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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 nonrepresentative). 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 $\pm 25\%$ to 4 t/yr $\pm 25\%$. 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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Full length article





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 (Petayratzi 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:

$$c_{j} = \frac{\sum_{i=1}^{k} c_{i} \dot{m}_{i}}{\sum_{i=1}^{k} \dot{m}_{i}}$$
(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.

$$S_{c_{j}}^{2} = \sum_{i=1}^{k} S_{\hat{m}_{i}}^{2} \left(c_{i} \left(\frac{1}{\sum_{i=1}^{k} \dot{m}_{i} \sum} - \frac{\sum \dot{m}_{i} c_{i}}{\left(\sum_{i=1}^{k} \dot{m}_{i}\right)^{2}} \right)^{2} \right) + \sum_{i=1}^{k} S_{c_{i}}^{2} \left(\frac{\dot{m}_{i}}{\sum_{i=1}^{k} \dot{m}_{i}} \right)^{2}$$
(4)

 S_{c_j} is the weighted relative standard uncertainty of the concentration c_j of the main flow j. $S_{\vec{m}_i}$ 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. <u>Fuels</u>: 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.

<u>Primary raw material</u>: 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			Concent	ration		Relevance
		Lipoit	Rating	Source	Comment	Rating	Source	Comment	
all	EM	EX	+	3	EMEP, industry reports are available in similar detail			not applicable	Ι
all	WW	EX	~	3,4	reporting biannually, pulp & paper: detailed yearly reporting in industry report	~	5		Ι
С	Р	EX	-	1	harmonized and aggregated	-	5,6		?
MET	Р	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	Р	EX	++	4	detailed reporting and publishing of production volumes for pulp & paper				III
WPP	Р	EX	++	4	detailed reporting and publishing of production volumes for cement	+	5,6		III
OTH	Р	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		Ι
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	Ι
all	WAT	IM	~	3,4	reported biannually until 2010; pulp & paper: detailed yearly reporting in industry report	~	5		III
С	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		П
MET	SRM	IM	~	4	steel scrap input reported since 2010; no data on non-ferrous metal scrap	~	5	little data available	п
NMM	PRM	IM	+	2,4	for cement inputs are available; for other sectors calculation on basis of production margins was applied	+	5,6		Ι
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	Ι
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.

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

<u>Secondary raw material</u>: 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.

<u>Water supply</u>: 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. <u>Products</u>: 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 PROD-COM (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).

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

<u>Waste</u>: 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.

<u>Wood pulp and paper</u>: 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).

<u>Metal industry</u>: 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.

<u>Chemical industry</u>: 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.

<u>Energy generation</u>: 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.

<u>Primary raw materials</u>: 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, PRM min	/kg] max	PF min	max	SRM min	max	SF min	max	WAT [m min	g/m ³] max	output [mg WW [mg/n min	g/kg] n ³] max	P min	max
С			$\begin{array}{c} \textbf{0.016} \pm \\ \textbf{87\%} \end{array}$	$\begin{array}{c} \textbf{0.017} \pm \\ \textbf{86\%} \end{array}$			$\begin{array}{c} 0.097 \\ \pm \ 91\% \end{array}$	$\begin{array}{c} \textbf{0.11} \pm \\ \textbf{94\%} \end{array}$	$\begin{array}{c} 0.01 \\ \pm 66\% \end{array}$	$\begin{array}{c} 0.01 \\ \pm 66\% \end{array}$	$\begin{array}{c} \textbf{0.013} \pm \\ \textbf{23\%} \end{array}$	$\begin{array}{c} \textbf{0.013} \pm \\ \textbf{23\%} \end{array}$	$\begin{array}{c} 0.0074 \\ \pm 44\% \end{array}$	$\begin{array}{c} 0.0087 \\ \pm \ 45\% \end{array}$
EG			$\begin{array}{c} \textbf{0.039} \pm \\ \textbf{61\%} \end{array}$	$\begin{array}{c} \textbf{0.092} \pm \\ \textbf{61\%} \end{array}$					$\begin{array}{c} 0.01 \\ \pm 66\% \end{array}$	$\begin{array}{c} 0.01 \\ \pm 66\% \end{array}$	$\begin{array}{c}\textbf{0.017} \pm \\ \textbf{75\%} \end{array}$	$\begin{array}{c} \textbf{0.017} \pm \\ \textbf{75\%} \end{array}$		
MET	$\begin{array}{c} \textbf{0.04} \pm \\ \textbf{66\%} \end{array}$	$\begin{array}{c} \textbf{0.043} \pm \\ \textbf{64\%} \end{array}$	$\begin{array}{c} \textbf{0.15} \pm \\ \textbf{62\%} \end{array}$	$\begin{array}{c} \textbf{0.16} \pm \\ \textbf{62\%} \end{array}$	$\begin{array}{c} 0.025 \\ \pm \ 92\% \end{array}$	$\begin{array}{c} 0.025 \\ \pm \ 92\% \end{array}$	$\begin{array}{c} 0.02 \pm \\ 96\% \end{array}$	$\begin{array}{c} \textbf{0.12} \pm \\ \textbf{96\%} \end{array}$	$\begin{array}{c} 0.01 \\ \pm \ 66\% \end{array}$	$\begin{array}{c} 0.01 \\ \pm 66\% \end{array}$	$\begin{array}{c} \textbf{0.016} \pm \\ \textbf{44\%} \end{array}$	$\begin{array}{c} \textbf{0.016} \pm \\ \textbf{44\%} \end{array}$	$\begin{array}{c} \textbf{0.011} \pm \\ \textbf{95\%} \end{array}$	$\begin{array}{c} \textbf{0.014} \pm \\ \textbf{95\%} \end{array}$
NMM	$\begin{array}{c} \textbf{0.12} \pm \\ \textbf{71\%} \end{array}$	$\begin{array}{c} 0.13 \pm \\ 90\% \end{array}$	$\begin{array}{c} \textbf{0.024} \pm \\ \textbf{45\%} \end{array}$	$\begin{array}{c} \textbf{0.049} \pm \\ \textbf{45\%} \end{array}$	$\begin{array}{c} \textbf{0.19} \pm \\ \textbf{74\%} \end{array}$	$\begin{array}{c} \textbf{0.29} \pm \\ \textbf{76\%} \end{array}$	$\begin{array}{c} \textbf{0.2} \pm \\ \textbf{44\%} \end{array}$	$\begin{array}{c} \textbf{0.29} \pm \\ \textbf{59\%} \end{array}$					$\begin{array}{c} \textbf{0.11} \pm \\ \textbf{87\%} \end{array}$	$\begin{array}{c} \textbf{0.16} \pm \\ \textbf{91\%} \end{array}$
OTH	5.8e-09 ± 36%	$\begin{array}{c} 0.0061 \\ \pm \ 38\% \end{array}$	$\begin{array}{c} 0.0034 \\ \pm \ 38\% \end{array}$	$\begin{array}{c} 0.0063 \\ \pm \ 30\% \end{array}$			$\begin{array}{c} \textbf{0.02} \pm \\ \textbf{95\%} \end{array}$	$\begin{array}{c} 0.043 \\ \pm \ 71\% \end{array}$	$\begin{array}{c} \textbf{0.01} \\ \pm \ \textbf{66\%} \end{array}$	$\begin{array}{c} 0.01 \\ \pm 66\% \end{array}$	$\begin{array}{c}\textbf{0.018} \pm \\ \textbf{76\%} \end{array}$	$\begin{array}{c}\textbf{0.018} \pm \\ \textbf{76\%} \end{array}$	$\begin{array}{c} 0.0058 \\ \pm \ 36\% \end{array}$	$\begin{array}{c} 0.0061 \\ \pm \ 38\% \end{array}$
WPP	$\begin{array}{c} \textbf{0.021} \pm \\ \textbf{70\%} \end{array}$	$\begin{array}{c} \textbf{0.022} \pm \\ \textbf{65\%} \end{array}$	$\begin{array}{c} \textbf{0.023} \pm \\ \textbf{50\%} \end{array}$	$\begin{array}{c} \textbf{0.032} \pm \\ \textbf{53\%} \end{array}$	$\begin{array}{c} 0.13 \pm \\ 19\% \end{array}$	$\begin{array}{c} \textbf{0.13} \pm \\ \textbf{19\%} \end{array}$	$\begin{array}{c} 0.022 \\ \pm \ 84\% \end{array}$	$\begin{array}{c} 0.032 \\ \pm \ 69\% \end{array}$	$\begin{array}{c} 0.01 \\ \pm 66\% \end{array}$	$\begin{array}{c} 0.01 \\ \pm \ 66\% \end{array}$	$\begin{array}{c} 0.0076 \\ \pm \ 45\% \end{array}$	$\begin{array}{c} \textbf{0.0076} \\ \pm \text{ 45\%} \end{array}$	$\begin{array}{c} \textbf{0.016} \pm \\ \textbf{62\%} \end{array}$	$\begin{array}{c} \textbf{0.017} \pm \\ \textbf{59\%} \end{array}$

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.

<u>Products</u>: 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 \pm 70% and 4.0 t/yr \pm 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 \pm 55%, whereas the main input (140 kg/yr \pm 60%) can be allocated to primary fuels. Further Hg input (around $13 \text{ kg/yr} \pm 90\%$) arises from water supply. On the output side, emissions accounted for 145 kg/ yr $\pm4\%$ and wastewater for around 6 kg/yr $\pm80\%$. Waste was calculated to be -10 kg/yr \pm 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 \pm 45%. Primary raw material was the biggest Hg input (700 kg/yr \pm 65%), followed by

primary fuels (390 kg/yr \pm 60%). Secondary raw materials, water supply and secondary fuels had the smallest Hg input (60 kg/yr \pm 90%, 9 kg/yr \pm 90%, 1.5 kg/yr \pm 90%, respectively). On the output side, emissions and products accounted for around 330 kg/yr \pm 4% and 100 kg/yr \pm 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 \pm 45%. Waste was calculated to be 700 kg/yr \pm 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 \pm 30%. Hg input via secondary raw materials was estimated at 300 kg/yr \pm 20%, and primary raw materials at 240 kg/yr \pm 70%. Secondary fuels and primary fuels were estimated at 60 kg/yr \pm 66% and 45 kg/yr \pm 50%, respectively. Products accounted for 270 kg/yr \pm 55% of the output, emissions and wastewater for 75 kg/ yr \pm 4% and around 0.5 kg/yr \pm 45%. Waste was calculated to be 300 kg/ yr \pm 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 \pm 65%, whereas the most significant input was associated with primary fuels (150 kg/yr \pm 90%), followed by secondary fuels (70 kg/yr \pm 100%). With regard to output, products accounted for 70 kg/yr \pm 43%, and emissions for 25 kg/yr \pm 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 \pm 50%. Primary raw materials are the most significant yearly Hg import (800 kg/yr \pm 75%), followed by secondary raw materials (400 kg/yr \pm 70%). Primary fuels were the smallest Hg Input (25 kg/yr \pm 45%), and secondary fuels (160 kg/yr \pm 60%) the second smallest. Products were the biggest output flow at 800 kg/yr \pm 90%, emissions accounted for around 130 kg/yr \pm 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 \pm 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 \pm 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 \pm 240% up to \pm 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: nonmetallic 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

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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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