ORIGINAL PAPER

Using Yield and Entropy-Based Characteristics for Circular Economy

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Abstract

Environmental impacts of the extant linear carbon economy and aspects of conservation of resources demand a transformation to a circular carbon economy (CCE). In view of this transformation, carbon-containing plastic products should be reused and recycled to prevent or minimize the release of their carbon content into the environment. Different plastic waste feedstock recycling strategies are applicable, with different degrees of feedstock destruction, depending on the degree of degradation and contamination of the intended recycle fractions. The evaluation of the effectiveness of recycling processes by substance and carbon-based yield and entropic characteristics could be a part of the overall evaluation strategy for recycling processes. Possible principles and base equations of such substance and carbon-based yield and entropic characteristics, extracted from the literature and adapted, are delineated in this article. Substance-based characteristics could be applied for physical recycling processes in which the aspired recovery substances remain preserved and are physically separated. A resort to carbon-based characteristics could be practiced for recycling and combustion processes, in which the feedstock is chemically destroyed, and new substances are possibly synthesized. Stylized process examples depict the way of a joint usage of yield and entropic characteristics.

Abbreviations

A, B	Shortcut for target substance A (recyclate) of mechanical recycling and for residual substance B
C	Carbon, as index: carbon-based characteristic

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c	Mass concentration [kg j/kg stream] of conserved item j in process inlet or outlet streams
CCE	Circular carbon economy
CE	Circular economy
CR	Chemical recycling
CRR	Carbon recycling rate
DAC	Direct air capture
DSD	Duales System Deutschland
eSEA	Extended statistical entropy analysis
f_in, f_out	Number of relevant process inlet respectively outlet streams for SEA
f_recov	Number of the intended process recovery streams
HDPE	High density polyethylene
i	Added index for the intended C-containing target substance i in recovery stream k_recov
j	Substance j (here j = PET or PP or carbon C) considered for SEA and for recovery
k_in, k_out	Indices of relevant process inlet respectively outlet streams for SEA
k_recov	Indices of aspired recovery streams (subset of process outlet streams k_out) for the considered j
$\dot{m}$	Relevant mass stream [kg/h] for SEA which pass over the considered process battery limits
M	Substance j based stream frequency
MFA	Material flow analysis
MTO	Methanol to olefins synthesis
PET	Polyethylene terephthalate
PP	Polypropylene
RDF	Refuse derived fuel
RE	Recycling effectiveness
RSE	Relative statistical entropy
RR	Recovery rate, recycling rate
SCE	Substance concentration efficiency (= substance collection efficiency)
SDN	Substance dilution number (= RSE)
SE	Statistical entropy
SEA	Statistical entropy analysis
SLF	Shredder light fraction
WTE	Waste-to-energy facility
X	Mass load of j via inlet or outlet stream k_in or k_out [kg j in stream / kg process throughput of j

Introduction

Different defined conventional yield characteristics are traditionally used to evaluate the success of physical separation and chemical synthesis processes. In addition, entropic characteristics based on the statistical "Shannon entropy" were developed in the last decades and likewise applied for the evaluation of separation and synthesis processes, especially in the context of waste treatment and the recovery and recycling of substances. The methodologies of statistical entropy analysis (SEA) feature different degrees of complexity for different scopes

of application (see, e.g., [1–37]). Conventional yield characteristics and entropic characteristics could be competitively juxtaposed as instruments for the evaluation of processes, but they could also be supplementary used for process optimization and quantification of substance losses. This conjunction is pursued in the "Waste4Future" project of the Fraunhofer Society as part of the evaluation of recycling processes. Outlines of conventional and entropic characteristics are presented in the following paragraphs, followed by the formulation of the entropy calculation methods as used in the project. Stylized calculation examples are showcased to depict the way of the application of SEA and yields in the project. Carbon-based entropic deteriorations were calculated in the project if the feedstocks are burnt in waste-to-energy facilities (WTE) or used in traditional chemical recycling processes with partial feedstock combustion.

Outline of Conventional Substance and Carbon Yield Characteristics for Physical and Chemical Recycling

Mechanical recycling processes and other physical recycling processes, like selective extraction, are used if comparably clean waste feedstocks with high content of nondegraded desired plastic substances are available [35]. Different conventional yield characteristics could be applied as indicator for the plastic substance separation success, as, e.g., recycling rates (RR) related to the complete recovery mass stream $\dot{m}_{k\_recov}$ containing the desired substance polyethylene terephthalate (PET) [32, 33]. Other recovery rates (RR) could also be applied, e.g., Eq. (1), based on Eq. (11), which only relates to the desired target plastic substance j mass stream $\dot{m}_{k\_recov,j}$ in one of the process outlet streams $\dot{m}_{k\_out}$ which is declared as the allotted recovery stream $\dot{m}_{k\_recov}$ for the substance j, and with $\dot{m}_{k\_recov,j}$ based on Eq. (6) applied to this declared recovery outlet $\dot{m}_{k\_out} = \dot{m}_{k\_recov}$. Undesired contaminations in $\dot{m}_{k\_recov}$ by other substances $\neq$ j is thereby excluded in the nominator of Eq. (1). The process inlet $\dot{m}_{in,j} = \dot{m}_j$ of the target substance j possibly occurs via more than one inlet stream $\dot{m}_{k\_in}$ according to Eqs. (5) and (7).

$$RR_{k\_recov,j} = \dot{m}_{k\_recov,j} / \dot{m}_j \tag{1}$$

Problems with plastic waste feedstock impurities, resulting necessary advanced pretreatments, degradation, and deterioration of mechanical properties of the recyclates (downcycling), are summed up, e.g., in [38].

For higher plastic waste feedstock contamination and degradation, or low feedstock content of desired recyclates, less or more destructive chemical recycling (CR) processes could be applied. Such processes would be, e.g., solvolysis, pyrolysis, and gasification [39]. As stated by Lange in [40], p. 15,734: "Chemical recycling will likely take over, where mechanical recycling is struggling, e.g., in delivering top-grade products and recycling mixed, contaminated, or degraded reject streams." Overviews about different physical and chemical recycling processes are given in [40–44], some realized CR processes and their capacities are included in [41, 42].

The recovery of carbon via desired main products and desired byproducts of CR processes can be judged by conventional process yield characteristics such as the carbon recycling rate (CRR) proposed in [42], which is the relation between carbon process outlet via target products i and carbon inlet via feedstock:

$$CRR = \dot{m}_{target\ product\ outlets,C} / \dot{m}_{waste\ feedstock\ inlet,C} \tag{2}$$

CRR is a measure for the carbon loop closure in the CCE context because of its inverse relationship to process CO_2 emissions [42]. In CR processes, the degraded plastic waste feedstock represents the main but possibly not the only carbon process inlet. Other carbon-containing inlets (support fuels, reactor solvents, absorbents, etc.) could also be involved, as far as these supplementary inlets materially participate in the chemical conversion and/or carbon redistribution processes. In view of embracing the complete process C source $\dot{m}_{in,C} = \dot{m}_C$, yield characteristics termed carbon recovery rates $RR_{k\_recov,C,i}$ by aspired carbon-containing target items i in allotted recovery streams $\dot{m}_{k\_recov}$ are proposed via Eq. (3), which are analogous to Eq. (1), for j = C (carbon). The supplementary index i in Eq. (3) shall imply that the only aspired C sink in these specified recovery streams is this target item i, a specified substance i or a carbon-containing defined substance mixture i. Carbon via possible and undesired carbon-containing contaminants ≠ i in this recovery stream are excluded in Eq. (3). Such undesired contaminations are aspired to be avoided, but nevertheless, they could unintentionally occur (e.g., caused by insufficient separation and purification operations within the investigated overall process). The process can have different allotted recovery streams for different C-containing target items i, each characterized by specific carbon recovery rates $RR_{k\_recov,C,i}$. It is conceivable that, e.g., for reasons of the need of different subsequent downstream processes, different process outlet recovery streams $\dot{m}_{k\_recov}$ for one and the same item i could exist, each with possibly different degree of quality, i.e., with possibly intentional parts of carbon-free associated diluting materials, each of these $\dot{m}_{k\_recov}$ lead to a different recovery rates $RR_{k\_recov,C,i}$ for the same item i according to Eq. (3). The summation of these different $RR_{k\_recov,C,i}$ (incl. possibly different $RR_{k,recov,C,i}$ for the same item i) would be a process overall carbon recovery rate as in Eq. (2), with all relevant C inlets in the denominator according to Eqs. (7) and (5), with j = C. Other conventional carbon yield characteristics could be defined. The inclusion of all carbon outlet parts in the outlet recovery streams $\dot{m}_{k\_out} = \dot{m}_{k\_recov}$, inclusively possible carbon-containing contaminants, would lead to the recovery stream carbon loads $X_{k\_recov,C}$ based on Eq. (11), applied to j = C. The yields $RR_{k\_recov,C,i}$ coincide with these $X_{k\_recov,C}$ if the item i is the only carbon-containing part in these recovery streams.

$$RR_{k\_recov,C,i} = \dot{m}_{k\_recov,C,i} / \dot{m}_C \tag{3}$$

Outline of SEA

SEA (with different complexity levels and extensions, different extent of the considered process battery limits) is used for the examination of the process distribution paths of different considered items, like chemical elements, plastic substances, components consisting of different substances, or complete products, since, "SEA is a method that quantifies the power of a system to concentrate or dilute substances," according to Brunner and Rechberger [3], p. 148, and [4], p. 195. Also, according to Sobańtka et al. [17], p. 1949: SEA quantifies the distribution of a substance among different material flows and after a process, and the change in the distribution of the substance indicates the concentrating power relative to the extent of dilution (dispersion) of the process. Examples for SEA application are the investigation of the distribution paths of metals [2, 5–12], nitrogen [12–18], phosphor [19, 20], carbon [21, 22], plastic recycling [32–37], and extensions to multicomponent products and product disassembling. incl. multilevel SEA [26–28, 34, 35], and the development of the Relative Product-inherent Recyclability RPR [29–31, 33]. An overview

of SEA levels and methods for plastic waste feedstock recycling is given, e.g., in [35]. SEA is also applied as tool for circular economy (CE) principles, cf., e.g., [23–35, 37].

The application of the SEA methodology requires the conservation of the considered item j within the process according to Eq. (7). The overall process input of the item j is mathematically balanced with its process outlet via process material flow analysis (MFA), e.g., [3, 4, 6, 7, 23]. In the Waste4Future project [39], this substance conservation (by non-destructive physical recycling processes) was given for the experiments for polypropylene (PP) recovery from shredder light fractions (SLF) und PET from refuse derived fuel (RDF) fractions, so that SEA analyses were carried out for these physical PP and PET recovery processes. For the examined and simulated CR processes with the complete destruction of the input feedstock substances and subsequent synthesis of new substances (simulated polyolefins namely high density polyethylene (HDPE) and PP synthesis in the project), the conservation of plastic substances in the feedstocks is not given: The investigated feedstocks according to Table 2 is destroyed in feedstock gasification units of the simulated CR processes, inclusively the destruction of the valuable feedstock parts such as PET and PP. In these cases, the SEA methodology was applied within the project for the distribution path of carbon (as considered item j) into (desired) valuable outlet products and byproducts i and the (undesired) distribution and dilution of carbon via process outlet exhaust and waste streams as losses, for usage in the carbon circular economy (CCE) context. Within the project, the SEA methodology is used together with the above-mentioned substance respectively carbon yield characteristics.

The process outlet distribution of the considered item j for (j = carbon for CR processes, j = PP, PET in case of physical recycling processes in the project) and possibly the process inlet state of this item contribute to entropic characteristics, contrary to conventional yield characteristics in which only a recovery of the items via allotted recovery streams is considered (disregarding possible contaminations in these recovery streams, the losses of recovery items by exhaust and waste side streams, furthermore disregarding different degrees of dilution of the item in these side streams).

The SEA methodology and its application to waste management and recycling processes was developed by Rechberger, et al. [1–4, 7, 9] based on the statistical Shannon entropy [45–47] of the information theory according to Eq. (4). The Shannon entropy (Greek symbol H, unit: Bit, respectively Shannon) is a measure of the uncertainty in the distribution of probabilities P_k of appearance of events k, and of the average information content of this distribution. The information content of the events k is presented by $-\log_2(P_k)$. The development of the Shannon entropy function in turn originates from Boltzmann's statistical description of entropy for physical applications [7]. There is a formal identity between the thermodynamic entropy as introduced by R. Clausius in 1856 and the statistical Shannon entropy; however, there is no direct physical relationship between the two entropy terms [7].

$$H = -\Sigma \, P_k \bullet \log_2(P_k) \qquad (\Sigma \, P_k = 1) \qquad (4)$$

The probabilities P_k of the events k are transformed into the probabilities of occurrence of the item j (chemical element or substance or defined substance mixtures) in the streams k (as events k), i.e., in those process inlets k = k_in, respectively, outlet streams k_out which are as sources and sinks related to the process internal item j redistribution, whereby the item j occurrence is expressed via item mass concentrations $c_{k\_in,j}$, respectively, $c_{k\_out,j}$ in these streams. The complexity of the derived expressions is influenced by the chosen units of the item j mass concentrations $c_{k,j}$ [7]. Simple

expressions could be achieved if $c_{k,j}$ are expressed as mass parts, kg/kg, as, e.g., in [13]. The transformation of Eq. (4) is separately to be done for the process inlet and outlet distribution of the considered item j. Two Eqs. (15) and (17) arise: for the relevant process inlet streams $\dot{m}_{k\_in}$ as well as for the relevant process outlet streams $\dot{m}_{k\_out}$. Inlet and outlet variables are indexed with "_in" and "_out" in the Eq. (5) and the subsequent equations. During the substitution of P_k by $c_{k\_in,j}$ and $c_{k\_out,j}$ a scaling is needed because, contrary to $\sum P_k = 1$, usually $\sum_{k\_in=1}^{f\_in} c_{k\_in,j} \neq 1$ and $\sum_{k\_out=1}^{f\_out} c_{k\_out,j} \neq 1$. An exceptional case is pictured in Fig. 10, in which the outlet mass streams are incidentally equal to the target substance j = A process throughput by mass, and subsequently the substance A outlet stream mass parts $c_{k\_out,A} = \dot{m}_{k\_out,A}/\dot{m}_{k\_out}$ in the f_out = 2 outlet streams coincide with the respective substance A loads of the streams $X_{k\_out,A} = \dot{m}_{k\_out,A}/\dot{m}_{out,A}$ according to Eqs. (1) and (11), here with the load in stream k_out = 1 as declared substance A recovery yield $RR_{1,A} = X_{1,A}$. This exceptionally results in $\sum_{k\_out=1}^{f\_out} c_{k\_out,A} = \sum_{k\_out=1}^{f\_out} X_{k\_out,A} = 1$. In this stylized process of Fig. 10, the matches $c_{k\_out,B} = X_{k\_out,B}$ and $\sum_{k\_out=1}^{f\_out} c_{k\_out,B} = \sum_{k\_out=1}^{f\_out} X_{k\_out,B} = 1$ also apply for the residual substance B. The item j loads $X_{k,j}$ of in the inlet and outlet streams k are defined according to Eqs. (10) and (11) [13, 16]. Exceptional cases $\sum_{k\_in=1}^{f\_in} c_{k\_in,j} = 1$ for process inlets are analogous, as depicted, e.g., in Fig. 1 in [13] or Fig. 2.1 in [16]. For general cases, the different amounts of mass streams $\dot{m}_{k\_in}$ and $\dot{m}_{k\_out}$ into and out of the considered process, which contain the target substance j, must be incorporated by a scaling. The scaling is carried out by means of standardized or specific mass fractions $M_{k,j}$ [7, 32, 33] = item j based frequencies $M_{k,j}$ of occurrence of the different mass streams $\dot{m}_k$ [13, 16]. Streams, each respectively imaginarily subdivided by such dedicated frequencies into imaginarily constructed process inlet bins with the bin size according to the target item j process throughput $\dot{m}_j$ by mass, and thereby repeated occurrences of the respective item j mass part $c_{k,j}$, are exemplarily pictured in Fig. 2 in [13] or Fig. 2.2 in [16]. These frequencies lead to a desired normalization $\sum_{k=1}^{f}(M_{k,j} \bullet c_{k,j}) = 1$, as in Eqs. (12) and (13).

Calculation Methods of the Used SEA

Used Set of Equations for Statistical Entropies (SE) and Entropic Substance Collection Efficiencies (SCE)

The target item j inlet and outlet mass streams $\dot{m}_{k\_in,j}$ and $\dot{m}_{k\_out,j}$ are related in Eqs. (5) and (6) to the respective inlets and outlets $\dot{m}_k$ via mass concentrations $c_{k,j}$ in kg/kg. The considered item must remain conserved in the process according to Eq. (7). The conserved throughput $\dot{m}_j$ is used in Eqs. (8) and (9) for calculating the item j-based frequencies $M_{k,j}$ of the occurrence of the relevant mass streams $\dot{m}_k$ into and out of the process, e.g., [13, 16]. The summation stream indices "q_in" and "q_out" for the relevant streams in Eqs. (8) and (9) are applied to distinct from the stream indices "k_in" and "k_out" which are fixed in these 2 equations. The indices "f_in" and "f_out" in Eqs. (7)–(17) represent at least the number of those relevant process inlets "k_in" respectively outlet streams "k_out" which contain the considered item j $(c_{k,j} > 0)$, and which are inlet and outlet parts for the process interior mass turnover with redistribution of item j. The inclusion (in the numbers "f_in" and "f_out") of further relevant inlets respectively outlet streams without content of item, $c_{k,j} = 0$, but which are source or sink parts of the process internal mass turnover and redistribution of item j, do not contribute to the

Eqs. (7)–(9), (12)–(17) because of $c_{k,j} = 0$ and Eqs. (19) and (20). However, an inclusion of such relevant inlets respectively outlet streams k with $c_{k,j} = 0$ is necessary in Eqs. (21)–(23) if the usage of target item related average concentrations and maximum statistical entropies $SE_{max,j}$ for a subsequent scaling to relative statistical entropies RSE's (substance dilution numbers SDN) according to Eqs. (24) and (25) is intended.

The specific loads $X_{k,j}$ in Eqs. (10) and (11) as interconnections between $M_{k,j}$ and $c_{k,j}$ (unit of $X_{k,j}$: kg item j in the specific inlet respectively outlet mass stream $\dot{m}_k$/kg overall process throughput $\dot{m}_j$ of the target item) provides the necessary scaling of the probability $c_{k,j}$ of appearance of item j in stream $\dot{m}_k$ with the frequency $M_{k,j}$ of occurrence of $\dot{m}_k$. The scaling results in the normalization according to Eqs. (12) and (13), separately for the relevant process inlets as well as outlet streams.

The normalization in Eqs. (12) and (13) is analog to the normalization of the prefactors P_k in Eq. (4). $X_{k,j}$ substitute these prefactors P_k of Eq. (4), which leads to the SE of item j, separately for process inlet and outlet, according to Eqs. (15) and (17). The information about the concentration (respectively the dilution) of item j as its probability of occurrence in the relevant streams k remains obvious via the information terms $-\log_2(c_{k,j})$.

Item j mass parts $c_{k\_in,j}$ and $c_{k\_out,j}$ in process inlets k_in and outlet mass streams k_out [kg item j / kg stream]:

$$c_{k\_in,j} = \dot{m}_{k\_in,j}/\dot{m}_{k\_in} \tag{5}$$

$$c_{k\_out,j} = \dot{m}_{k\_out,j}/\dot{m}_{k\_out} \tag{6}$$

The conservation of considered item j according to Eq. (7) must be fulfilled:

$$\dot{m}_j = \dot{m}_{in,j} = \sum_{k\_in=1}^{f\_in} \dot{m}_{k\_in,j} = \dot{m}_{out,j} = \sum_{k\_out=1}^{f\_out} \dot{m}_{k\_out,j} \tag{7}$$

Item j based stream frequencies $M_{k\_in,j}$ and $M_{k\_out,j}$ of process inlets k_in and outlet mass streams k_out:

$$M_{k\_in,j} = \dot{m}_{k\_in}/\dot{m}_j = \dot{m}_{k\_in}/\sum_{q\_in=1}^{f\_in} \dot{m}_{q\_in,j} = \dot{m}_{k\_in}/\sum_{q\_in=1}^{f\_in}\left(\dot{m}_{q\_in} \bullet c_{q\_in,j}\right) \tag{8}$$

$$M_{k\_out,j} = \dot{m}_{k\_out}/\dot{m}_j = \dot{m}_{k\_out}/\sum_{q\_out=1}^{f\_out} \dot{m}_{q\_out,j} = \dot{m}_{k\_out}/\sum_{q\_out=1}^{f\_out}\left(\dot{m}_{q\_out} \bullet c_{q\_out,j}\right) \tag{9}$$

Item j loads $X_{k\_in,j}$ and $X_{k\_out,j}$ of process inlets k_in and outlet mass streams k_out:

$$X_{k\_in,j} = M_{k\_in,j} \bullet c_{k\_in,j} = \dot{m}_{k\_in,j}/\dot{m}_j \tag{10}$$

$$X_{k\_out,j} = M_{k\_out,j} \bullet c_{k\_out,j} = \dot{m}_{k\_out,j}/\dot{m}_j \tag{11}$$

$$\sum_{k\_in=1}^{f\_in} X_{k\_in,j} = \sum_{k\_in=1}^{f\_in}(M_{k\_in,j} \bullet c_{k\_in,j}) = 1 \tag{12}$$

$$\sum_{k\_out=1}^{f\_out} X_{k\_out,j} = \sum_{k\_out=1}^{f\_out}(M_{k\_out,j} \bullet c_{k\_out,j}) = 1 \tag{13}$$

Item j-based statistical entropies SE (process inlet and outlet), substance collection efficiency SCE:

$$SE_{k\_in,j} = -X_{k\_in,j} \bullet \log_2(c_{k\_in,j}) \tag{14}$$

$$SE_{in,j} = \sum_{k\_in=1}^{f\_in} SE_{k\_in,j} = -\sum_{k\_in=1}^{f\_in} X_{k\_in,j} \bullet \log_2(c_{k\_in,j}) \tag{15}$$

$$SE_{k\_out,j} = -X_{k\_out,j} \bullet \log_2(c_{k\_out,j}) \tag{16}$$

$$SE_{out,j} = \sum_{k\_out=1}^{f\_out} SE_{k\_out,j} = -\sum_{k\_out=1}^{f\_out} X_{k\_out,j} \bullet \log_2(c_{k\_out,j}) \tag{17}$$

$$SCE_j = (SE_{in,j} - SE_{out,j})/SE_{in,j} = 1 - SE_{out,j}/SE_{in,j} \tag{18}$$

$$\lim_{c_{k,j}\downarrow 0} c_{k,j} \bullet \log_2(c_{k,j}) = \lim_{v\to\infty} 2^{-v} \bullet \log_2(2^{-v}) = -\lim_{v\to\infty} \bullet \frac{v}{2^v} \bullet \log_2(2) = -\lim_{v\to\infty} \bullet \frac{1}{v \bullet 2^{v-1}} = 0 \tag{19}$$

$$\lim_{c_{k,j}\downarrow 0} X_{k,j} \bullet \log_2(c_{k,j}) = M_{k,j} \bullet \lim_{c_{k,j}\downarrow 0} c_{k,j} \bullet \log_2(c_{k,j}) = 0 \tag{20}$$

Lowest item j-based inlet or outlet entropies $SE_{in,min,j} = 0$ Bit or $SE_{out,min,j} = 0$ Bit appear if item j only occurs enriched to 100wt.% concentration, $c_{k\_in,j} = 1kg/kg$ (or $c_{k\_out,j} = 1kg/kg$), in one or more of the inlets (or outlet streams), and is completely absent $(c_{k,j} = 0kg/kg)$ in other relevant inlet (or outlet streams). Minimum outlet values $SE_{out,min,PP} = 0$ Bit and $SE_{out,min,PET} = 0$ were practically achieved within the Waste4Future project in experiments for physical PP recovery from shredder light fractions (SLF) and PET from Duales System Deutschland (DSD) fractions. Because carbon (j=C) is part of chemical substances, $SE_{out,min,C} = 0$ Bit is only possible if pure carbon outlet streams should be the hypothetical goal and the only C-containing outlet streams of a chemical recovery process (see possible substance i related $SE_{out,min,C} = SE_{min,C,i}$ in Table 1 for different intended carbon-containing substances i as the only carbon-containing process outlet).

Scaling the Entropies with Substance j Average Concentrations and Resulting Maximum Statistical Entropy $SE_{max,j}$, Normalized Entropic Substance Dilution Numbers (SDN) (or Relative Statistical Entropies RSE)

Possible or hypothetical item j-based maximum statistical entropies $SE_{max,j}$, according to Eq. (23) for the process inlet or outlet turn up if item j is incidentally (or imaginarily assumed) equally distributed among all relevant process inlets k_in respectively all relevant outlet streams k_out, with an average concentration according to Eq. (22). The streams $\dot{m}_{k\_in}$ and $\dot{m}_{k\_out}$ in Eq. (22) and the item j-related stream frequencies $M_{k\_in,j}$ and $M_{k\_out,j}$ of Eqs. (8) and (9) must now—as a supplementary constraint—include all relevant process inlets and outlets $\dot{m}_k$, which are connected to and participate in the interior mass turnover within the process limits in which the redistribution of item j occur, not only inlets and outlets with $c_{k,j} \neq 0$ kg/kg, in order to calculate a correct average concentration via Eq. (22) and correct $SE_{max,j}$ via Eq. (23). Therefore, the usage of the set of Eq. (21)–(23) requires supplementary mathematical effort for an overall mass flow analysis, Eq. (21), beside the still necessary preservation of the target item j according to Eq. (7). Relevant inlet mass streams $\dot{m}_{k\_in}$,

which materially participate in the redistribution of item j in the process (but not necessarily contain this item j at the beginning, i.e., possibly $c_{k\_in,j} = 0$), are balanced with all relevant outlet mass streams $\dot{m}_{k\_out}$. For j = C (carbon), these inlets $\dot{m}_{k\_in}$ could include carbon-free inlet streams involved in mixing or chemical reactions with the relevant carbon-containing substances, e.g., pure O_2 inlet flows or other O_2-containing agents for combustion or gasification. The numbers "f_in" and "f_out" in Eqs. (21)–(23) now represent with this constraint the necessary extended number of relevant process inlets respectively outlet streams, including relevant streams k with $c_{k\_in,j} = 0\,kg/kg$. whereas the numbers "f_in" and "f_out" in Eqs. (7)–(17) as well as (24) and the following equations only need—as necessary minimum requirement and as justified by Eqs. (19) and (20)—to include the relevant inlets k and outlets k with $c_{k,j} \neq 0\,kg/kg$ to satisfy the conservation Eq. (7).

Overall mass balance (inlets and outlet mass streams which materially participate in the redistribution of item j):

$$\sum\nolimits_{k\_in=1}^{f\_in} \dot{m}_{k\_in} = \sum\nolimits_{k\_out=1}^{f\_out} \dot{m}_{k\_out} \tag{21}$$

Target item j-related average concentration $c_{average,j}$ and item j-based maximum statistical entropy $SE_{max,j}$:

$$c_{average,j} = \dot{m}_j / \sum\nolimits_{k\_in=1}^{f\_in} \dot{m}_{k\_in} = 1 / \sum\nolimits_{k\_in=1}^{f\_in} M_{k\_in,j} = \dot{m}_j / \sum\nolimits_{k\_out=1}^{f\_out} \dot{m}_{k\_out} = 1 / \sum\nolimits_{k\_out=1}^{f\_out} M_{k\_out,j} \tag{22}$$

$$SE_{max,j} = -\log_2(c_{average,j}) = \log_2\left(\sum\nolimits_{k\_in=1}^{f\_in} M_{k\_in,j}\right) = \log_2\left(\sum\nolimits_{k\_out=1}^{f\_out} M_{k\_out,j}\right) \tag{23}$$

Target item j-based RSE, (SDN), substance concentration (collection) efficiency (SCE), recycling effectiveness (RE):

$$RSE_{in,j}(= SDN_{in,j}) = SE_{in,j}/SE_{max,j} = \sum\nolimits_{k\_in=1}^{f\_in} X_{k\_in,j} \cdot \log_2(c_{k\_in,j})/\log_2(c_{average,j}) \tag{24}$$

$$RSE_{out,j}(= SDN_{out,j}) = SE_{out,j}/SE_{max,j} = \sum\nolimits_{k\_out=1}^{f\_out} X_{k\_out,j} \cdot \log_2(c_{k\_out,j})/\log_2(c_{average,j}) \tag{25}$$

$$SCE_j = (RSE_{in,j} - RSE_{out,j})/RSE_{in,j} = 1 - RSE_{out,j}/RSE_{in,j} \tag{26}$$

$$RE_j = (SE_{max,j} - SE_{out,j})/SE_{max,j} = 1 - RSE_{out,j} \tag{27}$$

Equation (23) as defined entropic maximum dilution of j is used to normalize Eqs. (15) and (17) resulting in scaled Eqs. (24) and (25) for RSE (SDN) [1] of item j, again separately for the process inlets and outlets. RSE's of Eqs. (24) and (25) range from 1 Bit/Bit (item j equipartitioned in the relevant inlets or outlets and thereby entropically most diluted, highest degree of homogeneity and mixing) to possibly 0 Bit/Bit (highest entropic degree of item j separation and concentration). In cases of $RSE_{in,j} = 0\,Bit/Bit$ or $RSE_{out,j} = 0\,Bit/Bit$, item j only appears to 100 wt.% enriched $(c_{k,j} = 1\,kg/kg)$ in one or some of the process inlets respectively outlets and is absent in the other relevant inlets or outlets. For carbon as considered item (j = C), $SE_{out,C} = 0\,Bit$ and $RSE_{out,C} = 0\,Bit/Bit$ are hypothetically only possible in cases of recovery streams of pure carbon and absence of carbon or carbon-containing substances in other side outlet streams. For carbon as part of an target item i, $RSE_{out,C}$ is > 0 Bit/Bit even for cases of item i-based $RSE_{out,i} = 0\,Bit/Bit$,

Table 1 Substance i specific carbon parts $c_{C,i}$ and minimum substance i related carbon-based process outlet entropies $SE_{min,C,i}$

Substance i	Formulae	$x_{C,i}$ / n	MW_i / n [g/mol]	$c_{C,i}$ [g C / g i]	$SE_{min,C,i}$ [Bit]
Carbon	C	1	12.01	1	0
Acetylene	C_2H_2	2	26.04	0.9226	0.1163
Polypropylene (PP)	$(C_3H_6)_n$	3	42.08	0.8563	0.2238
Polyethylene (PE)	$(C_2H_4)_n$	2	28.05	0.8563	0.2238
Butane	C_4H_{10}	4	58.12	0.8266	0.2748
Propane	C_3H_8	3	44.10	0.8171	0.2913
Ethane	C_2H_6	3	30.07	0.7989	0.3240
Methane	CH_4	1	16.04	0.7487	0.4176
Polyethylene tereph-thalate (PET)	$(C_{10}H_8O_4)_n$	10	192.17	0.6250	0.6780
Carbon monoxide	CO	1	28.01	0.4288	1.2216
Methanol	CH_3OH	1	32.04	0.3748	1.4156
Carbon dioxide	CO_2	1	44.01	0.2729	1.8735

Remark: Acetylene and lower alkanes are byproducts during the simulation of methanol to olefins synthesis (MTO) in the project

$SE_{min,C,i}$ according to Eqs. (32) and (33) belongs to pure substance i outlets, $c_{k\_recov,i} = 1$ kg i/kg allotted recovery stream, and to cases, in which this substance i is the only C sink of the process, hence belonging to cases of minimum substance i based outlet entropies $SE_{out,i} = SE_{min,i} = 0$ Bit and therefore $RSE_{out,i} = 0$ Bit/Bit. Carbon-containing substances i and carbon are thereby absent $c_{k\_out,i} = 0$ kg i/kg and $c_{k\_out,C} = 0$ kgC/kg in other outlet (side) streams

i.e., even for cases, in which the carbon-containing item i is the only process carbon sink, occur alone in recovery streams and is absent in other side outlet streams, as shown, e.g., in Table 1.

The scaled RSE's (SDN's) of Eqs. (24) and (25) are used for entropic characteristics for the item j redistribution in the process, resulting in Eqs. (26) and (27). Respectively, the unscaled SE are directly used as in Eq. (18), which the same result as Eq. (26). The substance concentration efficiency SCE [1, 3, 4] of Eqs. (18) and (26), also termed substance collection efficiency SCE [3, 4], is an entropic characteristic or "entropic process concentration power," which ranges between $SCE_j > 0$ (entropic concentration); $SCE_j = 0$ (target item j entropically unchanged by the process) and $SCE_j < 0$ (entropic dilution of item j via the process). $SCE_j = 1 \left(RSE_{out,j} = 0 \, \text{Bit/Bit} \right)$ indicates the highest entropic concentration or collection of item j in a process in which the considered item j occurs alone $\left(c_{k\_out,j} = 1 \text{kg/kg} \right)$ in one or more recovery outlet streams $\dot{m}_{k\_out}$ (aspired recovery streams $\dot{m}_{k\_recov}$) and is absent $\left(c_{k\_out,j} = 0 \text{kg/kg} \right)$ in other process relevant outlets $\dot{m}_{k\_out}$.

The RE of Eq. (27) is another entropic characteristic and was applied, e.g., for PET recycling [32, 33]. Complementarily to $RSE_{out,j} \left(= SDN_{out,j} \right)$ of Eq. (25), RE ranges from 0% (entropically most diluted outlet of item j, i.e., item j equipartitioned between all relevant outlets) to possibly 100% (most concentrated process outlet of item j, i.e., item only occurs enriched with 100 wt.% concentration in one or more recovery outlet streams). Contrary to SCE, RE does not consider the real process inlet state of the item j to recover, but references as worst case a maximum inlet entropy by equipartitioning target item j $\left(c_{average,j} \right)$ among all relevant process inlets according to Eqs. (22) and (23) and thereby require the mathematical effort for an overall mass flow analysis, Eq. (21). Conventional yield characteristics like the RR, Carbon Recycling Rate or Carbon Recovery Rates (CRR) as shown in Eqs. (1)−(3) do

not consider the inlet distribution state of the target item j (e.g., PET, PP, carbon); however, they refer to the process inlet $\dot{m}_{in,j}$ of target item j in the denominator and the conservation Eq. (7) of the target item throughput must be met. RE like SCE, and unlike conventional yields, take into consideration the yield and concentrations $c_{k\_recov,j}$ of item j in desired outlet recover streams $\dot{m}_{k\_recov}$, the item losses via other outlet side streams $\dot{m}_{k\_out}$ inclusively the item concentration $c_{k\_out,j}$ (dilution) in those side streams.

Restrictions of the Adapted Set of Equations

The focus of the Eqs. (5)–(27) is directed to have indices for the entropic concentration power of the considered process itself, respectively for its undesired entropic dilution. The equations only consider the distribution of target item j in the streams directly passing over the process battery limits (items j = PP, PET, carbon in the project), to gain entropic characteristics which exclusively judge entropic changes within the restricted limits of the considered process. In cases where j = C, carbon is aspired be recovered via valuable products and valuable byproducts with high C-content, or otherwise favorably also in high concentrated CO_2 streams for potentially further processing. Exhaust and waste streams, with their C dilution state as it is during passage over the process battery limits, participate in the entropic calculations. In this sense, the Eqs. (5)–(27) fit the entropic model of solid residuals presented in [1], or the methodology presented in [32, 33]. Other entropic models, such as the overall model [1], consider the further ongoing dilution of emitted species after passage over the process battery limits into the environments, to or up to near the geogenic background concentration of these species in the environment compartments [1, 2, 13, 14, 16–22, 26], resulting in outlet entropies which incorporate further entropic changes by these effects. In Eqs. (5)–(27), there is no consideration of the geogenic background concentrations of components in the system environment compartments in which gaseous, liquid, or solid process waste streams such as exhaust gases, waste water streams, with their carbon-containing emitted components such as CO_2, organics, are possibly emitted. The impact of undesired emission into the affected environment is considered otherwise in the overall process evaluation procedure of the Fraunhofer Waste4Future project [39].

SEA application to carbon is considered by Kaufman et al. [21, 22] in the context of municipal solid waste (MSW) management (landfills) and waste-to-energy (WTE) facilities. To intensify environmental impacts (beside considering above mentioned geogenic background concentrations), supplementary forcing factors for methane emission were introduced to further increase the entropic effect of greenhouse gas emissions as a penalty. Supplementary forcing factors are not incorporated in the set of Eqs. (5)–(27). The effect of emitted greenhouse gases (CO_2 footprints), as well as CO_2 equivalents and CO_2 savings of a process in comparison with other processes, are considered otherwise in the overall process evaluation procedure developed in the Fraunhofer Waste4Future project [39].

The chemical bounding of carbon, i.e., the carbon speciation into different chemical components, is not considered in Eqs. (5)–(27). This speciation is topic of an SEA expansion to extended statistical entropy analysis (eSEA), as applied by Sobańtka et al. for the distribution path of nitrogen via different nitrogen compounds, during drop farming, in catchments and wastewater treatment plants, cf., e.g., [13, 14, 16–18].

The set of Eqs. (5)–(27) bases upon mass streams k_in and k_out and target item mass concentrations $c_{k,j}$ in kg/kg. Different densities and aggregate states of the streams are not considered. A liquid waste outlet stream with a certain carbon content (per

mass) would have the same carbon-based entropic impact as a gaseous outlet stream with the same mass and the same C mass content (the above mentioned further dilutions in the respective environment compartments not considered). However, a gaseous outlet stream with the same mass would have a higher volume and consequently a lower "volumetric" C concentration, i.e., this stream would volumetrically be more "diluted" with respect to carbon. Volumetric effects are not respected in Eqs. (5)–(27) because of the derivation of these equations by means of mass streams und mass concentrations.

Remarks regarding relevant and irrelevant process inlet and outlet streams which contain the considered item j, and—for the Eqs. (21)–(23) and resulting $SE_{max,j}$ in Eqs. (24)–(27)—regarding relevant and irrelevant process inlet and outlet streams which do not contain the considered item j:

Only process inlet and outlet streams with item (substance) j content, which materially participate in the turnover and redistributions of the examined substance j amount (e.g., j = carbon) within the process, are to include in Eqs. (5)–(27). Cooling or heating or other auxiliary media with item j (carbon) content which, e.g., only take part in indirect heat exchanges in process units (heat exchanger with separated walls) and thereby materially bypass the actual distribution procedures of the considered substance j (carbon) without direct material contact, are irrelevant for the entropic examination of the considered substance redistribution and are to omit. Streams to be omitted would be cooling air stream inlets (with their 400 ppmv CO_2) which solely act in heat exchangers in the process. These air streams materially bypass the mass reservoirs within the process in which the redistribution and reaction processes of the really examined carbon amount take place. If such streams are erroneously included in equation set (5)–(27), then the calculated entropic characteristics of the concentration/dilution process (of the actually examined carbon amount) would be distorted to a small or sizeable degree. The degree of this distortion would depend on the amount CO_2 in these erroneously incorporated cooling airs in comparison to the processed C amount in question which is really subjected to the C redistribution process. Contrariwise, if air inlets directly contact and thereby materially participate in the redistribution of the considered carbon C, e.g., take part in mixing, combustion or gasification reactions with the considered carbon-containing substances, then these inlet streams $\dot{m}_{k\_in}$ must be included in Eqs. (5)–(27).

Streams without carbon content which pass through the process as auxiliary media and materially bypass the examined carbon redistribution processes are not to include in Eqs. (5)–(27). Examples are cooling water inlet and outlets, reboiler water supply inlets, corresponding reboiler bleed stream and steam outlets, etc., for indirect heat exchange, which materially do not contact and do not take part in the carbon redistribution process. The inclusion of such carbon-free stream passages would erroneously alter the overall mass throughput, subsequently erroneously alter the average concentration, $SE_{max,j}$, the relative statistical entropies RSE in Eqs. (21)–(25) and the entropic recycling effectiveness RE in Eq. (27), even if these effects would cancel each other out in Eq. (26).

Contrariwise, and relevant in case of usage of the set of Eqs. (21)–(23), if carbon-free stream process inlets take part in the carbon redistribution process, e.g., if O_2 inlets materially participate in the redistribution of the considered carbon by combustion or gasification reactions with these carbon-containing substances, then such carbon-free streams must be incorporated at least into Eqs. (21)–(23), and for this 3 equations considered in Eqs. (8) and (9), in order to fulfill the constraint of the overall mass conservation Eq. (21) and to calculate correct average concentrations and maximum entropies via Eqs. (22) and (23), if the following usage of Eqs. (24)–(27) is intended. For relevant process, battery limits streams without content of the considered substance j ($c_{k,j} = 0\,kg/kg$, here j = C), the limits

according to Eqs. (19) and (20) for $c_{k,j} \cdot \log_2(c_{k,j})$ then applies in Eqs. (14)–(17), (24), and (25).

Carbon-Based Minimum Entropies for Aspired Process Goals and Possible Process Optimization Strategies

Equations (28)–(30) express a priori and possibly idealized process aspiration in the sense that different allotted recovery streams $\dot{m}_{k\_recov}$, which are selected out of the available process outlet streams $\dot{m}_{k\_out}$, are idealized the only process sinks for carbon, each of these $\dot{m}_{k\_recov}$ with a dedicated carbon-containing target item i (substances or defined mixtures of carbon containing substances). The streams $\dot{m}_{k\_recov}$ therewith imply carbon streams $\dot{m}_{k\_recov,C,i}$, connected with these carbon-containing items i. Predefined aspired target item i mass parts $c_{k\_recov,i} = \dot{m}_{k\_recov,i} / \dot{m}_{k\_recov}$ and concomitantly aspired item i based carbon mass part $c_{k\_recov,C,i} = \dot{m}_{k\_recov,C,i} / \dot{m}_{k\_recov}$ be allotted to these recovery streams $\dot{m}_{k\_recov}$, furthermore predefined intended outlet carbon loads $X_{k\_recov,C,i} = \dot{m}_{k\_recov,C,i} / \dot{m}_C$ of the overall process carbon inlet $\dot{m}_{in,C} = \dot{m}_{out,C} = \dot{m}_C$, with these $X_{k\_recov,C,i} =$ aspired ideal carbon recovery rates $RR_{k\_recov,C,i}$ based on Eq. (3). Carbon-containing items and thereby carbon itself are ideally aspired to be absent ($c_{k\_out,i} = 0 kg/kg$, $c_{k\_out,C} = 0 kg/kg$) in other process side or waste outlets $\dot{m}_{k\_out}$. The supplementarily attached substance index "i" in Eqs. (28) – (33) and in Table 1 is provided to avoid confusion with the former substance index j in Eqs. (5)–(27), which is now reserved for $j = C$ (carbon), and to imply, that, in the respective variables, carbon is part of and specifically related to a specified item i which in turn is intended to be the only carbon-containing process outlet item in the specified allotted recovery stream $\dot{m}_{k\_recov}$. The carbon-containing item i mass parts $c_{k\_recov,i}$ could be < 1 kg item i / kg recovery stream if other carbon-free admixture or diluting substances are intentionally parts in these streams $\dot{m}_{k\_recov}$. The resulting aspired carbon-based recovery stream frequencies $M_{k\_recov,C}$ and item i-based carbon loads $X_{k\_recov,C,i}$ are calculated via Eqs. (7), (9), and (11) for $\dot{m}_{k\_out} = \dot{m}_{k\_recov}$ and $j = C$. Equation (30) is derived from Eq. (17) and expresses the a priori defined aspired statistical carbon outlet entropy for this idealized aspired carbon redistribution process. The aspired carbon concentrations in Eqs. (28) and (32) are related to the intended concentrations $c_{k\_recov,i}$ of the carbon-containing items i in these recovery streams $\dot{m}_{k\_recov}$ itself by carbon / aspired item (or defined substance mixture) molar weight relations adjusted by the number of carbon atoms $x_{C,i}$ in the substance (or defined substance mixture) formulae. The indices "k_recov" in Eqs. (29) and (30) for the allotted recovery streams of this process aspiration present a subset of the indices "k_out" of the outlet streams of the considered real process, which is represented by Eqs. (13) and (17). Equations (29) and (30) are valid if the other outlet streams $k\_out \neq k\_recov$ are carbon-free according to the idealized process aspiration and thus do not contribute to Eqs. (29) and (30), confer also Eq. (20). For example, in Figs. 1, 2, 3, and 4, the stream $k\_out = 1$ is the intended recovery stream $\dot{m}_{k\_recov}$ in which the complete carbon (in this process only presented in substance $i = CO_2$) is ideally aspired to exit the separation process, whereupon stream $k\_out = 2$ is ideally aspired to be carbon-free.

Idealized aspired carbon recovery rates (RR) $RR_{k\_recov,C,i}$ and carbon-based statistical entropy $SE_{goal,C}$ of an idealized comparison process: recovery of carbon-containing items i with concentrations $c_{k\_recov,i}$ only via allocated recovery streams $\dot{m}_{k\_recov}$, the remaining outlet streams $\dot{m}_{k\_out}$ are assumed to be carbon-free. Each recovery streams $\dot{m}_{k\_recov}$ only contains on dedicated carbon-containing items i (substances or defined mixtures of carbon-containing substances). The streams $\dot{m}_{k\_recov}$ can possibly contain carbon-free admixtures.

$$c_{k\_recov,C,i} = x_{C,i} \bullet (MW_C/MW_i) \bullet c_{k\_recov,i} \tag{28}$$

$$\sum\nolimits_{k\_recov}^{f\_recov} RR_{k\_recov,C,i} = \sum\nolimits_{k\_recov}^{f\_recov} X_{k\_recov,C,i} = \sum\nolimits_{k\_recov}^{f\_recov} \dot{m}_{k\_recov} \bullet c_{k\_recov,C,i}/\dot{m}_C =$$
$$= \sum\nolimits_{k\_recov}^{f\_recov} \dot{m}_{k\_recov,C,i}/\dot{m}_C = \sum\nolimits_{k\_recov}^{f\_recov} M_{k\_recov,C} \bullet c_{k\_recov,C,i} = 1 \tag{29}$$

$$SE_{goal,C} = -\sum\nolimits_{k\_recov}^{f\_recov} RR_{k\_recov,C,i} \bullet \log_2(c_{k\_recov,C,i}) \tag{30}$$

Equations (28) and (30) lead to Eq. (31) if only one carbon-containing substance i in one allotted recovery stream, or possibly more allotted recovery outlets with the same carbon content $c_{k\_recov,C,i}$, is the process goal.

$$SE_{goal,C,i} = -\log_2(c_{k\_recov,C,i}) = -\log_2\left(x_{C,i} \bullet \left(MW_C/MW_i\right) \bullet c_{k\_recov,i}\right) \tag{31}$$

Equations (28) and (31) further simplify to Eqs. (32) and (33) if this intended one carbon-containing outlet substance i (or defined substance mixture i) is aspired in pure form, $c_{k\_recov,i} = 1$ kg/kg, without associated carbon-free diluting constituents in the allotted recovery stream(s). The indices "k_recov" for the allotted recovery streams is subsequently omitted. Table 1 shows examples for substance i specified carbon parts $c_{C,i}$ and related process minimum entropies $SE_{min,C,i}$ according to Eq. (33), if this item i is aspired to be the only carbon-containing outlet and recovered in pure form.

$$c_{C,i} = x_{C,i} \bullet MW_C/MW_i \tag{32}$$

$$SE_{goal,C,i} = SE_{min,C,i} = -\log_2\left(c_{C,i}\right) = -\log_2\left(x_{C,i} \bullet MW_C/MW_i\right) \tag{33}$$

The intended outlet entropies $SE_{goal,C}$ of the idealized process aspiration according to Eqs. (30) or (31) or (33) are in turn inputs for aspired entropic characteristics $RSE_{goal,C}$ ($SDN_{goal,C}$), $SCE_{goal,C}$ and $RE_{goal,C}$ on the basis of Eqs. (25), (18) or (26), and (27), where index j is replaced by index C (carbon) and $SE_{out,C}$ is replaced by $SE_{goal,C}$, whereupon the carbon-based input and average entropies, Eqs. (15), (23), and (24), of the real process are used for these entropic outlet characteristics of the process aspiration. The aspired outlet entropy $SE_{goal,C}$ and aspired outlet entropic characteristics could be compared with the really achieved carbon-based $SE_{out,C}$ and real entropic characteristics of the considered real process with the same carbon and overall mass inlet, which is characterized by Eqs. (15), (17), (23), (24), (25), (18) or (26), and (27). The carbon throughput, Eq. (7), j = C, and the overall mass balance, Eq. (21) should —because of its influence on $c_{average,C}$, $SE_{max,C}$, RSE and RE—be the same or comparable between the idealized process aspiration and the real or simulated process under review.

A process optimization strategy could be to find configurations of interconnected units and operations within the process for which the carbon outlet entropy $SE_{out,C}$ of Eq. (17) (j = C) contracts to the desired low $SE_{goal,C}$ of Eqs. (30) or (31) or (33), and concomitantly the entropic characteristics $RSE_{out,C}$ ($SDN_{out,C}$), SCE_C and RE_C contracts to $RSE_{goal,C}$ ($SDN_{goal,C}$), $SCE_{goal,C}$, and $RE_{goal,C}$. However, undesired substances with high carbon content, possibly appearing highly concentrated and in great quantities in process outlets, could also influence (decrease) $SE_{out,C}$. Carbon-based entropic characteristics are complex and conglomerated parameters, which consider the overall fate of carbon within the carbon conversion and distribution process, i.e., which include interconnected entropic

effects via the amount and concentration of carbon-containing target items i in intended recovery streams, via valuable carbon-containing byproduct streams, and the amount and the concentration or dilution of carbon in process side streams. Consequently, an optimization via carbon-based entropies and resulting entropic characteristics should preferably be done in conjunction with conventional item i-related carbon yield characteristics, i.e., with aspired high item i-based carbon recycling or recovery rates according to Eqs. (2) or (3) in a priori allotted recovery streams k_recov, possibly aspiring a carbon yield via item i of $RR_{k\_recov,C,i} = 1$, in order to force such an optimization into the desired direction. Carbon-based entropic and conventional yield characteristics could be judged together for CR processes, as exemplified by Fig. 4 for the stylized direct air capture (DAC) process. Conventional carbon yield characteristics as, e.g., the carbon recycling rate (CRR)—as a measure for the carbon loop closure in the CCE context [42]—could be complementarily evaluated with carbon-based entropic characteristics which enable to locate and quantify the amount of the carbon-based entropic deteriorations (entropic dilutions) as, e.g., the entropic contributions of exhaust streams containing diluted CO_2 as losses, which are detrimental for the carbon recovery process via carbon-containing target substances and should be abated. In case of chemical recycling, this could be applied, e.g., for process internal optimization of CO_2 capture and reutilization strategies, or investigation of the effect of increased usage of electrical energy input for endothermic chemical conversion processes instead of partial combustion of the feedstock for endothermic reactions, e.g., gasification enhanced by plasma integration.

Analogous conjunctions between conventional yields (recovery rates) und entropic characteristics based on conserved items *j* in physical separation processes (instead of carbon) are exemplified in Figs. 2, 3, 6, and 8 for the stylized physical separation processes. Substance (item)-based entropic characteristics for physical separations are conglomerated penalty parameter as well. Roithner [32, 33] emphasized the integrated quantitative and qualitative information of the entropic recycling effectiveness RE as a recycling indicator and metric parameter in the context of CE for the assessment of the success of physical separation processes of, e.g., PET from plastic feedstock, beside a complementary usage of conventional recycling rates (RR) defined via complete recovery mass streams $\dot{m}_{k\_recov}$ which includes contaminations in $\dot{m}_{k\_recov}$, contrary to the recovery rate of Eq. (1). In the Wast4Future project, conventional target substance j yields, i.e., RR according to Eq. (1), and substance-based entropic characteristics are judged together and in conjunction for physical separations, analogous to the joint usage of carbon-based yields and entropic characteristics in cases of chemical recycling.

Stylized Process Examples

Stylized DAC Scheme, High Substance, and Carbon-Based Inlet Entropies (CO_2 Diluted Inlet)

Figures 2, 3, and 4 depict the results of a first test of the set of Eqs. (5)–(33) on a separation process regarding an inlet with very diluted target substance content, i.e., the attained CO_2-based entropic characteristics and concomitantly the entropic characteristics for carbon as part of CO_2 for the stylized DAC scheme of Fig. 1, with all auxiliary material streams neglected. The target substance CO_2 is preserved in this process, so that a resort to carbon-based characteristics is not required, but
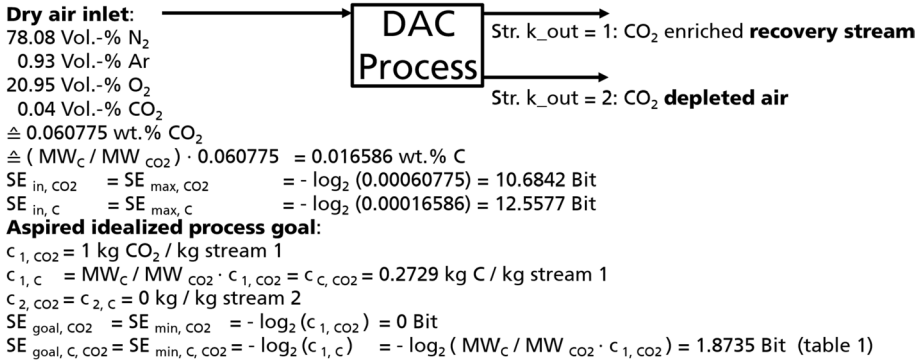
Dry air inlet:
78.08 Vol.-% N_2
  0.93 Vol.-% Ar
20.95 Vol.-% O_2
  0.04 Vol.-% CO_2
$\triangleq 0.060775$ wt.% CO_2
$\triangleq (MW_C / MW_{CO2}) \cdot 0.060775 = 0.016586$ wt.% C
$SE_{in, CO2} = SE_{max, CO2} = - \log_2 (0.00060775) = 10.6842$ Bit
$SE_{in, C} = SE_{max, C} = - \log_2 (0.00016586) = 12.5577$ Bit
Aspired idealized process goal:
$c_{1, CO2} = 1$ kg CO_2 / kg stream 1
$c_{1, C} = MW_C / MW_{CO2} \cdot c_{1, CO2} = c_{C, CO2} = 0.2729$ kg C / kg stream 1
$c_{2, CO2} = c_{2, C} = 0$ kg / kg stream 2
$SE_{goal, CO2} = SE_{min, CO2} = - \log_2 (c_{1, CO2}) = 0$ Bit
$SE_{goal, C, CO2} = SE_{min, C, CO2} = - \log_2 (c_{1, C}) = - \log_2 (MW_C / MW_{CO2} \cdot c_{1, CO2}) = 1.8735$ Bit (table 1)

The DAC Process block: Str. k_out = 1: CO_2 enriched **recovery stream**; Str. k_out = 2: CO_2 **depleted air**

Fig. 1 Stylized direct air capture (DAC) process

nevertheless, they are also depicted for comparison. Dry air CO_2, with high carbon-based inlet entropy, is the only carbon process source and sink. The CO_2 content in recovery stream 1 is aspired to be $c_{1,CO2} = 1 kgCO_2/kg$. The outlet 2 is aspired to be CO_2 and thereby carbon free, $c_{2,CO2} = 0\,kg\,CO_2/kg$. The substance CO_2-based inlet entropy $SE_{in,CO2} = SE_{max,CO2}$ 10.684 Bit and the carbon-based inlet entropy $SE_{in,C} = SE_{max,C} = 12.558$ Bit according to Eq. (15) and Fig. 1 are high because of the diluted CO_2 inlet with $c_{in,CO2} = c_{average,CO2} = 0.06077\,wt.\%CO_2$ and the concomitant carbon concentration $c_{in,C} = c_{average,C} = 0.01659\,wt.\%C$, which is connected to $c_{in,CO2}$ in analogy to Eq. (28). The maximum process separation of CO_2
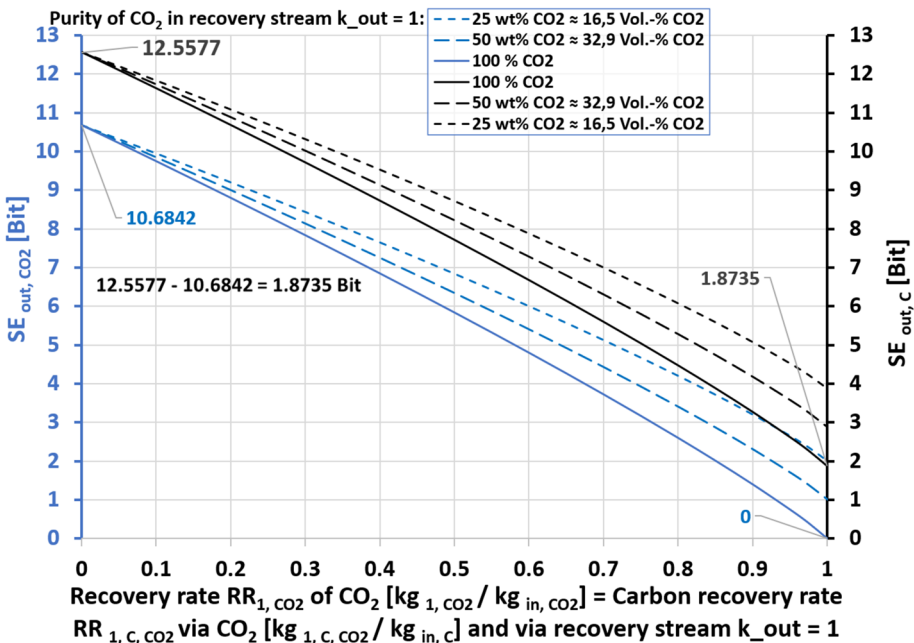
Fig. 2 Statistical entropies SE for CO_2 and C versus yield characteristics of the simple DAC process of Fig. 1
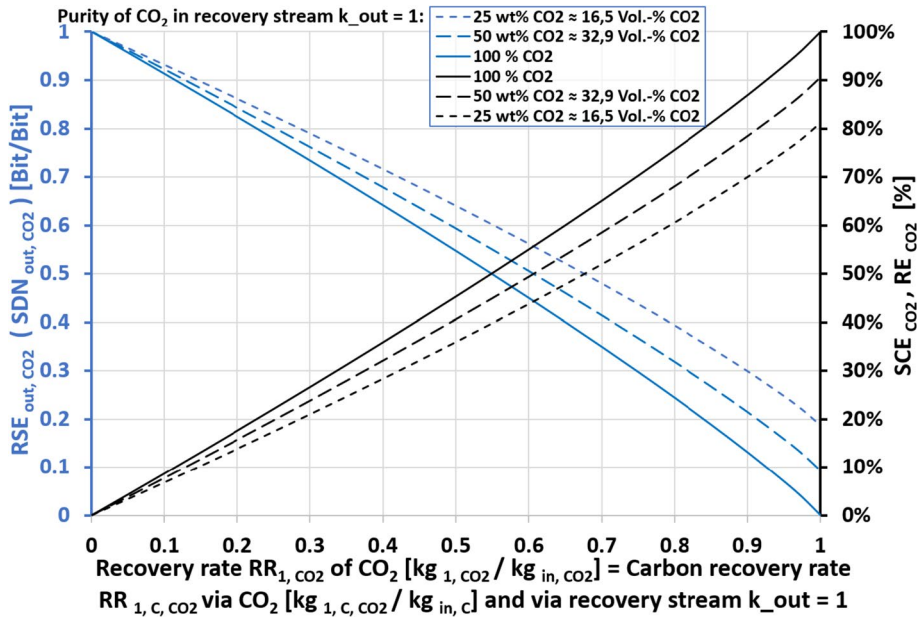
Fig. 3 Substance CO_2-based entropic characteristics versus yield characteristics for the stylized DAC process of Fig. 1
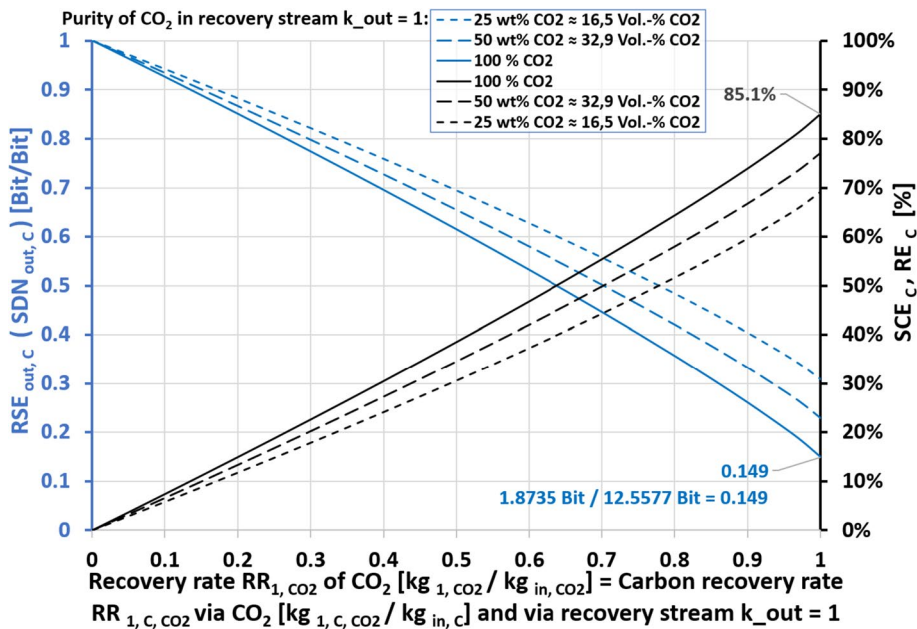

Fig. 4 Carbon-based entropic characteristics versus yield characteristics for the simplified DAC process of Fig. 1
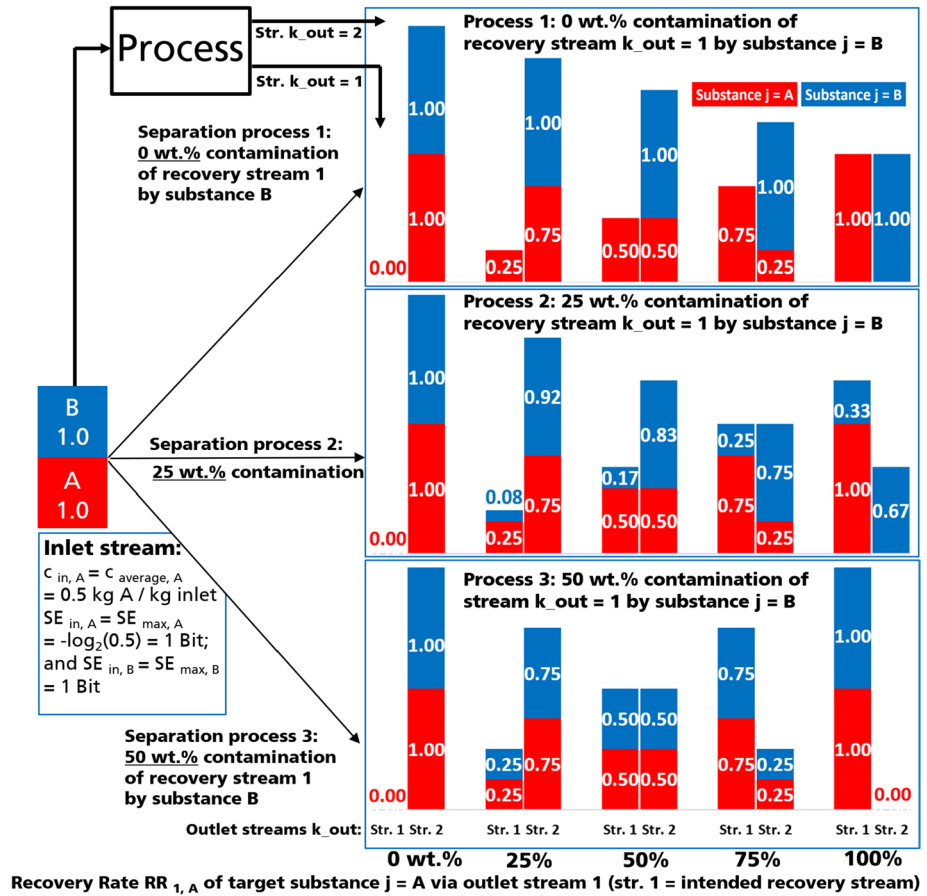
Fig. 5 Stylized separation processes 1, 2, and 3 for recycling target substance $j = A$

and aspired minimum CO_2 entropy is achieved if recovery stream k_out = 1 contains the complete CO_2 inlet in pure form, $c_{1,CO2} = 1\,kg\,CO_2/kg$ stream 1, resulting in $SE_{out,CO2} = SE_{min,CO2} = -\log_2(1) = 0\,Bit$, as well as $c_{1,C} = c_{C,CO2} = 0.2729\,kg\,C/kg$ stream 1 and $SE_{out,C} = SE_{min,C,CO2} = -\log_2(0.2729) = 1.8735\,Bit$ according to Eqs. (32), (17), and (33) and Table 1. Entropic penalties (higher SE, SDN, lower SCE and RE) with increasing contamination of the CO_2 recovery stream are depicted in Figs. 2, 3, and 4 via CO_2 purity curve parameters. The concave curvatures of the entropies SE and RSE (SDN) and the convex curvatures of the entropic characteristics SCE and RE illustrate supplementary entropic penalties of undesired CO_2 losses beyond the yield losses $1 - RR_{1,CO2}$ and $1 - RR_{1,C,CO2}$: Even for 100 wt.% CO_2 purity in recovery stream 1, the entropic CO_2 dilution numbers $SDN_{CO2}(RSE_{CO2})$ of Fig. 3 are above the diagonal which descend to the lower right corner of the diagram (excluding the curvature final points), whereas the entropic characteristics SCE_{CO2} and RE_{CO2} are $<$ than the conventional yield characteristics $RR_{1,CO2}$ and $RR_{1,C,CO2}$ (they are below the diagonal which arise from the diagram origin). These curvatures in Figs. 2 and 3 are not pronounced because of the high inlet entropy levels $SE_{in,CO2} = SE_{max,CO2} = 10.684\,Bit$ and
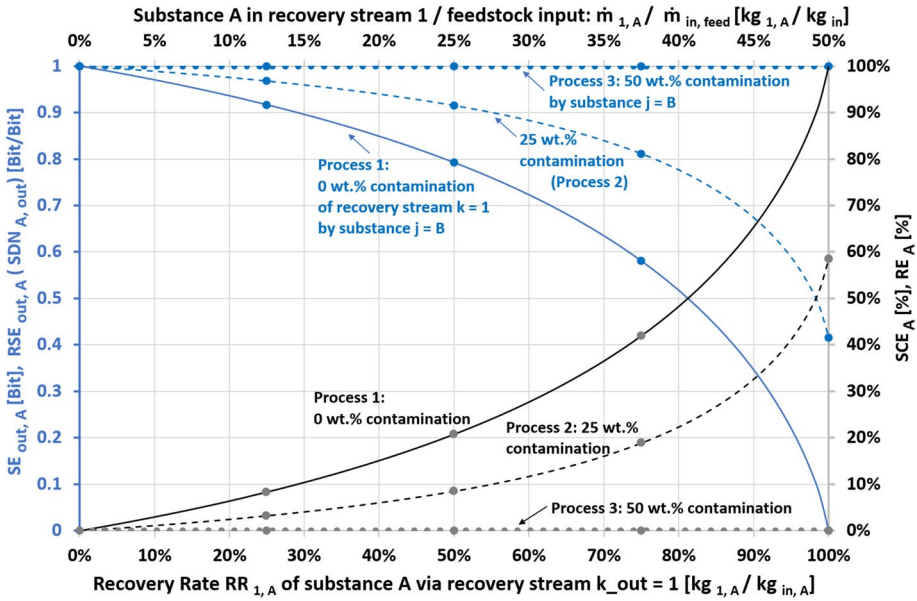
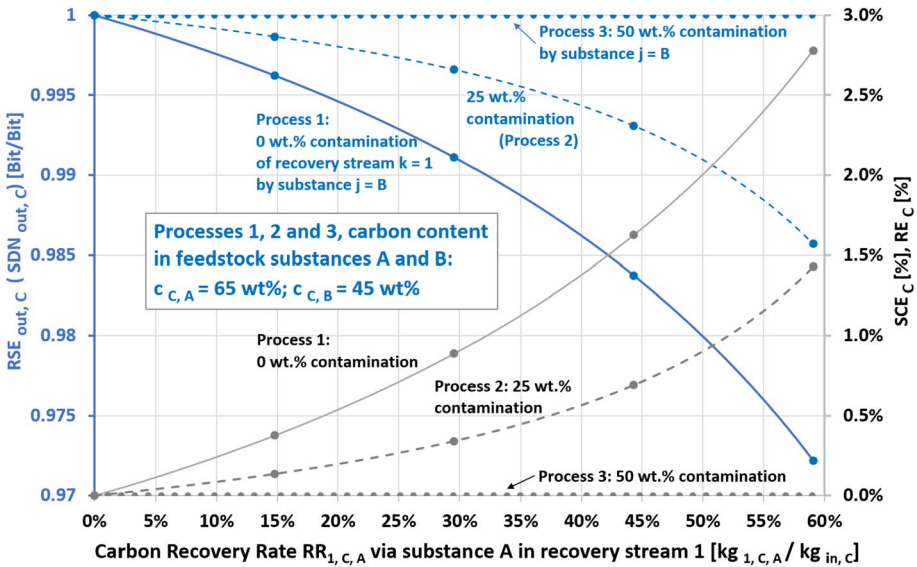**Fig. 6** Substance A-based entropic characteristics for separation processes 1, 2, and 3 in Fig. 5

Fig. 7 Carbon-based entropic characteristics for separation processes 1, 2, and 3 in Fig. 5

$SE_{in,C} = SE_{max,C} = 12.558$ Bit and the diluted CO_2 content in the depleted air stream (stream 2) in this example process. The concave and convex curvatures and thereby the entropic penalties for losses are more pronounced, if the substance content in the inlet stream is high and the substance inlet entropy would thereby be relatively low, e.g., for the 1 Bit recycling substance A inlet entropy $SE_{in,A}$ in Figs. 5 and 6.
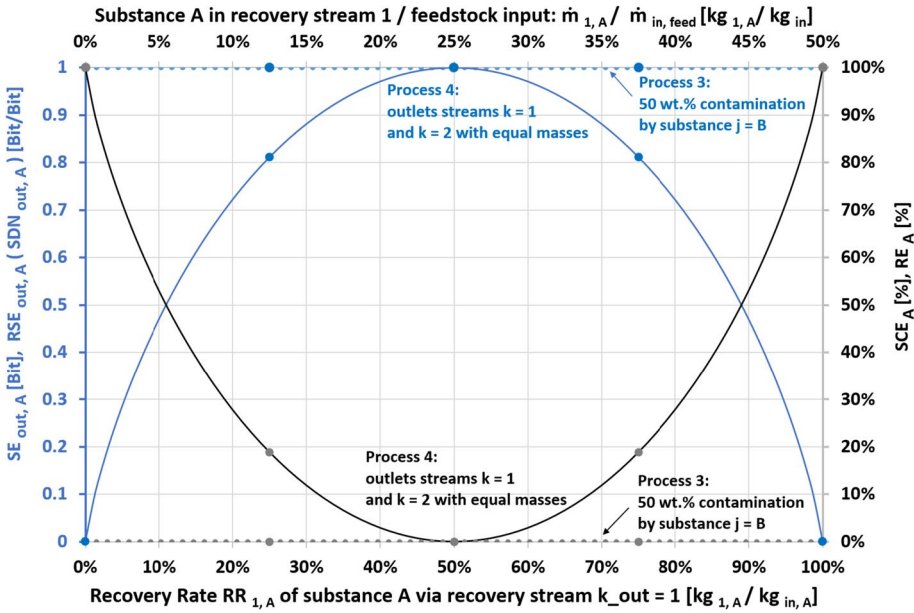
Fig. 8 Substance A-related entropic characteristics for separation processes 3 and 4 of Figs. 5 and 10

For the stylized separation scheme of Fig. 1 with the CO_2 as the only aspired carbon-containing target substance and the only aspired carbon sink, the carbon-based statistical entropies $SE_{in,C}$ of Eq. (15), $SE_{out,C}$ of Eq. (17) and in Fig. 2, and $SE_{max,C}$ of Eq. (23) could be downshifted by $SE_{min,C,CO2} = -\log_2(MW_c/MW_{CO2}) = 1.8735\,\text{Bit}$, derived via Eq. (33) and Table 1. The downshifted carbon outlet entropies $SE_{out,C,shift} = SE_{out,C} - SE_{min,C,CO2} = SE_{out,C} - 1.8735\,\text{Bit}$ would then coincide with the CO_2 outlet entropies $SE_{out,CO2}$ in Fig. 2. Subsequently, if these CO_2 specific $SE_{min,C,CO2}$
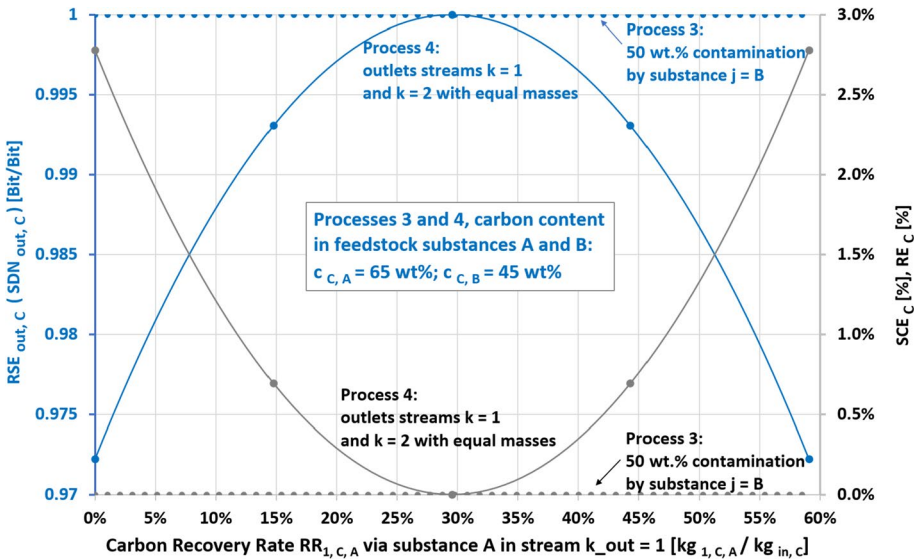
Fig. 9 Carbon-based entropic characteristics for separation processes 3 and 4 of Figs. 5 and 10

downshift of the carbon-based entropies in Eqs. (15), (17), and (23), with $j = C$, is applied in Eqs. (25), (18) or (26), and (27) for calculating shifted carbon-based entropic characteristics $RSE_{out,C,shift}$, $SCE_{C,shift}$ and $RE_{C,shift}$, then these shifted carbon-based entropic characteristics would coincide with the substance CO_2-based entropic characteristics in Fig. 3, i.e., the original carbon-based entropic characteristics of Fig. 4 would then change to modified (shifted) carbon-based entropic characteristics as in Fig. 3. For this stylized case, the DAC process could thus alternatively be evaluated via CO_2-based as well as via such shifted carbon-based entropic characteristics and the associated CO_2-based or carbon-based recovery rates.

Stylized Physical Separation Processes for Solid Feedstock

Contrary to the stylized DAC process in Figs. 1 and 3 with its high CO_2, respectively, carbon-based inlet entropies of 10.684 and 12.558 Bit, the solid feedstocks for recycling processes of the Waste4Future project exhibit a high carbon content and thereby low carbon-based inlet entropies $SE_{in,C}$ in low-single-digit range, as depicted in Table 2. Furthermore, during mechanical and other physical recycling processes, the carbon contents c_C and therewith $-\log_2(c_C)$ of the feedstock constituents often do not differ by several orders of magnitude, as exemplified by $-\log_2(c_{C,A})$ and $-\log_2(c_{C,B})$ in Table 2 for the recyclate part A and residual part B. Figures 5 and 6 illustrate a stylized mechanical recycling process with 2 outlets for a binary inlet mixture of equal mass parts of conserved substances $j = A$ and B, with target substance $j = A$ related outlet entropies $SE_{out,A}$ and $RSE_{out,A}$ in Fig. 6 according to Eqs. (17) and (25), as well as the entropic characteristics according to Eqs. (18) or (26), and (27). The horizontal axis presents the target substance A yields

Table 2 Used solid feedstock for mechanical / physical and chemical recycling (CR) processes in the Waste4Future project

Feedstock	Unit	RDF (DSD)	SLF
Feedstock = Σ (Recyclable A + Residual B)			
$^c c_{,Feedstock}$	[kg C / kg feedstock]	0.5623	0.5487
$SE_{in,C} = -\log_2(c_{C,Feedstock})$ *	[Bit]	0.831	0.866
Feedstock recyclable substance $j = A$ to recover by mechanical / physical recycling			
Recyclable $j = A$		PET	PP
Recyclable mass part, $c_{Recyclable}$	[kg recyclable $j = A$ / kg feedstock]	60.80%	22.78%
$SE_{in,Recyclable} = -\log_2(c_{Recyclable})$ **	[Bit]	0.718	2.134
Carbon part in recyclable, $c_{C,Recyclable}$	[kg carbon / kg recyclable $j = A$]	0.625	0.8563
$-\log_2(c_{C,Recyclable})$	[Bit]	0.678	0.224
Feedstock residual substance B (PE, PS, ABS, PC, PA, wood, metals, etc.)			
Residual mass part, $c_{Residual}$	[kg residual B / kg feedstock]	39.20%	77.22%
Carbon part in residual, $c_{C,Residual}$	[kg carbon / kg residual B]	0.4651	0.4579
$-\log_2(c_{C,Residual})$	[Bit]	1.104	1.127

RDF refuse derived fuel, SLF shredder light fraction; $c_{C,A}$ and $c_{C,B}$, carbon mass parts in substance A and B

*:= $SE_{in,C}$, if this feedstock is the only carbon-containing inlet of a separation process. $SE_{in,C}$ and carbon-based SEA methodology is considered for WTE and CR within the Waste4Future project, i.e., processes where the feedstock is destroyed

**: $SE_{in,j}$ ($j = A$ = recyclable PET or PP) and substance j-based SEA methodology is considered for mechanical / physical recycling within the project, i.e., separation processes where the substance j remains preserved
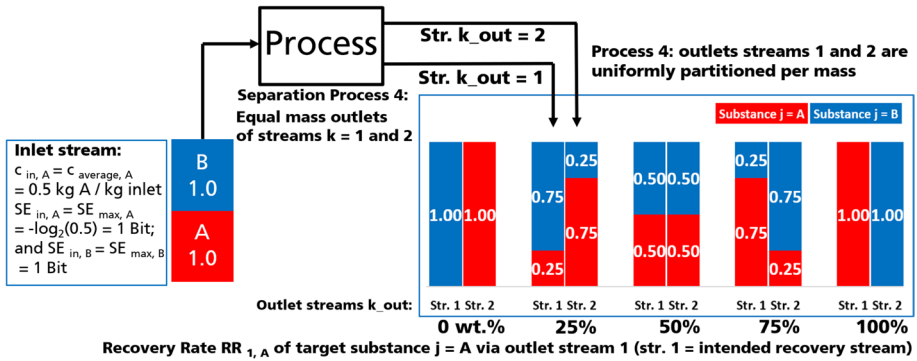
Fig. 10 Simplified separation process 4, mass symmetric case: outlets stream 1 and 2 with equal masses

(recovery rates) via allotted recovery stream k_out = 1 according to Eq. (1). As in Fig. 3, the entropic penalties—higher SE and SDN (RSE), lower SCE and RE—with increasing contamination of the recovery stream by substance B are visible. Different to Fig. 3, the entropic penalties of undesired substance A losses via residual stream k_out = 2 are more pronounced, i.e., more pronounced convex curvatures of SCE_A and RE_A in Fig. 6 because of the high substance A inlet concentration and low substance A inlet entropy level, here 1 Bit, and because of the possible high concentration of the lost recycling substance A in this residual stream 2. The target substance-based entropic characteristics for the recovery of the recyclates PET and PP of Table 2 were calculated for the complete recovery range as in Fig. 6 and show analogous qualitative behaviors as in Fig. 6. A complete separation of the target substances in pure form was practically achieved during the test plant experiments for physical PP recovery from SLF and PET from RDF in the Waste4Future project ($RR_{1,PP} = 1$ and $SCE_{PP} = 1$ as well as $RR_{1,PET} = 1$ and $SCE_{PET} = 1$). The steep gradients and sensitivities of the entropic characteristics in Fig. 6 in the range of the curvature final points near RR = 1 highlight, however, the demand of a high level of accuracy for the measurements of the achieved purity and recovery of the target recyclate in this RR range, and possibly of repeated separation experiments for statistical evaluations.

For purely mechanical recycling processes with preservation of the aspired substances, the yield and entropic analysis would be restricted to these target substance-based characteristics as in Fig. 6. The processes could be optimized in direction of high target substance A related entropic characteristics in conjunction with concomitant high substance A yields via the allotted recovery stream k_recov, e.g., in direction of upper right corner in Figs. 6 or 9. However, a further process objective could be to convert the residual carbon-containing substances B out of these mechanical or other physical recycling processes or alternatively the complete feedstock input itself via CR with its synthetic routes into (further) valuable substances j = A, thereby violating the substance conservation Eq. (7) for substance j = A. In these cases, the substance inherent carbon content is the selected subject of yield and entropic characteristics in the Waste4Future project. Exemplified, the substance j = A related characteristics in Fig. 6 of processes in Fig. 5 are complemented by j = carbon-based characteristics in Fig. 7. Analogous complements are repeated for Fig. 8 and shown in Fig. 9, concerning the processes 3 from Fig. 5 and processes 4 from Fig. 10. This complement by carbon-based entropic characteristics would not be necessary and meaningful for a mechanical separation process alone but could be done in preparation of possible

subsequent chemical conversion step of the residuals B into supplementary valuable substance A.

For the complemented carbon-based entropies in Figs. 7 and 9, a carbon content of $c_{C,A} = 65$ wt.% of the aspired substance A and $c_{C,B} = 45$ wt.% of residual substance B are exemplarily assumed, resulting in an overall C inlet content of 55 wt.% and carbon inlet entropy $SE_{in,C} = SE_{max,C} = -\log_2(0.55) = 0.8625$ Bit according to Eqs. (15) and (23). The maximum mechanical carbon recovery rate via substance A and via recovery stream k_out = 1 would be $65/(65 + 45) = 59\%$ for these examples, as observable from the curvature final points in Figs. 7 and 9. Figures 7 and 9 exemplarily illustrate that carbon-based entropic changes (SCE and RE improvements) are low or moderate during the stylized mechanical separation examples in Figs. 5 and 8 and for feedstocks with valuable low carbon-based inlet entropies (high carbon contents) as in Table 2, at least compared with processes as in Figs. 1 and 4. These low or moderate changes during mechanical separation depend on the relative carbon content differences between $c_{C,A}$ and $c_{C,B}$ of substances A and B, which are to separate via outlet streams k_out = 1 and 2. Carbon contents $c_{C,A}$ and $c_{C,B}$ (recyclables A and residuals B) of investigated mixtures in the project are depicted in Table 2. For $c_{C,A} = c_{C,B}$, there is even no carbon-based entropic changes for the stylized processes as illustrated in Fig. 11 in the addendum. The residual streams of physical separation processes preserve, if they have a high carbon content, low and valuable carbon-based entropies for possibly following processes (e.g., substances B in Table 2).

An aspired goal after the stylized separation process 1 of Figs. 5, 6, and 7, i.e., after complete substance A recovery without contamination by substance B in recovery stream k_out = 1 by physical separation, could be to convert the residual substance B with the remaining 41% carbon of residual stream k_out = 2 into new valuable substance A, now by subsequent CR procedures, like gasification with downstream cleaning and reactor units for CO shift and for synthesis of this aspired substance A out of this residual. The CR procedures of such entropically valuable residuals should be configured with the aspiration to minimize or ideally avoid carbon-based entropic dilutions. CR processes applied to such feedstock residuals after mechanical separation processes (or to a complete feedstock) are currently accompanied by undesired losses of significant carbon parts of these feedstock residuals in diluted form by CO_2-containing exhaust streams. This is reflected by unfavorably big carbon-based statistic outlet entropies $SE_{out,C}$ and negative entropic carbon collection efficiencies SCE_C during the CR processes. In Fig. 7, e.g., after complete physical recovery of substance A in pure form via recovery stream 1and therewith 59% of the inlet carbon which resides in this substance A, a chemical carbon conversion of the residuals of stream 2, i.e., of the residual 41% carbon inlet into supplementary synthesized valuable substance A, would currently be accompanied by a steep drop of the entropic carbon collection efficiencies SCE_C into negative ranges, caused by losses of an important carbon part of the processed residuals by exhaust gases with diluted CO_2 content. An aspiration in the CCE context could minimize such entropic losses and deteriorations, e.g., by minimizing in-process exhaust streams with diluted C contents or appropriating in-process CO_2 capture and utilization procedures, possibly carried out by conversion of this captured residual CO_2 with H_2 to additional synthesis gas in the process or, e.g., by electrification of CR (gasification enhanced by plasma integration for the necessary energy input) which reduces the in-process CO_2 release caused by partial feedstock combustion for the necessary energy for endothermic gasification reactions. The simulated CR processes up to now in the project of the complete feedstocks RDF and SLF of

Table 2 to produce polyolefins, i.e., without preceding physical separation of the target substance A (PET respectively PP), resulted in unfavorably big negative entropic carbon collection efficiencies $SCE_C < -200\%$ caused by process inherently occurring exhaust gases which contain > 50% of the feedstock carbon content as diluted CO_2 (overall CR process sequence: autothermic feedstock gasification via partial feedstock combustion, gas cleaning and CO shift units, methanol synthesis, MTO, polyolefin synthesis). These calculations up to now do not include an in-process capture of this diluted CO_2 and the reaction of imported or in-process produced H_2 with this captured CO_2 to produce additional synthesis gas. The biggest carbon-based entropic deterioration of $SCE_C < -500\%$, which served as the negative benchmark, occurred for the simulated WTE processes with complete combustion of the feedstocks of Table 2 and the release of the complete feedstock C content as diluted CO_2 via process exhaust gas.

Conclusion and Outlook

A joint usage of yield and entropy-based characteristics is applied as part of evaluating processes in the Waste4Future project and could be a useful instrument for CE and CCE.

SEA is applied as tool for CE principles [23–35, 37]. Substance-related entropic characteristics of SEA together with substance-based yields (RR) are applied in the project for physical recycling processes where the substance remains preserved (in the project: recyclables PP, PET) within the evaluated processes (e.g., in the project: test plant experiments for density separation and selective extraction), and where the feedstock is accompanied by favorably low recyclable-based input entropies [34] (the feedstock has high parts of the aspired recyclables).

Carbon-based entropic characteristics of SEA could be used as instrument in the CCE context in conjunction with carbon yield characteristics, e.g., CRR yield, Eq. (2), as proposed in [42], or carbon recovery rates, Eq. (3), to force an optimization strategy in the desired direction of the yield of the aspired carbon-containing substances and to localize and quantify carbon losses. A recourse to carbon-based entropic characteristics should preferably only take place for combustion or chemical conversion processes where substances are destroyed and possibly new target substances are yielded, and where the carbon-based entropic deterioration (entropic dilutions), e.g., exhaust stream containing diluted CO_2, is inherently significant and should be abated by amendments, optimization strategies, or new recycling process techniques (as, e.g., feedstock gasifiers enhanced by plasma integration). The simulation and following entropic evaluation of further combustion and CR processes as well as physical separations are parts of the project.

Item j (substance as well as carbon)-based SE and derived characteristics (RSE, SCE, RE) of the SEA are conglomerated penalty parameters respectively characteristics, which include interconnected entropic effects via the amount and concentration (purity) of the target item(s) in intended recovery streams, via the amount and the concentration (i.e., dilution) of the item(s) in process side streams (losses), and (for carbon) via valuable carbon-containing byproduct streams. For example, byproducts during methanol to olefins (MTO) synthesis are valuable natural gas ersatz C_1+C_2, LPG, C_{4+}, with their respective high carbon content. Entropic characteristics could prospectively and potentially be applied as engineering parameters (e.g., RSE [23]) which support decision-making in favor of CE [23] and CCE (design of new process principles and process chains). The conglomerated properties of entropic characteristics could necessitate the proposed conjunction of entropic characteristics with conventional yield characteristics (RR).

Entropic characteristics could combine quantitative and qualitative performance assessment indications for the success of recycling processes [32, 33]. SEA assessment methods could be coupled with other characteristics to overcome possible limitations of SEA. Within the Waste4Future project, the evaluation of the effectiveness of recycling processes by substance respectively carbon-based entropic characteristics according to the SEA methodology is coupled together with substance, respectively, carbon-based yields (RR) as well as other characteristics as CO_2 footprint, respectively, CO_2 equivalents, economic yields, also economic yields if electricity and heat production by WTE are considered. The couplings of characteristics is an ongoing investigation topic, e.g., [23, 33–35, 39]. To strengthen the case of resource conservation and recovery, characteristics of SEA could conceivably also play a role in assessment methods of product qualities and losses for such topics as the shift to recycling friendly and sustainable product designs, see, e.g., [34], closed-loop supply chain designs [48], and reverse supply chain practices [49].

Appendix

Addendum

If the carbon concentrations of the feedstock substances A and B are incidentally equal in the stylized physical separations of Figs. 5 and 10, e.g., $c_{C,A} = c_{C,B} = 50wt.\%$, then there would not be carbon-related entropic chances in these processes, as illustrated in Fig. 11. The carbon-based entropies SE_C according to Eqs. (15), (17), and (23) would remain $SE_{in,C} = SE_{out,C} = SE_{max,C} = -\log_2(0.5) = 1$ Bit for all processes in Figs. 5 and 10, irrespective of the substance A recovery amount via stream 1 and the recovery stream 1 contamination by substance B which influence the substance-based entropic characteristics in Figs. 6 and 8. The carbon-based entropic dilution numbers $SDN_C \left(= RSE_C\right)$ would therewith remain constant 1 Bit / Bit according to Eq. (25). The entropic carbon collection efficiency SCE_C and recycling effectiveness RE_C would all remain constant 0% for these stylized processes according to Eqs. (18) or (26), and (27). The exemplary carbon-based entropic curvatures for the processes in Figs. 7 and 9 with $c_{C,A} = 65wt.\%$ and $c_{C,B} = 45wt.\%$ would change to constant values as in Fig. 11 if $c_{C,A} = c_{C,B}$. The substance-based entropies of the stylized physical separations in Figs. 6 and 8 are completely unaffected by the carbon contents (and their difference) of the inlet mixture parts A and B and the resulting carbon-based entropies as exemplified in Figs. 7, 9, and 11. These constructed $c_{C,A} = c_{C,B}$ cases additionally emphasize, beside the explanations in the "Stylized Physical Separation Processes for Solid Feedstock" section, that a recourse to evaluating carbon-based entropic characteristics should only take place for combustion or chemical conversion processes, where substances are destroyed and new target substances with high carbon content are possibly built up, where the carbon-based entropic changes, entropy increase by entropic C dilutions, are inherently significant, and arrangements for the abatement of such undesired carbon-based entropic deterioration and negative entropic carbon collection efficiencies SCE_C, e.g., the mentioned carbon losses via exhaust stream with diluted CO_2, are the main focus. For physical separation processes with conservation of the considered carbon-containing target substances j according to Eq. (7), the main focus for entropic process evaluation should rest upon target substance yields (RR) combined with target substance-based entropic characteristics as, e.g., exemplified via Figs. 6 and 8 for the stylized processes in Figs. 5 and 10.
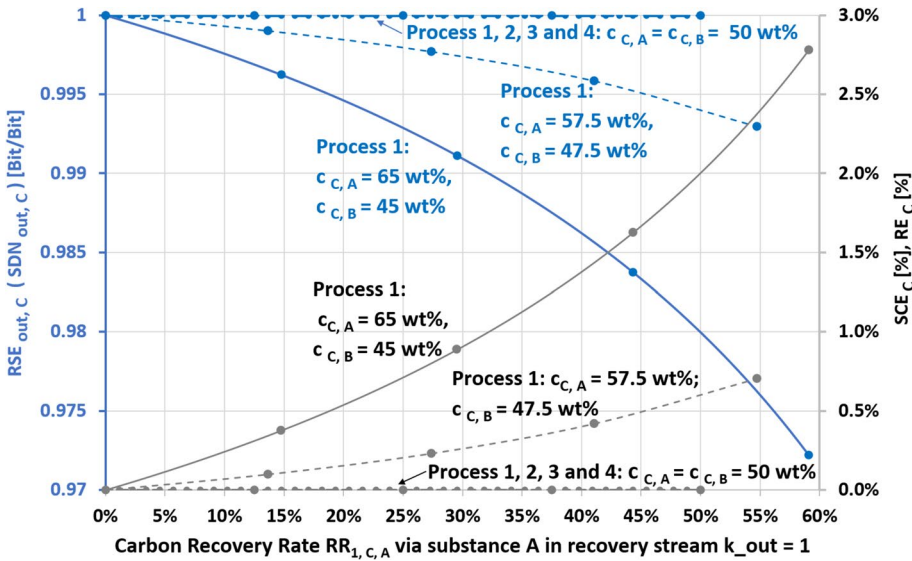
Fig. 11 Unchanged carbon-based entropic characteristics for the processes of Fig. 5 and Fig. 10 if substance A and B carbon contents approach $c_{C,A}=c_{C,B}$. Feedstock components A and B with equal carbon contents, decoupling of carbon-based entropic characteristics from substance based entropic characteristics

Data Availability Raw data cannot be shared at this time as the data forms part of ongoing work in the Waste4Future project. The Figures in the manuscript can be reproduced with the help of the formulae presented in this work.

Declarations

Conflict of Interest The authors declare no competing interests.

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