Doctoral Thesis

Material Flow Analyses of Steel and Mercury - Differences and implications of data availability

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Dissertation

Stoffflussanalysen von Stahl und Quecksilber - Unterschiede von Datenverfügbarkeit und deren Auswirkungen

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Kurzfassung

Stoffflussanalysen sind eine bekannte und weit verbreitete Methode, um Flüsse, Quellen und Senken von Ressourcen oder Schadstoffen in einem bestimmten System zu untersuchen und zu verstehen. Auf regionaler, nationaler oder globaler Ebene wird die Quantifizierung der Ströme häufig durch die unzureichende Verfügbarkeit von Daten erschwert. Die Datenverfügbarkeit für branchenweite Analysen ist meist besser. Stoffstromanalyse auf Unternehmensebene ist vergleichsweise einfach durchzuführen, da alle erforderlichen Daten gemessen werden können, falls sie nicht verfügbar sind. In der vorliegenden Arbeit wurden die Flüsse von zwei Materialien mit Hilfe der Stoffflussanalyse eingehend untersucht. Bei dem einen handelt es sich um einen der weltweit am meisten verwendeten Rohstoffe, Stahl. Der andere ist das Spurenmetall Quecksilber, das aufgrund seiner toxischen Eigenschaften strengen Restriktionen unterliegt. Sowohl für Quecksilber als auch für Stahl werden die Vorgehensweise bei der Erstellung einer Stoffstromanalyse sowie die Ergebnisse der Analyse detailliert untersucht und diskutiert. Ein weiterer Teil dieser Arbeit befasst sich mit der Datenverfügbarkeit für beide Materialien und der Notwendigkeit einer Unsicherheitsbewertung in beiden Bereichen.

Während die weltweite Stahlproduktion in den letzten Jahrzehnten enorm gestiegen ist (von 0,9 Mrd. Tonnen im Jahr 2000 auf 1,9 Mrd. Tonnen im Jahr 2020), ist in industrialisierten Ländern eine Sättigung des Stahlmarktes eingetreten, die sich durch Stagnation der Rohstahlproduktion und einem höheren Anteil an Altschrottarten äußert. Diese Entwicklung würde es ermöglichen, einen höheren Anteil des Stahls aus Schrott zu erzeugen. Aus veröffentlichten Statistiken geht jedoch hervor, dass die Stahlproduktion aus Sekundärrohstoffen in vielen dieser Region stagniert und ein erheblicher Anteil des Schrotts exportiert wird, während diese Länder gleichzeitig große Mengen an Eisenerz einführen. Ein limitierender Faktor für die schrottbasierte Stahlproduktion ist der Gehalt von unerwünschten Begleitelementen im Schrott. Daher wurde im Rahmen dieser Arbeit eine Qualitätsbewertung von Stahl und Stahlschrott anhand des Gesamtgehalts von fünf relevanten Begleitelementen, nämlich Cu, Sn, Cr, Ni und Mo, durchgeführt. Insbesondere wurde der europäische Stahlhaushalt quantitativ und qualitativ mittels Stoffstromanalyse modelliert. Für die historische Analyse (1946 - 2017) wurde eine retrospektive mehrjährige Stoffflussanalyse durchgeführt, während für die Prognose der zukünftigen Stahl- und Stahlschrottströme eine dynamische Stoffsflussanalyse für den Zeitraum 1910 bis 2050 durchgeführt wurde. Da detaillierte Informationen über den Gehalt der fünf Begleitelemente in den verschiedenen Stahlprodukten und Stahlschrotten kaum verfügbar waren, wurden vier Kategorien (Stahlqualitätsklassen) für den Gehalt an Begleitelementen eingeführt. Die so modellierten Schrottmengen und -qualitäten wurden anschließend mit den erforderlichen Toleranzwerten für Begleitelemente in verschiedenen Stahlprodukten verglichen. Eine Material-Pinch-Analyse wurde für den Vergleich zwischen der

Schrottverfügbarkeit und der Stahlnachfrage durchgeführt. Diese Methode berücksichtigt, dass Materialien mit geringer Verunreinigung (z.B. geringer Gehalt an unerwünschten Begleitelementen) auch für Produkte verwendet werden können, die höhere Verunreinigungsgrade zulassen (z.B. hoher Gehalt an unerwünschten Begleitelementen), aber nicht umgekehrt. Die Materialflussanalyse des europäischen Stahlschrotts und der Stahlnachfrage zeigt seit Mitte der 1990er Jahre einen zunehmenden Überschuss an Ressourcen mit niedrigem Reinheitsgrad (Schrott mit niedrigem Reinheitsgrad), der ohne weitere Maßnahmen (z. B. Verdünnung mit Stahlquellen mit hohem Reinheitsgrad, verbesserte Sortierung oder technologische Eingriffe bei der Stahlerzeugung) nicht genutzt werden kann.

Es wird prognostiziert, dass die Menge an Schrott mit niedrigem Reinheitsgrad in der EU von 20 Mio. t/Jahr heute auf 43 Mio. t/Jahr im Jahr 2050 weiter ansteigen wird, wenn keine Maßnahmen zur Verbesserung der Verwertbarkeit des Schrotts ergriffen werden. Das Verdünnungspotenzial (Verdünnung von Schrott mit niedrigem Reinheitsgrad durch Ressourcen mit hohem Reinheitsgrad) würde bis 2040 erschöpft sein, wenn das derzeitige Schrottsammelsystem beibehalten wird. Ein Vergleich der aktuellen Handelsdaten für Altschrott mit dem modellierten Überschuss an Schrott mit niedrigem Reinheitsgrad zeigt, dass der Überschuss an Stahlschrott mit niedrigem Reinheitsgrad mit hoher Wahrscheinlichkeit aus der EU exportiert wird, da die Daten nahezu übereinstimmen. Dies führt zu der Schlussfolgerung, dass derzeit keine nennenswerten Anstrengungen unternommen werden, um die schrottbasierte Stahlproduktion durch fortschrittliche Schrottverarbeitungsmethoden zu steigern.

In einer weiteren Fallstudie wurde der Quecksilber-Durchsatz der österreichischen verarbeitenden Industrie untersucht. Es ist die erste Studie, die versucht, eine mehrjährige Materialflussanalyse zu Quecksilber für die gesamte verarbeitende Industrie eines Landes zu erstellen. Dafür wurden die Daten hinsichtlich ihrer Verfügbarkeit, Qualität und deren Mangel bewertet. Die verfügbaren Daten wurden zu einer mehrjährigen Materialflussanalyse kombiniert, in der die Inputs (primäre und sekundäre Ressourcen und Brennstoffe, Wasserversorgung) und die Outputs (Emissionen in Luft und Wasser, Produkte) quantitativ und mit der entsprechenden Unsicherheit definiert sind. Die entsprechenden Abfälle aus den einzelnen Sektoren wurden bilanziert und die zugehörigen Unsicherheiten durch Fehlerfortpflanzung ermittelt. Die Berechnung ergibt einen Quecksilber-Durchsatz für den Zeitraum 2005 bis 2016 zwischen 3,4 t/Jahr pm25% bis 4 t/Jahr pm25%. Der größte Teil des Quecksilbers wurde über primäre Roh- und Brennstoffe importiert (75%), etwa 25% über sekundäre Roh- und Brennstoffe. Der Quecksilber-Export über Produkte wurde auf 35% bis 40% geschätzt, jener über Emissionen in die Luft auf 20% bis 25% und über Abfälle auf 40% bis 45%.

Stahl ist eine der ersten Waren deren Produktion und Verbrauch systematisch erfasst wurden. Daten über Stahlflüsse werden seit vielen Jahrzehnten dokumentiert und veröffentlicht, wenn auch in unterschiedlicher Auflösung. Bestehende Datenlücken können meist durch Interpolation geschlossen werden. Allerdings liegen die Datensätze in der Regel nicht in einem maschinenlesbaren Format vor und müssen aufbereitet werden. Daten über die Mengen von Spurenelementen sind dagegen eher spärlich. Quecksilber-Konzentrationen für die meisten Güter sind verfügbar, aber oft von geringer Qualität (unvollständig, teilweise nicht repräsentativ) wobei Daten über

Warenmengen meist verfügbar sind, insbesondere für Primärrohstoffe und Produktmengen. Die öffentlich verfügbaren Daten unterscheiden sich jedoch stark von Sektor zu Sektor und innerhalb der Sektoren. Die verfügbaren Daten erlauben daher eine Bilanzierung, allerdings nur mit hohen Unsicherheiten und unerfassten Flüssen.

Abstract

Material flow analysis is a well known and widely used method to investigate and understand flows, sources and sinks of resources or pollutants in a defined system. On regional, national or global level, the quantification of flows is often challenged by unsatisfactory data availability. Data availability for industry wide commodities is generally better. Material flow analysis at company level is comparably easy to be accomplished, as all necessary data can be measured if not available.

In the present thesis the flows of two materials were investigated thoroughly by means of material flow analysis. One represents one of the most used commodities worldwide, steel. The other one is the trace metal mercury, which is to be phased out from our economy due to its toxic characteristics. For both, mercury and steel, the procedure to set up a material flow analysis as well as the results of the analysis are discussed in detail. An additional part of this thesis discusses the data availability for both materials and the need for uncertainty assessment within both areas.

Whereas the global steel production grew immensely during the last decades (from 0.9 billion tonnes in 2000 to 1.9 billion tonnes in 2020), affluent countries have experienced a saturation of the steel market, indicated by stagnant crude steel production rates and higher post-consumer scrap rates. This development would allow a higher share of steel to be produced from scrap. However, reporting data shows that the rate of steel production from secondary resources in many affluent countries or economic areas is stagnant and a significant share of scrap is exported, whereas at the same time these countries import massive quantities of iron ore. One limiting factor for scrap based steel production is the content of tramp elements present in scrap. Hence, in the frame of this thesis a quality assessment of steel and steel scrap was conducted using information about the total content of five relevant tramp elements, namely Cu, Sn, Cr, Ni and Mo. In particular, the European steel household was modelled in a quantitative and qualitative way using material flow analysis. For the historical analysis (1946 – 2017) retrospective multi-annual material flow analysis was conducted, whereas for the prognosis of future steel and steel scrap flows a dynamic material flow analysis, covering the period 1910 to 2050, was conducted. Since detailed information about the content of the five tramp elements in the different steel products and steel scraps was hardly available, four categories (steel quality classes) for content of tramp elements were introduced. The so modelled scrap quantities and qualities were subsequently compared to the tolerance levels of tramp elements in different steel products demanded. For the comparison between scrap availability and steel demand, material pinch analysis was used. This method considers, that materials of low impurity (e.g., low content of tramp elements) can also be used for products which allow higher impurity levels (e.g., high content of tramp elements), but not the other way around. The material pinch analysis of the European steel scrap and steel demand shows an increasing surplus of low purity resources (low purity scrap) since the middle of the 1990s, which cannot be used without further interventions (e.g. dilution with high purity steel sources, enhanced sorting or technological interventions during steel production). For the future situation, it is predicted that the amount of low purity scrap in Europe will further increase from today's 20 Mt/yr to 43 Mt/yr in 2050, if no measures are put in place to improve scrap usability. The dilution potential (dilution of low purity scrap with high purity resources) would be exhausted by 2040 if the current scrap collection scheme is upheld. When comparing current trade data on post-consumer scrap with the modelled surplus of low purity scrap, it is shown that the surplus of low purity steel scrap is most likely exported out of the EU, as both data match quite well. This leads to the conclusion, that currently there is no significant effort to increase the scrap-based steel production by advanced scrap treating methods.

In a further case study, the Mercury-throughput of the Austrian manufacturing industry was investigated. It is the first study to attempt to put together a multi-annual material flow analysis on Mercury covering the whole manufacturing industry of a country. Therefore, data was assessed regarding its availability, quality and lack thereof. The data available was combined to a multi-annual material flow analysis where inputs (primary and secondary resources and fuels, water supply) and outputs (emissions to air and water, products) are defined in their amount and according uncertainty. The corresponding waste from each sector was balanced and the according uncertainty was determined with error propagation. The model estimates the Hg-throughput over the period 2005 to 2016 between 3.4 t/yr ±25% to 4 t/yr ±25%. Most mercury was imported via primary raw materials and fuels (75%), and about 25% was imported via secondary raw materials and fuels. Hg export via products was estimated at 35% to 40%, via emissions to air at 20% to 25% and via waste at 40% to 45%.

Steel is one of the first commodities which was reported in a systemic way. Data on steel flows have been documented and made publicly available for many decades, even though in different resolution. Existing gaps can mostly be filled via interpolation. Still, the data sets are usually not available in a machine-readable format and needs to be processed. Data on amounts of tramp elements, on the other hand, are rather scarce. Hg concentrations for most goods are available, but often of low quality (incomplete, partly non-representative). Data for quantities of goods are mostly available, especially for primary raw materials and production numbers. Nevertheless, publicly available data differs strongly from sector to sector and within sectors. Therefore, the data available allows a balance, but only with high uncertainties and unconsidered flows.

Contents

Acronyms

b RB reinforcing bar

C construction

C Bu buildings

C In infrastructure

Ch chemical industry

CrS crude steel

DMFA dynamic material flow analysis

EEoLP export of end-of-life products

EG energy generation

EM emissions to air

EoLP end-of-life products

F fuels

FS fabrication scrap

FSP finished steel products

IP intermediate products

MET metal industry

MFA material flow analysis

MPA material pinch analysis

NEUFSP net-end-use of finished steel products

NIFSP net-import of finished steel products

NIIP net-import of intermediate products

NIS net-import of ingots and semis

NMM non metallic mineral industry

NRF norm reporting format for air pollutant emission inventory

OTH other industry

P products

PFS production and forming scrap

PoCS post-consumer scrap

PoCSg post-consumer scrap generated

PoCSr post-consumer scrap recovered

PRM-F primary raw materials and fuels

RM raw material

SR scrap recycled

SRM-F secondary raw materials and fuels

T transport

T Ca cars

T Tr trucks

WAT water supply

WMS waste management system

WPP wood pulp and paper industry

WW waste water

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Published articles and contributions

This thesis is based on the three journal articles listed below, which can be found in the Appendices of this work.

Paper I

Dworak, S., & Fellner, J. (2021). Steel Scrap Generation in the EU-28 since 1946 – Sources and Composition. Resources, Conservation and Recycling, 173. https://doi.org/10.1016/j.resconrec.2021.105692

published, available online 5 June 2021

Contribution: Conceptualization, Data collection and data analysis, Investigation, Formal Analysis, Visualization, Writing

Paper II

Dworak, S., Fellner, J., & Rechberger, H. (2022). How Will Tramp Elements Affect Future Steel Recycling in Europe? – A Dynamic Material Flow Model for Steel in the EU-28 for the Period 1910 to 2050. Resources, Conservation and Recycling.

published, available online 21 November 2021

Contribution: Conceptualization, Data collection and data analysis, Investigation, Formal Analysis, Visualization, Writing

Paper III

Dworak, S., & Rechberger, H. (2021). Mercury throughput of the austrian manufacturing industry – discussion of data and data gaps. Resources, Conservation and Recycling, https://doi.org/166.10.1016/j.resconrec.2020.105344

available online 9 December 2020

Contribution: Data curation, Software, Investigation, Formal analysis, Visualization, Writing - original draft preparation

Chapter 1

Introduction

1.1 Data availability - Commodity with contamination vs. toxic trace element

Many data points are necessary for effective modelling, especially for the construction of dynamic or multi-year models. For commodities, the data base is usually rather solid in affluent economies. Especially the steel industry has been reporting systematically since the 1950s (see e.g. Eurostat, 1983; Melcher et al., 1952). Data in this field is at least reported in a superficial way (upper-level-grouping, see e.g., Eurofer, 2018; World Steel Association, 2020). The data is mostly complete and available for long time periods and many countries. Further, more detailed data might be available for research upon request. If a cooperation between researchers and industrial associations or companies themselves can be established, both partners can benefit. Inherently, the situation is very different for trace elements, especially unwanted ones. There is no internal motivation to study areas which do not concern the process itself, but rather cause additional challenges. External pressure has to be applied (social pressure, legislation). Monitoring systems, which are usually in place in areas presumed to be sensitive, can supply a solid and constant source for flow data (among others, e.g. Hg emissions into the atmosphere (CEIP, 2020)). The estimation of other flows relies on data for flows of goods and the corresponding concentration. In general, little original data is available, so proxy data from various studies and reports is applied. Data is kept confidential, especially regarding the composition of flows and the content of specific elements or compounds. Hansen analysis (Hansen, 2002) already indicated that *MFA* studies often rely heavily on data on material flows from companies, which are kept confidential, and therefore confidentiality is one of the main challenges faced when putting together an accurate and feasible *MFA*. For flows of goods, economy-wide material accounting (Eurostat, 2020) and industry reports with sector-wide material accounting (e.g., Austropapier, 2018; Mauschitz, 2012) improved data availability significantly. Still, detailed data is often not available.

1.2 Steel scrap as secondary resource

Steel is one of the most frequently used commodity in our society. Global crude steel production nearly doubled during the last decades (based on World Steel Association, 2020). In affluent regions (e.g. US or EU), the growth has come to a stop. Globally only moderate growth, or even no further growth, is predicted for the coming years (Hatayama et al., 2010). Stagnant steel production rates facilitate higher shares of scrap as resource for crude steel production. This would not only be beneficial from a resource point of view, but also from an environmental one (e.g. CO_2 emission reduction (Broadbent, 2016; Damgaard et al., 2009), lower eutrophication, acidification and photochemical oxidation (Hu et al., 2014; López et al., 2020))

Zhu et al., 2019 showed that about two thirds of the annual steel production in 2014 in the US was based on scrap. In the EU, the share of scrap in the crude steel production amounts to about 54% (see Fellner et al., 2018; Passarini et al., 2018) and was declining in recent decades. Cullen et al., 2012 estimated that about 36% of the global crude steel production was based on scrap.

Steel flows have been extensively investigated on different levels, for different regions and time frames (e.g. Cooper et al., 2020; Hatayama et al., 2010; Klinglmair and Fellner, 2011; Mueller et al., 2011; Mueller et al., 2006; Pauliuk, Wang, et al., 2013; T. Wang et al., 2007). On the European level, only rather superficial studies are available (Fellner et al., 2018; Passarini et al., 2018), whereas single European countries have been investigated more closely (Mueller et al., 2011; Pauliuk, Wang, et al., 2013).

In recent decades, qualitative aspects have been added to quantitative analyses of steel flows. Therefore, content of impurities in form of specific tramp elements (mostly Cu, Ni, Mo, Cr and/or Sn) and possible technological interventions to deal with them have been investigated (e.g., Daehn et al., 2019; Daigo et al., 2021; Noro et al., 1997; Sampson and Sridhar, 2013; Savov et al., 2003; Spitzer et al., 2003). However, only few studies investigate the available scrap and its composition in relation to the crude steel demand and its requirements (e.g., globally for Cu (Daehn et al., 2017), for Japan (Daigo, Iwata, et al., 2017; Igarashi et al., 2007; T. Oda et al., 2010)). What the results of these studies have in common is that they indicate a surplus of low purity scrap, which requires dilution with primary steel in order to be recycled. On a global scale, this surplus might arise in the near future (see Daehn et al., 2017). In Japan it is already prevalent, as highlighted by Igarashi et al., 2007.

Two case studies in the present thesis modelled the steel flows in the EU retrospective (multi-annual *MFA* to analyse the past and current system) and prospective (*DMFA*, to predict future development), all in all covering the period from 1910 to 2050. In the analysis, quality (sum of Cu, Ni, Mo, Cr and Sn) and corresponding quantities of available scrap and crude steel demand are compared to investigate to which extend the crude steel demand could be covered by secondary resources.

1.3 Mercury as unwanted trace element

Mercury (Hg) is known to be harmful to humans and nature, and risks of exposure were investigated in many studies (e.g. recently reviewed by Bjørklund et al., 2017). The use of Hg in products or as process agent is highly restricted and monitored in most countries, first harmonized in the Minamata treaty UNEP, 2017a, which is the basis and harmonized outcome of many of these internationally implemented regulations to monitor and restrict Hg use. In accordance with the treatment, most products containing Hg or processes using Hg will phase out in the years to come. The remaining ones will be monitored closely. Summarized, the intended use of Hg will be highly restricted and will be subject to mandatory reporting.

In addition to intended Hg use, Hg is present in almost every natural material (Adriano, 2001). Therefore, industry sectors with high material throughput are prone to a higher Hg-throughput. High temperature production or transformation processes potentially release Hg from the material to the environment, which is also reflected in the Minamata Convention (UNEP, 2017a) by listing potential sources of emissions such as coal-fired boilers, smelting and roasting processes, cement plants and waste incineration plants. Hence, the industrial system is of particular interest in regards to monitoring and controlling Hg for multiple reasons: i) the material throughput is high and potentially results in high Hg throughput. ii) The composition of output material (product, by-product such as waste or emission) might differ highly from the input material (resource) due to substantial transformation (mechanical, chemical, thermal) during processing which leads to potential release of Hg from the materials. iii) Highly industrialized processes are highly monitored, meaning data existence and possible data availability to facilitate the investigation of Hg's fate in these processes. iv) The processes might present a good opportunity to remove Hg from the system, as it is done in some processes already (e.g. Hg remediation in combustion and industrial plants by means of flue gas cleaning).

A detailed industry wide *MFA* is a feasible way to understand the sources, pathways and the fate of Hg in the industrial system, which was applied in various studies (e.g. Jasinski, 1995; Krook et al., 2004). Recent studies on Hg flows focus on overall emissions to the environment either on industrial, national or global levels (e.g. Amos et al., 2013; CEIP, 2020; Fukuda et al., 2011; Glodek et al., 2010; Pacyna et al., 2010; UNEP, 2003, 2013, 2019; Wilson and Futsaeter, 2013; Won and Lee, 2012; Q. Wu et al., 2017; Y. Wu et al., 2006; Xu et al., 2017), or on static one year analyses (e.g. nationally for Poland (Panasiuk and Głodek, 2013), China (Hui et al., 2017), Denmark (Christensen et al., 2003), Turkey (Civancik and Yetis, 2018) and Austria (Reisinger, Schöller, Müller, et al., 2009) and for industry sectors: cement (e.g. for Austria (Lederer et al., 2017) and Germany (Achternbosch et al., 2003; Achternbosch et al., 2005; Harraß et al., 2018; MUNLV, 2005), metals (e.g. for zinc (Chung et al., 2017) and iron (Fukuda et al., 2011; F. Wang, Wang, Zhang, Yang, Gao, et al., 2016)) and coal combustion (e.g. globally (Mukherjee et al., 2008)). What all these analysis lack, is a multi-annual approach and analysis.

The case study in the present thesis modelled the Hg-flows throughout the Austrian manufacturing industry, where Hg is present as unwanted trace element by use of a multi-annual *MFA*.

Chapter 2

Objectives and thesis structure

The overall objective of this thesis is to investigate flows and stocks of two, in terms of usage, very different materials (mercury and steel) through our anthropogenic systems and to assess the challenges (e.g. availability, usability and reliability of data, interpretation and meaningfulness of results) when conducting material flow analysis for materials of different interest (important commodity versus hazardous pollutant).

In reach of the objectives given above, the following general research questions are addressed:

1. Which data sources are available for steel, its respective tramp element content and mercury?
2. To what extent does the availability of the data sources impact the respective material flow analysis system investigated?
3. Do the current data sources allow a continuous monitoring of material flows?
4. How should data sources be made available in order to be easily usable for material flow analysis?

The following questions regarding the case studies themselves (steel and mercury) are addressed:

5. How did the steel scrap flows on the territory of the EU develop and how will they develop until 2050?
6. How did the composition of steel scrap develop with regards to relevant tramp elements and what will the composition be in the future?
7. To what extent can steel scrap generated in the EU be utilized by the domestic industry (in the past and in future)?
8. Which measure might be necessary to increase the domestic production of scrap-based steel?
9. How much and through which paths does Hg move through the manufacturing industry in Austria?

10. Does the data availability allow deeper understanding of the unintended Hg flows in the Austrian manufacturing industry?

By the use of material flow analysis the steel flows (from steel products to scrap) in the former EU-28 from WWII (1946) up to today (2017) are investigated (multi-annual material flow analysis). Further, the future development of these steel flows is investigated via dynamic material flow analysis, thereby covering the period from 1910 to 2050. To investigate the recyclability of the available scrap from a quality point of view, each steel flow is assigned to a steel intermediate (flats, longs, bars, shapes and their sub-groups) and a designated end-use sector (construction, transport, metal goods, industrial equipment and their sub-sectors). Each combination of steel intermediate and sector is assigned to one of four quality classes. The quality classes are defined by their tramp element contents of η (Cu, Sn, Cr, Ni, Mo). Subsequently to the material flow analysis modelling, material pinch analysis is used to compare the quantity and qualities of steel scrap available with the demand of steel production in terms of quantity and quality. Thereby, material pinch analysis allows assessing to what extent the scrap available meets the crude steel demand in regards to quality requirements. Based on the retrospective analysis (1946 – 2017), the future development of scrap availability and its composition is assessed via dynamic material flow analysis and compared with the future crude steel demand.

Besides steel and therein contained tramp elements, the flows of (undesired) Hg, throughout the Austrian manufacturing industry is investigated. The static, retrospective multi-annual material flow analysis model covers the trace element and heavy metal Hg as throughput in the Austrian manufacturing industry for the period 2005-2016. The modelling is based on good flows (mostly based on economic material accounting), which are relevant to the Hg throughput. Hg present in machines and other equipment is not considered in this model. The good flows are assigned the corresponding Hg-concentration.

This thesis builds on three peer-reviewed and published papers, which can be found in the Appendix. The papers cover the main areas of the research questions given above. The first paper (Steel Scrap Generation in the EU-28 since 1946 – Sources and Composition, Dworak and Fellner, 2021) analyses the steel flows in Europe retrospectively. The second paper (How Will Tramp Elements Affect Future Steel Recycling in Europe? – A Dynamic Material Flow Model for Steel in the EU-28 for the Period 1910 to 2050, Dworak et al., 2022) analyses the steel flows in Europe since 1910 to model their further development until 2050. Both studies combined tackle the research questions 5, 6, 7 and 8.

The third paper investigates the Hg throughput in the Austrian manufacturing industry by means of a multi-annual material flow analysis (Mercury throughput of the Austrian manufacturing industry – Discussion of data and data gaps, Dworak and Rechberger, 2021) and thereby covers the research questions 9 and 10 and the parts of research questions 1 to 4, which concern mercury.

Based on the results and experience collected while conducting the case studies of the introduced papers, research questions 1, 2, 3 and 4 are discussed and completed.

Chapter 3

Methods

In the following chapter, the used methods are described. Material flow analysis (*MFA*) and uncertainty analysis was used to compile the mercury balance. Dynamic material flow analysis (*DMFA*), quality assessment, validation, material pinch analysis (*MPA*) and sensitivity analysis were used to analyse the steel flows.

3.1 Material flow analysis (*MFA*)

MFA, as described by Brunner and Rechberger, 2016, is widely used to map material flows, their sources and sinks and how they are connected. Generally, *MFA* allows to assess the flows and stocks of materials throughout a system that is defined in space and time. *MFA* is based on the law of conservation of matter and uses material balances to compare all inputs, stocks and outputs of each process which is part of the assessed system. The unknown flows can be calculated by the mass balance formula:

$$\Sigma_{i=1}^{k}\dot{m}_{in,i} = \Sigma_{j=1}^{l}\dot{m}_{out,j} \pm \dot{m}_{stock,j} \tag{3.1}$$

with $\dot{m}$ given in mass per time unit. $\Sigma_{i=1}^{k}\dot{m}_{in,i}$ the total mass input $\dot{m}_{in}$ of k input flows i, $\Sigma_{j=1}^{l}\dot{m}_{out,j}$ the total mass output $\dot{m}_{out}$ of i output flows j, and $\dot{m}_{stock}$ represents a potential flow from or to a stock located in the process itself.
Otherwise, transfer coefficients can be used to calculate unknown flows:

$$TC_j = \frac{\dot{m}_{out,j}}{\Sigma_{i=1}^{k}\dot{m}_{in,i}} \tag{3.2}$$

where the transfer coefficient TC_j of an output flow j, $\dot{m}_{out,j}$ is its mass relative to the total mass input $\Sigma_{i=1}^{k}\dot{m}_{in,i}$ of k flows i. The transfer coefficients can be calculated based on the same equation if all flows are known.

The method can be applied to goods from a rather economic level, like commodities (as in this thesis done for steel) and/or for specific chemical compounds or elements (as in this thesis done for Hg and the tramp elements present in steel flows).

DMFA is a specific type of *MFA* which considers stock accumulation and depletion within processes based on lifetime functions.

3.1.1 Uncertainty analysis

Uncertainty analysis was conducted for the case study on Hg only. For that, a normal distribution was chosen, as applied by e.g. Lederer et al., 2017; Reisinger, Schöller, Jakl, et al., 2009.

To quantify concentrations and corresponding uncertainties, secondary data was screened. Depending on availability, multiple data points were combined to estimate a range for the concentration in specific goods. Outliers were identified based on the distribution and reliability of the values (considering description, source, sample size) and eliminated from the used database. The minimum and maximum of these estimates were set as standard deviation for the concentration of each flows. The data for goods was mainly obtained from official reporting (industrial reports and/or economic accounting). Due to lack of uncertainty estimates, uncertainty was estimated considering data quality requirements, based on Laner et al., 2016; Laner et al., 2014. Error propagation was conducted as described by Brunner and Rechberger, 2016.

3.2 Quality assessment of steel flows

The quality of steel and scrap was assessed via the level of tramp element contamination of five major tramp elements (Σ of Cu, Sn, Cr, Ni and Mo). The considered levels are shown in Tab. 3.1. Considering the sum of tramp elements for quality assessment was preferred to single elements, as some of the elements might have super positioning effects on the workability of steel (e.g., Daigo et al., 2020; Kim et al., 2003; Lee et al., 2004). Also, unlike to Japan (e.g., Daigo, Fujimura, et al., 2017; Daigo and Goto, 2015; Nakamura et al., 2017; T. Oda et al., 2010), detailed information about the actual presence of specific tramp elements in specific steel products or grades is not available for the European steel market. Therefore, this semi-quantitative consideration was chosen.

Tab. 3.1: *Steel qualities (tolerable content of tramp elements) considered, adopted from Dworak and Fellner, 2021*

Max. content of tramp elements Σ (Cu, Sn, Cr, Ni, Mo) in %	Quality categories	Typical steel intermediates
<0.18	Q1	most flat products (cold rolled coils) – deep drawing quality, interstitial-free steel
0.18 – 0.25	Q2	tubes, plates, hot rolled products in construction, wire rod (other than construction)
0.25 – 0.35	Q3	hot rolled bar, plates (construction), wire rod (construction)
> 0.35	Q4	heavy section, light section, rail section, reinforcing bar, hot rolled bar (construction)

The quality classification (see Tab. 3.1) is based on several studies (Bjørklund et al., 2017; Daehn et al., 2017; Noro et al., 1997; Schrade et al., 2006; Toi et al., 1997). High alloyed and stainless steel is not considered in this study.

3.3 Model validation for steel flows

Bottom-up assessment with independent data was used to partially validate the two steel models. For the retrospective model, the modelled input into the sectors cars (*T Ca*) and trucks (*T Tr*) was compared with production data of cars and trucks. The prospective approach was partially validated via stock assessment of reinforcing bar (*b RB*) based on independent data, and compared with the modelled stock of *b RB* in the sector buildings (*C Bu*). The same approach was followed for cars, where registration data on cars was compared with steel stock in the sector *T Ca*.

Further, a top-down approach was chosen to validate scrap quantities becoming available (modelled *PoCS* compared with reported).

3.4 Material pinch analysis (*MPA*)

Linnhoff and Hindmarsh, 1983 developed Pinch Analysis originally to optimize (minimize) energy demand in industries. The concept was further developed to be applied on material flows (*MPA*, e.g. Daehn et al., 2017; Ekvall et al., 2014; Hatayama et al., 2009, 2012), taking into account the requirements of different levels of purity for different processes and application of specific materials.

MPA is used in this thesis to analyse the results of the steel *MFA* per annum to asses how well the scrap availability meets the crude steel demand, considering tramp elements in scrap and tolerance of those in crude steel for specific applications.

3.5 Sensitivity analysis

The prospective *DMFA* was also analysed for its sensitivity to investigate the robustness of the models. Target parameter of the analysis was the surplus of low purity scrap (Q3 & Q4) in relation to the crude steel demand of this purity. The analysis was carried out for each year of the model (1910-2050), whereas the results are discussed for the last 5 decades (since 1980) up to 2050. The following parameters where adapted.

- *Sector split*: for the future prediction (2019-2050) we assumed varying splits of two end use sectors (construction and transport). We considered a variation of ±10% (relatively) for construction and transport (marked as “C+”, “C-“, “T+”, “T-“), each separately, while the remaining sectors splits were scaled accordingly.
- *Average lifetime*: a variation of the average life time ±20% (marked as “upper” and “lower”) was applied.

- *Export rate of end-of-life products*: The export rate of EoL products was varied by ±20% relative to the base value (marked as “upper” and “lower”)
- *Recovery rate of post-consumer scrap generated*: a reduction in losses of 20% relative to the base value (marked as “upper”) was assumed. As the efficiency of the waste management system in the future will rather increase than decrease, only an increase in recovery rates was considered.

Chapter 4

Design of case studies

4.1 European steel cycle

4.1.1 Model design

The European steel flows, covering carbon steel and cast iron and steel, were investigated with two approaches. First, a retrospective, multi-annual material flow analysis (*MFA*) was undertaken. It was used to investigate the composition of scrap available in the investigated period (1946-2017). It is mainly build on reported data and balancing thereof. Second, dynamic material flow analysis (*DMFA*) was used to investigate the future development of steel flows in Europe, projected until 2050. The designed models are slightly different, mainly concerning the system boundaries (see Fig. 4.1). The retrospective assessment considers the reported back-flow of post-consumer scrap (*PoCS*), whereas the *DMFA* (prospective) also considers the use itself and various import and export flows between delivery-to-gate and the resulting back-flow of scrap. The common aspects of the models for the case studies are described in the following. The details which differ are described in the subsequent sections.

The models are build up in two layers, the intermediate steel products and the steel end-use sectors. Altogether 19 intermediate steel products and 10 end-use sectors are distinguished according to Cullen et al., 2012 and Zhu et al., 2019, and summarized in Tab. 4.1 and Tab. 4.2.

For the distribution of the steel intermediates to the different end-use sectors intermediate steel product and end-use sector specific transfer coefficients were applied. Similarly, fabrication scrap

Tab. 4.1: *End-use sectors considered, according to Cullen et al., 2012; Zhu et al., 2019*

End Use Sectors	abbreviation	Grouped End-Use Sectors
Buildings	C Bu	Construction
Infrastructure	C In	
Cars	T Ca	Transport
Trucks	T Tr	
Other Transport	T OT	
Mechanical Engineering	I ME	Industrial Equipment
Electrical Engineering	I EE	
Other Metal Goods	MG OMG	Metal Goods
Appliances	MG Ap	
Packaging	MG Pa	

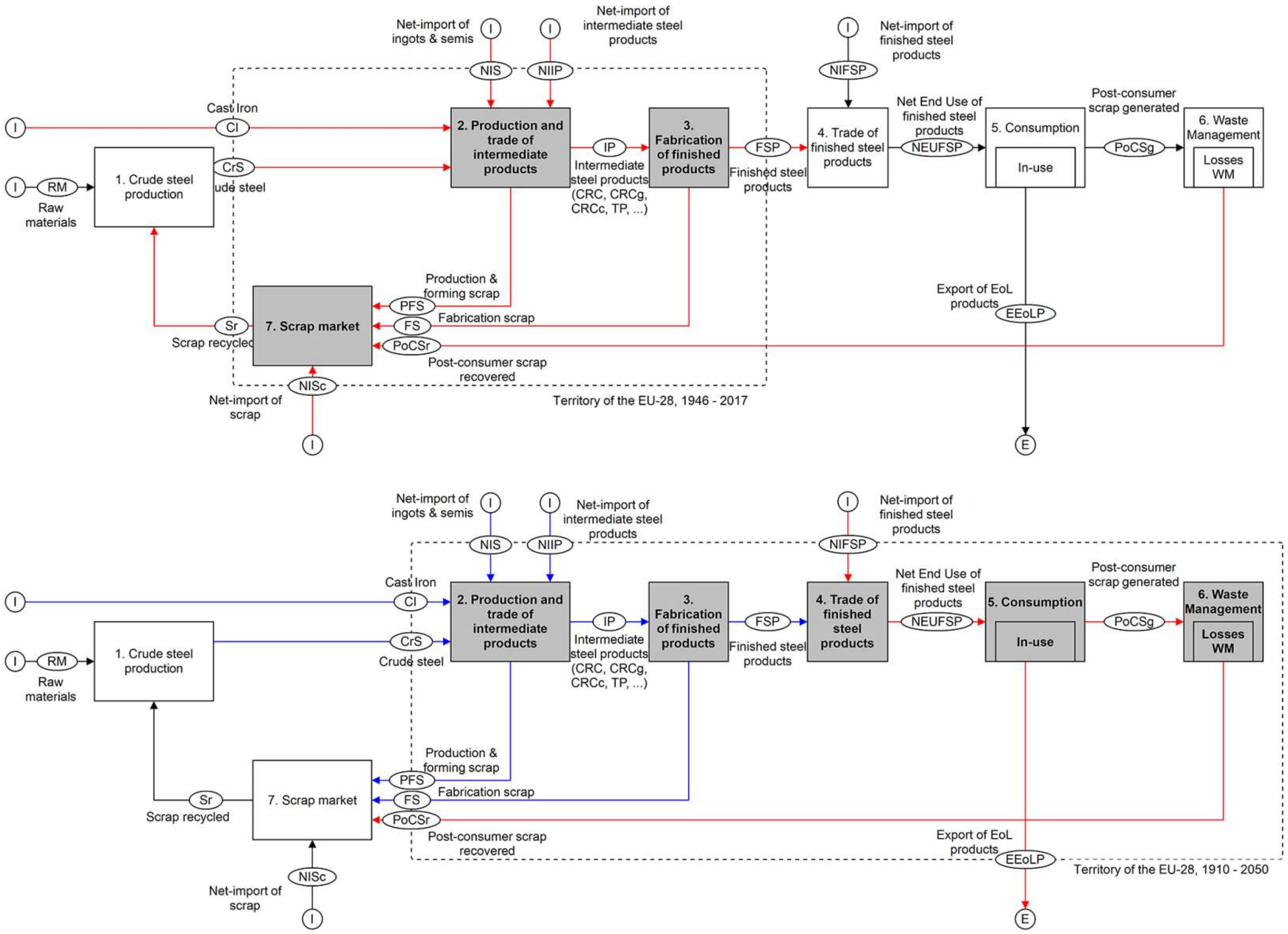

Fig. 4.1: *MFA system for assessing steel scrap flows in the EU-28 retrospective (left) and prospective (right), red steel flows are determined, blue flows are adopted from retrospective model, black flows are shown for completeness. Adapted from Dworak and Fellner, 2021; Dworak et al., 2022*

Tab. 4.2: *Steel Intermediates considered, according to Cullen et al., 2012; Zhu et al., 2019*

Intermediate Steel Products	abbreviation	Grouped Intermediate Steel Products
Cast steel	c CS	casts
Cast Iron	c CI	
Electrical Strip	f ES	flats
Tin Plated	f TP	
Plate (excl. plates used for welded tubes)	f P	
Cold Rolled Coil galvanized	f CRCg	
Cold Rolled Coil coated	f CRCc	
Cold Rolled Coil	f CRC	
Hot Rolled Coil galvanized	f HRg	
Hot Rolled Narrow Strip (excl. Strips used for welded tubes)	f HRNS	
Hot Rolled Coil	f HRC	
Welded Tubes	t WT	tubes
Seamless Tubes	t ST	
Wire Rod	b WR	bars
Reinforcing Bar	b RB	
Hot Rolled Bar	b HRB	
Heavy Section	s HS	shapes
Light Section	s LS	
Rail Section	s RS	

(*FS*) was calculated based on end-use sector and intermediate steel product specific material efficiency rates.

The domestic net-end-use of finished steel products (*NEUFSP*) is determined by balancing the process *4. Trade of finished steel products* based on the domestic fabrication of the finished steel products (*FSP*) and the trade thereof (net-import of finished steel products (*NIFSP*)). For net-import of finished steel products (*NIFSP*), the same sectoral composition (shares of steel intermediates per sector) as finished steel products (*FSP*) for the corresponding year was assumed. The amount of end-of-life products (*EoLP*) was determined on the basis of Weibull lifetime functions, depending on the corresponding end-use sector. Export of end-of-life products (*EEoLP*) is relevant for the transportation sector (cars (*T Ca*) and trucks (*T Tr*)) and is considered to determine post-consumer scrap generated (*PoCSg*). Losses due to processing in the waste management system (*WMS*) are considered to determine post-consumer scrap recovered (*PoCSr*) and are assigned sector-specific. It is assumed that losses and scrap are composed identically, meaning shares of intermediate products (*IP*) are the same for each sectoral output of *PoCS* and the corresponding losses. *PoCSr* is considered the output of the *WMS* and is then available for recycling or trade.

4.1.2 Data

In particular, the following steel flows are based on statistical data: crude steel (*CrS*), intermediate products (*IP*), net-import of ingots and semis (*NIS*), net-import of intermediate products (*NIIP*) and net-import of finished steel products (*NIFSP*). For the retrospective approach, also scrap recycled (*SR*) is based on reported data, whereas the other flows were determined either by balancing the single processes or applying transfer coefficients. In the prospective approach, *EoLP* (not shown in Fig. 4.1, represented by sum of *PoCSg* and *EEoLP*) and *PoCSg* are determined by applying Weibull lifetime distributions for the end-use phase. If not otherwise stated (e.g., recovery rate) the future projection from 2018 onwards is based on the 15 year average (considered period: 2003-2017). This applies mainly to data retrieved from statistical reporting (e.g., *CrS*, *IP*, *NIS*, *NIIP*, *NIFSP*, and the sectoral split of *NEUFSP*).

Reported data: The raw data was retrieved from official reports (Eurostat, 1970, 1977, 1985, 1994, 1998, 2002; UN Comtrade, 2020; World Steel Association, 2018, 2020). The data series were checked for plausibility and completed (extra- or interpolated).

Transfer coefficients: Transfer coefficients for the distribution of steel intermediates to specific end-use sectors were derived from Cullen et al., 2012 and calibrated based on EG, 1976, 1985, 1990; Eurofer, 2018. Fabrication losses (represented by *FS*) were assessed and applied, depending on according end-use sector and according steel intermediate, as provided by Cullen et al., 2012. As the material efficiency increased, and therefore fabrication losses decreased in the recent decades (see Pauliuk, Wang, et al., 2013), the fabrication losses were calibrated accordingly. *EEoLP* was assessed for *T Ca* and *T Tr* based on Oeko-Institut, 2017. To determine *PoCSr*, sector specific recovery rates are applied, based on EC, 2000; Pauliuk et al., 2019; UBA and BMU, 2019. Is was assumed, that the recovery rate increases, represented in the dynamic model in an constant increase from 2010 onwards. A summary of the input parameters are provided in 4.3.

Lifetime functions: End-use sector specific Weibull-distribution are used as lifetime-functions. Even though the lifetime is changing over time, there is no sector specific lifetime variation available. Therefore, the shape (shape: 5) and average lifetime is kept constant over the investigated period. The assumed lifetimes are provided in Tab. 4.3 and are derived based on Dahlström et al., 2004; Davis et al., 2007; Gauffin and Pistorius, 2018; Hatayama et al., 2010; Huuhka and Lahdensivu, 2016; Melo, 1999; Michaelis and Jackson, 2000; Mueller et al., 2011; Mueller et al., 2006; Neelis and Patel, 2006; J. Oda et al., 2013; Pauliuk, Milford, et al., 2013; T. Wang et al., 2007.

4.1.3 Steel and scrap quality assessment

For all modelled new scrap flows (production and forming scrap (*PFS*) and *FS*), the steel qualities with respect to the content of tramp elements were assigned according to the intermediates and end-use sector they arise from. For the *PoCS* in the retrospective *MFA* approach (reported data), Q3 and Q4 (each 40%-60%) was assumed, based on various studies Daehn et al., 2017; Davis et al., 2007; Pauliuk, Milford, et al., 2013; Savov et al., 2003; Schrade et al., 2006; Toi et al.,

Tab. 4.3: *Input parameters for the dynamic MFA model: lifetimes, EEoLP-rate, recovery rate*

End-use sector	Lifetime	EEoLP	Recovery rate	
	Average lifetime	Rate	Up to 2010	2050
C Bu	65	-	82%	87%
C In	65	-	82%	87%
T Ca	17	30%	82%	98%
T Tr	17	70%	82%	98%
T OT	55	-	82%	87%
I ME	17,5	-	87%	91%
I EE	15	-	87%	91%
MG OMG	14	-	58%	71%
MG Ap	14	-	58%	71%

1997; Wagner et al., 2012; Willmann et al., 2017. In the prospective *DMFA* model the three following options were assessed for the quality of *PoCSg*:

- *Option A*: The quality class was assigned based on the sector the *PoCS* arises from. The quality classes are based on literature data on the content of tramp elements (including contamination due to e.g. treatment in the *WMS*) and represents the usage of scrap without enhanced sorting or decontamination procedures in place. This scenario is considered the worst-case scenario investigated in this case study.

- *Option B*: The average tramp element content of each sector is determined and the according quality class is assigned. Similar to Option C no contamination is considered.

- *Option C*: The *PoCS* is sorted in the assigned quality classes. Dismantling and clean sorting of all parts is assumed to be possible and conducted. No contamination (e.g., copper cables) was considered. Option C qualifies as best-case scenario.

4.1.4 Model validation

For both models (retrospective *MFA* and prospective *DMFA*), several approaches based on independent data were used to validate the model at least partially.

The retrospective *MFA* was validated by bottom-up data of two sectors. Production data of cars & trucks and their respective average weight and steel content (Castellani et al., 2017; ICCT, 2011; Todor and Kiss, 2016) was compared with the modelled steel inputs in the respective sectors.

The prospective *DMFA* was validated by bottom-up stock assessment of *b RB* in buildings based on average concrete use and average steel use in reinforced concrete mostly based on Nemry et al., 2008. Further, a bottom-up stock assessment of cars, based on registered cars and average weight (Eurostat, 2021; Todor and Kiss, 2016), was applied. The model was also validated by comparing the modelled and reported scrap available, based on the scrap available retrieved from reported data (Eurostat, 1970, 1977, 1985, 1994, 1998, 2002; UN Comtrade, 2020; World Steel Association, 2018, 2020).

4.2 Mercury throughput in Austrian manufacturing industry

4.2.1 Model Design

The static, retrospective multi-annual model covers the trace element and heavy metal Hg as throughput in the Austrian manufacturing industry. The spatial boundaries are the Austrian state borders and the model extends over the years 2005-2016. The modelling is based on flows of goods (mostly based on economic accounting) which are relevant to the Hg throughput. Hg in machines and other equipment is not considered in this model. The processes in the developed model are grouped in three blocks: inputs (primary raw materials and fuels (*PRM-F*), secondary raw materials and fuels (*SRM-F*), water supply (*WAT*)), transformation (industrial sectors energy generation (*EG*), chemical industry (*Ch*), metal industry (*MET*), non metallic mineral industry (*NMM*), wood pulp and paper industry (*WPP*), other industry (*OTH*)) and outputs (emissions to air (*EM*), products (*P*), water supply (*WAT*), waste water (*WW*)). The sectors were chosen in accordance to the norm reporting format for air pollutant emission inventory (*NRF*) and NACE reporting categories. The flows are connecting all input collector-processes with the industrial sectors, whereas *PRM-F* and *SRM-F* are sub-categorised in raw material (*RM*) and fuels (*F*). Transformed flows (*EM*, *P*, *WAT*, *WW*) are then connected to the corresponding collector-processes in the output block. The flows are composed of fraction flows, to account for the heterogeneous composition of the flows (especially regarding *SRM-F*, but not only). The weighted Hg-concentration of the flows was calculated via Eq. 4.1. Solely *EG* does not provide products, as the product is energy, not matter. International trade flows play a major role in a small country as Austria. Therefore, the direct inter-correlation between the domestic sectors was represented only indirectly via the export of wastes & products out of the system and the import of raw material into the system.

$$c_j = \frac{\Sigma_{i=1}^{k} c_i \dot{m}_i}{\Sigma_{i=1}^{k} \dot{m}_i} \tag{4.1}$$

EG solely consists of energy generation plants, without waste incineration plants. Waste incineration plants are for one not part of the manufacturing industry and furthermore rather irrelevant to the all over Hg-emissions due to strict regulations in place, which is reflected in only about 2% Hg-emissions of overall emissions to air (Anderl et al., 2017; CEIP, 2020). The sector *Ch* refers to the oil refinery. For other parts of the chemical industry (e.g. plastic industry, pharmaceutical industry) sufficient data was not available. Therefore, only fuel was considered. *MET* includes relevant primary production (ferrous, tungsten) and coke production. Non-ferrous secondary production was assumed to be not relevant in regards to Hg-flows. *NMM* is composed of the cement industry, the brick and tile industry and the glass industry. The wood (timber and board production) and pulp & paper industry is summarised in *WPP*. *OTH* consolidates other industries (e.g., textiles and the food industry). In this process, similarly to *Ch*, mainly the consumed fuels were considered. The model can be seen in Fig. 4.2.

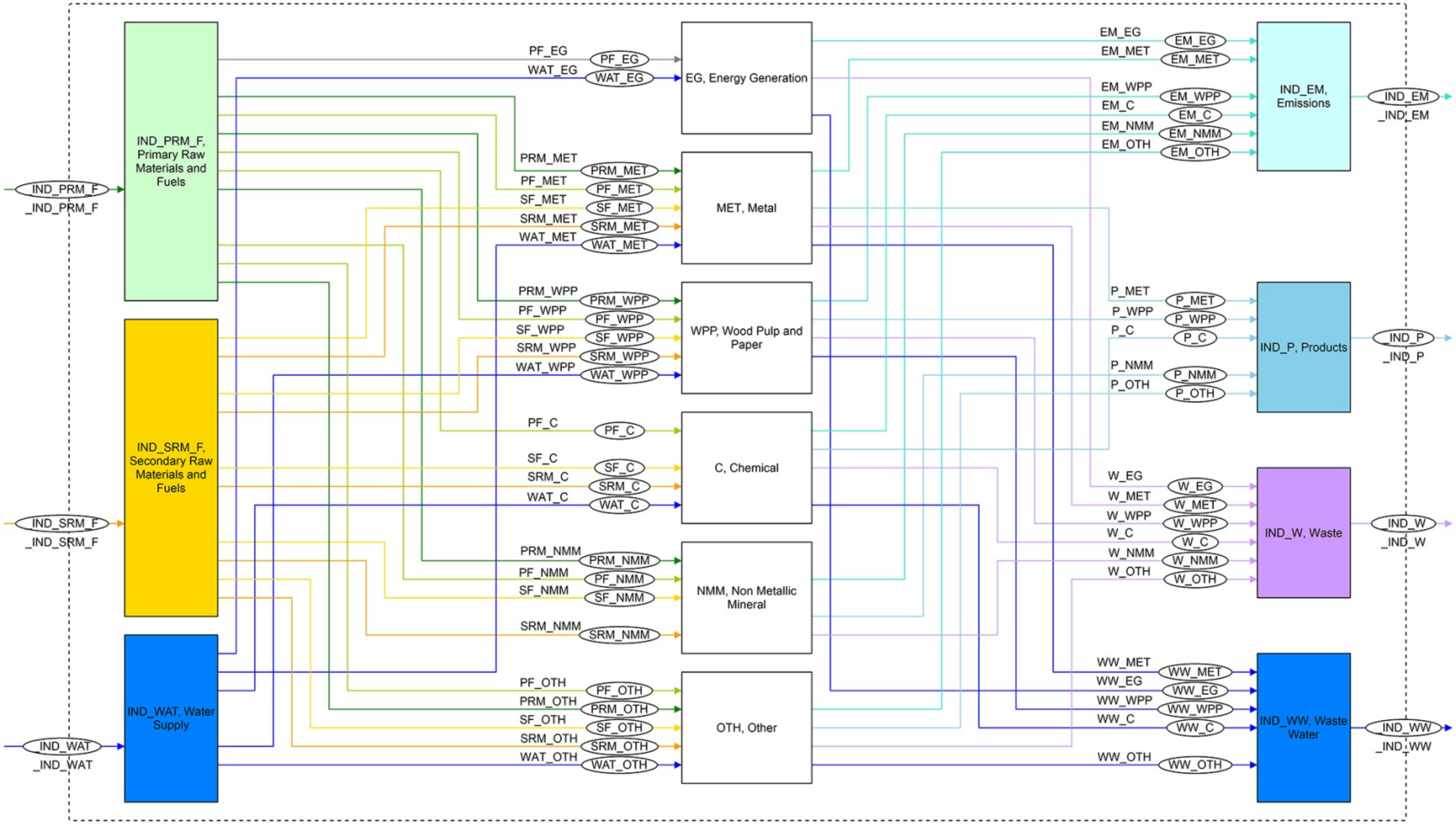

Fig. 4.2: *Hg throughput for the Austrian manufacturing industries: process and flow names (abbreviations: IND (summed up import and export flows); import main flow types: PRM (primary raw materials), PF (primary fuels), SRM (secondary raw materials), SF (secondary fuels), WAT (water supply); export main flow types: EM (emissions), P (products), W (wastes), WW (wastewater), adopted from Dworak and Fellner, 2021*

4.2.2 Data

The Hg flows are determined by applying good-specific Hg-concentrations on the reported good flows. The single exception is emission to air, where the absolute Hg-emissions to air are reported and applied directly (CEIP, 2020). The good flows are taken from official reporting data (economical accounting), from official statistics (Statistik Austria or eurostat) or from industry reports (e.g., Austropapier, 2018; Fachverband der Glasindustrie, 2019; Mauschitz, 2012; Pfeiler and Gradischnig, 2019). Hg-concentrations are derived from various sources, mainly peer-reviewed literature (e.g, Ali and Al-Qahtani, 2012; Bai et al., 2017; Diao et al., 2018; Fukuda et al., 2011; F. Wang, Wang, Zhang, Yang, Wu, et al., 2016; Q. Wu et al., 2017; Yang et al., 2018), but also scientific reports (e.g., Achternbosch et al., 2003; AGES, 2016; Christensen et al., 2003; Deutsch et al., 2012; Harraß et al., 2018; MUNLV, 2005; Szednyj and Schindler, 2004; Taverna et al., 2010; UNEP, 2017b; Wilhelm, 2001). Where applicable, the waste characteristic database ABANDA (LANUV, 2019) was consulted.

Chapter 5

Results and Discussion

In the present chapter the results of the case studies are summarized and further analysed regarding data availability and handling. In the first part, the European steel cycle is discussed. The second part deals with the Hg-throughput in the Austrian manufacturing industry. The third part contains the analysed data availability and quality in regards to the materials investigated.

5.1 The European Steel Cycle

The two approaches (retrospective *MFA* and prospective *DMFA*) for modelling the European steel cycle match quite well for the overlapping period, as one of the validation approaches for the *DMFA* shows (for details see 5.1.4). Therefore, in this summary, the results of both models are discussed collectively for for the most part.

5.1.1 Scrap available

The total amount of available scrap (sum of *PFS*, *FS* and *PoCSr*) constantly increases (from 20 Mt/yr in 1910 to 130 Mt/yr in 2050). The rate of *PFS* decreased tremendously due to the introduction of continuous casting between the 1970s and 1990s. The amount of *FS* has stayed rather constant since the 1970s. The amount of *PoCSr*, on the other hand, increases constantly. Accordingly, the share of new scrap (*PFS* and *FS*) in relation to total scrap available decreases constantly, where *PoCSr*'s rate increases from about 30% in the late 1940s to 60% nowadays (2020) and even reaching more than 70% in 2050.

5.1.2 Scrap quality

Since the 1970s, new scrap (*PFS* and *FS*) is composed mostly of high purity fractions (nowadays 55% Q1 and 25% Q2). The high share of high purity scrap is mainly contributed to flat products, which represent a major part of production volume and have a low material efficiency, which leads to higher *FS* yields. Hence, especially *FS* has a rather high share of Q1 scrap.

Old scrap (*PoCS*) on the other hand is, as currently handled (e.g. little sorting, no decontamination), mainly composed of low purity scrap of the classes Q3 and Q4. As mentioned above, the share of old scrap is constantly growing, which leads to majority shares of low purity scrap. Provided that the scrap handling scheme stays as it is currently (option A), these low purity

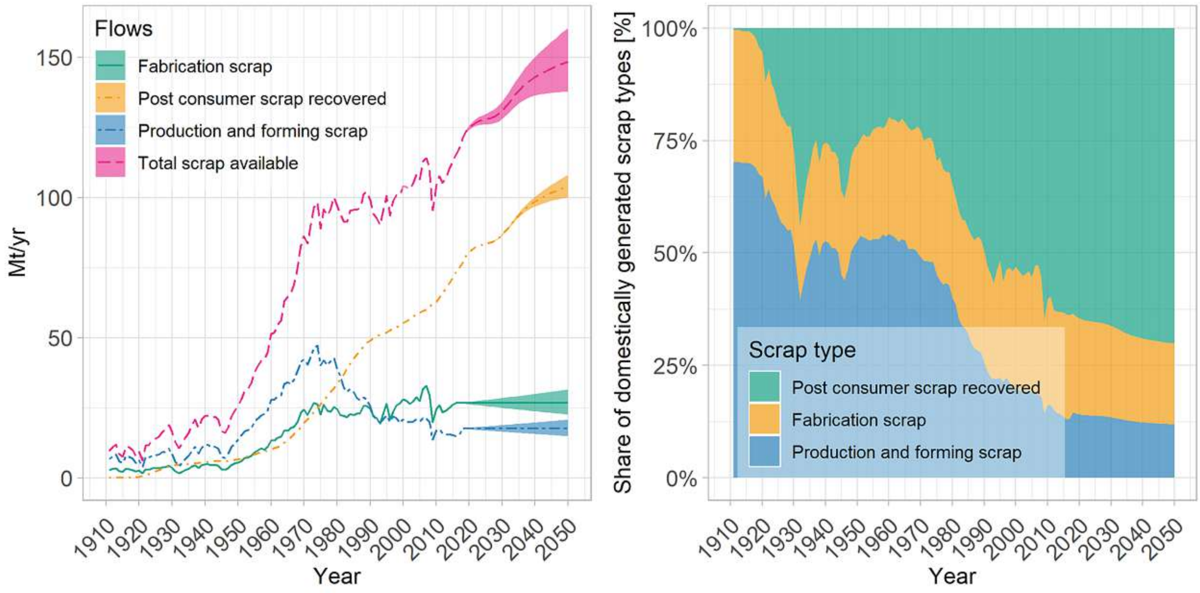

Fig. 5.1: *left: Modelled quantities of scrap flows becoming available. The ribbon indicates the three scenarios (upper bound: growth; line: zero growth; lower bound: de-growth); right: Shares of scrap types. Adopted from Dworak et al., 2022*

fractions of scrap already exceed the crude steel demand of these purity (see Fig. 5.3, based on *DMFA*). This is also reflected in the results of the m*MPA* (Fig. 5.4 and Fig. 5.6).

The determined scrap surplus of low purity scrap (Q3 & Q4) correlates with the net export of scrap (acc. to UN Comtrade, 2020) since the 2000s, when there is a higher availability of low purity scrap than crude steel demand (positive surplus). This allows the conclusion, that most of the low purity scrap is rather exported from the EU than utilised. Hence, it seems that interventions, such as dilution or enhanced sorting, to utilize scrap are currently not exploited to a significant extent.

The current practise in combination with the projected development of scrap amount and quality leads to two main issues. First, the share of suitable scrap (high purity, Q1 & Q2) is and will be decreasing, which means a smaller share of available scrap can be used, as shown in the *DMFA*. Hence, the scrap rate in steel production stagnates. Second, the demand of low purity scrap in regions beyond EU borders is only present as long as the receiving economies are still growing. At some point these economies will provide there own low purity scrap and will no longer be able or willing to handle the European low purity scrap.

The prospective *DMFA* shows that the surplus of low purity scrap is increasing, as more and more old scrap arises. If the handling of scrap continues in the current way (option A in *DMFA*), the surplus of low purity scrap increase even further. Also, the dilution potential will be exhausted by 2040 (without regarding eventual higher tramp element contents due to a higher share of tramp element loaded scrap and lower share of virgin steel).

Post-consumer scrap was modelled according to three options and all of them, as well as the sectoral composition of *PoCSr* can be seen in Fig. 5.2. For option A, the sector-specific quality classification of post-consumer steel scrap, the vast majority of *PoCSr* is of low purity. Only

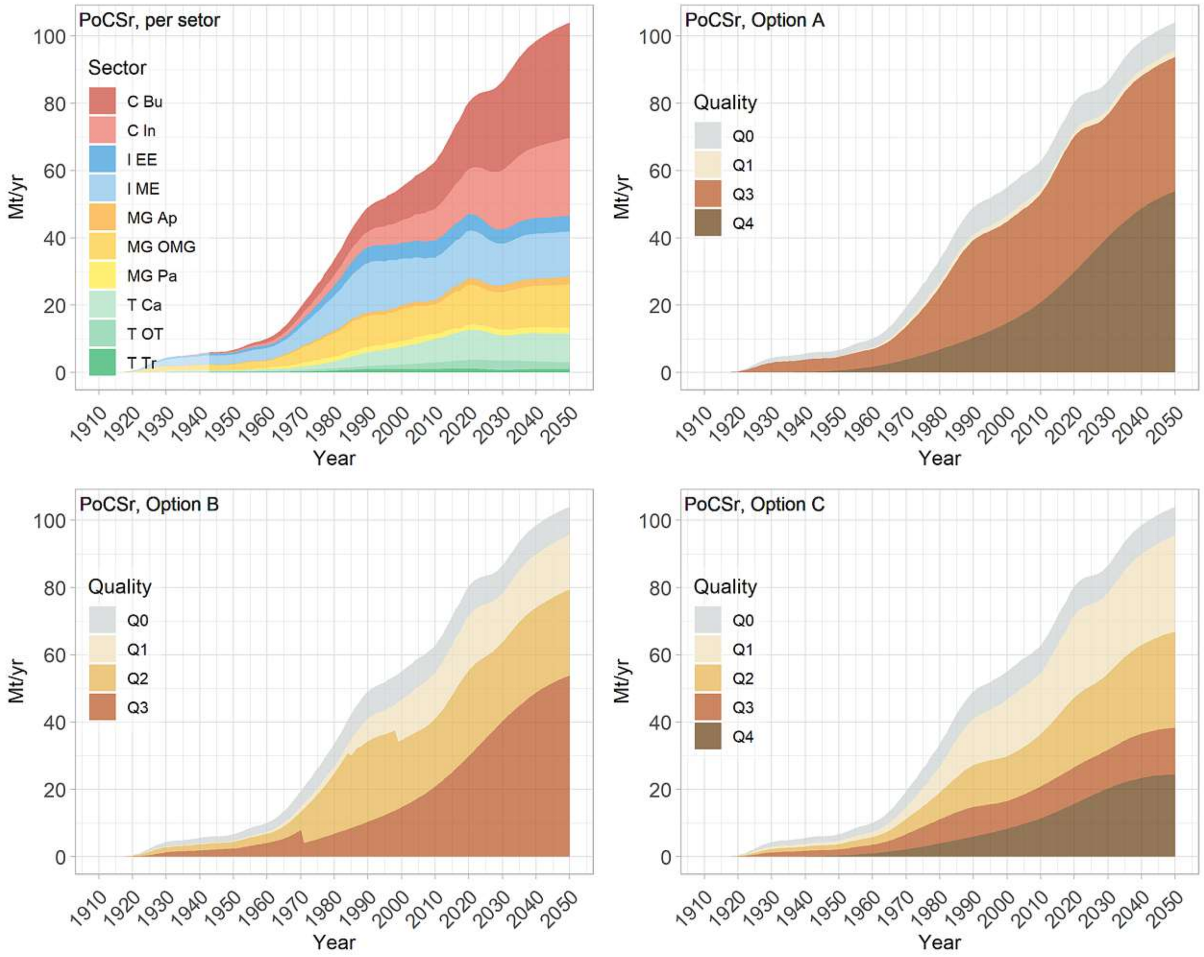

Fig. 5.2: *Post-consumer scrap recovered with sector origin (upper left), with quality classification according to option A (purity based on sector of origin, upper right), option B (average purity per sector based on inputs, no contamination considered, lower left), option C (purity of input = purity of output, lower right). Abbreviations of sectors: Construction: C Bu - Buildings, C In – Infrastructure; Industrial Equipment: I ME - Mechanical Engineering, I EE - Electrical Engineering; Transport: T Ca - Cars, T Tr - Trucks, T OT - Other Transport; Metal Goods: - MG OMG: Other Metal Goods, MG Ap - Appliances, MG Pa - Packaging. Adopted from Dworak et al., 2022*

separately collected food packaging can be recovered at quality class Q1 (see 5.3). The other streams of post-consumer scrap becoming available are classified as Q3 and Q4, whereas the share of Q4 is constantly increasing, driven by the increasing share of *PoCSr* from the sector construction (*C*).

Option B: The results for this approach are by definition somewhere in between the results of the two other options (A and C). Still, some results are remarkable: Even though some sectors (especially infrastructure (*C In*) and *C Bu*) receive most of the lowest steel quality class (Q4), the summed-up tramp elements of the sectors correspond to Q3. It should, however, be mentioned that the calculated average concentration is close to the threshold to Q4. The same principal applies to the higher share of Q1 in comparison to option A. The calculated mean values for the Q1 scrap fractions are rather close to the threshold for Q2. The steps in quality class changes can be attributed to the sharp distinction between the quality classes, which means that if the input quality of a sector changes, the whole subsector might switch to another quality class (as e.g. MG OMG from 1992 to 1993 and I ME from 2020 to 2021, with both switching from average Q2 to Q1). Specific data about the concentration of the tramp elements in the different scraps is provided as a spreadsheet in the Supplementary Information.

For option C, the post-consumer scrap exits the consumption process of the same quality as the intermediate steel products comprising the final steel products. Scrap of quality Q4 is mainly delivered and subsequently yielded as scrap by the Construction sector, therefore the share of Q4 is constantly rising similar to the yield of the scrap from the Construction sector, even if less intensively. The share of Q2 is rather constant (slightly above 25%).

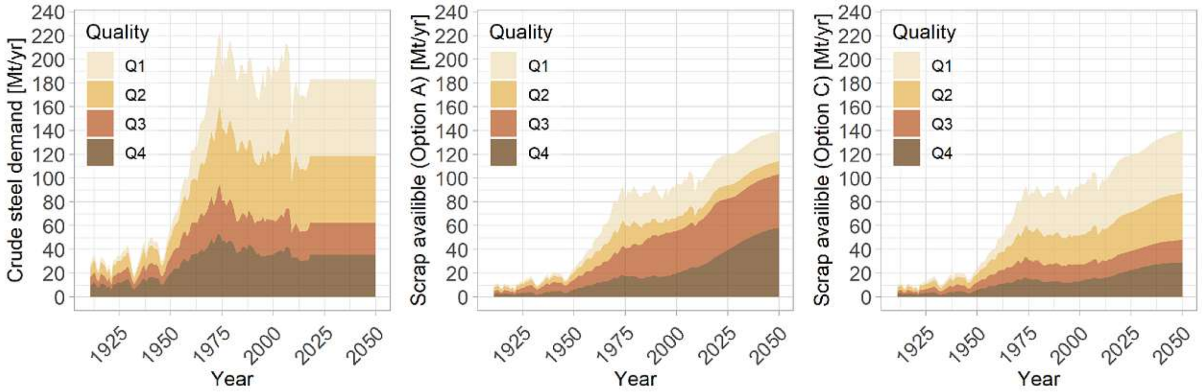

Fig. 5.3: *Crude steel demand taking quality classes for crude steel demand into account (left), scrap available with quality classes based on option A (middle) and option C (right). Adopted from Dworak et al., 2022*

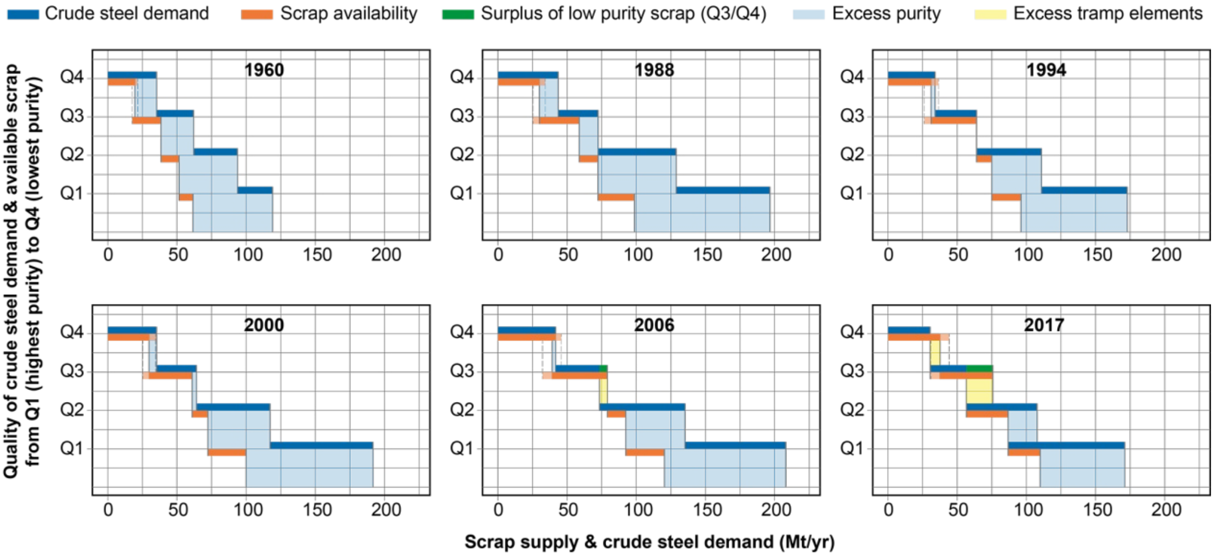

Fig. 5.4: *Material pinch analysis for the quantities and qualities of crude steel demand and available scrap in the EU for the years 1960, 1988, 1994, 2000, 2006 and 2017, orange-shaded areas show the excess tramp elements, blue-shaded areas the excess purities . Adopted from Dworak and Fellner, 2021*

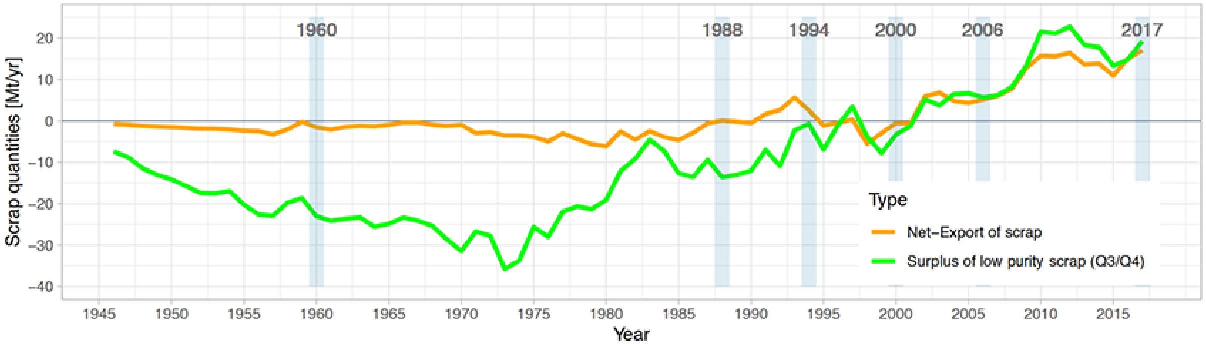

Fig. 5.5: *Assessed scrap surplus of categories Q3 & Q4 and net-export of scrap from the EU (data are given in Mt/yr). Starting from the time (mid-90s) when a surplus of low purity scrap (positive values) occurred, a good match with the net-export of scrap can be observed. Adopted from Dworak and Fellner, 2021*

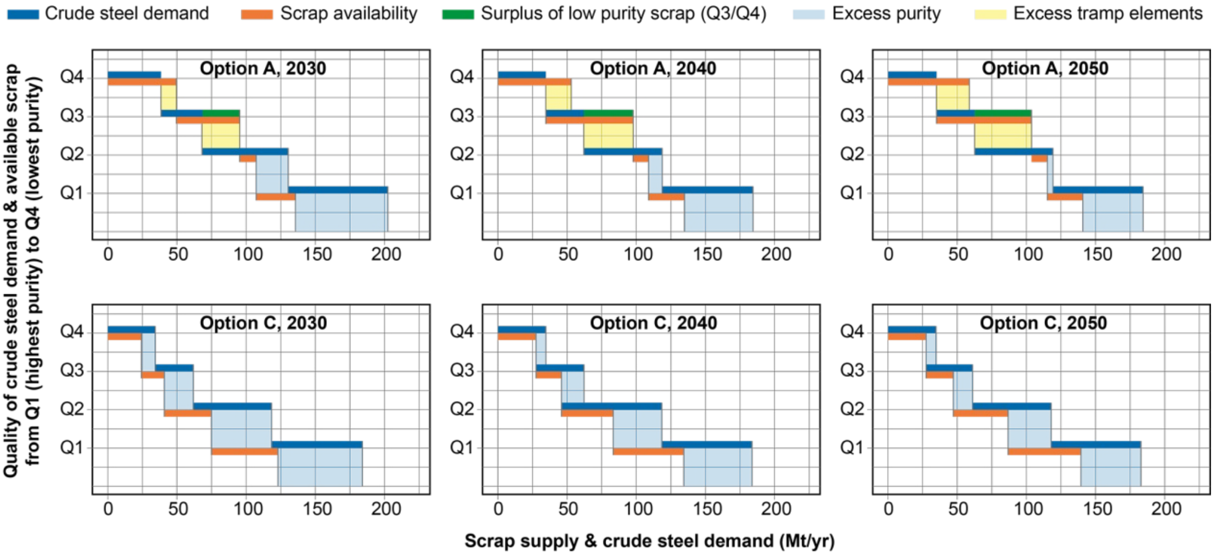

Fig. 5.6: *Material pinch analysis for the quantities and qualities (purities) of crude steel demand and available scrap in the former EU-28 for the years 2030, 2040, 2050. Adopted from Dworak et al., 2022*

5.1.3 Stock development

The in-use stock increases constantly, from about 6.7 t/cap (3,000 Mt) in 1985 to 9.5 t/cap (4,700 Mt) in 2010 and further up to 10.5 t/cap (5,300 Mt) in 2050. The results are in line with various other studies (e.g. Mueller et al., 2011; Pauliuk, Milford, et al., 2013; Pauliuk, Wang, et al., 2013).

In regards to composition, the following development can be observed: The major part of the steel in-use stock can be allocated to the sector *C*, which covered around 70% in 1970s. The results suggest a further rise up to 80% until 2050. This sector is also the major driver for stock increase, as the lifetimes are rather high.

Quality wise, the largest share of in-use stock is assigned to Q4 (lowest purity). In 2050, roughly 50% can be assigned to low purity scrap (Q3 & Q4). The high share of flat steel intermediates (mainly Q1 & Q2) in the European steel portfolio is not reflected in the stock composition, which may be attributed to the comparatively short lifetimes and low material efficiency in the corresponding sectors.

5.1.4 Validation

Both steel models (*MFA* and *DMFA*) were quantitatively validated with independent data. In case of the *MFA*, production data of cars and trucks was used. The bottom-up data fits well with the steel delivered in these end-use sectors, even though validation data estimates slightly lower amounts (see Fig. 5.7). This may be attributed to additional flows into these sectors besides mere production, such as spare parts for maintaining or repairing cars. Due to rather uncertain data of steel content in vehicles, the uncertainty of the validation data is quite high (± 10% cars, ± 20% for trucks).

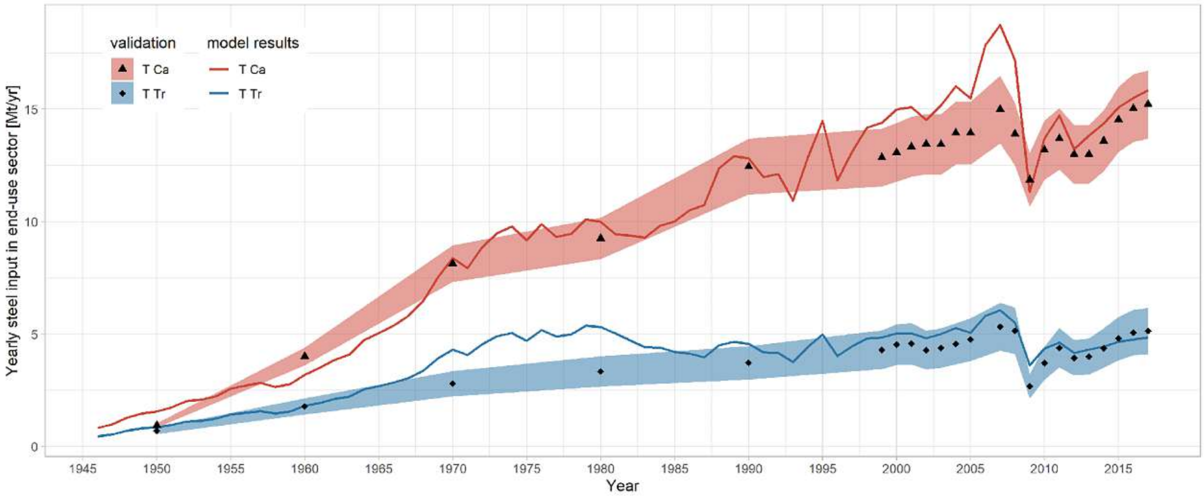

Fig. 5.7: *Comparison of annual steel flows into final cars (T Ca) and trucks (T Tr) manufactured in the EU-28 (model data are indicated by continuous lines; bottom-up data are indicated by dots and ribbons, which indicate the estimated uncertainty of the bottom up data). Adopted from Dworak and Fellner, 2021*

The *DMFA* was validated with two approaches. First, two bottom-up stock estimates based on independent data were compared with the present stock assessments. The steel content in cars in-use (based on statistics of registered cars, their weights and model results) are compared (see Fig. 5.8, most upper part). Especially until 2010, the bottom-up stock is underestimated, which may be attributed to cars which are not regularly registered (interchangeable number plates, non-registered cars on private property). Further, the steel in form of reinforcement bars in buildings was estimated based on reinforced concrete used for new construction and demolition statistics and compared with the models in-use stock of reinforcement bars in buildings (see Fig. 5.8, middle part). The development (increase and magnitude) of both approaches (model and bottom-up validation) are similar. Still, the model results slightly underestimate the steel content in reinforced concrete in comparison to the bottom-up stock assessment, which may be attributed to other steel intermediates used for reinforcement of concrete (e.g. hot rolled bars or wire rod).

Second, the *PoCSr* was compared with scrap available based on statistics (traded and used scrap). Fig. 5.8 (lowest part) shows that the reported scrap available fits well with the modelled scrap available. The underestimate during growth (until 1980s) can be at least partially attributed to the rather long life times of some steel products.

All in all, the validation approaches show, the rather simple and easily comprehensible modelling approaches still deliver fairly accurate results.

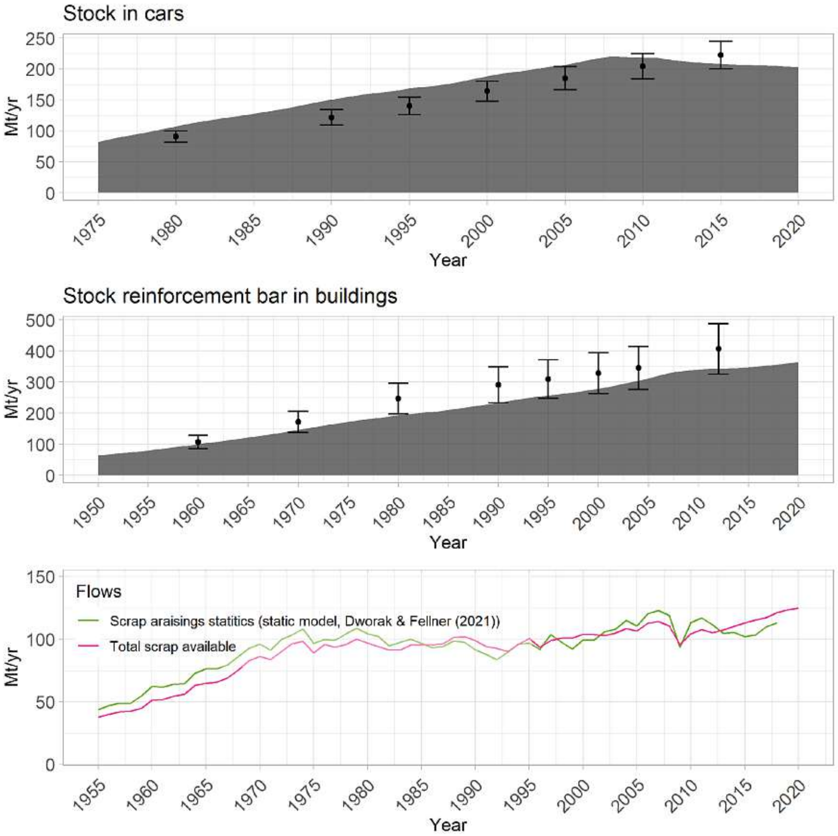

Fig. 5.8: *Validation of model, upper part: validation of in-use stock in the car sector, modelled stock compared with bottom-up data on cars in use; middle part: validation of in-use stock of reinforcement bars in the sector buildings, modelled stock compared with data on housing space statistics; lower part: validation of modelled scrap available with top-down data based on scrap statistics. Adopted from Dworak et al., 2022*

5.1.5 Sensitivity analysis

The sensitivity analysis for the chosen scenarios shows, that lifetime variations and changes in sector splits are the most influential on the surplus of low purity steel and are displayed in Fig. 5.9. As target value the surplus of low purity relative to the crude steel demand was chosen. Lifetime has the biggest impact (up to 9.3% for option A in 2015, 4% for option C in 2009, if the average lifetime is reduced by 20%). Changes in the sectoral split (±10% higher steel demand in the construction (*C*) sector) lead to up to ±4% of surplus of low purity scrap for option A. The remaining factor investigated (change in sectoral split for the sector transport (*T*), reduction of losses during recovery and export of end-of-life vehicles) have lesser impact (mostly below 1%, reduction of losses during recovery up to 1.9% in 2009 for option A).

The steel demand in the sector *C* is rather decisive in regard to the surplus of low purity scrap. In 2050, an increased demand in the sector *C* of 10% would reduce the scrap surplus by 2.1% and 4% (about 4 Mt/yr and 7.8 Mt/yr) for options A and C, respectively. In contrast, the export rate of end-of-live vehicles has little influence on the amount of surplus scrap. In the case of 20% less end-of-live vehicles exports, the surplus scrap quantities would only increase by less than 0.52% and 0.03% (about 0.06 Mt/yr and 1 Mt/yr) in 2050, for options A and C, respectively.

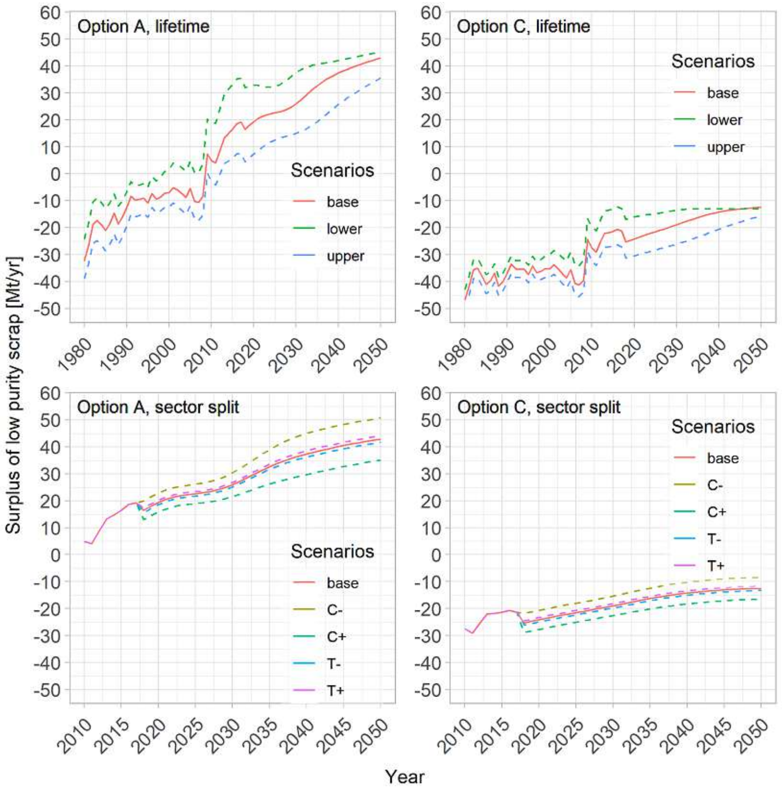

Fig. 5.9: *Sensitivity for lifetime (±20%, upper part) and sector split (±10% for either Construction (C+/C-) or Transportation (T+/T-) sector, lower part) for option A (purity of input = purity of output, left column) and option C (purity of output based on sector of origin, right column). The reference value is the surplus of low purity scrap (Q3 & Q4) relative to the crude steel demand. Adopted from Dworak et al., 2022*

5.2 Mercury throughput in the Austrian manufacturing industry

The multi-annual *MFA* investigates Hg concentrations in flows of goods. As mentioned in the model description (Chapter 4.2.2), the data for good flows is mainly reported data and therefore will not be analysed further here. The Hg concentration on the other hand are derived from various sources. Therefore, the applied Hg concentrations are discussed in the following chapter. Subsequently, the overall results of the model are discussed.

5.2.1 Hg-concentrations

Hg concentrations and uncertainties were assigned to each fraction flow, to account for the immense variability of Hg concentrations in goods. Hg-concentrations with up to 100% relative standard uncertainty are applied in the study. Two main reason may be given:

- Hg-concentrations of goods, vary a lot and are dependable on multiple factors. For coal, e.g., high difference in Hg content are reported dependant on deposit area and forming period (e.g., Bai et al., 2017; Lassen and Hansen, 2000; Mukherjee et al., 2008; Pirrone et al., 2010; Toole-O'Neil et al., 1999).
- The analysis of Hg in the laboratory is rather challenging. As the concentrations are quite low (usually way below 1 mg/kg), cross contamination can be an issue. Further, Hg is volatile at low temperatures, which is challenging for storage and sample preparation.

These circumstances lead to a paradoxical situation: often, if more data is available, the variability of the values becomes higher as well.

In Fig. 5.10 (left part) the distribution of the mean values of single fraction flows are displayed, grouped in their corresponding main flow. Especially *SRM-F* have a high variability of mean Hg-concentrations and, in average, a higher level of Hg-concentration in general. Also shown in Fig. 5.10 (right part) is the distribution of relative standard uncertainty of single fraction flows, again grouped in their corresponding main flow. Even though some relative standard uncertainties are below 50%, most of them can be found between 75% and 100%, whereas again, *SRM-F* show the highest uncertainty. Concluding, available data on the *SRM-F* is rather diverse depending on the specific material, but also within each material itself.

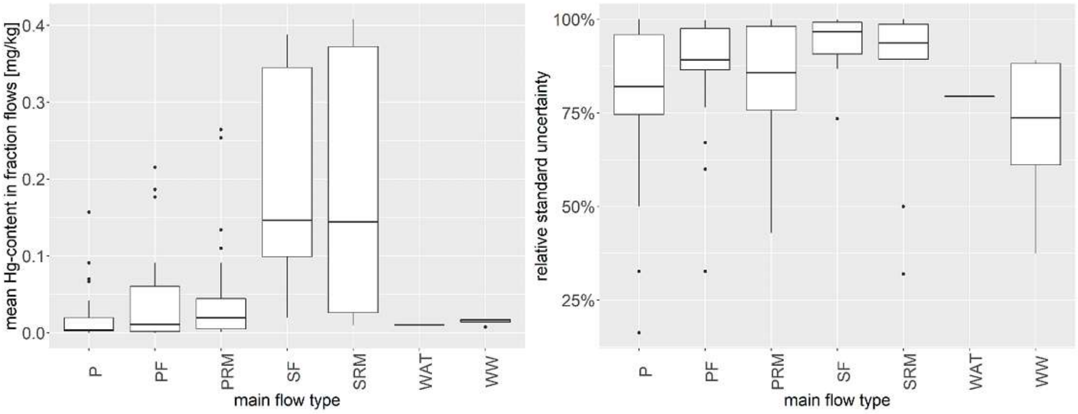

Fig. 5.10: *left: Distribution of mean Hg content of fraction flows in main flow types shown as absolute mean values of fraction flows in the corresponding main flow types: (products (P); primary fuels (PF); primary raw materials (PRM); secondary fuels (SF); secondary raw materials (SRM); water supply (WAT); wastewater (WW); right: relative standard uncertainty of Hg content of each fraction flow present in the corresponding main flow type. Adopted from Dworak and Rechberger, 2021*

5.2.2 Modelled throughput

The Hg-throughput was calculated between 3.5 t/yr ± 70% and 4.0 t/yr ± 70% over the period investigated. The biggest part (roughly 70% to 75%) was imported via *PRM-F*. *SRM-F* account for roughly 25% to 30% of the Hg-input. *WAT* accounts for around 1%. The export via products is determined around 35% to 40%, waste in a similar magnitude, at roughly 40% to 45%. Hg via emissions between 20% and 25%. The balance for 2016 can be seen in Fig. 5.11.

The highest share of throughput can be assigned to *NMM* and *MET* (roughly 30% to 40% and 30%, respectively). *WPP* contributes with 16% to 28% of the throughput, *EG* with 4% to 14%. Hg-throughput via *Ch* and *OTH* was determined much lower (around 5%). These rather low shares are mainly explainable by the low data availability on material flows (input and output of goods) for these sectors. For *Ch*, for instance, only data from the Austrian energy balance (Statistik Austria, 2019) was available, covering energy carriers only. The most significant single material input of Hg over all and for every sector individually, was hard coal (21% to 24% for most industries, up to 50% to 60% for *Ch*).

The structure of the Austrian industry did not change significantly in the investigated period (no new manufacturing sector, no suspension of manufacturing sectors). The model showed that the overall changes of Hg throughput into the system are not significant, as they did not exceed the estimated relative standard uncertainty.

5.3 Data availability

5.3.1 The case of steel

5.3.1.1 Commodity

Steel is one of the first commodities reported systemically with original records still available (e.g. Eurostat, 1970; Melcher et al., 1952) or compiled in various works (e.g. Kelly et al., 2014). Associations like Eurofer or the World Steel Association publish yearly data on steel production on a steel intermediate resolution. Even though the data is partially reported in slightly different categories, it is consistent. In regards to the Steel Statistical Yearbooks (e.g. World Steel Association, 2020), some data points are missing for specific years, but can generally be interpolated/scaled from provided data. For Europe, also some reported data is available on the shares of steel in specific sectors (e.g. EG, 1976, 1985, 1990; Eurofer, 2018). Less detailed reports are available for the deliveries of specific intermediate steel products to specific sectors. Here, we may relay on case studies, such as Cullen et al., 2012. All in all, the data availability is well suited for modelling a system, even though none of the data is available in a machine-readable format. Hence, data preparation has to be done manually, which reduces options for independent monitoring.

5.3.1.2 Tramp elements

The data situation for tramp elements is much more precarious. Officially, there are standards which quantify specific tramp elements for specific steel products which limit tramp elements to specific thresholds. These data is practically not linkable to the reported products and rather superficial. Specific amounts of alloy-elements are not disclosed and therefore no specific data is available on a detailed level. Multiple studies attempt to quantify the tramp element content in specific steel flows. Particularly for Japan, studies on this topic are available (e.g. Daigo and Goto, 2015; Daigo, Iwata, et al., 2017; Daigo et al., 2010; Igarashi et al., 2007; T. Oda et al., 2010). Globally, the Cu content in the steel cycle was investigated (Daehn et al., 2017). Combining all available data, it was not possible to assign specific tramp element contents to specific steel flows. Hence, a classification was applied to be able to investigate the system semi-quantitatively and draw conclusions from the data available.

5.3.2 The case of mercury

In Tab. 5.1 the data availability assessed in the case study is summarized. Further, the assesed relevance of the according flows and recommended monitoring measures are listed. The table underlines the rather divers availability and quality of data. Particularly data on the sectors *Ch* and *OTH* is rare and of low quality and it was not possible to give a recommendation on the relevance of the flows, even though the results of the model might suggest otherwise.

Tab. 5.1: *Data availability assessment by sectors (C: chemical industry, EG: energy generation, MET: metal industry, NMM: non-metallic minerals, OTH: other industries, WPP: wood, pulp and paper) and main flow types (PRM: primary raw materials, PF: primary fuels, SRM: secondary raw materials, SF: secondary fuels, WAT: water supply, WW: wastewater, P: products, EM: emissions)*

Sector	Flow	Import Export	Goods			Concentration			Relevance
			Rating	Source	Comment	Rating	Source	Comment	
all	EM	EX	+	3	EMEP, industry reports are available in similar detail			not applicable	I
all	WW	EX	~	3,4	reporting biannually, pulp & paper: detailed yearly reporting in industry report	~	5		I
C	P	EX	--	1	harmonized and aggregated	-	5,6		?
MET	P	EX	+	1	coke industry is counted as part of the metal industry	~	5,6	coke industry is counted as part of the metal industry; assumption: no Hg in finished metal products	III
NMM	P	EX	++	4	detailed reporting and publishing of production volumes for pulp & paper				III
WPP	P	EX	++	4	detailed reporting and publishing of production volumes for cement	+	5,6		III
OTH	P	EX	--	2	domestic material consumption	-	5,6	due to highly aggregated and missing flows, little specific data can be applied	?
all	PF	IM	+	1,4	harmonized and aggregated (statistical accounting), industry report for cement and pulp and paper	+	5,6		I
all	SF	IM	~	1,4	detailed reports for cement and pulp & paper	+	5,6,7,8	high variation, max for all Hg concentration based on allowed Hg concentration in cement industry	I
all	WAT	IM	~	3,4	reported biannually until 2010; pulp & paper: detailed yearly reporting in industry report	~	5		III
C	PRM	IM	-	1	crude oil statistical accounted in fuels; no further information on primary raw materials	+	5,6		?
C&OTH	SRM	IM	--		no information available			not applicable	?
MET	PRM	IM	-	2	according to domestic material consumption (ores)	~	5,6		II
MET	SRM	IM	~	4	steel scrap input reported since 2010; no data on non-ferrous metal scrap	~	5	little data available	II
NMM	PRM	IM	+	2,4	for cement inputs are available; for other sectors calculation on basis of production margins was applied	+	5,6		I
NMM&WPP	SRM	IM	+	4	detailed reporting and publishing of secondary raw material used for cement and pulp & paper	~	5,6,7,8	high variation, max for all Hg concentration based on allowed Hg concentration in cement industry	I
OTH	PRM	IM	--	2	rough estimate as material accounting is not explicitly related to industrial sector	-	5,6,7	due to highly aggregated and missing flows, little specific data can be applied	?
WPP	PRM	IM	+	2,4	detailed reporting, auxiliary materials and additives reported for pulp & paper, aggregated	-	5,6,7		III

rating: ++: detailed data available, +: aggregated data available, ~: fragmented data available, -: no specific data available, --: no data available

sources: 1: statistical accounting for fuels, 2: material accounting, 3: officially reported, 4: industry reports, 5: scientific reports, 6: peer-reviewed literature, 7: legislation, 8: waste characteristics data base

relevance: I: high relevance, continuous monitoring recommended, II: moderate relevance, periodic checking recommended, III: low relevance, occasional checking recommended, ?: not enough data to assess relevance

5.3.2.1 Goods

Based on the model design, flows of various goods were considered. Flows into the industry sectors are mostly well statistically documented, even though in an accumulated way and mostly without any uncertainty comments. These statistical datasets are machine-readable, which enables efficient updating and analysis of material flows for multiple time periods. More details (composition of the aggregated flows, details on uncertainty and plausibility) are sparsely available and if so, not in machine-readable format, which makes the processing demanding and labour intensive.

5.3.2.2 Mercury concentrations

As mentioned above, the data situation for Hg-concentrations is rather diverse. There are many data points available, but most are only limitedly applicable for a *MFA*. Often missing description of sampling process (origin, condition of sample), sample preparation process, specification of the samples (references to dry matter, solid matter, and overall sample) leads to low quality data points. Still, where no suitable data is available, also low quality data might be used. As visualized in Fig. 5.10, the diverse data quality leads to very high uncertainties, mostly over 75%. This situation leads to a balance with a limited meaningfulness

More insight could be provided by detailed datasets of Hg-related information of sectors or plants (e.g. composite sampling) published. Still, due the high natural variability, Hg-monitoring might be the only way to really grasp how Hg is moving through the industries, and also through society. A solid database is essential to understand and improve the system. A framework such as proposed by Petavratzi et al., 2018 could add value by locating data points in systems and thereby increase data quality by means of comparability.

In summary, better harmonization, as proposed by e.g. Petavratzi et al., 2018, and access to data on a detailed level would enable much more robust modelling.

5.3.3 Differences and implications

MFA, regardless if dynamic or static, are highly dependent on data available. Some missing or low quality data might be compensated by deep understanding of the system. But still, the core of a *MFA* will always be data. The two case studies "steel" and "mercury" are positioned diametrically on the spectrum of available data for an *MFA*. On one end, steel, a commodity that was one of the first ones reported systemically with records still available in original (e.g. Eurostat, 1970) or compiled in various works (e.g. Kelly et al., 2014). For Hg, the case is rather different. As a well known hazardous substance in most of its forms, nowadays mainly present as unwanted accompanying trace element in many resources, reporting of data is not in the interest of industrial sectors. Incentives for reporting Hg is mainly enforced based on regulations. Hence, data is mainly made publicly available by health and environmental organizations (public and private) and by the scientific community. Müller et al., 2014 reviews applied methodologies of *DMFA* of various metals, where the steel studies are represented the most (17 of 60) and for Hg (among others) just one.

The different situation requires adapted approaches in conducting an *MFA*. With a reporting system established (such as in the case of steel), it follows logically to work within this system as the data is available at specific data points. Further, there may be studies done according to the established system, which allow comparison and validation with the conducted study.

Even though the flows of steel are well documented, levels of tramp elements in steel are rarely available. Some general and superficial restrains can be derived from according standards, but to be able to model composition, much more detailed data is needed. One approach can be to specify classes, as was done in the case study for steel.

Looking at a trace element, which is only reported systemically were legally required, data points are often unconnected and available independent from the system itself (e.g. emissions to the environment CEIP, 2020) are available. Therefore, no established system can be adopted. In case of the manufacturing industry, economic wide material flow accounting (Eurostat, 2020) provides a suitable basis data wise, even though it might not be the most suitable system design to investigate Hg in the industry. Nevertheless, these reported flows can be applied with the according Hg-concentration, if it is available.

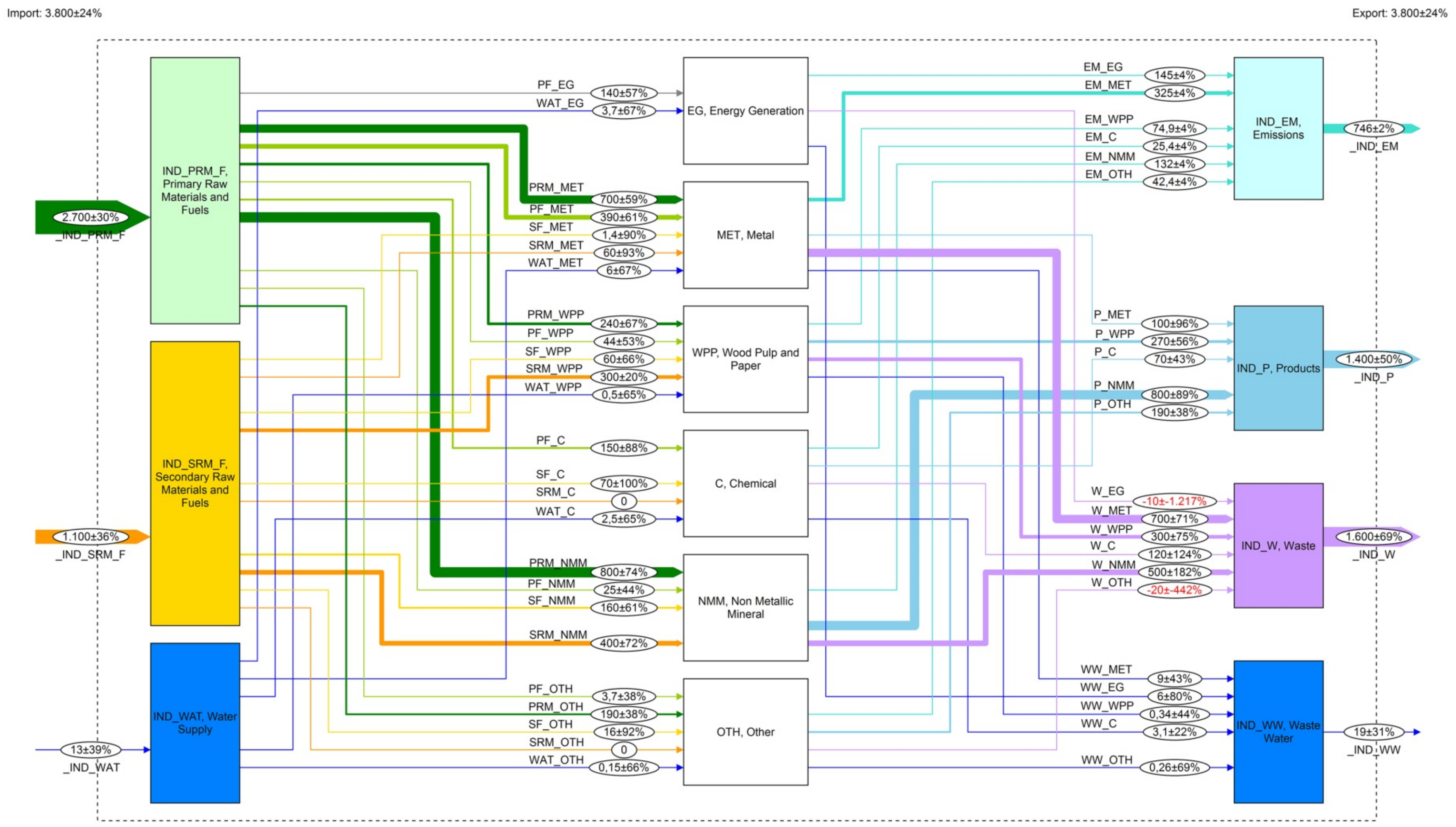

Fig. 5.11: *Hg throughput for the Austrian manufacturing industries: balance for 2016 in kg/yr (abbreviations: IND (summed up import and export flows); import main flow types: PRM (primary raw materials), PF (primary fuels), SRM (secondary raw materials), SF (secondary fuels), WAT (water supply); export main flow types: EM (emissions), P (products), W (wastes), WW (wastewater). Adopted from Dworak and Fellner, 2021*

Chapter 6

Conclusion

In the scope of this work, data availability and quality was investigated and discussed based on two case studies. The European steel circle and the throughput of mercury in the Austrian manufacturing industry were investigated via material flow analysis. In the case study for steel, two models were used to assess the scrap composition quality-wise (according to tramp element load) up to 2017 (static multi-annual material flow analysis) and to project the composition of scrap until 2050 (dynamic material flow analysis). The resulting scrap composition was compared with the steel amount of the according year, using material pinch analysis. In a second case study, a static multi-annual material flow analysis was applied to investigate the Hg-throughput in the Austrian manufacturing industry. Both case studies were used to derive conclusions regarding the handling of data with varying availability and quality.

The following chapter summarizes the results of this work. Details for methods (chapter 3), model description (chapter 4) and results (chapter 5) can be found in the chapters above.

- 1. Which data sources are available for steel, its respective tramp element content and mercury?

  For *steel*, various and mostly consistent data is publicly available. Regarding mass flows of steel, official reporting data is widely available. Existing gaps for single years and/or countries can usually be interpolated or scaled from available data. The data is publicly available, but not in a machine-readable format.

  The situation for *tramp elements* present in specific steel flows is rather different. Putting aside rough ranges for intermediate products specified in standards, mainly isolated scientific case studies are available. Precise tramp element levels in specific steel intermediates are rarely available.

  Hg-concentrations are available for most goods, but mostly of a rather low quality (incomplete, non-representative). Data on quantities of goods are mostly available, based on economic material accounting. Still, data availability on flows of goods, which are necessary to apply the Hg-concentrations on, vary strongly within and between sectors.

- 2. To what extent does the availability of the data sources impact the respective material flow analysis system investigated?

  The approach to such diverse data situations is fundamentally different. In case of commodities, like steel, consistent data sets are available for many areas. Especially production volumes are well documented and published. Data processing includes mainly finding data gaps in series and inter- or extrapolate from existent data points. There is an inherent motivation to understand steel flows to be able to manage them and exploit the product economically. It allows researchers and public entities to use the data and monitor flows. mercury, on the other hand, is an undesired trace element. Sectors with high Mercury load tend to keep the data confidential. To establish a working system, which allows conclusions, extensive data screening is required, often with contradicting data points. Consequently, conclusions based on the system are mostly less reliable. Hence, such a system is rather suitable to point out areas, where the pool of available data needs to be improved to investigate system dynamics themselves. For these systems, with such diverse data base, a thorough analysis of the data quality is necessary to prevent bias from low quality data. Further, all interpretation must come with background information with regards to data gaps.

- 3. Do the current data sources allow a continuous monitoring of material flows?

  With the data publicly available, no continuous monitoring is possible, especially no automated monitoring. The case of steel shows, that the flows can be monitored with intense data processing beforehand. Nevertheless, validation must be done individually and independent data might not be easy to find. Mercury, on the other hand, is quite hard to verify at all. In this case, a judgement about the viability of specific data points is rarely possible. Hence, the data available does not meet the criteria for continuous monitoring.

- 4. How should data sources be made available in order to be easily usable for material flow analysis?

  A standardized framework with underlying models could add value by locating data points in systems. Such an approach would support systematic data collection and application and enable each data point collected data to contribute to the overall picture.

  In the case of mercury, detailed datasets of Hg-related information of sectors or plants (e.g. composite sampling) would be most suitable to feed the mentioned system with viable data. Still, due the high natural variability, continuous monitoring of Hg-concentrations in relevant flows, might be the only way to really comprehend how Hg is moving through the industries, and also through society.

  In summary, a solid data base is essential to understand and improve the system. Better harmonization or even standardization, and access to data on a detailed level would enable much more robust modelling.

- 5. How did the steel scrap flows on the territory of the EU develop and how will they develop until 2050?

  The scrap available is continuously increasing. Up to the 1970s the increase is rather steep, whereas after that, the increase continues moderately. The composition of available scrap has shifted from predominately new scrap (production and forming scrap and fabrication scrap) to predominately old scrap (post-consumer scrap). The constant increase, especially from the 1970s onwards, can be contributed to the vastly increasing amount of the post-consumer scrap available. New scrap, particularly production and forming scrap decreased from the 1970s to the 1990s (from 50% of total scrap available in the 1950 to 12% in the 2010s), due to an immensely increased production and forming efficiency. The share of fabrication scrap is more or less constant, between 20% and 30% of the total scrap available. Based on the assumption of stagnant amount of crude steel production, an constant material efficiency up to 2050, the absolute amount of new scrap (production and forming scrap and fabrication scrap) is constant. Old scrap (post-consumer scrap), on the other hand, is constantly increasing, as the average lifetimes are close to or longer than the prospective period in many sectors. A further increase of the share of old scrap (from 65% in 2020 to almost 75% in 2050) is expected.

- 6. How did the composition of steel scrap develop with regards to relevant tramp elements and what will the composition be in the future?

  The share of high purity scrap (Q1 & Q2) in new scrap is constantly increasing over the investigated period up to now, from about 15% up to more than 50%. The production portfolio of the European steel industry changed, producing higher shares of flat products. These shift leads to the mentioned change in composition of the scrap flows quality wise. Especially fabrication scrap is composed of more high purity fractions, as flat products tend to demand high purity and additionally, the low material efficiency of flat products generates higher scrap volumes compared with long intermediate steel products. Still, the all-over composition of scrap is shifting to lower purity scrap (Q3 & Q4) due to the increasing share of post-consumer scrap, which is composed of low purity fractions (mostly mixed up or contaminated during use or recovery).

  The composition of new scrap is by design not changing for the projected period (2018 to 2050). But the share of old scrap is increasing further, which leads to higher shares of low purity fractions (Q3 & Q4), if the current recovery practices stay in place. The low purity share would reach more then 70% of the scrap available in 2050. Potentially, if the fractions could be recovered from the system as they are put in (each quality class can be recovered as it was put into the system), the share of low purity scrap could be lowered to around 35% of the available scrap.

- 7. To what extent can steel scrap generated in the EU be utilized by the domestic industry (in the past and in future)?

  Up to the 2000, all of domestic scrap could be and was used in the domestic steel industry. From that time on, the surplus of low purity scrap was mainly exported. If the current system stays in place, the scrap rate will soon stagnate around 55%. With interventions, which would allow the usage of low purity scrap, the scrap rate could increase similarly to the amount of scrap available to more then 70% in 2050.

- 8. Which measure might be necessary to increase the domestic production of scrap-based steel?

  Since a surplus of low purity steel has been available, it has been exported. Technological interventions, such as the removal of tramp elements, or dilution of low purity scrap with high purity resources (e.g. materials from primary resources) has not been practised on a significant scale in the EU. The surplus of low purity scrap is increasing if the current practice of scrap handling stays in place. It would increase from 20 Mt/yr in 2017 to 50 Mt/yr (around 1/3 of scrap available) in 2050.

  One approach to be able to utilize the surplus is dilution of low purity fractions with high purity fractions (e.g. crude steel from primary sources). Applying dilution efficiently would enable to dilute low purity fractions up to 2040, from than on, some of the low purity scrap could not be used for recycling as the tramp element content in the system would be too high.

  Further, technological interventions could raise the threshold of tramp elements present in scrap (e.g., secondary metallurgy, material and process design).

  But is to take action already during recovery by improving sorting would have the highest impact. It was shown, that the theoretical potential of precise alloy sorting would make the usage of all scrap fractions possible.

- 9. How much and through which paths does Hg move through the manufacturing industry in Austria?

  The Hg throughput in the Austrian manufacturing industry in the investigated years (2005-2016) was determined with 3.5 t/yr ±70% to 4 t/yr ±70%. About 70% to 75% are imported via primary raw materials and fuels and 25% to 30% via secondary raw materials and fuels. The import of Hg via the water supply accounts for around 1%. A slow shift from primary raw materials and fuels to secondary raw materials and fuels can be noted. A similar amount of Hg is exported via products and waste (35% to 40% and 40% to 45%, respectively). Around 20% ends up in the atmosphere. It can be noted, that the overall transfer coefficient to emissions to air was decreasing in the last years (2013-2016), which, at least partially, can be attributed to the fade out of coal for energy generation.

- 10. Does the data availability allow deeper understanding of the unintended Hg flows in the Austrian manufacturing industry?

  The data availability allows some general conclusions, but does not allow detailed investigations of the Austrian manufacturing industry. Furthermore, the study shows, that industries which publish specific data are prone to be investigated more thoroughly. In case of the investigation of hazardous substances, this situation is not favourable, as it may be seen as a barrier for open data policies. Due to high variability in Hg-contents it seems the only way to deepen understanding of the system and its development over time is to install permanent monitoring.

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Appendix

Paper I

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Steel scrap generation in the EU-28 since 1946 – Sources and composition

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ABSTRACT

A detailed understanding of scrap generation and utilization is needed to target increased material efficiencies and circular economy in the steel industry. In the present paper, the generation and composition of steel scrap (production & forming scrap, fabrication scrap and post-consumer scrap and their composition) in the former territory of the EU-28 have been assessed from 1946 to 2017 by means of Material Flow Analysis. The results reveal that the steel scrap composition in the EU-28 has changed significantly since 1946. Until 1980 scrap generation was dominated by new scrap (mostly production & forming scrap). Today, most of the overall steel scrap is post-consumer scrap. Most of the new scrap consists of fabrication scrap. Taking the presence and tolerance levels of major tramp elements in steel (Cu, Sn, Cr, Ni and Mo) into consideration, a material pinch analysis reveals a surplus of steel scrap with higher levels of tramp elements since the 1990ies. This scrap could only be utilized in the European steel industry by dilution with primary iron sources. At present, this surplus scrap seems to be largely exported as its quantity corresponds well to the net-exports of steel scrap from the EU-28. Transplanting the observed trends of steel scrap generation to emerging economies implies a significant increase in fabrication and post-consumer scrap in these countries in the near future. However, the utilization of scrap at higher rates, particularly post-consumer scrap, will challenge the local steel industry as tramp elements significantly increase along the production, use and end-of-life chain.

1. Introduction

After mineral construction materials, steel is the most frequently used commodity in our society. Global crude steel production amounted to almost 1.9 billion tonnes in 2019, and has doubled during the last two decades (based on World Steel Association, 2020). For the coming years only moderate growth, or even no further growth, in global steel production is expected according to Hatayama et al. (2010), a situation that affluent economies (such the United States or the European Union (EU)) have already experienced for several decades. In the case of stagnant steel production rates, the potential raw material supply of steel producers via scrap is gaining in importance since a higher share of the production can be covered by the scrap available and thus less iron ore needs to be used. Such a situation is not only preferable from a resource point of view, but also from an environmental perspective as steel production from scrap is less CO_2 intensive than from iron ore (according to Broadbent (2016) up to 75% CO_2 reduction) Furthermore, scrap based steel production is also associated with other environmental benefits (e.g., lower eutrophication, acidification and photochemical oxidation) in comparison to primary production (Hu et al. 2014, López et al. 2020).

For US steel production, Zhu et al. (2019) show that in 2014 about two-thirds of the annual production stemmed from scrap, including production & forming scrap, fabrication scrap and post-consumer scrap. For the EU, the share of steel production from secondary resources is estimated to be about 54% (see Fellner et al. 2018, Passarini et al. 2018), with a slightly declining trend in recent decades. At a global level, Cullen et al. (2012) estimated that in 2008 only about 36% of overall steel production was based on scrap.

Although steel flows have been extensively researched at different levels and for different regions and time frames (e.g. Cooper et al. 2020, Hatayama et al. 2010, Klinglmair and Fellner 2011, Müller et al. 2011, Müller et al. 2006, Pauliuk et al. 2013, Wang et al. 2007), works conducted for Europe or the EU are rather limited. Analyses of overall steel flows are available only at a rather superficial level (Fellner et al. 2018, Passarini et al. 2018), although the development of steel stocks for single European countries has been investigated in more detail (Müller et al. 2011, Pauliuk et al. 2013). Data on the amounts of the different "types" of steel scrap generated and utilized (e.g., production & forming, fabrication and post-consumer scrap), their origin with regard to finished steel products, the respective composition with respect to the

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content of tramp and alloying elements as well as the temporal development of scrap generation and composition is largely missing. Today only for Japan such data exists, as intensive research regarding tramp and alloying elements present in steel (including their sources and sinks) has been conducted during the recent years (e.g., Oda et al. 2010; Daigo & Goto 2015; Daigo et al. 2017; Nakamura et al. 2017).

However, in order to assess and understand the potential of steel scrap utilization for substituting primary steel production in the European Union, detailed knowledge about steel production, steel qualities, sectoral steel consumption and the scrap generated thereof is essential. Such knowledge would not only be beneficial for the European steel industry, but is also of global significance as historical trends observable for steel use and scrap generation in the EU-28 might help to indicate future developments of steel flows in other regions of the world (e.g., China, India, Africa).

Hence, the main objective of the present paper is to assess the development of steel flows for the territory of today's EU countries including the United Kingdom (in the following referred to EU-28) for the period 1946 to 2017, focusing on scrap generation (divided into production & forming scrap, fabrication scrap and post-consumer/End-of-life (EoL) scrap) and scrap utilization. Besides the assessment of the total flows of the different types of scrap, a qualitative assessment of the composition of the steel scraps with respect to major tramp elements (sum of Cu, Sn, Cr, Ni and Mo) is conducted and the scrap qualities and quantities determined in this manner are compared to the demand for steel in order to evaluate to what extent the steel portfolio of the European steel industry can be potentially produced out of scrap. Moreover, the question as to whether observable trends pertaining to the steel scrap trade might be explained by the qualities of the available scrap was investigated.

2. Methods

2.1. Material flow analysis MFA

The present study uses Material Flow Analysis (MFA) as described by Brunner and Rechberger (2016) to map the steel and steel scrap flows throughout the territory of the EU-28 for the period 1946 to 2017. MFA generally allows the flows and stocks of materials to be assessed through a system that is defined in space and time. The calculations are based on the law of conservation of matter by using a material balance to compare all inputs, stocks and outputs of a process. Hence, unknown flows can be calculated by the mass balance formula (in the present study, for instance, it is used to calculate the quantities of post-consumer scrap):

$$\sum_{i=1}^{k} \dot{m}_{IN,i} = \sum_{j=1}^{l} \dot{m}_{OUT,j} \pm \dot{m}_{STOCK}, \tag{1}$$

with $\dot{m}$ given in steel mass per time unit. $\sum_{i=1}^{k} \dot{m}_{IN,i}$ represents the total steel input $\dot{m}_{IN}$ of k input flows i, $\sum_{j=1}^{l} \dot{m}_{OUT,j}$ the total steel output $\dot{m}_{OUT}$ of i output flows j, and $\dot{m}_{STOCK}$ represents a potential flow from or to a stock located in the process itself. Otherwise, transfer coefficients can be used to calculate unknown flows, as done in the present work for the quantities of fabrication scrap and the distribution to the End-use sectors:

$$TC_j = \frac{\dot{m}_{OUT,\ j}}{\sum_{i=1}^{k} \dot{m}_{IN,\ i}}, \tag{2}$$

where the transfer coefficient TC_j of an output flow j, $\dot{m}_{OUT,\ j}$ is its mass relative to the total mass input $\sum_{i=1}^{k} \dot{m}_{IN,\ i}$ of k flows i. The transfer coefficients can be calculated based on the same equation if all flows are known, which was not, however, the case in the present study.

2.2. MFA model for steel scrap flows

The overall MFA system for the steel flows in the EU-28 is illustrated in Fig. 1. It is defined by six processes and 14 flows of steel and the temporal and spatial boundaries, which is the territory of the EU-28 in the period from 1946 to 2017. The study presents a static, retrospective top-down model for steel scrap flows where each year (72 years) is analysed separately. Since all relevant flows for the study can be derived from statistical data, we refrained from applying a dynamic material flow approach typically used for analyses over longer times.

To assess the flows of the different types of steel scrap (production & forming, fabrication and post-consumer scrap), only three processes out of the six processes shown in Fig. 1, namely *2. Production of intermediate products, 3. Fabrication of finished products* and *6. Scrap market* were analysed and balanced accordingly. Hence, only the flows indicated in red in Fig. 1 were determined, whereas the black flows are just given for the sake of completeness of the system.

For the process "*2. Production of intermediate products*", and hence also for the flows of intermediate products (IP), altogether 19 intermediates are distinguished and determined according to Cullen et al. (2012) and Zhu et al. (2019), which are summarized in Table 1.

Besides the domestic production of intermediate steel products, imports and exports (summarized as net-import of intermediate products (NIIP)) were considered in the present work. *Production & forming scrap* (PFS) include all scrap generated until the production of intermediate steel products and is thus composed of production scrap from steelmaking and forming scrap from the production of intermediates (rolling and forming). According to Cullen et al. (2012) the amount of forming scrap is generally dominant in comparison to the production scrap arising during steel making.

For the calculation of the *Production & forming scrap* (PFS), reported shares of production & forming scrap in relation to the crude steel amounts produced and formed (Eurostat 1970, 1985, 2002) were used together with information about the share of continuous casting steel production. The introduction of the continuous casting steel production led to a signficant decrease in the share of PFS generated (see Supplementary Information – Fig. A1). To portion the overall quantity of PFS to the different intermediates produced, information about the specific scrap generation of the different production and forming processes presented by Cullen et al. (2012) were used. Details about the intermediate specific PFS rates over time derived in this manner are summarized in the Supplementary information (SI) (Table A2). For the total amount of crude steel produced and formed by the European steel industry, the sum of domestic crude steel production (CrS) and net-import of ingots & semis (NIS) has been used. This simplification with regard to PFS generation (in particular concerning production scrap generation which only arises from domestic production) is justified by the fact that NIS is generally below 3% of CrS (see Table A1 of SI).

The 19 intermediate steel products were distributed to 4 different end-use sectors, subcategorized in 10 end-use sectors. For the calculation of *Fabrication scrap* (FS), sector and intermediate steel product specific material efficiency rates were applied. Similarly, intermediate steel product and sector specific transfer coefficients were applied for the distribution of the steel intermediates to the different end-use sectors. The sectors have been chosen in accordance with the works of the same authors cited above (Cullen et al. 2012, Zhu et al. 2019), which can be found in Table 1.

By balancing the process *6. Scrap market*, the quantity of post-consumer scrap recovered (PoCSr) was determined since all other input and output flows of this process were available - either derived from statistical data (net-import of scrap, scrap recycled) or calculated via transfer coefficients (production & forming and fabrication scrap). Losses of new scrap (PFS and FS) were neglected.

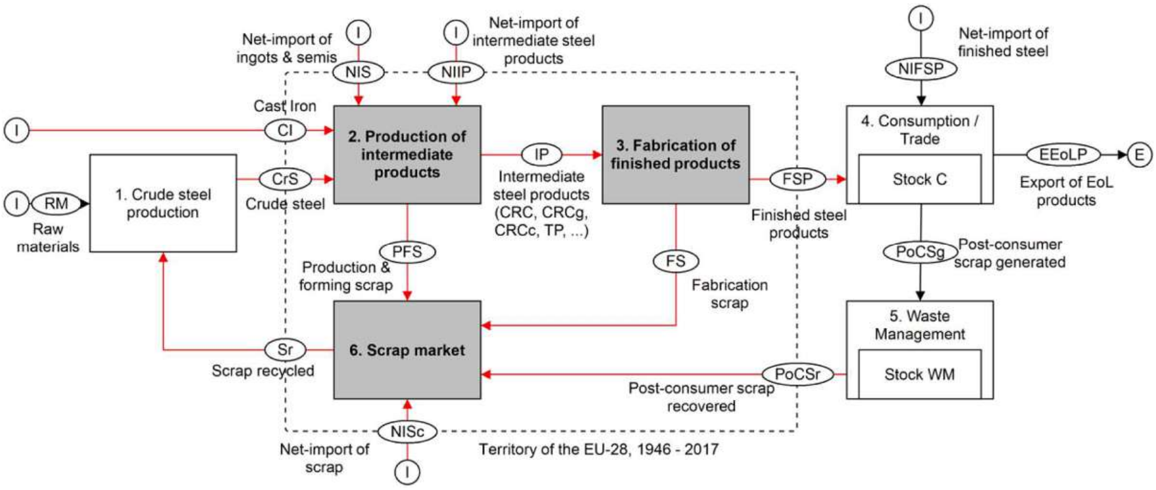

Fig. 1. Simplified MFA system for assessing steel scrap flows in the EU-28 (all red steel flows are determined, whereas black flows are just shown for completeness); the processes 1., . and 5. are also within the borders of the EU-28, but not considered/balanced within the framework of the present study and are hence located outside the system boundary; Stock C ... consumption stock; Stock WM … stock in waste management (e.g., landfills). In addition to steel, the flows of cast iron were considered as well.

Table 1
Intermediate steel products and end use sectors considered (according to Cullen et al., 2012)

Intermediate steel products			**End use sectors**		
Intermediate Steel Products	abbreviation	Grouped Intermediate Steel Products	End Use Sectors	abbreviation	Grouped End Use Sectors
Cast steel	c CS	casts			
Cast Iron	c CI				
Electrical Strip	f ES	flats	Buildings	C Bu	Construction
Tin Plated	f TP		Infrastructure		
Plate (excl. plates used for welded tubes)	f P			C In	
Cold Rolled Coil galvanized	f CRCg				
Cold Rolled Coil coated	f CRCc		Cars	T Ca	Transport
Cold Rolled Coil	f CRC				
Hot Rolled Coil galvanized	f HRg		Trucks	T Tr	
Hot Rolled Narrow Strip (excl. Strips used for welded tubes)	f HRNS		Other Transport	T OT	
Hot Rolled Coil	f HRC				
Welded Tubes	t WT	tubes	Mechanical Engineering	I ME	Industrial Equipment
Seamless Tubes	t ST				
Wire Rod	b WR	bars	Electrical Engineering	I EE	
Reinforcing Bar	b RB				
Hot Rolled Bar	b HRB		Other Metal Goods	MG OMG	Metal Goods
Heavy Section	s HS	shapes	Appliances	MG Ap	
Light Section	s LS		Packaging	MG Pa	
Rail Section	s RS				

2.3. *Data sources utilized for steel flows*

In the present chapter, the different data sources for assessing the respective steel flows of the MFA diagram are summarized. In particular, the following steel flows are based on statistical data: crude steel, intermediate products, net-import of semis and ingots, net-import of intermediate products, net-import of scrap and scrap domestically recycled, whereas the other flows (remaining red-indicated in Fig. 1) were determined by balancing the single processes or applying transfer coefficients.

Crude steel production (CrS): The total amount of domestic crude steel production was assessed via data published by the World Steel Association. Data for the EU-28 countries (except for Croatia and Slovenia) were available for the period 1967 until 2017. For the years 1946 to 1966 it was assumed that the total crude steel production in the “EU-28 countries” amounts to about 180% (based on the years 1967 - 1972) of the crude steel production of the EU-6. For the latter, statistical data were also available for the early years from Eurostat (Eurostat 1970).

Net-import of Semis & ingots (NIS) and Intermediate products (NIIP): The net-import of the semis and ingots and the different intermediates was derived from trade statistics provided by the United Nations Commodity Trade Statistics Database (UN Comtrade 2020) for the years 1994 to 2017 by balancing the trade of semis and the 19 different intermediate products for each member country. For the earlier years (1967 – 1993), only the overall amounts of net-imports of semis & intermediate products (difference between import and export of all steel semis and intermediate products, respectively) were available. For the years before 1972, information about the total net-imports were available for the EU-6, which was used, on the one hand, to assess the share of EU-6 net-imports relative to the total net-imports of the EU-28 (using the years 1967 – 1972, for which data for both, EU-6 and EU-28, were available). On the other hand, the data for the EU-6 were applied together with the aforementioned share to estimate the overall net-import of ingot & semis and intermediates into the EU-28 for the years before 1967. These total annual amounts of net-imports (reported and estimated data) were subsequently distributed to ingots & semis and the 19 different intermediates according to global trade statistics provided by the World Steel Association (International Iron and Steel

Institute 1978). Detailed information about the net-import of ingots & semis and intermediates and their assessment is given in the SI (see Chapter B and Table B1). In general, the net-import of the intermediates amounts to less than ±15% of the domestic production for most of them. Only for a few intermediates (mainly heavy sections, rail sections, and seamless and welded tubes) are higher shares of net-imports in relation to domestic intermediate production observable.

Intermediateproducts(IP): For the domestic production of the different intermediate products (particularly for long products, tubes and selected flat products), data were largely taken from the World Steel Association for the years 1984 – 2017. For the production volume of different flat products in the period 2004 till 2017, information was obtained from Eurofer (Eurofer 2018). It must be noted that Eurofer data were incorporated into the World Steel reporting scheme as they differ for some flat intermediates. For the earlier years, data from the iron & steel yearbooks of Eurostat (Eurostat 1970, 1977, 1985, 1994, 1998, 2002) for the EU-6, EU-9, EU-12 and EU-15 were used to assess the overall production in the EU-28. The production volumes of intermediates reported from the EU-6, EU-9, EU-12 and EU-15 were up-scaled with the respective ratios of crude steel production (e.g., crude steel production in EU-6 referred to crude steel in the EU-28). In order to account for the fact that the production share of the different intermediates in the EU-28 might differ from the data reported for a limited number of member countries (e.g. EU-6), correction factors for the productions volumes of the different intermediates were introduced. These correction factors were derived by comparing the up-scaled production data of intermediates for the EU-28 (based on Eurostat data) with production volumes reported for all EU-28 member countries by the World Steel Association or Eurofer. Detailed information on the annual production of the different semis and the assessment of the data are provided in the SI (see Chapter A and Table A1)

Net-importofscrap(NISc): For the net-import of scrap into the EU-28, data provided in the statistical yearbooks of the World Steel Association were used for the years 1971 - 1987. For the later and earlier years 1988 – 2017 and 1955 – 1970, respectively, data provided by Eurostat were used, whereas for the first decade considered (1946 – 1954), net-scrap imports were estimated assuming that the net-import amounts to about 5% (as observed for 1955 – 1958) of the overall scrap domestically recycled. All data and calculations for assessing the annual net-import of scrap are summarized in the SI (see Table A1).

Scraprecycled(SR): The annual quantity of scrap utilized by the steel industry of the EU-28 countries is reported by the World Steel Association for the period 1967 to 2017. From 1945 to 1966 a constant ratio of 50% for overall scrap consumption in relation to crude steel production was applied, an assumption justified by World Steel data reported for the late '60s and the beginning of the '70s and which is also in line with data provided by Eurostat on the typical ratio between overall scrap consumption and crude steel production for the EU-6 (Eurostat 1970). Annual data for SR are given in Table A1 of the SI.

Transfercoefficients: Transfer coefficients for the distribution of steel intermediates to specific end-use sectors were derived from different sources (Cullen et al. 2012, EG 1976, 1985, 1990, Eurofer 2018) and have been significantly changing over the last 70 years. For details about the temporal development of the transfer coefficients and their derivation, the reader is referred to the SI (chapter C). For the gross flows of steel intermediates into the 10 different end-use sectors determined in this manner (not displayed in Fig. 1), fabrication losses were assessed. To do so, sector- and semi-specific transfer coefficients for the fabrication scrap (determined by the material efficiencies) provided by Cullen et al. (2012) were applied (see Table D1 of SI). The fact that material efficiencies in the fabricating of final steel goods have improved in recent decades (see Pauliuk et al., 2013) was accounted for (It was assumed that between 1965 and 1995 the material efficiencies for flats and longs increased by 15% and 5%, respectively, see SI – Fig. D1).

2.4. Steel and scrap qualities

In order to assess the qualities of steel and steel scrap, four different levels of major tramp elements (impurities of Cu, Sn, Cr, Ni and Mo) in steel and thus steel qualities were considered (see Table 2). The sum of major tramp elements for assessing steel qualities was preferred over considering single elements, as for many steel grades the sum is limited due to the fact that the different elements may have super positioning effects on the mechanical properties and workability of steel (Kim et al. 2003; Lee et al. 2004; Daigo et al. 2020). Furthermore, detailed information about the actual presence of single tramp elements in different steel grades is not available for the European steel production (unlike to Japan, e.g., Oda et al. 2010; Daigo & Goto 2015; Daigo et al. 2017; Nakamura et al. 2017). Hence, a semi-quantitative consideration of steel and scrap qualities (considering the impurities of Cu, Sn, Cr, Ni and Mo) had to be chosen.

The classification of the four steel qualities was made based on information provided by several studies (Björkman & Samuelsson, 2014; Daehn et al., 2017; Huellen et al., 2006; Toi et al., 1997). Furthermore, it was assumed that the overall maximum content of tramp elements (sum of Cu, Sn, Cr, Ni and Mo) in steel products amounts to about 1.5 to 2.5 of the maximum Cu content allowed, based on Noro et al. (1997) and Toi et al (1997). In alloyed and stainless steel significantly higher amounts of the respective tramp elements are allowed or required. However, for simplicity reasons this fact has been disregarded in the present paper. According to the data of Eurofer, the overall share of alloyed and stainless steels amounts at present to about 20% of total crude production. Until 1990 their share was even below 10%, thereby justifying the simplification made. Moreover, the tramp elements Cu, which in most cases accounts for about 50% of the sum of the tramp elements considered, is also limited in almost all alloyed and stainless steel grades.

For the new scrap (production & forming and fabrication scrap), the steel qualities with respect to the content of tramp elements were assigned according to the intermediates and the respective steel qualities they arise from. With respect to the assignment of qualities to new scrap, it must be noted that in particular for the first 3 to 4 decades investigated, intermediates with higher tolerance levels of tramp elements (e.g. sections, hot rolled bars, reinforcing bars) have also been produced from primary steel and thus might contain lower levels of tramp elements, as assumed in the present paper. Hence, for this period the quantities of the lower steel quality categories (Q3 & Q4) might be overestimated.

For the EoL scrap, it was assumed based on data provided by different studies (Daehn et al., 2017; Davis et al., 2007; Huellen et al., 2006; Pauliuk et al., 2013; Sasov et al., 2003; Toi et al., 1997; Wagner et al., 2012; Willmann et al., 2017) that 40% to 60% of the scrap correspond to quality Q3 (max. content of tramp elements between 0.25% to 0.35%), whereas the remaining quantities of EoL scrap

Table 2
Steel qualities (tolerable content of tramp elements) considered. Detailed information about the assignment of the four quality categories to the different intermediates and their sectoral use is provided in SI, chapter E (Table E1).

Max. content of tramp elements $\sum$ *(Cu, Sn, Cr, Ni, Mo) in %*	**Quality categories**	**Typical steel intermediates**
<0.18	Q1	most flat products (cold rolled coils) – deep drawing quality, interstitial-free steel
0.18 – 0.25	Q2	tubes, plates, hot rolled products in construction, wire rod (other than construction)
0.25 – 0.35	Q3	hot rolled bar, plates (construction), wire rod (construction)
> 0.35	Q4	heavy section, light section, rail section, reinforcing bar, hot rolled bar (construction)

correspond to quality Q4 (tramp elements > 0.35%).

2.5. Material pinch analysis for steel and scrap flows

Pinch analysis represents a method that was developed to minimize the energy demand in industries (Linnhoff & Hindmarsh, 1983). In recent decades, the concept has also been increasingly applied to materials flows (Daehn et al., 2017; Ekvall et al., 2014; Hatayama et al., 2012; Hatayama et al., 2009), thereby taking into account the fact that different processes and products require materials with different purity.

In this study, a version of material pinch analysis is used per annum to assess how quality categories of steel scrap and crude steel demand in the EU-28 match. Unlike to similar works done for steel, excess scrap of lower qualities was not only considered to be diluted by primary steel but also exported out of the EU. The latter might reflect more the actual steel scrap management in case of open markets (no limitation of scrap import and exports). The steel scrap exports assessed in this manner were subsequently compared to the officially reported figures on steel scrap trade in order to evaluate whether scrap trade flows observable can be explained by the quality and quantity of steel scrap domestically available.

2.6. Validation of steel flows

To partially validate the mass flows of the steel scrap model presented, the steel flows into finished goods were cross-checked with bottom up data. Therefore, production figures of cars and trucks in the EU-28 countries (OICA 2020) were combined with estimates on the average steel content of cars and trucks. For cars, it was assumed that the average steel content increased from about 700 kg in the ‘60s and ‘70s to 900 kg today (Castellani et al. 2017, ICCT 2011, 2020, Todor and Kiss 2016), whereas for trucks (production mix of light, medium and heavy commercial vehicles) an almost constant steel content over the years of about 2,000 kg per truck was assumed. Further information on the validation data is provided in chapter F of the SI.

3. Results

3.1. Scrap quantities

3.1.1. Production & forming scrap

Production & forming scrap started at a rate of about 7 Million tonnes (Mt) per year in 1946 and peaked in 1974 at 47 Mt/yr (see Fig. 2). Today EU-28′s steel industry generates about 15.5 Mt of production & forming scrap per year. The relative scrap generation (PFS to domestic crude steel production plus net import of ingots & semis) amounts at present to about 8.7%, whereas in the 1950s almost one quarter of crude steel production ended up as PFS, indicating a strong improvement in terms of material efficiencies in the steel industry. Since 1970, when continuous casting steel production was introduced (see also Fig. A1 of the SI), significant improvements in the efficiencies are observable. Between 1970 and 1997 the share of PFS (relative to crude steel production) was halved (from 22% to 11%).

3.1.2. Fabrication scrap

The overall annual amount of fabrication scrap in the EU-28 countries rose from 3 Mt in 1946 to a maximum of about 33 Mt in around 2007, currently (in 2017) reaching a level of 26.5 Mt. In 2017 the biggest share of FS originates from the processing of Cold Rolled Coil galvanized (31%), followed by Hot Rolled Coil (20 %) and Cold Rolled Coil (11%). In general, the results of the MFA model clearly indicate the dominant role of flat products for the overall production of fabrication scrap (77%) as they are characterized by a significantly lower material efficiency during the fabrication of final goods in comparison to long products (see Fig. 3).

When the fabrication scrap is assigned to the end-use sectors of finished steel products (see SI - Fig. G2), it becomes obvious that the production of cars was responsible for 30% of the overall FS generated in the EU-28 in 2017, although only about 11% of final domestic steel use ends up in cars. Until the 1970s, the share of the car sector in total fabrication scrap was much smaller and amounted to only 8%-15%. The second most important end-use sector of steel regarding the generation of FS in 2017 represents Other Metal Goods (20%) followed by Mechanical Engineering (15%) and Buildings (13%).

The relative share of fabrication scrap (relative to crude steel production) amounts to 17% today (year 2017). This share has increased over time (in the 1950s it amounted to only about 10%) as more flat products are being produced and subsequently processed and manufactured to final goods.

3.1.3. Post-consumer scrap (End-of-Life scrap)

Balancing the process, “*6. Scrap market*” allowed the amount of End-of-Life (EoL) or post-consumer steel scrap actually recovered to be assessed. In particular, it was assumed that the sum of production & forming scrap, fabrication scrap, net-imports of scrap and the EoL scrap recovered must equal the total amount of steel scrap utilized by the EU-28′s steel industry, whereby all scrap quantities (except EoL scrap recovered) are either known by statistical data or calculated by means of the MFA model presented. The results thereby obtained for EoL scrap recovered show that the quantity increased from less than 5 Mt/yr in

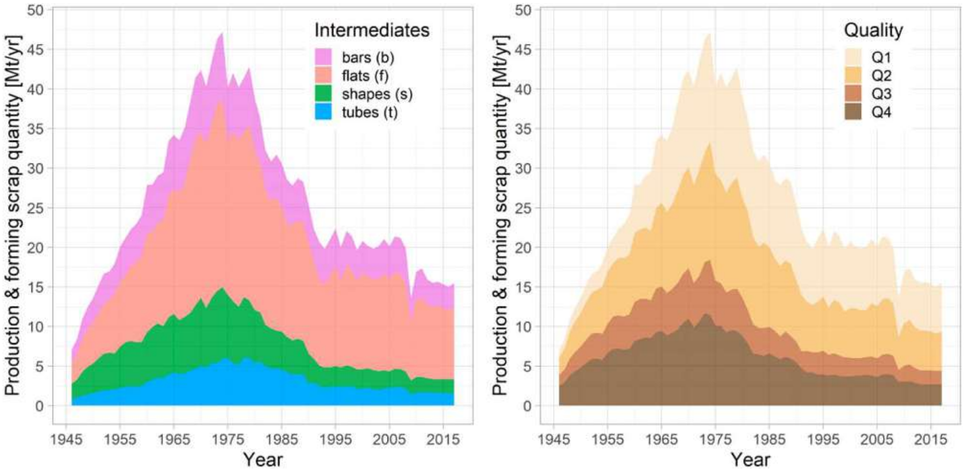

Fig. 2. Annual quantities (given in Mt/yr) of production & forming scrap generation in the EU-28 from 1946 to 2017, categorised by intermediate steel product process (left) and scrap quality based on estimated tramp element contents (right)

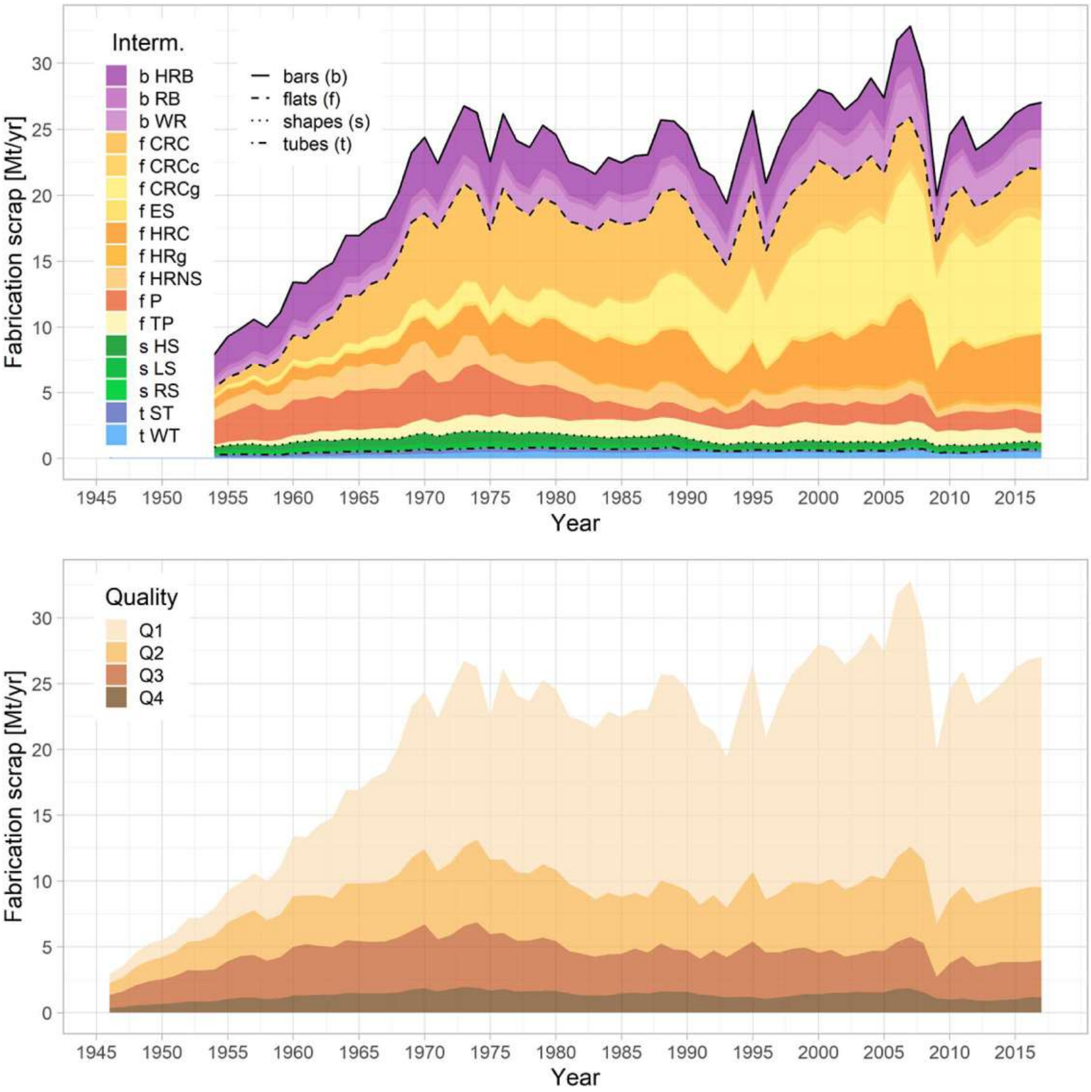

Fig. 3. Annual production of fabrication scrap (given in [Mt/yr]); quantities of fabrication scrap according to the intermediate steel products processed (upper part), according to the resulting qualities (lower part)

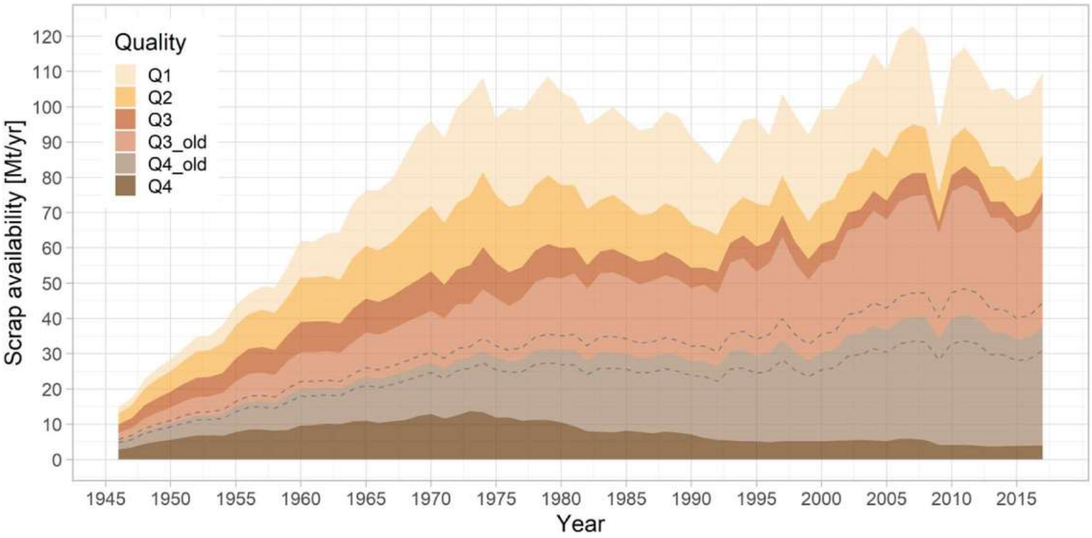

Fig. 4. Annual total of new (production & forming, fabrication) and old (end-of-life) scrap generated (given in [Mt/yr]) categorised by quality. The dashed line indicates possible variability of quality shares for old scrap (40% to 60% to Q3 and Q4, each).

1946 to almost 74 Mt/yr in 2011 (see Fig. 4). In 2017, the EoL scrap recovered amounted to approximately 68 Mt/yr. A steady increase in EoL scrap recovery can be observed over time, which might be interpreted as an indication that the steel stock in the EU-28 is still growing and not saturated yet. EoL scrap not recovered (e.g., lost via landfilling) is not considered in the data presented.

The share of EoL scrap recovered relative to the total scrap utilized increased from about 30% in the early 1950s to more than 70% nowadays, whereby until 1980 the share of EoL was still below 37%. A different trend is observable for the share of production & forming scrap. PFS amounted to more than 45% of the total scrap utilized in the initial years under investigation, while its share dropped to about 17% in 2017. The fabrication scrap share in relation to the total domestically utilized scrap in the 1950s was about 19% and increased to almost 30% in 2017.

3.2. *Scrap and steel qualities*

Besides the quantities of steel scrap flows, their qualities with respect to the content of tramp elements was assessed as well, taking four different categories into consideration (see Table 2). The results reveal that nowadays the vast majority of the new scrap (PFS and fabrication scrap) can be attributed to quality Q1 (55%) and Q2 (25%), with comparatively low levels of tramp elements (see SI – Fig. E1). Steel scrap quality categories Q3 and Q4 each account for about 10%. Until the beginning of the 60ties, the shares of the four quality categories in new scrap were almost equal.

It was assumed that EoL scrap generated only consists of the categories Q3 and Q4, with possible shares of both categories between 40% to 60%. The overall quantities of the different EoL scrap qualities are displayed in Fig. 4.

Considering the quality categories of crude steel demanded (see SI – Fig. E2), it becomes obvious that also for crude steel a shift towards category Q1 has occurred in recent decades, whereas the shares of categories Q3 and Q4 have continuously decreased. Both observations can be attributed to a shift in the European steel production from long products towards flat products.

The upper part of Fig. 5 provides snapshots of the material pinch analysis by comparing the masses of the different steel scrap qualities (potential material sources for steel) with the crude steel qualities demanded (sinks for the scrap) per annum for selected years. In particular, the masses of steel in each quality category are plotted to visualize the possible destiny of the scrap generated domestically to meet the demand. Until 1960 all scrap domestically generated could be used in steel products with tolerances greater than 0.25 % of tramp elements (category Q3 and Q4). Until the mid-90s scrap supply in the different categories was always significantly lower (at least more than 5 Mt/year) than crude steel demand in the respective quality category. In 1993 and 1994 scrap supply and steel demand for Q3 and Q4 became almost identical. This is also the period when significant net-exports of steel scrap from the EU-28 are observable for the first time (see Fig. 5, lower part). Before this time the EU-28 was a net-importer of steel scrap. For the subsequent years (until 2008) the scrap supply for the categories Q3 and Q4 increasingly surpassed the domestic steel demand in these categories, which corresponds with a growing net-export of steel scrap.

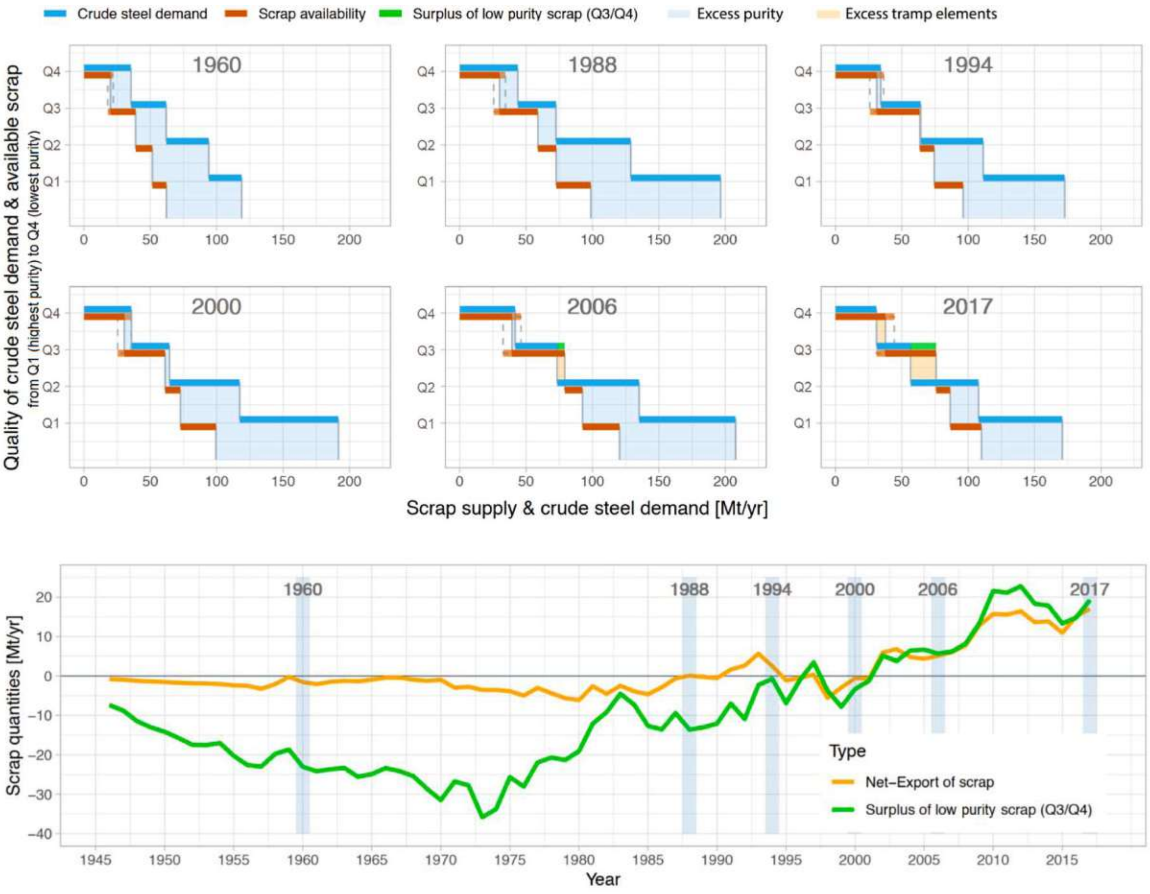

Fig. 5. Material pinch analysis for the quantities and qualities of crude steel demand and available scrap in the EU-28 for the years 1960, 1988, 1994, 2000, 2006 and 2017, orange-shaded areas show the excess tramp elements, blue-shaded areas the excess purities (upper part). Assessed scrap surplus of categories Q3 & Q4 and net-export of scrap from the EU-28 (data are given in Mt/yr). Starting from the time (mid-90s) when a surplus of scrap (positive values) occurred, a good match with the net-export of scrap can be observed (lower part). The overlap between Q3 & Q4 is based on the fact that the assignment of EoL scrap to these classes is uncertain (40% to 60% to Q3 and Q4, each).

From 2009 onwards, the annual amount of "surplus scrap" (for category Q3 and Q4) compared to crude steel demand amounted to 14 to 23 Mt/year. Since that time net-exports of steel scrap peaked and reached a maximum of 17 Mt/yr in 2017.

The temporal trend of the annual amount of "surplus scrap" modelled and the net-export of steel scrap (statistical data) match quite well, which can be seen as an indication that observed exports of steel scrap are mainly due to quality constraints. A detailed analysis of the steel scrap trade (see SI – Fig. E3) confirms that mainly EoL scrap (HS code 720449) is exported, whereas the trade of new scrap (HS code 720441) is largely balanced. The net trade of other steel scrap, such as stainless or alloyed steel scrap (720410, 720421, 720429, 720430), which contain beside new scrap also EoL scrap, is comparatively small.

Instead of exporting the excess scrap of lower purity (Q3 and Q4), diluting it with primary steel sources (e.g., pig iron, directly reduced iron) might be a suitable alternative. In Fig. 5, a comparison of excess tramp elements (shaded in orange) and excess purities (shaded in blue) is shown on a qualitative level (only steel grades are displayed on the y-axis and not the amounts of tramp elements). Comparing the quantities of tramp elements for the year 2017, it becomes obvious that the overall dilution capacity still significantly surpasses the quantities of excess impurities present in the scrap (24 kt/yr of excess tramp elements versus more than 70 kt/yr of excess purities – the respective calculations are provided in the supplementary information; see Table E2). Such a theoretical dilution of impurities, however, is difficult to implement into the steel industry at the European level, given the logistic challenges for scrap management, and the significant changes to current production routes that would be required. Furthermore, it has to be considered that PFS (and to some extent also FS) is obviously recycled at its production plant, thereby limiting the utilization of other steel scrap sources in case of production via basic oxygen furnace.

3.5. Validation of MFA model

The quantitative results of the model were partially validated by comparing the simulated net flows of steel intermediates into finished products, namely cars and trucks, with bottom up data using EU-28 production figures of cars and trucks and their typical steel contents. The results are shown in Fig. 6 and indicate a fairly good fit between model results and bottom up data. Some of the higher steel input modelled for both end-use sectors can be attributed to steel used for maintaining and repairing cars and trucks, which is not covered by the vehicle-based bottom up data used for model validation. Furthermore, the bottom up data are also characterized by a significant uncertainty (estimated to about ±10% for cars and ±20% for trucks, shown as ribbon in Fig. 6) due to limited information about the average steel contents of cars and trucks manufactured in the EU-28 countries and their temporal development.

In general, the outcomes of the comparison verify the assessed distribution of intermediates to the two finished products (cars and trucks) and the respective material efficiencies assumed for the fabrication of these goods. With respect to the scrap modelling efforts, this implies the reliability of the fabrication scrap quantities originating from the fabrication of cars and trucks, which today account for about one third of the overall fabrication scrap generated in the EU-28.

4. Discussion

Besides a tremendous increase in the overall scrap generation from 14 Mt/yr in 1946 to a maximum amount of 120 Mt/yr in 2007, the results further indicate a shift in the composition of the steel scrap generated. Whereas until 1980 most of the scrap generated was new scrap, nowadays the quantities of old scrap generated are dominant. Old scrap accounts for more than 60% of the overall steel scrap recovered in the EU-28. In the 1950s, the share of old scrap was only about 20% of the overall steel scrap generated (see SI Fig. G1).

The observed shift towards higher shares of old scrap in relation to new scrap resulted also in shift towards scrap with higher contents of tramp elements (Cu, Sn, Cr, Ni and Mo). This and the fact that in the EU-28 an increasing share of crude steel is used to produce flat intermediates (mainly driven by a significant increase in the steel end use of the transport sector), which are less tolerant of tramp elements, resulted in a surplus of steel scrap of lower purity in recent decades. This surplus of scrap with lower purity could be tackled by diluting it with new scrap or primary iron sources. Statistical data about scrap trade, however, suggests that the majority of this "surplus scrap" is exported from the EU. During the last decade, the net-export of scrap from the EU-28 amounted on average to about 14 Mt/yr, with EoL scrap representing the vast majority of the exported scrap. Until 1990, when the quantities of scrap with lower purity (Q3 & Q4) were smaller than their domestic demand, the EU-28 countries were net importers of steel scrap. A similar situation for the scrap trade is observable for the United States, where steel scrap is also net-exported to the extent of 9 to 20 Mt/yr or 10 to 20% of domestic steel scrap generation during the last decade, according to (World Steel Association 2018) and Zhu et al. (2019). Steel scrap generation in both the EU-28 and the US are, on the one hand, characterized by a large share of post-consumer scrap (>60%) enriched with tramp elements (e.g., Cu). On the other hand, the portfolio of the steel industry in both regions consists primarily of flat products with lower tolerance levels for tramp elements. Contrary to the EU, the US steel

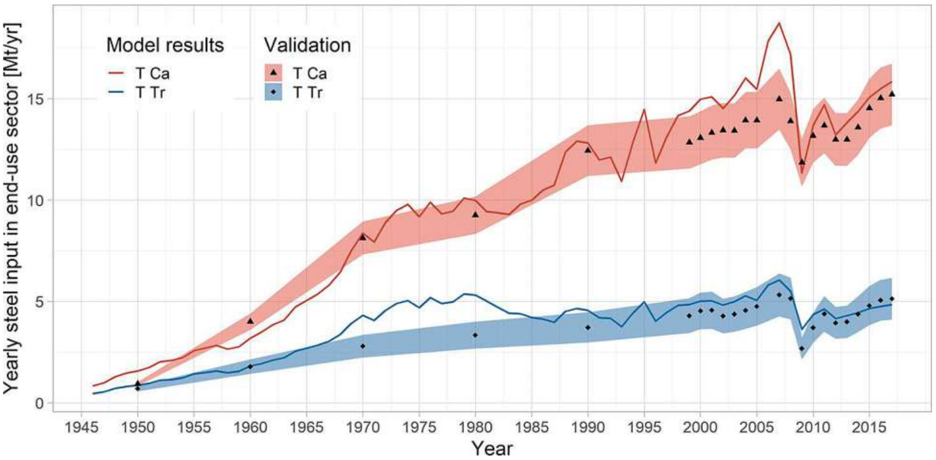

Fig. 6. Comparison of annual steel flows into final cars (T Ca) and trucks (T Tr) manufactured in the EU-28 (model data are indicated by continuous lines; bottom-up data are indicated by dots and ribbons, which indicate the uncertainty of the bottom up data)

industry is capable of producing a significant amount of flat products from electric arc furnace EAF and thus scrap based steel. However, as indicated by Zhu et al. (2019) this EAF steel for flats contains up to 50% of primary iron sources (pig iron and direct reduced iron) in order to control steel scrap impurities.

By comparison, Cullen et al. (2012) determined a share of post-consumer scrap of about 42% (relative to total scrap generation) at the global scale for the year 2008. The ratio of fabrication scrap in their study amounted to 41% and that of production scrap to 17%. In the EU-28 both shares are significantly lower today. 23% of the overall steel scrap generated in the last decade was fabrication scrap and 15% represents production & forming scrap.

Comparing the global shares of the different types of steel scrap to the historical development of scrap generation in the EU-28 countries, the situation in the EU at the beginning of the 1980s is somewhat comparable with the global situation in 2008. In addition, the distribution of steel semis to the different end-use sectors in the EU-28 40 years ago is also comparable to that of the global steel cycle in 2008. Hence, on the simple assumption that historical trends of steel use in Europe are similar to the ones at the global level in the near future, a global surplus of steel scrap with lower purity might be expectable in the coming 30 to 50 years, which is in line with results presented by Daehn et al. (2017). Scrap of this quality (with higher levels of tramp elements) is difficult to recycle without diluting it with primary steel. Today the surplus of old scrap generated in the EU-28 and in the US is counterbalanced by the international scrap trade. This, however, will not be feasible in the future if a global surplus of scrap with lower purity is produced. Hence, to improve the (domestic) circularity of steel, which is targeted by the EU circular economy package, a better separation of post-consumer scrap is mandatory (Daehn et al. 2019). Automated alloy sorting of old scrap or parts of steel-containing products, such as done in the case of EoL vehicles investigated by Ohno et al. (2015) and Willmann et al. (2017), seems to be capable of not only closing the circle of steel, but of saving valuable alloying elements present in old scrap. In addition, several technical interventions might be considered, such as the chemical removal of tramp elements from the melt (e.g., vacuum distillation, sulphide slagging) or redesign of production processes (e.g., direct strip casting (Spitzer et al., 2003)) and materials (e.g., adding interaction alloys for contra balance unfavourable properties) for a higher tolerance of tramp elements (Daigo et al., 2021). Due to practical (e.g., high energy demand, high investments costs) and technical (e.g., only prototypes or lab scale plants realized) barriers, the use of above-mentioned interventions is rarely employed so far.

The results of the study clearly indicate that higher recycling targets for steel products, as demand by the Circular Economy Action Plan of the European Union, (EC 2020), might not necessarily lead to a reduced consumption of primary iron sources in the EU, as quality constraints already today limit the domestic utilization of scrap. Hence, policy makers should consider such quality constraints, which are observable also for other commodities to be recycled at higher rates (e.g. aluminium, plastics). Besides regulating recycling rates, substitution rates should increasingly be incorporated into policies for an enhanced circular economy (see Fellner and Lederer (2020)).

5. Conclusion

The simplified MFA model for steel scrap flows presented allowed the different types of steel scrap generated (incl. their quality with respect to the content of tramp elements) in the EU-28 countries since 1946 to be assessed. Even though more precise modelling of tramp elements in steel flows, as was carried out for Japan (e.g., Oda et al. 2010; Daigo & Goto 2015; Daigo et al. 2017; Nakamura et al. 2017), would be favourable, the following can be concluded from the results of the present study:

A shift in scrap composition (towards EoL scrap and thus scrap with higher contents of impurities) can be observed over the last 75 years. At the same time, more flat steel products, which are less tolerant of tramp elements, are produced within the EU-28. Hence, since the 1990ties a surplus of scrap with higher contents of impurities can be observed which would need dilution with crude steel or primary iron sources (such as direct reduced iron) to be utilized by the European steel industry. At present however, a significant share of the scrap with lower purity is exported from the EU-28, rather than diluted with steel sources with low impurity levels.

In order to domestically recycle a higher share of steel scrap, i) the dilution potential should be exploited, ii) better post-consumer scrap separation (e.g., alloy sorting) and iii) advanced ferrous metallurgical processes (e.g., vacuum distillation, sulphide slagging) are required. The latter would not only facilitate the circularity of steel but would also allow for the recovery of valuable metals (Cr, Ni, Mo, Cu), which are currently non-functionally recycled, as demonstrated by Daigo et al. (2020).

CRediT authorship contribution statement

Sabine Dworak: Conceptualization, Data curtion, Investigation, Formal analysis, Visualization, Writing – original draft. **Johann Fellner:** Conceptualization, Supervision, Data curtion, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2021.105692.

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How will tramp elements affect future steel recycling in Europe? – A dynamic material flow model for steel in the EU-28 for the period 1910 to 2050

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ABSTRACT

Global steel production has undergone massive growth since WWII. In recent decades, however, affluent regions such as the US and the EU-28 have been experiencing a saturation of the steel market. Stagnant steel production volumes and increased post-consumer scrap volumes are the consequence. The increasing shares of post-consumer scrap provide the opportunity to increase the scrap rate (share of utilized scrap) in crude steel production. However, steel recycling has a major limiting factor: the content of specific tramp elements.

In the present study, a dynamic material flow model for steel is used to compare available scrap with crude steel demand on a quantitative and qualitative level (tramp element content of Cu, Ni, Mo, Cr and Sn). The results show that post-consumer scrap increases from 80 Mt/yr (65% of all scrap available) in 2020 to more than 100 Mt/yr (75% of all scrap available) in 2050. Based on the model, the development of the yearly surplus of low purity scrap (for which there is a higher supply than demand) was assessed via material pinch analysis. The low purity scrap surplus rises further, from today's 20 Mt/yr (2020) to 43 Mt/yr in 2050. Assuming that the current handling of scrap continues, the maximal scrap rate is shown to lie at around 55%, while the potential scrap rate (without quality constraints) could reach 75%. The dilution of low purity scrap with high purity resources would allow the utilization of all scrap until 2040 if the current collection scheme remains in place.

1. Introduction

Steel is not only one of the most commonly used commodities, but is also applied widely in all end-use sectors. On a global scale, the steel industry has experienced extensive growth in recent decades (World Steel Association, 2020), which has been mainly driven by emerging regions, such as Asia (Hatayama et al., 2010). In affluent economies, steel in-use stock is mostly saturated (Hatayama et al., 2010; Müller et al., 2011; Pauliuk et al., 2013b), which results in stagnant steel consumption rates in these regions. This stagnation makes scrap increasingly relevant as raw material for steel production. The usage of scrap for crude steel production is not only beneficial from a resource point of view, but also helps reduce CO_2 emissions (Broadbent, 2016) and brings other environmental benefits, such as lower eutrophication, acidification and photochemical oxidation, in comparison to steel production from primary resources (Hu et al., 2014; López et al., 2020).

Steel flows have been extensively investigated on various levels, in varied degrees of detail and in different timeframes (retrospectively and in terms of forecasting: e.g., Cooper et al. (2020); Cullen et al. (2012); Hatayama et al. (2010); Müller et al. (2011); Pauliuk et al. (2013b, 2013a); Zhu et al. (2019)). However, analyses of steel flows on an European level are rather rare (e.g., Dworak and Fellner (2021); Passarini et al. (2018)).

In recent decades, quantitative analyses of steel flows have been extended to include qualitative aspects. To do so, the presence of impurities in steel and steel scrap (tramp elements such as Cu, Ni, Mo, Cr or Sn), their impact on recycling and possible technical interventions to deal with such impurities have been investigated (e.g., Daehn et al. (2019); Daigo et al. (2021); Noro et al. (1997); Sampson and Sridhar (2013); Savov et al. (2003); Spitzer et al. (2003)). However, only very few studies are available which quantify the scrap flows and associated tramp element flows and put the scrap availability in relation to crude steel demand (e.g., globally for Cu (Daehn et al., 2017), retrospectively for the former EU-28 (Dworak and Fellner, 2021), and for Japan (Daigo

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et al., 2017; Igarashi et al., 2007; Oda et al., 2010)). What the results of these studies have in common is that they indicate a surplus of low purity scrap, which requires dilution with primary steel in order to be recycled. On a global scale, this surplus might arise in the near future (see Daehn et al. (2017)). In Japan and Europe it is already prevalent, as highlighted by Igarashi et al. (2007) and Dworak and Fellner (2021). At present, this existing surplus is largely balanced by the export of scrap as both the European Union and Japan are net exporters of steel scrap (+16 Mt/yr and +8 Mt/yr). For the US the situation is similar.

The extent to which this current surplus of low purity scrap might increase in the future, and thus further limit the domestic recycling of steel scrap in affluent economies, has not been investigated so far.

Hence, the objective of the present paper is to assess the long-term development (until 2050) of steel scrap generation and its composition with regards to the total contents of tramp elements (Cu, Sn, Cr, Ni and Mo) in the European Union and to compare the domestic steel scrap supply with the crude steel demand in terms of both quantity and quality. To do so, a dynamic top-down material flow model for steel (taking steel impurities into account) is developed and applied for the territory of the former EU-28 and the period from 1910 to 2050. In order to enhance the significance of the results, the model is partially validated with bottom-up data, which represents an advance relative to the existing dynamic steel models.

2. Material and methods

2.1. Model structure

The proposed system boundaries correspond to the geographical border of the former EU-28 over the time period of 1910 to 2050. The start of the timeframe is chosen to take stock accumulation and resulting post-consumer scrap for sectors with long lifetimes into account. The flows in the model are balanced, based on the law of mass conservation. Fig. 1 displays the simplified steel flow model (covering carbon steel and cast iron and steel, but not considering stainless steel) for the former EU-28. Stainless steel is not considered since the accompanying elements in these sorts are seen as resources for alloying rather than as tramp elements. Hence, stainless steel is managed differently. Furthermore, the average share of stainless steel in relation to total steel production is less than 10% for the period considered. It contains 7 processes ((1). Crude steel production, (2). Production and trade of intermediate products, (3). Fabrication of finished products, (4). Trade in finished steel products, (5). Consumption, (6). Waste Management and (7). Scrap market) and 16 flows of steel and steel scrap (CrS - Crude steel, IP - Intermediate steel products, FSP - Finished steel products, PoCSg - Post-consumer scrap generated, PoCSr - Post-consumer scrap recovered, Sr - Scrap recycled, PFS - Production & forming scrap, FS - Fabrication scrap, NIS - Net-import of ingots & semis, RM - Raw materials, NIFSP - Net-import of finished steel products, EEoLP - Export of end-of-life products, NISc - Net-import of scrap, CI - Cast Iron, NIIP - Net-import of intermediate steel products, NEUFSP - Net End Use of finished steel products). Process 1 (Crude steel production) and Process 7 (Scrap market) are not balanced and are therefore located outside of the system boundaries. For the sake of completeness, however, they are shown in Fig. 1. The steel flow model is largely based on the work of Dworak and Fellner (2021). All blue flows are based on statistical data or determined via the application of transfer coefficients. Transfer coefficients are used to partition specific inputs to specific outputs. In this study, the transfer coefficients are applied to determine specific steel intermediate products to the end-use sectors, and also to determine the fabrication scrap based on steel product and end-use sector specific material efficiencies. A detailed description of their derivation is given in Dworak and Fellner (2021). The current work focuses on the determination of the red flows, especially on the amount and composition of PoCSr as well as the in-use stock. The steel flow model is built up in multiple layers. On the one hand, the flows of specific intermediates can be mapped throughout the system. Further, each intermediate steel product is split up into the corresponding end-use sectors. Altogether 19 intermediates (casts - Cast Steel (c CS), Cast Iron (c CI); flats - Electrical Strip (f ES), Tin Plated (f TP), Plate (excl. plates used for welded tubes) (f P), Cold Rolled Coil galvanized (f CRCg), Cold Rolled Coil coated (f CRCc), Cold Rolled Coil (f CRC), Hot Rolled Coil galvanized (f HRg), Hot Rolled Narrow Strip (excl. Strips used for welded tubes) (f HRNS), Hot Rolled Coil (f HRC); tubes - Welded Tubes (t WT), Seamless Tubes (t ST); bars - Wire Rod (b WR), Reinforcing Bar (b RB), Hot Rolled Bar (b HRB); shapes - Heavy Section (s HS), Light Section (s LS), Rail Section (s RS)) and 4 end-use sectors with altogether 10 sub-end-use sectors (Construction - Buildings (C Bu), Infrastructure (C In); Industrial Equipment - Mechanical Engineering (I ME), Electrical Engineering (I EE); Transport – Cars (T Ca), Trucks (T Tr), Other Transport (T OT); Metal Goods - Other Metal Goods (MG OMG), Appliances (MG Ap), Packaging (MG Pa)) are distinguished according to similar studies (Cullen et al., 2012; Zhu et al., 2019).

This multilevel approach allows quality classes to be assigned to the

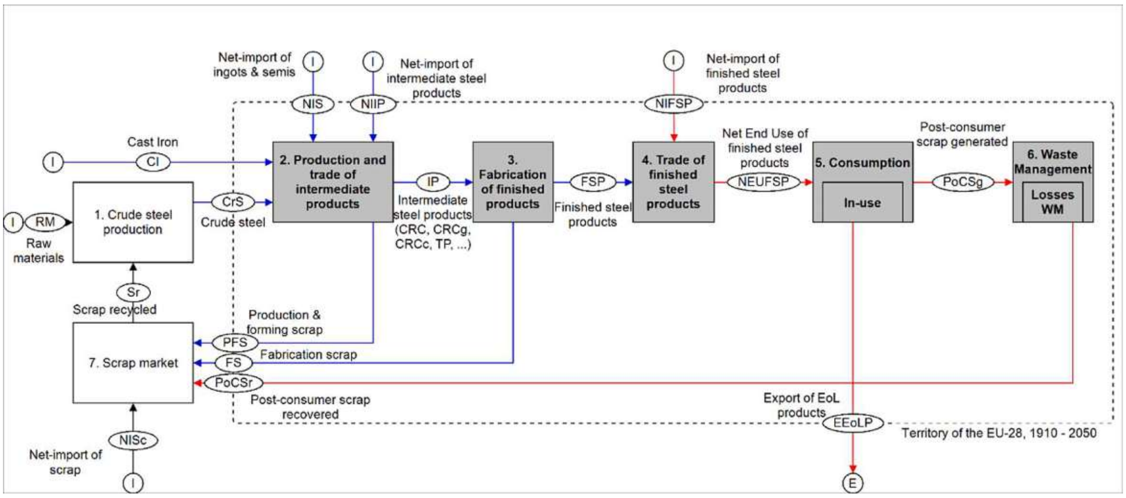

Fig. 1. Simplified MFA system for assessing steel and scrap flows in the former EU-28 (the red flows are determined in this study via dynamic modelling, whereas the blue flows are based on statistics and transfer coefficients (Dworak and Fellner, 2021)); processes 1 and 7 are within the borders of the former EU-28, but not considered/balanced within the framework of the present study and are therefore located outside the system boundary; In-use: steel stock in use, Losses WM: Losses in waste management system to e.g. landfills. In addition to steel, the flows of cast iron were considered as well.

specific steel intermediates with regards to allowed levels of impurities depending on the assigned end-use sector. Four classes of steel and steel scrap are distinguished (Q1-Q4) based on total content of the tramp elements Cu, Sn, Cr, Ni and Mo (Daigo et al., 2010; Dworak and Fellner, 2021), plus an additional quality class for cast products (Q0). Following this principle, we can estimate the quality of all flows and stocks within the system boundaries.

Based on the domestic fabrication of the final steel products (FSP) and the trade thereof (NIFSP, where the same sectoral composition as for FSP for the corresponding year is assumed), the domestic end-use of finished steel products (NEUFSP) is determined. End-of-life products (EoLP) are determined on the basis of Weibull life-time functions, depending on the corresponding end-use sector. Taking the export of end-of-life products (EEoLP) into consideration, which is especially relevant regarding cars, the post-consumer scrap generated (PoCSg) can be determined. Further processing in the waste management system leads to losses, which are considered equally distributed within each sector (meaning, of all steel intermediates exiting the sector in the corresponding year, the same share is regarded as loss and the sectoral composition is identical in PoCSg and PoCSr). The output of the waste management system is the PoCSr, which then becomes available for recycling or trade.

2.2. *Input data*

The input data was compiled from various data sources, which are briefly described in the following. The blue flows in Fig. 1 are mostly based on statistics and transfer coefficients (for details see Dworak and Fellner (2021)).

2.3. *Historical data and basic input data*

Intermediate steel products (IP): The starting point of the calculation for the period from 1946 to 2017 is the reported production data of IP (adapted from Dworak and Fellner (2021)). From 1910 up to 1945, shares of steel intermediates were assumed to correlate with the average shares of the years 1946–1950, which is definitely a gross simplification as two world wars significantly influenced steel use during this time. Nonetheless, the overall impact of this simplification on the present and future scrap supply is negligible. The total amount of steel production of the EU correlates with world steel production (Kelly and Matos, 2014), again using the relation between EU-28 and world steel production for the period from 1946 to 1950 for scaling.

Production and forming scrap (PFS): Quantities of PFS were taken from Dworak and Fellner (2021), who applied time- and production route dependant PFS rates to estimate the quantities of the crude steel production (CrS) and the amount of PFS.

Final steel products (FSP) and fabrication scrap (FS): We applied transfer coefficients dependant on (i) time, (ii) end-use sector and (iii) steel intermediate to determine FSP. Material efficiencies, likewise dependant on time, end-use sector and steel intermediate, were applied to determine FS. Both transfer coefficients and material efficacy are based on Cullen et al. (2012) and calibrated according to Dworak and Fellner (2021).

Net-import of finished steel products (NIFSP) and net end use of finished steel products (NEUFSP): Quantities of traded final steel products were derived from the UN Comtrade database (UN Comtrade, 2020) and the corresponding steel contents were adopted from the World Steel Association (World Steel Association, 2018). For the NIFSP, the same composition as for FSP (in terms steel qualities) for the corresponding year is assumed. Detailed information about the determination of the trade flows is provided in the Supplementary Information. By balancing *Process 4. Trade of finished steel products*, the net end use of finished steel products NEUFSP was determined.

End-of-life products (EoLP equals the sum of EEoLP and PoCSg and is not shown as a separate flow): As mentioned above, EoLP was determined by applying Weibull lifetime-functions to the NEUFSP. Weibull functions are quite well suited for use as lifetime-function as they are finite (no negative values) and their form is easily adjustable (width and skewness, e.g. (Melo, 1999)). Even though some studies aim to estimate the variance of lifetimes over the years in general (e.g. via the volume correlation model (Gauffin et al., 2015)), data on the variation of lifetimes for specific sectors is rarely available. For the sake of consistency and simplicity, we decided to keep the lifetime functions for the different sectors constant over time. The parameters applied for the lifetime functions are summarized in Table 1.

In-use stock (In-use): The initial steel stock in 1910 was assumed to be zero, which is a simplification as Klinglmair and Fellner (2011) and Pauliuk et al. (2013a) have shown in their work that the per capita steel stock in the Austro-Hungarian Empire and Western Europe amounted to approximately 1.2 and 2 t/cap in 1910. Nonetheless, this simplification is justified by the fact that the stock present 100 years ago is of minor significance for the steel stocks today and thus also for steel scrap generation today. Nevertheless, the model might underestimate the in-use stock for the first few decades.

Export of end-of-life products (EEoLP): Unknown whereabouts in the Transportation sector (Cars and Trucks) is a well-known issue in the former EU-28 (e.g. manifested in the ELV-directive (EC, 2000)). In this model, vehicles (cars or trucks) with unknown whereabouts (possibly due to illegal export, vehicle theft, or vintage vehicles kept by car enthusiasts on private properties (Oeko-Institut e.V., 2017)) are considered via a constant rate. The determined share of end-of-life vehicles with unknown whereabouts is a little above 30% of the deregistered and not re-registered or reported as end-of-life vehicles in waste statistics. The corresponding reports do not discriminate between cars and trucks. We assumed that 30% of steel contained in all end-of life cars and 70% of steel contained in all end-of-life trucks is of unknown whereabouts. The rates applied are also provided in Table 1.

Post-consumer scrap generated (PoCSg): By subtracting the corresponding EEoLP from the EoLP, the PoCSg was determined.

Post-consumer scrap recovered (PoCSr): To determine PoCSr, the corresponding losses through processing in waste management were determined by means of sector-specific recovery rates. We chose to decrease the recovery losses between 2010 and 2050 to account for efforts to improve recovery rates. To do so, we assumed that the rate of losses decreases linearly until 2050 by 30% in comparison to the losses in 2010. Only for the Transport sector (Cars and Trucks) is more specific data available. Therefore, based on these data, the recovery rate of steel from shredded EoL cars and trucks is adjusted at several points in time (2006, 2009, 2018, 2030). Detailed sources and derivation of the values

Table 1
Input parameters for the dynamic MFA model; lifetimes derived based on (Dahlström et al., 2004; Davis et al., 2007; Gauffin et al., 2015; Hatayama et al., 2010; Huuhka and Lahdensivu, 2016; Melo, 1999; Michaelis and Jackson, 2000; Müller et al., 2011, 2006; Neelis and Patel, 2006; Oda et al., 2013; Pauliuk et al., 2013a; Wang et al., 2007); EEoLP derived based on (Oeko-Institut e.V., 2017); recovery rate based on (EC, 2000; Pauliuk et al., 2019; UBA and BMU, 2019). The recovery rate is constant up until 2010 and then liniarly intropolated to increased recovery rate (30% less losses), except for Cars and Trucks (for details see Supplementary Information).

End-use sector	Lifetime	EEoLP	Recovery rate	
	Average lifetime	Rate	Up to 2010	2050
C Bu	65	–	82%	87%
C In	65	–	82%	87%
T Ca	17	30%	82%	98%
T Tr	17	70%	82%	98%
T OT	55	–	82%	87%
I ME	17,5	–	87%	91%
I EE	15	–	87%	91%
MG OMG	14	–	58%	71%
MG Ap	14	–	58%	71%
MG Pa	1	–	58%	71%

are provided in the Supplementary Information. The recovery rates applied are provided in Table 1.

2.4. *Future projection of steel and scrap flows*

The projection is based on the mean value of the period 2003 to 2017 to account for the intense fluctuations in recent years. A stagnant crude steel production rate was assumed based on the fact, that steel production rates are more or less stagnant for the last decades in Europe. It was assumed that the quantity of each specific flow (in every layer) stays in the same relation (no significant changes in composition of flows).

Three simple scenarios for the future development of steel use in the former EU-28 are considered. They assume either a moderate growth in steel use of 0.5% per year (Scenario “growth”), a constant steel use (Scenario “zero growth”) or a reduction in steel use of 0.5% per year (Scenario “de-growth”). Modelling based on stock saturation, which is a common approach for dynamic models, comes with high uncertainties, especially in lights of expected societal changes (e.g., transformation to a low carbon society). To evade additional uncertainties, and for simplicity reasons, only the above-mentioned scenarios are considered.

A stock-driven model for dynamic steel MFA as, for instance, applied by Pauliuk et al. (2013a) and Hu et al. (2010) was not considered as too many factors (e.g. implementation of shared economy in the Car sector, increase in public transport due to climate mitigation targets, development of infrastructure, etc.) make assessments of future stocks of steel in the different sectors highly uncertain. Therefore, we decided to use a flow-driven model as was also applied by Igarashi et al. (2007).

2.5. *Steel quality assessment*

2.5.1. *Quality assessment of steel products and new scrap (production and forming scrap, fabrication scrap)*

The quality assessment was adopted from Dworak and Fellner (2021). Four quality classes (Q1-Q4) of steel are defined on the basis of the total content of five major tramp elements (Cu, Sn, Cr, Ni and Mo), and an additional class for cast iron and cast steel products (Q0) was introduced. The classes are assigned based on the steel intermediate and the designated end-use sector (for details, see Dworak and Fellner (2021)). As (Dworak and Fellner, 2021) argue, the data availability regarding tramp elements is not sufficient to follow another, more precise, approach.

2.5.2. *Quality assessment of post-consumer scrap*

The post-consumer scrap was categorized using three different options:

Option A: The post-consumer scrap generated was assigned a sector-specific quality class (for instance, scrap from EoL cars was assigned quality Q4 due to the potential contamination with copper during EoL processing). For the sectoral assignment of the steel qualities to the different EoL products, literature data was applied (Q1: Metal Goods – Packaging, Q2: none, Q3: Transportation, Industrial Equipment, Metal Goods (except Packaging), Q4: Construction; based on (Daehn et al., 2017; Daigo et al., 2017, 2005; Eurofer, 2016; Hatayama et al., 2014; Igarashi et al., 2007; Savov et al., 2003; Schrade et al., 2006), for details see Supplementary Information). Option A considers contamination which might occur during use or during waste management when no enhanced sorting or decontamination procedure is in place. It represents in many respects the “worst case scenario” regarding the post-consumer scrap quality investigated in this paper.

Option B: The assignment of quality classes of steel scrap to the different EoL products was based on the average content of tramp elements $c_{Tramp,av}$ present in the steel of the sector. Contamination of the steel scrap via other metals (e.g., copper cables) during dismantling and processing of EoL products have been disregarded for option B. In determining the average content of tramp elements in the sector-specific steel mix of EoL products, normal distribution for the tramp elements in each quality class was assumed.

$$\overline{c_{Tramp,x}} = \frac{\sum_{i=1}^{n} c_{Tramp,ix} \cdot IM_{ix}}{\sum_{i=1}^{n} IM_{ix}}$$

With

$\overline{c_{Tramp,x}}$ Average content of tramp elements in the steel mix of the respective sector [%]

$c_{Tramp,ix}$ Content of tramp elements in the steel intermediate i for the product x [%]

IM_{ix} Mass of steel intermediate i for the product x [Mt/yr]

Option C: For this option, total disassembly of steel products, subsequent alloy/steel quality sorting and no contamination is assumed. This means that the post-consumer scrap can be sorted into the different steel intermediates (and their respective qualities) used to manufacture the finished steel products. This option represents a hypothetical “best case scenario” regarding the post-consumer scrap quality investigated.

2.6. *Sensitivity analysis*

To evaluate the robustness of the model, a sensitivity analysis was performed. The surplus of low purity scrap (Q3 & Q4, tramp element content higher than 0.25%) in relation to crude steel demand serves as the major results, whose changes are to be assessed. The sensitivity was analysed for the following four groups of parameters:

- Sector split: for the future prediction (2019–2050) we assumed varying splits of two end use sectors (Construction and Transport). We considered a variation of ±10% (relatively) for Construction and Transport (marked as “C+”, “C-”, “*T*+”, “T-”), each separately, while the remaining sectors splits were scaled accordingly.
- average lifetime (shape of lifetime function was kept constant): a variation of the average life time ±20% (marked as “upper” and “lower”) was applied.
- Export rate of end-of-life products: the export rate of EoL products was varied by ±20% relative to the base value (marked as “upper” and “lower”).
- recovery rate of post-consumer scrap generated: a reduction in losses of 20% relative to the base value (marked as “upper”) was assumed. As the efficiency of the waste management system in the future will rather increase than decrease, only an increase in recovery rates was considered.

The sensitivity analysis was conducted for each year modelled (1910–2050). Due to rather poor data (only rough estimates via projection) and not yet accumulated in-use stock, the results of the first few decades are not meaningful. Therefore, and for reasons of relevance, the sensitivity analysis will be discussed only from the year 1980 onwards.

2.7. *Material pinch analysis*

Pinch analysis was originally established to minimize energy demand in industries (Linnhoff and Hindmarsh, 1983) by taking into consideration the fact that different processes require different heat and pressure levels. It was further developed and has been used for some decades now to analyse material flows (Daehn et al., 2017; Ekvall et al., 2014; Hatayama et al., 2012, 2009). In this context the method takes into account that different materials and processes require different purities. In this study a semi-quantitative material pinch analysis (analogue to the work of Dworak and Fellner (2021)) is applied to compare the annually available scrap with crude steel demand in terms of the required purity. The results of this semi-quantitative analysis show to what extent the quantity and composition of the scrap (regarding tramp elements) meets the demand for crude steel.

2.8. *Model validation*

In order to increase the reliability of the model, specific stocks and flows were independently validated with bottom-up data. In particular, bottom-up stock assessments were conducted for cars and for reinforcement bars in buildings. Both together account for up to 18% of the total steel stock. A top-down approach was chosen for the validation of scrap quantities becoming available.

2.8.1. *Bottom-up stock assessment for buildings and cars*

For the Transport sector (subsector Cars) official registration numbers (eurostat, 2021) were used to validate the stock in the end-use sectors. The car weight was derived via estimated average weights of the reported weight classes (less than 1000 kg, from 1000 kg to 1249 kg, from 1250 kg to 1499 kg, 1500 kg or over). For each weight class the average weight was assumed (less than 1000 kg: 950 kg; from 1000 kg to 1249 kg: 1125 kg; from 1250 kg to 1499 kg: 1375 kg; 1500 and over: 2100 kg). A steel content of 65% (based on (Todor and Kiss, 2016)) was applied. The uncertainty of the stock determined in this manner was estimated at ±10%. This bottom-up based assessment of steel stock in the car fleet was then compared with the dynamically modelled stock of the Car sector.

The in-use stock of reinforcement bars was determined based on average concrete use and steel use in reinforced concrete based on newly built and demolished buildings. Most data were taken from Nemry et al., 2008).

A detailed description of data sources and the procedures applied for the bottom-up stock assessments is provided in the Supplementary Information.

2.8.2. *Comparison with reported scrap data*

The outcome of the dynamic MFA model (basically the scrap quantities generated) was validated by reported scrap quantities becoming available. Therefore, the total scrap quantities determined by the dynamic model, including production and forming scrap, fabrication scrap and post-consumer scrap, were compared with officially reported data on scrap generation. For the latter, the reader is referred to the work of (Dwork and Fellner, 2021), who assessed the scrap becoming available based on data provided by Eurostat (1946–1966) and Worldsteel (1967–2017).

3. Results

3.1. *Quantitative analysis*

3.1.1. *Scrap*

Over the period investigated (1910–2050), the total amount of available scrap (sum of production & forming scrap, fabrication scrap and post-consumer scrap recovered) constantly increases from about 20 Mt/yr in the beginning of the 20th century to 130 Mt/yr in 2050. In the first few decades (from 1910 onwards), post-consumer scrap may be underestimated as the in-use stock was assumed to be zero at the beginning of dynamic modelling in 1910.

As shown by Dwork and Fellner (2021), production & forming efficiency increased tremendously between the 1970s and the 1990s, which led to a decrease in production and forming scrap (from about 50% of the total scrap available in the 1950s to 12% from the 2020s onwards). The share of fabrication scrap is more or less constant (the share varies between 20% and 30%, trending toward 20% since the 2020s). Nevertheless, the amount of total scrap available is constantly increasing due to the vastly increasing post-consumer scrap recovered (PoCSr). PoCSr quantities rose from about 10 Mt/yr in 1960 (about 20% of the overall scrap quantity) to 80 Mt/yr in 2020 (or 65% of total amount) and will further increase to more than 100 Mt/yr (slightly less than 75%) in 2050 (see Fig. 2), which is in line with results of (Oda et al., 2013).

3.1.2. *In-use stock*

The in-use stock constantly increases, although with less and less intensity. In the zero-growth scenario, the expected plateau is not yet reached (around 2075 the stock would be saturated). While an in-use stock of 3000 Mt (6.7 t/cap) can be quantified in 1980, it rises up to 4700 Mt (9.5 t/cap) in 2010 and 5300 Mt (10.5 t/cap) in 2050, which is in line with results of Müller et al. (2011) and Pauliuk et al. (2013b, 2013a).

3.2. *Export of end-of-life products*

Based on the applied rates of export of end-of-life vehicles (30% for cars, 70% for trucks), the share of steel exiting the system this way is about 5% to 8% of the post-consumer scrap generated (PoCSg). The amount rises constantly up to 2020, when it peaks at around 7 Mt/yr.

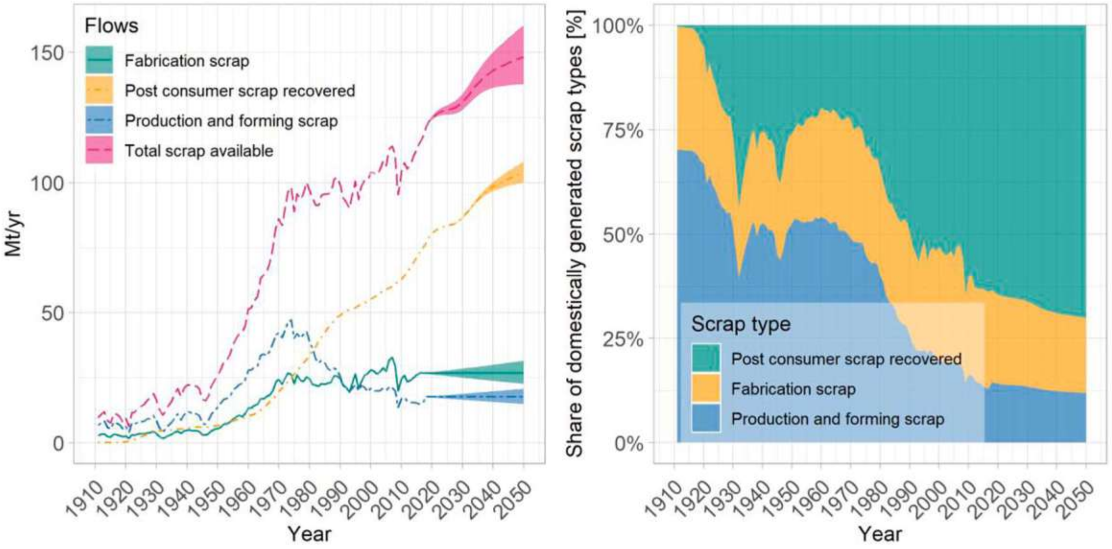

Fig. 2. left: Modelled quantities of flows scrap becoming available, categorized into forming & production scrap, fabrication scrap and post-consumer scrap as well as the total amount (sum of all scrap cathegories). The ribbon indicates the three scenarios (upper bound: growth; line: zero growth; lower bound: de-growth); right: Shares of scrap types: production & forming scrap, fabrication scrap and post-consumer scrap.

Until 2050, a slight decrease in the steel exported by EEoLP to 6 Mt/yr is expected due to the production peak of cars in the early 2000s.

3.3. *Steel qualities and sector shares – scrap, stock and crude steel*

3.3.1. *New scrap (production & forming scrap, fabrication scrap)*

For the period 1946 to 2017, the quantities and qualities of new scrap (production & forming scrap, fabrication scrap) were adopted from Dworak and Fellner (2021). The composition for the preceding period (1910–1945) was projected back with shares based on the period 1946–1950 covered by the investigation of Dworak and Fellner (2021). Hence, the shares of steel qualities Q1, Q2, Q3 and Q4 present in new scrap during the period 1910–1945 are 17%, 31%, 23% and 29%, respectively. Similarly, for future prediction, shares of the different qualities are based on the average shares of the years 2003 to 2017. Hence, for the period 2018–2050 the shares of Q1, Q2, Q3 and Q4 in new scrap are 53%, 25%, 11% and 10%, respectively.

3.3.2. *In-use stock*

Sector split: The major part of the steel in-use stock is allocated in the Construction sector (Buildings and Infrastructure). The high share of the steel stock present in the Construction sector of about 70% in the late 1970s will further increase to over 80% in 2050. This sector is also the driver for the overall stock increase due to its rather long lifetimes. The stock in the remaining sectors rises in varying degrees of intensity until the late 2000s, whereas afterwards they either slightly decrease or subsequently flatten, depending on their lifetime. Only the subsector Other Transport is characterized by a further stock increase due to its comparatively long average lifetime (55 years). The share of the overall steel stock amongst the Industrial Equipment, Metal Goods and Transport sectors decreases from 11%, 10% and 8%, respectively, in the 1980s to 6% (Transport and Industrial Equipment) and 5.5% (Metal Goods).

Quality: The steel qualities (impurity levels) assigned to the in-use stock are based on the quality of the steel intermediates comprising the final steel products. The share of Q0 (cast products) constantly decreases, from 14% (350 Mt/yr) in the mid 1970s to 6% (300 Mt/yr) in 2050. Steel of the lowest purity level (Q4) assumes the largest share in the total in-use stock by 2050 (about 30%). From then on, steel quality Q2 becomes dominant; both amount to about 1600 Mt. The share of Q3 steel relative to the overall stock is rather stable at about 15%. Nonetheless, stock quantities of Q3 almost double from the 1980s (500 Mt) to 2050 (850 Mt). Q1's share in the stock increases most, rising from 5% (80 Mt) in 1960 to 19% (1000 Mt) in 2050. In general, a shift from lower purity steel (Q3 & Q4, tramp element content higher than 0.25%) to higher purity steel (Q1 & Q2, impurities below 0.25%) can be observed for the in-use stock.

3.3.3. *Post-consumer scrap*

Up to the 1990s, all sectors deliver constantly increasing amounts of PoCSr. The biggest share can be attributed to the Mechanical Equipment sector, which yields the highest amount of PoCSr up until the mid-2000s. The share of Industrial Equipment in PoCSr constantly decreases, from 40% in 1950 to less than 20% of the total PoCsr in 2050. From 2010 onwards, the Construction sector (Buildings and Infrastructure) becomes the dominant source of PoCSr. In 2050, it is forecasted that more than 50% of PoCSr originates from Construction. The Transport sector (Car, Truck and Other Transport) accounts for a constant share of around 10% up until the 1980s, after which the share increases and reaches the highest value in the 2010s, with slightly more than 15%. By 2050, its share will again decrease to around 10%.

The share of cast products constantly decreases from 25% in 1970 to 6% in 2050. As scrap from cast products (Q0) is considered separately and the amount is the same for all options, the following results focus on the remaining qualities (Q1-Q4) only.

For Option A, the sector-specific quality classification of post-consumer steel scrap, the vast majority of PoCSr is low purity scrap. Only separately collected food packaging can be recovered at quality class Q1 (see Fig. 3). The other streams of post-consumer scrap becoming available are classified as Q3 and Q4, whereas the share of Q4 is constantly increasing, driven by the increasing share of PoCSr from the Construction sector. Until the mid-2020s Q3 dominates the PoCSr, after which Q4 assumes the largest share.

Option B: The results for this approach are by definition somewhere in between the results of the two other options (A and C). Still, some results are remarkable: Even though some sectors (especially C In and C Bu) receive most of the lowest steel quality class (Q4), the summed-up tramp elements of the sectors correspond to Q3. It should, however, be mentioned that the calculated average concentration is close to the threshold to Q4. The same principle applies to the higher share of Q1 in comparison to Option C. The calculated mean values for the Q1 scrap fractions are rather close to the threshold for Q2. Specific data about the concentration of the tramp elements in the different scraps is provided as a spreadsheet in the Supplementary Information.

For option C, the post-consumer scrap exits the consumption process of the same quality as the intermediate steel products comprising the final steel products. Scrap of quality Q4 is mainly delivered and subsequently yielded as scrap by the Construction sector, therefore the share of Q4 is constantly rising similar to the yield of the scrap from the Construction sector, even if less intensively. The share of Q2 is rather constant (slightly above 25%).

3.4. *Comparison of scrap availability and crude steel demand*

Besides the quantity and quality of available scrap, the crude steel requirements (quantity and quality) were determined in order to compare scrap availability with crude steel demand in the territory of the former EU-28. As discussed above, the PoCSr are becoming increasingly relevant as a major source for steel scrap. Crude steel demand, on the contrary, is dominated by increasing quantities of high purity steel (due to an ever-increasing demand for flats). In the upper row of Fig. 4, crude steel demand and scrap availability for Option A and Option C, with regard to their quantity and quality, are presented. From a quantitative perspective, an increasing share of the EU's crude steel demand might be produced out of scrap. In the 1990s, the potential scrap-based production rate of steel amounted to 50%, increased in 2020 to 65% and will further increase to 75% by 2050. After 2050, the potential scrap-based production rate of steel will further increase.

However, the results of material pinch analysis also clearly indicate that without implementing alloy/steel quality sorting, the quantity of low purity scrap (Q3 & Q4) exceeds the respective demand of steel from 2009 onwards. The lower part of Fig. 4 shows snapshots of the material pinch analysis for the corresponding years for Option A (upper row) and Option C (lower row). While up to 2008, the crude steel demand could take up all of the low purity scrap (tolerance greater than 0.25% of tramp elements, Q3 & Q4) becoming available in Option A, starting from 2009 the surplus of low purity scrap constantly rises, up to almost 50 Mt/yr by 2050 (which equals more than 1/3 of the overall scrap generated). By 2020, this surplus is almost exclusively composed of Q3 steel scrap. However, later on Q4 scrap also exceeds the respective crude steel demand (24 Mt/yr in 2050). When calculating the dilution potential for the tramp elements present in steel scrap (for option A), it turns out that by 2040, steel scrap generated could theoretically be diluted by the necessary quantities of crude steel from primary sources. However, later on, there is a surplus of tramp elements present in the steel cycle, which cannot be managed by simply diluting the scrap. The respective results are given in the Supplementary Information in the form of a spreadsheet.

3.5. *Sensitivity analysis*

The sensitivity analysis was performed in reference to the low purity (Q3 & Q4) scrap surplus on four parameters (lifetime, EEoLP, recovery

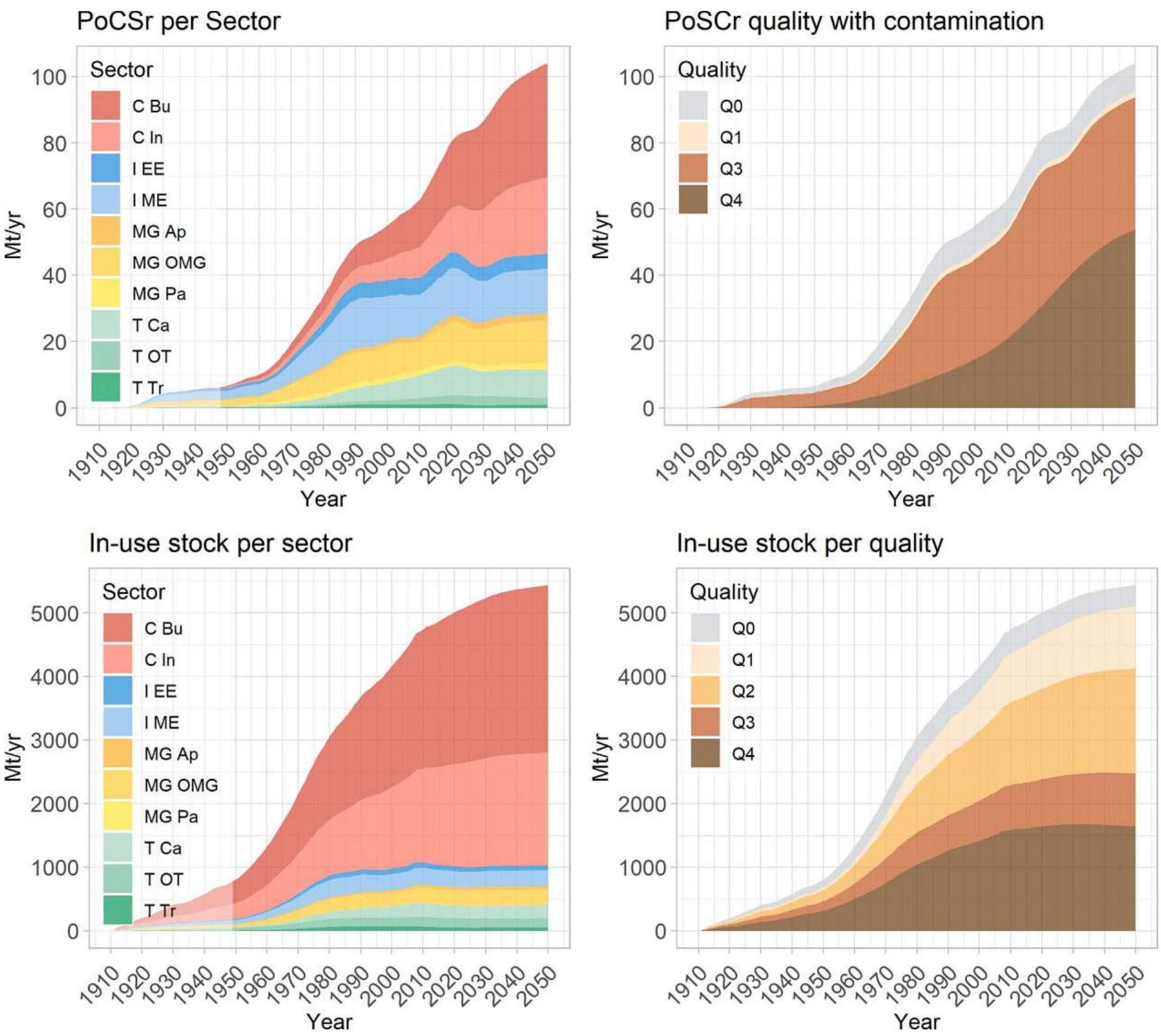

Fig. 3. Post-consumer scrap recovered (PoCSr) with sector origin (upper left), with quality classification according to Option A (purity based on sector of origin, lower right), Option B (average purity per sector based on inputs, no contamination considered, lower left), Option C (purity of input = purity of output, upper right). The steps in quality class changes can be attributed to the sharp distinction between the quality classes, which means that if the input quality of a sector changes, the whole subsector might switch to another quality class (as e.g. MG OMG from 1992 to 1993 and I ME from 2020 to 2021, with both switching from average Q2 to Q1). Abbreviations of sectors: Construction: C Bu - Buildings, C In – Infrastructure; Industrial Equipment: I ME - Mechanical Engineering, I EE - Electrical Engineering; Transport: T Ca - Cars, T Tr - Trucks, T OT - Other Transport; Metal Goods: - MG OMG: Other Metal Goods, MG Ap - Appliances, MG Pa - Packaging.

rate, sector split (forecast from 2018 onwards)) for all options. In the following, only Option A and Option C are discussed. Fig. 5 shows the sensitivity of the core results of the study (the surplus of low purity scrap Q3 & Q4) for the most influential parameters (lifetime, sector split). The sensitivity analysis indicates that the average lifetime of the different sectors has the highest impact (up to 9.3% for option A in 2015, 4% for option C in 2009, if the average lifetime is reduced by 20%) on the surplus of low purity scrap (see Fig. 5). This can be mainly attributed to the faster backflow of low purity scrap from the Construction sector. Furthermore, the steel demand in the Construction sectors is rather decisive for the surplus of low purity scrap. An increased demand in the Construction sector of 10% would reduce the scrap surplus by 2.1% and 4% (about 4 Mt/yr and 7.8 Mt/yr) in 2050, respectively, for Options A and C. In contrast, the export rate of ELV has little influence on the amount of surplus scrap. In the case of 20% less ELV exports, the surplus scrap quantities would only increase by less than 0.52% and 0.03% (about 1 Mt/yr and 0.06 Mt/yr) in 2050, respectively, for Options A and C.

3.6. Validation

The model was partially validated by comparing three of the model's stocks/flows (steel stock of cars, reinforcement bars in buildings, and available scrap) with independent data.

Steel stock in the Car sector (see Fig. 6, upper part): Bottom-up data of registered cars in the former EU-28 was compared with the steel stock modelled for the Car sector. Until 2010, the modelled stock figures are higher than the stock determined via bottom-up data. This apparent overestimation of the steel stock by the MFA model can be partly attributed to the fact that not all cars in the stock are registered (either due to interchangeable number plates or because of non-registered vehicles kept on private property).

Reinforcement bars in buildings (see Fig. 6, middle part): The modelled stock is below determined bottom-up stock, but still within the uncertainty range. The increase is similar in both datasets. The underestimation of the modelled stock can at least partly be explained by the use of other steel products (e.g., hot rolled bars or wire rod) as reinforcement in concrete.

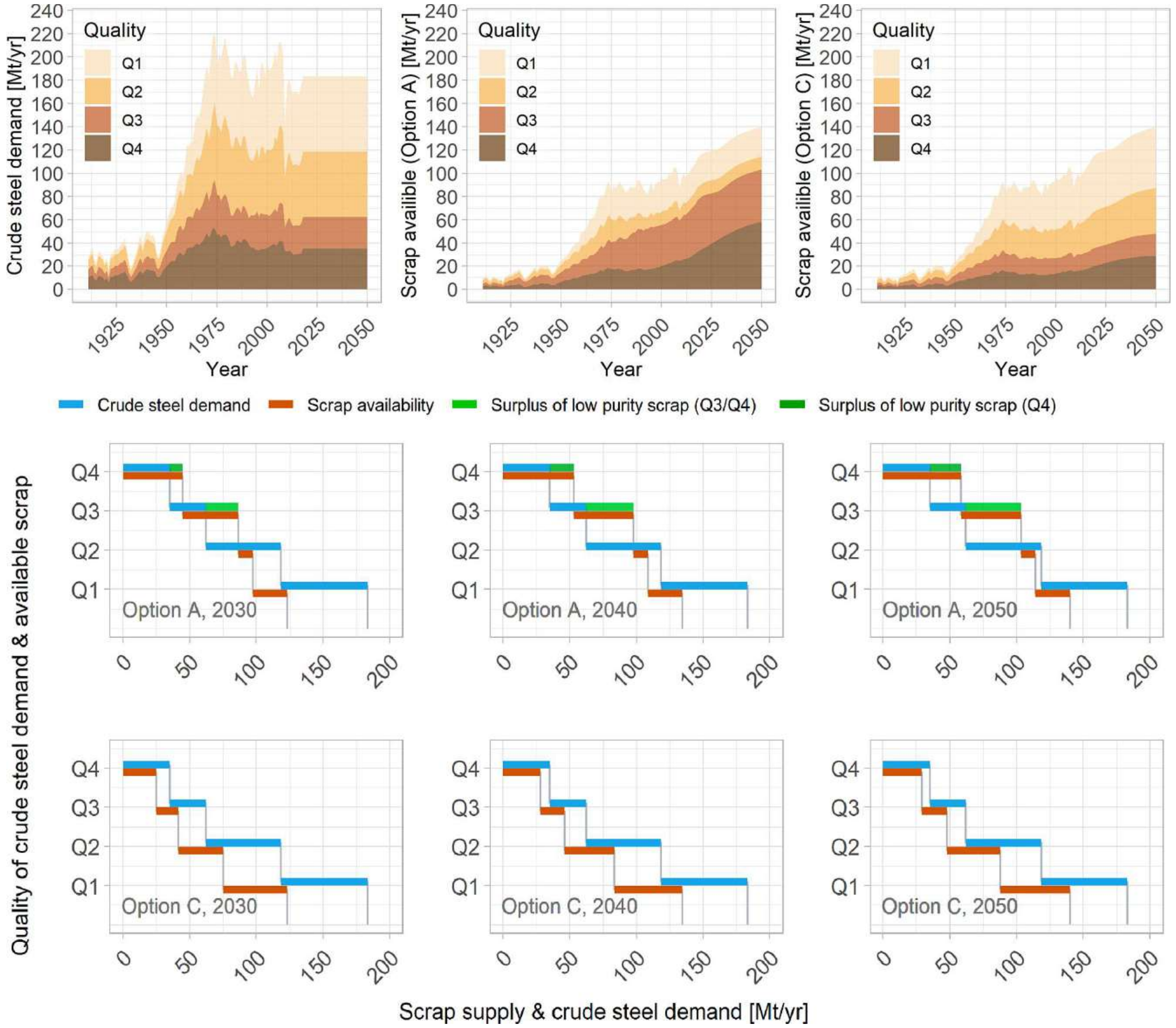

Fig. 4. upper part: Crude steel demand taking quality classes for crude steel demand into account (left), scrap available with quality classes based on Option A (middle) and Option C (right), considering Q1 to Q4, without Q0 (cast products); lower part: Material pinch analysis for the quantities and qualities (purities) of crude steel demand and available scrap in the former EU-28 for the years 2030, 2040, 2050.

Scrap availability (see Fig. 6, lower part): The increase of available scrap modelled is similar to that derived from statistics, but the model slightly underestimates the available scrap from 1950 to 1985, which can be at least partially attributed to the long average lifetimes of some steel products (up to 65 years). From 1985 to 2017, a good fit can be observed, which suggests that the model is fit for future assessment as propagated underestimates no longer influence the result.

To summarize, the approach of the various validation methods do not result in a perfect fit, but key parameters such as magnitude and the rate of change fit rather well with the model. In general, the outcomes of the comparisons verify the model approach and the data used.

4. Discussion

The top-down approach, employing the assumption of stagnant crude steel demand and constant sector splits and composition, is rather simple. It does not allow presumed saturation as well as economic and societal development to be accounted for. Still, in comparison with very sophisticated models, it performs quite well and reaches similar results in many respects. Additionally, it yields insights into future challenges regarding the quality of steel scrap and its handling:

In-use Stock: Stock assessment is not the focus of this study, but such evaluation allows - in comparison with much more sophisticated models on steel stocks (e.g., Hatayama et al. (2010); Müller et al. (2011;) Pauliuk et al. (2013b, 2013a)) - for plausibility checks. The in-use stock is dominated by steel in the Construction sector, whose share will increase even further to up to 80% (8.7 t/cap) in 2050, whereas the Metal Goods, Transportation and Industrial Equipment sectors will have similar magnitudes in 2050 (0.58 t/cap, 0.79 t/cap, 0.64 t/cap, respectively). The in-use stock will amount to about 10.7 t/cap in the former EU-28 in 2050. The stock values are below the mean results, but within the uncertainty margin for stock saturation determined in Pauliuk et al. (2013b) (Construction:10 ± 2 t/cap, Metal Goods: 0.6 ± 0.2 t/cap, Transportation: 1.5 ± 0.7 t/cap, Industrial Equipment 1.3 ± 0.3 t/cap). In comparison with other stock investigations in various regions in affluent economies (Böhmer et al., 2010; Müller et al., 2011), the results are in line with the results of total in-use stock. Furthermore, the bottom-up stock assessment for cars in use and for reinforcement bars in buildings as well as the top-down validation based on historic scrap output both validate the model results at least partially. The model also

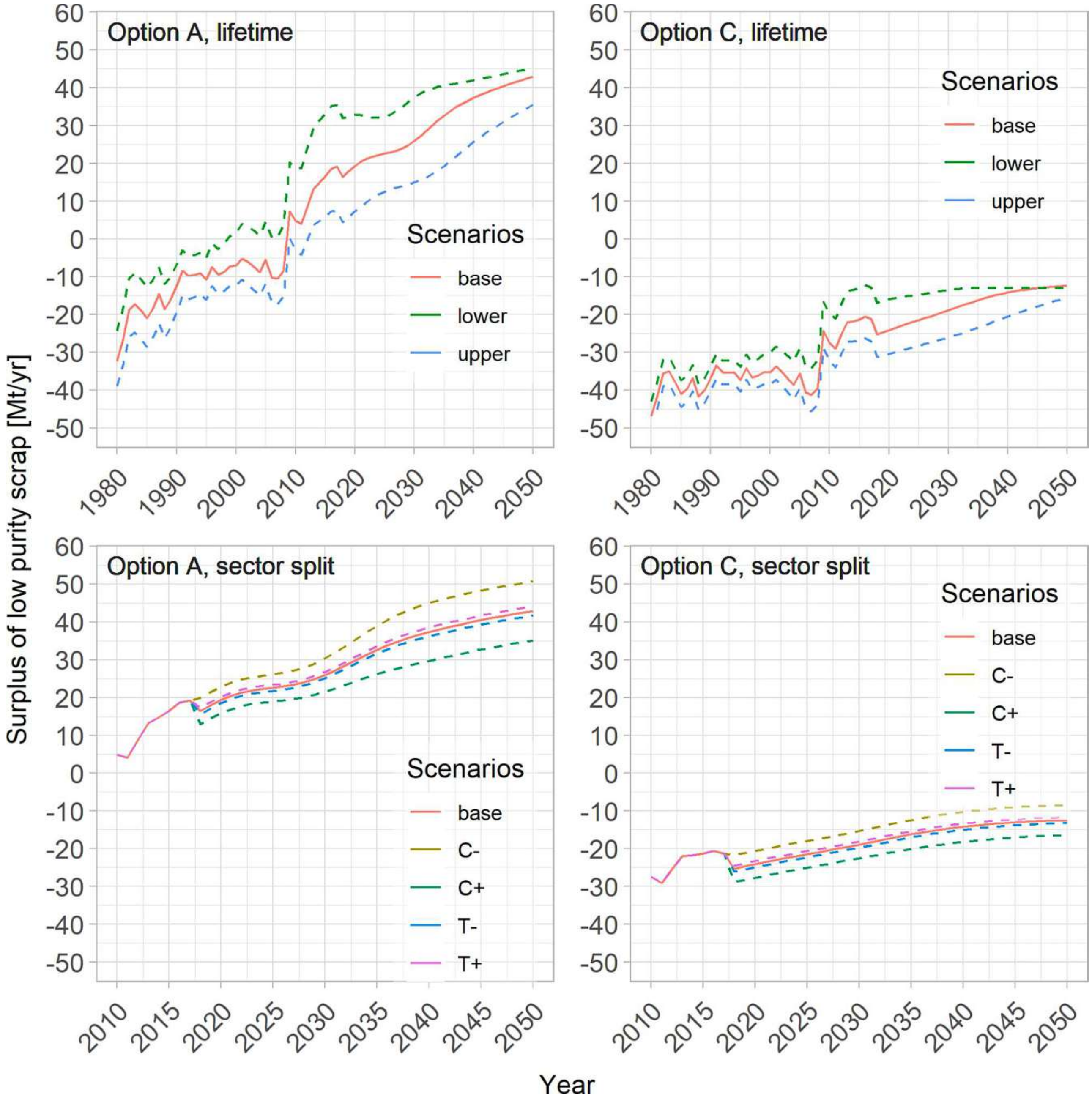

Fig. 5. Sensitivity for lifetime (±20%, upper part) and sector split (±10% for either Construction (C+/C-) or Transportation (*T*+/T-) sector, lower part) for Option A (purity of output based on sector of origin, right column) and Option C (purity of input = purity of output, left column). The reference value is the surplus of low purity scrap (Q3 & Q4, up to 0.25% of tramp elements considered) relative to the crude steel demand.

shows that for in-use stock to be saturated in the near future, crude steel demand (input into the system) and therefore steel use would have to decrease (as shown by Pauliuk et al. (2013a)). In our linear projection with constant input, the total stock (without taking population projections into account) will reach a plateau well after 2050, around 2075. That would mean a per capita stock of roughly 11 t/cap.

Scrap: The role of scrap is becoming more important. Dworak and Fellner (2021) showed with a static model that in the past (1945–2017) the composition of scrap shifted significantly from predominantly new scrap to predominantly old scrap (60% in 2017). The dynamic modelling approach comes to a similar conclusion and suggests that this trend will continue for several decades, even if less intensely (see Fig. 2). In 2050, the share of old scrap reaches close to 75%. If we assume that the efficiency in steel making and fabricating of final steel products (which means less new scrap) as well as the recovery rate (which was conservatively assumed to be low in this study) of steel from end-of-life products both increase, the old scrap ratio may be even higher. Currently, low purity scrap is mainly exported and not used in the former EU-28 (Dworak and Fellner, 2021). Similarly, scrap in the range of 10 to 20% of crude steel production is net exported from the US (according to the World Steel Association, 2017; Zhu et al., 2019). As global steel production volumes are still increasing, demand for scrap, regardless of the quality, is still high, especially in emerging economies. Therefore, the exported scrap was and is currently used to substitute primary raw materials abroad (as shown by Dworak and Fellner (2021)). But it can be expected that the demand for external low purity scrap will diminish since the emerging economies will also reach saturation at some point and generate their own post-consumer (low purity) scrap.

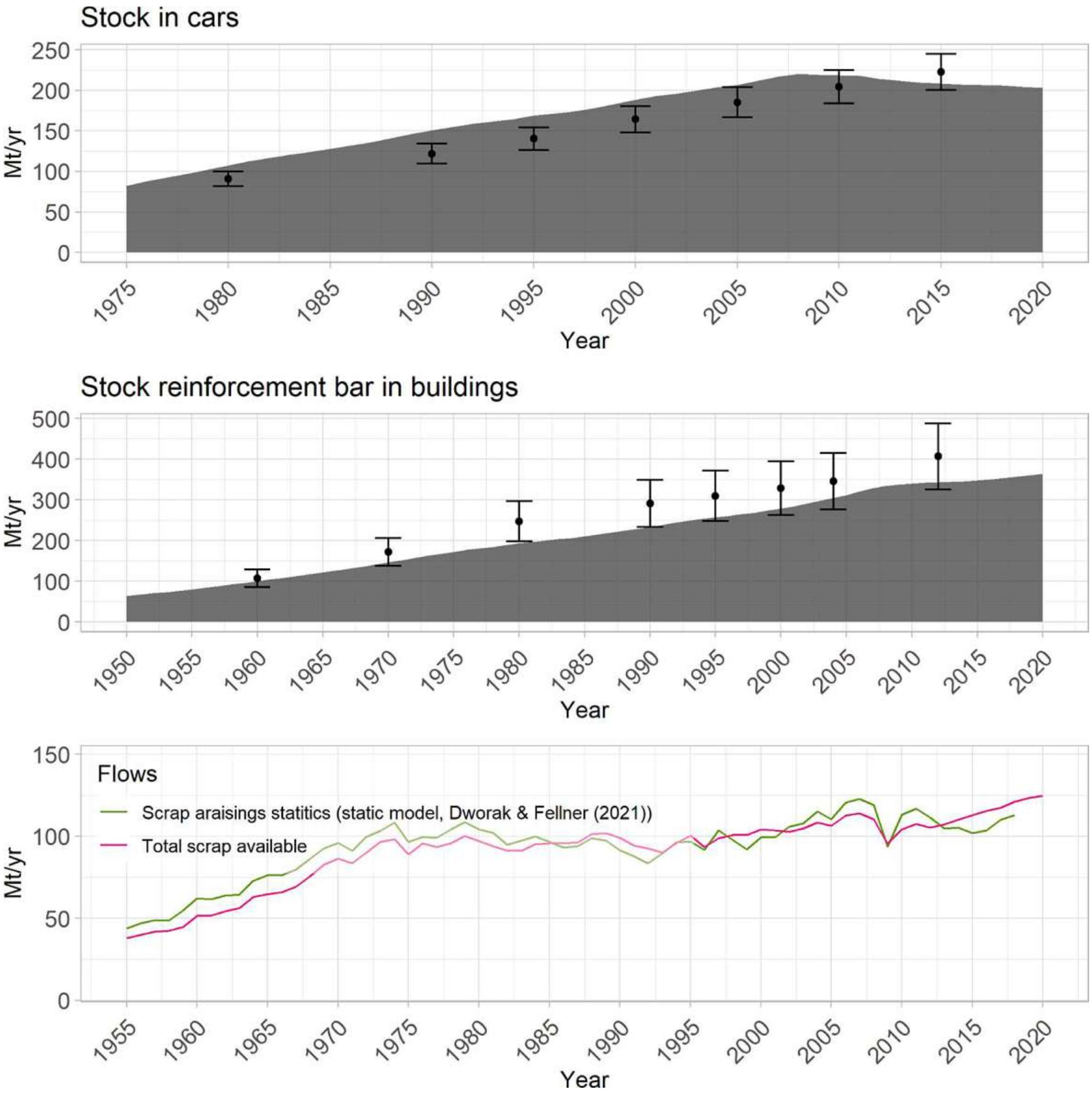

Fig. 6. Validation of model, upper part: validation of in-use stock in the Car sector, modelled stock compared with bottom-up data on cars in use; middle part: validation of in-use stock of reinforcement bars in the sector Buildings, modelled stock compared with data on housing space statistics; lower part: validation of modelled scrap available with top-down data based on scrap statistics.

Hence, the handling of steel scrap must undergo changes for it to be profitably employed in steel production. For one, by diluting available scrap with crude steel from primary sources, the surplus of low purity scrap could be balanced until 2040, potentially. Still, one of the main challenges remains the common lack of knowledge as regards the composition of single scrap batches. To potentially reach the goals of the circular economy package, alloy sorting is crucial (Daehn et al., 2019). The latter is also of importance to reduce the climate impact of the European steel industry. Utilizing the predicted 1000 Mio tons of surplus scrap (sum until 2050) domestically would reduce the EU's CO2 emissions by 560 to 2360 Mio tons (estimate based on (Damgaard et al., 2009)) and at the same time reduce the import of almost 1300 Mio tons of iron ore (estimate based on (Broadbent, 2016)). For end-of-life vehicles, alloy sorting was investigated by Ohno et al. (2015) and Willmann et al. (2017) and seems to have the potential to sort the available scrap sufficiently enough to close the steel cycle while simultaneously allowing present alloy elements to be preserved. Moreover, low purity scrap could be diluted with high purity resources (e.g., steel from primary sources, iron sponge) to exhaust the dilution potential. Proper alloy sorting would facilitate efforts in this direction as the composition of specific batches of scrap would come to be known better and could be applied accordingly. Further, several technical interventions could be considered to (i) reduce the tramp element content from the melt (e.g., sulphide slagging or vacuum distillation for Cu removal); (ii) improve processes for more tramp element tolerance (e.g., direct strip casting (Spitzer et al., 2003)) (iii) redesign materials for higher tramp element tolerance (e.g., counterbalance negative properties by adding interacting alloys (Daigo et al., 2021)).

For policymakers the results potentially imply that in order to foster a higher domestic circularity of steel, a tax on exports of scrap might need to be considered since valuable resources are more likely to be domestically recovered and alloy sorting technologies or technical innovations in the production process (reducing tramp element content in the melt) may only pay off with the help of such a measure.

5. Conclusion

We investigated the European steel cycle with a practicable dynamic MFA model to assess scrap availability until 2050 with regard to quantity and quality, and validated it partially. The top-down approach, with its assumption of stagnant crude steel demand and constant sector splits and composition, is straight forward and can be applied without taking many variables, such as presumed saturation or economic and societal developments, into account. Still, it offers robust results with the data available.

The share of post-consumer scrap is increasing in overall scrap composition (up to 75% of total scrap available). This scrap is mostly of low purity, especially if no countermeasures (e.g. alloy sorting) are put in place. Low purity scrap (tramp element content above 0.25%) is therefore gaining in importance as regards overall scrap composition (post-consumer, production & forming and fabrication scrap). The potential scrap rate (including old and new scrap) for crude steel production will reach more than 75% in 2050. Seen from a circularity point of view, this seems to be very good news. But if scrap continues to be handled as it usually is at the moment (e.g., little sorting, little dilution), the quality requirements for crude steel in the EU will not be able to be met and the European steel industry will also in future have to produce more than 45% of crude steel from primary sources to satisfy the qualitative (purity) requirements of crude steel, which is approximately the rate currently attained. This means that an increase in the scrap rate is not achievable under current practices, while simultaneously the amount of post-consumer-scrap is constantly increasing.

CRediT authorship contribution statement

Sabine Dworak: Conceptualization, Data curation, Investigation, Formal analysis, Visualization. **Helmut Rechberger:** Supervision, Writing – review & editing. **Johann Fellner:** Conceptualization, Supervision, Data curation, Writing – review & editing.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2021.106072.

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Full length article

Mercury throughput of the Austrian manufacturing industry – Discussion of data and data gaps

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ABSTRACT

Mercury (Hg) in the anthroposphere and its emission into the environment have been increasingly attracting attention. It is agreed upon that a substantial share of Hg is caused by anthropogenic activities. Comprehensive insight into Hg flows is crucial to minimizing exposure risk to humans and the environment.

This study is the first to put together a detailed, multiannual (2005 to 2016) Hg balance for the whole manufacturing industry of an industrialized country, namely Austria. It investigates data availability, quality, and the lack thereof by following a systematic approach. Assessed data is combined in a material flow analysis model to estimate and discuss the relevant Hg flows through inputs (raw materials) and outputs (products, wastes, emissions) of the individual industry sectors. Uncertainty ranges are estimated and processed according to error propagation.

Hg concentrations for most goods are available, but often of low quality (incomplete, partly non-representative). Data for quantities of goods are mostly available, especially for primary raw materials and production numbers. Nevertheless, publicly available data differs strongly from sector to sector and within sectors.

Over the period 2005 to 2016 the Hg throughput varied from 3.4 t/yr ±25% to 4 t/yr ±25%. Primary raw materials and fuels accounted for 70% to 75% of the Hg import, secondary ones for 25% to 30%. Hg export via products was estimated at 35% to 40%, via emissions to air at 20% to 25% and via waste at 40% to 45%.

1. Introduction

1.1. *The relevance of understanding the metabolism of anthropogenic mercury*

Mercury (Hg) is known to be harmful to humans and nature hence its use and handling are strictly regulated in most countries. The risks of Hg exposure have been investigated in peer reviewed literature (e.g. recently reviewed by Bjørklund et al. (2017)) and scientific reports (e.g. WHO, 2007), resulting in various legislation to restrict Hg use and regulate its occurrence (e.g. UNEP, 2017a). The Minamata treaty (UNEP, 2017a) is the basis and harmonized outcome of many of these internationally implemented regulations to monitor and restrict Hg use. In accordance with such regulations, Hg flows due to products containing Hg will be mostly phased out, except for dental amalgam. Moreover, the use of Hg in specific production processes (chlor-alkali production (2025); acetaldehyde production, in which Hg or Hg compounds are used as a catalyst (2018)) is no longer allowed in all countries which ratified the treaty. Other production processes that use Hg (for vinyl chloride monomer production, sodium, potassium methylate and ethylate production, as well as for the production of polyurethane using catalysts containing Hg) are to be highly regulated or restricted. These developments aim to reduce Hg flows of a larger magnitude to “minimize the potential exposure of human health and the environment from anthropogenic emissions and releases of Hg and Hg compounds” (UNEP, 2017a). Furthermore, anthropogenic emissions of Hg into the air in North America and Europe have decreased significantly over the last few decades (Guerreiro et al., 2017; Zhang et al., 2016). Nevertheless, various indicators, such as Hg content in the biota, are not significantly decreasing and still exceed national thresholds (e.g. fish in the Austrian biota (Uhl et al., 2010)), indicating that perhaps not all sources of Hg emissions are known and under control. Estimates of Hg flows are subject to considerable uncertainties as Hg is mainly present as a trace element and therefore its content is not always easily quantifiable. For example, Hg quantification in water requires its own analytical method capable of measuring minimal levels of content, in

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contrast to possible bulk testing of other substances such as e.g. applied in Zoboli et al. (2019). Tracking Hg throughout the natural and anthropogenic system can contribute to a better understanding of paths and fates of Hg. Direct anthropogenic emissions contribute with 30% significantly to the overall emission of Hg into the atmosphere (UNEP, 2019). In addition to direct anthropogenic emissions, mainly environmental processes (much of which involves recycling from formerly disposed of anthropogenic Hg) and natural (geogenic) emissions contribute to the overall Hg emissions (UNEP, 2019). UNEP concludes that understanding and influencing the anthropogenic system is playing a key role in tackling challenges arising from Hg circulation and exposure.

1.2. *Mercury balances for industrial systems and industry sectors*

Traces of Hg can be found in almost every natural material (Adriano, 2001). Therefore, industry sectors which produce high volume products and thus have a high material input are prone to a higher amount of Hg throughput. The application of high temperatures in the production processes leads to potential Hg releases into the environment if no appropriate measures are taken. This is reflected in the Minamata Convention (UNEP, 2017a), which lists the following sectors and processes as being potential sources of Hg emissions: coal-fired power plants, coal-fired industrial boilers, smelting and roasting processes used in the production of non-ferrous metals, waste incineration facilities and cement clinker production facilities. Thus, the industrial system is of particular interest regarding Hg throughput for the following reasons: First, the throughput of material is proportionally high, which potentially results in significant Hg flows, as mentioned above. Second, due to substantial transformations of materials (mechanical, chemical, thermal) during industrial processing, the physical composition of raw materials (input) usually differs from that of the corresponding products (output). Hence the Hg concentration may vary considerably between input material and output material (products and by-products, such as waste and emissions). Third, the highly controlled processes, and consequently favourable data situation (data existence and possible availability), provide the opportunity to closely investigate the fate of the Hg. Last but not least, this sector and these processes might constitute an optimal place from which to remove Hg from the system, as has already been done in many cases (e.g. Hg remediation in combustion and industrial plants by means of flue gas cleaning). In the field of Hg, sophisticated models are available to simulate Hg circulation in the environment or parts thereof (e.g. Amos et al., 2015; Gustin et al., 2015; Horowitz et al., 2017; Obrist et al., 2018; Zhu et al., 2016). Detailed industry wide Hg flow analyses are a feasible way of understanding the sources, paths and fate of Hg, which in turn facilitate the application of effective measures to influence the system in accordance with desired outcomes. Material Flow Analysis (MFA) has been used on many occasions to investigate and gain deeper understanding of the flows of goods and substances (e.g. waste management (Allesch and Brunner, 2015), plastics (Van Eygen et al., 2018), pollutants (DEPA, 2004a, 2004b; Zoboli et al., 2015) and Hg (Jasinski, 1995; Krook et al., 2004)). The majority of studies on anthropogenic Hg flows focus on the overall Hg emissions into the environment, either on industrial, national or global levels (e.g. Amos et al., 2013; CEIP, 2020; Fukuda et al., 2011; Glodek et al., 2010; Pacyna et al., 2010; UNEP, 2019, UNEP, 2010, UNEP, 2008; Won and Lee, 2012; Wu et al., 2006, 2017; Xu et al., 2017). Until recently, most studies have focused on static, one-year analysis (e.g. for Poland (Panasiuk and Głodek, 2013) or China (Hui et al., 2017)). UNEP has been making efforts to assess and compare Hg releases with high resolution (categorized in industrial sectors and countries) into the environment periodically with the Global Mercury Assessment Report (UNEP, 2019, 2013a, UNEP, 2008; 2003). Furthermore, Hg balances have been conducted for single industry sectors or branches, with different goals and varying levels of detail, e.g. for cement (e.g. for Austria (Lederer et al., 2017) and Germany (Achternbosch et al., 2005, 2003; Harraß et al., 2018; MUNLV, 2005)), metals (e.g. for zinc (Chung et al., 2017) and iron (Fukuda et al., 2011; Wang et al., 2016)) and coal combustion (e.g. globally (Mukherjee et al., 2008)). Some studies examined the industrial system as a whole as part of wider national studies without, however, investigating the specific details of individual industrial sectors (e.g. for Denmark (Christensen et al., 2003), Turkey (Civancik and Yetis, 2018) and Austria (Reisinger et al., 2009b)). A paucity of dynamic or multiyear models can be identified.

1.3. *Data availability*

Many data points are necessary for effective modelling, especially for the construction of dynamic or multiyear models. Monitoring systems, which are usually in place in areas assumed to be especially sensitive, can supply a solid and constant source for Hg flow data (e.g. emissions into the atmosphere (CEIP, 2020)). The MinFuture framework (minfuture.eu), a recent EU-funded project to improve the monitoring of the physical economy, proposes a systematic approach to designing MFA studies. Therefore, a hierarchical structure was developed which causes the system itself to serve as the foundation of any MFA. Data should then be collected according to the system design. Thereby data should be put into a system context, which allows consistency to be checked (Petavratzi et al., 2018). Economy-wide material accounting (eurostat, 2020a) and industry reports with sector-wide material accounting (e.g. Austropapier, 2018; Mauschitz, 2018) improved data availability significantly. However, the approach proposed is not established yet, so for now the model was designed to fit the data available. Most data are kept confidential, especially regarding the composition of flows and the content of specific elements or compounds. Already Hansen's (2002) analysis indicated that, MFA studies often rely heavily on data on material flows from companies, which are kept confidential, and therefore confidentiality is one of the main challenges faced when putting together an accurate and feasible MFA.

The present study aims to investigate, map, and understand the Hg throughput in the industrial sector and its evolution over more than a decade in Austria. Challenges regarding data availability, reliability and usability were investigated and evaluated. The pathways and fate of the Hg introduced into the system were investigated based on the available data. It involved quantifying and visualizing the relevant flows, their import into the system, their paths through the system and their fate/export beyond the system boundaries. This study is the first to put together a detailed multiannual Hg balance for the whole manufacturing industry of an industrialized country.

2. Materials and methods

2.1. *Material flow analysis*

Material Flow Analysis (MFA) as described by Brunner and Rechberger (2016) was used in the present study to map Hg flows throughout the Austrian industry. MFA balances flow on the basis of the law of mass conservation. It is applied to goods (e.g. raw materials, fuels, wastes) and the substances contained therein (e.g. chemical compounds or elements, such as Hg). Temporal and spatial boundaries for the system are set and relevant flows and processes are identified. Data is usually derived from the literature (peer reviewed literature, reports), stakeholders (e.g. plant owners) or own measurements. In addition to the most probable value of a flow, the relative standard uncertainty can be considered. Usually normal distribution is assumed. Error propagation is based on Gauss's Law of error propagation, as proposed in Brunner and Rechberger (2016).

2.2. *Model design and data*

The model developed can be seen in Fig. 1.

Materials: The study covers the trace element and heavy metal Hg.

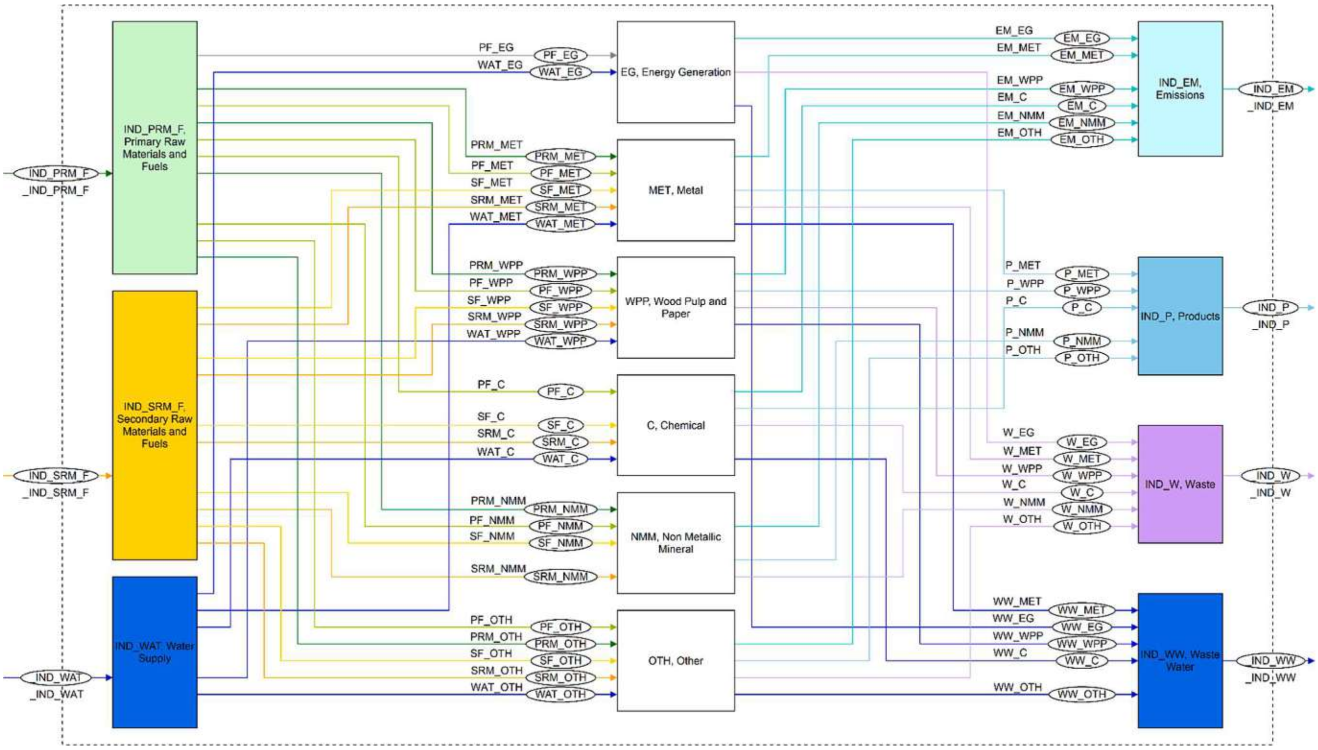

Fig. 1.. Hg throughput for the Austrian manufacturing industries: process and flow names (abbreviations: IND (summed up import and export flows); import main flow types: PRM (primary raw materials), PF (primary fuels), SRM (secondary raw materials), SF (secondary fuels), WAT (water supply); export main flow types: EM (emissions), P (products), W (wastes), WW (wastewater).

Good flows (mainly economic accounting) relevant to Hg throughput were considered, whereas Hg content in equipment, machinery and infrastructure was not considered. The goods were summarized in three input categories (Primary Raw Materials and Fuels (PRM_F), Secondary Raw Materials and Fuels (SRM_F), and Water Supply (WAT, water abstraction in form of ground and surface water)). PRM_F and SRM_F were further subcategorized into materials (Primary Raw Materials (PRM), Secondary Raw Materials (SRM, inter alia recovered glass and paper, various slags, scrap)) and Fuels (Primary Fuels (PF), Secondary Fuels (SF, inter alia plastic waste, used tires, solvents, waste oil and various residues)). The output flows were categorized according to Emissions into air (E), Products (P), solid Wastes (W) and Wastewater (WW). For details on flow composition and materials considered, the reader is referred to the supplementary information (see tab “data_goods”).

Processes: Processes were grouped into two collector blocks (Imports and Exports) and a transformation block (industrial sectors). The imports (according to the above-mentioned materials, Primary Raw Materials and Fuels (IND_PRM_F), Secondary Raw Materials and Fuels (IND_SRM_F), Water Supply (IND_WAT)) transport the raw materials into the system, which are transformed into products, by-products and wastes in the industrial sectors (Energy Generation (EG), Chemical Industry (C), Metal Industry inclusive coke production (MET), Non-Metal Minerals (NMM), Wood Pulp and Paper (WPP), Other Industries (OTH). The industrial sectors were chosen in accordance with the NRF (Norm Reporting Format for air pollutant emission inventory (Ntziachristos et al., 2014)) and NACE (European Commission, 2008) reporting categories. This categorisation focuses on the economic structure and economically relevant material flows, but not on the tracking and understanding of trace elements or like Hg. Therefore, it might not be ideal for the tracing of Hg through the industrial system. Nevertheless, this approach was chosen as the most complete dataset (reporting data) and is structured accordingly. As Austria is a rather small country, the international trade of raw material (primary and secondary) plays a significant role. Therefore, the direct intercorrelation between the domestic sectors was represented only indirectly via the export of wastes out of the system and the import of secondary raw material into the system. The second collector block (Emissions (EM), Products (P), Wastes (W), Wastewater (WW)) adds these outputs and transports them as accumulated flows beyond the system boundaries and hence out of the system investigated. EG solely consists of energy generation plants, without waste incineration plants. Although waste incineration and waste treatment plants are known to be a significant source of Hg emissions to air, this cannot be stated for Austria. Hg emissions from this sector only amount to around 2% of the overall Hg emissions to air (Anderl et al., 2017; CEIP, 2020). This is due to the advanced Air Pollution Control devices installed at these facilities. Therefore, and as they are not part of the manufacturing industry, waste incineration plants were not considered in this study. C refers to the oil refinery. Other parts of the chemical industry (e.g. plastic industry, pharmaceutical industry) were only considered regarding fuel consumption. Sufficient data was not available for raw materials and products. MET includes ferrous, primary non-ferrous (tungsten) industry and coke production. Other non-ferrous metal production was not considered as only secondary production is conducted in Austria. This was assumed to be not relevant in terms of Hg flows. NMM consists of the cement industry, the brick and tile industry and the glass industry, whereas WPP summarises the pulp and paper industry and the wood industry (timber and board production). Besides this, OTH takes other industries, such as textiles and the food industry, into account. In this process, mainly the consumed fuels were considered.

Spatial and Temporal Scale and Extent: The Austrian manufacturing industry was investigated. The spatial boundaries are the Austrian state borders. The model extends over the years 2005–2016.

Basic principles: A retrospective, bottom-up model was put together. The available data on flows of goods was applied with the respective Hg concentrations for fraction flows and subsequently aggregated to main flows. By way of example, the main flow of secondary fuels for the sector non-metal minerals consists, amongst others, of fly ash, used foundry sand and blast furnace slag. A corresponding Hg concentration was assigned to each of these mass flows (fraction flows). These fraction flows were then aggregated to main flows. For this purpose, the mass flows were summed up and the Hg concentrations were weighted with regard to their corresponding mass flow according to the following equation:

$$\mathrm{c_j} = \frac{\sum_{i=1}^{k} c_i \dot{m}_i}{\sum_{i=1}^{k} \dot{m}_i} \tag{1}$$

with c_j as the weighted concentration of the main flow j, c_i as the specific concentration corresponding to the specific mass flow $\dot{m}_i$ of the fraction flow i.

Static or dynamic modelling approaches: The modelling approach was a static, multiyear one (as applied by Zoboli et al., (2015)), which balances 13 time nodes separately, resulting in 13 yearly snapshots. It was assumed that the system is in a steady state and no accumulation or depletion of stocks occurs.

Dissipation: Dissipation was considered and quantified via emissions into air and water. Reported data was used for quantifying the emissions into air. Emissions into water were estimated based on the wastewater generation and concentrations measured in effluents (discharges into water bodies after treatment) from specific industries. Specific effluent types were not considered as no detail data is available. Dissipation via transport, handling and transformation, which is not officially reported, was not considered.

Uncertainty, concentrations: For trace metals such as Hg, skewed or composed distributions might be more accurate, as indicated for various trace metals in e.g. various studies (Hernández-Crespo and Martín, 2015; Martyniuk et al., 2020; Tobías et al., 1997; Yan et al., 2020), and for Hg specifically e.g. in Hylander and Herbert (2008), Wu et al., (2016), and Zhang et al., (2012). Nevertheless, in the model proposed it was decided to assume normal distribution for further calculations and error propagation, such as applied in various publications (e.g. Lederer et al., 2017; Reisinger et al., 2009a). The reasons for this were as follows: First, not enough data on Hg concentration distribution was available in enough detail for all the mass flows applied. Second, for reasons of simplicity as this is the first study which aimed to put together a timeline with detailed Hg flows of the industrial system of a country. Third, even though the normal distribution may have an impact on the absolute Hg throughput, it has a minor impact on the distribution of Hg to the individual outputs, which is essential to understand the Hg throughput in the industrial sector. Fourth, the focus of the present study lies not solely on the model, but on the assessment of the available data and its applicability. Moreover, e.g. Marrugo-Negrete et al., (2008) indicates, that skewed distribution does not have to be the case for Hg concentrations in materials. To quantify the concentrations, the literature data (see further under Model input data) was used. Where possible, multiple datapoints were combined to estimate a range for the concentration in a specific good. Depending on availability, the following statistical parameters were regarded as mean-value estimates in order of priority: median, arithmetic mean, published value without further statistical description. Outliers were identified based on the distribution and reliability (considering description, source, sample size) of the values. The outliers identified were not considered in further calculations. Due to the diverse quality (available information regarding e.g. statistical parameters, number of specimens for statistical estimates, number of independent data sources), identifying outliers was conducted visually from the distribution of all available values. The standard deviation was set to the minimum to maximum of these mean-value estimates (as mentioned, preferably the median of each individual dataset, followed by the arithmetic mean and the published value without further statistical description) for the concentration of each fraction flow, thus the considered mean of the assumed normal distribution was set to the arithmetic mean of this range of mean-value estimates.

Uncertainty, goods: Data for the flows of goods were mainly obtained from official reporting (statistics (economic accounting) and/or industrial reports). As the uncertainty is usually not quantified, the uncertainty assessment was conducted according to the proposals of Laner et al., (2016, 2014), with 4% for officially reported data from statistical offices, as applied in Zoboli et al., (2015).

Error propagation: Error propagation as described in Brunner and Rechberger (2016) was applied. Error propagation for main flows was performed within the MFA software STAN, as elaborated in Cencic (2016). Data preparation for STAN-input involved the calculation of the weighted concentration and corresponding uncertainty to calculate the uncertainty of goods and concentrations from detailed level (fraction flows) to analysing level (main flows). Therefore, the weighted relative standard uncertainty was derived from Gauss's Law of error propagation to calculate it for the corresponding main flow.

$$S_{c_j}{}^2 = \sum_{i=1}^{k} S_{\dot{m}_i}{}^2 \left(\mathrm{c}_i \left(\frac{1}{\sum_{i=1}^{k} \dot{m}_i \sum} - \frac{\sum \dot{m}_i c_i}{\left(\sum_{i=1}^{k} \dot{m}_i \right)^2} \right)^2 \right) + \sum_{i=1}^{k} \mathrm{S}_{c_i}{}^2 \left(\frac{\dot{m}_i}{\sum_{i=1}^{k} \dot{m}_i} \right)^2 \tag{4}$$

S_{c_j} is the weighted relative standard uncertainty of the concentration c_j of the main flow j. $S_{\dot{m}_i}$ is the weighted relative standard uncertainty of the mass flow $\dot{m}_i$ of the flow i.

Model Input Data: The input data consists of flows for goods and concentrations for the Hg content. As a single exception, the Hg emissions into air were employed as total Hg mass flows as officially reported (European Monitoring and Evaluation Programme by (CEIP, 2020)). Data for the flows of goods was mainly taken from official statistical databases (Eurostat and Statistik Austria) or reporting from industrial sectors (e.g. Austropapier, 2018; Fachverband der Glasindustrie, 2019; Mauschitz, 2018; Pfeiler and Gradischnig, 2019; World Steel Association, 2017) if available. Data for Hg concentrations was, as mentioned above, put together from various data sources, but mainly from scientific surveys, which were either published as scientific reports (e.g. Achternbosch et al., 2003; AGES, 2016; Christensen et al., 2003; Deutsch et al., 2012; Harraß et al., 2018; MUNLV, 2005; Szednyj and Schindler, 2004; Taverna et al., 2010; UNEP, 2017b; Wilhelm, 2001) or peer reviewed literature from various fields (e.g. Ali and Al-Qahtani, 2012; Bai et al., 2017; Diao et al., 2018; Fukuda et al., 2011; Wang et al., 2016; Wu et al., 2017; Yang et al., 2018). Furthermore, where applicable, the ABANDA-database (German database for waste characteristics (LANUV, 2019)) was considered. Detailed documentation regarding applied data can be found in the supplementary information (SI).

Model Output Data: On the one hand, all good flows considered were linked with the corresponding Hg concentrations. Hence, the consequential mercury flows and the corresponding relative standard uncertainty were determined. On the other hand, the Hg output flows via solid waste were determined by balancing the respective industry sector on the basis of the available data. This approach was chosen since the estimate of Hg flows via solid wastes on the basis of reported data was not feasible due to confidentiality constraints with respect to reported waste flows (some reported flows were only publicly available in a highly aggregated form, while others were confidential altogether).

Evaluation: The system was calculated and visualized via the MFA software STAN (Cencic and Rechberger, 2008). A sensitivity analysis was applied for each process to assess and document the most influential Hg concentration applied. The temporal development of all main flows was investigated and visualized with the corresponding relative standard uncertainty. Reported quantities of wastes were used to validate the model's – or rather the data – consistency.

3. Results

3.1. Data availability

A short overview of the data availability assessment can be seen in Table 1. In summary, better harmonization, as proposed by Petavratzi et al., (2018), and access to data on a detailed level would enable much more robust modelling. In what follows, all categories and industrial sectors are described in more detail.

3.1.1. Goods

Accumulated flows of goods into the industry are statistically well documented for most industrial sectors. Statistical data, supplied mainly on a highly aggregated level, is machine-readable, which enables updates and analysis of material flows for multiple time periods. Detailed volumes for specific industries or materials are sparsely available. The data processing is demanding and labour intensive as most reports are structured differently and mostly not available in machine-readable format.

3.1.1.1. Imports into industrial system. Fuels: The fuels used were derived from the Austrian energy balance (Statistik Austria, 2019). The data set is publicly available in different formats, i.e., as a spreadsheet and consistent over years. Data on fuel consumption is therefore available for all sectors in similar quality. Where available, more detailed data from industrial reports was used (e.g. from the cement industry (Mauschitz, 2018) or the pulp and paper industry (Austropapier, 2018)). Reported incineration of municipal solid waste and sludge were not considered as the waste management is not part of the present study.

Primary raw material: Data for the material input were taken from official statistics (domestic material consumption, and material flow accounting (eurostat, 2020a)) where no more detailed data from industrial reports are available. Data from industrial reports mostly coincide with data from the material accounting data set. Therefore, a

Table 1.
Data availability assessment by sectors (C: chemical industry, EG: energy generation, MET: metal industry, NMM: non-metallic minerals, OTH: other industries, WPP: wood, pulp and paper) and main flow types (PRM: primary raw materials, PF: primary fuels, SRM: secondary raw materials, SF: secondary fuels, WAT: water supply, WW: wastewater, P: products, EM: emissions).

Sector	Flow	Import Export	Goods			Concentration			Relevance
			Rating	Source	Comment	Rating	Source	Comment	
all	EM	EX	+	3	EMEP, industry reports are available in similar detail			not applicable	I
all	WW	EX	~	3,4	reporting biannually, pulp & paper: detailed yearly reporting in industry report	~	5		I
C	P	EX	–	1	harmonized and aggregated	–	5,6		?
MET	P	EX	+	1	coke industry is counted as part of the metal industry	~	5,6	coke industry is counted as part of the metal industry; assumption: no Hg in finished metal products	III
NMM	P	EX	++	4	detailed reporting and publishing of production volumes for pulp & paper				III
WPP	P	EX	++	4	detailed reporting and publishing of production volumes for cement	+	5,6		III
OTH	P	EX	–	2	domestic material consumption	–	5,6	due to highly aggregated and missing flows, little specific data can be applied	?
all	PF	IM	+	1,4	harmonized and aggregated (statistical accounting), industry report for cement and pulp and paper	+	5,6		I
all	SF	IM	~	1,4	detailed reports for cement and pulp & paper	+	5,6,7,8	high variation, max for all Hg concentration based on allowed Hg concentration in cement industry	I
all	WAT	IM	~	3,4	reported biannually until 2010; pulp & paper: detailed yearly reporting in industry report	~	5		III
C	PRM	IM	–	1	crude oil statistical accounted in fuels; no further information on primary raw materials	+	5,6		?
C&OTH	SRM	IM	–		no information available			not applicable	?
MET	PRM	IM	–	2	according to domestic material consumption (ores)	~	5,6		II
MET	SRM	IM	~	4	steel scrap input reported since 2010; no data on non-ferrous metal scrap	~	5	little data available	II
NMM	PRM	IM	+	2,4	for cement inputs are available; for other sectors calculation on basis of production margins was applied	+	5,6		I
NMM&WPP	SRM	IM	+	4	detailed reporting and publishing of secondary raw material used for cement and pulp & paper	~	5,6,7,8	high variation, max for all Hg concentration based on allowed Hg concentration in cement industry	I
OTH	PRM	IM	–	2	rough estimate as material accounting is not explicitly related to industrial sector	–	5,6,7	due to highly aggregated and missing flows, little specific data can be applied	?
WPP	PRM	IM	+	2,4	detailed reporting, auxiliary materials and additives reported for pulp & paper, aggregated	–	5,6,7		III

rating: ++: detailed data available, +: aggregated data available, ~: fragmented data available, -: no specific data available, –: no data available.
sources: 1: statistical accounting for fuels, 2: material accounting, 3: officially reported, 4: industry reports, 5: scientific reports, 6: peer-reviewed literature, 7: legislation, 8: waste characteristics data base.
relevance: I: high relevance, continuous monitoring recommended, II: moderate relevance, periodic checking recommended, III: low relevance, occasional checking recommended, ?: not enough data to assess relevance.

rather solid database is available, but the available level of detail varies significantly between different sectors.

Secondary raw material: Published records of secondary raw materials are diverse. Some industries report in detail (the cement industry (Mauschitz, 2018) and the pulp and paper industry (Austropapier, 2018)), while others do not publish any data regarding secondary raw materials. Hence the database regarding secondary raw materials is not satisfactory.

Water supply: Data on water use was taken from statistical reporting. The data is highly aggregated and is reported biannually until 2010. For subsequent years, the wastewater volumes were applied for water abstraction as well. As the water supply is not a significant import of Hg into the system, this assumption was deemed acceptable.

3.1.1.2. Exports out of industrial system. Products: Production margins are reported in detail. Aggregated production volumes are publicly available for most industries on some level. Detailed data is publicly available mainly in industrial reports. Confidentiality restrictions of production volumes are in place for many sectors, preventing the use of machine-readable data from the industrial production survey PRODCOM (eurostat, 2020b).

Emissions: Emissions into air are reported and published according to Ntziachristos (Ntziachristos et al., 2014) for all sectors investigated. The dataset is available in detail and machine-readable (CEIP, 2020). Similar and higher emission rates are estimated in the Global Mercury Assessments Report (UNEP, 2013b, 2003; UNEP and AMAP, 2019).

Wastewater: Wastewater generation is reported biannually (eurostat, 2020c) and was applied accordingly.

Waste: Estimated waste flows were the targeted outcome of the model. For some sectors, environmental reporting was conducted in a more detailed manner (the paper industry, e.g. (Austropapier, 2018)). Furthermore, data on reported waste generated is available in a machine-readable format (eurostat, 2020d). The available statistical data are very coarse and not consistent. Inconsistency seems to result from changing of i) reporting modes, ii) availability and iii) confidentiality. Hence, these data are not applicable for a consistent balance or for robust validation.

3.1.1.3. Sectors. Non-metal minerals: The Austrian cement industry publishes its material input, production volumes and emissions in detail yearly. The report includes a detailed accounting for the primary and secondary fuels and raw materials used. Overall, raw meal input is reported as well. Primary raw materials (such as limestone, silica sand, clay or iron ore, as is done e.g. for Germany (VDZ, 2018)) are not reported in detail. However, the reports on the material flows of the Austrian cement industry are one of the most detailed industrial reports available. The glass industry reports on production numbers but only provides approximate data on the share of secondary materials (recovered glass, e.g. Fachverband der Glasindustrie, 2017). The brick and tile industry reports on their production (Pfeiler and Gradischnig, 2019) without further information on raw materials and fuels. None of the data is publicly available in machine-readable form, but the reports of the cement industry are well structured and can therefore be transformed into a suitable format with comparably little effort.

Wood pulp and paper: The Austrian pulp and paper industry publishes a detailed report on material in- and outputs (e.g. Austropapier, 2018), and products volumes are available in machine-readable form in eurostat (eurostat, 2020e). In addition to the materials used, additives and auxiliary agents are also reported on in some detail. Environmental reports also state volumes of waste and wastewater generation in detail. The wood industry (wood and wood-based materials) reports timber harvesting and board production, which is also available in similar detail and machine-readable in statistical reporting (eurostat, 2020f).

Metal industry: Production volumes are well documented and published for the ferrous metal industry by the European and global steel associations (eurofer and worldsteel). There is no machine-readable form available, but the reports are well structured and can be transformed into a suitable format with comparably little effort. Ferrous metal scrap usage has been documented since 2008 (BIR, 2018). Fuels are not documented in specific industrial reports. Raw material usage is not publicly available. In Austria, primary production of tungsten represents the main primary non-ferrous metal production. Data is available for the tungsten mining industry. Otherwise, data on the use of non-ferrous metal ores is contradictory (e.g. copper (BMWFW, 2017; eurostat, 2020a)). As the primary production of most of the non-ferrous metal ores are not specifically declared in eurostat (eurostat, 2020a), only the domestic consumption of highly aggregated categories of other non-ferrous ores (eurostat, 2020a) was applied to at least cover tungsten production. Raw material input for secondary non-ferrous metal production was largely disregarded as only primary production is considered relevant with respect to Hg. Fuel consumption was considered for all metal production.

Chemical industry: All flows into the chemical industry were derived from the Austrian energy balance (Statistik Austria, 2019). Therefore, only the oil refinery was assessed. Production volumes of the refinery can be derived from the energy balance. Fuel consumption of the remaining chemical industry was considered as it can be derived for the whole sector from the energy balance.

Energy generation: All material inputs into the energy generation sector were derived from the Austrian energy balance. As mentioned above, no waste incineration plants were considered.

3.1.2. Hg concentrations

Values of Hg concentrations vary highly with relative standard uncertainty up to 100% (see Fig. 2). Mainly two reasons may be given: (i) The Hg content is highly dependant on the specific raw material and the location of the natural mineral deposit. Different sources report high variations in Hg content in coal due to e.g. coal-types, coal-forming periods and coal-accumulating areas (Bai et al., 2017; Lassen and Hansen, 2000; Mukherjee et al., 2008; Pirrone et al., 2010; Toole-O'Neil et al., 1999). (ii) The laboratory analysis of Hg is challenging. The risk of cross contamination must be considered due to the low concentrations in materials (trace elements). Furthermore, Hg's volatility (Holleman et al., 2007) demands especially careful handling of samples (e.g. low drying temperatures, no heat production during sample processing). These circumstances sometimes lead to a paradox situation: the more data is available, the higher is the potential uncertainty.

Missing specifications of the sample condition (references to dry mater, solid matter, and overall sample) is another reason for uncertainty. This lack of definition does not have a great influence on most minerals and fossil fuels as their moister content is mostly insignificant in relation to the natural uncertainty of Hg content. For organic materials such as wood or even lignite, the contrary may be the case as the reference to the sample condition (particularly moister content) can be quite influential. In this study, the relative standard uncertainty of the Hg concentrations derived range between 10% (new paper and paper board) and 100% (e.g. bricks and tiles, blast furnace slag or used tires). Table 2 shows the maximum and minimum aggregated Hg concentration of input and output main flows with corresponding standard normal uncertainty. Each value shown corresponds to the weighted mean concentration applied and the corresponding standard normal uncertainty for the main flow of one specific year. The variation is based on the varying composition of fraction flows and therefore varying weighted Hg concentrations.

Primary raw materials: Data on the Hg-content is available for most minerals. The variations in the Hg-content are high, with most of them ranging between one and two, with some even up to three orders of magnitude. Generally, little data is available for the tungsten industry in Austria. Primary tungsten production is the only primary non-ferrous metal production. There is no data available on Hg in tungsten ores despite it being deemed potentially relevant by UNEP (UNEP, 2019).

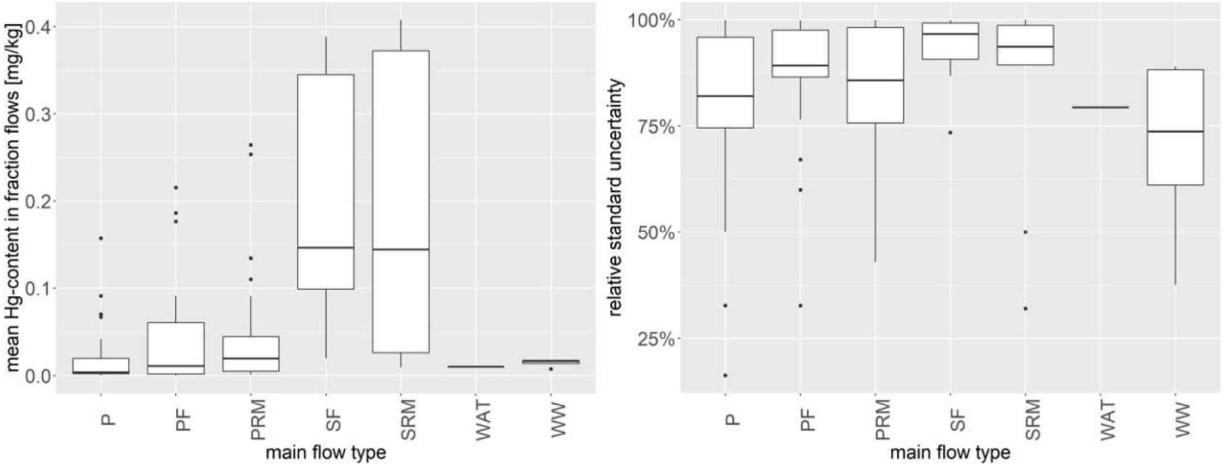

Fig. 2.. left: Distribution of mean Hg content of fraction flows in main flow types shown as absolute mean values of fraction flows in the corresponding main flow types: (products (P); primary fuels (PF); primary raw materials (PRM); secondary fuels (SF); secondary raw materials (SRM); water supply (WAT); wastewater (WW); right: relative standard uncertainty of Hg content of each fraction flow present in the corresponding main flow type.

Table 2.
Aggregated maximum and minimum Hg concentration with corresponding standard normal uncertainty of main flows (flow types and corresponding sector) for the period investigated, 2005–2016. The values displayed represent the applied weighted mean concentration and corresponding standard normal uncertainty for the main flow of the year with the lowest (min) and the year with the highest (max) Hg concentration, in mg/kg where not indicated differently.

	input [mg/kg]										output [mg/kg]			
	PRM		PF		SRM		SF		WAT [mg/m^3]		WW [mg/m^3]		P	
	min	max	min	max	min	max	min	max	min	max	min	max	min	max
C			0.016 ± 87%	0.017 ± 86%			0.097 ± 91%	0.11 ± 94%	0.01 ± 66%	0.01 ± 66%	0.013 ± 23%	0.013 ± 23%	0.0074 ± 44%	0.0087 ± 45%
EG			0.039 ± 61%	0.092 ± 61%					0.01 ± 66%	0.01 ± 66%	0.017 ± 75%	0.017 ± 75%		
MET	0.04 ± 66%	0.043 ± 64%	0.15 ± 62%	0.16 ± 62%	0.025 ± 92%	0.025 ± 92%	0.02 ± 96%	0.12 ± 96%	0.01 ± 66%	0.01 ± 66%	0.016 ± 44%	0.016 ± 44%	0.011 ± 95%	0.014 ± 95%
NMM	0.12 ± 71%	0.13 ± 90%	0.024 ± 45%	0.049 ± 45%	0.19 ± 74%	0.29 ± 76%	0.2 ± 44%	0.29 ± 59%					0.11 ± 87%	0.16 ± 91%
OTH	5.8e-09 ± 36%	0.0061 ± 38%	0.0034 ± 38%	0.0063 ± 30%			0.02 ± 95%	0.043 ± 71%	0.01 ± 66%	0.01 ± 66%	0.018 ± 76%	0.018 ± 76%	0.0058 ± 36%	0.0061 ± 38%
WPP	0.021 ± 70%	0.022 ± 65%	0.023 ± 50%	0.032 ± 53%	0.13 ± 19%	0.13 ± 19%	0.022 ± 84%	0.032 ± 69%	0.01 ± 66%	0.01 ± 66%	0.0076 ± 45%	0.0076 ± 45%	0.016 ± 62%	0.017 ± 59%

Even in China, where tungsten production is quite significant, there are no detailed studies, as there are of the primary production of other non-ferrous metals (e.g. zinc (Chung et al., 2017)) or ferrous metals (e.g. (Wang et al., 2016; Wu et al., 2017)).

Secondary raw material: For most secondary materials, Hg data is available. Secondary raw material was investigated in several different projects to evaluate the suitability of the material to replace primary raw material (Achternbosch et al., 2005, 2003; Finster et al., 2015; Lederer et al., 2017; Louch, 2005; Morf et al., 2007). Furthermore, waste needs to meet certain standards (e.g. limit for total Hg content or leaching limits (BMLFUW, 2017)) to be used as secondary material or to be disposed of in a certain way. This leads to multiple datapoints for Hg concentrations in secondary raw materials and secondary fuels. Furthermore, there is no solid indication to suspect that the Hg content of materials applied exceed the required limits for use. Nevertheless, it should be mentioned that the data should be treated with caution as the Hg concentration can vary significantly over time. This high uncertainty can be seen in the right part of Fig. 2, where the uncertainty ranges for secondary raw materials and fuels are notably higher than for the other investigated material groups.

Products: Data on Hg contents in various products are sparsely available. In particular, studies and data for industries where Hg is not known to be a prevailing problem are only sporadically available, e.g. for the wood pulp and paper industry (e.g. Louch (2005)).

3.2. *Mercury throughput*

The calculated Hg import into the system ranged between 3.5 t/yr ±70% and 4.0 t/yr ±70% over the period investigated. Roughly 70% to 75% are imported via primary raw materials and primary fuels, 25% to 30% via secondary raw materials and secondary fuels, and around 1% via water supply (see Fig. 4). As we assumed the system to be static, with no stock accumulation or depletion, the Hg export out of it amounts to the same value. Hg export via P was estimated at roughly 35% to 40%, via emissions between roughly 20% and 25%. Hg export via waste was estimated at roughly 40% to 45% (see Fig. 4). The results of the balance for the year 2016 are displayed in Fig. 3.

The overall Hg input into the systems did not change significantly over the time investigated. This is due to the few changes in the industrial sector (no new manufacturing sector, no suspension of manufacturing sectors) and the lack of manufacturing of products utilizing Hg which would potentially be phased out during this period. Furthermore, our model shows that the overall changes of Hg input into the system does not exceed the estimated relative standard uncertainty. Therefore, the changes are not significant.

Hg introduced into the system by hard coal was the most significant flow in each of the sectors investigated, ranging from 21% to 24% for other industries and up to 50% to 60% for the chemical industry. Sensitivity levels for all flows can be found in the supplementary information (SI).

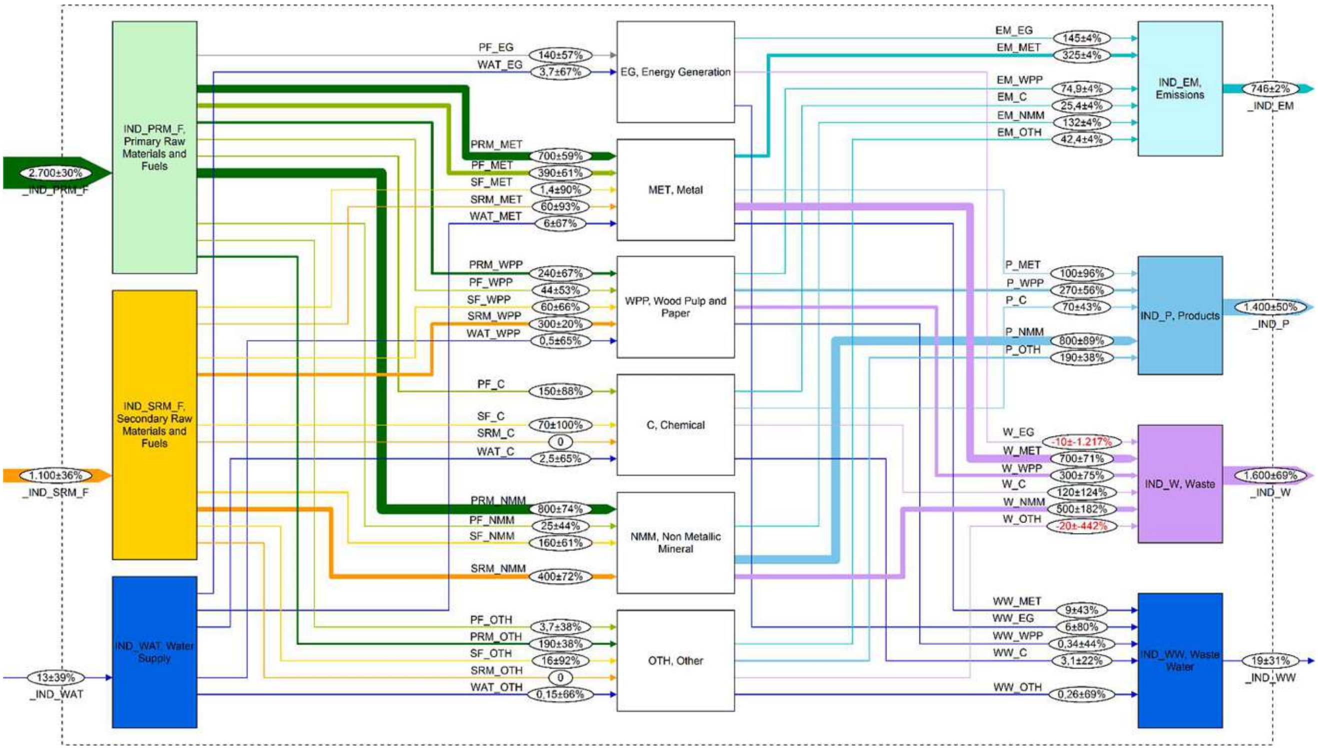

Fig. 3.. Hg throughput for the Austrian manufacturing industries: balance for 2016 in kg/yr (abbreviations: IND (summed up import and export flows); import main flow types: PRM (primary raw materials), PF (primary fuels), SRM (secondary raw materials), SF (secondary fuels), WAT (water supply); export main flow types: EM (emissions), P (products), W (wastes), WW (wastewater)Non-metal minerals and metal had the highest share in Hg throughput (roughly 30% to 40% and 30%, respectively), followed by wood pulp and paper and energy generation (roughly 16% to 28% and 4% to 14%, respectively). We calculated the lowest throughput in the chemical industry and other industries (approximately 6% and 4% to 6%, respectively). The significant lower shares of the chemical industry and other industries resulted from lack of data on raw material supply and product output for these industries. Therefore, the Hg throughputs of the chemical industry and other industries are most certainly underestimated.

3.2.1. Energy generation (EG)

EG consists of energy supply companies dedicated to supplying energy. Industrial enterprise internal energy production is allocated to the corresponding sector. Waste incineration plants with energy recovery are not included. In 2016 the Hg throughput was determined to be 140 kg/yr ±55%, whereas the main input (140 kg/yr ±60%) can be allocated to primary fuels. Further Hg input (around 13 kg/yr ±90%) arises from water supply. On the output side, emissions accounted for 145 kg/yr ±4% and wastewater for around 6 kg/yr ±80%. Waste was calculated to be -10 kg/yr ±1200%. As the Hg content is assumed to be normally distributed, theoretically negative values are possible. In particular, values relatively close to zero with high relative uncertainty might be negative even if physically not possible. It also might be an indicator of missing or underestimated (in this case input) flows. EG is the only sector where we observe a continuous reduction in Hg throughput. This is mainly due to the decommissioning of coal combustion plants for energy generation, which is part of the Austrian fossil fuel phase-out process. It is expected that future studies will confirm even lower Hg emissions from energy generation for the 2017–2020 period in coming years as the last coal combustion plant in Austria was decommissioned in 2020.

3.2.2. Metals (MET)

MET consists of the non-ferrous and ferrous metal industry, including coke production. The non-ferrous industry was considered mainly with regard to fuel consumption. The sole primary raw material of the primary non-ferrous production of tungsten (the sole primary non-ferrous metal production in Austria) was considered as well. The Hg throughput in 2016 was determined to be 1100 kg/yr ±45%. Primary raw material was the biggest Hg input (700 kg/yr ±65%), followed by primary fuels (390 kg/yr ±60%). Secondary raw materials, water supply and secondary fuels had the smallest Hg input (60 kg/yr ±90%, 9 kg/yr ±90%, 1.5 kg/yr ±90%, respectively). On the output side, emissions and products accounted for around 330 kg/yr ±4% and 100 kg/yr ±95%, respectively. As described above, coke production is balanced as part of the metal industry. Coke accounts for the Hg output via products of the metal industry. Wastewater accounted for 9 kg/yr ±45%. Waste was calculated to be 700 kg/yr ±70%.

3.2.3. Wood pulp and paper (WPP)

WPP consists of the Pulp and Paper industry and the wood processing industry (e.g. wood and wood-based panels). The Hg throughput in 2016 was determined to be 650 kg/yr ±30%. Hg input via secondary raw materials was estimated at 300 kg/yr ±20%, and primary raw materials at 240 kg/yr ±70%. Secondary fuels and primary fuels were estimated at 60 kg/yr ±66% and 45 kg/yr ±50%, respectively. Products accounted for 270 kg/yr ±55% of the output, emissions and wastewater for 75 kg/yr ±4% and around 0.5 kg/yr ±45%. Waste was calculated to be 300 kg/yr ±70%.

3.2.4. Chemical industry (C)

C consists of chemical industry and the fossil fuel refinery. Data is only available for refinery in- and output and fuel consumption. Hg throughput in 2016 was determined to be 220 kg/yr ±65%, whereas the most significant input was associated with primary fuels (150 kg/yr ±90%), followed by secondary fuels (70 kg/yr ±100%). With regard to output, products accounted for 70 kg/yr ±43%, and emissions for 25 kg/yr ±4%. Waste was calculated to be 120 kg/yr 125%.

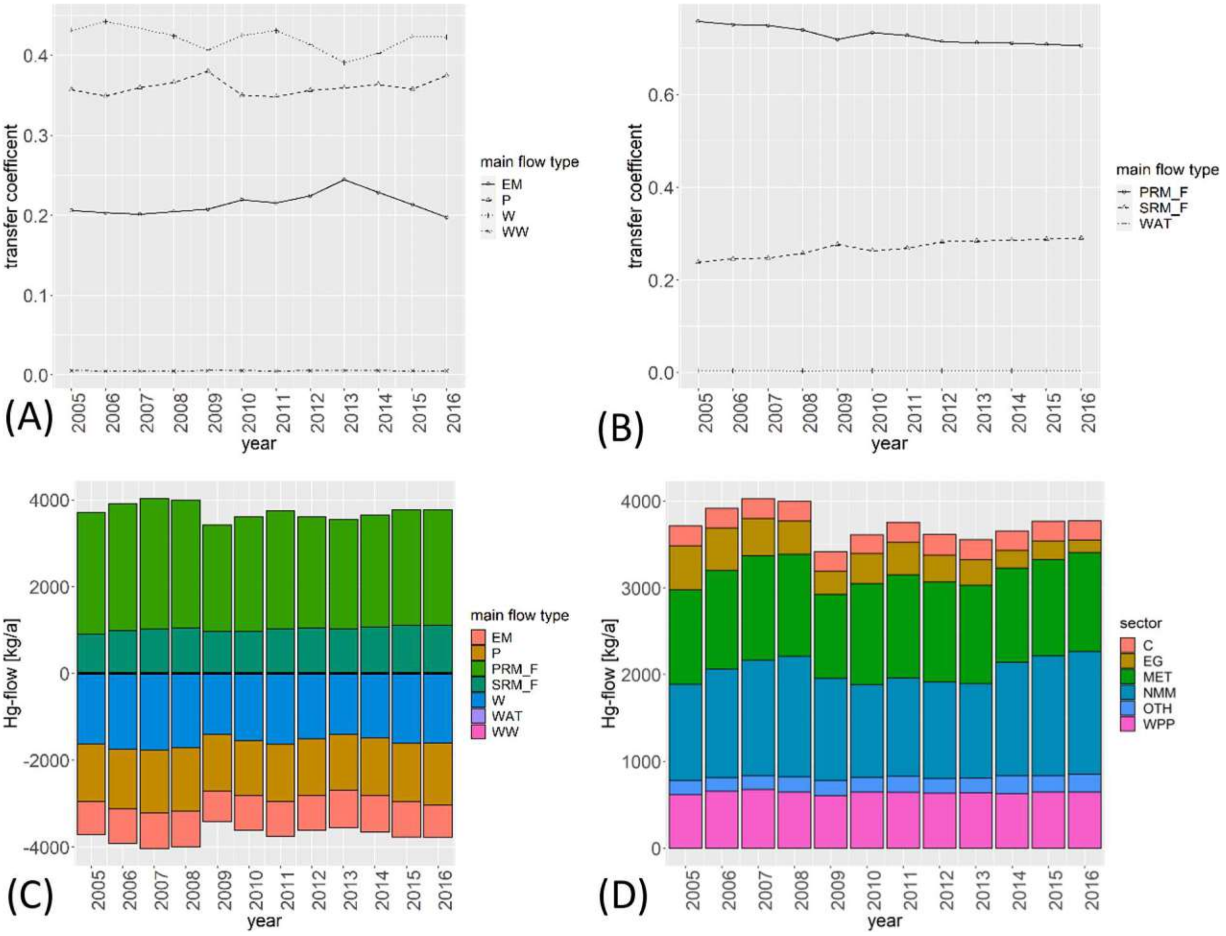

Fig. 4.. (A) Estimated shares of Hg import; (B) transfer coefficients to export flows; (C) Hg import and export categorized by flows (PRM_F: primary raw materials and fuels, SRM_F: secondary raw materials and fuels, WAT: water supply, WW: wastewater, W: waste, P: products, EM: emissions); (D) Hg throughput categorized by sectors (C: chemical industry, EG: energy generation, MET: metal industry, NMM: non-metallic minerals, OTH: other industries, WPP: wood, pulp and paper).

3.2.5. Non-metal minerals (NMM)

NMM consists of the cement industry, the tile and brick industry and the glass industry. In 2016 the Hg throughput was determined to be 1400 kg/yr ±50%. Primary raw materials are the most significant yearly Hg import (800 kg/yr ±75%), followed by secondary raw materials (400 kg/yr ±70%). Primary fuels were the smallest Hg Input (25 kg/yr ±45%), and secondary fuels (160 kg/yr ±60%) the second smallest. Products were the biggest output flow at 800 kg/yr ±90%, emissions accounted for around 130 kg/yr ±4%. Waste was calculated to be 500 kg/yr 180%.

3.2.6. Other industries (OTH)

OTH encompasses those industrial sectors assumed not to be as relevant in terms of Hg throughput and which are hence not further investigated. The estimated throughput in 2017 was 210 kg/yr ±35%. It accounts for the food industry, textile industry, machinery making and other industries. Primary raw materials and products were the biggest flows (190 kg/yr ±35%). The biggest share of these flows is related to food and products thereof as there is no data available for the other sectors (e.g. textile). In general, these industry sectors were not investigated in detail as they are not assumed to be as relevant as other sectors. This is also the case for waste streams, which the model estimated to be negative, with relative standard uncertainties ranging from ±240% up to ±1000%. The high relative standard uncertainties also resulted from the low values close to zero.

3.2.7. Wastes (W)

Over the timeline investigated, the estimated amount of Hg exported via waste did not change significantly (Fig. 5). On the one hand, the fluctuation was not high. However, the relative standard uncertainties were quite high, ranging from 70% for the individual waste flows (wood pulp and paper and metal) up to 3800% (EG). The Hg output via waste from energy generation constantly (but not significantly) decreased over this period, reaching a low of -10 kg/yr in 2016, which is the reason for the extraordinarily high relative standard uncertainty. The high relative standard uncertainties in all sectors and the negative values in other industries and with respect to energy generation are due to the following factors: i) natural variability of Hg content, ii) challenging laboratory analysis due to Hgs volatility and, iii) lack of process data in specific sectors and subsectors.

The waste flows were calculated from the balance of inputs (primary raw materials, primary fuels, secondary raw materials, secondary fuels, water supply) and outputs (emissions, products, wastewater). Therefore, high uncertainties and negative values indicate the lack of sufficient data quantity and/or quality. In the case of the energy generation sector, it indicates that the input of Hg was underestimated, hence flows of a good might be missing or the Hg concentration of the input materials might be estimated too low. It is also possible that emissions and/or wastewater are overestimated, but due to the more solid data base on the output side, this seems less likely.

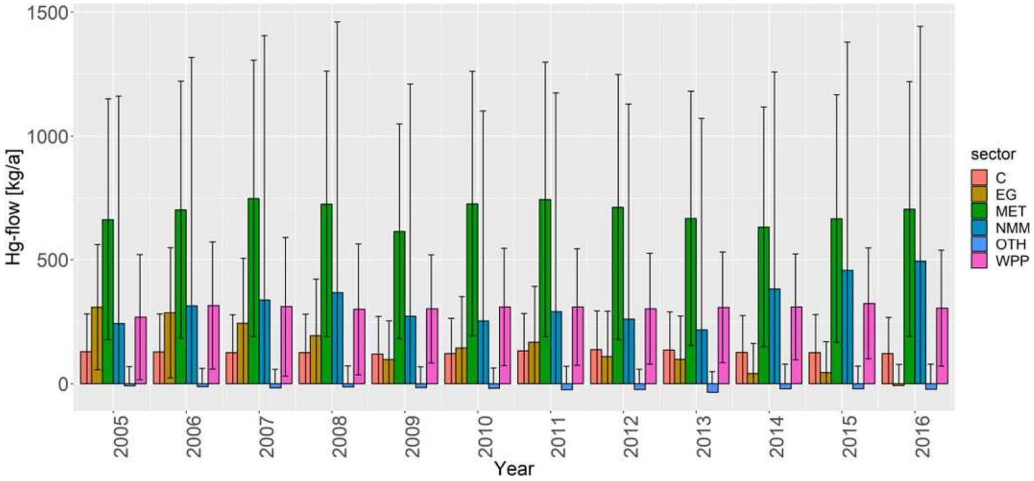

Fig. 5.. Development of Hg output via waste calculated for the sectors investigated (C: chemical industry, EG: energy generation, MET: metal industry, NMM: non-metallic minerals, OTH: other industries, WPP: wood, pulp and paper) for the years 2005–2016.

4. Discussion

The results of this study show that various data on Hg from different sources can be assembled in a balance model and that the balance conflicts are medium to moderate. This indicates that such surveys and studies are feasible. However, high uncertainties, negatively balanced flows as well as obviously existing data gaps indicate that no coherent balance of Hg for Austria is currently possible. The following reasons can be given: i) lack of data on flows of goods, partially due to confidentiality, ii) lack of detail on material composition of flows of goods, iii) inconsistency due to changes in reporting modes, iv) lack of accuracy of Hg concentrations due to high natural variability (see Fig. 2) and due to challenging lab analysis (sample preparation).

Nevertheless, the results allow some general conclusions regarding Hg throughput in the Austrian industry: Most Hg is imported via primary raw materials and primary fuels (70%-75%). A similar amount of Hg is exported via waste and products (40% to 45% and 35% to 40%, respectively). Around 20% ends up in the atmosphere. The overall Hg emissions to the atmosphere from industrial activity did not significantly decline over the period investigated. The detailed investigation of the industrial system thereby confirms findings of several studies regarding materials relevant to Hg import into the industrial system. The high share of Hg import via primary raw materials and primary fuels is also caused by the high-volume throughput of primary raw materials and primary fuels goods. The lower share of Hg imported by secondary raw materials and secondary fuels is caused by the lower shares in goods. Regarding Hg content, it can be stated that the current mix of secondary raw materials and secondary fuels is loaded with a higher Hg content than the primary raw materials and primary fuels mix. Hence a higher share of secondary material resulting from increased utilization of secondary raw materials has the potential to increase Hg throughput and therefore emissions to the environment. The higher Hg content in secondary raw material and fuels, and its consequences, are discussed in various publications (e.g. Achternbosch et al., 2005; Hayes et al., 2015; Krook et al., 2004). One option to avoid increasing Hg throughput involves establishing a threshold for the Hg content in secondary materials, as is in place for the Austrian cement industry (BMLFUW, 2017). However, this threshold (maximal permitted median) is higher than the estimated Hg content in the primary raw materials and primary fuels in this study. Therefore, even if it is met, it might not fully prevent a higher Hg input into the sector and therefore might lead to a potential increase in emissions. Moreover, it has to be acknowledged that the performance of reliable Hg analysis of secondary materials and fuels is difficult. Hence, the routine and reliable control of thresholds remains a challenge for every day practice. In other words, there is a big need for better sampling and analysis procedures and methods. Furthermore, the investigation of timelines is essential in terms of verifying the flows of goods as it is possible that changes in the reporting structure related to socio-economic accounting data are inevitable (e.g. change of reporting modes and parties, change of sections underlying confidentiality).

Public access to detailed datasets of Hg related information for specific industries or industrial plants (e.g. composite samples) would provide more insight into the system. Nevertheless, due to high variability, permanent Hg monitoring data seems to be the only means of deepening understanding of the dynamic of the Hg system over time. Original monitoring data on the inputs and outputs of materials with respect to Hg content would be required to determine reasonable Hg thresholds for secondary raw materials and fuels. Investigating and discovering potentially problematic flows is essential to understand and improve the system. Therefore, we are dependant on a solid database and the cooperation of industrial sectors. The application of a framework as proposed in the MinFuture project (Petavratzi et al., 2018) would also increase the value of such reported data since the explicit system context adds value to single isolated data. In other words, the sum of all the parts is more valuable than their individual parts.

However, as this balance shows exemplarily, industrial sectors that share their data in more detail are more likely to be investigated and part of such a study. By contrast, sectors which are more reserved and less inclined to share data are investigated less thoroughly as it is impossible to investigate without corresponding data. This circumstance does not encourage the sharing of information. Therefore, regulations calling for obligatory generation and reporting of specific data are required to guarantee a level playing field in this respect. Furthermore, the data collected in this manner should be put into context with the underlying system so that the dataset can be fully exploited, thereby avoiding the need to introduce assumptions into their definitions (Petavratzi et al., 2018). The risk of wrongly interpreting data would thereby be eliminated and subsequently allow material cycle monitoring via MFA (Petavratzi et al., 2018).

5. Conclusions

This study makes a scientific contribution by showing that it is generally possible to generate an Hg balance for the manufacturing industry, even though there are moderate data conflicts. Nevertheless, the results of the present study allow some general conclusions regarding the Austrian industry to be drawn: Most Hg is imported via primary raw materials and primary fuels (70%-75%). A similar amount of Hg is exported via waste and products (40% to 45% and 35% to 40%, respectively). Around 20% ends up in the atmosphere. The overall Hg emissions into the atmosphere from industrial activity did not significantly decline over the period investigated. The current mix of secondary raw materials and fuels is burdened with a higher Hg concentration than that of primary raw materials and fuels, which leads to the conclusion that the utilization of secondary raw materials and fuels needs to be well regulated and monitored to prevent rises in Hg emissions to the environment.

The study presented shows that industry sectors which share their data publicly are prone to be investigated more thoroughly. Therefore, regulations calling for obligatory generation and reporting of specific data are required to guarantee a level playing field in this respect. Therefore, Table 1 may serve as a guideline where improvements in regulations concerned with data collection and monitoring are needed.

In a next step we want to include other sectors (e.g. waste management) and the consumer sector (private households) in order to gain insight into the full anthropogenic Hg household of Austria, which can then be linked to the relevant environmental compartments. From this we expect to enhance our understanding and to generate more suitable measures to optimize Hg management. Furthermore, we would like to apply different, e.g. skewed probability distributions to investigate the influence on the result of the balance given.

Credit author statement

Sabine DWORAK: Conceptualization, Data Curation, Software, Investigation, Formal Analysis, Visualization, Writing - Original Draft Preparation, Writing - Review & Editing

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2020.105344.

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