A New, Entropy Based Method To Support Waste and Resource Management Decisions

2 AUTHORS:

Helmut Rechberger
TU Wien

104 PUBLICATIONS **1,449** CITATIONS

SEE PROFILE

Paul H. Brunner
TU Wien

548 PUBLICATIONS **2,706** CITATIONS

SEE PROFILE

A New, Entropy Based Method To Support Waste and Resource Management Decisions

HELMUT RECHBERGER* AND
PAUL H. BRUNNER

Vienna University of Technology, Institute for Water Quality and Waste Management, Karlsplatz 13/226.4, A-1040 Vienna, Austria

A new method is presented to quantify the potential of a system to concentrate or dilute substances. The approach is based on a comprehensive material flow analysis and Shannon's statistical entropy function that is transformed by a three-step procedure. The result is a new function that can be applied to any defined system with known mass-flows and substance concentrations. In combination with materials balances, the method yields quantitatively the Relative Statistical Entropy (RSE) and the Substance Concentrating Efficiency (SCE) of a given system. When applied to compare five solid waste incinerators of increasing technology levels, the method (1) gave evidence of the potential of modern incinerators to control material flows and (2) pointed to future directions for further improvement of incineration technology. Case studies demonstrate that the new tool is well suited to support and complement existing methods for decision making in waste and resource management such as LCA.

Introduction

Traditional evaluation of waste management and treatment is often based on emission standards, recycling rates, product quality, costs, and public acceptance. There does not yet exist a quantitative method to assess the potential of a specific waste collection and/or treatment system to concentrate or dilute substances. Hence, waste management processes, which accomplish to concentrate or dilute substances in one of their residues receive a bonus only if the economic value of the residue increases due to the change in concentration. Examples for such an economic advantage are construction waste sorting plants, which produce "clean" secondary raw materials, or recycling processes for car batteries separating and concentrating lead as a valuable raw material.

Some waste treatment processes do not concentrate or dilute substances to an economically beneficial level. Still, from the point of view of resource conservation and environmental protection, such concentration changes can yield a significant advantage. E.g., incineration of municipal solid waste (MSW) results in two solid products: highly enriched filter ash, concentrating 90% of cadmium and 50−80% of other atmophilic heavy metals in only 2.5% of the mass of MSW (*1−3*), and a comparatively "clean" bottom ash that may be further upgraded to a construction material. Thus, incineration increases the resource potential and

decreases the hazardous potential, both of which are not reflected in economic terms yet. The necessity for a concentrating waste management system can also be derived from when investigating the total metabolism of resources such as heavy metals. After producing metals of high purity from ores (concentrating process) these metals are alloyed, installed in products, buildings, and infrastructure (dilution process). Once entering the waste management system the concentrations of metals in the various waste streams are usually low (<1%). To make them available for recycling processes a concentrating step is necessary. Hence, waste streams with a relevant metal content should be treated in concentrating processes (*4*).

The objective of this paper is to present a new method to quantify the potential of a single process or of combinations of processes to concentrate and/or dilute substances. In addition, a case study serves to exemplify how the method can be used as a complementary tool to analyze, evaluate, and optimize the design of waste incineration. This tool is intended as a necessary supplement to the existing array of evaluation instruments such as LCA (*5, 6*) for decisions regarding the management of wastes, resources, and the environment.

In the first part of this paper, the method, which is based on the methodologies of material flow analysis (*7*) and statistical entropy (*8*), is described in detail. In the second part, a case study on the development of MSW incineration technology during the last 80 years is used to show the potential of the method for evaluation and design purposes.

Method

Terminology of MFA. Material Flow Analysis (MFA) is a method to assess flows and stocks of materials within a defined system. It is based on the principle of the conservation of mass and has been developed to support the analysis, evaluation, and design of the so-called "metabolism of the anthroposphere" (*7*). An MFA of a system comprises a mass balance of all inputs, outputs, internal flows, and stocks of a system. A system consists of one or more processes and material flows. A process denotes the transformation, transport, or storage of materials. Processes are linked by flows of goods. A "good" consists of substances and has a positive (e.g. drinking water) or negative (e.g. wastewater) economic value. A "substance" is an element (e.g. hydrogen, oxygen) or a compound (e.g. water) (*9*). The distinction between goods and substances is important: Traditional economic data comprises mainly quantities for goods, while decisions regarding the management of resources and the environment require data on substances. The term "material" includes both goods and substances. So far, most MFAs consider the total flows and stocks of substances but neither their transformations nor speciations. Reasons for this fact are given in ref *7*. The "system boundary" is defined (a) in space by the "area" where the investigated processes are located and (b) by the time period of mass-balancing, ranging from a sampling period to the lifetime of a product, a waste treatment or production facility, depending on the goal of the MFA.

Overview of the Evaluation Problem. A material balance is established by combining mass-flows of all goods with the substance concentrations in these goods. In Figure 1 an exemplary MFA is displayed for a selected substance. For simplicity, the system is in an ideal steady-state. There is neither an exchange with an internal stock nor a stock. The evaluation of systems with internal stocks by the method presented here is possible but not discussed in this paper. An application to a system comprising stocks is demonstrated in ref *4*. The in- and output-goods of the system are defined

* Corresponding author phone: +41 1 823 5495; fax: +41 1 823 5226; e-mail: helmut.rechberger@eawag.ch. Present address: Swiss Federal Institute of Technology Zurich, Chair of Resource and Waste Management, P.O. Box 162, ETH Hoenggerberg HIF E21, CH-8093 Zurich, Switzerland.
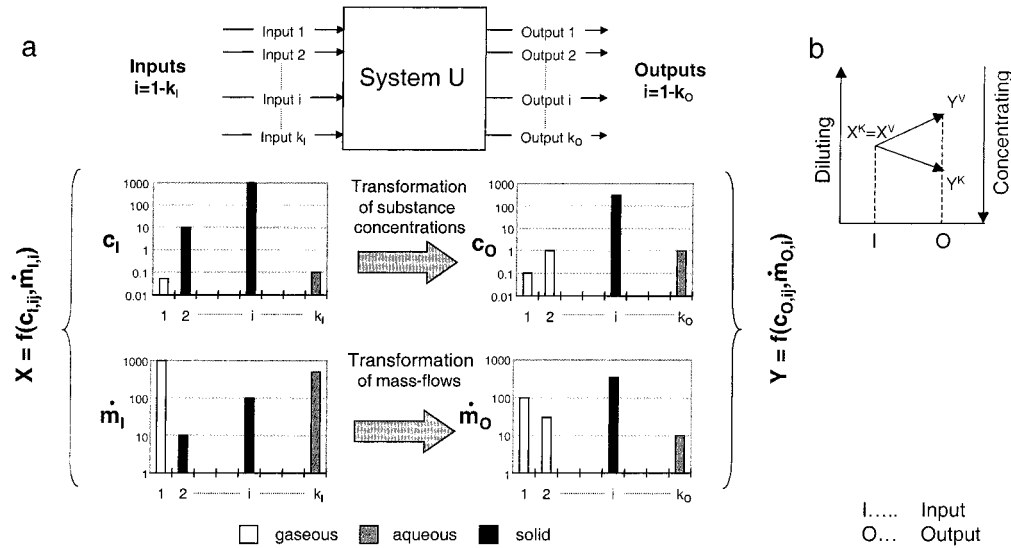
FIGURE 1. (a) A balance for substance j of a system U is determined by substance concentrations (c_{ij}) and mass-flows ($\dot{m}_i$) of all in- and output-goods. For all j and for steady-state conditions $\sum_{i=1}^{k_I} \dot{m}_{I,i} \cdot c_{I,ij} = \sum_{i=1}^{k_O} \dot{m}_{O,i} \cdot c_{O,ij}$. The system can be regarded as an algorithm that transforms sets of concentrations ($[c_I],[c_O]$) and mass-flows ($[\dot{m}_I],[\dot{m}_O]$) from the input (I) to the output (O). X and Y are functions that quantify the sets of concentrations and mass-flows. (b) Comparison of a concentrating system K (X^K-Y^K>0) and a diluting system V (X^V-Y^V<0).

by a *set of elemental concentrations* ($[c_I],[c_O]$) and a *set of mass-flows* ($[\dot{m}_I],[\dot{m}_O]$). Hence, a system can be regarded as a procedure that transforms an input-set of concentrations into an output-set of concentrations; the same applies to the mass-flows. Each system can be viewed as a unit that concentrates, dilutes, or leaves unchanged its throughput of a substance. To measure this transformation an appropriate function that quantifies the various sets is required. The transformation can be defined as the difference between the quantities for the input (X) and the output (Y). This allows determination of whether a system concentrates (X−Y>0) or dilutes (X−Y<0) substances.

Mathematical Solution of the Evaluation Problem

The Statistical Entropy Function. To calculate X and Y, a mathematical function from the field of Information Theory is used (*8*). This function originates from L. Boltzmann's statistical description of entropy. It is formally and mathematically identical with Boltzmann's well-known H-Theorem (*10*). In Information Theory, which was founded by C. E. Shannon in the 1940s (*11*), it is used to measure the loss or gain of information about a system. In statistics, the so-called Shannon-entropy is used to measure the variance of a probability distribution; the greater the variance, the less the information about the quantity of interest. It is important to note that the thermodynamic entropy denoted "S" [Joule per mole and Kelvin] as introduced by R. Clausius in 1865 (*12*) is *formally* identical with the statistical entropy "H" [bit] of Shannon; however, there is no physical relationship between the two entropy terms. The present paper is based on Shannon's statistical entropy and not on thermodynamic entropy.

The statistical entropy H of a finite probability distribution is defined by eqs 1 and 2 (*8*):

$$H(P_i) = -\lambda \cdot \sum_{i=1}^{k} P_i \cdot \ln(P_i) \geq 0 \qquad (1)$$

$$\sum_{i=1}^{k} P_i = 1 \qquad (2)$$

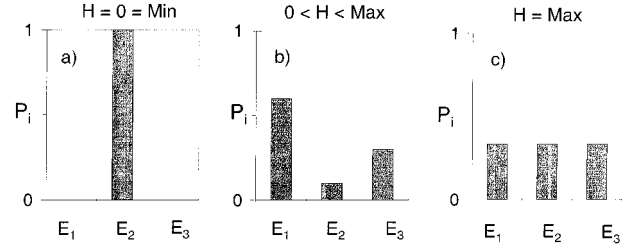where P_i is the probability that event i happens.

FIGURE 2. Probability distributions for a case where one of three events E_i can happen: extreme (a,c) and arbitrary (b) values of H. P_i probability for event i, $\sum P_i = 1$.

In statistical mechanics, λ is defined by Boltzmann as the ratio of the gas constant per mole (R) to Avogadro's number (N_0): $k_B = R/N_0$, unit J/K. In Information Theory, λ is replaced by the term $\{1/(\ln 2)\}$. This converts the natural logarithm in eq 1 to the logarithm to the base 2 (indicated as ld($\cdot$) in the following equations). The unit of H then becomes 1 bit (short for "binary digit"). For two events with equal probability ($P_1 = P_2 = 1/2$) H is 1 bit, and the connection to Coding and Information Theory becomes evident. The term $\{0 \cdot ld(0)\}$ is defined to be zero (*8*). Figure 2 illustrates three different distributions with extreme as well as arbitrary values of H. A case is presented in which one of three events (E_i) can happen. In Figure 2a, the probability of event two is unity ($P_2 = 1$). The statistical entropy of such a distribution is zero. In Figure 2c, the probabilities for all three events are identical. The entropy of such a distribution becomes a maximum. This can be proven using the Lagrange multiplier theorem (*13*). Since H is a positive definite function, the distribution in Figure 2a must yield the minimum value of H. Hence all other possible combinations of probabilities (e.g. the distribution in Figure 2b) must yield a value of H in the range between [0, Max].

Application of the Statistical Entropy Function. To be applied to sets of concentrations and mass-flows, the statistical entropy function is transformed in three steps (I–III).

(I) The statistical entropy function is applied to both the input and the output of the investigated system (cf. Figure 1). During this first step we assume that the mass-flows of the investigated set of goods are identical and unity ($\dot{m}_i = 1$). To quantify the variance of the attribute "concentration"

(instead of "probability"), eq 1 with $\lambda = 1/(\ln 2)$ is transformed replacing P_i by c_{ij}/c_j. The relative concentrations c_{ij}/c_j range between [0,1]. They can be regarded as a measure for the distribution of substance j among the goods. Eq 4 has no practical meaning and only serves to normalize the concentrations c_{ij}.

$$H^I(c_{ij}) = \mathrm{ld}(c_j) - \frac{1}{c_j} \cdot \sum_{i=1}^{k} c_{ij} \cdot \mathrm{ld}(c_{ij}) \qquad (3)$$

$$c_j = \sum_{i=1}^{k} c_{ij} \qquad (4)$$

where $i = 1, ..., k$; $j = 1, ..., n$; and c_{ij} is the concentration of substance j in good i.

Index k gives the number of goods in the set and n gives the number of investigated substances. As stated above, the maximum of H^I is found for $c_{1j} = c_{2j} = ... = c_{kj} = c_j/k$. Eq 3 consequently changes to

$$H_{\max}{}^I = \mathrm{ld}(k) \qquad (5)$$

Thus, H^I ranges from 0 to $\mathrm{ld}(k)$ for all possible sets of concentrations. Since the maximum of H^I is a function of the number of goods (k), the *relative* statistical entropy H_{rel}^I is used to compare sets with different numbers of goods. H_{rel}^I is defined as follows:

$$H_{\mathrm{rel}}{}^I(c_{ij}) = \frac{H^I(c_{ij})}{H_{\max}^I} = \frac{H^I(c_{ij})}{\mathrm{ld}(k)} \qquad (6)$$

The value of H_{rel}^I ranges between 0 and 1 for all possible sets of concentrations; it is dimensionless ([-]).

(II) To quantify additionally the attribute "mass-flow", eq 3 is modified. The mean concentration in a good (c_{ij}) is weighted with the mass-flow ($\dot{m}_i$) of this good; $\dot{m}_i$ can be regarded as the frequency of "occurrence" of the concentration c_{ij}. The entropy H^{II} of a mass-weighted set of concentrations therefore is (eqs 7 and 8 correspond to eqs 3 and 4)

$$H^{II}(c_{ij}, \dot{m}_i) = \mathrm{ld}(\dot{X}_j) - \frac{1}{\dot{X}_j}\sum_{i=1}^{k}\dot{m}_i \cdot c_{ij} \cdot \mathrm{ld}(c_{ij}) \qquad (7)$$

$$\dot{X}_j = \sum_{i=1}^{k}\dot{m}_i \cdot c_{ij} \qquad (8)$$

where $\dot{m}_i$ is the mass-flow of good i and $\dot{X}_j$ is the total substance flow induced by the set of goods.

(III) The last modification of the initial function concerns the gaseous and aqueous output-goods (emissions). In contrast to the products (e.g. solid residues) of the investigated system, the emissions are diluted in air and receiving waters, which results in an increase in entropy. In eqs 10 and 11, the index "geog" indicates the "natural" or geogenic concentrations of substances in the atmosphere and the hydrosphere. These concentrations serve as reference values to describe the dilution process. Factor "100" in eqs 10 and 11 means that the emission (c_{ij}, $\dot{m}_i$; e.g. measured in a stack or wastewater pipe) is mixed with a geogenic flow ($c_{j,\mathrm{geog}}$, $\dot{m}_{\mathrm{geog}}$), so that the concentration of the resulting flow ($\dot{m}_i + \dot{m}_{\mathrm{geog}}$) is 1% above $c_{j,\mathrm{geog}}$. It has been demonstrated that this approximation reflects the actual (unlimited) dilution in the environmental compartment sufficiently for $c_{ij} \gg c_{j,\mathrm{geog}}$ (13). For $c_{ij} = c_{j,\mathrm{geog}}$ there is apparently no dilution. Hence, as a rule of thumb in cases where $c_{ij}/c_{j,\mathrm{geog}} < 10$, eqs 10 and 11 have to be replaced by more complex terms that cover the

total range $c_{j,\mathrm{geog}} < c_{ij} < c = 1$ [g/g] (13). The applicability of eqs 10 and 11 has to be checked in any case.

$$H^{III}(\underline{c}_{ij}, \underline{\dot{m}}_i) = \mathrm{ld}(\dot{X}_j) - \frac{1}{\dot{X}_j}\sum_{i=1}^{k}\underline{\dot{m}}_i \cdot \underline{c}_{ij} \cdot \mathrm{ld}(\underline{c}_{ij}) \qquad (9)$$

where $\underline{c}_{ij}$ is defined as

$$\underline{c}_{ij} = \begin{cases} c_{j,\mathrm{geog,g}}/100 \\ c_{j,\mathrm{geog,a}}/100 \\ c_{ij} \end{cases} \qquad (10)$$

$$\text{for} \begin{cases} i = 1,...,k_g \\ i = k_g + 1,...,k_g + k_a \\ i = k_g + k_a + 1,...,k \end{cases} \text{for} \begin{cases} \text{gaseous} \\ \text{aqueous} \\ \text{solid} \end{cases} \text{outputs}$$

where k is the number of total output-goods, k_g is the number of gaseous output-goods, k_a is the number of aqueous output-goods, g stands for gaseous, a stands for aqueous, and $\underline{\dot{m}}_i$ is defined as

$$\underline{\dot{m}}_i = \begin{cases} \dfrac{\dot{X}_{ij}}{c_{j,\mathrm{geog,g}}} \cdot 100 \\ \dfrac{\dot{X}_{ij}}{c_{j,\mathrm{geog,a}}} \cdot 100 \\ \dot{m}_i \end{cases} \qquad (11)$$

$$\text{for} \begin{cases} i = 1,...,k_g \\ i = k_g + 1,...,k_g + k_a \\ i = k_g + k_a + 1,...,k \end{cases} \text{for} \begin{cases} \text{gaseous} \\ \text{aqueous} \\ \text{solid} \end{cases} \text{outputs}$$

with $\dot{X}_{ij}$ being the substance flow of output i.

The geogenic concentrations in eqs 10–12 can be replaced by more realistic background concentrations. In this case the influence of the actual surrounding environment is better reflected in the evaluation process of the investigated system. For a simple comparison of options this is usually not necessary.

Applied to the output of a system, eq 9 quantifies the distribution of substance j. The maximum of H^{III} is reached when all of substance j is directed to the environmental compartment with the lowest geogenic concentration ($c_{j,\mathrm{geog,min}}$). For heavy metals this is usually the atmosphere as there is $c_{j,\mathrm{geog,a}} > c_{j,\mathrm{geog,g}}$. The maximum of H^{III} is given by eq 12.

$$H_{\max,j}^{III} = \mathrm{ld}\left(\frac{\dot{X}_j}{c_{j,\mathrm{geog,min}}} \cdot 100\right) \qquad (12)$$

Using eqs 6 and 9–12, the *Relative Statistical Entropy* $\mathrm{RSE}_{j,O} \equiv H_{\mathrm{rel}}^{III}$ of the output (index O, measure Y in Figure 1) can be calculated. In the same way using eqs 6–8 and 12, the $\mathrm{RSE}_{j,I} \equiv H_{\mathrm{rel}}^{II}$ for the input (index I, measure X in Figure 1) of a system is obtained. The difference in the RSE_j between the in- and output of a system can be defined as Substance Concentrating Efficiency (SCE) of the system (13).

$$\mathrm{SCE}_j \equiv \frac{\mathrm{RSE}_{j,I} - \mathrm{RSE}_{j,O}}{\mathrm{RSE}_{j,I}} \cdot 100 \qquad (13)$$

The SCE_j is given as percentage and ranges between a negative value, which is a function of the input, and 100%. An SCE_j-value of 100% for substance j means the following: Substance j is transferred to 100% into one pure output-good. $\mathrm{SCE}_j = 0$ results if RSE_j-values of the in- and output are identical. This means that the system neither concentrates nor dilutes substance j. However, this does not imply identical
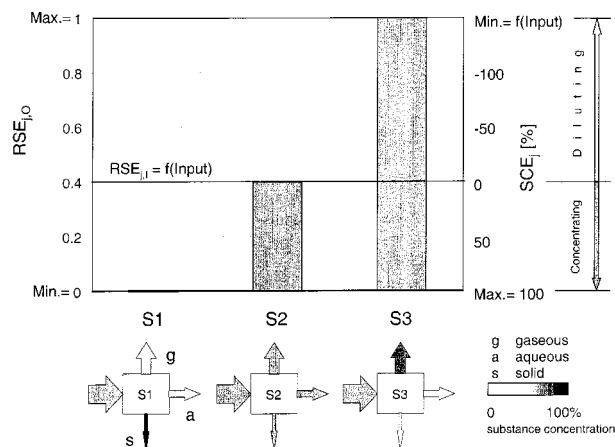
FIGURE 3. Relationship between Relative Statistical Entropies RSE_j and Substance Concentrating Efficiency SCE_j. S1 symbolizes a system that achieves maximum concentrating by producing a pure residue containing all of substance j. S2 is an example for a system that does not chemically discriminate between its outputs ($c_i = [c_0]$). S3 transfers the substance entirely into the atmosphere, which means maximum dilution.

sets of mass-flows and concentrations in in- and outputs. The SCE_j equals a minimum if all of substance j is emitted into that environmental compartment that allows for maximum dilution (in general the atmosphere). These relationships are illustrated in Figure 3. An example for the calculation of the SCE of a simple system is given in the Supporting Information.

Application and Results

In the following a case study of five MSW incinerators of different technological levels is presented. This example demonstrates the application of the RSE-method and shows how it can be used as a tool for the evaluation and design of waste treatment systems. The main advantage of the approach is that it becomes possible to quantify and value the change in concentrating and diluting of heavy metals during the course of incinerator development. This information, which was not available so far, is crucial for the further development of waste incinerators.

The five incinerators reflect selected furnace and air pollution control technology levels of the last 80 years. The data used in the study originate from investigations into state-of-the-art incinerators at different time periods. If measured quantities were not available, the missing data were assessed based on the technology used at the time.

First, the partitioning of four heavy metals by the incinerators was investigated. Cadmium, mercury, lead, and zinc have been chosen for this case study since MSW is an important carrier for these metals on a national level (*14*). For a comprehensive evaluation the focus on a limited set of metals is not sufficient. Other heavy metals, inorganic substances (e.g. S, Cl, F, N), and organic compounds have to be taken into account as well. However, priority should be given to those substances that are characteristic for the investigated waste stream.

Second, the relevance of emissions of state-of-the-art incinerators from the entropy point of view is investigated (exemplarily for Cd and Pb), and a combination of RSE and energy efficiency is proposed in order to exemplify the use of the method for design purposes.

The incinerator technologies are as follows (cf. Figure 4); all specifications are related to 1000 kg of MSW incinerated if not stated otherwise:

A: Incineration with energy recovery, no flue gas treatment, resulting in about 270 kg bottom ash and 22 kg boiler

ash. For this comparison, it is assumed that the amount of bottom ash is roughly the same for all five technologies investigated. Dust concentrations in the stack are about 2000 mg/Nm³. This technology can be regarded as the incineration standard ca. 1930.

B: Incineration with energy recovery and electrostatic precipitator (ESP) to remove particulates, stack emissions of about 150 mg/Nm³ (state-of-the-art ~1970), about 270 kg bottom ash, and 30 kg of mixed ash from the boiler and the ESP.

C: Same as B, but equipped with a more efficient ESP and supplemented by a two-stage wet scrubber and catalytic reduction of NO_x. The incineration residues are as follows: 270 kg bottom ash, and 31 kg mixed boiler and ESP ash (designated as fly ash); 2 kg filter cake resulting from the chemical-physical wastewater treatment, and 620 L of purified wastewater. The flue gas particulate emissions amount to 1–2 mg/Nm³. Data for case C have been taken from a comprehensive mass-balance study on the MSW incinerator of the city of Vienna that was upgraded to the described standard in 1989 (*15*).

D: In the 1990s, design efforts for the optimization of waste incineration were directed toward the improvement of the quality of the solid incineration residues. An example is the incinerator in Wels/Austria that was put into operation in 1995: The fly ash is chemically and thermally treated in order to remove and concentrate heavy metals in the filter cake for recycling purposes. The outputs of this incinerator are as follows: 270 kg bottom ash, 50 kg of treated fly ash (wet), 6 kg of filter cake, and 620 L of purified wastewater (*16*). The emissions are slightly lower than those of incinerator C due to a supplementary activated carbon filter.

E: To assess the effect of additional treatments of incineration residues, a "best possible" treatment process is defined and evaluated, too. This fictitious treatment E results in low emissions (observing stringent emission limits) and produces two solid residues with the composition of the average earth crust and the composition of a mineral ore. According to the objectives of waste management, E would be a most goal-oriented process since the protection of human health and the environment is guaranteed and all solid residues are suited either for recycling or land-filling in a so-called "final storage landfill" (no after care required) (cf. refs *1* and *17*). The specific mass flows, concentrations, and transfer coefficients of the materials investigated for incinerators A–E are given in Table 1.

To calculate the RSE_j-values, the following boundary conditions were chosen: The same MSW is supplied to all five incinerators. Hence, the change in the MSW composition over time is not considered. The following concentrations for heavy metals in MSW were taken for the present study: Cd = 10 mg/kg, Hg = 1.5 mg/kg, Pb = 600 mg/kg, and Zn = 1200 mg/kg. These values correspond to data published by several authors during the last two decades (*1, 15, 16, 18, 19*). Masses and concentrations of material flows for the technologies around 1930 (A) and 1970 (B) were assessed based on the corresponding material flows in modern incinerators. For example, it was presumed that a single boiler around 1930 had a similar fly ash removal as a boiler of today. Values for technologies A and B are rough figures. However, it can be shown that the uncertainty of these figures does not affect results and conclusions. The resulting Relative Statistical Entropy values are given in Figure 5. The mean RSE was calculated using the weighting procedure as described in the Supporting Information.

The improvement in the RSE from incinerator A–C is evident for all four heavy metals. While early incinerators with inappropriate flue gas cleaning (A and B) have been important sources for metals in the environment (*20, 21*),
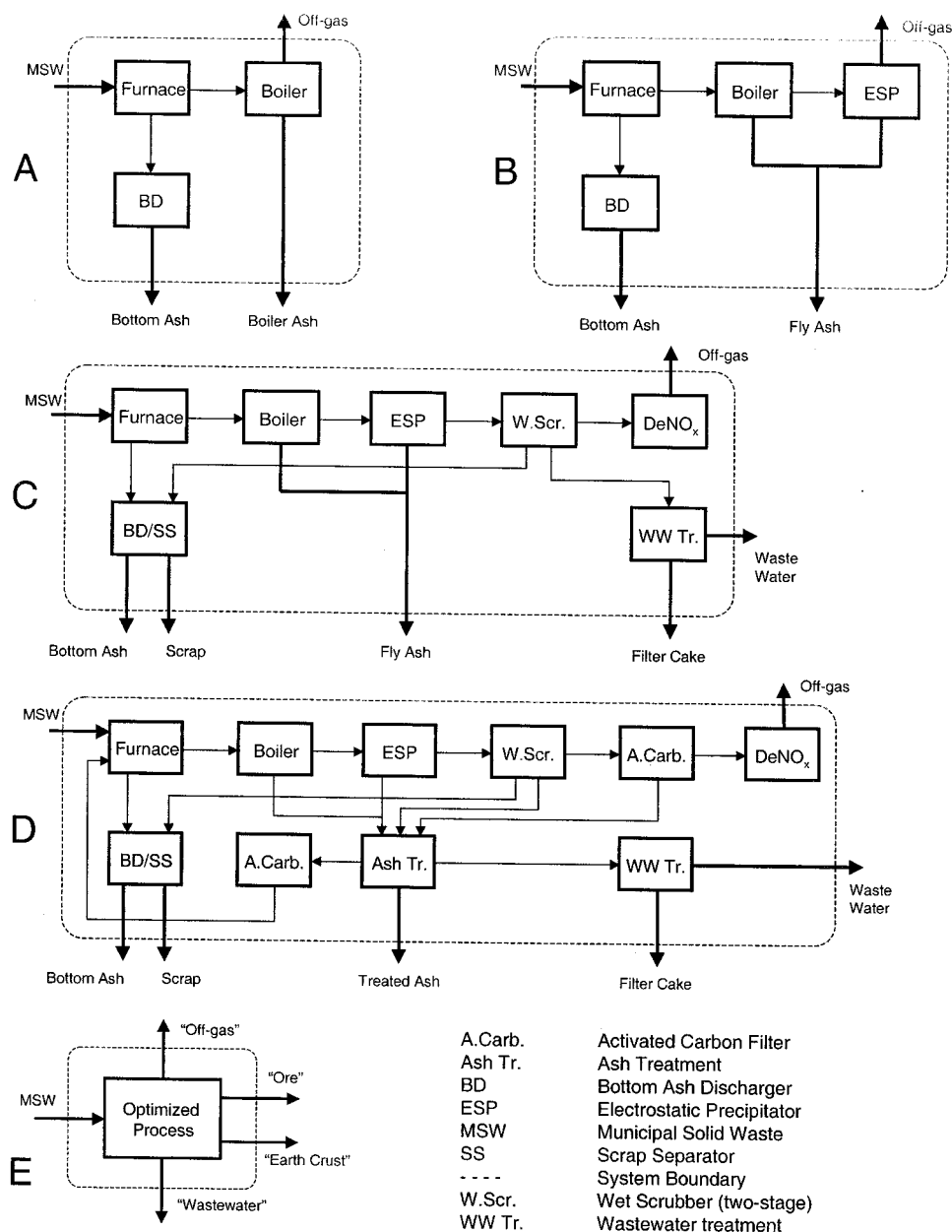
FIGURE 4. Flowcharts of incinerator technologies A—E. Flue gas treatment has been highly improved over the years (A: first half of the 20th century, D: past decade). New technologies (D) focus on posttreatment of solid residues. A most favorable incineration process (E) produces environmentally compatible emissions as well as earth crust or ore-like residues.

modern facilities (C and D) concentrate these metals in their solid residues. To investigate the relevance of the flue gas emissions, the RSE for Cd and Pb of technology C are presented in Figure 6 as a function of the metal concentration in the flue gas. The calculations are based on the assumption that the partitioning between bottom ash (BA) and raw gas (RG) remains constant ($TK_{BA} + TK_{RG} = 1$) and that the transfer-coefficient (TK) of the entire flue gas cleaning process ranges from 0 to TK_{RG}. This means that a maximum of $100 \cdot TK_{RG}$ % of a substance is transferred into the atmosphere; it explains why the curves in Figure 6 do not end at RSE = 1 (= 100% into the atmosphere). The actual emissions of state-of-the-art incinerators (Cd < 0.001 mg/Nm³; Pb <0.1 mg/Nm³) are in the horizontal area of the curves ($y' \cong 0$). Hence the impact of these emissions on the entropy is insignificant. This means that the emissions are not relevant for the material management. The emission limits, which are based on human and environmental toxicity, are located in the transition area

where y' becomes > 0. This means that they are reasonable from an entropy point of view, too.

Due to efficient wastewater treatment, emissions are not shifted from the atmosphere to the hydrosphere by the wet flue gas cleaning technologies applied in C and D (cf. transfer-coefficients in Table 1). Gaseous and aqueous emissions are similarly low. Hence, the crucial differences between technologies are the amount and composition of the solid residues (22). Figure 5 reveals that in the state-of-the-art incinerators C and D, Zn and partly Pb are already controlled in an optimum way (small difference to E). Nevertheless, manu-facturers are investigating new methods for the treatment of bottom ash and fly ash (23). Additionally, new process technologies based on pyrolysis, gasification, vitrification, or high-temperature treatment appear on the market (23–28). These new technologies have different energy efficiencies (ratio of useful energy to energy input [-]) since producing useful residues from MSW is an energy consuming process.

TABLE 1. Specific Masses ($\dot{m}_i$), Selected Metal Concentrations (c_{ij}), and Transfer Coefficients (TK_{ij}) for Incinerators A–E[a]

solid: mg/kg; *aqueous*: mg/L; *gaseous*: mg/Nm³; *transfer coefficient*: -

	mass kg/t MSW $\dot{m}_i$	Cd $c_{i,Cd}$	$TK_{i,Cd}$	Hg $c_{i,Hg}$	$TK_{i,Hg}$	Pb $c_{i,Pb}$	$TK_{i,Pb}$	Zn $c_{i,Zn}$	$TK_{i,Zn}$
Technology A									
bottom ash	268	3	0.08	0.4	0.07	1000	0.45	1800	0.4
boiler ash	22	56	0.12	0.3	0.004	2000	0.072	8600	0.16
off-gas	7000	1.4	0.80	0.25	0.92	52	0.48	95	0.44
Technology B									
bottom ash	268	3	0.08	0.4	0.07	1000	0.45	1800	0.4
fly ash	30	210	0.67	18	0.37	8800	0.46	20000	0.53
off-gas	7000	0.45	0.25	0.15	0.56	11	0.1	15	0.07
Technology C									
bottom ash	270	3	0.08	0.4	0.07	1500	0.67	1 800	0.4
scrap	3	n.d.	0.0	n.d.	0.0	n.d.	0.0	n.d.	0.0
fly ash	31	300	0.92	25	0.51	6500	0.33	24000	0.6
filter cake	2	5	0.001	300	0.4	300	0.001	300	0.001
wastewater	620	0.01	0.001	0.001	0.0004	0.005	0.00001	0.1	0.00005
off-gas	6000	0.01	0.006	0.01	0.04	0.006	0.0001	0.01	0.00005
Technology D									
bottom ash	270	3	0.08	0.4	0.07	1 000	0.45	1 500	0.34
scrap	3	n.d.	0.0	n.d.	0.0	n.d.	0.0	n.d.	0.0
treated fly ash	50	67	0.34	0.05	0.002	6150	0.51	13 000	0.54
filter cake	6	970	0.58	230	0.91	3 700	0.04	26 000	0.13
wastewater	620	0.01	0.001	0.001	0.0004	0.005	0.00001	0.1	0.00005
off-gas	6250	0.005	0.003	0.002	0.008	0.005	0.0001	0.005	0.00003
Technology E									
"earth crust"	270–299	0.2	0.006	0.08	0.02	13	0.006	70	0.02
"ore"	1–30	4400	0.99	2000	0.964	50000	0.994	40000	0.98
wastewater off-gas		*same (or lower) transfer coefficients as incinerator technologies C and D*							

[a] Due to advancing Air Pollution Control Technologies the metal transfer into the solid residues increased (e.g. for Cd from 20% to over 99%, for Hg from 8% to over 98%). Differences between modern incinerator technologies are no longer in their emissions but in the composition of their residues (cf. composition of fly ashes and filter cake of C and D).
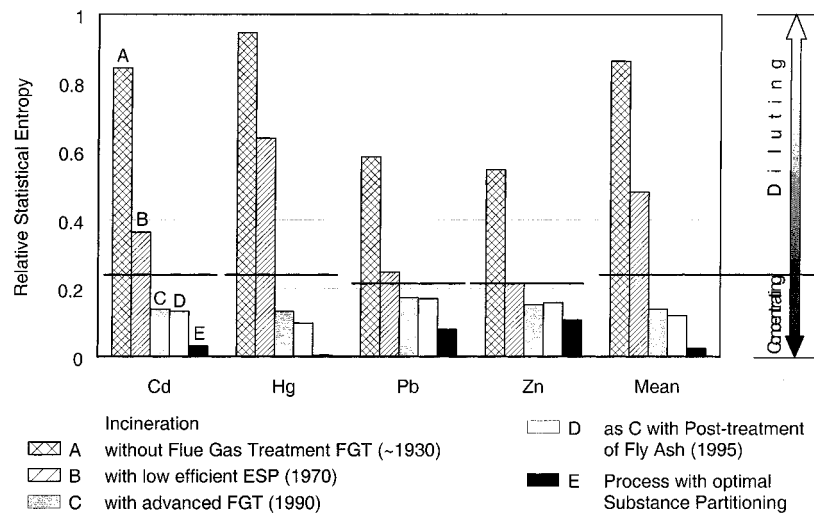

FIGURE 5. Relative Statistical Entropy values for the partitioning of Cd, Hg, Pb, Zn, and their weighted mean values for incineration technologies A–E. The horizontal lines indicate the calculated RSE values for the input (MSW). Incinerators without advanced flue gas treatment (A and B) dilute heavy metals into the environment. Modern incinerators (C and D) concentrate these substances. Technologies C and D partition Pb and Zn nearly as good as the most favorable process E.

The nonweighted ratio between energy efficiency and relative statistical entropy for selected substances can serve as one indicator for the efficiency of the materials management of the technologies.

Discussion

The method as described in this paper is useful to determine the distribution of conservative, inorganic substances that are not transformed by the processes investigated. As mentioned in the method chapter, most MFA do not consider the speciation of elements and their transformations yet. Nevertheless, an investigation into the extension of the method to include different compounds of an element (CO_2, CO, CH_4, C_xH_y, etc.) seems attractive; it will be the subject of a subsequent paper. A second restriction of the approach is that it does not explicitly consider the toxicity of a substance. Implicitly this can be achieved by using toxicity based threshold values instead of geogenic concentrations in eqs
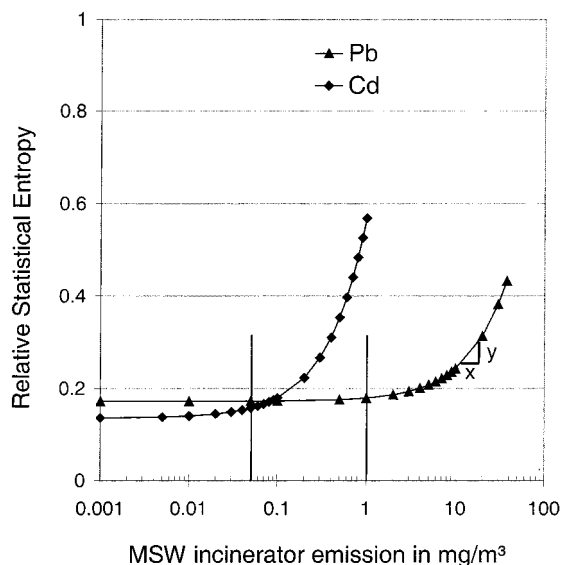
FIGURE 6. Relative Statistical Entropy of substance partitioning as a function of emission into the atmosphere. The vertical lines indicate the emission limits for Cd and Pb (Cd: 0.05 mg/Nm^3; Pb+Zn+Cr: 2 mg/Nm^3 (29)). Actual emissions of state-of-the-art incinerators (Cd <0.001 mg/Nm^3; Pb <0.1 mg/Nm^3) are in the horizontal areas of the curves ($y' = 0$).

10−12 (cf. also the Supporting Information). However, in ref *13* examples are given that suggest a correlation and a possible link between the toxicity centered evaluation system and the entropy approach: Cases where toxicity based emission limits are exceeded show a significant increase in entropy as well (cf. also Figure 6). This will be subject to further investigations. An advantage of the proposed method is that emissions into different environmental compartments and via solid products are weighted against each other. The indicator statistical entropy is highly sensitive relating to whether a substance is directed into air, groundwater, or a solid product. Of course, the "indicator" RSE is as meaningful as the data it is based on. If information about the uncertainty of data (standard deviations) is available, the variances of RSE-values can be calculated by a Gaussian approximation, and confidence intervals can be established. If this is not possible, the RSE can be used to determine the necessary accuracy of the basic data (sensitivity analysis).

Up to now the RSE-method has been used mainly to evaluate the management of wastes in *single facilities* such as MSW incinerators, mechanical separation plants for MSW (*30*), recycling plants for construction and demolition wastes (*13*), cocombustion of wastes in cement kilns, and utilization of incinerator ashes as secondary raw materials to produce cement (*31, 32*). In addition the RSE-method can be applied to *systems* that consist of several processes, e.g. the combination of MSW incineration and land-filling of incinerator residues. Then it is also possible to investigate the contribution of each single process within the system to the overall performance. This indicates that the method is not limited to the evaluation of waste treatment systems but can be applied to all kinds of materials management systems (e.g. mining, manufacturing) (*4*). The only prerequisites are consistent data sets for each process and some practice in the application of the method. Data sets can often be found in environmental impact statements or in reports of manufacturers and operators on compliance testing. In the future, it is recommended to collect data for auditing, submission, and testing in a way that allows calculating RSE and SCE, too.

A major advantage of the method is that the flows of many materials through complex systems can be quantified by a single measure (per substance) and thus become easy to compare. The new indicator may well be combined with other evaluation criteria like energy efficiency (% SCE/kWh) and costs (% SCE/$). If time series of the RSE are established, the impact of measures such as new recycling schemes or prevention measures on the material flows through waste management can be assessed. The case studies quoted above (*13, 30−32*) point out that the RSE method improves the understanding of the "metabolism" of a system. By assessing the concentrating and diluting potential, the RSE method provides new insights that are complementary to the results of existing methods for decision making in environmental and resource management such as LCA. We suggest that this entropy based evaluation method be used more widely to assess the performance of systems that manage resources and wastes.

Supporting Information Available

An example of how to calculate the SCE of a simple system and information about the weighting of substance-specific RSE values. This material is available free of charge via the Internet at http://pubs.acs.org.

Nomenclature

λ	constant
$[c_i]$	set of concentrations of k goods; $[c_i] = [c_1, c_2, ..., c_k]$
BA	bottom ash
c	concentration [mg/kg; %; etc.]
E	event
EC	index for earth crust
g	weighting factor [-]
geog	index for geogenic concentration
H	Statistical Entropy [bit]
H_{rel}	Relative Statistical Entropy RSE, standardized between [0,1]
i	index for goods
I	index for input
I, II, III	index for steps in the derivation of the RSE
j	index for substances
k	number of goods
k_B	Boltzmann's constant: $1.380658 \cdot 10^{-23}$ [J/K]
ld	logarithm to the base 2
$\dot{m}$	mass-flow of a good [kg/h; kg/t input; etc.]
n	number of substances
N_0	Avogadro's number: $6.02214 \cdot 10^{23}$ [1/mole]
O	index for output and ore
P_i	probability for event i
R	gas constant per mole: 8.31451 [J/(mole·K)]
RG	raw gas
RSE	Relative Statistical Entropy [-]
$\overline{RSE}$	mean Relative Statistical Entropy [-]
SCE	Substance Concentrating Efficiency [%]
$\dot{X}$	substance-flow [kg/h; g/t input; etc.]
X, Y	functions
y'	first derivative, $y = f(x)$

TK   transfer-coefficient [-]

w   index for a weighted quantity

Literature Cited

(1) Belevi, H.; Mönch, H. *Environ. Sci. Technol.* **2000**, *34*, 2501–2506.

(2) Morf, L. S.; Brunner, P. H.; Spaun, S. *Waste Manage. Res.* **2000**, *18*, 4–15.

(3) Morf, L. S.; Brunner, P. H. *Environ. Sci. Technol.* **1998**, *32*, 1825–1831.

(4) Rechberger, H.; Graedel, T. E. Submitted to *Ecological Economics*.

(5) Finnveden, G.; Albertsson, A.-Ch.; Berendson, J.; Eriksson, E.; Höglund, L.; Karlsson, S.; Sundqvist, J.-O. *J. Cleaner Prod.* **1995**, *3*, 189–199.

(6) White, P. R.; Franke, M.; Hindle, P. *Integrated Solid Waste Management. A Life Cycle Inventory*; Blackie A&P: Glasgow, U.K., 1995.

(7) Baccini, P.; Brunner, P. H. *Metabolism of the Anthroposphere*; Springer: New York, 1991.

(8) Vogel, F. *Beschreibende und schliessende Statistik*; R. Oldenbourg: Vienna, 1996.

(9) Atkins, P. W.; Beran, J. A. *General Chemistry*, 2nd ed.; Scientific American Books: New York, 1992.

(10) Boltzmann, L. *Vorlesungen über Gastheorie*; Barth: Leipzig, 1923.

(11) Shannon, C. E. *Bell System Technol. J.* **1948**, *27*, 379–443, 623–656.

(12) Clausius, R. *Pogg. Ann.* **1856**, *125*, 353–400.

(13) Rechberger, H. Ph.D. Thesis, Vienna University of Technology, Institute for Water Quality and Waste Management, 1999.

(14) Morf, L. S. Ph.D. Thesis, Vienna University of Technology, Institute for Water Quality and Waste Management, 1998.

(15) Schachermayer, E.; Bauer, G.; Ritter, E.; Brunner, P. H. *Messung der Güter- und Stoffbilanz einer Müllverbrennungsanlage*; Federal Environment Agency, Monographien Band 56, Vienna, 1995.

(16) Morf, L.; Ritter, E.; Brunner, P. H. *Güter- und Stoffbilanz der MVA Wels*; Final Report, Institute for Water Quality and Waste Management, Vienna University of Technology, Austria, 1997.

(17) Belevi, H.; Langmeier, M. *Environ. Sci. Technol.* **2000**, *34*, 2507–2512.

(18) Brunner, P. H.; Mönch, H. *Waste Manage. Res.* **1986**, *4*, 105–119.

(19) Vehlow, J. *Umwelt Technologie Aktuell – International Edition* **1996**, *3*, 144–160.

(20) Greenberg, R. R.; Zoller, W. H.; Gordon, G. E. *Environ. Sci. Technol.* **1978**, *12*, 566–573.

(21) Chillrud, S. N.; Bopp, R. F.; Simpson, H. J.; Ross, J. M.; Shuster, E. L.; Chaky, D. A.; Walsh, D. C.; Chin Choy, C.; Tolley, L.-R.; Yarme, A. *Environ. Sci. Technol.* **1999**, *33*, 657–662.

(22) Sawell, S. E.; Chandler, A. J.; Eighmy, T. T.; Hartlen, J.; Hjelmar, O.; Kosson, K.; van der Sloot, H. A.; Vehlow, J. *Biomass Bioenergy* **1995**, *9*, 377–386.

(23) *Thermische Abfallbehandlung*; Thomé-Kozmiensky, K. J., Ed.; EF-Verlag für Energie- und Umwelttechnik GmbH: Berlin, 1994.

(24) *Thermoselect-Verfahren zur Ent- und Vergasung von Abfällen*; Schweitzer, F. J., Ed.; EF-Verlag für Energie- und Umwelttechnik: Berlin, 1994.

(25) Calaminus, B.; Stahlberg, R. *Waste Manage. Res.* **1998**, *18*, 547–556.

(26) Baumgärtel, G.; May, K.-W. Das Siemens Schwel-Brenn-Verfahren. In *Reaktoren zur thermischen Abfallbehandlung*; Thomé-Kozmiensky, K. J., Ed.; EF-Verlag für Energie- und Umwelttechnik: Berlin, 1993; pp 189–200.

(27) Mayer, A.; Rey, T. *Waste Manage. Res.* **1996**, *16*, 27–33.

(28) Faaij, A.; Hekkert, M.; Worrell, E.; von Wijk, A. *Resour. Conserv. Recy.* **1998**, *22*, 47–82.

(29) Federal Ministry for Environment, Youth, and Family. *Luftreinhalteverordnung für Kesselanlagen*; BGBl. Nr. 1988/360 i.d. F. BGBl. 1993/185, Federal Ministry for Environment, Youth, and Family: Vienna, 1993.

(30) Rechberger, H. *Waste Manage. Res.* **2001**, *19*, 186–192.

(31) Rechberger, H. *Waste Manage. Res.* **2001**, *19*, 262–268.

(32) Rechberger, H.; Brunner, P. H. *Österreichische Wasser- Abfallwirtschaft* **2000**, *52*, 3/4, 53–57.