Journal of
Industrial Ecology

A Unifying Theory of Hybrid Life-Cycle Assessment

Journal:	*Journal of Industrial Ecology*
Manuscript ID	24-JIE-8494.R1
Wiley - Manuscript type:	Methods Article
Date Submitted by the Author:	30-Apr-2025

Complete List of Authors:	Weinold, Michael; Paul Scherrer Institut PSI, PSI Center for Energy and Environmental Sciences Majeau-Bettez, Guillaume; Ecole Polytechnique de Montreal, CIRAIG, Chemical Engineering Department

Keywords:	hybrid life-cycle assessment, life cycle assessment (LCA), input-output life cycle assessment (IO-LCA), methods, environmental impact assessment
User-Supplied Keywords:	rumsfeld, unknown unknown, monetary balance

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A Unifying Theory of Hybrid Life-Cycle Assessment

Michael P. Weinold[1,2,†], Guillaume Majeau-Bettez[3]

[1]Laboratory for Energy Systems Analysis, PSI Center for Energy and Environmental Sciences, Paul Scherrer Institute PSI Villigen, Switzerland

[2]Chair of Energy Systems Analysis

Department of Mechanical and Process Engineering, ETH Zurich, Zurich, Switzerland

[3]Department of Chemical Engineering, Polytechnique Montréal, Montréal, Canada

[†]Corresponding Author: `michael.weinold@psi.ch`

Abstract

Life-cycle assessment (LCA) is a key method for analyzing decarbonization strategies of techno-economic systems, with results forming the basis for environmental decision-making. In this context, hybrid life-cycle assessment (HLCA) methods seek to combine bottom-up data from process-based life-cycle inventories with top-down data from environmentally extended input-output tables. This is done to overcome limitations of data coverage and aggregation: Process inventory data, though detailed, may underestimate flow magnitudes or miss flows entirely, potentially underestimating environmental impacts. On the other hand, input-output data is by definition complete for intermediary flows, but highly aggregated into a small number of economic sectors and generally does not capture the use-phase of products. Combining these data sources would give a more complete picture of the environmental impact associated with products or services. Four main methods are currently recognized in literature: The integrated, tiered, matrix-augmentation and path-exchange hybrid methods. However, no consensus has emerged on the underlying mathematical differences and their applicability. Here, we propose a unifying theory of HLCA. First, we show that all methods can be expressed in the same matrix form. Using this insight, we show that instead of four, only two distinct methods exist: the tiered and the integrated methods. We show how both can be used to account for "known unknown" and "unknown unknown" data and how economic equilibrium can be maintained through balancing conditions. Finally, we offer recommendations on the optimal selection and application of each method. This constitutes an important step toward a unified methodological framework for hybrid life-cycle assessment.

1 Introduction

Life-cycle assessment (LCA) is a standardized [1] method designed to evaluate the environmental impacts of products or services. Today, it is used as a key evidence-based environmental decision support tool by industry and policy makers alike [2][3][4]. When used prospectively, the method allows for an evaluation of the effectiveness of different climate change prevention and mitigation strategies [5]. Ensuring accurate results is crucial, as they form the foundation for making reliable and effective environmental decisions that can significantly influence policy and industrial practices. **??** shows selected core concepts of the method using the example of a simplified automotive supply chain.

Life-cycle assessment based on a process-based inventory (PLCA) constitutes the majority of studies conducted today. The process-based inventory is a structured collection of data that contains information about different production processes in the economy. It includes information on the type and amount of physical flows between different processes, and the associated environmental burdens [6]. Usually, only a small subset of this data - the *foreground* - is compiled by life-cycle practitioners themselves, while the vast majority of processes - the *background* - is taken from commercially available general-purpose life-cycle inventory databases, such as *Ecoinvent* [7] or *GaBi* [8]. This is because the effort associated with the compilation of such large-scale process databases is considerable [9]. In reality, the complete supply chain of products or services can be infinite, since each production process connects to numerous upstream and downstream processes, creating an extensive graph of inter-dependencies. For practical reasons, a *system boundary* is drawn during assessment, describing the subset of processes which is considered by the practitioner. A consistent determination of the system boundary remains an unresolved problem of life-cycle assessment [10, Sec.2.2]. The resulting inevitable difference between the computed result of an assessment and the inaccessible [11, Theorem 5] *true* value is known as the *truncation error* [12]. What is more, unless complete coverage of monetary and physical flows related to a production process has been achieved, it is possible that important flows or environmental burdens have been omitted from the process description. This is true regardless of whether a production process is part of the foreground system or the background system. Errors of this kind can accumulate through the supply chain and lead to underestimation of the ultimate environmental burdens associated with the life-cycle of a product.

On the other hand, the same kind of analysis can be conducted on the basis of environmentally-extended multi-regional input-output (EE-MRIO) tables. These tables record the monetary flow between different sectors of the economy and are prepared on an annual basis by national governments or international organizations [13, Sec.13.1]. They were originally proposed by Wassily Leontief as a tool to study the relationships between different sectors and their contributions to overall economic output [14], and in the 1970s were extended to include sectoral environmental burdens [15]. While input-output tables fully account for all monetary or material flows between sectors in the production process, making their coverage complete for economic flows, they do not incorporate information on the use-phase of products. Care must also be taken in recognizing that all sectoral data, both economic flows and associated environmental burdens, describe sectoral averages, with associated resolution varying widely between countries. The Japanese table, for instance, provides high resolution data for the electronics industry, with *"sectors that include 'personal computers', 'commercial residential air conditioners', and 'cameras'* [16, Sec.3.2]. On the other hand, such processes would all be contained in a single sector *'manufacture of computer, electronic and optical products'* in the Swiss table [17]. The utility of input-output tables in the life-cycle assessment of specific products is therefore limited. This is primarily due to the *aggregation bias* which is inherent to these tables [18]. Instead, it is frequently recommended that the approach be used *"as a template for [more detailed process-based] LCA"* [19, Sec.14.3.3] or as *"a first proxy"* [20, Sec.4]. A direct comparison between the carbon footprints obtained from IOLCA and PLCA as recently performed by Steubing et al. [21] is therefore also not directly meaningful, given the contrasting levels of detail and scope inherent in each approach. The governing equations of this approach are quite similar to those for the process-based approach, although all flows are generally in monetary units [22]. Life-cycle assessment based on environmentally-exended input-output tables (IOLCA) is often referred to using different abbreviations, including MRIO-LCA [23], IO-LCA [24], EIO-LCA [25], EIOLCI [26], EEIOA [27] or simply EEIO [28]. It should be noted that the use of the abbreviations EEIOA and EEIO to designate life-cycle assessment based on environmentally extended input-output data can be confusing, since they have historically referred to input-output analysis in a more general sense, which differ from life-cycle assessment in their scope and application.

2 Graphical Abstract

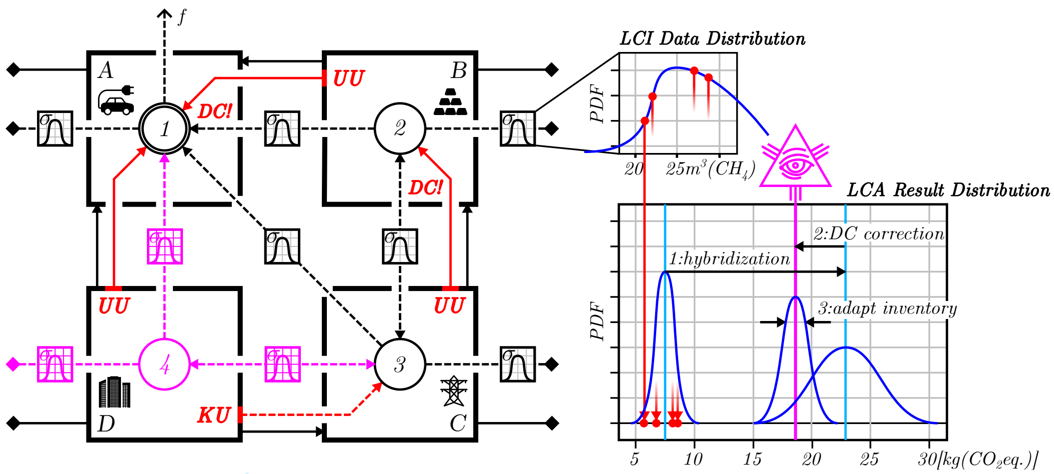

Figure 1: **Visual abstract of selected core concepts in hybrid life-cycle assessment (HLCA).** For a complete legend of the diagrammatic notation used, compare Fig. 6. Specific manufacturing steps are represented as circles, with corresponding industry sectors as enclosing squares. Dashed arrows indicate that flow data is derived from a process database, while solid arrows indicate that flow data is derived from an input-output table. Distribution symbols indicate that flow data may be statistically distributed due to inherent uncertainty. Red color indicates flow from sectors to processes. Pink color indicates processes and flows which exist in reality, but of which the practitioner is unaware. On the left, the highly simplified supply chain for the production of a car is shown, including assembly (1), metals production (2), electricity production (3) and the service sector (4). The overall environmental impact of automotive production can be evaluated by adding the environmental burden of every step in the supply chain. In the case where data is distributed, repeated random sampling of life-cycle inventory (LCI) data can be used to derive a probability density function (PDF) for the life-cycle assessment (LCA) environmental impact result. From this representation it is evident that total emissions will be underestimated, because the practitioner has no knowledge about flows from process (4) in the service sector. It may also be the case that inputs derived from a process database may be incomplete. For instance, a greater amount of metal may be required to build the car than the one stored in the life-cycle inventory database. The Eye of Providence indicates this *true* emissions value, which is accessible only to an omniscient observer. Hybrid life-cycle assessment can now be used to mitigate this limitation by using information from flows between sectors of the economy, which are captured in input-output tables. For example, if the magnitude of a flow into a process is known but not the details of its upstream production (*"known unknown", KU*), we can simply add a flow from the relevant sector into our process. On the other hand, if the practitioner has no knowledge about either the flow or its upstream production (*"unknown unknown", UU*), it could be inferred from the structure of the input-output table. We provide a detailed discussion of these epistemic categories in Section 3.3. Using data from the sectoral system ("hybridization") will broaden the system boundary of the process-based assessment and therefore capture more environmental emissions. However, the probability density function of the assessment result will also broaden, because additional uncertainty is introduced. For instance, price data for all hybridized processes is required for hybridization. Also, some material flows may now be captured twice - once in a process flow and once in a sectoral flow. This is known as *double-counting* and must be corrected for ("DC correction"). Finally, some of the sectoral data can be replaced with specific process data in an iterative process, narrowing the results distribution and ultimately providing an emissions value more closely aligned with reality. Note that this presentation follows a conceptualization of hybrid life-cycle assessment originally proposed by Perkins & Suh [29] as well as Agez et al. [30].

3 Hybrid Life-Cycle Assessment (HLCA)

3.1 The Promises of HLCA

In this context, researchers have attempted to combine the aggregated top-down input-output-based inventory and the dis-aggregated bottom-up process-based inventory. The primary motivation for using hybrid methods in life-cycle assessment is to obtain a more accurate and comprehensive environmental impact assessment. The first attempt was made in the 1970s [31], with most subsequent improvements made from the 1990s [32]. The umbrella term *hybrid life-cycle assessment* for these methods first saw use after 2000, for instance in a review by Lenzen et al. [12]. Fundamentally, hybrid life-cycle assessment can be framed in two ways. First, as a way to extend the system boundary [33] of a process-based life-cycle inventory using sectoral data, thereby reducing the truncation error of the impact assessment result [12]. Second, as a way to improve the specificity of an input-output based life-cycle inventory (input-output table) using process data, thereby reducint aggregation error.

It has been shown that the use of hybrid methods can significantly improve the coverage of purely process-based life-cycle assessment. This is most frequently done by estimating the truncation error associated with the assessment result of different production processes in large process-based background databases. Yu and Wiedmann for processes in the Australian Life Cycle Inventory Database (AusLCI) found a 21-32% underestimation in carbon emissions [27, Sec.3.2], while Crawford for another Australian database in the same year found an average of 50% [34]. Ward in his meta-analysis asserts that *"that mean truncation error estimates are likely to be in the range of 30% to 80%"*[35]. More recently, Agez et al. through their hybridization of the entire Ecoinvent database with a conservative approach for avoiding double-counting were able to provide a lower estimate of 5-16% underestimation in global warming potential (GWP100) across processes, with significantly higher values for other environmental burdens, such as freshwater ecotoxicity [36, Fig.2].

3.2 Double-Counting and Data Fusion Problems in HLCA

Hybrid life-cycle assessment aims to quantify the environmental impacts along a supply chain by drawing on data sources which are partly complementary and partly overlapping. In this context, the problem of *double-counting* arises. This refers to the unintentional duplication of environmental impacts or resource flows within the system boundaries of a hybrid life-cycle assessment, arising from the integration of partly overlapping data sources. However, this is not the only concern in hybrid systems— overcounting of flows not required by the system and undercounting of essential flows may introduce additional inaccuracies. We provide a comprehensive illustration of double-counting and other data fusion issues in Fig. 2.

The mitigation of issues arising from this data fusion constitutes an active field of inquiry. However, such efforts can be regarded as separate from hybridization methods themselves. We therefore refer the reader to a recent review by Agez et al. [37] for a discussion of mitigation methods.

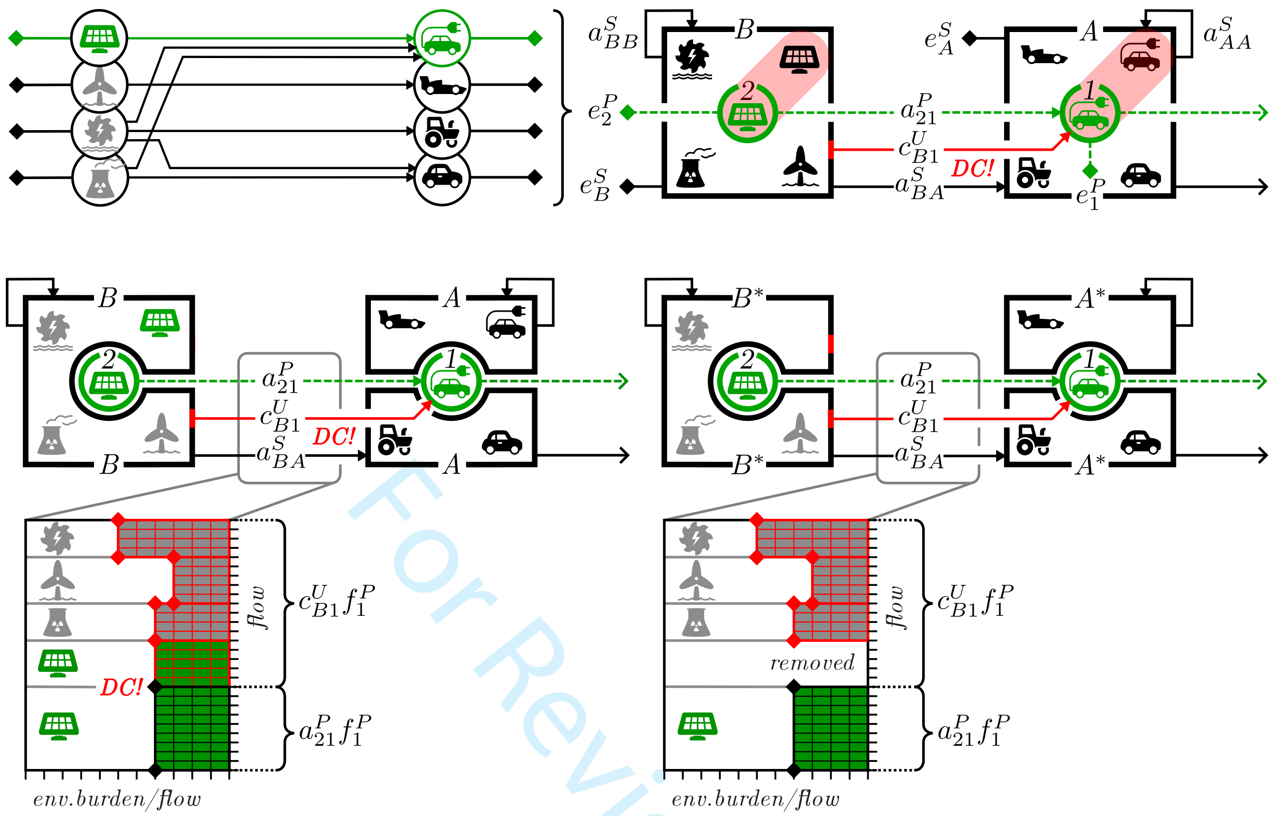

Figure 2: **Comprehensive diagrammatic illustration of "double-counting" and associated data fusion issues in hybrid life-cycle assessment.** *Top left:* Representation of the *"real supply chain"* of this example system. Different electricity production processes provide input to different automotive manufacturing processes. Here, we investigate the electric vehicle assembly process highlighted in green. It consumes electricity from latest-generation ("new") solar cells, wind and hydro power. Electricity sources not providing input to the process are highlighted in pink. This includes previous-generation ("old") solar cells and nuclear power. *Top right:* Diagrammatic representation of the example system. The practitioner has specific information on the new solar cell electricity production process. In addition, he knows the amount of input required from the other two relevant electricity production processes. In order to approximate associated emissions, he places this demand on the electricity sector through c^U_{B1}. *Bottom left:* The environmental burdens resulting from flows $A \rightarrow B$ and $1 \rightarrow 2$ are shown in detail. The total amount of upstream flow from sector B to process 1 is determined by $c^U_{B1} f^P_1$. Note the composition of the flow $c^U_{B1} = a^S_{BA}$, which due to the aggregated nature of the sectoral system includes flows not actually required by the automotive process, highlighted in pink. Note also that the electricity production sector contains an average of all solar cells, both old and new. *Bottom right:* Removing the new solar cells from the electricity sector B leaves a disaggregated sector B^*, which now contains only the old solar cells. There is now no *double-counting* of the flow from new solar cells. However, we can see that flows from electricity sources not required by the automotive process are still *"overcounted"*, while the flows from electricity sources required by the automotive process are *"undercounted"*. This constitutes an example of the well-known aggregation bias inherent to input-output systems [38][39][40]. Only in a fully disaggregated system would there be no problems of double-counting, overcounting and undercounting. Note that while all concepts in this figure apply to both the integrated hybrid and the tiered hybrid methods, only upstream flows are shown in the system to maintain a managable level of visual complexity. For a legend of the diagrammatic notation used, compare Fig. 6.

3.3 Different Scopes of HLCA

While the primary motivation for using hybrid methods has not changed over time, the scope of hybrid methods has recently expanded from addressing only *known unknown* inputs to the process system to also uncovering *unknown unknown* inputs by inferring them from the input-output system. This is a crucial difference, which also requires distinct mathematical approaches. Unfortunately, we find that of the publications we surveyed for Section 3.4, only one takes note of this important development: Mattila in a section of the textbook by Hausschild et al. [19, Sec.14.3]. Here, we have illustrated the conceptual difference in Fig. 3 and describe the two approaches below. A more formal definition is provided in Section 10.2.

In general, knowledge can be classified according the *Rumsfeld matrix*. This conceptual framework categorizes knowledge into four distinct types based on what is *known* and *unknown*. It is named after U.S. secretary of defense Donald H. Rumsfeld, who popularized the framework in a 2002 press conference [41]. It has since been adopted in the literature of policy-making [42] and strategic management [43][44][45], as well as the natural sciences [46].

In our context, we define as *knowns* those flows to processes which are covered by the bottom-up process inventory. Conversely, we define as *unknowns* those flows to processes which are not covered by the bottom-up process inventory. With these definitions, we introduce a version of the *Rumsfeld matrix* adapted for hybrid life-cycle assessment in Table 1.

Table 1: The *Rumsfeld matrix* [42] of knowledge classification adapted for identifying the different kinds of knowledge practitioners can have on flows from different production processes or economic sectors to a specific production process.

known knowns (KK (zero))	*known unknowns (KU)*
We know the quantity of each flow into our specific production process and have an associated flow production process. We also know that our specific production process does not require upstream inputs from certain sectors. This could be because we have complete mass and/or cost coverage in our process inventory. These flows are know to have magnitude zero.	We know there is a flow from an economic sector to our production process. This could be because the bill-of-goods of the reference product lists input from a supplier, which we know to be belonging to a specific economic sector. However, we do not have a process description for its production. Therefore, we we fall back on the economic sector by placing demand on in through an upstream flow. Practitioners sometimes also use proxy datasets from the process inventory as an alternative.
unknown knowns (UK)	*unknown unknowns (UU)*
In the context of the original *Rumsfeld matrix*, this would describes knowledge that is internalized by the practitioner, albeit without explicit awareness. Since we have adapted the matrix to specifically reflect *knowledge about data*, rather than human knowledge in the context of decision-making, this concept is not applicable to our case of flows in hybrid life-cycle assessment.	We do not know of any flow into our specific production process. We therefore also do not know anything about the associated flow production process. However, we suspect that the inputs to our process are *not* complete. This could be because we know that the cost of the inputs is *not* equal to the sum of the output price plus the known added value of the process. Therefore, we infer this flow from the inherent structure of the input-output table.

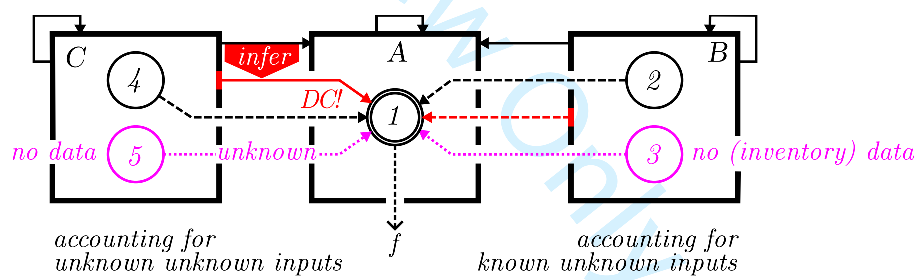

Figure 3: Example system illustrating the two scopes of hybrid life-cycle assessment, described in Section 3.3 with mathematical definitions provided in Section 10.2. Right: Case of a *known unknown* input $3 \rightarrow 1$, where no process-level data is available to the practitioner in existing databases. Instead, demand is placed on sector B, resulting in the *manually added* upstream flow $B \rightarrow 1$. Left: Case of an *unknown unknown* input $5 \rightarrow 1$, where the practitioner is unaware of the existence of the upstream process. By inferring flows from the inherent structure of the sectoral system, this flow can be approximated into an upstream flow $C \rightarrow 1$. It is evident that this relies on the matching between processes and sectors. Also, there might be double-counting of flows of emissions, as indicated by DC!. For a legend of the diagrammatic notation used, compare Fig. 6.

Note that the inference of upstream flows introduces potential instances of *double-counting*. This describes instances in the hybrid system where an upstream flow from a sector into a process is already covered by a process flow. This is illustrated in the example system of Fig. 3.

3.3.1 Using Input-Output Data as a Background Database ("known unknowns")

Early publications using hybrid methods sought to use input-output data in the way practitioners today use generic background databases. While a foreground inventory for the reference product is compiled, any inputs into this foreground inventory were taken directly from different sectors of the economy. For instance, a product of interest might require an input of steel, the demand for which is placed directly on the economic sector of steel production of the sectoral system. Input-output data was, at that time, sometimes described as a *"shortcut to life-cycle data"* [47]. Aftera all, large-scale generic life-cycle databases were still in their infancy in the late 1990s. The historical growth of the number of unit processes in these databases is illustrated in Fig. 4. It should be noted that this growth in process coverage is not uniform across the economy, and that some sectors remain highly underrepresented [28, Fig.1].

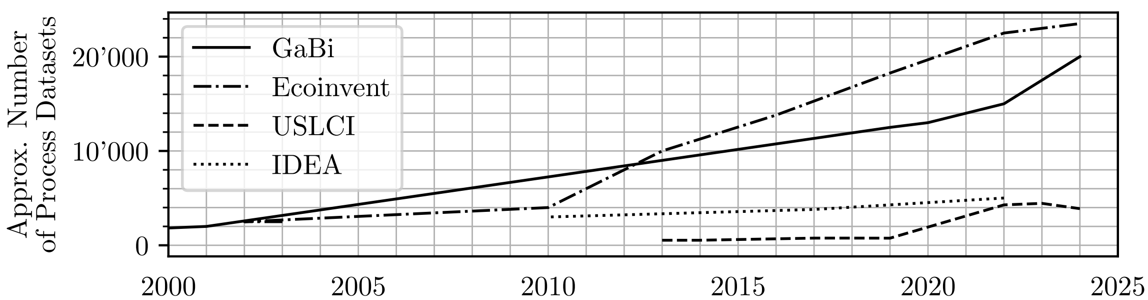

Figure 4: **Historical growth in the number of unit process datasets included in large generic process-based life-cycle databases.** Sources: IDEA [48][49][50], USLCI [51][52], Ecoinvent [53] and downloaded database versions, GaBi [54] and [55][8][56].

3.3.2 Using Input-Output Data to infer Missing Upstream Flows ("unknown unknowns")

As background databases have become more comprehensive in their coverage of important processes in the global economy, their use by life-cycle assessment practitioners has become more established [57, Sec.6]. Still, as discussed in Section 1, it is likely that processes described in the databases are not complete with respect to associated flows and environmental burdens. In this context, more recent publications using hybrid methods have aimed to account for these flows which may have *unknowingly* been omitted during the compilation of the life-cycle inventory.

To our knowledge, this approach was first explicitly proposed by Strømman in 2008 [58]. There, the inherent structure of the input-output table is used to infer upstream flows from sectors to processes. What is more, the amount of upstream flows added to processes is made dependent on the amount of flows still missing. This requires information on the basic prices of all products in the inventory. For instance, in Fig. 3, process 1 could be the manufacturing of a specific automobile. It may therefore be contained in the *vehicle manufacturing* sector. From this, we can infer that an economic flow from the *service industry* sector will likely also be consumed by the specific car manufacturing process - even though we are unaware of the existence of the associated flow or process.

It should be noted that the *path-exchange* method of Treloar, Lenzen & Crawford [59][60] also implicitly infers upstream flows from the input-output table, albeit without scaling flows not to exceed the amount of missing flows. Both Strømman as well as Treloar and Lenzen have suggested to perform the matching in an *automated* way, thereby allowing the hybridization of entire background databases. It should be noted, however, that this was *"(...) not meant as a substitute for gathering original inventory as such."* [58].

3.4 Present Issues of HLCA

Four main methods are currently recognized in literature [32][61]: The *path-exchange (PXC) method*, proposed in 1997 by Treloar [59] and formalized in 2009 by Lenzen et al. [60]. The *matrix augmentation method*, introduced in 1999 by Joshi [62]. The *integrated method*, introduced in 2000 by Suh and Huppes [63]. The *tiered method* proposed in 1978 by Bullard et al. [31] and formalized by Heijungs et al. in 2002 [6], which was further improved to avoid double counting by Agez et al. between 2019 and 2022 [37][30][36].

Despite recent efforts aimed at establishing a standardized taxonomy of methods [32], we find that a considerable amount of confusion still remains both over the naming and even the mathematical distinctions between methods. For instance, a great diversity of terms is used by practitioners when self-describing the method used in their publications, shown in Fig. 5.

A varying subset of hybrid methods has been covered in life-cycle assessment textbooks since at least 2002 [6, Sec.5.4], albeit often only superficially [64, P.80][65, Sec.3.4][66, Sec.4.4.5][67, Sec.14][68, P.185], without a mathematical definition [69, Sec.9] or simply as a short preface to sections on more well-established input-output assessment methods [70, Sec.3.5][9, Sec.2.5][71, Sec.7.9.2]. Of the textbooks surveyed, only Nakamura and Nansai [16] and Crawford [72, Sec.3.3.2.2 & Sec.4.4.2] discuss the different methods in detail, albeit without making any specific recommendation on their use. In fact, even these latter two works refer the reader to the original publications for the definition of matrices and equations.

Hybrid methods have also been reviewed in a number of recent scientific publications, including the reviews of Hagenaars et al. [73], Bakindi et al. [74], Crawford et al. [32] and Islam et al. [61], or the earlier review by Suh and Huppes [20, Table 1] and the doctoral thesis of Cruze [26]. The shared verdict of the authors is that the *integrated hybrid* method would provide the most accurate results, while also being the most difficult to implement, due to its extensive data requirements. The *tiered hybrid* method is often described as the most straightforward to apply. Still, *"there is no real consensus over the preferred method."* [32, Sec.1]. For instance, Islam et al. assert that *"for long term decision[s] tiered hybrid or [matrix-augmentation] (...) hybrid is appropriate. On the contrary, with time and money available, the choice (...) should clearly be integrated hybrid."* [61, Sec.5], thereby mirroring the earlier contention of Suh et al. that *"The tiered hybrid analysis has the appeal of easy extension on existing simple partial LCA systems in filling in the gaps. (...) With time and money available, the choice clearly is for the integrated hybrid analysis."* [20, Sec.4]. Cruze makes no such recommendation, providing only his observation that *"interestingly, the tiered hybrid result sometimes understates life cycle emissions relative to the more comprehensive methods."* [26, Sec.7]. Finally, Crawford state that *"the PXC and Integrated hybrid methods are the most fit for [mathematical] normalisation, as a strict framework is already in place to apply these methods. Additionally, (...) these two provide the most comprehensive approach for hybridising process and input-output data."* [32]

More generally, this is illustrative of life-cycle assessment as a "young field" of techno-economic and environmental modeling. While going back to the 1960s, it was formalized only in the 1990s [75][76][77] along with the present-day mathematical framework [78][79].

4 Aim and Scope

Here, we derive the governing equations of the four main methods for hybrid life-cycle assessment using a consistent mathematical framework. In the process, we show that despite previous report in literature, only two distinct methods exist. We then introduce an epistemic classification of the flows between the process system and the sectoral system, allowing us to describe classify methods according to the way in which they use the input-output data. Finally, we provide guidance to practitioners on when to use which method.

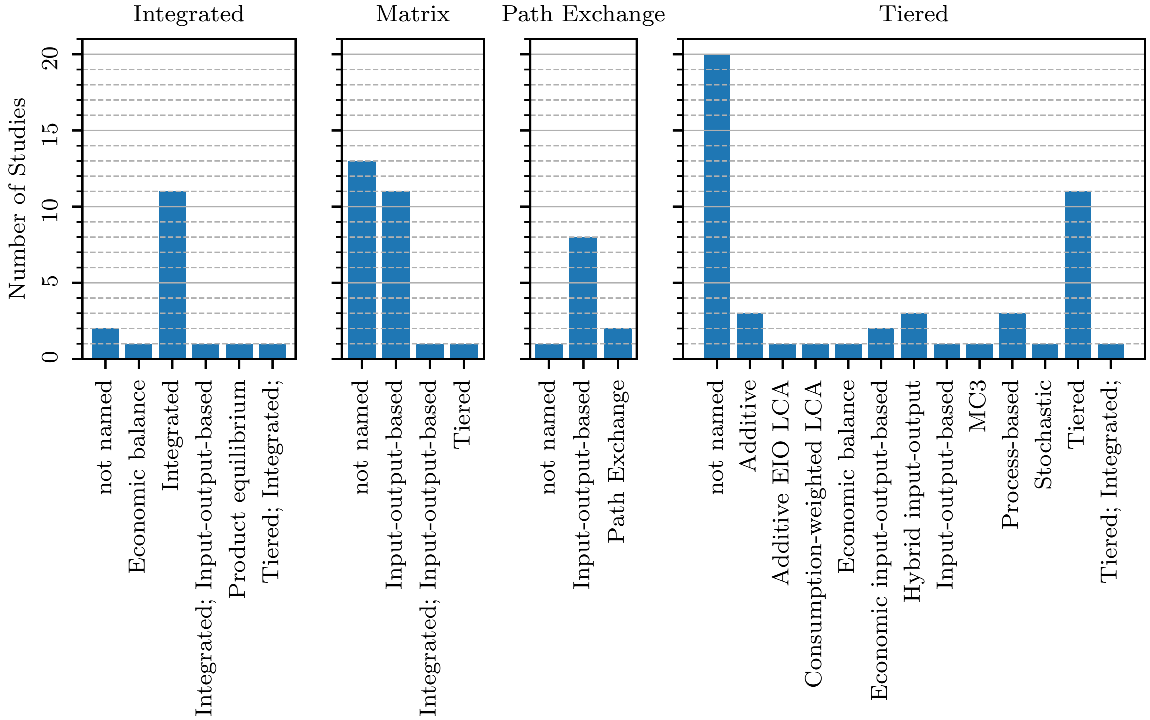

Figure 5: **Histogram showing the diversity in nomenclature used by authors of different life-cycle assessment case studies to self-define the methods used to hybridize life-cycle inventory.** Shown are peer-reviewed publications between 2010 and 2024. The terminology used in recent reviews [20][61][32] is used to classify different methods: *integrated*, *matrix(-augmentation)*, *path-exchange* and *tiered*. If a method has simply been designated as *hybrid* by the authors of a study, the figure assigns this to the label *not named*. Data based on [32, Appendix A], updated with more recent publications based on a systematic literature review detailed in the Supplementary Information.

We do not consider in detail different methods to avoid double-counting, even though we provide illustrations of the issue in Fig. 6. For a detailed discussion, we refer the reader to a recent review by Agez et al. [37] and the preceding work of Strømman [80]. We also do not provide numerical results from a large-scale hybridization of a process-based inventory database. Investigations of this nature are reserved for future work. We refer instead to first results by Agez et al. [36] or Jakobs et al. [24]. Similarly, no numerically founded discussion of the potential uncertainties introduced through the use of input-output data is provided. A comprehensive treatment of these issues can be obtained from Jakobs et al. [24][81].

5 Convention and Notation

5.1 Mathematical Convention

In the following, we assume that unless explicitly stated, the A-matrix of the process system $\mathbf{A}_P$ is given in the process-based life-cycle assessment convention [22, Table 1]

$$(\mathbf{A}_P)_{ij} = (+\text{production}/-\text{consumption}) \text{ of product } i \text{ by process } j \tag{1}$$

We also assume that the A-matrix of the sectoral system $\mathbf{A}_S$ is given in the input-output convention [22, Table 1]

$$(\mathbf{A}_S)_{ij} = \frac{\text{flow from node } i \text{ to node } j}{\text{output of node } j} \tag{2}$$

All derivations remain valid for input-output data, whether structured as industry×industry or product×product tables, as this choice does not influence the computed economic flows or multipliers. We discuss differences in convention and resulting differences in presentation of the hybrid matrix in detail in the Supplementary Information.

5.2 Mathematical Notation

In this manuscript, the subscript P denotes the process system, the subscript S denotes the sectoral system and the subscript H denotes the hybrid system. To help the reader distinguish between matrices and coefficients, we use a representative notation to indicate units frequently encountered when using hybrid life-cycle assessment. We use $[kg]$ to indicate physical units, $[kg(CO_2)]$ is to indicate environmental burdens and [\$] is used to indicate monetary units. Note that this is for illustrative purposes only - in reality, coefficients in the process system may have monetary units and coefficients in the sectoral system may have physical units, etc.

5.3 Diagrammatic Notation

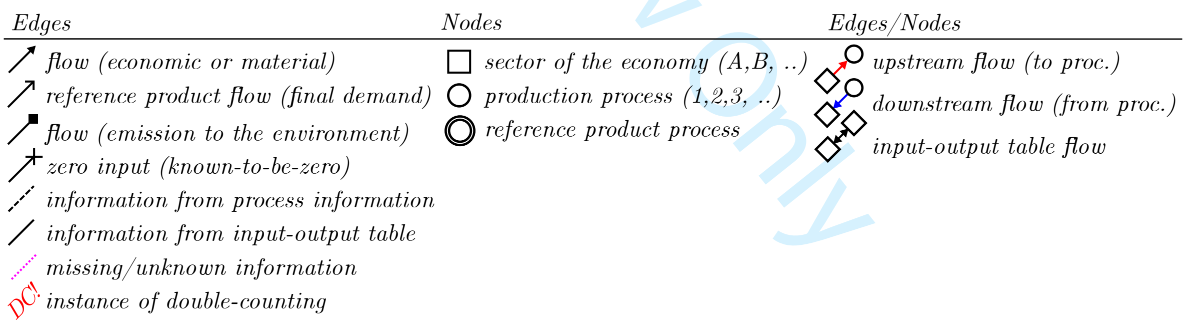

Figure 6: Legend for the novel diagrammatic notation used in this publication.

Table 2: Mathematical notation for vectors and matrices used throughout this article. For a complete derivation of the associated governing equations, refer to the supplementary information.

Index	*System*	*Description*
$i \in \mathbb{N}$, for $(1 \leq i \leq M)$	process system	production process ("activity")
$i \in \mathbb{N}$, for $(1 \leq i \leq E)$	process system	product ("technosphere flow")
$j \in \mathbb{N}$, for $(1 \leq j \leq N)$	sectoral system	economic sector
$j \in \mathbb{N}$, for $(1 \leq j \leq F)$	sectoral system	economic commodity
$k \in \mathbb{N}$, for $(1 \leq k \leq R)$	process system	biosphere flow
$l \in \mathbb{N}$, for $(1 \leq k \leq P)$	sectoral system	environmental satellites

Matrix or Vector	*Unit*	*Description*
$\mathbf{A}_P \in M_{M \times M}(\mathbb{R})$	$[kg]$	technology matrix
$\mathbf{B}_P \in M_{R \times M}(\mathbb{R})$	$[CO_2]$	environmental flow matrix
$\vec{f}_P \in M_{M \times 1}(\mathbb{R})$	$[kg]$	final demand vector
$\vec{s}_P \in M_{M \times 1}(\mathbb{R})$	$[1]$	scaling vector
$\vec{e}_P \in M_{R \times 1}(\mathbb{R})$	$[CO_2]$	environmental flow vector
$\vec{x}_P \in M_{M \times 1}(\mathbb{R})$	$[kg]$	output vector, process system
$\vec{x}_S \in M_{N \times 1}(\mathbb{R})$	$[\$]$	output vector, sectoral system
$\vec{x}_P^D \in M_{M \times 1}(\mathbb{R})$	$[kg]$	output vector, process system, induced by downstream demand from the sectoral system
$\vec{x}_P \in M_{N \times 1}(\mathbb{R})$	$[\$]$	output vector, sectoral system, induced by upstream demand from the process system
$\mathbf{C}^u \in M_{N \times M}(\mathbb{R})$	$[\$]$	upstream cut-off matrix
$\mathbf{C}^d \in M_{M \times N}(\mathbb{R})$	$[kg/\$]$	downstream cut-off matrix
$\mathbf{A}_S \in M_{N \times N}(\mathbb{R})$	$[\$/\$]$	technical coefficient matrix
$\mathbf{A}_S^* \in M_{N \times N}(\mathbb{R})$	$[\$/\$]$	technical coefficient matrix, with process flows removed
$\mathbf{B}_S \in M_{P \times N}(\mathbb{R})$	$[CO_2/\$]$	environmental satellite matrix

6 Integrated Hybrid Method

6.1 Description

Processes consume inputs from other processes. In addition, they consume inputs from different sectors of the economy. The corresponding flow into the process system is termed the *upstream flow*. Sectors of the economy consume inputs from other sectors. In addition, they consume inputs from different processes. The corresponding flow is termed the *downstream flow*. Since the sectoral system describes the average of all processes in the economy, both systems overlap, as shown in Fig. 7. This means that monetary equilibrium must be maintained in the sectoral system by subtracting process flows from corresponding sectoral flows. The *integrated hybrid method* aims to dis-aggregate some sectors with process data while preserving the overall economic balances of the input-output table.

To write this, we start from the governing equations of process-based life-cycle assessment and environmental input-output analysis, which we also derive in the Supplementary Information:

$$\vec{f}_P[\text{kg}] = \mathbf{A}_P[\text{kg}]\vec{s}_P[1] \tag{3}$$

$$\vec{f}_S[\$] = (\mathbf{I} - \mathbf{A}_S)[\$/\$]\vec{x}_S[\$] \tag{4}$$

We include the additional output of the process system $\vec{x}_P^D$, which is generated through *downstream flows* to the sectoral system and the additional output of the sectoral system $\vec{x}_S^U$, which is generated through *upstream flows* to the process system:

$$\vec{f}_P[\text{kg}] + \vec{x}_P^D[\text{kg}] = \mathbf{A}_P[\text{kg}]\vec{s}_P[1] \tag{5}$$

$$\vec{f}_S[\$] + \vec{x}_S^U[\$] = (\mathbf{I} - \mathbf{A}_S)[\$/\$]\vec{x}_S[\$] \tag{6}$$

The magnitude of these upstream and downstream flows can be described by introducing two new matrices, which Suh named the *cut-off matrices* [82, Sec. 5.1]. They are defined as:

$$\mathbf{C}^D\vec{x}_S = \vec{x}_P^D \tag{7}$$

$$\mathbf{C}^U\vec{s}_P = \vec{x}_S^U \tag{8}$$

where

$$\mathbf{C}_U[\$] \in M_{N\times M}(\mathbb{R}) \ldots \text{upstream cutoff matrix}$$

$$c_{ij}^U = \frac{\text{flow [\$] from sector } i \rightarrow \text{activity } j}{\text{scale of activity } j}$$

$$\mathbf{C}_D[\text{kg}/\$] \in M_{M\times N}(\mathbb{R}) \ldots \text{downstream cutoff matrix}$$

$$c_{ij}^D = \frac{\text{flow [kg] from activity } i \rightarrow \text{sector } j}{\text{output of sector } j \text{ [\$]}}$$

The construction of these matrices is detailed in Section 10.

> ***Nota bene!*** Some authors use different conventions for these matrices, including the signs of the matrix coefficients. A detailed discussion is provided in section *Hybrid Matrix Connections* of the supplementary information.

We must now consider the fact that our process system and our sectoral system overlap. First, we recognize that the sectoral A-matrix (technical coefficient matrix) is derived based on annual intermediate flows between sectors $z_{(X,Y)}(t = 1a)$[1]. Shown in Fig. 7 is an example system with two sectors A, B. If we now add two processes $1, 2$, a share of the annual intermediate flow $z_{(B,A)}$ is covered by the annual

[1] In the following, we omit the indicator of time, implicitly assuming that all elements refer to $t = 1a$.

intermediate flows $z^P_{(2,1)}, z^U_{(B^*,1)}, z^D_{(2,A^*)}$. We must therefore calculate the adjusted sectoral matrix $\mathbf{A}^*_S$ and the adjusted environmental burden matrix $\mathbf{B}^*_S$. In these adjusted matrices, the economic and environmental flows already accounted for in the process system are removed. For this, we introduce three conditions to maintain economic balance in the sectoral system:

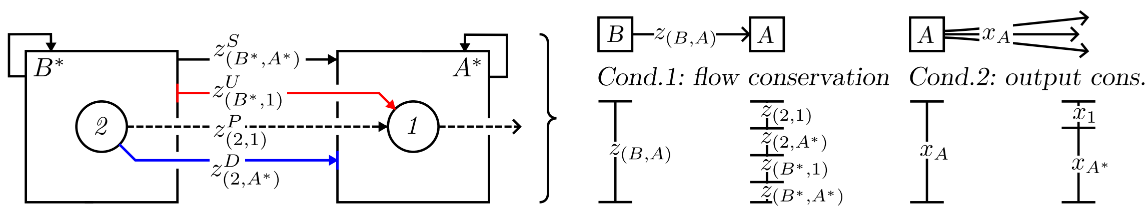

Figure 7: **Diagrammatic representation of the conditions for economic balance inherent to the *integrated hybrid* method.** Failure to remove the relevant annual flows $z^P_{(2,1)}, z^U_{(B^*,1)}, z^D_{(2,A^*)}$ from the sectoral flow $z^S_{(B,A)}$ therefore disturbs the monetary balance of the sectoral system and effectively leads to double-counting. Bottom left: System diagram featuring two manufacturing processes $(1,2)$ and two corresponding sectors of the economy (A,B). Shown are annual intermediate flows z from $B \rightarrow A$. The superscript asterisk on a sectoral index indicates that the corresponding process has been subtracted (eg. subtracting process 1 from sector A gives A^*). Bottom right: The two conditions for maintaining economic balance in the A-matrix of the sectoral system, used in both the *integrated hybrid* and the *matrix augmentation hybrid* methods. For a diagram legend, see Fig. 6.

Condition 1 is the conservation of total (annual) sectoral outputs $\vec{x}_X$. For sector B, this is:

$$x_B[\$] = x_{B^*}[\$] + x_2[\$] \tag{9}$$

Condition 2 is the conservation of (annual) intermediate flows $\vec{z}_{XY}$. For the flows $B \rightarrow A$, this is:

$$z_{(B,A)}[\$] = z_{(B^*,A^*)}[\$] + z_{(2,1)}[\$] + z_{(B^*,1)}[\$] + z_{(2,A^*)}[\$] \tag{10}$$

Condition 3 is the conservation of total (annual) environmental burdens $\vec{e}_X$. For sector B, this is:

$$e_B[CO_2] = e_{B^*} + e_2 = b_{B^*}[CO_2/\$]x_{B^*}[\$] + b_2[CO_2/\text{kg}]x_2[\text{kg}] \tag{11}$$

For a more complex system, the conditions Eq. (9)-Eq. (11) can also be expressed in matrix form:

$$\begin{aligned}
\vec{x}_S[\$] &= \vec{x}^*_S[\$] + \vec{x}_{P(S)}[\$] \\
&= \vec{x}^*_S[\$] + \mathbf{H}(\text{diag}(\vec{p})[\$/\text{kg}]\vec{x}_P[\text{kg}]) && (12)\\
\mathbf{Z}_S[\$] &= \mathbf{Z}^*_S[\$] - \mathbf{Z}_{P(S)}[\$] - \mathbf{Z}_{U(S)}[\$] - \mathbf{Z}_{D(S)}[\$] \\
&= \mathbf{Z}^*_S[\$] - \mathbf{H}(\text{diag}(\vec{p})[\$/\text{kg}]\mathbf{Z}_P[\text{kg}])\mathbf{H}^T - \mathbf{Z}_U\mathbf{H}^T[\$] - \mathbf{H}(\text{diag}(\vec{p})[\$/\text{kg}]\mathbf{Z}_D[\text{kg}]) && (13)\\
\vec{e}_S[CO_2] &= \vec{e}^*_S[CO_2] + \vec{e}_P[CO_2] \\
\mathbf{B}_S[CO_2/\$]\vec{x}_S[\$] &= \mathbf{B}^*_S[CO_2/\$]\vec{x}^*_S[\$] + \mathbf{B}_{P(S)}[CO_2/\text{kg}]\vec{x}_{P(S)}[\text{kg}] \\
&= \mathbf{B}^*_S[CO_2/\$]\vec{x}^*_S[\$] + \mathbf{B}_{P(S)}[CO_2/\text{kg}]\mathbf{H}^T\mathbf{H}\vec{x}_P[\text{kg}] && (14)
\end{aligned}$$

where the corrected sectoral A-matrix (technical coefficient matrix) $\mathbf{A}^*_S$ can be quickly derived from the corresponding flow matrix $\mathbf{Z}^*_S$ according to

$$\mathbf{A}^*_S = \mathbf{Z}^*_S \ \text{diag}(\vec{x}^*_S)^{-1} \tag{15}$$

The intermediate flow matrices can be derived by using the annual output vector of the process system

$$\mathbf{Z}_P = \mathbf{A}_P\text{diag}(\vec{x}_P) - \text{diag}(\vec{x}_P) \tag{16}$$

$$\mathbf{Z}_U = \mathbf{C}^U\text{diag}(\vec{x}_P) \tag{17}$$

$$\mathbf{Z}_D(t = 1a) = \mathbf{C}^D\text{diag}(\vec{x}_S) \tag{18}$$

Notably, the discussion of these important balancing conditions is absent in recent reviews. While Crawford et al. mention that *"The [$\mathbf{A}_S$] system is (...) rebalanced by subtracting the upstream and downstream cut-offs."* [32, Sec. 5.4], the equation they later provide does not include this rebalancing step [32, (B.16)]. Islam et al. similarly mention the rebalancing briefly [61, Sec. 3.3.3], but do not provide equations. We find this to be indicative of the application of this method by practitioners more broadly. The majority of authors using this method have neglected to apply this correction. Publications that explicitly cite Suh and use the term *integrated hybrid* for their method, but do not remove flows include: [83, (3)], [84, (1)], [27, (1)], [85, (1)]. Wiedmann et al. [86] raises the hopes of the reader by using the notation $\mathbf{A}^*$ in their *integrated hybrid* governing equation. Unfortunately, the supplementary information of their publication reveals that process flows have not in fact been subtracted in their matrix.

Using the definition of the cut-off matrices Eq. (7)-Eq. (8) and the corrected sectoral matrices $\mathbf{A}_S^*, \mathbf{B}_S^*$ derived with the help of the rebalancing conditions Eq. (12)-Eq. (14), we can now write the governing equation of the *interated hybrid* method:

$$\vec{f}_P[\text{kg}] = \mathbf{A}_P[\text{kg}]\vec{s}_P[1] - \mathbf{C}^D[\text{kg}/\$]\vec{x}_S[\$] \tag{19}$$

$$\vec{f}_S[\$] = (\mathbf{I} - \mathbf{A}^*{}_S)[\$/\$]\vec{x}_S[\$] - \mathbf{C}^U[\text{kg}]\vec{s}_P[1] \tag{20}$$

Nota bene! The final demand vector $\vec{f}$ is an exogenous variable. It represents the choice of the functional unit of the life-cycle assessment [6, (2.9)]. If the output of an activity from the process-based system is selected as the functional unit of the analysis, we can write $\vec{f}_P \neq 0 \wedge \vec{f}_S = 0$. We can therefore write Eq. (20) in the form most frequently used in literature:

$$\vec{f}_S = 0 = (\mathbf{I} - \mathbf{A}_S)\vec{x}_S - \mathbf{C}^u\vec{s}_P \tag{21}$$

Rewriting Eq. (19)-Eq. (20)

$$\vec{f}_P[\text{kg}] = \mathbf{A}_P[\text{kg}]\vec{s}_P[1] - \mathbf{C}^D[\text{kg}/\$]\vec{x}_S[\$] \tag{22}$$

$$\vec{0} = -\mathbf{C}^U[\text{kg}]\vec{s}_P[1] + (\mathbf{I} - \mathbf{A}^*{}_S)[\$/\$]\vec{x}_S[\$] \tag{23}$$

We find that we can combine this into a single hybrid matrix and formulate the governing equation of hybrid life-cycle assessment as

$$\vec{e}_{H(int)} = \begin{pmatrix}\mathbf{B}_P[CO_2/\text{kg}] & \mathbf{B}_S^*[CO_2/\$]\end{pmatrix} \begin{pmatrix}\mathbf{A}_P[\text{kg}] & -\mathbf{C}_D[\text{kg}/\$] \\ -\mathbf{C}_U[\$] & \mathbf{I} - \mathbf{A}_S^*[\$]\end{pmatrix}^{-1} \begin{pmatrix}\vec{f}_P \\ 0\end{pmatrix} \tag{24}$$

Note that some authors have used a different convention for the inverse matrix of Eq. (24). For an helpful discussion of differences in the notation, compare [22].

Nota bene! Only if the number of biosphere flows is equal to the number of environmental satellites ($P = R$), as for instance in Crawford [32, Sec. B.3], we can write

$$\vec{e}_H = ((e_S)_1 + (e_P)_1, (e_S)_2 + (e_P)_2, \ldots, (e_S)_{P=R} + (e_P)_{R=P})^T = [(P = R) \times 1] \tag{25}$$

instead of

$$\vec{e}_H = ((e_S)_1, (e_S)_2, \ldots, (e_S)_P; (e_P)_1, (e_P)_2, \ldots, (e_P)_R)^T = [(P + R) \times 1] \tag{26}$$

and we can define

$$\vec{e}_H = \begin{pmatrix} \mathbf{B}_P & \mathbf{B}_S \end{pmatrix} \begin{pmatrix} \mathbf{A}_P & 0 \\ 0 & \mathbf{I} - \mathbf{A}_S \end{pmatrix}^{-1} \begin{pmatrix} \vec{f}_P \\ \vec{f}_S \end{pmatrix} \tag{27}$$

$$= \begin{pmatrix} \mathbf{B}_P & 0 \\ 0 & \mathbf{B}_S \end{pmatrix} \begin{pmatrix} \mathbf{A}_P & 0 \\ 0 & \mathbf{I} - \mathbf{A}_S \end{pmatrix}^{-1} \begin{pmatrix} \vec{f}_P \\ \vec{f}_S \end{pmatrix} \tag{28}$$

The *integrated hybrid* methods is essentially dis-aggregating the input-output table based on data from the process inventory. As we have shown in Section 4, the upstream and downstream matrices may be augmented by inferring data from the inherent structure of the input-output table. A mathematical description is provided in Section 10.2.

7 Matrix Augmentation Hybrid Method

7.1 Description

The fundamental assumption and goal is equivalent to the *integrated hybrid* method [62], described in Section 6.1. The only difference is in the units of the input data. The original publication of the *integrated hybrid* method by Suh provided the process data in physical units [82], while Joshi in the *matrix augmentation* provided the process data in monetary units [62, "Model II"]. Of course, a conversion between the two can be performed using the price vector [87].

Here, we use monetary data to show the equivalence of the two methods. We can then adapt the main equation of environmentally-extended input-output analysis, which we already introduced in Eq. (4) and derive in the Supplementary Information:

$$\vec{x}_S[\$] = (\mathbf{I} - \mathbf{A}_S)^{-1}[\$/\$]\vec{f}_S[\$] \tag{29}$$

by adding processes as new sectors to the sectoral A-matrix (technical coefficient matrix):

$$\mathbf{A}_{H(aug)} = \begin{pmatrix} \mathbf{A}_P[\$/\$] & \mathbf{C}_D[\$/\$] \\ \mathbf{C}_U[\$/\$] & \mathbf{A}_S^*[\$/\$] \end{pmatrix} \tag{30}$$

Since all elements of the matrix are dimensionless, based on economic units, we hereafter omit explicit units from the equations. Following Eq. (30), the new basis for our vector space is

$$B_{(M,N)} = \{\vec{b}_1, \ldots, \vec{b}_M;\ \vec{b}_1, \ldots, \vec{b}_N\} \tag{31}$$

Nota bene! This choice of vector space is slightly different from the original publication of Joshi. There, the process and sectoral basis vector were swapped:

$$B^{Joshi}_{(N,M)} = \{\vec{b}_1, \ldots, \vec{b}_N;\ \vec{b}_1, \ldots, \vec{b}_M\} \tag{32}$$

Our choice makes the comparison and proof of equivalence with the *integrated hybrid* method trivial.

Just like in the *integrated hybrid* method in Section 6, we consider the fact that our process system and our sectoral system overlap, as illustrated in Fig. 7. We must therefore calculate the adjusted sectoral matrix $\mathbf{A}_S^*$ and the adjusted environmental burden matrix $\mathbf{B}_S^*$. For this, we use the same conditions we already introduced in Eq. (9)-Eq. (11) for the *integrated hybrid* method to maintain economic balance in the sectoral system.

For a general system, the conditions can be expressed in matrix form:

$$\begin{aligned} \vec{x}_S &= \vec{x}_S^* + \vec{x}_{P(S)} \\ &= \vec{x}_S^* + \mathbf{H}\vec{x}_P && (33) \\ \mathbf{Z}_S &= \mathbf{Z}_S^* - \mathbf{Z}_{P(S)} - \mathbf{Z}_{U(S)} - \mathbf{Z}_{D(S)} \\ &= \mathbf{Z}_S^* - \mathbf{H}\mathbf{Z}_P\mathbf{H}^T - \mathbf{Z}_U\mathbf{H}^T - \mathbf{H}\mathbf{Z}_D && (34) \\ \mathbf{B}_S\vec{x}_S &= \mathbf{B}_S^*\vec{x}_S^* + \mathbf{B}_{P(S)}\vec{x}_{P(S)} \\ &= \mathbf{B}_S^*\vec{x}_S^* + \mathbf{B}_P\mathbf{H}^T\mathbf{H}\vec{x}_P && (35) \end{aligned}$$

The intermediate flow matrices $\mathbf{Z}_S^*, \mathbf{Z}_P, \mathbf{Z}_U, \mathbf{Z}_D$ are defined through the relation

$$\mathbf{A}\text{diag}(\vec{x}) = \mathbf{Z} \tag{36}$$

We finally introduce a final demand vector $\vec{f}^H$ in the new basis of our hybrid vector space Eq. (32). Using the above matrices, we then arrive at the governing equation of the *matrix augmentation* method.

$$\vec{x}_{H(aug)} = (\mathbf{I} - \mathbf{A}^*_{H(aug)})^{-1} \vec{f}_H \tag{37}$$

$$\vec{e}_{H(aug)} = \mathbf{B}_{H(aug)} (\mathbf{I} - \mathbf{A}^*_{H(aug)})^{-1} \vec{f}_{H(aug)} \tag{38}$$

7.2 Mathematical Equivalence with Integrated Hybrid Method

Despite previous reports in literature [32][61], the *matrix-augmentation hybrid* method does not constitute a distinct mathematical model. This was first observed by Peters and Hertwich without proof by explicitly writing out the corresponding matrices [88, Sec. 2.2.1]. A partial proof, albeit needlessly involved through the use of block-matrix inversion formulas, was provided by by Cruze [26, Sec. 6.2, Sec. A.1]. However, neither authors showed that the removal of process flows from the sectoral system is equivalent in both methods. Here we show more succinctly that the method is equivalent to the *integrated hybrid* method.

Starting from the governing equation of the *matrix augmentation* method in Eq. (38) and the governing equation of the *integrated hybrid* method in Eq. (24) we need to show that

$$\vec{e}_{H(aug)} = \begin{pmatrix} \mathbf{B}_P[\mathrm{CO_2}/\$] & \mathbf{B}^*_S[\mathrm{CO_2}/\$] \end{pmatrix} \begin{pmatrix} \mathbf{1} - \mathbf{A}_P[\$/\$] & -\mathbf{C}_D[\$/\$] \\ -\mathbf{C}_U[\$/\$] & \mathbf{I} - \mathbf{A}^*_S[\$/\$] \end{pmatrix}^{-1} \begin{pmatrix} \vec{f}_P[\$] \\ 0 \end{pmatrix} \tag{39}$$

$$= \vec{e}_{H(int)} = \begin{pmatrix} \mathbf{B}_P[\mathrm{CO_2}/\mathrm{kg}] & \mathbf{B}^*_S[\mathrm{CO_2}/\$] \end{pmatrix} \begin{pmatrix} \mathbf{A}_P[\mathrm{kg}] & -\mathbf{C}_D[\mathrm{kg}/\$] \\ -\mathbf{C}_U[\$] & \mathbf{I} - \mathbf{A}^*_S[\$/\$] \end{pmatrix}^{-1} \begin{pmatrix} \vec{f}_P[\mathrm{kg}] \\ 0 \end{pmatrix} \tag{40}$$

Having already taken care in the presentation of the mathematical notation and convention of these methods in Section 6-Section 7, we can see that they are equivalent, differing only in the units of $\mathbf{A}_P, \mathbf{B}_P, \mathbf{C}_U, \mathbf{C}_D$.

To complete the proof, we must show that the removal of process flows from the sectoral system is equivalent in both methods. Unfortunately, the authors used different approaches and notation to describe the computation of these matrices. To simplify the comparison, we have already introduced the corrected matrices using our unified notation and illustrated the motivation in Fig. 7.

We need to show that the equations of Suh [82, adapted from (A6)-(A7)] in Eq. (12)-Eq. (14)

$$\vec{x}_S[\$] = \vec{x}^*_S[\$] + \mathbf{H}(\mathrm{diag}(\vec{p})[\$/\mathrm{kg}]\vec{x}_P[\mathrm{kg}])$$

$$\mathbf{Z}_S[\$] = \mathbf{Z}^*_S[\$] - \mathbf{H}(\mathrm{diag}(\vec{p})[\$/\mathrm{kg}]\mathbf{Z}_P[\mathrm{kg}])\mathbf{H}^T - \mathbf{Z}_U\mathbf{H}^T[\$] - \mathbf{H}(\mathrm{diag}(\vec{p})[\$/\mathrm{kg}]\mathbf{Z}_D[\mathrm{kg}])$$

$$\mathbf{B}_S[\mathrm{CO_2}/\$]\vec{x}_S[\$] = \mathbf{B}^*_S[\mathrm{CO_2}/\$]\vec{x}^*_S[\$] + \mathbf{B}_{P(S)}[\mathrm{CO_2}/\mathrm{kg}]\mathbf{H}^T\mathbf{H}\vec{x}_P[\mathrm{kg}]$$

are equivalent to the equations of Joshi [62, adapted from (6)-(8)] in Eq. (33)-Eq. (35)

$$\vec{x}_S[\$] = \vec{x}^*_S[\$] + \mathbf{H}\vec{x}_P[\$]$$

$$\mathbf{Z}_S[\$] = \mathbf{Z}^*_S[\$] - \mathbf{H}\mathbf{Z}_P[\$]\mathbf{H}^T - \mathbf{Z}_U[\$]\mathbf{H}^T - \mathbf{H}\mathbf{Z}_D[\$]$$

$$\mathbf{B}_S[\mathrm{CO_2}]\vec{x}_S[\$] = \mathbf{B}^*_S[\mathrm{CO_2}]\vec{x}^*_S[\$] + \mathbf{B}_P[\mathrm{CO_2}]\mathbf{H}^T\mathbf{H}\vec{x}_P[\$]$$

Just like above, we can see that the equations differ only in the units of the process system matrices. We find, therefore, that the methods are equivalent in both the mathematical structure and the correction of the sectoral system.

8 Tiered Hybrid Method

8.1 Description

Processes consume inputs from other processes. In addition, they consume inputs from different sectors of the economy. The final demand on the process system $\vec{f}_P$ therefore induces "upstream" output $\vec{u}_S$ of the sectors of the economy, with associated additional environmental burdens $\vec{b}_S$. Contrary to the *integrated hybrid method*, it explicitly does not aim to provide a balanced representation of the entire economy. This is because the foreground outputs are assumed to not be sufficiently important to alter the upstream impacts of the sectors used as complements. We can write this as:

$$\vec{e}_H = \vec{e}_P + \vec{e}_S \tag{41}$$

Using the governing equations of process-based life-cycle assessment in monetary units from Eq. (3) and environmentally-extended input-output analysis from Eq. (4)

$$\vec{e}_S[CO_2] = \mathbf{B}_S[CO_2/\$]\vec{x}_S[\$] = \mathbf{B}_S[CO_2/\$](\mathbf{I} - \mathbf{A}_S)^{-1}[\$/\$]\vec{u}_S[\$] \tag{42}$$

$$\vec{e}_P[CO_2] = \mathbf{B}_P[CO_2]\vec{x}_P[kg] = \mathbf{B}_P[[CO_2]/kg]\mathbf{A}_P^{-1}[kg]\vec{f}_P[1] \tag{43}$$

we can write the governing equation of the tiered hybrid method:

$$\vec{e}_{H(tiered)}[CO_2] = \vec{e}_S + \vec{e}_P = \mathbf{B}_S[CO_2/\$]\mathbf{A}_S^{-1}[\$/\$]\vec{u}_S[\$] + \mathbf{B}_P[CO_2](\mathbf{I} - \mathbf{A}_P)^{-1}[kg]\vec{f}_P[1] \tag{44}$$

$$\vec{e}_{H(tiered)} = \begin{pmatrix} \mathbf{B}_P[CO_2/\$] & \mathbf{B}_S[CO_2] \end{pmatrix} \begin{pmatrix} \mathbf{A}_P[kg] & 0 \\ 0 & \mathbf{I} - \mathbf{A}_S[\$/\$] \end{pmatrix}^{-1} \begin{pmatrix} \vec{f}_P[1] \\ \vec{u}_S[\$] \end{pmatrix} \tag{45}$$

The *tiered hybrid* methods is essentially expanding the system boundary of the process-based inventory by adding upstream flows from the input-output table. As we have shown in Section 4, the upstream matrix may be augmented by inferring data from the inherent structure of the input-output table. A mathematical description is provided in Section 10.2.

8.2 Relation to Integrated Hybrid Method

If we place final demand *only* on the process system through $\vec{f}_P$, the additional upstream demand on the sectoral system $\vec{u}_S$ is induced by the process system alone. This means that we can again introduce an *upstream cut-off* matrix describing the amount of flow from sectors to processes. Using this matrix, the additional upstream demand on the sectoral system $\vec{u}_S$ induced by the process system can be expressed as

$$\mathbf{C}_U[\$/kg]\vec{x}_P[kg] = \vec{u}_S[\$] \tag{46}$$

where

$$\mathbf{C}_U[\$/kg] \in M_{N\times M}(\mathbb{R}) \ldots \text{upstream cutoff matrix} \tag{47}$$

$$c_{ij}^U = \frac{\text{flow [\$] from sector } i \rightarrow \text{activity } j}{x_j[kg]} \tag{48}$$

With this, the governing equation of the tiered hybrid matrix method takes the form used in literature more recently:

$$\vec{e}_{H(tiered)} = \begin{pmatrix} \mathbf{B}_P & \mathbf{B}_S \end{pmatrix} \begin{pmatrix} \mathbf{A}_P & 0 \\ \mathbf{C}_U^{uncorr} & \mathbf{I} - \mathbf{A}_S \end{pmatrix}^{-1} \begin{pmatrix} \vec{f}_P \\ 0 \end{pmatrix} \tag{49}$$

As we can see, the *tiered hybrid* method of Eq. (49) can be expressed in the same matrix framework as the *integrated hybrid* method of Eq. (24). This was previously observed by Peters and Hertwich [88, Sec. 2], Heijungs et al. [6, Sec. 5.4.1] and Suh et al. [82, Sec. 2.4.1].

9 Path-Exchange Hybrid Method

We demonstrate elsewhere [89] that the algorithm of the *path-exchange method* can always be expressed through a hybrid matrix, where the downstream matrix is zero and the flows of the upstream matrix are inferred from the sectoral system as described in Fig. 3, with binary double-counting correction applied. It is therefore equivalent to the *tiered hybrid method*. The full mathematical proof of equivalence is rather involved and can therefore not be integrated in the present work.

Note to Reviewers: We provide the latest version of this manuscript as a supplementary review file for your consideration.

10 Building the Upstream/Downstream Matrices

In Eq. (7)-Eq. (8), we introduced the cut-off matrices, describing the flow from the sectoral system to the process sytem and vice versa. Here, we summarize how they can be constructed, either *manually* by adding flow data, or by *inferring* flow data from the sectoral system itself.

10.1 Practitioners adding Flow Data

Upstream and downstream matrices can be compiled by the practitioner based on original research. Suh et al. described this process briefly: *"The upstream cut-off by processes matrix is derived by dividing the total bill of goods for the inputs that are not covered by a processes in a process-based system during the period of steady-state approximation by the total unit operation time of each process."* [82, Sec. 5.1]. In contrast, *"The downstream cut-off by functional flow matrix is derived by dividing the annual sales of functional flow (in physical units that are relevant to each functional flow) by the production of each total commodity."* [82, Sec. 5.1].

10.2 Inferring Flow Data

On the other hand, building the upstream matrix by inferring flows from the input-output table, as originally proposed by Strømman [58], is best understood with the help of a diagrammatic representation of the involved monetary flows, shown in Fig. 8. Using this elegant inference method *"an inventory with no cutoff with respect to costs can be obtained (...) increasing the consistency and robustness of eco-efficiency results."* [58].

As we can see from the example system in Fig. 8, sectors B, C, D and E all provide input to sector A. Similarly, processes 2 and 3 provide input to process 1. We also know explicitly that some input of sector B is required by process 1. Unfortunately, the cost of inputs from 2, 3 and B together with the known value added of the process does not sum to the basic price of the product produced by process 1. This amount of *"missing flow"* can now be inferred. First, all processes are matched to economic sectors. For instance, process 1 is contained in sector A. Based on this matching, additional flows are inferred from the inherent structure of the technical coefficient matrix of the input-output table. The matrix expression for this process can be written as

$$\mathbf{C}_{U(scaled)} = \mathbf{C}_{U(K)} + \mathbf{C}_{U(unscaled)}\text{diag}(\vec{\gamma}) \tag{50}$$

For an in-depth derivation of the original equations of Strømman [58], we refer to the Supplementary Information.

A similar approach can be employed to derive information on how to feed output of processes back into the sectoral system. To our knowledge, this has not previously been described. This inference method is again best understood with the help of a diagrammatic representation of the involved monetary flows, shown in Fig. 9.

First, information on *known downstream flows* can be collected in the manually compiled downstream cut-off matrix $\mathbf{C}_{D(K)}$. Then, the remainder of the residual of different processes can be distributed to sectors of the economy in same ratio that the sector containing each process provides its output to other sectors.

$$\begin{aligned}\mathbf{Z}_D =& \mathbf{Z}_{D(K)} + \qquad (51)\\ & [\mathbf{\Theta} \stackrel{?}{=} 0] \odot [\mathbf{Z_{D(K)}} \stackrel{?}{=} \mathbf{0}] \odot \text{diag}(\vec{x}^P - \mathbf{Z}_P \cdot \vec{1} - \mathbf{Z}_{D(K)} \cdot \vec{1}) \cdot \qquad (52)\\ & \text{diag}(\mathbf{H}^T \vec{x}^s - \mathbf{Z}_{D(K)} \cdot \vec{1} - \mathbf{\Theta} \odot \mathbf{H}^T \mathbf{Z}_S)^{-1} \cdot \mathbf{H}^T \mathbf{Z}_S\end{aligned}$$

Here, the square brackets refer to the Iverson bracket, which allows for a concise representation of

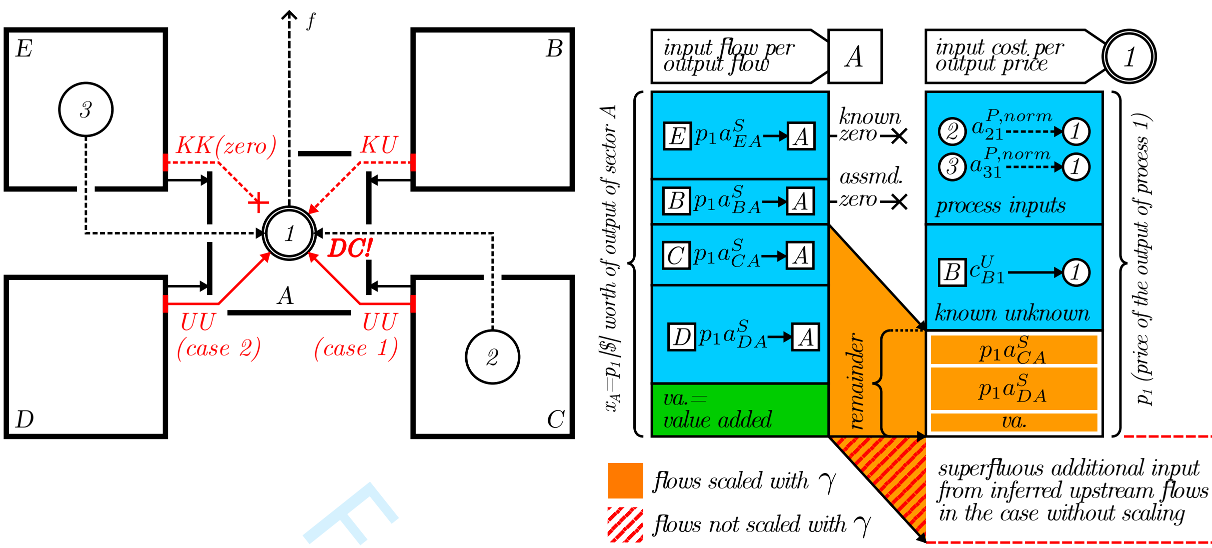

Figure 8: **Diagrammatic representation of the logic underlying the construction of the upstream cut-off matrix in Eq. (50).** *Left:* An example system illustrating the different possible kinds of inputs from an economic sector to a production process, which are introduced in Table 1. Abbreviations: KK - known known; UU (case 1) - unknown unknown, with process flow from sector (included to show an instance of double counting); UU (case 2) - unknown unknown, without any process flow from sector; KU - known unknown; KK (zero) - known-to-be-zero, describing a case where input from a sector is known not to be required because a corresponding process covers all input of a certain kind; DC! - double counting. *Right:* A schematic illustration of the inference method proposed by Strømman in 2008 [58]. Flow $E \to 1$ is "known to be zero", because the flow $3 \to 1$ covers all relevant inputs of this kind. Flow $B \to 1$ is added by the practitioner as a proxy. Flows $C \to 1$ and $D \to 1$ are inferred from the structure of the sectoral system and scaled down to the remainder of the input cost per output price of process 1. If the upstream inputs were not scaled down, input costs could exceed the output price. For a legend of the diagrammatic notation used, compare Fig. 6. For a description of the different flow types, compare Table 1.

logical conditions. It is defined as

$$[x \stackrel{?}{=} 0] = \begin{cases} 1 & \text{if } x = 0 \\ 0 & \text{else} \end{cases} \tag{53}$$

For further details on the Iverson bracket and an in-depth derivation of this equation, we refer to the Supplementary Information. Using this inference method, outputs of processes can be distributed to sectors of the economy, based on the inherent structure of the sectoral system.

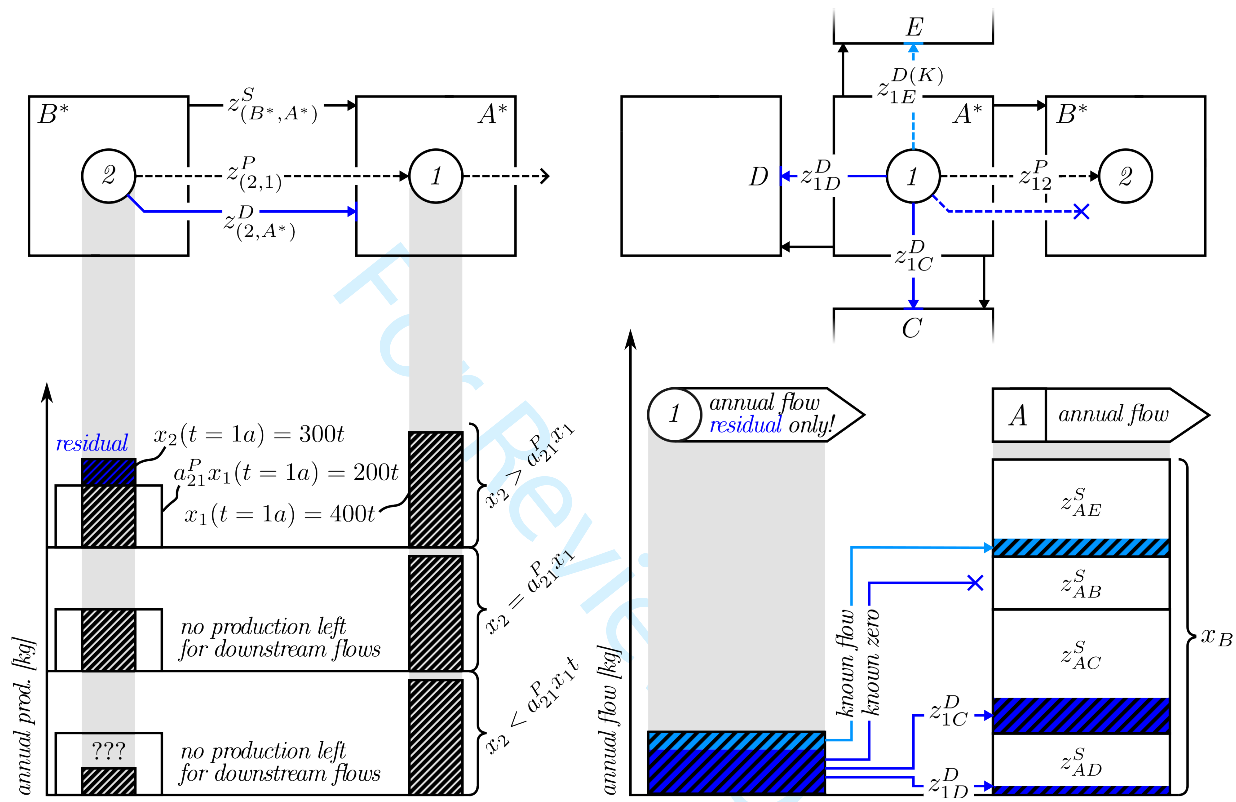

Figure 9: **Diagrammatic representation of the logic underlying the construction of the downstream matrix in Eq. (7).** *Left:* An example system illustrating the different possible scenarios of annual production volume data for processes. For example, the bottom chart shows that production data of process 2 indicates smaller annual production than would be required according to data on the flow a^P_{21} and the annual production of process 1. Of the three scenarios shown, only in the top case where the known annual production x_2 of process 2 exceeds the total annual production requirement $a^P_{21}x_1$ on process 2 can a residual be distributed to the sectoral system through downstream flows. *Right:* An example system illustrating the process by which "market shares" of the input-output table can be used to distribute a residual to different sectors. The part of the a residual for which the practitioner has no downstream flow information is distributed to sectors in the same ratio that sector $\boxed{B}$ provides its output to sectors.

11 Discussion

As we have shown in Section 3.1, hybrid life-cycle assessment offers the enticing promise of reducing the truncation error inherent to purely process-based life-cycle assessment. This would improve the accuracy of assessment results and strengthen the empirical basis of the method as an environmental decision support tool. Unfortunately, we have shown further in Section 3.4 that significant confusion still surrounds the nomenclature and distinction between different hybrid methods. We have therefore first shown in Section 4 how hybrid life-cycle assessment can be used to treat *known unknowns*, *unknown unknowns* or a combination of both with respect to the process inventory. A detailed mathematical and diagrammatic treatment is provided in Section 10. Finally, in Section 6-Section 9, we have then shown by using a consistent mathematical framework that presently only two distinct methods for hybrid life-cycle assessment exist: the *tiered hybrid* and the *integrated hybrid* method.

11.1 Summary and Classification of Methods

As we have shown in Section 6-Section 9, only the tiered hybrid method and the integrated hybrid method are mathematically distinct. They differ in the direction of flows between the sectoral system and the process system. The tiered hybrid method has only *uni-directional* flows into the process system, while the integrated hybrid method has *bi-directional* flows between the process system and the sectoral system. This necessitates a dis-aggregation of the sectoral system in order to maintain economic balance.

On the basis of the mathematical derivation of each method in Section 6-Section 9 and the Rumsfeldian classification of flows introduced in Section 4 and Table 1, we can provide a classification scheme for the methods associated with specific nomenclature historically chosen by practitioners in Table 3.

Table 3: Complete classification of the historical description of methods for hybrid life-cycle assessment, as used in recent reviews [20][61][32] according to the presence of upstream/downstream flows and the Rumsfeld matrix of Table 1. This table augments the diagrammatic representation of different hybridization methods in Fig. 10 and the histogram of nomenclature used by practitioners in Fig. 5. Abbreviations: class. - classification

historical terminology (cf. Fig. 5)	*flow data class.*	*Rumsfeld class.*	*examples*
tiered hybrid (manually added flows)	$\mathbf{C}^D = 0 \wedge \mathbf{C}^U \neq 0$	*KU*	[90][91][23]
tiered hybrid (inferred flows)	$\mathbf{C}^D = 0 \wedge \mathbf{C}^U \neq 0$	$KU \wedge UU$	[30]
path-exchange hybrid	$\mathbf{C}^D = 0 \wedge \mathbf{C}^U \neq 0$	*UU*	[92][93][94]
integrated hybrid	$\mathbf{C}^D \neq 0 \wedge \mathbf{C}^U \neq 0$	*KU*	[82][84][85]

From our derivation in Section 6-Section 9, we recognize that both distinct hybrid life-cycle assessment methods (tiered and integrated) can deal with *known unknowns* and *unknown unknowns*, and are only distinguished in principle by their compilation (or not) of their downstream cutoff matrix.

If, for instance, the *tiered hybrid* method is used in a case where both *known unknowns and unknown unknowns* are considered, part of the uncertainty inherent to the method is transformed from an *inaccessible* to an *accessible* state. While additional uncertainties may be introduced through price or geographic variance, these can then be treated mathematically [24][81].

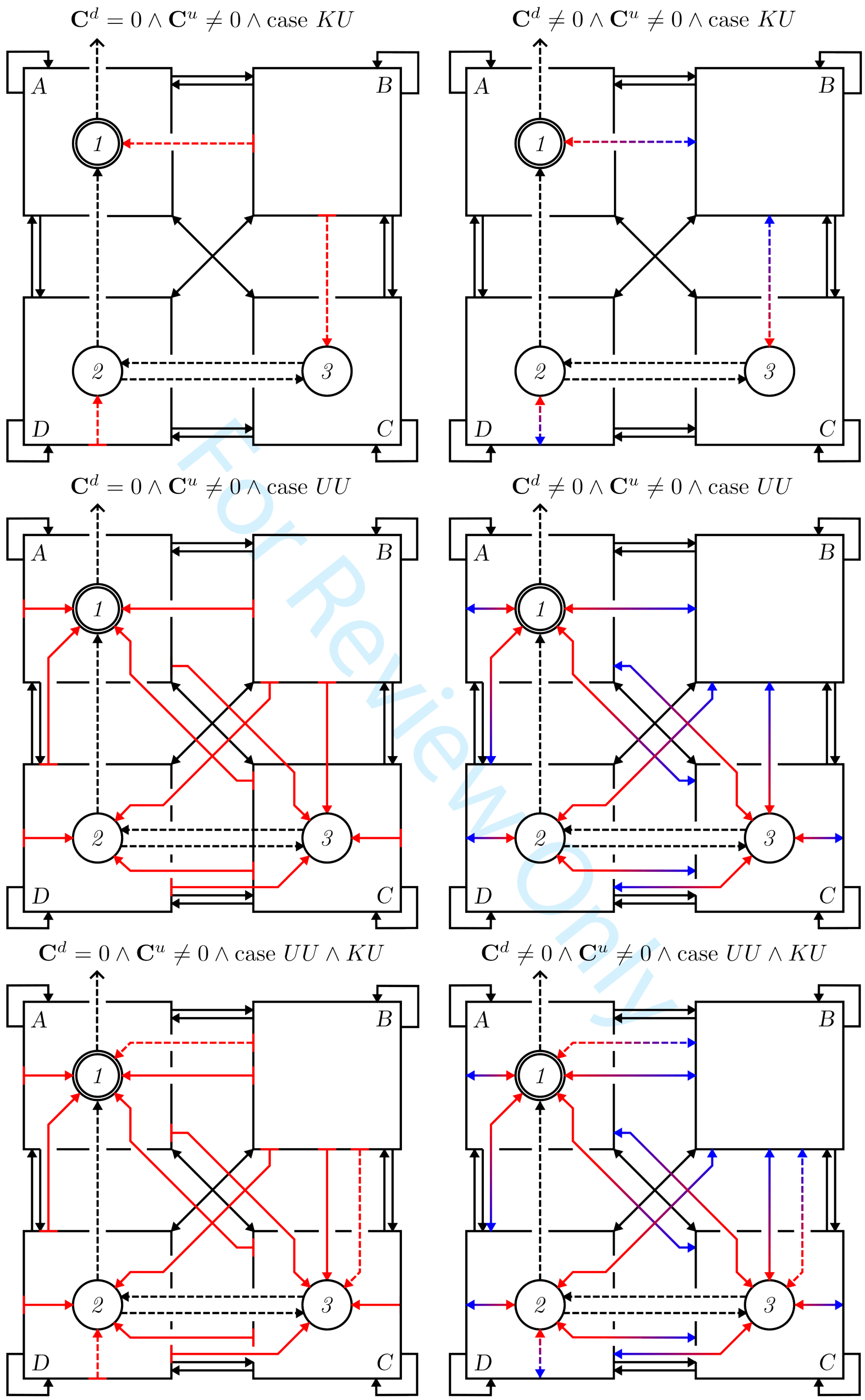

Figure 10: Example systems to illustrate the different methods (Section 6-Section 9) and scopes (Section 3.3 and Table 1). For a legend of the diagrammatic notation used, compare Fig. 6.

11.2 Recommendation of Methods

The *tiered hybrid* method constitutes a useful tool for expanding the system boundary of process-based life-cycle assessment for more accurate and comprehensive results. On the other hand, the *integrated hybrid* method effectively dis-aggregates the sectoral system based on information from the process system. It is therefore best suited for investigating the effects of technological choices on the environmental impact across the economy at large. Especially in the latter case, a double-counting correction technique will be required to ensure emissions are not over-estimated. As we note in Section 4, a coverage of these is beyond the scope of this publication. In both cases, flows can be added manually from data compiled by the practitioners, or inferred from the inherent structure of the sectoral system. Notably, the data requirements between methods differ significantly:

The *integrated hybrid* method can only be applied if (annual) production volumes for the output of production processes can be estimated. This is a result of the need for maintaining economic balance in the sectoral system, which we describe mathematically in Section 6. As described in Section 6, this method requires the matrix of technical coefficients, derived from the input-output table, to be corrected in order to maintain economic balance. The contents of both upstream and downstream matrices mus be compiled by hand. As previously noted in Section 3.4, authors have rightly cautioned that this can make the method potentially very resource intensive, while also providing the most accurate assessment result.

If no such sales records are available, the *tiered hybrid* method can be applied. If the practitioner compiles the matrix of upstream flows exclusively by hand, he is using the *tiered hybrid* method to account for *known unknowns*. As we describe in Section 3.3.1, this is the way in which hybrid life-cycle assessment was originally used. If instead he augments compiles the matrix by additionally inferring flows from the input-output system, he is using the *tiered hybrid* method to account for both *known unknowns and unknown unknowns*.

12 Conclusion and Outlook

We believe that our publication provides some much needed clarity one of the core issues of hybrid life-cycle assessment. We hope that the primary beneficiary of our work will be the research software development community. Developers might extend the functionality of existing solutions, such as the `pylcaio` software package of Agez [95] to build a robust ecosystem of open-source software for hybrid life-cycle assessment.

Once software solutions are available to practitioners, we believe that the most immediately beneficial application of hybrid life-cycle assessment could be the iterative improvement of process-based inventories, in line with what has been already proposed as *"iterative HLCA"* [96][97]. This also aligns most closely with how the application of hybrid methods had been envisioned when different methods were first proposed. For instance, Lenzen originally only intended for the *path-exchange* method to inform the life-cycle assessment process [98, Sec.2]. Practically, this means that experts at *Ecoinvent* or *GaBi* could use hybrid methods to prioritize future data gathering efforts in their database. If large amounts of upstream flows are found to be present in a production process, it likely warrants further investigation.

We further expect that important questions regarding the utility of hybrid life-cycle assessment in general [99][100] or in the context of a trade-off between accuracy and precision [29] will be re-invigorated. After all, large-scale quantitative comparisons between the assessment results obtained from using different hybrid methods have only been performed very recently [101][102][103][104]. Using the consistent description we have provided, future comparisons are likely to yield much more meaningful results than the superficial comparisons sometimes performed on illustrative systems of less than ten sectors or processes [61][105][100].

Finally, we hope that our formalism will foster greater confidence in hybrid life-cycle assessment as a reliable environmental decision-making method for synthesizing diverse data sources. To make the best possible use of process inventories and input-output data, we must ensure clarity in the data fusion process to eliminate inconsistencies, unacknowledged assumptions, and methodological confusion. We invite researchers and practitioners to embrace our formalism, operationalize it through collaborative open-source tool development, and build a robust community-driven ecosystem for hybrid life-cycle assessment.

13 Funding

This work has been supported by the Swiss Innovation Agency Innosuisse in the context of the WISER flagship project (PFFS-21-72). In addition, Michael P. Weinold gratefully acknowledges the support of the Swiss Study Foundation.

14 Acknowledgements

The authors would like to thank Dr. Arthur Jakobs (Center for Energy and Environmental Sciences, Paul Scherrer Institute) for helpful discussions related to all aspects of hybrid life-cycle assessment. The authors would also like to thank Dr. Maxime Agez (International Reference Center for Life Cycle Assessment and Sustainable Transition (CIRAIG)) for helpful discussions related to his `pylcaio` package as well as various aspects of hybrid life-cycle assessment. Finally, the main author would like to thank Dr. Chris Mutel (Départ de Sentier Association) for originally sparking his interest in the methodology underlying hybrid life-cycle assessment.

15 Data Availability Statement

Data sharing not applicable to this article as no datasets were generated or analyzed during the current study.

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Journal of
Industrial Ecology

Critical Analysis of the Path-Exchange Method for Hybrid Life-Cycle Assessment

Journal:	*Journal of Industrial Ecology*
Manuscript ID	24-JIE-8428.R1
Wiley - Manuscript type:	Methods Article
Date Submitted by the Author:	20-Mar-2025
Complete List of Authors:	Weinold, Michael; Paul Scherrer Institut PSI, Laboratory for Energy Systems Analysis PSI Centers for Nuclear Engineering & Sciences and Energy & Environmental Sciences; ETH Zurich, Chair of Energy Systems Analysis Institute of Energy and Process Engineering Department of Mechanical and Process Engineering Majeau-Bettez, Guillaume; Ecole Polytechnique de Montreal, CIRAIG, Chemical Engineering Department
Keywords:	hybrid life-cycle assessment, input-output life cycle assessment (IO-LCA), life cycle assessment (LCA), structural path analysis, environmental input-output analysis
User-Supplied Keywords:	
Abstract:	Hybrid life-cycle assessment (HLCA) methods combine bottom-up data from process-based inventories with top-down data from environmentally extended input-output tables. This is done to overcome limitations of data coverage and aggregation: While process inventory data is very detailed, it can never be complete. On the other hand, input-output data is by definition complete, but highly aggregated into economic sectors. Combining this data gives a more complete picture of the environmental impact associated with products or services. To this end, different mathematical methods have been proposed. Of the four main methods currently recognized in literature, three combine this data into a hybrid matrix. The path-exchange method instead works at the graph-level by combining the supply chain paths of both systems. This method is used most frequently in the environmental assessment of construction and the build environment. Unlike matrix-based hybrid methods, the accuracy of results of the graph-based method is limited by the number of paths considered. For the first time, we provide a concise mathematical description of the path-exchange algorithm and conduct a proof that this method is mathematically equivalent to the tiered-hybrid matrix method where upstream flows are inferred from the sectoral system. Based on this novel finding, we recommend the use of the method be discontinued in favor of the more accurate matrix-based method, in combination with a structural path analysis of the resulting hybrid matrix. Our proof and

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Note: The following files were submitted by the author for peer review, but cannot be converted to PDF. You must view these files (e.g. movies) online.	
main.tex references.bib	

Critical Analysis of the Path-Exchange Method for Hybrid Life-Cycle Assessment

Michael P. Weinold[1,2,†], Guillaume Majeau-Bettez[3]

[1]Laboratory for Energy Systems Analysis, PSI Centers for Nuclear Engineering & Sciences and Energy & Environmental Sciences, Villigen, Switzerland
[2]Chair of Energy Systems Analysis, Institute of Energy and Process Engineering, Department of Mechanical and Process Engineering, ETH Zurich, Zurich, Switzerland
[3]Department of Chemical Engineering, Polytechnique Montréal, Montréal, Canada
[†]Corresponding Author: michael.weinold@psi.ch

Abstract

Hybrid life-cycle assessment (HLCA) methods combine bottom-up data from process-based inventories with top-down data from environmentally extended input-output tables. This is done to overcome limitations of data coverage and aggregation: While process inventory data is more detailed, it can never be complete. On the other hand, input-output tables offer full cost-coverage for economic inventories that are complete, but highly aggregated into broad economic sectors. Combining these complementary datasets gives a more complete picture of the environmental impact associated with products or services. To this end, different mathematical methods have been proposed. Of the four main methods currently recognized in literature, three combine this data into a hybrid matrix. The path-exchange method instead works at the graph-level by combining the supply-chain paths of both systems. Unlike matrix-based hybrid methods, the accuracy of results of the graph-based method is limited by the number of paths considered. For the first time, we provide a concise mathematical description of the path-exchange algorithm and conduct a proof that this method is mathematically equivalent to the tiered-hybrid matrix method where upstream flows are inferred from the sectoral system. Based on this finding, we recommend the use of the method be discontinued in favor of the more accurate and computationally favorable matrix-based method, in combination with a structural path analysis of the resulting hybrid matrix. Our proof and the resulting guidance for practitioners is an important step toward a unified methodological framework for hybrid life-cycle assessment.

Keywords: hybrid life-cycle assessment, input-output life cycle assessment (IO-LCA), life cycle assessment (LCA), structural path analysis, environmental input-output analysis

1 Introduction

Since the early days of life-cycle assessment (LCA), researchers have attempted to combine data from complete but highly aggregated input-output tables of the economy with the incomplete but high-resolution process-based life-cycle inventory into a hybrid inventory. The first attempt was made in the 1970s [1], with most subsequent improvements made from the 1990s [2]. The umbrella term *hybrid life-cycle assessment* first saw use after 2000, for instance in a review by Lenzen et al. [3]. Today, it designates a number of distinct methods.

Four main methods have been recognized in literature [2][4]: The tiered method proposed in 1978 by Bullard et al. [1] and formalized by Heijungs et al. in 2002 [5], which was further formalized with respect to its harmonization of system boundaries to avoid double counting by Agez et al. between 2019 and 2022 [6]. The matrix augmentation method, introduced in 1999 by Joshi [7]. The integrated method, introduced in 2000 by Suh and Hupped [8]. Finally, the path-exchange (PXC) method, proposed in 1997 by Treloar [9] and formalized in 2009 by Lenzen et al. [10]. Despite recent publications aimed at establishing a standardized taxonomy [2], disagreement remains over the distinctions between methods.

Unlike the other three established methods, the path-exchange method operates at the level of the supply chain graph. This sets it apart from any matrix-based method. Its purported unique benefits, however, have remained somewhat elusive. For instance, the original authors have repeated statements of the kind *"Unlike other hybridisation methods, modifications to the supply chain are performed solely on discrete nodes, and thus do not require other changes within the overall matrix."* [11, Sec.2]. What this means for the utility of the method, for instance in the context of double-counting as described most recently by Agez et al. [6], has remained ambiguous. For instance, while some authors have described the path-exchange methods as employing *"algorithmic corrections for double-counting"* [12, Sec.6.1], others go further by claiming that it *"solves problems of double counting"* [4, Table 4] or that it *"cannot create any double-counting incident à la Strømman ([13])."* [14, Sec.2.6.3].

The original authors of the method in 2017 still observed that *"its application has been limited to a small group of scientists."* [11] and in 2018 *"(...) its application is rare and often limited to the group of researchers behind its development."* [2, Sec.5.2]. A systematic literature review shows that the path-exchange method for hybrid life-cycle assessment today is finding use primarily in the environmental assessment of the built environment, as shown in Table 2 of the Supplementary Information. Most recently, Stephan et al. were awarded the Graedel Best Paper Prize by the Journal of Industrial Ecology for their work on a *"multiscale framework for modeling and improving the life cycle environmental performance of built stocks"* [15], which employs this method.

Here, we describe the method in concise mathematical fashion, which allows us to illustrate the algorithm for a simple example system. Finally, we show that the method is, *in theory*, equivalent to the tiered hybrid matrix-based method for hybrid life-cycle assessment. We highlight the limitations inherent to any path-based algorithm, which mean that *in practice*, the path-exchange method will necessarily be inferior in accuracy to the matrix-based method. Based on this discussion, we caution practitioners against the use of this method. This comprehensive treatment will bring much-needed clarity to the ongoing discussion around the development of methods for hybrid life-cycle assessment.

2 Mathematical Framework

2.1 Example System and Diagrammatic Notation

We introduce an example system in Fig. 1, which we use extensively in Section 5. It consists of four economic sectors, and four production processes. We render the example system in a novel diagrammatic notation which allows for simple identification of process/sector correspondence, instances of double-counting, upstream flows from sectors to processes and the origin of data for every flow. This notation allows for an intuitive understanding of the hybridization of processes and sectors. It also allows us to augment the mathematical definition of the path-exchange method in Eq. (49) with a diagrammatic illustration in Fig. 3.

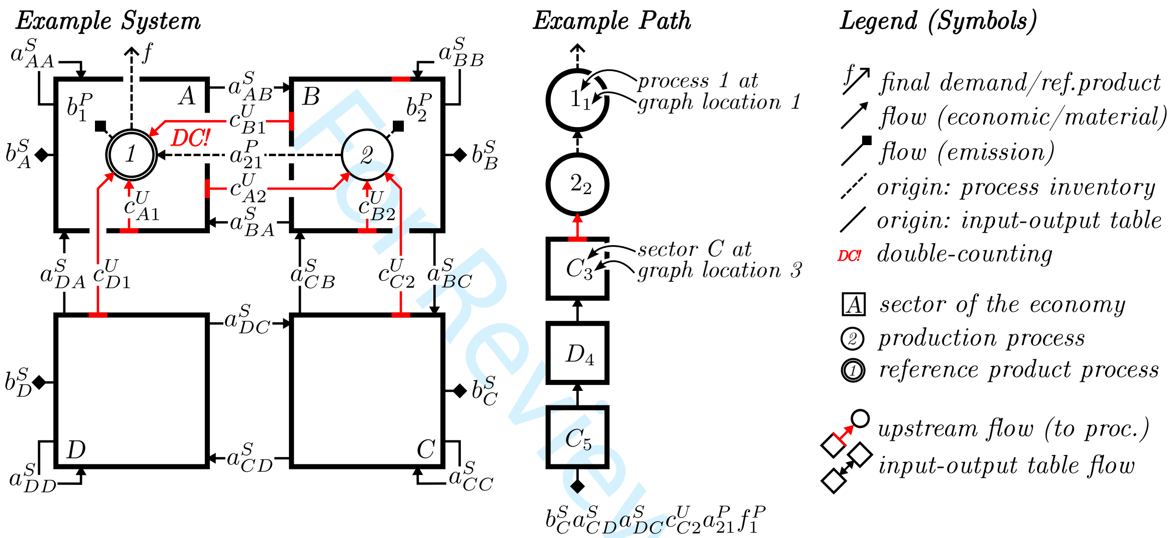

Figure 1: An example system of four sectors (A-D) and two processes (1,2), with corresponding matrices as defined in Eq. (1)-Eq. (3). Flows and emissions are annotated explicitly for the benefit of the reader. Note that not all sectors are connected to limit the complexity of the example. The diagrammatic notation is described in a legend presented in the right panel of the figure. This system is used in Fig. 3 and the mathematical proof of Section 4.

$$\mathbf{A}_S = \begin{array}{c} \\ A \\ B \\ C \\ D \end{array} \begin{array}{c} \begin{array}{cccc} A & B & C & D \end{array} \\ \begin{bmatrix} a^S_{AA} & a^S_{AB} & 0 & 0 \\ a^S_{BA} & a^S_{BB} & a^S_{BC} & 0 \\ 0 & a^S_{CB} & a^S_{CC} & a^S_{CD} \\ a^S_{DA} & 0 & a^S_{DC} & a^S_{DD} \end{bmatrix} \end{array} \tag{1}$$

$$\mathbf{A}_P = \begin{array}{c} \\ 1 \\ 2 \end{array} \begin{array}{c} \begin{array}{cc} 1 & 2 \end{array} \\ \begin{bmatrix} 0 & 0 \\ a^P_{21} & 0 \end{bmatrix} \end{array} \tag{2}$$

$$\mathbf{H} = \begin{array}{c} \\ A \\ B \\ C \\ D \end{array} \begin{array}{c} \begin{array}{cc} 1 & 2 \end{array} \\ \begin{bmatrix} 1 & 0 \\ 0 & 1 \\ 0 & 0 \\ 0 & 0 \end{bmatrix} \end{array} \tag{3}$$

2.2 Mathematical Convention and Table of Symbols

For the sake of simplicity in the equations of our proof but without loss of generality, we assume that both the process system and the sectoral system are already defined in the same units - either monetary or physical. For a detailed description of the conversion between these, we refer the reader to the comprehensive treatment by Weisz at al. [16].

For the sake of simplicity in our hybrid matrix representation, we have adopted the $\vec{e} = \mathbf{B}_P(\mathbf{I} - \mathbf{A_P})^{-1}\vec{f}_P$ convention for the governing equation of process-based life-cycle assessment. For a helpful discussion of the two different conventions, compare the comprehensive treatment by Heijungs et al. [17].

We employ the terms *node* and *edge* consistent with their standard usage in graph theory [18, P.9], where an *edge* is the flow between two *nodes*. In our case, a node could be a production process or economic sector, while an edge is the monetary of physical flow between them.

Table 1: Mathematical notation for vectors and matrices used throughout this article. The subscript P denotes the process system, the subscript S denotes the sectoral system and the subscript H denotes the hybrid system. For a complete derivation of the associated governing equations, refer to the supplementary information. Following formal notation in linear algebra [19, P.26], $\mathbf{A} \in \mathbb{R}^{R \times M}$ designates a matrix $\mathbf{A}$ of size $R \times M$ with all coefficients being elements of the real number field $\mathbb{R}$.

Index	*System*	*Description*
$i \in \mathbb{N}, \text{for } (1 \leq i \leq M)$	process system	production process ("activity")
$j \in \mathbb{N}, \text{for } (1 \leq j \leq N)$	sectoral system	economic sector
$k \in \mathbb{N}, \text{for } (1 \leq k \leq R)$	process system	environmental burdens (biosphere flows)
$l \in \mathbb{N}, \text{for } (1 \leq k \leq P)$	sectoral system	environmental burdens (env. satellite categories)

Matrix or Vector	*Description*
$\mathbf{A}_P \in \mathbb{R}_{M \times M}$	process system A-matrix (technology matrix [5])
$\mathbf{B}_P \in \mathbb{R}_{R \times M}$	process system environmental flow matrix (intervention matrix [5])
$\vec{f}_P \in \mathbb{R}_{M \times 1}$	final demand vector
$\vec{e}_P \in \mathbb{R}_{R \times 1}$	environmental flow vector
$\vec{x}_P \in \mathbb{R}_{M \times 1}$	output vector
$\mathbf{C}^u \in \mathbb{R}_{N \times M}$	upstream cut-off matrix
$\mathbf{A}_S \in \mathbb{R}_{N \times N}$	technical coefficient matrix
$\mathbf{B}_S \in \mathbb{R}_{P \times N}$	environmental satellite matrix
$\mathbf{H} \in \mathbb{R}_{N \times M}$	concordance matrix
$\vec{p} \in \mathbb{R}_{N \times 1}$	price vector

2.3 Matrix-Based HLCA Framework

Recent reviews recognize three distinct methods for hybrid life-cycle assessment which can be expressed in matrix form: the tiered hybrid method, the integrated hybrid method and the matrix-augmentation hybrid method [20][4][2]. While we must defer to these publications for detailed treatment of the different methods, we recapitulate the derivation of the tiered hybrid matrix method in the Supplementary Information for the benefit of the reader.

The basic assumption of the tiered hybrid method is that production processes consume inputs from other production processes. In addition, they consume "upstream" inputs from different sectors of the economy. Bearing in mind that a coefficient a_{ij} of the A-matrices defined in Table 1 describes the flow from node $i \rightarrow j$, we can consider a column of the A-matrix as the "production recipe" for the associated process or sector. With this, the governing equation of the tiered hybrid matrix method can therefore be written in the form most frequently used in literature:

$$\vec{e}_{H(tiered)} = \begin{pmatrix} \mathbf{B}_P & 0 \\ 0 & \mathbf{B}_S \end{pmatrix} \begin{pmatrix} \mathbf{I}-\mathbf{A}_P & 0 \\ \mathbf{C}_U^{uncorr} & \mathbf{I}-\mathbf{A}_S \end{pmatrix}^{-1} \begin{pmatrix} \vec{f}_P \\ 0 \end{pmatrix} \tag{4}$$

The uncorrected upstream cutoff matrix $\mathbf{C}_U$ contains all flows from sectors to processes. It can be populated *manually* by the practitioner. It can also be populated *automatically*, by inferring flows from the sectoral system, as first described by Strømman [21]. An intuitive illustration of this process is provided in Fig. 2. Here, process 1 originally only has inputs from process 2, which might reflect an incomplete system boundary, considering that its corresponding sector A requires inputs from both sector B and sector D. If the upstream cutoff matrix is automatically populated based on the requirements of sector A, it may compensate for missing flows from sector D. It also runs the risk, however, of doubly-counting some inputs. In this case from process 2 *and* sector B. This is instance of double counting illustrates the need for automated harmonization of the boundaries of the process and sectoral inventories through an automated correction for double counting.

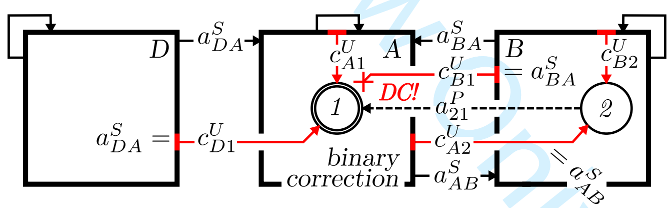

Figure 2: A simple example system consisting of three sectors (A,B,C) and two processes (1,2), taken the larger system in Fig. 1. Shown is the process by which upstream flows c^U are inferred from the sectoral system, as well as an instance of double-counting in a hybrid system ("DC!"). If both the upstream flow c^U_{C1} and the process flow a^P_{21} are retained, the environmental impact upstream of process 1 will be overestimated. Under the binary double-counting correction technique, the upstream flow c^U_{B1} is removed altogether. The upstream flow c^U_{D1} is not removed, since no process flow to process 1 originates in sector B. For a legend of the diagrammatic notation used, compare the right panel of Fig. 1.

The construction of the upstream matrix in this way can be formally defined as [21, Eqn.(4)ff.][6, Eqn.(7)]

$$\mathbf{C}_U^{uncorr} = \mathbf{A}_S \mathbf{H} \tag{5}$$

$$= \begin{array}{c} \\ A \\ B \\ D \end{array} \begin{array}{c} \begin{array}{ccc} A & B & D \end{array} \\ \begin{bmatrix} a^S_{AA} & a^S_{AB} & 0 \\ a^S_{BA} & a^S_{BB} & 0 \\ a^S_{DA} & 0 & a^S_{DD} \end{bmatrix} \end{array} \begin{array}{c} \\ A \\ B \\ D \end{array} \begin{array}{c} \begin{array}{cc} 1 & 2 \end{array} \\ \begin{bmatrix} 1 & 0 \\ 0 & 1 \\ 0 & 0 \end{bmatrix} \end{array} \tag{6}$$

$$= \begin{array}{c} \\ A \\ B \\ D \end{array} \begin{array}{c} \begin{array}{cc} 1 & 2 \end{array} \\ \begin{bmatrix} c^U_{A1} = a^S_{AA} & c^U_{A2} = a^S_{AB} \\ c^U_{B1} = a^S_{BA} & c^U_{B2} = a^S_{BB} \\ c^U_{D1} = a^S_{DA} & 0 \end{bmatrix} \end{array} \tag{7}$$

Here, we first use the concordance matrix $\mathbf{H}$. As detailed in the Supplementary Information, a concordance matrix contains the information required to assign each process to one or more sectors of the economy. It can be used to convert vectors or matrices from the process-basis into the sector-basis. We define it as

$$\mathbf{H} \rightarrow h_{ij} = \begin{cases} 1 & \text{if sector } i \text{ contains process } j \\ 0 & \text{else} \end{cases} \tag{8}$$

As indicated in Fig. 2, this leads to potential cases of *double-counting*. These are instances in the hybrid system where an upstream flow from a sector into a process is already covered by a process flow. The upstream cut-off matrix must therefore be corrected to avoid double-counting. For a comprehensive discussion of different double-counting correction methods, compare the recent review by Agez et al. [6]. Here, we use the binary double-counting correction method[1]. Under this correction method, the upstream input from sector i into process j is set to zero, if any process flow to process j originates from a process contained in sector i. The concept is illustrated in Fig. 2. As we will see later, the path-exchange method essentially employs an identical logic.

In order to streamline the notation, we first introduce the Iverson bracket, which is a generalization of the Kronecker delta [23]. It is defined such that it evaluates to 1 if the condition in the bracket is true and evaluates to 0 otherwise. In our case, we use the condition

$$[x \stackrel{?}{=} 0] = \begin{cases} 1 & \text{if } x = 0 \\ 0 & \text{else} \end{cases} \tag{9}$$

where the question mark above the equality indicates that the expression is a logical condition evaluated by the Iverson bracket, rather than an assignment. Using the Iverson bracket notation and the definition of the concordance matrix, we can now formalize the equation which applies the binary double-counting correction technique to the uncorrected upstream flow matrix.

$$c_{ij}^{U,corr} = \left[\sum_{k=1}^{M} h_{ik} a^P_{kj} \stackrel{?}{=} 0 \right] c_{ij}^{U,uncorr} \tag{10}$$

According to the definition of the concordance matrix in Eq. (8)

$$h_{ik} a^P_{kj} = \begin{cases} a^P_{kj} & \text{if flow from process } k \rightarrow j \text{ originates in sector } i \\ 0 & \text{else} \end{cases} \tag{11}$$

and

$$M < i \leq N \ldots \text{index } i \text{ iterates over all } N \text{ sectors}$$
$$1 \leq j \leq M \ldots \text{index } j \text{ iterates over all } M \text{ processes}$$
$$1 \leq k \leq M \ldots \text{index } k \text{ iterates over all } M \text{ processes}$$

[1] Note that a small error is present in the equations for the corrected upstream cut-off matrix $\mathbf{C}_U^{corr}$ provided by Agez et al. [6, (8)-(8')]. Instead of the standard matrix multiplication the authors meant to use the Hadamard product [22].

We can see that the expression in the Iverson bracket evaluates to 1, only if no process in sector i has a flow a^P that terminates in process j. This is the definition of binary double-counting correction for hybrid life-cycle assessment. Using this notation, we can write corrected coefficients of the upstream flow matrix from the simple example in Fig. 2.

$$c_{B1}^{U,corr} = [0 \stackrel{?}{=} 0] c_{B1}^{U,uncorr} = c_{B1}^{U,uncorr} \tag{12}$$

$$c_{C1}^{U,corr} = [a_{21}^P \stackrel{?}{=} 0] c_{C1}^{U,uncorr} = 0 \tag{13}$$

In matrix form, Eq. (10) can be expressed as

$$\mathbf{C}_U^{corr} = [\mathbf{H}\mathbf{A}_P \stackrel{?}{=} 0] \otimes \mathbf{C}_U^{uncorr} \tag{14}$$

and the governing equation of the tiered hybrid matrix method becomes

$$\vec{e}_{H(tiered)} = \begin{pmatrix} \mathbf{B}_c^P & 0 \\ 0 & \mathbf{B}_c^S \end{pmatrix} \begin{pmatrix} \mathbf{I} - \mathbf{A}_P & 0 \\ \mathbf{C}_U^{corr} & \mathbf{I} - \mathbf{A}_S \end{pmatrix}^{-1} \begin{pmatrix} \vec{f}_P \\ 0 \end{pmatrix} \tag{15}$$

2.4 Path-Based HLCA Framework

As per the governing equation of both environmentally-extended input-output analysis and process-based life-cycle assessment, the environmental burden vector $\vec{e}$, which gives the total environmental burdens incurred from an arbitrary final demand $\vec{f}$, can be written as

$$\vec{e} = \mathbf{B}_c(\mathbf{I} - \mathbf{A})^{-1}\vec{f} = \mathbf{B}_c\mathbf{L}\vec{f} \tag{16}$$

In this context, $\mathbf{L}$ is known as the Leontief inverse. In the following, we will limit the discussion to the case of a single environmental burden (eg. carbon dioxide emissions). The above equation then becomes

$$e = \vec{b}_c^T\mathbf{L}\vec{f} \tag{17}$$

The first part of the equation is also known as the multiplier vector [24, Sec.6.2.3, Sec.8.5.1]

$$\vec{m}^T = \vec{b}_c^T\mathbf{L} \tag{18}$$

As originally proposed by Waugh [25] and reported by Miller & Blair from 1985 [26][27][24], the Leontief inverse $\mathbf{L} = (\mathbf{I} - \mathbf{A})^{-1}$ can be approximated by a power series, since $\sum_{i=1}^{n} a_{ij} < 1 \land a_{ij} \geq 0$ [24, Sec. 2.4.2].

$$\mathbf{L} = (\mathbf{I} - \mathbf{A})^{-1} = (\mathbf{I} + \mathbf{A} + \mathbf{A}^2 + \mathbf{A}^3 + \dots) \tag{19}$$

This is sometimes called *production layer decomposition* [10]. It forms the basis of both *structural path analysis*[2] in general and the path-exchange method for the *hybrid method* for life-cycle assessment in particular. A path in this context describes a product of coefficients of the kind $b_3a_{32}a_{21}$. These paths are formally known as directed acyclic graphs [18, P.200ff.]. With this, the environmental burden multiplier vector $\vec{m}^T$ in Eq. (18) can be expressed using the product layer decomposition in Eq. (19) as

$$m_i = b_i + \sum_{j=1}^{N} b_j a_{ji} + \sum_{j=1}^{N}\sum_{k=1}^{N} b_j a_{jk} a_{ki} + \sum_{j=1}^{N}\sum_{k=1}^{N}\sum_{l=1}^{N} b_j a_{jk} a_{kl} a_{li} + \dots \tag{20}$$

The path-exchange method now operates by combining paths from the production layer decomposition of a process system and a sectoral system. The associated algorithm is described in detail in Section 3.1. Similar to Eq. (15), we introduce the following notation to describe operation of the path-exchange method on both the sectoral and process paths

$$e = \mathrm{PXC}\{\mathbf{B}_c^S(\mathbf{I} - \mathbf{A_S}), \mathbf{B}_c^P(\mathbf{I} - \mathbf{A_P})^{-1}\}\vec{f}_P \tag{21}$$

[2]A detailed discussion of the evolving use of the term *structural path analysis* is provided in the Supplementary Information

3 The Path-Exchange Method for Hybrid Life-Cycle Assessment

3.1 General Description and Illustration

As detailed in Section 3.2, the path-exchange algorithm was first proposed by Lenzen et al. in 2009 [10]. Unfortunately, a formal mathematical or pseudo-code definition of this first implementation of the algorithm (hereafter named "PXC(2009)") was not provided. Instead, the different steps were traced out explicitly using a practical example.

Subsequent publications on the methodology of the path-exchange method by Crawford and Stephan et al. in 2017 [11] and 2019 [28] (hereafter named "PXC(2017/2019)") changed the definition of the method slightly [11, Sec.3.2][28, Sec.2.1]. No formal definition of the new implementation was provided either, although the method was visually illustrated in [11, Figure 2] and [28, Figure 1]. The method consists of multiple discrete steps, which we have illustrated in Fig. 3 using our novel diagrammatic notation.

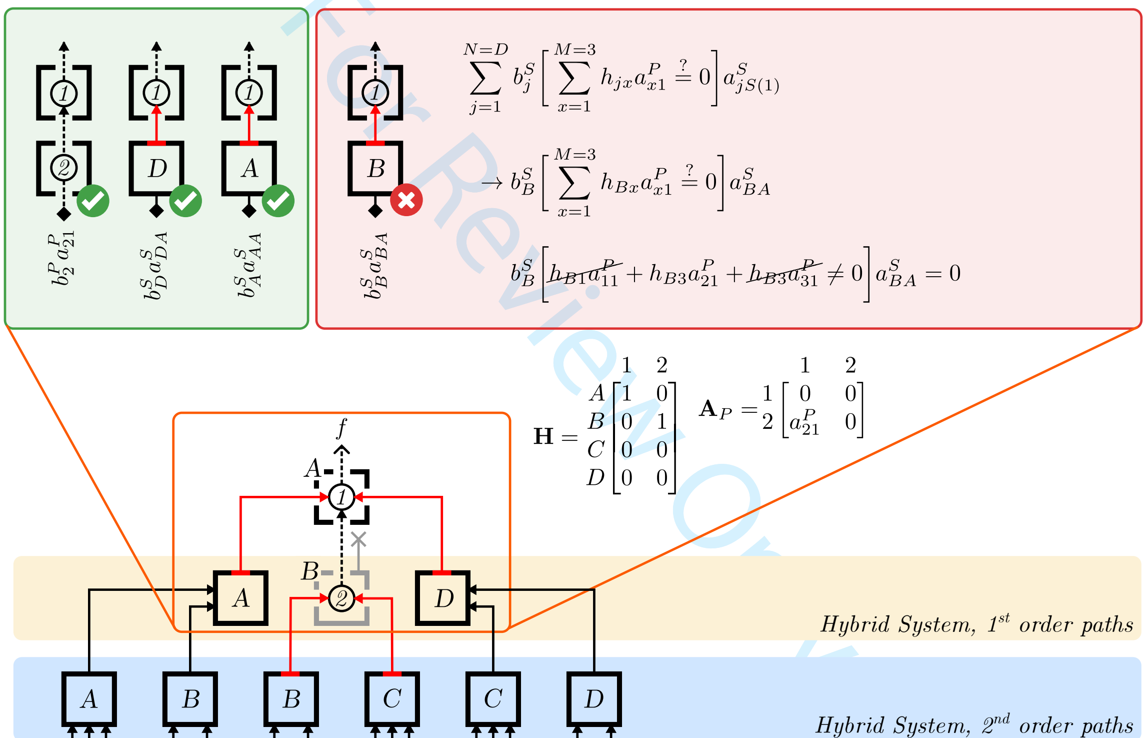

Figure 3: Visual representation of the PXC(2017/2019) algorithm, using the example system of Fig. 1. First, the algorithm conducts a structural path analysis of both the process system and the sectoral system. Of this, here we show only paths of orders 1-2 terminating in process 1. For order 1, we obtain one process path $b_2^P a_{21}^P$ and three sectoral paths $b_D^S a_{DA}^S, b_A^S a_{AA}^S, b_B^S a_{BA}^S$. Now, the algorithm *"matches"* the paths based on a concordance matrix $\mathbf{H}$. In our diagrammatic notation, this is intuitively shown by process symbols being contained in sector symbols. Now, those sectoral paths are removed for which there is a direct process path equivalent. In this example, this is the case only for path $b_B^S a_{BA}^S$, which has the equivalent $b_2^P a_{21}^P$. Summing all paths according to Section 2.4 now yields the "hybrid" environmental impact. The mathematical formulation of this condition is shown next to the removed path. For a definition of the Iverson bracket operator used, compare Eq. (48). For a legend of the diagrammatic notation, compare the right panel of Fig. 1.

3.2 Historical Development and Motivation

Lenzen in his first complete formulation of the path-exchange method for hybrid life-cycle assessment describes his work [29, Sec.1] as building on the earlier method of Treloar [30, Sec.4], then referred to as *"an innovative input-output based hybrid analysis method"* [31, P.205]. He then asserts that *"The general decomposition approach* [used in the path-exchange method] *was introduced into economics and regional science in 1984 under the name Structural Path Analysis."*, citing Defourny et al. [32] and Crama et al. [33]. We find this representation of the lineage of the method to be incorrect, and present a more accurate version in Table 2. A detailed discussion of this lineage is presented in the Supplementary Information.

Table 2: Milestones in the historical development of the path-exchange method for HLCA.

authors	*year*	*contribution*	
Waugh	1950	power series expansion of the Leontief inverse	[25]
Bullard et al.	1975	power series expansion of the Leontief inverse and coefficient replacement	[34]
Seeman	1984	power series expansion of the Leontief inverse and coefficient replacement	[35]
Treloar	1997	refined method for coefficient replacement	[9]
Lenzen et al.	2009	first formalization in the context of life-cycle assessment	[36]
Crawford et al.	2017	workflow formalization	[11]
Stephan et al.	2019	software implementation	[28]

3.3 Motivation and Misconceptions

To support our formal proof in Section 4, we are providing a detailed discussion of three major misconceptions associated with the path-exchange method for hybrid life-cycle assessment in Section 3.3.1-Section 3.3.3. These misconceptions have all been used to motivate the introduction and use of the method and are among the reasons why the path-exchange method has so far been listed as a separate hybrid life-cycle assessment method in reviews publications.

3.3.1 "Avoiding Changing Coefficients in the Sectoral Matrix"

Treloar in his 1998 thesis motivated the introduction of his first version of the path-exchange method from two different perspectives: *"The process analysis framework cannot be used as a basis for hybrid analysis because of its incompleteness (ie, regardless of the greater reliability of the process analysis data). The comprehensive input-output framework cannot currently be used as basis for hybrid analysis because the substitution of process analysis data into the input-output model causes unwanted indirect effects."* [30, Sec.1].

The first point relates to the lack of available data for building the background inventory of a process-based life-cycle assessment. At that time, the *Ecoinvent* database predecessor *ETH 96* had just been released and featured less than 2'500 individual processes [37][38] - a number which has since increased to over 20'000 in the latest version [39][40]. The hope of Treloar was therefore to utilize readily available and up-to-date government-compiled input-output tables for better data coverage.

The interpretation of the second point is more involved and reveals an important misconception which has been frequently repeated since then. Treloar goes on to specify that *"(...) input-output-based hybrid analysis (...) involves the substitution of process analysis data for coefficients in the direct input-output matrix (Bullard et al., 1978 [1]; Seeman, 1984 [35])."* [30, Sec.2.3.2].

The concern here is that changes made to a technical coefficient of the input-output matrix in the context of one specific supply chain affect all other supply chains involving this technical coefficient.

This is an understandable concern in principle. However, the referenced publication by Bullard et al. does not modify the input-output matrix at all. It is instead an early example of input-output-based life-cycle assessment, as described most recently by Heijungs [17]. We must stress that the publication of Bullard et al. does not attempt to hybridize data, nor replace any coefficients in the input-output matrix, as claimed[3].

Similarly, the referenced thesis of Seeman does not modify the input-output matrix with more specific coefficients either. He briefly suggests a way to dis-aggregate sectors into more specific sub-sectors, thereby adding new columns and rows to the technical coefficient matrix. This approach would later be described in detail by Joshi [7] and is now commonly referred to as the *matrix-augmentation* hybrid method [2]. The calculations of Seeman, however, do not actually employ this dis-aggregation[4]. Instead, he describes a method for using the power series expansion of the Leontief inverse where input-output technical coefficients are replaced with more process-specific coefficients. This was already suggested by Bullard some 10 years earlier [34]. We must stress again that the publications of Seeman and Bullard et al. do not replace any coefficients in the input-output matrix, as claimed.

Apart from the integrated method for hybrid life-cycle assessment, where this is done deliberately, no hybrid analysis modifies the coefficients of the sectoral matrix. This includes the publications cited by Treloar. Note also that when Treloar made his proposal in 1997, the integrated method for hybrid life-cycle assessment had not yet been developed - and could therefore not have been referenced by him. Only two early publications by Bullard from 1976 [42] and 1978 [1] on what is today designated the tiered hybrid method had been published at the time.

Even so, this second point of the original motivation for the path-exchange method continued to be cited in subsequent publications: *"Treloar observed that changing the transaction coefficient for a particular element, or node, in an input-output matrix used for LCA would affect all supply chain paths that contain that node, even if the changed coefficients applied only to a particular path. Treloar correctly recognized that SPA provides a means to avoid such undesired "global" effects."* [10, Sec.1].

In summary, the second of two key issues cited as the original motivation for the introduction of the path-exchange method is not supported by the literature cited. It is plausible that it is based on a misinterpretation of the referenced publications.

[3] In the last paragraph, the theoretical possibility of "integrating" process data and input-output data is mentioned: *"With more extensive data, such as that from a conventional LCA, and a two-step process that integrates these data into the input-output matrix, we believe the two approaches can be integrated."* [41]. However, this *"integration"* is not described there in any further detail, nor is its implementation within the scope of the publication.

[4] The only change to the technical coefficient matrix that is made as part Seeman's calculations is the conversion of *"(...) the outputs of the energy sectors from dollars to megajoules (...)"* to facilitate the analysis of energy flows rather than economic flows.

3.3.2 "Working on Mutually Exclusive Nodes"

More recent publications on the path-exchange method have built on the misconception of Section 3.3.1 and listed as the main advantage of this approach that *"(...) it operates on mutually exclusive process and input output nodes"* [28, P.240] and that *"Unlike other hybridisation methods, modifications to the supply chain are performed solely on discrete nodes, and thus do not require other changes within the overall matrix."* [11, P.159].

This suggests that the path-exchange hybrid method, unlike matrix methods, can selectively modify nodes or edges in the supply chain pathways of a sectoral system and replace them with more precise nodes or edges from another system.

However, this is another misconception about the path-exchange method. In fact, selective replacement of sectoral information for process information at arbitrary locations in the supply chain graph can easily be achieved using a matrix-based system. As an example, consider a system illustrated in Fig. 4.

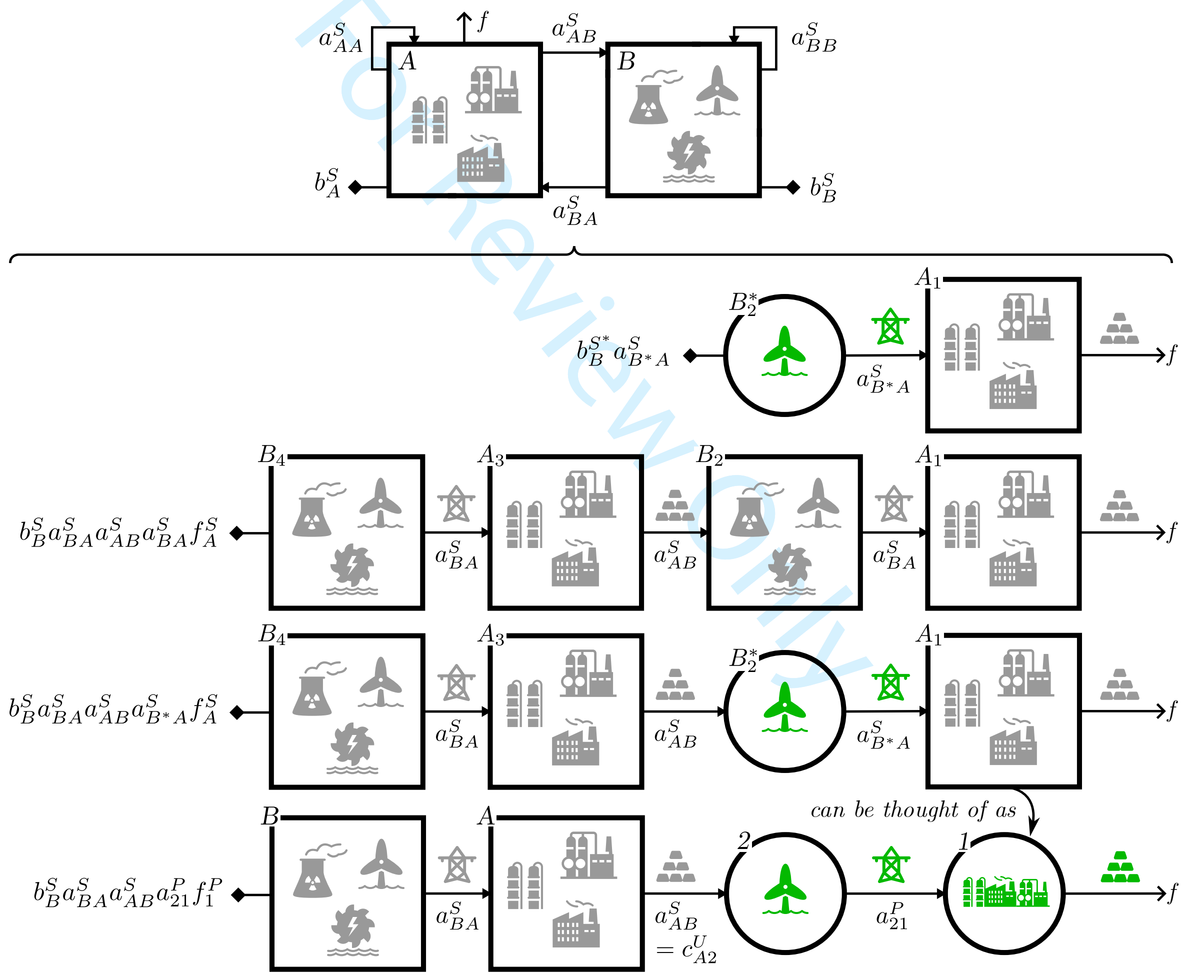

Figure 4: Top: A simple example system consisting of two sectors (A,B), taken the larger system in Fig. 1. Here, we assume sector A produces metals, while sector B supplies electricity. Bottom: An example path of order three: $b^S_B a^S_{BA} A^S_{AB} A^S_{BA} f^S_A$, in which a coefficient a^S_{BA} is exchanged for a more specific coefficient $a^S_{B^*A}$. For a legend of the diagrammatic notation, compare the right panel of Fig. 1.

We first posit that we have obtained specific information on edge $a^S_{B_2A_1}$, the flow from the electricity sector into the metals sectors. In the following, we designate this specific node B^*. This includes data on the specific environmental burden of sector B $b^S_{B^*_2}$ and the amount of flow supplied to sector A $a^S_{B^*_2 A_1}$. Note that according to the legend in Fig. 1, the integer subscripts here indicate the position of the node or edge in the supply chain. The corresponding path

$$b^S_{B^*_2} a^S_{B^*_2 A_1} f^S_A \tag{22}$$

is shown in Fig. 4. Now, we consider an arbitrary third order path of the sectoral system. Note that the edge a^S_{BA} appears in different locations of this supply chain. It is underlined for emphasis.

$$b^S_{B_4} \underline{a^S_{B_4 A_3}} a^S_{A_3 B_2} \underline{a^S_{B_2 A_1}} f^S_A \tag{23}$$

According to Section 3.1, this path is now altered by the path-exchange method to

$$b^S_{B_4} a^S_{B_4 A_3} a^S_{A_3 B_2} a^S_{B^*_2 A_1} f^S_A \tag{24}$$

The path-exchange method has targeted a specific edge, denoted as $a^S_{B_2 A_1}$, at a particular point in the supply chain and replaced it with another edge instance, $a^S_{B^*_2 A_1}$, without altering other instances of the node-edge pair, such as $a^S_{B_4 A_3}$.

However, such a scenario can be easily captured in matrix form. The key to understanding this equivalence lies in the abstract notion that *specific information always lives on a specific supply chain.*

First, it is important to realize that in this example, we have information *only* on the electricity production node which feeds directly into the metal production node $B_2 \rightarrow A_1$. We have no information on electricity production nodes which appear *further upstream* in the supply chain, such as $B_4 \rightarrow A_3$.

To elaborate, consider again the first-order path of Eq. (22). In our scenario, we have specific information on the environmental burden coefficient $b^{S*}_{B_2}$ of node B at position 2 in the graph:

$$B_2 \rightarrow A_1 \rightarrow f \tag{25}$$
$$B^*_2 \rightarrow A_1 \rightarrow f \tag{26}$$

From Eq. (26) we can see that in our scenario we know *not only* something specific about node B_2, but *also* something about node A_1. At the very least, we know that in our specific supply chain, node A_1 does not consume the average input of node B_2, but the input of a specific node B^*_2. In our scenario, this is the only thing we know about node A_1. All other properties of this node we simply *infer* from the input-output system. These properties are the technical coefficient to this node a^S_{BA} and the environmental burden coefficient b^S_A.

We can therefore think of node A_1 as a *specific* node instance of sector A, much like B^*_2 is a *specific* node instance of sector B. For consistency, we therefore denote it A^*_1. This means that in our specific example, we have taken the metal production sector as a *proxy* for the metal production process under investigation.

How can we collect this specific information? One way to do so is in a process matrix $\mathbf{A}_P$ and an environmental burden coefficient vector $\vec{B}_P$. We can also think of these specific sectoral node instances (A_*, B_*) as processes $(1, 2)$.

$$\mathbf{A}_P = \begin{array}{c} \\ A^* \\ B^* \end{array} \begin{array}{c} \begin{array}{cc} A^* & B^* \end{array} \\ \left[\begin{array}{cc} 0 & 0 \\ a^S_{BA} & 0 \end{array} \right] \end{array} = \begin{array}{c} \\ 1 \\ 2 \end{array} \begin{array}{c} \begin{array}{cc} 1 & 2 \end{array} \\ \left[\begin{array}{cc} 0 & 0 \\ a^P_{21} = a^S_{BA} & 0 \end{array} \right] \end{array} \tag{27}$$

$$\vec{B}_P = \begin{array}{c} A^* \\ B^* \end{array} \left[\begin{array}{c} b^S_A \\ b^{S*}_B \end{array} \right] = \begin{array}{c} 1 \\ 2 \end{array} \left[\begin{array}{c} b^P_1 = b^S_A \\ b^P_2 = b^{S*}_B \end{array} \right] \tag{28}$$

We can see that in Eq. (27), the technical coefficient a_{21}^P between nodes B_2 and A_1 is simply the technical coefficient A_{BA}^S. Similarly, we can see that in Eq. (28), the environmental burden coefficient b_2^P for node B_2 is simply the *specific* coefficient b_B^{S*} of which we have knowledge. The environmental burden coefficient b_1^P for node A_1 is simply the sectoral average environmental burden coefficient b_A^S.

We know one more thing about node B_2. Since it is a specific instance of sector B, we can see from Fig. 4 that is takes input from sector A. We record this information in an upstream flow matrix $\mathbf{C}^U$.

$$\mathbf{C}^U = \begin{array}{c} \\ A \\ B \end{array} \begin{array}{c} \begin{array}{cc} 1 & 2 \end{array} \\ \left[\begin{array}{cc} 0 & 0 \\ 0 & c_{A2}^U = a_{AB}^S \end{array} \right] \end{array} \tag{29}$$

We can now, according to the definition of matrix-based methods for hybrid life-cycle assessment [2], combine this process matrix $\mathbf{A}_P$ with the input-output technical coefficient matrix $\mathbf{A}_S$. We do the same for the environmental burden coefficient vectors.

$$\mathbf{A}_H = \begin{array}{c} \\ 1 \\ 2 \\ A \\ B \end{array} \begin{array}{c} \begin{array}{cccc} 1 & 2 & A & B \end{array} \\ \left[\begin{array}{cc|cc} 0 & 0 & 0 & 0 \\ a_{21}^P = a_{BA}^S & 0 & 0 & 0 \\ \hline 0 & c_{A2}^U = a_{AB}^S & a_{AA}^S & a_{AB}^S \\ 0 & 0 & a_{BA}^S & a_{BB}^S \end{array} \right] \end{array} \tag{30}$$

$$\vec{B}_H = \begin{array}{c} 1 \\ 2 \\ A \\ B \end{array} \left[\begin{array}{c} b_1^P = b_A^S \\ b_2^P = b_B^{S*} \\ b_A^S \\ b_B^S \end{array} \right] \tag{31}$$

Conducting a structural path analysis on the hybrid matrix in Eq. (30), we get:

$$m_1 \sim b_2^P a_{21}^P + a_B^S a_{BA}^S c_{A2}^U a_{21}^P + \ldots \tag{32}$$

This is equivalent to the path-exchange approach. As we can see, the matrix-based solution retains

$$B_4 \rightarrow A_3 \rightarrow B_3^* \rightarrow A_1^* \rightarrow f \tag{33}$$

In summary, it is key to understand that we *can* make modifications of specific nodes in a supply chain graph. However, this means that *all nodes downstream* of this modified node become specific node instances. We can infer their parameters from the input-output system and record them in a process matrix. This matrix can then be solved exactly according to the governing equation of input-output analysis $(\mathbf{I} - \mathbf{A})^{-1}\vec{f} = \vec{x}$ [24, Eqn.(2.11)].

3.3.3 "Avoiding Double Counting"

Another claim made by recent publications about the path-exchange method is that it *"avoids double-counting"* [10, Sec.4] as a result of *"exchange by definition"* [10, Sec.2.1]. This refers to the step of the path-exchange method that replaces some sectoral nodes with process nodes, leaving no *"ambivalence"* or double flows in these pathways.

However, this is another misconception about the path-exchange method. It does not somehow inherently avoid the problem of double counting. Instead, it simply employs the well-established binary double-counting correction method at the graph-level. As an example, consider a system illustrated in Fig. 5.

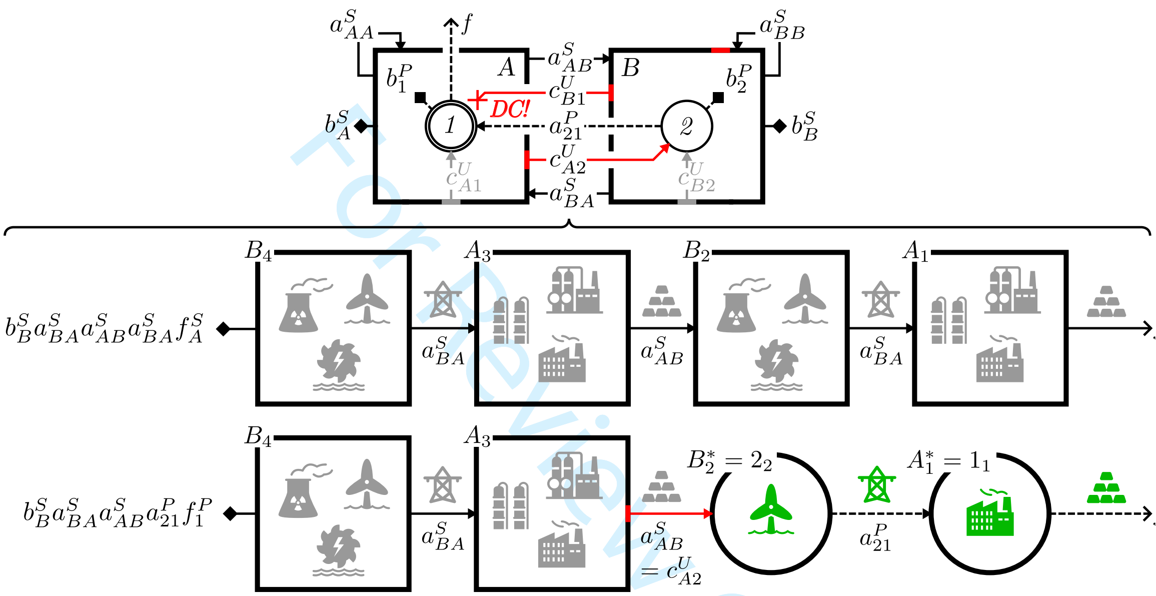

Figure 5: Top: A simple example system consisting of two sectors (A,B) and two processes (1,2), taken the larger system in Fig. 1. For the corresponding hybrid matrix, compare Eq. (37). Note that the two upstream flows c_{A1}^U and c_{B2}^U are not considered in the hybrid matrix for reasons of simlicity and are therefore shown in grey. As indicated by the line-style of the two red "upstream flow" arrows, they have been deduced from the underlying sectoral system. The resulting instance of double-counting is marked "DC!". As indicated by the terminating "X", flow c_{B1}^U is removed completely, therefore constituting binary double counting correction. For a legend of the diagrammatic notation, compare the right panel of Fig. 1. Bottom: Two example paths of the system: $b_2^P a_{21}^P f_1^P$ and $b_B^S a_{BA}^S a_{AB}^S a_{21}^P f_1^P$.

Here, we again consider an arbitrary third order path of the sectoral system:

$$b_{B_4}^S a_{B_4 A_3}^S a_{A_3 B_2}^S a_{B_2 A_1}^S f_A^S \tag{34}$$

Note that according to the legend in Fig. 1, the integer subscripts here indicate the position of the node in the supply chain. As indicated in Fig. 5, specific information on two processes (1,2) is available. According to Section 3.1, the path is therefore altered by the path-exchange method to

$$b_{B_4}^S a_{B_4 A_3}^S a_{A_3 B_2}^S a_{2_2 1_1}^S f_1^P = \tag{35}$$
$$b_{B_4}^S a_{B_4 A_3}^S c_{A_3 2_2}^S a_{2_2 1_1}^S f_1^P \tag{36}$$

Note that here only the coefficient $a_{B_2 A_1}^S$ was changes, but not $a_{B_4 A_3}^S$.

The system of Fig. 5 can be represented through a hybrid technical coefficient matrix $\mathbf{A}_H$ and a hybrid environmental burden coefficient vector $\vec{B}$.

$$\mathbf{A}_H = \begin{array}{c} \\ 1 \\ 2 \\ A \\ B \end{array} \begin{array}{cccc} 1 & 2 & A & B \\ \left[\begin{array}{cc|cc} 0 & 0 & 0 & 0 \\ a_{21}^P & 0 & 0 & 0 \\ \hline 0(def!) & c_{A2}^U = a_{AB}^S & a_{AA}^S & a_{AB}^S \\ 0(DC!) & 0(def!) & a_{BA}^S & a_{BB}^S \end{array}\right] \end{array} \tag{37}$$

$$\vec{B}_H^T = \begin{array}{c} \begin{array}{cccc} 1 & 2 & A & B \end{array} \\ \left[\begin{array}{cccc} b_1^P & b_2^P & b_A^S & b_B^S \end{array}\right] \end{array} \tag{38}$$

As we can see from Eq. (37), the upstream flow c_{B1}^U into process 1 has been removed, because there is already a process flow a_{21}^P into process 1. This is the definition of binary double-counting correction. In the matrix this is indicated through "$0(DC!)$".

To *trace the supply chain paths* of this system, we now conduct a structural path analysis, following the description in Section 2.4. In the governing equation of environmentally extended input-output analysis, the environmental burden e associated with a final demand vector $\vec{f}_H$ can be expressed as

$$e = \vec{B}_H^T(\mathbf{I} - \mathbf{A}_H)^{-1}\vec{f}_H \tag{39}$$

The first part of this equation is known as the multiplier vector $\vec{m}$

$$\vec{m} = \vec{B}^T(\mathbf{I} - \mathbf{A})^{-1} \tag{40}$$

Using the power series expansion of a matrix inverse

$$(\mathbf{I} - \mathbf{A}_H)^{-1} = \mathbf{I} + \mathbf{A}_H + \mathbf{A}_H^2 + \dots \tag{41}$$

we can expand the multiplier vector of Eq. (40). In our example, we consider the case where the functional unit is the output of process 1, as indicated in Fig. 5. The final demand vector is therefore simply

$$\vec{f}_H = \begin{array}{c} 1 \\ 2 \\ A \\ B \end{array} \left[\begin{array}{c} 1 \\ 0 \\ 0 \\ 0 \end{array}\right] \tag{42}$$

According to Eq. (39) and Eq. (40), in this case the environmental burden e can be written as

$$e = \vec{m}\vec{f}_H \tag{43}$$

$$e = m_1 \tag{44}$$

and the power series expansion can be written as

$$m_1 = b_1^P + \sum_{j=1}^{4} b_j^H a_{j1}^H + \sum_{j=1}^{4}\sum_{k=1}^{4} b_k^H a_{kj}^H a_{j1}^H + \sum_{l=1}^{4}\sum_{j=1}^{4}\sum_{k=1}^{4} b_l^H a_{lk}^H a_{kj}^H a_{j1}^H + \dots \tag{45}$$

We can now take a single third-order pathway from this expansion

$$b_4^H a_{43}^H a_{32}^H a_{21}^H = b_B^S a_{BA}^S c_{A2}^U a_{21}^P \tag{46}$$

This path is visualized in the bottom section of Fig. 5. As we can see, the binary double-counting correction in Eq. (37) ensures that no flow from sector B to process 1 is added. However, it does not impede the flow between sectors B and A elsewhere in the supply chain.

3.4 Limitations

In the structural path analysis of Eq. (20), the upper bound for the number of possible paths of order n for a system of N economic sectors is N^n. This holds true only if every node in the system is connected to every other node. Note that the exact number of possible paths in a more realistic system can only be computed by means of the adjacency matrix of the hybrid system, which we describe in the Supplementary Information. The equation for the upper bound of paths is frequently used incorrectly when referring to the number of possible paths [9, P.378][43, Footnote 1][36, P.8252][14, P.25]. Even limiting the investigation of paths to an arbitrary maximum path order may therefore prove computationally prohibitive, depending on the size of the system. To mitigate this issue, pruning techniques are used by practitioners, disregarding paths below a threshold contribution to overall impact [44, Sec.2.3]. These pruning techniques can be effective, reducing the number of relevant paths to a number much smaller than the total number of paths [45]. However, as practitioners note, a specific cut-of values is often chosen *"(...) for convenience, and because it was expected that it would provide a sufficiently detailed model to be used as the basis for an I–O-based hybrid analysis without providing too many energy paths."* [31]. And since most studies use systems of different size and scope, *"(...) subjective choices are unavoidable during the computational process."* [44].

One study combined the input-output table of the United Kingdom with the *Ecoinvent* database using the path-exchange method to investigate emissions of wind power. It was found that 23% of emissions were associated with paths that each contributed less than 0.034% [46, Supplement Sec.5]. As other authors observed, *"These small paths are often neglected in SPA studies ([47][29][48])."* [49]. One interesting example from a specific case study was provided: *"environmental impacts of electricity production in developing countries arise from numerous small contributions and not a few single, but large, contributions."* [49]. Any method working at the path-level is therefore subject to the above limitations. This is also the case for the path-exchange method for hybrid life-cycle assessment.

In addition, the error introduced by cutting of paths cannot be readily quantified. Error here refers to the missing amount of upstream input from the sectoral system into the process system, which is not accounted for due to the path cutoffs inherent to the path-exchange method. Since the number of possible paths grows exponentially with the size of the hybrid system, the traversal of these paths must be cut off after a specific threshold t of contribution to total emissions, or a specific path length d. In practice, authors have used varying parameters, depending on the study context $d = 5/t = 0.005 - 0.01\%$ [44], $d = 6/t = 5 - 1\%$ [48], $d = 8/t = 0.001\%$ [50], $d = 9/t = 0.001\%$ [49] or $d = 10/t = 0.1\%$ [51].

3.5 Mathematical Definition of the Path-Exchange Algorithm

Here, we provide the first ever formal mathematical description of the path-exchange algorithm. In Section 4, we will use this description to prove that the path-exchange method is equivalent to the tiered hybrid method. To show this, we will simply perform a structural path analysis on a matrix-based hybrid system and compare it to the formal description below.

As described in Section 3.1, a structural path analysis of the sectoral system and the process system is first performed. The key to a formal mathematical description of the algorithm is now the description of the logic it employs to add only some of the nodes of the sectoral system to the process system. The resulting paths then contain only the *"mutually exclusive nodes"* often cited as the distinction of the path-exchange method [11, Sec. 2][2, Sec.4][28, Sec.2.1].

As we can see in Fig. 3, only those paths from the sectoral SPA for which there is no corresponding process path are added. Consider, for instance, all first-order paths into sector $\boxed{A}$ in the example system of Fig. 1:

$$\sum_{j=1}^{N} b_j^S a_{jS(1)}^S = b_D^S a_{DA}^S + b_B^S a_{BA}^S \tag{47}$$

Here, the index subscript notation $S(1)$ refers to the sector which contains process 1. In the example system of Fig. 1, $S(1) \rightarrow A$ and therefore the sum $\sum_{j=1}^{N} a_{jS(1)}^S = a_{DA} + a_{BA}$.

First, we must determine whether there is any process with flows to process ① originating in sector $\boxed{B}$. As we can see from Fig. 1, process path a_{21}^P corresponds to sectoral path a_{BA}^S. Under the definition of the algorithm, this sectoral path must therefore not be added.

Again availing ourselves of the Iverson bracket notation Eq. (9) and the definition of the concordance matrix Eq. (8), we can write this condition as

$$\left[\sum_{i=1}^{M} h_{Bi} a_{i1}^P \stackrel{?}{=} 0\right] = \begin{cases} 1 & \text{if no process flow } a^P \text{ to process 1 originates in sector } B \\ 0 & \text{else} \end{cases} \tag{48}$$

The formal definition of the algorithm is now provided in Eq. (49). As we can see, the condition of Eq. (48) is used whenever sectoral paths are appended to a process path.

$$
\begin{aligned}
m_{1,pxc} = {} & \underbrace{\sum_{j=1}^{M} b_j^P \delta_{j1}}_{P:0^{\mathrm{th}}-\mathrm{order}} + \qquad (49) \\
& + \underbrace{\sum_{j=1}^{M} b_j^P a_{j1}^P}_{P:1^{\mathrm{st}}-\mathrm{order}} + \underbrace{\sum_{j=1}^{N} b_j^S \left[\sum_{x=1}^{M} h_{jx} a_{x1}^P \stackrel{?}{=} 0\right] a_{jS(1)}^S}_{S:1^{\mathrm{st}}-\mathrm{order}} + \\
& + \underbrace{\sum_{k=1}^{M}\sum_{j=1}^{M} b_k^P a_{kj}^P a_{j1}^P}_{P:2^{\mathrm{nd}}-\mathrm{order}} + \underbrace{\sum_{k=1}^{N}\sum_{j=1}^{M} b_k^S \left[\sum_{x=1}^{M} h_{kx} a_{xj}^P \stackrel{?}{=} 0\right] a_{kS(j)}^S a_{j1}^P}_{S/P:2^{\mathrm{nd}}-\mathrm{order}} + \underbrace{\sum_{k=1}^{N}\sum_{j=1}^{N} b_k^S a_{kj}^S \left[\sum_{x=1}^{M} h_{jx} a_{x1}^P \stackrel{?}{=} 0\right] a_{jS(1)}^S}_{S:2^{\mathrm{nd}}-\mathrm{order}} + \\
& + \underbrace{\sum_{l=1}^{M}\sum_{k=1}^{M}\sum_{j=1}^{M} b_l^P a_{lk}^P a_{kj}^P a_{j1}^P}_{P:3^{\mathrm{rd}}-\mathrm{order}} + \underbrace{\sum_{l=1}^{N}\sum_{k=1}^{N}\sum_{j=1}^{N} b_l^S a_{lk}^S a_{kj}^S \left[\sum_{x=1}^{M} h_{jx} a_{x1}^P \stackrel{?}{=} 0\right] a_{jS(1)}^S}_{S:3^{\mathrm{rd}}-\mathrm{order}} + \\
& + \underbrace{\sum_{l=1}^{N}\sum_{k=1}^{N}\sum_{j=1}^{M} b_l^S a_{lk}^S \left[\sum_{x=1}^{M} h_{kx} a_{xj}^P \stackrel{?}{=} 0\right] a_{kS(j)}^S a_{j1}^P + \sum_{l=1}^{N}\sum_{k=1}^{M}\sum_{j=1}^{M} b_l^S \left[\sum_{x=1}^{M} h_{lx} a_{xk}^P \stackrel{?}{=} 0\right] a_{lS(k)}^S a_{kj}^P a_{j1}^P}_{S/P:3^{\mathrm{rd}}-\mathrm{order}} + \dots
\end{aligned}
$$

4 Proof of Mathematical Equivalency with Matrix Method

Despite previous reports in literature, the path-exchange method for hybrid life-cycle assessment does not constitute a distinct mathematical approach for integrating process and sectoral data. This has not been observed previously, as is evident by the treatment of this method in recent review publications [4][2]. In this section, we show the mathematical equivalence between the path-exchange method we formally defined in Section 2.4 and the matrix method we introduced in Section 2.3.

More specifically, we show the equivalence of the matrix method with binary double-counting correction where $\mathbf{C}^d = 0 \wedge \mathbf{C}^u \neq 0$.

Starting from the governing equation of the integrated hybrid matrix method in Eq. (15) and the governing equation of the path-exchange hybrid method in Eq. (21), we need to show that

$$\begin{aligned}\vec{e}_{H(pxc)} &= \mathrm{PXC}(\mathbf{B}_c^S(\mathbf{I}-\mathbf{A}^S)^{-1})\vec{f}^P = \\ &= \mathrm{PXC}(\mathbf{B}_c^S(\mathbf{I}+\mathbf{A}^S+(\mathbf{A}^S)^2+(\mathbf{A}^S)^3+\dots))\vec{f}^P \qquad (50)\end{aligned}$$

$$= \vec{e}_{H(mx)} = \begin{pmatrix}\mathbf{B}^P & 0\\ 0 & \mathbf{B}_c^S\end{pmatrix}\begin{pmatrix}\mathbf{A}^P & 0\\ \mathbf{C}_{corr}^U & \mathbf{I}-\mathbf{A}^S\end{pmatrix}^{-1}\begin{pmatrix}\vec{f}^P\\ 0\end{pmatrix} \qquad (51)$$

In order to show the equivalence of Eq. (50) and Eq. (51), we must conduct a structural path analysis of the hybrid matrix in Eq. (51). The trick is to simply split up the row/column index into two indices, which go over $1 \leq i < M$ (processes) and $M \leq i \leq N$ (sectors). Note also that by the definition of the tiered hybrid matrix introduced in Section 2.3, the downstream flow matrix is zero and therefore all terms $\sum_{i=1}^{M}\sum_{j=M+1}^{N} a_{ij} = 0$. For convenience, the range of the row/column index combinations (i, j) is shown in Eq. (52):

$$\left[\begin{array}{c|c} 1 \leq i \leq M & 1 \leq i \leq M \\ 1 \leq j \leq M & M < j \leq N \\ \hline M < i \leq N & M < i \leq N \\ 1 \leq j \leq M & M < j \leq N \end{array}\right] \qquad (52)$$

Note that in the upstream quarter of the matrix, where $M < i \leq N \wedge 1 \leq j \leq M$ the binary double-counting correction method from Eq. (10) must be applied. This means, that instead of the term

$$\sum_{i=M+1}^{N}\sum_{j=1}^{M} a_{ij}^H \qquad (53)$$

in the decomposition, we get

$$\sum_{i=M+1}^{N}\sum_{j=1}^{M}\left[\sum_{k=1}^{M} h_{ik}a_{kj}^P \stackrel{?}{=} 0\right]a_{ij}^H \qquad (54)$$

Splitting up the indices, we can write:

$$
\begin{aligned}
m_{1,mx} = & \underbrace{\sum_{j=1}^{M} b_j^H \delta_{j1} + \cancel{\sum_{j=M+1}^{N} b_j^H \delta_{j1}}}_{H:0^{\mathrm{th}}-\mathrm{order}} \\
& + \underbrace{\sum_{j=1}^{M} b_j^H a_{j1}^H + \sum_{j=M+1}^{N} b_j^H \left[\sum_{x=1}^{M} h_{jx} a_{x1}^P \overset{?}{=} 0 \right] a_{j1}^H}_{H:1^{\mathrm{st}}-\mathrm{order}} \\
& + \underbrace{\sum_{k=1}^{M} \sum_{j=1}^{M} b_k^H a_{kj}^H a_{j1}^H + \sum_{k=M+1}^{N} \sum_{j=1}^{M} b_k^H \left[\sum_{x=1}^{M} h_{kx} a_{xj}^P \overset{?}{=} 0 \right] a_{kj}^H a_{j1}^H + \sum_{k=M+1}^{N} \sum_{j=M+1}^{N} b_k^H a_{kj}^H \left[\sum_{x=1}^{M} h_{jx} a_{x1}^P \overset{?}{=} 0 \right] a_{j1}^H}_{H:2^{\mathrm{nd}}-\mathrm{order}} \\
& + \underbrace{\sum_{l=1}^{M} \sum_{k=1}^{M} \sum_{j=1}^{M} b_l^H a_{lk}^H a_{kj}^H a_{j1}^H + \sum_{j=M+1}^{N} \sum_{k=M+1}^{N} \sum_{l=M+1}^{N} b_l^H a_{lk}^H a_{kj}^H \left[\sum_{x=1}^{M} h_{jx} a_{x1}^P \overset{?}{=} 0 \right] a_{j1}^H}_{H:3^{\mathrm{rd}}-\mathrm{order}} \\
& + \underbrace{\sum_{l=M+1}^{N} \sum_{k=M+1}^{N} \sum_{j=1}^{M} b_l^H a_{lk}^H \left[\sum_{x=1}^{M} h_{kx} a_{xj}^P \overset{?}{=} 0 \right] a_{kj}^H a_{j1}^H + \sum_{l=M+1}^{N} \sum_{k=1}^{M} \sum_{j=1}^{M} b_l^H \left[\sum_{x=1}^{M} h_{lx} a_{xk}^P \overset{?}{=} 0 \right] a_{lk}^H a_{kj}^H a_{j1}^H}_{H:3^{\mathrm{rd}}-\mathrm{order}} + \ldots
\end{aligned}
\tag{55}
$$

Comparing this to Eq. (49), we find all paths to be equal. □

This means that the power-series expansion of the system in Eq. (19) of a hybrid matrix like Eq. (15), which is constructed according the definition of the *tiered hybrid* method with binary double-counting correction, is equivalent to the paths returned by the *path-exchange hybrid* method.

5 Example

We illustrate the proof of mathematical equivalency from Section 4 by explicitly writing out all possible paths up to order 2 for the example system in Fig. 1. To simplify the equations, we assume the case of only a single environmental burden of interest (eg. carbon dioxide emissions). In addition, we place only a single unit of final demand on process 1, which is contained in sector A through a final demand vector

$$\vec{f}_P = \begin{bmatrix} 1 \\ 0 \end{bmatrix}$$

Using the definition of the path-exchange algorithm we introduced in Eq. (49) of Section 3.5, for paths up to order 2, the governing equation of the method can be written as

$$e_{1,H(pxc)} \sim \mathrm{PXC}\left\{ \sum_{j=1}^{N=4} b_j \left(\delta_{j1} + a_{j1}^S + \sum_{k=1}^{N=4} a_{jk}^S a_{k1}^S \right) \right\} \tag{56}$$

For illustrative purposes, we first separately write out the results of the structural path analyses of the process system and the sectoral system. The sub-paths of the sectoral system for which there is no correspondence with any sub-paths of the process system are underlined for better visibility.

$$\begin{aligned} \mathrm{SPA}(S) \sim & b_A^S + \\ & + \underline{b_A^S a_{AA}^S} + b_B^S a_{BA}^S + \underline{b_D^S a_{DA}^S} + \\ & + \underline{b_A^S a_{AA}^S a_{AA}^S} + \underline{b_A^S a_{AB}^S} a_{BA}^S + \underline{b_B^S a_{BA}^S a_{AA}^S} + \underline{b_B^S a_{BB}^S} a_{BA}^S + \underline{b_C^S a_{CB}^S} a_{BA}^S + \underline{b_C^S a_{CD}^S a_{DA}^S} + \underline{b_D^S a_{DD}^S a_{DA}^S} \end{aligned} \tag{57}$$

$$\begin{aligned} \mathrm{SPA}(P) \sim & b_1^P + \\ & + b_2^P a_{21}^P + \\ & + b_3^P a_{32}^P a_{21}^P \end{aligned} \tag{58}$$

The path-exchange algorithm now goes to work and returns:

$$\begin{aligned} e_{1,H(pxc)} \sim & b_1^P + \\ & + \underline{b_A^S a_{AA}^S} + b_2^P a_{21}^P + \underline{b_D^S a_{DA}^S} \\ & + \underline{b_A^S a_{AA}^S a_{AA}^S} + \underline{b_A^S a_{AB}^S} a_{21}^P + \underline{b_B^S a_{BA}^S a_{AA}^S} + \underline{b_B^S a_{BB}^S} a_{21}^P + \underline{b_C^S a_{CB}^P} a_{21}^P + \underline{b_C^S a_{CD}^S a_{DA}^S} + \underline{b_D^S a_{DD}^S a_{DA}^S} \end{aligned} \tag{59}$$

Now, we construct the hybrid matrix of the example. This will allow us to perform a structural path analysis on the hybrid matrix and compare it to Eq. (59).

$$\mathbf{A}_H = \begin{array}{c} \\ 1 \\ 2 \\ A \\ B \\ C \\ D \end{array} \left[\begin{array}{cc|cccc} 1 & 2 & A & B & C & D \\ 0 & 0 & 0 & 0 & 0 & 0 \\ a_{21}^P & 0 & 0 & 0 & 0 & 0 \\ \hline c_{A1}^U = a_{AA}^S & c_{A2}^U = a_{AB}^S & a_{AA}^S & a_{BA}^S & 0 & 0 \\ c_{B1}^U = 0 (DC!) & c_{B2}^U = a_{BB}^S & a_{BA}^S & a_{BB}^S & a_{BC}^S & 0 \\ 0 & c_{C2}^U = a_{CB}^S & 0 & a_{CB}^S & a_{CC}^S & a_{CD}^S \\ c_{D1}^U = a_{DA}^S & 0 & a_{DA}^S & 0 & a_{DC}^S & a_{DD}^S \end{array} \right]$$

We can now perform the structural path analysis on this matrix:

$$e_{1,H(mx)} \sim \sum_{j=1}^{M+N=2+4} b_j^H \left(\delta_{j1} + a_{j1}^H + \sum_{k=1}^{M+N=2+4} a_{jk}^H a_{k1}^H \right) \tag{60}$$

$$\begin{aligned} =&b_1^H + \\ &+ b_2^H a_{21}^H + b_3^H a_{31}^H + b_6^H a_{61}^H + \\ &+ b_3^H a_{32}^H a_{21}^H + b_4^H a_{42}^H a_{21}^H + b_5^H a_{52}^H a_{21}^H + b_4^H a_{43}^H a_{31}^H + b_3^H a_{33}^H a_{31}^H + b_5^H a_{56}^H a_{61}^H + b_6^H a_{66}^H a_{61}^H \end{aligned} \tag{61}$$

$$\begin{aligned} =&b_1^P + \\ &+ b_2^P a_{21}^P + b_A^S c_{A1}^U + b_D^S c_{D1}^U + \\ &+ b_A^S c_{A2}^U a_{21}^P + b_B^S c_{B2}^U a_{21}^P + b_C^S c_{C2}^U a_{21}^P + b_A^S a_{AA}^S c_{A1}^U + b_B^S a_{BA}^S c_{A1}^U + b_C^S a_{CD}^S c_{D1}^U + b_D^S a_{DD}^S c_{D1}^U \end{aligned} \tag{62}$$

$$\begin{aligned} =&b_1^P + \\ &+ b_2^P a_{21}^P + b_A^S a_{AA}^S + b_D^S a_{DA}^S \\ &+ b_A^S a_{AB}^S a_{21}^P + b_B^S a_{BB}^S a_{21}^P + b_C^S a_{CB}^S a_{21}^P + b_A^S a_{AA}^S a_{AA}^S + b_B^S a_{BA}^S a_{AA}^S + b_C^S a_{CD}^S a_{DA}^S + b_D^S a_{DD}^S a_{DA}^S \end{aligned} \tag{63}$$

Comparing Eq. (59) and Eq. (63), we find them to be equal. Note that the paths listed in these equations can be also visually traced in the diagrammatic representation of the example system in Fig. 1.

6 Computational Intensity

Finally, we find that the path-exchange method for hybrid life-cycle assessment is significantly more computationally expensive than any matrix-based method. This is because covering a large enough number paths to obtain a high degree of emissions coverage in life-cycle assessment is essential in the context of decision-making [43]. As we have previously discussed, the number of paths grows exponentially with the size of the system. What is more, number of paths required to obtain reasonable coverage of total emissions Number of required paths depends strongly on the system under investigation [45].

In Panel A of Fig. 6, we show that for the simple case of a single-region input-output table of only 114 sectors, computation on current high-end consumer hardware may for some sectors take $2hrs$ while covering only 50% of total emissions in the computed paths. While is evident that for some sectors the structural path analysis does indeed converge quickly, for others the convergence behavior is very poor. Even computational optimizations such as parallelization cannot compensate for poor convergence behavior. In Panel B of Fig. 6 on the other hand, we show that the exact solution of a hybrid system system combining Ecoinvent and the Exiobase multi-regional input-output table of a combined 30'800 rows/columns can be computed exactly within $\sim 3min$ on the same hardware. For these solutions, we found excellent numerical stability to within floating-point precision for all calculations. The use of a path-based method, such as the path-exchange method, therefore confers no computational advantage.

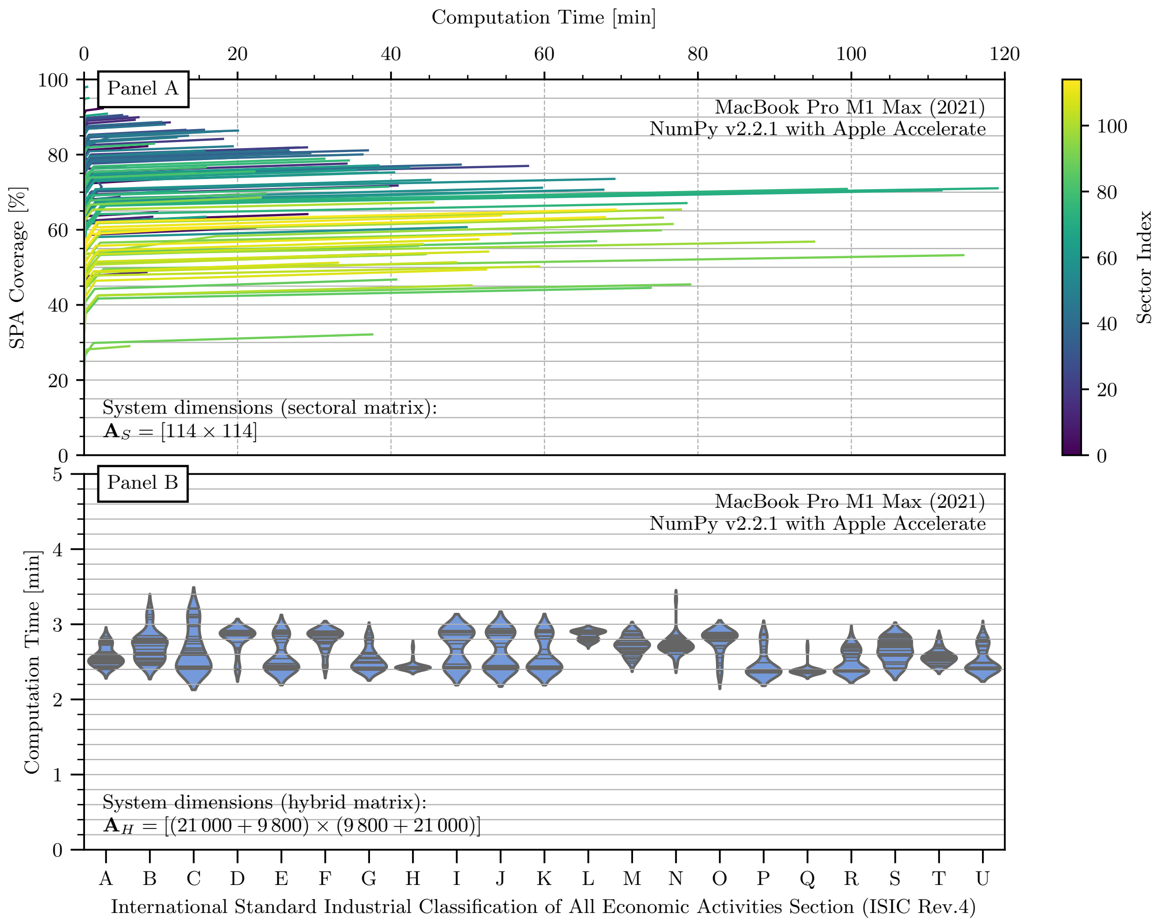

Figure 6: Computational intensity of the path-exchange (graph-based) hybrid life-cycle method and the tiered (matrix-based) life hybrid life-cycle method. *Panel A*: Convergence behavior of the environmental burden coverage from a structural path analysis for every sector in the input-output table of Australia. Every line represents a single sector. The maximum path length was set to 20, with the cut-off criteria varied between [0.1%, 0.01%, 0.001%, 0.0001%, 0.00001%]. In general, high SPA coverage in short computation time is desirable. It is evident that the convergence behavior strongly depends on the sector and can vary between $> 90\%$ in $\sim 5min$ to $< 50\%$ in $\sim 2hrs$ on current hardware. Note that this system is two orders of magnitude smaller than the hybrid system of Panel B. Computations were preformed using the `pyspa` [52] package (v2.4) on a MacBook Pro with an M1 Max CPU and NumPy (v2.2.1). *Panel B*: Computation time for the solution of the governing equation of hybrid life-cycle assessment $e = \mathbf{Q}_H \cdot \mathbf{B}_H \cdot \mathbf{A}_H^{-1} \cdot \vec{f}$ using the `numpy.linalg.solve(a,b)` function. The hybrid matrix was constructed by combining the Ecoinvent technosphere matrix of $\dim(\mathbf{A}_P) = [21'000 \times 21'000]$ and the multi-regional input-output table of the Exiobase 3 database [53] of $\dim(\mathbf{A}_S) = [9'800 \times 9'800]$. A sample of 10 Ecoinvent processes per ISIC section (A-U) was selected at random to serve as final demand. The numerical stability of every solution was checked by repeating the same computation 4 times. For every computation, the solution was found to be stable within the precision of the standard NumPy floating point data type. Note that this system is two orders of magnitude larger than the single-region input-output table of Panel A. Computations were preformed using NumPy v2.2.1. built against the Apple Accelerate BLAS framework on a MacBook Pro with an M1 Max CPU. All underlying data is available in a Zenodo repository [54].

7 Discussion

In Section 3.2, we provide a description of the path-exchange method supported by our novel diagrammatic notation introduced in Fig. 1. Following a short historical overview in Section 3.2, we show in Section 3.3 that the key assumption underlying the motivation for the development of the algorithm was incorrect. In Eq. (49) of Section 3.5 we then provide a concise mathematical description of the path-exchange method [10][11][28] for hybrid life-cycle assessment. This description makes use of a concordance matrix and the Iverson bracket. To our knowledge, no such definition has been provided to date. In order to augment the original visual illustrations of the algorithm provided by the authors in [11, Figure 2] and [28, Figure 1], a novel diagrammatic illustration is provided in Fig. 3. Finally, in Section 4 we show that the path-exchange (=graph-based) method for hybrid life-cycle assessment is mathematically equivalent to the tiered hybrid (=matrix-based) method for hybrid life-cycle assessment. The core of the proof is straightforward: A structural path analysis of the hybrid matrix compiled according to the tiered hybrid method is performed. Splitting up the indices of the matrix multiplication in the power-series expansion, it then becomes clear that the resulting paths are equivalent to those of the path-exchange method. An explicit example based on the system illustrated in Fig. 1 is provided in Section 5.

From Section 3.3 and Section 4, we can see that the frequently invoked argument of the path-exchange method working on *"mutually exclusive paths"* [11][28] is not an inherent property of the method. Instead it is a result of the algorithm making implicit use of binary double-counting correction. By extension, the claim that the method somehow avoids the problem of double-counting [14, Sec. 2.6.3][4, Table 4] is insubstantial. The method avoids instances of double-counting only through its implicit use of binary double counting correction. Finally, we can see that the purported advantage of avoiding *(...) the need to collect data and make assumptions that would be needed to populate the so-called upstream and downstream cut-off matrices (...) [which] makes the process more efficient as only the most significant nodes are modified."* [28, Sec. 2.1] is void: Downstream cut-off coefficients are not considered simply by definition of the path-exchange method algorithm. On the other hand, all information which the practitioner of the path-exchange method has on specific processes can easily be arranged into a matrix - the upstream cut-off matrix.

8 Conclusion

Ultimately, practitioners should be acutely aware of the inherent limitations of the path-exchange method we discuss in Section 3.4 and Section 6. While we have shown the tiered hybrid method and the path-exchange hybrid methods to be equivalent *in principle*, this holds true only in the case where the power series expansion of Eq. (49) is considered *ad infinitum* - a practical impossibility. We therefore suggest that the use of the former method is more prudent, since it avoids all these limitations by definition. This use of a matrix-based hybrid life-cycle assessment method should be preferred by practitioners, even in the case where individual paths are of interest, rather than just numerical value of the environmental burden. This is because a matrix method can also capture modifications at specific locations in the supply chain - and is computationally superior. If required, a structural path analysis can always be conducted on the tiered hybrid matrix, as we have shown in Section 4. This allows practitioners to determine the supply chain nodes with the highest overall environmental impact.

We hope that our formal treatment of the path-exchange method will provide some much-needed clarity in the ongoing discussion surrounding the specific properties and applicability of methods for hybrid life-cycle assessment. It is our hope that work toward a unified theory of methods will continue, ultimately providing a sound mathematical for the development of open-source tools, which can be integrated into mainstream software for life-cycle assessment.

9 Funding

This work has been supported by the Swiss Innovation Agency Innosuisse in the context of the WISER flagship project (PFFS-21-72). In addition, Michael P. Weinold gratefully acknowledges the support of the Swiss Study Foundation.

10 Code Availability

The source code used for generating the data visualized in panels A and B of Fig. 6 is available under a permissive license from a Zenodo repository [54].

11 Data Availability Statement

Part of the data that support the findings of this study are openly available, with links provided in a Zenodo repository [54]. Part of the data that support the findings of this study are available from Ecoinvent. Restrictions apply to the availability of these data, which were used under license for this study.

12 Acknowledgements

The authors would like to thank Dr. Arthur Jakobs (Laboratory for Energy Systems Analysis, PSI Centers for Nuclear Engineering & Sciences and Energy & Environmental Sciences, Paul Scherrer Institute (PSI)) for helpful discussions related to the definition of the path-exchange method. The authors would also like to thank Dr. Benedict Bauer (formerly Vienna University of Technology and University of Vienna) for helpful discussions related to the properties of elementary paths and walks in the graph of economics sectors. The authors would also like to thank the team of the joint library for the Research Institutes within the ETH Domain (Lib4RI) for their tireless assistance in obtaining digitized copies of many early publications of hybrid life-cycle assessment. Finally, the authors would also like to thank Mr. Philipp Seeman for making a complete scan of his 1984 thesis available to us with the help of Ms. Rachel Rogers of the Department of Chemical & Process Engineering at the University of Canterbury, New Zealand.

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