Integration of Life Cycle Assessment into Environmental Process Engineering Practices

A thesis submitted to the
Budapest University of Technology and Economics
for the degree of

Doctor of Philosophy in Chemical Engineering

by

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under the supervision of

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Doctor of Science

Budapest 2008
ACKNOWLEDGEMENTS

I would like to thank my supervisor Prof. Dr. Peter Mizsey for his guidance and support during the course of this research. I am grateful for his encouragement and for giving me an opportunity to use all the facilities available at the Department of Chemical and Environmental Process Engineering.

I would like to thank Dr. Daniela Jacob and the colleagues at the Max-Planck Institute for Meteorology for their help and support in the field of atmospheric simulations and modelling during my visit at the Institute in Hamburg, Germany. I also would like to thank Professor Sandor Kemeny for his help with statistical problems.

Moreover, I would like to thank all the colleagues at the department, especially to Mrs. Gabriella Ling-Mihalovics, for helping me even with technical, scientific, and intellectual questions, and for maintaining a friendly and family atmosphere at the department.

I am forever indebted to my parents, my brother Peter, and Viola for encouraging and supporting my studies and helping me get through the difficulties I encountered.

I would like to thank the Deutsche Bundesstiftung Umwelt, the Hungarian Scientific Research Foundation (OTKA), and the Pro Progressio Foundation for the financial support they provided during my PhD studies.
ABSTRACT

During the last decades, the need for environmentally-consciousness and the establishment of sustainable practices have become guiding principles in the milieu of chemical engineering. These new challenges for engineering are referred to as the ‘fourth paradigm’ of the chemical process design.

During process design there must be an objective function that enables designers to rank design alternatives. If environmentally-conscious design is to be performed, the design alternatives should be evaluated according to how they meet environmental targets. To realise such design action, however, quantitative environmental measures are sought.

Life Cycle Assessment (LCA) is the only standardized and thus widely-accepted tool currently used to assess the environmental loads of products and/or processes. The Life Cycle Impact Assessment (LCIA), as a part of LCA, is the scientific technique for assessing the potential environmental impacts of industrial systems and their associated products.

The main motivation of this thesis work is to investigate the applicability of LCA in process engineering in order to support environmentally-conscious decision making. The thesis deals with the data uncertainties of different LCIA methods and promotes the suitability of damage oriented methods in decision making. Moreover, environmental problems related to air pollution and waste solvent treatment are analysed in order to show the advantages of environmentally-conscious process evaluation over classic, economic evaluation methods.

The single score impact indicators of the two important LCIA methods (Eco-indicator 99, and the European Union’s CAFE CBA method) are investigated and a clear linear dependency is detected between them. The detected similarity might help and support the work of both LCIA tools and mutually exploit the merits of them.

I demonstrate through a case study referring to the environmental evaluation of the annual airborne emission inventory of an industrialized city, that, contrary to quantitative analysis of the emission inventory, damage-oriented LCIA tools such as EI-99 can successfully be applied to identify and rank air pollutants and their sources with highest environmental loads.

It is found that the application of the single score indicators of damage-oriented LCIA methods allows the determination of clear environmental preferences – something that would not be possible if full spectrums of single scores’ uncertainties are included in the analysis.

The right selection of the proper air pollution abatement techniques is also a challenge for environmentally-conscious process design. The investigation of three flue gas desulphurization (FGD) techniques proves that, with the application of FGD processes at the emission sites, environmental impacts can be reduced by about 80% as compared to the uncontrolled release of sulphur oxides into air.

A ranking system is set up for the investigated FGD techniques according to their environmental performance. The results show that intra-furnace limestone addition and wet scrubbing processes (techniques using similar physical and chemical principles) have similar environmental indices; however, FGD with wet-limestone scrubbing is found to be slightly better from an environmental viewpoint. The regenerative process which utilises the sorption/reduction/oxidation cycle for SO\(_2\) removal shows better environmental performance. This means (according to design heuristics) that recovery and recycling of SO\(_2\) is the most preferable option from the environmental viewpoint.

In connection with the FGD techniques, the effect of supplementary installed FGD units at high capacity power plants on regional air pollution in the Carpathian Basin is investigated.
The results show that FGD units significantly reduce both horizontal and vertical dispersion of the emitted SO₂, as well as its transboundary transport. Besides SO₂ removal efficiency, dispersion and accumulation also depend on the seasonal weather conditions. During winter, dispersion and accumulation are higher than in other seasons. Due to this phenomenon, higher SO₂ removal efficiency is needed in winter to guarantee similar air quality features to other seasons.

The preservation of natural resources is also an important challenge of the process design. The economic treatment of chemical solvents is one of the important issues in European Union’s environmental policies.

In this work, the treatment alternatives of a non-ideal solvent mixture containing azeotropes are investigated to determine the preferable option. For the recovery of the solvent mixture, two different separation alternatives are evaluated: a less effective alternative and a novel design based on hybrid separation tools. The third investigated waste solvent treatment alternative is incineration with heat utilization. Contradictions between environmental and economic evaluations are detected: economic features clearly favour total recovery; however, the environmental evaluation shows that if a recovery process of low efficiency is applied, its environmental burden can be similar or even higher than that of incineration. This motivates engineers to design more effective recovery processes and to reconsider the evaluation of process alternatives during environmental decision making.
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CHAPTER 1  INTRODUCTION

1.1 Motivation

Chemical process engineering is a complex engineering task that involves synthesis, analysis, optimisation activities, and the evaluation of design alternatives. Traditionally, system optimisation in chemical and process engineering applications has focused on maximising economic outcomes. The development of industrial technologies has enabled the transformation of the environment in different ways which change the nature and extent of the environmental impacts of industrial activities. Resource depletion, air, water and land pollution are examples of environmental problems which have arisen as a result of intensified interventions into the environment.

One of the main problems associated with these activities is that they may not have an immediate effect and some may have global impacts on the environment. This is becoming apparent with the increasing scientific awareness of the cumulative and synergistic effects of some of the environmental impacts over space and time.

Industry plays a paramount role with respect to the environment, not only as one of the main sources of environmental impacts but also as one of the main actors regarding new solutions. Industry-related environmental policy was originally intended to control emissions of various environmental elements. It was widely thought that corrective technical measures at the end of the pipe would sufficiently reduce environmental impact. However, as we have seen through the years, this is insufficient to halt progressive environmental degradation and also lacks the flexibility essential to an evolving industry (Sonnemann, 2002).

During the last decades, the emphasis has shifted from the end-of-pipe waste reduction techniques to the pollution prevention. The rise in environmentally-consciousness and a move towards more sustainable practices have become guiding principles in the milieu of chemical engineering. This is a new challenge for engineers and is referred to as the ‘fourth paradigm’ of chemical process design (Fonyo, 2004).

On the other hand, during process design there must be an objective function that enables designers to rank design alternatives. If environmentally-conscious design is to be performed, design alternatives should be evaluated according to how they meet environmental targets. To support such design, however, quantitative environmental measures are sought.

This work focuses on the designing and planning phase of the process engineering. The principles of prevention of environmental problems are fully recognized and accepted; moreover, it is also accepted that future environmental problems should be avoided and prevented in the present. Environmental evaluations and comparison of alternative solutions for existing industrial and common engineering problems may avoid problems for future generations.

The main motivation of this thesis work is to investigate the applicability of Life Cycle Assessment (LCA) to process engineering in order to support environmentally-conscious decision making and process design.
1.2 The aims of this work

After the emergence of the fourth paradigm in the chemical process design, several classes of concepts and techniques which support environmentally-conscious decision making have been formulated. However, many questions and issues are still open and under research.

Main aim of my study is to show that the concept of Life Cycle Assessment can be successfully integrated in environmentally-conscious process design. The emphasis is laid on numerical tools applying aggregated, single score impact indicators for the expression of environmental impacts.

Two important types of environmental evaluation used in chemical engineering are investigated:
- the construction and design selection, where alternative technical solutions of a prescribed problem are compared. In this case, environmental performance is determined for continuous and consistent operational parameters of the alternative techniques, and;
- environmentally-conscious process engineering, where the optimal operational parameters of the selected technical solution or process are determined in order to enable selection of the option with the lowest environmental load.

The applicability and suitability of LCA to this end is demonstrated in several case studies related to real environmental problems.

According to this, the aims of the study can be outlined as follows:

1. Investigation of two important and frequently-used environmental impact assessment methods under the consideration of their data uncertainties in order to show where there are dependencies and similarities between them. If similarities and dependencies can be found that might help by the selection of the proper impact assessment tool from the numerous ones.

2. Environmental problems due to air pollution in industrialized cities are investigated. The single score impact assessment method is used in order to rank different air pollutants to help determine their sources.

3. The environmental performances of basically different air pollution abatement techniques are determined with the help of LCA in order to support selection of the process type with the lowest environmental load.

4. Investigation of effects and efficiency of flue gas desulphurization techniques on air quality on regional scale are detailed.

5. Selection of the process alternative, and furthermore, the optimal process performance, is determined with the help of LCA integrated into the process-design stage. The study is connected with the problem of treatment of waste solvents. Moreover, differences between the environmental and economic evaluation of treatment options are presented in order to show the obvious necessity of environmental evaluations in process engineering.
1.3 Approach

The main numerical environmental impact assessment tool applied in this study is the Eco-indicator 99 methodology which uses aggregated, single score impact indicators. Work with impact indicators is supported by the software SimaPro. In addition, the Cost-benefit Analysis environmental impact assessment tool of the European Union’s CAFE Programme is used and investigated.

Investigation a) of the uncertainties, b) the assessment of the aggregated indicators with the highest probability, and c) the statistical calculations related to these problems are carried out by using Monte Carlo simulations.

According to the investigation of effectiveness and efficiency of air pollution control techniques, atmospheric transport of the pollutants is modelled and simulated. Simulations are carried out by the regional atmospheric model REMOTE coupled with the chemistry package RADM II.

1.4 Scope and contribution

The scope of this thesis is to demonstrate the applicability and suitability of LCA to environmental process engineering. Two major environmental problems are addressed during the investigations in order to present real cases and problems for environmentally-conscious evaluations, studies, and analyses. These fields are, namely:

- Air pollution
- Waste solvent treatment

In the field of air pollution, a major and pressing environmental problem that shows need for research and analysis is selected. This issue is the improvement of air quality in Europe; an issue that requires a knowledge-based approach combined with technical and scientific analyses and policy development which will lead to the adoption of a thematic strategy on air pollution. This work may assist stakeholders and policy makers in making better decisions for the sake of environmental protection.

First, the effects of the uncertainties of two different impact assessment tools are investigated. The uncertainties can influence evaluation of the results and may make the interpretation of the results too difficult for decision makers. It is therefore necessary to have a clear picture of how far the uncertainties should be considered, and how the results of the impact assessment should be interpreted.

Moreover, existing air pollution abatement techniques - especially for the reduction of SO\(_2\) emissions - are studied and analysed. There are numerous alternative options for the reduction of SO\(_2\) emission; however, evaluation based on their environmental aspects has not yet presented. This work aims at helping decision makers to select from alternatives.

The effect of air pollution abatement techniques on the dispersion of air pollutants is also studied to help elucidate the proper policy for the application of these techniques.
Chapter 1  Introduction

The recovery and recycling of waste solvents is covered in one of the most important environmental directives of the European Union. It is not clear, however, under what circumstances solvent recovery should be preferred over incineration. This work presents a model set up for the environmental evaluation of different alternatives. Moreover, differences between environmentally-driven evaluation and economic evaluation are presented.

1.5 Outline of the dissertation

To develop the objectives mentioned in the previous sections, the thesis work is distributed into 6 Chapters which cover the following aspects:

Chapter 1 is an introduction to the dissertation and includes a motivation statement, aims of the work, approach, the scope and contributions, and an outline of the dissertation.

Chapter 2 reviews the literature, gives an overview about the tools and techniques generally applied for environmental assessment and a detailed description of the elements of Life Cycle Assessment especially important to process design.

Chapter 3 presents a comparison of two environmental assessment methods. The investigation provides the basis for the selection of the life cycle impact assessment method applied to environmental evaluation throughout the thesis.

Chapter 4 shows an example of the application of Life Cycle Assessment during chemical processes. The investigated field is air pollution prevention, with special regard paid to flue gas desulphurization.

Chapter 5, in connection with Chapter 4, is a study concerning the importance and effectiveness of flue gas desulphurisation on regional air pollution.

Chapter 6 presents an example of the application of Life Cycle Assessment for process design in the field of waste solvent treatment. Thermal treatment of waste solvents with incineration and material recovery with distillation are investigated related to an existing industrial problem. Model results are compared in order to select the optimal process performance. Similarities and differences between economic and environmental evaluation are also presented.

Chapter 7 comprises the major new results determined during the different studies of this work.
CHAPTER 2 LITERATURE REVIEW

2.1 Techniques for environmental assessment

The chemical industry one of the keys to a nation’s economic health; however, it is also one of the main sources of pollution (Pereira, 1999). Over the past several decades, significant efforts have addressed how to reduce industrial pollution and the focus has gradually shifted from downstream pollution control to more aggressive practices of trying to prevent pollution (Allen and Rosselot, 1997; El-Halwagi, 1997). Pollution prevention requires environmentally-conscious process engineering and design.

Since the early 1990s, many works have been focused on environmentally-conscious process integration and design. Douglas (1992) introduced the hierarchy approach to pollution prevention research. Smith and Petela (1991a-b, 1992) proposed pollution prevention considering process waste and utility waste. Nourelding et al. (1999) elaborated a concept to derive cost optimal mass exchange networks (MENs) with minimum emissions. Wang and Smith (1995) proposed design targets for minimum wastewater generation in process plants based on MENs. Recently, Foo et al. (2006) proposed a graphical technique called the property surplus diagram and cascade analysis technique to establish a targeting property-based material reuse network. However, in each of these works, detailed environmental impact assessments were not studied systematically, and minimization of pollutants was solely taken into account (Zhang et al., 2008).

Later on, quantitative evaluation of environmental impacts of chemical processes became the focus. Pistikopoulos and Stefanis (1998) proposed a methodology for estimating various environmental impacts using the cost function concept. Cave and Edwards (1997) proposed the environmental hazard index (EHI) method for chemical process selection in the early design stage to avoid processes with a high potential of pollution. Gunasekera and Edwards (2006) introduced the method of the atmosphere hazard index (AHI) based on five environmental impact categories which was used to assess the inherent environmental friendliness of chemical processes in order to rank alternative routes. Further methods and criteria for evaluating chemical processes are available in the systematic review of Sharratt (1999).

Burgess and Brennan (2001) also conducted an overview of important techniques for environmental assessment related to chemical processes. The Environmental Impact Assessment allows the identification of the environmental effects of one economic activity, usually at a specific location and at a single point in time (UNEP, 1996). The Best Practicable Environmental Option Assessment (BPEO) technique utilises an ‘integrated environmental index’ (IEI), which is calculated from pollutant releases to air, water and land (Carlyle, 1995). Environmental Risk Assessment (ERA) involves the estimation and evaluation of risk to the environment caused by a particular activity or exposure (Burgess and Brennan, 2001). This activity or exposure may be linked to any part of a product life cycle; for example in the use or disposal of a product, but also from processing, transport and storage of materials during product manufacture and distribution.

Life Cycle Assessment (LCA) is also a technique for environmental evaluation of processes/products. Contrary to different classes of techniques in the field of support systems for making environmental decisions LCA is currently the only standardized tool for this purpose. LCA was originally developed to assess the environmental burdens of products. However, it became the most accepted and applied technique of environmental evaluation of chemical processes (Azapagic, 1999; Azapagic and Clift, 1999; Burgess and Brennan, 2001).
For this reason, a detailed description of the structure and main points of LCA are presented in Chapter 2.2.

2.2 Life Cycle Assessment

The International Standards Organization (ISO) was founded in 1946 in Geneva, Switzerland. ISO has established non-mandatory international standards for the manufacturing, communication, trade and administrative sectors. For environmental management, ISO has created the ISO 14000 series, a new generation of standards to foster national and international trade in compliance with international standards to protect the environment. In this way, some common guidelines and similarities between environmental management and business management are established for all businesses regardless of size, activity or geographical location. Several of the ISO 14000 standards refer to the previously mentioned procedural and analytical tools (Sonnemann et al., 2004).

In order to consider environmental impacts of a product’s life cycle systematically, the life cycle assessment methodology has been developed.

2.2.1 Definition of Life Cycle Assessment

Life Cycle Assessment (LCA) of a product comprises the evaluation of environmental effects produced during its entire life cycle, from its origin as a raw material until its end, usually as waste (Sonnemann et al. 2004). This concept is also referred as the ‘cradle to grave’ approach.

The origins of the LCA methodology can be traced to the late 1960s (Miettinen and Hamaleinen, 1997). Initial studies were simple and generally restricted to calculating energy requirements and solid wastes, with little attention given to evaluating potential environmental effects. By the end of the 1980s, numerous studies had been performed, but with different methods and without a common theoretical framework.

Since 1990, attempts have been made to develop and standardise the LCA methodology under the coordination of the Society of Environmental Toxicology and Chemistry (SETAC) (Udo de Haes, 1993). In 1993 SETAC published a ‘Code of Practice’, which presents general principles and a framework for the conduct, review, presentation, and use of LCA findings.

ISO composed an international standard for LCA which is similar to that of SETAC; however, it includes some differences in the interpretation phase: ISO has included further analysis and sensitivity studies. Thus LCA is the only standardized tool currently used to assess the environmental loads of a product (Burgess and Brennan, 2001).

The procedural steps of LCA are described in different standards of the ISO 14040 series for environmental management. ISO 14040 (1997) provides the general framework for LCA. The standards divide the LCA procedure into four separate steps:

1. Goal and scope definition
2. Inventory analysis
3. Impact assessment
4. Interpretation

Each step is detailed described in different ISO standards; Figure 2.1 shows the connections between them.

LCA is not necessarily carried out in a single sequence. It is an iterative process where subsequent rounds may result in increasing levels of detail (from screening LCA to full LCA) or lead to changes in the first phase promoted by the results of the last phase (Sonnemann et al. 2004).

2.2.1.1 Goal and scope definition

The goal and scope definition is designed to obtain the required specifications for the LCA study. During this step, the intended audience and the strategic aspects are defined and answered. To carry out the goal and scope of the LCA study, the following procedures have to be followed:

1. Define the purpose of the LCA study, ending with the definition of the functional unit, which is the quantitative reference for the study.
2. Define the scope of the study, which embraces two main tasks:
   2.1. Establish the spatial limits between the product system under study and its neighbourhood that will be generally called ‘environment’; see Figure 2.2.
   2.2. Detail the system through drawing up its unit processes flowchart, taking into account a first estimation of inputs from and outputs to the environment.
3. Define the data required, which includes a specification of the data necessary for the inventory analysis and for the subsequent impact assessment phase.
Definition of system boundaries is a crucial point of the LCA. Environmental impacts of only those elementary flows which cross the system boundary are considered and evaluated during the LCA study. The boundaries are in close relation with the scope of the study; however, their definition is based on individual choices.

The life cycle of the product/process under study is connected to the life cycle of other processes and products; see Figure 2.3. The more detail (sub-products and sub-processes) included in the system boundary the more data, and thus time and money, required for the LCA study. Therefore, it is desirable to reduce the extent of the system boundaries as much as possible; however, improperly defined system boundaries may result in misleading results.

One example is presented here for the so called Limited Life Cycle Assessment (LLCA) which makes possible a saving of the time and money needed to perform the study by reducing the system boundary. Boundary conditions are defined so that only one or several stages of the whole life cycle are considered, see Figure 2.3.

In the selected example (Vignes, 2001), an attempt is made to select the proper treatment alternative for pesticide-containing waste waters. Three alternatives are compared: incineration (EU-supported solution), biological treatment and release of the untreated waste water to nature.

If system boundaries are restricted to the direct emissions of the treatment alternatives (release of pesticides, TOC and chloride content of the waste water), off-site incineration is the most favourable alternative. However, if indirect environmental burdens of the treatment process (transportation, fuel oil, energy requirements) are also included, the study shows that biological treatment is a significantly better alternative for the treatment than incineration; moreover, incineration causes even more environmental impacts than the release of the untreated waste water to nature.

It can be concluded that the results of the Limited Life Cycle Assessment should be carefully interpreted.
2.2.1.2 Inventory analysis

During inventory analysis all the data of the unit processes within a product system are collected and related to the functional unit of the study. In this case, the following steps must be considered:

1. Data collection, which includes the specification of all input and output flows of the processes within the product system (product flows, i.e., flows to the other unit processes, and elementary flows from and to the environment).
2. Normalization to the functional unit, which means that all data collected are quantitatively related to one quantitative output of the product system under study; usually 1 kg of material is chosen, but often other units such as a car or 1 km of mobility are preferable.
3. Allocation, which means the distribution of emission and resource extractions within a given process throughout its different products, e.g., petroleum refining providing naphtha, gasoline, heavy oils, etc.
4. Data evaluation, which involves a quality assessment of the data (e.g., through a sensitivity analysis).

The result of the inventory analysis, consisting of the elementary flows related to the functional unit, is often called the Life Cycle Inventory (LCI) table.
2.2.1.3 Impact assessment

The impact assessment phase aims at making the results from the inventory analysis more understandable and more manageable in relation to human health, the availability of resources, and the natural environment. To accomplish this, the inventory table will be converted into a smaller number of indicators.

The mandatory elements of LCIA are (ISO 14042, 2002):
1. Selection of impact categories, indicators, and models. Impact categories are classes of a selected number of environmental impacts such as global warming, acidification, etc.
2. Classification of environmental loads within the different categories of environmental impact.
3. Characterisation of environmental loads by means of a reference pollutant typical of each environmental impact category. The results of the characterization step are known as the environmental profile or environmental performance of the product system.

Optional elements are (ISO 14042, 2002):
- Calculating the magnitude of category indicator results relative to reference values (normalisation) means that all impact scores (contribution of a product system to one impact category) are related to a reference situation.
- Grouping indicators (sorting and possibly ranking).
- Weighting (across impact categories) is a quantitative comparison of the ‘seriousness’ of the different resource consumption or impact potential of the product, aimed at covering and possibly aggregating indicator results across impact categories.
- Data analysis to better understand the reliability of the LCIA results.

Figure 2.4 illustrates relationships between the results of the life cycle inventory analysis, indicators, and category endpoints for one impact category for the example of acidification. In the first step, relevant information is selected from the life cycle inventory (‘materials causing acidification’). In the second step, mathematical models are applied to assess the magnitude of the impact referring to the category indicator (in this case, proton release). The third step is assessment of the environmental relevance or potential for damage referring to the category (in this case, assessment of the damage to an ecosystem due to acidification).
2.2.1.4 Interpretation

The interpretation phase aims at evaluating the results from the inventory analysis or impact assessment and compares them with the goal of the study defined in the first phase. The following steps can be distinguished within this phase:

1. Identification of the most important results of the inventory analysis and impact assessment.
2. Evaluation of the study’s outcomes, consisting of a number of the following routines: completeness check, sensitivity analysis, uncertainty analysis and consistency check.
3. Conclusions, recommendations and reports, including a definition of the final outcome, comparing outcomes with the original goal of the study, drawing up recommendations, procedures for a critical review, and the final reporting of the results.

The results of the interpretation may lead to a new iteration step in the study to include a possible judgement of the original goal.

2.3 Two main schools: mid- and endpoints based methods

The impact assessment phase of the LCA requires the modelling of the environmental impacts caused by the materials and energy flows collected in the inventory table. This can be carried out with the help of life cycle impact assessment methods. According to Jolliet et al. (2003 and 2004), LCIA methods can be classified as:

1. classical or midpoints-based impact assessment methods, and
2. damage oriented or endpoints-based (single score) methods.

As shown in Figure 2.5, LCI results with similar impact pathways (e.g. all elementary flows influencing stratospheric ozone concentration) can be grouped into impact categories at midpoint level, also called midpoint categories.
Figure 2.5 Overall scheme of linking LCI results via midpoint categories to damage categories (Jolliet et al., 2003).

A midpoint indicator characterizes the elementary flows and other environmental interventions that contribute to the same impact. The characterisation generally occurs via well-known mathematical functions. The term ‘midpoint’ expresses the fact that this point is located somewhere on an intermediate position between the LCI results and the damage (or endpoint) on the impact pathway. Owing to this, midpoints-based methods give complex, multi-faceted information about the impact potential of the system under study; however, the different impacts are not ranked and are not comparable with each other (it can not be decided which system has lower environmental loads if system A has higher global warming potential and system B causes more ionizing radiation). Therefore this kind of analysis does not really support comparison of different systems from an environmental perspective.

A further step may allocate the midpoint categories to one or more damage categories. This step requires modelling of the damages caused by the potentials calculated in the different impact categories. In practice, a damage indicator result is a simplified model of a very complex reality, giving a coarse approximation to the quality status of the item or to the change in this quality. It is a big advantage of the endpoints-based method that the results in the different damage categories can be aggregated. The aggregation of different impacts into a single number makes the interpretation and communication of the results to stakeholders easier, and this method, therefore, is often more desirable to decision-makers (Lenzen, 2006).

Although users may choose to work at either the midpoint or damage levels, a current tendency in LCIA method development aims at reconciling these two approaches. Both of them have their merits, and optimal solutions can be expected if the midpoint-oriented methods and the damage-oriented methods are fitted into a consistent framework (Bare et al., 2000).
For instance, Life Cycle Initiative, the joint project of UNEP (United Nations Environment Programme) and SETAC also recognised the need for shifting the interpretation of LCA results in the direction of the more easily-interpretable endpoint-based approaches instead of the more complex midpoint approaches. A comprehensive LCA framework is under construction which combines classical methods with damage-oriented methods in order to utilize the benefits of both approach types (Jolliet et al., 2004 and 2005).

2.4 Available LCIA tools and methods

A number of impact assessment methodologies are available for the LCA practitioner. They differ, and often there is not a single one obvious choice between them.

The Life Cycle Impact Assessment Programme, as part of the Life Cycle Initiative, aims at the enhancement of the availability of sound LCA data and methods and at guidance about their use. Within the framework of the Programme, the most used LCIA methods are collected and listed. According to this list, the most important LCIA methods are listed below with a short description obtained from the Programme’s homepage (http://lcinitiative.unep.fr/).

2.4.1 Eco-Indicator 99

Eco-indicator 99 is a damage-oriented LCIA method focusing on the weighting step as the key problem to solve. Weighting has been simplified by:

- using just three endpoints; this minimizes mental stress among LCA-panellists regarding the need to take into account too many issues;
- defining these three issues as endpoints that are reasonably easy to understand.

The weighting problem has not been solved, but weighting and interpretation of results without weighting has been made easier. Other new ideas in the methods are the consistent management of subjective choices using the concept of cultural perspective. This has lead to a good documentation of the choices and to the publication of three versions, each with a different set of choices. Other issues are the introduction of the DALY approach, the introduction of the PAF and PDF approach, as well as the surplus energy approach. A detailed description is presented in Chapter 2.5.

2.4.2 EDIP97 and EDIP2003

EDIP97 (Environmental Design of Industrial Products) is a thoroughly documented midpoint approach covering most of the emission-related impacts, resource use and working environment impacts (Wenzel et al., 1997; Hauschild and Wenzel, 1998) with normalization based on person equivalents and weighting based on political reduction targets for environmental impacts and working environment impacts, and a supply horizon for resources. Ecotoxicity and human toxicity are modelled using a simple key-property approach wherein the most important fate characteristics are included in a simple modular framework requiring relatively few substance data for calculation of characterization factors.

The updated version of the methodology, EDIP2003, supports spatially-differentiated characterization modelling which covers a larger part of the environmental mechanism than EDIP97 and lies closer to a damage-oriented approach. This part of the general method development and consensus programme covers investigations of the possibilities for inclusion.
of exposure in the LCIA of non-global impact categories (photochemical ozone formation, acidification, nutrient enrichment, ecotoxicity, human toxicity and noise).

2.4.3 EPS 2000d

The EPS (Environmental Priority Strategies) 2000d impact assessment method was developed for use in supporting choices between two product concepts. Category indicators are chosen for this purpose, which represent actual environmental impacts on any or several of five safeguard subjects: human health, ecosystem production capacity, biodiversity, abiotic resources and recreational and cultural values.

The characterization factor is the sum of a number of pathway-specific characterization factors describing the average change in category indicator units per unit of an emission. Characterization factors are only available where there are known and likely effects.

Weighting factors for the category indicators are determined according to people’s willingness to pay to avoid one category indicator unit of change in the safeguard subjects.

2.4.4 IMPACT 2002+

The IMPACT 2002+ life cycle impact assessment methodology proposes a feasible implementation of a combined midpoint/damage approach, linking all types of life cycle inventory results (elementary flows and other interventions) via 14 midpoint categories to four damage categories. For IMPACT 2002+ new concepts and methods have been developed, especially for the comparative assessment of human toxicity and eco-toxicity.

Human Damage Factors are calculated for carcinogens and non-carcinogens, employing intake fractions, best estimates of dose-response slope factors as well as severities. The transfer of contaminants to human food is no longer based on consumption surveys, but accounts for agricultural and livestock production levels. Indoor and outdoor air emissions can be compared and the intermittent character of rainfall is considered. Both human toxicity and ecotoxicity effect factors are based on mean responses rather than on conservative assumptions.

Other midpoint categories are adapted from existing characterizing methods (Eco-indicator 99 and CML 2002). All midpoint scores are expressed in units of a reference substance and related to the four damage categories of human health, ecosystem quality, climate change, and resources.

Normalization can be performed either at midpoint or at damage level. The IMPACT 2002+ method presently provides characterization factors for almost 1500 different LCI-results, and can be downloaded at http://www.epfl.ch/impact.
2.4.5 Swiss Ecoscarcity Method (Ecopoints)

The method of environmental scarcity – sometimes called Swiss Ecopoints method – allows a comparative weighting and aggregation of various environmental interventions by use of so-called eco-factors. The method supplies weighting factors for different emissions into air, water and topsoil/groundwater as well as for the use of energy resources. The eco-factors are based on the annual actual flows (current flows) and on the annual flow considered as critical (critical flows) in a defined area (country or region).

Eco-factors were originally developed for the area of Switzerland (see references below). There, current flows are obtained from the newest available statistical data, while critical flows are deduced from the scientifically-supported goals of Swiss environmental policy, each as of publication date. Later, sets of eco-factors were also made available for other countries, such as Belgium and Japan.

The ecopoints method contains common characterization/classification approaches (for climate change, ozone depletion and acidification). Other interventions are assessed individually (e.g. various heavy metals) or as a group (e.g. NMVOC, or pesticides).

The method is planned to be used for standard environmental assessments, e.g., with specific products or processes. In addition, it is often used as an element of companies’ environmental management systems where assessment of the company's environmental aspects is supported by such a weighting method.

The method was first published in Switzerland in 1990. A first amendment and update was made for 1997. A next version, based on 2004 data, has been published in 2005.

2.4.6 TRACI

TRACI (Tool for the Reduction and Assessment of Chemical Impacts) is an impact assessment methodology, developed by the U.S. Environmental Protection Agency, which facilitates the characterization of environmental stressors that have potential effects, including:

- ozone depletion,
- global warming,
- acidification,
- eutrophication,
- tropospheric ozone (smog) formation,
- ecotoxicity,
- human health criteria–related effects,
- human health cancer effects,
- human health noncancer effects, and
- fossil fuel depletion.

TRACI was originally designed for use with life cycle assessment, but it is expected to find wider application to pollution prevention and sustainability metrics.
2.5 Eco-indicator 99

The following section contains a short description of EI-99 methodology. The content of the section is based on the EI-99 methodology manual (Goedkoop et al., 2000) as well as on the works of Sonnemann et al. (2004) and Koning et al. (2002).

The Eco-indicator 99 (EI-99) is a damage-oriented approach for LCIA. It models the cause-effect chain up to the damage (endpoint) and expresses the environmental impact with a single score, the so-called ‘Eco-indicator point’. The higher the impact, the higher the EI-99 point. The EI-99 method assesses the environmental impacts of individual substances; however, since the EI-99 points are additive they can be used for calculation/assessment of the environmental impacts of complex process or product systems.

There are over 200 predefined Eco-indicator 99 scores for commonly used substances and materials available. These can be grouped as:

- raw materials,
- airborne emissions,
- waterborne emissions,
- emissions to soils,
- final waste flows,
- nonmaterial emissions.

There is no absolute value for the eco-indicator scores. They only have a relative value: similar processes might be compared based on the eco-indicator points. The scale of Eco-indicators is chosen in such a way that the value of one point is representative for one thousandth of the yearly environmental load of one average European inhabitant.

The working environment of the EI-99 is provided by the software SimaPro (System for Integrated Environmental Assessment of Products) which supports the set up of the LCI and the evaluation (characterization, normalization, weighting, and single scores) with the EI-99 method.

It is a very helpful feature of the software package that it also includes large databases describing the input/output data of common industrial processes such as:

- production of chemicals,
- production of energy carriers and fuels,
- production of energy (electricity and heat from several fuels, import from several countries),
- extraction and processing of minerals,
- several types of transportation, etc.

Detailed documentation, large implemented LCI databases and a high level of flexibility make the EI-99 method a powerful and effective tool of environmental process engineering.

EI-99 is acknowledged as being a standard investigation tool for LCA and is applied in about one hundred countries. The method is frequently used and referred to both in the LCA praxis, and scientific life. Numerous studies have been published in the literature where EI-99 is chosen for environmental evaluation (i.e. Hofstetter, 2002; Hofstetter et al., 2003; Raluy et al., 2003; Erol et al., 2005; Ortiz et al., 2007; Ribeiro et al., 2007).
2.5.1 Structure of the method

The methodology of EI-99 has been developed with regard to the fact that the most critical and controversial step in Life Cycle Assessment is the weighting step. If weighting between impact categories has to be done by the evaluating panel of the LCA study it is better if (1) the number of impact categories to be weighted is as small as possible and (2) the impact categories to be weighted are easy to explain to a panel.

According to this, three impact categories are considered in the EI-99 methodology:

1. damage to human health,
2. damage to ecosystem quality, and
3. damage to resources.

User defined weighting of the impact categories during the determination of the final Eco-indicator point makes the methodology very flexible, and subjective choices can be clearly explained.

The EI-99 methodology is designed to link the inventory results to the indicator system. The core concept of the methodology is shown in Figure 2.6.

Figure 2.6 The core concept of the EI-99 methodology.

The first step includes the construction of the life cycle model and the preparation of the life cycle inventory, including data about resource-requirements, land-use, and emissions.

In the second step, the probable effects and damages linked to the material and energy flows determined in the inventory phase are modelled through a scientific calculation step. This step contains the damage modelling and the scientific assessment of the three forms of damages.

1) In the model for Human Health, four sub-steps are used:
   a) Fate analysis, linking an emission (expressed as mass) to a temporary change in concentration.
   b) Exposure analysis, linking this temporary concentration to a dose.
   c) Effect analysis, linking the dose to a number of health effects (such as number and types of cancers, and respiratory effects).
   d) Damage analysis, linking health effects to the number of Years Lived Disabled (YLD) and Years of Life Lost (YLL).

The indicator of Human Health is DALY (number of Disability-Adjusted Life Years). This indicator, also used by the World Bank and the WHO, measures the total amount of illness,
due to disability and premature death attributable to specific diseases and injuries. The DALY concept thus compares YLD and YLL. Health is simply added across individuals. That is, two people each losing 10 years of disability-free life are treated as the same loss as one person who loses 20 years (Murray et al., 1996).

2) In the model for Ecosystem Quality two different approaches are used:
   a) Toxic emissions and emissions that change acidity and nutrients levels go through the procedure of:
      i. Fate analysis, linking emissions to concentrations
      ii. Effect analysis, linking concentrations to toxic stress or increased nutrient or acidity levels.
      iii. Damage analysis. Linking these effects to the increased potentially disappeared fraction for plants.
   b) Land-use and land transformation is modelled on the basis of empirical data on the quality of ecosystems as a function of the land-use type and the area size.

For measuring toxic stress, PAF (Potentially Affected Fraction of species, Hamers et al., 1996) is typically used as dimension which can be used to interpret the fraction of species that is exposed to a concentration equal to or higher than the NOEC.

For measuring the effects of acidification, eutrophication and land-use, the PDF (Potentially Disappeared Fraction) of species is selected in EI-99. The PDF is used to expresses the effects of pollutants on vascular plant populations in an area; and can be interpreted as the fraction of species that has a high probability of no occurrence in a region due to unfavourable conditions.

3) In the model for Resource extraction two steps are included:
   a) Resource analysis, which can be regarded as a similar step to fate analysis as it links extraction of a resource to a decrease in resource concentration.
   b) Damage analysis, linking lower concentration to the increased efforts to extract the resource in the future.

The seriousness of the extraction of natural resources is assessed on the basis of the relation between the concentration of the resource in nature and the energy needed for its extraction.

The unit of Resources damage category is the ‘surplus energy’ in MJ per kg extracted material. This is the expected increase in extraction energy per kg of extracted material, assuming that the deposits with the highest concentration of a given resource are depleted first, leaving future generations to deal with the lower concentrations.

As a summary, Table 2.1 gives an overview of the damage and impact categories used in the EI-99 methodology.
Chapter 2  Literature review

<table>
<thead>
<tr>
<th>Damage category</th>
<th>Name</th>
<th>Impact category</th>
<th>Name</th>
<th>Dimension</th>
</tr>
</thead>
<tbody>
<tr>
<td>Human Health</td>
<td>Carcinogens</td>
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<td>DALY</td>
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<td></td>
<td>Respiratory, organics</td>
<td></td>
<td>DALY</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Respiratory, inorganics</td>
<td></td>
<td>DALY</td>
<td></td>
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<td></td>
<td>Climate change</td>
<td></td>
<td>DALY</td>
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<td>DALY</td>
<td></td>
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<td></td>
<td>Ozone layer depletion</td>
<td></td>
<td>DALY</td>
<td></td>
</tr>
<tr>
<td>Ecosystem Quality</td>
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<td>PDF<em>m²</em>yr</td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>Ecotoxicity</td>
<td></td>
<td>PAF<em>m²</em>yr</td>
<td></td>
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<td></td>
<td>Land use</td>
<td></td>
<td>PDF<em>m²</em>yr</td>
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<td>MJ surplus</td>
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<td></td>
<td>Fossil fuels</td>
<td></td>
<td>MJ surplus</td>
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</tbody>
</table>

Table 2.1 Damage categories and impact categories considered in the EI-99 method.

The last step of the concept is the valuation procedure which aims to establish the significance of the damage. The valuation procedure includes the normalisation and the weighting of the results in the three damage categories. These steps are detailed discussed in the following two chapters.

2.5.1.1 Normalization

The three damage categories all have different units. In normalisation, the relative contribution of the calculated damages to the total damage caused by a reference system is determined. The purpose of normalisation in the EI-99 is to prepare the environmental impact data for additional procedures (grouping and weighting).

EI-99 has been developed for Europe, therefore damage categories are normalised on a European level: damage caused by 1 European per year European during a reference period (mostly based on 1993 as base year, with some updates for the most important emissions) is determined and used as reference. European normalisation values are shown in Figure 2.7. Normalisation means in practice that environmental impact indicators determined by the model for a certain substance are divided by the normalisation factors (bold numbers) in each damage category.

At the end of the normalisation, environmental impacts of the certain substance are characterized by three dimensionless numbers ($D_k$) referring to the three damage categories ($k$).
Figure 2.7 Normalisation values for Europe in the EI-99 methodology. (Goedkoop et al., 2000).
2.5.1.2 Weighting

Weighting of the results obtained in the three damage categories makes possible the expression of the environmental impact with one single score. The weighting supports the evaluation of the normalised damage indicators from the several aspects based on the individual choices of LCA experts regarding the individual weighting of the three damage categories.

The result of the LCA study after the weighting step is a single score per functional unit which can be calculated as shown in Equations (2.1) and (2.2).

\[
\begin{align*}
    i &= \sum_{k=1}^{3} (\delta_k \cdot D_k) \\
    \delta_{HH} + \delta_{EQ} + \delta_{R} &= 1000
\end{align*}
\]

where

- \(i\): eco indicator point referring to the functional unit of the LCA study [EI-99 point/functional unit]
- \(k\): damage category: Human Health (HH), Ecosystem Quality (EQ), or Resources (R)
- \(\delta_k\): weighting factor of the \(k^{th}\) damage category [-],
- \(D_k\): normalised impact indicator in the \(k^{th}\) damage category [-].

Normalised impact indicators \((D_k)\) are constant in EI-99. However, the indicators obtained in the damage categories (also referred as damage category results or shortly category results) depend on the selected weighting set, see Equation (2.1).

According to this, the ranking of compared systems may depend on the selected weighting set which expresses the individual choice of the LCA expert carrying out the study.

The effects of the selection of the weighting set can be studied with the help of weighting triangles. The weighting triangle can be used for the comparison of two processes based on the results of the LCA without knowing the weighting factors. The weighting triangle is a mixing triangle with the three damage categories in each corner (Figure 2.8). Each point within the triangle represents a combination of weights that add up to a 100%.
Figure 2.8 Weighting triangle: The marked weighting point is positioned where Human Health is weighted 50\%, Ecosystem Quality 40\% and Resources 10\% (Goedkoop and Spriensma, 2000).

A key feature is the possibility to draw lines of indifference (Figure 2.9). These are lines which represent weighting factors for which products $A$ and $B$ have the same environmental loads. The lines of indifference divide the triangle into areas of weighting sets for which product $A$ is favourable to product $B$ and vice versa.

![Weighting triangle](image)

Figure 2.9 The line of indifference in the weighting triangle and the sub-areas with their specific ranking orders. (B>A means that alternative B is environmentally superior to A and the eco-index A is higher than B), (Goedkoop and Spriensma, 2000)
2.5.1.3 Uncertainties

The EI-99 method applies several models and model equations for the assessment of the environmental impacts. These types of calculations always include different uncertainties. Three types of uncertainties can be distinguished in the EI-99 methodology:

1. Operational or data uncertainty.
2. Fundamental or model uncertainties.
3. Uncertainty about the completeness of the methodology.

Operational uncertainties

Operational or data uncertainties refer to the technical problems of measuring and assessing factors in the calculation step of the methodology, or in other words, technical uncertainties in the data. This type of uncertainty can be assumed as the variation in the results of the calculations caused by the variation of the parameters involved.

In the EI-99 it is intended to give quantitative uncertainty estimates for operational uncertainty whenever they are relevant. Uncertainty data is presented quantitatively as squared geometric standard deviation ($\sigma_g^2$) assuming a log-normal distribution for each EI-99 indicator. The squared geometric standard deviation expresses the variation between the best estimate (‘best guess’) of the indicators and the upper and lower confidence limits (97.5% and 0.5%).

Lower and upper limits of the confidence interval (CI) at the 95% level can be calculated as shown in Equation (2.3) and (2.4). A brief description of the log-normal distribution is presented in Appendix A.

\[
\text{Lower limit of the 95\% CI} = i \cdot \sigma_g^2 \\
\text{Upper limit of the 95\% CI} = i \cdot \sigma_g^2
\]  

where

\(i\): impact indicator of a given pollutant (best guess).

\(\sigma_g^2\): squared geometric standard deviation.

The uncertainties are intended for use in software tools that apply Monte Carlo type analysis.

Fundamental or model uncertainties

During the development of the EI-99 methodology many modelling choices have to be made on issues like:

- What should be included and excluded in the model?
- What level of scientific proof is required to accept a theory or hypothesis?
- What time frame should be taken into account?
- Are health problems among young people as serious as health problems among older people?
- Are future damages just as serious as damages that occur today or, in other words, should there be discounting?
- Are potential damages which could be avoided if proper management were applied less serious, or should manageable problems be disregarded?

The basis for making such choices is often rather subjective. The fundamental or model uncertainty is a reflection of the doubt about the correctness of choices made in the development of the method. The choice of a concept implies that the assumptions that are the
basis of this concept are fixed. Model uncertainties refer to the uncertainty if the model is configured correctly.

These types of uncertainties can not be expressed as a range; the assumptions of a model are either correct or they are not. In order to cope with this type of uncertainty, a system, referred to as Cultural Theory is used to separate three versions of the damage models. A simplified characterisation of the three versions is the following:

- **E (Egalitarian):** long time perspective; even minimal scientific proof justifies inclusion.
- **I (Individualist):** short time perspective; only proven effects are included.
- **H (Hierarchist):** balanced time perspective, consensus among scientists determines inclusion of effects (Goedkoop and Spriensma, 2000; Koning et al., 2002).

**Model completeness**
In the Eco-indicator 99 methodology a third type of uncertainty must be added. This is the uncertainty concerning whether the model includes all important damages that fall under our definition of the term ‘Eco’. There are several impact categories which are important and probably relevant, but for which no adequate damage model or sufficient data has been found. These categories have been excluded from the methodology. Moreover, within some impact categories more damage types can be found than has been successfully described and included in the methodology. For instance in climate change only a limited set of all the health problems that can probably be related to this impact category could be modelled, and thus included in the methodology.

Uncertainty on completeness cannot be documented at all, except for providing a specification of possibly important, but not included damages.
CHAPTER 3  INVESTIGATION OF LCIA METHODS
UNCERTAINTIES OF THE IMPACT INDICATORS AND DEPENDENCIES BETWEEN THE METHODS

The environmental performance of systems involved in a process engineering task can be determined by Life Cycle Assessment. Life Cycle Impact Assessment, being part of LCA, supports this kind of evaluation; however, the selection between the different available methods is often a difficult task and subjective choices provide the basis for that.

There are numerous LCIA methods available on the market. These differ in the impact pathway approaches and impact indicators they use. Based on the target of the impact pathway, the methods can be classified as midpoints-based approaches and endpoints-based (single score) approaches. All LCA experts agree in that midpoints based methods have lower uncertainty than endpoints-based methods; however, it is also obvious that single score impact indicators, analogically to money, are more suitable for environmentally-conscious process engineering. Moreover, different LCIA methods utilise different mathematical models; it often therefore occurs that different methods give different results from the evaluation of the same case study. Examples for this can be found in Hayo and Petit (2002), Dreyer et al. (2003), and Boeva and Gallardo (2006).

With the help of comparative studies - meaning the comparison and analysis of the results obtained by the evaluation of the same case study by different LCIA methods - the differences or even similarities between the methods can be found. If similarities and dependencies are detected, this may help and support the work of mutually exploiting the merits of both methods. On the other hand, uncertainty performances of the methods have to be analysed as well before they are selected for a certain environmental process engineering task. Actual air quality problems provide the basis for comparative studies.

3.1  The air quality improvement problem in the EU

The environment is exposed to several pollutants and among them air pollution is also an important factor to be considered. Recently, Krupa (2003) has provided an overview about air pollution source categories and the effects of these pollutants are being studied by many other scientists. There are several incentives to reduce air pollution since it seriously damages human health and the environment: premature deaths, respiratory diseases, eutrophication, and damage to ecosystems are some of the consequences of this problem. The problem is local, national, international, and transboundary in nature.

The need for cleaner air has been recognised for several decades with action having been taken at national and European Union (EU) levels and also through active participation in international conventions. EU action has focused on establishing minimum quality standards for ambient air and tackling the problems of acid rain and ground level ozone. Polluting emissions from large combustion plants and mobile sources have been reduced; fuel quality has been improved and environmental protection requirements have been integrated into the transport and energy sectors.

To improve air quality in Europe, the Commission of European Communities launched the Clean Air for Europe Programme (CAFE) in May 2001– a knowledge based approach with technical/scientific analyses and policy development that will lead to the adoption of a thematic strategy on air pollution, fulfilling the requirements of the Sixth Environmental Action Programme (6th EAP).
The 6th EAP, ‘Environment 2010: Our future, our choice’, includes ‘environment and health’ as one of the main target areas where new effort is needed. Air pollution is one of the issues included under environment and health. Whilst overall air quality trends in the Community are encouraging, continued efforts and vigilance are still needed. The Community is acting at many levels to reduce exposure to air pollution: through European Community legislation, through work at the wider international level in order to reduce cross-border pollution, through working with sectors responsible for air pollution such as national and regional authorities, and through research. The focus for the next ten years will be implementation of air quality standards and coherency of all air legislation and related policy initiatives (Clean air and transport, CAFE Programme).

The aim of the CAFE Programme – in agreement with the objective of the 6th EAP – is to develop a long term, strategic and integrated policy advice for ‘achieving levels of air quality that do not give rise to significant negative impacts on and risks to human health and the environment’; including ‘no exceedance of critical loads and levels for acidification or eutrophication’ (Watkiss et al., 2005).

During air quality improvement incentives, however, it is desirable to rank pollutants according to their detrimental impact either on human health or environment prior to creation of the relevant policies. Such a ranking of pollutants should highlight the most dangerous ones and it is also desirable to know their origins; that is, the activity that results in the emission of such pollutants. The basis of such a ranking is a scientific assessment of the environmental impacts caused by the pollutants.

### 3.2 Comparison and analysis of two LCIA methods

Two significant and important incentives for the assessment and evaluation of environmental impacts of polluting materials are compared and analysed. The investigation is connected to a real environmentally-oriented project; namely, the evaluation of the pollution registers of Polish cities. The case study aims at the identification of the pollution sources with the highest environmental impact.

The two investigated methods are: the results of the cost-benefit analysis within the framework of the EU’s CAFE Programme (CAFE CBA) and the Eco-indicator 99 Life Cycle Impact Assessment Method. Since both the EI-99 points and CAFE CBA results consider the impacts of the pollutants on human health and ecosystem quality (basically exposure of crops to ozone), it is logical to believe they can be compared.

Impact indicators of the two methods are compared in order to show whether similarities can be found between the two methods. If similarities and dependencies are detected, this information may help and support the work of mutually exploiting the merits of both methods (that is, the EI-99 and the marginal damage estimation of pollutants in the CAFE Programme).

The investigation includes the following steps:

1. An arithmetic comparison of the impact indicators used by the two methods (without any case study) in order to compare the relative pollutant-ranking properties of the two impact assessment methods.
2. Application of the two methods for the evaluation of the same task - the assessment of environmental impacts caused by air pollution in several industrialized cities. The investigation is extended and not only single score indicators but even uncertainties of results are included in the investigation.
3. In the final step, the original environmental project is completed. The emission inventory of a Polish city is analysed in order to identify the most significant air...
pollutants and their sources. This step of the investigation is carried out through the use of the EI-99 method.

3.2.1 Emission inventories

The basis for an environmental investigation is provided by the annual airborne emission inventories of several industrialized cities. The emission data is obtained from literature sources defined below.

The project of the Emission and Health Unit, Institute for Environmental and Sustainability, JRC, Ispra, entitled ‘From toxic emissions to health effects – an integrated emissions, air quality and health impacts case study Krakow’, delivers real existing data about air quality in Polish cities (Jimenez, 2006).

Jimenez (2006) collected air pollution data and took measurements to determine pollution levels and their health effects. The pollution data are summarised in an emission inventory.

The methodology used to set up the emission inventory is the recommendation of the UN ECE Task Force on Emission Inventory and Projections. Different data bases are used for the determination of the emission inventory such as, for example, the EMEP/CORINAIR Emission Inventory Guidebook, State Centre of Emission Inventory in Warsaw, Main Statistical Office of Poland (NILU Polska and IEIA 2005).

Table 3.1 shows the pollutants included in the emission inventory and the pollutant sources according to Selected Nomenclature for Air Pollution (SNAP) codes. The emission inventory was set up for the year 2002 on the level of administrative units. The emission inventory completed for the selected cities consists of data gathered for the source split based on SNAP codes.

<table>
<thead>
<tr>
<th>Pollutants</th>
<th>Pollutant sources</th>
</tr>
</thead>
<tbody>
<tr>
<td>P1. SO₂</td>
<td>S1. District heating</td>
</tr>
<tr>
<td>P2. NOₓ</td>
<td>S2. Combustion in the housing and commercial sector, individual heating</td>
</tr>
<tr>
<td>P3. CO</td>
<td>S3. Combustion in agriculture</td>
</tr>
<tr>
<td>P4. NMVOC</td>
<td>S4. Road transport</td>
</tr>
<tr>
<td></td>
<td>S5. Other transport</td>
</tr>
<tr>
<td></td>
<td>S6. Household use of paints and solvents</td>
</tr>
<tr>
<td></td>
<td>S7. Land filling</td>
</tr>
<tr>
<td>P5. CH₄</td>
<td>S8. Agricultural waste burning</td>
</tr>
<tr>
<td>P6. NH₃</td>
<td>S9. Emission from crops</td>
</tr>
<tr>
<td>P7. Dioxins and furans</td>
<td>S10. Farming</td>
</tr>
<tr>
<td>P8. PAH (including benzo(a)pirene)</td>
<td></td>
</tr>
<tr>
<td>P9. PM (d&gt;10 µm, 10&gt;d&gt;2.5 µm, d&lt;2.5 µm); indicated as TSP, PM₁₀, and PM₂.₅.</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.1 Pollutants included in the emission inventory and their sources according to their SNAP codes

Emission factors for all processes listed according to the SNAP codes are developed on the basis of Polish data taken from a country level emission inventory. For the calculation of the individual emissions, the regional data of different activities are also determined and used.
Table 3.2 shows the emission inventories for the selected Polish cities. The pollutants are listed according to their sources (NILU Polska and IEIA 2005). Pollutant source identifiers can be found in Table 3.1. Pollutants highlighted with grey background are involved in the comparison of CAFE CBA and EI-99 methods. It should be noted that approximately 10% of the total number of homes are heated with individual heating alternatives using coal. This is classified under combustion in housing activity.

![Image of Table 3.2 showing emission inventories for several cities in the neighbourhood of Krakow. Units are in t/year.](image-url)

Table 3.2 Emission inventories of several cities in the neighbourhood of Krakow. [t/year]
3.2.2 LCIA methods applied and investigated

3.2.2.1 Eco-indicator 99

For the investigation, hierarchist version of the EI-99 method with a custom weighting set is selected, that is, both the Human Health and the Ecosystem Quality damage categories are weighted by 50%, respectively. Since air pollution has no direct effect on natural resources this damage category is not included.

EI-99 indicator points (also referred to as ‘best guess’) per tonne airborne emissions applied in this study as well as the squared geometric standard deviation data obtained from the manuals of the EI-99 method (Goedkoop et al. 2000) are presented in Table 3.3. The EI-99 indicators refer to those pollutants which are included in Table 3.2.

<table>
<thead>
<tr>
<th></th>
<th>best guess</th>
<th>$\sigma_g^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO$_2$</td>
<td>1,878</td>
<td>78</td>
</tr>
<tr>
<td>NO$_x$</td>
<td>3,446</td>
<td>116</td>
</tr>
<tr>
<td>VOC</td>
<td>17</td>
<td>23</td>
</tr>
<tr>
<td>NH$_3$</td>
<td>4,285</td>
<td>98</td>
</tr>
<tr>
<td>PM$_{2.5}$</td>
<td>22,785</td>
<td>19</td>
</tr>
<tr>
<td>CO</td>
<td>11</td>
<td>n/a</td>
</tr>
<tr>
<td>NMVOC</td>
<td>42</td>
<td>n/a</td>
</tr>
<tr>
<td>CH$_4$</td>
<td>144</td>
<td>n/a</td>
</tr>
<tr>
<td>Dioxin</td>
<td>5.9E+09</td>
<td>n/a</td>
</tr>
<tr>
<td>PAH</td>
<td>5,600</td>
<td>n/a</td>
</tr>
<tr>
<td>TSP</td>
<td>3,580</td>
<td>n/a</td>
</tr>
<tr>
<td>PM$_{10}$</td>
<td>12,200</td>
<td>n/a</td>
</tr>
</tbody>
</table>

Table 3.3 Eco-indicator points of 1 tonne airborne emissions and squared standard deviation values.
(n/a= not applied in the study). Eco-indicator 99 hierarchist version, with 50-50% weighting for damage categories human Health and Ecosystem Quality.

3.2.2.2 CAFE marginal damage costs

The Service Contract for Cost-Benefit Analysis of Air Quality Related Issues, in particular in the Clean Air for Europe Programme, has been established in order to carry out the assessment of the costs and benefits of air pollution policies (estimation of the impacts of air pollutants and their control options), and to conduct analysis on scenarios generated within the CAFE programme. A short description of the technique of cost-benefit analysis is presented in Appendix B. CAFE CBA supports future political decision making in the European region. Extended CBA which supplies decision makers with more contextual information on impacts has been carried out, in order to aid in better understanding and, if desired, factoring in of decision-maker views on the importance of unquantified impacts into the CBA (Holland et al. 2005a).
Since CBA is an economic tool which supports decision making through monetary valuation, environmental damages are converted into costs – generally on the basis of Paretian’s welfare theory, in which individuals confronted with external effects judge their importance on their quality of life. The evaluation of damages is done in one single category: currency. Position of CBA indicators in the cause-effect chain is at the damage level.

Holland et al. (2005b) presented results of the CAFE CBA which assessed and evaluated the external costs and benefits of anthropogenic air pollution referring to five major air pollutants (SO\textsubscript{2}, NO\textsubscript{x}, VOC, NH\textsubscript{3} and PM\textsubscript{2.5}). They provided data on the marginal damages per tonne of pollutants accounting for the variation in the site of emission by providing estimates for each country in the EU 25 (excluding Cyprus) and for the surrounding seas. Results are based on modelling a uniform relative reduction in emissions of each pollutant within each country. Analysis contained in their report follows the impact pathway methodology developed in the ExternE project funded by the European Commission Directorate-General for Research (ExternE). The considered and quantified impacts include:

i. human exposure to PM\textsubscript{2.5},
ii. human exposure to ozone, and
iii. exposure of crops to ozone.

Holland et al. (2005b) presented total damages from each of the five pollutants with four sets of assumptions at the impact assessment, including different assumptions at the estimation of mortality, core health and crop functions, sensitivity of health functions and ozone impacts. The change in magnitude of marginal damages for the central scenarios is largely a reflection of unit values used for mortality valuation, rather than a response to other sensitivities explored. Therefore, the arithmetic mean of the four marginal damage values referring to one pollutant is also considered and used as best estimation of the marginal damages caused by the several pollutants. The lower and upper ends of the analyses are used as minimum and maximum values referring to each pollutant. Table 3.4 shows the marginal damage per tonne emission with the different sets of sensitivity analyses and the arithmetic mean of the four marginal damage values. CAFE CBA marginal damage values in Table 3.4 refer to Poland.

<table>
<thead>
<tr>
<th>CAFE marginal damage values</th>
<th>PM mortality</th>
<th>VOLY-median</th>
<th>VSL-median</th>
<th>VOLY-mean</th>
<th>VSL-mean</th>
<th>Arithmetic mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO\textsubscript{2}</td>
<td>5,600</td>
<td>8,600</td>
<td>11,000</td>
<td>16,000</td>
<td>10,300</td>
<td></td>
</tr>
<tr>
<td>NO\textsubscript{x}</td>
<td>3,900</td>
<td>5,800</td>
<td>7,100</td>
<td>10,300</td>
<td>6,775</td>
<td></td>
</tr>
<tr>
<td>VOCs</td>
<td>630</td>
<td>960</td>
<td>1,500</td>
<td>2,000</td>
<td>1,273</td>
<td></td>
</tr>
<tr>
<td>NH\textsubscript{3}</td>
<td>10,000</td>
<td>15,000</td>
<td>20,000</td>
<td>29,000</td>
<td>18,500</td>
<td></td>
</tr>
<tr>
<td>PM\textsubscript{2.5}</td>
<td>29,000</td>
<td>44,000</td>
<td>57,000</td>
<td>83,000</td>
<td>53,250</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.4 CAFE marginal damage per tonne emission (Euro/tonne) with different sets of sensitivity analyses, and the overall mean value. Data refer to Poland.

SOMO 0/35: sum of ozone concentration means over 0 or 35 ppb; VOLY: value of life year; VSL: value of statistical life

3.3 Discussion and Results
3.3.1 Arithmetic comparison

First, arithmetic comparison of the impact indicators is presented: CAFE marginal damage values (X-axis) are plotted against EI-99 impact indicators (Y-axis) for the five air pollutants. In the case of EI-99 impact indicators, the ‘best guess’ values (Table 3.3) are applied. In the case of CAFE CBA, the

1. minimum (diamonds),
2. maximum (triangles), and
3. arithmetic mean (squares)

marginal damage values are considered for the five pollutants (Table 3.4). This type of comparison helps to detect similarities and differences in the relative ranking properties of the two methods referring to the five pollutants. The results are shown in Figure 3.1.

![Figure 3.1](image-url)  
**Figure 3.1** EI-99 points against CAFE CBA results (minimum, maximum and overall mean values). Impacts per tonne emission of air pollutants.

It can be seen that the data points are arranged along straight lines. It is more obvious if trend lines are fitted to the data points. Based on graphical analysis it can be concluded that there is a linear correlation between the data points which indicates coherence and correlation between the impact indicators and thus between the valuation and ranking properties of EI-99 and CAFE CBA methods. Moreover, this coherence does not depend on the version of the CAFE CBA method.

It has to be noted that, since the data points are not distributed uniformly along the fitted lines (impact indicators of PM$_{2.5}$ are quite far from the other four pollutants), and there are no more data points available, linear correlation can not be proven by numerical statistical manners like the coefficient of determination ($R^2$). However, nor can the assumption of a linear correlation be refused.

Since the EI-99 has a consistent model framework and CAFE CBA is an accepted tool for environmental evaluation, the detected similarity in the relative ranking properties of the two methods assume that EI-99 gives trustable estimations for the environmental impacts of other air pollutants. However, the coherence between the two methods can be demonstrated better if they are applied to evaluate the same environmental case studies. This is demonstrated in the next section which is also extended with an investigation into data uncertainties.
3.3.2 Investigation of uncertainties

In the second step of the comparison of the two methods, a comparative study is carried out. The air pollution inventories of five industrialized cities are evaluated with both EI-99 and CAFE CBA methods. However, in this case the uncertainties of the impact indicators are considered too.

This comparative study is aimed at showing and comparing magnitude and the influence of uncertainty on the results. On the other hand, the comparison of the aggregated impact indicators obtained during evaluation of the real case studies can help to detect similarities and differences between the two methods.

Emission inventories included in the investigation are shown in Table 3.2. The pollutants are the same as in the previous section (SO\textsubscript{2}, NO\textsubscript{x}, VOC, NH\textsubscript{3}, and PM\textsubscript{2.5}).

Uncertainty regarding impact indicators is studied with Monte Carlo simulation. The population of impact indicators is generated based on available uncertainty data. Each simulation includes 1,000 steps of valuation and the confidence level of the evaluation is 90%.

**EI-99**

In the case of EI-99 impact indicators, the interval of the impact indicators referring to one pollutant is generated using the ‘best guess’ value and standard deviation data, shown in Table 3.3. Samples of a lognormal distribution are generated by the LOGINV function of Excel. LOGINV function returns the inverse of the lognormal cumulative distribution function of X with the syntax:

\[
\text{LOGINV} (\text{probability}, \mu_L, \sigma_L)
\]

where \(\ln(X)\) is normally distributed with parameters mean of \(\ln(X) \mu_L\), and standard deviation of \(\ln(X): \sigma_L\). ‘Probability’ is associated with the lognormal distribution (a brief description of the log-normal distribution is presented in Appendix A).

The parameters \(\mu_L\) and \(\sigma_L\) correspond to the parameters ‘best guess’ value and standard deviation of the eco-indicators, as shown in Table 3.3. One sample of the population is calculated by Equation (3.1); this calculation is carried out for all pollutants 1000 times.
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\[ i_{EI-99,j}^{MC} = \text{LOGINV} \left( \mu_{L,j}, \sigma_{L,j}, \text{rand} \right) \]  \hspace{1cm} (3.1)

\[ \mu_{L,j} = \ln(i_j) \]  \hspace{1cm} (3.2)

\[ \sigma_{L,j} = \ln(\sigma_{\mu,j}) \]  \hspace{1cm} (3.3)

\( i_{EI-99,j} \): impact indicator of the \( j \)th pollutant generated corresponding to its distribution function

\( \mu_L \): parameter of the lognormal distribution function, mean

\( \sigma_L \): parameter of the lognormal distribution function, standard deviation

\( \text{rand} \): random number

\( i_j \): impact indicator (‘best guess’) of the \( j \)th pollutant [EI-99 point / tonne]

\( \sigma_{\mu,j} \): standard deviation of the lognormal distribution referring to the \( j \)th pollutant

The total environmental impact (\( \bar{I}_{EI-99}^c \)) of one city (\( c \)) is calculated as shown by Equation (3.4).

\[ I_{EI-99}^c = \sum_j \left( i_{EI-99,j}^{MC} \cdot \bar{m}_j \right) \]  \hspace{1cm} (3.4)

**Figure 3.2** shows the results of the Monte Carlo simulation obtained for the evaluation of the emission inventory of Bielsko Biala. The diagram shows the frequency of the possible EI-99 indicator values. It can be determined from the diagram that the total environmental impacts in Bielsko Biala can be characterised with the highest probability with approx. \( 18 \cdot 10^6 \) EI-99 points/year. However, since the Monte Carlo simulation includes random numbers, the values indicated on the diagram slightly change for each new simulation.

**Figure 3.2** Probability of impact indicators obtained by the evaluation of the emission inventory of Bielsko Biala with Eco-indicator 99.
CAFE CBA
In the case of CAFE CBA marginal damage values, samples of a uniform distribution are generated between an upper limit (UL) and a lower limit (LL) for each air pollutant with the help of a random number generator. Since no quantitative uncertainty data is published for CAFE CBA results, the minimum and maximum marginal damage values are considered as lower and upper limits of the interval, assessing the environmental impacts. One sample of the population is calculated by Equation (3.5), this calculation is carried out for all pollutants 1000 times.

\[ i_{CAFE,j}^{MC} = rand \times (UL_j - LL_j) + LL_j \] (3.5)

\( i_{CAFE,j}^{MC} \): impact indicator of the \( j^{\text{th}} \) pollutant generated corresponding to its distribution function

The total environmental impact (\( I_{CAFE}^{C} \)) of one city (\( c \)) can be calculated as shown by Equation (3.6).

\[ I_{CAFE}^{C} = \sum_{j} (i_{CAFE,j}^{MC} \cdot \vec{m}_j) \] (3.6)

**Figure 3.3** shows the frequency of the possible CAFE CBA marginal damage values expressing the environmental impacts due to the air pollution in Bielsko Biala. The average of the generated population is at approx. 33 × 10^6 Euro/year.

**Figure 3.3** Probability of impact indicators obtained by the evaluation of the emission inventory of Bielsko Biala with CAFE CBA marginal damage values.
Comparison of the results

Through the use of Equations (3.4) and (3.6), total environmental impacts of the studied cities have been calculated and the impact indicators with the highest probability and the limits of the confidence interval (5% and 95% percentile) have been determined. These values are shown in Figure 3.4. The horizontal and vertical intervals cross each other at their means (in the case of lognormal distribution, the mean value is not in the middle of the interval).

CAFE CBA results give discrete impact assessment intervals for the annual air pollution in Nowy Sacz, Katowice, and Krakow which could support assessments on overall air pollution in the different cities. In the case of Bielsko Biała and Kielce, impact indicator intervals partially overlap.

Intervals of environmental impacts assessed by EI-99 are widespread and substantially overlapping which makes the distinction of clear preferences between the alternatives (cities) impossible.

![Figure 3.4](image)

**Figure 3.4** Impact indicator intervals obtained by Monte Carlo simulation. Results are shown for five Polish cities.

According to former studies (Lenzen 2006, Basson and Petri 2007), propagation of uncertainty in damage-oriented impact indicators can lead to the situation that no ordinal ranking between different alternatives can be established.

In this study, inclusion of uncertainty in the evaluation of the emission registers does not facilitate decision making, since impact indicator intervals mostly overlap. Being aware of the problem caused by the valuation uncertainty in EI-99 and CAFÉ CBA results, the delivery of interpretable results to stakeholders should be attempted.

If the overall mean values of the impact indicator populations (for both the EI-99 method and CAFÉ CBA) generated by the Monte Carlo simulation are also compared, strong correlation between the results can be found. The overall mean values are plotted against each other as shown in Figure 3.5.
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The strong correlation between these data is obvious. If a linear trend line is fitted to the points the coefficient of determination ($R^2$) is higher than 0.99, which shows the reliability of the fit and the strong correlation between the two methods. Data shown in Figure 3.5 changes with new Monte Carlo simulations; however, the value of $R^2$ is always found to be greater than 0.95. This means that the applied version of EI-99 and CAFE CBA do not show significant difference in the praxis - the two methods give practically the same results for identical cases.

In addition, the emission inventories are also evaluated with the single score indicators (in the case of EI-99 indicators the ‘best guess’ values are considered; in the case of the CAFE CBA the arithmetic mean marginal damage values are considered). The results of the calculation using single scores are compared with one set of results obtained by Monte Carlo simulations for the air pollution inventory of the cities in the study. The values are shown in Table 3.5.

![Figure 3.5](image)

Figure 3.5 Overall mean values of population of impact indicators obtained by Monte Carlo simulation. Results are shown for five Polish cities.

<table>
<thead>
<tr>
<th></th>
<th>Bielsko Biala</th>
<th>Krakow</th>
<th>Nowy Sacz</th>
<th>Katowice</th>
<th>Kielce</th>
<th>Opole</th>
</tr>
</thead>
<tbody>
<tr>
<td>EI-99</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>points/yr</td>
<td>10,768,249</td>
<td>48,814,532</td>
<td>6,259,465</td>
<td>32,682,985</td>
<td>11,129,995</td>
<td>10,109,567</td>
</tr>
<tr>
<td>evaluation with single scores</td>
<td>10,575,149</td>
<td>51,188,312</td>
<td>6,385,141</td>
<td>32,824,310</td>
<td>11,570,233</td>
<td>11,087,928</td>
</tr>
<tr>
<td>Relative error (%)</td>
<td>-2</td>
<td>5</td>
<td>2</td>
<td>0.4</td>
<td>4</td>
<td>9</td>
</tr>
<tr>
<td>CAFE CBA</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Euro/year</td>
<td>29,200,741</td>
<td>141,564,508</td>
<td>20,752,548</td>
<td>96,324,224</td>
<td>33,055,354</td>
<td>30,301,785</td>
</tr>
<tr>
<td>evaluation with single scores</td>
<td>31,967,664</td>
<td>155,281,965</td>
<td>22,456,756</td>
<td>102,748,008</td>
<td>36,185,245</td>
<td>32,823,478</td>
</tr>
<tr>
<td>Relative error (%)</td>
<td>9</td>
<td>9</td>
<td>8</td>
<td>6</td>
<td>9</td>
<td>8</td>
</tr>
</tbody>
</table>

*:mean values of the generated population of indicators.

Table 3.5 Comparison of the results obtained during evaluation of air pollution

a) with single score indicators, and
b) with Monte Carlo simulation. (Mean values of the generated populations of the indicators are presented).
It is obvious that the two methods of calculation give very similar results. This means that the indicators with the highest probability obtained by the Monte Carlo simulation are approximately the same as those obtained with the single score values. Due to this, it can be concluded that the application of single score indicators is sufficient for the environmental evaluation of a case study; the inclusion of the uncertainty data is neither helpful nor necessary.

According to the results, it can be concluded that:

- The EI-99 method evaluates environmental damages similarly to the impact assessment method supported by the EU (that is, the CAFE CBA method).
- Since the EI-99 method has a consequential structured framework, it is a logical assumption that all EI-99 impact indicators are in agreement with the EU’s environmental policy. In consequence, the application of EI-99 indicators in environmental evaluation is a well-grounded choice. Since the EI-99 includes not only the impact indicators of five pollutants (SO$_2$, NO$_x$, VOC, NH$_3$, and PM$_{2.5}$) but of more than one hundred pollutants, this significantly increases the quality of any environmental evaluation.
- Integrating the uncertainty of the indicators into the results does not improve the quality of the environmental evaluation.

3.3.3 Analysis of pollution and ranking of pollution sources with Eco-indicator points; a real case study

Due to factors bought up in the previous sections, the impact indicators of the EI-99 method are selected for use in an environmental evaluation. The investigated problem is an environmental evaluation of the air pollution of an industrialized city. However, not only the five air pollutants available in the CAFE CBA method are involved in the analysis, but all eleven pollutants included in the NILU database. For the analysis, Krakow city is selected; the pollutants for analysis and their annual emission data are shown in Table 3.2.

The results of such a study can serve several purposes:

- the identification of the polluting sectors, and
- identification of air pollutants with the highest environmental load.

For the evaluation of the results, it should be emphasised again that these data relate to a city environment and in this city individual heating is also used which means that residents operate their own coal-based domestic heating systems.

Total environmental impact caused by the emitted air pollutants ($I_{Krakow}^{EI-99}$) is calculated by Equation (3.7).

$$I_{Krakow}^{EI-99} = \sum_j (m_j \cdot i_j)$$  \hspace{1cm} (3.7)

where

- $j$ : the $j^{th}$ air pollutant
- $m_j$ : emission of the $j^{th}$ pollutant [tonne/year]
- $i_j$ : impact indicator of the $j^{th}$ pollutant [EI-99 point/tonne]
Total environmental impacts are calculated and presented for each pollutant (Figure 3.6), and each pollution source, see on Figure 3.7.

EI-99 impact indicators make possible the comprehensive environmental evaluation of the emission inventory and comparison and ranking of different pollutants.

It is clearly shown in Figure 3.6 that environmental impacts are mostly caused in the damage category of Human Health. According to EI-99,

- damage to Ecosystem Quality accounts for less then 10% of the total damage,
- the most dangerous pollutants to Human Health are fine particulates, NO\(_x\), and SO\(_2\),
- the most polluting sectors and their emissions are also identified: environmental impacts caused by particulate matter is mainly caused by the private heating sector (56%), and road transport (33%); impacts related to NO\(_x\) originate mainly from road transport (81%); environmental impacts of SO\(_2\) are caused mainly by the private (44%) and district (37%) heating sectors.

![Figure 3.6](image)

**Figure 3.6** Environmental impacts of Krakow city relating to air pollutants. Results are obtained throughout the use of EI-99 (evaluation of 11 pollutants).

**Figure 3.7** shows the environmental impacts caused by the different pollution sources. A ranking of the polluting activities can be done: the most polluting activities are traffic (road transport, S4), and individual heating (S2). Special attention should be paid to the fact that district heating (S1) causes significantly lower environmental impacts than the effects of combustion from the housing and commercial sector.

This evaluation, on the one hand, helps to locate the most polluting activities and, on the other hand, helps policy makers to make better decisions about the installation of air quality improvement facilities.
Figure 3.7 Environmental impacts of Krakow city relating to pollution sources. Results are obtained by EI-99.

3.4 Conclusions

It can be concluded that the comparison of the two environmental impact assessment methods (EI-99 and CAFE CBA) highlights a clear dependency between them. If the uncertainties of the methods are also considered, it is proven (through the examples of real case studies) that their dependency is founded and the overall mean values and the best guess data are in correlation. According to these results, the best guess data of EI-99 can be used for the evaluation of environmental problems with high probability. This helps decision makers to have a clearer overview about environmental performance compared to those cases when the results are presented in the analysis with the full spectrum of single score uncertainties.

On the other hand, the best guess of the EI-99 indicators can be used to rank emissions according to their relative effects on human health and ecosystems, and to locate emission sources and polluting activities. These results can help decision makers to in efficient environmentally-conscious policy-making.
CHAPTER 4 INVESTIGATION OF AIR POLLUTION PREVENTION WITH LCA

An important application area of LCA is the evaluation and comparison of technologies and processes to generate ranking of elements from environmental point of view. Such evaluations are usually used alongside or instead of economic analyses to assist in selecting from different process alternatives.

Focusing on engineering problems associated with air quality improvements, SO\textsubscript{2} emission reduction is a technology of industrial importance where there is a wide variety of solution alternatives. However, a comprehensive evaluation of the different solution alternatives based on LCA has not been completed yet.

4.1 Introduction

Coal plays a significant role in the generation of electricity. In 2003, about 110,000 TWh primary energy was consumed worldwide and, on a global basis, coal-fired processes provided about 26% of the net electricity generated (BP Statistical Review, 2004). The global coal consumption rate is about 5,100 Mt coal per year, and this value is expected to grow over the course of this century due to its relative abundance (McFarland et al., 2004).

Due to the sulphur content of coal (that varies normally between 0.3 and 1.2 wt%) (EMEP/CORINAIR, 2006), sulphur-oxides (SO\textsubscript{x}) - mainly SO\textsubscript{2} - are formed through oxidation of sulphur during high temperature coal combustion. Atmospheric SO\textsubscript{2} is a long range transboundary air pollutant responsible for respiratory problems and acid rain.

The uncontrolled release of SO\textsubscript{2} from coal-fuelled power plants would raise the amount of anthropogenic SO\textsubscript{2} emissions by about 150% therefore several attempts have been made to regulate SO\textsubscript{2} emissions such as the Helsinki Protocol (‘Protocol on the Reduction Sulphur Emissions or their Transboundary Fluxes by at least 30 per cent’) and the Oslo Protocol (‘Protocol on Further Reduction of Sulphur Emissions’) submitted by the United Nations Economic Commission for Europe, and the Clean Air Act Amendments 1990 (CAAA) passed by the U.S. Congress (Metz, 1998).

There are several techniques for the abatement of SO\textsubscript{2} emissions. Selection from the abatement alternatives is usually made on the basis of economic considerations. Environmental performance of the technologies is usually characterized by measuring the concentration of the SO\textsubscript{2} remaining in the exhaust gas. However, control technologies have also a significant environmental load. LCA provides a framework for identifying and evaluating environmental burdens associated with the life cycles of materials and services in a ‘cradle-to-grave’ approach and providing the possibility of an environmentally-focused comparison end evaluation.

Chevalier et al. (2003) studied flue gas cleaning processes (a typical wet process and the new transported droplets column) of municipal solid waste incinerators with an LCA approach. They found that the global environmental burden is similar regarding use of the two processes which conforms the viability of the transported droplets columns process. Benetto et al. (2004) investigated the environmental issues of electricity production scenarios which promote the design of new production scenarios. Meyer (2002) studied the rate of greenhouse gas emission originating from electricity generation, offering an accurate means for evaluating greenhouse gas emission reduction strategies for U.S. electricity generation. Environmental consciousness has to be integrated into process engineering too; however, it requires the numerical expression of environmental impacts. Several attempts have already
been made in this field (Chen and Shonnard, 2004; Burgess and Brennan, 2001; Alexander et al., 2000; Azapagic and Clift, 1999; Azapagic, 1999); however, a comprehensive solution for the integration of LCA results into the process engineering has not yet been presented.

4.1.1 SO$_2$ abatement techniques

Techniques for reducing emissions of SO$_2$ during the combustion of fossil fuels can be distinguished as
1. pre-combustion,
2. intra-furnace sulphur removal, and
3. end-of-pipe abatement technologies (flue gas desulphurization).

Pre-combustion sulphur removal includes a wide range of technologies, i.e.
- microbial desulphurization,
- halogenation,
- pyrolysis,
- electrochemical oxidation,
- irradiation (Thoms, 1995),
- liquid phase methanol process and coal gasification (CCTP 2006).

Intra-furnace sulphur removal is possible with the addition of alkaline sorbent such as calcium oxide or calcium carbonate to the coal therewith removing SO$_2$ through dry-adsorption. The most commonly-used furnace type is the circulating fluidized bed boiler which provides a long residence time for sorbent particles. This process type has inherent environmental benefits over end-of-pipe flue gas desulphurization processes since there is no need for expensive FGD equipment in addition to the boiler; however, the retrofitting of an existing boiler is difficult and may require new apparatus.

End-of-pipe flue gas desulphurization (FGD) is an effective control of SO$_2$ emissions. In the last few decades, FGD processes have undergone considerable developments in terms of improved removal efficiency and reliability, as well as reduced costs. Wet scrubbers are the most commonly used FGD system, accounting for 87% of the total FGD capacity world wide and wet limestone is the predominant process, accounting for 82% of all installed wet FGD capacity worldwide.

A great advantage of wet limestone scrubbing is its relatively easy adaptability to existing plants and low operating costs because of the low price of limestone (Wu, 2001; Royal Academy of Engineering, 2004; Rajaram, 1999).

Formerly-discussed SO$_2$ removal techniques are called ‘once-through’ processes since a continuous delivery of fresh sorbent is required for the operation. Simultaneously, a huge amount of by-product is generated and has to be disposed of. But FGD processes with regenerable sorbent offer a solution to that problem.
A relatively new technology developed by the US Department of Energy Federal Energy Center is called Copper Oxide Technology (CuO), which is able to reduce SO$_x$ and NO$_x$ in a single unit. The CuO process is a dry regenerable process that has many advantages over wet scrubbers:

- it does not produce landfill waste, thus
- it avoids concerns over the limited landfill space and does not increase landfill costs related to SO$_2$ removal, and
- raises public awareness of the environmental impact.

CuO process also provides an effective way to obtain a concentrated SO$_2$ stream that can be used to produce sulphuric acid, elementary sulphur, fertilizer, etc. The valuable by-products partially compensate for the relatively higher operating costs when compared to other technologies (Daraguzas et al., 1999).

4.1.2 Comparison of SO$_2$ removal techniques with LCA

Three SO$_2$ removal techniques used in coal-fuelled power plants are compared:

1. intra-furnace sulphur removal with limestone addition in an atmospheric circulating fluidized bed combustor (ACFBC);
2. flue gas desulphurization with wet lime scrubbing, and
3. SO$_2$ removal with the regenerable CuO process.

The three options use different physical and chemical principles to flue gas clean-up. The comparison is made on the basis of environmental impacts derived from energy and mass balances of the investigated processes considering the cradle to grave approach.

Environmental impact is assessed by EI-99 impact indicators (best guess values). The egalitarian version of the method is selected with the default weighting set (egalitarian) which is 30%, 50% and 20% for the damage categories Human Health, Ecosystem Quality and Resources, respectively. Calculations are supported by the software SimaPro 6.0.

The environmental evaluation is carried out using annual input-output databases obtained form the literature.

System boundaries of the studied processes (see Figure 4.1) include:

- sorbent production (mining of raw materials, manufacturing and transportation of sorbents);
- electricity consumption;
- discharge of the purified flue gas;
- discharge of solid wastes and valuable by-products.

![Figure 4.1](image-url) System boundaries applied during comparison of SO$_2$ abatement techniques.
Life cycle inventories of sub-processes, like sorbent production and electricity generation are obtained from the built-in inventories (ETH-ESU 1996) of SimaPro 6.0. For our assumption, the auxiliary energy requirement of the studied processes is covered under ‘lignite fuelled power plants’; for further details see the SimaPro database.

Disposal of flue gas cleaning by-products can be done by landfilling in highly active chemical landfills or by its industrial utilization (material recycling). The three SO₂ removal techniques produce different valuable by-products.

ACFBC systems produce so called FBC ashes (bed- and flying ashes from ACFBC systems) which can be used in cement production.

FGD with wet-limestone scrubbing produces so called FGD gypsum that can be utilized in the building industry.

The by-product of the CuO process is pure SO₂ which can be utilized in the chemical industry.

Three possible disposal scenarios which represent the less and the most desired situations in the field of the utilization are considered and investigated; the third scenario aims to represent the situation today:
- 0% of the by-product is utilized by the industry;
- 100% of the by-product is utilized by the industry, and
- the current utilization rate as given according to industrial statistics.

Industrial statistics about the utilization rate of FGD by-products are obtained from the USGS Minerals Yearbook (Kalyoncu, 2000). An extract of the yearbook is shown in Table 4.1. In the environmental evaluation, utilized by-products reduce the total environmental impact, since they replace the production of new materials which consume the Earth’s resources. These valuable and utilizable by-products are called ‘avoided products’ which reduce the total environmental impacts in the calculations.
<table>
<thead>
<tr>
<th></th>
<th>Fly ash</th>
<th>Boiler slag</th>
<th>FBC ashes</th>
<th>FGD gypsum</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cement raw material</td>
<td>20.6</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Blended cement</td>
<td>10.6</td>
<td>-</td>
<td>2.2</td>
<td>-</td>
</tr>
<tr>
<td>Concrete addition</td>
<td>29.9</td>
<td>6.6</td>
<td>6.7</td>
<td>-</td>
</tr>
<tr>
<td>Aerated concrete blocks</td>
<td>3.7</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Nonaerated concrete</td>
<td>3.2</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Lightweight aggregate</td>
<td>1.3</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Bricks and ceramics</td>
<td>0.4</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Grouting</td>
<td>2.9</td>
<td>6.6</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Asphalt filler</td>
<td>1.0</td>
<td>-</td>
<td>11.1</td>
<td>-</td>
</tr>
<tr>
<td>Subgrade stabilization</td>
<td>1.8</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Pavement base course</td>
<td>1.2</td>
<td>51.7</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>General engineering fill</td>
<td>7.2</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Structural fill</td>
<td>7.6</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Infill</td>
<td>7.6</td>
<td>-</td>
<td>80.0</td>
<td>-</td>
</tr>
<tr>
<td>Blasting grit</td>
<td>-</td>
<td>30.2</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Plant nutrition</td>
<td>-</td>
<td>1.7</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Set retarder for cement</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>7.1</td>
</tr>
<tr>
<td>Projection plaster</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>9.4</td>
</tr>
<tr>
<td>Plaster boards</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>61.0</td>
</tr>
<tr>
<td>Gypsum blocks</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>3.6</td>
</tr>
<tr>
<td>Self levelling floor</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>18.9</td>
</tr>
<tr>
<td>Screeds</td>
<td>1.1</td>
<td>3.7</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 4.1 Utilization ratios [%] of several coal combustion by-products, based on the data of the European Coal Combustion Products Association (Kalyoncu, 2000).
4.1.3 SO\(_2\) removal in ACFBC

In the investigated atmospheric circulating fluidised bed combustion boiler, limestone is contacted with the flue gas in a circulating fluidized bed and SO\(_2\) is captured by the calcinated limestone in a sulphation reaction, as shown in Equations (4.1)-(4.3). The process is schematically shown in Figure 4.2.

![Schematic diagram of the atmospheric circulating fluidised bed combustion.](image)

**Figure 4.2** Schematic diagram of the atmospheric circulating fluidised bed combustion.

The fluidized bed is formed as a result of pure air and/or flue gas flowing upward through a bed of sorbent solids. ACFBC ensures a long contact time between the sorbent and flue gas because sorbent passes through the bed several times. The flue gas, laden with reaction products, then flows to a particulate control device, in this case an electrostatic precipitator (ESP). Bed ash produced in the furnace is removed and sent for disposal.

An additional benefit of the fluidised combustion is that low furnace temperature and air factor makes possible the reduction of thermal NO\(_x\) formation.

Based on Equations (4.1)-(4.3) the theoretical Ca/S ratio is 1. Because of steric problems, the inner parts of the CaO particles can not be utilized by the sulphation reaction, therefore the Ca/S ratio should be set at least between 1.4 and 2 (Srivastava, 2000). Due to the relatively low furnace temperature, the sulphation reaction is strongly shifted to the right side and the product (gypsum) is thermally stable. The gypsum is obtained from either the fly or bottom ash.

- calcination: \( CaCO_3 \rightarrow CaO + CO_2 \) (4.1)
- sulphation: \( CaO + SO_2 \rightarrow CaSO_3 \) (4.2)
  \[ CaSO_3 + \frac{1}{2}O_2 \rightarrow CaSO_4 \] (4.3)

Environmental evaluation of an ACFBF system is carried out on the basis of the annual input-output database of an existing European power plant. Operational and technical
parameters of the investigated ACFBC system are in reasonable agreement with the generic technological descriptions found in the Oslo Protocol.

Operational data are obtained from the literature (Rentz and Gütling, 2002a, 2002b). In the reference year (1999), 295 kilotons of lignite were burnt with an average sulphur content of 1.7%. The molar Ca/S ratio was 1.7/1. NO$_x$ was controlled with primary measures: low combustion temperatures and air staging reduced the formation of thermal NO$_x$ to the desired low level. For dust control an ESP with a separation efficiency of >99.9% was installed. In my consideration, bed ash and fly ash forms FBC ash.

### 4.1.4 SO$_2$ removal with wet-limestone scrubbing

A generalized flow diagram of a baseline wet FGD system is shown in Figure 4.3. Fly ash is removed from the flue gas by a particulate control device - in this case ESP. The SO$_2$-containing flue gas is then contacted with limestone slurry in an absorber. Limestone slurry is prepared in two consecutive steps. First, limestone is crushed into a fine powder with the desired particle size distribution. This takes place at a crushing station. Next, this fine powder is mixed with water in a slurry preparation tank. Sorbent slurry from this tank is then pumped into the reaction tank and the absorber where limestone slurry is sprayed downwards by an array of spray nozzles. In the absorber, SO$_2$ is removed by both sorption and reaction with the slurry.

![Figure 4.3 Schematic diagram of wet-limestone scrubbing.](image-url)
Reactions initiated in the absorber are completed in a reaction tank, which provides retention time for finely ground limestone particles to dissolve and to react with the dissolved \( \text{SO}_2 \). The main reaction in the absorber and in the reaction tank can be summarized in Equation (4.4).

\[
\text{SO}_2 + \frac{1}{2} \text{O}_2 + \text{CaCO}_3 + 2\text{H}_2\text{O} \rightarrow \text{CaSO}_4.2\text{H}_2\text{O} + \text{CO}_2
\]  

(4.4)

Normally, the required stochiometry of a limestone wet FGD system varies from 1.01 to 1.1 moles of \( \text{CaCO}_3 \) per mole of \( \text{SO}_2 \). Spent sorbent from the reaction tank (slurry bleed) is dewatered and disposed of.

In this study, a power plant which utilises a pulverized lignite fired dry bottom boiler is investigated. Operational data and technical parameters obtained from Rentz and Güting (2002a, 2002b) are in reasonable agreement with the generic technological descriptions found in the Oslo Protocol.

In the reference year (1999) 12,068 kilotons of lignite were fired with an average sulphur content of 0.73%. Sulphur oxides were removed with wet limestone scrubbing consuming limestone, water and auxiliary energy and producing FGD gypsum. Waste water produced by the FGD unit was utilized by fly ash sedimentation. \( \text{NO}_x \) control was performed with primary measures (fuel and air staging). Dedusting was done with ESP which consumes auxiliary energy and produces fly ash.

4.1.5 \( \text{SO}_2 \) removal with the Copper Oxide System

The investigated regenerable FGD technology with CuO system was developed by DB Riley Inc. (Ruth, 1997). The technology was under research supported by the Department of Energy.

CuO technology uses a regenerable sorbent, removing both \( \text{SO}_2 \) and \( \text{NO}_x \) from the flue gas and producing pure \( \text{SO}_2 \) instead of creating a solid waste. The process is shown schematically in Figure 4.4.

![Figure 4.4 Schematic diagram of flue gas desulphurization with regenerable copper oxide sorbent.](image-url)
The basis of the technology is that CuO can readily react with SO$_2$ in flue gas at temperatures of around 350 to 400°C to form CuSO$_4$, see Equation (4.5). CuSO$_4$ then can be reduced to Cu with methane or other reducing gases, releasing SO$_2$ in a concentrated form that can be used in various processes, see Equation (4.6).

The regenerated sorbent is exposed to the flue gas so that the elemental copper is converted to copper oxide that can be again used to react with SO$_2$, see Equation (4.7). The temperature in the reaction unit is 350°C while regeneration takes place at 500°C. The absorbent (CuO) is layered on an Al$_2$O$_3$ carrier with approx. 7% CuO content (Daraguzas et al., 1999).

The main reactions can be expressed as follows.

\[
\begin{align*}
CuO + SO_2 + \frac{1}{2}O_2 & \rightarrow CuSO_4 \\
CuSO_4 + \frac{1}{2}CH_4 & \rightarrow Cu + SO_2 + \frac{1}{2}CO_2 + H_2O \\
Cu + \frac{1}{2}O_2 & \rightarrow CuO
\end{align*}
\]

(4.5)\hspace{2cm} (4.6)\hspace{2cm} (4.7)

An interesting feature of the process is that both CuSO$_4$ and CuO can serve as catalyst for reducing the NO$_x$ content of the flue gas to N$_2$ using NH$_3$. By injecting NH$_3$ into the flue gas before it contacts CuO impregnated sorbent, both NO$_x$ and SO$_2$ in the flue gas can be removed. The NO$_x$ reduction reactions can be written as:

\[
\begin{align*}
4 NO + 4 NH_3 + O_2 = 4 N_2 + 6 H_2O \\
6 NO + 4 NH_3 = 5 N_2 + 6 H_2O \\
2 NO_2 + 4 NH_3 + O_2 = 3 N_2 + 6 H_2O \\
6NO_2 + 8 NH_3 = 7 N_2 + 12 H_2O
\end{align*}
\]

(4.8)\hspace{2cm} (4.9)\hspace{2cm} (4.10)\hspace{2cm} (4.11)

Operational data of this CuO process applied in this study is obtained from the literature (Darguzas et al., 1999; Spath et al., 1999; Ruth, 1997). The CuO technology studied here was applied as a secondary FGD and DeNOx measure at a Low Emission Boiler System (LEBS).

LEBS is projected to have significantly higher thermal efficiency, better performance, and require less electricity.

Emissions for this system are those forecasted from future plant utilizing a LEBS. Sulphur oxides and nitrogen oxides coming from the boiler are removed in the CuO absorber unit, while dust is removed in the fabric filter following the absorber unit. Theoretical consideration is given to CO$_2$ emission, see Equation (4.6).

The LEBS system uses a U-fired slagging boiler which converts the coal ash and fly ash into slag. As it is quenched, the slag transforms into a low volume, inert, vitreous granulate.

The system consumes electricity for the pneumatic transport of the sorbent, NH$_3$ and CH$_4$ as reducing agents and air.

By-products of the system are highly concentrated SO$_2$ and boiler slag generated in the bed of the boiler. There are no statistical data about the utilization rate of the recovered SO$_2$; however, it may be considered a valuable product. Fly ash is recycled to the boiler and leaves the system in form of boiler slag.
4.2 Discussion and results

4.2.1 Identification of damage and its sources

Firstly, the LCIs of the studied processes are prepared on the basis of the operational data obtained from the literature, see Table 4.2.

<table>
<thead>
<tr>
<th>Coal used:</th>
<th>ACFBC system</th>
<th>Wet Lime Scrubbing</th>
<th>CuO process</th>
</tr>
</thead>
<tbody>
<tr>
<td>Amount, kt/a</td>
<td>Lignite</td>
<td>lignite</td>
<td>Illinois coal</td>
</tr>
<tr>
<td>S%</td>
<td>291</td>
<td>12,068</td>
<td>1,215</td>
</tr>
<tr>
<td>Plant capacity, MW</td>
<td>43</td>
<td>1,500</td>
<td>407</td>
</tr>
<tr>
<td>DeNOx:</td>
<td>Primary</td>
<td>primary</td>
<td>SCR</td>
</tr>
<tr>
<td>DeSOx:</td>
<td>Limestone</td>
<td>wet scrubbing</td>
<td>CuO cycle</td>
</tr>
<tr>
<td>Dedusting:</td>
<td>ESP</td>
<td>ESP</td>
<td>fabric filter</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Input</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Limestone, kg/kgS</td>
<td>8.1</td>
<td>2.1</td>
<td>-</td>
</tr>
<tr>
<td>Electricity, kWh/kgS</td>
<td>3.8</td>
<td>6.0</td>
<td>1.6</td>
</tr>
<tr>
<td>Water demand, kg/kgS</td>
<td>-</td>
<td>39</td>
<td>-</td>
</tr>
<tr>
<td>CuO, g/kgS</td>
<td>-</td>
<td>-</td>
<td>19.5</td>
</tr>
<tr>
<td>Al₂O₃, g/kgS</td>
<td>-</td>
<td>-</td>
<td>279</td>
</tr>
<tr>
<td>Ammonia, g/kgS</td>
<td>-</td>
<td>-</td>
<td>9.9</td>
</tr>
<tr>
<td>Natural gas, m³/kgS</td>
<td>-</td>
<td>-</td>
<td>0.3</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Output</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Emission to air, g/kgS</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SO₂</td>
<td>116.1</td>
<td>21.2</td>
<td>10.9</td>
</tr>
<tr>
<td>NOₓ</td>
<td>49.8</td>
<td>44.7</td>
<td>10.9</td>
</tr>
<tr>
<td>PM</td>
<td>2.9</td>
<td>0.7</td>
<td>1.1</td>
</tr>
<tr>
<td>CO₂</td>
<td>-</td>
<td>-</td>
<td>684</td>
</tr>
<tr>
<td>Solid waste, kg/kgS</td>
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<td></td>
</tr>
<tr>
<td>FBC ash</td>
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<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Fly ash</td>
<td>-</td>
<td>4.7</td>
<td>0.9</td>
</tr>
<tr>
<td>FGD gypsum</td>
<td>-</td>
<td>3.7</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 4.2 Input-output database of the studied systems, referring to the treatment of flue gas containing 1 kilogram of sulphur.

The amounts of valuable by-products are calculated on the basis of the three disposal scenarios, explained above. Table 4.3 shows the utilization rates referring to the three considered disposal scenarios.
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Based on the life cycle inventories (Table 4.2) and the disposal scenarios of the by-products, the environmental impacts of the studied processes are assessed with EI-99 impact indicators. The results are shown in Table 4.4.

### Table 4.3 Utilization ratios of FGD by-products considered in the study.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Disposal</th>
<th>ACFBC FBC ash</th>
<th>FGD with Wet Lime Scrubbing Fly ash</th>
<th>FGD with Wet Lime Scrubbing FGD gypsum</th>
<th>FGD with CuO Boiler slag</th>
<th>SO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>0% utilization</td>
<td>utilization</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
</tr>
<tr>
<td></td>
<td>landfill</td>
<td>100%</td>
<td>100%</td>
<td>100%</td>
<td>100%</td>
<td>0%</td>
</tr>
<tr>
<td>100% utilization</td>
<td>utilization</td>
<td>100%</td>
<td>100%</td>
<td>100%</td>
<td>100%</td>
<td>0%</td>
</tr>
<tr>
<td></td>
<td>landfill</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
</tr>
<tr>
<td>statistical data</td>
<td>utilization</td>
<td>45%</td>
<td>48%</td>
<td>87%</td>
<td>100%</td>
<td>100%</td>
</tr>
<tr>
<td></td>
<td>landfill</td>
<td>55%</td>
<td>52%</td>
<td>13%</td>
<td>0%</td>
<td>0%</td>
</tr>
</tbody>
</table>

### Table 4.4 Environmental impacts of the studied systems referring to 1 kg sulphur contained in the flue gas [10⁻³ EI point/kgS].

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Disposal</th>
<th>ACFBC FBC ash</th>
<th>Wet-limestone scrubbing Fly ash</th>
<th>FGD with Wet Lime Scrubbing FGD gypsum</th>
<th>FGD with CuO Boiler slag</th>
<th>SO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Input</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Limestone</td>
<td>3</td>
<td>0.75</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Electricity</td>
<td>164</td>
<td>260</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Water demand</td>
<td>-</td>
<td>0.001</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CuO</td>
<td>-</td>
<td>-</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>-</td>
<td>-</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ammonia</td>
<td>-</td>
<td>-</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Natural gas</td>
<td>-</td>
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</tr>
<tr>
<td>Output</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Emission to air</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SO₂</td>
<td>135</td>
<td>25</td>
<td></td>
<td></td>
<td>13</td>
<td></td>
</tr>
<tr>
<td>NOₓ</td>
<td>114</td>
<td>102</td>
<td></td>
<td></td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>PM</td>
<td>6</td>
<td>1</td>
<td></td>
<td></td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>CO₂</td>
<td>-</td>
<td>-</td>
<td></td>
<td></td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>By-products</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>utilization</td>
<td>0% 100% stat. util.</td>
<td>0% 100% stat. util.</td>
<td>0% 100% stat. util.</td>
<td>0% 100% stat. util.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>FBC ash</td>
<td>0 -55 -25</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Fly ash</td>
<td>- - -</td>
<td>0 -49 -24</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>FGD gypsum</td>
<td>- - -</td>
<td>0 -35 -30</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Boiler slag</td>
<td>- - -</td>
<td>-</td>
<td>-</td>
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</tr>
<tr>
<td>SO₂</td>
<td>- - -</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0 -2 -2</td>
<td></td>
</tr>
<tr>
<td>Landfill</td>
<td>40 0 22</td>
<td>31 0 11</td>
<td>3</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>TOTAL</td>
<td>462</td>
<td>367</td>
<td>419</td>
<td>419</td>
<td>305</td>
<td>345</td>
</tr>
</tbody>
</table>

util.: utilization; stat. data: statistical data
With the help of the EI-99 scores, the environmental performance of the three systems can be assessed, and a ranking can be set up. It is worthy of note that EI-99 makes it possible to compare the environmental impacts of different pollutants and materials.

As it can be seen, environmental impacts caused by the systems vary between 0.189 and 0.462 EI-99 points per 1 kg sulphur for the three systems. According to EI-99, the simple release of SO\textsubscript{2} into air causes 2.32 EI-99 points/kgS damage to the environment. This shows the reasonability of the flue gas treatment: at least 80% reduction in environmental impact can be achieved by flue gas desulphurization.

The ranking of the three processes does not depend on the utilization scenarios: the best option is the CuO process, followed by wet-limestone scrubbing, and the worst option from an environmental viewpoint is the ACFBC system.

However, the analysis also shows that from input side, the best option is the ACFBC system. In this ranking, the CuO process comes second and third is wet-limestone scrubbing.

In the case of the airborne emissions, the ACFBC system shows the worst performance (0.26 EI-99 points/kg S) Wet-limestone scrubbing causes 50% less environmental impact according to airborne emissions (0.13 EI-99 points/kg S); however, the truly environmentally-friendly option is the CuO process with 0.05 EI-99 points/kg S). These results are in agreement with the SO\textsubscript{2} removal efficiencies of the techniques.

The study of by-product utilization shows that the by-products (also including landfilling) are responsible for about 10% of the total environmental impact (see the 0% by-product utilization scenario). However, it can not be concluded that it does not make sense to increase the by-product utilization ratios. In the case of the ACFBC system, the 100% by-product utilisation ratio decreases the total impact by 20%; in the case of the wet-limestone scrubbing by 27%, and in the case of the CuO process by 12%. Therefore, it is recommended and highly desirable to increase the utilization ratio of the by-products in order to improve the environmental performance of energy production with coal combustion.
4.2.2 Analysis of the damage categories, effect of weighting

The EI-99 methodology makes it possible to analyse and rank the three SO\textsubscript{2} removal options based on the results obtained for the three damage categories (Human Health, Ecosystem Quality, and Resources).

**Figure 4.5** shows the environmental impacts in the three damage categories if by-product utilization rate according to statistical data is considered.

![Environmental impacts caused by the studied systems in the damage categories; by-product utilization rate is based on statistical data.](image)

The results show that:

- the highest environmental impacts are caused in the damage category Human Health, expect for the CuO process;
- from the viewpoint of the Ecosystem Quality, the ACFBC and wet-limestone scrubbing options are identical; however, the CuO process has slightly better performance in this damage category;
- analysis of damage categories shows that the wet-limestone scrubbing and the CuO process cause higher damages in the category for Resources than the ACFBC system.

The environmental evaluation of the studied SO\textsubscript{2} removal techniques is carried out using the egalitarian weighting set (n.b. 30%, 50%, and 20% for the damage categories Human Health, Ecosystem Quality and Resources, respectively). The change in rank order due to different weighting sets can be compared using three triangles (**Figure 4.6**).
Figure 4.6 Comparison of the studied SO2 removal options with the weighting triangles.

On the first triangle it can be seen that approx. 2/3 of all possible weighting sets lead to the conclusion that the wet-limestone scrubbing option has lower environmental load than the ACFBC system - which supports the former conclusion: SO₂ removal with wet-limestone scrubbing is preferable from an environmental viewpoint.

However, the triangle also shows that ACFBC system would be considered better from environmental viewpoint, if the damage category Human Health was weighted lower (lower than 30%) and the Resources category weighed higher.

In the second triangle, the ACFBC system and CuO process are compared. It is clearly shown that the CuO process receives in almost all weighting sets a higher ranking than the ACFBC system.

The ACFBC system would get a better ranking if the damage category Human Health was weighted considerably lower (lower than 12%) and Resources higher (at least 33%). However, a weighting preference with a lower weight for Human Health than 12% is very unlikely.
The third triangle show that the ranking between CuO process and wet-limestone scrubbing does not depend on the selected weighting set. The CuO process has in every case a lower environmental load, in agreement with the former results.

4.3 Conclusions

According to the results, the following statements can be concluded:

- The environmental performance of alternative SO\textsubscript{2} removal techniques can be determined with the help of EI-99 impact indicators. This makes possible the comprehensive environmental evaluation and comparison of the alternatives and moreover permits their ranking. These results help engineers during the process selection problem.
- The environmental evaluation shows that the application of SO\textsubscript{2} abatement techniques is able to reduce the total environmental impact by at least 80%; moreover, a further increase in the utilization ratio of by-products is desirable in order to achieve a further decrease in environmental impact.
- The environmental-based comparison of the three different processes shows that there is only a small difference between the ACFBC and wet-limestone scrubbing from an environmental viewpoint. The study shows that wet-limestone scrubbing is preferred if human health aspects are featured and ACFBC if raw material reserves are featured. The higher efficiency and the easier way of utilization of the by-products of the novel technique result in a significantly lower environmental impact than those of the conventional techniques. The CuO process causes less damage to human health and ecosystem quality; however, it consumes a similar amount of raw materials for the treatment of the same amount of sulphur in the flue gas as the other investigated processes.
- The most attractive process is the flue gas cleaning system using regenerable sorbent, where SO\textsubscript{2} is not converted into a new compound. This result is in agreement with the design heuristic that different materials should not be mixed or unnecessarily transformed where possible.
CHAPTER 5  REGIONAL EFFECTS AND EFFICIENCY OF SO$_2$ CONTROL IN THE CARPATHIAN BASIN

Despite the efforts of expansive national and international environmental measures and available technical potential, air pollution remains an important global problem. Air pollutants have high mobility; therefore, they cause environmental problems not only at a local but even at the regional scale.

In connection with the previous chapter, SO$_2$ – as one of the most important air pollutants – is considered and investigated. However, while a continuous decrease in the atmospheric level of SO$_2$ can be seen in Europe, localized SO$_2$ problems still exist related to local emission, meteorological, and topographical factors.

Due to this fact, the effect and efficiency of installed SO$_2$ emission reduction processes are investigated on a regional scale. Atmospheric transport and chemistry is simulated and the simulation results are analysed to show the change in air pollution both in horizontal and vertical directions.

5.1  Introduction

The atmospheric fate and transport of air pollutants such as SO$_2$ is very complex from the point of chemical and physical analysis; however, high capacity computers facilitate their modelling and simulation. The relation between emitted SO$_2$ and its airborne concentration can be simulated by atmospheric transport and chemistry models applied in tandem to meteorological data.

The modelling of SO$_2$ air pollution is a complex task which has drawn the attention of many scientists from all over the world since the early 1960s. Several deterministic and statistical modelling approaches have been proposed. Statistical approaches are frequently considered for short-term forecasting. They are developed and calibrated using local data, while deterministic models are portable to different areas of the world. They are therefore more difficult to apply.

Langmann and Graf (1997) established a model environment which allows estimation of the influence of global climate change on the chemistry of the polluted atmosphere over Europe. The model is adaptable to input data from a regional climate model, which is nested in a global atmospheric circulation model. Furthermore, Langmann (2000) developed a three-dimensional regional scale atmosphere-chemistry model to contribute to an improved understanding of atmospheric photochemical processes. This model calculates meteorological processes jointly with tracer transport and photochemistry. The model was validated and evaluated using a 10-day simulation of a summer smog episode over Europe in July 1994. The comparison of observations and simulation results of an off-line model showed that the on-line model was able to reproduce measured near-surface concentrations more accurately than the off-line model.

Recently, several studies of atmospheric transport of SO$_2$ were made public, e.g. Zunckel et al. (2000). In the absence of a major network of direct measurements of dry deposition, regional-scale dry deposition estimates from the MATCH (Multiscale Atmospheric Transport and Chemistry Model) were compared at five sites to those derived previously from the inferential method of Hicks et al. (1987, 1991), by Zunckel et al. (1999) and Zunckel (1999). MATCH and the inferential model estimates of dry deposition were in reasonable agreement. Furthermore, MATCH-modelled wet deposition rates compared favourably with network measurements. Marmer and Langmann (2005) investigated the impact of ship emissions on the Mediterranean summertime pollution and climate through a case study. They determined
the seasonal variability of secondary trace gases and aerosols, their origin and impact on climate. Siniarovina and Engardt (2005) carried out a one year simulation of anthropogenic sulphate and sulphur dioxide with a high-resolution model for South-East Africa. Their work showed that the calculated sulphate concentrations (on atmospheric aerosols and in precipitation) compared reasonably with observations, while atmospheric SO$_2$ mixing ratios were not in agreement.

5.2 Investigation of SO$_2$ control, a case study

The effect of flue gas desulphurization (FGD) systems applied at large capacity coal-fuelled power plants on atmospheric SO$_2$ concentration is studied on a regional scale. For emission sources, large Hungarian capacity power plants are considered. The investigated area is the Carpathian Basin.

The numerical tool used for the simulation of the atmospheric fate of emitted SO$_2$ is REMOTE (Regional Model with Tracer Extension) which has been successfully in the cases of atmospheric transport, deposition and chemical transformation of air pollutants (Bauer and Langmann, 2000, 2002; Langmann, 2000; Marmer and Langmann, 2005).

The model results are specifically analysed to determine the effect of FGD systems on the atmospheric dispersion of SO$_2$, which is shown by parallel simulations with a single modified parameter (SO$_2$ emission rate). In our study, two emission scenarios are constructed: the first one is based on reported emission data of high capacity power plants; the second emission scenario considers the supplementary installation and application of FGD units at all investigated power plants.

5.3 Data and methods

5.3.1 Atmospheric model

The atmospheric transport of the emitted SO$_2$ is simulated by the hydrostatic, regional three-dimensional atmosphere-chemistry model REMOTE (Langmann, 2000) which determines the physical and chemical state of the model atmosphere at every time step of the simulation. REMOTE is a coupled model consisting of the regional atmospheric circulation model REMO 5.0 (Jacob, 2001) and the gas-phase chemistry module RADM II (Stockwell et al., 1990).

The model uses a terrain-following hybrid vertical coordinate with 27 layers between the surface and a 10 hPa pressure level. The horizontal resolution is 1/6° on a spherical rotated grid with a model timestep of 100s. Meteorological processes are described with second order horizontal and vertical differences with leap-frog time stepping with semi-implicit correction and Asselin-filtering. The finite-difference equations are written in advective form on the Arakawa C-Grid. The dynamic part of the model REMO is based on the former weather forecast model EM/DM of the German Weather Service (Majewski, 1991). The prognostic atmospheric variables of the model are: surface pressure, temperature, horizontal wind components, water vapour content and cloud water content. They are adjusted towards the large-scale forcing (ECMWF analysis, see below) in a lateral sponge zone of 8 grid boxes according to Davies (1976). The model includes fractional surface cover for land, water, and sea ice (Semmler, 2002). A monthly variation of vegetation parameters (background albedo, leaf area index, vegetation fraction) is considered in the model (Rechid and Jacob, 2006). A detailed description of the model referring to physical parameterization,
cumulus convection, stratiform cloud water content, radiative transfer scheme, can be found in Benko et al. (2007).

In the current model set-up (Marmer and Langmann, 2005), species transport is determined by horizontal and vertical advection according to the algorithm of Smolarkiewitz (1983). Convective up- and down-draft is calculated by a modified scheme of Tiedtke (1989) and vertical diffusion is calculated according to Mellor and Yamada (1974). Dry deposition velocities are computed as in Wesley (1989) dependent on the friction velocities and stability of the lowest model layer. Wet deposition is computed according to Waleck and Taylor (1986) by integrating the product of the grid-averaged precipitation rate and the mean cloud water concentration. REMOTE includes the gas-phase chemistry package RADM II implemented with a quasi-steady-state approximation solver (Hestvedt et al., 1978). The chemistry package describes the atmospheric chemistry of 63 chemical species: 43 prognostic species and 20 short-lived diagnostic molecules. The photochemical gas phase mechanism consists of 158 reactions. The clear sky photolysis rates are calculated by a climatological pre-processor model (Madronich, 1987). They are modified in the presence of clouds.

The gas phase chemistry of SO\(_2\) is described by RADM II with the chain mechanism shown in Equations (5.1)-(5.4) (Stockwell et al., 1990). SO\(_2\) reacts with hydroxy radicals (HO) and produces sulphuric acid (H\(_2\)SO\(_4\)) at the end of the chain. In aqueous phase, SO\(_2\) is oxidized by H\(_2\)O\(_2\), O\(_3\), MHP (methyl-hydrogen-peroxide), PAA (peroxyacetic acid), and O\(_2\) (catalyzed by Fe\(^{3+}\) and Mn\(^{2+}\)). Other sinks of SO\(_2\) are dry and wet deposition. REMOTE calculates chemical species concentration data for each grid box of the three-dimensional model domain, each chemical compound and at every time step.

\[
\begin{align*}
\text{SO}_2 + \text{HO (+M)} & \rightarrow \text{HOSO}_2 (+M) & (5.1) \\
\text{HOSO}_2 + \text{O}_3 & \rightarrow \text{SO}_3 + \text{HO}_2 & (5.2) \\
\text{SO}_3 + \text{H}_2\text{O} & \rightarrow \text{H}_2\text{SO}_4 & (5.3) \\
\text{HO}_2 + \text{NO} & \rightarrow \text{HO} + \text{NO}_2 & (5.4)
\end{align*}
\]

In this study, the REMOTE model is run in 'climate mode'. That means that the model is initialised with meteorological and chemical data over the whole three dimensional model domain at the first time step. Then the model runs continuously till the end of the simulation period with chemical and meteorological updates at the lateral boundaries every six hours. Simulation results are recorded every six hours.

5.3.2 Initial and boundary conditions

For the initialisation and the 6-hourly updates of the meteorological field in the model, analysis data of the European Centre for Medium-Range Weather Forecasts (ECMWF) are used (http://www.ecmwf.int/). The chemical fields are initialised and updated at the lateral boundary with simulation results from the global chemistry transport model MOZART 4 (MOZART 4, 2006), which include: SO\(_2\), sulphate, NO, NO\(_2\), O\(_3\), HNO\(_2\), HNO\(_3\), HO\(_2\)NO\(_2\), NH\(_3\), N\(_2\)O\(_5\), NO\(_3\), PAN (peroxyacetyl nitrate), TPAN (H-(CO)-CH=CH-CO\(_3\)-NO\(_2\)), organic nitrogens, CO, H\(_2\)O\(_2\), organic peroxydes, organic acids, alkanes, alkenes, BTX, cresol, glyoxal, methylglyoxal, aldehydes, ketones, unsaturated dicarbonyls and isoprene. The concentration of methane is held constant throughout the simulation.
5.3.3 Emission data

Anthropogenic emission fields including $\text{SO}_2$, $\text{NO}_x$, CO, ethane, propane, butanes, ethene, propene, ethyne, alcohols, benzene, toluene, and xylene are considered. Emission fields are defined only inside the country border of Hungary; emission data enter the model at the lowest model level (approx. 60m).

$\text{SO}_2$ emission data are obtained from the Hungarian Energy Office (HEO) in form of official annual emission reports submitted by Hungarian high capacity power plants - see Table 5.1. The reported annual emission rate has been distributed equally over the year. Emission data of the other species are obtained from the ‘REanalysis of the TROpospheric chemical composition over the past 40 years’ (RETO) project in the form of a 0.5°x0.5° gridded monthly mean emission rate data set (http://www.reto Romero/). In our simulations, $\text{NO}_x$ emission is released in form of 96% $\text{NO}_2$ and 4% NO. The RETRO data is remapped on the REMOTE model grid before simulations take place.

<table>
<thead>
<tr>
<th>Point sources</th>
<th>Longitude [°E]</th>
<th>Latitude [°N]</th>
<th>Emission rate [ton/year]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Oroszlányi Erőmű</td>
<td>18.271</td>
<td>47.502</td>
<td>102,974</td>
</tr>
<tr>
<td>Mátrai Erőmű</td>
<td>20.067</td>
<td>47.790</td>
<td>98,920</td>
</tr>
<tr>
<td>Dunamenti Erőmű</td>
<td>18.918</td>
<td>47.328</td>
<td>26,055</td>
</tr>
<tr>
<td>Bánhidai Erőmű</td>
<td>18.373</td>
<td>47.573</td>
<td>21,566</td>
</tr>
<tr>
<td>Pécsi Hőerőmű</td>
<td>18.263</td>
<td>46.064</td>
<td>28,596</td>
</tr>
<tr>
<td>Borsodi Hőerőmű</td>
<td>20.684</td>
<td>48.235</td>
<td>25,016</td>
</tr>
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<td>47.906</td>
<td>13,806</td>
</tr>
<tr>
<td>Tisza Erőmű</td>
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<td>47.917</td>
<td>13,821</td>
</tr>
<tr>
<td>Ajkai Hőerőmű</td>
<td>17.558</td>
<td>47.096</td>
<td>9,294</td>
</tr>
</tbody>
</table>

Table 5.1 Localization and emission rate of the investigated power plants.

5.3.4 Model domain

The investigated region is the Carpathian Basin which includes mostly Hungary and its neighbours. The model domain of the simulation is shown in Figure 5.1. Data analysis is performed only for Section A in order to decrease the influence of the boundary data. The simulations are carried out on a rotated grid. Geographical coordinates are therefore not shown in the plots and in the figures. National borders help orientation on the maps.
5.4 Discussion

The aim of this study is to show the influence of the application of SO$_2$ emission control systems on air quality in the Carpathian Basin. Therefore, parallel simulations are carried out with REMOTE using identical meteorological and atmospheric chemistry (emissions and initial background concentrations) boundary data, except the SO$_2$ emission field.

In the case of SO$_2$, two emission scenarios are constructed: in the Base Scenario (1), the SO$_2$ emission field of the year 2000 is prepared on the basis of HEO reports. Hungarian coal-fuelled power plants responsible for more than 90% of the total annual SO$_2$ emissions of the power sector are selected for this study. Emission rates and geographical location of the selected power plants are shown in Table 5.1.

For the Alternative Scenario (2), it is assumed that all investigated power plants are fitted with FGD units with an SO$_2$ removal efficiency of 90%. Based on HEO data and the geographical locations of the investigated power plants, the emission data are placed as point source emissions on the spherical rotated REMOTE grid (see Figure 5.1). The effect of the application of FGD units is shown by the differences found during analysis of the results of the parallel REMOTE simulations. REMOTE delivers simulation results in for each grid box.

A grid box is considered to be affected if the SO$_2$ concentration exceeds 0.001 ppm(v). The time frame of the simulations include one month of each season (respectively, January, April, August, and November of the selected year 2000) with the aim of capturing the effect of the annual cycle of meteorological conditions on the atmospheric dispersion of SO$_2$. The simulations start with the appropriate initial values prepared from ECMWF, RETRO and HEO data on the first day of the month and end at the 30th day.

Table 5.2 shows seasonal meteorological data and the square of the standard deviation ($s^2$) for the following parameters: monthly mean temperature, 10 meter wind, daily mean and monthly sum of precipitation as simulated by REMOTE. In April, there is relatively large...
variability in the daily mean temperature. Daily precipitation varies from 1-3 mm/day in average in each month. January is the driest and November is the most humid month (see monthly sum of precipitation). The monthly mean of the 10m wind velocity varies from 2.0 to 3.1 m/s.

<table>
<thead>
<tr>
<th></th>
<th>Temperature</th>
<th>Precipitation</th>
<th>10m wind</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Monthly mean</td>
<td>Daily mean</td>
<td>Monthly</td>
</tr>
<tr>
<td></td>
<td>°C</td>
<td>mm/day</td>
<td>sum/mm/month</td>
</tr>
<tr>
<td></td>
<td>s²</td>
<td>s²</td>
<td>m/s</td>
</tr>
<tr>
<td>January</td>
<td>-3.8</td>
<td>8.9</td>
<td>1</td>
</tr>
<tr>
<td>April</td>
<td>12.9</td>
<td>22.8</td>
<td>2</td>
</tr>
<tr>
<td>August</td>
<td>21.6</td>
<td>7.6</td>
<td>1</td>
</tr>
<tr>
<td>November</td>
<td>7.8</td>
<td>7.3</td>
<td>3</td>
</tr>
</tbody>
</table>

Table 5.2 Meteorological data of the investigated months in 2000.

As the aim of this study is to analyse the changes in the atmospheric dispersion of the emitted SO₂ driven by the supplementary installed FGD units, the following terms are investigated: (1) horizontal and (2) vertical distribution and accumulation of emitted SO₂.
5.5 Results

5.5.1 Horizontal dispersion

Figure 5.2 shows the monthly mean horizontal dispersion and accumulation of SO$_2$ in the lowest atmospheric level (approx. 60m). Figure 5.2 (a)-(d) show the monthly mean SO$_2$ concentrations in the different grid boxes if no FGD units are considered. Figure 5.2 (e)-(h) show the concentrations, if FGD units are installed. A continuous black line marks the border of the affected region.

In the base emission scenario (Base Scenario), SO$_2$ disperses and accumulates strongly in the air in January; however, air pollution is lower in the other seasons. The size of the affected region is similar (56,271 – 67,094 km$^2$) in April, August and November, and it is two to three times larger in January (160,817 km$^2$). SO$_2$ emissions cause transboundary air pollution. The size of the affected region outside Hungary is shown in Table 5.3.

<table>
<thead>
<tr>
<th>Month</th>
<th>Emission scenario</th>
<th>Reduction</th>
<th>Total size of the affected region</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Base</td>
<td>Alternative</td>
<td></td>
</tr>
<tr>
<td>January</td>
<td>81,627</td>
<td>23,771</td>
<td>71</td>
</tr>
<tr>
<td>April</td>
<td>9,153</td>
<td>0</td>
<td>100</td>
</tr>
<tr>
<td>August</td>
<td>10,162</td>
<td>678</td>
<td>93</td>
</tr>
<tr>
<td>November</td>
<td>21,388</td>
<td>678</td>
<td>97</td>
</tr>
<tr>
<td>Total</td>
<td>122,330</td>
<td>25,127</td>
<td>79</td>
</tr>
</tbody>
</table>

Table 5.3 Numerical results of the horizontal analysis in Section A [km$^2$].

The transboundary transport of air pollution spreads furthest in the Base Scenario, especially in January. It hardly reaches the borders in the Alternative Scenario for all other months. In January, large amounts of SO$_2$ accumulate outside Hungary. The application of FGD units strongly reduces the transboundary air pollution, as expected. The annual mean relative reduction is 79%.

The reduction in the size of the affected region is also presented in Table 5.3. There is a large difference in the reduction ratios of the affected region size in the different seasons: the reduction ratio is relatively high in April, August, and November (87-89%), but it has a significantly lower value in January (68%). The emission rates of the point sources are the same for all months, therefore the regional distribution and accumulation of the SO$_2$ is forced by the meteorological conditions. The same level of technological SO$_2$ removal efficiency can lead to significantly different reductions in air pollution in winter compared to the other seasons. In conclusion, SO$_2$ removal efficiency should be enhanced in winter time in order to achieve similarly low levels of air pollution to other seasons.
Figure 5.2 Horizontal distribution of monthly mean SO$_2$ concentration at the first model level.
5.5.2 Vertical dispersion

Vertical distribution of SO₂ is displayed by plotting the daily field mean concentration in Section A at the different model levels (see Figure 5.3). Only the 15 lowest model levels of the total 27 model levels are shown to aid visualization.

Simulation results according to the base emission scenario (Base Scenario) are presented in Figure 5.3 (a)-(d) and according to the Alternative Scenario in Figure 5.3 (e)-(h).

The numerical information referring to the vertical distribution analysis is presented in Table 5.4 which shows the number of grid boxes affected, maximal altitude above sea level reached by the of the affected region in a month, and the mean of the highest daily level data for each month (since mean of integers is determined, the result is a decimal number).

<table>
<thead>
<tr>
<th></th>
<th>Number of grid boxes affected by SO₂</th>
<th>Maximal altitude reached by the affected region</th>
<th>Mean height of the affected region</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>[number of grid boxes]</td>
<td>[m]</td>
<td>[model level]</td>
</tr>
<tr>
<td>Base Scenario</td>
<td>136</td>
<td>2,600</td>
<td>4.2</td>
</tr>
<tr>
<td>Alternative Scenario</td>
<td>90</td>
<td>2,600</td>
<td>3.1</td>
</tr>
<tr>
<td>Reduction [%]</td>
<td>44</td>
<td></td>
<td>26</td>
</tr>
<tr>
<td>Base Scenario</td>
<td>42</td>
<td>750</td>
<td>1.3</td>
</tr>
<tr>
<td>Alternative Scenario</td>
<td>2</td>
<td>500</td>
<td>0.1</td>
</tr>
<tr>
<td>Reduction [%]</td>
<td>95</td>
<td></td>
<td>92</td>
</tr>
<tr>
<td>Base Scenario</td>
<td>59</td>
<td>1,100</td>
<td>1.9</td>
</tr>
<tr>
<td>Alternative Scenario</td>
<td>3</td>
<td>800</td>
<td>0.1</td>
</tr>
<tr>
<td>Reduction [%]</td>
<td>95</td>
<td></td>
<td>94</td>
</tr>
<tr>
<td>Base Scenario</td>
<td>44</td>
<td>750</td>
<td>1.4</td>
</tr>
<tr>
<td>Alternative Scenario</td>
<td>1</td>
<td>450</td>
<td>0.3</td>
</tr>
<tr>
<td>Reduction [%]</td>
<td>98</td>
<td></td>
<td>98</td>
</tr>
<tr>
<td>Sum:</td>
<td>281</td>
<td>Mean:</td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td>96</td>
<td></td>
<td>0.78</td>
</tr>
<tr>
<td></td>
<td>66</td>
<td></td>
<td>65</td>
</tr>
</tbody>
</table>

Table 5.4 Numerical results of the vertical analysis in Section A.

The analysis of Figure 5.3 and Table 5.4 shows the changes in vertical distribution of SO₂ due to the application of FGD units. The number of grid boxes affected (Table 5.4, column 1) is proportional to the volume of the affected air mass. Emission reduced by FGD units result in lower air pollution: the volume of affected air mass is reduced by 66% on a yearly average. In January, this reduction ratio is significantly lower (44%) than in the other three months (95-98%). The mean height of the affected region in a month is also determined (Table 5.4, column 3) which is proportional to the vertical transport of SO₂. On a yearly mean, the relative reduction in height of the affected region due to FGD units is 65%. In January, the relative reduction is significantly lower (26%) than in the other three months (92-98%). In general, due the application of the FGD units, the vertical air column is free from strong SO₂ pollution all the time, except January.

Table 5.4 (column 2) shows the maximal altitude above sea level reached by the affected region in each simulated month. Extreme high altitude is reached just once or just several times per month. A relative comparison of altitude values in the case of the two emission scenarios shows that altitude is not strongly affected by the FGD units. However, significant intra annual variability can be found in altitude values: the highest altitude is 2600m in January and 450m in November. This shows that these extreme vertical transport and accumulation events are not really influenced by the FGD units, but are mainly driven by meteorological factors. The analysis of the vertical distributions confirms that winter time SO₂ removal efficiency should be enhanced in order to achieve similarly low air pollution to other seasons.
Figure 5.3 Vertical distribution of daily mean SO$_2$ concentration in Section A.
5.6 Conclusions

The results show that FGD units have a significant influence both on the vertical and the horizontal distribution and accumulation of the emitted SO\textsubscript{2}. FGD units reduce the transboundary transport of SO\textsubscript{2}, whereas they have no strong effect on the maximal altitude reached by SO\textsubscript{2} within one month.

Based on the simulation results, FGD units with 90\% SO\textsubscript{2} removal efficiency reduce horizontal distribution by 79\%, and vertical distribution by 66\% for the yearly average.

Significant intra-annual variability is found in the efficiency of FGD units on the air pollution ratio: significantly worse air quality can be detected in winter than in the other seasons with the same SO\textsubscript{2} removal efficiency at pollution sources.

The detected intra-annual variability in the reduction ratios is due to meteorological conditions, which shows that winter weather strongly decreases the efficiency of FGD units on air pollution prevention. To overcome this problem an initiative such an SO\textsubscript{2} removal policy with greater efficiency (higher than 90\%) in winter should be followed. The weather circumstances of the other seasons are more favourable for the full utilization and realisation of SO\textsubscript{2} removal benefits.
CHAPTER 6 APPLICATION OF LCA TO DETERMINE THE PREFERABLE WASTE SOLVENT TREATMENT OPTION

The recovery and recycling of waste solvents is one of the important environmental directives of the European Union. It is not clear, however, under what circumstances solvent recovery should be preferred over incineration.

However, with the proper application of the LCA this question can be answered. It might be also interesting to compare the results with the results of economic analysis.

6.1 Introduction

A typical environmental dilemma is the problem of used solvents creating waste streams. In different industries, usually chemical ones, organic solvents are used in large amounts for synthesis processes as well as for work-up and purification of products and, as a consequence, a huge amount of waste solvent is produced every year. Since many solvents show high volatility, considerable environmental persistence and high toxicity, the handling of solvents in the chemical industry clearly represents a high priority environmental issue (Sardessai and Bhosle, 2002.)

Nowadays, experts are making major efforts to consider environmental impact by process optimization and the selection of an optimal solvent (Pistikopoulus and Stefanis, 1998; Stefanis et al., 1996; Sinha et al., 1999). However, the treatment of waste solvents from existing technologies is still a problem of high priority.

An industrial case study on the waste solvent treatment alternatives of a printing company is selected for demonstration of LCA modelling. In the studied case, a waste solvent mixture containing mainly ethanol (ETOH), isopropyl acetate (IPAC), ethyl acetate (ETAC), and water (H₂O) is generated as by-product which requires treatment before release to the wider environment.

The quantity of the waste stream is about 40,000 tons/year which explains the need for continuous operation. The main physical properties of the components and the composition are shown in Table 6.1.

Two types of waste solvent treatment options are considered:

- incineration with heat utilization,
- component recovery.

Each option has its own advantages and disadvantages: in the case of the incineration, thermal energy is recovered but new solvents are needed, consuming natural resources. In the case of recovery carried out by separation processes like distillation, membrane separation or other alternatives, the recovered solvents can be reused; however, a huge amount of steam is consumed in this case.

Selection between solvent treatment alternatives has been usually made on the basis of economic analysis, however, the problem is sometimes complicated since sometimes re-use of the solvent is strictly prescribed (e.g. in pharmaceutical industry) and environmental considerations add to complexity. Moreover, if an on-site incinerator is used next to recovery apparatus (i.e. distillation columns) the waste solvent stream can be split between the two apparatus.
Table 6.1 The physical properties of the waste solvent mixture components.

<table>
<thead>
<tr>
<th></th>
<th>Ethanol</th>
<th>Ethyl acetate</th>
<th>i-Propyl acetate</th>
<th>Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weight fraction</td>
<td>0.309</td>
<td>0.261</td>
<td>0.221</td>
<td>0.209</td>
</tr>
<tr>
<td>Formula</td>
<td>C₂H₆O</td>
<td>C₄H₈O₂</td>
<td>C₅H₁₀O₂</td>
<td>H₂O</td>
</tr>
<tr>
<td>Boiling points [°C]</td>
<td>78</td>
<td>77</td>
<td>88</td>
<td>100</td>
</tr>
<tr>
<td>Mol. weight, used for calculation [g/mol]</td>
<td>46</td>
<td>88</td>
<td>102</td>
<td>18</td>
</tr>
<tr>
<td>Heat of combustion, ( \Delta H_{\text{comb}} ) [MJ/kg]</td>
<td>32.33</td>
<td>24.35</td>
<td>20.16</td>
<td>-5°</td>
</tr>
</tbody>
</table>

\( ^{a} \): \( \Delta H_{\text{H₂O}}^{\text{imp}} (1200^\circ C) = 5 \text{ MJ/kg} \)

6.2 Waste solvent treatment options

6.2.1 Incineration

Incineration is a typical end-of-pipe waste solvent treatment option. Waste solvents are burnt in a furnace with optimized combustion performance for total oxidation of the solvent components. The incineration process is usually supported with easy combustible fuels (i.e. light fuel oils) in order to maintain the desired temperature in the furnace. Energy released by combustion can be converted to steam and electricity.

The scheme of such a waste solvent incinerator is obtained from Seyler (2003), see Figure 6.1. Combustion takes place in the furnace; the flue gas is purified in a washer and with a catalytic flue gas cleaner before its release to air. Slag and ash produced in the furnace, and particulates removed from the flue gas are collected and treated as solid wastes. Electricity and steam are produced during the incineration process; the extent of the obtained energy depends on the heat of combustion of the inflow waste solvent stream.
For the preparation of the LCA, detailed literature data of an existing waste solvent incinerator are used (Hofstetter, 2002). Input-output data of the incinerator are shown in Appendix C. In 1998, the reference year for data used in this study, the incinerator produced 191 GWh$_{net}$ thermal energy (steam) and 14 GWh$_{net}$ electric energy (electricity). More than 81% of the energy produced originated from the incineration of waste solvents (33,500 tons). The burning process required support oil and natural gas in order to maintain the required temperature in the incinerator and also for system start up. Low pressure steam and electricity can be produced during operation at 86% efficiency.

6.2.2 Solvent recovery with distillation based on hybrid separation processes

Recovery of the solvent components is possible with several techniques. According to the investigated waste solvent treatment case study, two possible alternatives, recently published in the literature (Mizsey et al, 2002; Szanyi et al., 2004) are considered and investigated. Organic components of the waste solvent stream are recovered with 95w/w% purity. The water phase leaving the system must not contain any solvent components. Both investigated processes fulfil these purity requirements.

In the studied procedures, water is used as separating or extractive agent – this corresponds with international environmental policies on green chemistry (Anastas and Warner, 1998) which propose that the use of new materials as separating agents is avoided.

In an earlier design, a quite complicated separation process (the so called ‘ternary cut scheme’) was elaborated for the recovery of the studied solvent mixture. The process consists of seven distillation columns (C1-C7) and two liquid-liquid extractors (E1-E2), see Figure 6.2. First, the quaternary mixture is separated into two ternary ones. The top product contains no IPAC and the bottom product contains no ETAC. The two ternary mixtures can be processed simultaneously in a similar way.

Process performance and simulation results used in this study are obtained from the literature. The input/output data of the ternary cut scheme obtained from Raab (2001) is shown in Appendix C.
This separation scheme can be significantly simplified if the so-called extractive heterogeneous-azeotropic distillation (EHAD) (Szanyi et al., 2004) is applied and used as a basis for this separation problem (Figure 6.3). In this novel design, ordinary distillations and internal recycles are also applied. This separation scheme consists of only four distillation columns (C1-C4) and a phase separator (S), so this scheme is much simpler than the previous one. Input-output data of the distillation are shown in Appendix C.
6.2.3 Simultaneous thermal and component recovery

If the waste solvent incinerator operates directly on or close to the recovery plant, the energy demand of the recovery can be covered by on-site produced steam.

The incinerator can be fuelled either by waste solvent or by light fuel oil in order to capture a sufficient amount of steam to cover the energy demand of recovery. Thus a connection between the two systems is possible: a fraction of the waste solvent stream can be fed into the incinerator in order to produce energy that can be utilized at the recovery stage and the rest of the solvents can be recovered. If energy obtained by the incineration of the waste solvent fraction being incinerated exceeds the energy demand of the recovery surplus, energy can be utilized in the grid. If waste solvent-based energy production is not sufficient, the missing steam can be generated by the incineration of light fuel oil.

6.3 LCA modelling

A model of the waste solvent treatment is prepared. The functional unit of the LCA study is the treatment of the waste water stream during a one hour operation.

The two alternative systems are defined and represented with dotted lines, considered system boundaries, which include:
1. solvent production (for solvent make-up),
2. technological application of the solvent, and
3. disposal (waste solvent treatment).

The symbolic flow sheet for the LCA of the two treatment alternatives and the possible connection between recovery and incineration are shown in Figure 6.4.

The environmental impacts of the treatment processes are assessed using EI-99 impact indicators (best guess values). The ‘egalitarian’ version of the method is used with the egalitarian weighting set (30%, 50%, and 20% weighting for the damage categories Human Health, Ecosystem Quality, and Resources, respectively).
The environmental impact of the functional unit \( I_{\text{treatment\_option}} \) can be calculated on a standard basis as represented by Equation (6.1); the dimension is EI-99 points/h.

\[
I_{\text{treatment\_option}} = I_{\text{solv\_prod}} + I_{\text{ws\_treatment}} \tag{6.1}
\]

\( I_{\text{solv\_prod}} \) represents the environmental impacts caused by the industrial production of the components in the waste solvent mixture. This is the product of the massflow of the \( i \)th solvent component \( \dot{m}_i \) in the waste solvent stream and the specific impact indicator \( i_{\text{prod}} \) referring to the petrochemical production of the \( i \)th component, as shown in Equation (6.2).

\[
I_{\text{solv\_prod}} = \sum (\dot{m}_i \cdot i_{\text{prod}}) \tag{6.2}
\]

\( I_{\text{ws\_treatment}} \) represents the environmental impacts during a one hour operation of the investigated treatment plant. To determine the accurate Eco-points referring to the different treatment options a detailed model of each processing unit of the solvent treatment alternatives (recovery and/or incineration) should be elaborated. The preparation of the LCA requires detailed life cycle inventories of the treatment alternatives.
6.3.1 Assessment procedure of the recovery

Environmental impacts of the recovery processes are calculated using the same equation including the same parameters, see Equation (6.3). All parameters are functions of the mass flow of the waste solvent stream, indicated as $\dot{m}$.

$$I_{ws,\text{recovery}} = I_{\text{basic, dist}} + I_{\text{investment, dist}} + I_{\text{operation}} + I_{\text{residue}} - I_{\text{rec.ws}}$$ (6.3)

**Basic environmental impacts of solvent recovery**

$I_{\text{basic, dist}}$ represents the contribution to the total environmental impact of the recovery process caused by solvents and chemicals (methanol and N$_2$) used for the cleaning and the maintenance of the columns (by breakdown). Equation (6.4) represents the environmental impacts related to these activities: $i_{\text{basic, dist}}$ represents the environmental impacts of the production of these chemicals and the emissions of the recovery plants to air and water referring to 1 kg waste solvent.

$$I_{\text{basic, dist}} = \dot{m} \cdot i_{\text{basic, dist}}$$ (6.4)

**Environmental impacts due to equipment installation**

$I_{\text{investment, dist}}$ stands for installed material of the recovery plant, assessed based on the size and material costs of the installed equipment, see Equation (6.5). The environmental impact of the installation referring to 1 kilogram of waste solvent mixture is denoted by $i_{\text{investment, dist}}$ which has different values for the two recovery alternatives. Project life is 10 years. An interaction between the environmental impact of investment and waste solvent stream with a power of 0.6 is supposed.

$$I_{\text{investment, dist}} = (\dot{m})^{0.6} \cdot i_{\text{investment, dist}}$$ (6.5)

**Environmental impacts related to operation**

$I_{\text{operation}}$ represents the environmental impacts of the steam and cooling water consumption of the distillation steps. The amount of cooling water depends only on the mass flow of the waste solvent stream. However, steam has two possible sources in our consideration: heat obtained form the incineration of waste solvents or incineration of light fuel oil. Therefore, in Equation (6.6) only the environmental impacts of the extra steam produced by the incineration of oil are considered -since the environmental impacts of solvent combustion are considered at the incinerator.

According to this, in Equation (6.6), $\dot{m}_{\text{steam,oil}} \ [kg / h]$ represents the mass flow of the steam obtained from the incineration of light fuel oil and $i_{\text{steam,oil}}$ stands for the environmental impacts of the incineration light fuel referring to 1 kilogram of steam obtained.

The environmental impacts of the cooling water consumed by the recovery are calculated as the product of the cooling water demand referring to 1 kilogram of waste solvent ($i_{\text{cooling water}}$), the mass flow ($\dot{m}$) of the waste solvent stream being recovered, and the impact indicator referring to the production and delivery of 1 kilogram of cooling water ($i_{\text{cooling water}}$).

$$I_{\text{operation}} = \dot{m}_{\text{steam,oil}} \cdot i_{\text{steam,oil}} + r_{\text{cooling water}} \cdot \dot{m} \cdot i_{\text{cooling water}}$$ (6.6)
Environmental impacts due residue treatment

Environmental impacts due to the treatment of the residues are also considered. It is assumed that the disposal happens in a municipal incinerator. Environmental impacts related to the disposal are expressed as in Equation (6.7). The specific impact of the residue treatment referring to 1 kilogram of waste solvent is denoted by \( i_{\text{residue}} \).

The yield of the recovery (\( \alpha \)) is 0.95, in this study.

\[
I_{\text{residue}} = (1 - \alpha) \cdot \dot{m} \cdot i_{\text{residue}} \tag{6.7}
\]

Environmental impacts of solvent make-up

Recovered solvents replace fresh solvents and thus the environmental impacts of their industrial production. Therefore the environmental impacts of the recovered solvents (\( I_{\text{rec.ws}} \)) are equal to the environmental impacts of the industrial production of the studied waste solvent mixture (\( I_{\text{solv.prod}} \)) multiplied by the yield and have a negative sign, as in Equation (6.3). The calculation, therefore, needs iteration.

\[
I_{\text{rec.ws}} = \alpha \cdot I_{\text{solv.prod}} \tag{6.8}
\]

6.3.2 Assessment procedure of the incineration

Environmental impacts caused by the treatment of the waste solvent stream with incineration are calculated by Equation (6.9).

\[
I_{\text{ws,inc}} = I_{\text{basic,inc}} + I_{\text{investment,inc}} + I_{\text{CO}_2} + I_{\text{oil}} - I_{\text{gain}} \tag{6.9}
\]

All elements are functions of the mass flow of the waste solvent stream; moreover, some functions depend also on the composition of the waste solvent. According to this,

- Equations (6.10) and (6.11), are considered composition dependent, while
- Equations (6.12)-(6.14) are composition independent functions.

Basic environmental impacts of incineration

\( I_{\text{basic,inc}} \) refers to the composition-independent material and energy flows of the incinerator, Equation (6.10). It includes the environmental impacts of the production of chemicals consumed at the plant, emissions to air and water, and solid emissions. The aggregated environmental indicator of these terms referring to 1 kilogram of waste solvent is denoted by \( i_{\text{basic,inc}} \).

\[
I_{\text{basic,inc}} = \dot{m} \cdot i_{\text{basic,inc}} \tag{6.10}
\]

Environmental impacts due to equipment installation

\( I_{\text{investment,inc}} \) stands for the installed material of the incinerator plant; \( i_{\text{investment,inc}} \) represents the environmental impacts of the construction materials referring to 1 kilogram of waste solvent. An interaction between the environmental impact of investment and waste solvent stream with a power of 0.6 is supposed (according to process design heuristics). Project life is 10 years.

\[
I_{\text{investment,inc}} = (\dot{m})^{0.6} \cdot i_{\text{investment,inc}} \tag{6.11}
\]
Environmental impacts due to CO₂ emission

Total oxidation of the waste solvent in the furnace is assumed, therefore CO₂ emission can be calculated from the carbon-mass-balance, see Equation (6.12). The environmental impact of the CO₂ released to air is denoted by \( I_{\text{CO}_2} \) which includes carbon entering the system from the waste solvent components \( (\dot{m}_{C,i} \cdot i_{C-CO_2}) \) and the applied light fuel oils: support oil and start-up oil \( (\dot{m}_{\text{oil, support}} + \dot{m}_{\text{oil, start-up}}) \). The carbon content of light oil is assumed to be 86w/w%. The specific environmental impact indicator of CO₂ emission is denoted by \( i_{C-CO_2} \)

\[
I_{CO_2} = \sum (\dot{m}_{C,i} \cdot i_{C-CO_2} + (\dot{m}_{\text{oil, support}} + \dot{m}_{\text{oil, start-up}}) \cdot i_{C-CO_2})
\]  
(6.12)

Environmental impacts due to support oil

Total oxidation of the solvent components is ensured if the required high temperature (1,200°C) in the furnace is also guaranteed. Therefore, support oil is added to the waste solvents to reach the desired heat input of 23 MJ/kg. Since the heat of combustion of the waste solvent mixture is only about 20 MJ/kg, \( r_{\text{oil, ws}} = 0.177 \) kg oil is added to each kilogram of waste solvent mixture.

\( I_{\text{oil}} \) represents the environmental impacts of the industrial production of light fuel oil consumed during operation in form of start-up oil (829 t/year) and support oil. The environmental impacts of the industrial production of 1 kilogram of light fuel oil are indicated as \( i_{\text{oil}} \)

\[
I_{\text{oil}} = (\dot{m}_{\text{oil, start-up}} + r_{\text{oil, ws}} \cdot \dot{m}) \cdot i_{\text{oil}}
\]  
(6.13)

Environmental impacts avoided by heat utilization

\( I_{\text{gain}} \) stands for the avoided environmental impacts through the production of steam and electricity, see Equation (6.14).

In our consideration, the produced energy is either added to the grid or it is consumed by the simultaneously working regenerating plants. Therefore, the extent of the valuable products (energy sent to the grid) is the difference between produced energy and energy consumption of the possible simultaneous working recovery plant.

Considering the efficiency of the incinerator \( (\eta) \) is 86% and the heat of combustion of the waste solvent mixture and of the support oil (together 23 MJ/kg, denoted by \( \Delta H_{\text{Feed}} \)), about 19.8 MJ of energy can be obtained by the incineration of one kilogram of waste solvent. The heat energy requirement of the recovery is denoted by \( \Delta H_{\text{recovery}} [MJ / h] \). The environmental impacts of the valuable products can be calculated if the massflow of the waste solvent stream sent to recovery \( (\dot{m}_{\text{recovery}} [kg / h]) \) and to incineration \( (\dot{m}_{\text{inc}} [kg / h]) \) are determined.

The environmental impacts of the heat energy production that are avoided by the delivery of the surplus heat to the grid are denoted by \( i_{\text{gain}} \) [EI-99 points/MJ energy]. If the heat requirement of the recovery plant is higher than the heat energy produced by the incineration of the waste solvent, the value of \( I_{\text{gain}} \) is zero.

\[
I_{\text{gain}} = \eta \cdot (\dot{m}_{\text{inc}} \cdot \Delta H_{\text{Feed}} - \dot{m}_{\text{recovery}} \cdot \Delta H_{\text{recovery}} \cdot i_{\text{gain}})
\]  
(6.14)
6.3.3 Assessment procedure of the simultaneous incineration and recovery

A model of the treatment options is prepared in order to estimate the total environmental impact as if the two studied treatment options are working simultaneously. The model equations are direct or indirect functions of the waste solvent mass flow entering the treatment options which can be incineration or recovery. The total environmental impact referring to the functional unit is calculated as the superposition of Equations (6.3) and (6.9).

6.3.4 Economic calculation

The economic calculation needs the complete engineering model of the treatment alternatives to determine their major parameters needed.

For the recovery alternatives, these data are obtained from the literature (Raab, 2001; Szanyi et al., 2004) including operational data and the main geometric data for the sizing of the unit operations. Operational costs include steam (low pressure steam) and cooling water consumption. Capital costs include the material demand of the unit operations and installation costs calculated on the basis of geometrical data with the Marshall and Swift indices (Economic indicators, 2004). The project life is 10 years. Prices for calculations are shown in Table 6.2.

In the case of the incineration, utility cost is calculated based on the operational datasheet of the incinerator found in the literature (Hofstetter, 2002) which includes energy and material requirements. The quantity and cost of support oil and oil used for steam production is added to the utility cost too.

According to process design heuristics, the capital cost of an incinerator treating 40,000 t/yr waste solvent is about 24 million dollars.

<table>
<thead>
<tr>
<th>Material cost</th>
<th>Utility cost</th>
<th>Capital cost</th>
</tr>
</thead>
<tbody>
<tr>
<td>ETOH (USD/kg)</td>
<td>1.23 Steam (USD/t)</td>
<td>18.1 Marshall and Swift index (2003)</td>
</tr>
<tr>
<td>IPAC (USD/kg)</td>
<td>0.9 Electricity (USD/MWh)</td>
<td>43 Project life (years)</td>
</tr>
<tr>
<td>ETAC (USD/kg)</td>
<td>1.28 Light fuel oil (USD/t)</td>
<td>600</td>
</tr>
<tr>
<td></td>
<td>Water (USD/t)</td>
<td>0.042</td>
</tr>
</tbody>
</table>

**Table 6.2** Data used in the economic assessment.
6.4 Results and discussion

6.4.1 Results of the life cycle assessment

The calculation of the impact indicators related to a one hour operation of the waste solvent treatment alternatives requires the determination of the specific impact indicators discussed above. This means that emission inventories shown in Appendix C are used to form LCIs of the treatment alternatives (material and energy streams have to be related to the functional unit). In a consecutive step, the LCI data can be evaluated by EI-99 indicators. There results are shown in Table 6.3. The impact indicators of investment of the distillation plants are divided into two groups according to the two separation schemes selected: the first value stands for the EHAD scheme and the second one for the ternary cut scheme.

<table>
<thead>
<tr>
<th>Incineration</th>
<th>( i_{\text{production}} ) [pt/kg ws]</th>
<th>( i_{\text{basic, inc}} ) [pt/kg ws]</th>
<th>( i_{\text{investment, inc}} ) [pt/kg ws]</th>
<th>( i_{\text{C-CO}_2} ) [pt/kg]</th>
<th>( i_{\text{gain}} ) [pt/MJ]</th>
<th>( i_{\text{oil}} ) [pt/kg oil]</th>
</tr>
</thead>
<tbody>
<tr>
<td>EtOH</td>
<td>0.241</td>
<td>0.0253</td>
<td>0.166</td>
<td>0.0054</td>
<td>0.0177</td>
<td>0.129</td>
</tr>
<tr>
<td>ETAC</td>
<td>0.289</td>
<td></td>
<td></td>
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<tr>
<td>IPAC</td>
<td>0.292</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>water</td>
<td>0.0000302</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Recovery</td>
<td>( i_{\text{ws mix, production}} ) [pt/kg ws]</td>
<td>( i_{\text{basic, dist}} ) [pt/kg ws]</td>
<td>( i_{\text{investment, dist}} ) [pt/kg ws]</td>
<td>( i_{\text{steam, external}} ) [pt/kg steam]</td>
<td>( i_{\text{cooling water}} ) [pt/kg water]</td>
<td>( i_{\text{inc}} ) [pt/kg ws]</td>
</tr>
<tr>
<td>EHAD</td>
<td>0.214</td>
<td>1.22 \times 10^{-6}</td>
<td>0.0518</td>
<td>0.0184</td>
<td>3.32 \times 10^{-5}</td>
<td>0.131</td>
</tr>
<tr>
<td>Tern. cut</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
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</table>

Table 6.3 Specific impact factors applied by the calculation of the total environmental impacts.

Abbreviations: pt: EI-99 points; ws: waste solvent

First the Eco-indicator points for the total incineration and total recovery are determined using Equations (6.1)-(6.14). The different elements of the environmental contributions and the total impacts are shown in Table 6.4.

<table>
<thead>
<tr>
<th>Incineration</th>
<th>( I_{\text{solvent, production}} )</th>
<th>( I_{\text{basic, incineration}} )</th>
<th>( I_{\text{investment, inc}} )</th>
<th>( I_{\text{C-CO}_2} )</th>
<th>( I_{\text{oil, support}} )</th>
<th>( I_{\text{gain}} )</th>
<th>Total</th>
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<tr>
<td></td>
<td>913</td>
<td>108</td>
<td>25</td>
<td>11</td>
<td>51</td>
<td>-477</td>
<td>632</td>
</tr>
<tr>
<td>Recovery</td>
<td>( I_{\text{solvent, ws mix}} )</td>
<td>( I_{\text{basic distillation}} )</td>
<td>( I_{\text{investment, dist}} )</td>
<td>( I_{\text{residue}} )</td>
<td>( I_{\text{operation}} )</td>
<td>( I_{\text{recovered ws}} )</td>
<td>Total</td>
</tr>
<tr>
<td>Tern. cut scheme</td>
<td>913</td>
<td>0.01</td>
<td>17</td>
<td>29</td>
<td>568</td>
<td>-868</td>
<td>661</td>
</tr>
<tr>
<td>EHAD scheme</td>
<td>913</td>
<td>0.01</td>
<td>8</td>
<td>29</td>
<td>256</td>
<td>-868</td>
<td>338</td>
</tr>
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</table>

Table 6.4 LCA results of the total recovery and total incineration of the waste solvents referring to the functional unit. (EI-99 point/hour).

Due to the evaluation using EI-99, a ranking of different alternatives can be done. Solvent recovery with the EHAD scheme has the best environmental performance; recovery with the ternary cut scheme and incineration with heat utilization have environmental burdens of the similar magnitude.

Moreover, the sources of the environmental impacts with the highest contributions can also be identified. Most environmental impacts in this system are related to crude oil.
manufacturing and usage (also including fresh solvent production). However, environmental impacts due to the emissions of the incineration are also high.

Additionally, simultaneous treatment of the waste solvent mixture with both recovery and incineration is considered.

The total environmental impact referring to the treatment of the waste solvent mixture during one hour is the superposition of the impacts caused by the two processes: incineration and recovery. Figure 6.5 shows the total impact and the contributions of the different treatment options to the total value if simultaneous incineration and recovery with ternary cut scheme are assumed.

![Environmental impacts of the waste solvent treatment](image)

**Figure 6.5** Environmental impacts of simultaneously working recovery, applying ternary cut scheme and incineration.

It is clearly shown that with a low fraction of solvent recovery, incineration covers the steam demand of recovery; therefore the environmental impacts created by recovery are low. Meanwhile, the environmental impacts of incineration are mainly determined by the solvent make-up production which is balanced with the surplus avoided products (steam sent to the grid). If the fraction of solvent recovery exceeds 0.5, steam production of the incineration does not cover the steam demand of recovery. The required steam is produced by the incineration of light fuel oil which strongly increases the environmental impacts.

The environmental impacts of the combustion of fuel oil are considered for regeneration since the steam is used there. Therefore, if the fraction of solvent recovery exceeds 0.5, the environmental impacts of the recovery increase rapidly since it includes the impacts of light fuel oil combustion. The total impact of waste solvent treatment with simultaneously working recovery (ternary cut scheme and incineration) is minimal if total incineration is undertaken.

Figure 6.6 shows the environmental impacts caused by simultaneously working incineration and recovery with the EHAD scheme. Incineration covers the steam demand of the recovery if at least 30% of the waste solvent mixture is sent to incineration and the rest is recovered. If more fractions are sent to recovery than the 70% of the waste solvent mixture, the combustion of light fuel oil is required which increases the environmental impacts of the recovery.

The EHAD scheme is a more effective recovery alternative and has lower heat energy demand. According to this, total environmental impacts show that the most attractive solution
is total recovery of the waste solvent mixture with distillation through application of EHAD. Moreover, the results show that environmental impacts cannot be reduced by the division of the waste solvent mixture between the two treatment plants.

**Figure 6.6** Environmental impacts of the simultaneously working recovery applying EHAD scheme and incineration.
6.4.2 Economic analysis

Selection between the waste treatment options is generally carried out on the basis of economic evaluation. Therefore, the environmental evaluation presented above is completed with an economic analysis. The annual cost of operations (including investment costs) is calculated and determined for each treatment alternative. Table 6.5 shows the results of the economic evaluation.

Recovery is significantly better from an economic viewpoint than end-of-pipe treatment due to high solvent make-up costs. If the waste solvent treatment is carried out by recovery with the EHAD scheme the total cost can be reduced by almost 90% compared to the end-of-pipe treatment. Based on the economic evaluation, the recommended waste solvent treatment option is total recovery with the EHAD scheme followed by recovery with the ternary cut method and the least attractive option is end-of-pipe treatment with incineration.

<table>
<thead>
<tr>
<th></th>
<th>Total annual cost [1,000 USD/year]</th>
<th>Relative value [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incineration</td>
<td>26,400</td>
<td>100</td>
</tr>
<tr>
<td>Recovery with tern. Cut</td>
<td>6,430</td>
<td>24</td>
</tr>
<tr>
<td>Recovery with EHAD</td>
<td>2,860</td>
<td>11</td>
</tr>
</tbody>
</table>

Table 6.5 Total annual costs of the waste solvent treatment options.

6.5 Comparison of results

If economic evaluation and environmental impact assessment are compared, the environmental impact assessment shows that the most preferable solution for the treatment of the investigated waste solvent mixture is recovery with the EHAD method, followed by incineration and the least attractive solution from an environmental point of view is recovery with the ternary cut scheme, see Table 6.6.

Economic assessment does not give the same results; since from an economic point of view, recovery with the ternary cut scheme is significantly better than incineration - see Table 6.6. The difference can mainly be explained as being due to the relative high prices of the pure solvent components and low prices of fuel oil (providing cheap energy for recovery); prices which do not express the real environmental impacts of their industrial production.

<table>
<thead>
<tr>
<th></th>
<th>Total annual cost [1,000 USD/year]</th>
<th>Environmental impacts [EI-99 point/hour]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Incineration</td>
<td>26,400</td>
<td>632</td>
</tr>
<tr>
<td>Recovery with ternary cut scheme</td>
<td>6,430</td>
<td>660</td>
</tr>
<tr>
<td>Regeneration with EHAD scheme</td>
<td>2,860</td>
<td>332</td>
</tr>
</tbody>
</table>

Table 6.6 Total annual cost and environmental impacts of the waste solvent treatment options.

This example shows that, since strategic decisions of companies and governments are usually made on the basis of economic calculations, such decisions can be misleading and,
according to the concept of sustainable development, it is highly desirable that total cost and environmental impacts which refer to the same process are reconciled with each other.

6.6 Conclusions

The results highlighted a contradiction between economic and environmental evaluation of treatment options.

Based on the economic evaluation, recovery of solvents is preferred to incineration; however, recovery with the ternary cut method shows higher environmental impacts than incineration. In this case, this difference emerges because of the relatively high prices of pure solvents in comparison to the price of light fuel oil. This conclusion raises economic and environmental management problems.

As a conclusion, it can be stated that economic evaluation for decision-making in the field of waste solvent treatment is not sufficient. Decision making should be also preceded by environmental evaluation. Economic and environmental evaluations agree in that the more efficient solvent recovery process (EHAD) is the most attractive treatment option. This can serve to motivate engineers to design more effective recovery processes and proves the importance of ‘green engineering’ which better suits the concepts of sustainability. However, according to economic evaluations, decision-making on environmental grounds is often unsupported; therefore, both the economic and environmental features of design alternatives should be considered when making decisions.
CHAPTER 7  MAJOR NEW RESULTS

7.1 INVESTIGATION OF ENVIRONMENTAL IMPACT ASSESSMENT METHODS CONSIDERING THEIR UNCERTAINTIES

Life Cycle Assessment (LCA) is the only standardized and widely accepted tool currently used to assess environmental loads of products and/or processes. The Life Cycle Impact Assessment (LCIA), as a part of LCA, is the scientific technique for the qualitative assessment of the potential environmental impacts of industrial systems and their associated products.

The single score impact indicators of two different environmental impact assessment tools are the Eco-indicator 99 (EI-99) method and the European Union’s CAFE CBA method. They are compared referring to the environmental impacts of certain air pollutants.

Thesis 1) Clear linear dependency is detected between the single score impact indicators. This indicates a similarity in the relative ranking features of the two impact assessment methods, and a strong connection and dependencies between them. It is revealed that the detected similarities in the relative environmental valuations of environmental impacts do not depend on the assumptions used in the calculations of the CAFE CBA marginal damage values.

Data uncertainties of the two impact assessment methods (EI-99 and CAFE CBA) are investigated. The population of impact indicators which express environmental impacts due to annual air pollution is generated by Monte Carlo simulations for five industrialized cities.

Thesis 2) It is detected that propagation of data uncertainties results in barely distinguishable and interpretable environmental impact indicator intervals at the 90% confidence level. On the other hand, strong correlation ($R^2$ higher than 0.95) is found between the most likely values (determined as overall mean values of the generated populations) of the impact indicator populations if the results of the two different LCIA tools are compared. The detected correlation between the two impact assessment tools might help and support the work in their areas mutually exploiting the merits of both methods.

Thesis 3) The most likely impact indicator values obtained from the uncertainty analysis and the aggregated single score impact indicators (without uncertainty data) are compared to the same environmental problem. It is determined that the difference between them is less than 10%. This allows the application of the single score indicators (best guesses in the case of EI-99, and marginal damage values in the case of CAFE CBA) in order to obtain clear environmental preferences that would not be possible if full spectrums of single scores’ uncertainties were included in the analysis.
Thesis 4) It is detected and demonstrated through a comprehensive case study referring to the environmental evaluation of the annual airborne emission inventory of an industrialized city that aggregated, single score impact indicators such as EI-99 can successfully be applied to identify and rank air pollutants and their sources according to their environmental loads.

7.2 APPLICATION OF LIFE CYCLE ASSESSMENT: AIR POLLUTION

A.) Environmentally-conscious process design: selection of the proper air pollution abatement technique

An environmental impact assessment according to the EI-99 method is prepared for three basically different flue gas desulphurization (FGD) processes: (1) intra-furnace sulphur removal during coal combustion with limestone addition, (2) FGD with wet-limestone scrubbing, and (3) regenerative copper oxide flue gas clean-up process.

Thesis 5) It is determined that the three FGD processes investigated can create less environmental impacts than the uncontrolled release of sulphur oxides into air. The reductions range between 80 and 92%.

The results show that intra-furnace limestone addition and the wet scrubbing processes, techniques using similar physical and chemical principles, have similar environmental indices; however, FGD with wet-limestone scrubbing is found to be slightly better from an environmental viewpoint.

The basis of the regenerative process is a sorption/reduction/oxidation cycle that has higher SO$_2$ removal efficiency than the two other processes. This higher efficiency results in significantly lower environmental impacts. This means that recovery and recycling of SO$_2$ is the most preferable option from an environmental viewpoint.

B.) Study of effectiveness of flue gas desulphurization at a regional scale

The effect of supplementary installed flue gas desulphurization (FGD) units at high capacity power plants on regional air pollution in the Carpathian Basin is investigated. The dispersion and accumulation of the SO$_2$ air pollutant are studied with the regional three-dimensional on-line atmosphere-chemistry model REMOTE. Changes in the SO$_2$ air pollution are investigated by parallel simulations in a case study, where the single modified parameter is SO$_2$ removal rate.

Thesis 6) It is found that FGD units operating with constant 90% SO$_2$ removal efficiency significantly reduce both the annual mean horizontal (by 79%) and vertical (by 66%) dispersion of emitted SO$_2$, as well as its transboundary transport.

Besides the efficiency of removal of SO$_2$, dispersion and accumulation also depend on seasonal weather conditions. During winter, dispersion and accumulation are by at least 25% higher than in other seasons. Due to this phenomenon, higher SO$_2$ removal efficiency is needed during winter to guarantee similar air quality to other seasons.
7.3 APPLICATION OF LIFE CYCLE ASSESSMENT: WASTE SOLVENT TREATMENT

Based on an industrial case study, environmental and economic evaluations of treatment alternatives are completed and compared for a non-ideal solvent mixture containing azeotropes to determine the preferable option. For the recovery of the solvent mixture, two different separation alternatives are evaluated: a less effective alternative and a novel design based on hybrid separation tools. The third investigated waste solvent treatment alternative is incineration with heat utilization.

Thesis 7)
Contradictions between environmental and economic evaluations are detected: economic appraisal clearly favours total recovery; however, an environmental evaluation shows that if a recovery process of low efficiency is applied, its environmental burden can be similar or even higher to that of incineration. This finding may motivate engineers to design more effective recovery processes and to reconsider the evaluation of process alternatives at the environmental decision-making stage.
PUBLICATIONS

Papers published in scientific journals


Conference papers


Oral lectures and presentations


Posters


REFERENCES


CAFE Programme, home page: http://europa.eu.int/comm/environment/air/cafe/


Clean air and transport, home page: http://ec.europa.eu/environment/air/index.htm


ExternE, home page: http://www.externe.info/


Rentz O.K., Güttling U.K., 2002a. Exemplary Investigation into the State of Practical Realisation of Integrated Environmental Protection with regard to Large Combustion
Plants in Germany, French-German Institute for Environmental Research University of Karlsruhe, Germany.
## NOMENCLATURE

### Abbreviations

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<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>ACFBC</td>
<td>Atmospheric Circulating Fluidized Bed Combustor</td>
</tr>
<tr>
<td>AHI</td>
<td>Atmosphere Hazard Index</td>
</tr>
<tr>
<td>BPEO</td>
<td>Best Practicable Environmental Option Assessment</td>
</tr>
<tr>
<td>CAFE</td>
<td>Clean Air for Europe Programme</td>
</tr>
<tr>
<td>CAFE CBA</td>
<td>Cost-benefit analysis carried out within the framework of the EU’s CAFE programme</td>
</tr>
<tr>
<td>CBA</td>
<td>Cost-benefit analysis</td>
</tr>
<tr>
<td>CI</td>
<td>Confidence Interval</td>
</tr>
<tr>
<td>CML</td>
<td>Institute of Environmental Sciences, Leiden University</td>
</tr>
<tr>
<td>CORINAIR</td>
<td>Co-ordination of Information on Air Emissions Programme</td>
</tr>
<tr>
<td>DALY</td>
<td>Disability-Adjusted Life Years</td>
</tr>
<tr>
<td>EAP</td>
<td>Environmental Action Programme</td>
</tr>
<tr>
<td>ECMWF</td>
<td>European Centre for Medium-Range Weather Forecast</td>
</tr>
<tr>
<td>EDIP</td>
<td>Environmental Design of Industrial Products</td>
</tr>
<tr>
<td>EDIP</td>
<td>Environmental Design of Industrial Products</td>
</tr>
<tr>
<td>EHAD</td>
<td>Extractive heterogeneous-azeotropic distillation</td>
</tr>
<tr>
<td>EHI</td>
<td>Environmental Hazard Index</td>
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<tr>
<td>EI-99</td>
<td>Eco-indicator 99 methodology</td>
</tr>
<tr>
<td>EMEP</td>
<td>Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe</td>
</tr>
<tr>
<td>EPS</td>
<td>Environmental Priority Strategies</td>
</tr>
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<td>EPS</td>
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<tr>
<td>EQ</td>
<td>Ecosystem Quality</td>
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<td>ERA</td>
<td>Environmental Risk Assessment</td>
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<td>ESP</td>
<td>Electrostatic Precipitator</td>
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<tr>
<td>EU</td>
<td>European Union</td>
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<td>FBC</td>
<td>Fluidized Bed Combustor</td>
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<td>FGD</td>
<td>Flue gas desulphurization</td>
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<td>HEO</td>
<td>Hungarian Energy Office</td>
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<tr>
<td>HH</td>
<td>Human Health</td>
</tr>
<tr>
<td>IEI</td>
<td>Integrated Environmental Index</td>
</tr>
<tr>
<td>ISO</td>
<td>International Standards Organization</td>
</tr>
<tr>
<td>JRC</td>
<td>Joint Research Centre</td>
</tr>
<tr>
<td>LCA</td>
<td>Life Cycle Assessment</td>
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<td>LCI</td>
<td>Life Cycle Inventory</td>
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<td>LCIA</td>
<td>Life Cycle Impact Assessment</td>
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<td>LEBS</td>
<td>Low Emission Boiler System</td>
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<tr>
<td>LLCA</td>
<td>Limited Life Cycle Assessment</td>
</tr>
<tr>
<td>MATCH</td>
<td>Multiscale Atmospheric Transport and Chemistry Model</td>
</tr>
<tr>
<td>MEN</td>
<td>Mass Exchange Network</td>
</tr>
<tr>
<td>MHP</td>
<td>Methylhydrogenperoxide</td>
</tr>
<tr>
<td>MOZART 4</td>
<td>Model for Ozone and Related Chemical Tracers</td>
</tr>
<tr>
<td>NMVOC</td>
<td>Non-Methane Volatile Organic Carbons</td>
</tr>
<tr>
<td>NOEC</td>
<td>No Observed Effect Concentration</td>
</tr>
<tr>
<td>PAA</td>
<td>Peroxyacetic acid</td>
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**Nomenclature**

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<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tr>
<td>PAF</td>
<td>Potentially Affected Fraction of species</td>
</tr>
<tr>
<td>PDF</td>
<td>Potentially Disappeared Fraction of species</td>
</tr>
<tr>
<td>R</td>
<td>Resources</td>
</tr>
<tr>
<td>RADM II</td>
<td>Gas-phase Chemistry Module Applied in REMOTE</td>
</tr>
<tr>
<td>REMO</td>
<td>Regional Atmospheric Circulation Model</td>
</tr>
<tr>
<td>REMOTE</td>
<td>Regional Model with Tracer Extension</td>
</tr>
<tr>
<td>SCR</td>
<td>Selective Catalytic Reduction of NOx</td>
</tr>
<tr>
<td>SNAP</td>
<td>Selected Nomenclature for Air Pollution</td>
</tr>
<tr>
<td>TRACI</td>
<td>Tool for the Reduction and Assessment of Chemical Impacts</td>
</tr>
<tr>
<td>UN ECE</td>
<td>United Nations Economic Commission for Europe</td>
</tr>
<tr>
<td>UNEP</td>
<td>United Nations Environmental Programme</td>
</tr>
<tr>
<td>WHO</td>
<td>World Health Organization</td>
</tr>
<tr>
<td>ws</td>
<td>waste solvent</td>
</tr>
<tr>
<td>YLD</td>
<td>Years Lived Disabled</td>
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<tr>
<td>YLL</td>
<td>Year of Life Lost</td>
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**Symbols**

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<tr>
<td>$D$</td>
<td>normalised impact indicator, [-]</td>
</tr>
<tr>
<td>$\delta$</td>
<td>weighting factor, [-]</td>
</tr>
<tr>
<td>$i$</td>
<td>eco-indicator [EI-99 point]</td>
</tr>
<tr>
<td>$I$</td>
<td>Environmental impacts related to the functional unit [EI-99 point/functional unit]</td>
</tr>
<tr>
<td>$\dot{m}$</td>
<td>mass flow [kg/h] or emission [ton/year]</td>
</tr>
<tr>
<td>$\mu_L$</td>
<td>mean of ln(X) assuming normal distribution</td>
</tr>
<tr>
<td>$\sigma^2_g$</td>
<td>squared geometric standard deviation</td>
</tr>
<tr>
<td>$\sigma_L$</td>
<td>standard deviation of ln(X) assuming normal distribution</td>
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**Subscript**

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<tr>
<td>$i$</td>
<td>solvent component</td>
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<tr>
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<td>pollutant type</td>
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**Superscript**

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<th>Description</th>
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<tbody>
<tr>
<td>MC</td>
<td>Monte Carlo Simulation</td>
</tr>
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</tbody>
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APPENDIX A  LOG-NORMAL DISTRIBUTION

In probability and statistics, the log-normal distribution is the single-tailed probability distribution of any random variable whose logarithm is normally distributed. According to this,

the $X$ random variable is log-normally distributed with $\mu$ and $\sigma^2$ parameters,

if $Y = \log(X)$ random variable is normally distributed with $\mu_L$ and $\sigma_L^2$ parameters ($X \in \mathbb{R}^+$).

**Probability density function**

$$f(t, \mu_L, \sigma_L) = \begin{cases} 0 & \text{if } t \leq 0 \\ \frac{1}{t\sigma_L\sqrt{2\pi}} \exp\left(-\frac{(\log t - \mu_L)^2}{2\sigma_L^2}\right) & \text{if } t > 0 \end{cases}$$

**Cumulative distribution function**

$$F(x) = \begin{cases} 0 & \text{if } x \leq 0 \\ \frac{1}{\sigma_L\sqrt{2\pi}} \int_{-\infty}^{x} \exp\left(-\frac{(t - \mu_L)^2}{2\sigma_L^2}\right) dt & \text{if } x > 0 \end{cases}$$

**Expected value (mean)**

$$\mu = \exp\left(\mu_L + \frac{\sigma_L^2}{2}\right)$$

**Median**

$$F(\mu_c) = 0.5$$

$$\mu_c = \exp(\mu_L)$$

**Mode**

$$f(\mu_m) = \max$$

$$\mu_m = \exp(\mu_L - \sigma_L^2)$$

**Variance**

$$\sigma^2 = (\exp(\sigma_L^2) - 1) \exp\left(2\mu_L + \sigma_L^2\right)$$

**Geometric mean and geometric standard deviation**

The geometric mean of the log-normal distribution is $\exp(\mu_L)$, and the geometric standard deviation is equal to $\exp(\sigma_L)$.

If a sample of data is determined to come from a log-normally population, the geometric mean and geometric standard deviation may be used to estimate confidence intervals similar to the way the arithmetic mean and standard deviation are used to estimate confidence intervals for a normally distributed sample of data.
Confidence interval bounds | Log space | Geometric space
--- | --- | ---
3σ lower bound | $\mu_L - 3\sigma_L$ | $\mu_{\text{geo}} / \sigma_{\text{geo}}^3$
2σ lower bound | $\mu_L - 2\sigma_L$ | $\mu_{\text{geo}} / \sigma_{\text{geo}}^2$
1σ lower bound | $\mu_L - \sigma_L$ | $\mu_{\text{geo}} / \sigma_{\text{geo}}$
1σ upper bound | $\mu_L + \sigma_L$ | $\mu_{\text{geo}} \cdot \sigma_{\text{geo}}$
2σ upper bound | $\mu_L + 2\sigma_L$ | $\mu_{\text{geo}} \cdot \sigma_{\text{geo}}^2$
3σ upper bound | $\mu_L + 3\sigma_L$ | $\mu_{\text{geo}} \cdot \sigma_{\text{geo}}^3$

Table A1 Determination of confidence interval bounds for log-normal distribution.

Remarks
- $\mu_L$ and $\sigma_L^2$ are the expected value and standard deviation of the $Y = \log(X)$ random variable, while $\mu$ and $\sigma$ refer to the $X$ random variable.
- Figure A1 shows the probability density function of log-normally distributed random variables generated with the parameters $\mu_L = \ln(0.5)$ and $\sigma_L = 0.5$. The figure shows also the mean, median and mode values of the variables as well as the $\pm 2\sigma_L$ interval which is approximately equivalent to the 95% confidence interval.

![Probability density function](image)

Figure A1 Probability density function of a log-normally distributed sample.

APPENDIX B  COST-BENEFIT ANALYSIS

Cost-benefit analysis is an economic tool for determining whether or not the benefits of an investment or policy exceed its costs.

The tool has a very broad scope and aims at expressing all positive and negative effects of an activity in a common unit, namely money, from a social point of view. Usually whole production and consumption systems are examined. Thus, in a world of perfect markets, costs and benefits would indicate to any decision-maker every relevant information for economic welfare. Economic and environmental elements are likewise expressed in monetary values – as far as possible and depending on the level of details. In terms of methodological steps CBA involves first of all a determination of which costs and benefits are examined, then tries to identify these costs and benefits and finally weights them against each other (Wrisberg at al., 2002). The concept of cost-benefit analysis is shown in Figure B1.

![Figure B1](image-url)  The concept of cost-benefit analysis.
## APPENDIX C  I/O DATABASES OF WASTE SOLVENT TREATMENT ALTERNATIVES

<table>
<thead>
<tr>
<th>Input</th>
<th>Unit</th>
<th>Quantity</th>
<th>Emission to air</th>
<th>Unit</th>
<th>Quantity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste solvents</td>
<td>t</td>
<td>33,350</td>
<td>Stack gases</td>
<td>Nm$^3$</td>
<td>280*10$^6$</td>
</tr>
<tr>
<td>Light fuel oil</td>
<td>t</td>
<td>3,735</td>
<td>CO$_2$</td>
<td>t</td>
<td>66,000</td>
</tr>
<tr>
<td>Natural gas</td>
<td>Nm$^3$</td>
<td>47,000</td>
<td>NO$_x$</td>
<td>t</td>
<td>14</td>
</tr>
<tr>
<td>Electricity</td>
<td>MWh</td>
<td>10,700</td>
<td>SO$_2$</td>
<td>t</td>
<td>1</td>
</tr>
<tr>
<td>Water</td>
<td>t</td>
<td>325,000</td>
<td>dust</td>
<td>t</td>
<td>1.1</td>
</tr>
<tr>
<td>Process water</td>
<td>m$^3$</td>
<td>290,000</td>
<td>NH$_3$</td>
<td>t</td>
<td>0.8</td>
</tr>
<tr>
<td>Exhaust gas</td>
<td>Nm$^3$</td>
<td>1.25*10$^6$</td>
<td>CO</td>
<td>t</td>
<td>0.3</td>
</tr>
<tr>
<td>NaOH (30%)</td>
<td>t</td>
<td>5680</td>
<td>HCl</td>
<td>t</td>
<td>0.4</td>
</tr>
<tr>
<td>HCl (32%)</td>
<td>t</td>
<td>290</td>
<td>HBr</td>
<td>t</td>
<td>0.05</td>
</tr>
<tr>
<td>Sulphur suspension</td>
<td>t</td>
<td>4</td>
<td>HI</td>
<td>t</td>
<td>0.002</td>
</tr>
<tr>
<td>NaCl</td>
<td>t</td>
<td>5</td>
<td>Ni</td>
<td>t</td>
<td>8.5*10$^{-5}$</td>
</tr>
<tr>
<td>Na$_3$P</td>
<td>t</td>
<td>0.07</td>
<td>Cu</td>
<td>t</td>
<td>1.2*10$^{-4}$</td>
</tr>
<tr>
<td>H$_2$O$_2$ (35%)</td>
<td>t</td>
<td>3</td>
<td>Co</td>
<td>t</td>
<td>8.8*10$^{-5}$</td>
</tr>
<tr>
<td>NH$_4$OH (25%)</td>
<td>t</td>
<td>240</td>
<td>Zn</td>
<td>t</td>
<td>2.2*10$^{-3}$</td>
</tr>
<tr>
<td>NH$<em>4$OH$</em>{rec}$ (80%)</td>
<td>t</td>
<td>300</td>
<td>Fe</td>
<td>t</td>
<td>2.0*10$^{-3}$</td>
</tr>
<tr>
<td>CaCl$_2$ (77%)</td>
<td>t</td>
<td>170</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Polyelectrolyte</td>
<td>t</td>
<td>0.7</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Trimercaptotriazin</td>
<td>t</td>
<td>7</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>FeCl$_3$</td>
<td>t</td>
<td>2</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Emission to water</th>
<th>Unit</th>
<th>Quantity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste water</td>
<td>m$^3$</td>
<td>107,000</td>
</tr>
<tr>
<td>SO$_4^{2-}$</td>
<td>t</td>
<td>840</td>
</tr>
<tr>
<td>Cl$^-$</td>
<td>t</td>
<td>1,100</td>
</tr>
<tr>
<td>Br$^-$</td>
<td>t</td>
<td>110</td>
</tr>
<tr>
<td>I$^-$</td>
<td>t</td>
<td>3.8</td>
</tr>
<tr>
<td>F$^-$</td>
<td>t</td>
<td>1.5</td>
</tr>
<tr>
<td>Ni</td>
<td>t</td>
<td>0.04</td>
</tr>
<tr>
<td>Cu</td>
<td>t</td>
<td>0.02</td>
</tr>
<tr>
<td>Co</td>
<td>t</td>
<td>0.13</td>
</tr>
<tr>
<td>Zn</td>
<td>t</td>
<td>0.05</td>
</tr>
<tr>
<td>Fe</td>
<td>t</td>
<td>2.5</td>
</tr>
</tbody>
</table>

| Table C1 Input-output data of the investigated incinerator. |
### Table C2  Input-output data of the ternary cut scheme.

<table>
<thead>
<tr>
<th>Input</th>
<th>Unit</th>
<th>Quantity</th>
<th>Output</th>
<th>Unit</th>
<th>Quantity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste solvent</td>
<td>kmol/h</td>
<td>100</td>
<td>ETOH (95 wt%)</td>
<td>kg/h</td>
<td>1,353</td>
</tr>
<tr>
<td></td>
<td>kg/h</td>
<td>4,261</td>
<td>ETAC (99 wt%)</td>
<td>kg/h</td>
<td>1,107</td>
</tr>
<tr>
<td>ETOH</td>
<td>kg/h</td>
<td>1,317</td>
<td>IPAC (99 wt%)</td>
<td>kg/h</td>
<td>937</td>
</tr>
<tr>
<td>ETAC</td>
<td>kg/h</td>
<td>1,112</td>
<td>Waste water to</td>
<td>kg/h</td>
<td>4,462</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>treatment</td>
<td></td>
<td></td>
</tr>
<tr>
<td>IPAC</td>
<td>kg/h</td>
<td>942</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Water</td>
<td>kg/h</td>
<td>891</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Extracting agent (water)</td>
<td>kg/h</td>
<td>3,604</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reboiler duty</td>
<td>MJ/kg</td>
<td>19.7</td>
<td></td>
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</tr>
<tr>
<td>Cooling water demand</td>
<td>kg/kg</td>
<td>235</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of columns</td>
<td>piece</td>
<td>7</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of separators</td>
<td>piece</td>
<td>2</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Table C3  Input-output data of EHAD scheme.

<table>
<thead>
<tr>
<th>Input</th>
<th>Unit</th>
<th>Quantity</th>
<th>Output</th>
<th>Unit</th>
<th>Quantity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste solvent</td>
<td>kmol/h</td>
<td>100</td>
<td>ETOH (95 wt%)</td>
<td>kg/h</td>
<td>1,241</td>
</tr>
<tr>
<td></td>
<td>kg/h</td>
<td>4,261</td>
<td>ETAC (99 wt%)</td>
<td>kg/h</td>
<td>1,105</td>
</tr>
<tr>
<td>ETOH</td>
<td>kg/h</td>
<td>1,317</td>
<td>IPAC (99 wt%)</td>
<td>kg/h</td>
<td>939</td>
</tr>
<tr>
<td>ETAC</td>
<td>kg/h</td>
<td>1,112</td>
<td>Waste water to</td>
<td>kg/h</td>
<td>11,786</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>treatment</td>
<td></td>
<td></td>
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<tr>
<td>IPAC</td>
<td>kg/h</td>
<td>942</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>Water</td>
<td>kg/h</td>
<td>891</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Extracting agent (water)</td>
<td>kg/h</td>
<td>10,809</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reboiler duty</td>
<td>MJ/kg</td>
<td>8.8</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cooling water demand</td>
<td>kg/kg</td>
<td>105</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of columns</td>
<td>piece</td>
<td>4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of separators</td>
<td>piece</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
DECLARATION

The research work reported in this thesis has been carried out under the supervision of Prof. Dr. Peter Mízsey, Head of the Chemical and Environmental Process Engineering Department at Budapest University of Technology and Economics.

I declare that no portion of the work referred to in this thesis has been submitted in support of an application for another degree or qualification of this or any other university, or other institute of learning.

NYILATKOZAT

Alulírott Benkő Tamás kijelentem, hogy ezt a doktori értekezést magam készítettem és abban csak a megadott forrásokat használtam fel. Minden olyan részt, amelyet szó szerint, vagy azonos tartalomban, de átfogalmazva más forrásból átvettem, egyértelműen, a forrás megadásával megjelölttem.


.....................................

Tamás Benkő