Environmental Science & Technology
Critical Review
pubs.acs.org/est

Modeling Metal Stocks and Flows: A Review of Dynamic Material Flow Analysis Methods

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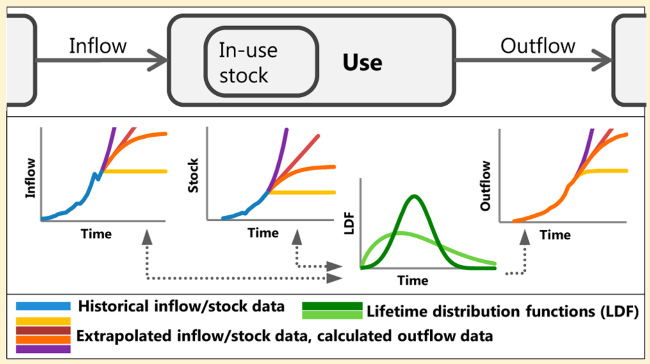
Supporting Information

ABSTRACT: Dynamic material flow analysis (MFA) is a frequently used method to assess past, present, and future stocks and flows of metals in the anthroposphere. Over the past fifteen years, dynamic MFA has contributed to increased knowledge about the quantities, qualities, and locations of metal-containing goods. This article presents a literature review of the methodologies applied in 60 dynamic MFAs of metals. The review is based on a standardized model description format, the ODD (overview, design concepts, details) protocol. We focus on giving a comprehensive overview of modeling approaches and structure them according to essential aspects, such as their treatment of material dissipation, spatial dimension of flows, or data uncertainty. The reviewed literature features similar basic modeling principles but very diverse extrapolation methods. Basic principles include the calculation of outflows of the in-use stock based on inflow or stock data and a lifetime distribution function. For extrapolating stocks and flows, authors apply constant, linear, exponential, and logistic models or approaches based on socioeconomic variables, such as regression models or the intensity-of-use hypothesis. The consideration and treatment of further aspects, such as dissipation, spatial distribution, and data uncertainty, vary significantly and highly depends on the objectives of each study.



INTRODUCTION

The industrial application of metals increased continually during the 20th century, with around 60 metallic elements in use today.[1] In particular, the use of metals, such as indium, platinum group metals, rare earth metals, or tantalum, which play a crucial role in many emerging technologies, has grown rapidly in recent years (e.g., refs 2 and 3). Increased consumption has led to an accumulation of significant stocks of metals in the anthroposphere, and the collection and recycling of metals from these secondary resources has become more and more important.[4] These activities rely on knowledge of anthropogenic material cycles regarding quantities, qualities, and locations of metal-containing goods that have accumulated in the past. Bulk metals (such as iron, copper, or aluminum) entering the anthroposphere remain largely concentrated, and dissipative losses to the environment are rather small.[5] Other metals, however, are often used at very low concentrations, which leads to sparsely distributed stocks and flows that can hardly be concentrated and recovered in current recycling systems.[3] Efforts to specifically recover these metals through recycling are in most cases only just beginning, the metals are thus often lost to recovered bulk materials or dissipated to the environment.

Many studies analyzing the material cycles of metals in the anthroposphere are based on material flow analysis (MFA) as introduced and defined, for example, by Baccini and Brunner.[6] In a critical review, Chen and Graedel[7] give an overview of the existing information on anthropogenic cycles, including those of more than 60 metals. The major engineering metals iron/steel, copper, lead, zinc, and aluminum, as well as silver and chromium, have been studied most often and their material cycles are thus the most well-understood. In recent years, some MFAs were also conducted for metals, such as antimony, cobalt, gold, platinum group metals (PMG), rare earth elements (REE), indium, tantalum, tin, and tungsten.[7] Most

Received: August 8, 2013
Revised: January 17, 2014
Accepted: January 17, 2014
Published: January 17, 2014

Table 1. Elements of the ODD (Overview, Design Concepts, Details) Protocol for MFA

overview	purpose	What is the purpose and general framework of the model?
	materials (goods, substances)	What materials (goods/substances) are included? Are materials further divided into material categories (and subcategories)?
	processes	What processes are included? Do they transform, transport, or store materials? Are processes further divided into process categories (and subcategories)?
	spatial and temporal scale and extent	What is the spatial and temporal scale and extent of the study?
	system overview	What is the structure of the system regarding processes, stocks, and flows?
design concepts	basic principles	Static or dynamic, top−down or bottom−up, retrospective or prospective?
	static or dynamic modeling approaches	How are stocks and flows modeled? What are the extrapolation methods for exogenous variables?
	dissipation	How does the model account for dissipation?
	spatial dimension	How does the model account for the spatial distribution of stocks and flows?
	uncertainty	How does the model account for data and model uncertainty?
details	initial condition	How is the initial state (e.g., the initial stocks and flows) of the model set?
	model input data	What data is used as input to the model?
	model output data	What data is generated as model output?
	evaluation	What methods (e.g., for data aggregation and visualization) are used to evaluate the results?
	detailed model description	What, in detail, is the formal description (e.g., equations) of the system and what are the algorithms (e.g., solution procedures) used for the calculations?
		What are exogenous and endogenous model variables? What are the model parameters, their dimensions, and reference values?

of these MFAs use static models with a time scale of one year, thus providing only snapshots in time. They offer some insights into the anthropogenic metabolisms of metals, but provide no information about the dynamics of resource use and resulting changes in stocks and flows. Estimations of past and future flows can provide insights on factors influencing resource use and early warnings of environmental problems, or they can support investment planning in infrastructures for mining, production, and waste management.[8] After Baccini and Bader[9] developed the methodology of dynamic MFAs, the first studies on metals were published in 1999 for copper[10] in the United States and for aluminum[11] in Germany. Since then, various methods to dynamically model past and future stocks and flows of metals, which provide information about the behavior of the system as a function of time, have become well-established. The existing dynamic metal flow models differ in terms of their modeling approach, their temporal scale, or the inclusion of processes, end-use sectors, or trade and loss flows, depending on the study's purpose and data availability. So far, the methodological approaches to model the dynamics of metals, or metal cycles, have not been standardized. This makes it difficult to compare studies and combine their results.[7]

In this article, we present a Critical Review of the methodologies applied in the literature on dynamic MFAs of metals. We focus on giving a comprehensive selection of modeling approaches that can be used as a basis for future studies on the dynamics of metal cycles. We further identify distinguishing aspects of MFAs, such as the treatment of material dissipation, the spatial dimension of flows, or data uncertainty. The review covers the published literature in English on dynamic MFAs of metals. One German reference[12] is included because it provides background data used by Bader et al.[13] Literature with only rudimentary or incomplete model descriptions is not included. We thus compile information from 60 studies published between 1999 and 2013 and covering a total of 34 metallic elements.

METHOD

The review is structured based on the standardized description ODD (overview, design concepts, details) protocol that was originally developed for the documentation of individual-based and agent-based models.[14,15] Although the studies we review use a fundamentally different type of models, ODD has proven to be useful for structuring them. The main objective of the ODD protocol is to provide a complete, understandable, and reproducible description of the models to make their complexity manageable for the human reader.[14,15] MFAs are in general less complex than agent-based models, we therefore simplified the ODD protocol slightly to adapt it to the field of MFA. The adapted structure is provided in Table 1. Each element of the protocol is further specified with one or more questions.

The protocol is grouped into three parts. The first part gives an overview of the study, including the purpose, the scope, the system boundaries, and the structure of the MFA. The second part describes the generic concepts and modeling approaches of the research. The third part provides the details necessary to ensure the reproducibility of the study.

MFA-specific terms in the ODD are used as defined by Brunner and Rechberger.[16] In the following, we will further clarify some terms: static versus dynamic MFA, top−down versus bottom−up approach to MFA, prospective versus retrospective MFA, endogenous versus exogenous model variable, and material dissipation.

An MFA is *static* if it describes a "snapshot" of a system in time. An MFA is *dynamic* if it describes the behavior of a system over a time interval.[7]

The material stock of a process can be measured by two different methods. The first method, usually referred to as the *top−down approach*, derives the stock from the net flow: the difference between inflows (consumption) and outflows (discard). The second method, the *bottom−up approach*, directly estimates the stock by summing up the material in question present within the system boundary at a certain time.[17] Most authors define stock as the in-use stock and do not include "hibernating" materials, that is, those that have been retired and remain somewhere in storage. Hibernating or obsolete stock is explicitly included only by Daigo et al.[18]

An MFA can be either *retrospective*, analyzing past stocks and flows based on historical data, or *prospective*, looking into the future using data extrapolation, or a combination of both approaches.

An *endogenous model variable* is a variable whose value is determined by one of the functional relationships in the model, for example, the outflow of a given product as waste, determined by the inflow and lifetime of that product. An *exogenous model variable* is an independent variable that affects endogenous model variables without being affected by any of them. It represents a quantity that exists outside the chosen system boundary. For simulation, an exogenous variable needs input data, for example, data of the inflow of a certain product or socioeconomic data such as time series of the gross domestic product (GDP).

According to Ayres,[19] who was one of the first to use the concept of *dissipation* associated with material flows,[5] "there are only two possible long-run fates for materials—dissipative loss and recycling or reuse." He argues that materials are recycled or reused if economically and technologically feasible, otherwise they are eventually dissipated. In Ayres et al.,[20] he later specifies four categories of metal stocks: long-lived goods in use, short-lived goods in use, landfill and identifiable mine waste dumps, and finally metals that have been irrecoverably dissipated into soil, groundwater, or surface water. In this definition, only the last category accounts for material dissipation.

RESULTS

Purpose. Most of the reviewed studies (43 of 60) aim at understanding the pathways of metals in the anthroposphere, the magnitudes of their stocks and flows, and how they evolve as a function of time. This involves quantifying and visualizing the dynamics of relevant stocks and flows of metals and their use in specific product groups or end-use sectors. Additional purposes include to specifically examine the recycling potential of metals, including recycling efficiency[21,22] and future recycling flows,[23−25] to evaluate future scenarios of resource availability,[20,26−28] to assess changes in environmental impacts related to changes in material flows,[29−31] and to compare different methodological approaches.[11,32−36]

Materials. The studies assessed cover 34 metallic elements as summarized in Table 2, with iron, aluminum, and copper being the most frequently investigated elements. Dynamic MFAs are still lacking for more than 30 metals.[7]

Table 2. Metallic Elements Covered in the Reviewed Literature

element (alloy)	covered in no. of studies
Fe/steel	17
Al	12
Cu	11
Pb	6
Zn	4
Cr, Ni	3
Cd, Ce, Dy, Eu, Gd, In, La, Nd, Pt, Pr, Sm, Te, Tm, Y	2
Ag, Co, Er, Hg, Ho, Lu, Pd, Rh, Se, Sn, Tb, W, Yb	1

Thirty-four studies consider metal use in some or all of the following end-use sector categories (sometimes disaggregated into subcategories): transportation, buildings and construction, infrastructure and telecommunication, machinery, electric appliances, and consumer goods, and containers and packaging.

Instead of end-use categories, 20 studies assess metal use in products. Among these, 13 studies cover the most relevant products containing the investigated metal and 7 studies cover the metal use only of individual products: CRT screens,[33] vehicles,[37,38] catalytic converters in automobiles,[31] photovoltaic systems,[23,27] and products containing indium tin oxide (ITO).[39] Three further studies include both end-use sectors and products,[24,34,40] and three studies do not categorize metal use.[20,41,42]

The metal content of stocks and flows is calculated either directly by computing metal stocks and flows from input data or indirectly by computing material stocks of end-use sectors or products and then calculating metal quantities based on the assumed metal share in an end-use sector or content in a product. In the indirect case, the metal share or content is usually considered time-variant.

Processes. The processes most commonly included in MFAs of metals cover the whole life cycle of a metal, from primary mining to raw material production to product manufacturing to use and finally waste management. In most of the models, the use phase is the only process that stores materials, while the other phases transform them without accumulating stocks. Potential stocks outside the use phase are neglected because they are assumed to be stationary over the smallest time interval considered, usually the period of one year (this assumption may be challenged by stocks of very valuable metals created for speculation). Additional processes such as landfill, environment, or other repositories are often included to illustrate the final sink of the assessed metals. Table S1 in the Supporting Information (SI) gives an overview of the processes covered by the studies.

Spatial and Temporal Scale and Extent. The spatial extent ranges from urban to global system boundaries, though most literature (38 studies) assesses metal stocks and flows of a specific country. Figure S1 in the SI shows the percentage distribution of the reviewed studies by spatial extent. Regional or national studies exist mainly for industrial countries. Global studies often extrapolate data from industrial countries because of the lack of domestic data in developing countries.[7]

Thirty-one studies model both retrospective and prospective flows, examining temporal extents in the time frame from 1700 to 2100. Twenty-six studies analyze only past flows and three studies include only prospective flows. The temporal scale of input and output data is usually one year. Hence, discrete-time calculations are also carried out with time steps of one year.

System Overview. The structure of most studies is based on a generic system with processes graphically represented in a sequence or a loop.[7] Although some studies include subprocesses containing more details than the top-level processes listed above, the general structure remains the same (see Figure 1). The topology of flows between the processes depends on the purpose, the characteristics of a considered metal (e.g., potential toxicity), and the complexity of the study; for example, some studies consider metal emissions of all processes (e.g., refs 20 and 33), some only of the use phase (e.g., refs 13 and 43), and other studies do not take emissions into account at all (e.g., refs 11, 41, and 42).

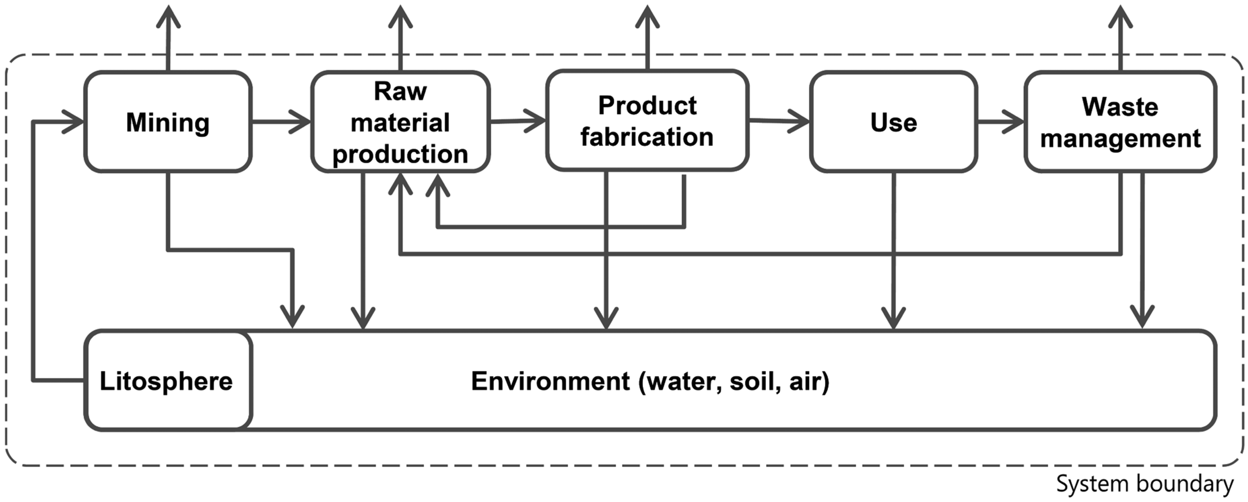


Figure 1. System overview of a generic dynamic material flow model of metals.

Basic Principles. The dynamic MFAs of metals generally assume that in the production, manufacturing, and waste management processes, no material is stored or the net flow during the sample time is zero, that is, that this part of the system can be treated as static. Hence, the dynamic modeling approaches focus on the use phase (which has nonzero net flows) and the resulting in-use stock changes. Stocks and flows are often modeled as time series with a constant sampling rate T, that is, $f[n] = f(nT)$, typically with T = 1 year.

The in-use stocks are quantified by one of the following two methods. The top−down approach derives the in-use stock S from the net flow by using the balance of masses as shown in eqs 1a, 1b, and 1c.[17]

$$\mathrm{d}S(t) = (\mathrm{inflow}(t) - \mathrm{outflow}(t))\cdot \mathrm{d}t = \mathrm{net\ flow}(t)\cdot \mathrm{d}t \tag{1a}$$

$$S[n] = (\mathrm{inflow}[n] - \mathrm{outflow}[n])\cdot T + S[n-1] \tag{1b}$$

$$S[N] = S[0] + T\cdot \sum_{n=1}^{N} (\mathrm{inflow}[n] - \mathrm{outflow}[n]) \tag{1c}$$

The second method, referred to as the bottom−up approach, derives the in-use stock $S[n]$ at a time n by summing all the metals contents c_i in their respective products or end-use sectors P_i according to eq 2[17]

$$S[n] = \sum_{i=1}^{I} P_i[n]\cdot c_i[n] \tag{2}$$

where I is the total number of products or end-use sectors considered. To construct time series of in-use stock, $S[n]$ is computed for every requested year n. If required, net flow can be calculated by introducing eq 2 into eq 1b. Almost 90% of the reviewed literature applies the top−down approach and 10% the bottom−up approach.

For inflows, historical data (e.g., on long-term consumption) is often accessible, but outflows are rarely measured. Most authors use and adapt methods developed in the field of system reliability to quantify the outflow of discarded items. The frequently used quantitative measures to describe this process[44] are listed in the SI.

Most authors choose to quantify outflows by assigning lifetime distribution functions to specific products or end-use sectors, with the relationship between inflows and outflows corresponding to a convolution (eq 3 with "*" denoting the convolution; this approach is also called the residence time model or population balance model[11,45−47]). Since it is rarely possible to solve this convolution analytically, it is integrated numerically according to eq 4.

$$\mathrm{outflow}(t) = (\mathrm{inflow}*f)(t) = \int_{-\infty}^{\infty} \mathrm{inflow}(t-u)\cdot f(u)\, \mathrm{d}u \tag{3}$$

$$\mathrm{outflow}[n] = \sum_{m=-\infty}^{\infty} \mathrm{inflow}[n-m]\cdot f[m] \tag{4}$$

where $f(t)$ and $f[m]$ are the probability densities of the lifetime distribution function for the continuous and the time discrete case, respectively.

The lifetime distribution functions most frequently used are the Dirac delta distribution, which represents average and constant lifetime, and the Weibull distribution. Other distributions used are the normal, log-normal, beta, and gamma distributions. In 23 of the reviewed studies, authors use two or more distributions. They either choose this approach according to available lifetime data for their considered products or end-use sectors (e.g., refs 18, 48, and 49) or to explore the effect of applying different lifetime distributions on the model output.[11,21,50−55] Melo,[11] for example, uses the delta, Weibull, normal, and beta distributions for modeling scrap flows. He concludes that by applying the delta distribution, scrap flows are highly influenced by fluctuations of the inflows, which can lead to significant under- or overestimations of the scrap potential. All of the other distributions lead to a smooth progress of outflows, but compared to the normal distribution, the Weibull and beta distributions can assume a wide variety of shapes. The results thus show no significant difference between the two lifetime distributions. Dahlström et al.[50] compare the delta, Weibull, and log-normal distributions and reach similar conclusions, as the log-normal distribution can be adapted as well. Other authors tested the sensitivity of their models regarding different lifetime distributions, mean values of lifetimes, and deviations. In addition to the findings described above, they find that their models are most sensitive to the mean values of lifetimes.[35,51,53,55]

Table 3. Dynamic Modeling Approaches Implemented in the Reviewed Literature

	retrospective	retrospective and prospective	prospective
top−down	historical data and lifetime distribution[8,18,22,34,35,39−41,45,46,50,52−55,59,61−71]	historical data and lifetime distribution + constant consumption model[30,42,58,72,73] linear consumption model[10,25,74] exponential consumption model[11,32,37] logistic consumption model[10,73] regression model[33,43,73,75] intensity of use[20,26] consumption scenarios according to existing models[38] individual consumption models for each product group[29] logistic stock/capita model[24,47−49,76,77]	individual consumption scenarios[23,27,28]
bottom−up	historical data and lifetime distribution[34]	historical data and lifetime distribution + exponential stock model[78] stock scenarios according to existing models[31,79] individual stock models for each metal-containing technology[13,80]	

Instead of using a lifetime distribution, Cheah et al.[37] choose a logistic survival rate function. In their review of methodologies for estimating lifetime distributions of commodities, Murakami et al.[56] and Oguchi et al.[57] give a comprehensive overview of how a lifetime distribution and a survival rate distribution, among others, are related. Only a few studies use time varying, nonparametric lifetime data, for example, for passenger vehicles and trucks in Japan[18,34,58,59] and lead-containing products in a global stock analysis.[60]

Table S3 in the SI summarizes the characteristics and implementations of the different distributions.

Dynamic Modeling Approaches. The dynamic modeling approaches can be grouped according to their temporal extent and basic modeling principles as shown in Table 3.

The first dynamic MFAs of metals were modeled using retrospective and partly also prospective top−down approaches. With the exception of Van Beers and Graedel,[78] bottom−up approaches have been applied only since 2009. Likewise, solely prospective dynamic MFAs of metals based on scenario analyses have only been established recently. Figure 2 summarizes the development of modeling approaches over time.

Retrospective Top−Down Approach. Probably the simplest approach is the retrospective top−down dynamic MFA. It analyzes past stocks and flows based on time series of historical inflow data, such as trade, import, or consumption statistics. Given the past inflows, the outflows are calculated according to eq 4, and subsequently, stocks are calculated using eq 1b. This approach is the most frequently chosen in the existing literature on dynamic MFA of metals (see Table 3), probably because of the better availability of inflow data compared to the stock data needed for bottom−up approaches.

In a recent study, Pauliuk et al. extend the top−down approach by calibrating its results based on the assumption that the old scrap supply equals the apparent old scrap demand, given a balanced scrap market, a homogeneous stock, and a perfectly closed steel cycle.

Retrospective Bottom−Up Approach. A retrospective bottom−up model produces time series of historical stock data based on eq 2. If lifetime distributions and an initial stock value $S[0]$ are known, past inflows and outflows can be calculated iteratively by applying eq 4 and eq 1c. These latter calculation steps overlap with the retrospective top−down approach, so

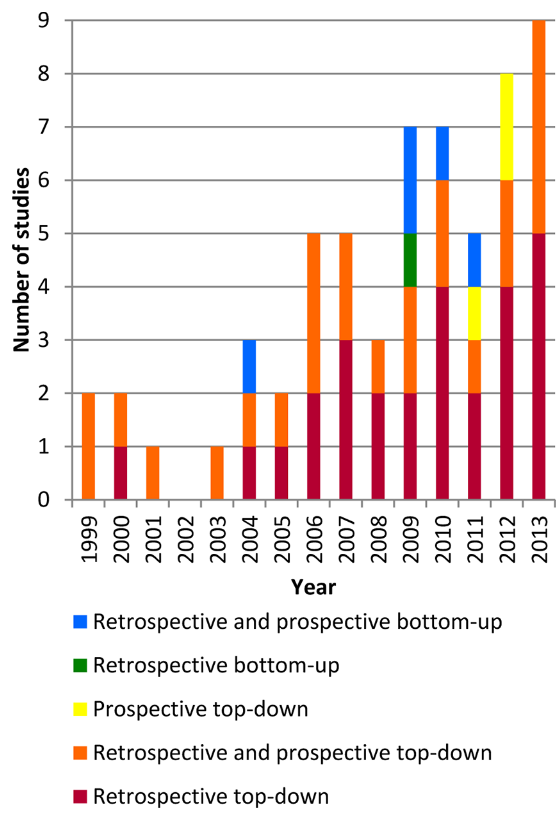


Figure 2. Development of modeling approaches used in dynamic MFAs of metals from 1999 to 2013.

both approaches can be used to calculate the missing time series; given the inflow and a lifetime distribution, the outflow and stock are calculated (thus an input-driven model) and given the stock and a lifetime distribution, the outflow and inflow are calculated (thus a stock-driven model). Hirato et al.[34] use both retrospective top−down and bottom−up models for automobiles in Japan and compare the results of the two approaches.

Retrospective and Prospective Top−Down Approach. Given that retrospective dynamic MFAs provide insights only on resource use in the past, the top−down approach is often combined with extrapolating time series of historical inflow data

dx.doi.org/10.1021/es403506a | Environ. Sci. Technol. 2014, 48, 2102−2113

by fitting an appropriate continuous time function. The same approach can be used for a stock-driven model, as first introduced by Müller,[8] who proposes using the service provided by the in-use stock as the main driver of a material cycle, especially for materials with a long service lifetime. Past stocks are first calculated using an input-driven model; then the stocks are extrapolated to finally calculate future inflows using a stock-driven model.

The models used by Michaelis and Jackson, Hatayama et al., Igarashi et al., and Oda et al.[30,42,58,72,73] assume that metal consumption is in a steady state at a level of a specific reference year t_0

$$\mathrm{inflow}(t) = \mathrm{inflow}(t_0) \tag{5}$$

According to Michaelis and Jackson[42] and Oda et al.,[58] this simplification is justified when the metal stock has already reached or is going to reach saturation.

Zeltner et al.,[10] Park et al.,[74] and Yan et al.[25] use a linear model to extrapolate total metal consumption

$$\begin{aligned}\mathrm{inflow}(t) &= p_1 \cdot t + p_0 \text{ (continuous time)}\\ \mathrm{inflow}[n] &= p_1 \cdot nT + p_0 \text{ (discrete time)}\end{aligned} \tag{6}$$

with p_0 = initial value [kg/s] (all dimensions are given in SI units) and p_1 = gradient [kg/s^2].

Models with an exponential consumption rate use a constant consumption growth rate which can be based on, for example, market reports or expert judgments[11,32,37]

$$\begin{aligned}&\mathrm{inflow}(t) = \mathrm{inflow}(t_0)\cdot(1 + p_1)^{t/\tau}\\ &\mathrm{inflow}[n] = \mathrm{inflow}(n_0T)\cdot(1 + p_1)^{nT/\tau}\\ &\text{if } \tau = T, \text{ then } \mathrm{inflow}[n] = \mathrm{inflow}(n_0T)\cdot(1 + p_1)^{n}\end{aligned} \tag{7}$$

with p_1 = constant growth rate [-] in one period τ. Zeltner[10] and Igarashi et al.[73] also model future metal consumption with a logistic function that takes growth limits of a system into account

$$\begin{aligned}\mathrm{inflow}(t) &= \frac{p_1}{1 + \mathrm{e}^{-p_2(t-p_3)}}\\ \mathrm{inflow}[n] &= \frac{p_1}{1 + \mathrm{e}^{-p_2(nT-p_3)}}\end{aligned} \tag{8}$$

with p_1 = saturation value of inflow [kg/s], p_2 = steepness of the sigmoidal curve [-], and p_3 = midpoint of the growth trajectory [-]. The parameters are determined using fitting algorithms (e.g., the ordinary least-squares method).

Elshkaki et al.[33,75] and Yamaguchi and Ueta[43] model inflows of lead-containing products as a function of socioeconomic explanatory variables, such as GDP, population size, product price, etc.

$$\mathrm{inflow}(t) = p_r + \sum_{i=1}^{n} p_i \cdot X_i(t) + \varepsilon(t) \tag{9}$$

with p_r = regression parameter [kg/s], n = number of socioeconomic explanatory variables, p_i = regression parameter [kg/(s·U)] (U = unit of explanatory variable), $X_i(t)$ = time series of socioeconomic variables [U], and $\varepsilon(t)$ = the model error [kg/s].

Through regression analysis, the most significant socioeconomic variables can be found and evaluated based on statistical tests, such as the adjusted coefficient of determination, t test, and F-statistics.[33] The regression parameters are determined via fitting algorithms. With extrapolations of the explanatory variables, the regression model is then used to estimate future inflows. Igarashi et al.[73] apply linear and nonlinear regression models in a similar approach.

Ayres et al.[20] and Kapur[26] use extrapolative scenarios based on the intensity-of-use hypothesis, which describes a metal's intensity of use (metal demand per unit GDP) as a function of per capita income with a general form of an inverse U-shaped curve

$$\mathrm{IU}(y(t)) = \frac{p_1}{y(t) + \frac{p_2}{y(t)^{p_3}}}\cdot p_4^{(t-t_0)/T} \tag{10}$$

with $y(t)$ = GDP per capita [US\$/p], p_1 = parameter [kg/p], p_2 = parameter [US\$/p], p_3 = parameter [-], p_4 = factor that scales down the intensity of use with time) [-], and t_0 = first year [s]. The inflow of metal is then

$$\mathrm{inflow}(t) = \mathrm{IU}(y(t))\cdot w(t) \tag{11}$$

with $w(t)$ = GDP [US\$].

The curve illustrates the development from an agricultural to an industrial, more resource-intensive economy, and eventually to a high-income, service-oriented economy, which in turn has lower resource use. The extrapolation of population and GDP is based on scenarios developed by the Intergovernmental Panel on Climate Change (IPCC).[20]

Yano et al.[38] model the current and future end-of-life vehicle flows and their lead content based on Japanese car registration statistics and forecasts.

In a study on platinum use for new technologies, Elshkaki and Van Der Voet[29] present individual production and consumption models for each product group, often as functions of the exogenous variables GDP and population. Future GDP and population data is also taken from existing IPCC scenarios.

Instead of extrapolating inflows, Hatayama et al. and Pauliuk et al.[47−49] use forecasts of the in-use stock for their prospective dynamic MFAs.

Pauliuk et al.[49] apply logistic stock per capita models with scenario-dependent saturation levels

$$\text{per capita stock}(t) = \frac{p_1}{1 + \left(\frac{p_1}{p_0} - 1\right)\mathrm{e}^{-p_2 p_1 t}} \tag{12}$$

with p_0 = initial value [kg], p_1 = saturation value of total stock per capita [kg], and p_2 = constant [1/(kg·s)]. They choose the parameters by assuming that the future per capita stock is tangent to the actual development and will ultimately reach the saturation level. Future inflows can be iteratively calculated based on future stock data with an initial value for inflow ($t = t_0$) and eq 4 being introduced into eq 1b.

The models applied by Hatayama et al.[24,47,48] are based on per capita GDP as the only exogenous variable, allowing for different growth rates between regions

$$\text{per capita stock}(t) = \frac{p_1}{1 + \mathrm{e}^{(p_2 - p_3\cdot(\mathrm{GDP}(t)/\mathrm{cap}(t))}} \tag{13}$$

with p_1 = saturation value of total stock/capita [kg], p_2 = parameter [-], and p_3 = parameter [1/(US\$)]. The model parameters are determined using nonlinear regression on the historical relationship between the per capita stock and GDP.

dx.doi.org/10.1021/es403506a | Environ. Sci. Technol. 2014, 48, 2102−2113

In their most recent studies, Pauliuk et al.[76] and Liu et al.[77] use a generalized five-parameter logistic function as a synthesis of the logistic curve and a Gompertz model to choose the saturation level and time independently for different world regions.

Retrospective and Prospective Bottom–Up Approach. Van Beers and Graedel[78] use a retrospective and prospective bottom–up approach to model zinc in-use stocks in Cape Town. They apply constant annual stock growth rates based on literature data for extrapolating past and future in-use stocks and flows.

$$\text{stock}(t) = \text{stock}(t_0)\cdot(1 + p_1)^{t/\tau} \tag{14}$$

with p_1 = constant growth rate [-] in 1 period τ. Stock-driven copper flows were dynamically modeled by Bader et al.[13] The model comprises detailed analyses of historical copper stocks, including individual models for different end-use sectors and their copper-containing technologies.[12] For the extrapolation of stocks, Bader and his colleagues apply and fit logistic, linear-logistic, or double-logistic stock growth models, depending on the historical growth patterns. Gerst[80] also proposes a dynamic bottom–up model of global copper stocks. He derives his models from the historical stock of copper-containing technologies based on macrolevel socioeconomic variables such as GDP per capita, population, average household size, and level of urbanization. These time-dependent variables are then extrapolated, partly based on existing models and scenarios, to compute future stocks.

Saurat and Bringezu[31] model the use of platinum group metals in catalytic converters in automobiles in Europe based on a bottom–up simulation of the European fleet of passenger cars, including the expected evolution of the future fleet of fuel cell vehicles. For their dynamic analysis of iron and steel in Chinese residential buildings, Hu et al.[79] use an existing dynamic MFA model simulating the development of the floor area stocks in China's urban and rural housing systems.

Prospective Top–Down Approach. Zuser and Rechberger,[27] Marwede and Reller,[23] and Alonso et al.[28] model the future consumption of metals used in emerging technologies with a prospective top–down approach. Alonso et al.[28] model five demand scenarios for REE, basing the demand either on historical production or demand growth rates or on expected demand growth rates for emerging technologies according to expert knowledge or existing scenarios.

Zuser and Rechberger[27] analyze material demand for four different photovoltaic technologies according to three demand scenarios. In a similar approach, Marwede and Reller[23] develop three demand scenarios for tellurium in cadmium telluride photovoltaic cells.

Dissipation. The focus of the dynamic MFAs of metals reviewed is on bulk metal flows incorporated in durable goods and infrastructure. Metals, however, are also dissipated to the environment throughout their life cycle, with material dissipation being understood as defined by Ayres and colleagues[19,20] (see also Method section). In about half of the reviewed studies, dissipative flows are described as such or referred to as emissions, loss flows, stock leakage or specific flows to landfills or the environment. In 18 studies, the concept is an inherent part of the methodology, with 11 of these considering dissipative flows for all life phases and seven for only the use or disposal phase. Dissipative outflows of a specific process are calculated either from inflows and transfer coefficients or loss rates (e.g., refs 13, 20, and 53), from stocks and leaching/emission factors or corrosion coefficients (e.g., refs 13 and 75), from mass balances,[59] or based on historical data (e.g., slag sales[51]). Only three authors[13,20,71] consider time-variant coefficients; in all other studies, the share of dissipation remains constant over time.

Some literature specifically focuses on time-variant dissipative flows. In a recent study, Lifset et al.[5] assess dissipative copper flows in the United States based on historical data and individual models for the different copper flows. They further categorize these flows into "intentional and unintentional release", as well as "intentional and unintentional use". They also define a dissipation index that quantifies the ratio of dissipative flows to bulk flows as a measure of resource efficiency. Elshkaki et al.[81] model the nonintentional flows of lead in the Dutch economic system using a regression model approach, Sundset et al.[82] illustrate the mercury flows in the European Union, and Yamasue et al.[83] evaluate the potential amounts of dissipated rare metals from waste electrical and electronic equipment in Japan.

Spatial Dimension. It is important to know the location of a resource in addition to its quantity and quality to consider it for future mining.[78] Thus, some studies include the spatial distribution of in-use stocks, based on statistical or remote sensing data and usually processed in geographic information systems (GIS).[84] Van Beers and Graedel[78] link GIS data sets from a population census in Cape Town with zinc densities per area by applying weighting factors related to household income, dwelling type, or length of roads. In combination with annual stock growth rates, they also calculate the retrospective and prospective zinc distributions. Pauliuk et al.[70,76] and Liu and Müller[36] analyze steel and aluminum stocks and flows, respectively, for all countries in the world, based on statistical data. In addition, Pauliuk et al.[76] include capacity models to show how extensive tradeing of finished steel could prolong the lifetime of the steelmaking assets in different world regions, and Liu and Müller[85] develop a trade-linked multilevel MFA to map the global pathways of aluminum between countries. Remote sensing methods are used by Takahashi et al.,[86] who analyze in-use copper stocks using satellite nighttime light observation data.

Other studies that include the spatial dimension of metal stocks are static MFAs.[87−89]

Uncertainty. Data included in an MFA of metals are acquired from many different sources with varying data reliability. If only individual values from measurements, expert interviews, or historical sources are available, it is often difficult to quantify the uncertainty of input data and parameters. The reviewed literature can be roughly divided into four groups according to how uncertainties are handled (see also Figure S2 in the SI).

The first group, comprising approximately half of the studies, does not consider data uncertainty. The second group, 37% of all studies, applies sensitivity analysis. A sensitivity analysis helps to assess the relevance of uncertainties of the model parameters by providing knowledge of how the model output reacts to parameter changes. Many studies test the sensitivity of the model to different average lifetimes[22,35,45,51,67] or different lifetime distributions and standard deviations,[22,53,55,68,69] concluding that varying average lifetimes has a greater influence on model results than varying standard deviations or lifetime distributions. In addition to lifetime distributions, authors also carry out sensitivity analyses for other key parameters, such as steel intensity,[34,50,52,63,79] scrap recovery rate,[37,50,52,59] population size, stock saturation level, and saturation time.[76] In their most recent studies, Liu et al.[36,77] and Pauliuk et al.[70] carry out full sensitivity analyses for all model parameters according to their estimated data uncertainty. Besides the average lifetime, they found that the trade data estimation (based either on reported import or export data) and the metal concentration in commodities also have a high impact on resulting in-use stock calculations.

McMillan et al.[54] quantify the sensitivity of the lifetime distribution, recycling rate, and metallic recovery by using the Fourier Amplitude Sensitivity Test method, which provides a measure of input sensitivity defined as the fraction of total model variance.

The third group, 6% of all studies, uses uncertainty intervals. In particular, Kapur[26] assigns confidence levels to copper flows according to a confidence scale developed by Moss and Schneider.[90] He states that, as a general rule, the data quality decreases along the life cycle of a metal, that is, data for production, manufacturing, and the inflows into the use phase is more reliable than data for the waste management and recycling processes. Hedbrant and Sörme include uncertainty intervals as proposed in a comprehensive article on data uncertainty in urban heavy metal data collection. They assign uncertainty levels to sources of information, with associated uncertainty intervals based on factors (e.g., the value x could be as much as $3x$ or as little as $1/3x$, annotated with "*/" analogue to "±"), which is especially useful for large uncertainties.[91] Van Beers and Graedel[78] apply asymmetrical uncertainty ranges for the zinc stocks per end-use sector.

Finally, the fourth group (5% of all studies) uses the Gaussian error propagation to calculate the standard deviation of stocks and flows based on standard deviations that were defined for each input variable and parameter.[10,13,36]

A different approach to handling uncertainty, which has not, however, been applied to MFAs of metals thus far, is probabilistic MFA, as proposed by Gottschalk et al.[92] They model inflows, transfer coefficients, and concentrations as probability distributions. The shape of the distributions (e.g., uniform, triangular, or log-normal) is chosen based on the characteristics of the available data. The dependent variables are calculated by means of Monte Carlo simulation and are therefore again provided as probability distributions. Bornhöft et al.[93] review existing modeling approaches and tools with regard to the requirements of probabilistic MFA.

Initial Condition. The initial condition of an MFA model depends primarily on the temporal extent chosen. If analyses go far back in time, initial stocks and flows are often considered zero at $t = t_0$. If the temporal extent is short or starts in the present, initial stocks, and flows are defined based on available data or the authors' assumptions.

Model Input Data. Model input data includes time series for exogenous model variables such as metal inflow and stock data, socioeconomic data such as GDP or population, and model parameters such as the average lifetime of products or end-use categories. A detailed discussion of the data sources used by the studies reviewed is beyond the scope of this article.

Model Output Data. Model output data of the reviewed dynamic MFAs of metals comprise time series of those stocks and flows under investigation. Some studies quantify only the in-use stock (e.g., refs 68 and 80), while others provide information on all stocks and flows in their system from extraction to landfilling.[45] The resulting stocks and flows are often further divided into end-use sectors or products (most studies), disaggregated for different regions or countries,[40,48] or include details such as the chemical composition of scrap flows or a breakdown into different alloy types (e.g., refs 59 and 72).

Evaluation. Besides a visualization and discussion of the output data, some studies include further evaluation. Various indicators can be applied that condense the results for better explication and communication.[16] Examples include the recycling rate, defined as the ratio between the actual scrap consumption and the scrap arising,[52] the scrap self-sufficiency ratio as the ratio of scrap recycling to scrap demand,[74] or other recycling indicators as applied by Glöser et al.[22] or Yan et al.[25] Zeltner et al.[10] introduce the separation efficiency as the fraction of recycling in the total waste flow and Bader et al.[13] present the consumption loss as the sum of all metal flows to landfills or the soil/aquatic system.

Some evaluations are based on the relationship between material stocks and flows and socioeconomic indicators. McMillan et al.[54] analyze the relationship between the net addition to stock and GDP, and Mao and Graedel[66] and Liu and Müller[36] relate per capita stock to per capita GDP.

Comparisons of natural resources with anthropogenic stocks and flows are performed by Müller et al.,[51] Gerst[80] Alonso et al.,[28] and Liu and Müller.[36]

Some authors evaluate results by comparing them with the outcome of other studies (e.g., refs 13 and 80).

Further approaches evaluate the energy consumption or environmental impacts of the metal flows. Dahlström et al.[50] use value chain analysis to examine the material- and energy-related resource productivity and efficiency of the iron, steel, and aluminum industries in the United Kingdom, and Cheah et al.[37] analyze the embodied energy demand of automotive aluminum. For the United Kingdom steel sector, Michaelis and Jackson[41,42] calculate the consumption and development of exergy (available work). Hu et al.[79] assess resource depletion and global climate change by the accumulated net steel use and the net CO_2-equivalent emissions. The CO_2 emission volume reduction potential resulting from an enhanced collection of postconsumer steel was analyzed by Igarashi et al.[30] Saurat and Bringezu[31] model the SO_2 emissions related to PGM production and use in Europe. Liu et al.[35,77] analyze the energy use and greenhouse gas (GHG) emissions of the U.S. aluminum cycle and the GHG emission pathways of the global aluminum cycle.

Additional analyses include, for example, multimaterial pinch analyses to derive optimized recycling,[24] material intensity per service unit, life-cycle assessment, cost-benefit analysis, statistical entropy analysis,[16] and entropy analysis.[94−96]

Detailed Model Description. The detailed model description in the (adapted) ODD protocol comprises details about the model's formalisms (e.g., equations) and algorithms (e.g., solution procedures), the exogenous and endogenous variables, and the model parameters. Providing detailed model descriptions beyond the generic equations already discussed is beyond the scope of this article.

DISCUSSION

We reviewed 60 studies of anthropogenic metal flows, comparing them with regard to their purpose, the materials, products, and sectors investigated, the coverage of processes, their spatial and temporal scale and extent, and the way they conceptualize and delimit the system under study. We extracted and summarized the basic concepts, principles, and methodological approaches underlying the models used and showed how the approaches developed over time.

The adapted ODD protocol proved to be beneficial for structuring the review process as well as this article. The literature fit well into the protocol's structure, which helped us to provide a one-to-one comparison of corresponding elements of the models despite their high diversity. The ODD protocol in our adapted form could hence provide a basis for the standardized description of MFA models in general, providing better orientation to the reader and supporting the completeness of model documentation and reproducibility of the results.

Most studies apply a top−down approach that could be used for any material. The required time series of inflow data are often provided by production, trade, or consumption statistics. Data is mainly available for bulk metals or, in the form of global production figures, also for less widely used metals. Because available inflow data is often highly aggregated, the top−down approach may be less suitable for specific products or smaller regions.

Only 10% of the studies apply a bottom−up approach. Bottom-up models are far less generic since the entire stock of a specific metal has to be assembled from all product groups containing that metal. These may all show different growth patterns, requiring a specific stock model for each product group as well as extensive data collection.[13,78,80] This approach is thus most suitable for analyzing metals that are only used in a few products, or for focusing on a specific product. Bottom−up models can also benefit from existing models that provide past and future time series of stock data that can be directly applied in an MFA.[31,79]

The bottom−up approach, although it has not been widely applied to date, could provide important insights on consumer behavior, which, for example, influences the product lifetime or the disposal pathways, sociocultural and spatial differences in patterns of metal use,[36] the split of metals to different end-use sectors, or the share of obsolete stock (e.g., stored products, abandoned infrastructure) in the in-use stock[70] by investigating in detail the in-use stock, for example, through consumer surveys.

Especially for studies with a long time horizon, the assumption of constant model parameters, for example, the lifetime distribution parameters, is a far-reaching simplification that could add a significant error to the results.[97,98] Sinha-Khetriwal et al.[99] also point out that forecasts of outflows can be improved by introducing product mass functions incorporating the changing weight of products over time as in Gregory et al.[100] Such data can often only be derived from bottom−up models. Detailed data generated by bottom−up models can also be used to calibrate and validate top−down models, an issue not yet often addressed in the literature. Future research should therefore investigate time-variant systems and how top−down and bottom−up models could be combined or complemented.

The diverse extrapolation methods reveal various challenges. Inflow data often fluctuates, depending on economic and technological developments, such as market crises or product substitutions, which are difficult to predict. Furthermore, linear and exponential consumption models, since they do not take into account resource scarcity and market saturation, are only valid within a short time frame. Extrapolation of inflow data is therefore prone to oversimplification. Stocks, however, are less affected by short-term market fluctuations and thus provide a more robust basis for forecasts.[8,77] For regression models, too, the forecasts based on socioeconomic variables are only valid as long as no unpredicted societal or economic changes occur.[33]

The majority of studies analyze bulk metals such as iron/steel, aluminum, copper, and zinc with increasingly detailed information on their stocks and flows. Literature on less widely used metals such as indium, tantalum or REE is still scarce. The use of these metals highly depends on new emerging technologies. At the end of product life, most of the metals are not recovered but lost from the system considered, since there is not yet any technically or economically feasible recycling option for many of them. It is thus highly advisable to further develop data and models with a special focus on metal dissipation. Earlier, dissipation, losses or emissions of metals to the environment were included in dynamic MFA models focusing on heavy metal pollution. In recent years, dissipation has also been addressed also from a resource point of view, but data is extremely scarce and many authors have still not taken up the issue. Moreover, metals may not only be dissipated, but also distributed in low concentrations to many products, and, within these products, scattered all over the globe. These metals are not irrecoverable, but great efforts are needed to concentrate them again. Rechberger and Graedel[101] developed statistical entropy analysis to measure the distribution pattern of a substance over its life cycle, that is, to describe how a system concentrates or distributes substances. It appears promising to apply statistical entropy analysis to metals in the anthroposphere for the measurement and illustration of their distribution as a basis for improved resource management. Future research should also investigate other, possibly new indicators of dissipation and material distribution.

The most recent literature[36,70,77] has shown that besides lifetime distribution parameters there might be many other parameters or variables with a strong influence on the model's output. Performing uncertainty analysis such as Gaussian Error Propagation, or, if the data uncertainty is unknown, a full sensitivity analysis, is therefore important to understand the effect of uncertain model input. Probabilistic MFA by Gottschalk et al.,[92] which models all data as probability distributions and thus accounts for the influence of the model input's uncertainty on the model output, is a comprehensive approach for dealing with uncertainty.

Dynamic MFA is a useful method for providing knowledge of metal stocks and flows in the anthroposphere in a simple and comprehensible way. Many studies also include further evaluations of their results or serve as a basis for further assessments (e.g., ref 102). However, with the exception of Kapur et al.,[26] the results and conclusions of the reviewed literature do not directly support environmental policy making. Some studies indirectly give recommendations, for example, regarding how to increase recycling rates (e.g., refs 23, 39, 49, 54, 62, and 65), reduce environmental impacts (e.g., refs 31 and 67) or mitigate climate change (e.g., refs 41 and 77), but without a clear target audience. We suggest that future studies that intend to provide environmental policy recommendations, identify their target audience and their purpose from the beginning. MFA could also be recognized as a necessary element of impact assessments used for new regulations, such as the sustainability impact assessment used for new trade agreements by the European Commission.[103] The mid- and long-term impacts of political decisions on material stocks and flows may become one of the most important economic and environmental concerns in the coming decades. To fulfill the requirements of policy support, MFA models should be embedded in an environment of scenario definition and simulation that easily connects the models to socioeconomic data taken from statistical databases and geographic information systems.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Summary and overview of all reviewed studies, overview of the processes covered by the reviewed literature, figures showing the percentage distribution of the reviewed studies by spatial extent and by treatment of data uncertainty, more details on the basic modeling principles, a summary of lifetime distribution functions, and additional information on the relationship between different measures from the field of system reliability. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We thank Prof. Dr. Matthias Reuter and Dr. Patrick Wäger for their helpful comments on the article. The project was funded by Empa.

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