anthroposphere with potential negative effects on a) product quality, and b) environment:

- ad a) Some "tramp elements" accumulate finely dispersed in the product cycle and lower the quality of second generation goods made up from secondary resources. This becomes apparent for example in the plastic industry, where stabilizers like cadmium, lead and tin contaminate the recycling products (Fehringer et al., 1997), or in the steel industry, where copper contaminates the steel cycle (Gleich et al., 2004), or in packaging industry, where mineral oils contaminate paperboards used for food packaging (Kappen et al., 2012). In short, the goals of high recycling rates and high product qualities often contradict each other.
- ad b) The accumulation of toxic "tramp elements" in the product cycle presents an increasing potential for the release of hazardous substances. The reason is that some fractions of a material are inevitable released throughout the entire life cycle (see Fig. 1). In other words, substances from the stock-in-use are converted into unrecoverable forms and dissipated into the environment (Ayres et al., 2002). For example, in 1983 about 10%–30% of refined copper left the product cycle and ended up in environment or landfills (Bureau of Mines, 1983; Nriagu and Pacyna, 1988). Exemplary, emission sources are roofs and brake wear. The products are exposed to abrasion, corrosion and weathering processes which effect dissipative material releases. The great majority of these dissipated materials accumulate in specific environmental compartments and may threaten to impact natural processes (Geiser, 2001). A case study on urban surfaces shows that about 1/3 of the diffusively emitted copper enters the waste management system. In contrast, roughly 2/3 is directly lost in an uncontrolled form to the environment (Rebernik, 2007). While the scientific community is increasingly aware of these kinds of material losses (e.g. Arx, 2006; Bergbäck et al., 2001; Burkhardt et al., 2007, 2008; Obernosterer et al.,

2003; Sörme et al., 2001), few guidelines were developed to keep dissipative losses from urban surfaces and abrasion on low levels (Hoffmann and Rudolphi, 2005; Zysset et al., 2002). In the future, dissipative material losses from all sources and their effects on the environment must be taken into account to determine sustainable resource use.

In order to establish a sustainable resource use with no burden for future generations, hazardous substances have to be eliminated from material cycles. Otherwise recycling strategies run the risk to a) support a qualitative down-cycling of materials in a large scale format, and b) raise the potential for harmful material losses throughout the material life cycle.

There are two conclusions regarding a sustainable recycling policy which incorporates a "clean cycle" strategy:

- For environmental protection and product quality reasons, the recycling quotes have to take qualitative characteristics regarding secondary materials into account. The dilution of unwanted substances in second generation products as well as the total release of harmful substances to the environment has to be taken under control.
- If "clean cycles" are established, the safe disposal of specific substances removed from the cycles is mandatory. These contaminated material flows have to enter safe "final sinks". Consequently, a safe "final sink" concept with corresponding policy instruments has to be developed.

Fig. 2 displays the resulting material flows based on a "clean cycle" strategy. An optimum mix of primary and clean secondary material keeps the material cycle alive. Impurities and dissipative material losses are directed to safe final sinks.

3. The need for final sinks

In 1965 Albert Wolman was the first who analyzed cities as metabolic systems (Wolman, 1965). He highlighted the fact, that material use is directly linked with emissions and waste. The discussion about the need for sinks to take up substance flows became public awareness in 1987, where the Brundtland report states the importance of "ultimate sinks for the by-products of human activities" (United Nations, 1987). In 1996, Joel Tarr – an environmental historian – has drawn the attention to "the search of the ultimate sink" from a historical perspective (Tarr, 1996). At the same time Marina Alberti highlighted the fact that "cities cannot sustain themselves without drawing on the carrying capacity of their hinterland or region at the

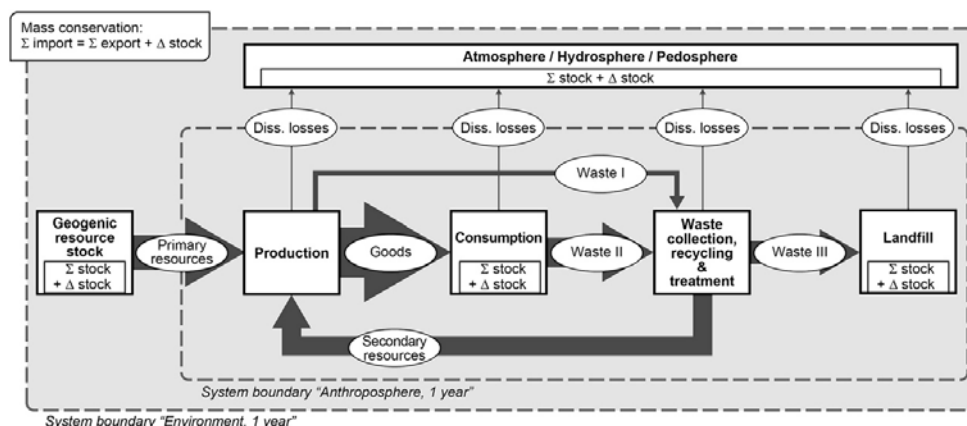


Fig. 1. Anthropogenic material stocks and flows.

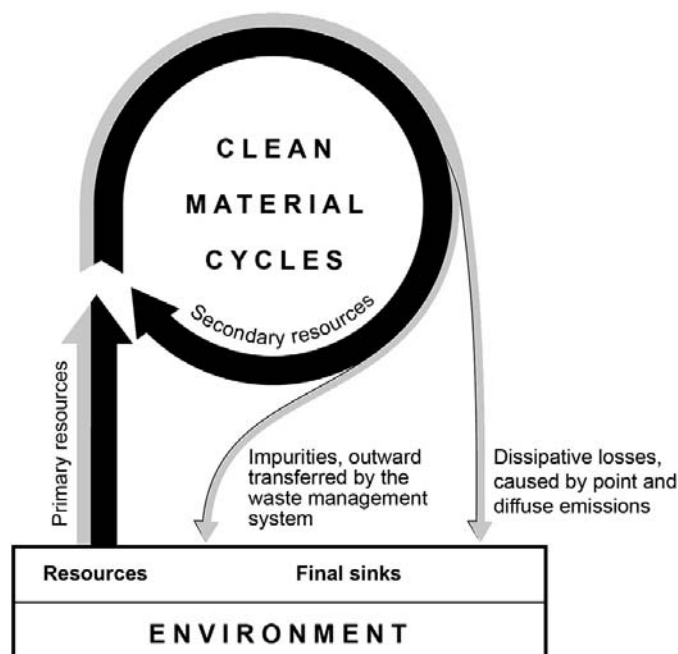


Fig. 2. Material flows based on a "clean cycle" strategy (Stumm and Davis, 1974, modified).

back end of their metabolism" (Alberti, 1996). Now it's time to develop a systematic "sink concept" that links urban output flows with manmade or natural sinks. The definition of sink indicators can also support the evaluation of ecological sustainability (Döberl and Brunner, 2004), and thus contribute to the reduction of the risk of overloading sinks, such as today's excessive flow of greenhouse gases to the sink "atmosphere" (Brunner, 2010).

From a materials management point of view, the anthroposphere can be seen as flow through reactor with a distinctive storage function. Material flows leave the anthropogenic material cycle (see Fig. 2), due to a) the second law of thermodynamics (dissipative losses occur) and, b) the diminishing returns (secondary resources). The core question is: Where should these off-flows end up? The answer on a general level is: They have to end up in final sinks. A more concrete answer remains to be developed because at present, the sink concept is still a vague framework. Based on previous definitions, the authors put the following working hypothesis forward for discussion:

- A "sink" is defined as a process that receives anthropogenic material flows that have no positive value for present societies.
- A "final sink" is a sink that either destroys a substance completely, or that holds a substance for a very long time period.

In order to exemplify the term "sink" and "final sink", two case studies are discussed. They focus on: a) the retention in the environment of anthropogenic materials that have been previously lost by dissipation, and b) the elimination of copper from the steel cycle and the deposit of recycling residues.

ad a) Glaciers act as receptor for a broad range of dissipative, airborne losses like heavy metals or persistent organic pollutants (POPs). Thus glacier ice is a "sink" that can become a secondary source of pollutants (Salomons, 1998). Recently, a Swiss study (Bogdal et al., 2009) identified an accelerated release of pollutants from melting Alpine glaciers. This might result in an

accumulation of hazardous substances in the food chain up to fishes or plants growing in glacier water. The case study exemplifies the term sink as used here for the temporary storage process "glacier ice", and reveals that so far many anthropogenic flows of hazardous substances have not yet reached an environmentally safe "final sink".

ad b) Due to technological constraints, it is difficult and costly to remove copper during steel recycling. So, traces of copper remain and accumulate in the product cycle. This results in down cycling and lower product quality. In order to achieve appropriate product qualities, steel companies select scrap fractions on the resource market depending on the copper concentration. A study about sustainable metal management (Gleich et al., 2004) indicates, that in the future, copper concentrations will become so high that specific scrap fractions cannot be used as secondary resource in the steel industry anymore. Scenarios were identified as reasonable that remove part of the scrap from the recycling stream in order to dispose them off in manmade sinks like landfills.

The example shows that a) steel is a temporary sink for copper and the steel cycle eventually stresses long term sinks like landfills, and b) the selection of scrap depends on quality criteria. So, steel recycling quotes are quality driven and can't be achieved by prescribed recycling rates. The exclusive definition of recycling rates on a quantitative base blinds out the qualitative constraints of recycling. In order to guarantee a clean steel cycle, recycling policies have to consider product qualities too.

Concluding, both the whole range of dissipative material losses¹ and impurities within products have to be directed to safe final sinks. If a safe final sink can't be identified for ecological, technical

¹ Dissipative material losses cover point and diffuse emissions with no option for recovery.

or economic reasons, material design strategies have to be developed that consider final sink limitations, such as in the case of fossil fuels where a safe mediate term sink for carbon is missing.

4. Conclusion

In order to achieve sustainable resource management, a strategy directed towards clean cycles and safe final sinks needs to be developed. A clean cycle strategy delivers an optimum instead of a maximum of secondary resources through elimination of hazardous substances from material cycles. This ensures the generation of quality proven recyclables for multiple life cycles of products without the risk to shift problems into the future. Removing hazardous substances from material cycles requires final sinks where these substances can be stored safely for geological time periods.

Acknowledgments

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Article II

Kral, U., Lin, C.Y., Kellner, K., Ma, H.-W., Brunner, P.H., 2014. The copper balance of cities: Exploratory insights into a European and an Asian city. *Journal of Industrial Ecology*. Online: <http://dx.doi.org/10.1111/jiec.12088>. In press.

The Copper Balance of Cities

Exploratory Insights into a European and an Asian City

Ulrich Kral, Chih-Yi Lin, Katharina Kellner, Hwong-wen Ma, and Paul H. Brunner

Keywords:

cities
environmental protection
industrial ecology
resource efficiency
substance flow analysis (SFA)
urban metabolism

 Supporting information is available on the JIE Web site

Summary

Material management faces a dual challenge: on the one hand satisfying large and increasing demands for goods and on the other hand accommodating wastes and emissions in sinks. Hence, the characterization of material flows and stocks is relevant for both improving resource efficiency and environmental protection. This article focuses on the urban scale, a dimension rarely investigated in past metal flow studies. We compare the copper (Cu) metabolism of two cities in different economic states, namely, Vienna (Europe) and Taipei (Asia). Substance flow analysis is used to calculate urban Cu balances in a comprehensive and transparent form. The main difference between Cu in the two cities appears to be the stock: Vienna seems close to saturation with 180 kilograms per capita (kg/cap) and a growth rate of 2% per year. In contrast, the Taipei stock of 30 kg/cap grows rapidly by 26% per year. Even though most Cu is recycled in both cities, bottom ash from municipal solid waste incineration represents an unused Cu potential accounting for 1% to 5% of annual demand. Nonpoint emissions are predominant; up to 50% of the loadings into the sewer system are from nonpoint sources. The results of this research are instrumental for the design of the Cu metabolism in each city. The outcomes serve as a base for identification and recovery of recyclables as well as for directing nonrecyclables to appropriate sinks, avoiding sensitive environmental pathways. The methodology applied is well suited for city benchmarking if sufficient data are available.

Introduction

One of the main tasks of managing human settlements during the past 10,000 years of urban history was the sufficient supply and disposal of materials. Driven by technology and socioeconomic factors, the interaction of humans with natural resources and the environment has been in continuous change over the years (Agudelo-Vera et al. 2011). Key developments of the past were the rise of cities and empires after 3000 B.C. and the start of the industrial revolution in the eighteenth century. Global population has been increasing and resource-intensive lifestyles became predominant in the modern world. One of the pioneers who recognized the consequences of ongoing urbanization at an early stage was Patrick Geddes (Geddes 1885). At

the transition from the nineteenth to the twentieth century, he created awareness for the massive flows of resources in cities. By pointing out material losses in the production chain through input-output (I/O) balancing of material budgets, he was a forerunner of today's material flow modeler and accountants.

Since that time, several research frameworks have been developed and put forward. Approaches such as material flow analysis (MFA) (Brunner and Rechberger 2004; Baccini and Bader 1996), physical I/O analysis (e.g., Nakamura and Kondo 2009; Nakamura et al. 2007; Hoekstra and Van den Bergh 2006), and environmental-extended I/O analysis (e.g., Hertwich and Peters 2010; Leontief and Ford 1972) are utilized to investigate the anthropogenic metabolism (Baccini and Brunner 2012). In particular, substance flow analysis (SFA) has been

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proven to be a practical tool for analyzing urban metal pathways (e.g., Månsson et al. 2009; Henseler et al. 1992; Lindqvist and von Malmborg 2004). SFA tracks the pathway of selected substances through systems such as households, enterprises, cities, or regions. Concerning metal flows in urban areas, a literature review yields two characteristics: First, urban metal flow studies are rare. Recently, Chen and Graedel (2012) reviewed more than 350 SFA articles in a comprehensive manner. They found five cities with insights into metal metabolism. Stockholm is the only city that takes copper (Cu) into account (Bergbäck et al. 2001; Sörme et al. 2001a, 2001b). Additional cities are unexploited even though Cu is a subject of interest because of its relevance from both a resource and environmental point of view. Second, individual city studies are hardly comparable to each other. We extended the scope of literature research and compiled 15 exemplary studies for Cu on an urban scale (see section 1 of the supporting information available on the Journal's website). A common feature is that they are selective in their scope and that they vary in methodology, such as in terms of system boundaries, modeling approaches, and data acquisition and allocation.

To fill the gap of rare urban Cu studies on the one hand, and comparative city assessments on the other hand, we give exploratory insights into urban Cu balances of different cities. The aims of this study are to (1) develop a methodology to analyze and evaluate the Cu flows and stocks on an urban scale, (2) present and compare the results of a Cu flow and stock analysis for two cities, (3) discuss the differences between the two cities on the basis of selected indicators, and, finally, (4) test the hypothesis that comparing metabolic differences between cities is instrumental for improving decision making regarding resource management and environmental protection.

To reach these objectives, a case-study approach is applied. Two cities in Europe and Asia, namely, Vienna and Taipei, are chosen as study objects because of their distinct differences, such as population densities and trends, economic developments, culture and lifestyles, and geographical and environmental settings. Explorative data analysis and MFA are used to summarize the characteristics of urban Cu balances in rigid, transparent, and comprehensible form. The procedure chosen reduces complexity of Cu flows and stocks and facilitates the comparison of different cities.

The work contributes to the field of industrial ecology. It gives a substance-specific understanding of urban resource flows and stocks for city planners and researchers, pointing out the total flows from import into stocks as well as to export out of a city. The individual process descriptions and indicators might appear insufficient in content and level of detail. But, exploring full-substance balances facilitates the interpretation of dynamic substance turnover in a comprehensive manner. The results serve well the improvement of resource efficiency and environmental performance from an urban systems point of view.

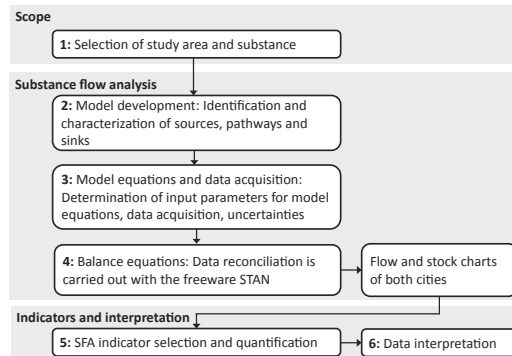


Figure 1 Research framework.

Materials and Methods

The framework of the study is summarized in figure 1. First, we give reasons for city and substance selection. Second, a generic substance flow model is set up. Third, we use an accounting scheme in order to define model equations on an individual city base. Input parameters are processed with Monte Carlo simulation. Uncertainty ranges of input parameters are considered. Fourth, static modeling is applied for balancing flows and stocks. This yields reconciled data visualized with stock and flow charts. Fifth, substance flow indicators are selected and calculated based on the balanced stock and flow charts. Sixth and last, the differences in the score of the indicators are used as a starting point for interpretation and discussion of distinct urban patterns in each city.

Scope

Choice of Cities and City Characteristics

We have chosen Vienna and Taipei as case-study regions because of their characteristics in the following areas:

1. Population density in Taipei is more than twice that of Vienna (96 vs. 40 inhabitants per hectare [ha]). Vienna hosts 36% less citizens (1.7 vs. 2.6 million capita) on an area that is 53% larger than in Taipei (41,487 vs. 27,180 ha). The population outlook for the next 50 years expects a 19% decrease for Taipei and a 27% increase for Vienna (Statistik Austria 2013; CEPD 2012).
2. From a cultural and lifestyle point of view, Vienna, as a traditional European city, differs from Taipei, as an Asian city with rather little Western influence. Nevertheless, anthropogenic activities in both cities are service oriented and modern, lacking of heavy industry.
3. The economic power of Vienna and Taipei show similarities such as the magnitude of gross regional product (Vienna, 43,900 Euros per capita; Taipei, 34,800 Euros per capita). This affects the turnover of consumer and

Table 1 Process characterization

<i>Process name</i>	<i>Characterization</i>
External anthroposphere	Stands for the anthropogenic hinterland of the city; it delivers products and construction material to the city and receives exported products, waste, and recyclables.
Industry, business, services, and forestry	Covers economic activities as well as related buildings; economic activities refer to the trade of goods, material processing, and distribution for final consumption. The buildings are addressed for stock calculation, including construction material and installations.
Transport, energy, and communication infrastructure	Covers immobile infrastructure and corresponding copper stock in transport networks, power grids, and telecommunication networks.
Vehicles	Covers the mobile copper stocks, such as in cars, trucks, bikes, buses, trams, and trains.
Private households	Covers anthropogenic activities of daily life and related buildings; anthropogenic activities refer to residing, nourishing, cleaning, and communication. Related buildings, such as flats and houses, are used for stock calculation of construction material and installations.
Waste management system	Covers the collection, treatment, and disposal of solid waste; the process is disaggregated, which gives further insights into fluxes in view of incineration, composting, and landfilling.
Wastewater management system	Covers the collection and treatment of wastewater; material stocks are not taken into account.
Underground storage	External salt mines out of use act as final storage for hazardous residues from incineration.
Planetary boundary layer	Stands for the lowest part of the atmosphere that is influenced by its contact with the earth's surface, usually several hundred meters high
Urban pedosphere and vegetation	Consisting of urban soil and vegetation in parks, green areas, and agricultural fields
Urban hydrosphere	Urban water bodies, mainly rivers, groundwater, ponds, and small lakes
Receiving waters	The hydrosphere that takes up both wastewater treatment effluents and combined sewer overflow from the city, such as the river Danube (Vienna) and the Taiwan Straits (Taipei)

investment goods as well as the need for facilities for waste and wastewater treatment and disposal.

- The geographical and environmental settings of the two cities are different: Vienna is a landlocked city dewatering by the river Danube to the far-away Black Sea; Taipei is situated on a Pacific island close to the South China Sea. Thus, the so-called disposal hinterland for the two cities is quite diverse, with an abundant dilution potential for liquid emissions in Taipei and limited capacity for effluents in Vienna.

The two cities are representative of many other cities in the world: If global population is grouped according to the surrounding ecosystems, 65% of people in coastal ecosystems live in urban areas like Taipei. Approximately 45% of people in cultivated ecosystems live in urban areas like Vienna (Marcotullio et al. 2008).

Choice of Substance

Resource and environmental aspects are in focus when selecting Cu: This metal is a relevant resource for modern humans. The lifestyle in both cities depends essentially on this technological metal. It is used in many consumer goods such as electric appliances, private and public transport systems, and in infrastructure systems for supply and disposal of water, energy, and information. Because of the high costs of producing Cu from ores, the recovery and recycling of Cu is attractive and is a widely used practice.

Concerning the environment, Cu acts as a tracer for urban emissions. For instance, Cu concentrations in Viennese sewage sludge are significantly larger than in rural areas (Kroiss et al. 2008), and Cu concentrations in Vienna soils are higher than

in surrounding rural areas (Pfleiderer 2011). Sörme (2003) describes how modern cities are faced with nonpoint metal emissions. They play an increasing role, compared to emissions from point sources. Because of their high number of sources, they are more difficult to control by regulation than point sources.

Substance Flow Analysis

We apply a static mass-balance approach based on materials accounting for two reasons. First, accounting requires reported or measured data sets. In general, quantity and quality of data are better available for past years. So, we selected the years 2008 (Vienna) and 2009 (Taipei) for the ex-post assessment of material flows. Second, a descriptive framework is most appropriate because of a lack of knowledge regarding deterministic linkages between inflows, stocks, and outflows. From a methodological viewpoint, our approach combines SFA and exploratory data analysis.

Model Development

The aim is to present a generic stock and flow chart that allows for comparing the two cities. This requires a common understanding of the key flows and stocks of Cu in a city. Therefore, we set up a model that meets individual urban characteristics of Vienna and Taipei without losing sight of the need to finally compare the data of the two cities. The spatial boundary is set by the administrative city limits, and the data are compiled on an annual basis. The urban systems comprising processes and flows are defined based on previous studies, literature investigations, reports by local municipalities, and expert interviews. All processes and flows are roughly characterized in table 1. The Cu flow models are represented in figures 2 and 3. The

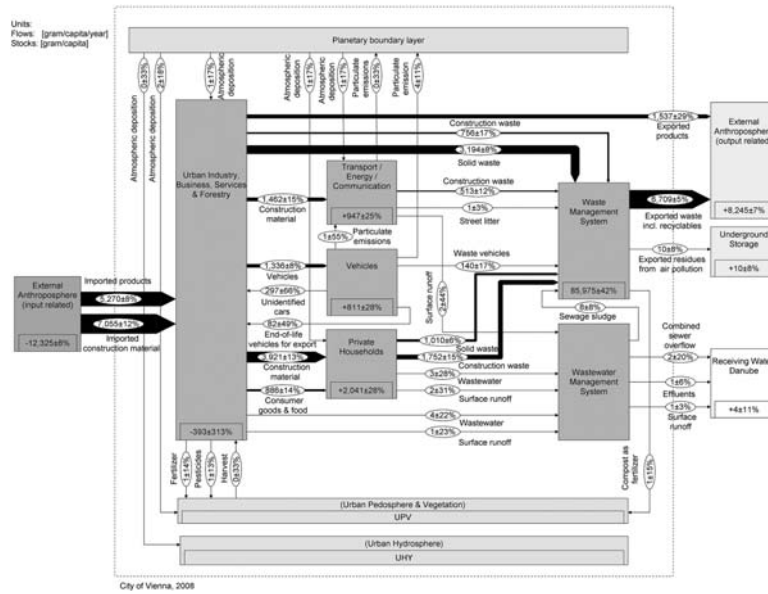


Figure 2 System “copper flows and stocks in Vienna” for the year 2008. Values for flows and changes in stocks are given in grams per capita per year (g/cap/yr) and for stocks in grams per capita (g/cap). The flows are represented as Sankey arrows proportional to the flow rate; figures for stocks are given within the “process” boxes. Numbers have been rounded.

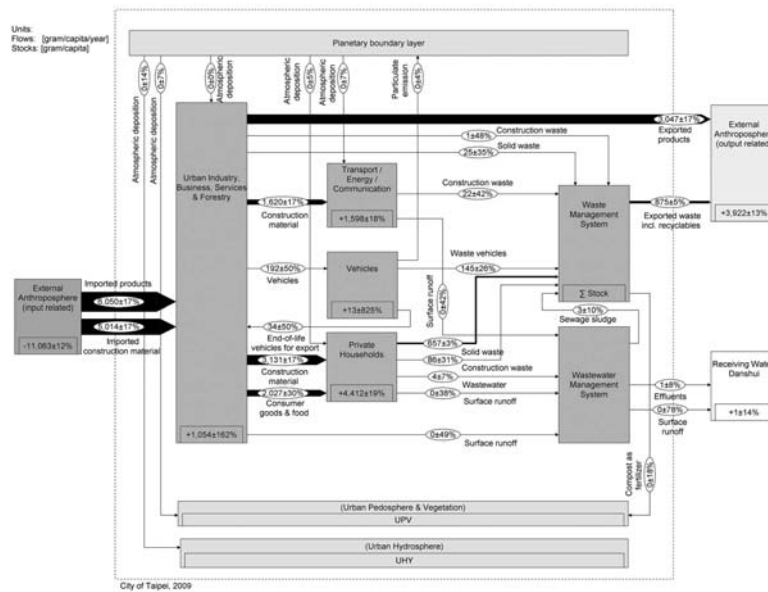


Figure 3 System “copper flows and stocks in Taipei” for the year 2009. Values for flows and changes in stocks are given in grams per capita per year (g/cap/yr) and for stocks in grams per capita (g/cap). The flows are represented as Sankey arrows proportional to the flow rate; figures for stocks are given within the “process” boxes. Numbers have been rounded.

Supporting Information on the Web (see section 2.2.2.1) provides additional, comprehensive descriptions for all flows and stocks.

Model Equations and Data Acquisition

The following model equations are used to calculate each flow and stock:

$$\dot{m}_{flow} = f(p_1, p_2, p_3, \dots, p_i)$$

$$m_{stock} = f(q_1, q_2, q_3, \dots, q_i)$$

where \dot{m}_{flow} is the Cu flux rate [mass/time], m_{stock} is the Cu stock [mass], p_i is the input parameter for \dot{m}_{flow} , and q_i is the input parameter for m_{stock} . The input parameters p_i and q_i are assumed to be normally distributed with $N(m_{p_i}, s_{p_i})$ and $N(m_{q_i}, s_{q_i})$. The mean values m and the standard deviation s are determined as follows:

- m is determined by data mining according to Månsson (2009). The data acquisition procedure prioritizes a bottom-up approach for both cities. Data availability, quantity, and quality vary between each city. They are even manifold within each city depending on the type of flow and stock. As a common denominator, input parameters representing city characteristics are documented in official statistics. Import data are established through downscaling national import and export statistics. The allocation to city internal processes is based on the global sector share of Cu products and estimations based on local waste statistics. Cu content in products is compiled from literature data, local consumption, and waste statistics. Waste flows are documented in statistics provided by public and private waste companies. Cu fluxes entering and leaving waste management plants, such as incinerators and waste water treatment plants, are documented in scientific reports conducted by the city authority. Emission flows are estimated by the compilation of literature data and inventory databases, such as Ecoinvent. Cu stocks in technical infrastructures are estimated with network lengths and corresponding specific masses. In Vienna, stocks in buildings are based on Swiss per capita data and those in Taipei are based on proxy data from other Taiwanese cities.
- s is derived from the uncertainty factor uf according to data vagueness concept from Hedbrant and Sörme (2001): $s_p = m_{p_i} * \frac{uf-1}{2}$, $s_q = m_{q_i} * \frac{uf-1}{2}$ with $uf = 1 + 0.0036 * e^{1.105 * l}$. The uncertainty level l ranges from “1” to “5” and depends on the classification of the data source. For example, official statistics on local level are assumed to have low uncertainties ($l = 1$). Another example is official statistics on the national level down-scaled to the local level with a higher level of uncertainty ($l = 3$).

The mean value and standard deviation of each flow ($m_{\dot{m}_{flow}}, s_{\dot{m}_{flow}}$) and stock ($m_{m_{stock}}, s_{m_{stock}}$) is computed with

Monte Carlo simulation by taking into account the model equations and the distribution functions of the input parameters p_i and q_i .

The documentation of the SFA model is given in the Supporting Information on the Web. It includes section 2.2.2.1 with a comprehensive description of flows and stocks for both cities. Sections 2.2.2.2 and 2.2.2.3 address the city of Vienna, including two tables: one for the model equations and one for the input parameters. Section 2.2.2.4 provides the background data for the city of Taipei.

Balance Equations

We use static model architecture and apply the mass balance principle on each process:

$$0 = \dot{m}_{input} - \dot{m}_{output} + \dot{m}_{stock}$$

where \dot{m}_{input} is the annual input flow, \dot{m}_{output} is the annual output flow, and \dot{m}_{stock} is the alteration of stock. Because multiple data sources are used, data quality and quantity are heterogeneous. Consequently, contradictions in fulfilling the mass balance criteria occur. To overcome this gap, we applied the freeware, STAN (Cencic 2012). It uses data reconciliation with an algorithm based on the error propagation law.

Stock and Flow Charts of Vienna and Taipei

Figures 2 and 3 show the annual Cu SFA charts. A full list of unbalanced flows and balanced results is provided in the Supporting Information on the Web (see section 2.3).

Data Analysis and Indicator Selection

Exploratory data analysis stands for analyzing data sets to summarize their main characteristics in an easy-to-understand form. We use this tool in combination with indicators for comparative assessment of individual Cu flow data. An indicator is defined to be one or several “observed variables that are used to report a non observable reality” (Loiseau et al. 2012, 214). Our set of indicators represents the interaction of substances within and between the anthroposphere and the environment and form, in part, a base for policy support and decision making for substance management, recycling, and waste management. Table 2 compiles eight indicator groups, including 13 indicators in total. Seven relate to resource efficiency (RE), six relate to environmental protection (EP). The calculation routine is based on the final Cu balances in each city, which comprise 42 flows and four stocks each (figures 2 and 3).

Results and Discussion

Overview

Table 2 presents the computed indicator results, including mean value and standard deviations as well as normalized indices on a per capita basis.

Table 2 Indicators and their values for Vienna and Taipei

No.	Indicator	Scope	Absolute values						Normalized values on a per capita base					
			Vienna			Taipei			Vienna			Taipei		
			Unit	Mean	Dev (%)	Mean	Dev (%)	Unit	Mean	Dev	Mean	Dev		
I	Imports into the cities	RE	t/yr	20,644	8	—	28,847	12	—	kg/cap/yr	12.3	0.9	11.1	1.4
II	Stocks and changes in stocks	—	—	—	—	—	—	—	—	—	—	—	—	—
	Present urban stock	RE	t	298,000	13	72,051	21	178	24	kg/cap	—	—	28	6
	Absolute change in stock	RE	t/yr	5,535	43	18,453	27	3.3	1.4	kg/cap/yr	—	—	7.1	1.9
III	Relative change in stock	RE	%	2	n.q.	26	n.q.	—	—	—	—	—	—	—
	Wastes and emissions	RE	t/yr	12,370	5	2,450	6	7.4	0.4	kg/cap/yr	—	—	0.9	0.1
	Solid waste	RE	t/yr	12,337	5	2,437	6	7.4	0.4	kg/cap/yr	—	—	0.9	0.1
IV	Unintentional emissions	EP	t/yr	28	10	13	6	0.016	2 × 10 ⁻³	kg/cap/yr	—	—	0.005	3 × 10 ⁻⁴
	Intentional emissions	EP	t/yr	5	9	0	0	0.003	3 × 10 ⁻⁴	kg/cap/yr	—	—	0	0
	Ratio nonpoint emissions to total emissions	EP	%	51	n.q.	12	n.q.	—	—	—	—	—	—	—
V	Flows to sinks	RE	t/yr	14,935	6	10,402	13	8.9	0.5	kg/cap/yr	—	—	4.0	0.5
	Anthropogenic	RE	t/yr	14,924	6	10,390	13	8.9	0.5	kg/cap/yr	—	—	4.0	0.5
	Environmental	EP	t/yr	12	8	4	13	7.0 × 10 ⁻³	0.6 × 10 ⁻³	kg/cap/yr	—	—	1 × 10 ⁻³	0.2 × 10 ⁻³
VI	Accumulation in urban soil	EP	%	0.07	n.q.	0.03	n.q.	—	—	—	—	—	—	—
VII	Removal efficiency by wastewater management	EP	%	65%	n.q.	74%	n.q.	—	—	—	—	—	—	—
VIII	Copper content in bottom ash	RE	t/yr	1,097	7	165	34	0.655	0.043	kg/cap/yr	—	—	0.063	0.021

Notes: RE = resource efficiency; EP = environmental protection; mean = mean value; dev = standard deviation; n.q. = not quantified; — = not relevant; t = metric tons; t/yr = metric tons per year; % = percent; kg/cap/yr = kilograms per capita per year.

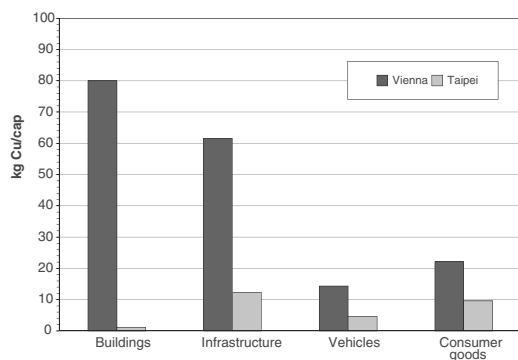


Figure 4 Comparison of copper stocks in Vienna (total stock: 178 kilograms per capita [kg/cap]) and Taipei (total stock: 28 kg/cap). Whereas the residence time of copper in buildings and infrastructure is long (10 to 100 years), it is shorter in vehicles and consumer goods (less than 10 years).

Comparative Assessment and Interpretation of Copper Balances

In the following sections, indicators and their relevance are explained in detail, and results as well as conclusions are presented.

Imports into the Cities (I)

Net imports represent the demand and consumption patterns and relate to resource supply as well as to the city's economic situation. In both consumption-oriented cities with a comparatively high gross domestic product (GDP), Cu is mainly used in infrastructure and consumer goods. Similar amounts of Cu are imported for Vienna (12 ± 1 kilograms per capita per year [kg/cap/yr]) and Taipei (11 ± 1 kg/cap/yr); both cities rely heavily on Cu import from outside regions. The observed net import rates are six times higher than the average global per capita consumption (Graedel et al. 2004; United Nations 2010).

Stocks and Changes in Stocks (II)

Three indicators focus on stocks: (1) the present urban stock and (2) the absolute and (3) relative change in stock.

A large part of Cu imports turns into stocks. There are Cu stocks of 178 ± 23 kg/cap in Vienna and of 28 ± 6 kg/cap in Taipei. Similar stocks have been found for other cities in Europe (Sörme et al. 2001a) and in Asia (Zhang et al. 2012). The two cities show different shares of individual Cu stocks (figure 4). Vienna hosts 80% of Cu in long-term assets. Heating systems as well as networks for electricity and telecommunication are the main Cu carriers. In contrast, Taipei (1) has a much shorter history in urban development and (2) buildings lack heating systems. Forty-eight percent of the Cu is stored in long-term goods, such as infrastructures and buildings, and 52% are related to consumer goods, such as air conditioners, cars, and scooters. As a summary, the two cities show marked

differences in quantity and relative share in individual sector: Vienna is rich in Cu, and urban mining hotspots are identified in long-term assets, such as infrastructure and building components. In Taipei, consumer goods have more importance for recovering secondary Cu.

Stock changes denote the annual accumulation or depletion of Cu in various stocks, such as buildings, infrastructure, vehicles, and consumer goods. The indicator represents the economic and technological pattern of a city: As long as a city grows, the input will always be bigger than the output, resulting in a stock increase. An exception is given for a material that is substituted by another, or that is phased out, such as cadmium. For such substances, the input can be smaller than the output, resulting in stock depletion.

The total annual growth rate of the Cu stock is 7 ± 2 kg/cap in Taipei and 3 ± 1 kg/cap in Vienna. In both cities, consumer stocks increase. In Taipei, more Cu is accumulated in private households than infrastructure and vehicles, with sales in the household electronic sector and in electronic appliances in vehicles as the main drivers.

The relative change in stock puts the absolute change in stock (in kg/cap/yr) in relation to present stock (in kg/cap). The relative accumulation of Cu in the stock of Taipei (26%) is more than 10 times higher than in Vienna (2%). In other words, the Cu stock in Vienna is already on a high level and grows only moderately. In Taipei, there is a backlog of demand in the city, resulting in a much faster stock increase, which is mainly a result of private households. For Taipei, the Cu consumption in private households is five times higher than in Vienna, where Cu turnover is determined by maintaining relatively large stocks.

Wastes and Emissions (III)

This category comprises three indicators: (1) the amount of Cu in solid wastes, including scrap for recycling, and (2) intentional and (3) unintentional emissions. "Intentional emissions" denominate Cu flows resulting from applications that transport, on purpose, Cu into the environment, such as Cu use in agriculture as a fungicide. "Unintentional emissions" are by-products resulting from other processes, such as wastewater from households and industry, the release of Cu from catenary wires, or brake pads during the operation of vehicles. These emissions pose a potential threat to the environment.

In Vienna, the total flow of Cu resulting from wastes and emissions is eight times higher than in Taipei (7.4 ± 0.4 vs. 0.9 ± 0.1 kg/cap/yr). In both cities, solid-waste-borne Cu dominates emissions by more than 99%. Unintentional emissions follow next with less than 1%. These findings are in line with results from a study on dissipative emissions in the United States (Lifset et al. 2012). Lifset and colleagues point out that the recycling rate would increase only by 0.5% if dissipative losses would be recovered and included in recycling, too.

The household waste fractions of Vienna and Taipei are similar (1.0 ± 0.06 vs. 0.7 ± 0.02 kg/cap/yr). In contrast, the fractions from the industrial and construction sector and waste vehicles differ significantly between Vienna (6.4 ± 0.4 kg/cap/yr)

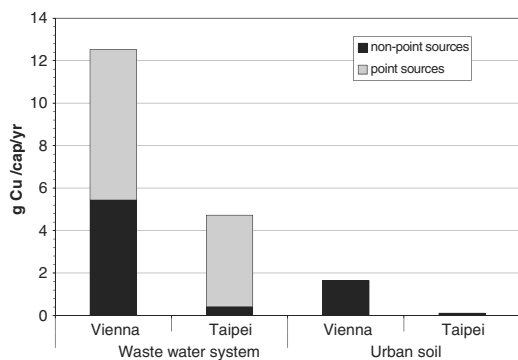


Figure 5 Comparison of copper flows into sinks in Vienna (14 grams per capita per year [g/cap/yr]) and Taipei (5 g/cap/yr) divided into flows entering the wastewater system and soil. Nonpoint sources address waterborne copper in surface runoff and include roof runoff, brake and tire wear from cars, catenary wear, and atmospheric deposition. Point sources relate to waterborne emissions from industry and household, such as urine, feces, consumer products, and pipe corrosion.

and Taipei (0.2 ± 0.1 kg/cap/yr). These findings may be the result of the following three reasons. First, the large and comparatively old stock of Cu in infrastructure and buildings of Vienna must be continuously replaced and acts a source of Cu waste. Second, Taipei has installed a zero waste policy attempting to reduce Cu waste from production. As a third source of uncertainty, statistical data from the two cities about waste flows from the industrial sector have been collected by differing methodologies.

Unintentional Cu emissions in Vienna are three times larger than in Taipei (16 ± 2 grams per capita per year [g/cap/yr] vs. 5 ± 0.3 g/cap/yr). Both airborne emissions and surface runoff in Vienna are approximately 12 times higher than in Taipei. This is because of a higher amount of car mileage and corresponding brake wear, catenary wear which is inexistent in Taipei, and the utilization of Cu as a roof and gutter material in Vienna.

Ratio of Nonpoint Emissions to Total Emissions (IV)

Bergbäck (1992) and Sörme and colleagues (2001b) refer to the increasing relevance of nonpoint emissions from the use phase of goods when compared to industrial point source emissions. They report a significant amount of nonpoint emissions for Stockholm and Sweden. The investigators state the difficulties when attempting to regulate nonpoint emissions, such as abrasion from brake pads or corrosion from roofs and gutters. Therefore, we choose as an indicator the proportion of nonpoint emissions to total emissions. Results show that the ratio of nonpoint emissions to total emissions differs between 51% in Vienna and 12% in Taipei (figure 5).

In both cities, the wastewater system receives (1) most of the Cu emissions in town (Vienna, 87%; Taipei, 97%) and (2) more Cu from point sources than from nonpoint sources (Vienna,

57%; Taipei, 91%). Point sources include Cu inputs into the wastewater from private and commercial facilities. In Vienna, approximately 20% of point emissions originate from feces, urine, consumer products, and kitchen waste, 20% from corroding water pipes, and the remaining 60% are related to industrial activities. In Taipei, the relevance of individual point sources is not fully determined because of a lack of local information.

Nonpoint sources cover brake and tire wear, catenary wear, and roof runoff. In Vienna, the annual Cu flow from nonpoint sources is 13 times larger than in Taipei (7.3 vs. 0.6 g/cap/yr). The higher rate is the result of the presence of an extensive tram network, the popularity of Cu roofs and gutters, and higher emissions from low-duty vehicles. On a per capita basis, the Viennese Cu flow into soil is approximately 15 larger than that in Taipei (1.65 vs. 0.11 g/cap/yr).

Two conclusions can be drawn: First, nonpoint emission patterns play an important role in the case of Vienna, confirming the result of the Swedish studies (Sörme et al. 2001b; Hjortenkrans et al. 2007). This requires that city authorities develop specific long-term strategies for protecting the environment from nonpoint emissions. Second, because of high ratios of sealed and drained area, the majority of Cu flows is waterborne and collected by the sewer system. Effective end-of-pipe technologies, such as appropriate sewage treatment plants, are needed for separating heavy metals in order to control the impact on receiving waters.

Flows to Sinks (V)

For this study, anthropogenic sinks and environmental sinks have been taken into account as indicators. In both cities, the main Cu flows to sinks stay within the anthroposphere (more than 99.9%), and less than 0.1% of the total processed output accumulates in the environment (Vienna, 0.08%; Taipei, 0.03%). Figure 6 summarizes the flows to the main sinks.

From the city point of view, anthropogenic sinks consist of (1) landfills within the city limits and of (2) goods, wastes, and recyclables that are processed outside of Vienna and Taipei, respectively. Landfills containing Cu in municipal solid waste (MSW) and bottom ash from MSW incineration (see discussion below on "Copper content in bottom ash") belong to the internal anthroposphere. Cu from recyclables is recovered in the external anthroposphere because there are no Cu recycling facilities within the two investigated cities. The largest amount of Cu leaves the city as a product or solid waste. Taipei exports 78% of Cu in a product form and Vienna 81% of Cu as waste, including recyclables.

Environmental sinks consist of receiving waters and the urban soil within the city limits. The efficiency of the total wastewater system, in terms of separating and directing Cu to appropriate sinks and its relevance for city planning, is further examined when the indicator "Technical efficiency of the wastewater system" is discussed below. The soil accumulating Cu by deliberate, as well as unintentional, emissions is included in the following discussion of the indicator "Accumulation in urban soil." The atmosphere as a sink has been neglected as a result of the short residence time of Cu in the air.

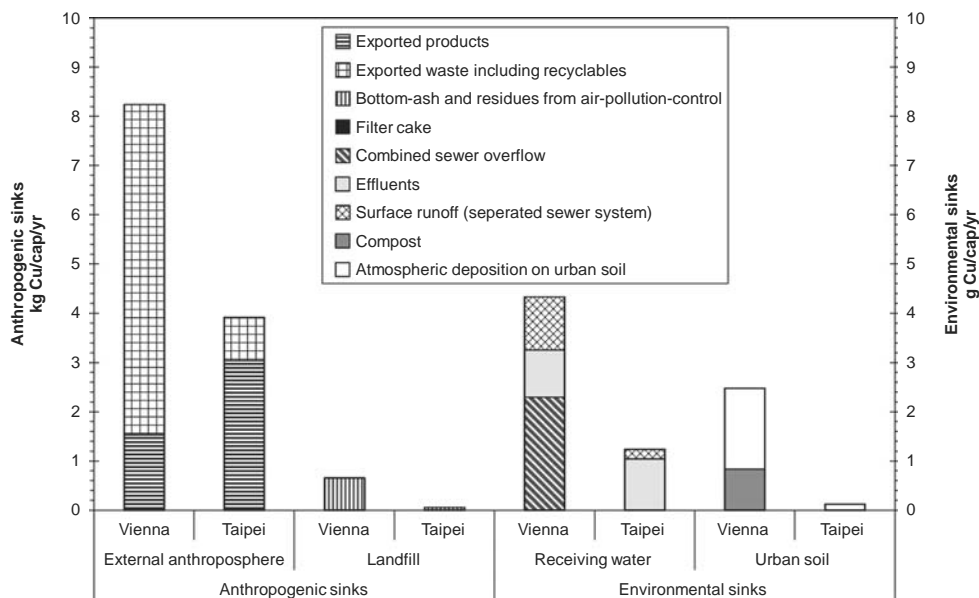


Figure 6 Comparison of copper flows to anthropogenic and environmental sinks in Vienna. The bars for the “anthropogenic sinks” are given in kilograms per capita per year (kg/cap/yr) and those for the “environmental sinks” in grams per capita per year (g/cap/yr). The “External anthroposphere” consists of the hinterland and includes exterior consumption, recycling processes, and underground storage sites outside of Vienna and Taipei. “Receiving water” stands for the river Danube and the Danshui River, respectively.

Accumulation in Urban Soil (VI)

The disparity between the levels of Cu in urban and rural areas has been known for many decades. See, for instance Purves (1966, p. 1077) who is referring to “evidence of slow poisoning of the soil environment in built-up areas.” The need for observing urban soil conditions for sustainable, long-term land management is well established (e.g., Wong et al. 2006; Johnson et al. 2011), mostly because analysis of soil samples occasionally showed elevated concentrations compared to rural areas (Pfleiderer 2011; Jien et al. 2011). Because of the lack of such systematic monitoring in Vienna and Taipei, we used a simplified indicator by relating the annual Cu load to the existing Cu stock in the top 30 centimeters of soil of greenspace areas within the city boundaries. Results indicate slow Cu accumulation for the two city soils. The higher accumulation rate in Vienna (0.07% per year) versus Taipei (0.03% per year) may well resemble the bigger emission rate into the air by the Austrian city. Both countries and cities fall short of legal threshold values regarding substance concentrations in urban soil.

Based on the SFA data and the fact that both cities are situated in service-based and not industrial regions, it can be concluded that the accumulation of Cu in the soil of the two cities is small. Little effects on the pedosphere are expected in the next few centuries. Nevertheless, because there are no appropriate surveying programs in place, it may be that hotspots in the soil exist either by local emissions, geological anomalies,

or former anthropogenic inputs. Because the Cu metabolism of Vienna is distinctly different, with more Cu on the surface of buildings and more incorporated in traffic systems, an effective monitoring program should be established first in Vienna in order to ensure that reference values are not surpassed. Site-specific reference values, based on geogenic background concentrations, are actually being developed in a project called “urban geochemistry of Vienna” (Pfleiderer 2011).

Removal Efficiency by Wastewater Management (VII)

In Germany, approximately 35% of Cu input into rivers originates from urban areas (Blondzik et al. 2004). The type of sewer system, its leakage rates, and the treatment technology determine the heavy metal discharge into the hydrosphere. To estimate the efficiency of the wastewater system regarding Cu removal, we relate Cu removed by the wastewater treatment (wwt) system and contained in sewage sludge to the total amount of Cu that has been introduced into the sewer system. SFA calculations yield Cu removal efficiencies of 74% for Taipei and 65% for Vienna. The remaining 26% and, respectively, 35% enter receiving waters by combined sewer overflow, wwt effluent, and surface runoff. Both surface runoff collected by separate sewer systems and combined sewer overflow reach receiving waters directly without treatment. The lower removal efficiency in Vienna is likely the result of larger nonpoint emissions and additional Cu loads from the combined sewer

overflow. Based on these results, the following conclusions can be drawn:

- *Wwt systems design:* In order to decrease Cu loadings into the hydrosphere, reduction as well as collection of nonpoint Cu emissions and high removal rates by wwt is important. The comprehensive design of the entire wastewater collection and treatment system becomes crucial for service-oriented cities where nonpoint sources are the dominant cause of emissions.
- *Monitoring:* There is a lack of accurate data with known uncertainties regarding surface runoff, combined sewer overflow, and stormwater overflow. Few cities establish complete water balances, including collected and uncollected as well as treated and untreated waters and wastewaters. We recommend establishing water balances for cities and using them as a base for decisions regarding environmental management. The city of Berlin serves as an example for a sophisticated precipitation runoff model based on land use and resulting in estimations of location-based surface runoff (City of Berlin 2012b, 2012a).
- *Environmental risk assessment:* Cities in general depend on dilution potentials in their hinterland. Vienna uses the river Danube and Taipei the Danshui River for dissipation of Cu contained in purified sewage. Despite that one third to one fourth of sewage-borne Cu is released to the receiving waters, existing environmental quality standards are observed in both cases. For sediments of the river and sea, there is a lack of both legal standards as well as consistent monitoring data about Cu concentrations (e.g., for Vienna, Kavka et al. 2000). In order to prevent future overloading, environmental fate and impact models could be used for predicting the evolution of heavy metal concentrations over time.

Copper Content in Bottom Ash (VIII)

In both cities, municipal solid waste is incinerated in waste to energy plants. During incineration, most of the Cu contained in MSW is transferred to bottom ash. To recover Cu from bottom ash offers two advantages: First, it contributes to resource conservation. Second, it is instrumental for minimizing Cu flows to the environment in case bottom ash is landfilled or reused as a construction material.

In Vienna, bottom ash is stabilized with cement and disposed of in the municipal landfill. In Taipei, bottom ash is used as a base material for road construction as well as a fine aggregate in asphalt (Taipei County Government 2010). The per capita Cu flux in Vienna is approximately 10 times larger than in Taipei (655 ± 43 vs. 63 ± 21 g/cap/yr), corresponding to approximately 5% (Vienna) and 1% (Taipei) of annual Cu consumption. The market value of Cu contained in bottom ash equals roughly US\$8.8 million per year in Vienna and US\$1.4 million per year in Taipei.

As a result of the value of Cu and other metals, economic incentives exist to recover valuable elements, such as Cu, aluminum, gold, and silver, from bottom ash. Substance concen-

trations are several times higher than in natural ores (Simon 1996; Jordi 2004) and can be considerably increased by bottom ash treatment (e.g., Muchova et al. 2009; Morf et al. 2013; Shen and Forssberg 2003). Recent experiences favor dry discharge of bottom ash for efficient recovery of nonferrous metals (Morf et al. 2013). This technology is not yet implemented in Vienna and Taipei.

Another means to recover valuable metals from MSW is landfill mining, that is, the excavation of materials from old MSW or bottom ash landfills. From an economic point of view, landfill mining appears to be attractive only if additional values are created. This could be driven by gaining new land for building sites or reduced costs for long-term landfill after care. Thus, recovery projects that aim exclusively at recovering resources are rare (Hölzle 2010). Assuming a period of 20 years and a waste generation rate of one kilogram of MSW per person and day, 24,000 tonnes¹ of Cu with an economic value of US\$200 million have been accumulated in Viennese landfills.

In addition to recovery of Cu from bottom ash and landfills, Cu can also be recovered by separate collection of Cu containing waste fractions, such as waste electrical and electronic equipment. In fact, this practice is widely applied and is the most favored by European waste policy. For best effectiveness of a comprehensive Cu recovery system, the main carriers of Cu (Morf and Taverna 2006), as well as the efficiencies and costs of recovery technologies, must be known. It remains to be determined which of the recovery pathways reaches the goals of waste management “resource recovery” and “environmental protection” at the least costs.

Conclusions

This article presents the results of investigations into urban Cu flows and stocks in two different cities. It provides a starting point for comparing additional cities and metals. The mass balance approach, focusing on the main processes, Cu flows, and stocks, goes beyond the city as a black-box model. To our knowledge, this study is the most comprehensive Cu balance on the urban level, encompassing Cu flows from imports to stocks in use and exports. Also, it is the first comparison of the Cu metabolism of two cities. Based on the results, we discuss reasons for differences in the Cu balances of Vienna and Taipei and give recommendations for the management of Cu as a resource and potential environmental pollutant.

First, we find typical characteristics in the dynamics of Cu stock changes. Rapid growth in a young city such as Taipei is characterized by low amounts of Cu stocks and relatively high annual stock increases. In contrast, Cu stocks in older Vienna are relatively high, and thus the relative stock change is smaller than in Taipei. Cu demand and disposal in Vienna is mainly the result of maintenance and replacement. The relative importance of Cu in consumer goods decreases.

Second, much Cu is recycled, but there are still recovery potentials available. Bottom ash from waste incineration is an

example for both cities. In order to reach sustainability goals such as resource conservation and long-term environmental protection, recovering more Cu from MSW and other wastes is mandatory. On the one hand, especially if combined with the recovery of additional metals such as aluminum, gold, and silver, this may result in economic benefits. On the other hand, separation of Cu together with other metals, such as chromium, lead, and cadmium reduces the concentrations of heavy metals in bottom ash, making this material more suitable for utilization as a construction material.

Third, some Cu is emitted diffusively by wear, corrosion, and weathering of Cu built into infrastructure, transport systems, and others. These emissions partly enter the wastewater system and partly accumulate in the urban soil. Nonpoint emissions are not yet in the focus of urban governance, even though, for example, the European Union urges member states to take nonpoint emission into account. Based on the experiences of the studies in Stockholm, Vienna, and Taipei, it is recommended to monitor the concentrations of Cu and other heavy metals in urban soils and sediments by a combination of SFA and direct measurements.

Fourth, designing urban wastewater systems in a comprehensive way is crucial for minimizing the loading of receiving waters with Cu and other heavy metals. The combination of wastewater collection systems and treatment technologies determine the total efficiency in terms of removing pollutants. For effective control of heavy metal flows to receiving waters, the entire systems performance is more important than the removal efficiency of wastewater treatment alone.

Fifth, this study has shown that transnational collaboration yields new insights into the substance balances of cities. Authorities and researchers profit from each other through a common research framework for comparing urban metabolism data. We recommend sharing experiences and discussing methodology for urban metabolism studies and developing common generic models as well as data acquisition procedures.

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Note

1. The term tonne refers to metric ton. One tonne (t) = 10³ kilograms (kg, SI) ≈ 1.1 short tons.

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Supporting Information

Additional Supporting Information may be found in the online version of this article at the publisher's web site:

Supporting Information S1: This supporting information provides a large quantity of background material that complements the methods and findings included in the main research article, including a compilation of 15 exemplary studies for copper (Cu) on an urban scale, comprehensive descriptions of all flows and stocks for the study, documentation on the substance flow analysis (SFA) model used, and a full list of unbalanced flows and balanced results for this study.

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Kral, U., C.-Y. Lin, K. Kellner, H.-W. Ma, P.H. Brunner. 2013. The copper balance of cities: Exploratory insights into a European and an Asian city. *Journal of Industrial Ecology*.

Summary

This supporting information provides a large quantity of background material that complements the methods and findings included in the main research article, including: a compilation of 15 exemplary studies for copper (Cu) on an urban scale; comprehensive descriptions of all flows and stocks for the study; documentation on the substance flow analysis (SFA) model used; and a full list of unbalanced flows and balanced results for this study.

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1. INTRODUCTION

Table S1: Examples of Cu flow studies on urban scale. They vary in terms of scope, modeling and data acquisition framework.

Analytical entities	City	Focus	Reference
stocks	Nanjing (CN)	Supply-restrictions of Cu in China lead to the identification of potential reservoirs of secondary Cu resources in urban infrastructure.	(Zhang et al. 2012)
	Stockholm (SE)	Pathways and stock are investigated comprehensively.	(Sörme et al. 2001a; Bergbäck et al. 2001; Sörme et al. 2001b)
	Linköping (SE)	Stocks-in-use and hibernating stocks in the telecommunication network are estimated in view of the recovering potential.	(Krook et al. 2011)
	Cape Town (ZA)	Stocks-in-use of major appliances are linked with product lifetimes in order to predict future waste flows and recovering potentials.	(van Beers and Graedel 2003)
emissions and/or waste flows	Nanjing (CN)	Cu impacts on road sediments are analyzed.	(Zuo et al. 2012)
	Stockholm (SE)	The pathway analysis of diffuse emissions is driven by increased concentrations in Stockholm's receiving sediments.	(Sörme and Lagerkvist 2002; Sörme et al. 2001b)
	Urban catchment (UK)	Water quality is assessed by diffuse emissions entering waste water systems.	(Rule et al. 2006)
	Sofia (BG), New Heaven (USA)	Cu discard, reuse and recovery fluxes and rates are compared between two cities.	(Dimitrova et al. 2007)
	Villach (AUT)	The fate of diffuse emissions is determined in order to assess environmental risks.	(Rebernig 2007)
End-of-Pipe plants	Vienna (AUT)	- Waste water treatment plant: Heavy metal flow ratios on plant level demonstrate the separation efficiency. High separation rates improve environmental performance. - Incinerator: Monitoring of heavy metal in residues reveals the temporal evolution of concentrations in household waste.	(Kroiss et al. 2008; Morf and Taverna 2006)
	Hinwil (CH)	Incinerator: Residues were analyzed in order to estimate recovering potential of precious metals and rare earth elements.	(Morf et al. 2013)
Urban Soil	Edinburgh, Dundee (UK)	Historical early warning of slow-poisoning of urban soils.	(Purves 1966)
	Oslo (SE)	Urban transactions are used to demonstrate the influence of urbanization of chemical soil quality.	(Reimann et al. 2011)
	Vienna (AUT)	Estimation of geogenic background values (relevant for legislation) based on geochemical patterns in urban soil.	(Pfleiderer 2011)
	Taipei (TW)	Identification of toxic contaminants in urban top soil layers.	(Jien et al. 2011)

2. MATERIAL AND METHODS

2.1 City selection and city characteristics

Table S2: City parameters

Parameter	Unit	Vienna 2008	Taipei 2009
Inhabitants	capita	1,674,909	2,607,428
City area	hectare	41,487	27,180
Population Density	capita/hectare	40	96
Gross Regional Product (GDP)	Euro/capita	43,900	34,800

2.2 Substance flow modeling

2.2.1 Model development

Figure S1 displays the generic stock and flow chart.

Figure S1: Generic Cu flow model on 1st level. It covers 9 city internal processes of which 6 represent mainly anthropogenic activities (dark grey boxes) and 3 stand for environmental media (light grey boxes). Exterior processes are splitted in the supply and export as well as receiving waters in the hinterland. Regarding nomenclature, the flow acronyms refer to the type of flow (first three letters), to the source process (second three letters and to the sink process (last three letters). The stock acronyms refer to the type of stock only.

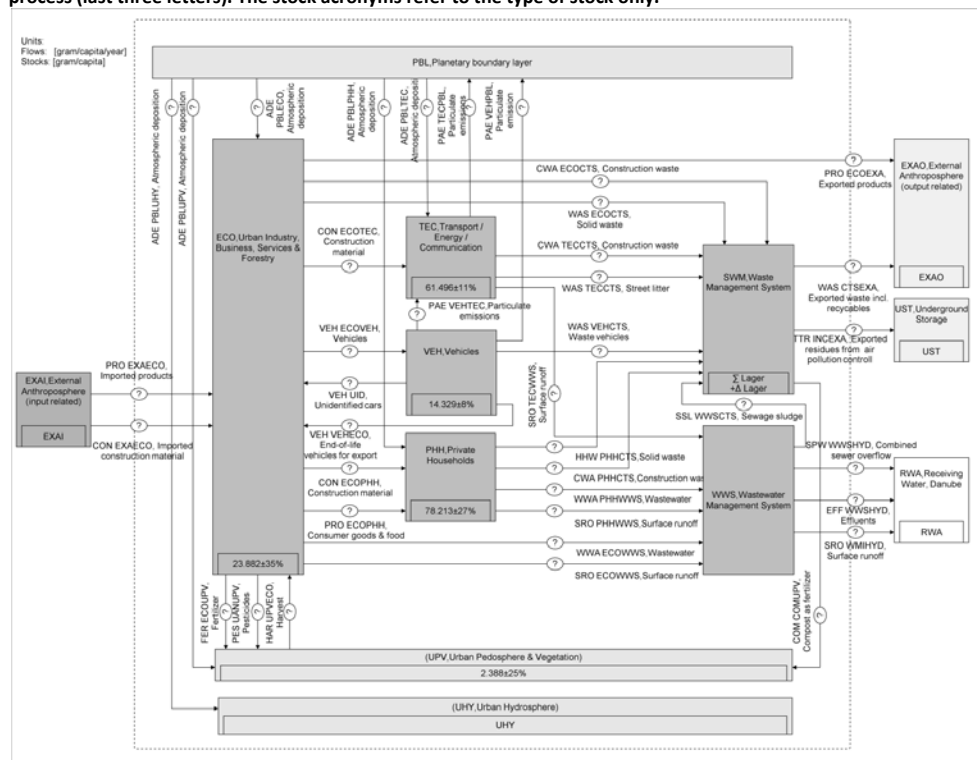
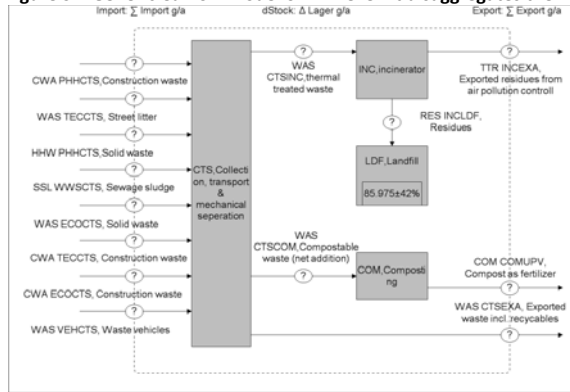


Figure S2: Generic Cu flow model on 2nd level. It disaggregates the “Waste Management System”.



2.2.2 Model equations and data acquisition

This section provides a compilation of flow and stock calculations as well as insights into data acquisition. Additionally, we documented the mathematical model equations, input parameters, uncertainty classification and references for Vienna p. 15, ff. and background data for Taipei on p. 19, ff.

2.2.2.1 Flows & stock description for Vienna and Taipei

2.2.2.1.1 Atmospheric Deposition (ADE PBLUHY, ADE PBLUPV, ADE PBLECO, ADE PBLPHH, ADE PBLTEC)

Urban areas are affected by depositions caused by fireworks, fossil fuel burning, break and catenary wear.

In Vienna, total deposition rate was measured with an rate of 148,7 g Cu/ha/yr on arable land close to Vienna (Spiegel 2003), the dry deposition was measured in town with an average rate of 7,7 g Cu/ha/yr (Kalina et al. 2000). City wide extrapolation on an area base results about 6.6 t Cu/yr, from which 3.5 tons are allocated to building area, 2.8 t Cu/yr to green space (ADE PBLUPV), 0.3 t Cu/yr to open water bodies (ADE PBLUHY) and 0.9 t Cu/yr to traffic surfaces (ADE PBLTEC).

In Taipei, one research was conducted that sampled and analysed the content, source, and transportation mechanism of metals in atmospheric deposition. Dry deposition rate was 3.8 mg/m²/yr. It was related to public facilities surface (ADE PBLTEC) as 0.3 t Cu/yr, private households (ADE PBLPHH) 0.2 t Cu/yr, open water bodies (ADE PBLUHY) 0.1 t Cu/yr, and 0.3 t Cu/yr to green space (ADE PBLUPV).

2.2.2.1.2 Products (PRO EXAECO, PRO ECOPHH)

The import of products stands for consumer goods and pre-products. Both are finally consumed or processed by trade and skilled labor.

In Vienna, we have chosen four main products groups that ended up in the urban production sector or final consumption. (1) Cu containing goods like sheet metals, wires, cable pies and pre-products were taken from a national Cu flow study (Daxbeck et al. 2006). To allocate the flows on different sectors (buildings, infrastructure, production industries), we used global market shares of specific Cu products (ICF 2001). The number of employers in the construction and production sector (Hauptverband der österreichischen Sozialversicherungsträger SV 2011; Statistik Austria 2009c) were used as proxy to downscale from the national to the urban level. Therefore, 4,600 t Cu/yr are allocated to the production industry and is part of the product flow (PRO EXAECO). 11,800 t Cu/yr were allocated to the construction sector (follow sec. 2.2.2.1.3). (2) Electrical and electronic appliances (EEA) are monitored when they placed on the market and when they turn to waste. To estimate the flow, we used the difference of imports and exports on EEA from the regional, mass based trade statistic (Magistratsabteilung 05 2011) and a mean Cu content based on a

basket of 11 representative goods (Truttmann et al. 2005; Oguchi et al. 2011; Hausmann 2005; Wittmer 2006). To estimate the allocation, we used the national sale statistics for household consumption, the population ratio as proxy for downscaling to the regional level (EAK 2009). This resulted about 1,500 t Cu/yr for PHH and 500 t Cu/yr for ECO, respectively. (3) Vehicles were calculated in sec. 2.2.2.1.4 and resulted 2,200 t Cu/yr (VEH ECOVEH), and (4) Food contains essential Cu for human dietary and varies with the type of food. To estimate the flow, we used sixteen food categories (grain, oilseeds, and so on) from the Viennese trade statistic (Magistratsabteilung 05 2011) and given Cu concentration from the National Nutrient Database (USDA 2011). Therefore, about 2 t Cu/yr were imported with food and fully allocated to PHH. Totally, 8,800 t Cu/yr are imported via products (PRO EXAECO) of which 1,500 kg Cu/yr (PRO ECOPHH) enters the PHH as EEA and food.

In Taipei, top-down and bottom-up approaches were adopted to estimate of Cu in products. Since “import” generally happens at a national level, there was no urban statistics of imported goods into the city. Hence, the proxy of total retail sales of consumer goods and areas of factories were used to downscale to the urban level for the imported products (PRO EXAECO), as 16,000 t/yr of Cu contained.

Because there is lacking statistics of products consumption within the city, we used data of recycling to estimate backward. In Taiwan, people can recycle household products with certain recycling fee as feedback. Therefore, bottom-up method was adopted to estimate the quantity of consumer goods, with the assumption that Taiwanese purchase a new product when they discard the old one. In terms of Cu-based products, two categories were selected: Waste Electrical and Electronic Equipment (WEEE) and Waste Computer Appliances (WCA). We took statistics from Recycling Fund Management Board, which is an official organization in charge of executing the recycling fund system. Estimation showed that around 5,000 t Cu/yr was consumed in household commodities (PRO ECOPHH).

2.2.2.1.3 Construction material (CON EXAECO, CON ECOTEC, CON ECOPHH, CWA ECOCTS, CWA TECCTS, CWA PHHCTS, CWA CTSLDF)

The construction sector heavily uses Cu for coatings, plumbing equipments and telecommunication and electricity grids in buildings.

In Vienna, the imported Cu as construction material including pipes for plumbing and heating, sheets for roof and outdoor applications as well as cables and wires like building wires for electric currents and telecommunications. As calculated in sec. 2.2.2.1.2, 11,800 t Cu/yr (CON EXAECO) enter the construction sector, of which 9,400 t Cu/yr accumulates in buildings and 2,400 t Cu/yr infrastructure (CON ECOTEC). Specific land use categories were used to add 6,600 t Cu/yr to the PHH (CON ECOPHH) and 2,800 t Cu/yr to industrial and business buildings.

If it comes to construction waste, the total flow was categorized in three groups: (1) Demolition Material amounted 1.8 Mio t/yr of which the Cu fractions are mostly unknown. So, we picked out the top 5 construction waste flows from the regional waste statistic (Wiener Umweltschutzabteilung MA22 2011) and multiplied them with corresponding Cu contents from literature (Brunner and Stampfli 1993; Schnöller et al. 2010; Arx 2006). So, 800 t Cu/yr were estimated in demolition material. Allocation of flows to the waste generators PHH, ECO and TEC was done by a sector based consumption ratio and the land use ratio. Direct disposals were neglected after the economic benefit of recycling omits landfilling. (2) Cu Scrap was collected with an rate of 3,200 t Cu/yr (Wiener Umweltschutzabteilung MA22 2011) and is estimated to be fully recycled. Allocation followed the same routine as for the demolition material. (3) Collected cables were explicit recorded in the local waste statistics. Hence, we used measured Cu contents in cables (Skutan 2008) and the same allocation proxy as for the demolition material. The total construction waste flows were 3,000 t Cu/yr from private households (CWA PHHCTS), 1,300 t Cu/yr from industry and business (CWA ECOCTS) and 700 t Cu/yr from technical infrastructure (CWA TECCTS).

In Taipei, we determined the imported construction material from infrastructure and buildings import in Taiwan. The economy and transportation budget for infrastructure and the newly built floor area for

buildings were included to downscale statistics from Taiwan to Taipei. The two categories contributed to 45.3% of the total Cu consumed in Taipei which was 13,000 t/yr (CON EXAECO).

We allocated the imported construction material to construction material consumption in economic sectors, private households and infrastructure by the proportion of land-use area. Cu consumed in households and infrastructure was then estimated to be 8,000 t/yr (CON ECOPHH) and 4,000 t/yr (CON ECOTEC), respectively.

For construction waste, construction and demolition waste (C&D waste) and total demolished floor area of the year were considered. Allocation for households, infrastructure and economy sectors were determined by the corresponding area of land-use in Taipei. Results show that Cu in construction waste are 150 t/yr from private households (CWA PHHCTS), 50 t/yr from infrastructure (CWA TECCTS) and 3 t/yr from economy sectors (CWA ECOCTS). The relatively low amount of construction waste produced from infrastructure and economy sectors may be due to the longer lifespan of the infrastructure and buildings.

2.2.2.1.4 Vehicles (VEH ECOVEH, WAS VEHCTS, VEH UID, VEH VEHECO)

Cu is extensively used in vehicles and rolling stocks. It is part of electrical components, heat transfer devices, and bronze sleeve bearings. Hence, countless fittings, fasteners, and screws are made from brass (European Copper Institute (ECI) 2011).

In Vienna, the modal split in the traffic sectors allocates 1/3 of all trips to public transport, 1/3 to private cars and 1/3 to non motorized traffic like cycling and walking. So, private and public transport fleets are of equal importance for Cu calculations. To estimate the inflow, we used official registration statistics for vehicles like cars, busses, lorries and motor bikes (Statistik Austria 2008), the rolling stock alteration of the public transport provider (Lebhart 2010), and the average Cu content per vehicle found in the literature (European Copper Institute (ECI) 2011; Bertram et al. 2002; Struckl 2007). This results an inflow of 2,200 t Cu/yr (VEH ECOVEH). The total outflow has not been statistically reported. So, the whereabouts of EOL cars can't be determined at full scale. We carried out a national car balance in order to estimate the number of unidentified cars. Hence, the Viennese car ownership of 15.3% acts as proxy for regional scaling (Statistik Austria 2008) and an average Cu content per car was used to estimate Cu flows. This results an outflow of 880 t Cu/yr of which 240 t Cu/yr enter a shredder (WAS VEHCTS), 140 t Cu/yr are legally exported to foreign countries (VEH VEHECO) and 500 t Cu/yr remain unidentified in terms of their whereabouts (VEH UID).

In Taipei, private vehicles dominate transportation, especially the sedans and scooters. Since statistics of newly sold vehicles were absent, Cu estimation was top-down from the national data. Vehicles which are ready to be in use were assumed to be the sales volume of cars and scooters/motorcycles of the study year. By the Taipei-Taiwan ratio of newly registered vehicles and Cu content 1.4% (Bertram et al. 2002), sales volumes in Taipei was estimated to be 500 t/yr of Cu (VEH ECOVEH).

We assumed that all end-of-life vehicles (ELV) were treated as recyclable waste. Since data of recycled ELV in Taipei was lacking, we used statistics of recycled ELV in Taiwan and Taipei-Taiwan ratio of newly registered vehicles to top down the estimation, as a result of 200 t/yr of Cu in waste vehicles (WAS VEHCTS).

Amount of registered vehicles difference between 2008 and 2009 was assumed to be the exported ELV, since the change of ELV was unknown and unclear. Hence, 90 t Cu/yr in ELV (VEH VEHECO) was estimated to leave the city boundary.

2.2.2.1.5 Particulate emissions (PAE VEHTEC, PAE VEHPBL, PAE TECPBL)

Particulate emissions are driven by wearing, weathering and corrosion processes. Diffuse emission patterns are characteristic for urban areas. They end up in urban soil and surface runoff.

In Vienna, we considered three main emission sources. First, brake wears from low duty vehicles was estimated according to the fleet mileage (Holzapfel and Riedel 2011), average total wear rates (Winther and Sleno 2010) and Cu concentrations in brake linings representing the Austrian fleet (Figli et al. 2010). Finally we used transfer coefficients from the Netherlands National Emission Inventory (Hulskotte et al. 2006; Oonk et al. 2005) in order to examine the flows towards ambient air, road surfaces and vehicle depositions.

Second, brakes and wheels from rolling stock (trams, trains) emit Cu too. To estimate the flows, we processed the individual network lengths (ÖBB-Holding AG 2008; Wiener Linien 2008), normalized Cu abrasion rates from railways (Burkhardt et al. 2005) and the transfer coefficients from individual sources to ambient air, railroad, surrounding and vehicle deposits (Müller et al. 2008). Third, catenary wear contains 99.5-99.9% Cu on a mass basis. To estimate the wear entering urban sinks, we used the track lengths, corresponding cross sections at the time of installation and replacement, an average lifetime of 40 years and transfer coefficients were derived from a SFA study that balanced Cu flows on 1 kilometer railroad (Müller et al. 2008). Fourth, flows from roofs were estimated according to (Odnevall Wallinder et al. 2007) with a rate of 1.5 g Cu/m²/yr. The Cu roof area was gathered from local tin smiths (Wocilka and Höfner 2011), the total roof area and average roof slope was delivered by city authorities who once plotted the inclinations for estimating solar energy potentials (Kubu 2011), the regional precipitation rate (Lebhart 2010), the SO₂ concentration in precipitation was equally set with monitoring data in ambient air (Augustyn et al. 2010), and the pH value with 5.

Allocating the single results that a) the vehicles release 5 t Cu/yr to the ambient air (PAE VEHPBL), 2.2 t Cu/yr occur as road debris (PAE VEHTEC), and b) the catenaries release 0.6 t Cu/yr to ambient air (PAE TECPBL), 2.0 t Cu/yr to surrounding urban surfaces like roads and facades. Cu deposits on vehicles were not taken into further account.

Taipei City is a rather young Asian urban system and materials used in roof are different from those in Europe. Cu flows of trains, trams and roofs were not taken into account. Airborne particulates from light and high duty vehicles were considered in break wear estimation, with mileage (Department of Statistics 2007) and the wear rate which was the same as in Vienna. Mass of Cu in tire wear were estimated through the top-down data of registered vehicles in Taipei, the tire wear factor, and Cu content in the tire wear emission. Around 6 t Cu/yr were emitted due to break wear and tire wears (PAE VEHPBL).

2.2.2.1.6 Waste Water flows (SRO ECOCTS, SRO PHHWWS, SRO TECWWS, SRO WMIHYD WWS ECOWWS, WWS PHHWWS, SSL WWSCTS, SPW WWSHYD, EFF WWSHYD)

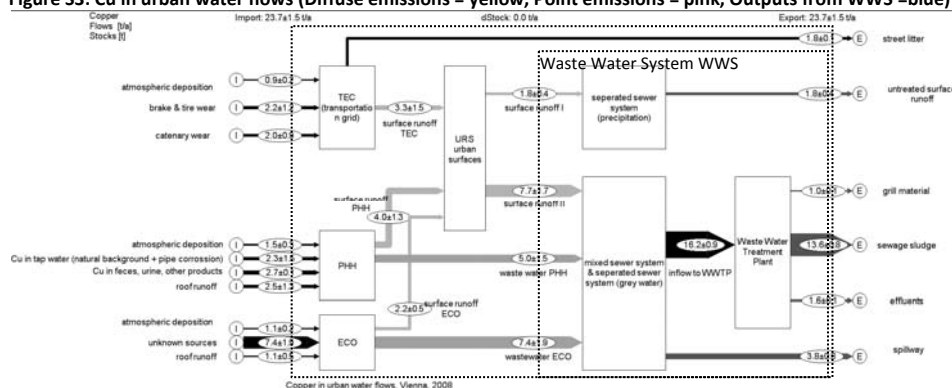
Waterborne Cu flows receive inputs from diffuse emissions, surface runoff and grey water from private households, Cu processing industry and skilled labor.

In Vienna, the majority of collected water flows entered the mixed sewer network and the waste water treatment plant (WWTP) downstream. Those flows become part of grill material, sewage sludge or effluent. Combined sewer overflow enters the receiving water without treatment. The separated sewer network takes up parts of the surface runoff and transports it to the receiving water. We established a separate SFA model (Figure S3) in order to balance waterborne Cu flows.

- Measured flows were available for the WWTP (Kroiss et al. 2008). Therefore, 16.2 t Cu/yr enter the plant of which 13.6 t Cu/yr (SSL WWSCTS) are transferred to the sewage sludge, 1.6 t Cu/yr to the effluent (EFF WWSHYD) and 1.0 t Cu/yr to the grill material.
- Diffuse emissions enter urban surfaces via surface runoff. They calculation of particulate emissions from the transport sector (low duty cars, trams and trains, catenary), building sector (Cu roofs) and atmospheric deposition is described at sec. 2.2.2.1.5. The surface runoff from the transportation grid covers brake & tire wear and catenary with an amount of 3.3 t Cu/yr (SRO TECWWS). The surface runoff from PHH carries atmospheric deposition and roof runoff with a flux rate of 4.0 t Cu/yr (SRO PHHWWS). The surface runoff from ECO covers 2.2 t Cu/yr (SRO ECOWWS). The ratio of separated and mixed sewer network length of 19:81 (Lehmann 2011) is used as proxy to allocate the surface runoffs to the two sewer types. 1.8 t Cu/yr enter the separate sewer system which transports the Cu to the receiving water. 8.9 t Cu/yr enter the mixed sewer system which of 50% enter receiving water as spillover (Fenz 1999) with a flux rate of 3.8 t Cu/yr (SPW WWSHYD).
- Two point sources are relevant. (1) Waste water in PHH covers the final use of tap water as well as Cu from anthropogenic activities. (1.1) Viennese tap water covers the geogenic Cu content and Cu corrosion from pipes. Data on local water consumption statistics excluding the losses (Daxbeck et al. 1996; Tomenendal 2011) and measurements of Cu concentrations at the network endpoints in

households (Magistratsabteilung 31 2008) result a flux rate of 2.3 t Cu/yr. (1.2) Human off-flows like feces, urine and skin particles result about 760 mg Cu/cap/yr (I C Consultants Ltd 2001; Lampert et al. 1997). Other emission sources like residues from food preparation, washing dishes, toilet papers, washing clothes and cleaning activities count for 843 mg Cu/cap/yr (Baccini et al. 1993; I C Consultants Ltd 2001). The multiplication with Viennese population size results 2.7 t Cu/yr. In total, the flux rate is 5.0 t Cu/yr (WWS PHHWS). (2) Waste water data from business and industries were restricted for access. As a consequence, we calculated the flow by stressing the mass balance principle with a flux rate of 7.4 t Cu/yr (WWS ECOWWS).

Figure S3: Cu in urban water flows (Diffuse emissions = yellow, Point emissions = pink, Outputs from WWS =blue)



In Taipei, the percentage of houses connected to public sanitary sewers is 100%. There are three wastewater treatment plants in operation for Taipei: Dihua, Neihu, and Bali. Local statistics showed that Cu in untreated and treated wastewater from Neihu WWTP were not detectable (< 0.02 mg/L). To estimate the flow, we set the detection limit of 0.02 mg Cu/l as maximum concentration. The estimation turned out to be 9.8 t/yr (WWA PHHWS) and 2.8t/yr (TWA WWSHYD). By the ratio of different land uses, Cu in surface runoff was 0.02 t/yr (SRO ECOWWS), 0.5 t Cu/yr (SRO PHHWS) and 0.4 t Cu/yr (SRO TECWWS). Cu in sewage sludge was then estimated to be 60 t Cu/yr (SSL WWSCTS). To estimate the flow, we used the sewage sludge flow with 65,700 tons (sum of Bali, Dihua, and Neihu sewage farms) and Cu concentrations with 887 ppm from Bali Sewage Farm (Shih 2007).

2.2.2.1.7 Solid waste (HHW PHHCTS, WAS ECOCTS)

Cu in solid waste streams are much sought after by stockholders in the recycling and recovering business. Due to the economic incentivizes waste collection strategies and technologies that gain valuable metals are continuously improved.

In Vienna, waste from private households, industry and business covered four waste streams: (1) Municipal solid waste and likewise fractions from business and industry was collected by public service providers. It amounts 579,888 t/yr, while recent studies in Austria determine the concentration from 1,800 – 2,200 mg Cu/kg (Skutan and Rechberger 2007). (2) Waste from industry and business contains four industrial waste fractions like waste from punching and planking, Cu chloride, non-ferrous metal scrap and electroplating sludge. To estimate the flow, we used local waste statistics (Wiener Umweltschutzabteilung MA22 2011) and waste specific Cu contents derived from personal correspondence (Daxbeck et al. 2006). (3) Bulky waste composition varies all over Austria due to different collection schemes. In Vienna, about 70.000 t are collected as composite materials, wood and metals (Wiener Umweltschutzabteilung MA22 2011). After the Cu content is varying to a large extent, we used a broad bandwidth from 1,800-3,300 mg/kg (Skutan and Brunner 2006). (4) Waste electrical and electronic equipment (WEEE) covers large apparatuses, fridges, and freezers, monitors, small WEEE and lamps. To estimate the Cu flow from private households, we used collection data from the national coordination center for WEEE and Cu concentrations from a Swiss Cu flow

study (Wittmer 2006). Those from business were not exclusively reported for Vienna. Therefore, the national amount was downscaled by the ratio of WEEE from private households to total WEEE flow in Vienna. Summarizing, about 1,700 t Cu/yr were generated by households (HHW PHHCTS) and 5,400 t Cu/yr by industry and business (WAS ECOCTS).

In Taipei, household waste includes the trash collected by cleaning squad and by people themselves, as well as the recyclables. Result shows that 1,700 t/yr of Cu contained in household waste (HHW PHHCTS). In Taiwan, industrial waste comprises from industries and service sectors. Since there was nearly no factory in Taipei, industrial waste was presumed to come from service sectors, whose composition is similar to household waste. Because the process ECO was defined as the industrial sector, the amount of industrial waste transported by industrial or waste management institutes to incinerators and landfill sites were considered. Cu concentration in industrial waste was assumed to be as the same as bottom ash and fly ash from incinerators with a rate of 60 t Cu/yr (WAS ECOCTS).

2.2.2.1.8 Exported products (PRO ECOEXA)

Exported products stand for valuable goods which cross the city boundary.

In Vienna, exported products cover three sub-flows. First, the exported vehicles are traded as goods and were estimated to be 140 t Cu/yr (VEH VEHECO). Second, the number of unregistered EOL vehicles was assumed to be exported with 500 t Cu/yr (VEH UID). Third, fabricated goods from the production industry lack of sound data after the import/export statistics are based on the headquarter approach instead of a territorial allocation. So, we estimated 2,000 t Cu/yr with the help of import/export statistics and subtracted residues from production. In total, 2,600 t Cu/yr leave Vienna in a physical product form (PRO ANTEXA).

In Taipei, the same estimation assumption was made as in Vienna, and products exported from Taipei City were estimated to be 8,000 t Cu/yr.

2.2.2.1.9 Exported waste including recyclables (WAS CTSEX A)

Exported waste flows are directed to recycling or disposal facilities out of the city. They cover valuable Cu which is recovered as well unrecovered Cu in residues from incineration.

In Vienna, we combined the generated waste flows, individual information regarding the whereabouts in recycling facilities and specific Cu contents. The flow is summarized by individual flows like recyclables within construction waste, waste from industry and business, and recycled EOL vehicles to an extend of 11,100 t Cu/yr (WAS CTSEX A).

Taipei City incinerated household waste from Keelung City too and stored the bottom ash temporarily during the period of constructing new landfill in Keelung. Likewise, Keelung City would provide landfill service of waste from Taipei City and temporary depository fly ash monolith. The temporary repository of bottom ash and fly ash will be sent back to the original city after the new landfill is constructed. Statistics of Taipei City showed that in 2003 total weight of bottom ash conveyed to Keelung City was 96,000 tons. We also assumed that ELV, scrap, and recyclables were included in exported waste, result in total 2,700 t Cu/yr (WAS CTSEX A).

2.2.2.1.10 Incineration: Mixed waste, exported residues for underground storage, residues, disposable waste (WAS CTSINC, TTR INCEXA, RES INCLDF)

Waste incinerators are used to treat household and toxic industrial waste by temperatures up to 1,200 °C. While microorganisms like bacteria, fungi and virus as well as organic compounds are mineralized, heavy metals like Cu are transferred to bottom ash and APC residues. The whereabouts of residues depends on national policies and varies from bottom ash as construction material to underground disposal for filter APC residues.

In Vienna, four plants secure the treatment of household waste, residues from mechanical sorting, bulky waste fractions, street litter and some minor waste fractions with an amount of 600,000 t/yr. To estimate the plant load, we used Cu concentrations for six different waste flows that stem from plant specific monitoring reports and waste flow measurements (Boller 2002; Skutan and Brunner 2006; Skutan and Rechberger 2007; Umweltbundesamt 2000; Arx 2006; Hausmann 2005). Therefore, about 1,140 t Cu/yr enter the incinerators (WAS CTSINC). Public secondary waste flow data (Wiener Umweltschutzabteilung MA22 2011) and measured Cu concentrations in a representative Austrian waste to energy plants (Skutan and Rechberger 2007; Taverna et al. 2011) result 990 t Cu/yr in bottom ash (RES INCLDF) and 16 t Cu/yr in APC residues (TTR INCEXA). The bottom ash is used as aggregate to construct concrete blocks for landfill stabilization that ends up at a Viennese landfill. APC residues are heavily contaminated with salts and heavy metals. In 2008, they were exported to Heilbronn, an underground storage facility in Germany.

In Taipei, three incinerators operate for Taipei: Beitou, Neihu, and Muzha. According to local statistics, waste transported into incinerator was 640,000 ton in 2009. Cu content was estimated from the Cu concentration in fly ash and bottom ash. Cu in municipal solid waste (MSW) was 200 t/yr (WAS CTSINC). Residues from incinerators consist of fly ash and bottom ash, which contained 200 t/yr of Cu (RES INCLDF). Due to the policy “Zero Waste” of the local government, the waste amount in Taipei has decreased, and no statistics showed that residues from incinerators were exported from the city. Hence, Cu in exported residue for underground storage is zero (TTR INCEXA) in Taipei’s case.

2.2.2.1.11 Composting: Compostable waste, residues, compost as fertilizer (WAS CTSCOM, RES COMCTS, COM COMUPV)

Composting degrades biogenic waste in terms of volume and mass. Quality proven compost can be used as nutrient supplier for plants and soil improver.

In Vienna, one compost plant has been operated. The inputs stem from separated biomass collection and cover materials such as greencut and uncooked vegetables from households. To estimate the Cu flows throughout the plant, we used monitored bulk flows, and derived average dry matter contents as well as Cu concentrations from the literature. The inflow covers compostable raw materials and residues from anaerobic digestion in a biogas plant (Magistratsabteilung 48 2009). Data lacks prevented full Cu balance on plant level. So we set the output as net addition to plant. We assumed a dry matter ratio of 30% and a Cu concentration of 83 mg/kg dry matter and results 1.4 t Cu/yr (WAS CTSCOM). About 33.000 t valuable compost is used for land applications in Vienna (Republik Österreich 2001b; Weinmar 2011). Legal Cu concentrations for high quality compost A+ was set with 70 mg/kg dry substance (Republik Österreich 2001a) and resulted 1.4 t Cu/yr (COM CTSCOM = COM COMUPV).

In Taipei, incinerator plants in Taipei have auxiliary spaces to process compost, which has nothing to do with incinerators themselves. Uncooked food waste scraps are collected to incinerator plants and through the process of dehydration, fermentation, etc., they become compost as product. People or companies can take the compost if they need. Since there are few livestock industries or agricultural activities in Taipei, the compost here was assumed to be the treated uncooked food waste without residues. Due to limited data, the official statistics of uncooked food waste scraps in Taipei City and compost from Muzha Refuse Incineration Plant were adopted to estimate total compost in Taipei, which included 0.04 t Cu/yr (WAS CTSCOM, COM COMUPV).

2.2.2.1.12 Pesticides (PES ECOUPV)

The agricultural sector applies Cu as purposeful fungicides and bactericide on fruits and vegetables. The utilization of Cu dates back to beginning of the 19th century and was known as “Bordeaux-Brühe” It is a mixture of chalk and aquatic Cu-sulphate-dissolution and bans the evolution of mildew. In 2009, the European Commission decided to constrain Cu form agricultural appliances after 2016 (Berger et al. 2011). Member states are enforced to establish national risk assessments in order to regulate Cu use.

In Vienna, the flow was calculated based on data on Austrian pesticide and plant protection product use (Berger et al. 2011) for wine, fruits and vegetables & crops and Viennese agricultural land use areas. In total, about 2.4 t Cu/yr are released by pesticides (PES ECOUPV).

In Taipei, there are barely agricultural activities; the agricultural area is 5.31 km², which is around 2% of total area of the city. Hence, estimation of Cu in pesticides was not included for Taipei's case.

2.2.2.1.13 Fertilizer (FER ECOUPV)

Manure, mineral fertilizer and harvest residues are common fertilizer inputs into agricultural soils. Especially manure is known to have a significant Cu concentration. Concentrations vary from 40-300 mg Cu/kg manure depending on the type of animal and foodstuff (Zethner et al. 2007). As an example, Cu is given to piglets in high doses to protect them from various diseases in the first weeks. Mineral fertilizer and harvest residues cover minor Cu as trace element.

In Vienna, the agricultural sector plays a minor role in terms of land use and production rates. About 15% of urban area is used to produce working animals, crops, wine and vegetables for local supply primarily. The Cu release was estimated by multiplying normalized Austrian and German Cu inputs (Fricke and Höhl 2000; Umweltbundesamt 2001; Zethner et al. 2007; Berger et al. 2011) with agricultural area classified into viticulture production, fruit production, and agriculture & horticulture (Fitzthum 2009). In total about 2.2 t Cu/yr are released by fertilizer applications (FER ECOUPV).

In Taipei, there are barely agricultural activities; the agricultural area is 5.31 km², which is around 2% of total area of the city. Hence, estimation of Cu in fertilizer was not included for Taipei's case.

2.2.2.1.14 Technical Infrastructure (TECstock)

Cu is mainly used for electricity transmission and information transfer.

In Vienna, three types of networks are taken into account. First, the electricity grid covers ~3.700 km of overhead lines and ~ 22.300 km of underground cables (Wien Energie 2010). The stock was estimated based on voltage classes, cable lengths, cross sections and specific masses according to the Swiss network (Wittmer 2006). In total, about 89,200 t Cu provide the public energy supply. Second, Cu in the telecommunication network is estimated by a proxy based on the number of business units. Combining data from Australia and Sydney (van Beers and Graedel 2007) with Viennese business statistics resulted about 13,300 t Cu. Third, catenaries supply the 2nd largest tram network worldwide as well as national and local railway tracks with electricity. The Cu stock of 1,000 t is estimated based on the track length, cross sections, Cu content, and lifetime data. Summarizing, the infrastructure covered about 103,000 t Cu. In reality, the stock is larger than estimated due to Cu cables in subway lines and power transfer stations which were not taken into account.

In Taipei, the electricity transmission system and telecommunication system were taken into account. The electricity grid included 2,100 km overhead power lines and 310 km underground power lines (Lin 2003). For the telecommunication network, estimation was made according to case of Australia and statistics of household and business number in Taipei which gave a result of 24,000 t Cu. Cu stock in infrastructure was aggregated as 32,000 t Cu. Since Cu stock in cables and wires of MRT (Mass Rapid Transit) was not included, the actual value stock maybe larger than the estimated one.

2.2.2.1.15 Economy and private households (ECOstock, PHHstock)

The process economy and private household covers stocks like roofs, water pipes, heating systems, telecommunication and electricity networks as well as consumer goods.

In Vienna, the building structure and housing technology is comparable to Swiss standards. Using per capita data from Switzerland (Wittmer 2006) result about 134,000 t Cu in buildings. The land use area is used as proxy to allocate 94,000 t Cu to private households and 40,000 to economy. Cu in consumer goods like washing machine, dryer, electronics equipment and minor items like keys and coins stands for 37,000 t Cu. They were allocated to private households.

In Taipei, we selected items based on products listed in recycling fund system to estimate Cu stock in consumer goods. Assumption was made that those Cu-contained products included in recycling fund system were those comparatively highly used in households in Taiwan. The lifespan of different products were presumed to be uniform distribution. These consumer goods were either electrical and electronic appliances (EEA) or computer appliances. Five items for Cu stock calculation were color TV, air conditioner, washing machine, laptop, and desktop computer. With numbers of hundred households in Taipei as 9,628.31 in 2009, Cu consumed annually in household appliances and computer appliances in Taipei City were 17,000 t Cu. In economy sectors (ECO), building materials were assumed to be the stocks in it while consumer products were all input to private households (PHH). Electricity and telecommunication wire and cables were taken into account for Cu stock of buildings. In this study, the average length of in-use line in a case study in Tainan (Ou et al. 2007) and the total floor area in Taipei were integrated to do estimation. The Tainan case was reasonable and suitable for estimating because its structure and space usage are common around Taiwan, and hence the case could represent the general housing in Taiwan as well as in Taipei. We used ratio of land-use to allocate Cu stocks of buildings to ECO and PHH. The result shows that 200 t Cu existed in the form of building materials in ECO.

2.2.2.1.16 Vehicles (VEHstock)

The vehicle pool covers private and commercial wheelers as well as rolling stocks.

In Vienna, the calculation is based on the number of vehicles (Statistik Austria 2008; Wiener Linien 2009) and corresponding Cu contents from literature (Bertram et al. 2002; European Copper Institute (ECI) 2011; Hoock 2008; Struckl 2007). The private and public owned fleet consist registered motorized vehicles like cars, lorries and busses as well as rolling stock such as subways and trams. In total, the Cu stock in the transport sector covers 24.000 t Cu.

In Taipei, the estimation included the number of registered cars, buses and scooters in operation. 12,000 t Cu was estimated to be placed in vehicles.

2.2.2.1.17 Urban Soil (UPVstock)

Urban soil covers Cu, which compromises geogenic backgrounds and former anthropogenic inputs.

In Vienna, soil sampling data were reported by the City Authority. We printed a box plot including the detected Cu concentrations in parks and playgrounds. Therefore, about 46 mg Cu/kg soil were multiplied with the area of green space and a soil depth of 30 cm. This results about 4,000 t Cu in urban soil (UPVstock).

In Taipei, there is no local report of the soil sampling data, and hence estimation was made according to EPA public health declaration that Cu concentration in soil generally ranges from 2 to 250 ppm. With the area of green space of Taipei City and 30-cm depth of soil, Cu stock in urban pedosphere was obtained to be 830 t (UPVstock).

2.2.2.2 Vienna – List of mathematical model equations and input parameters

Table S3: Model Equations and flow results

Flow Acronym	Description	Equation	Expected Value [t/yr]	Deviation [t/yr]
01 ADE PBLECO	Atmospheric Deposition ECO	$Dep * A_{ECO}$	1.1	0.19
02 ADE PBLPHH	Atmospheric Deposition PHH	$Dep * A_{PHH}$	1.5	0.26
03 ADE PBLTEC	Atmospheric Deposition TEC	$Dep * A_{TEC}$	0.9	0.15
04 ADE PBLUHY	Atmospheric Deposition UHY	$Dep * A_{UHY}$	0.3	0.05
05 ADE PBLUPV	Atmospheric deposition UPV	$Dep * A_{UPV}$	2.8	0.48
06 COM ANTUPV	Compost, applied on land	$m_{comp} * W_{comp} * C_{comp}$	1.4	0.24
07 CON ECOPHH	Construction Material	$Cons_{AUT} * Proxy_{Market Share} * Proxy_{Employees, bui} * Proxy_{Area}$	6,567	938
08 CON ECOTEC	Construction material	$Cons_{AUT} * Proxy_{Market Share} * Proxy_{Employees, bui}$	2,448	376
09 CON EXAECO	Imported construction material	$Cons_{AUT} * Proxy_{Market Share} * Proxy_{Employees, bui}$	11,817	1,390
10 CWA ECOCTS	Construction waste ECO	$DEM+SCR+CAB$	1,275	217
	Demolition waste (DEM)	$DEM=Dem * C_{dem} * (1-Proxy_{area})$	223	53
	Scrap metal (SCR)	$SCR=Scrab * C_{scrabl} * Proxy_{pre-products, scrap} * (1-Proxy_{area})$	894	207
	Cables (Cab)	$CAB=Cable * C_{cable} * Proxy_{pre-products, cable} * (1-Proxy_{area})$	158	37
11 CWA PHHCTS	Construction waste PHH	$DEM+SCR+CAB$	2,983	509
		$DEM=Dem * C_{dem} * Proxy_{area}$	522	123
		$SCR=Scrab * C_{scrabl} * Proxy_{pre-products, scrap} * Proxy_{area}$	2,092	486
		$CAB=Cable * C_{cable} * Proxy_{pre-products, cable} * Proxy_{area}$	369	86
12 CWA TECCTS	Construction waste TEC	$DEM+SCR+CAB$	682	107
		$DEM=Dem * C_{dem}$	90	14
		$SCR=Scrab * C_{scrabl} * Proxy_{pre-products, scrap}$	249	62
		$CAB=Cable * C_{cable} * Proxy_{pre-products, cable}$	343	86
13 EFF WWSHYD	Effluents	$m_{Cu, inflow} * TC_{Cu, WWTP}$	1.6	0.12
14 FER ECOUPV	Fertilizer	$A_{agr} * m_{fertilizer}$	2.2	0.3
15 HAR UPVECO	Harvest	$m_{uptake} * Proxy_{Land use, agriculture}$	0.3	0.05
16 HHW PHHCTS	Household waste	$MSW+BW+WEE$	1,693	100
	Municipal Solid Waste (MSW)	$MSW=m_{MSW} * C_{MSW}$	1,123	79
	Bulky Waste (BW)	$BW=m_{BW} * C_{BW}$	170	34
	WEE	$WEE=m_{WEE, PHH} * C_{WEE}$	400	52
17 PAE TECPBL	Particulate emissions	$I_{train, tram} * A_{CAT} * 1/\beta_{CAT} * A_{loss} * TC_{MOV, TEC}$	0.6	0.2
18 PAE VEHPBL	Particulate emissions	$LDV+ROL$	5.0	2.5
	Low Duty Vehicles (LDV)	$LDV=mil * e_{LDV} * C_{brake} * TC_{MOV, TEC}$	4.9	2.4
	Rolling Stock (ROL)	$ROL=(I_{train} + I_{tram}) * e_{ROL} * TC_{MOV, TEC}$	0.1	0.3
19 PAE VEHTEC	Particulate emissions	$LDV+ROL$	2.2	1.2
		$LDV=mil * e_{LDV} * C_{brake} * TC_{MOV}$	2	1
		$ROL=(I_{train} + I_{tram}) * e_{ROL} * TC_{MOV}$	0.1	0.6
20 PES UANUPV	Pesticides	$A_{agr} * p_{rate}$	2.4	0.3
21 PRO ECOEXA	Exported products	$VEH_{VEHECO} + VEH_{UID} + FG$	2,574	746
	EOL vehicles for export (VEH VEHECO)	$VEH_{VEHECO} = car_{dereg, exp} * C_{car}$	138	68
	Unidentified Vehicles (VEH UID)	$VEH_{UID} = \text{see row 33}$	498	330
	Fabricated goods from production sector (FG)	$FG=Imp_{good} - Proxy_{exp, good} * Exp_{good}$	1,938	666
22 PRO ECOPHH	Consumer goods & food	$EEA+FOO$	1,484	205
	Electrical and Electronic Appliances (EEA)	$EEA = EEA_{PHH} * C_{EEA}$	1,482	205
	Food (FOO)	$FOO=Food * C_{food}$	2	0
23 PRO EXAECO	Imported products	$CCG+EEA+VEH_{ECOVEH}+FOO$	8,827	728
	Cu containing goods (CCG)	$CCG=Cons_{AUT} * Proxy_{Market Share} * Proxy_{Employees, ind}$	4,619	625
	Electrical and Electronic Appliances (EEA _{total})	$EEA_{total} = EEA_{PHH} * C_{EEA} * EEA_{VE} / \sum EEA_{PHH}$	1,969	325
	Vehicles (VEH ECOVEH)	$VEH_{ECOVEH} = VEH * C_{veh}$	2,237	185
	Food (FOO)	$FOO=Food * C_{food}$	2	0
24 RES INCLDF	Residues from incineration	$m_{bottom ash} * C_{bottom ash}$	986	170
25 SPW WWSHYD	Combined sewer overflow	$Ratio_{SPW} * Ratio_{SST} * (SRO_{TECWWS} + SRO_{PHHWWS} + SRO_{ECOWWS})$	3.8	0.8
26 SRO ECOWWS	Surface runoff ECO	$ADE_{PBLECO} + PAE_{ECOURS}$	2.2	0.5
	Atmospheric Deposition PHH (ADE PBLECO)	$ADE_{PBLPHH} = \text{see row 01}$	1.1	0.19
	Roof runoff (PAE ECOURS)	$PAE_{PHHURS} = Area_{Roof} * Ratio_{Curoof} * r_{roof} * (100 - Proxy_{PHH, roof})$	1.1	0.50

27	SRO PHHWS	Surface runoff PHH	ADE PBLPHH + PAE PHHURS	4.0	1.3
		Atmospheric Deposition PHH (ADE)	ADE PBLPHH=see row 02	1.5	0.26
		PBLPHH)	PAE PHHURS= $Area_{Roof} * Ratio_{Curroof} * r_{roof} *$	2.5	1.26
		Roof runoff (PAE PHHURS)	$PROXY_{PHH,roof}$		
28	SRO TECWWS	Surface runoff TEC	ADE PBLTEC + PAE MOVTEC + PAE TECTEC - WAS	3.3	1.5
		Atmospheric Deposition TEC	TECCTS	0.9	0.2
		Brake & Tire wear on TEC (PAE VEHTEC)	ADE PBLTEC = see row 03	2.2	1.2
		Catenary wear on TEC (PAE TECTEC)	PAE VEHTEC= see row 19	2.0	0.9
		Street litter (WAS TECCTS)	PAE TECTEC= $I_{train, tram} * A_{CAT} * 1/a_{CAT} * A_{loss} *$	1.8	0.05
			$TC_{Mov, TEC}$ WAS TECCTS= see row 40		
29	SRO WMIHYD	Surface Runoff	SRO TECWWS * (1-Ratio _{SST})	1.8	0.05
30	SSL WWSCTS	sewage sludge	$m_{cu, inflow} * TC_{cu, WWTP}$	13.6	1.1
31	TTR INCEXA	Exported APC residues	$m_{APC, res} * CAPC_{res}$	16.4	1.3
32	VEH ECOVEH	Vehicles, consumed in Vienna	Vehicles * $c_{vehicles}$	2,237	185
33	VEH UID	Unidentified cars	$(Car_{dereg} + Car_{impreused} - Car_{rereg} - Car_{shred} - Car_{dereg, exp}) * m_{cu, car}$	498	330
34	VEH VEHECO	EOL vehicles for export	$Car_{dereg, exp} + ccar$	138	68
35	WAS CTSCOM	Compostable waste (net addition)	$m_{comp} * W_{comp} * C_{comp}$	1.4	0.3
36	WAS CTSEXA	Exported Waste and Recyclables	m_{WR}	11,135	729
37	WAS CTSINC	Thermal Treated Waste	MW+SSL	1,137	80
		Mixed waste (MW)	$MW = m_{mw} * C_{mw}$	1,123	80
		Sewage Sludge (SSL)	SSL=see row 30	13.6	1.1
38	WAS ECOCTS	Solid waste ECO	MSW+WIB+BW+WEE	5,390	457
		Waste from industry and business (WIB)	WIB= $m_{wib} * C_{wib}$	5,363	457
		WEE	WEE= $m_{WEE, PHH} * CWEE * PROXY_{WEE, ECO}$	27	3
39	WAS TECCTS	street litter	$m_{street} * C_{street}$	1.8	0.05
40	WAS VEHCTS	Waste vehicles	$Car_{shred} * ccar$	235	39
42	WWA ECOWWS	Wastewater ECO	$m_{cu, inflow} + SPW$ WWSHYD - SRO URSMSW - WWA	7.4	1.9
41	WWA PHHWS	Unknown sources	PHHWS		
		Wastewater PHH	TAB + FUO	5.0	1.5
		Cu in tap water (TAB)	TAB= $m_{tabwater} * Loss_{water} * C_{cu, tabwater}$	2.3	1.52
	Cu in feces, urine, other products (FUO)	FUO= $Cap_{VIE} * (C_{ww} + C_{df})$	2.7	0.11	

Note: t = metric tons per year

Table S4: Model Equations and stock results

Stock Acronym	Description	Equation	Expected Value [t]	Deviation [t]
LDFstock	Landfill	$waste_{LDF} * C_{LDF}$	144,000	61,000
PHHstock	Private Households	BUI+CONS	131,000	35,000
	Buildings (BUI_PHH)	BUI_PHH= $stock_{bui} * Cap_{VIE} * Proxy_{area}$	94,000	32,000
	Consumer Goods (CONS)	CONS= $cons_{cap} * Cap_{VIE}$	37,000	13,000
RDAstock	River Danube	Not identified	-	-
TECstock	Transport / Energy / Communication Infrastructure	CAT+EG+TEL	103,000	11,000
	Catenary (CAT)	CAT= $I_{train, tram} * A_{CAT}$	1,000	50
	Electricity Grid (EG)	EG= $grid_{length} * C_{grid}$	89,000	10,100
	Telecommunication (TEL)	TEL= $Unit * C_{unit}$	13,000	4,100
USTstock	Underground Storage	Not identified	-	-
ECOstock	Urban Industry, Business, Services & Forestry /	BUI_ECO= $stock_{bui} * Cap_{VIE} * (1-Proxy_{area})$	40,000	14,000
VEHstock	Vehicles	$VEH_{stock} * C_{veh, stock}$	24,000	2,000
UPVstock	Urban Pedosphere	$A_{UPV} * t_{soil} * d_{soil} * C_{soil}$	4,000	1,000

Note: t = metric tons per year

Table S5: List of input parameter (non bold=single values, bold=matrices)

Input Parameter	Description of data	Unit	Value	Uncertainty level	Reference	Compiled data
Cap_{DAUT}	Inhabitants Austria	#	8,347,341	1	(Statistik Austria 2012)	
Cap_{VIE}	Inhabitants Vienna	#	1,674,909	1	(Lebhart 2010)	
Dep	Deposition Rate	g Cu/ha/yr	148,7	2	(Spiegel 2003)	
A	Total City Area	ha	41,487	1	(Lebhart 2010)	
A_{UH}	Urban Hydrosphere (Open Water Bodies)	ha	1,933	1	(Lebhart 2010)	
A_{TEC}	Traffic Surface	ha	5,981	1	(Lebhart 2010)	
A_{PHH}	Residential Area	ha	10,267	1	(Lebhart 2010)	
A_{ECCO}	Public Facilities and production areas	ha	4,381	1	(Lebhart 2010)	
A_{UPV}	Green Space Area	ha	18,925	1	(Lebhart 2010)	
$CONS_{AUT}$	National Cu Consumption in 2006	t Cu/yr	$\Sigma=108,000$	2	(Daxbeck et al. 2006)	Table S7

Input Parameter	Description of data	Unit	Value	Uncertainty level	Reference	Compiled data
PROXY _{Market Share}	Global Market Share of Cu use	%	-	-	(ICF 2001)	Table S8
PROXY _{pre-products, scrap}	Ratio of sector based pre-product consumption for Cu scrap	%	-	-		Table S8
PROXY _{pre-products, Cable}	Ratio of sector based pre-product consumption for cables	%	-	-		Figure S4 Table S8 Figure S5
PROXY _{employees, ind}	Ratio of Employees in industries using metals such as Cu on Viennese and Austrian scale	-	0.12	1.5		Table S9
PROXY _{employees, bul}	Ratio of Employees in building industry on Viennese and Austrian scale	-	0.17	1.5		Table S9
EEA _{VE}	Difference of Viennese EEA imports and exports of EEA	t	43,600	-	(Magistratsabteilung 05 2011)	
EEA _{PHH}	EEA consumed by PHH in Vienna	t	32,813	1	(EAK 2009)	
C _{EEA}	Average Cu concentration in EEA	%	0.1-8.0	1.6-3.0	(Truttmann et al. 2005; Wittmer 2006; Hausmann 2005)	Table S11 Table S12, 13
VEH	Number of new registered vehicles and new rolling stock in Vienna	#	80,445	1	(Statistik Austria 2008; Lebhart 2010)	Table S15
C _{veh}	Cu concentration of specific vehicle types	kg Cu/#	4-80	1.3-2.5	(Wittmer 2006; ICF 2000; European Copper Institute (ECI) 2011; Bertram et al. 2002; Struckl 2007; Graedel et al. 2002)	Table S14 Table S15
Food	Food imports to Vienna	t	1,236	1	(Magistratsabteilung 05 2011)	Table S16
C _{food}	Cu concentration in different food types	mg/100 g	0.002-0.900	2	(USDA 2011)	Table S16
Dem	Demolition waste (5 categories)	t	1,770,091	-		Table S21
C _{dem}	Cu concentrations in demolition waste (5 categories)	mg/kg	22-670	-	(Brunner and Stampfli 1993; Schnöller et al. 2010)	Table S21
PROXY _{area}	Area Ratio of residential land use in contrast to total built area	%	70	2	(Lebhart 2010)	Table S18
Scrap	Amount of Cu scrap	t	4,982	-	(Wiener Umweltschutzabteilung MA22 2011)	
C _{scrap}	Cu concentration in cooper scrap	%	90-99.9	-		
Cable	Amount of cable waste	t	2,400	-	(Wiener Umweltschutzabteilung MA22 2011)	-
C _{cable}	Cu content of cables from demolished buildings	%	37	-	(Skutan 2008)	-
Vehicles	Number of vehicles signed on in Vienna and new rolling stock	#	80,445	-		Table S15
C _{vehicles}	Vehicle specific Cu content	kg Cu/vehicle	5-69	-		Table S15
Car _{dereg}	Deregistered Cars in Austria	#	993,354	-		
Car _{imp, reused}	Imported second hand cars in Austria	#	28,696	-	(Wirtschaftskammer Österreich 2012)	
Car _{re-reg}	Re-Registered Cars in Austria	#	738,690	-		
Car _{dereg, exp}	Deregistered and exported cars in Austria	#	37,629	-		
Car _{shred}	Shredded cars in Austria	#	63,975	-	(BMLFUW 2011)	
f _{car}	Proxy representing Viennese and Austrian car ownership	%	0.1534	-		
C _{car}	Cu content in cars	kg Cu/car	24	-		
m _{hhw}	Mixed waste from households and similar institutions	t/yr	579,888	-		
C _{hhw}	Cu concentration in mixed waste	mg/kg	1,990	-		
m _{wib}	Five Waste fractions from industry and business	t/yr	15,216	-	(Wiener Umweltschutzabteilung MA22 2011)	Table S38
C _{wib}	Cu concentrations in specific waste flows	%	12-99.9	-	(Daxbeck et al. 2006)	Table S38
m _{bw}	Collected bulky waste flow	t/yr	69,422	1.5	(Wiener Umweltschutzabteilung MA22 2011)	

Input Parameter	Description of data	Unit	Value	Uncertainty level	Reference	Compiled data
C_{BW}	Average Cu concentration in bulky waste	mg/kg	2,437	2.07	(Skutan and Brunner 2006)	
$m_{WEE, PHH}$	Collected WEEE in PHH of Vienna	t/a	9,044	1	(EAK 2009)	Table S40
C_{WEE}	Cu concentrations of 4 different WEEE fractions	%	1-8	2-5,6	(Wittmer 2006)	Table S40
$Proxy_{WEE, ECO}$	Proxy for allocating WEEE to ECO	-	0.066		(EAK 2009)	Table S42
m_{uptake}	Cu uptake	kg Cu/ha/yr	0.01-0.045		(Berger et al. 2011)	Table S45
$Proxy_{land use, agriculture}$	Agricultural area splitted into different land use categories	ha	$\Sigma=6,281$		(Fitzthum 2009)	Table S45
P_{rate}	Pesticide rate	kg Cu/ha/yr	0-3.0	2	(Berger et al. 2011)	Table S46
A_{agr}	Agricultural area splitted into different land use categories and organic/traditional farming	ha	$\Sigma=6,281$	1	(Fitzthum 2009)	Table S47
$m_{fertilizer}$	Fertilizer rate	kg Cu/ha/yr	0.03-0.55	2	(Fricke and Höhl 2000; Umweltbundesamt 2001; Zethner et al. 2007; Berger et al. 2011)	Table S47
mil	Mileage of low duty vehicles in Vienna	Veh-km	5,694,000.00	-	(Holzapfel and Riedel 2011)	
e_{LDV}	Total wear rate of brake lining in low duty vehicles	g /veh-km	12.5	-	(Winther and Slento 2010)	
C_{brake}	Cu content of brake linings in low duty vehicles	%	14.231	-	(Figl et al. 2010, own calculations to represent the Austrian vehicle fleet)	Table S26
$TC_{VEH, TEC}$	Transfer coefficients describing the ratio of particulate emissions entering individual sinks	%		-	(Müller et al. 2008)	Table S25
l_{train}	Track length of the train network, owned by National Railway Company ÖBB	track km	761	-	(ÖBB-Holding AG 2008)	
l_{tram}	Track length of the tram network, owned by Wiener Linien	track km	363	-	(Wiener Linien 2008)	
e_{ROL}	Cu abrasion rate of break and wheel wear in trams and trains	g/track km/yr	1,200	-	(Burkhardt et al. 2005)	
$l_{TRAM, TRAIN}$	Track length of train and tram network according to the catenary diameter	track km	$\Sigma=1,124$	-	(ÖBB-Holding AG 2008; Wiener Linien 2008)	Table S28
A_{CAT}	Cross section area of individual catenaries	mm	65-120	-		Table S28
a_{CAT}	Average lifetime of a catenary	yr	40	-		
A_{loss}	Average cross section loss over the whole lifetime	%	20	-		
$Proxy_{exp_good}$	Proxy for estimating physical exports based on reported flow data (headquarter approach)	%	50		Assumption	
Imp_{good}	Imported Cu for producing industries		4,619		see flow PRO EXAECO	
Exp_{good}	Cu waste from production		5,363		see flow WAS ECOCTS	
m_{WR}	Sum of seven Cu waste flows that contain potential recyclables (demolition material, Cu scrap, cables, waste from industry and business, bulky waste, WEEE, Recycled EOL vehicles)	t/yr	$\Sigma=11,071$	-		Table S44
$m_{bottom ash}$	Amount of bottom ash and fly ash	t/yr	125,016	1	(Wiener Umweltschutzabteilung MA22 2011)	
$C_{bottom ash}$	Cu concentration in bottom ash	kg Cu/t	7,885	2	(Skutan and Rechberger 2007; Mitterbauer et al. 2009)	
$m_{APC res}$	Amount of APC residues	t/yr	18,189	1	(Wiener Umweltschutzabteilung MA22 2011)	
$C_{APC res}$	Cu concentration in APC residues	t Cu/t	0.0009011	1.009	(Skutan and Rechberger 2007; Taverna et al. 2011)	
$m_{Cu, inflow}$	Cu Inflow to Waste Water Treatment Plant	t Cu/yr	16.2	1	(Kroiss et al. 2008)	
$TC_{Cu, WWTP}$	Transfer Coefficients for Cu in Waste Water Treatment Plant	%	6-85	1	(Kroiss et al. 2008)	Table S32
$Area_{Roof}$	Total Roof Area	m^2	52.000.000	3	(Kubu 2011)	
$Ratio_{CuRoof}$	Percentage of Cu roofs	%	5	-	(Wocilka and Höfner)	

Input Parameter	Description of data	Unit	Value	Uncertainty level	Reference	Compiled data
					2011)	
r_{roof}	Cu Runoff rate	$g/m^2/yr$	1.365	0.654		Table S30
$PROXY_{PHH, \text{roof}}$	Allocation of roof runoff to PHH	%	70	-	(Lebhart 2010)	Table S31
m_{tabwater}	Revenue water	m^3/yr	122.775.000	1	(Tomenendal 2011)	
$LOSS_{\text{water}}$	Water Loss rate	%	10	-	(Daxbeck et al. 1996, adopted for 2008 data)	
$C_{\text{Cu, tabwater}}$	Cu concentration in tap water at the network endpoints (geometric mean of five measurements in private households)	$\mu g/l$	20.675	3.26	(Magistratsabteilung 31 2008)	
C_{ww}	Average Cu disposal in waste water	$mg \text{ Cu}/cap/yr$	843	1	See table	Table S33
C_{bf}	Cu in bodily fluids	$mg \text{ Cu}/cap/yr$	760	1	See table	Table S34
m_{street}	Mass Flow of collected street litter	t/yr	39,495	1	(Wiener Umweltschutzabteilung MA22 2011)	
C_{street}	Cu concentration in street litter	$mg \text{ Cu}/t$	46	2	(Boller 2002)	
m_{mw}	Six mixed waste fractions, thermally treated in Vienna	t/yr	$\Sigma=598,088$	1	(Kronberger 2011)	Table S36
C_{mw}	Average Cu content in six mixed waste fractions	$mg \text{ Cu}/t$	45-1990	0.97-2	(Skutan and Brunner 2006; Skutan and Rechberger 2007; Umweltbundesamt 2000; Boller 2002; Hausmann 2005; Arx 2006)	Table S37
m_{comp}	Applied compost on land	t/yr	33,000	1	(Weinmar 2011)	
W_{comp}	Water content of compost	%	41	2	(Umweltbundesamt 2000)	
C_{comp}	Legal limit of Cu concentration in compost, quality level A+	$mg \text{ Cu}/kg$	70	2	(Republik Österreich 2001a)	
$RATIO_{SPW}$	Percentage of surface runoff ending up as combined sewer overflow	%	50	-	(Fenz 1999)	
$RATIO_{SST}$	Ratio surface water collected by mixed sewer system based on network lengths	%	81	-	(Lehmann 2011; I C Consultants Ltd 2001; Lampert et al. 1997)	
$stock_{\text{bui}}$	Cu stock in buildings	$kg \text{ Cu}/cap$	80	2	(Wittmer 2006)	
$CONS_{\text{cap}}$	Cu stock in consumer goods	$kg \text{ Cu}/cap$	22	2	(Wittmer 2006)	
VEH_{stock}	Number of registered vehicles	#	$\Sigma=850,784$	1	(Statistik Austria 2008; Wiener Linien 2009)	Table S50
$C_{\text{veh, stock}}$	Cu content of different vehicle types	$kg \text{ Cu}/\#$	4-1,500	0.6-12.47	(Bertram et al. 2002; European Copper Institute (ECI) 2011; Hoock 2008; Struckl 2007)	Table S50
$grid_{\text{length}}$	Cable lengths in the electricity grid	km	$\Sigma=22,477$	1	(Wien Energie 2010)	Table S48
C_{grid}	Cu content of different cable types	$t \text{ Cu}/km$	4-11	2	(Wittmer 2006)	Table S48
Unit	Number of different business units and private households	#	$\Sigma=859,397$	1		Table S49
C_{unit}	Cu stock per business unit and private households	$kg \text{ Cu}/\#$	7-620	3		Table S49
t_{soil}	Soil depth	m	0.30	-	own assumption	
d_{soil}	Soil density	t/m^3	1.7	-	own assumption	
C_{soil}	Average Cu concentration in Viennas top soil layer (land use category park and playgrounds)	$kg \text{ Cu}/kg$	46		(Kreiner 2004, data analysis based on published data)	Table S53 Figure S10
$waste_{\text{LDF}}$	Mass of waste, disposed off in Viennas landfills and dumpsites	t	$\Sigma=81 \text{ Mio.}$	1	(Ableidinger et al. 2007)	Table S51 Table S52
C_{LDF}	Average Cu content in old dumped waste fractions	mg/kg	28-2600	2-4	(Brunner and Stampfli 1993; Woisetschlaeger et al. 2000)	Table S51 Table S52

Notes: # = quantity; t = metric ton; kg Cu/# = kilogram copper per quantity; ha = hectare; g Cu/ha/yr = gram copper per hectare per year; Veh-km = vehicle kilometer; track-km = track kilometer; $g/m^2/yr$ = gram per square meter per year; m^2 = square meter; $\mu g/l$ = microgram per liter

2.2.2.3 Vienna - Background data

2.2.2.3.1 Products (PRO EXAECO)

2.2.2.3.1.1 Overview

Four groups of products are estimated to represent more than 90% of Cu in imported products:

1. Cu containing goods (Sheet metal, Wires, Cables, Pipes, Pre-Products etc.)
2. Electrical and electronic appliances
3. Vehicles
4. Food

Table S6: Imported products (PRO EXAECO)

Goods	Flux rate [t Cu/yr]	Deviation [t Cu/yr]	Allocation to ECO	Allocation to VEH	Allocation to PHH
Cu containing goods	4,619	625	4,619	-	
Electrical and Electronic Appliances	1,969	287	486	-	1,482
Vehicles	2,237	185	-	2,237	-
Food	2	0.25	-	-	2
Sum	8,827	341	5,105	2,237	1,484

Note: t Cu/yr = metric tons copper per year; ECO = Industry, business, services and forestry; VEH = Vehicles; PHH = Private Households

2.2.2.3.1.2 Cu containing goods

Table S7: Cu consumption in Austria (Daxbeck et al. 2006)

N°	Austrian Cu Consumption	Flux rate [t/yr]
1	Electrical conductors, cables	33,000
2	Pipes	26,000
3	Sheets	15,000
4	Wires, Cables, Ropes	12,000
5	Rest	22,000
Sum of Cu consumed in Austria		108,000

Note: t/yr = metric tons per year

Table S8: Proxy: Cu Market share (ICF 2001)

	Cables & Wires	Pre-product Cu	Pre-product Cu-Alloy	Σ
Global Cu Consumption	[%]	[%]	[%]	[%]
Building Sector [%]	20	12	8	40
Infrastructure [%]	13	1	1	15
Other Production Industries [%]	19	7	19	45
Total Product Share	52	20	28	100

Table S9: Proxy: Employees

	Austria	Vienna	Proxy	Reference
	[#]	[#]	[]	
Employed people in building industry	275,266	46,736	0.17	(Statistik Austria 2009c; Hauptverband der österreichischen Sozialversicherungsträger SV 2011)
Employed people in industries using metals such as Cu (ÖNACE 2008 C24-C30 and C33)	345,926	42,081	0.12	

Note: # = quantity

Table S10: Vienna Cu consumption by sector: Downscaling and allocation of Cu consumption

	Austria				Vienna				Note
	Cables & Wires [t Cu/yr] (1)+(4)	Pre-product [t Cu/yr] (2)+(3)+(5)	Sum [t Cu/yr]	Proxy: Employees	Cables and Wires [t/yr]	Pre-product [t Cu/yr]	Sum [t Cu/yr]	Allocation to flow	
Building Sector	17,308	37,800	55,108	0.17	2,942	6,426	9,368	CON EXAECO	Follow sec. 2.2.2.1.3
Infrastructure	11,250	3,150	14,400	0.17	1,913	536	2,449	CON EXAECO	Follow sec. 2.2.2.1.3
Other producing Industries	16,442	22,050	38,492	0.12	1,973	2,646	4,619	PRO EXAECO	-
SUM	45,000	63,000	108,000		6,828	9,608	16,436		

Note: t Cu/cap/yr = metric tons copper per capita per year;

2.2.2.3.1.3 Electrical and electronic appliances

Table S11: Cu in electrical and electronic appliances (EEA) and allocation to PHH and ECO

	Cu in electrical and electronic appliances (EEA)	Flux rate [t/yr]	Ratio [%]	Reference
EEA _{JE}	Difference of import and export of EEA	43,586	100	(Magistratsabteilung 05 2011)
ΣEEA _{PHH}	EEA consumed by PHH in Vienna	32,813	75.3	(EAK 2009)
	Large EEA	14,508		
	Fridges & Freezers	4,465		
	Monitors	4,022		
	Small EEA	9,442		
	Lamps	376		
	EEA consumed by ECO	10,774	24.7	

Note: t/yr = metric tons per year;

Table S12: EEA consumed by Private Households in Vienna

EEA consumed by PHH in Vienna	Flux rate [t/yr]	Cu concentration (min) [%]	Cu concentration (max) [%]	Cu concentration (geometric mean) [%]	Uncertainty Level	Flux rate [t Cu/yr]	Deviation [t Cu/yr]
Large EEA	14,508	3.90	8.00	5.586	2.249	810	181
Fridges & Freezers	4,465	1.00	4.00	2.000	3.008	89	20
Monitors	4,022	3.90	7.80	5.515	2.211	222	50
Small EEA	9,442	2.90	5.00	3.808	1.957	360	80
Lamps	376	0.10	0.15	0.122	1.657	0	0
Total (ΣEEA _{PHH})	32,813					1,482	205
Total (ΣEEA _{ECO})						486	134
Total						1,969	287

Note: t/yr = metric tons per year; t Cu/yr = metric tons copper per year;

Table S13: Cu content of EEA

Cu content of electrical and electronic appliances	Reference	Cu concentration	
		[kg/#]	[%]
EEA in general	(Truttmann et al. 2005)	-	3,4
	Hausmann, 2005	-	4,3
Fridges & Freezers	(Truttmann et al. 2005)	2	4,0
	Wittmer, 2006	-	1,0
	Oguchi, Murakami et al., 2011	-	3,4
Washing Machine	(Truttmann et al. 2005)	1,8	2,4
	Oguchi, Murakami et al., 2011	-	3,1
Dishwasher	(Truttmann et al. 2005)	1,3	2,5
	Wittmer, 2006	-	2,0
Microwave	Truttmann et al., 2003	0,9	3,9
TV Set	Truttmann et al., 2003	1,5	5,0
	Oguchi, Murakami et al., 2011	-	0,9 - 3,9
Monitor CRT	Truttmann et al., 2003	1,1	7,8
	Oguchi, Murakami et al., 2011	-	3,9
Computer excl. Monitor	Truttmann et al., 2003	0,5	4,8
	Wittmer, 2006	-	4,0
	Oguchi, Murakami et al., 2011	-	0,9 - 1,0
Video recorder	Truttmann et al., 2003	0,3	6,0
	Wittmer, 2006	-	4,0
	Oguchi, Murakami et al., 2011	-	2,9
Telephone	Oguchi, Murakami et al., 2011	-	10,3
Air conditioner	Oguchi, Murakami et al., 2011	-	17,8

Note: kg/# = kilogram per item

2.2.2.3.1.4 Vehicles (VEH ECOVEH)

Table S14: Cu contents of vehicles – literature overview

Vehicle type & Reference	Cu Content		Notes
	[Mass %]	[kg/#]	
Cars			
<i>(Keoleian et al. 1997)</i>	1,4	19	Cu and Brass in U.S.-Built Car 1994
<i>(ICF 2000)</i>	n.a	17	Wires and Cables in Cars
<i>(Bertram et al. 2002)</i>	1,4	14	PKW from 80's average weight: 1000 kg
<i>(Graedel et al. 2002)</i>	1,4	20	Car produced between 1990-1994 average weight 1395 kg
<i>(Hoock 2008)</i>	2	29	BMW 3 series
<i>(European Copper Institute (ECI) 2011)</i>	n.a.	25	
Busses/Trucks			
<i>(Bertram et al. 2002)</i>	0,5	60 - 80	0,5% assumed total Mass 12.000 kg or more
Locomotive			
<i>(European Copper Institute (ECI) 2011)</i>	n.a.	8000	
Airplane			
<i>(European Copper Institute (ECI) 2011)</i>	2	n.a.	
Motorbikes			
<i>Assumption</i>	2	n.a.	Same as in modern cars
Subway cars			
<i>(Struckl 2007)</i>	3	1.300 – 1.500	Average weight of subway car in Vienna: 42 t
Trams			
<i>Calculation based on (Struckl 2007)</i>	3	700 - 800	Average weight of tram car in Vienna 23 t

Note: kg/# = kilogram per item

Table S15: Import of vehicles and transportation items

Vehicles and transportation items	Number of new registered vehicles in Vienna and new rolling stock	Uncertainty Level	Cu (min)	Cu (max)	Cu (geom. mean)	Uncertainty Level	Flux rate	Deviation
			[kg Cu/#]	[kg Cu/#]	[kg Cu/#]			
New registered vehicles	#		[kg Cu/#]	[kg Cu/#]	[kg Cu/#]		[t Cu/a]	
<i>Cars</i>	63.473	1	20	29	24	1.570	1.529	176
<i>Trucks</i>	8.088	1	60	80	69	1.319	650	53
<i>Motor Bikes</i>	7.856	1	4	6	5	1.657	38	5
<i>Others (mainly heavy machinery, busses etc.)</i>	976	1	25	60	39	2.466	38	11
Change in stock of Wr. Linien 2007/2008 - public transport system								
<i>Subway (Triebwagen)</i>	<i>Cars</i>	1				0.783		
	52		1273	1500	1382		72	5
Total (VEH ECOVEH)	80,445						2,237	185

Note: kg Cu/# = kilogram copper per item

2.2.2.3.1.5 Food

Table S16: Cu imports via food

	Food	C_{food}	Cu	Standard Deviation
	[t/yr]	[mg/100g]	[kg Cu/yr]	[kg Cu/yr]
Fische, Krebstiere, Weichtiere	5.74	0.062	4	
Fleisch, Innereien, genießbarer Schlachtanfall	21.07	0.060	13	
Früchte	33.47	0.057	19	
Gemüse, Wurzeln, Knollen	33.30	0.079	26	
Getränke, alkoholische Flüssigkeiten, Essig	133.25	0.007	9	
Getreide	220.31	0.490	1,080	
Kaffee, Tee, Mate, Gewürze	53.44	0.011	6	
Milch, Milcherzeugnisse, Vogeleier, Honig	42.23	0.013	5	
Müllereierzeugnisse, Malz, Stärke	6.64	0.270	18	
Ölsaaten, Samen, Stroh, Futter	106.12	0.900	955	
Tierische und pflanzliche Öle, Fette, Wachse	167.70	0.002	3	
Verschiedene eßbare Zubereitungen	50.56	0.206	104	
Zubereit. v. Fleisch, Fisch, Krebs u. Weichtieren	7.87	0.061	5	
Zubereit. v. Gemüse, Früchte, and. Pflanzenteilen	40.49	0.068	27	
Zubereitungen v. Getreide, Mehl, Stärke, Milch	34.40	0.490	169	
Zucker und Zuckerwaren	279.10	0.007	20	
Total	1,236		2,463	252

Note: t/yr = metric tons/year; kg Cu/yr = kilogram copper per year; mg/100g = milligram per 100 gram

2.2.2.3.2 Construction material (CON EXAECO, CON ECOPHH, CON ECOTEC)

Table S17: Cu in construction material

Acronym	Sector	Flux rate		Calculation routine
		Expected Value [t Cu/yr]	Deviation [t Cu/yr]	
	Building Sector	9,368	1,338	See Table S10
CON ECOPHH	Private Households	6,567	938	Allocation of construction material in the building sector to PHH: $9368 * 70\% \dots = 6,567 \text{ t/yr}$ $70\% = \text{Proxy}_{\text{Area}}$
	Economy, Business	2,802	400	
CON ECO TEC	Infrastructure	2,449	376	See Table S10
CON EXAECO	SUM	11,817		

Note: t Cu/yr = metric tons per year

Table S18: Proxy area

N°	Land use category	Area [ha]	Allocation to process ...	Reference
1	Residential Area	10,267	PHH	(Lebhart 2010)
2	Public facilities	1,888	ECO	(Lebhart 2010)
3	Industry and Business	2,492	ECO	(Lebhart 2010)
4	Construction Areas Total	14,647		
Proxy = (1)/(4) *100		70%		

Notes: ha = hectare; PHH = Private Households; ECO = Industry, business, services and forestry

2.2.2.3.3 Construction waste (CWA ECOCTS, CWA PHHCTS, CWA TECCTS)

2.2.2.3.3.1 Overview

Table S19: Cu in construction materials

Flow acronym	Waste generator	Demolition material		Scrap metal		Cables		Sum	
		Expected Value [t Cu/yr]	Deviation [t Cu/yr]	Expected Value [t Cu/yr]	Deviation [t Cu/yr]	Expected Value [t Cu/yr]	Deviation [t Cu/yr]	Expected Value [t Cu/yr]	Deviation [t Cu/yr]
CWA PHHCTS	PHH	522	123	2,092	486	369	86	2,983	509
CWA ECOCTS	ECO	223	53	894	207	158	37	1,275	217
CWA TECCTS	TEC	90	14	249	62	343	86	682	107
Total amount of Cu in construction material		835	135	3,235	532	870	127	4,940	563

Note: t Cu/yr = metric tons copper per year; PHH = Private Household; ECO = Industry, business, services and forestry; TEC = Transport, energy, and communication infrastructure

2.2.2.3.3.2 Demolition Material

Table S20: Cu in construction material

Cu content in construction materials	Cu content	
	[mg/kg]	Reference
Total construction waste	670	(Brunner and Stampfli 1993)
Mineral fraction (Average)	22	(Schnöller et al. 2010)
Fine dust from construction waste	34	(Townsend et al. 2004)
Construction waste (literature research)	650 - 1950	(Arx 2006)
Construction timber	65	(Arx 2006)
Tar and Bitumen (Average)	80	(Schnöller et al. 2010)

Notes: mg/kg = milligram per kilogram

Table S21: Cu in construction waste

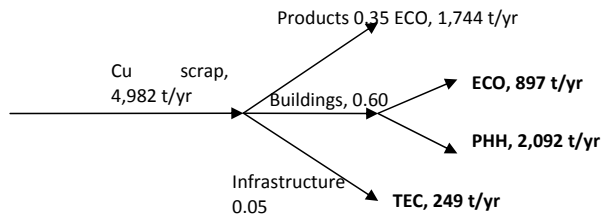
Construction waste in Vienna 2008	Attribution	Cu content		Amount of Waste [t/yr]	Cu in Waste [t/yr]	Proxy _{area}	ECO	PHH	TEC
		[mg/kg]	[t/yr]						
Construction waste	PHH&ECO	670	1.090.976	731	30% for ECO, 70% for PHH	219	512		
Concrete demolition	PHH&ECO	22-34	432.296	12	30% for ECO, 70% for PHH	4	8		
Road construction waste	TEC	670	121.182	80					80
Tar and Bitumen	TEC	80	83.543	10					10
Construction and demolition timber	PHH&ECO	65	42.094	3	30% for ECO, 70% for PHH	1	2		
Sum			1.770.091	~ 830		223	522		90

Notes: mg/kg = milligram per kilogram; t/yr = metric tons/year; PHH = Private Households; ECO = Industry, business, services and forestry; TEC = Transport, energy, and communication infrastructure

2.2.2.3.3.3 Scrap metal

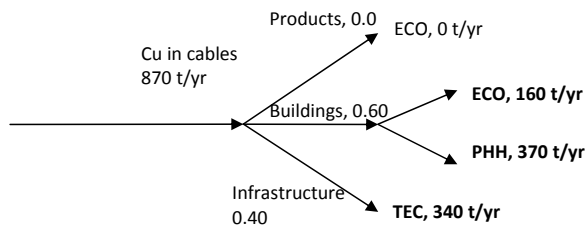
Viennese wastes statistics report that 5,254 t Cu scrap with a Cu concentration of 90-99,9% was collected (Wiener Umweltschutzabteilung MA22 2011). That amounts 4,982 t Cu/yr. We allocated the flows to sectors based on the consumption ratio of “pre-products” first, and to the buildings in ECO and PHH with the land use area.

Figure S4: Allocation routine for Cu scrap (t/yr = metric tons per year; PHH = Private Households; ECO = Industry, business, services and forestry; TEC = Transport, energy, and communication infrastructure)



2.2.2.3.3.4 Cables

Figure S5: Allocation routine for cables (t/yr = metric tons per year; PHH = Private Households; ECO = Industry, business, services and forestry; TEC = Transport, energy, and communication infrastructure)



2.2.2.3.4 Vehicles

Figure S6: National car balances. Flows are given in number of cars per year, stocks are given in number of cars.

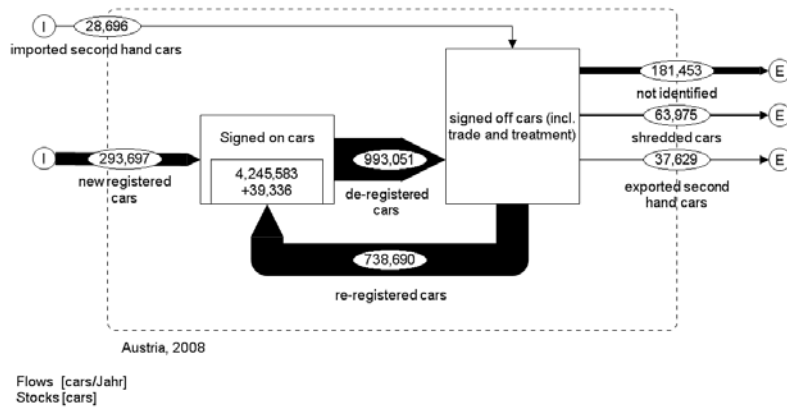


Table S22: National car balance

N°	Flow/stock name	#	Reference / Calculation routine
1	Imported second hand cars	28,696	(Wirtschaftskammer Österreich 2012)
2	New registered cars	293,697	(Statistik Austria 2009a)
3	De-registered cars	993,354	Calculated based on mass balance principle
4	Re-registered cars	738,690	(Statistik Austria 2009b)
5	Stock of signed on cars (31.12.2007)	4,245,583	(Statistik Austria 2008)
6	Stock of signed on cars (31.12.2008)	4,284,919	(Statistik Austria 2008)
7	Stock alteration of signed on cars	39,336	(6)-(5)
8	Shredded cars	63,975	(BMLFUW 2011)
9	Exported second hand cars	37,629	(Wirtschaftskammer Österreich 2012)
10	Not identified	181,756	Calculated based on mass balance principle

Note: # = number of items

Table S23: Downscaling of car numbers from the national to regional scale

Flow acronym	Flow name	Austria	Vienna	Cu [t/yr]	Uncertainty Factor	Deviation [t/yr]
WAS VEHCTS	Shredded cars	63,975	9,788	235	2	39
VEH VEHECO	Exported cars	37,629	5,757	138	3	68
VEH UID	Unidentified cars	181,756	27,809	667	3	330
TOTAL				1,040		339

Note: t/yr = metric tons per year

2.2.2.3.5 Particulate Emissions

2.2.2.3.6 Overview

Table S24: Compiled Results of particulate emissions and the whereabouts

source	sink		Deposition on vehicle	deposition on track	sum
	PBL	TEC			
	[t Cu/yr]	[t Cu/yr]	[t Cu/yr]	[t Cu/yr]	[t Cu/yr]
Process VEH					
Low duty vehicles	4.9±2.4	2±1	3.1	-	10.0
Rolling stock	0.1±0.3	0.2±0.6	0.2	0.9	1.4
<i>Sum</i>	<i>5±2.5</i>	<i>2.2±1.2</i>			
<i>Flow Acronym</i>	<i>PAE VEHPBL</i>	<i>PAE VEHTEC</i>	3.3	0.9	11.4
Process TEC					
Catenary Wiener Linien	0.2	0.2	1.4	-	1.9
Catenary ÖBB	0.4	1.8	0.3	0.6	3.1
<i>Sum</i>	<i>0.6±0.2</i>	<i>2.0±0.9</i>			
<i>Flow Acronym</i>	<i>PAE TECPBL</i>	<i>PAE TECTEC</i>	1.7	0.6	5

Note: t Cu/yr = metric tons copper per year; VEH = Vehicles; TEC = Transport, energy, and communication infrastructure

Table S25: Transfer coefficients

process	Low duty vehicles, brake wear	Train & trams, brake and wheel wear	Train catenary, ÖBB	Tram catenary, Wiener Linien
Ambient air	0.49	0.05	0.14	0.14
Deposition on surroundings (road debris, urban surfaces)	0.20	0.1	0.57	0.76
Deposition on vehicle	0.31	0.15	0.10	0.10
Deposition on track	-	0.7	0.19	0.00
Reference	(Oonk et al. 2005)	(Müller et al. 2008)	(Müller et al. 2008, modified)	(Müller et al. 2008, modified)

Notes: The transfer coefficients are adopted for urban areas

2.2.2.3.7 Brake wear

Table S26: Cu concentration in brake pads (low duty vehicles)

Market share of Car brands in Austria	Cu in brake pads (Figi et al. 2010)	Weighted average Cu content in brake pads of Austrian fleet
[%]	[µg Cu / g brake pad]	[µg Cu / g brake pad]
VW	20.4	138,935
Opel	8.2	169,080
Audi	6.6	106,080
Ford	6.2	151,420
Renault	5.6	139,980
Mercedes	5.4	190,240
BMW	4.9	93,140
Peugeot	4.5	135,375
Toyota	4.4	156,040
Mazda	4.3	142,254
Others	29.5	142,254
	100	142,312

Notes: [µg Cu / g brake pad = microgram copper per gram brake pad

Figure S7: Particulate emissions from vehicles and the whereabouts. Flows are given in metric tons per year.

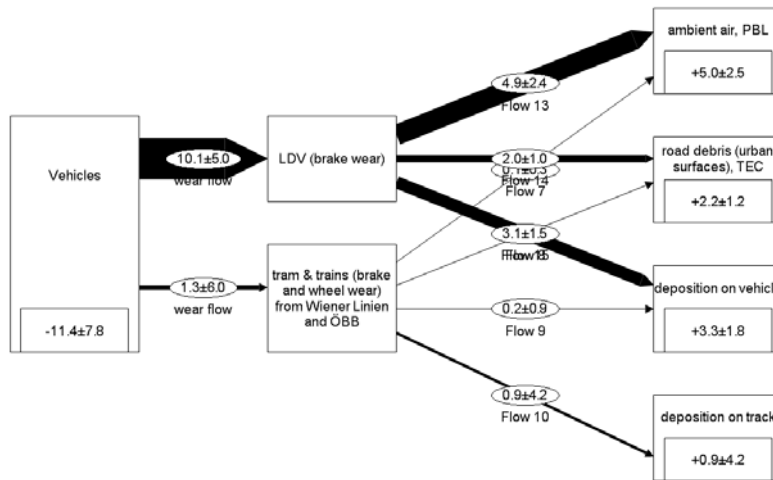


Table S27: Cu emissions from vehicles

Source	Flux Rate [t Cu/yr]	Uncertainty Level	Uncertainty Factor	Uncertainty
low duty vehicles	10.1	3	1.991	5.0
tram & trains	1.3	3	1.991	0.6

Note: t Cu/yr = tons copper per year

2.2.2.3.8 Catenary wear

The National Railway Company ÖBB operates 761 km electrified track with varying catenary diameters. The catenary material is made up from 99,5 – 99,9% Cu and 0,1-0,5% Ag or Cd. The catenary is replaced after 40 years; the remaining cross section is about 80%. The urban Public Transport Provider Wiener Linien operates 363 km electrified tram tracks (main tracks + station tracks). The cross section is assumed with 120mm². Lifetime data are equally to ÖBB.

Table S28: Catenary wear in Vienna

Catenary stock & wear	Length	Cross section	Cu stock	Catenary wear
	[km]	[mm ²]	[t]	[t/yr]
ÖBB, Section 1	272	65	158	0,1
ÖBB, Section 2	345	100	307	0,4
ÖBB, Section 3	144	120	154	0,8
ÖBB, Sum	761		619	3,1
Wiener Linien	363	120	389	1,9
Total	1.124		1.008	5,0

Note: km = kilometer; mm² = square millimeter; t = metric tons; t/yr = metric tons per year

Figure S8: Catenary wear and the whereabouts. Flows are given in metric tons per year.

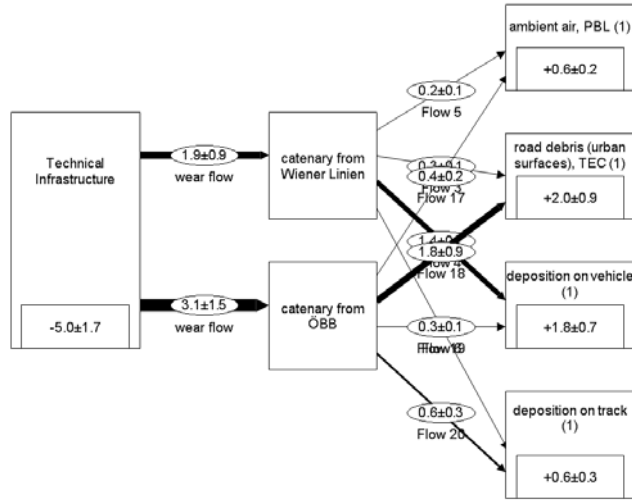


Table S29: Cu emissions from rolling stock

Source, catenary	Flux Rate [t Cu/yr]	Uncertainty Level	Uncertainty Factor	Uncertainty
Wiener Linien	1.9	3	1.991	0.9
ÖBB	3.1	3	1.991	1.5

Note: t Cu/yr = tons copper per year

2.2.2.3.9 Roof runoff

$$R = (0.37SO_2^{0.5} + 0.96 \text{ rain}10^{-0.62 \text{ pH}}) \left(\frac{\cos(\theta)}{\cos(45^\circ)} \right)$$

Formula 1. Formula for the calculation of R, the Cu run-off rate from roofs (Odnevall Wallinder et al. 2007).

Table S30: Climate and air comparison Stockholm / Vienna

Corrosion rate of city surfaces	Year	pH rain [-]	Surface Inclination [°]	SO ₂ [µg/m ³]	O ₃ [µg/m ³]	Precipitation [mm]	Corrosion rate [g/m ² /yr]
Stockholm	1995/1998	-	-	3,8 - 4,1	53 - 63	540	1,1 - 1,7
Vienna	2009	4,9-5,1	26	3	50	669	1,3-1,5

Notes: pH = pH value; µg/m³ = microgram per cubic meter; mm = millimeter; g/m²/yr = gram per square meter per year

Table S31: Land use as proxy for allocating roof runoff

Land use	Area [ha]	Proxy	Related to process
Residential Area (Wohngebiete)	10,267	0.70	PHH
Public Institution (Öffentliche Einrichtungen)	1,888	0.13	ECO
Commercial Area (Betriebsbaugebiete)	2,492	0.17	ECO
Total	14,647		

Notes: ha = hectare; PHH = Private Households; ECO = Industry, business, services and forestry;

2.2.2.3.10 Waste water flows

Table S32: Transfer Coefficients for Cu in WWTP

Material flow	Transfer Coefficients	Flux Rate
	[%]	[t Cu/yr]
Grill Material	6.0	1.0
Sewage Sludge	84.2	13.6
Effluents	9.8	1.6
Total	100	16.2

Notes: t Cu/yr = metric tons copper per year

Table S33: Cu in waste water coming from anthropogenic activities in PHH

Source	Flux Rate [mg/cap/yr]	Reference
Cu from food preparation	70	(Baccini et al. 1993)
Cu from washing dishes	25	(Baccini et al. 1993)
Cu in toilet paper (from recycling paper)	64	(Baccini et al. 1993)
Cu from product use	30	(I C Consultants Ltd 2001)
Cu from washing clothes	218	(Baccini et al. 1993)
Cu from cleaning living areas	436	(Baccini et al. 1993)
Total	843	

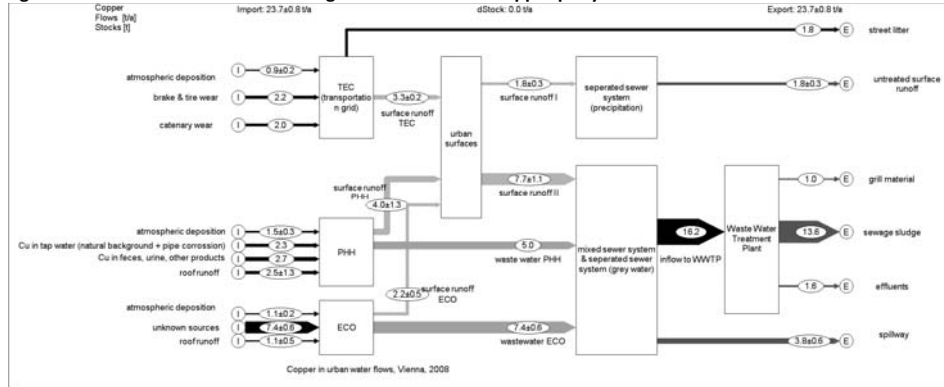
Note: mg/cap/yr = milligram per capita per year

Table S34: Cu in waste water coming from humans

Source	Flux Rate [mg/cap/yr]	Reference
Cu in feces	708	(I C Consultants Ltd 2001)
Cu in urin	5	(Lampert et al. 1997)
Cu in skin particles	47	(Lampert et al. 1997)
Cu in human off-flows	760	

Notes: mg/cap/yr = milligram per capita per year

Figure S9: Cu in waterflows. Flows are given in metric tons copper per year.



2.2.2.3.11 Solid Waste

2.2.2.3.11.1 Overview

Table S35: Compiled solid waste flow data

Composition of Solid Waste flow	PHH (HHW PHHCTS)		ECO (WAS ECOCTS)		total	
	Expected Value [t Cu/yr]	Deviation [t Cu/yr]	Expected Value [t Cu/yr]	Deviation [t Cu/yr]	Expected Value [t Cu/yr]	Deviation [t Cu/yr]
Municipal Solid Waste	1,123	79	-	-	1,123	79
Waste from industry and business	-	-	5,363	457	5,237	457
Bulky Waste	170	34	-	-	170	34
WEEE	400	52	27	3	427	52
Total	1,693	100	5,390	457	6,957	

Notes: t Cu/yr = metric tons copper per year; PHH = Private Households; ECO = Industry, business, services and forestry;

2.2.2.3.11.2 Municipal Solid Waste

Table S36: Cu flows in mixed waste

Cu waste input for incineration	Flux Rate [t]	Lower level [mg Cu/kg dm]	Upper level [mg Cu/kg dm]	Expected Value [mg/kg dm]	Cu Flux [t Cu/yr]	Deviation [t Cu/yr]
Municipal Solid Waste	525,982	1800	2200	1989.975	1,046.7	79.3
Organic waste collection	4,548			76	0.3	0.1
Plant waste collection	1,576			104.1	0.2	0.0
Bulky waste (Sperrmüll)	37,164	1800	2200	1989.975	74.0	5.6
Street litter	27,136			46	1.2	0.2
Paper & Plastic	1,682			65	0.1	0.0
total	598,088				1,122.5	79.5

Notes: t = metric tons; mg Cu/kg ds = milligram copper per kilogram dry matter; t Cu/yr = metric tons copper per year

Table S37: Cu concentrations in various mixed waste fractions

Cu content in waste	Cu content [mg/kg]	Reference
Municipal solid waste	2000	(Skutan and Rechberger 2007, p. 82; Skutan and Brunner 2006, p. 236)
Average Organic Waste Collection (Biotonne)	76	(Umweltbundesamt 2000)
Average Plant Waste Collection (Grünschnitt)	104	(Umweltbundesamt 2000)
Cu concentration in street litter	46	(Boller 2002)
Cu concentration in bulky waste	2.000	(Hausmann 2005)
Cu concentration in paper and plastic	65	(Arx 2006)

Note: mg/kg = milligram per kilogram

2.2.2.3.11.3 Waste from industry and business

Table S38: Waste from industry and business

key N°	Waste from industry and business	Amount of Waste in Vienna [t/yr]	Cu Content [%]	Cu in waste flow		Note
				Expected Value [t Cu/yr]	Deviation [t Cu/yr]	
35301	Waste from punching, blanking (is recycled)	8,831	12.5%	1,104	191	
35315	Non-ferrous metal scrap (is recycled)	2,993	55%	1,646	285	
51104	Electroplating sludge	142	15-12%	37	14	
51530	Cu chloride	1,506	47-65%	832	86	
35310	Cu scrap from production industry	1,839	90-99,9%	1,744	290	See sec. 2.2.2.3.3
Total				5,363	457	

Notes: t/yr = metric tons per year; t Cu/yr = metric tons copper per year; key N° = key number according to the Austrian waste management directive

2.2.2.3.11.4 Bulky waste

Table S39: Allocation of bulky waste flows

Flow	Collected bulky waste in Vienna, from PHH and ECO [t/yr]	Cu concentration Lower range [mg/kg]	Cu concentration Upper range [mg/kg]	Geometric mean [mg/kg]	Cu Flux [t Cu/yr]
Bulky Waste	69,422	1800.00	3300.00	2437.212	169

Notes: t/yr = metric tons per year; mg/kg = milligram per kilogram; t Cu/yr = metric tons copper per year;

2.2.2.3.12 WEEE

Table S40: WEEE from PHH

WEEE from PHH	Flow of goods, expected value [t]	Cu concentration Lower range [%]	Cu concentration Upper range [%]	Geometric mean [%]	Cu Flux rate [t Cu/yr]	Deviation [t Cu / yr]
Large WEEE	2,983	3.90	8.00	5.586	167	37
Fridges & Freezers	2,103	1.00	4.00	2.000	42	21
Monitors	2,387	3.90	7.80	5.515	132	28
Small WEEE	1,572	2.90	5.00	3.808	60	10
Sum Waste PHH - WEEE	9,044				400	52

Notes: t/yr = metric tons per year; t Cu/yr = metric tons copper per year;

Table S41: WEEE from ECO

WEEE from ECO	Flow of goods, expected value [t]	Cu concentration Lower range [%]	Cu concentration Upper range [%]	Geometric mean [%]	Cu Flux rate [t Cu/yr]	Deviation [t Cu / yr]
Small WEEE	198	3.90	8.00	5.586	11	2
Cu in WEEE Waste	139	1.00	4.00	2.000	3	1
Large WEEE	158	3.90	7.80	5.515	9	2
Fridges & Freezers	104	2.90	5.00	3.808	4	1
Sum Waste PHH - WEEE	599				27	3

Notes: t = metric tons; t Cu/yr = metric tons copper per year;

Table S42: Allocation of WEEE flows

Description	value	unit
Collected from private households	61,390,135	t/yr
Collected from economy	4,067,869	t/yr
Ratio = Proxy ECO/PHH	0,07	

Note: t/yr = metric tons per year; PHH = Private Households; ECO = Industry, business, services and forestry;

2.2.2.3.13 Exported products (PRO ANTEXA)

Table S43: Compiled data for exported flows

Flow Name	Flux rate [t Cu/yr]	Deviation [t Cu/yr]	Calculation procedure and references
Flow acronym			
VEH VEHECO	Exported vehicles 138	68	See flow "Exported Vehicles", sec. 2.2.2.3.4
-	Products from producing industry 950		After data are not available, we assumed that 50% of the difference from imported Cu for producing industries (see flow PRO EXAECO) and the waste from production (see flow WAS ECOCTS). $0.5 \cdot (4,600 - 2,700) = 950$
PRO ANTEXA	1,088		

Note: t Cu/yr = metric tons copper per year;

2.2.2.3.14 Exported waste including recyclables (REC CTSECO)

To estimate the recycling flow, we used the recycling potential of six generated waste flows, combined them with individual information regarding the whereabouts in recycling facilities.

Table S44: Cu waste including potential recyclables

Recyclables and waste	Potential recyclables in waste flow		Calculation procedure and references
	Expected Value [t Cu/yr]	Deviation [t Cu/yr]	
Demolition Material (construction waste, concrete demolition, Construction and demolition timber)	835 PHH (522) ECO (223) TEC (90)	135 (123) (53) (14)	See flow "Construction material", sec. 2.2.2.3.2. 835 t Cu/yr is estimated for demolition material. Even 75% (Ableidinger et al. 2007) of demolition material was sent out of the city, we assumed that 100% of Cu is sent out to recycling facilities out of Vienna.
Cu scrap	3,235 PHH (2,092) ECO (894) TEC (249)	532 (486) (207) (62)	See flow "Construction material", sec. 2.2.2.3.2. Cu scrap was reported in the Viennese waste statistics with an amount of 5,254 t/yr. An average Cu content of 90-99.5% results 4,982 t Cu/yr of which 3,235 are related to construction waste that goes to recycling facilities out of Vienna.
Cables	870 PHH (369) ECO (158) TEC (343)	127 (86) (37) (86)	See flow "Construction material" sec. 2.2.2.3.2. The Viennese waste statistics accounted 2,400 t cables [42]; measurements reveal a Cu content of 37% in cables within demolition material [47]. Combining the two factors results 870 t Cu/a.
Subtotal, construction waste	4,940	563	
Waste from industry and business	5,363	457	See flow "solid waste", sec. 2.2.2.3.11
Bulky Waste	170	34	See flow "solid waste", sec. 2.2.2.3.11
WEEE	427	52	See flow "solid waste", sec. 2.2.2.3.11 WEEE is collected and partly dismantled in Vienna but separation to the metal fractions occurs in various specialized treatment plants outside the city (Spitzbart 2012). The total amount of Cu in WEEE collected in Vienna is ~430 t/yr.
Recycled End-of-Life Vehicles	235	39	See flow "EOL vehicles", sec. 2.2.2.3.4
Total	11,135	729	

Note: t Cu/yr = metric tons copper per year

2.2.2.3.15 Harvest

Plant uptake of Cu from soil range are estimated to be 43 g/(ha*a) in farmland to 51 g/(ha*a) in grassland (Zethner et al. 2007; Kühnen and Goldbach 2004). If the plants are further harvested, this amount of Cu is actively removed from the soil. With an agricultural area of ~ 6.000 ha, the Cu flow in harvest is estimated to be ~ 0,3 t/yr. To test this values probability, detailed harvest information (Fitzthum 2009) combined with Cu take-up values for Crops, Wine and Fruit production which was published in an Austrian study (Berger et al. 2011). The Sum of Cu take-up in harvest is therefore estimated to be 0,3 t/yr (uncertainty factor: 1,34).

Table S45: Agricultural production

Cu in harvest products	Harvest 2008 [ha]	Cu take-up [kg Cu/ha/yr]	Cu [kg]
Agriculture & Horticulture	5,600	0,045	252
Wine production	600	0.01	6
Fruit production	70	0.03	2
Sum	6,270		~ 260

Notes: ha = hectare; kg Cu/ha/yr = kilogram copper per hectare per year; kg = kilogram;

2.2.2.3.16 Pesticides

Table S46: Pesticide inputs on soil

Pesticide Input	Area	Pesticide rates	Cu Flux rate
	[ha]	[kg Cu/ha/yr]	[t/yr]
Viticulture organic	109	3.000	0.327
Fruit Production organic	12	3.000	0.036
Agriculture & Horticulture organic	1,009	1.400	1.413
Viticulture	497	1.000	0.497
Fruit Production	55	1.500	0.082
Agriculture & Horticulture	4,599	0.000	0.000
Total	6,281		2.356

Notes: ha = hectare; kg Cu/ha/yr = kilogram copper per hectare per year; t = metric tons;

2.2.2.3.17 Fertilizer

Table S47: Fertilizer inputs on soil

Fertilizer Input	Area	Fertilizer rate	Cu flux rate
	[ha]	[kg Cu/(ha/a)]	[t/yr]
Viticulture organic	109	0.200	0.022
Fruit Production organic	12	0.200	0.002
Agriculture & Horticulture organic	1,009	0.550	0.555
Viticulture	497	0.030	0.015
Fruit Production	55	0.030	0.002
Agriculture & Horticulture	4,599	0.350	1.609
Total	6,281		2.205

Notes: ha = hectare; kg Cu/ha/yr = kilogram copper per hectare per year; t/yr = metric tons per year;

2.2.2.3.18 Cu stock in technical infrastructure (TECstock)

Table S48: Cu stocks in electricity grid

Electricity grid	Length of network [km]	Cu content in [t/km]		Amount of Cu [t]	
		min	max	min	max
		Highest voltage	54	10.72	583
High voltage	375	10.72	4,015	4,015	
Medium voltage	6,844	5.09	34,837	34,837	
Low voltage	11,255	4.07	45,809	45,809	
Overhead power lines	3,747	0.75	1.34	2,807	5,012
Sum	22,276			88,052	90,257
				89,154	

Notes: km = kilometer; t/km = metric tons per kilometer; min = minimum value; max = maximum value

Table S49: Cu stocks in telecommunication grid

Unit	Number of Units [#]	Cu in telecommunication [kg/#]	Cu stock [t]
Vienna businesses <5 employees	64,631	12	776
Vienna businesses >100 employees	1,142	620	708
Vienna businesses 5-99 employees	21,918	294	6,444
Households in Vienna	77,1706	7	5,402
Total			13,329

Notes: # = number of items; kg/# = kilogram per item; t = metric tons

2.2.2.3.19 Cu stock in Vehicles (VEHstock)

The total stock of Cu in transportation utilities (e.g. cars, busses, trucks, subway, trams etc.) is calculated to be ~24.000 t Cu (uncertainty: 17%) or ~ 15 kg/cap. This value fits well with the above described literature. The calculation is based on data on the number of registered vehicles with the city of Vienna and data on public transportation vehicles such as the number of subways and trams (Wiener Linien 2009; Statistik Austria 2008) uncertainty level 1,0 and corresponding Cu contents from literature (Struckl 2007; Bertram et al. 2002; Hooch 2008; European Copper Institute (ECI) 2011). Used literature values can be found at flow calculations VEH ECOVEH. Uncertainty factors of Cu content were calculated by using the range found in literature.

Table S50: Cu stock in vehicles

Item	Number of vehicles [#]	Cu content			Cu Flux Rate	
		Min [kg/#]	Max [kg/#]	Geomittel [kg/#]	Expected Value [t Cu/yr]	Deviation [t Cu/yr]
Cars	657192	20.00	29.00	24.0831892	15,827	1,616
Trucks	61488	60.00	80.00	69.2820323	4,260	330
Motor Bikes	73938	4.00	6.00	4.89897949	362	41
Others (mainly heavy machinery, busses incl. Wr. Linien etc.)	56857	25.00	60.00	38.7298335	2,202	605
Trams	832	700.00	800.00	748.331477	623	21
Subway Cars (Triebwagen)	477	1272.78	1500.00	1381.72655	659	28
					23,933	1,757

Notes: # = number of items, kg/# = kilogram per number of item; t Cu/yr = metric tons copper per year

2.2.2.3.20 Cu stock in Landfills (LDFstock)

In 2005, there were six operated landfills located in Vienna (Ableidinger et al. 2007). Another 36 (excluding remediation sites) old unofficial dumps and landfills, filled with household or construction waste can be found in official datasets (Umweltbundesamt Österreich 2012). The total amount of Cu stocks in Viennese landfills is estimated to be ~ 144.000 t of Cu (uncertainty factor 1,84). This value fits well with the first estimation of 100.000 t Cu in landfills (without unofficial waste dumps) quite well.

Table S51: Cu stock in landfills

	Amount of Cu		Uncertainty factor	
	[t] Min	[t] Max		
Excavated Soil (1)+(2)+(3)+(4)	6,845,900	200	400	1,49
Construction Waste (5)	11,300,000	900	8.000	2,90
Treated municipal solid waste	-	27.000	71.000	1,62
Slag concrete as construction material (6)+(7)+(8)	27,240,000	30.000	35.000	3,24
Waste dumps (unofficial)	35,690,400	24.000	93.000	2,02
Total sum of Cu stock in landfills		82.000	210.000	1,84
Mean Value			144.000	1,84

Notes: t = metric tons

Table S52: Landfills in Vienna

Landfill	Landfill Type	Approved volume	Available capacity	Deposited waste volume	Density*)	Mass	N°
		[m3]	[m3]	[m3]	[t/m3]	[t]	
Deponie Max	Excavated soil	97.000	0	97.000	1,7	164.900	(1)
Nassbaggerung Transportbeton	Excavated soil	2.900.000	300.000	2.600.000	1,7	4.420.000	(2)
Nassbaggerung Readymix	Excavated soil	1.600.000	350.000	1.250.000	1,7	2.125.000	(3)
Deponie Rendezvousberg (Kleedorfer)	Excavated soil	250.000	170.000	80.000	1,7	136.000	(4)
Deponie Langes Feld (3 Kompartimente)	Construction w.	7.400.000	1.750.000	5.650.000	2	11.300.000	(5)
	Mass waste	2.000.000	1.650.000	350.000	2,4	840.000	(6)
	Municipal solid w.	1.600.000	1.500.000	100.000	2,8	280.000	(7)
Deponie Rautenweg	Mass waste	14.000.000	3.000.000	11.000.000	2,4	26.400.000	(8)
total		29.847.000	8.720.000	21.127.000		45.665.900	

Note: * = own estimation; m3 = cubic meter; t/m3 = metric tons per cubic meter; t = metric tons

Deposited wastes include excavated soil (usually from construction sites), construction waste and treated municipal waste according to Austrian landfill regulations and furthermore some municipal solid waste is stored temporarily. Using Cu concentrations from earlier calculations for excavated soil (28–61 mg/kg), construction waste (80–670 mg/kg) and municipal solid waste (1000–2600 mg/kg) the total Cu stock was calculated. Especially for the landfills “Langes Feld” and “Rautenweg” calculations are difficult. Due to their age, it is difficult to determine a Cu content of the former input. For example, “Langes Feld” was originally built to receive construction and demolition waste from World War II. Additionally to the type of landfill, slags which are used as building material in Viennese landfills and their input since 1964 (when the first MSW incineration plant in Vienna was established) were estimated. Here, the input of 2008, calculated with ~ 1.100 t/yr as well as capacities of incineration plants since 1964 were used as a base value.

Additionally to official landfills, waste dumps or suspected waste dumps with a total volume of ~ 30 Mio m³ were included into calculations (Umweltbundesamt Österreich 2012). With an average density of 1,0–1,2 t/m³ (Fellner 2012) and contents (partly suspected, partly tested) as a mixture of untreated municipal solid waste and construction waste (with unknown percentages), Cu stocks are estimated to be between 24.000 and 90.000 t.

2.2.2.3.21 Cu stock in urban soil (UPVstock)

Figure S10: Substance concentration in parks with playgrounds, given in milligram per kilogram (mg/kg). The box is defined by the 10% and 90% percentile and the median. The red and blue bars indicate orientation values.

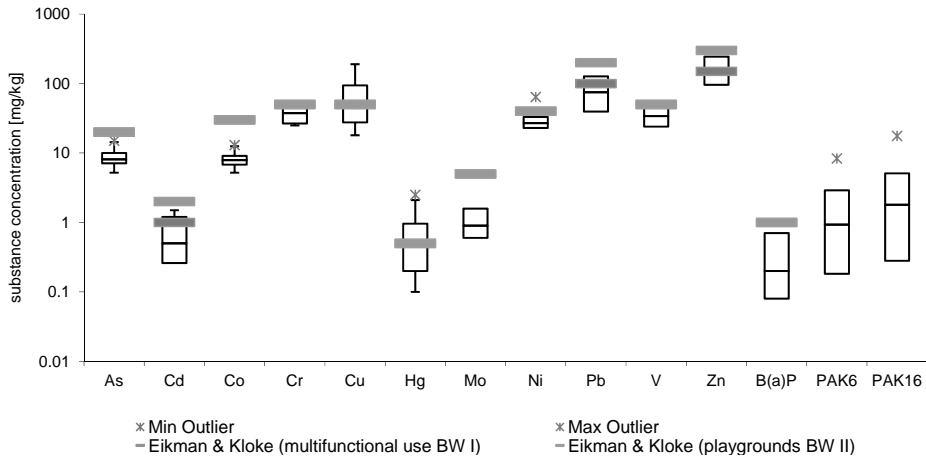


Table S53: Median concentrations in parks with playgrounds given in milligram per kilogram (mg/kg)

	As	Cd	Co	Cr	Cu	Hg	Mo	Ni	Pb	V	Zn	B(a)P	PAK6	PAK16
median	8,1	0,5	7,9	37,5	46,0	0,5	0,9	27,0	75,0	34,0	158,0	0,2	0,9	1,8
Eikman & Kloke playgrounds, BW2	20	2	30	50	50	0,5	5	40	200	50	300	1	-	-

2.2.2.4 Taipei – Background data

2.2.2.4.1 Atmospheric Deposition (ADE PBLUHY, ADE PBLUPV, ADE PBLECO, ADE PBLPHH, ADE PBLTEC)

In Huang (2004), metals in atmosphere within Taipei City were sampled and analyzed in the form of particulate matters and precipitation. Both wet and dry depositions of Cu were measured, as the rate of 3.4 mg/m²/yr and 3.8 mg/m²/yr respectively. Total Cu dry deposition rate was estimated by multiplying total area of Taipei, which is 0.92 t/yr. Also, atmospheric deposition of Cu through precipitation was calculated as 1.03 t/yr.

2.2.2.4.2 Products (PRO EXAECO, PRO ECOPHH)

Table S44: Ratio of Taipei and Taiwan for Imported Goods Estimation

Cu Application Category	Consumer Goods Total Retail Sales (NTD)	Industrial Equipment Areas of Factories (m ²)
Taipei	1,282,965,935,000	24,052,521
Taiwan	3,081,553,000,000	164,959,000
Ratio	0.42	0.15
Ratio in Taiwan	18%	21%
Ratio in Taipei	38.8%	15.9%

Notes: NTD = New Taiwan Dollar; m2 = square meter

Figure S15: Mechanism of Recycling Fund in Taiwan (Recycling Fund Management Board Taiwan 2010b)

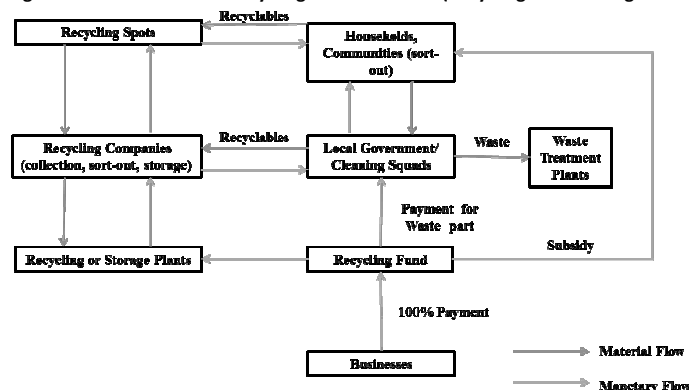


Table S45: Estimation of Household Appliances Number Consumed in Taiwan (2009)

WEEE Item	Recycling Fee (NTD/#) (EPA 2004)	Amount of Audited Recyclables (unit) (Recycling Fund Management Board Taiwan 2010a)	Estimated Recycling income (NTD)	Ratio in Recycling Fund	Fund Distribution (NTD)	Estimated Amount of Products (#)
TV		504,978	123,467,121	28 %	257,518,274	1,053,244
CRT TV						
Over 25"	371					
Under 25"	247					
LCD, PDP						
TV						
Over 25"	233					

Under 25"	127					
Air Conditioner	248	326,283	80,918,184	18%	168,772,957	680,536
Washing Machine	317	292,000	92,564,000	21%	193,062,908	609,031
Refrigerator		293,869	148,403,845	33%	309,529,384	612,929
Over 250 L	606					
Under 250 L	404					

Note: WEEE = waste electrical and electronic equipment; NTD = New Taiwan Dollar; # = number of items; NTD/# = New Taiwan Dollar per item

Table S46: Estimation of Cu Mass in Household Appliances in Taiwan

Item	Conversion Rate (kg/#)	Conversion Weight (t)	Cu Content (Truttmann et al. 2005) (%)
TV	25.0	26,331.11	
CRT TV			3.9
LCD, PDP TV			0.8~1.2
Air Conditioner	60.0	40,832.17	17.8
Washing Machine	40.0	24,361.25	3.1
Refrigerator	50.0	30,646.47	3.4
Sum			

Note: kg/# = kilogram per item; # = number of items; t = metric tons;

Table S47: Estimation of Computer Appliances Number Consumed in Taiwan (2009)

WCA	Recycling fund (2010): \$ 775,608,054 NTD							
Notebook	Sales volume in Taiwan (2008): 1,042,772 units							
	Recycling Fee (NTD/unit): 39							
	Estimated recycling income: \$ 40,668,108 NTD							
	Recycling Fund without Notebook (NTD): 734,939,946							
Item	Recycling (NTD/#) 2004)	Fee (EPA)	Amount Audited (Recycling Fund Management Board Taiwan 2010a)	of (unit)	Ratio in WCA w/o Notebook	Estimated Recycling Amount (NTD)	Estimated income	Estimated Amount of Products (#)
Monitor			822,353		37%	269,264,506		1,658,713
CRT Monitor	127							
LCD Monitor over 25"	233							
under 25"	127							
Desktop PC	8.2, 49.2		839,120		37%	274,754,554		9,573,329
Printer			583,086		26%	190,920,886		1,552,202
Inkjet Printer	81							
Laser Printer	137							
Dot-matrix Printer	151							
Sum			2,244,559					

Notes: WCA = waste computer appliances; kg/# = kilogram per item; # = number of items; t = metric tons;

Table S48: Estimation of Cu in Computer Appliances in Taiwan

Item	Conversion (kg/#)	Rate	Conversion Weight (t)	Cu Content (%)
Notebook	4.0		4,171.09	1.00% (Oguchi et al. 2011)
Monitor	12.0		19,904.56	7.83% (Truttmann et al. 2005)
Desktop PC	12.0		114,879.95	0.90% ^a
Printer	8.5		13,193.72	3.20% ^a
Sum				

Notes: kg/# = kilogram per item; t = metric tons;

2.2.2.4.3 Construction Material (CON EXAECO, CON ECOTEC, CONECOPHH, CWA ECOCTS, CWA TECCTS, CWA PHHCTS)

Table S49: Estimation of Cu in Construction Waste

Taipei	Value	Unit
Total demolished floor area	25,000	m ²
Waste from C&D waste (Huang 1998)	1~1.28	t
Cu content in C&D waste (Huang 1998)	670~2,587	mg/kg

Notes: m² = square meter; t = metric tons; mg/kg = milligram per kilogram

Table S50: Results of Cu in Construction Waste from PHH, CTS, and ECO

Land Use in Taipei	Area (ha)	Proportion
Urbanized Area	12,993.11	100%
PHH		41%
Residential District	3,793.06	
Commercial District	866.36	
Administration District	80.97	
Cultural District	73	
Special District	190	
Others	260.45	
ECO		3%
Industrial District	419.49	
TEC		56%
Public Facilities	7,309.78	

Notes: ha = hectare

2.2.2.4.4 Vehicles (VEH ECOVEH, WAS VEHCTS, VEH UID, VEH VEHECO)

Table S51: Estimation of Sales Volume of Vehicles in Taipei 2009

	Newly Registered Number	Sales Volume	Average weighted weights per unit (kg) (Liu 2009)	Mass of Sales Volume (t/yr)
Taiwan				
Cars	6,769,845	294,423		
Scooters/Motorcycles	14,604,330	478,268		
Taipei				
Cars	721,326	31,371	1,034	32,437,332.57
Scooters/Motorcycles	1,092,788	35,787	90.85	3,251,251.27
Sum				35,688,583.84

Notes: kg = kilogram; t/yr = metric tons per year

Table S52: Estimation of Recycled Vehicles in Taipei

	Newly Registered Number [#]	Average weighted weights per unit (kg) (Liu 2009)	Recycled Mass (t)
Taiwan			
Cars	6,769,845	1,034	130,791
Scooters/Motorcycles	14,604,330	90.85	21,994
Taipei			
Cars	721,326	1,034	195
Scooters/Motorcycles	1,092,788	90.85	23
Sum			

Notes: t = metric tons; # = number of items

Table S53: Estimation of Exported Vehicles

	Newly Registered Number [#]	Change of Registration (2009-2008)
Taiwan		
Cars	6,769,845	42,929
Scooters/Motorcycles	14,604,330	238,888
Taipei		
Cars	721,326	4,574
Scooters/Motorcycles	1,092,788	17,875
Sum		

Note: # = Number of items

2.2.2.4.5 Solid waste (HWW PHHCTS, WAS ECOCTS)

Table S54: Result of Cu Mass in Household Waste

Source of Waste	Cu Content	Amount (t/yr)
By Cleaning Squads		368,253.67
By People Themselves	184.92~577.13 mg/kg	13,974.27
Recyclables	(%)	
TV	0.8~3.9	5,302.27
Air Conditioner	17.8	8,222.33
Washing Machine	3.1	4,906
Refrigerator	3.4	6,171.25
Laptop	1	75.29
Monitor	7.83	4,144.66
Motherboard	0.9	4,229.16
Printer	3.2	2,081.62
Sum		

Note: t/yr = metric tons per year

2.2.2.4.6 Incineration: Mixed waste, exported residues for underground storage, residues Disposable waste (WAS CTSINC, TTR INCEXA, RES INCLDF)

Table S55: Residues of Incinerators

	Amount (t)	Cu Content (mg/kg) (Chen 2008)
Fly Ash	20,000	1028~1220
Bottom Ash	110,000	889~3136

Note: t = metric ton; mg/kg = milligram per kilogram

2.2.2.4.7 Composting: Compostable waste, residues, compost as fertilizer (WAS CTSCOM, RES COMCTS, COM COMUPV)

Table S56: Estimation of Compost Generated in Taipei City

Data source	Uncooked food waste (t/yr)	Compost (t/yr)	Cu content (mg/kg)
Muzha Incinerator	16,625.3	221.15	0.69
Waste in Taipei City	59,371.23 (Department of Environmental Protection of Taipei City Government 2009)	789.76	0.69

Note: t/yr = metric tons per year; mg/kg = milligram per kilogram

2.2.2.4.8 Stocks in ECO, PHH

Table S57: Estimation of the In-Use Household Appliances

Item	Average Number Per Hundred Households in Taipei (#)	Estimated Amount of In-use Stock (#)	Conversion Rate (kg/#)	Conversion Weight (t)	Cu Content (%) (Oguchi et al. 2011)
TV	155.87	1,500,764.68	25.0	37,519.12	0.8~3.9
Air Conditioner	224.39	2,160,496.48	60.0	129,629.79	17.8
Washing Machine	99.26	955,706.05	40.0	38,228.24	3.1
Laptop	123.03 (IDC Taiwan 2008)	1,184,570.98	4.0	4,738.28	1.0
Desktop	123.03	1,184,570.98	12.0	14,214.85	0.9

Note: # = number of items; kg/# = kilogram per item; t = metric ton;

Table S58: Information of the Tainan Case (construction completed in 1997)

Structure	Floors	Usage
Aboveground	13	1F 2F~13F
Underground	3	B1~B3 Parking Lot

Table S59: Estimation of Wire and Cable Usage in Buildings

The Tainan Case	Amount	Unit
Wire and Cable Used	17,763.84	kg
Total Floor Area	1.79	kg/m ²
Total Floor Area in Taipei	171,185,000	m ²
Estimated Wire and Cable Used	306,372,155	kg
Cu Content	40~60 (Ruhrberg 2006; Spatari et al. 2002)	%

Note: kg = kilogram; kg/m² = kilogram per square meter;

2.2.2.4.9 Stocks in VEH

Table S60: Estimation of Cu Stock in Transportation

Vehicles	Number of Registration/In Operation (Environmental Protection Administration - Executive Yuan 2010)	Average Weight Per Unit (kg) (Liu 2009)	Weighted	Cu Content
Cars	721,326	1,034		1.4%
Scooters	1,092,788	90.85		1.4 %
Buses	2,628	--		60~80 kg/#

2.3 Stock and flow results

Table S54 shows the model inputs for each city in column (3), (4), (7), (8) and the balanced results in column (5), (6), (9), (10).

Table S54: Flow list.

Flow	Flow name	TAIPEI				VIENNA			
		Mass flow [t/yr]	± Mass flow [t/yr]	Mass flow (calculated) [t/yr]	± Mass flow (calculated) [t/yr]	Mass flow [t/yr]	± Mass flow [t/yr]	Mass flow (calculated) [t/yr]	± Mass flow (calculated) [t/yr]
(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)
ADE PBLECO	Atmospheric Deposition ECO	0.02	0.00	0.02	0.00	1.10	0.19	1.09	0.19
ADE PBLPHH	Atmospheric Deposition PHH	0.19	0.01	0.19	0.01	1.50	0.26	1.49	0.26
ADE PBLTEC	Atmospheric Deposition TEC	0.28	0.02	0.28	0.02	0.90	0.15	0.90	0.15
ADE PBLUHY	Atmospheric Deposition UHY	0.07	0.01	0.07	0.00	0.30	0.10	0.30	0.10
ADE PBLUPV	Atmospheric deposition PV	0.29	0.02	0.29	0.02	2.80	0.50	2.76	0.49
COM ANTUPV	Compost	0.04	0.01	0.04	0.00	1.40	0.30	1.40	0.21
CON ECOPHH	Construction Material	8,164.59	1,420.10	8,164.59	1,420.10	6,567.00	838.00	6,567.00	838.00
CON ECOTEC	Construction material	4,224.21	734.73	4,224.21	734.73	2,448.00	376.00	2,448.00	376.00
CON EXAECO	Imported construction material	13,073.32	2,273.89	13,073.32	2,273.89	11,817.00	1,390.00	11,817.00	1,390.00
CWA ECOCTS	Construction waste ECO	2.79	1.36	2.82	1.36	1,275.00	217.00	1,266.04	212.19
CWA PHHCTS	Construction waste PHH	150.86	73.44	223.07	69.99	2,983.00	509.00	2,933.70	443.42
CWA TECCTS	Construction waste TEC	48.66	23.64	56.15	23.53	862.00	107.00	859.82	106.43
EFF WWSHYD	Effluents	2.85	0.22	2.73	0.22	1.60	0.10	1.60	0.10
FER ECOUPV	Fertilizer	0.00		0.00		2.20	0.30	2.20	0.30
HAR UPVECO	Harvest	0.00		0.00		0.30	0.10	0.30	0.10
HHW PHHCTS	Household waste	1,672.34	54.77	1,712.52	53.36	1,693.00	100.00	1,691.10	99.53
PAE TECPBL	Particulate emissions	0.00		0.00		0.60	0.20	0.61	0.20
PAE VEHPBL	Particulate emissions	6.10	2.67	0.84	0.03	5.00	2.50	5.94	0.63
PAE VEHTEC	Particulate emissions	0.00		0.00		2.20	1.20	2.20	1.20
PES UANUPV	Pesticides	0.00		0.00		2.40	0.30	2.40	0.30
PRO ECOEXA	Exported Products	7,944.63	1,359.73	7,944.63	1,359.73	2,574.00	746.00	2,574.00	746.00
PRO ECOPHH	Consumer goods & food	5,286.44	1,610.12	5,286.44	1,610.12	1,484.00	205.00	1,484.00	205.00
PRO EXAECO	Imported products	15,773.73	2,743.58	15,773.73	2,743.58	8,827.00	728.00	8,827.00	728.00
RES INCLDF	Residues	209.08	81.53	164.72	55.91	986.00	170.00	1,097.19	72.22
SPW WWSHYD	Combined sewer overflow	0.00		0.00		3.80	0.80	3.86	0.78
SRO ECOWWS	Surface runoff	0.02	0.01	0.02	0.01	2.20	0.50	2.18	0.49
SRO PHHWWS	Surface runoff PHH	0.51	0.25	0.66	0.25	4.00	1.30	3.84	1.20
SRO TECWWS	Surface runoff	0.35	0.18	0.43	0.18	3.30	1.50	3.09	1.35
SRO WMIHYD	Surface Runoff	0.90	0.40	0.51	0.40	1.80	0.05	1.80	0.05
SSL WWSCTS	sewage sludge	58.28	4.51	9.08	0.92	13.60	1.10	13.71	1.04
TTR INCEXA	Exported residues for underground	0.00		0.00		16.40	1.30	16.41	1.30
VEH ECOVEH	vehicles	499.64	250.02	499.64	250.02	2,237.00	185.00	2,237.00	185.00
VEH UID	Unidentified cars	0.00		0.00		498.00	330.00	498.00	330.00
VEH VEHECO	EOL vehicles for export	88.95	44.42	88.95	44.42	138.00	68.00	138.00	68.00

WAS CTSCOM	Compostable waste	0.04	0.01	0.04	0.00	1.40	0.30	1.40	0.21
WAS CTSEXA	Exported waste incl. recyclables	2,738.32	184.80	2,281.03	119.63	11,135.00	729.00	11,236.14	518.30
WAS CTSINC	waste	209.08	81.26	164.72	55.91	1,137.00	80.00	1,113.59	72.21
WAS ECOCTS	Solid waste ECO	58.42	22.64	65.28	22.54	5,390.00	457.00	5,350.26	410.20
WAS TECCTS	street litter	0.00		0.00		1.80	0.05	1.80	0.05
WAS VEHCTS	Waste vehicles	218.14	108.88	376.88	97.28	235.00	39.00	234.71	38.97
WWA ECOWWS	Wastewater ECO	0.00		0.00		7.40	1.90	7.07	1.59
WWA PHHWWS	Wastewater PHH	9.80	0.76	11.21	0.75	5.00	1.50	4.79	1.35

Note: t/yr = metric tons per year

Table S55: Stock list

Process	Process name	Taipei		Vienna	
		Mass [t]	± Mass [t]	Mass [t]	± Mass [t]
EXA	External Anthroposphere (input related)				
EXAO	External Anthroposphere (output related)				
LDF	Landfill			144,000	61,000
PHH	Private Households	27,795	11,579	131,000	35,000
DSR	Dansuie/Danube River				
TEC	Transport / Energy / Communication Infrastructure	32,027	8,325	103,000	11,000
UST	Underground Storage				
UHY	Urban Hydrosphere				
ECO	Urban Industry, Business, Services & Forestry /	215	27	40,000	14,000
UPV	Urban Pedosphere & Vegetation	831	145	4,000	1,000
VEH	Vehicles	12,014	5,911	24,000	2,000

Notes: t = metric tons

Figure S11: Cu flow system of Vienna on 1st level: Snapshot for the year 2008. The stocks are given in gram per capita (g/cap) and the flows in gram per capita per year (g/cap/yr). The stock values as well as the alteration of stocks are shown in the boxes; the flows are represented as Sankey arrows proportional to the flux rate.

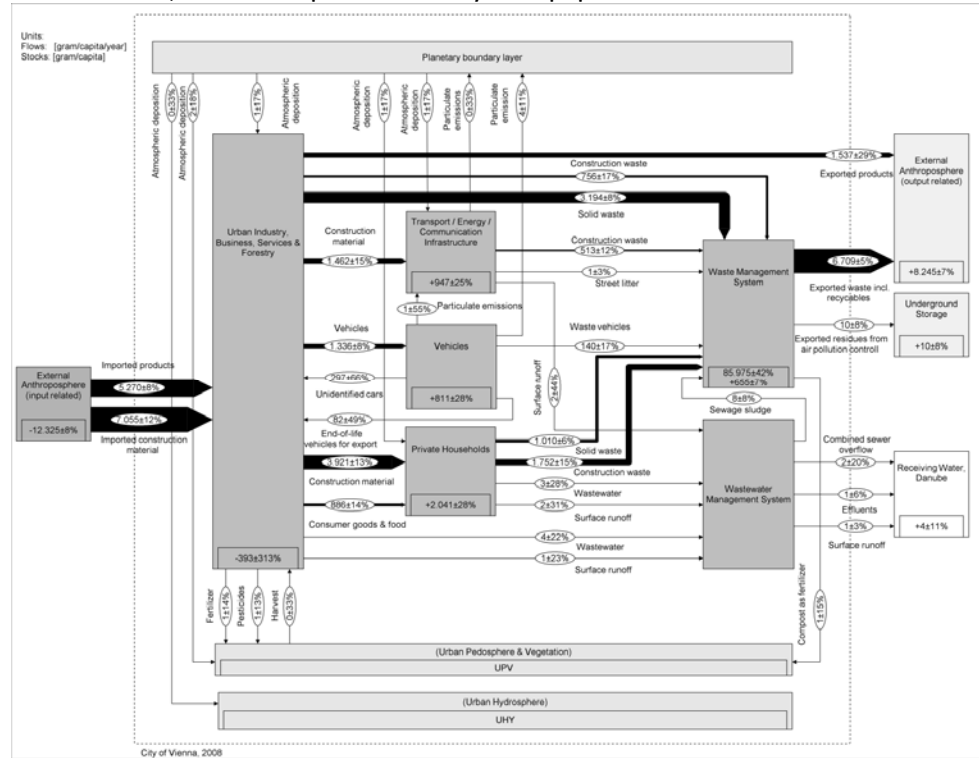


Figure S12: Cu flow system of Vienna on 2nd level: Snapshot for the year 2008. The stocks are given in gram per capita (g/cap) and the flows in gram per capita per year (g/cap/yr). The stock values as well as the alteration of stocks are shown in the boxes; the flows are represented as Sankey arrows proportional to the flux rate.

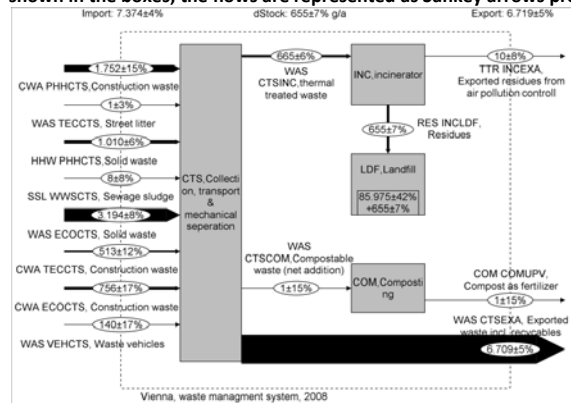


Figure S13: Cu flow system of Taipei on 1st level: Snapshot for the year 2009. The stocks are given in gram per capita (g/cap) and the flows in gram per capita per year (g/cap/yr). The stock values as well as the alteration of stocks are shown in the boxes; the flows are represented as Sankey arrows proportional to the flux rate.

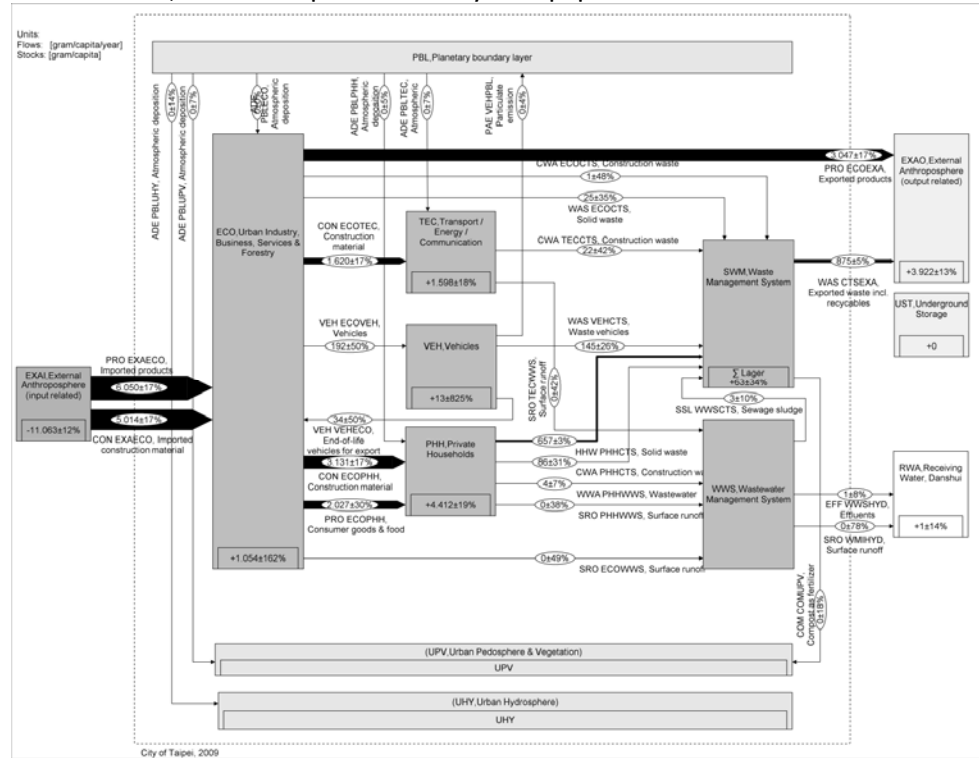
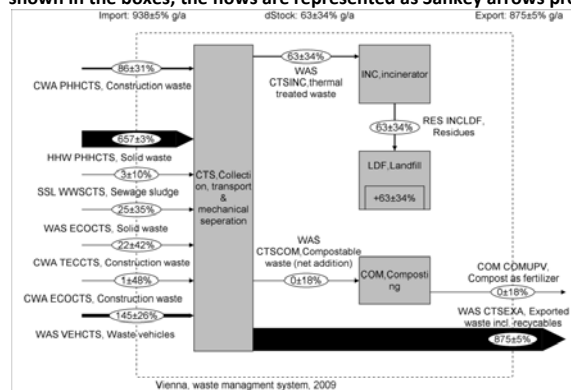


Figure S14: Cu flow system of Taipei on 2nd level: Snapshot for the year 2009. The stocks are given in gram per capita (g/cap) and the flows in gram per capita per year (g/cap/yr). The stock values as well as the alteration of stocks are shown in the boxes; the flows are represented as Sankey arrows proportional to the flux rate.



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Article III

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Sinks as limited resources? - A new indicator for evaluating anthropogenic material flows.

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Abstract

Besides recyclables, the use of materials inevitably yields non-recyclable materials such as emissions and wastes for disposal. These flows must be directed to sinks in a way that no adverse effects arise for humans and the environment. The objective of this paper is to present a new indicator for the assessment of substance flows to sinks on a regional scale. The indicator quantifies the environmentally acceptable mass share of a substance in actual waste and emission flows, ranging from 0% as worst case to 100% as best case. This paper consists of three parts: First, the indicator is defined. Second, a methodology to determine the indicator score is presented, including (i) substance flows analysis, and (ii) a distant-to-target approach based on an adaptation of the Ecological Scarcity Method 2006. Third, the metric developed is applied in three case studies including copper (Cu) and lead (Pb) in the city of Vienna, and Perfluorooctane Sulfonate (PFOS) in Switzerland. The following results were obtained: In Vienna, 99% of Cu flows to geogenic and anthropogenic sinks are acceptable when evaluated by the distant-to-target approach. However, the 0.7% of Cu entering urban soils and the 0.3% entering receiving waters are beyond the acceptable level. In the case of Pb, 92% of all flows into sinks prove to be acceptable, and 8% are disposed of in local landfills with limited capacity. For PFOS, 96% of all flows into sinks are acceptable. 4% cannot be evaluated due to a lack of normative criteria, despite posing a risk for human health and the environment. The examples demonstrate the need (i) for appropriate data of good quality to calculate the sink indicator, and (ii) for standards, needed for the assessment of substance flows to urban soils and receiving waters. This study corroborates that the new indicator is well suited as a base for decisions regarding the control of hazardous substances in waste and environmental management.

Keywords: Sink, substance flow analysis, resource management, environmental management, PFOS, copper, lead.

Highlights

- Sinks are required to accommodate wastes and emissions.
- Anthropogenic sinks are provided where capacities of geogenic sinks are lacking.
- A new indicator discriminates between acceptable and unacceptable flows to sinks.
- The indicator is applied in three case studies on urban and national scales.
- Implications for resource, waste and environmental management are discussed.

1 Introduction

“I do not worry about peak oil whatsoever. We have plenty of oil, gas, and coal to last for hundreds of years, and we are not running out. But we are running out of room in the atmosphere to store our exhaust.” Schnoor (2013) highlights the sink “atmosphere” as constraint for anthropogenic carbon before the sources run dry. The overriding question is if we are running out of “room in sinks” for other substances, too. Annually, millions of tons of materials are exploited from the earth crust or are produced synthetically, and processed into consumer and investment goods. After years or decades in use, the materials are discarded and meet their fate in terms of recycling or disposal in sinks. Therefore, geogenic sinks are available to a certain extent and anthropogenic sinks have to be provided where geogenic sinks are lacking. Geogenic sinks are part of biogeochemical cycles (e.g. Abeles et al., 1971; Berg and Dise, 2004; Feichter, 2008; Fong and Zedler, 2000; ICSU, 1989; Molina and Rowland, 1974; Paterson et al., 1996; Yanai et al., 2013). Anthropogenic sinks are manmade and refer to technologies such as incinerators, sanitary landfills, and sewage treatment plants (e.g. Brunner, 1999; Brunner and Tjell, 2012; ISWA, 2013; Morf and Brunner, 2005; Vogg, 2004; Zeschmar-Lahl, 2004). In general, materials must be directed to sinks in a way that no adverse effects arise for humans and the environment (Tarr, 1996).

To avoid unacceptable overloads, several authors have suggested metrics that focus on the relation between anthropogenic off-flows and potential impacts (table 1). In common, these metrics (i) operate on a substance specific level, (ii) focus on human activities within regions, and (iii) work with a set of indicators. To calculate the indicator, a combination of descriptive and normative assessment methods is needed:

- Descriptive methods analyze the fate and behavior of substances through the anthroposphere and the environment. For this purpose, the tools substance flow analysis (SFA) and environmental fate modeling (EFM) have been developed (e.g. Brunner and Rechberger, 2004; Mackay et al., 2006; OECD, 2007; UNEP, 2002). To calculate pressure indicators, researchers devoted much effort to quantify substance flows from human activities into

geogenic and anthropogenic sinks (e.g. Buser and Morf, 2009; Chen and Graedel, 2012; Henseler et al., 1992; Ott and Rechberger, 2012).

- Normative methods focus on the cause-effect chain of substances. Depending on the available knowledge, they either refer to “known damage due to known causalities”, or “known damages due to unknown causalities”, or “unknown damage due to unknown causalities”(adopted from Hofstetter, 1998). If damage and causalities are known, impact indicators can be provided. Therefore the tools risk assessment (RA) and life cycle impact assessment (LCIA) have been developed. LCIA focuses on the assessment of emissions along the whole life cycle chain of products and services rather than on emissions from entire human activities within regions (Loiseau et al., 2012). In general, LCIA methods rely on the scientific treatment of cause-effect relations from the intervention level towards the impact level. The LCIA method “Ecological Scarcity 2006” is an exception, because it considers the definition of critical flows into sinks based on legal limits and political agreements (Jungbluth et al., 2012). However, for the majority of substances placed on the market, the damages and causalities are partly or totally unknown (Berg and Scheringer, 1994; Grandjean, 2013). In this case, proxy indicators with more or less predictive power are used to approximate potential impacts.

Table 1: Selected studies applying pressure, proxy and impact oriented indicators characterizing environmental sustainability.

Reference	Spatial level	Pressure indicators ¹⁾	Proxy indicators ²⁾	Impact indicators ³⁾
Alfsen et al. (1993)	Norway		X	
Gilbert et al. (1994)	Netherlands		X	
Nilsson et al. (1995)	Sewage Treatment Plant	X		
Azar et al. (1996)	World	X	X	
UNCSD (1996)	Not specified	X	X	X
Van der Voet (1996)	European Union	X	X	
Guinée, van den Bergh et al. (1999)	Netherlands	X	X	X
Umweltbundesamt (1999)	Austria	X	X	
UNCHS (2001)	World	X		
Graymore et al. (2010)	World	X	X	
EEA (2012)	European Union	X	X	X

Notes:

1) Examples for pressure indicators are the amount of waste and emission flows.

2) Examples for proxy indicators are (i) the spatial and temporal range of substances (Scheringer et al. 1994), (ii) the persistence, bio-accumulation, and toxicity of substances (European Parliament 2006), (iii) legal limits or political agreements (Frischknecht, Steiner et al. 2009), (iv) the ratio of anthropogenic to geogenic substance flows (Förstner et al. 1973, Reimann et al. 2005), and (v) exposure assessments (U.S. EPA 2011).

3) Examples for impact indicators are the number of human deaths due to certain substance flows into geogenic sinks.

Summarizing, the indicators developed so far focus on certain levels along the cause-effect chain. This includes the intervention level (pressure indicators), the effect level (impact indicators) or a level between intervention and effect (proxy indicators towards impacts). To our knowledge, individual indicators have not been linked yet systematically in view of ecological and human health assessment of regions. At present, the question “Which amounts of waste and emission flows are

acceptable and unacceptable, respectively?” cannot be answered with a single indicator. To overcome this gap, Döberl and Brunner (2004) proposed to amend the tool box of sustainability metrics by the following indicator:

$$\frac{\text{Amount of substances a region or process directs into appropriate final sinks}}{\text{Total amount of substances emitted by a region or process}} \quad \text{Equation 1}$$

Beyond the definition of the indicator, there is no operationalization in terms of assessment methods presented. However, the denominator of Equation 1 refers to the intervention level and the numerator of Equation 1 refers to a final level along the life cycle chain.

The present paper is inspired by Equation 1, and advances it further to make it operational for application. The aim of the paper is to develop an assessment method that

- is able to consider specific substances,
- takes into account discarded material flows (wastes, emissions, substance flows from wear, corrosion, and weathering) from human activities within a spatial unit,
- covers geogenic and anthropogenic sinks for discarded material flows,
- allows the integration of normative criteria such as proxy and impact criteria,
- consists of a quantifiable indicator.

To achieve this goal, we relate acceptable to actual substance flows into sinks. Actual flows are determined by regional SFA, usually on an annual base. Acceptable flows can be determined by any environmental assessment method. We have chosen a distant-to-target approach according to the Ecological Scarcity (ES) method, and apply this framework in three case studies. The indicator score is determined for (1) copper (Cu) in the city of Vienna, (2) lead (Pb) in the city of Vienna, and (3) perfluorooctanesulfonate (PFOS) in Switzerland. Based on the findings, we present options to control the indicator score. The resulting indicator serves as a guide to identify potential constraints for sinks to accommodate waste and emission flows. The indicator is intended to support material management in view of potential sink limitation. Accordingly, we propose to add this indicator to existing metrics for characterizing the environmental dimension of sustainability.

2. Material and methods

In the following sections, we (i) define the indicator, (ii) present the methods for calculating the indicator score, and (iii) apply the metric in three case studies.

2.1 Indicator definition

The sink indicator (λ) quantifies the environmentally acceptable mass share of a substance in actual waste and emission flows. The score ranges between 0% and 100% and is displayed as in Figure 5.

The sink indicator is defined by

$$\lambda = \frac{F_a}{F} * 100 \quad \text{Equation 2}$$

where F_a is the sum of acceptable actual flows in a region (see Equation 6) and F is the sum of actual flows in a region (see Equation 3).

$$F = \sum_{i=1}^n F_i \quad \text{with} \quad \text{Equation 3}$$

$$F_i = \begin{cases} \beta_i & \text{for } \beta_i > 0 \\ 0 & \text{for } \beta_i \leq 0 \end{cases} \quad \text{with} \quad \text{Equation 4}$$

$$\beta_i = \dot{m}_{in,i} - \dot{m}_{out,i} \quad \text{Equation 5}$$

where F_i is an actual flow in a region and i is an index for a process. Hence, β_i is the net flow of a process, $\dot{m}_{in,i}$ is the sum of flows into a process, and $\dot{m}_{out,i}$ is the sum of flows out of a process (see figure 2). If $\beta_i > 0$, then F_i is equal to the positive net flow of a process. In this case, the net flow could either be a net addition to stock or the transformed mass share of the substance. The method for calculating the actual flows is presented in section 2.2.2.

$$F_a = \sum_{i=1}^n F_{a,i} \quad \text{with} \quad \text{Equation 6}$$

$$F_{a,i} = \begin{cases} F_{c,i} & \text{for } \alpha_i \geq 0 \\ F_i & \text{for } \alpha_i < 0 \end{cases} \quad \text{with} \quad \text{Equation 7}$$

$$\alpha_i = F_i - F_{c,i} \quad \text{Equation 8}$$

where $F_{a,i}$ is an acceptable flow in a region, and i is an index for each processes. Hence, α_i is the distance-to-target value, F_i is the actual flow, and $F_{c,i}$ is the critical flow. A critical flow represents *proxy criteria* such as political targets or *damage criteria* such as accepted human health risks. The method for calculating the critical flow is presented in section 2.2.3.

Figure 1(a) shows the acceptable flow $F_{a,i}$ as a function of the distance-to-target value α_i . If $\alpha_i < 0$ then the actual flow F_i represents the acceptable flow $F_{a,i}$. If $\alpha_i \geq 0$ then the critical flow $F_{c,i}$ represents the acceptable flow $F_{a,i}$. Figure 1(b) plots the actual flow F_i on both axes in combination with the distant-to-target value α_i . This produces three potential sub-flows: First, the acceptable flow is the mass flow below the actual flow (for $\alpha_i < 0$) or below the critical flow (for $\alpha_i \geq 0$). Second, the unacceptable flow is the mass flow above the critical flow. Third, the tolerable flow is the mass flow below the critical flow and above the actual flow. In other words, the tolerable flow expresses the potential to increase the actual flow without violating normative criteria.

Figure 1 (a): Acceptable flow $F_{a,i}$ as a function of the distant-to-target value α_i . Example 1 demonstrates that for $\alpha_i < 0$ the acceptable flow $F_{a,i}$ is equal to the actual flow F_i . Example 2 demonstrates that for $\alpha_i \geq 0$ the acceptable flow $F_{a,i}$ is equal to the critical flow $F_{c,i}$. Figure 1 (b): The combination of the actual flow F_i and the distant-to-target value α_i yields two potential sub-flows: acceptable and unacceptable flows. Example 1 demonstrates for $\alpha_i < 0$ the results of an acceptable flow. Example 2 demonstrates for $\alpha_i \geq 0$ the result of acceptable and unacceptable flows.

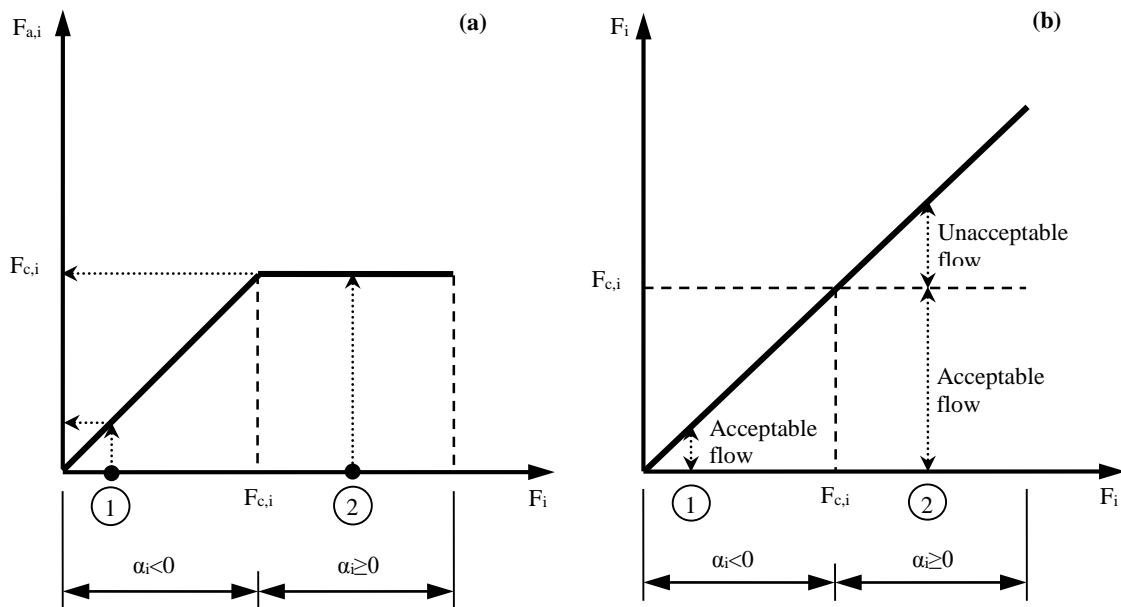
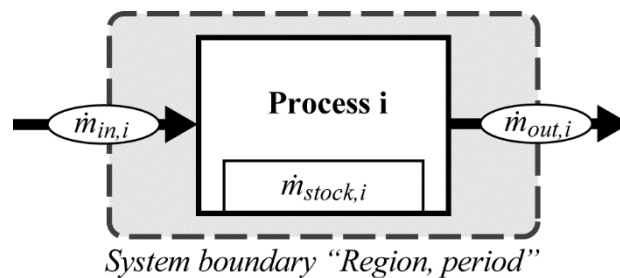


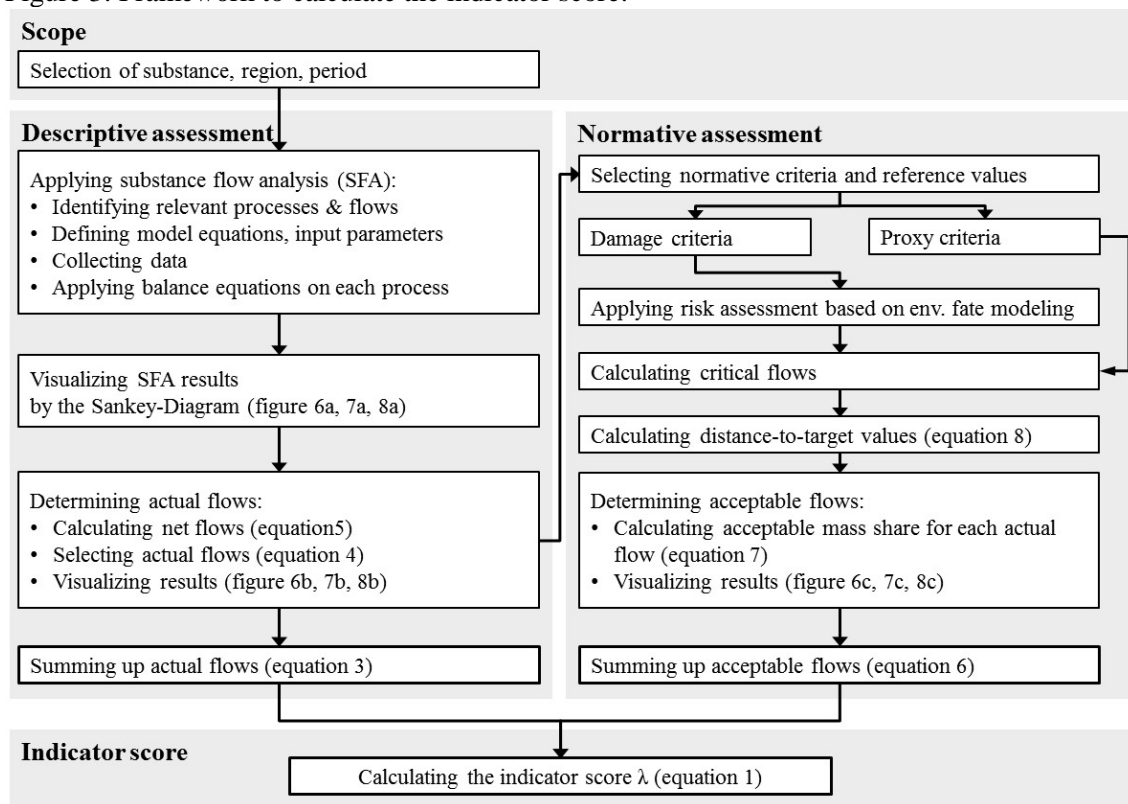
Figure 2: Plot of a generic process i , where $\dot{m}_{in,i}$ is the substance flow [mass per time] entering process i , $\dot{m}_{out,i}$ is the substance flow [mass per time] leaving process i , and $\dot{m}_{stock,i}$ is the resulting alteration of mass [mass/time] within process i .



2.2 Methods for calculating the indicator score

To calculate the indicator score four steps are required (figure 3): First, the scope of assessment is defined. Second, a descriptive assessment of flows yields the sum of actual flows F . Third, a normative assessment of flows yields the sum of acceptable flows F_a . Fourth, the indicator score is calculated.

Figure 3: Framework to calculate the indicator score.



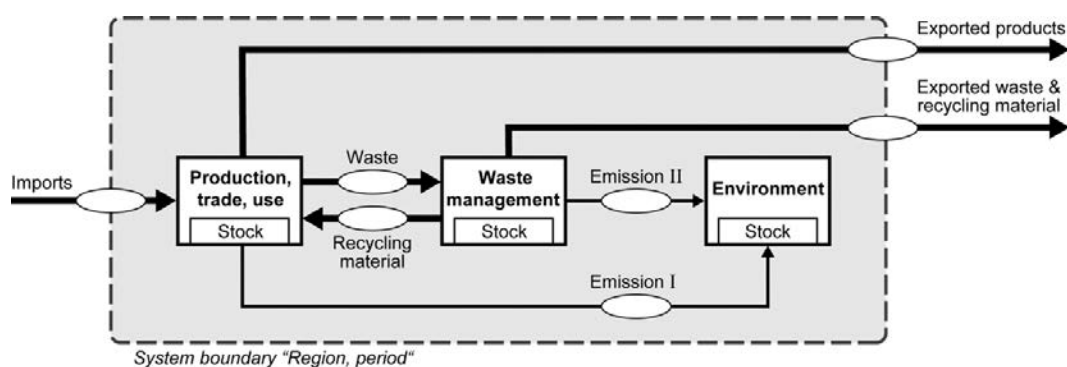
2.2.1 Scope

Setting the scope of the assessment includes the selection of a substance, a reference region and a period of interest. First, the notion “substance” is defined as “Matter of constant composition best characterized by the entities (atoms, molecules, formula units) it is composed of” (Nic et al., 2012). This is in line with the SFA framework used in this study, which defines the notion as “any (chemical) element or compound composed of uniform units” (Brunner and Rechberger, 2004). Second, the assessment deliberately focuses on regional flows instead of all flows along the life cycle chain of products. A region is bounded by administrative limits. Accordingly, communities, cities, federal states, nations or continents are subjects of the assessment. Third, setting the system boundary in time includes the selection of a reference period (day, week, month, year, decade, and so on) and a reference point in time (e.g. the year 2013). Temporal variations of flows within the reference period are often neglected because of lack of data.

2.2.2 Descriptive assessment

To quantify the sum of actual flows F (equation 3), four steps are needed: First, to investigate into the anthropogenic metabolism, SFA has been proven to be a practical tool. It tracks the pathway of selected substances through systems such as households, enterprises, cities or regions. The applied methodology is in accordance with Baccini and Bader (1996); Brunner and Rechberger (2004). The model development focuses on the identification of relevant processes and their links in terms of flows. Figure 4 present a framework for developing the SFA model. Model equations define the flows and stocks with the help of input parameters. Next, balance equations are applied for each process (figure 2, figure 4). The software STAN is used for data reconciliation and error propagation in order to balance mass flows and stocks (Cencic, 2012). Second, Sankey-Diagrams are elaborated to present SFA results (Schmidt, 2008). Third, the actual flows are determined. Therefore, the net flow β_i of each process (equation 5) is calculated and displayed. This kind of plot allows the comparisons of various Sankey-Diagrams in a comparable manner. A positive net flow ($\beta_i > 0$) indicates a sink process, and a negative net flow ($\beta_i < 0$) indicates a source process. The actual flow F_i is defined as a positive net flow into a sub-process within the process “Waste management” and “Environment” (equation 4). Fourth, the actual flows are summed up (equation 3).

Figure 4: The SFA model refers to a specific substance, region and period of time. It includes processes in the, production, trade and use phase, in the waste management sector, and in the environment. Flows link the processes. The flows assessed by the indicator λ include the actual flows within the flows “Waste”, “Emissions I”, and “Emissions II”.



For two out of the three case studies, we use SFA data that have already been published: Cu in Vienna for the year 2008 (Kral et al., 2014) and PFOS in Switzerland for the year 2007 (Buser and Morf, 2009). The third case study focuses on Pb in Vienna for the year 2008. Background datasets can be found in the supplement information. In common, the Sankey-Diagram serves as starting point for calculating the net flows β_i and for filtering the actual flows F_i .

2.2.3 Normative assessment

To determine the acceptable flow F_a (equation 6), four steps are needed: First, normative criteria and reference values define the acceptance of flows into sinks. Criteria and reference values are derived from goal oriented frameworks with respect to waste and emissions, such as regulations, standards, political agreements, or concepts for sustainable resource use like “clean cycles and final sinks” (Brunner, 2010; Kral et al., 2013), and “gradle to gradle” (Mulhall and Braungart, 2010). The definition of criteria depends on circumstances in the case study region. The circumstances might change over time, for example, as a consequence of new scientific knowledge, by improved data availability, and by changes in the ethical value-sphere. The circumstance might also vary from region to region, for example, as a consequence of initiatives to increase the recycling rate, different political agreements for accepted emission rates, and environmental quality standards. To select normative criteria and reference values in a specific region, the outcome of stakeholders’ involvement might be considered, without being further discussed in this article. To consider various criteria in the indicator framework, a criteria is related at any stage throughout the cause-effect chain of a substance, and a reference value determines the normative criteria in a quantitative manner. Table 2 gives some examples for proxy criteria at the stage of pressure, state, exposure and effect, and damage criteria at the stage of damage. Second, the critical flows are calculated based on models that establish causal links between the actual flow and the reference value attributed. Hence, the actual flow F_i is varied as long as the reference value is achieved. The resulting flow is called critical flow $F_{c,i}$. In the case of multiple criteria for a single actual flow, the most stringent criteria is selected. Third, the distant-to-target value (equation 8) determines the acceptable flow $F_{a,i}$ (equation 7). Fourth, the acceptable flows are summed up (equation 6).

Table 2: Examples for normative criteria and reference values in context of the cause-effect chain, from the source towards the damage (adopted from Hofstetter 1998, Frischknecht 2009).

Stage	Stage description	Normative criteria	Reference values
Source	Potential of waste and emissions	n.r.	n.r.
Pressure	Flow into sink	Emission rates Waste into landfill	Legal limits, political agreements
State	Substance fate in a) geogenic and b) anthropogenic sinks.	Substance concentration, accumulation or transformation rate in a) environmental media, and b) recycling goods, landfills, underground storage facilities.	Geogenic reference values in soil; Approved landfill capacity and disposal time;
Exposure	Standard characteristics of exposed organism	Exposed dose, collective effective dose	MAK values, BAT values
Effect	Dose-Response-Relationship	Number and type of human diseases, Number of vanishing plant species	Accepted number of cases regarding an human disease
Damage	Damage to human health or ecosystem quality	DALYs, QALYs, Share of vanishing plant species per area and time unit.	Value weighted DALYs, Accepted share of vanishing plant species per area and time unit

Notes: n.r. = not relevant; MAK values = Maximum Concentrations at the Workplace, BAT values = Biological Tolerance Values; DALY = disability-adjusted life years or disease-adjusted life years; QALY = quality adjusted life year;

In view of the case studies, the selection of normative criteria and references values is based on the Ecological Scarcity (ES) method (Frischknecht et al., 2009) including the following adoptions:

- ES method provides critical flow data for Switzerland. We adopt the data according to local circumstances in the case study region. For example, critical metal flows into surface waters refer to local environmental quality standards for surface waters of the specific case study country.
- ES method uses proxy criteria such as national reduction targets for greenhouse gas emissions and legal standards for heavy metal concentrations in surface waters. In addition, we introduce impact criteria such as human health risk. The integration of impact criteria demonstrates an additional option for critical flow determination. For human health risk, the cancer risk level (RL) and the non-cancer hazard-index (HI) represent two impact criteria.
- The ES method does not include PFOS as substance of interest. Legal limits in terms of concentrations in various emission flows have not been published yet. To assess flows into surface waters, we use an U.S. based reference concentration as proxy criteria.
- ES method assesses waste flows into landfills based on standards for the carbon content. We replace this proxy criterion with the constraints given by the official permission for each landfill. Therefore, the remaining landfill volume is divided by the approved, remaining time for disposal. The conversion of the annual volume flow into mass yields the annual critical flow into landfills.
- ES method aims at assessing impacts of waste and emission flows. Flows without impacts are not taken into account. But, they are of concern for the applied method. For example, organic substances are transformed mainly into carbon dioxide and water in incinerators, and are not present in their original form anymore. Accordingly, we introduce proxy criteria for organic substances in incineration. The critical flow is defined with the capacity of the incinerator in mass per year.

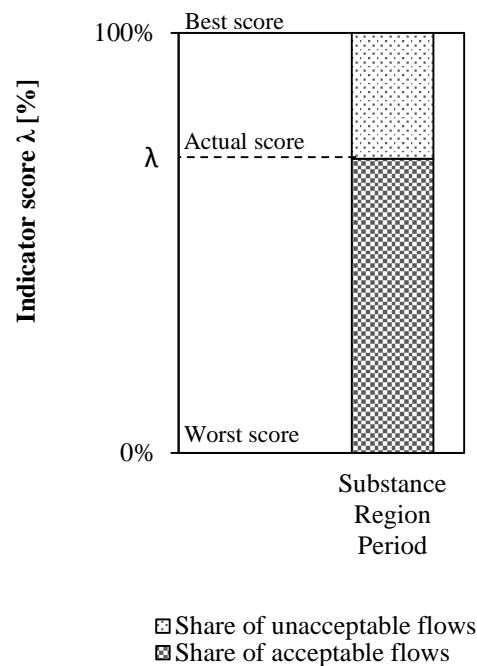
The applied method is in line with ES methodology as follows:

- Heavy metals to soils: The Swiss Regulation on the Impact on Soils (Schweizer Bundesrat, 1998) aims to ensure long-term soil fertility. Accumulation of heavy metals in soil is not accepted. ES method defines the critical flow as the heavy metal uptake through plants. This simplified approach neglecting leaching from the soil might be justified due to a lack of more precise regional data, but should be amended in the future.
- Hazardous waste to underground storage facilities: Switzerland has no appropriate storage facilities and thus exports hazardous waste to foreign underground storages facilities. In consultation with Swiss authorities, ES method sets the actual flow equal to the critical flow. Just as Switzerland, Austria exports waste into underground storage facilities. This justifies the same definition of the critical flow.

2.2.4 Indicator score

Applying descriptive and normative assessment methods result in an indicator score, ranging from 0% to 100% (Figure 5). The indicator takes 100% of actual flows into account, and discriminates between acceptable and unacceptable flows. Either all actual flows are fulfilling criteria of acceptability ($\lambda=100\%$), at least one flow is unacceptable ($0\%<\lambda<100\%$), or all actual flows are unacceptable ($\lambda=0\%$). Accordingly, the positive connotation of the score can be seen as the more the better.

Figure 5: Generic plot of the indicator score λ shows both, the amount of acceptable and unacceptable flows into sinks. The score refers to the total flows into sinks of a given substance, within a given region and within a given time period.



In general, the interpretation of the indicator score depends on the selection of (i) actual flows, (ii) normative criteria and references values, and (iii) data availability.

The export flow “Exported waste & recycling material” crosses the spatial system boundary and enters external regions (see figure 4). It might be that the flow is not acceptable due to local circumstances in the export region, for instance, as a consequence of missing standards for pollution control, a lack of environmental sound treatment and recycling facilities, and sanitary landfills. The relevance of waste exports fraught with risk are well documented, for example, for waste electrical and electronic and equipment (e.g. Sthiannopkao and Wong, 2013; Widmer et al., 2005). In view of the present case studies, the flow “Exported waste & recycling material” is allocated to the external region and not taken into account, except those into underground storage facilities. To

allocate flows into external sinks to the export region, critical flows for “Exported waste & recycling material” have to be defined.

Normative criteria and reference values can be derived from goal oriented frameworks such as regulations, standards, and political agreements. If the goals cannot be operationalized in terms of normative criteria and reference values, or if criteria are not considered in the indicator score calculation, the indicator score lacks of interpretational power regarding the goal oriented concepts. For example, conclusions regarding the effectiveness of recycling initiatives fail, if a normative criteria regarding recycling is missing. To overcome this gap, the critical flow into landfill might be 10% lower than the actual flow, if the recycling rate should be enhanced by 10%.

Data acquisition is based on a bottom-up approach, supposing appropriate data quality and quantity. If data is lacking, the outcomes point to data requirements that have to be met before implications for environmental and waste management can be identified.

2.3 Case studies

The following three case studies are used to demonstrate the application of the sink indicator: Cu in Vienna, Pb in Vienna, and PFOS in Switzerland (table 2). First, we highlight the motivation for the case study selection and briefly explain the background. Second, we present detailed Sankey-Diagrams including substance flows and calculate the sum of actual flows F according to Equation 3. Third, we calculate the critical flows in order to discriminate between acceptable and unacceptable flows, respectively. The sum of acceptable flow yields F_a according to Equation 6. Results are presented in table 3.

Table 3: Case study overview

Substance	Region	Year	Descriptive method	Number of actual flows	Number of sink processes	Type of criteria	SFA data reference
Cu	Vienna	2008	SFA	8	4	Proxy criteria	Kral, Lin et al. (2014)
Pb	Vienna	2008	SFA; EFM	9	7	Proxy and damage criteria	Supplement information
PFOS	Switzerland	2007	SFA	6	5	Proxy criteria	Buser et al. (2009)

Notes: Cu = copper; Pb = lead; PFOS = perfluorooctanesulfonate; SFA = substance flow analysis; EFM = environmental fate modeling;

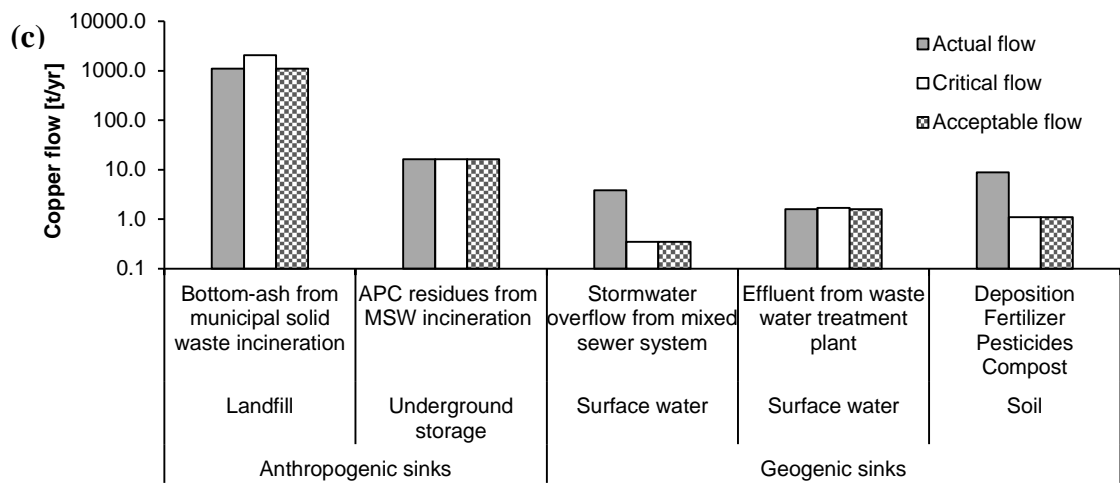
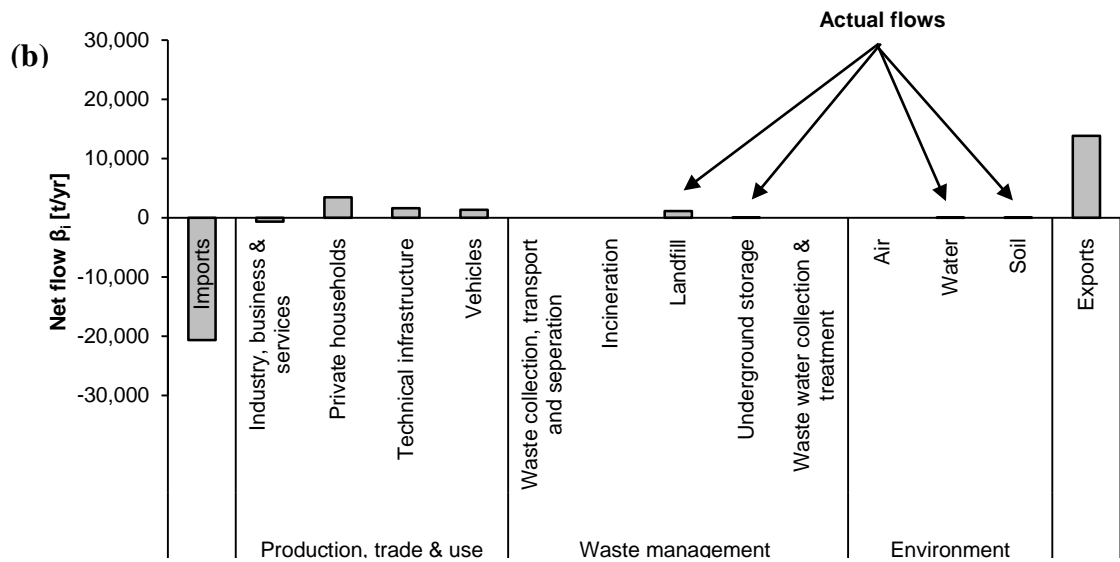
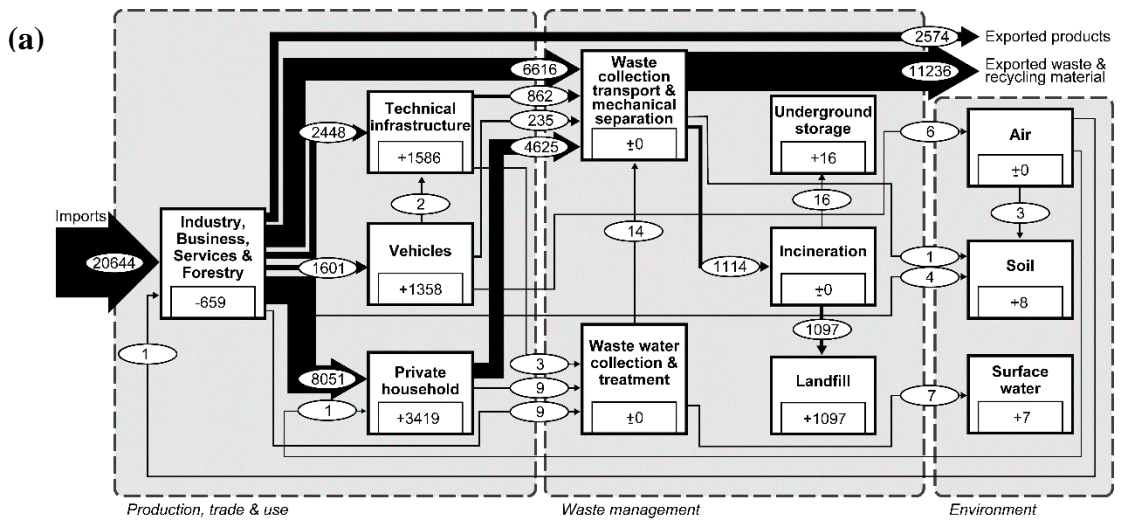
2.3.1 Cu in Vienna

Copper is relevant from both a resource use and environmental impact viewpoint. On the one hand, Cu is essential for modern lifestyles, resulting in Cu waste fractions that have to be disposed of. From 1900-2000, about 0.7% of the Swiss Cu stock have been annually discarded in landfills (Wittmer, 2006). On the other hand, Cu is emitted from point and non-point sources and poses a risk for aquatic life. In Germany, it has been estimated that about 30% of total Cu loadings in receiving waters originate from urban areas (Böhm et al., 2001). In Vienna, Cu concentrations in sewage sludge are significantly larger than in rural areas (Kroiss et al., 2008), and Cu concentrations in urban soils are higher than in surrounding rural areas (Pfleiderer, 2011).

The Sankey-Diagram in figure 6a represents the annual Cu flows for the city of Vienna for the year 2008. Details about the SFA study have been published by (Kral et al., 2014). Calculating the sum of actual flows yields 1,129 t Cu/yr (figure 6b). Thereof, 97.3% are disposed of in a local landfill, 1.5% are shipped to a foreign underground storage facility, 0.8% are deposited on urban soil, and 0.4% are entering receiving waters.

To determine the critical flows, we use proxy criteria. The critical flow of bottom-ash into landfill results from the available landfill volume of 3.45 million cubic meter, a density of 1.8 tons per cubic meter, the approved remaining time for disposal with 19 year (Ableidinger et al., 2007), and the actual Cu mass in bottom ash with 990 kg/yr. The critical flow for exported air-pollution-control (APC) residues into an underground storage facility is set equal to the actual flow. This assessment is justified, because the Viennese disposal practice meets the Swiss practice. The critical flow into soil is taken as equal to the Cu uptake through plants, which has been calculated for green and agricultural areas in Vienna (Kral et al., 2014). The critical flow into receiving water is based on environmental standards for surface waters with 9.3 microgram Cu per liter ($\mu\text{g Cu/l}$) (Bundesrepublik Österreich, 2006). There are two flows in total: The actual overflow from mixed sewer system is 37.5 million tons of water per year (Leitner, 2013). The actual Cu concentration of effluents from WTP is $8.8 \mu\text{g Cu/l}$ (Kroiss et al., 2008).

Figure 6: **(a)** Sankey-Diagram for copper in Vienna for the year 2008. Annual flow rates and changes in stocks are given in tons per year (t/yr), for stocks in tons (t). The flows are represented as Sankey diagrams proportional to the flow rate; figures for stocks are given within the process boxes. Deviations from mass balance are due to rounding. **(b)** The Source-Sink-Diagram presents the net flows β_i for each process i . The actual flows are positive net flows into the waste management sector and the environment. **(c)** The plot shows the results from normative assessment, namely the rated flows into sinks.



2.3.2 Pb in Vienna

Human health is directly affected by emissions of Pb and Pb compounds. In Austria, Pb emissions to air decreased from 218 t/yr in the year 1990 to 13 t/yr in 2009 (Anderl et al., 2011). In 1993, lead has been banned from the Austrian petrol market. It is still used in accumulators, building coatings, tires and paints. Due to former Pb depositions and present diffusive losses, anthropogenic Pb is found in urban soils (Kreiner, 2004). Hence, it can be transferred to fodder and food, and may affect human health (WHO, 2007). Up to now, the Pb content in Austrian soils lacks of systematic nationwide monitoring (Umweltbundesamt, 2010).

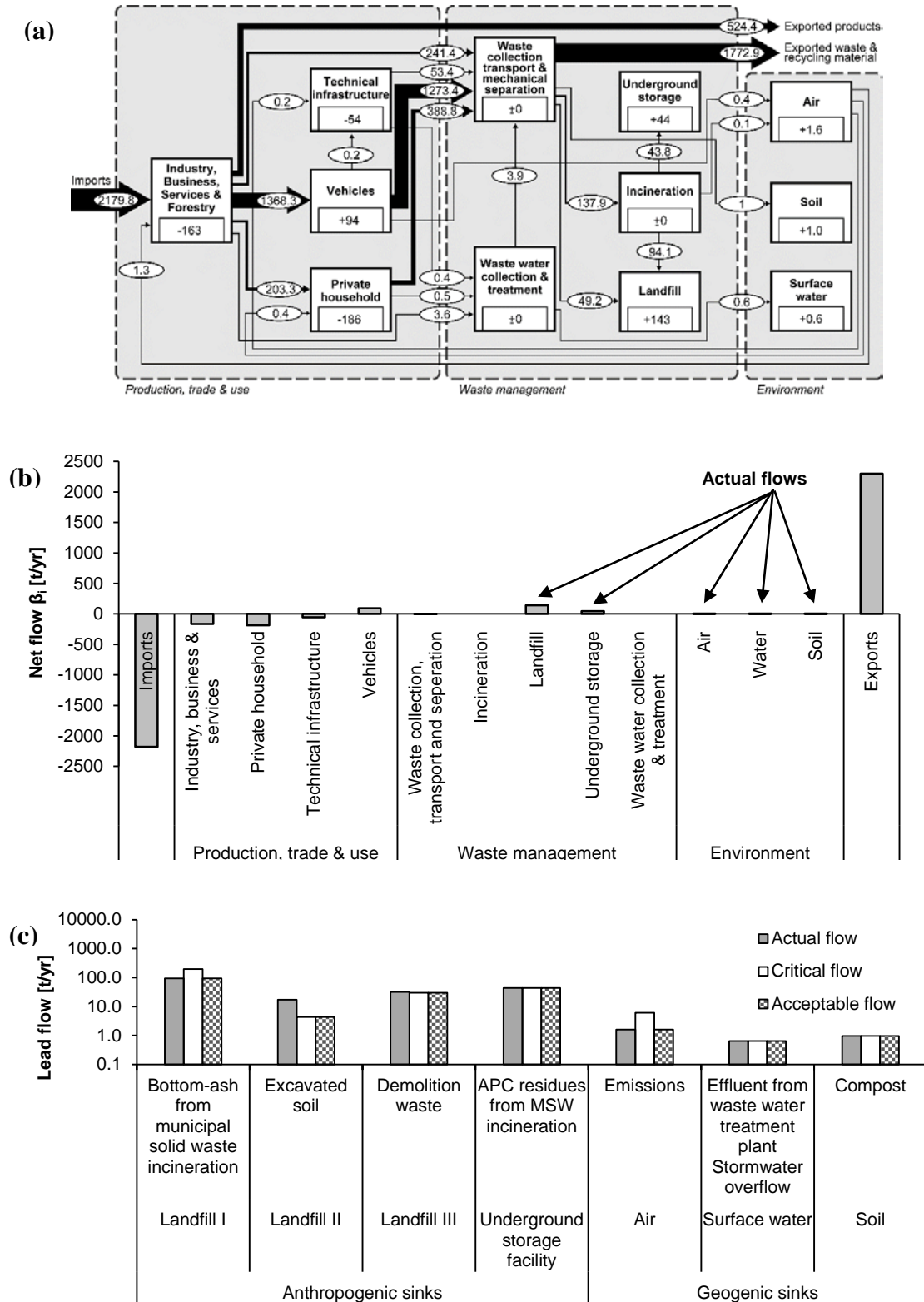
The Sankey-Diagram in figure 7a represents the annual Pb flows for the city of Vienna for the year 2008. Details about the SFA are given in the supplemental information. Calculating the sum of actual flows yields the sum of actual flows with 191 t Pb/yr, of which 75.4% entered a local landfill, 22.9% entered a foreign underground storage facility, 0.8% entered ambient air, 0.5% entered urban soil, and 0.3% entered receiving waters (figure 7b).

To determine the critical flows, we use (a) damage criteria for flows into geogenic sinks and (b) proxy criteria for flows into anthropogenic sinks.

ad (a) The impact of flows into geogenic sinks is assessed in view of human health risks. Therefore, we apply the risk assessment model CalTOX 4.0 beta (McKone and Enoch, 2002). CalTOX has been developed to assess human exposures from continuous emissions to multiple environmental media. Background datasets can be found in the supplemental information. The method quantifies two damage criteria, namely the RL and the HI. To calculate the critical flows, we varied the actual flows as long as the damage criteria result acceptable risks. To demonstrate the method, widely-used acceptable risks of 10^{-6} for RL and 1 for HI are selected, without discussing further acceptable risks (Kelly, 1991). Each actual flow is varied in a single scenario. Each variation results in two critical flows. One meets the acceptable RL, another meets the acceptable HI. Accordingly, three actual flows result in six scenarios. We picked out a stringent scenario, representing the minimum ratio between the critical flow and the actual flow.

ad (b) Flows into anthropogenic sinks are determined with proxy criteria in accordance to the Cu case study. The critical flow into landfills takes into account three different flows: Bottom ash from incineration, excavated soil, and demolition waste. An average annual flow is calculated with respect to landfill capacity. Therefore, the approved remaining landfill volume is divided by the approved disposal time (Ableidinger et al., 2007). The flow into the foreign underground storage facility is assessed in accordance with ES method. Therefore, the actual flow equals the critical flow. This approach has been chosen because the Viennese disposal practice is equal to the Swiss practice.

Figure 7: (a) Sankey-Diagram for lead in Vienna for the year 2008. Annual flow rates and changes in stocks are given in tons per year (t/yr), for stocks in tons (t). The flows are represented as Sankey diagrams proportional to the flow rate; figures for stocks are given within the process boxes. Deviations from mass balance are due to rounding. (b) The Source-Sink-Diagram presents the net flows β_i for each process i . The actual flows are positive net flows into processes within the waste management sector and the environment. (c) The plot shows the results from normative assessment, namely the rated flows into sinks.



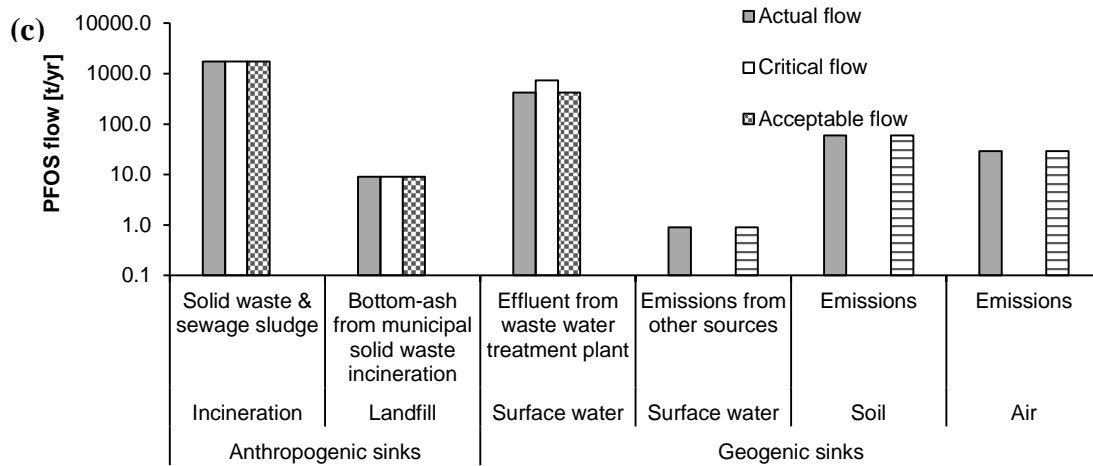
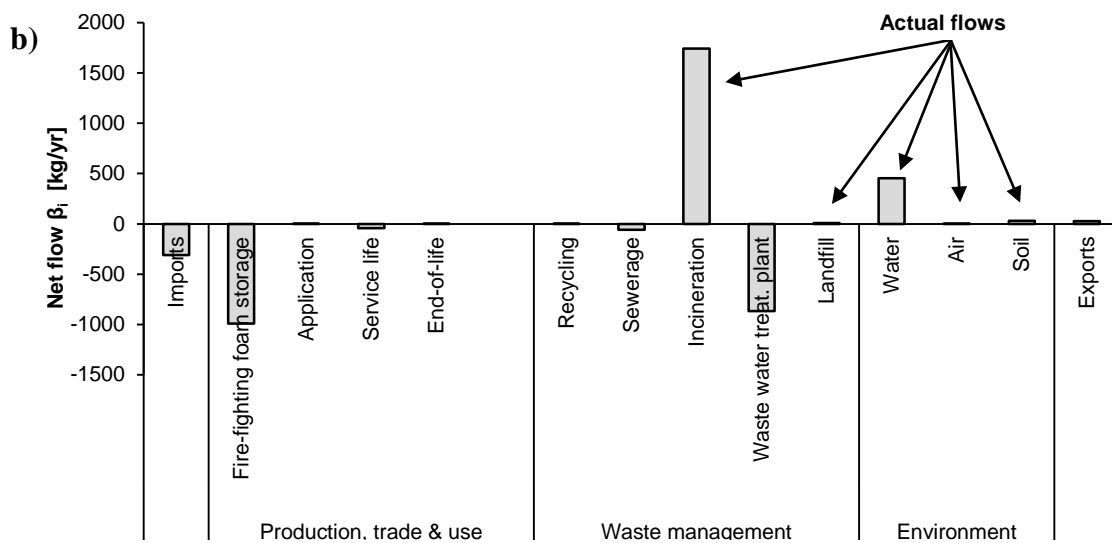
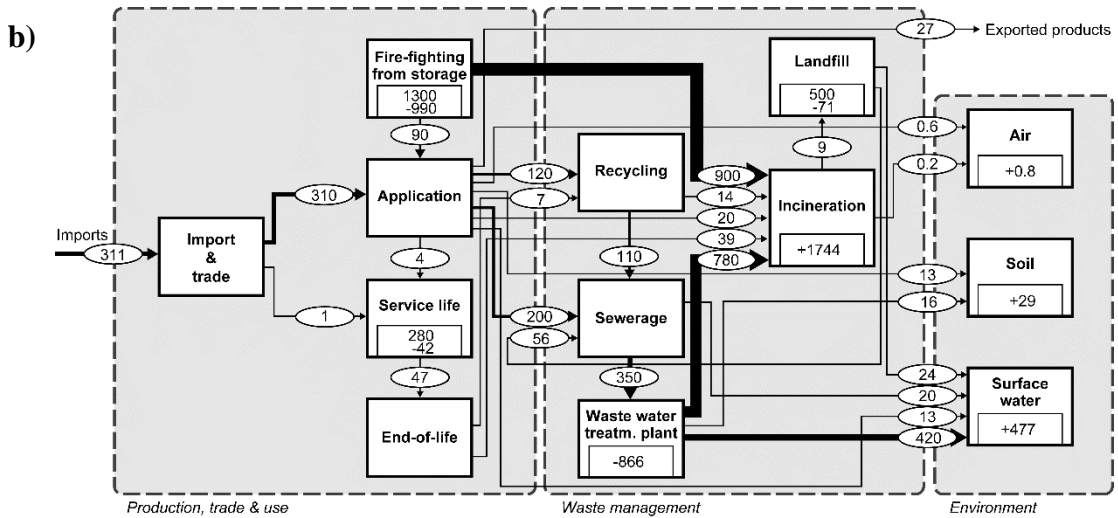
2.3.3 PFOS in Switzerland

Perfluorooctane sulfuric acid and its derivatives, collectively named PFOS, are persistent, bio-accumulative and toxic substances. They are regulated under the Persistent Organic Pollutants Regulation 850/2004 (European Parliament, 2004) and Regulation 2006/122/EG (European Parliament, 2006). In 2010, PFOS has been added to the convention with some exemptions for specific applications (European Commission, 2010). Hence, the European Union urges member states to implement strategies for careful PFOS management.

In Switzerland, the Federal Office of the Environment conducted a national study regarding the determination of stockpiles and waste fractions containing PFOS for the year 2007 (Buser and Morf, 2009). The corresponding Sankey-Diagram can be found in figure 8a. The production and use phase as well as - in consequence of - the waste water treatment plant (WWTP) act as main PFOS sources. Waste management provides the “anthropogenic sink” incineration, which mineralizes PFOS into carbon dioxide, water and HF. Environment provides the geogenic sinks hydrosphere, soil and atmosphere. Calculating the sum of actual flows yields 2,260 kg PFOS/yr, of which 77.1% entered incineration, 0.4% entered landfills, and 22.5% entered geogenic sinks within hydrosphere, atmosphere and soil (figure 8b). The flow from WWTP to hydrosphere is determined with a concentration of $114 \cdot 10^{-9}$ gram PFOS per liter (ng/l) (Götz et al., 2011).

To determine the critical flows, we classified the actual PFOS flows into incineration as acceptable flow. This is in accordance to the EU Regulation, because PFOS is mineralized if it undergoes thermal treatment. Beyond the mineralized fraction, there is a very small PFOS fraction in incineration ashes. Due to the large concentrations of heavy metals and certain organic refractory substances, the ashes are classified as hazardous waste and are deposited in landfills or foreign underground storage facilities. According to ES method, this actual flow is set equal to the critical flow. This assessment is justified, because the Viennese disposal practice meets the Swiss practice. For flows into geogenic sinks, there is a lack of normative criteria. To estimate the critical flow from WWTP into surface waters, national standards are actually missing but under development (Götz et al., 2011). We used the critical concentration of 200 ng/l PFOS. This value is a provisional health advisory for drinking water, published by US EPA (2009). Due to the lack of regulations and standards, other flows to the environment cannot be assessed and thus are excluded from normative assessment.

Figure 8: **(a)** Sankey-Diagram for PFOS in Switzerland for the year 2007. Annual flow rates and changes in stocks are given in kilogram per year (kg/yr), for stocks in kilogram (kg). The flows are represented as Sankey arrows proportional to the flow rate; figures for stocks are given within the process boxes. Deviations from mass balance are due to rounding. **(b)** The Source-Sink-Diagram presents the net flows β_i for each process i . The actual flows are positive net flows into processes within the waste management sector and the environment. **(c)** The plot shows the results from normative assessment, namely the rated flows into sinks.



3. Results

The following sections give an overview of the results, and discuss the composition of the indicator score λ for each case study.

3.1 Overview

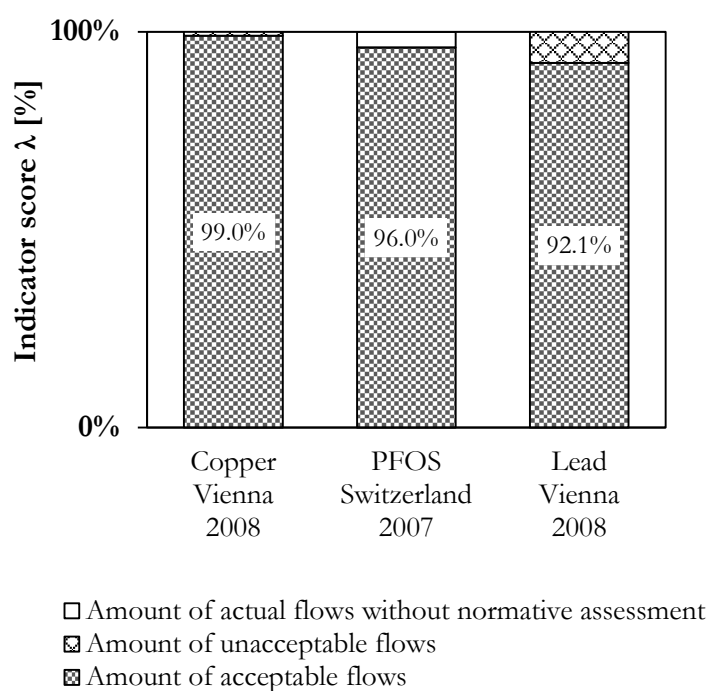
The indicator score λ for each case studies is visualized in figure 9. It quantifies the share of acceptable flows in entire flows into sinks. The Cu metabolism in Vienna is limited by flows into the geogenic sink urban soil and receiving waters. The PFOS metabolism in Switzerland partly lacks of normative assessment due to lack of data, and the Pb metabolism in Vienna is restrained by flows into anthropogenic sinks such as local landfills. To compute the indicator score, the sum of acceptable and actual flows is needed and compiled in table 4.

Table 4: Substance flow data required for calculating the indicator score.

Case study	Sink	Flow	Actual flow F_i [t/yr]	Critical flow $F_{c,i}$ [t/yr]	Acceptable flow $F_{a,i}$ [t/yr]
Cu	Landfill	Bottom-ash from MSW incineration	1,097.2	2,075.5	1,097.2
	Underground storage facility	APC residues from MSW incineration (exported filter cake)	16.4	16.4	16.4
	Surface water	Stormwater overflow from mixed sewer system	3.9	0.3	0.3
	Surface water	Effluent from WWTP	1.6	1.7	1.6
	Soil	Deposition, Fertilizer, Pesticides, Compost	8.8	1.1	1.1
		Sum		$F = 1,127.9$	-
Pb	Landfill I	Bottom-ash from MSW incineration	94.7	198.5	94.7
	Landfill II	Excavated soil	17.3	4.3	4.3
	Landfill III	Demolition waste	31.9	29.8	29.8
	Underground storage facility	APC residues from MSW incineration (exported filter cake)	43.9	43.9	43.9
	Air	Emissions	1.6	6.1	1.6
	Surface water	Stormwater overflow from mixed sewer system Runoff from separated sewer system	0.6	0.6	0.6
	Soil	Effluent from WWTP Compost	1.0	1.0	1.0
	Sum		$F = 190.9$	-	$F_a = 175.8$
PFOS	Incineration	Solid waste and sewage sludge	1.744	1.744	1.744
	Landfill	Bottom-ash from MSW incineration	0.009	0.009	0.009
	Surface water	Effluent from WWTP	0.420	0.737	0.420
	Surface water	Emissions from other sources	0.060	n.a.	n.a.
	Soil	Emissions	0.029	n.a.	n.a.
	Air	Emissions	0.001	n.a.	n.a.
	Sum		$F = 2.263$	-	$F_a = 2.173$

Note: -=not relevant; n.a.=not available due to a lack of normative criteria; APC=Air pollution control; WWTP=Waste water treatment plant; t/yr=tons per year, F_a =sum of acceptable flows; F =sum of actual flows; Cu=copper, Pb=lead; PFOS=perfluorooctanesulfonate

Figure 9: Compilation of Indicator scores λ for the three case studies.



3.2 Cu in Vienna

99.0% of all actual Cu flows into sinks are acceptable. The sink “soil” poses a constraint for 0.7% of all actual Cu flows only. The sink “surface water” poses a constraint for 0.3% of all actual Cu flows. In detail, the following results have been obtained (figure 6c), whereas the percentage numbers refer to the sum of actual Cu flows ($100\% \cong 1.128 \text{ t/yr}$):

- Landfill: 97.3% of flows are due to bottom-ash from MSW incineration. This is in compliance with landfill regulation. Calculated by the approved volume and service time of the landfill, the critical flow is 189% larger than the actual flow. Consequently, there is no constraint for the disposal of bottom-ash until the end of the approved service time. However, recycling is one option to disburden the landfill. If the stakeholders strive to increase the recycling rate of bottom-ash, the indicator calculation has to include a criteria with respect to both, the flow into the landfill and the recycled material flow (see chapter 2.2.4).
- Underground storage facility: 1.5% of flows are due to APC residues from MSW incineration. These residues are exported and disposed of in approved underground storage facilities.
- Water: 0.1% of flows are from effluents from WWTP, fulfilling quality standards for surface waters. However, the actual flow is only 6% below the critical flow. Beside WWTP effluents, Cu in stormwater overflow from mixed sewer systems enters receiving waters too. Applying the same standards as for effluents shows that the actual flow is 11 times

larger than the critical flow. From an impact point of view, the flow complies with the quality standards in the receiving water. For the future, the indicator score might increase due to ongoing measures for reducing Cu bypassing waste water treatment via storm water overflow. Retention reservoirs and collection sewers are constructed at present in order to direct more urban surface waters including diffusive Cu losses towards WWTP (e.g. Stadt Wien, 2013).

- Soil: 0.1% of flows are acceptable, and 0.7% of flows are unacceptable because they exceed the critical level by a factor of eight. Cu flows into the soil are larger than the removal by plants. If no Cu leaching from soil is assumed (cf. ES methodology), Cu will be accumulating in specific soil compartments. The determination of the critical flow was rather crude due to a lack of accurate local data. Consequently, the quantity and quality of data has to be improved. This finding is in agreement with the recommendation of the Austrian Environmental Agency for a nation-wide soil metal monitoring program (Umweltbundesamt, 2010). Improved sampling with respect to horizontal and vertical sample collection and including the identification of potential hot spots would allow decreasing the uncertainty when determining the amount of unacceptable flows to the soil.

3.3 Pb in Vienna

92.1% of all actual Pb flows into sinks are acceptable. The sink “landfill” poses a constraint for 7.9% of all actual Pb flows. In detail, the following results have been obtained (figure 7c), whereas the percentage numbers refer to the sum of actual Pb flows (100% \cong 191 t/yr):

- Landfills: 49.6% of flows is due to bottom-ash from MSW incineration. Due to the permit of the landfill, the critical flow is twice as large as the actual flow. 15.9% of all actual Pb flows originates from excavated soil to local landfills. If the landfill capacity should be fully utilized at the end of approved disposal time, only 9.1% of all actual Pb flows (instead of 15.9%) can be disposed of. The same pattern was found for Pb in demolition waste. 17.8% of Pb origins from demolition waste to landfills. If the landfill capacity should be utilized at the end of approved disposal time, only 16.7% of all actual Pb flows (instead of 17.8) can be disposed of. If the disposal practice continues, landfills for excavated soil and for demolition waste will exceed their approved landfill volume within the approved time for disposal. In other words, the disposal of actual flows is in accordance with legal limits, but the disposal practice faces constraints. To overcome these constraints, landfill permissions have to be extended in time. Alternatively, waste fractions can be recycled complying with advanced quality standards (BRV 2009), or wastes are directed to remote landfills beyond the system limits, which increases transport distances and costs.

- Underground storage: 22.9% of flows originate from APC residues from MSW incineration. The fractions are acceptable and exported into foreign underground storage facilities.
- Air, soil, water: 0.8% of flows enter ambient air, 0.5% enters urban soil, and 0.3% enters the water. The actual flows yield acceptable risks (RL: $7 \cdot 10^{-6}$, HI: 0.26). Increasing the actual flow into air by a factor of three results in a critical flow (HI: 1). Even though the results yield acceptable human health risks, it has to be noted that the method is based on a uniform approach without spatial resolution. Thus, accidental hot spots representing possible risks for the local population are not included by this approach.

3.4 PFOS in Switzerland

96.0% of all actual PFOS flows into sinks are acceptable. 4% of all actual PFOS flows lack of normative assessment due to insufficient knowledge. In detail, the following results have been obtained (figure 8c), whereas the percentage numbers refer to the sum of actual PFOS flows ($100\% \cong 2.260 \text{ t/yr}$):

- Incineration: 77.1% of PFOS flows are originating from solid waste and sewage sludge and are treated by MSW incineration. This flow is mineralized and is classified as acceptable flow, which is accordance to the EU Regulation (European Commission, 2010).
- Landfill: 0.4% of flows derives from residues of waste incineration and enters landfills in an acceptable manner. Today's PFOS emissions from landfills result from former rather than from present waste disposal. They have been assessed too.
- Water: Two flows enter the aquatic sphere. 18.6% of PFOS are within WWTP effluents and fulfill provisional drinking water standards (US EPA, 2009). Up to now in Switzerland, legal limits are missing but under development (Götz et al., 2011). Even though standards will be available in the future, the monitoring of PFOS is rather challenging and expensive (e.g. Becker et al., 2008; Schultz et al., 2005). 2.7% of PFOS originate from additional sources. This flow has not been assessed, because data and standards were missing.
- Soil and air: 1.3% of PFOS enter the soil, and 0.04% enter the air. These flows, together with the 2.7% of flows into water have not been assessed. They pose potential threats on human and ecological health without a clear understanding about the fate and effects.

4. Conclusions

A new methodology is presented to assess if sinks are a constraint for waste and emission flows. The methodology is based on an indicator, ranging from 0% to 100%, and representing the ratio between the amount of environmentally acceptable and unacceptable flows into sinks. To our knowledge, it is the first indicator that indicates possible constraints for regional waste and emission flows to sinks in a region-wide perspective. The methodology is tested by three case studies on Cu and Pb in Vienna, and PFOS in Switzerland. The findings have several implications for material, environmental and waste management: (i) As long as the indicator score stays below 100%, there are unacceptable substance flows to geogenic and/or anthropogenic sinks. The information gained while determining the new indicator is highly instrumental for developing strategies and measures to decrease these flows and to raise the score up to a maximum value of 100%. (ii) The study shows the important part waste management (incineration and landfilling) plays as a relevant and necessary sink for anthropogenic material flows. (iii) For the three substances taken into account by the case studies, there are fractions (roughly 1 to 10 %) that flow to inappropriate sinks. Still, these flows can pose an environmental problem and should be further investigated

5. Outlook

The article starts with a statement, highlighting the constraints for anthropogenic carbon flows not only at the supply but also at the disposal side. In the case of carbon, the United Nations Framework Convention on Climate Change (UNFCCC) aims at managing the carbon content of the atmosphere. So, nations periodically update and publish national inventories of anthropogenic carbon emissions and removals by sinks. In the future, the systematic provision of source/sink inventories beyond carbon facilitates informed decisions about substance flow management in a comprehensive manner. This supports a sustainable resource management strategy with respect to safe sinks as both, need and constraint for waste and emissions.

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Supplement information for:

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Summary

This supplement information includes datasets for the case study about lead in Vienna. It comprises the scope, the descriptive and normative assessment of lead flows. Figure 1 highlights the framework to calculate the indicator score and the content of the supplement information in grey colored boxes.

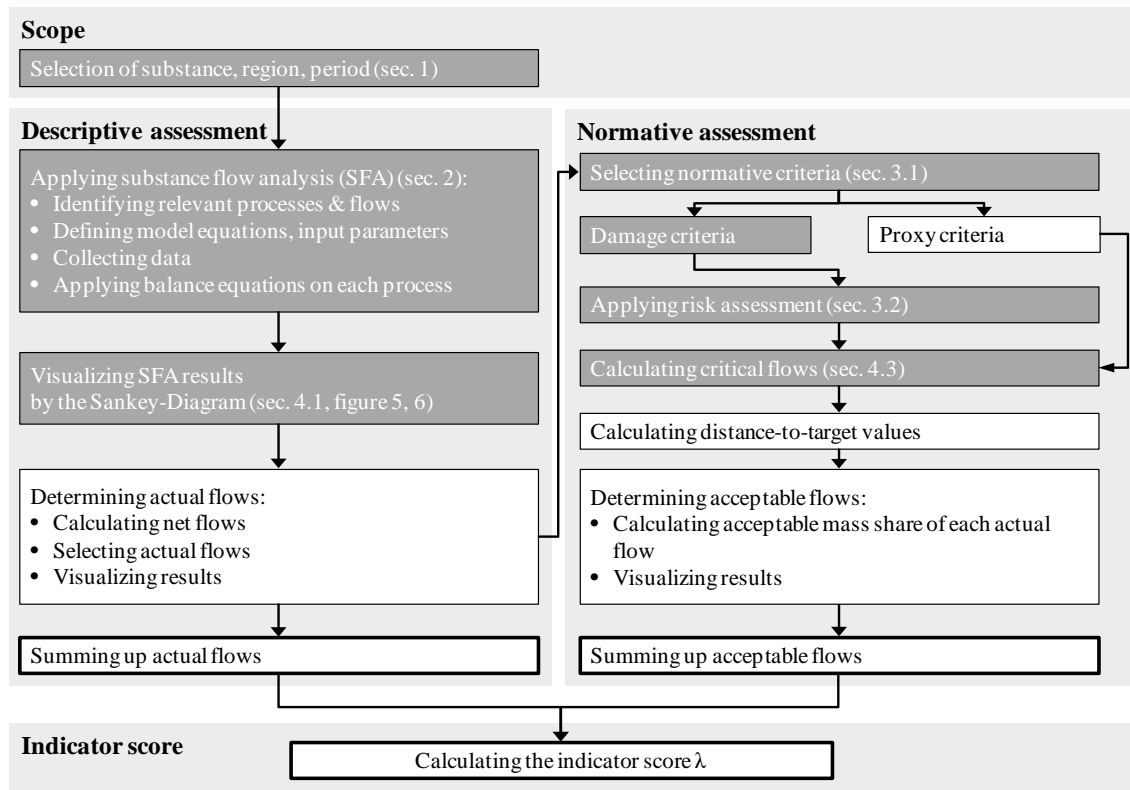


Figure 1: Framework to calculate the indicator score. Tasks documented within the supplement information are colored grey, including the section numbers. Tasks documented within the supplement information are dark grey colored.

1 Scope

Lead is selected as substance of interest. The system boundary in space is set with the administrative city limits. The system boundary in time is set with the year 2008.

2 Descriptive assessment

In the sections that follow, the calculation of actual lead flows is documented. First, the model development includes the identification of processes and flows. Second, the model equations are defined. Third, the flows are balanced using the software STAN.

2.1. Model development

The aim is to present a generic Pb flow model that allows the visualization of actual flows. This requires an understanding of the key Pb flows in the city of Vienna. The determination of processes and flows is based on previous studies, literature investigations, reports by local municipalities, and expert interviews. The generic Pb flow model can be found in Figure 2 and Figure 3. The process description is in accordance with Kral et al. (2013) and can be found in Table 1.

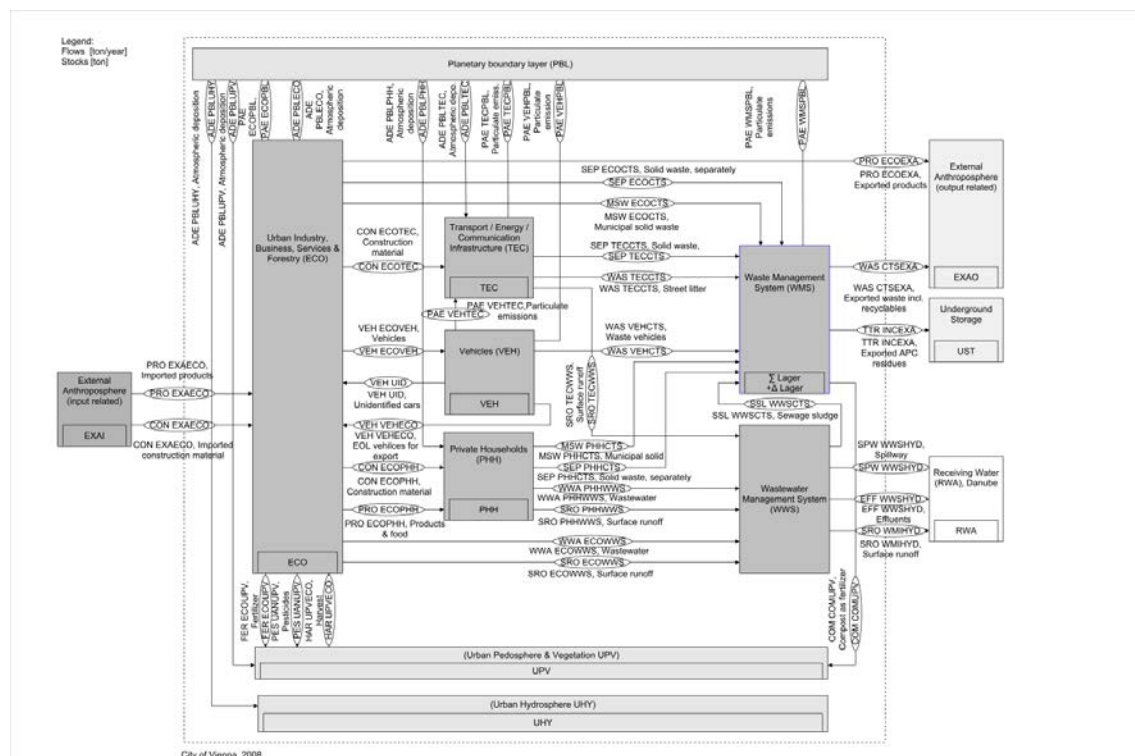


Figure 2: Generic lead flow model on top level. It covers 9 city internal processes of which 6 represent mainly anthropogenic activities (dark grey boxes) and 3 stand for environmental media (light grey boxes). Exterior processes are splitted in the supply and export as well as receiving waters in the hinterland. Regarding nomenclature, the flow acronyms refer to the type of flow (first three letters), to the source process (second three letters and to the sink process (last three letters). The stock acronyms refer to the type of stock only.

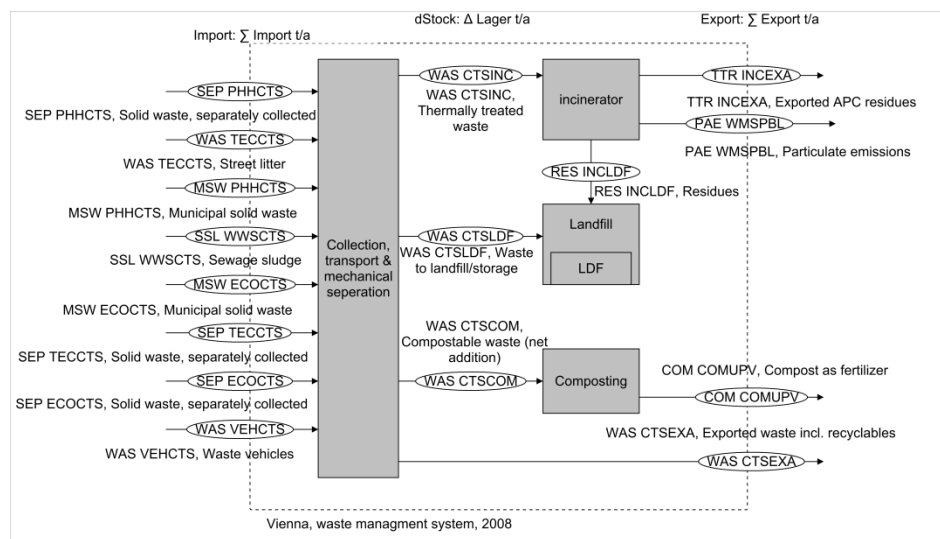


Figure 3: Generic lead flow model on 2nd level. It disaggregates the “Waste Management System”.

Table 1: Process characterization.

Process name	Characterization
External anthroposphere	Stands for the anthropogenic hinterland of the city. It delivers products and construction material to the city and receives exported products, waste and recyclables.
Industry, business, services and forestry	Covers economic activities as well as related buildings. Economic activities refer to the trade of goods, material processing and distribution for final consumption. The buildings are addressed for stock calculation including construction material and installations.
Transport, energy, and communication infrastructure	Covers immobile infrastructure and corresponding lead stock in transport networks, power grids and telecommunication networks.
Vehicles	Covers the mobile lead stocks such in cars, lorries, bikes, busses, trams, and trains.
Private Households	Covers anthropogenic activities of daily life and related buildings. Anthropogenic activities refer to residing, nourishing, cleaning, and communication. Related buildings like flats and houses are used for stock calculation of construction material and installations.
Waste Management System	Covers the collection, treatment and disposal of solid waste. The process is disaggregated which gives further insights into fluxes in view of incineration, composting and landfilling.
Waste Water Management System	Covers the collection and treatment of waste water. Material stocks are not taken into account.
Underground storage	External salt mines out of use act as final storage for hazardous residues from incineration.
Planetary boundary layer	Stands for the lowest part of the atmosphere that is influenced by its contact with the earth surface, usually several hundred meters high.
Urban pedosphere and vegetation	Consisting of urban soil and vegetation in parks, green areas and agricultural fields.
Urban hydrosphere	Urban water bodies, mainly rivers, groundwater, ponds, and small lakes.
Receiving waters	The hydrosphere that takes up both, waste water treatment effluents and combined sewer overflow from the city, such as the river Danube.

2.2 Model equations and data acquisition

This section provides a compilation of lead flow calculations. Table 2 includes the model equations and the flow results, Table 3 includes all input parameters.

Table 2: Model Equations and flow results

N°	Flow Acronym	Description	Equation	Mean Value [t/a]	Standard deviation [t/a]
01	ADE PBLECO	Atmospheric Deposition ECO	$(Dep_wet_dry - Dep_wet) * ADECO$	0,17	0,08
02	ADE PBLPHH	Atmospheric Deposition PHH	$(Dep_wet_dry - Dep_wet) * ABBH$	0,39	0,19
03	ADE PBLTEC	Atmospheric Deposition TEC	$(Dep_wet_dry - Dep_wet) * ATEC$	0,23	0,11
04	ADE PBLUHY	Atmospheric Deposition UHY	$(Dep_wet_dry - Dep_wet) * AUHY$	0,07	0,04
05	ADE PBLUPV	Atmospheric Deposition UPV	$(Dep_wet_dry - Dep_wet) * AUUV$	0,73	0,35
06	COM ANTUPV	Compost as fertilizer	$m_{comp} * W_{comp} * C_{comp}$	0,97	1,08
07	EFF WWSHYD	Effluents	$m_{nb_efflow} * TC_{CWTP}$	0,05	0,01
08	MSW ECOCTS	Municipal solid waste	$(m_{MSW_total} - m_{MSW_PHH}) * C_{MSW}$	70,64	16,39
09	MSW PHHCTS	Municipal solid waste	$C_{PHH} * m_{MSW_PHH} * C_{MSW}$	66,59	15,45
10	PAE ECOECPBL	Particulate emissions	Calculated based on mass balance principle	1,13	0,41
11	PAE VEHPBL	Particulate emission	$LDV + HDV$	0,40	0,12
12	PAE VEHTEC	Low Duty Vehicles (LDV) High Duty Vehicles (HDV)	$LDV = m_{LDV_LDV} * (C_{LDV_brake} * C_{brake} + C_{LDV_tire} * C_{tire}) * TC_{VEH}$ $HDV = m_{LDV_HDV} * (C_{HDV_brake} * C_{brake} + C_{HDV_tire} * C_{tire}) * TC_{VEH}$	0,23	0,05
13	PAE WMSPBL	Particulate emissions	$LDV = m_{LDV_LDV} * (C_{LDV_brake} * C_{brake} + C_{LDV_tire} * C_{tire}) * TC_{VEH}$ $LDV + HDV$	0,06	0,01
14	PRO ECOEXA	Exported products	$m_{inwaste} * C_{waste}$	524,38	203,83
15	PRO ECOPHH	End-of-life vehicles for export (VEH VEHECO) Unidentified cars (VEH UID) Lead and commodities out of lead (LC _{exp}) Products & food	$VEH\ VEHECO = car_{usage_exp} * m_{battery} * Proxy_{car}$ $VEH\ UID = (car_{usage} + car_{mp_reuse} - car_{usage} - car_{dereg}) * m_{battery} * Proxy_{car}$ $LC_{exp} = m_{goods_exp} * C_{goods}$ $EEA_{A2HH} = m_{EEA_PHH_ECO} * C_{EEA}$ $EEA_{A2HH} = m_{EEA_PHH_ECO} * C_{EEA} * Proxy_{exp} * Proxy_{area}$ $BUI = (m_{goods_imp} - m_{goods_exp}) * C_{goods} * Proxy_{BUI}$ $FO = m_{food} * C_{food}$	203,33	91,81
16	PRO EXAEEO	Building material for roof and facades (BUI) Food (FO) Imported products Batteries (BAT) = VEH ECOVEH Electric & electronic appliances (EEA _{total}) Lead and commodities out of lead (LC _{imp}) Energy carriers (EC) Food (FO)	$BAT = EEA + LC + EC + FO$ $BAT = (car_{usage} * Proxy_{car} + car_{stock} / lifetime_{battery}) * m_{battery}$ $EEA_{total} = (m_{EEA_PHH_ECO} + m_{EEA_ECO}) * C_{EEA} * Proxy_{exp}$ $LC_{imp} = m_{goods_imp} * C_{goods}$ $EC = d_{usage} * C_{usage} * C_{usage}$ $FO = m_{food} * C_{food}$	2179,76	313,74
17	RES INCLDF	Bottom ash	$m_{MSW_total} * C_{waste} * TC_{inc}$	94,69	21,98
18	SEP ECOCTS	Solid waste, separate collected Excavated soil (EXS) Demolition waste (DEM) PVC products (PVC) Lead scrap (SCR) Concrete waste (CON) Waste electronics and equipment (WEE) Lead mud (MUD)	$EXS + DEM$ $EXS = m_{soil} * C_{soil} * RC_{soil} * Proxy_{soil}$ $DEM = m_{dem} * C_{dem} * RC_{dem} * Proxy_{dem}$ $PVC = (m_{window} * C_{window} + m_{figus} * C_{figus} + m_{tables} * C_{tables}) * Proxy_{pvc}$ $SCR = m_{scrap} * C_{scrap} * (1 - Proxy_{area})$ $CON = m_{con} * C_{con} * (1 - Proxy_{area})$ $WEE = m_{wef} * C_{wef} * (1 - Proxy_{area})$ $MUD = m_{mud} * C_{mud}$	170,90	45,07
19	SEP PHHCTS	Solid waste, separate collected Excavated soil (EXS) Demolition waste (DEM) PVC products (PVC)	$EXS + DEM$ $EXS = m_{soil} * C_{soil} * RC_{soil} * Proxy_{soil}$ $DEM = m_{dem} * C_{dem} * RC_{dem} * Proxy_{dem}$ $PVC = (m_{window} * C_{window} + m_{figus} * C_{figus} + m_{tables} * C_{tables}) * Proxy_{pvc}$	322,61	97,60

Table 3: List of input parameters (non bold=single values, bold=matrices)

N°	Flow Acronym	Description	Equation	Mean Value [t/a]	Standard deviation [t/a]
20	SEP TECCTS	Lead scrap (SCR) Concrete waste (CON) Waste electronics and equipment (WEE) Solid waste, separate collected Excavated soil (EXS) Demolition waste (DEM)	$SCR = m_{scr} * C_{comp} * Proxy_{area}$ $CON = m_{con} * C_{con} * Proxy_{area}$ $WEE = m_{wve} * C_{wve} * Proxy_{area}$ $EXS = m_{soil} * C_{soil} * RC_{soil} * Proxy_{soil}$ $DEM = m_{dem} * C_{dem} * RC_{dem} * Proxy_{dem}$ $(SRO\ TECWWS + SRO\ PHHWWS + SRO\ ECOWWS) * Ratio_{SST} * Ratio_{overflow}$	53,36	18,57
21	SPW	Spillway	$(Dep\ wet\ dry - Dep\ wet) * A_{sc0}$	0,40	0,04
22	SRO ECOWWS	Surface runoff (ADE PBLECO)	$(Dep\ wet\ dry - Dep\ wet) * A_{sfr}$	0,39	0,19
23	SRO PHHWWS	Surface runoff (ADE PBLPHH)	$ADE\ PBLTEC = (Dep\ wet\ dry - Dep\ wet) * A_{TEC}$	0,23	0,11
24	SRO TECWWS	Surface runoff Atmospheric depositions (ADE PBLTEC) Particulate emissions Low Duty Vehicles (LDV) Particulate emissions High Duty Vehicles (HDV)	$LDV = m_{LDV} * LDV * (C_{LDV_brake} * C_{brake} + C_{LDV_tire} * C_{tire}) * IC_{VEH}$ $HDV = m_{HDV} * HDV * (C_{HDV_brake} * C_{brake} + C_{HDV_tire} * C_{tire}) * IC_{VEH}$ $WAS\ TECCTS = (ADE\ PBLTEC + LDV + HDV) * Ratio_{particulate}$ $(SRO\ ECOWWS + SRO\ PHHWWS + SRO\ TECWWS) * (1 - Ratio_{SST})$	0,42	0,06
25	SRO WMHYD	Surface runoff	$m_{pbl_allow} * IC_{WWRP}$	0,19	0,02
26	SSL WWSCTS	Sewage sludge	$m_{pbl_total} * C_{WWS} * IC_{RC}$	3,75	0,87
27	TTR INCXA	Exported APC residues	$(C_{org} * Proxy_{org} + C_{carbo} / lifetime_{battery}) * m_{battery}$	43,91	9,88
28	VEH ECOVEH	Vehicles	$(C_{deng} + C_{deng_resid} - C_{deng} - C_{deng_em}) * m_{battery} * Proxy_{car}$	1804,65	291,13
29	VEH UID	Unidentified cars	$C_{deng_exp} * m_{battery} * Proxy_{car}$	361,52	190,24
30	VEH VEHECO	EOL vehicles for export	$m_{comp} * W_{comp} * C_{comp}$	74,84	39,38
31	WAS CTSCOM	Compostable waste (net addition)	$EXS + DEM + CON +$	0,97	1,08
32	WAS CTSEXA	Exported waste incl. recyclables	$EXS = m_{soil} * C_{soil} * (1 - RC_{soil})$ $DEM = m_{dem} * C_{dem} * (1 - RC_{dem})$ $CON = m_{con} * C_{con}$	1698,12	1369,46
33	WAS CTSINC	Concrete waste (CON)	$WAS\ VEHCCTS = C_{deng} * m_{battery} + C_{carbo} / lifetime_{battery} * m_{battery}$	137,23	22,53
34	WAS CTSLDF	Waste vehicles (WAS VEHCCTS) Lead scrap (SCR) Thermal treated waste Waste to landfill/storage	$SCR = m_{scr} * C_{scr}$ $m_{MSW_total} * C_{MSW}$ $EXS = m_{soil} * C_{soil} * (1 - RC_{soil})$ $DEM = m_{dem} * C_{dem} * (1 - RC_{dem})$	49,14	20,41
35	WAS TECCTS	Excavated soil (EXS)	See line 24	0,05	0,01
36	WAS VEHCCTS	Demolition waste (DEM) Concrete waste (CON) Waste vehicles (WAS VEHCCTS)	$SRE + MAI$	1347,74	1365,32
37	WWA ECOWWS	Waste vehicles Batteries from shredded cars (SRE) Exported batteries due to maintenance (MAI)	$SRE = C_{deng} * m_{battery}$ $MAI = C_{deng} / lifetime_{battery} * m_{battery}$	3,37	0,88
38	WWA PHHWWS	Wastewater (unknown sources) Wastewater	$m_{pbl_allow} + SPW\ WWSHYD - WWA\ PHHWWS - (SRO\ TECWWS + SRO\ PHHWWS + SRO\ ECOWWS) * Ratio_{SST}$ $m_{water} * C_{water}$	0,32	0,06

Input Parameter	Description of data	Unit	Value	Uncertainty level	Reference
CapAUT	Inhabitants Austria	#	8,347,341	1	(Statistik Austria, 2012)
CapVIE	Inhabitants Vienna	#	1,674,909	1	(Lebhart, 2010)
Dep_wet_dry	Deposition rate (wet & dry)	g Pb/(ha*yr)	44.60	3	(Spiegel, 2003)
Dep_dry	Deposition rate (wet)	g Pb/(ha*yr)	7.20	3	(Kalina et al., 2000)
A	Total City Area	ha	41,487	1	(Lebhart, 2010)
AURH	Urban Hydrosphere (Open Water Bodies)	ha	1,933	1	(Lebhart, 2010)
ATEC	Traffic Surface	ha	5,981	1	(Lebhart, 2010)
AHEH	Residential Area	ha	10,267	1	(Lebhart, 2010)
AECO	Public Facilities and production areas	ha	4,381	1	(Lebhart, 2010)
AUPV	Green Space Area	ha	18,925	1	(Lebhart, 2010)
PROXY _{exp}	Ratio of inhabitants Vienna/Austria	-	0.201	-	-
PROXY _{area}	Area Ratio of residential land use in contrast to total built area = A _{HEH} / (A _{HEH} + A _{ECO})	-	0.701	-	-
IMEA_PHH_ECO	National sale of electric & electronic appliances (household appliances, consumed in PHH and ECO)	kg	Σ=32,436,269	2	(EAK, 2009)
IMEA_ECO	National sale of electric & electronic appliances (commercial appliances)	kg	Σ=1,624,894	2	(EAK, 2009)
CEEA	Average lead concentration in electric & electronic appliances	%	0-1.6	3	(EMPA, 2009)
m _{battery}	Lead content per car battery	kg Pb/#	13	3	assumption
m _{food}	Food imports to Vienna	t	1,236	1	(Magistratsabteilung 05, 2011)
C _{food}	Lead concentration in different food types	mg Pb/kg food	0.013-0.263	2	(USDA, 2011)
m _{goods_imp}	Imported "Lead and commodities out of lead"	t/yr	136	3	(Magistratsabteilung 05, 2011)
m _{goods_exp}	Exported "Lead and commodities out of lead"	t/yr	98	3	(Magistratsabteilung 05, 2011)
PROXY _{BUT}	Amount of lead that is consumed the building sector PHH in relation to the net difference of imported and exported "Lead and commodities out of lead"	%	15	3	(Forum Nachhaltiges Bauen, 2013)
E _{goods}	Lead concentration for goods within the category "Lead and commodities out of lead"	%	89.9	3	(Raisinger et al., 2009)
d _{energy}	Final energy demand in Vienna, categorized into various energy carriers.	TJ	Σ=1,259,22	1	
e _{energy}	Colorific values of various energy carriers	TJ/t	0.015-0.063	-	
Energy	Lead concentration in energy carriers	mg/kg	293,697	-	(Statistik Austria, 2009b)
car _{reg}	New registered cars in Austria	#	993,354	-	Calculated based on mass balance principle
car _{dereg}	Deregistered Cars in Austria	#	28,696	-	(Wirtschaftskammer Österreich, 2012)
car _{imp_road}	Imported second hand cars in Austria	#	738,690	-	(Statistik Austria, 2009a)
car _{reg}	Re-Registered Cars in Austria	#	37,629	-	(Wirtschaftskammer Österreich, 2012)
car _{dereg_exp}	Deregistered and exported cars in Austria	#	63,975	-	(BMLFUW, 2011a)
car _{dereg}	Shredded cars in Austria	#	0.1534	-	(Statistik Austria, 2008)
PROXY _{car}	Proxy representing Viennese and Austrian car ownership	#	657,192	1	assumption
car _{stock}	Number of registered cars in Vienna	Years	7	3	(Wiener Umweltschutzabteilung MA22, 2011)
lifetime _{battery}	Lifetime of a battery	t/yr	579,888	2	(BMLFUW, 2011a)
m _{MSW_total}	Mixed waste from households and similar institutions	kg/cap/yr	168	-	(Morf and Taverna, 2006; Taverna et al., 2011)
m _{MSW_PHH}	Mixed waste from private households, national average	mg/kg	geomet(200-280)	2	(Wiener Umweltschutzabteilung MA22, 2011)
MSW	Lead concentration in mixed waste	t/yr	2,978,726.89	1	(Wiener Umweltschutzabteilung MA22, 2011)
m _{soil}	Amount of excavated soil	mg/kg	16.1	3	(Woitschlaeger et al., 2000)
E _{soil}	Lead concentration in excavated soil	%	64	3	(BMLFUW, 2011b)
RC _{soil}	Recycling quote for excavated soil	-	1/3	2	assumption
PROXY _{soil}	Allocation proxy for soil to PHH, ECO and TEC	-			

Input Parameter	Description of data	Unit	Value	Uncertainty level	Reference
mDem	Amount of demolition waste	t/yr	1,090,976	1	(Wiener Umweltschutzabteilung MA22, 2011)
RCdem	Lead concentration in demolition waste	mg/kg	85	3	(König, 2006)
RCdem	Recycling quote for demolition waste	%	65.6	3	(BMLFUW, 2011a)
Proxydem	Allocation proxy for demolition waste to PHH, ECO and TEC	-	1/3	2	Assumption
mWindow	Amount of window frames	t/yr	geomittel(2,000;4000)	1	(BMLFUW, 2002a)
Cwindow	Lead concentration in window frames	%	0.7	2	(AgPU, 2004)
mPipes	Amount of pipes	t/yr	geomittel(1,000;1000)	1	(BMLFUW, 2002a)
CPipes	Lead concentration in pipes	%	2.7	2	(AgPU, 2004)
mCables	Amount of cables	t/yr	geomittel(4,000;8000)	1	(BMLFUW, 2002a)
Ccables	Lead concentration in cables	%	2.0	2	(AgPU, 2004)
ProxyPVC	Allocation proxy for PVC waste fractions to PHH and ECO	-	various	3	(BMLFUW, 2002a)
mScrap	Amount of lead scrap	t/yr	263.57	2	(Wiener Umweltschutzabteilung MA22, 2011)
Cscrap	Lead concentration of lead scrap	%	90	3	(König, 2006)
mCon	Amount of concrete waste	t/yr	432,294.65	2	(Wiener Umweltschutzabteilung MA22, 2011)
Ccon	Lead concentration in concrete waste	g/t	50	3	Assumed to be 50% of maximal listed Pb concentration (VDZ, 1996)
mWEEE	Collected WEEE in PHH of Vienna	t/a	9,044	1	(EAK, 2009)
mil_LDV	Mileage of low duty vehicles in Vienna	veh-km	5,694,000,000	1	(Holzapfel and Riedel, 2011)
mil_HDV	Mileage of high duty vehicles in Vienna	veh-km	803,000,000	1	(Holzapfel and Riedel, 2011)
emDV_brake	Total wear rate of brake lining in low duty vehicles	mg /veh-km	12.5	2	(Winther and Slento, 2010)
emDV_brake	Total wear rate of brake lining in high duty vehicles	mg /veh-km	54.7	2	(Winther and Slento, 2010)
Qbrake_LDV	Lead content of brake linings in low duty vehicles	mg Pb/kg brake pad	11,381 mg	2	(Westerlund, 2001)
Qbrake_HDV	Lead content of brake linings in high duty vehicles	mg Pb/kg brake pad	geomittel(158,656)	3,04	(Westerlund, 2001)
emDV_tire	Total wear rate of tires in low duty vehicles	mg/veh-km	136.38	2	(Luhana et al., 2004; Winther and Slento, 2010)
emDV_tire	Total wear rate of tires in high duty vehicles	mg/veh-km	136.38	2	(Luhana et al., 2004; Winther and Slento, 2010)
Ctire_LDV	Lead content of tires in low duty vehicles	mg/kg	80.5	2	(Luhana et al., 2004; Winther and Slento, 2010)
Ctire_HDV	Lead content of tires in high duty vehicles	mg/kg	80.5	2	(Luhana et al., 2004; Winther and Slento, 2010)
TCMOV	Transfer coefficients describing the ratio of particulate emissions entering individual sinks	%	20; 31; 49	2	(Hulskotte et al., 2006)
mLead	Amount of lead mud in Vienna	t/yr	0.18	1	(Wiener Umweltschutzabteilung MA22, 2011)
Cmud	Lead concentration of lead mud	%	50	3	assumption
mPb_inflow	Lead inflow to Waste Water Treatment Plant	t Pb/yr	4.09	2	(Kroiss et al., 2008)
TCWWTP	Transfer coefficients for lead in the Viennese waste water treatment plant	%	1.3; 7.0; 91.7	2	(Kroiss et al., 2008)
TCRC	Transfer coefficients for lead in the Viennese incinerator plants	%	1; 31; 69	2	(Favera et al., 2011)
mWater	Revenue water	m ³ /yr	122,773,000	1	(Tomenendal, 2011)
Cwater	Lead concentration in Viennese drinking water	µg/l	2.6	2	(Haider et al., 2002)
mLeadest	Amount of collected street litter	t/yr	39,495	1	(Wiener Umweltschutzabteilung MA22, 2011)
mComp	Applied compost on land	t/yr	33,000	1	(Weinmar, 2011)
mComp	Water content of compost	%	41	3	(Umwelbundesamt, 2000)
Ccomp	Lead concentration in compost	mg Pb/kg	71.6	3	(Umwelbundesamt, 2000)
RatioOverflow	Percentage of surface runoff ending up as stormwater overflow	%	50	-	(Fenz, 1999)
RaftOBS	Ratio surface water collected by mixed sewer system based on network lengths	%	81	-	(I C Consultants Ltd, 2001; Lampert et al., 1997; Lehmann, 2011)

Input Parameter	Description of data	Unit	Value	Uncertainty level	Reference
m_{waste}	Mixed waste flows into municipal solid waste incinerators in Vienna	t/yr	$\Sigma=508,254$	1	(Kronberger, 2011)
e_{waste}	Lead emission factors for municipal solid waste incinerators in Vienna	$g Pb/t waste$	0.220; 0.054	2	(BMLFUW, 2002b)
$Rat_{CO_{2waste}}$	Ration of Pb in street litter in relation to Pb deposition on traffic surfaces	%	10	-	assumption

2.3 Balance equations

The quantified flows from Table 2 do not fulfill the mass balance on each process. Consequently, we balanced the flows by error propagation and data reconciliation. This step was carried out by the software STAN (Cencic, 2012). The balanced flows are used to determine actual flows into sinks (Table 5).

Table 4: Unbalanced versus balanced flows in t/yr.

Acronym	Flow name	Mass flow (unbalanced)	± Mass flow (unbalanced)	Mass flow (balanced)	± Mass flow (balanced)
ADE PBLECO	Atmospheric deposition	0,17	0,03	0,17	0,03
ADE PBLPHH	Atmospheric deposition	0,39	0,07	0,39	0,07
ADE PBLTEC	Atmospheric deposition	0,23	0,04	0,23	0,04
ADE PBLUHY	Atmospheric deposition	0,07	0,01	0,07	0,01
ADE PBLUPV	Atmospheric deposition	0,73	0,12	0,73	0,10
COM COMUPV	Compost as fertilizer	0,97	1,08	0,97	0,76
CON ECOPHH	Construction material	0,00	0,00	0,00	0,00
CON ECOTEC	Construction material	0,00	0,00	0,00	0,00
CON EXAECO	Imported construction material	0,00	0,00	0,00	0,00
EFF WWSHYD	Effluents	0,05	0,01	0,05	0,01
FER ECOUPV	Fertilizer	0,00	0,00	0,00	0,00
HAR UPVECO	Harvest	0,00	0,00	0,00	0,00
MSW ECOCTS	Municipal solid waste	70,64	16,39	70,63	16,39
MSW PHHCTS	Municipal solid waste	66,59	15,45	66,58	15,45
PAE ECOPBL	Particulate emissions	1,13	0,08	1,13	0,07
PAE TECPBL	Particulate emissions	0,00	0,00	0,00	0,00
PAE VEHPBL	Particulate emission	0,40	0,12	0,40	0,10
PAE VEHTEC	Particulate emissions	0,24	0,05	0,24	0,05
PAE WMSPBL	Particulate emissions	0,06	0,01	0,06	0,01
PES UANUPV	Pesticides	0,00	0,00	0,00	0,00
PRO ECOEXA	Exported products	524,38	203,83	524,38	203,83
PRO ECOPHH	Products & food	203,33	91,81	203,33	91,81
PRO EXAECO	Imported products	2179,76	313,74	2179,76	313,74
RES INCLDF	Residues	94,69	21,98	94,06	16,39
SEP ECOCTS	Solid waste, separate collected	170,90	45,07	170,82	45,06
SEP PHHCTS	Solid waste, separate collected	322,61	97,60	322,23	97,48
SEP TECCTS	Solid waste, separate collected	53,36	18,57	53,35	18,57
SPW WWSHYD	Spillway	0,40	0,04	0,40	0,04
SRO ECOWWS	Surface runoff	0,39	0,07	0,39	0,07
SRO PHHWWS	Surface runoff	0,23	0,04	0,23	0,04
SRO TECWWS	Surface runoff	0,42	0,06	0,42	0,06
SRO WMIHYD	Surface runoff	0,19	0,02	0,19	0,02
SSL WWSCTS	Sewage sludge	3,75	0,87	3,92	0,62
TTR INCEXA	Exported APC residues	43,91	9,88	43,78	9,43
VEH ECOVEH	Vehicles	1804,65	291,13	1804,65	291,13
VEH UID	Unidentified cars	361,52	190,24	361,52	190,24
VEH VEHECO	EOL vehicles for export	74,84	39,38	74,84	39,38
WAS CTSCOM	Compostable waste (net addition)	0,97	1,08	0,97	0,76
WAS CTSEXA	Exported waste incl. recyclables	1698,12	1369,46	1772,92	968,58
WAS CTSINC	Thermal treated waste	137,23	22,53	137,91	16,46
WAS CTSLDF	Waste to landfill/storage	49,14	20,41	49,16	20,41
WAS TECCTS	Street litter	0,05	0,01	0,05	0,01
WAS VEHCTS	Waste vehicles	1347,74	1365,32	1273,39	968,56
WWA ECOWWS	Wastewater	3,37	0,88	3,20	0,62
WWA PHHWWS	Wastewater	0,32	0,06	0,32	0,06

3 Normative assessment

The sections that follow include the selection of the criteria, the methodology to calculate the criteria, as well the data needed to apply the methodology.

3.1 Selection of criteria

To assess the actual flows into natural sinks, two impact indices are selected in view of human health. The “risk level” stands for the carcinogenic risk and the “hazard-index” stands for the non-carcinogenic risk.

3.2 Risk assessment

The sections that follow include a brief overview of the risk assessment approach and details about the background data.

3.2.1 Approach

To compute the two impact indices, the software tool CalTOX 4.0 has been applied. The model has been developed by Lawrence Berkeley National Laboratory. CalTOX is designed to run in Microsoft Excel® and is available online (State of California, 2007). The CalTOX model structure is visualized in Figure 5. The approach covers three steps in total, though methodological details can be found in the user manual (University of California, 1994):

- First, multimedia transport simulation is used to compute the fate of lead in the environment. The model turns out the result of substance flows and stocks in each compartment when the system reaches an equilibrium state.
- Second, exposure assessment defines the relation between the environmental medium and the exposure medium. The result is the “average daily dose rate” for each exposure route.
- Third, the impact indices were calculated with the dose-response relationship of lead.

Figure 4: CalTOX model structure (adopted from State of California, 1993; State of California, 2007). It consist of (a) multimedia transport simulation, (b) exposure assessment, and (c) the risk algorithm that yields two impact indices.

3.2.2 Data acquisition

In general, CalTOX requires four types of input data. The points that follow include the abbreviation from the default values, provided by the CalTOX spreadsheet.

1. Emission flows in mol/day. These data are provided by the SFA model. Therefore, 1,59 t Pb/yr enter air, 0,97 t Pb/yr enter soil and 0,64 t Pb enter water (sec. 2, Table 17).

Table 5: Anthropogenic flows for CalTOX model inputs

Flow short name	Flow name	Sink	Actual flow [ton/year]	Actual flow into sink [ton/year]	Actual flow into sink* [mol/day]
PAE ECOPBL	Particulate emissions	Air	1,12		
PAE VEHBPBL	Particulate emission	Air	0,41	1.59	21.02
PAE WMSPBL	Particulate emissions	Air	0,06		
COM COMUPV	Compost as fertilizer	Soil	0,97	0.97	12.81
EFF WWSHYD	Effluents	Water	0,05		
SPW WWSHYD	Spillway	Water	0,40	0.64	8.50
SRO WMIHYD	Surface runoff	Water	0,19		

Note: *=Conversion factor from [ton/year] to [mol/day]: $10^6[\text{g/t}] * 1\text{mol}/207.21 [\text{g*t}] * 1 [\text{year}/365\text{days}]$

2. Chemical properties. In total 36 parameters are needed. Default values for Pb are provided in CalTOX and used to run the model.
3. 58 landscape properties. In total 58 parameters are needed. 11 out of 58 landscape parameters are adopted for Vienna (Table 18). 47 out of 58 parameters are default values, provided in CalTOX, and were used to run the model.
4. Exposure factors. In total 52 parameters are needed. 21 out of 52 exposure factors are adopted for Vienna (Table 19). 31 out of 52 exposure factors are default values, provided in CalTOX, and were used to run the model.

Table 6: Vienna's landscape properties in CalTOX

Landscape properties	Variable name	value
Contaminated area in m2	Area	414871000
Annual average precipitation (m/d)	rain	0.002
Land surface runoff (m/d)	runoff	0.0008
Atmospheric dust load (kg/m3)	rhob_a	2.39E-08
Ground-water recharge (m/d)	recharge	0.00003
Thickness of the ground soil layer (m)	d_g	0.01
Soil particle density (kg/m3)	rhos_s	1600
Water content in surface soil (vol fraction)	beta_g	0.15
Fraction of land area in surface water	f_arw	0.05
Ambient environmental temperature (K)	Temp	285
Yearly average wind speed (m/d)	v_w	319200

Table 7: Vienna's exposure properties in CalTOX

Human Exposure Factors	Variable name	value
Body weight (kg)	BW	74
Surface area (m ² /kg)	SAb	0.0254
Fluid Intake (L/kg-d)	Ifl	0.0208
Fruit and vegetable intake (kg/kg-d)	Ifv	0.0055
Grain intake (kg/kg-d)	Ig	0.0000
Milk intake (kg/kg-d)	Imk	0.0095
Meat intake (kg/kg-d)	Imt	0.0021
Egg intake (kg/kg-d)	Iegg	0.0004
Fish intake (kg/kg-d)	Ifsh	0.0002
Soil ingestion (kg/d)	Isl	0.0002
Fraction of water needs from ground water	fw_gw	0.0251
Fraction of water needs from surface water	fw_sw	0
Frcn frts & vgtbls that are exposed produce	fabv_grd_v	0.4
Fraction of fruits and vegetables local	flocal_v	0.364
Fraction of grains local	flocal_g	0.069
Fraction of milk local	flocal_mk	0
Fraction of meat local	flocal_mt	0
Fraction of eggs local	flocal_egg	0
Fraction of fish local	flocal_fsh	0
Exposure duration (years)	ED	30
Averaging time (days)	AT	28878.8

4 Results

The sections that follow include the Sankey-Diagram for actual flows, the actual risk based on the actual flows, and the critical flows in view of acceptable risks.

4.1 Sankey-Diagram

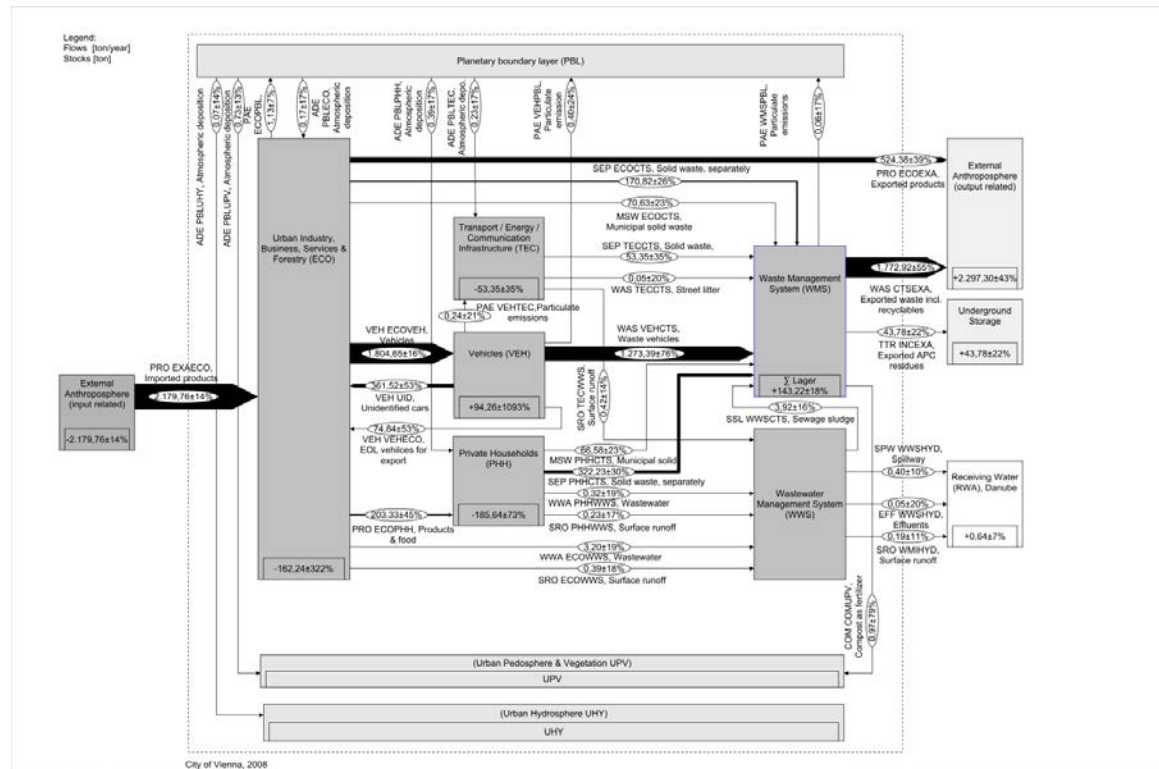


Figure 5: The Sankey diagram for lead in Vienna on an annual base for the year 2008. Flow rates and changes in stocks are given in mass/time, for stocks in mass. The flows are represented as Sankey arrows proportional to the flow rate; figures for stocks are given within the process boxes.

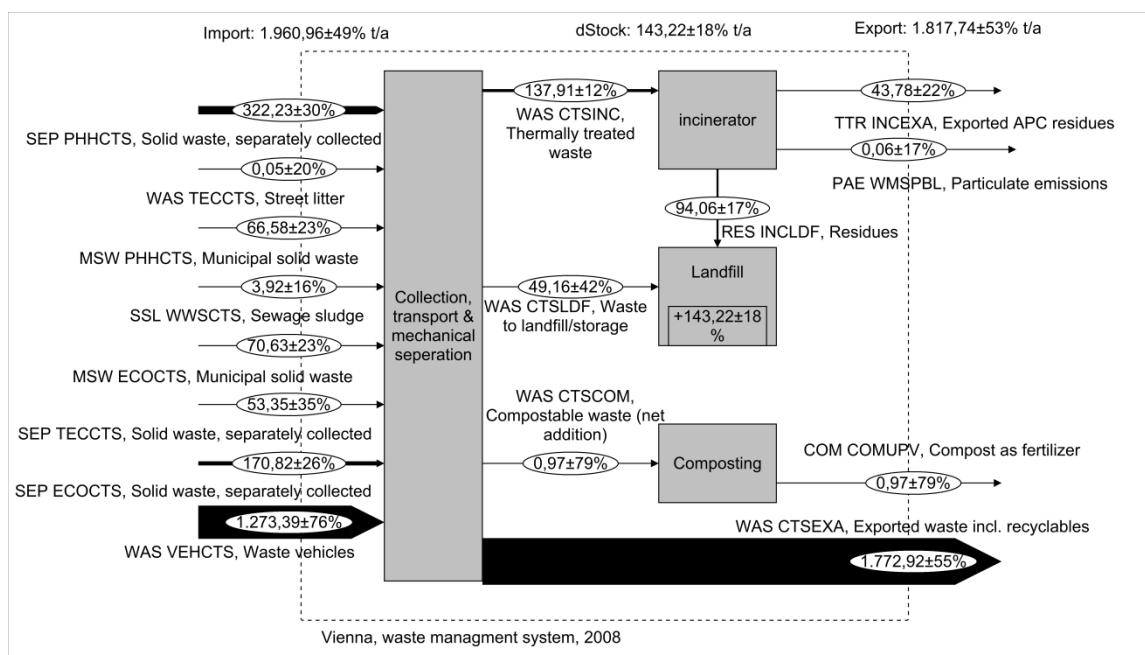


Figure 6: The Sankey diagram for lead in the Viennese waste management sector, on an annual base for the year 2008. Flow rates and changes in stocks are given in mass/time, for stocks in mass. The flows are represented as Sankey arrows proportional to the flow rate; figures for stocks are given within the process boxes.

4.2 Actual risks

The actual flows into natural sinks yield a risk level of $7.41 \text{ E-}8$ (Carcinogenic risk) and a hazard-index of 0.26 (non-carcinogenic risk). Table 20 and Table 21 shows that the risk level and the hazard-index are mainly driven by the ingestion of lead due to vegetables grown above the ground (exposed produce).

Table 8: The analysis of carcinogenic risk

Exposure route	Risk	Contribution rate
Inhalation	1.25 E-8	16.82%
Ingestion	6.15 E-8	83.01%
Water	1.34 E-9	1.81%
Exposed produce	6.01 E-8	81.03%
Unexposed produce	2.54 E-14	0.00%
Meat	0	0%
Milk	0	0%
Eggs	0	0%
Fish	0	0%
Soil	1.23 E-10	0.17%
Dermal	1.33 E-10	0.18%
Total Risk	7.41 E-8	100%

Table 9: The analysis of hazard-index

Exposure route	Hazard-index	Contribution rate
Inhalation	1.83E-02	6.98%
Ingestion	2.43E-01	92.82%
Water	5.30E-03	2.03%
Exposed produce	2.37E-01	90.61%
Unexposed produce	1.00E-07	0.00%
Meat	0	0%
Milk	0	0%
Eggs	0	0%
Fish	0	0%
Soil	4.87E-04	0.19%
Dermal	5.24E-04	0.20%
HQ	2.62E-01	100%

4.3 Critical flows

Scenario 4 yields the minimum ratio between the critical flow and the actual flow (Table 22 and Table 23).

Table 10: Critical flows in view of the risk level.

Scenario N°	Sink	Actual flow (tons/year)	Critical flow (tons/year)	Tolerable flow (tons/year)	Ratio critical / actual flow
1	Air	1,59	21,93	20,34	14
	Water	0,64	0,64	0,00	
	Soil	0,97	0,97	0,00	
2	Air	1,59	1,59	0,00	
	Water	0,64	461,35	460,74	717
	Soil	0,97	0,97	0,00	
3	Air	1,59	1,59	0,00	
	Water	0,64	0,64	0,00	
	Soil	0,97	2.647,11	2.646,14	2.733

Table 11: Critical flows in view of the hazard-index.

	Sink	Actual flow (tons/year)	Critical flow (tons/year)	Tolerable flow (tons/year)	Ratio critical / actual flow
4	Air	1.59	6.13	4.54	4
	Water	0.64	0.64	0.00	
	Soil	0.97	0.97	0.00	
5	Air	1.59	1.59	0.00	
	Water	0.64	90.76	90.15	141
	Soil	0.97	0.97	0.00	
6	Air	1.59	1.59	0.00	
	Water	0.64	0.64	0.00	
	Soil	0.97	605.05	604.08	625

References

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