LAPPEENRANTA UNIVERSITY OF TECHNOLOGY
LUT School of Engineering Science
Computational Engineering

Anastasia Pavlova

Carbon footprint of recycled paper

Examiners: Professor Ph.D. Tuomo Kauranne
D.Sc.(Tech.) Virpi Junttila

ABSTRACT

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Industrial production of pulp and paper is an intensive consumer of energy, natural resources, and chemicals that result in a big carbon footprint of the final product. At present companies and industries aspire to calculate their gas emissions into the atmosphere in order to afterwards reduce atmospheric contamination. One of the approaches allowing to increase carbon burden from the pulp and paper manufacture is paper recycling. The general purpose of the current paper is to establish methods of quantifying and minimizing the carbon footprint of paper. The first target of this research is to derive a mathematical relationship between virgin fibre requirements with respect to the amount of recycled paper used in the pulp. One more purpose is to establish a model to be used to clarify the contribution of recycling and transportation to decreasing carbon dioxide emissions. For this study sensitivity analysis is used to investigate the robustness of obtained results. The results of the present study show that an increasing of recycling rate does not always lead to minimizing the carbon footprint. Additionally, we derived that transportation of waste paper throughout distances longer than 5800 km has no sense because the use of that paper will only increase carbon dioxide emissions and it is better to reject recycling at all. Finally, we designed the model for organization of a new supply chain of paper product to a customer. The models were implemented as reusable MATLAB frameworks.

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Anastasia Pavlova
Lappeenranta, September, 2015.

Contents

List of Symbols and Abbreviations

CO_2	Carbon Dioxide
CH_4	Methane
GHG	Greenhouse Gas
CO_2eq	Carbon Dioxide equivalent
IPCC	the Intergovernmental Panel on Climate Change
GWP	Global-Warming Potential
LCA	Life Cycle Assessment
ISO	International Organization for Standardization
PAS	Publicly Available Specification
CEPI	Confederation of European Paper Industries
BAT	Best Available Techniques
EDTA	Ethylenediaminetetraacetic Acid
DTPA	Diethylene Triamine Pentaacetic Acid
ODE	Ordinary Differential Equation
N	Nitrogen
P_2O_5	Phosphorus Pentoxide
K_2O	Potassium Oxide
CO	Carbon Monoxide
NaOH	Sodium Hydroxide
O_2	Oxygen Gas
$NaClO_3$	Sodium Chlorate
SO_2	Sulfuric Dioxide
H_2O_2	Hydrogen Peroxide
O_3	Ozone
$MgSO_4$	Magnesium Sulfate
CaO	Calcium Oxide (quicklime)
$Na_2S_2O_4$	Sodium Dithionite
H_2SO_4	Sulphuric Acid
RPU	Recovered Paper Use

CF	Carbon footprint
km	Kilometer - 1000 meters - unit of length
GJ	Gigajoule - 10^9 joule - unit of energy
kWh	Kilowatt hour - 1000 watt-hours - unit of energy
EF	Emission Factor
OAT	One-At-A-Time approach of sensitivity analysis

1 INTRODUCTION

Industrial production of pulp and paper is an intensive consumer of energy (fossil fuels, electricity), natural resources (wood, water) and chemicals, that results in a large carbon footprint of the final product. Paper mills use principally wood pulp combined with pulp from recycled paper for production of paper. Paper recycling is a process of recovering waste paper and remaking it into new paper products. Nowadays it is well known that the production of one tonne of paper from discarded "waste" paper uses much less energy and water in comparison with a production of a tonne of paper from virgin wood pulp. It conduces to less air and water pollution, reduces solid waste going to landfills, and decreases the number of trees supposed to be cut.

When the cost efficiency of producing fibre based products - such as pulp, paper, cardboard or bioenergy - is analyzed, the tallying is most often based on transport costs, bulk masses and the value of both raw materials and corresponding products. However, in modern production plans also environmental costs and values play an increasingly big role. Environmental cost can include things such as the carbon footprint and the cost of corresponding emission rights, but also positive brand value associated with sustainable production processes. With the political driver of battling climate change, the importance of environmental cost factors is assuming an ever-larger role in profit calculus.

A concrete example of such a cost factor is the cost of the carbon footprint of production through the entire logistic cycle. There is substantial political pressure in Europe and elsewhere to increase the current price of an emitted tonne of carbon dioxide. This can weight heavily as an additional cost on production, but it can also be turned into a competitive advantage.

So, what is a carbon footprint exactly? It is a measure of the total amount of carbon dioxide (CO_2) and methane (CH_4) emissions of a defined population, system or activity, considering all relevant sources, sinks and storage within the spatial and temporal boundary of the population, system or activity of interest [1]. In other words, a term "carbon footprint" is commonly used to describe the total amount of greenhouse gas (GHG) emissions caused by an organization, event, product or person. In order to calculate carbon footprint, emissions from other than CO_2 gases (GHG) are normalized to the mass of CO_2 that is converted to the measurement of carbon dioxide equivalents (CO_2eq) using the Intergovernmental Panel on Climate Change (IPCC) 100-year Global Warming Potential (GWP) factors [2]. This allows to have one common unit for reporting results.

Greenhouse gases affect climate change, which is considered to be one of the gravest problems facing humanity. It is worth mentioning that many research reports have linked these emissions to global warming, displacement of the forest boundaries and natural habitat of animals, species extinction, floods, and spreading deadly diseases [3].

Nowadays, there are many types of paper, ranging from printing paper, packaging papers, tissue, newspapers, and boards to specialty papers. As a matter of fact, different types of fibre based products are traditionally made out of different amounts of recycled and virgin fibres. Clearly, consumption of pulp and quantity of recycled paper in that pulp vary from mill to mill, that significantly influences the amount of natural resources used. Therefore, the first objective of the current study is to establish a mathematical relationship between virgin fibre requirement with respect to the amount of recycled paper used in the pulp.

As mentioned earlier, manufacturing paper on an industrialized scale has enormous effects on the environment and one of the main ways to diminish a carbon footprint is paper recycling. In addition, it is believed that higher recycling rates lead to a reduction of carbon footprint. However, the fundamental question is: how much pulp from the waste paper should be added for paper production so that the carbon footprint is minimized? Clarification of this issue is the second task of this work. In addition, the robustness of this optimum is investigated by sensitivity analysis.

One of the biggest carbon dioxide emission sources of the paper life cycle is transportation throughout the supply chain. Thereby, an organization of a new supply chain of manufactured paper to a consumer is one of the crucial problems. The quantity of emissions emitted by transportation depends on such factors as the mode of shipment, the mass of cargo, and distance. The last part of this research is devoted to a model, used to choose the logistic path so that the carbon footprint is minimum.

The major part of current work was implemented in MatLab, which is a numerical computing environment. In addition, computer algebra system Maple was employed for deriving equations from models and dealing with them by symbolic computation.

The structure of the thesis is as follows. The next section briefly goes through the theoretical background for the study: approaches for calculating a total carbon footprint, the specificity of pulp and paper making processes, recycling of waste paper. Section 3 moves on to the investigation of the model for a recycling process and establishing a relationship between a recycling rate and virgin pulp requirements. Section 4 covers the model of the total carbon footprint assessment and describes how this model can be used to estimate the optimal carbon footprint in terms of carbon dioxide emissions. In addition, this section investigates the influence of transportation distance on the optimal recycling rate and implementation of the transportation model in order to find the optimal route for paper's supply chain. Finally, section 5 concludes and gives proposals for future work.

2 Background

2.1 Life cycle assessment

Life cycle assessment (LCA) is a technique which was developed to understand and address impacts associated with products, both manufactured and consumed throughout a product's life cycle [4]. It became internationally standardized by the International Organization for Standardization (ISO). The use of LCA is important for companies because it helps to ensure compliance with government regulations, to reshape a company strategy, and to decrease the environmental impact of a given product. In the current work we are interested in the product carbon footprint which is a globally accepted tool for assessment of the global warming potential of an organization, project or products, whereas LCA estimates multiple environmental impact categories. The carbon footprint is a subset of LCA because it focuses only on the climate change impact category [5]. The calculation of the carbon footprint is standardized by the specification PAS 2050 [6], where a method for accounting for the GHG emissions in the life cycle of goods and services is provided. The carbon footprint quantification is expressed in CO_2 equivalent (using the latest IPCC 100-year global warming potential (GWP) [6]) that is in a unit for expressing the irradiative forcing of a GHG to carbon dioxide which is has become a common indicator for environmental assessment [7].

Defined by ISO, there are two approaches of the LCA method: cradle-to-grave and cradle-to-gate. The first mentioned approach considers everything from harvesting materials to the disposal of the finished goods; the second one considers all activities starting with the extraction of materials from the earth (the cradle), their transportation, refining, processing and fabrication activities until the material or product is ready to leave the factory gate.

2.2 Functional unit and system boundary

This study is a cradle-to-gate LCA of the paper production. The function of the system under study is the production of paper and carbon dioxide emissions. The functional unit for wood products is 1 tonne of finished paper. The system boundary encompasses forestry and product manufacturing process, including inputs (logs, electricity, fuels) and transport to each production facility (Figure 1). Resources needed for the production of co-products are not considered.

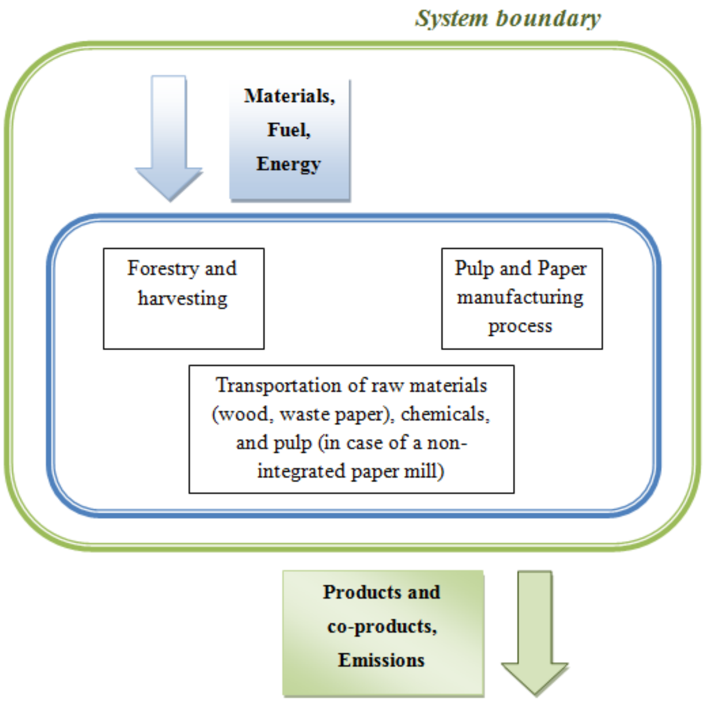

Figure 1: System boundary.

2.3 Pulping process from virgin fibres

Pulp and paper are produced in a mill which can be either non-integrated or integrated. A non-integrated installation means that pulp- and papermaking processes are undertaken in different places, i.e. the pulp is produced and dried in a pulp mill, and then the pulp is sold to a paper mill which manufactures paper. Integrated production means that pulp and paper are produced in the same place, i.e. a mill manufactures pulp and runs a paper machine to produce paper. The most commonly used such types of integrated mills as chemical pulp mills (kraft or sulphite pulp) with papermaking; mechanical pulping with papermaking; mills processing paper for recycling with papermaking; mixture of mechanical pulping and processing paper for recycling with papermaking; other mixtures, e.g. chemical pulp and paper for recycling can be used at the same site for the manufacture of a single product. [8]

According to Key Statistics of The Confederation of European Paper Industries (CEPI) [9] and The European Integrated Pollution Prevention and Control (IPPC)

[8], the kraft process is currently the most applied production method, which covers about 80% of the world pulp production. The sulphate pulping process has definite advantages over the sulphite process: superior pulp strength properties, application to all wood species, efficient chemical recovery system. However, in contrast to the sulphite process, unbleached kraft pulp has a considerably lower initial brightness than unbleached sulphite pulp. [8]

IPPC BAT 2001 [10] has a thorough description of applied processes and techniques of a kraft pulp mill. For the investigation, chemical kraft (sulphate) pulping technique and processing paper for recycling are selected. Therefore, we do not describe and discuss features of mechanical, sulphite or other pulping techniques we are not interested in. Storage of wood in a mill is not considered due to lack of information and a small carbon burden.

As denoted by IPPC BAT 2001 [10], the manufacture of paper consists of a number of stages such as an acquisition of wood; debarking, chipping and/or recycling; pulp preparation; paper formation; paper finishing. To begin the process, logs are passed through a debarker and then through a chipper. As a result, logs are transformed into uniform sized chips without bark. It is noted by writers of BAT 2001 [10] that "the more uniform the chips are after the chipper, the lower the raw material consumption". Next, the chips are screened in order to remove oversized chips and sawdust. Then the stage of delignification and cooking are carried out. The chips are placed in the cooking plant where such active chemicals as sodium hydroxide and sodium sulphide are added in order to liberate the fibres. The pulp coming from the digester contains both fibres and spent cooking liquor (black liquor) and approximately half of the wood is dissolved during the cooking process. Then, in the subsequent washing stages the black liquor, spent cooking chemicals, and organic substances are removed from the fibres. It is worth mentioning that modern systems usually recover at least 99% of the chemicals applied in the digester. After washing the pulp is screened to separate knots and fibre bundles. If a final paper product must be clean strong and white, it is necessary to remove all the lignin and impurities in the pulp by bleaching. Normally, chlorine dioxide, oxygen, ozone, alkali, and peroxide are used. Chlorine dioxide and ozone have to be produced on site; peroxide, oxygen and alkali can be delivered to mills. In order to take metal ions away to avoid degradation of the hydrogen peroxide metal chelating agents (i.e. EDTA or DTPA) or acid washing are added into the pulp. After bleaching the secondary screening takes place. Next, there are two options: for an integrated pulp and paper mill and for a non-integrated mill. In the first case, the finished pulp is transferred forward to papermaking in a wet state. For a non-integrated pulp mill the finished pulp is first pressed, then dried and, finally, cut into sheets bales for shipment to a paper mill.

2.4 Pulping process of recovered paper

Used paper is another significant source of fibre for the paper manufacturing industry. Paper recycling refers to the process of collecting used paper materials which are usually considered as "waste" and reprocessing them to be used as "raw materials" for the manufacture of new paper products. "So paper recycling is the process for making a paper used to the new material with the aim of preventing any waste that could be something useful, reducing the use of new raw materials, reduce energy use, reduce pollution, land and forest destruction, and greenhouse gas emissions compared to new manufacturing processes" [11]. In Europe, approximately 71.7% of consumed paper and paperboard was recycled in 2014 [9]. However, there is no type of paper production where a solely recycled paper is used. Wood fibres possess such property as a reduction in a length during the recycling process. It results in a loss of fibres' strength that in turn conduces to a poor quality of a final paper product. Therefore, a certain amount of primary wood fibres is mixed with the recycled fibres to maintain to fibre cycle and to ensure an appropriate strength of the paper. [8], [12]

Used paper and board are collected for recycling and delivered to collection yards where paper products are sorted and released from non-paper elements. Next, the paper for recycling is delivered to paper mills and manually sorted [8]. In order to disintegrate the paper into fibres, the paper is placed into a pulper with hot water and process water. When brightness and cleanliness are important, the resulting pulp is deinked with some chemicals such as sodium hydroxide and sodium silicate. After that, the fibres are cleaned the pulp is filtered and screened several times to reach required quality of pulp [10], [13]. Next, the pulp is often bleached by use of bleaching chemicals. Different types of fine screens and cleaners remove residual contaminants before the diluted pulp is fed to the paper machine. [8]

2.5 Papermaking process

In Europe, paper is usually made from a mixture of virgin pulp and pulp from recycled paper. Process steps differ considerably depending on the raw stock used and on the required quality of finished products. As a rule, the first step of a paper-making process is stock preparation. It removes impurities and air. Then different pulps together with water and chemicals are blended. Undissolved impurities are withdrawn from the slurry by screening and cleaning. Furthermore, the fibre suspension is cleaned in a centrifugal cleaner. To improve the strength of paper and to create the required properties of paper, refining is carried out. The last stock preparation's stage is plumping the pulp to the storage chests. After that, a series of prepared stocks and a required quantity of additives are mixed so that a desired grade of paper can be produced. The paper is formed in a paper machine. To make paper, the paper machine is supplied by a dilute suspension of fibres, fillers (optional), dyes and other chemicals. As a result, the machine gives a dry paper sheet. The higher an amount of recycled paper used in comparison with virgin wood fibres in the mixed pulp for paper production the lower the quality of finished paper [8].

3 Model of paper recycling

Nowadays paper mills use both recycled and virgin wood fibre in their production. There are economics and environmental reasons for adding recovered fibre to the virgin wood pulp such as reducing landfill waste, preventing water pollution, global warming, and saving energy. The first target of the present study is to establish a mathematical relationship between virgin fibre requirement and the amount of recycled paper in mixed pulp. In other words, the task is to ascertain the virgin fibre pulp requirement needed for a production of one tonne of paper, providing that recycling rate is known.

3.1 Discrete model of paper recycling

Schenk et al. [14] proposed a model of closed-system fibre recycling process, which describes paper manufacturing from virgin and recovered pulp. This model is used in order to derive a mathematical dependence between recycling rate and quantity of virgin fibre in the total amount of pulp needed for paper production. A description of the papermaking process model with four stocks is as follows (Figure 2). Virgin fibres enter the system through the flow Z into the stock S1. The stocks S$_i$, $i = 1, 2, 3, 4$, represent fibre stocks of different qualities in the system so that S1 contains the longest fibres and S4 contains the shortest ones. The arrows in Figure 2 represent flows F$_i$, $i = 1, 2, .., 8$. F1 through F4 represent nonrecycled fibre flows exiting the system. F5 through F8 represent damaged fibre flows. Z represents the new fibre requirement for the system. Because the system is closed, the total quantity of fibres in stocks has to be a constant r, which indicates the required quantity of mixed pulp for producing 1 tonne of new paper. Thereby, the input flow must compensate all output flows: Z = F1 + F2 + F3 + F4 + F8, and the value of Z in dynamic equilibrium is the model output [14]. The model can be expanded to any number of stocks. The number of stocks N symbolizes also how many times paper can be recycled.

At the first iteration, no recovered paper is used in a mill, so the new paper is made only from virgin fibres. At the second iteration, there are virgin fibres in S1 and nothing in S2 - SN. When the fibre is not recycled, it leaves the system through flow F1. The amount of fibres in each stock Si depends on recycling rate (or recovered paper use (RPU)) x, which is the use of recovered paper in a papermaking industry as a percentage of the total paper production in that sector [15]. Since x is recovered

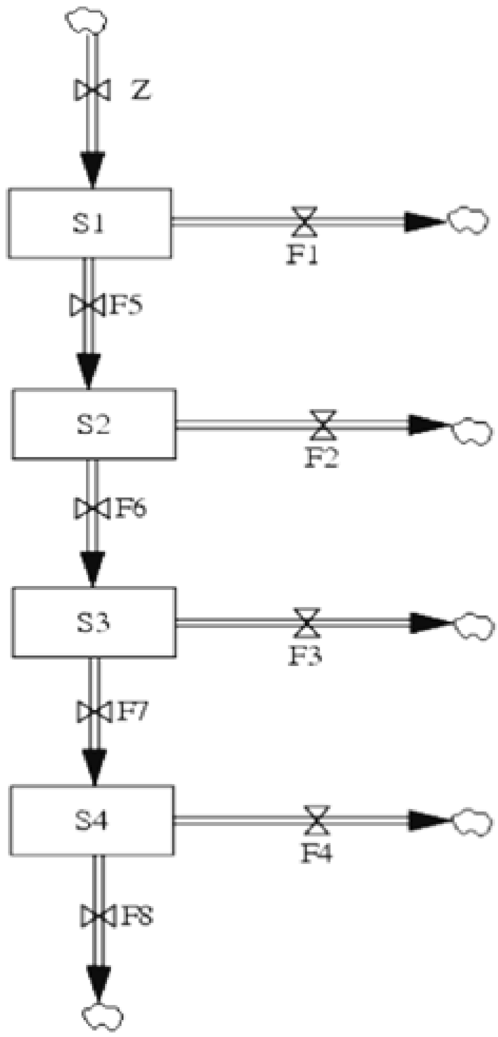

Figure 2: Closed-system fibre recycling model. Stocks S1 - S4 represent fibre stocks of different qualities in the system. Flows F1 - F4 represent nonrecycled fibre flows exiting the system. Flows F5 - F8 represent damaged fibre flows. Flow Z represents the new fibre requirement for the system.[14]

paper use, quantity x of paper goes back to the system (from a consumer) and part $(1 - x)$ leaves the system. In other words, from the second iteration to the end, a number of virgin fibres in each Si is multiplied by x. In the model, this action is represented by flows F1-F4. When the fibre is recycled, we assume the fibre to have a certain probability that it will be shortened during the repulping process, expressed in the damage rate y $(0 \leq y \leq 1)$. When the fibre is damaged, it leaves S1 and enters S2 through F5. When the fibre is recycled and not damaged it stays in S1 for use and, eventually, for further recycling. In the model some amount of fibres in each stock which is not shortened is multiplied by $(1 - y)$, and the other part of fibres is multiplied by y and goes to the next stock. The stocks S1 to S4 represent fibre stocks of different qualities in the system. Note that paper can contain fibres of mixed qualities. Fibres in S4 which are recycled but damaged are assumed to be disposed of in the washing stage of the recycling process.

The model can be run with chosen values of recycling rate x, damage rate y, and the total fibre stock r as a discrete evolution process. The graph in Figure 3 shows the evolution of the quantity of fibres in five stocks with given recycling rate x and damage rate y (Table 1). These quantities fluctuate at first time steps and after that the system stabilizes with time. In addition, it can be noticed that the quantity of fibres of the best quality is decreasing with respect to time and the quantity of fibres with worse quality is increasing. It is the result of the damaging process during the repulping process.

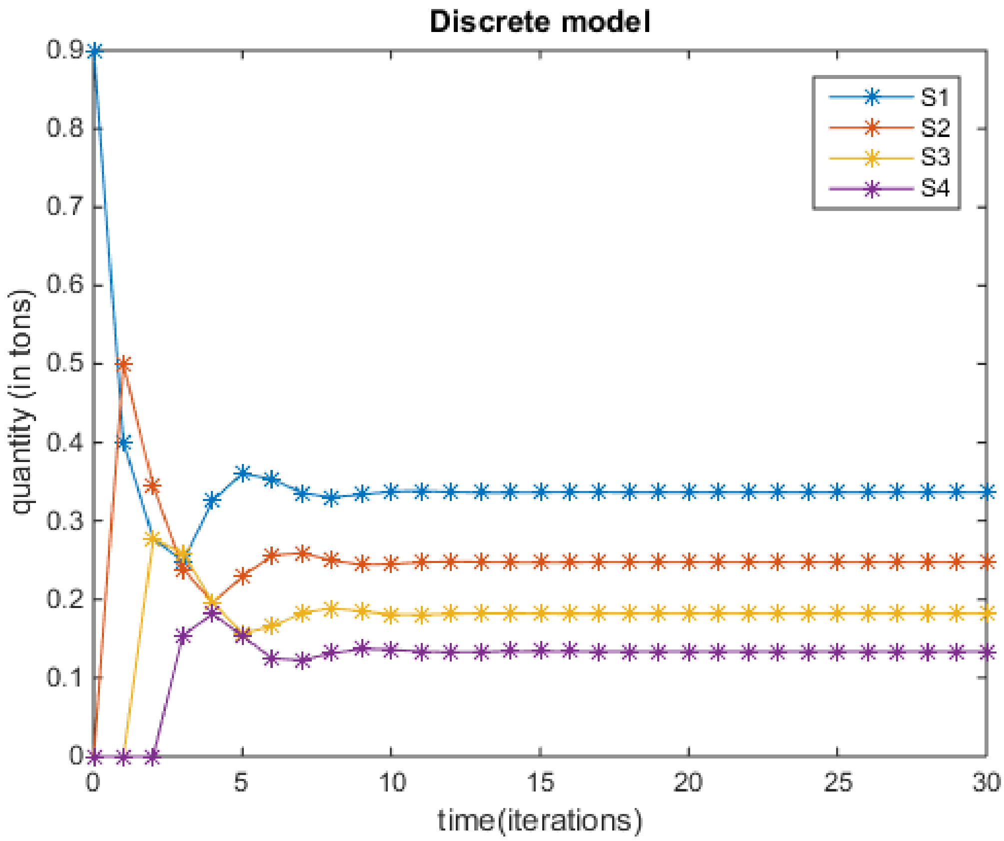

Figure 3: Discrete evolution of the number of fibres in stocks. N = 4.

The graph in Figure 4 shows the virgin fibre requirement at each time step, which evolves such that a dynamic equilibrium is finally achieved. The leap at the fifth iteration occurs because by this time all stocks have been filled by some amount of fibres and a considerable portion of fibres has been disposed of during the repulping process.

Recycling rate, x	Damage rate, y	Total fibre stock, r
1.1200	0.8534	0.9000

Table 1: Data for the discrete scheme of the paper recycling process.

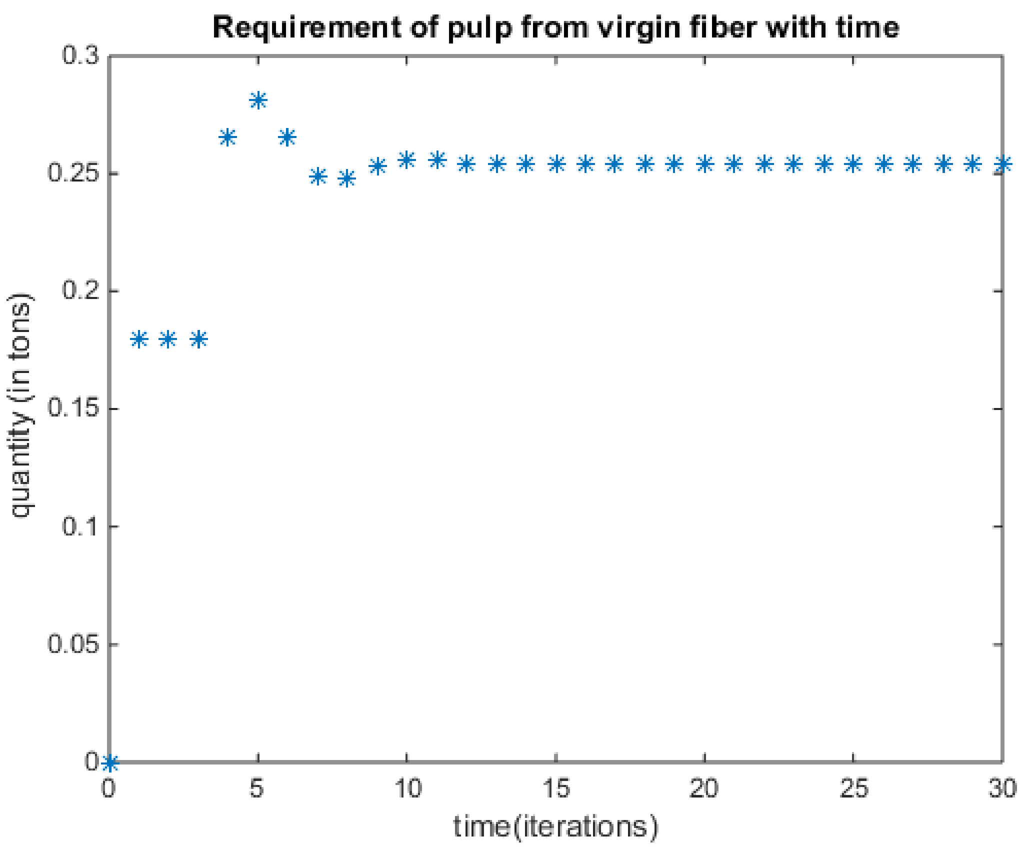

Figure 4: Requirements of pulp from virgin fibre with time.

Schenk et al. [14] derived an equation which represents a mathematical relationship between virgin fibre requirement and recycling rate. However, the equation does not fit well with virgin pulp requirements which can be obtained from the discrete evolution process. Therefore, we decided to find a more accurate equation for virgin pulp requirement by employing the theory of ordinary differential equations. Further, we illustrate the derivation of the system of differential equations in the case of four stocks (N = 4). It is worth mentioning that these equations can be expanded to any number of stocks.

3.2 Continuous model of paper recycling

According to the model (Figure 2), each flow F1, F2, F3, F4 represents the leaving of fibres out the system due to recycling rate x. These flows can be expressed by the following equations.

For the case when the number of stocks is four (N = 4):

$$\begin{cases} F_1 &= S_1(1-x), \\ F_2 &= S_2(1-x), \\ F_3 &= S_3(1-x), \\ F_4 &= S_4(1-x). \end{cases} \tag{1}$$

Flows F5, F6, F7, F8 represent the damaging process of fibres during the recycling process. As stated by the model, it is a multiplication of the present value in the stock by the damage rate y. Hence,

$$\begin{cases} F_5 &= S_1xy, \\ F_6 &= S_2xy, \\ F_7 &= S_3xy, \\ F_8 &= S_4xy. \end{cases} \tag{2}$$

The system of differential equations, explaining the model of the process, is

$$\begin{cases} \frac{dS_1}{dt} &= z(x,y) - F_1 - F_5, \\ \frac{dS_2}{dt} &= F_5 - F_2 - F_6, \\ \frac{dS_3}{dt} &= F_6 - F_3 - F_7, \\ \frac{dS_4}{dt} &= F_7 - F_4 - F_8. \end{cases} \tag{3}$$

As it was noted earlier, paper initially is made from only virgin pulp. This statement can be written as initial values for the ODE system:

$$S(0) = \begin{bmatrix} r & 0 & 0 & 0 \end{bmatrix}.$$

Since the model describes the closed system, the law of conservation of mass should hold:

$$\frac{dS_1}{dt} + \frac{dS_2}{dt} + \frac{dS_3}{dt} + \frac{dS_4}{dt} = 0. \tag{4}$$

The equation for virgin pulp requirement z can be derived in the following way: substitute (3) into (4) and, then, (1) and (2):

$$z - F_1 \underbrace{-F_5 + F_5}_{=0} - F_2 \underbrace{-F_6 + F_6}_{=0} - F_3 \underbrace{-F_7 + F_7}_{=0} - F_4 - F_8 = 0$$

$$\Rightarrow z - S_1(1-x) - S_2(1-x) - S_3(1-x) - S_4(1-x) - S_4xy = 0$$

$$\Rightarrow z(x,y) = (1-x)(S_1 + S_2 + S_3 + S_4) + S_4xy. \tag{5}$$

The system of the ordinary differential equations is established by substituting (5), (1), and (2) into the system (3):

$$\begin{cases} \frac{dS_1}{dt} &= (1-x)S_2 + (1-x)S_3 + (1-x)S_4 + S_4xy - S_1xy, \\ \frac{dS_2}{dt} &= S_1xy - (1-x)S_2 - S_2xy, \\ \frac{dS_3}{dt} &= S_2xy - (1-x)S_3 - S_3xy, \\ \frac{dS_4}{dt} &= S_3xy - (1-x)S_4 - S_4xy. \end{cases} \tag{6}$$

Now we have a continuous version of the model (6). It can be seen in the picture (Figure 5) that the models provide similar patterns in dynamics of stocks under the same conditions (x, y). Moreover, they converge to the same stable state. Consequently, we can conclude that the obtained system of equations is correct.

Next, we are going to solve the system of ODEs (6) in order to derive an equation for virgin pulp requirement. The system under consideration is linear. Therefore, we can rewrite it in matrix notations: $\frac{d\mathbf{s}}{dt} = \mathbf{A}\mathbf{s}$, $\mathbf{s} = (S1, S2, S3, S4)^{\mathrm{T}}$. Where $\mathbf{A}$ is a matrix form of the system of ODEs:

$$\mathbf{A} = \begin{bmatrix} -xy & 1-x & 1-x & xy - x + 1 \\ xy & -xy + x - 1 & 0 & 0 \\ 0 & xy & -xy + x - 1 & 0 \\ 0 & 0 & xy & -xy + x - 1 \end{bmatrix}.$$

It is well known from the theory of ordinary differential equations that the solution of the system of linear ODEs can be written as: $\mathbf{s} = \sum_{i=1}^{N} C_i e^{\lambda_i t} \bar{a}_i$, where λ_i is an eigenvalue associated with an eigenvector $\bar{a}_i$, and C_i is a constant.

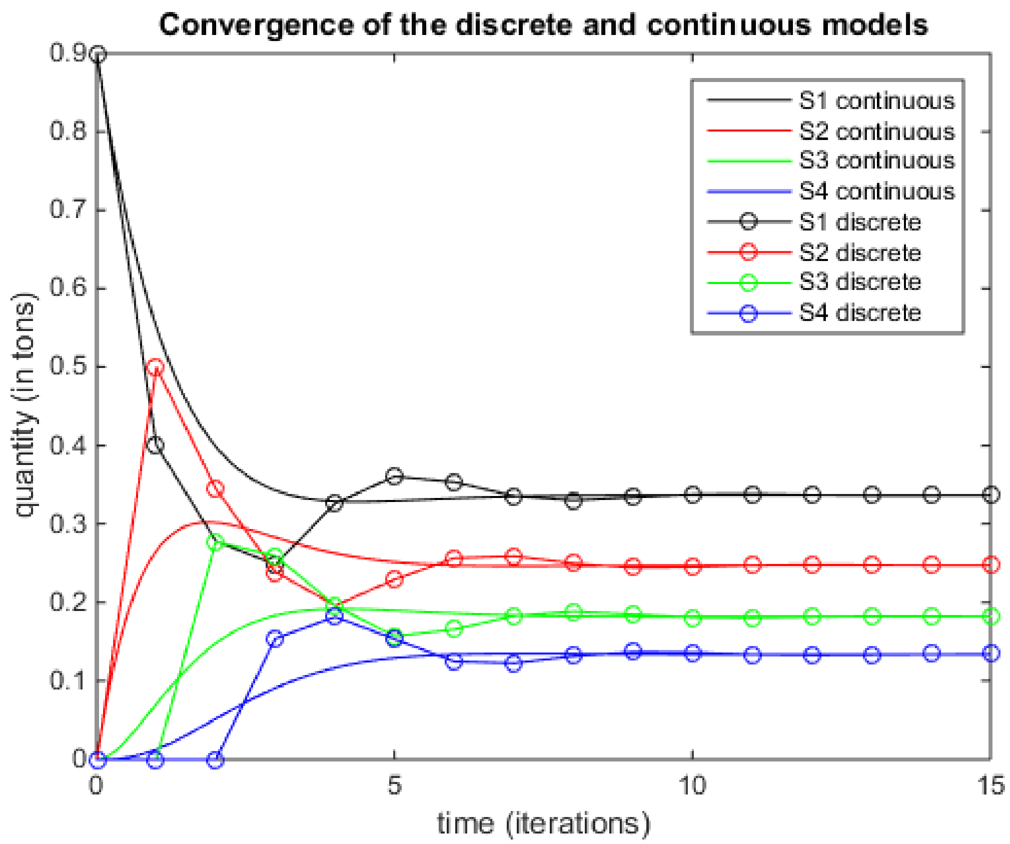

Figure 5: Convergence of the discrete and continuous models for the number of stocks N = 4.

Here

$$\lambda = \begin{pmatrix} -2xy + x - 1 \\ 0 \\ -xy + x - 1 + ixy \\ -xy + x - 1 - ixy \end{pmatrix}. \tag{7}$$

Four eigenvalues are obtained: two real and two complex. We can assume that all eigenvalues except the zero eigenvalue have negative real parts because $x, y > 0$. It is an appropriate assumption because, according to Schenk et al. [14], damage rate y tends to be more than 0.7 and it cannot exceed value 1 since damage rate is a probability. We assume also that the quantity of fibres from waste paper (x) lies in the range of 0 - 1.5 tonnes because it is impossible to add a negative amount of material into the pulp and adding more than 1.5 tonnes fibre is senseless since there are plenty of short fibres which will be disposed of during the papermaking process.

Hence,

$$-2xy + x - 1 \le -xy + x - 1 < 0.$$

As it was written earlier, the discrete model stabilizes with time. It means that, probably, there is a stable point for the system of ODEs. Now, when eigenvalues are known, we use the theorem to analyze the stability of the linear system of differential equations [16, 17].

Theorem: Stability of the Linear System
1. Every solution is stable if all the eigenvalues of $\mathbf{A}$ have negative real part.
2. Every solution is unstable if at least one eigenvalue of $\mathbf{A}$ has positive real part.
3. Suppose that the eigenvalues of $\mathbf{A}$ all have real parts that are zero or negative. List those eigenvalues with zero real part as $\lambda_j = i\beta_j$ for $1 \le j \le l$ where l is a number of eigenvalues with zero real part. Let the multiplicity of λ_j be m_j; that is, $p(\lambda) = (\lambda - \lambda_j)^{m_j} q(\lambda)$ where $q(\lambda_j) \ne 0$. Every solution is stable if $\mathbf{A}$ has m_j linearly independent eigenvectors for each λ_j. Otherwise, every solution is unstable.

The obtained eigenvalues satisfy the third condition of the stability theorem since the second eigenvalue has zero real part and, consequently, is always linearly independent; and other eigenvalues have a negative real part. Thus, we have proved that the system has one stable point. Next, we can derive it by letting t go to infinity.

In our case, all summands will tend to zero except the summand which relates to the zero eigenvalue because t goes to infinity. Therefore, the stable point of the solution of the system of differential equations is $\mathbf{s} = C_1 e^{0t} \bar{a}_1 = C_1 \bar{a}_1$, where $\bar{a}_1$ is the eigenvector associated with the zero eigenvalue, C_1 is a constant.

Next, the eigenvector and the constant can be calculated in accordance with the initial values. Finally, the solution is:

$$\begin{cases} S_1 &= \frac{r(xy+1-x)^3}{\sum\limits_{i=0}^{3}(xy+1-x)^{3-i}(xy)^i}, \\ S_2 &= \frac{r(xy+1-x)^2 xy}{\sum\limits_{i=0}^{3}(xy+1-x)^{3-i}(xy)^i}, \\ S_3 &= \frac{r(xy+1-x)(xy)^2}{\sum\limits_{i=0}^{3}(xy+1-x)^{3-i}(xy)^i}, \\ S_4 &= \frac{r(xy)^3}{\sum\limits_{i=0}^{3}(xy+1-x)^{3-i}(xy)^i}. \end{cases} \tag{8}$$

Return to the formula for finding virgin pulp requirement (5) and insert the derived formulas for stocks (8) into the equation:

$$z(x, y) = (1 - x)(S_1 + S_2 + S_3 + S_4) + S_4 xy.$$

For the case $N = 4$:

$$z(x, y) = \frac{r(1 + xy - x)^4}{\sum_{i=0}^{3} (1 + xy - x)^i (xy)^{3-i}},$$

z - virgin fibre pulp requirement (in tonnes),
x - recycling rate (recovered paper use) (constant),
y - damage rate (constant),
r - number of cellulose fibres in stock as a percentage of the amount of produced paper (in tonnes).

We have obtained the equation for virgin pulp requirement for a series of stocks: $N = 3, 4, 5, 6$ and have noticed that all these equations have a common pattern. Hence, we decided to prove that the general form of the equation is:

$$z(x, y) = \frac{r(1 + xy - x)^N}{\sum_{i=0}^{N-1} (1 + xy - x)^i (xy)^{N-1-i}}. \tag{9}$$

3.3 Derivation of the equation for an arbitrary number of stocks

In this section, we are going to prove that virgin pulp requirements for different numbers of stocks can be expressed as

$$z(x, y) = \frac{r(1 + xy - x)^N}{\sum_{i=0}^{N-1} (1 + xy - x)^i (xy)^{N-1-i}}.$$

Let us denote $xy = a$ and $1 - x = b$. Next, we write the matrix of the system of linear ODEs for N stocks:

$$\mathbf{A} = \begin{bmatrix} -a & b & b & \cdots & b & a+b \\ a & -b-a & 0 & 0 & \cdots & 0 \\ 0 & a & -b-a & 0 & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ 0 & \cdots & 0 & a & -b-a & 0 \\ 0 & \cdots & 0 & 0 & a & -b-a \end{bmatrix}. \tag{10}$$

This system (10) always has the zero eigenvalue because we require $\sum_{i=1}^{N} \frac{ds_i}{dt} = 0$.

Now we are going to find the eigenvector which relates to the zero eigenvalue. We may eliminate the first row in the matrix $\mathbf{A}$ in order to find the eigenvector because the multiplicity of the zero eigenvalue is one. The multiplicity is one because rows from 2 to N are linearly independent.

We have the matrix:

$$\hat{\mathbf{A}} = \begin{bmatrix} a & -b-a & 0 & 0 & \cdots & 0 \\ 0 & a & -b-a & 0 & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ 0 & \cdots & 0 & a & -b-a & 0 \\ 0 & \cdots & 0 & 0 & a & -b-a \end{bmatrix}.$$

Let us denote the eigenvector by

$$x = \begin{pmatrix} x_1 \\ \vdots \\ x_N \end{pmatrix}.$$

From the eigenvector definition we have to find the vector x so that $\hat{\mathbf{A}}x = 0_{N\times 1}$. From this equation we can derive:

$$\begin{cases} x_1 a - (b+a)x_2 & = 0, \\ x_2 a - (b+a)x_3 & = 0, \\ \vdots & \\ x_{N-1} a - (b+a)x_N & = 0. \end{cases}$$

$\Rightarrow$

$$\begin{cases} x_1 a & = x_2(b+a), \\ x_2 a & = x_3(b+a), \\ \vdots & \\ x_{N-1} a & = x_N(b+a). \end{cases}$$

$\Rightarrow$

$$\begin{cases} x_1 & = x_2 \frac{(b+a)}{a}, \\ x_2 & = x_3 \frac{(b+a)}{a}, \\ \vdots \\ x_{N-1} & = x_N \frac{(b+a)}{a}. \end{cases} \tag{11}$$

Next, we substitute the last obtained equation (11) in the previous one (x_{N-1} in x_{N-2}) and so on (the last pair is: x_2 in x_1):

$$\begin{cases} x_1 & = x_N \frac{(b+a)}{a}^{N-1}, \\ x_2 & = x_N \frac{(b+a)}{a}^{N-2}, \\ \vdots \\ x_{N-1} & = x_N \frac{(b+a)}{a}^{1}. \end{cases}$$

x_N is an arbitrary value. Let us assign $x_N = a^{N-1}$, then:

$$\begin{cases} x_1 & = a^{N-1} \frac{(b+a)}{a}^{N-1} = (a+b)^{N-1}, \\ x_2 & = a^{N-1} \frac{(b+a)}{a}^{N-2} = a(a+b)^{N-2}, \\ \vdots \\ x_{N-1} & = a^{N-1} \frac{(b+a)}{a}^{1} = a^{N-2}(a+b). \end{cases}$$

As a result, we get the eigenvector:

$$x = \begin{bmatrix} (a+b)^{N-1} \\ a(a+b)^{N-2} \\ a^2(a+b)^{N-3} \\ \vdots \\ a^{N-2}(a+b) \\ a^{N-1} \end{bmatrix}.$$

Under assumptions we have made that the other eigenvalues have negative real parts, the solution of the system is $s = C_1 e^{0t} x$ or in index notation $S_i = C_1 x_i$. Actually, all eigenvalues for a different number of stocks (N = 2,..,9) have been calculated and the assumption holds.

The next target is to estimate the constant C_1. We can use the property that the sum of all stocks should be equal to r:

$$S_1 + S_2 + \cdots + S_N = r$$

$\Rightarrow$

$$r = \sum_{i=1}^{N} S_i = C_1 \sum_{i=1}^{N} x_i \Rightarrow C_1 = \frac{r}{\sum\limits_{i=1}^{N} x_i} = \frac{r}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i}.$$

Then, we can write the formula for each component:

$$S_k = \frac{r(a+b)^{N-k} a^{k-1}}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i}. \tag{12}$$

Also, we remind the equation for virgin pulp requirements:

$$z(x,y) = (1-x)(S_1 + S_2 + \cdots + S_N) + S_N xy,$$

and rewrite it as:

$$z(a,b) = b(S_1 + S_2 + \cdots + S_N) + S_N a. \tag{13}$$

Substitute our equations for S_k (12) into $z(a,b)$ (13):

$$\begin{aligned}
z(a,b) &= \frac{r \sum\limits_{k=1}^{N} (a+b)^{N-k} a^{k-1}}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i} b + \frac{r a^{N-1}}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i} a = \\
&= \frac{r \sum\limits_{k=0}^{N-1} (a+b)^{N-1-k} a^{k}}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i} b + \frac{r a^{N-1}}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i} a = \\
&= \frac{r \left(\sum\limits_{k=0}^{N-1} (a+b)^{N-1-k} a^{k} \right) b + r a^N}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i} = r \frac{\left(\sum\limits_{k=0}^{N-1} (a+b)^{N-1-k} a^{k} \right) b + a^N}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i} = \\
&\underbrace{=}_{\text{Lemma 1}} r \frac{(a+b)^N}{\sum\limits_{i=0}^{N-1} (a+b)^{N-1-i} a^i}.
\end{aligned}$$

Lemma 1.

$$\left(\sum_{k=0}^{N-1} (a+b)^{N-1-k} a^k\right) b + a^N = (a+b)^N \tag{14}$$

holds for all positive natural numbers.

Proof. Mathematical induction will be used to prove the statement (14).
Basis: Show that the statement holds for $N = 1$.

$$\left(\sum_{k=0}^{0} (a+b)^{1-1-k} a^k\right) b + a = a + b.$$

The left-hand side of the equation is equal to the right-hand side of the equation. Thus, it has been shown that the equation is correct for $N = 1$.

Inductive step: Show that if the equation holds for N, then it also holds for $N + 1$.

We have to show that:

$$\left(\sum_{k=0}^{N} (a+b)^{N-k} a^k\right) b + a^{N+1} = (a+b)^{N+1}.$$

Using the induction hypothesis that the equation holds for N, the right-hand side can be rewritten as:

$$\begin{aligned}
(a+b)^{N+1} &= (a+b)^N (a+b) = \left(\left(\sum_{k=0}^{N-1} (a+b)^{N-1-k} a^k\right) b + a^N\right)(a+b) = \\
&= \left(\sum_{k=0}^{N-1} (a+b)^{N-1-k} a^k\right) b\,(a+b) + a^N (a+b) = \\
&= \left(\sum_{k=0}^{N-1} (a+b)^{N-k} a^k\right) b + a^N b + a^{N+1} = \\
&= \left(\sum_{k=0}^{N-1} (a+b)^{N-k} a^k + a^N\right) b + a^{N+1} = \left(\sum_{k=0}^{N} (a+b)^{N-k} a^k\right) b + a^{N+1}.
\end{aligned}$$

Thereby, we have shown that the equation holds for $N+1$. Since both the basis and the inductive step have been performed by mathematical induction, the statement holds for all positive natural numbers N. □

3.4 Application of the recycling model

The following part of the chapter is devoted to the application of the derived formula (9). It is assumed that the damage rate is specific for every mill. It means that it is a constant for a recycling process and does not depend on recycling rate. Therefore, we can estimate a damage rate for some known values of recycling rate and virgin pulp requirements by assuming different values for the number of stocks. In other words, in order to use the formula we have to know some information about paper production such as the recycling rate, the damage rate, the number of times that paper can be recycled and the total amount of pulp a mill needs to make one tonne of paper. Damage rate always lies in the interval (0 1] because it is a probability. It is worth mentioning that inappropriate guesses for the number of stocks result in an inappropriate damage rate (for example, more than one). Hence, we can calculate the maximal number of times that paper can be recycled.

Data for model launch was found in the literature. There are four required values: recovered paper use, virgin pulp use, and pulp requirements for manufacturing of one tonne of paper; 1.120 tonnes of waste paper and 0.113 tonnes of virgin pulp are used to produce 0.9 tonnes of mixed pulp, which is enough for manufacturing one tonne of new paper [10]. Next, we are able to determine a damage rate y by fitting the equation for virgin pulp requirement for different numbers of stocks. Results of estimating the unknown y are illustrated in the Table 2. According to the fact that damage rate y is a probability, it cannot be greater than 1, so the maximum number of stocks for the particular case is 5. As a matter of fact, since just one set of parameters (x, z, r) is used, we cannot conclude which number of stocks approximates the recycling model best.

Number of stocks, N	2	3	4	5	6	7
Damage rate, y	0.3760	0.537	0.6940	0.8534	1.0129	1.1725

Table 2: Damage rates depending on the number of stocks.

Next, it is possible to plot a virgin pulp requirement for a given damage rate y and a total fibre stock r in case of five stocks, see Figure 6. It becomes visible that the relationship between a recycling rate x and a virgin fibre requirement z is nonlinear rather than linear (in a theoretic, dynamic equilibrium situation). The dotted line in the Figure 6 represents a fictive situation with no fibre damage during the recycling process. It can be seen that for a small amount of recovered paper

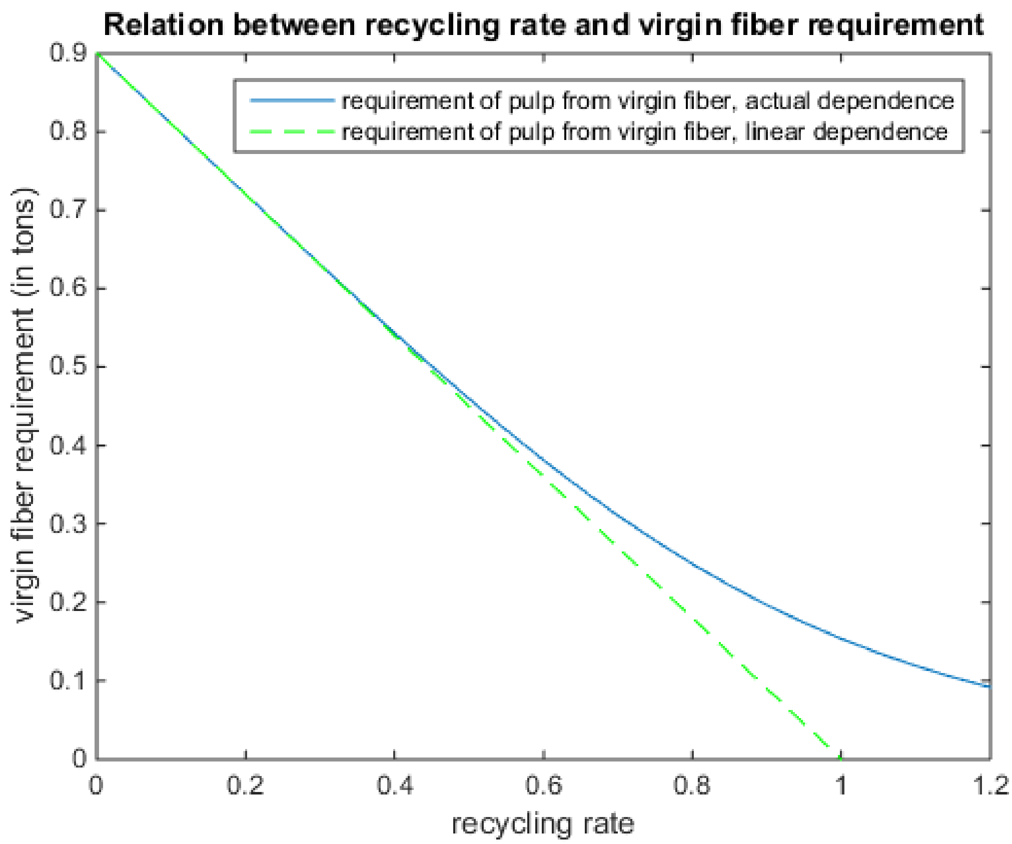

Figure 6: Relationship between a recycling rate and a virgin fibre requirement. If the recycling rate is known, it is possible to find out how much pulp from virgin material one has to add in order to obtain one tonne of new paper.

in the recipe the relationship appears to be approximately linear. However, with increasing recovered paper use the relationship starts to deviate from the dotted line and becomes nonlinear. It proves the fact that new paper cannot be produced without a permanent input of virgin fibres to the system.

In the Figure 7 graphs of virgin pulp requirements for a different number of stocks are depicted. As we can see, virgin pulp requirements differ from each other but not so much. It can be noticed that curvature of lines increases with the number of stocks. Therefore, we can conclude that the nonlinearity depends on the number of stocks. Finally, according to the graph, different assumptions about the number of stocks lead to approximately the same curve.

It is worth mentioning that the current model can be used to recognize the quality of the paper with respect to recycling rate (see Figure 8). As was mentioned earlier, the stock S1 represents a number of the largest fibres and SN - of the smallest ones. The

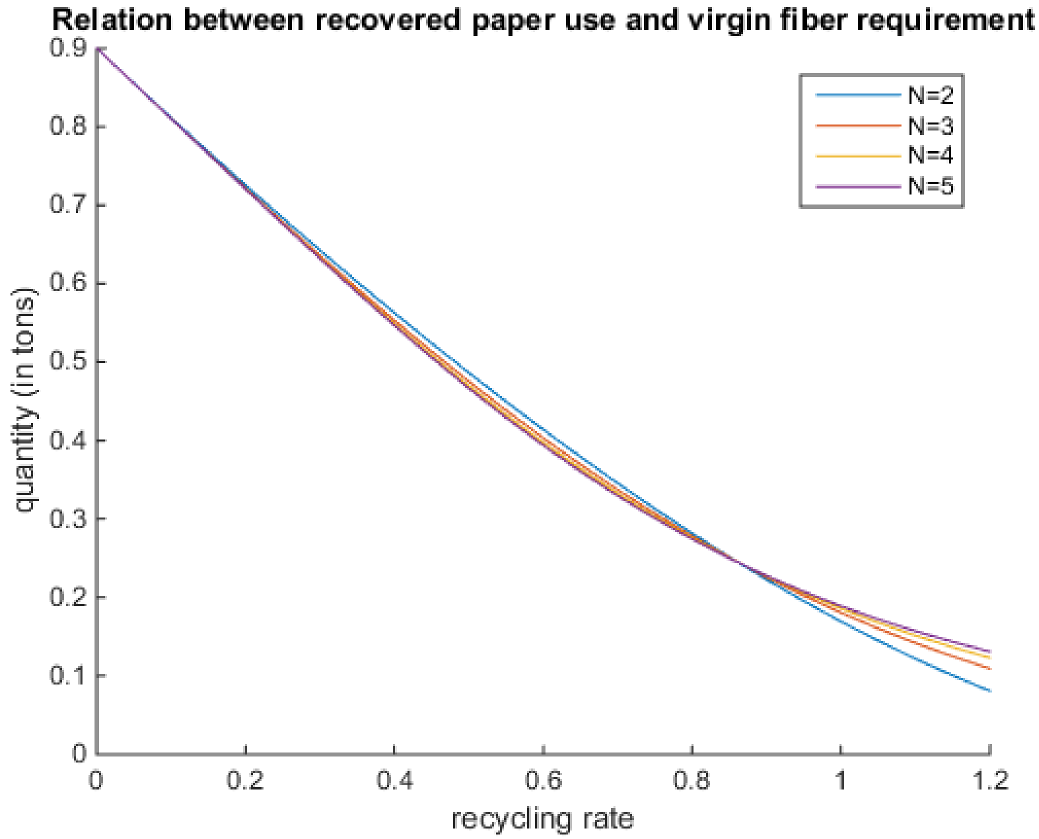

Figure 7: Relationship between recycling rate and virgin fibre requirement with respect to number of stocks.

quality of paper can be determined by the size of fibres which were used for paper production. Since the equations for stocks are derived, we can calculate the sizes of stocks for different recycling rates. Thereby, the grade of paper can be obtained by estimating the portions of fibre's size categories. For instance, when a recycling rate is equal to 0.2 then approximately 82.2% of fibres in resulted paper have the largest size and 14.5% of fibres belong to the second grade, the rest of fibres, which is 3.3%, are the shorter ones.

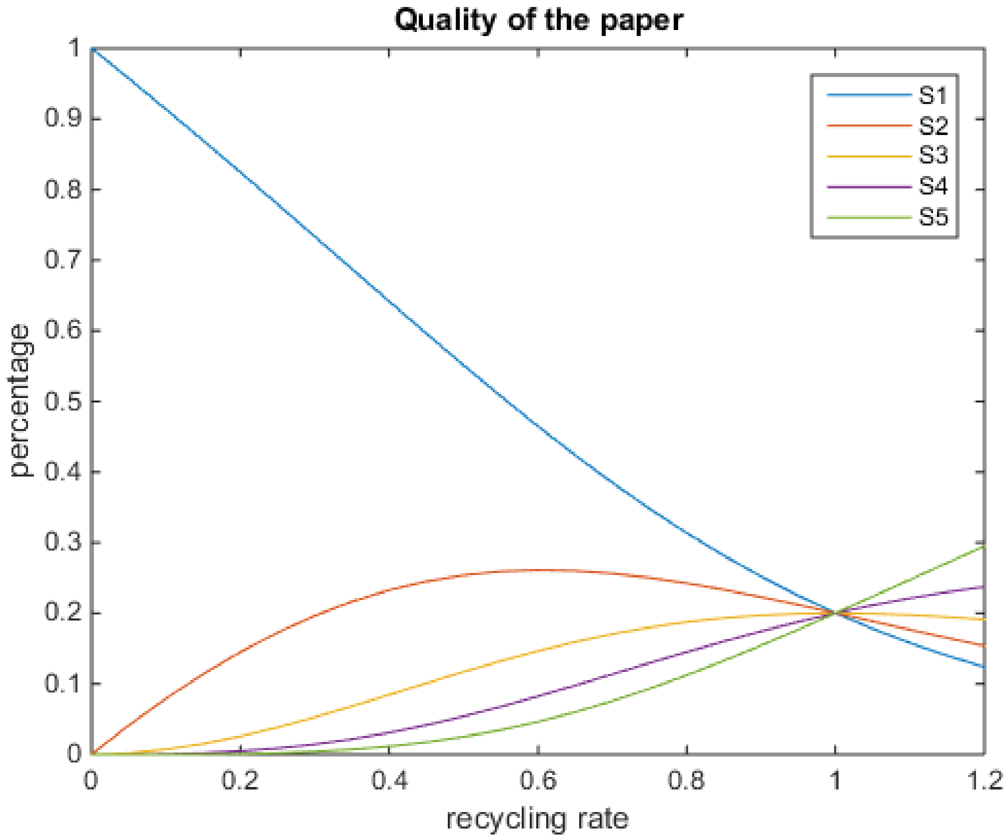

Figure 8: Quality of the paper with respect to recycling rate.

4 Optimal amount of recovered paper

A total carbon footprint consists of many components. Depending on materials and the amount used in production and the type of production itself, the value of carbon footprint varies. For analyzing and quantifying the environmental impacts, life cycle assessment (LCA) methodology is used. The system boundary of the comparative LCA study was cradle-to-gate, which means that all emissions associated with produced paper were considered from raw materials acquisition, through processing stages, accounting for the production and use of fuels, electricity, and heat, as well as taking into consideration transportation impacts along the product supply chain. In other words, the boundaries of the study begin at raw material extraction (the cradle) and finish when the manufactured products are prepared to leave the factory gate.

It is worth mentioning that transportation plays an essential role in the estimate of the carbon footprint since the production of a tonne of paper usually includes thousands of kilometers of transporting. The main aim of the current study described in this chapter is to estimate the optimal recycling rate in terms of carbon footprint and analyze its dependence on transportation distance.

4.1 Total carbon footprint

First of all, the total carbon footprint comprises emissions of pulp and paper production from raw material and emissions from recycled paper. The CO_2 emission from virgin fibre consists of the CO_2 content of wood, emissions made through transportation of raw materials, chemicals, process emissions which involve forest operations, pulpmaking and papermaking processes. It is reputed that fuel, steam, and electricity consumption make the largest contribution to the total carbon footprint associated with forest products [18]. The carbon footprint emitted by pulp production from recycled paper includes the CO_2 content of waste paper, emissions made through transportation of waste paper and chemicals, process emissions which involve the stock preparation, pulpmaking and papermaking processes.

4.1.1 Forest operations

Forest activities are considered as a first impact category. It includes emissions associated with the use of fertilizers (N, P_2O_5, and K_2O) and herbicides used on the land during planting and growth; energy-related emissions associated to the combustion of fossil fuels by harvesting equipment in forest operations; and burning which is used for site preparation or undergrowth control ([19], [20] , [8], [21]). Pine tree forests were assumed as they are the only virgin material processed in the pulp mill. Pine tree seedling production and transportation of workers are excluded due to the lack of data.

4.1.2 CO_2 content

It is well known that compounds with carbon such as carbon dioxide (CO_2) and carbon monoxide (CO) conduce to air pollution and play a role in climate change if they are contained in the atmosphere in high concentrations. Carbon dioxide gas can be eliminated from the atmosphere by trees through photosynthesis. The process of photosynthesis converts absorbed carbon dioxide and water into hexose sugars, which contain carbon and oxygen. After sawing down a tree, carbon is still stored within wood. Also, the products which are made of wood also contain carbon. Nevertheless, carbon finally returns to the atmosphere as carbon dioxide when products are not capable of storing it. Stored carbon can be released as a result of burning or feeding of heterotrophic organisms. They break hexose sugars backwards into water and carbon dioxide. The process is closed and exactly the same amount of CO_2 is returned to the atmosphere. Therefore, we decided to take that value into account.

The amount of carbon stored in trees was already estimated by many researchers. However, the carbon content is sometimes confused with the carbon dioxide equivalent as a given quantity of carbon. One molecule of carbon generates into one molecule of carbon dioxide (CO_2). The molecular mass of carbon is 12 and the molecular mass of CO_2 is 44. Thus, 12 grams of carbon is resulting in 44 grams of carbon dioxide. Therefore, carbon dioxide equivalent can be estimated as follows:

$$CO2eq = 44/12 \times \text{mass of carbon}.$$

4.1.3 Chemical consumption in paper industry

For both pulp and paper manufacturing, various chemicals and chemical additives are used. In present work, we examine the production of both bleached kraft pulp from virgin material and pulp form recovered paper after a recycling process. Therefore, chemicals used for such types of pulp such as sodium hydroxide (NaOH), oxygen gas (O_2), sodium chlorate ($NaClO_3$), ethylenediaminetetraacetic acid (EDTA), sulfuric dioxide (SO_2), hydrogen peroxide (H_2O_2), ozone (O_3),magnesium sulfate ($MgSO_4$), quicklime (CaO), sodium silicate, soap, talc, sodium dithionite ($Na_2S_2O_4$), sulphuric acid (H_2SO_4) and bentonite are taken into account. The amount of carbon dioxide emitted during the use of chemicals is calculated by multiplying the mean quantity of chemicals by the emission factor. [8]

4.1.4 Transportation

The research includes a study of carbon burden from transportation throughout the supply chain because transportation is considered one of the biggest carbon dioxide emissions sources. There is a need for conveyance of raw wood material from forests to pulp mills; next, pulp from a pulp mill's gate has to be transported to paper mills (in cases of non-integrated production); also, collected waste paper, that is paper for recycling, should be delivered to paper mills; and, finally, finished paper is taken from a paper mill's gate to the consumer. For the current study, the consumer of paper is defined at the level of the printer or merchant. In addition, the transportation of chemicals is taken into account. Distances are classified by the mode of travel (ship, rail, and truck). The wood supply structure is principally locally sourced. Raw fibres are received by harvesting from native forests. We consider that a radius of each facility to forests is within approximately 200 kilometers. In contrast, waste paper for further recycling is usually transported from various places which are located even far away from a paper mill.

GHG Protocol [22] states that there are two methods to calculate emissions from transportation: fuel-based method and distance-based method. The first mentioned approach involves determining the amount of fuel consumed and applying the appropriate emissions factor for that fuel [22]. However, data on the types and quantities of fuels consumed by the vehicle is unavailable. As a result, the fuel-based method cannot be employed. The distance-based technique involves determining the mass, distance, and the mode of each shipment, and then applying the appropriate mass-distance emissions factor for the vehicle used [22]. Data for this technique is known. Therefore, in present work we will use the distance-based method.

To calculate a carbon footprint of transportation by the chosen method, distance (km) is multiplied by mass of goods transported (tonnes or volume) and relevant emission factor (kg CO_2eq/tonne/km).

4.1.5 Stock preparation

Stock preparation is used to prepare waste paper for the production of new paper. However, various products require different cleanliness and brightness properties from the recovered fibres pulp and the process concepts vary accordingly. For example, stock preparation for newsprint and simple printing and writing papers differs from stock preparation for packaging paper and paperboards. Thereby, they have different impacts on the environment with respect to energy use. Therefore, it is not reasonable to describe "one typical" recovered paper processing system. The main recovered paper processing systems may include such stages as raw material handling, pulping, cleaning, screening / fractionation, flotation, washing / thickening / dewatering, refining / deflaking, dispersion / bleaching.

4.1.6 Pulping process

The primary targets of a pulping process are to free fibres in wood from the lignin that binds them together and to make the pulp suitable for papermaking by blending wood fibres with water. To complete these actions, large quantities of steam and electricity are required [18]. The writers of BAT stated that heating fluids, evaporation of water, acceleration and control of chemical reactions, the operation of the paper machine as well as other industrial activities related to pulp production (debarking, chipping, cooking, pulp washing, bleaching) demand high power inputs in the form of heat and power. In order to generate electrical power in turbo generators heat energy in the form of high-pressure steam is used. During fuel combustion which is made for the acquisition of steam and electricity, carbon dioxide is emitting in abundance.

For calculation of CO_2 emission, we have to find the total energy consumption for pulping and then multiply the obtained value by the appropriate emission factor. First, we transform measurement units of heat to measurement units of electric power by multiplying the amount of heat by 277.8 since it is known that 1 GJ is equal to 277.8 kWh. Next, we add the amount of electric power to the result to derive the total energy consumption in kWh measurement unit. By multiplying the obtained value to the emission factor, the carbon footprint of pulping can be found (equation 15). We decided to use the specific emission factor (EF) which represents CO_2 emissions per kWh from electricity and heat generation which is typical for Finland [23].

$$CO_2 = (\text{heat} \times 277.8 + \text{electricity}) \times \text{EF} \tag{15}$$

As was mentioned earlier, recycling technology includes various combinations of the treatment processes which are performed to produce pulp from recovered paper. One of the most crucial steps involved in recycling is the production of deinked pulp from recovered paper and manufacturing paper by utilizing this pulp alone or mixed with other pulps which can be also virgin or recycled. These two approaches are very similar, but the techniques used for the production of deinked pulp are completely different than those used for the production of pulp from wood [24]. As a result, pulp production from recovered paper requires considerably less total energy for processing than is needed for chemical and especially for mechanical pulping, because the secondary fibres have already passed through stock-preparation equipment when the original paper was made.

4.1.7 Paper production

When pulp is ready, it can be either used for the production of paper right away in case a mill is integrated or delivered to a paper mill for production of paper in case a mill is non-integrated. However, the paper production is quite similar to the pulp production in terms of calculating carbon dioxide emissions. Various technologies of paper manufacturing consume different quantities of heat energy and electrical energy. The energy consumption depends on the process configuration, process equipment and process control efficiency. It is worth mentioning that integrated pulp mills consume considerably less energy for paper manufacturing in comparison with non-integrated mills.

4.2 Model of the total carbon footprint

A carbon footprint can be calculated for the particular set of parameters that is for a known recycling rate, number of stocks and pulp requirements for producing one tonne of paper. However, it does not reveal how the chosen recycling rate influences total carbon dioxide emissions. Therefore, the goal of the current research is to estimate the total carbon footprint as a function of recycling rate. The next step is to ascertain whether the taken recycling rate is an optimal one, which leads to minimal carbon dioxide emission. It can be determined by employing the equation for a nonlinear relationship between virgin pulp requirement and recycling rate which has been established earlier. Further, we clarify how to calculate the total carbon footprint for paper manufacturing as a function of recycling rate. In order to calculate the total quantity of emission we collect together all components that can be seen in the Figure 9.

The total carbon footprint is expressed as a sum of emissions which are related to manufacturing paper from virgin fibres i.e. wood and emissions which are related to secondary fibres i.e. waste paper. Mathematically, it can be written as:

$$TCF = CF_{vf} + CF_{rf},$$

where TCF is the total carbon footprint, CF is the carbon footprint, vf is the subscript referring to virgin fibres and rf is the subscript referring to recycled fibres. Next, according to the scheme in the Figure 9, the carbon footprint associated with virgin fibres is equals to the sum of emissions from transportation, a carbon content,

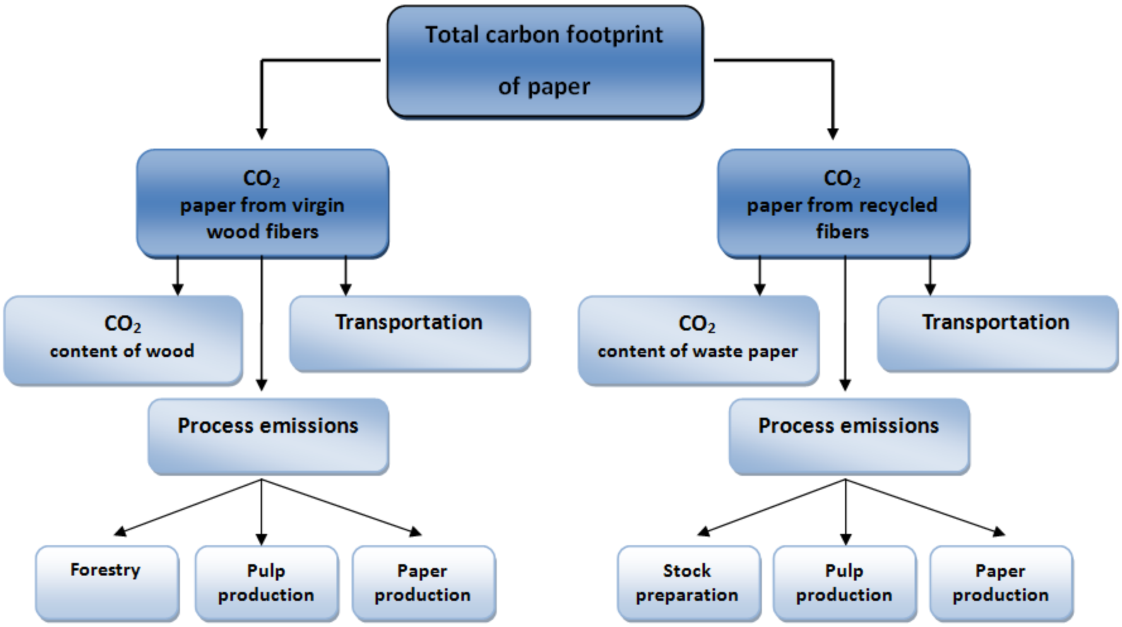

Figure 9: Model for assessment of the total carbon footprint of paper.

and manufacturing processes:

$$CF_{vf} = z(x) \times RWE \times CC_{wood} + PE_{vf} + TT_{vf},$$

where CC_{wood} is the carbon content of wood, PE is the process emission, and RWE is the roundwood equivalents, that is the amount of roundwood required to produce one tonne of pulp. TT_{vf} is the total transportation which is related to virgin fibres. PE_{vf} includes harvesting, pulp and paper production from virgin fibres.

The same way as the carbon footprint associated with virgin fibres was divided, carbon dioxide emission related to recycled paper is introduced. As was mentioned earlier, recycling rate is defined as recovered paper use (RPU) and the functional unit is one tonne of paper. Therefore, the amount of used paper is equal to the recycling rate. For example, if the recycling rate is 0.8, then 80% of total paper production, which is one tonne in our model, is used for paper production and it equals to 0.8 tonne of recycled paper.

$$CF_{rf} = x \times CC_{paper} + PE_{rf} + TT_{rf},$$

where CC_{paper} is the carbon content of paper, PE is the process emission, TT is total transportation.

Figure 9 depicts that PE_{rf} includes stock preparation, pulp and paper production from recovered fibres. Hence, process emissions associated with virgin materials are expanded as follows:

$$PE_{vf} = z(x) \times RWE \times HW + z(x) \times PulpP_{vf} + \frac{z(x)}{r} PaperP_{vf}.$$

Here HW is harvesting of wood, $PulpP_{vf}$ is the carbon footprint of pulp production from virgin fibres, $PaperP_{vf}$ is the carbon dioxide emissions which are emitted during paper production from pulp from virgin fibres.

Similarly, process emissions for paper manufacturing from waste paper can be derived as:

$$PE_{rf} = x \times SP + (r - z(x)) \times PulpP_{vf} + \frac{r - z(x)}{r} PaperP_{vf}$$

Here SP is the stock preparation, $PulpP_{vf}$ is the carbon footprint of pulp production from recycled fibres, and $PaperP_{vf}$ is the carbon footprint of paper production from pulp from recycled fibres.

Finally, the equations for a calculation of emissions of two components of the total carbon footprint are derived:

$$CF_{vf} = z(x) \times RWE \times (CC_{wood} + HW) + z(x) \times PulpP_{vf} + \frac{z(x)}{r} PaperP_{vf} + TT_{vf},$$

$$CF_{rf} = x \times CC_{paper} + x \times SP + (r - z(x)) \times PulpP_{vf} + \frac{r - z(x)}{r} PaperP_{vf} + TT_{rf}.$$

Now we are going to analyze the dependence of recycling rate on the amount of carbon burden released into the atmosphere. It is illustrated by the Figure 10. According to the graph, with raising a quantity of recovered paper use, the carbon footprint related to recycled paper increases. In contrast to this, the carbon footprint related to virgin material declines.

The graph (Figure 11) of the total carbon footprint shows that there is a point where the total carbon footprint has a minimum. We call this point the optimal point which is the optimal amount of recovered paper in terms of a carbon footprint. According to the data, it is possible to reduce the carbon footprint by approximately 735 kg (21% of reduction) of CO_2eq if the recycling rate is equal to 0.83. In other words, 0.83 is the optimal value in terms of carbon footprint for this case.

4.3 Sensitivity analysis

Calculations above are based on average values, therefore the sensitivity analysis is required. Sensitivity analysis is defined as the study of how uncertain is the

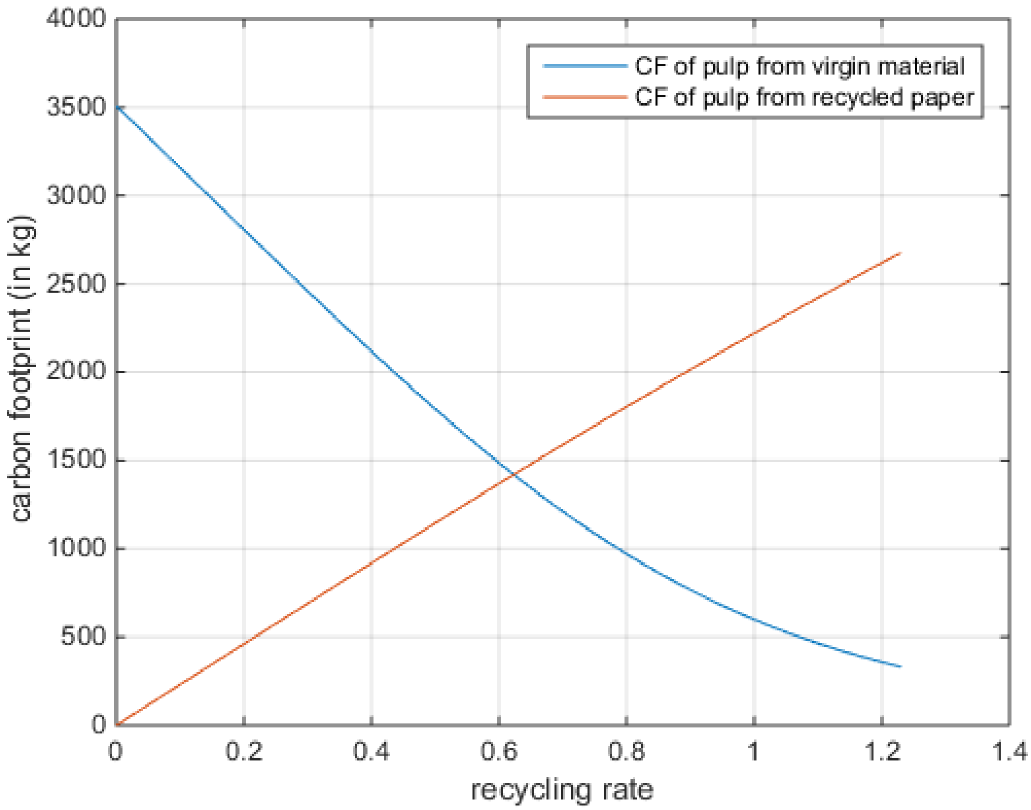

Figure 10: Carbon footprints associated with virgin fibres and recycled fibres with respect to recycling rate.

output of a model attributed to different sources of uncertainty in the model input [25]. There is a wide range of purposes to which a sensitivity analysis is employed, including testing the robustness of the results of a model or system in the presence of uncertainty and increased understanding of the relationships between input and output variables in a system or model.

A local sensitivity analysis method applies a one-at-a-time (OAT) approach to quantify uncertainty in the output of a model. This approach is one of simplest and most common approaches that is based on changing one value at a time of each model parameter in order to assess the impact on the model outputs. It means that sequentially each of the model parameters is varied and others are fixed for estimating model output. One of the main advantages of the current method is the low computational cost, however it cannot be applied to complex models. In addition, OAT method does not take into account any interactions between model parameters. Clearly, in the model for assessment of the total carbon footprint all parameters are

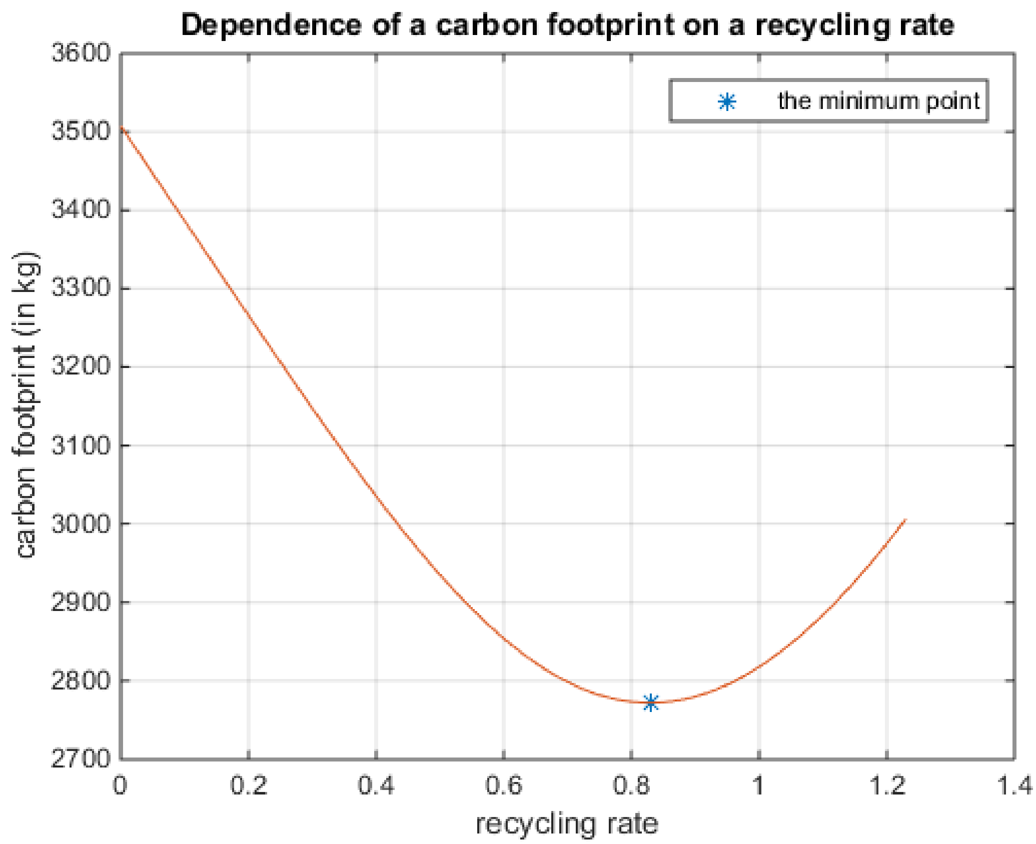

Figure 11: Dependence of a carbon footprint on recycling rate.

independent.

We perform the sensitivity analysis (Figure 12) for the model under study by changing typical mean values for following model parameters: carbon content of wood; roundwood equivalent; harvesting of wood; carbon footprints of production processes such as paper and pulp manufacturing; transportation's distances.

Sensitivity analysis revealed that the optimal recycling rate has an expectation 0.8215 with standard deviation 0.0886. Obtained results are subject to a lack of data. Hence, the confidence interval for the optimal recycling rate can be diminished by specifying more exact values for contributors in total dioxide emissions. According to the graph in Figure 12, we can say that an amount of wood needed for the pulpmaking process has the biggest influence on the optimal amount of recovered paper.

It is known that wood and chemicals are transported to a pulp mill from neighborhood areas i.e. 100-200 km. In contrast to this, waste paper can be located far

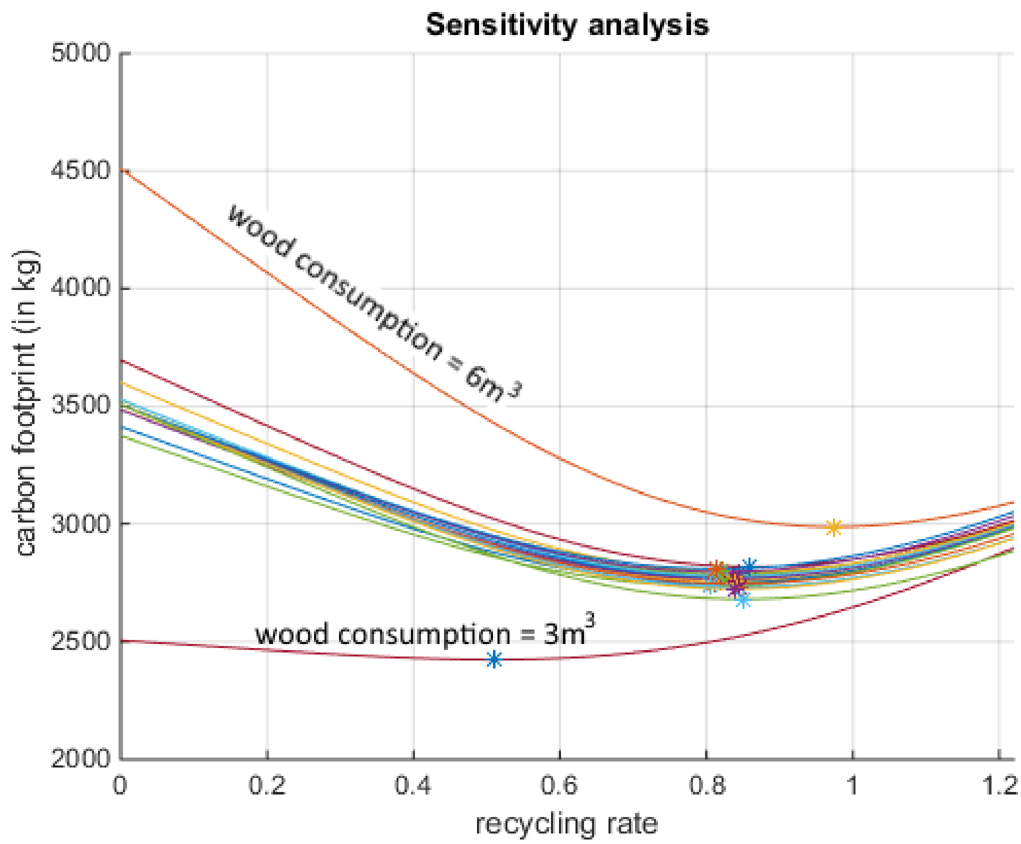

Figure 12: The result of the sensitivity analysis.

away from a paper mill where pulp from recycled paper is made. For studying a dependence between distances of transportation, an amount of recovered paper, and a carbon footprint we took following distances as an example (Figure 13):
Distance from a forest to a pulp mill = 150 km;
Distance from a chemicals storehouse to a pulp mill = 150 km;
Distance from a waste paper location to a paper mill = 200, 300, ... , 7000 km;

As is illustrated by the next graph (Figure 14), there is a clear relation of transportation distance to the optimal quantity of recovered paper. In addition, we can see that there is no reason (in terms of a carbon footprint) to use waste paper instead of virgin material when waste paper is located too far away (more than 5800 km in our case) from a paper mill.

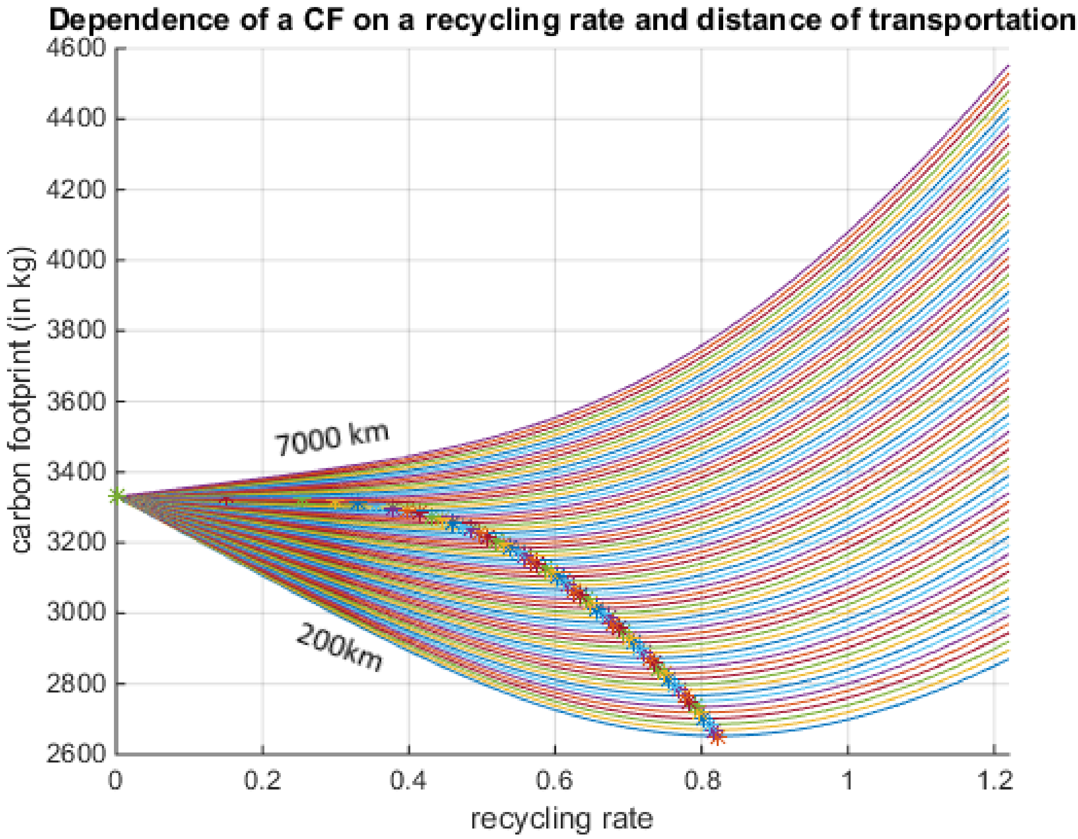

Figure 13: Dependence of a carbon footprint on recycling rate and distance of transportation.

4.4 Influence of transportation on paper's supply chain

Organization of a new supply chain of paper to a customer is one of the crucial problems. At the present time, companies tend to take into account, in addition, carbon dioxide emissions which are released into the atmosphere during transportation and paper production. Carbon dioxide emission from transportation throughout a product's life cycle also depend on where the product is produced, where the raw materials for the product are extracted and processed, and where it is sold. Therefore, the third aim of the present study is to devise a framework for calculating the optimal route of a supply chain.

Let us introduce the first transportation model. A customer can get paper from several different paper mills situated at distinct locations. During the transportation some quantity of CO_2 is emitted into the atmosphere. In turn, a paper mill requires pulp which can be manufactured at the same mill or at another one. It means

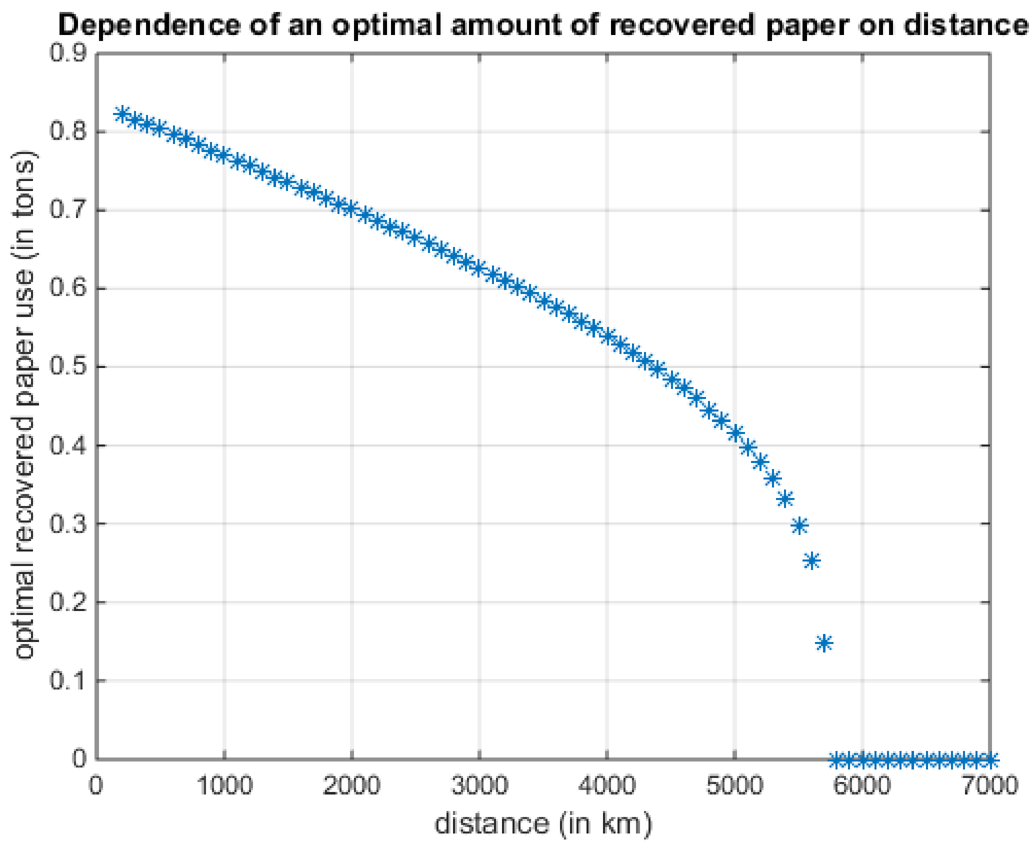

Figure 14: Dependence of an optimal recycling rate on distance of transportation.

that pulp also should be transported to the paper mill and that pollutes the air. Consequently, pulp mills are in need of wood which is transported from one of forests. There is an assumption that all pulp mills require the same amount of wood for manufacturing one tonne of pulp, however all paper mills may require different amounts of pulp for production of one tonne of paper and have different recycling rates. One more assumption is that chemicals are shipped to mills from a short distance about 100 km. The goal is to calculate the optimal path so that the amount of CO_2 is minimal.

The problem under consideration can be described by a scheme in the Figure 15 where all possible routes can be seen. It is worth mentioning that the problem at first glance resembles a graph, however, it is not a graph. There are two reasons for that. The first reason is that there are weights on nodes which represent the carbon footprint of mills and forests. The second reason is that weights on edges and nodes depend on the chosen route. Nonetheless, the current scheme can be transformed into a graph, but it will be enormous for fast finding the optimal supply chain. A better way to obtain the result rapidly is to solve the problem by total enumeration

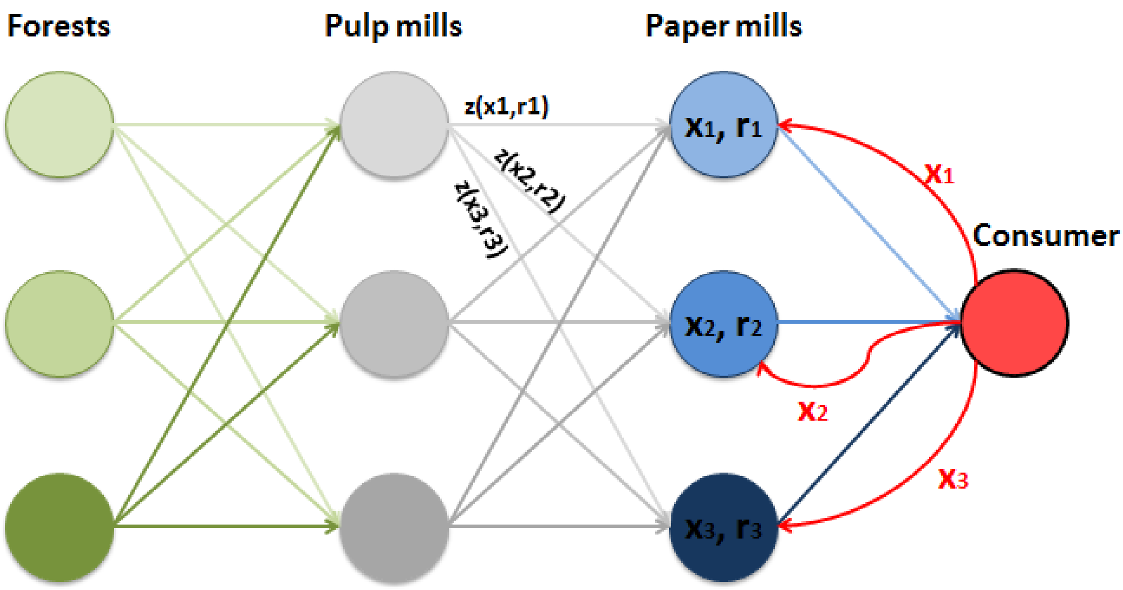

Figure 15: Model of the supply chain for manufacturing one tonne of paper and shipping it to a consumer. Red arrows represent transportation of waste paper from consumer to paper mills for recycling. Values x_i is the amount of waste paper, which should be shipped to a corresponding mill. $z(x_i, r_i)$ is virgin pulp requirements, which should be shipped to a corresponding paper mill from any pulp mill.

of all possible cases which is known as the brute-force method.

For using the model, we need to know distances between all forests and pulp mills, pulp mills and paper mills, paper mills and the customer. In case of fully integrated mills, pulp and paper are produced at the same location, thereby eliminating a need for transportation of pulp from distant locations. Also, we have to know a carbon footprint of each forest, each pulp mill, and each paper mill which are defined as specific emission factors. In addition, paper mills have their specific properties such as recycling rate and pulp requirement for producing one tonne of paper. As was mentioned earlier in the present study, it is possible to calculate virgin pulp requirements in order to divide the carbon footprint into two groups: the first one associated with virgin fibres, the second one associated with recycled paper. Here we also assume that the damage rate of each mill is known, and all mills have the same value.

Usually, waste paper is transported from the place where the consumer is. So, we make an assumption that the distance between a paper mill and a source of waste paper is equal to the distance between the mill and the consumer. Knowing all distances, types of vehicles, and the weight of cargo, we can calculate the optimal route for transportation. In reality various paper mills have diverse demands on resources and various pulp mills have different demands on quantity of wood. For example, the first paper mill requires r_1 tonnes of pulp, the second one r_2 tonnes and so on. It means that carbon burden released by the mill depends on pulp requirements, i.e. it is not constant and should be calculated. In turn, pulp mills have different demands on wood. What is more, paper mills can manufacture pulp with different amounts of recovered paper, that influences on virgin pulp requirements (wood from a forest).

Appropriate framework is designed to utilize the established model under consideration. User should indicate properties of the model. First, the number of forestry activities spots, paper mills, and pulp mills have to be specified. Further, framework requires data on special characteristics of forestry, pulp, and paper mills such as emission factors, recycling rates and etc. When all information about all places of activity is obtained, the route map can be established. The well-known way to specify route graphs is to employ a matrix of distances. Initially, distances between spots are assigned to infinity, which represents that there are no connections between any nodes. Hence, the matrix of distances should be filled with distances. Also, user is able to specify the type of vehicle (truck, train or vessel) which is used to ship cargo via the specific road. The result of the current framework's execution is the supply chain which is optimal in terms of carbon footprint among all possible choices.

We performed a test to verify the framework. In the Appendix 2, the input data for the framework is situated. The input describes an artificial situation when the final customer is located in the northern Germany. In addition, there are many possibilities where paper can be manufactured. We suggested three paper and pulp mills which are located in Finland, in Germany, in China. In addition, we assume that the pulp mill in Brazil is able to ship pulp to Europe. Finally, four spots for forest activities are placed in our model: Brazil, Finland, China, Russia. Also, the distances between pseudo-nodes are estimated and appropriate shipping methods are assumed. More exact information about all models parameters can be found in the Appendix 2. The result of the implementation of the established approach to current data depicts that the optimal route is $2 \rightarrow 2 \rightarrow 1 \rightarrow 1$. In model's notations, it can be represented as follows $Finland \rightarrow Finland \rightarrow Finland \rightarrow Germany$. The result states that the optimal route is sourced from Finland, where the paper is manufactured from local raw materials, and after that it ships to the consumer. The estimated optimal carbon footprint, in this case, is 3359.6 kg CO_2eq and the recycling rate of the paper mill in Finland is 0.8.

Sometimes paper industries would like to estimate not only the optimal route but also the optimal recycling rate. Therefore, we decided to combine the current model and an idea to estimate the optimal recycling rate in terms of carbon dioxide emissions in order to minimize a carbon footprint of paper. As a result, we should not only minimize the objective function by choosing a route, but also by estimating an appropriate recycling rate. The settings for the current model are almost the same as for the previous one, but the new model no longer requires specification of recycling rates of paper mills. That makes this approach more flexible than the previous one.

Current problem can be also solved by the brute-force method with an additional optimization of the recycling rate so that the amount of carbon dioxide emissions is minimum for each route as was described in Section 4.2. As a result, it takes more computational time, but it provides the solution which guarantees smaller carbon footprint than the previous scheme. We apply the current model to the data which is mentioned above (Appendix 2). The output of the second transportation model is $Russia \rightarrow Germany \rightarrow Germany \rightarrow Germany$, which is different from the result of the first model. It states that forest should be cut in Russia and then shipped to Germany for the consequent paper manufacturing. As we can see, the preferable route was altered and the optimal recycling rate was calculated. Thereby, the present result indicates lower carbon dioxide emission which is 2970.2 kg CO_2eq. Also, the model states that the optimal recycling rate of the paper mill in Germany should be 0.7366 instead of 0.1, which was assumed initially. It means that the route for the supply chain should be $Finland \rightarrow Finland \rightarrow Finland \rightarrow Germany$ when recycling rates cannot be changed and specified by particular mills. However, in case of flexible recycling rates the route can be modified dramatically to $Russia \rightarrow Germany \rightarrow Germany \rightarrow Germany$. What is more, it results in a diminishing the carbon footprint of manufactured paper in comparing to the model with fixed recycling rates.

5 Conclusions and future work

In the present paper, we studied the process of paper manufacturing and its influence on the environment in terms of carbon footprint. Generally, we discussed the importance of both paper recycling process and transportation on the amount of carbon dioxide emissions.

The analysis started from establishing a model of paper recycling. According to the model, a mathematical relationship between virgin fibre requirements with respect to the quantity of recycled paper in the pulp was found. As a side achievement of the present research, we mathematically derived the exact equation for estimating virgin pulp requirements for different numbers of stocks.

As a core of this project, the model of papermaking process was devised. It includes all stages of the process: from cutting trees until pulp and paper production. Moreover, the designed model was used to clarify the contribution of recycling to decreasing carbon dioxide emissions. In addition, we stated that increasing of recycling rate does not always lead to minimizing the carbon footprint. As a result, we considered that there was an optimal recycling rate which resulted in the lowest level of emissions. In addition, the robustness of this optimum was investigated by sensitivity analysis.

Next, we studied how transportation of waste paper to a paper mill influences on the optimal recycling rate. It was declared that shipping of paper for future recycling throughout long distances has a harmful effect on the environment. We derived also that transportation of waste paper throughout distances longer than 5800 km has no sense because use of that paper will only increase carbon dioxide emissions and it is better to reject recycling at all.

Finally, we designed a model for organization of a new supply chain of paper to a customer. The first established framework was designed to estimate the optimal route of a supply chain in terms of carbon footprint. The second framework provides also the optimal route, but it estimates, in addition, the optimal recycling rate for initiating a new supply chain so that emissions associated with paper production are minimized.

The main objective of further investigations is to apply derived equations to a real case since the current research utilized common values for models' parameters from a wide range of resources. Therefore, it seems reasonable to collaborate with a particular paper manufacturing company and collect necessary data in order to clarify the established models. In addition, we suggest expanding the model under study from cradle-to-gate to cradle-to-grave life cycle analysis. We assume that a cradle-to-grave model will help to make the calculated carbon footprint more accurate and comprehensive, and improve the value of the optimal recycling rate.

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6 Appendix 1

Substance	Consumption [kg/t]	Emission factor [kg CO_2eq/kg]	Sources
NaOH	20-25	1.2012639	[8] [21]
O_2	20-25	0.48166558	[8] [21]
$NaClO_3$	15-50	3.6563215	[8] [21]
EDTA	0-4	4.8107386	[8] [21]
SO_2	2-10	0.44	[8] [26]
H_2O_2	2-30	1.1279307	[8] [21]
O_3	0-5	8.01	[8] [26]
$MgSO_4$	2-3	0.3	[8] [26]
CaO	5-10	1.0064599	[8] [21]

Table 3: Consumption of main chemicals for kraft pulp production and their emission factors.

TO DO: tables with used data for calculations and references on sources

Substance	Consumption [kg/t]	Emission factor [kg CO_2eq/kg]	Sources
NaOH	2-10	1.2012639	[8] [21]
Na_2SiO_3	12-25	1.5295925	[8] [21]
Soap	3-8	1.6880499	[8] [21]
Talc	0-15	0.030415572	[8] [21]
H_2O_2	5-25	1.1279307	[8] [21]
EDTA	0-3	4.8107386	[8] [21]
$Na_2S_2O_4$	6-10	3.5629278	[8] [21]
H_2SO_4	8-10	0.089763527	[8] [21]
Bentonite	0-4	0.24	[8] [27]

Table 4: Consumption of main chemicals for processing waste paper and their emission factors.

Item	Consumption	Emission Factor	Source
Wood	3-6 m^3	708 kg CO_2eq/kg	[8], [28], [29]
Waste Paper		1569 kg CO_2eq/kg	[30]
Harvesting		15-26 kg CO_2eq/m^3	[21]
Stock Preparation	400-600 kWh and 0.450-0.9 GJ	0.191 kg CO_2eq/kWh	[8],[31]
Pulp and Paper Production related to virgin fibres	706 kWh and 3.5 GJ	0.191 kg CO_2eq/kWh	[10], [31]
Pulp and Paper Production related to waste paper	917 kWh and 5.5 GJ	0.191 kg CO_2eq/kWh	[10], [31]
Transportation by a truck		203.0007 kg CO_2eq/km/kg	[2]
Transportation by a ship		384.4257 kg CO_2eq/km/kg	[2]
Transportation by a train		17.1918 kg CO_2eq/km/kg	[2]

Table 5: Data used for the current study (for production of one tonne of paper)

7 Appendix 2

```
%Framework for finding the optimum route
%All data relate to production of 1 tonne of paper (kraft technique)
clear; close all; clc;
%Quantity(1) = number of forests
%Quantity(2) = number of pulp mills
%Quantity(3) = number of paper mills
%Quantity(3) = number of customers

Quantity = [4 4 3 1];
%Assume that the main consumer is in Germany

%Variable "Forest" contains different places of forest operations
%Essentially, all places can have different technologies for forest operations
%Forest(i) contains the specific emission factor of the i-th forestry in
%CO2kg/m3 that is CO2 emission for production of one m3 of wood
Forest = zeros(1,Quantity(1));
Forest(1) = 728;   %located in Brazil
Forest(2) = 750;   %located in Finland
Forest(3) = 800;   %located in China
Forest(4) = 750;   %located in Russia

%Variable "PulpMill" contains different places for pulp production
%Essentially, carbon footprint depends on the type of a mill
%(integrated / non-integrated) and technologies used by the mill
%PulpMill(i) contains the specific emission factor of the i-th pulp mill
PulpMill = zeros(1,Quantity(2));
PulpMill(1) = 250 ;   %located in Brazil
PulpMill(2) = 240 ;   %located in Finland
PulpMill(3) = 285 ;   %located in China
PulpMill(4) = 257 ;   %located in Germany

%Variable "PaperMill" contains different places for paper production
%Essentially, carbon footprint depends on the type of a mill
%(integrated / non-integrated) and technologies used by the mill
% PaperMill(i) contains the specific emission factor of the i-th paper mill and its
% pulp requirement for production of 1 ton of paper and
% recycling rate
PaperMill = cell(1,Quantity(3));
PaperMill(1) = {[512 0.853 0.8]};  %located in Finland
PaperMill(2) = {[500 0.902 0.1]};  %located in Germany
PaperMill(3) = {[560 0.940 0.5]};  %located in China

%%%%%%   TRANSPORTATION   %%%%%%
% 1 - truck
% 2 - train
% 3 - ship
```

```
%Transportation from a forestry to a pulp mill
%Cij - from the forestry i to the pulp mill j
C = [{[45 1]} {[9500 3]} {[10000 3]} {[7000 3]};
     {[9500 3]} {[37 1]} {[7000 3]} {[1000 3]};
     {[10000 3]} {[6200 3]} {[40 1]} {[8600 3]};
     {[10000 3]} {[150 1]} {[3500 3]} {[2000 2]};];

%Transportation from a pulp mill to a paper mill
%Dij - from the pulp i to the paper mill j
D = [{[9500 3]} {[7000 3]} {[10000 3]};
     {[0 1]} {[1000 3]} {[7000 3]};
     {[7000 3]} {[8600 3]} {[0 1]};
     {[1000 2]} {[0 1]} {[8600 3]};];

%Transportation from a paper mill to a consumer
%Ei - from the paper mill i to the consumer
E = [{[900 3]};
     {[500 1]};
     {[8600 3]};];

data.Quantity = Quantity;
data.Forest = Forest;
data.PulpMill = PulpMill;
data.PaperMill = PaperMill;
data.C = C;
data.D = D;
data.E = E;
save('params.mat','data');
clear all;
```