



Saeed Rahimpour Golroudbary

SUSTAINABLE RECYCLING OF CRITICAL MATERIALS



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Abstract

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The world's supply of critical materials such as phosphorus (P), niobium (Nb), lithium (Li) and other strategically important elements is under increasing pressure due to the rapidly growing global demand in the recent years and limited possibilities of substitution. These materials are used in producing a broad range of products in everyday life and forming an integral part of many advanced and clean energy technologies. Hence, such materials are significant for many industrial sectors and essential to societal well-being. Therefore, the steady supply of critical materials starts to be one of the key economic and environmental questions. Moreover, the analysis of flows of those materials coming from mining and recycling starts to evoke the growing interest.

A systematic understanding of how such materials flow through the industrial and residential sectors is required. Such awareness of materials' inclusion in various products and their current stocks in the anthroposphere improve the potential of recycling and reuse of those materials as well as minimize overall waste.

This dissertation presents dynamic models for critical materials such as P, Nb, and Li by using system dynamics methodology. It considers all stages of supply chain by addressing material and energy flows as well as greenhouse gas emissions. The main finding assists in optimizing for environmentally sustainable operations in designing and modelling of the critical materials supply chain.

The findings indicate a clear need to analyse the recycling processes carefully. The obtained results show that recycling of used products containing critical materials, in some cases, aims to prevent the shortage of those materials and contributes to developing a robust circular economy. However, the environmental sustainability of recycling procedures for all materials could not be taken for granted, because it could differ based on the type of the waste stream. For some critical materials, recycling can cause more environmental damage than mining. Therefore, we should not treat critical materials as a homogeneous group. Recycling carried out using the existing technologies is a partial solution for some materials. In addition, there are physical limitations to the increasing of the recycling rate for some materials. The main limiting conditions of recycling can be economic, environmental, and physical by nature. The lattermost means that even if recycling is both more profitable and "greener" than mining, it is still impossible to completely replace primary production with the secondary one.

Keywords: Critical raw materials, recycling, environmental aspect, sustainability, supply chain, dynamic simulation

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Saeed Rahimpour
April 2020
Lappeenranta, Finland

“Systems thinking is a discipline for seeing wholes. It is a framework for seeing interrelationships rather than things, for seeing 'patterns of change' rather than static 'snapshots'.”

– Peter Senge –

Contents

Abstract

Acknowledgments

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Publications

List of publications

This dissertation is based on the following four articles included in Part II. The rights have been granted by publishers to include the articles in the dissertation.

- I. El Wali, M., Rahimpour Golroudbary, S., and Kraslawski, A. (2019). Impact of recycling improvement on the life cycle of phosphorus. *Chinese Journal of Chemical Engineering*, Vol. 27, pp. 1219-1229.
- II. Rahimpour Golroudbary, S., El Wali, M., and Kraslawski, A. (2019). Environmental sustainability of phosphorus recycling from wastewater, manure and solid wastes. *Science of the Total Environment*, 672, pp. 515-524.2
- III. Rahimpour Golroudbary, S., Krekhovetckii, N., El Wali, M., and Kraslawski, A. (2019). Environmental sustainability of niobium recycling: The case of the automotive industry. *Recycling*, 4(1), 5.
- IV. Rahimpour Golroudbary, S., Calisaya-Azpilcueta, D. and Kraslawski, A. (2019). The Life Cycle of Energy Consumption and Greenhouse Gas Emissions from Critical Minerals Recycling: Case of Lithium-ion Batteries. *Procedia Cirp*, 80, pp. 316-321.

Author's contribution

Saeed Rahimpour Golroudbary is the principal author and investigator in papers II, III and IV, co-author in paper I, and the corresponding author in papers I, II, III and IV.

In paper I, Mohammad El Wali conducted data collection, literature, modelling and writing original draft and review. Saeed Rahimpour Golroudbary was responsible for conceptualization of idea, methodology, analysis, data collection, writing — preparation, review, and editing of original draft. Andrzej Kraslawski supervised conceptualization of idea and was involved in writing review and editing draft.

In paper II, Saeed Rahimpour Golroudbary was responsible for conceptualization of idea, methodology, software, modelling, formal analysis, data collection, writing — preparation, review, and editing of original draft. Mohammad El Wali facilitated literature survey, data collection, and modelling. Andrzej Kraslawski supervised conceptualization of idea and methodology as well as was involved in manuscript preparation and writing — review, and editing.

In paper III, Saeed Rahimpour Golroudbary was responsible for conceptualization of idea, methodology, software, modelling, analysis, data collection, writing — preparation, review, and editing of original draft. Nikita Krekhovetckii and Mohammad El Wali facilitated literature survey, validation, and data collection. Andrzej Kraslawski supervised methodology and was involved in writing — review and editing draft.

In paper IV, Saeed Rahimpour Golroudbary was responsible for conceptualization of idea, methodology, software, modelling, analysis, data collection, writing—preparation, review, and editing of original draft. Daniel Calisaya-Azpilcueta facilitated data collection and modelling. Andrzej Kraslawski supervised conceptualization of idea.

Nomenclature

CRM	Critical raw material
P	Phosphorus
PR	Phosphate rock
Nb	Niobium
Li	Lithium
Co	Cobalt
Mn	Manganese
P ₄	White phosphorus
SD	System dynamics
SC	Supply chain
SCM	Supply chain management
EV	Electric vehicle
EOL	End-of-life
ELV	End-of-life vehicle
GHG	Greenhouse gas
LIBs	Lithium-ion batteries
LMO	Lithium-ion manganese oxide
LCO	Lithium-ion cobalt oxide
LFP	Lithium-ion iron phosphate
NMC	Lithium-ion nickel manganese cobalt oxide
LiNCA	Lithium-ion nickel cobalt aluminum oxide
USGS	United States Geological Survey
EU	European Union
EU-28	28-member states of the European Union
IEA	International Energy Agency
HSS	High-strength steel
HSLA	High-strength low alloy
METI	Ministry of Economy, Trade, and Industry in Japan
MOFA	Ministry of Foreign Affairs in Japan
SEI	Stockholm Environment Institute
CLD	Causal loop diagram
SFD	Stock and flow diagram
CO ₂	Carbon dioxide
SO _x	Sulphur oxides
NO _x	Nitrogen oxides
CH ₄	Methane
V (t)	An exogenous variable in time t
P (t)	A set of parameters of the system
LCE	Lithium-carbonate equivalent
SDS	Sustainable development scenario

1 Introduction

1.1 Background

Critical raw materials (CRMs) are subject to supply risks, no substitution, environmental concerns, economic importance, recycling restriction, vulnerability to supply and demand growth (EU Commission, 2010a; Graedel et al., 2015; Northey et al., 2017; Schwegler, 2017). Such materials are fundamental to a broad range of industries such as food, automotive, battery, wind, and lighting, etc. Therefore, those materials are essential to societal well-being and ensure specific characteristics to advanced goods or systems.

Criticality of materials can be viewed at four levels: local (e.g., an enterprise) and national for the time horizon 1-5 years, economic region (e.g., Europe) in the time horizon 5-10 years, and a global scale in the time horizon 10-100 years (Glöser et al., 2015; Graedel et al., 2012). The criticality of a material strongly depends on what factors are considered in the assessment and how “criticality” is defined. For example, one of the primary issues regarding raw materials is the supply risk, which according to previous research was the most prevalent element of the material's criticality (Graedel et al., 2012; Jin et al., 2016).

In most cases, differences between the assessment of the criticality of the materials result from considering various parameters for assessing supply risks (Achzet and Helbig, 2013). For example, different parameters of supply risks were considered in previous studies, e.g.: geological availability (Angerer et al., 2009; Bauer et al., 2010; Buchert et al., 2009; Eggert et al., 2008; Graedel et al., 2012; Helbig et al., 2017; Hollins, 2008), by-product dependency (Achzet and Helbig, 2013; Bauer et al., 2010; Buchert et al., 2009; Eggert et al., 2008; Graedel et al., 2011a; Moss et al., 2011a), import dependency (Achzet and Helbig, 2013; Consult, 2011; Eggert et al., 2008; EU Commission, 2014; Frondel et al., 2007; Graedel et al., 2011b, 2012; Helbig et al., 2017; Moss et al., 2011a; Rosenau-Tornow et al., 2009; Thomason et al., 2010), potential of recycling (Eggert et al., 2008; EU Commission, 2010b, 2017a; Helbig et al., 2017; Ku et al., 2017), political risk (Bauer et al., 2010; EU Commission, 2017b; Helbig et al., 2017; Hollins, 2008; Ku et al., 2017), concentration of supply (Achzet and Helbig, 2013; Bauer et al., 2010; Buchert et al., 2009; Erdmann and Graedel, 2011; EU Commission, 2010b, 2014, 2017b; Frondel et al., 2007; Graedel et al., 2011a; Helbig et al., 2017; Hollins, 2008; Moss et al., 2011a; Rosenau-Tornow et al., 2009), vulnerability to climate change (Hollins, 2008), lead times to expand production (Buchert et al., 2009; Eggert et al., 2008), competing demand (Bauer et al., 2010; Helbig et al., 2017; Ku et al., 2017), technological factor (Angerer et al., 2009; Graedel et al., 2012), economic importance (Angerer et al., 2009; EU Commission, 2010b, 2014, 2017b; Graedel et al., 2012), social and regulatory (Graedel et al., 2012), time scale (Graedel et al., 2012; Peck et al., 2015), substitutability (Achzet and Helbig, 2013; Consult, 2011; EU Commission, 2010b, 2017a; Ku et al., 2017), depletion time (Achzet and Helbig, 2013; Consult, 2011; Erdmann and Graedel, 2011; Graedel et al., 2011b; Rosenau-Tornow et al., 2009), price volatility (Achzet and Helbig, 2013; Erdmann et al., 2011; Ku et al., 2017), demand growth (Angerer et al., 2009; Consult,

2011; Helbig et al., 2017; Moss et al., 2011a), risk of strategic use (Consult, 2011), and exploration degree (Rosenau-Tornow et al., 2009).

In literature, there are differences in criticality assessment methodologies (Frenzel et al., 2017; Jin et al., 2016). Therefore, critical materials are not generally defined on grounds of one common aspect. For instance, the US National Research Council uses the indicator of supply risk to assess the criticality of materials (Eggert et al., 2008). Notably, this assessment considers several factors such as the significance of a material for a specific area, the impact of the material on the decrease in economic output and decelerating or even stopping technological advances (Hofmann et al., 2018). In another assessment, Yale University assessed the criticality of materials by adding environmental implications to supply risk and vulnerability to supply restriction (Graedel et al., 2015). Moreover, material criticality is assessed by the EU Commission based on three leading aggregated indicators: supply risk, economic importance, and environmental country risk (EU Commission, 2017a). Those indicators are defined based on sustainability aspects (social, economic, and environmental). Looking from the social perspective, the indicator of supply risks is mainly determined by three factors including a high share of global production (stability/instability and level of concentration of producing countries), low substitutability and low recycling rate. At economic level we should look at the significance of a material through the lenses of its impact on European industrial sectors. This method focuses on the European manufacturing sectors, mainly on the role of each material. Finally, from the environmental point of view, the value of the indicator for the environmental country risk depends on several factors, such as the environmental performance index of the producing countries, sustainability of the raw material, and recycling rate. It means that producing countries might adopt regulations concerning the supply of raw materials to Europe to reduce their environmental impact. Therefore, this indicator assesses environmental issues, which may limit access to reserves or the supply of raw materials (EU Commission, 2010b).

This dissertation discusses critical materials selected in a two-stage approach: in the first stage materials are considered based on their significance to industry and technological development, e.g. phosphorus importance to food industry, niobium to renewable energy sector and automotive industry, and lithium to the electrification of transportation sector. The second stage identifies critical materials based on their high supply risk and low recycling rate from the circular economy perspective.

Therefore, three critical materials including phosphorus (P), niobium (Nb), and lithium (Li) are studied. Criticality of these materials is high if we look at it from several perspectives, such as clean technology requirements — e.g., advanced batteries and electric vehicles (EV); national security requirements — materials required to ensure the continuity of a country's fundamental operations and security; and general economic requirements — covering general industrial activities affecting the economy of the country (Jin et al., 2016).

Literature review carried out in the next chapter will be used to explain the importance of phosphorus, niobium, and lithium. The following have been the main reasons why these materials are selected for this dissertation:

- *Phosphorus (P)*: According to many evaluations, it was recognized as an important critical material (EU Commission, 2017a; Ober, 2018; Scholz and Wellmer, 2013). The main reasons for choosing phosphorus in this research are as follows: a) it is associated with a vital sector, i.e., production of fertilizers for agriculture and food industries, b) it is available on an oligopoly market, in which 58% of phosphorus and 44% of phosphate rock (PR) supply depends on one country - China, and c) it has a low end-of-life recycling input (around 15% in 2017) which will be a big challenge for the circular economy in the future.
- *Niobium (Nb)*: Different assessment bodies, such as U.S. Geological Survey (USGS) (Magyar and Petty, 2018), EU Commission 2014-2017 (EU Commission, 2010b, 2014, 2017a), and the United States National Research Council (NRC) committee (Graedel et al., 2012) have identified niobium as an important critical material. Also, the Ministry of Economy, Trade, and Industry (METI) and the Ministry of Foreign Affairs (MOFA) in Japan identified niobium as a highly critical metal (Diemer et al., 2018).
There are several reasons for choosing niobium for this study: a) it is associated with high-strength low-alloy (HSLA) steel and superalloys, which are very important components in automotive industry b) it is available on a monopoly market, i.e., 92% of niobium supplies depend on one country (Brazil); c) currently it has very low end-of-life recycling input in Europe, around 0.3% in 2017, which is a big challenge for the circular economy in the future (EU Commission, 2017a).
- *Lithium (Li)*: In a number of evaluations, lithium was catalogued as a critical mineral / material by distinct methods (Fortier et al., 2018; Magyar and Petty, 2018). Lithium has been identified as an important critical material in studies focused on vehicles (Mancini et al., 2013). According to criticality assessment carried out in Japan by METI and MOFA, lithium, together with other 30 minerals, was determined as a highly critical metal (Diemer et al., 2018).
The main reasons for selecting lithium for this dissertation are: a) it is essential for lithium-ion batteries (LIBs) which are very important for developing rechargeable batteries in different industries, such as portable electronic devices and electric vehicles b) its market structure is an oligopoly where four countries – Chile, Australia, Argentina, and China – account for more than 90% of global production of lithium; c) currently it has got a very low end-of-life recycling input (less than 1%) which is a big challenge for the circular economy in the future (Buchert et al., 2009; Wellmer and Hagelüken, 2015).

It is worth stressing that the degree of criticality of materials changes over time for several reasons, e.g. changes in the supply- and demand-side structure. Therefore, continuous updates of assessments of criticality of materials are needed.

1.2 Motivation behind the study

Many modern technologies require unique chemical and physical properties, which can be found in critical materials (Nuss et al., 2014). Critical materials in both the forward and reverse chains, from mining extraction to end-of-life product recycling, have impact on a global scale. Economic sectors in which critical materials are typically used, e.g. construction, automotive, metal and green technology industries are crucial to progress. Therefore, on the one hand, ensuring their continued availability is vital for future societal development (Savage, 2012). Moreover, global demand for critical material resources has increased ten-fold since 1900 and is expected to double by 2030 (Alves and Coutinho, 2015; Krausmann et al., 2009; Moss et al., 2011b). Keeping in mind this growing demand, Sverdrup et al. (2017) highlighted that many of the most critical materials for human society might be at risk of scarcity in the coming decades. Therefore, reliable and unconstrained access to critical raw materials is a growing concern within the world economy. On the other hand, ecological problems are the main drivers for choosing between virgin or recycled materials (Ferreira et al., 2012). Managers should therefore be conscious of possible environmental issues of the supply chain (SC) stages, e.g. fast increase in demand for LIBs for transport and industrial applications has increased worries about the negative environmental impact of battery manufacturing (Romare and Dahllöf, 2017; Xu et al., 2008). Environmental concerns influence the supply of CRMs because of environmental regulations and requirements (Glöser et al., 2015).

This dissertation is motivated by the wish to contribute to the growing literature on assessment of physical and environmental sustainability of critical materials such as phosphorus, niobium, and lithium. First, I analysed the physical limitations of supply of such materials from mining to production. Then, I discuss how the recycling stage prevents the shortage of CRMs. Also, I analysed environmental sustainability of CRMs recycling by considering energy consumption and greenhouse gas (GHG) emissions within the SC.

1.3 Problem

Demand for critical materials has increased abruptly over the past decades. It is expected to increase further over the next decades as a result of industrial development and population growth. According to literature, from linear economy perspective, the supply chain of CRMs generates several challenges such as rising prices of materials or limited opportunities to increase supply while by improving the efficiency of mining and processing we can get only short-term gains. Besides, the CRM supply chain in some cases additionally poses significant environmental concerns, such as environmental damage caused by extraction, landfilling, and waste disposal. In waste disposal, we face several issues. For example, CRMs contained in end of life (EOL) products are lost as oxides, or get diluted in recycling, so that their functional property is lost. In case of critical metals, there are some problems as dissolved ions of lead or chromium leak from landfills into the environment. Also, in case of non-metals, for example, excessive

presence of phosphorus in water leads to eutrophication and, ultimately, to the collapse of the entire ecosystem (Bradford-Hartke et al., 2015; Chen and Graedel, 2016; Xu et al., 2017). Therefore, the motivation for sustainable use of CRMs in the world is driven by the need to conserve resources inside the supply chain.

To address both supply risk and environmental issues, one of the possible solutions is to promote closing the loop for the materials by recycling end-of-life products. Circular economy represents the same approach to the increase of material recycling, waste reduction, and balancing economic growth with how we use the environment and its resources (Zhu et al., 2010). Hence, countries start to implement the principles of circular economy in industries and in the management of critical materials supply chain.

However, recycling cannot completely satisfy the demand for materials due to the efficiency of treatment technologies and the loss of some material stock. Generally, recycling can significantly reduce the need for mining, import dependency, and pressure on the environment (Pietrzyk-Sokulska et al., 2015). However, the environmental and economic issues made the network trade-off for decision makers about using recycling. Therefore, it is necessary to rethink the recycling network and move towards more sustainable recycling that becomes part of a circular economy. Efficient recycling may lead to a substantial decrease in the supply rates needed for society. This is, however, only a conditional part of what may constitute a solution (Graedel et al., 2011b). We need to look at sustainable recycling because past experiences demonstrate that efficiency gains are typically used to increase the overall quantity rather than save resources (Sverdrup et al., 2017).

Following the above discussion, there is a lack of analysis of critical material flows from mining to recycling in a holistic picture of supply chain. Moreover, there is a lack of assessment of environmental sustainability of critical material recycling from a long-term perspective.

1.4 Objectives of study and research questions

As mentioned in the previous section, recycling is primarily an industrial economic activity. This study emphasizes that a systematic view of recycling can deal with the complexity of interactions in the supply chain of critical materials in a long term. Therefore, the focus needs to be on rethinking recycling in lifecycle stages of products containing critical materials. This study aims to accomplish the following objectives to achieve environmentally sustainable recycling of critical materials:

- *Objective 1:* To determine the contribution of recycling of critical materials on decreasing the dependency on mining or imports.
- *Objective 2:* To analyze environmental concerns including energy consumption and GHG emissions of the recycling stage compared to other stages in supply chain.

To achieve the objectives mentioned above, this dissertation addresses the following research questions:

- *RQ 1*: To what extent can recycling mitigate the problem of criticality of materials?
- *RQ 2*: Can recycling mitigate the environmental concerns of the supply chain of critical materials?

1.5 Key definitions

A list of the key definitions used in the dissertation is presented in Table 1.

Table 1: List of definitions of terms.

Terminology	Definition	Reference
Circular economy	A system in which resource, waste, emission, and energy are minimized by closing, and narrowing the loops of material and energy.	(Geissdoerfer et al., 2017)
Recycling	A stage in a supply chain includes activities such as collection of wastes, sorting, processing, and using materials in the production of new products.	(King et al., 2006)
Sustainability	The balanced and systemic integration of economic, social, and environmental performance.	(Geissdoerfer et al., 2017)
System dynamics	A method of understanding the nonlinear behaviour of complex systems over time considering stocks, flows, interactions between various components and time delays.	(Forrester, 1997)
Supply Chain	“Supply chains are the multifaceted systems that comprise several independent establishments by different objects with focusing on the integration of all the factors from raw material to end products.”	(Özbayrak et al., 2007)
Raw material	Natural or processed resources to be used as an input into the production of semi-finished or finished products.	(EU Commission, 2017b)
Extraction stage	A stage includes the process of obtaining (extracting), mining or harvesting raw materials from our environment.	(EU Commission, 2017b)
Scrap	Materials contained in end of life products.	(EU Commission, 2017b)

Terminology	Definition	Reference
Primary/secondary raw material	Virgin materials, natural inorganic or organic substance used for the first time are considered primary raw materials. Secondary raw materials can be obtained from the recycling of end of life product.	(EU Commission, 2017b)

1.6 Structure of the dissertation

This dissertation includes two main parts. Part I consists of five chapters. The first chapter includes an introduction which describes the research background, the motivation of the study, problem, the objective of the study, research questions, key definition of terms, and structure of the dissertation. The second chapter discusses literature review related to the importance of critical materials such as phosphorus, niobium, and lithium. The third chapter explains the research methodology. System dynamics (SD), modelling process including causal loop diagram and stock and flow diagram are presented. Then mathematical formulas of the model are described. Next, primary data sources will be presented in this chapter. The fourth chapter summarizes key results of each published article included in this dissertation and discusses the analysis of recycling critical materials. Finally, the fifth chapter concludes by summarizing the main findings and suggestions for future research. Table 2 provides the structure of the dissertation in the input-output perspective. Part II includes four articles published in relation with this dissertation and their supplementary materials.

Table 2: Structure of the dissertation.

Chapter	Title	Input	Output
Chapter 1	Introduction	<ul style="list-style-type: none"> Background of the study Motivation of the study 	<ul style="list-style-type: none"> Problem Objectives Research questions
Chapter 2	Literature review	<ul style="list-style-type: none"> Challenges in supply chain of critical materials Phosphorus, niobium, and lithium as important critical materials 	<ul style="list-style-type: none"> Research gap Global dynamic market situation of critical materials Environmental sustainability of recycling of critical materials
Chapter 3	Research method	<ul style="list-style-type: none"> How can system dynamics method be used to analyze 	<ul style="list-style-type: none"> Applying system dynamics methodology in

Chapter	Title	Input	Output
		supply chain of critical materials?	analysis of supply chain of critical materials
Chapter 4	Publications and results	<ul style="list-style-type: none"> • Dynamic modeling of supply chain of critical materials • Environmental concerns of recycling of critical materials 	<ul style="list-style-type: none"> • Data collection • Recycling of phosphorus, niobium, and lithium • Assessment of energy use through all phases of SC in a long term • Assessment of GHG emissions through all phases of SC in a long term • Sustainability of recycling of critical materials
Chapter 5	Conclusions	<ul style="list-style-type: none"> • Answering research questions • Insight from a dynamic simulation model • Analysis from a long term perspective 	<ul style="list-style-type: none"> • Summary of the contributions of the study • Main findings • Managerial implications • Suggestion for future research

2 Literature Review

Recycling has emerged as a major factor in the development of closed-loop supply chains (Trochu et al., 2018). Recycling is very important in the case of critical materials, it is also in line with circular economy model (Busch et al., 2014). On the one hand, the role of recycling will be more highlighted due to concerns over future access to critical materials and their limited substitution in many applications. On the other hand, recycling would be very important due to minimizing waste and mitigating environmental issues through the supply chain of critical materials.

Building on literature review, this section explains the importance of critical materials such as P, Nb, and Li as well as the situation on their markets and recycling.

2.1 Importance of phosphorus as a critical raw material

Phosphorus is mainly obtained from the phosphate rock (PR) (El Wali et al., 2019). Phosphorus is one of the non-replaceable resources in food supply chain (Jacobs et al., 2017). It is an essential nutrient for all life forms, which plays a significant role in the bio-based economic processes that take place in the global economy (Golroudbary, El Wali, et al., 2019). Therefore, its sustainable supply is vital (Diallo et al., 2015).

Around 95% of global phosphate production goes to fertilizers and animal production (animal feed) (Van Vuuren et al., 2010). The remaining 5% of global phosphate production is used in a broad range of industrial applications, e.g. cleaning and water treatment, fire safety, electronics and batteries, lubricants, agro-chemicals, medical applications, and human food additives including several applications where phosphorus compounds are non-substitutable (EU Commission, 2017a).

PR is finite and non-renewable. Besides, it should be noted that the growing world population is the main driver of phosphorus consumption. The ever-increasing need for food leads to an increased demand for fertilizers based on phosphorus for agricultural use. Mainly, these issues have led to a high-level supply risk and economic importance for phosphorus, which makes it an important critical raw material.

2.1.1 Market situation of phosphorus

Phosphorus demand will continue to rise in the future owing to an increasing worldwide population, nutritional changes and an increasing proportion of biofuels (Cordell, Schmid-Neseta, et al., 2009). An extensive literature has been written on the limited availability of phosphorus, and there are widespread concerns that phosphorus production will soon peak (Cordell, Drangert, et al., 2009). Globally, phosphate rock reserves are limited to a few particular regions in the world. Figure 1 shows the top phosphate producers in the world in 2018, based on the USGS data source (Ober, 2017). Such geological distribution of phosphorus may influence fertilizer and food prices

(Schoumans et al., 2015), or may disrupt the continuous supply of phosphorus (de Ridder et al., 2012). The major geographic concentration of PR in a few countries — such as China, Morocco and Western Sahara, United States, Russia, Jordan and Brazil — is also raising in vulnerability of areas like Europe, which depends on imports of P, up to 90% (EU Commission, 2017a). The demand for phosphorus, mining of P, and its rapid loss from terrestrial ecosystem are globally increasing at the same time. As a result, there is a great threat to the security of phosphorus supply and its sustainability (Smil, 2000).

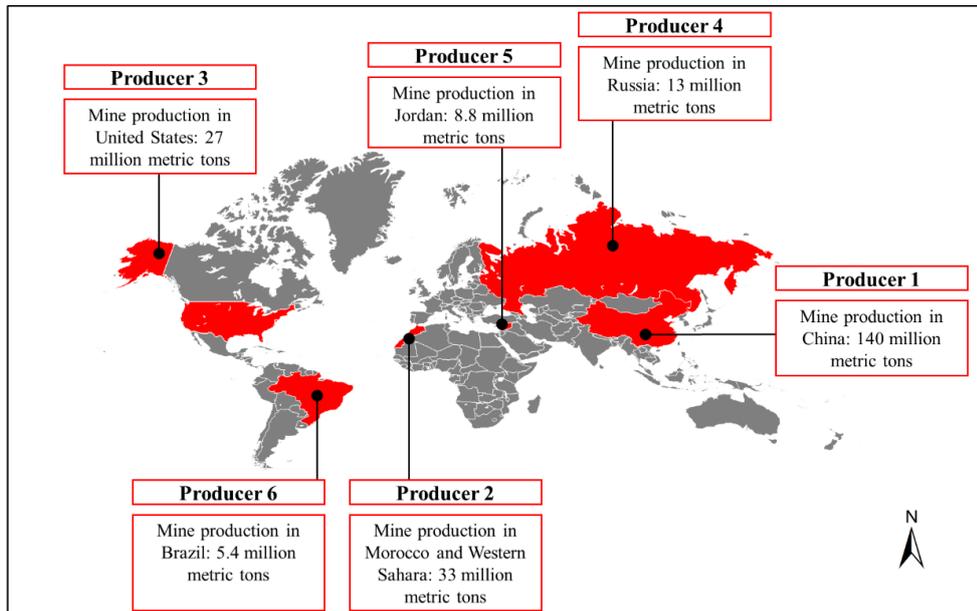


Figure 1: Top phosphate-producing countries in 2018.

2.1.2 Significance of phosphorus recycling

Most of the phosphorus usage in the world is linear and there is a considerable inefficiency in its production and use (Reijnders, 2014; Van Vuuren et al., 2010). Ecological, geopolitical, and economic issues of phosphorus demand its recycling (Cooper et al., 2011; Cordell, Drangert, et al., 2009). Hence, there is a worldwide trend towards improving phosphorus recovery from various waste streams.

It has been shown that worldwide collaboration is urgently needed to recycle and reuse phosphorus (Dawson and Hilton, 2011; Elser and Bennett, 2011). Hence, phosphorus recycling must be regarded as an essential aspect of phosphorus management strategies. Otherwise, there will be a significant proportion of phosphorus permanently lost in the

waste streams. To tackle this problem, we should use phosphorus in circular way and thus immediate measures and policies that contribute to the recycling of phosphorus should be put in place (Cornel and Schaum, 2009). For example, one of the major priorities of the EU 2020 Strategy is the improvement of recycling and sustainable phosphorous management (EU Commission, 2010a; Fischer and Kjaer, 2012). Also, new strategies regarding recycling are being developed in Europe to reduce phosphorus import dependency owing to the above-mentioned problems (Van Dijk et al., 2016; Scholz et al., 2013; Schoumans et al., 2015; Withers, Elser, et al., 2015). Therefore, tracking phosphorus reversibility has become a main research area for sustainable phosphorus supply chain management (SCM). In this regard, phosphorus recycling has already been suggested as one of the feasible solutions to mining (Childers et al., 2011; Cordell, Drangert, et al., 2009; Roy, 2016). Both solid waste and wastewater provide an outstanding chance to use phosphorus more efficiently (Kalmykova et al., 2012; Mateo-Sagasta et al., 2015).

Practically, numerous works have recognized the need to develop new techniques for phosphorous recovery (Caddarao et al., 2018; Suzuki et al., 2007; Zou and Wang, 2016). Several research studied the feasibility of attaining a greater level of phosphorus recycling, e.g. Shu et al. (2006) and Cordell (2010). In Europe, the rate of phosphorus recycling is very low. For instance, approximately 37% of P is presently recovered from municipal sewage sludge and reused in agriculture (Fischer and Kjaer, 2012). New procedures and technologies are under development to recover phosphorus from excess manure (Schoumans et al., 2010). In Sweden, the improvement of P recycling is expected to be one of the major alternatives to satisfy at least 30 percent of demand (Cordell, 2010). The current reliance on imported PR ('3 kg P per European citizen per year') cannot be sustained for a very long time. The survey conducted by Stockholm Environment Institute (SEI) demonstrated that in order to achieve sustainability in each phase of the agricultural and food SC phosphorus use effectiveness must be close to 100%. Therefore, full recycling of phosphorous should be suggested in Europe (Schroder et al., 2010).

2.1.3 Environmental sustainability of phosphorus recycling and research gap

Environmental performance at each stage of phosphorus SC is one of important dimensions in assessing its sustainability. Also, the effect of energy use on supply chain environmental sustainability is well known (Azadeh and Arani, 2016). It is demonstrated by the depletion of non-renewable energy resources and GHG emissions. It should be noted that fast increases in agricultural production have a significant effect on the growth of phosphorus mining and recycling (Wu et al., 2017). The scale of phosphorus SC motivates to evaluate its effect on the environment, especially by making a quantitative assessment of energy consumption and GHG emissions.

Considering the issues mentioned above, there are two research gaps regarding recycling of phosphorus. First, it is not determined to what extent phosphorus recycling mitigates the dependence on primary supply resources. Second, in the face of the significance of phosphorous recycling, its environmental sustainability is not widely researched.

Therefore, we need to find out and learn whether phosphorus recycling is an environmentally sustainable alternative.

2.2 Importance of niobium as a critical raw material

Niobium (Nb) is an essential alloying element for the production of steels and superalloys, electronic components, superconductors, medical implants, construction and petroleum industry (Rahimpour Golroudbary et al., 2019). Niobium is one of the most highly critical materials since only a few countries in the world produce this metal used in strategic energy technologies, such as the production of nuclear or wind energy, carbon capture and storage (Moss et al., 2011a). The lack of substitutes of niobium and its oligopoly market create a high supply risk of this element (Achzet et al., 2011; EU Commission, 2010b; Nuss et al., 2014; Ober, 2018). Also, the peak of the production and scarcity of niobium is estimated to take place after 2020 (Sverdrup and Olafsdottir, 2018).

2.2.1 Market situation of niobium

The global market of niobium has been increasing annually by 10% since 2000 (Alves and Coutinho, 2015; Mackay and Simandl, 2014; Zednicek et al., 2002). As shown in Figure 2, niobium resources and production are concentrated in Brazil, which accounts for 92% of the world's supply. Canada is the second largest producer of niobium (Ober, 2018). These limited reserves of niobium in the world have led other regions, e.g., Europe to 100% dependency on imports. Significant importers of niobium include Germany, the United States, Japan, and China (Sustainability, 2017).

Around 89% of niobium is used as a micro alloy for high-strength steels (HSS) alloy. Global demand for ferroniobium has increased at a compound average growth rate of 6.9% per year since 2001. Currently, about 10% of steel produced globally contains niobium, which is expected to increase by about 20% in the future (Rahimpour Golroudbary et al., 2019).

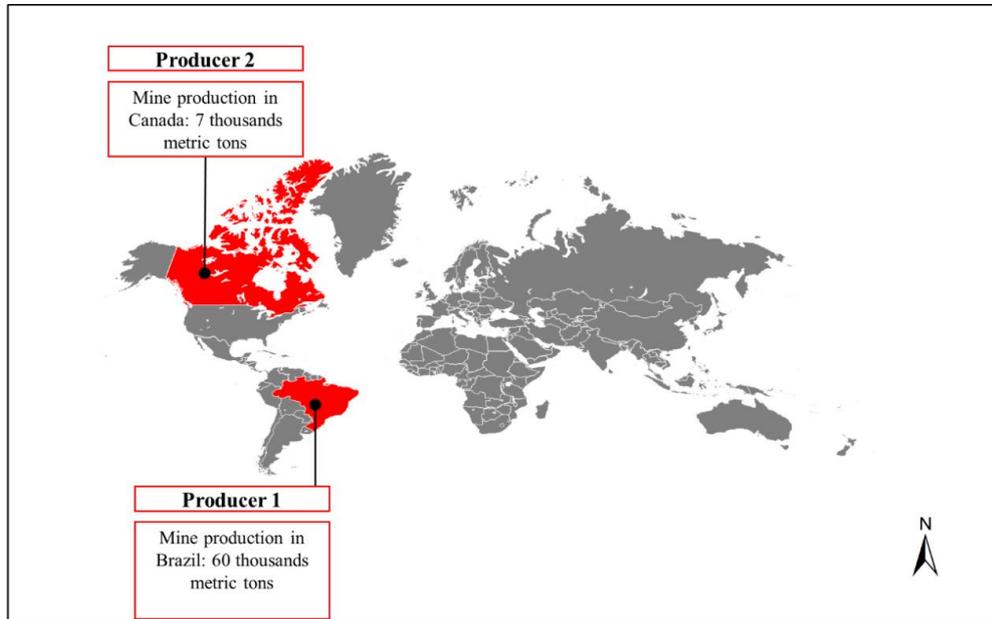


Figure 2: Top niobium-producing countries in 2018.

2.2.2 Significance of niobium recycling

Given the rapid increase in primary and secondary niobium production over the last 15 years (Mackay and Simandl, 2014), the production rate will peak in 2025 with around 60% recycling rate (Sverdrup et al., 2017). A detailed review conducted by the United Nations Environmental Program estimated end-of-life recycling rate and recycling content of niobium to be more than 50% between 2000 and 2005 (Graedel et al., 2011b). Such recycling rate results from the fact that the elements used in major amounts in recoverable products have high recycling rates (e.g., niobium in end-of-life vehicles (ELVs)). However, the recycling rate is quite low for the elements used in minor quantities in complicated products, e.g., tantalum in electronics (Graedel and Reck, 2014).

Recycling of products containing niobium contributes to the recovery of a significant amount of this metal from waste. Hence, recycling of niobium is a key strategy for its sustainable future. In other words, niobium recycling is a measure to mitigate adverse impacts of raising demand and exploit the potential of its impact on economic growth. For instance, ELVs have become a significant waste stream in the world (Widmer et al., 2015). Hence, the maximization of recyclability is one of the main trends in the automotive industry. It contributes significantly to the reduction of waste. Also, it constitutes a substantial source of critical raw materials such as niobium. For instance, the total number of ELVs was around 40 million in 2010, mainly in countries such as

Germany, France, Italy, UK, Spain, Canada, USA, Brazil, China, Japan, Australia, and Korea (Sakai et al., 2014).

It is worth mentioning that the concentration of Nb in steel alloy is on average low, generally lower than 0.1 wt % (Schulz et al., 2017). However, a significant amount of niobium used in typical cars (around 36,000 tonnes) is estimated by considering the number of ELVs in 2010. The calculation shows a quite significant amount of available niobium from secondary sources compared to 49,100 tonnes of niobium mined globally (Jaskula, 2013). In addition, the annual production of niobium alloyed steel was estimated at around 50 million tonnes globally (Patel and Khul'ka, 2001). The steel content in a passenger car will increase from 54% to 64% in 2020 (Sullivan et al., 2010). Therefore, niobium can be recovered from waste metals and scrap, which account for up to 20% of total supply (Rahimpour Golroudbary et al., 2019).

2.2.3 Environmental sustainability of niobium recycling and research gap

For several decades, supply chain research has attempted to address the issues of environmental impact and social sustainability (Brandenburg et al., 2014; Eskandarpour et al., 2015; Hutchins and Sutherland, 2008). Moreover, new areas of research have been studied focused on developing sustainability of the supply chain. From this perspective, several studies have investigated the available options for achieving both higher economic growth and lower GHG emissions (Mercure et al., 2016; Mirzaei and Bekri, 2017). In this case, the effect of energy use on sustainability and its impact on generating GHG emissions are very significant issues across all stages of the supply chain (Azadeh and Arani, 2016; Kuipers et al., 2018). However, the question remains what proportion of energy consumption and GHG emissions could be saved through niobium recycling also seen as a step towards ensuring environmental sustainability. Therefore, there is a gap in literature for quantitative analysis of supply chain of niobium including its mass flow, energy consumption and GHG emissions.

2.3 Importance of lithium as a critical raw material

Lithium plays an important role in the development of a low-carbon economy (de Koning et al., 2018). Lithium compounds are also used in systems such as, among others, military, communication, and especially in the production of several industrial applications such as ceramics and glasses, lubricants and greases, pharmaceutical products, and aluminium products (Martin et al., 2017).

Globally, the size of lithium product markets is estimated as follows: 35% for batteries, 32% for ceramics and glass, 9% for lubricating greases, 5% for air treatment and 5% for continuous casting mould flux powders, 4% for polymer production, 1% for primary aluminium production, and 9% for other uses. In recent years, the consumption of lithium for batteries has increased significantly due to the extensive use of rechargeable lithium batteries in electric vehicles, portable electronic devices, and electric tools (Ober, 2017).

Temporary relief in metal markets may lead to price rises or even to a period of upcoming supply shortages. For a variety of metals, such as lithium, there are presently first indications of increasing prices (Graedel and Reck, 2016). Therefore, lithium is considered an important critical material from many assessments.

2.3.1 Market situation of lithium

Because lithium cannot be substituted in most applications, therefore its supply risk is influenced by demand increase. It is anticipated that demand for lithium will be growing by 8–11% annually (Martin et al., 2017). For example, lithium use in battery industry increased from zero to 80% of the market share between 1991 and 2012. The leading consumer of lithium is China with 35% of the total global consumption, followed by Europe, Japan, the Republic of Korea, and North America with 24%, 12%, 10%, and 9%, respectively (Lv et al., 2018). Furthermore, the demand for lithium will increase significantly in the forthcoming years, mainly owing to the rapid growth in demand for lithium-ion batteries (Hao, Liu, et al., 2017; Lv et al., 2018). Consequently, in the coming years, this phenomenon will result in several challenges to the suppliers. Hence, the identification of the capacity of each supplier is of primordial importance to evaluate if it will be possible to meet the increasing demand in the coming future (Pehlken et al., 2017).

Figure 3 shows the top lithium-producing countries in 2018 which are: Australia, Chile, China, and Argentina. Global production of lithium increased slightly in 2015 to meet the growing demand for lithium represented by battery industry. Lithium manufacturing in Argentina, for instance, grew by 17% and only slightly in Chile and Australia. Major manufacturers of lithium anticipated around 32,500 tons of worldwide lithium consumption in 2015, an increase of 5 percent from 31,000 tons in 2014. After 2014, lithium carbonate prices rose by about 10-15 percent due to enhanced global demand.

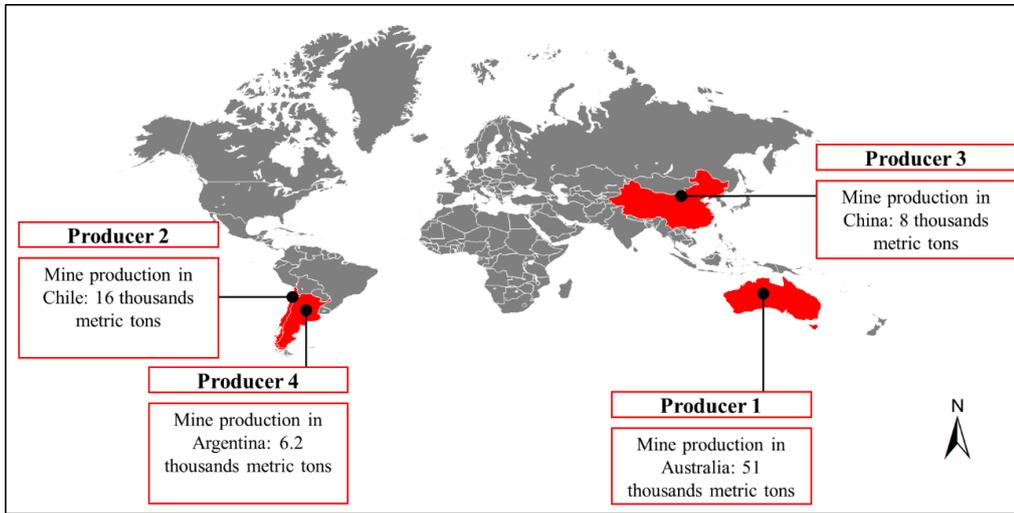


Figure 3: Top lithium-producing countries in 2018.

2.3.2 Significance of lithium recycling

Historically, there was no functional recycling of lithium, as its separation from end of life products was not possible or it was very costly. Lithium was either sent to disposal with other materials or recycled in a large magnitude material stream. Lithium recycling was therefore very low but steadily improved owing to the increase in lithium battery consumption.

Several studies have confirmed that the improvement of waste management systems globally and developing new lithium recycling technologies are crucial (Sun et al., 2017). In other studies, several aspects of lithium recycling, especially from LIBs, have been discussed. Previous studies have shown that the extension of automobiles lifetime increases the quantity of material in the stock. Consequently, we will face a small EOL flow of LIBs annually (Wang and Wu, 2017). In addition, the complex chemical interaction between various materials implies high energy consumption (Gratz et al., 2014). However, from mass flow perspective, recycling of lithium could become very significant when lithium batteries reach the end of life between 2020 and 2025. On the other hand, LIBs recycling is essential from the environmental point of view. It prevents hazardous effects produced by heavy metals included EOL products. Therefore, the importance of Li recycling lies not only in the conservation of resources, but also in avoiding environmental burdens (Ordoñez et al., 2016).

2.3.3 Environmental sustainability of lithium recycling and research gap

In literature, applications in batteries are marked as “zero emissions”. However, according to Dunn et al. (2012), no consideration is given to emissions generated by the production of batteries in the supply chain. Battery manufacturing is one of the major contributors to air pollution through the production of EVs (Notter et al., 2010).

Environmental concerns are the main drivers for choosing between either virgin ores or recycled materials (Ferreira et al., 2012). Therefore, in various phases of lithium SC, decision-makers should be conscious of possible undesirable environmental impacts. In this case, the quick increase of LIBs demand for transportation and industrial application has caused a growing concern regarding the negative environmental impact of their production (Xu et al., 2008). Considering potential ecological regulations, environmental issues may disturb the security of supply for CRMs (Glöser et al., 2015).

Mitigation of environmental problems, e.g. high energy consumption and GHG emissions within the supply chain, enables industries to gain a competitive advantage (Basiri and Heydari, 2017). However, reducing emissions at a specified phase of the supply chain can be harmful as it may raise emissions in other phases, e.g. by using more emission-intensive equipment (Modaresi et al., 2014). The need to evaluate particular changes in the supply chain requires a system thinking approach. It involves detailed analysis of dynamic interactions between various components over time.

Detailed assessment of the dynamic life cycle of lithium as well as the flow of consumed energy and generated emissions is essential for measuring its supply chain sustainability. In literature, various aspects of lithium life cycle have been researched (Lv et al., 2018). However, a holistic view has not yet been given of the life cycle of energy consumption and GHG emissions from all phases of the lithium SC and their change over time. Several studies included environmental assessment of Li by focusing on the production of various kinds of LIBs. They primarily have a product view and concentrate on just one phase of SC, e.g. the manufacturing phase (Hao, Mu, et al., 2017) or recycling stage (Dunn et al., 2015; Rahman and Afroz, 2017). The absence of appropriate systemic environmental analysis for lithium recycling exacerbates their supply chain's threat of environmental impacts. A broader knowledge of the worldwide lithium cycle is therefore needed.

One of the primary gaps in literature is its failure to discuss a dynamic lithium SC model to assess energy consumption and GHG emissions at all stages. It is also essential to realise whether lithium recovery from waste, such as spent LIBs, can contribute to long-term energy savings.

3 Research methodology

After having collected knowledge from literature, dynamic models were designed using system dynamics methodology to examine problems identified in the project. Then, the required quantitative data were collected from research papers, data sheets, and open sources data banks. Finally, different scenarios were designed to assess the supply chain of critical materials. Next, each step will be explained in detail.

3.1 System dynamics method

Systems dynamics (SD) is a methodology that focuses on modelling complex systems as is the case with supply chain. Supply chain operations and activities are the function of a number of main factors that often seem to have a solid interrelationship. Several reasons such as understanding and analysing the interactions between many factors over time, as well as providing the feedback for each part of the system make this methodology very useful for modelling supply chain networks (Towill, 1996). A framework for transforming the system from a mental model to a computer-based level and rate model is given in this method. Then, the model is used for further experiments based on several constructed scenarios. Results are discussed against the behaviour exhibited in different conditions.

The main reason why system dynamics modelling is used in this dissertation is because of its ability to answer the following questions:

- How a technical system co-evolves with changes in society, environment and economy?
- What unexpected behavior of the supply chain could be observed in the long-term?
- How to react to such unexpected conditions?

Forrester (1997) defines, examines, and clarifies several subjects relating to the supply chain management by the expansion and use of system dynamics methodology. Using the method of system dynamics introduced by Forrester (1997), this dissertation assists in optimizing environmentally sustainable operations in designing and modelling the supply chain of critical materials.

To address this objective, all processes along the stages of a SC, such as mining, processing, production, use, waste, and recycling have to be analysed and modelled. The systematic view employed in dynamic modelling to assess recycling stage to secure environmentally-friendly supply of critical materials from secondary sources. Three main layers of the supply chain of critical materials were considered in the analysis — mass flow, energy consumption and GHG emissions.

3.2 Dynamics modelling

According to Richardson and Pugh (1981, 1989), system dynamics modelling is a system stage method that starts and ends with understanding. The structure of system dynamics model contains; a) understanding of a system, b) problem definition, c) system conceptualization, d) mathematical formulation, e) simulation model, and f) policy analysis (Golroudbary and Zahraee, 2015; Richardson and Pugh, 1981).

The application of a SD model is based on a clearly defined cause for concern. It is motivated by the need to enhance the understanding of the system. The definition of the problem sets the study's focus. After identifying and characterizing the problem over a set time, a dynamic model is designed to address the relevant issue. To create insights into strategies to enhance system behaviour, an appropriate dynamic model is vital. Next, it is converted into a simulated quantitative representation. Finally, the policy analysis phase seeks to transfer the ideas and understanding acquired from the system dynamics models to those who may be concerned. It is essential to note that the method of modelling between phases is iterative and the steps continue in a conceptual way. Results of any step can provide ideas that lead to the revision and modification of the previous step.

3.2.1 Understanding the system (causal loop diagram)

One of the key diagrams in SD modelling is a casual loop diagram (CLD), which helps in the understanding of the system structure. Variables are rarely independent because they generally have solid interrelationships and the impact is one-way in most cases (Golroudbary, Zahraee, et al., 2019; Hatami et al., 2014; Kamrani et al., 2014). This creates a loop, in which variables affect each other (Özbayrak et al., 2007). Therefore, a CLD aims to identify significant factors influencing the system and the causal effect among them. A CLD comprises of arrow-connected variables that denote the hypotheses of the model to represent the feedback structure of systems (Sternan, 2000). A causal loop includes positive or negative feedback interrelationships. For example, in this dissertation, Figure 4 shows the causal loop diagram representing the dynamic models of supply chain of critical materials.

The system considered in this dissertation for each critical material focuses on the complex intervention between stages in the SC (mining, processing, production and recycling). I analysed the global supply chain of CRMs based on their application in most significant industries. Figure 4 demonstrates a general CLD of the supply chain of critical materials. This diagram helps in the development of the model as the first step of causal hypotheses. A causal link demonstrates how variables affect each other in the model. Next to the arrowhead, the positive (+) or negative (-) signs show if linked components change in the same or reverse direction. For example, the increasing *demand from market* causes higher *demand from industry* while its decrease produces lower *demand from industry*. Hence, market and industrial demands have a positive relationship. In this dissertation, demand is defined as a difference between the amount of critical material that is available and required in various processing and manufacturing sectors over a year. Therefore, total

demand for a critical material equals existing supply of this material which is available in different sectors of production and deficit of material based on the consumption of products.

The main feedback loops are also identified in the diagram. There are two types of feedback loops: the reinforcing loop, which is the source of growth or the accelerating collapse, and the balancing loop, which exhibits a goal-seeking behaviour. As shown in Figure 4, *B* sign displays five balancing feedback loops. Also, there is one reinforcing feedback loop presented by *R*. The loops are as follows:

- *B*₁ as the first balancing feedback loop includes variables such as *mining of critical materials, processing, manufacturing, waste generation, collection of wastes, recycling of used products, and recovery of critical materials.*
- *B*₂ as the second balancing feedback loop corresponds to *processing, manufacturing, demand from market, and demand from industry.*
- *B*₃ as the third balancing feedback loop includes *manufacturing and demand from market.*
- *B*₄ as the fourth balancing feedback loop consists of two variables such as *processing and demand from industry.*
- *B*₅ as the fifth balancing feedback loop is based on variables such as *waste generation, collection of wastes, and recycling of used products.*
- The reinforcing loop (*R*₁) includes *recovery of critical materials, manufacturing, waste generation, collection of wastes, and recycling of used products.*

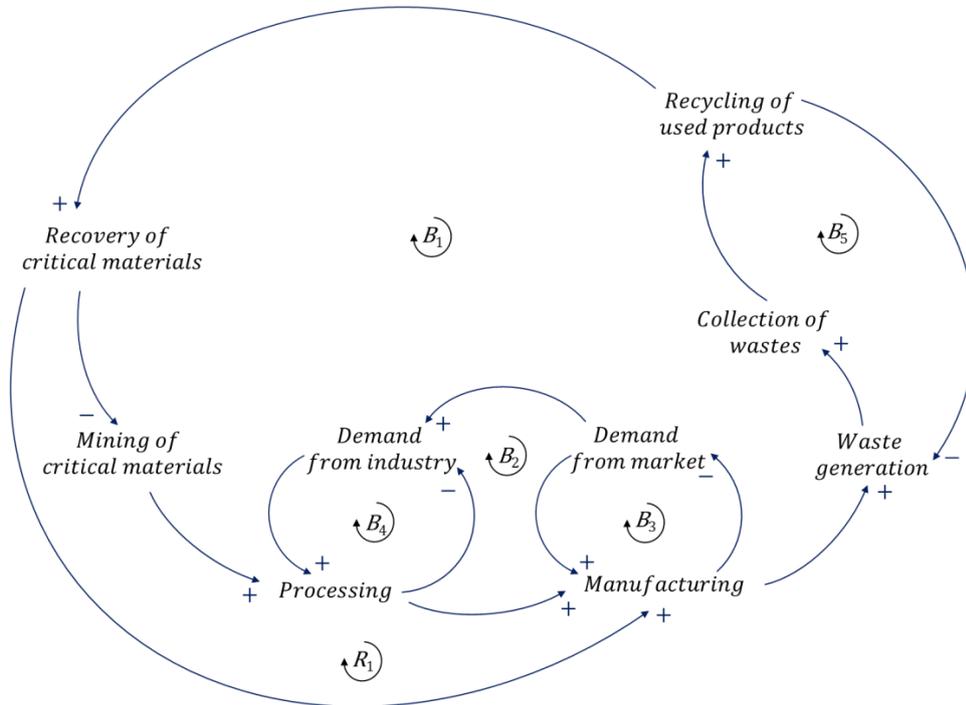


Figure 4: Causal loop diagram for supply chain of critical materials.

3.2.2 Problem definition

Considering the causal loop diagram (Figure 4), the problem of shortage of critical materials is intensified by increasing the demand and restricting supply sources (e.g., mining and recovery of materials from end of life products). Consequently, the system faces higher demand for materials and energy consumption, as well as environmental challenges such as GHG emissions. The reinforcing loop (R_1) in Figure 4 highlights that it is necessary to examine the supply chain of critical materials from environmental perspective, including energy consumption and GHG emissions, in terms of sustainability. This is the result of the interrelationship between mass flow and energy consumption which leads to GHG emissions. The use of recycling and recovery to supply a certain part of required critical materials does not guarantee the reduction of energy needed for different production processes and air emissions that they generate. The R_1 reinforcing loop shows the continuous increase in mass flows between stages. As a result, the required energy will increase in different stages of the supply chain. It means that the supply chain tries to meet the demand by supplying critical materials from different sources (mining or recycling); however, the question remains which source of supply guarantees smaller energy consumption and GHG emissions in the long-term.

3.2.3 System conceptualization

After recognizing the main variables and their interactions in all stages of critical materials (P, Nb, and Li) life cycle, their structural models are designed (Figure 5, 6, and 7). The models show three primary layers which are material and energy flows, as well as GHG emissions. Material flow includes several modules such as suppliers, mining, processing, manufacturing, use, collection, recycling, and CRMs recovery. Energy flow shows energy used at each stage, i.e., mining and processing, production, and recycling. Finally, the GHG emissions layer is mainly associated with energy flow; hence, its structure is identical.

Several industries, such as fertilizers, food and feed additives, detergent, and other uses are considered in the model of the global supply chain of phosphorus (Figure 5). In the model of the global supply chain of niobium (Figure 6), the focus is on high strength steel alloys applied in the automotive industry. Finally, in the global supply chain of lithium (Figure 7), the focus of analysis is on LIBs manufacturing.

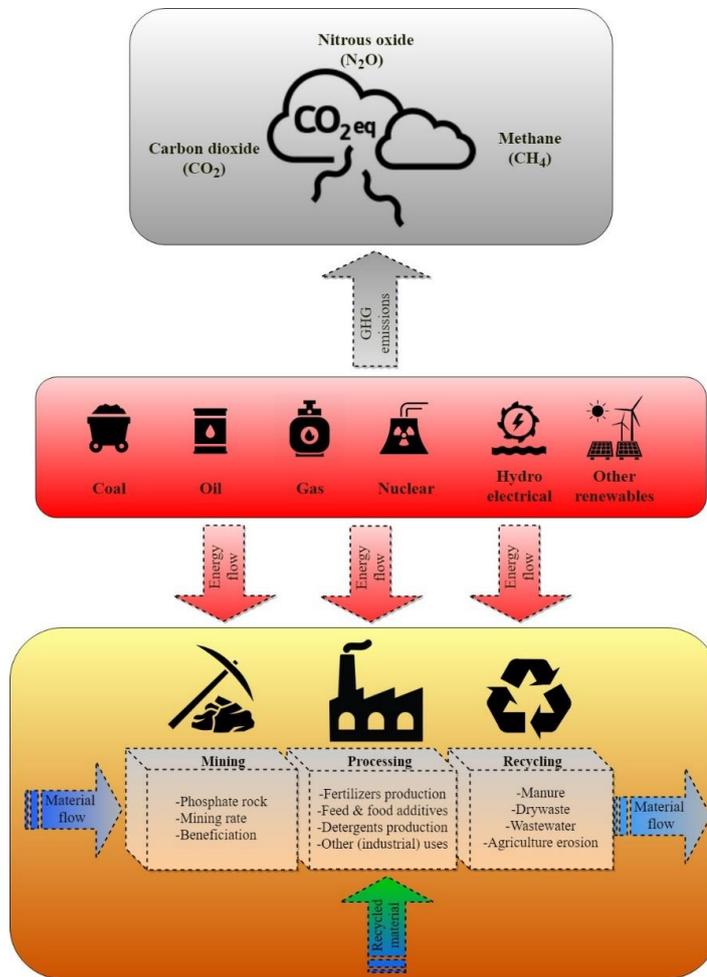


Figure 5: Model of global supply chain of phosphorus (Golroudbary, El Wali, et al., 2019).

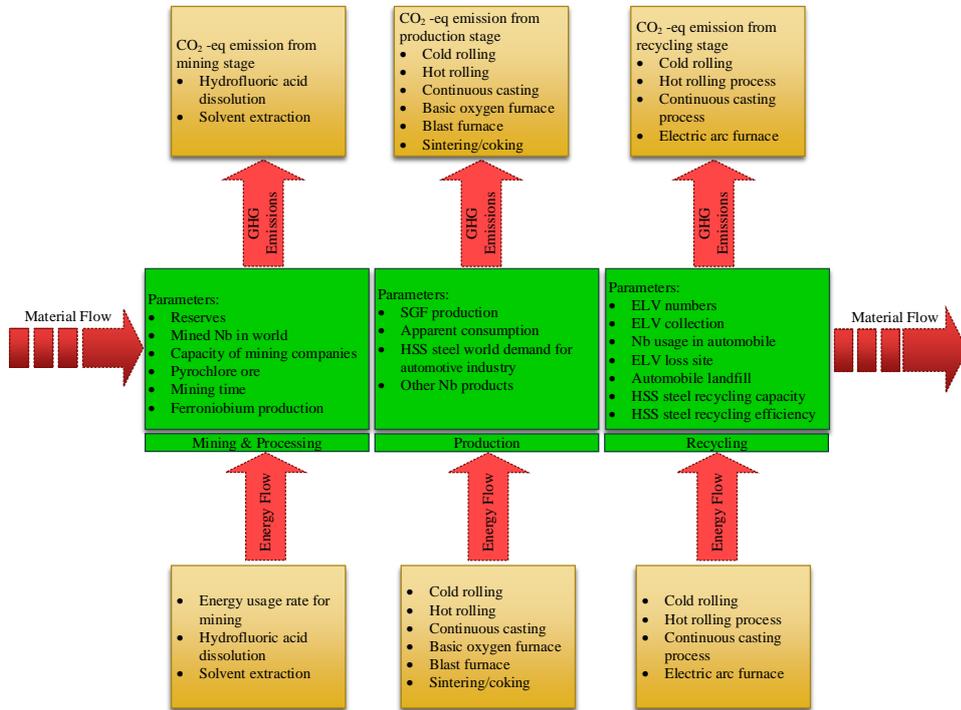


Figure 6: Model of global supply chain of niobium (Rahimpour Golroudbary et al., 2019).

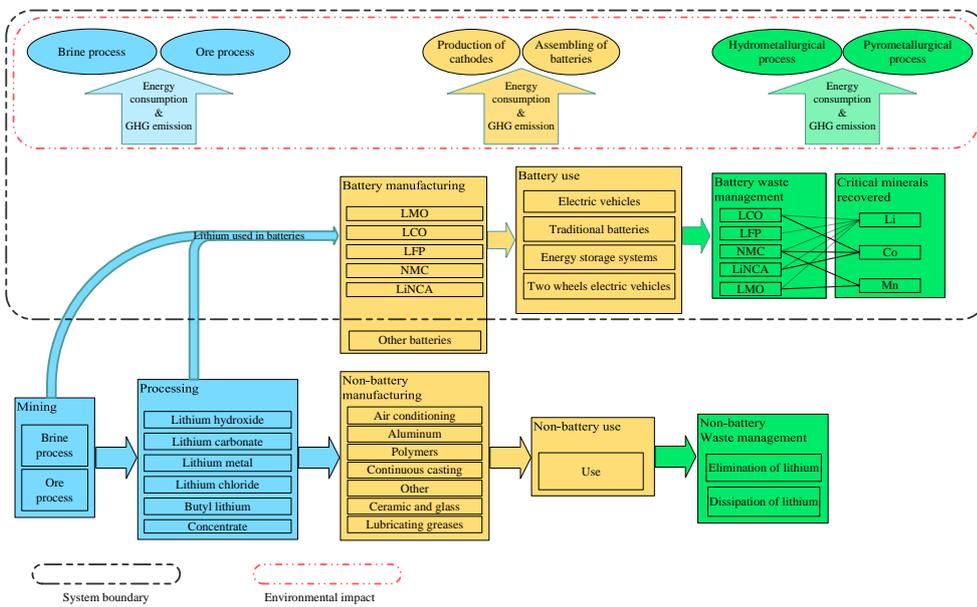


Figure 7: Model of global supply chain of lithium.

3.2.4 Simulation model (stock and flow diagram)

Next step in modelling consists in the transformation of the causal loop diagram and conceptual model into rate and level diagram, which in the systems dynamics methodology is called stock and flow diagram (SFD) (Özbayrak et al., 2007). A SFD is used to study the characteristics of the process. First of all, it implies analysing the dynamic features between variables of rate and level. Then, the connections are described between the model's factors. In order to run model simulations, these connections are used to create mathematical equations. In this study, dynamic model simulation was conducted using the AnyLogic (University 8.3.3) software as a leading simulation tool for SCM.

Besides the availability of AnyLogic software for a broad range of researchers, it has been used in many research studies focusing on the simulation and modelling of different stages of the supply chain, especially environmental sustainability analysis (Golroudbary, Calisaya-Azpilcueta, et al., 2019; Golroudbary, El Wali, et al., 2019; Rahimpour Golroudbary et al., 2019; El Wali et al., 2019). AnyLogic has been designed to simulate a model for one or more variables over time. It offers a graphical interface for modelling complex environments, the opportunity to test and explore various scenarios, as well as observe changes in the system behaviour over time at a detailed level. Also, it gives the opportunity for making a more precise forecast by capturing details across different processes. Therefore, AnyLogic software was chosen to fulfil the objectives of this dissertation by constructing a dynamic simulation.

The simplified dynamic model structure and the interrelationships among the main variables are presented in Figure 8. The general stock and flow diagram for supply chain of critical materials is shown in Figure 8. Based on the conceptual model of each material (P, Nb and Li), the SFD designed and developed separately, which are presented in the articles published in this dissertation. Variables classified as stocks, flows, connectors, and converters can be seen in Figure 8. Level or stock variables (symbolized by rectangles) represent the state of the system, accumulating the difference between input and output flows of the box. Flow variables (symbolized by valves) are the rate of change in stock variables over time. They constitute operations that either fill the stocks or drain them. Converters are intermediate variables used for various calculations (symbolized by circles). Lastly, the connectors (symbolized by simple arrows) are the connections within the model structure that represent the dependencies between factors.

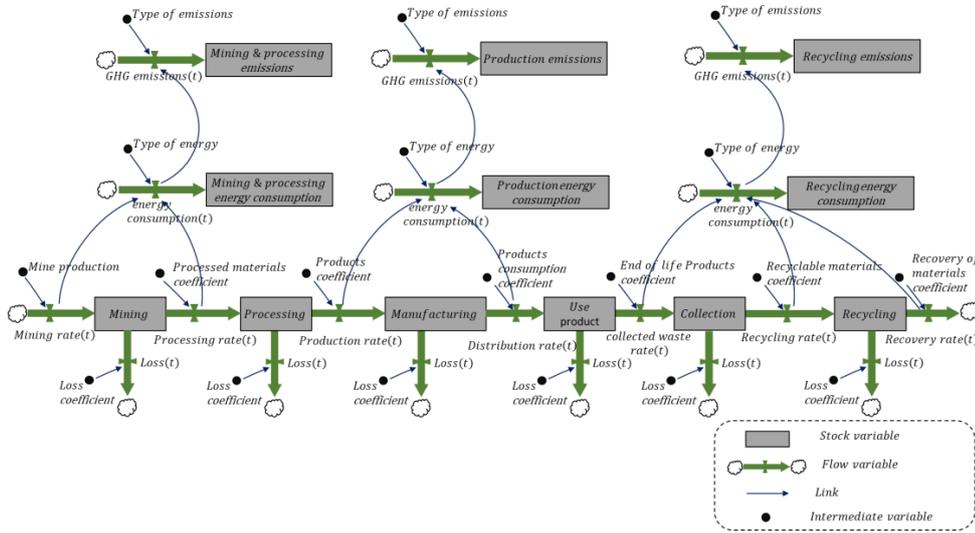


Figure 8: Stock and flow diagram of global supply chain of critical materials.

3.2.5 Mathematical formulation

The next stage in the SD methodology is the creation of the mathematical model. The SFD is converted into a collection of differential equations that are solved through simulation. Continuous system dynamics models are equal to differential equations for flows and integral equations for stocks (Sterman, 2000). Stocks integrate the flows, modifying and complicating the dynamics involved in the system (Barlas, 2007). Therefore, the formulas for all stock variables in the proposed system dynamics models are given by a time integral. Stocks accumulate or integrate their flows. It means that *stock* at time t equals the initial value of *stock* at time $t = 0$ plus the integral of the flows into *stock* minus the flows out of *stock* (Kirkwood, 1998). For example, according to the Figure 8, the dynamic behaviour of the level variables such as *mining*, *processing*, *manufacturing*, *use product*, *collection*, *recycling*, *mining & processing energy consumption*, *production energy consumption*, *recycling energy consumption*, *mining & processing emissions*, *production emissions*, and *recycling emissions* are calculated by a time integral of the inflows minus the outflows.

The overall equation of stocks and flows after Forrester (1997) is as follows:

$$Stock(t) = \int_{t_0}^t [Inflow(t) - Outflow(t)]dt + Stock(t_0) \quad (1)$$

where t_0 corresponds to the initial year, t represents the final year and $Stock(t)$ is a mass accumulated by $Inflow(t)$ and loss $Outflow(t)$ at the moment t of the given period.

$$Inflow(t) = f(Stock(t), V(t), P); Outflow(t) = f(Stock(t), V(t), P) \quad (2)$$

where, $V(t)$ is an exogenous variable in time t . For example, according to Figure 8, the *mining rate* relies on the quantity of material extracted from *mine production*. P is a set of parameters of the system, e.g. *processed materials coefficient*, *products coefficient*, *products consumption coefficient*, *end of life products coefficient*, *recyclable material coefficient*, *recovery of materials coefficient*, and *loss coefficient*.

All equations formulated for different stocks and flows were derived from Equations 1 and 2 (for detailed model and mathematical formulation, see the publications and their supplementary materials).

3.2.5.1 Calculation of energy consumption

The boundary model deals with the consumption of energy and materials in processes taking place at all stages of the supply chain. Energy consumption is calculated for three main stages of the supply chain of critical materials: mining, production and recycling.

In this study, I do not aggregate various types of energy, but consider electricity coming from various sources which I explain below.

For the phosphorus supply chain, during the mining of phosphate ores, energy consumption is mainly associated with the use of mining machines and equipment. That equipment includes draglines, pumps, pit cars, and equipment necessary for water treatment, pumping, beneficiation and transport of raw material to phosphate processing sites. In the production stage of the phosphorus supply chain, energy demand for feed and fertilizers production are estimated separately. In the recycling stage of the phosphorus supply chain, the analysis is limited to the estimation of energy demand for the recycling of phosphorus from waste streams coming from wastewater, food, animal, human excreta, and households.

For the niobium supply chain, in the mining stage required energy in hydrofluoric acid dissolution and solvent extraction are considered. In the production of HSS steels for automotive industry, six main processes (cold rolling, hot rolling, continuous casting, basic oxygen furnace, blast furnace, sintering/coking) are considered. In the recycling stage of HSS steels from ELVs, energy required in cold rolling, hot rolling, continuous casting, and electric arc furnace processes are calculated.

For the lithium supply chain, in the mining stage, the energy required for ore and brine processes is considered. Production of LIBs for different industries such as EV, energy storage system, traditional batteries, and two wheel electric vehicles is analysed in the model to quantify energy consumption. In recycling of LIBs, pyro-metallurgical process is the main technology that is used at near commercial scale followed by hydrometallurgical process. To calculate energy consumption, I considered different types of energy sources including non-fossil fuel, coal fuel, natural gas fuel, fossil fuel, petroleum fuel, as well as renewable, nuclear and biomass energy sources.

Generally, the calculation of energy consumption in a dynamic model is based on the mass flow of the material and sources of energy which are used through the processes.

Considering the energy required for processes in all stages of critical material supply chain, I calculated the total cumulative energy consumption.

Equation 3 shows the calculation corresponding to energy consumption in phosphorus supply chain:

$$EC_m(t) = P_m(t) \times \sum_{n=1}^6 \sigma_{m,n} \quad (3)$$

where, $EC_m(t)$ is energy consumption in stage m in the year t for the period 2000-2050; $m= 1,2,3$ represents stage of the phosphorus supply chain: mining, processing, and recycling; $n= 1,2,\dots,6$ corresponds to the type of energy obtainable from different energy sources: coal, nuclear, gas, oil, hydroelectricity and other renewables; $P_m(t)$ is the amount of phosphorus in stage m in the moment t ; $\sigma_{m,n}$ is the energy required per one ton of phosphorus flow in a given stage m from each energy source $n= 1,2,\dots,6$ (coal, nuclear, gas, oil, hydroelectricity and other renewables).

Equations 4, 5 and 6 show calculations corresponding to energy consumption in niobium supply chain:

Energy consumption in the mining stage of the niobium supply chain is calculated as follows:

$$E_{M,R}(t) = Nb_{M,R}(t) \times \sum_{n=1}^2 \sigma_n \quad (4)$$

where, $E_{M,R}(t)$ is energy consumption in the mining stage in the year $t = 2000,2001,\dots,2050$. $Nb_{M,R}(t)$ is the amount of material in the mining flow in the year t . σ_n is the energy (gigajoule) required per one tonne of niobium in the mining stage through each process $n = 1,2$ which corresponds to the amount of energy required in hydrofluoric acid dissolution and solvent extraction.

Energy consumption in the production stage of niobium supply chain is estimated as follows:

$$E_{P,R}(t) = Nb_{P,R}(t) \times \sum_{n=1}^6 \alpha_n \quad (5)$$

where, $E_{P_R}(t)$ is energy consumption in the production stage in the year $t = 2000, 2001, \dots, 2050$. $Nb_{P_R}(t)$ is the amount of material in the production flow in the year t . α_n is the energy (gigajoule) required per one tonne of niobium in the production stage through each process $n = 1, 2, \dots, 6$ which corresponds to six main processes: cold rolling, hot rolling, continuous casting, basic oxygen furnace, blast furnace, and sintering/coking.

Energy consumption in the recycling stage of niobium supply chain is estimated as follows:

$$E_{R_R}(t) = Nb_{R_R}(t) \times \sum_{n=1}^4 \rho_n \quad (6)$$

where, $E_{R_R}(t)$ is energy consumption in the recycling stage in the year $t = 2000, 2001, \dots, 2050$. $Nb_{R_R}(t)$ is the amount of material in recycling flow in the year t . ρ_n is energy (gigajoule) required per one tonne of niobium flow in the recycling stage through each process $n = 1, 2, \dots, 4$ which corresponds to the amount of energy required in cold rolling, hot rolling, continuous casting, and electric arc furnace.

Equation 7 shows calculations corresponding to energy consumption in different processes of the lithium supply chain:

$$E_f(t) = M_f(t) \times \sum_{n=1}^8 \sigma_n \quad (7)$$

Where, $E_f(t)$ is the flow of energy consumption in different processes such as mining, processing, production and recycling in the year $t = 2015, 2016, \dots, 2025$. $M_f(t)$ is the amount of material in the flow in the year t . σ_n is the energy (gigajoule) required per one tonne of LCE through each process from n type of energy sources ($n = 1, 2, \dots, 8$). Different types of energy sources include non-fossil fuel, coal fuel, natural gas fuel, fossil fuel, petroleum fuel, and energy generated from renewable and nuclear sources and biomass.

The main limitations to the calculation of energy consumption in this dissertation come from the fact that external consumption of energy was not considered, and I failed to analyse the demand for energy in transportation along the supply chain.

3.2.5.2 Calculation of greenhouse gas emissions

The GHG emission sub-system is primarily related to energy consumption and therefore the structure of both subsystems is identical.

Equation 8 shows GHG flows in the phosphorus supply chain as follows:

$$GHG_{x,m}(t) = EC_m(t) \times \sum_{n=1}^6 \delta_{x,n} \quad (8)$$

where, $GHG_{x,m}(t)$ represents greenhouse gas emissions of type $x = 1, 2, 3$ (CO₂, CH₄ or N₂O) in m stage of the supply chain, $m = 1, 2, 3$ (mining, processing, and recycling) in the year t of the period 2000-2050; $EC_m(t)$ is the energy consumption in stage m in the year t ; $\delta_{x,n}$ is the amount of greenhouse gas intensity (ton CO₂-eq) as type x per one joule of energy consumed using the n resource, $n = 1, 2, \dots, 6$ (coal, nuclear, gas, oil, hydroelectricity and other renewables).

In the niobium supply chain, the GHG emissions from the mining stage are estimated based on energy consumption in hydrofluoric acid dissolution and solvent extraction. They are assessed as follows (Eq. 9):

$$GHG_{M,R}(t) = E_{M,R}(t) \times \sum_{n=1}^2 \gamma_n \quad (9)$$

where, $GHG_{M,R}(t)$ represents greenhouse gas emissions (CO₂-eq) for the mining stage in the year $t = 2000, 2001, \dots, 2050$. $E_{M,R}(t)$ is the energy consumption in the mining stage in the year t . γ_n is the amount of greenhouse gas intensity (tonne CO₂-eq) for each process $n = 1, 2$ (hydrofluoric acid dissolution and solvent extraction).

The GHG emissions from the niobium production stage are estimated based on energy consumption in cold rolling, hot rolling, continuous casting, basic oxygen furnace, blast furnace, and sintering/coking. In the third submodel, they are assessed as follows (Eq. 10):

$$GHG_{P,R}(t) = E_{P,R}(t) \times \sum_{n=1}^6 \beta_n \quad (10)$$

where, $GHG_{P,R}(t)$ represents greenhouse gas emissions (CO₂-eq) for the production stage in the year $t = 2000, 2001, \dots, 2050$. $E_{P,R}(t)$ is energy consumption in the production stage in the year t . β_n is the amount of greenhouse gas intensity (tonne CO₂-eq) from each process $n = 1, 2, \dots, 6$ (cold rolling, hot rolling, continuous casting, basic oxygen furnace, blast furnace, and sintering/coking).

The GHG emissions from the niobium recycling stage are estimated based on energy consumption in cold rolling, hot rolling, continuous casting, and electric arc furnace. They are assessed as follows (Eq. 11):

$$GHG_{R,R}(t) = E_{R,R}(t) \times \sum_{n=1}^4 \lambda_n \quad (11)$$

where, $GHG_{R,R}(t)$ represents greenhouse gas emissions (CO₂-eq) for the recycling stage in the year $t = 2000, 2001, \dots, 2050$. $E_{R,R}(t)$ is energy consumption in the recycling stage in the year t . λ_n is the greenhouse gas emitted from each process $n = 1, 2, \dots, 4$ (cold rolling, hot rolling, continuous casting, and electric arc furnace).

Equation 12 shows calculations corresponding to GHG emissions in different processes of the lithium supply chain:

$$GHG_f(t) = E_f(t) \times \sum_{n=1}^8 \gamma_n \quad (12)$$

Where, $GHG_f(t)$ represents the flow of greenhouse gas emissions (CO₂-eq) for each process in the year $t = 2015, 2016, \dots, 2025$. $E_f(t)$ is the flow of energy consumption in different processes such as mining, processing, production and recycling in the year t . γ_n is the amount of greenhouse gas intensity (tonne CO₂-eq) from each process by using different type of energy sources $n = 1, 2, \dots, 8$ (non-fossil fuel, coal fuel, natural gas fuel, fossil fuel, petroleum fuel, renewable, nuclear sources and from biomass).

3.3 Data collection

The next step to construct the simulation model would be collecting data for the rate variables and constant variables that occur in the model. Therefore, the required data are collected from several sources within the global supply chain of critical materials. All input data corresponding to the models are presented in publications and materials supplementary to them.

3.4 Scenario design

Dynamic models are used in supply chain to investigate how the system behaves when conditions such as demand and supply are changing over time (Zahraee et al., 2014, 2019). In this situation, scenario studies which are created for different circumstances are particularly helpful.

In Publication I (El Wali et al., 2019), I have analyzed different alternatives for phosphorus recycling improvement and its impact on criticality of P in Europe. In Publication II (Golroudbary, El Wali, et al., 2019), I assessed various phosphorus consumption scenarios and their worldwide environmental impact. In Publication III (Rahimpour Golroudbary et al., 2019), different scenarios were assumed for the growth rate of ELVs. Those scenarios are used to demonstrate changes in cumulative energy

consumption and GHG emissions in the automotive industry from primary and secondary niobium HSS alloy manufacturing.

4 Publications and review of the results

This chapter summarizes the objectives, research questions, results and contributions of four publications of this dissertation.

4.1 Publication I: Impact of recycling improvement on the life cycle of phosphorus

4.1.1 Overview

The European Union (EU) has only a few phosphorus deposits (de Ridder et al., 2012). This restricted phosphorus availability could possibly create supply problems for European governments and companies (Van Dijk et al., 2016).

In Publication I, European perspective on phosphorus management was analysed. In this paper, I analysed the range of possible improvement of recycling of phosphorus in the long term and its impact on the P criticality in Europe until 2050. The main reason is the increasing vulnerability of this region as it depends on phosphorus imports. Also, in literature, there is not enough quantitative analysis to assess the impact of recycling improvement on phosphorus life cycle over time.

The proposed dynamic model focuses on the recycling of wastewater, solid waste and manure for P recovery. To estimate the current quantity of phosphorus recycling in Europe (EU-28 as a case of study in this paper), the different processes were considered as the input of the model. For example, the processes for P recovery from wastewater assumed anaerobic digestion. The processes for P recovery from solid waste assumed composting. Recycling from manure in agriculture is focused on anaerobic digestion and composting. They are the most often used methods for phosphorus treatment and recycling (Cesaro et al., 2015; Schoumans et al., 2015).

A SD model was built and implemented to analyse distinct situations in order to quantify the impacts of recycling effectiveness on phosphorus life cycle. Three scenarios help to explore how phosphorous flows in Europe change by improving recycling of P from solid waste, wastewater, and manure. Various possible methods of P recycling could be considered for future estimation of improvement of phosphorous recycling. For example, the processes for P recycling from sludge could be electrolytic process and nanofiltration, from wastewater - enhanced biotechnological approaches and from solid waste - acid and base leaching precipitation procedures.

4.1.2 Objectives and research questions

The first objective of Publication I is to estimate the actual level of phosphorus recycling from waste streams and estimate its trend in the future. Then, another objective is to assess

the role of recycling in the reduction of the reliance on P imports in Europe and its potential benefits.

To achieve the above-mentioned objectives, this article addresses the first research question of this dissertation:

- To what extent can recycling mitigate the problem of the criticality of materials?

Therefore, I designed a dynamic model for analysing phosphorus flows in Europe over the years 2000-2050.

4.1.3 Results and discussion

The results of Publication I indicate that the reduction of about 1 million tonnes (mt) P annually from total phosphorus imports can be achieved by improved recycling. As a result, this contributes to the reduction of the EU's dependence on P imports by about 17%. Figure 9 presents trends in recycling and import flows of phosphorus in different scenarios between 2000 and 2050.

Three recycling scenarios (15%, 30%, and 45%) were assumed to assess the impact of recycling improvement on phosphorus life cycle. Those scenarios define a range of future possibilities for the improvement of phosphorus recycling. Technological possibilities of economic recycling of phosphorus are the main reason for considering the range 15-45%. Recently introduced techniques, such as struvite fertilizers (Kabbe et al., 2015; Kabbe and Kraus, 2017), enable 15% phosphorous recycling. Therefore, economic phosphorus recycling is likely to reach the rate of 50% over the next 12 to 15 years (Scholz and Wellmer, 2018).

Scenario A (15%) and scenario B (30%) were chosen to examine the trends of P flows at different levels of recycling improvement. These trends correspond to cumulative and annual estimations of recycling and imports in the EU. I assumed the maximum rate of recycling improvement to be 45% (scenario C). The main reason for this scenario was the physical limitation of recycling. Based on the results, more than 45% improvement in recycling efficiency would not be physically possible. It would result in an unbalanced system.

The results of this publication could contribute to decision making at a continental scale where choices are intended to confront the challenge of the phosphorus SC in the EU. Moreover, the results of this paper show the trends in phosphorous flows in 28 EU Member States, dynamic interactions between multiple sectors, limits of recycling, the estimation of recycling and the EU reliance on phosphorus imports in different scenarios until 2050. Phosphorus flows from non-food and non-agricultural industries are taken into consideration in Publication I. The model introduces the effect of the P flow dynamic features and EU population demographic changes. The proposed model estimates the

quantity of P mined, imported, exported, produced, consumed, lost, and recycled. Figure 9 shows the trends of P in different scenarios between 2000 and 2050.

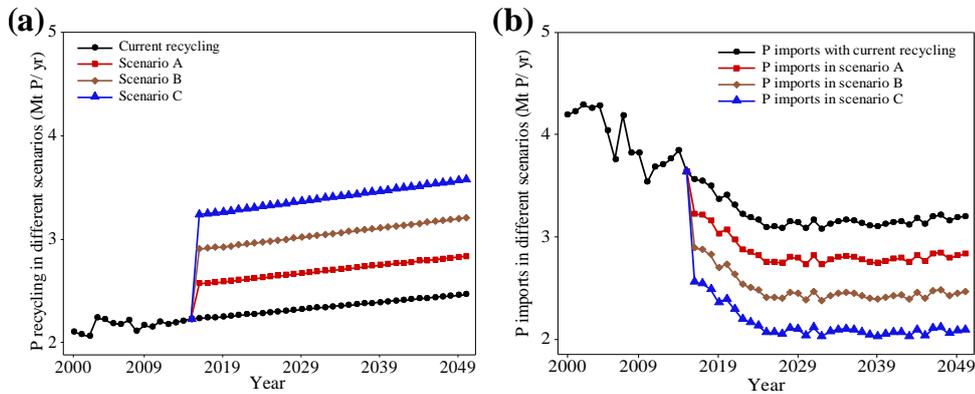


Figure 9. Estimations of P annual flows in different scenarios (El Wali et al., 2019).

4.1.4 Contributions

The EU Commission has identified high level of dependence on imports of P (EU Commission, 2017a). Their assessment relates, however, to the PR trade particularly. The findings of Publication I indicate the general level of dependence on P imports for 28 EU Member States by considering various industries, including mining, phosphorus processing into phosphoric acid, agricultural manufacturing, and manufacturing of detergents. The suggested dynamic model could be helpful for policymakers to be aware of the present P recycling level. Also, it helps to explore different alternatives for further phosphorus recovery improvements. The proposed model quantifies the prospective effect on the phosphorous supply chain with future innovations in recycling technology.

4.1.5 Clarification of further details for paper I

- Continuous system dynamics models are mathematically equivalent to differential and integral equations for flows and stocks, respectively (Sterman, 2000). Stocks integrate the flows, modifying and complicating the dynamics involved (Barlas, 2007). Therefore, the formula for all stock variables in the purposed system dynamics model are given by a time integral. Stocks accumulate or integrate their flows. It means that the stock at time t is equal to the initial value of the stock at time $t = 0$ plus the integral of the flows into the stock minus the flows out of the stock (Kirkwood, 1998). In this regard, Equation 2 in paper I describes the rate of change of the flows as their function in a system. It means Equation 2 helps us to understand how a flow grows or shrinks over time. Differential equation models

are often additionally specified by providing the values of the state variables at a specific point in time.

- In the model proposed in paper I, the initial amount of stock for total imports to the life cycle of phosphorus ($TIS(t_0)$) in the year 2000 is assumed zero. Therefore, the cumulative amount of total P imports to the system ($TIS(t)$) calculated by a time integral of the mined P import rate, phosphoric acid import rate, fertilizers import rate, the detergents import rate, the crops import rate, the food additives import rate, the feed additives import rate, the livestock import rate, the animal food import rate, the fish products import rate and the iron ores import rate.
- I divided the variables of the simulation model in paper I into two groups to further specify a model boundary as follows:
 - Endogenous variable — dynamic variables affect and are affected by other system variables in the feedback loops of the system.
 - Exogenous variable — Variables which are not directly affected by the system

As shown in Tables 3 and 4, I have also specified the type of variables (stock, flow and auxiliary).

Table 3: Endogenous variables for the proposed phosphorus supply chain modelling

Variable	Term	Unit	Type of Variables
$MS(t)$	Mined P stock	Tonnes P	Stock
$M_R(t)$	Mining rate	Tonnes P/yr	Flow
$B_R(t)$	Beneficiating rate	Tonnes P/yr	Flow
$BS(t)$	Beneficiated P stock	Tonnes P	Stock
$PS(t)$	Phosphoric acid stock	Tonnes P	Stock
$P_R(t)$	Processing rate	Tonnes P/yr	Flow
$FEP_R(t)$	Fertilizer production rate	Tonnes P/yr	Flow
$FEA_R(t)$	Feed production rate	Tonnes P/yr	Flow
$FA_R(t)$	Food additives production rate	Tonnes P/yr	Flow
$DP_R(t)$	Detergents production rate	Tonnes P/yr	Flow
$DS(t)$	Detergents stock	Tonnes P	Stock
$FS(t)$	Fertilizers stock	Tonnes P	Stock
$IP_R(t)$	Fertilizers from iron ores flow rate	Tonnes P/yr	Flow
$IPP_R(t)$	Iron ores production rate	Tonnes P/yr	Flow

Variable	Term	Unit	Type of Variables
$DF_R(t)$	Deposition flow rate	Tonnes P/yr	Flow
$CP_R(t)$	Crop production rate	Tonnes P/yr	Flow
P_5	Production variable for crops	Tonnes P/yr	Flow
$GC_R(t)$	Grass cut rate	Tonnes P/yr	Flow
$FAS(t)$	Feed Stock	Tonnes P	Stock
$CS(t)$	Crops Stock	Tonnes P	Stock
$CF_R(t)$	Crops to feed rate	Tonnes P/yr	Flow
$NC_R(t)$	Net crops flow rate	Tonnes P/yr	Flow
$CFO_R(t)$	Crops flow rate to food	Tonnes P/yr	Flow
$FSH_R(t)$	Fish flow rate	Tonnes P/yr	Flow
$FSHP_R(t)$	Fish production rate	Tonnes P/yr	Flow
$NFSH_R(t)$	Net fish catch rate	Tonnes P/yr	Flow
$AFP_R(t)$	Animal food production rate	Tonnes P/yr	Flow
P_{12}	Production variable for animal food	Tonnes P/yr	Flow
$FAS(t)$	Food additives stock	Tonnes P	Stock
$MI_R(t)$	Mined P import rate	Tonnes P/yr	Flow
$PI_R(t)$	Phosphoric acid import rate	Tonnes P/yr	Flow
$IPI_R(t)$	Iron ores import rate	Tonnes P/yr	Flow
$FI_R(t)$	Fertilizers import rate	Tonnes P/yr	Flow
$FEI_R(t)$	Feed import rate	Tonnes P/yr	Flow
$CI_R(t)$	Crops import rate	Tonnes P/yr	Flow
$DI_R(t)$	Detergents import rate	Tonnes P/yr	Flow
$FAI_R(t)$	Food additives import rate	Tonnes P/yr	Flow
$FSHI_R(t)$	Fish import rate	Tonnes P/yr	Flow
$MI_R(t)$	Mined P import rate	Tonnes P/yr	Flow
$LI_R(t)$	Livestock import rate	Tonnes P/yr	Flow
$ME_R(t)$	Mined P export rate	Tonnes P/yr	Flow
$PE_R(t)$	Phosphoric acid export rate	Tonnes P/yr	Flow
$IPE_R(t)$	Iron ores export rate	Tonnes P/yr	Flow
$FE_R(t)$	Fertilizers export rate	Tonnes P/yr	Flow
$FEE_R(t)$	Feed export rate	Tonnes P/yr	Flow
$CE_R(t)$	Crops export rate	Tonnes P/yr	Flow
$FAE_R(t)$	Food additives export rate	Tonnes P/yr	Flow

Variable	Term	Unit	Type of Variables
$DE_R(t)$	Detergents export rate	Tonnes P/yr	Flow
$FSHE_R(t)$	Fish export rate	Tonnes P/yr	Flow
$LE_R(t)$	Livestock export rate	Tonnes P/yr	Flow
$LS(t)$	Livestock	Tonnes P	Stock
$AFS(t)$	Animal food stock	Tonnes P	Stock
$FEC_R(t)$	Feed consumption rate	Tonnes P/yr	Flow
$FOC_R(t)$	Food consumption rate	Tonnes P/yr	Flow
$VFD_R(t)$	Vegetable food Demand rate	Tonnes P/yr	Flow
$AFD_R(t)$	Animal food demand rate	Tonnes P/yr	Flow
$DC_R(t)$	Detergent consumption rate	Tonnes P/yr	Flow
$FOS(t)$	Food Stock	Tonnes P	Stock
$FAC_R(t)$	Food additives consumption rate	Tonnes P/yr	Flow
$NCS(t)$	Net crops stock	Tonnes P	Stock
$AW_R(t)$	Animal waste rate	Tonnes P/yr	Flow
$HE_R(t)$	Human excreta flow rate	Tonnes P/yr	Flow
$SWS(t)$	Solid wastes stock	Tonnes	Stock
$NSW_R(t)$	Non treated solid waste rate	Tonnes P/yr	Flow
$TSW_R(t)$	Treated solid waste flow rate	Tonnes P/yr	Flow
$TSW(t)$	Treated solid waste stock	Tonnes P/yr	Stock
$SWL_R(t)$	Solid waste landfilling rate	Tonnes P/yr	Flow
$WWS(t)$	Wastewater stock	Tonnes P	Stock
$CWW_R(t)$	Wastewater collecting rate	Tonnes P/yr	Flow
$CWW(t)$	Collected wastewater	Tonnes P	Stock
$FW_R(t)$	Food waste generation rate	Tonnes P/yr	Flow
$TWW_R(t)$	Wastewater treating rate	Tonnes P/yr	Flow
$TWW(t)$	Treated wastewater stock	Tonnes P	Stock
$SL_R(t)$	Sludge landfilling rate	Tonnes P/yr	Flow
$SO_R(t)$	Sludge flow rate for other uses	Yonnes P/yr	Flow
$MR_R(t)$	Manure recycling rate	Tonnes P/yr	Flow

Variable	Term	Unit	Type of Variables
$SWR_R(t)$	Solid waste recycling rate	Tonnes P/yr	Flow
$WWR_R(t)$	Wastewater recycling rate	Tonnes P/yr	Flow
$TR_R(t)$	Total recycling rate	Tonnes P/yr	Flow
$ML_R(t)$	Mining loss rate	Tonnes P/yr	Flow
$PML_R(t)$	Phosphorus mining, beneficiating and processing loss rate	Tonnes P/yr	Flow
$BL_R(t)$	Beneficiation loss rate	Tonnes P/yr	Flow
$PL_R(t)$	Processing waste rate	Tonnes P/yr	Flow
$PR_R(t)$	Phosphorus runoff rate	Tonnes P/yr	Flow
$FL_R(t)$	Food loss rate	Tonnes P/yr	Flow
$AFL_R(t)$	Animal food loss rate	Tonnes P/yr	Flow
$CL_R(t)$	Crops loss rate	Tonnes P/yr	Flow
$FSHL_R(t)$	Fish loss rate	Tonnes P/yr	Flow
$NWW_R(t)$	Non treated wastewater rate	Tonnes P/yr	Flow
$UWW_R(t)$	Uncollected wastewater rate	Tonnes P/yr	Flow
$TL_R(t)$	Total loss rate	Tonnes P/yr	Flow

Table 4: Exogenous variables for the proposed phosphorus supply chain modelling (Type of variables: Auxiliary)

Variable	Term	Unit
k_0	P content in phosphate rock	%
k_1	P content in phosphoric acid	%
k_2	P content in P_2O_5	%
k_3	P content in food additives	%
k_4	P content in STPP	%
k_5	P content in iron ores	%
k_6	P content in Feed	%
$k_{7(1)}$	P content in Rice	Tonnes P/ tonnes crop
$k_{7(2)}$	P content in Wheat	Tonnes P/ tonnes crop
$k_{7(3)}$	P content in Barley	Tonnes P/ tonnes crop

Variable	Term	Unit
$k_{7(4)}$	P content in Sorghum	Tonnes P/ tonnes crop
$k_{7(5)}$	P content in Millet	Tonnes P/ tonnes crop
$k_{7(6)}$	P content in Rye	Tonnes P/ tonnes crop
$k_{7(7)}$	P content in Rye	Tonnes P/ tonnes crop
$k_{7(8)}$	P content in Oat	Tonnes P/ tonnes crop
$k_{7(9)}$	P content in Potato	Tonnes P/ tonnes crop
$k_{7(10)}$	P content in sweet potato	Tonnes P/ tonnes crop
$k_{7(11)}$	P content in Soybean	Tonnes P/ tonnes crop
$k_{7(12)}$	P content in Rapeseed	Tonnes P/ tonnes crop
$k_{7(13)}$	P content in Olive	Tonnes P/ tonnes crop
$k_{7(14)}$	P content in Sugar beet	Tonnes P/ tonnes crop
$k_{7(15)}$	P content in Sugar crane	Tonnes P/ tonnes crop
$k_{7(16)}$	P content in seed cotton	Tonnes P/ tonnes crop
$k_{7(17)}$	P content in Fruit	Tonnes P/ tonnes crop
$k_{7(18)}$	P content in vegetable and melon	Tonnes P/ tonnes crop
k_8	P content in fish	%
k_9	P content in Livestock	Tonnes P/ tonnes livestock
k_{10}	P content in Food waste	gP /Kg waste
$k_{11(1)}$	P content in cattle manure	%
$k_{11(2)}$	P content in Sheep manure	%
$k_{11(3)}$	P content in poultry manure	%
$k_{11(4)}$	P content in Horse manure	%
$k_{11(5)}$	P content in swine manure	%
M_0	Mining variable	Tonnes P rock/ yr
M_1	Beneficiating coefficient	Dimensionless
M_2	processing coefficient	Dimensionless
P_0	Production variable for fertilizers	Tonnes P ₂ O ₅ / yr
P_1	production variable for feed	Tonnes feed/ yr
P_2	production variable for food additives	Tonnes polyphosphates /yr
P_3	production variable for detergents	Tonnes P ₂ O ₅ / yr
P_4	production variable of phosphates from iron ores	Tonnes Iron ores/ yr
P_5	production variable for crops	Tonnes crops/ yr

Variable	Term	Unit
P_6	Grass cutting coefficient	Dimensionless
P_7	Crops flow coefficient to feed stock	Dimensionless
P_8	Deposition variable	Tonnes P/ yr
P_9	net crops flow coefficient	Dimensionless
P_{10}	Crops flow coefficient to food	Dimensionless
P_{11}	Fish production variable	Tonnes P/ yr
P_{12}	Animal food production variable	Tonnes P/ yr
I_0	Mined P import variable	Tonnes P rock/ yr
I_1	Phosphoric acid import variable	Tonnes phosphoric acid/yr
I_2	Import variable for iron ores	Tonnes iron ores/ yr
I_3	Import variable for fertilizers	Tonnes P_2O_5 / yr
I_4	Import variable for feed	Tonnes feed/ yr
I_5	Import variable for crops	Tonnes crops/ yr
I_6	Import variable for food additives	Tonnes polyphosphates/ yr
I_7	Import variable for STPP	Tonnes STPP/ yr
I_8	Fish import variable	Tonnes fish/ yr
I_9	Import variable for livestock	Tonnes livestock /yr
E_0	Mined P export variable	Tonnes P rock/ yr
E_1	Phosphoric acid export variable	T phosphoric acid/yr
E_2	Export variable for iron ores	Tonnes iron ores/ yr
E_3	Export variable for fertilizers	Tonnes P_2O_5 /yr
E_4	Export variable for feed	Tonnes feed/ yr
E_5	Export variable for crops	Tonnes crops/ yr
E_6	Export variable for food additives	Tonnes polyphosphates/ yr
E_7	Export variable for detergents	Tonnes STPP/yr
E_8	Export variable for fish	Tonnes fish/ yr
E_9	Export variable for livestock	Tonnes livestock /yr
C_0	Feed consumption coefficient	Dimensionless
C_1	Detergent consumption coefficient	Dimensionless
C_2	Demand of vegetable food coefficient per one person	Kg P/capita/yr
C_3	Demand of animal food coefficient per one person	Kg P/capita/yr
C_4	Food additives consumption coefficient	Dimensionless

Variable	Term	Unit
W_0	Waste flow variable from animals	Tonnes P/yr
W_1	Waste flow coefficient from human bodies	Dimensionless
W_2	Non-treated solid waste flow coefficient	Dimensionless
W_3	Solid waste treating coefficient	Dimensionless
W_4	Solid waste landfilling coefficient	Dimensionless
W_5	Wastewater collecting coefficient	Dimensionless
W_6	Wastewater treating coefficient	Dimensionless
W_7	Food waste generated per capita	Kg / capita
W_8	Sludge use for other uses coefficient	Dimensionless
$R_{0(1)}$	Manure amount from cattle	T/ head/ yr
$R_{0(2)}$	Manure amount from sheep	T/ head/ yr
$R_{0(3)}$	Manure amount from poultry	T/ head/ yr
$R_{0(4)}$	Manure amount from horse	T/ head/ yr
$R_{0(5)}$	Manure amount from swine	T/ head/ yr
R_1	Recycling coefficient for wastewater	Dimensionless
R_2	Recycling coefficient for solid waste	Dimensionless
L_0	Beneficiating loss coefficient	Dimensionless
L_1	Loss coefficient during processing to phosphoric acid	Dimensionless
L_2	Loss coefficient from fertilizers due to runoff of P	Kg P/ha
L_3	Food loss coefficient	Dimensionless
L_4	Animal food loss coefficient	Dimensionless
L_5	Crops loss coefficient	Dimensionless
L_6	Fish loss coefficient	Dimensionless
L_7	Uncollected wastewater coefficient	Dimensionless
L_8	Non treated wastewater coefficient	Dimensionless
L_9	P loss coefficient from mining rate	Dimensionless
A	Arable land % of total land in EU	%

Variable	Term	Unit
T	Total land in EU	Sq km
$P(t)$	EU population	Capita /yr

- In Table 2, paper I, relative variables are defined as follows:
 - The mining stage includes trade in the material corresponding to imports and exports of mined phosphate ores. In the processing stage, trade in the material covers imports and exports of phosphoric acid. The production stage is defined for specific sectors, such as phosphate fertilizers, feed additives, food additives, laundry detergents, and phosphate obtained from iron ores and concentrates. The consumption stage includes trade in the material representing imports and exports of crops, livestock, dairy products, fish and food and laundry detergents by human.
 - Imports correspond to the mined P import rate, phosphoric acid import rate, fertilizers import rate, the detergents import rate, the crops import rate, the food additives import rate, the feed additives import rate, the livestock import rate, the animal food import rate, the fish products import rate and the iron ores import rate.
 - Different processes were considered to calculate the current level of P recycling in Europe (EU-28 as a case of study in this paper). The processes for P recovery from wastewater use anaerobic digestion. P recovery from solid waste takes place based on composting. The recycling processes for manure to agriculture use anaerobic digestion and composting.
- Considering the closed loop supply chain of phosphorus, the size of recycling is calculated based on waste available from “production”, “consumption” and “loss”.
- Results of this study show not only the dynamic interaction between multiple sectors, but also present trends in P flows in the EU-28, estimation for recycling and imports in the EU and the reliance of the EU on P imports in different scenarios over time until 2050. In our system imports and mining are considered as inputs into the model. Therefore, the recycling of P should not exceed cumulative P imports in the considered period. It is also worth mentioning that the maximum improvement of recycling rate is assumed as 45%, which results from the turning point reached in scenario C, which shows P recycling exceeding cumulative P imports in a 50 years interval. The systematic view taken in this study helps in estimating to what extent bringing the recycled P back to the system will reduce the required amount of P imports.

4.2 Publication II: Environmental sustainability of phosphorus recycling from wastewater, manure and solid wastes

4.2.1 Overview

Long-term supply of phosphorus and its present pattern of consumption is unsustainable (Childers et al., 2011; Daneshgar et al., 2018). In literature, the main issues related to phosphorus sustainability are its scarcity (George et al., 2016), recycling (Morse et al., 1998), and environmental problems (Daneshgar et al., 2018). Also, a need for new sustainable policies and strategic frameworks was highlighted to address the above-mentioned issues (Cordell et al., 2011). Recycling of phosphorus from waste streams has therefore been suggested as a feasible strategy to its sustainable use (EU Commission, 2017a; Withers, Elser, et al., 2015; Withers, van Dijk, et al., 2015). However, there is a lack of assessment of environmental sustainability of phosphorus recycling.

In Publication II, I presented a comprehensive picture of the global phosphorus supply chain. The proposed dynamic model provides the estimation of energy consumed and GHG emitted through phosphorus life cycle, from mining to recycling.

4.2.2 Objectives and research questions

In this study, I developed the dynamic simulation model of Publication I at a global scale. The first objective of Publication II is to develop a dynamic model focusing on environmental aspects (energy consumption and GHG emissions) to assess their change over time until 2050. The second objective is to quantify the possible amount of phosphorus to be recovered from wastewater, manure, and solid waste. Finally, the third objective is to show the effect of present and future phosphorus use on the trend of GHG emissions at various phases of the supply chain over the period 2000-2050.

This article addresses both research questions of this dissertation:

- To what extent can recycling mitigate the problem of the criticality of materials?
- Can recycling mitigate the environmental concerns of the supply chain of critical materials?

4.2.3 Results and discussion

The results show, on average, there is a possibility for providing 14% of the total required phosphorus from secondary sources. The main findings indicate that approximately 82 percent of recycled phosphorus comes from manure. Calculations show that around 70 percent of total GHG generated from phosphorous recycling is caused by the processing of wastewater. Also, results indicate that phosphorus from recycled wastewater accounts for only 2 percent of the total quantity recovered during recycling.

Four situations have been researched in Publication II. Scenario A represents phosphorus current usage rate. Scenarios B, C, and D represent 10%, 30% and 40% rise in phosphorus usage, respectively. Technological feasibility of economic recycling of phosphorus is the primary reason for considering the range of values in different scenarios. Analysis of various situations (Figure 10) indicates that the growth of worldwide phosphorus consumption creates a slow rise in mining environmental issues which accelerate in the recycling phase. It creates a significant problem in a longer perspective leading to an unsustainable phosphorus supply chain from environmental perspective.

The findings show a clear need for an environmental assessment of the processes of phosphorus recycling. Phosphorus recycling processes could not be recommended from environmental sustainability viewpoint because they generate more emissions in comparison to the primary production. However, the results of this paper are limited to energy consumption and GHG emissions and do not deal with other environmental issues, such as water demand and noxious substances like sulfur oxides (SO_x) and nitrogen oxides (NO_x). Therefore, further investigation is needed into other environmental aspects of the performance of phosphorus recycling to get a systematic insight into its sustainability.

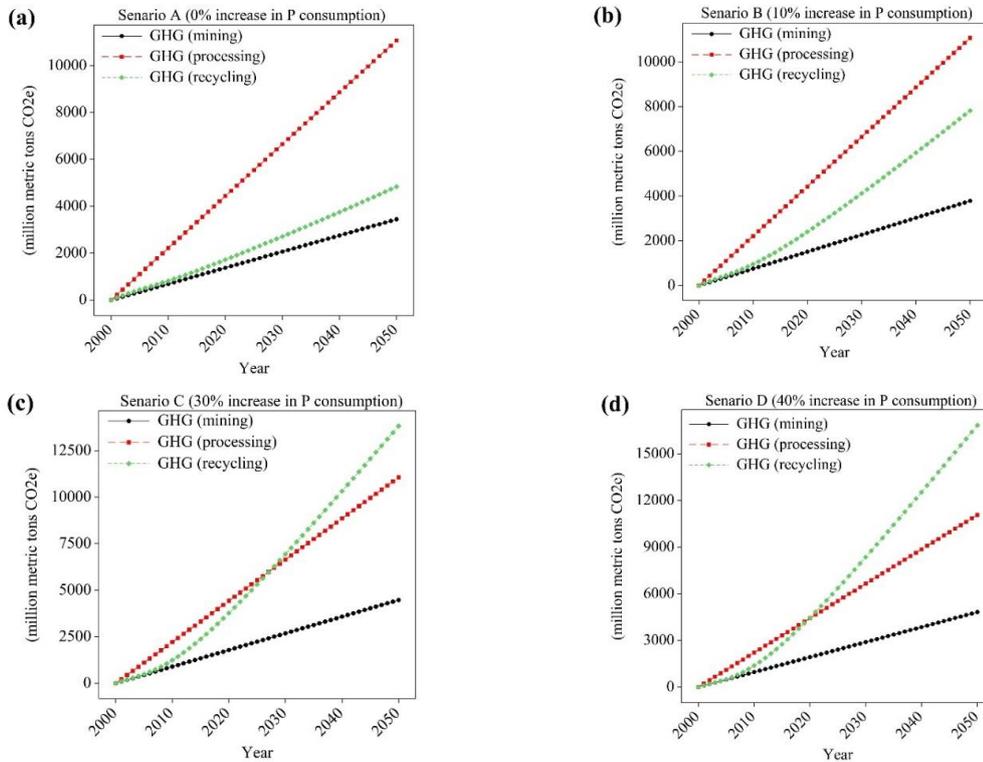


Figure 10. Greenhouse gas emissions trends within phosphorus supply chain (2000-2050) (Golroudbary, El Wali, et al., 2019).

4.2.4 Contributions

The main contribution of Publication II is to present a quantitative analysis to help securing environmentally sustainable supply of phosphorus. Assessment of phosphorus consumption scenarios demonstrates that GHG emissions are steadily increasing in the mining phase and exponentially increasing in the recycling phase. The primary finding of Publication II contradicts the overall view of recycling as being environmentally friendly. The analysis indicates that the recycling of phosphorus is not a long-term sustainable solution. Consequently, mining or phosphorus substitution must still be regarded as valid alternatives to recycling in order to achieve environmental sustainability in the years 2000-2050.

4.2.5 Clarification of further details for paper II

- Wet chemical approach and thermo-chemical treatment are the two main technologies for P recovery from sludge produced in wastewater treatment plants (Appels et al., 2010). In wet chemical process, strong acids are added to extract P from the sludge phase. Thermo-chemical treatment refers to the addition of chloride additives to remove heavy metals from the sludge, thus facilitating the chemical removal of P (Ye et al., 2017). It is worth mentioning that phosphorous recovery from wastewater is difficult due to different sludge compositions (Amann et al., 2018). The data given by Buratti et al. (2015) were used to determine energy consumption in the recycling of phosphorus from solid waste. There are mainly two treatment technologies of waste for P recovery, undifferentiated and source separated collections. In both cases, the treatment of waste and recovery of phosphorous is performed using aerobic biological facility. The undifferentiated collection is meant primarily for the landfilling of waste. In this paper, we consider the second option, source separated collection, in which treated waste is composted as fertilizers.
- Input data for the assessment of energy flow come from calculations based on the ratio of different sources used for energy production (i.e. coal, gas, oil, nuclear, hydro and other energy sources) (Petroleum, 2016). In the proposed model, we assumed those ratios as constant. The ratios could be adopted in the model based on a new identified technology or energy sources. In each stage (mining, processing and recycling) of our model, energy flow is influenced by the dynamic mass flow occurring in the different processes.
- Model validation: As highlighted by Oberkampff et al. (2002), a simulation model is always a simplified representation of reality. In addition, it is always a subject to uncertainty and errors (Biller and Gunes, 2010; Marelli and Sudret, 2014). The main sources of uncertainty/error in simulation models could be classified as epistemic (also known as ignorance, subjective, or reducible uncertainty) or ontological (also known as, random or irreducible uncertainty) (Roy and Oberkampff, 2011).

Epistemic uncertainty occurs due to lack of knowledge and it refers to the uncertainty that could be possibly reduced by improved experiments, more data, and further research (Scheidegger et al., 2018). Ontological uncertainty refers to the uncertainty that is very difficult, if not impossible, to be captured due to the natural variability of the systems and probabilistic occurrence of the event (Chen et al., 2013; Christley et al., 2013). Therefore, in Table 1 at paper II, I quantified the percentage accuracy of the model by comparing the data gathered in the real world with those generated by the simulation model.

To quantify those errors (E_i), I compared the mean value of each variable obtained from the simulation model (\bar{S}) with the mean values of real data (\bar{A}) in a given period as shown by Equations 13 and 14 (Barlas, 1996):

$$E_1 = \frac{|\bar{S}-\bar{A}|}{\bar{A}} \quad (13)$$

where

$$\bar{S} = \frac{1}{N} \times \sum_{i=1}^N S_i, \quad \bar{A} = \frac{1}{N} \times \sum_{i=1}^N A_i \quad (14)$$

Our study took account of the following limitations to calculations and considered specific periods:

First, although the technology for recovering phosphorus has been known since 1960s, quantification of phosphorus recycling from all waste streams before 2000 was not feasible due to the missing data. The main reason is very low efficiency performance of phosphorous recovery from the wastes (Neset et al., 2008; Senthilkumar et al., 2014).

Furthermore, in the recycling stage, one of the most important factors is the delay existing between the production phase and collection of waste. We have indicated that the conditions which caused the reduction of phosphorus production in 1990s (Chen and Graedel, 2016; Cordell, Drangert, et al., 2009) affected the supply of phosphorous to the recycling stage until 2006.

- Phosphorus recovery from manure is seen as more viable than from wastewater but energy and GHG emissions potential of both these recycling approaches is greater than that of the mining of phosphate rock. The results for wastewater recycling show the potential to undermine available technologies for phosphorus recovery, an example that reinforces the need for total life cycle and technoeconomic assessment. Presented results show inexpediency of phosphorus recycling from wastewater if only the amount of the recycled phosphorus and GHG emissions would be taken into account. However, it is obvious that the removal of phosphorus from wastewater must be carried out for a different reason. Excessive presence of phosphorus in water causes eutrophication, and eventually leads to the collapse of ecosystems. On the other hand, avoiding costly and energy-intensive transport of manure, mitigating eutrophication, economic savings from energy recovery and reduced costs of waste management are recurring elements in the framing of P recovery. It is worth mentioning that manure generated in high population and livestock density areas is often exported or used locally due to the combined high P application rates and increased levels of P saturation in agricultural soils.

4.3 Publication III: Environmental sustainability of niobium recycling: The case of the automotive industry

4.3.1 Overview

In the production of steel for automotive industry, niobium is used in the form of ferroniobium. Ferroniobium represents the most substantial portion of the global demand for niobium; around 87% (Schulz et al., 2017). The growing use of ferroniobium in automotive industry is driven by a trend to reduce the weight of vehicles (Das et al., 2016; Modaresi et al., 2014).

The end of life vehicles have become a significant waste stream (Widmer et al., 2015). Therefore, one of the dominant trends in car manufacturing is maximization of recyclability. Recycling of ELVs can contribute considerably to the reduction of waste and it constitutes a significant source of raw materials such as niobium.

Publication III presents the assessment of environmental impact of the global supply chain of niobium by using system dynamics modelling. Quantitative analysis for niobium recycling has been studied focusing on energy consumption and GHG emissions. The ELVs waste stream is considered in this study. ELVs flow is an important potential source of critical metal recycling, particularly niobium. The proposed model estimates annual energy savings and GHG emission mitigation by using niobium in secondary production until 2050.

4.3.2 Objectives and research questions

The first objective of Publication III is to assess energy consumption and GHG emissions at each stage of niobium supply chain including mining, production, and recycling. The second objective is to identify the impact of growth in global demand of niobium on environmental issues.

This paper, presenting a dynamic model, answers the second research question of this dissertation:

- Can recycling mitigate environmental concerns of the supply chain of critical materials?

4.3.3 Results and discussion

The results show that 45 percent of energy demand in niobium supply chain — the majority of total used energy — is consumed in the primary production stage (Figure 11). This stage generates 72 percent of total GHG emissions of niobium SC between 2010 and 2050 (Figure 12). Mining of niobium uses around 36% of energy and contributes to about

21% of GHG emissions. However, the secondary production of niobium in recycling stage, consumes 19% of SC energy and emits 7% of GHG emissions.

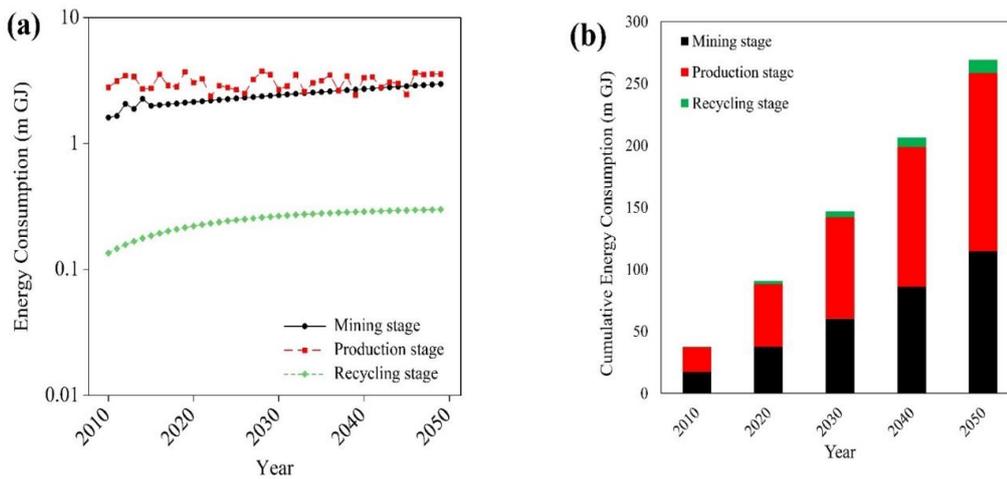


Figure 11. Energy consumption in mining, production and recycling stages of niobium supply chain (2010-2050). (a): annual energy consumption. (b): cumulative energy consumption (Rahimpour Golroudbary et al., 2019).

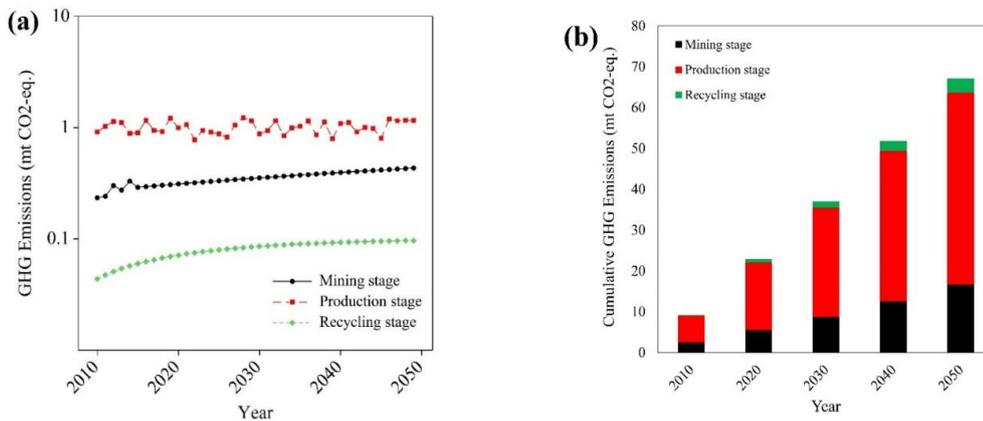


Figure 12. Greenhouse gas emissions in mining, production and recycling stages of niobium supply chain (2010-2050). (a): annual GHG emissions. (b): cumulative GHG emissions (Rahimpour Golroudbary et al., 2019).

Three scenarios are assumed for the growth rate of niobium applied in automotive industry. Those scenarios are to indicate the changes in energy consumption and GHG

emissions from primary and secondary production of niobium. Scenario A represents 10% increase in the number of ELVs and scenarios B and C correspond to 25% and 50%. The highest value of scenarios considered, 50%, is based on the statistical analysis of population and car ownership until 2050 through literature.

Calculations indicate that niobium recycling could save ca. 133-161 m GJ energy and would also contribute to the mitigation of 44-53 mt CO₂-eq over the period 2010-2050. The result shows around 18% reduction of annual emissions between 2010 and 2050 from the reuse of niobium in secondary production compared to the primary one.

4.3.4 Contributions

Publication III reveals the implications of niobium supply chain management strategies and focuses on recycling. Thus, it helps prioritize opportunities for reducing risks associated with the supply of critical materials and mitigating environmental harm. It shows the contribution of niobium recycling energy savings and to the reduction of emissions in a long term.

4.3.5 Clarification of further details for paper III

- In paper III, the calculation of input data of the simulation model is based on the amount of standard grade ferroniobium added to HSS steel in automotive industry. In the processing stage, according to the data from the U.S. Geological Survey, more than 90% of niobium is transformed in standard grade Ferroniobium by adding iron and aluminium. In the recycling stage, the calculation is based on niobium contained in HSS steel of collected end of life vehicles. Therefore, recovery or reuse of niobium from recycling of HSS steel is the subject of this study.
- In Table 2 of paper III, the initial flows considered for the stock of extracted materials are based on the global production of ferroniobium. In the processing stage two main flows are considered based on the percentage of global niobium production used to produce ferroniobium used in high strength low alloy steels and the percentage of global niobium production used in manufacture of niobium alloys, niobium chemicals and carbides, high purity ferroniobium, and other niobium metal products.
In the recycling stage, the flow of stock is based on automobile recycling rate and the efficiency of automobile scrap recycling. After estimating the possible available amount of niobium from end of life vehicles, the calculation of the possible supply or reuse of niobium from recycling for the HSS steel would be possible. The available amount of niobium from scrap contributes to the reduction of the mining of niobium as a primary resource of niobium supply.

- The amount of future mined mineral is calculated based on the estimation of the annual growth rate of niobium world production. This growth rate is affected by the increase in demand for ferroniobium.
- Calculations in Table 4 for saving in energy consumption by countries are based on the cumulative amount of energy consumption by primary production and recycling stages until 2050. In a similar approach, estimation of the reduction of GHG emissions by countries are based on the cumulative amount of GHG emitted from primary production and recycling stages until 2050.

4.4 Publication IV: The life cycle of energy consumption and greenhouse gas emissions from critical minerals recycling: Case of lithium-ion batteries

4.4.1 Overview

Growing demand and limited possibilities of substitution cause the increase of supply risk of lithium. Between 1991 and 2012, the use of lithium in the battery sector rose from zero to 80% of the battery market. LIBs recycling is essential to avoid hazardous outcomes of the post-consumer products, including heavy metals. Therefore, the importance of Li recycling arises not only from conserving the resources, but also from avoiding creating environmental burdens (Ordoñez et al., 2016). Dunn et al. (2012) have highlighted the lack of analysis of emissions generated from the production of batteries in its supply chain. Production of batteries has been identified as a primary contributor to GHG emissions of EV manufacturing (Notter et al., 2010).

Publication IV analyses energy consumption and GHG emissions during lithium-ion batteries production by simulating a model according to the principles of SD methodology. Publication IV provided a detailed discussion on the recovery of lithium from five different types of batteries including lithium-ion manganese oxide (LMO), lithium-ion cobalt oxide (LCO), lithium-ion iron phosphate (LFP), lithium-ion nickel manganese cobalt oxide (NMC), and lithium-ion nickel cobalt aluminium oxide (LiNCA).

4.4.2 Objectives and research questions

The key objective of Publication IV is to investigate material and energy flows, as well as GHG emissions at each stage of lithium supply chain including mining, production, and recycling. Besides, it analyses how much recovery of critical minerals such as lithium, cobalt, and manganese from spent LIBs may contribute to energy savings and emission mitigation.

This paper answers the first and second research question of this dissertation by presenting a dynamic model of lithium supply chain.

- To what extent can recycling mitigate the problem of criticality of materials?
- Can recycling mitigate environmental concerns of the supply chain of critical materials?

4.4.3 Results and discussion

Findings show that recycling of LIBs from a mass flow view helps to avoid the shortage of critical minerals such as cobalt (Co) and manganese (Mn). However, the contribution of lithium supplies from secondary sources is not significant. Also, the recovery of lithium from LIBs is not environmentally recommended. It leads to energy consumption higher by ca. 38-45%. It also generates around 16-20% more GHG emissions than its primary production.

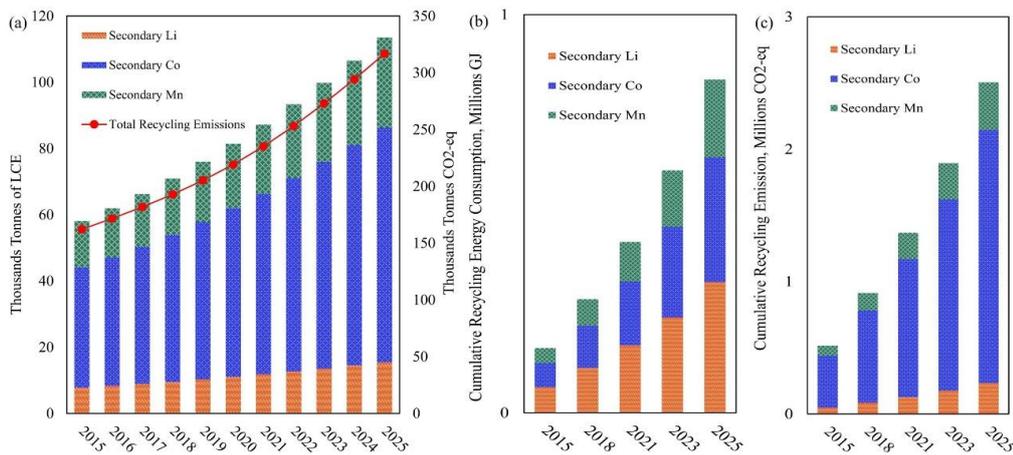


Figure 13. Environmental impact of production stage by using secondary materials between 2015 and 2025. (a) Annual; (b) Cumulative energy consumption; (c) Cumulative emissions (Golroudbary, Calisaya-Azpilcueta, et al., 2019).

4.4.4 Contributions

The main contribution of Publication IV is the quantification of the global closed-loop supply chain of lithium focused on LIBs production, which helps analyzing the environmental sustainability of its recycling. The main finding shows that recovery of lithium is not environmentally sustainable. It implies recovering Li from spent LIBs leads to higher energy consumption and air emissions compared to primary production.

4.4.5 Clarification of further details for paper IV

- Results of recycling: Calculation is based on the proportion of energy consumed and GHG emitted by the recovery of materials such as Li, CO, and Mn from Cathode production of different type of batteries (LMO, LCO, LFP, NMC and LiNCA) and assembly of the battery. The increased percentage of energy is calculated based on the cumulative energy consumed by secondary production in comparison to primary production per total amount of energy required until 2025. The increased percentage of generated GHG emissions is estimated based on cumulative GHG emissions by secondary production in compare to Cathode production per total amount of GHG emitted until 2025. Therefore, these increased percentages are not in the same range.
- The results of recycling: Besides solid waste reduction and material scarcity concerns, it worth mentioning that if LIBs recycling can recover cathodes at a lower required energy level and emit less emissions than producing virgin cathode materials, it is important to pursue battery recycling. The results of paper IV show that the recycling of LIBs based on the current technology do not recover an adequate amount of lithium to meet future demand from the automotive industry. Also, the analysis highlights that Li recovery processes are energy intensive and complicated.

In literature there are several research studies regarding the adoption of LIBs recycling processes. For example, Sonoc et al. (2015) suggested that LIBs could be disassembled by automated processes to recover valuable electronic components. Therefore, their cells can be safely discharged to recover residual energy and to render them safe to shred and treat by hydrometallurgical and pyrometallurgical methods. Dunn et al. (2015) focused on battery assembly stage to analyse the merits of LIBs recycling stage. However, further research is needed to find out if adoption of the recycling processes of LIBs at the level of components will lead to the recovery of lithium in a sustainable economic and environmental way.

4.5 Comparison of results obtained for phosphorus, niobium and lithium

Material from recycling and corresponding production from the end-of-life stage could become an alternative future resource. Furthermore, in the mining stage, some critical materials are produced as by-/co-products and require a long lead period (i.e. 5–10 years to start a new mine). Therefore, high supply risk may be involved in expanding the mining capacity following rapid deployment of technologies.

Figure 14 shows the comparison between critical materials (P, Nb and Li) available annually from primary and secondary productions. According to the current best available recycling technologies, results of the simulation model show that the possible amount of recycled phosphorus from wastewater, manure and solid waste could be estimated on average at the level of 26% in the period 2000-2050. The possible amount of recycled/reused niobium from automotive industry is estimated on average around 50% in the period 2000-2050. The possible amount of lithium recycled from batteries could be estimated on average around 4% in the period 2000-2025.

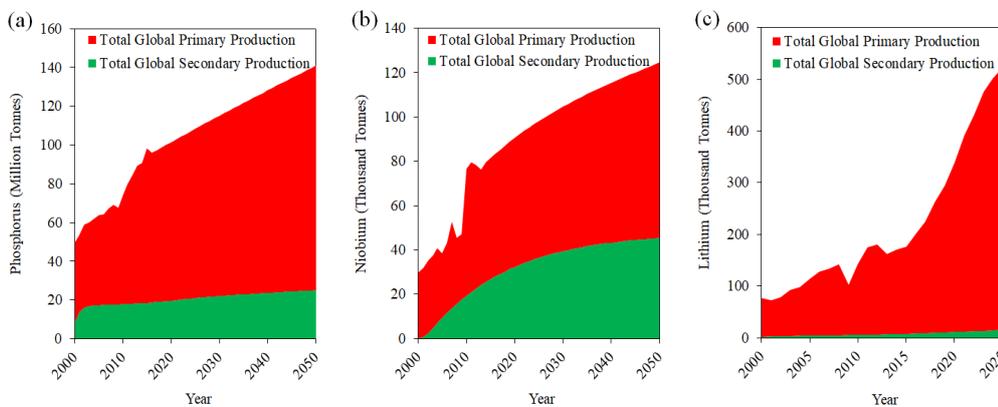


Figure 14: comparison of phosphorus, niobium and lithium available from primary and secondary productions.

This dissertation is designed to present a systematic view on supplying CRMs as virgin or recycled materials. Therefore, applying such materials in primary production and their recovery or reuse as a result of recycling processes were analysed considering the required energy and emitted GHG along the stages of supply chain. Table 5 presents the comparison of energy consumption and GHG emissions between primary and secondary production for 1 kg of a critical material.

Table 5: Comparison of annual energy consumption and GHG emissions between primary and secondary production for 1 kg of a critical material.

CRM	Product	Primary production		Secondary production	
		Energy (GJ)	GHG (t CO ₂ -eq)	Energy (GJ)	GHG (t CO ₂ -eq)
Phosphorus	Fertiliser	0.034	0.005	0.011	0.002

	Food & Feed	0.450	0.079	1.206	0.213
Niobium	HSS steel	60.200	1.980	6.510	2.100
Lithium	LCO	28.528	1.275	93.017	1.073
	LFP	16.846	0.761	117.918	1.249
	LMO	16.457	0.734	277.438	2.261
	LiNCA	4.616	2.606	393.235	38.091
	NMC	20.270	1.012	94.544	1.225

GHG emissions associated with electricity generation have been accounted for the analysis in this dissertation. The IEA report shows that global average intensity of carbon emissions of electricity generation was 524,263 (gCO₂/kwh) between 2000 and 2018. Also, the target of sustainable development scenario (SDS) has shown carbon intensity of 220 (gCO₂/kWh) to be achieved in 2030; the measure was at the level of 478 (gCO₂/kwh) in 2018. However, the trend in carbon intensity of electricity generation shows the current development is not on track to meet the SDS target. Table 6 presents results of this study based on carbon emissions intensity of electricity generation compared to estimation adjusted to the current and sustainable development scenario (SDS) carbon intensity levels.

Table 6: Comparison of the results of total carbon emissions intensity of electricity generation through material production (Mt CO₂-eq/MJ).

Material		Phosphorus	Niobium	Lithium
Simulation model results (2010)	Primary	195.43	0.23	0.02
	Secondary	68.14	0.04	0.12
Model results adjusted to IEA (2010)	Primary	187.65	0.24	0.02
	Secondary	65.42	0.02	0.06
Difference (2010)	(%)	3.98	0.45	2.45
Simulation model results (2030)	Primary	199.39	0.35	0.11
	Secondary	91.38	0.09	0.32
Model results adjusted to IEA's SDS (2030)	Primary	80.44	0.02	0.05
	Secondary	36.86	0.02	0.01
Difference (2030)	(%)	59.65	99.99	57.48

There are several research studies showing potential pathways to future energy mix, and these pathways vary greatly from one another, including emissions that they forecast. However, one often common denominator to these simulations is a scenario commonly referred to as “Business as usual” (BAU). This scenario assumes that the current energy mix continues into the future and it is the one I have used as a reference point.

Although there are several realistically optimistic scenarios for the reduction of carbon emissions from the energy sector, there is no actual certainty as to whether the energy sector will evolve in one specific way or another, and, therefore, I believe the selection of either scenario, including BAU, as a reference for the work is as good any other.

5 Conclusions

Minimizing energy consumption and attaining low or zero GHG emission, as well as protecting the environment are becoming increasingly crucial for critical materials supply chain. Addressing environmental sustainability issues in CRM models enables decision makers to decrease risk and exploit the possibilities that sustain the life cycle of materials. Despite these benefits, there are very few investigations that have been carried out to handle this key issue of sustainability in terms of critical materials. Therefore, it is important to assess and analyse the entire life cycle of CRMs focusing on environmental aspects including energy consumption and GHG emissions of supply chain in cradle-to-gate assessment. Next, I will explain the contribution of this dissertation by presenting the main findings of the study. Then, I will clarify the limitations of this research and will provide my suggestions for future studies.

5.1 Contribution of the study

A supply chain of critical materials has been modelled using systems dynamics. This dissertation has been an attempt to analyse the role of recycling in the continuity of supply of critical materials and mitigating the environmental issues through their supply chain.

I extend findings from previous literature on critical raw material flows in all stages of supply chain from mining to recycling. I quantified physical limitations to the supply of these materials to shed light on their criticality. From this perspective, continuity of supply of those materials is investigated for a long-term period. Then, I designed a dynamic model to investigate environmental issues (energy consumption and greenhouse gas emissions) along the supply chain of critical materials, such as phosphorus, niobium and lithium. This step aims at developing holistic perspectives on supply chain sustainability. From this perspective, the available options for reaching both higher economic growth and lower GHG emissions are investigated.

This research examines the appropriateness of simulation approaches (such as system dynamics methodology) to assist decision makers in enhancing the efficiency of their investment decisions and growth programs throughout the supply chain of critical materials in a closed loop. The model of this study is unique in that it captures the dynamics of interactions among the effective factors between different stages of the supply chain of critical materials. Moreover, considering challenges regarding critical materials lifecycle stages, this study opens an interesting window on the recycling stage and identifies its environmental challenges.

It is worth mentioning that the supply chain of each critical material is a troublesome issue to apprehend for it includes several levels of complexity. Therefore, the assessment of supply and environmental concerns should be conducted on a case by case basis, rather than as part of a general framework. As for generalization, this study supports researchers

in recognizing the importance of variables by dynamic modelling and aligning improved sustainability requirements with circular economy.

5.2 Managerial implications

Recycling of materials is important for their sustainability as well as for the advancement of circular economy. Legislation for waste management focuses on the possible amount of recycling and critical materials to be recovered from waste for specified target years. The legislation is limited due to its exclusive focus on the total quantity of critical materials to be recovered. It is important to measure the sustainability of recycling strategies against existing regulations and to make improvements in the policies. The results of Publication II and IV indicated that new recycling strategies would be needed to avoid more environmental issues in the future.

Dynamic models presented in this dissertation allow for an increased focus on environmental sustainability of recycling. Besides, the proposed dynamic models help managers in assessing the supply chain of critical materials and have an integrated mass flow and environmental analysis for all stages.

On the one hand, system dynamics model can be used on a case by case basis to explore the behaviour of the supply chain of critical materials at regional scale for different cultural systems, environmental and economic conditions, which has been shown in Publication I. On the other hand, system dynamics model can be applied in the analysis of the supply chain of critical materials at a global scale and used in analysing various options of supply from primary and secondary production, which has been shown in Publications II, III, and IV.

The results of this dissertation include a quantitative analysis that can be used as a policy decision support in the fields of resource efficiency, waste management, and environmental protection. Also, this dissertation attends to help managers to decide on using primary or secondary supply sources in line with environmental sustainability principles. Publications II and IV have demonstrated the recycling of critical materials such as phosphorus and lithium is not environmentally sustainable in the long term. Publication III showed that the recycling of niobium is environmentally sustainable if we consider energy consumption and greenhouse gas emissions. Besides, the analysis in this dissertation shows to what extent recovery of critical materials is possible from recycling and which options of supply (primary or secondary production) would produce fewer environmental issues in the long term.

This research is generally performed to direct the attention of decision makers at government and industry levels towards issues related to critical raw materials supply and environmental issues within their supply chain. Therefore, the analysis of this dissertation could be generalized into two perspectives. Firstly, from a mass flow perspective by focusing on the life cycle of critical materials through their supply chain. In this line, this dissertation shows a significant potential of supplying such materials by focusing on the

recycling stage to target a sustainable closed-loop supply chain. As a result, it contributes to reducing the supply risk of critical materials and improving waste management. Secondly, environmental assessment of the performance of all stages of the supply chain of critical materials targets managers to help them better cope with environmental issues such as energy consumption and GHG emissions.

The comparison of environmental behaviour of the recycling stage of the supply chain of each material highlights the necessity to improve technologies and processes deployed in the recovery of these materials from waste. Considering the chemical and physical properties of critical materials based on the periodic table, lithium from the group of alkali metals, niobium from the group of transition metals and phosphorus from the group of non-metals were considered in this dissertation. By applying the analysis proposed in this dissertation to other critical materials decision-makers will become aware of actual and potential environmental consequences of their decisions concerning the supply needed from primary or secondary sources to meet future demand for critical materials.

In line with circular economy guidelines, this dissertation aims to improve the decision-making process in using recycling of critical materials by applying a systematic approach. As indicated in this dissertation, sustainability of recycling procedures cannot be taken for granted as it may differ depending on the type of waste stream. Therefore, decision-makers and managers should consider the sustainability of recycling process for critical materials on a case by case basis instead of accepting a general view on recycling as being environmentally friendly. This dissertation can inform policy makers and make them aware of future challenges involved in recycling critical materials.

5.3 Limitation of study and suggestion for future research

The main limitation of this research is the availability of data for all industries in which critical materials are applied. Therefore, in this thesis, presented models focus on the most significant industries. For example, in the phosphorus model, I considered agricultural production. Other industries such as detergent production and other chemical uses were not considered in the global phosphorus model. In the niobium model, steel production was the main considered stream. Due to the lack of data, other uses of niobium, such as vacuum grade ferroniobium, chemical production and pipe industries were not considered in the presented model. In the lithium model, the focus was on lithium-ion battery industries. Non-battery manufacturing, such as air conditioning, aluminium, polymers, ceramic and glass lubricant gases and others were not considered in the presented model.

Another limitation of this dissertation is the lack of analysis for by-product of the critical materials value chain. For example, in the case of phosphorus, there is a lack of assessment of white phosphorus (P₄) stream generated from phosphate rock, which is an elementary form of phosphorus. White phosphorus was not assessed in the EU Commission studies on critical raw materials in 2011 and 2014. As a result, there is not enough information and data accessible to analyse its flow in detail. In the case of niobium, we lack tantalum analysis, which is a significant by-product in the value chain

of niobium. This limitation results from restricting the proposed dynamic model to the use of niobium in high strength steel for the automotive industry.

The absence of technical and economic evaluation of the improvement of recycling of critical materials as well as its effect on the environment is another restriction of this study. Such analysis could be suggested for future research.

Also, the change in future energy mix is not considered in this dissertation for analyses running until 2050. For such analyses more research would be needed based on fieldworks, which are beyond the aim of this study. It is worth mentioning that the uncertainty of each projection for the future energy mix justifies my assumption to consider BAU as a reference scenario. As limitation of my analysis, changes in the future emission intensity of the energy sector would influence largely the estimated results presented in this dissertation.

Moreover, the recycling of products containing critical materials that can be re-used in the same function in the system is of significant concern. Integrating extensive recycling from the design of products into developing a closed-loop supply chain would make a major contribution to sustainability and to circular economy. It is recommended that future study should focus on developing an extensive recyclability index for complicated products and their complex supply chain to close the loop of critical materials.

Another suggestion for future work would be, given stocks and flows of critical materials are uniformly analysed by considering environmental impacts as shown in this dissertation, and information included in many individual dynamic models is linked together in large databases on material sources, flows, stocks and final sinks, to propose sustainable use of supply sources.

Finally, dynamic models such as the ones presented in this dissertation can help to design long-term emission mitigation strategies that consider all stages of the supply chain. Focusing on one stage of the supply chain is a significant limitation in environmental assessment considering energy consumption and GHG emissions. Therefore, expanding this type of study to cover other critical materials is suggested.

Moreover, measures for climate change mitigation and adaptation are becoming the number one priority in the world, particularly due to the increased awareness of negative consequences of climate change among citizens. Greenhouse gases account for the major portion of air pollution. Based on the results presented in this dissertation, under the current shape of supply chains of some materials, it extracting virgin material is more environmentally-friendly than engaging in secondary production. For example, in the case of phosphorus and lithium results of this dissertation show that energy consumption and greenhouse gas emissions from current recycling technologies may make it less sustainable than originally hoped, a result that has the potential to undermine available technologies for such materials recycling. These results highlight the need for environmental assessment of the total life cycle.

It is also worth mentioning that recycling is a must from the waste management perspective. For example, recovering phosphorous from wastewater could considerably reduce eutrophication and create a supplementary source of fertilizers. Pollutions caused by waste can deteriorate human health. Therefore, the achieving of low or zero GHG emission, optimized water and energy demand, and a well-protected environment are challenges that have become increasingly more significant for the critical materials supply chain. The focus of this dissertation is on energy consumption and GHG emissions rather than on other issues, such as water demand and noxious substances like SO_x and NO_x. Therefore, further investigation is needed into other environmental issues at all stages of critical materials supply chain to get a systematic insight into their sustainability.

6 References

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Publication I

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Article

Impact of recycling improvement on the life cycle of phosphorus

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ABSTRACT

Access to natural resources is increasingly more difficult and more costly, partly due to their economic significance and to continuous increase of their global consumption in the recent years. In the case of phosphorus (P), which is a critical raw material, geological distribution of its primary nonrenewable source (phosphate rock) is concentrated in particular regions leading to high supply risk of this raw material. In Europe (EU-28), where phosphate rock reserves are scarce, import of phosphorus has been the main source of supply. It means that Europe relies highly on the foreign exporters. From decision makers' perspective, recycling of phosphorus was taken into account as one of the possible solutions to decrease the dependence on imports and extraction of reserves. The question, however, is to what extent does the recycling of phosphorus help in reducing the reliance on typical supply resources? Hence, the main objective of this paper is to quantify the dynamic flow of phosphorus and show potential benefits of its recycling in Europe. This article presents a system dynamics model for representation of the element P flow and helps to quantify to what extent the recycled phosphorus could mitigate its criticality. Analysis of the results supports previous studies indicating the high reliance of EU on P imports, estimating around 96% as the reliance percentage on imports. The results imply that improving P recycling has the potential to decrease the level of P imports to a certain extent, which may reach 79%.

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1. Introduction

1.1. Phosphorus as a critical material

One of the main concerns for raw materials is the supply risk which has been the most common aspect of criticality of a material according to the previous studies [1–3]. It is a result of disruptions in the supply chain due to physical shortage of resources [4]. They are often a result of continuous extraction of non-renewable natural resources leading to the scarcity of resources [5]. These concerns are driving for new methodologies for assessing criticality of materials [6]. Critical materials (a) ensure the specific properties of the advanced products or systems, (b) are subject to supply risk, (c) have no obvious substitutes, and (d) are substantial for environment-friendly technologies [7–9]. However, due to the differences in methodologies applied for criticality assessment, there is no overall definition of critical materials [1,10]. The EU assessment of material criticality is based on two factors: supply risk and economic importance [11]. The economic importance is determined by assessing the contribution of a given material to the industrial mega sectors in Europe. This process focuses primarily on the role of each material in the European manufacturing industries [3].

Phosphorus (P) plays a prominent role for all living creatures and organisms [12,13]. The essentiality and unequal distribution on global scale through regional surplus of phosphorus (P) highlight significant challenges in phosphorus life cycle for worldwide community [14]. Globally, a potential scarcity of phosphorus does not seem to occur [15–19]. However, considering the lack of any substantial reserves in Europe, the main challenge faced by decision makers is the complete dependency on raw phosphorous exporters. The European Union remarkably is a region with only a few deposits, in which the main part is found in Finland [20]. This imbalanced distribution of P could potentially cause geopolitical issues for European governments and firms [14].

Globally, phosphate rock (PR) reserves are concentrated in particular regions. Morocco holds 73% of the global reserves [21]. Such geological distribution of PR stock might affect the price of fertilizers and food [22], or it can also disrupt the continuous supply of the ore material [20]. The geographic concentration of current PR supply in a few major producing countries is also underlying the increase in the vulnerability of a region such as Europe (EU-28) that relies on imports for its primary P inputs.

Europe is about to develop new strategies to lessen the dependency on import of phosphorus due to the already mentioned issues and very limited substitutability of phosphorus [14,23]. It is planned to emphasize the role of recycling and recovery as the major factor reducing Europe dependency on imports [22,24]. Therefore, the research on

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sustainable supply chain management has largely focused on monitoring its reversibility.

Increasing recycling and improving sustainable management of phosphorous is one of the top priorities of the EU, as emphasised in the EU 2020 Strategy [25,26]. Hence, the main motivation of this paper is to calculate the current level of recycling through the various waste streams (solid waste, water treatment and manure) and to estimate the degree to which it could be enhanced. Also, we present the possible impact of this improvement on the criticality of P in Europe.

1.2. State of the art phosphorus recycling

Recycling has recently emerged to be one of the main possible factors for developing closed-loop supply chains [27], particularly for critical materials, which might be subject to supply disruptions if a circular economy is not effectively planned and implemented [28]. The recycling of phosphorus has been already proposed as one of the viable alternatives to mining [29–31]. Both solid wastes and wastewaters offer an excellent opportunity for more efficient use of phosphorus [32,33]. Recycling of phosphorus from the waste streams has a potential for decreasing the reliance on other sources and could significantly reduce this material's imports [18,33–35].

Most phosphorus flow studies are commonly conducted using methods such as substance/material flow analysis (SFA/MFA) [14]. To quantify the P stocks and flows, researches on the material flow of phosphorus were done in different studies considering various geographical scales, e.g., regional [36,37], national [34,38,39] and global [40–43]. The potential and the need for improving P recycling were the common recommendations among all these researches.

1.3. Objective of the study

Considering material flow analysis in the European community scale, the structural model of the phosphorus life cycle to quantify the flows has been studied by several researches [14,24,44]. Ott and Rechberger [44] use the SFA to quantify the flow of P by considering the 15 state members of the EU. Withers *et al.* [24] suggest a 5R stewardship to resolve the European concerns on the P life cycle. Van Dijk *et al.* [14] go further by considering the P flow in the 27 member states of the EU, which concludes the high dependency on primary P imports, long-term P accumulations in the soil, P losses due to societal consumption, limited recycling and low use efficiency of P.

In this study, using system dynamics modelling, we analyse and quantify the phosphorus flows in detail for the European Union (EU-28). The necessity of systems approach to phosphorous supply chain management has been presented in several studies [40–42]. System dynamics modelling is a useful approach to show evolution trends of the given system [45]. In the case of phosphorus, system dynamics models have been used to examine dynamic interactions among various components of the system being studied, e.g. solid waste management [46], phosphorus mass flow [47], a decoupled aquaponics system [48] and phosphorus flows in food and waste chains [49]. In this study, a system dynamics model is built to show the dynamics of phosphorous supply chain in years 2000–2050. System dynamics modelling adopts a top-down approach, conceptualizing a complex system at aggregate level. This method is useful for modelling dynamic changes over time as well as modelling large and complex systems [50,51]. It offers an approach supporting businesses and government institutions in development of strategy and designing policy interventions [52]. System dynamics modelling helps to consider that many objects interact with one another (complex), their interaction produces the distinctive behaviour of the system (interactive) and their distinctive behaviour varies over time (dynamics). The methodology will be explained in the next section. In this study, P flows from

non-food and non-agricultural sectors are also considered. The model also presents the impact of the dynamic characteristics of the P flow and the demographic changes of the EU population. The results estimate the amount of P mined, imported, exported, produced, consumed, lost and recycled. Previous suggestions on improving P recycling [24,34,53,54] are taken into consideration through scenario development of the model. It provides a scenario-based analysis to assess the impact of recycling on the P life cycle in the EU.

2. Materials and Methods

Necessary data input for the model was obtained from various database sources in addition to related literature (Supplementary Material Table S2). The required data should represent phosphorus content inside the materials. The available datasets show data on phosphorus in the form of chemical compounds (e.g. P pentoxide, phosphoric acid, and sodium triphosphates). The approach was to calculate the exact phosphorus composition inside each compound considering the total molar mass of the compound and the molar mass of P element (Supplementary Material, Table S2).

The system dynamics method introduced by Forrester [55] is used in this study due to the high level of complexity and interdependencies between the stages and variables [51]. The need of system dynamics is important to understand the behaviour of the model and build a structural representation of stocks, flows and interacting parameters [56] with the passage of time under different scenarios [57]. The model of the life cycle of phosphorus in Europe has been constructed to enable quantitative estimation of the amount of phosphorus recycled from solid wastes and wastewater streams. The model focuses on the main predictors of the flow changes and thus leads to dynamic behaviour of the whole system.

2.1. Model description and system definition

The developed model is a dynamic stock-flow diagram. All the equations associated with every component are provided in the Supplementary Material (Table S1–S3). This model is dynamic and is subject to change with time. Historical data starts from different years (2000 the latest) and ends in 2015 or 2016. Therefore, the model duration corresponds to two periods, the historical period (2000–2015) and the future estimated period (2016–2050). Historical data were used to form the structure of the variable variation, and to determine the behaviour of these variables over time (Fig. 1).

The model has two major assumptions. First, the system boundaries were fixed by taking into account only 28 member states of European Union. The system covers seven main stages of phosphorous life cycle. Those stages are: mining, beneficiation, processing, production, consumption, waste generation and recycling. In addition, import and export are considered in this study. Import and export correspond to the material traded between the EU and countries or groups outside the EU. The material trade within the system boundaries (within the EU) is not considered. The material trade (import and export) is analysed in mining, processing, production and consumption. In the mining stage, the material trade corresponds to import and export of mined phosphate ores. In the processing stage, the material trade corresponds to import and export of phosphoric acid. In the production stage, the material trade is shown separately for each production sector. These sectors are: phosphate fertilizers, feed additives, food additives, laundry detergents, and phosphate obtained from iron ores and concentrates. In the consumption stage, the material trade represents the import and export of crops, livestock, dairy products, fish and food. The second basic assumption concerns the time of analysis. Basing on historical data, we assumed the distribution of phosphorus in the different sectors. Table S3 in Supplementary Material represents distributions for the production of feed, crop and animals.

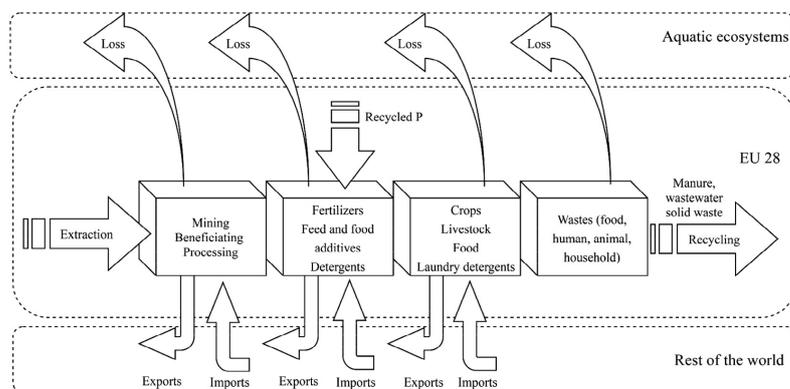


Fig. 1. Overview of the phosphorus life cycle in the EU.

2.2. Phosphorus life cycle in the EU

2.2.1. Mining and processing of phosphorus

The life cycle of phosphorus is strongly affected by human activities, where over 90% of P intakes go for the agro-food production, including fertilizers and food additives [58]. The first stage is the mining of raw materials (apatite) from phosphate rock. Europe does not extract any significant amounts of phosphorus, except a small amount (about 54800 t P) extracted in 2015. The major suppliers of phosphate ores are based in Morocco, Tunisia, the US, Jordan, Russia, and China [21]. Supply of phosphate ores corresponds to the mining of phosphate rock, which has been so far the largest source after WWII, containing around 11%–15% of P [59]. Besides, iron ores represent a secondary source for obtaining phosphorus. The phosphorus obtained from iron ores is used as a phosphate fertilizer [60]. Europe is highly dependent on imports with a reliance percentage of 88% [11]. The trend of primary material imports is essential to identify the pattern of EU imports from other countries. Data analysis shows, in 2009, the imports of apatite to the EU experienced a dramatic decline because of the global economic crisis, when the amount of imported apatite accounted for 389269 t. Then, the traded amount recovered rapidly until 2016. The reduction in 2009 is considered a sudden rather than a gradual change.

After the mining process, the mined material goes for beneficiation. The beneficiation process results in P effluents to the water [13], and the beneficiated material goes for the next stage (processing stage). This stage corresponds to the processing of phosphorus into phosphoric acid [43]. In this stage, P loss also occurs; where around 10% of the processed material is lost [61]. The domestic production is not the only source for obtaining phosphoric acid. The European trade of phosphoric acid appears in the imports that have been stable for the past ten years (2006–2016), except for the sharp decline in 2009, when the imported amount accounted for 301523 t. The exports, however, have been considerably stable in this period, without any significant increases or decreases. Trends in import and export activities are equally useful to understand the EU trade behaviour regarding the material processed in the life cycle of phosphorus.

2.2.2. Production stage of phosphorus life cycle

After processing, the next stage is the production of different products. Fertilizers is the major sector where phosphorus is used for [62]. Other sectors include feed additives, food additives, detergents and other industrial uses [63]. In the food additives sector, phosphorus

is added as preservatives into processed food [64]. They are used as preservatives for different categories of food including meat preparations, cheese products, vegetables, fruits and different kind of beverages [65]. In the feed additives sector, phosphorus is used for production of compound feeds for the livestock. The use of phosphorus in detergents corresponds to the production of sodium triphosphates (STPP) [66]. The production of STPP in Europe is subject to change in the future. Even though there exists STPP production in Europe, there is a serious plan to completely ban the use of this material in detergents and replace it with other detergent builders because of the damage it causes towards surface water. Thus, the use of STPP will no more occur after several years [67]. The trend of the European STPP trade significantly reflects the effect of policies adopted to ban the use of this material in the detergent industry. Trade in this material (exports and imports) has been dramatically decreasing. This process is important in this study particularly because it helps to estimate the contribution of the detergent industry to the European life cycle of phosphorus in the upcoming years.

2.2.3. Phosphorus consumption

The use of phosphorus in agriculture starts from fertilizer production, crop production, agri-commodities, food and human consumption [68]. Schoumans *et al.* [22] show a comparison between 1961–2012 and 2008–2012, when the average consumption of phosphorus fertilizers in Europe (1 kg P_2O_5 per hectare) was 13 and 7, respectively. After the application of fertilizers, a significant amount of phosphorus is lost into aquatic systems while being in the soil. This loss is due to the runoff of fertilizers [69]. Throughout the use of phosphorus in the soil, the element drains or leaks because there is a surplus of phosphorus after its required amount is consumed by agriculture [70]. Besides the produced compound feed, grass cut and some crops are used as feed for the livestock consumption [71,72]. In addition, livestock is an important contributor to the food industry. The amount of phosphorus going for food comes from crops, fishery and animal production. In the EU, some of this food reaches the end consumers, whereas the rest is lost [73].

2.2.4. Phosphorus waste generation

The amount of P in waste comes from food wastes, animal wastes and wastes from households. Food waste originates from six main sectors including primary production, processing, wholesale and retail, food service and households [74]. The EU Commission is trying to reduce food waste generation to enhance a circular economy [75]. Also,

P in wastes comes from human excreta and household waste included in the wastewater stream [76]. Waste coming from wastewater goes into a wastewater collecting system that can be divided into urban wastewater collecting systems (UWWCS) and independent wastewater collecting systems (IWWCS) depending on the source of waste.

2.2.5. Phosphorus treatment and recycling

Waste treating is performed differently for different types of waste. Waste from the UWWCS goes to urban wastewater treatment plants (UWWTP), and waste from IWWCS goes to either the UWWTP or the independent wastewater treatment plants (IWWTP). The recycling of P from wastewater is done through treatment operations from WWTP. So far, the main method applied is the anaerobic digestion method [22]. Production of sludge occurs in wastewater treatment plants after wastewater has already been treated. Sludge is disposed in a variety of ways including its use in agriculture. Collected solid waste is immediately treated and then either recycled or used in other applications (e.g. landfilling). Composting is one of the main methods applied for solid waste recycling to obtain necessary nutrients for fertilizers [77].

After the treatment of phosphorus originated from waste, some of them go for recycling. Recycled P is used in fertilizers, where the largest amount of recycled P comes from manure. The recycling processes for manure to agriculture include anaerobic digestion and composting [78]. Other sources (wastewater and solid waste) are also considerable sources of recycled P. They contain a considerable amount of phosphorus ranging from 2% to 6% in wastewater sludge and around 4 g of P per 1 kg of total food waste generated [35]. Mateo-Sagasta et al. [32] estimate the amount of P₂O₅ included in sludge to be from 1.5% to 4% of dry sludge mass. In sewerage systems, however, where dry sludge is not yet extracted, the concentration of phosphorus ranges from 1 to 2 mg·L⁻¹ [79]. The recycling of phosphorus takes place through wastewater treatment and recycling of sewage sludge, which is then applied to soil as fertilizer. Phosphorus originating from solid waste is also used in fertilizers [35,80].

2.3. Mathematical formulations

The mathematical representation of the equations is based on the system dynamics model of the P life cycle in the EU. Stocks and flows are so far the foundations of the system dynamics modelling. In system dynamics, a stock (i.e. stock of material) corresponds to an entity that is subject to accumulate or drain with the passage of time. The dynamic behaviour of stock in the period $t - t_0$ is given by a time integral of the net inflows of input rate minus the net outflows of output rate. The general mathematical representation of stocks and flows is given by the following equation:

$$\text{Stock}(t) = \int_{t_0}^t (\text{Inflow}(t) - \text{Outflow}(t)) dt + \text{Stock}(t_0) \quad (1)$$

Flows correspond to the rates at which a stock changes at time t . Some flows lead to the increase in a stock (Inflow), while other flows lead to the decrease in the stock (Outflow). The general dynamic behaviour of a flow is given by a time derivative of the amount of units (i.e. material) in the stock at time t .

$$\text{Flow}(t) = \frac{dx}{dt} \quad (2)$$

where x is the amount of units in the stock. In this section, the equations that were used to obtain the graphical and statistical estimations are explained. The complete list of mathematical formulations and equations can be seen in the Supplementary Material (Table S1).

The first activity that occurs in the P life cycle is the mining production from phosphate rock ($M_R(t)$). Mathematical representation of the

total imports to the life cycle of phosphorus ($TI_R(t)$) corresponds to the summation of all imports of P into the system.

$$TI_R(t) = MI_R(t) + PI_R(t) + FI_R(t) + DI_R(t) + CI_R(t) + FAI_R(t) + FEI_R(t) + LI_R(t) + AFI_R(t) + FSHI_R(t) + IPI_R(t) \quad (3)$$

where, $MI_R(t)$ is the mined P import rate, $PI_R(t)$ is the phosphoric acid import rate, $FI_R(t)$ is the fertilizers import rate, $DI_R(t)$ is the detergents import rate, $CI_R(t)$ is the crops import rate, $FAI_R(t)$ is the food additives import rate, $FEI_R(t)$ is the feed additives import rate, $LI_R(t)$ is the live-stock import rate, $AFI_R(t)$ is the animal food import rate, $FSHI_R(t)$ is the fish products import rate and $IPI_R(t)$ is the iron ores import rate. The cumulative amount of total P imports to the system follows this mathematical formulation:

$$TIS(t) = \int_{t_0}^t TI_R(t) dt + TIS(t_0) \quad (4)$$

Material that exits the system boundaries ($TE_R(t)$) corresponds to the total exports of P at different life cycle stages. The mathematical formulation of the total exports is as follows:

$$TE_R(t) = ME_R(t) + PE_R(t) + FE_R(t) + DE_R(t) + CE_R(t) + FAE_R(t) + FEE_R(t) + LE_R(t) + AFE_R(t) + FSHE_R(t) + IPE_R(t) \quad (5)$$

where, $ME_R(t)$ is the mined P export rate, $PE_R(t)$ is the phosphoric acid export rate, $FE_R(t)$ is the fertilizers export rate, $DE_R(t)$ is the detergents export rate, $CE_R(t)$ is the crops export rate, $FAE_R(t)$ is the food additives export rate, $FEE_R(t)$ is the feed additives export rate, $LE_R(t)$ is the live-stock export rate, $AFE_R(t)$ is the animal food export rate, $FSHE_R(t)$ is the fish products export rate and $IPE_R(t)$ is the iron ores export rate.

In this study, the total recycled material rate ($TR_R(t)$) represents rate of recycled material from solid wastes ($SWR_R(t)$), wastewater ($WWR_R(t)$) and manure ($MR_R(t)$). The recycled material rate is given by the following equation:

$$TR_R(t) = SWR_R(t) + WWR_R(t) + MR_R(t) \quad (6)$$

The total flow of material loss ($TL_R(t)$) corresponds to the loss of P from different sources. Those are mining losses ($ML_R(t)$), beneficiating loss ($BL_R(t)$), processing loss ($PL_R(t)$), P runoff from the soil ($PR_R(t)$), animal food loss ($AFL_R(t)$), crops loss ($CL_R(t)$), fish loss ($FSHL_R(t)$), uncollected wastewater ($UWW_R(t)$), and non-treated wastewater ($NWW_R(t)$).

$$TL_R(t) = ML_R(t) + BL_R(t) + PL_R(t) + PR_R(t) + AFL_R(t) + CL_R(t) + FSHL_R(t) + UWW_R(t) + NWW_R(t) \quad (7)$$

The consumption of phosphorus in the P life cycle is considered through the human consumption of food commodities and laundry detergents. Thus, the mathematical representation of the P consumption in the EU ($PC_R(t)$) is as follows:

$$PC_R(t) = FOC_R(t) + DC_R(t) \quad (8)$$

where $FOC_R(t)$ is the total food consumption rate, and $DC_R(t)$ is the detergents consumption rate. To determine the reliance of the EU on the P imports, the following mathematical formula which was provided by the EU Commission is used:

$$\frac{\text{Imports} - \text{Exports}}{\text{Imports} - \text{Exports} + \text{Domestic Production}} \times 100\% \quad (9)$$

2.4. Verification and validation of the model

The verification of the model was done through checking each component in the model, its definition, its description and its purpose for

Table 1
Comparison table between historical data (tonne) and model estimations

Year	Historical data			Model estimations		
	Feed production	Crop production	Animal production	Feed production	Crop production	Animal production
2000	589650	1595874	190272	626784	1740370	194385
2001	598895	1599706	188926	690066	1661700	193327
2002	613455	1633232	189723	630757	1700240	194545
2003	540161	1426056	189753	749929	1580210	196742
2004	704618	1809815	187366	672258	1704470	195146
2005	638798	1628047	188011	642702	1628050	196319
2006	613201	1532775	187231	653657	1643060	194787
2007	609075	1519230	187222	690321	1640990	199099
2008	717569	1766737	188600	604794	1732720	194148
2009	693059	1699996	187030	604460	1677070	198824
2010	660191	1598925	188163	640484	1593190	197599
2011	673757	1657020	190554	673306	1613030	192983
2012	649301	1598555	190466	595372	1657000	204672
2013	706524	1725552	191677	702369	1579410	197616
2014	777078	1867480	198569	592381	1664340	193147
2015	734048	1781130	202676	661445	1705590	202943
Average	657461	1652508	190390	651943	1657590	196643

this study. Next, the logic of the flows was investigated. After having all inputs to the developed model, the model was run and analysed simultaneously.

The aim for the validation of the model is to make sure that the model represents the system under study. In system dynamics, the outputs of the model represent the trend of the dynamic behaviour of the actual system in real life. In this study, the validation method applied is the behavioural validation. The outcomes of the main variables in the system are compared with historical data of the same variables. The aim of this method is to check how closely do model outcomes reflect the real historical data [81]. The validation process was conducted by considering three variables; those are feed production, crop production and animal production. The outcomes of the model are obtained and compared with real historical data between the years 2000 and 2015 (Table 1).

3. Results and Discussion

The results of the simulation model show a quantitative estimation of phosphorus flows in the EU-28 between 2000 and 2050. These results are based on the current recycling efficiencies in the EU for manure, solid waste and wastewater. Table 2 demonstrates the estimated results of the model regarding the major P flows in the system.

Between the years 2000 and 2015, the results estimate that P imports correspond to the largest P flow to the EU, where mined and processed P corresponds to around 42% of total imports, and fertilizers imports correspond to around 48% of the total P imported. The recycling rates come next to the P imports with around 87% coming from manure recycling, and the rest comes from wastewater and

solid waste recycling. The model estimations show that P production mainly comes from fertilizers production (60% of the total P production). The rest comes from feed compounds, food additives and sodium triphosphates production. P exports from the EU are mainly characterized by fertilizers, contributing to around 60% of the total P exported. Around 15% of the total exports come from domestically mined and processed P. The rest (around 25% of total P exports) come from exports of food commodities (crops, fish, livestock, animal dairy), chemical derivatives (feed and food additives) and laundry detergents. The results show a significant loss of phosphorus throughout the P life cycle. This loss is estimated to be around 1.3 Mt P, where around 40% is caused by food, crops and animal production loss, and around 22% is caused by phosphorus runoff from the soil. The loss of P caused by mining, beneficiating and processing of primary phosphorus is estimated around 16% of the total P loss. Households, human wastes, animal wastes and wastewater treatment activities also contribute around 22% of P loss.

Table 2 provides the quantitative estimations of the P flows in a time interval of 50 years (2000–2050). Based on these results, the fertilizer imports continue to be the largest amount among other imports, with around 53% of the total P imports. Around 28% of the total imports is expected to come from mined and processed P. Whereas the rest (19%) comes from imports of detergents, crops, fish, livestock, animal dairy, feed and food additives. The loss of P is estimated to decrease, reaching an average of around 1 Mt P. the model results show that around 53% of P loss comes from crops, food and animal production. The runoff of P from the soil is estimated to cover around 24% of the total material loss. On the other hand, the mining, beneficiating and processing loss is estimated to decrease reaching around 8% of total P loss. The rest of P loss (up to 15%) comes from households, human wastes, animal

Table 2
Model estimations of the major P flows (tonne) in the EU (2000–2050)

Year	Mining	Production	Consumption	Imports	Exports	Loss	Recycling
2000	101424	2247980	363289	4191550	1150740	1240290	2103070
2005	111103	2317430	372405	4040520	1225550	1316150	2177210
2010	110334	2098740	389985	3536540	1301950	1286200	2145750
2015	129136	2065790	386289	3636620	1377460	1185510	2223120
2020	127346	1967780	384934	3407530	1490990	1135330	2252250
2025	133631	1915200	384334	3097140	1464100	1086780	2287910
2030	139916	1793900	381483	3082880	1479890	1069350	2323050
2035	146202	1625900	376819	3162810	1527600	1070440	2358590
2040	152487	1567750	374827	3125310	1596180	1079880	2392630
2045	158772	1502600	372621	3127000	1666690	1081240	2426580
2050	165057	1470870	371234	3201600	1746520	1087610	2461050

wastes and wastewater treatment activities. In the upcoming years, the P production is estimated to decrease, having an average of 1.7 Mt P annually. From which around 50% comes from fertilizers production, 34% from feed production, 14% from food additives production and the rest comes from sodium triphosphates production. The results of this study show an increase of the total P exports starting from 2016. Up to 57% of total exports comes from fertilizer exports, whereas only around 8% comes from mined and processed P. The rest comes from exports of crops, livestock, fish, detergents, feed and food additives. The recycling of P is estimated to grow in the future, reaching an average of around 2.3 Mt P annually. Manure is estimated to be the largest source of recycled P, with around 89% of the total recycled phosphorus.

Fig. 2 shows the dynamic behaviour of the annual major flows of P by considering the total population and *per capita*. From 2015 afterwards, the results correspond to the quantitative estimations based on the developed model of the P life cycle in the EU. The imports of P are predominantly the largest flows in the EU. Recycling is the next largest flow of P in the EU. Fig. 2(a) shows a declining trend for the imports and a growing trend for recycling. This can be clearly shown in Fig. 2(c,d) as the cumulative flow of material recycling is growing exponentially. Whereas the cumulative flow of imports shows a logarithmic growth. The year-by-year change of the annual P consumed is considerably stable compared with other flows of P in the EU. The P consumption is primarily related to the population growth in the EU. Although the estimation of P mined shows an increasing trend, the mining activities are not enough to cover the P demand in production and consumption, as can be shown from Fig. 2(a,b). Fig. 2(c,d) shows the growing gap between different P flows in the EU. There is a growing

difference between the amount of P loss and the P consumption. This gap has been addressed by Withers *et al.* [24] because of inefficient use of P. Therefore, policies on reducing the material loss are recommended. Considering food losses from the food supply chain, there is a potential to decrease waste losses from the food chain to a significant level by changing the consumption behaviour and food chain adjustments [13]. In addition, reducing P losses from fertilizer runoff helps in dealing with the total loss in the life cycle stages. P loss from fertilizer runoff has been already discussed in several researches [36,82–84]. Suggestions have been made to treat P rich soils and to regulate the application of fertilizers to the soil [36,84].

4. Improving Recycling Outputs

Higher recycling rate is probably the best solution to handle the phosphorous challenge in Europe [85]. As shown in other works, there are many possibilities for recycling waste and by-products, including animal manure, human excreta, food waste, crop losses, and sewage sludge [41], which were presented in other works. For example, the scale of P recovery in wastewater treatment plants was analysed in detail by Schoumans *et al.* [22]. The comparison of 19 relevant P recovery technologies was discussed in detail by Egle [86]. Moreover, different possible processes and technologies for obtaining P from recycling were discussed by a range of studies [22,24,35,87]. There are several options to recover P. However, many techniques still have to be tested in practice at pilot and full scale, especially those for recovering P from manure [22]. In this paper, we do not make any statement regarding the likelihood of technology implementation or the costs associated with the different recycling options. The technological and economic

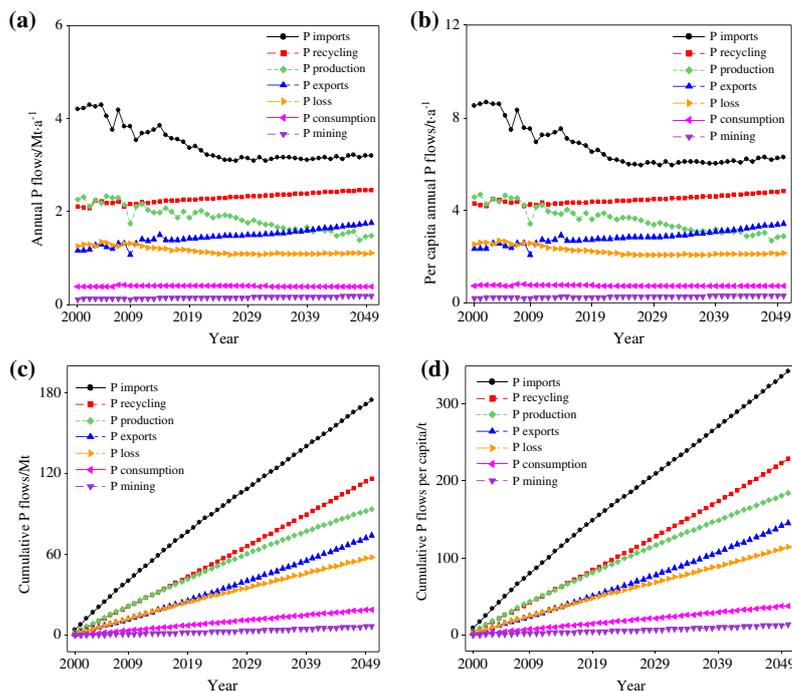


Fig. 2. Estimations of the major P flows from 2000 to 2050. (a) Annual flows. (b) Annual per capita flows. (c) Cumulative flows. (d) Cumulative per capita flows.

evaluation of phosphorus recovery has been carried out by other researchers [86,88].

4.1. Scenario development

Several studies have shown the significance of improving P recycling to lessen the dependency on phosphate rock extraction. The continuous emergence of new technologies shows the significance of improving P recycling [53]. This improvement can be done on the various P recovery sources [54]. In the EU particularly, improving the quality of P recycling is one of the potential options to resolve the concerns on the European phosphorus life cycle. This includes the implementation of new technologies to remove P from household wastes and improving the quality of manure treatment and separation [24]. Moreover, Cooper and Carliell-Marquet [34] suggested that reducing the reliance on imports of fertilizers can be done through developing new methods for recycling manure, food waste and sewage sludge.

The need to develop and establish new technologies for phosphorous recovery has been identified in many works [89–91]. The feasibility of achieving a higher rate of P recycling has been studied by several authors, e.g. Shu [88] and Cordell [92]. In Europe, the recycling of phosphorus from waste streams is low. For example, about 37% of P in municipal sewage sludge is currently recovered and reused in agriculture [25]. New processes and technologies to recover phosphorus from surplus manure are under development in the Netherlands [93]. In Sweden, it is anticipated that improving P recycling is one of the main solutions to meet at least 30% of future demand [92]. Based on the survey by the Stockholm Environment Institute (SEI), the current reliance on imported rock-based phosphorus ("3 kg P per European citizen per year") cannot be continued for a very long time. They suggested that for achieving sustainability in each step of the agriculture and food supply chain, the efficiency of phosphorus use must approach a level close to 100%. Therefore, there should be recommended a full recycling of phosphorous in Europe [94].

The scenario-based approach has been deployed by Mourad *et al.* [95] to determine the impact of recycling on greenhouse gas emissions through applying different recycling scenarios. Evaluation of various recycling scenarios was also performed by Giannis *et al.* [96]. They assessed alternative strategies for solid waste management by implementing a system dynamics model. In this study, modifications to the model include three different scenarios for improving recycling by: 15% (Scenario A), 30% (Scenario B) and 45% (Scenario C). These scenarios take into consideration the current recycling output of P from solid waste, wastewater and manure. The main reason for considering these values is the technological

possibilities of phosphorus economic recycling. The recently introduced technologies, e.g. struvite fertilizers [97,98], allow obtaining phosphorous recycling at the level of 15%. Therefore, it is probable that economic recycling of phosphorus reaches the 50% level in the next 12 to 15 years [99].

4.2. Results after application of scenarios

The improvements in recycling applied to the model are the same in each scenario because recycling efficiencies are modified with the same proportion by adding 15% after each iteration. Improving recycling streams of the P life cycle in the EU lessens the dependency on P imports to a certain extent. Fig. 3 shows the changes on the annual P flow following the different scenario applications. The annual flow of P recycling increases after each scenario by around 335000 t P (Fig. 3(a)). Consequently, the annual P imports decrease by the same proportion following the increase of P recycling (Fig. 3(b)). The analysis of the flow estimations in different scenarios shows that in scenario A, the annual imports still exceed the annual P recycled, with an average amount of imports more than P recycled by around 65000 t P. In scenario B, the annual P recycling rate is estimated to exceed the annual imports starting from the year 2016. This improvement continues in scenario C.

Fig. 4 shows the estimated results of P cumulative flows with current recycling outputs and under developed scenarios in a time range of 50 years (2000–2050). Fig. 4(a) corresponds to the cumulative P flow estimations given the current recycling efficiencies. Fig. 4(b,c) illustrates the effect of recycling improvement in a longer perspective. Despite the significant increase of P recycling in scenarios A and B, the total P imports are still contributing more material to the P life cycle in the EU following a logarithmic growth. In scenario C, the annual increase on P recycling is estimated to reach around 1 mt P. Considering this growth in recycling, the cumulative amount of P recycled is subject to exceed that of the total P imports following an exponential growth. This result is possible given a long time horizon of 50 years (Fig. 4(d)).

Several techniques and methods for improving P recovery and recycling have been discussed in the literature. Guedes *et al.* [100] and Schütte *et al.* [101] show techniques related to the P recovery from sludge, such as electrodialytic process and nanofiltration. Methods on P recovery from wastewater has been also discussed. Enhanced biotechnological approaches for the P recovery from wastewater is an efficient process and can be utilized [87]. In addition, Kalmykova and Fedje [35] illustrate the acid and base leaching precipitation procedures to obtain P from solid waste incineration residues.

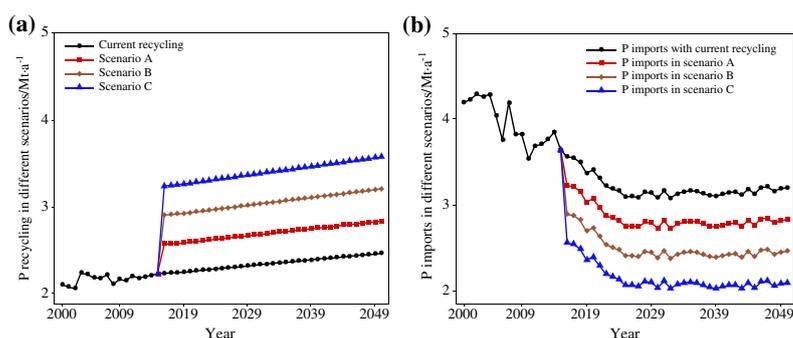


Fig. 3. P annual flow estimations, with current recycling efficiencies and in different scenarios.

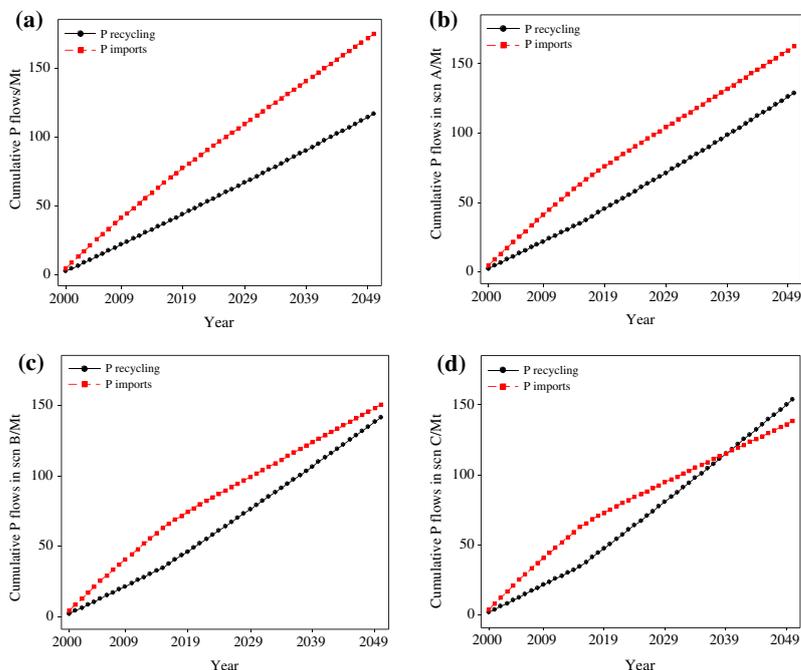


Fig. 4. Cumulative P flow estimations for recycling and imports in the EU (2000–2050) with current recycling efficiencies and in different scenarios.

4.3. The EU reliance on imports

The results of this study go side by side with previous studies that show the high reliance of the EU on the P imports. The main aim of the EU is to lessen the reliance level on the P imports. The model estimates the dynamic behaviour of the reliance percentage with the

Table 3
Percentage of the EU reliance on P imports

Year	Current/%	Scenario A/%	Scenario B/%	Scenario C/%
2000	97			
2001	97			
2002	97			
2003	97			
2004	96			
2005	96			
2006	96			
2007	96			
2008	96			
2009	97			
2010	95			
2011	95			
2012	95			
2013	95			
2014	95			
2015	95			
Ave. (2000–2015)	96			
2016	95	94	93	91
2020	94	93	91	88
2030	92	90	87	80
2040	91	88	84	75
2050	90	87	81	68
Ave. (2016–2050)	92	90	86	79

passage of time by considering equation 9. These estimates are given under the two periods, historical period (2000–2015) and future period (2016–2050). The model estimations on P import reliance are shown in Table 3. This includes estimations given current recycling efficiencies, and estimations given the different scenarios. Between the years 2000 and 2015, the results of the model show a decreasing trend in the reliance percentage on P imports. This decreasing trend is estimated to continue with all given scenarios from 2016 to 2050 (Fig. 5).

Currently, the model estimates the reliance percentage on P imports is around 96% between 2000 and 2015. By considering the historical data for the developed model, the model also shows that this percentage

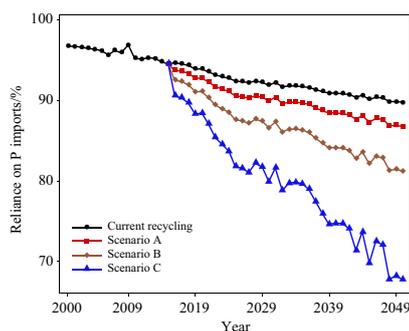


Fig. 5. Estimations for the EU reliance on P imports in different scenarios.

decreases with the passage of time between 2016 and 2050, with an average of 92% (Table 3). With the application of scenarios, this percentage decreases more to 90%, 86% and 79% in scenarios A, B and C respectively.

The results of this paper support previous studies on the European phosphorus balance. Our calculations indicate a high inefficiency in the phosphorous supply chain with around 1.3 million tonnes (Mt) phosphorous lost in 2005. Previous studies e.g. Dijk et al. [14] estimated 1.22 Mt phosphorous loss in the same year. The total trade balance of phosphorous was estimated to be around 2.1 Mt in 2005. In our study, the trade balance of phosphorous reaches up to 2.8 Mt in the same year. The results of this study show an average of 86% reliance on phosphate rock between the years 2010 and 2014, compared with a reliance of 88% based on a report by the EU Commission considering the same time interval [11]. Overall, the obtained results confirm previous research indicating a high dependency on P imports and a high inefficiency of the European phosphorous supply chain [22,44].

The EU Commission has already presented the reliance level [11]. However, this corresponds particularly to the trade of phosphate rock. The results presented in this study determine the overall reliance level on P imports to 28 member countries of the EU by considering multiple sectors, including mining, processing of phosphorous into phosphoric acid, agriculture production, and detergents production. Our model could be useful not only to inform policy makers about the current situation of P recycling, but also to explore new options for further improvements of phosphorous recovery. It quantifies the potential impact of future developments in recycling technology on the phosphorous supply chain.

4.4. Limitations of the study

The limitation of the study is lack of the detailed analysis of white phosphorous (P₄) flow. At present, phosphate rock is the only primary input material for phosphorous production [102]. Based on the study by the EU Commission [11] on the list of critical raw materials, P₄ is an elemental form of phosphorous, produced from phosphate rock. The EU does not produce P₄. Therefore, white phosphorous was not analysed in the EU study on critical raw materials in years 2011 and 2014. In consequence, there is not enough available data for a detailed analysis of its flow. Another limitation of this study is lack of the technical and economic assessment of technologies for improvement of phosphorous recycling as well as their environmental impact. These limitations are being addressed in research currently underway.

5. Conclusions

Due to the risk associated with the main source of the supply of phosphorous (phosphate rock) and its growing economic importance, recycling is considered one of the options to decrease the supply risk as much as possible, especially in Europe, the region that lacks the phosphate rock reserves. The aim of this study is to calculate the current level of recycling through the various waste streams (solid waste, water treatment and manure) and to estimate the enhancement degree of this level, which impacts on the criticality of the element P in Europe. To quantify the effects of recycling efficiency, a system dynamics model was constructed and applied to analyse different scenarios. The results show that with improving recycling, there is a potential to reach an annual decrease of around 1 million t P from total P imports. This achievement can lessen the EU reliance on imports from 96% to around 79%. Results of this study may potentially bring decision-making to a continental scale where decisions are designed to face the challenge of phosphorous supply chain in Europe. Consequently, it helps to tackle the problem of dependency on imports of phosphorous and the continuous resource extraction.

Supplementary Material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cjche.2018.09.004>.

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1 Supplementary Material

2 Table S1

3 Equations and calculation processes for the proposed phosphorus supply chain modelling.

Variable	Term	Unit	Equation	Coefficients and variables
I. Mining and Processing				
$MS(t)$	Mined P stock	Tonnes P	$\int_0^t (M_R(t) + M_{Rk}(t) - B_R(t) - BL_R(t) - ME_R(t))dt + MS(t_0)$	k_0 : P content in phosphate rock M_0 : Mining variable
$M_R(t)$	Mining rate	Tonnes P.a ⁻¹	$M_0 \times k_0$	
$B_R(t)$	Beneficiating rate	Tonnes P.a ⁻¹	$M_1 \times MS(t)$	M_1 : Beneficiating coefficient
$BS(t)$	Beneficiated P stock	Tonnes P	$\int_0^t (B_R(t) - PL_R(t) - P_R(t))dt + MS(t_0)$	
$PS(t)$	Phosphoric acid stock	Tonnes P	$\int_0^t (P_R(t) - FEPR(t) - FAAR(t) - DP_R(t))dt + PS(t_0)$	
$PR(t)$	Processing rate	Tonnes P.a ⁻¹	$M_2 \times BS(t)$	M_2 : Processing coefficient from primary material to phosphoric acid
II. Production				
$FEPR(t)$	Fertilizer production rate	Tonnes P.a ⁻¹	$P_0 \times k_2$	P_0 : Production variable for fertilizers k_2 : P content in P-fertilizers
$FAAR(t)$	Feed production rate	Tonnes P.a ⁻¹	$P_1 \times k_6$	P_1 : Production variable for feed
$FA_R(t)$	Food additives production rate	Tonnes P.a ⁻¹	$P_2 \times k_3$	P_2 : Production variable for food additives k_3 : P content in food additives
$DP_R(t)$	Detergents production rate	Tonnes P.a ⁻¹	$P_3 \times k_4$	P_3 : Production variable for detergents k_4 : P content in STPP
$DS(t)$	Detergents stock	Tonnes P	$\int_0^t (DP_R(t) + DL_R(t) - DE_R(t) - DC_R(t))dt + DS(t_0)$	

Variable	Term	Unit	Equation	Coefficients and variables
$FS(t)$	Fertilizers stock	Tonnes P	$\int_0^t (FEP_R(t) + IP_R(t) + DF_R(t) + FI_R(t) + FE_R(t) + TR_R(t) - CR_R(t) - PR_R(t) - GC_R(t))dt + FS(t_0)$	
$IP_R(t)$	Fertilizers from iron ores flow rate	Tonnes P.a ⁻¹	$IP_R(t) + IP_I_R(t) - IP_I_R(t)$	P_1 : Production variable of phosphates from iron ores
$IPP_R(t)$	Iron ores production rate	Tonnes P.a ⁻¹	$P_4 \times k_5$	P_4 : Deposition variable
$DF_R(t)$	Deposition flow rate	Tonnes P.a ⁻¹	P_8	P_8 : Production variable for crops
$CP_R(t)$	Crop production rate	Tonnes P.a ⁻¹	P_5	P_5 : Production variable for crops
P_5	Production variable for crops	Tonnes P.a ⁻¹	$\sum_{i=1}^{18} P_{5(i)} \times k_{5(i)}$	$P_{5(i)}$: Production rate of crop i $k_{5(i)}$: P content in crop i
$GC_R(t)$	Grass cut rate	Tonnes P.a ⁻¹	$P_6 \times FS(t)$	P_6 : Grass cutting coefficient
$FAS(t)$	Feed Stock	Tonnes P	$\int_0^t (FAP_R(t) + GC_R(t) + CF_R(t) + CF_I_R(t) - FEC_R(t))dt + FAS(t_0)$	
$CS(t)$	Crops Stock	Tonnes P	$\int_0^t (CP_R(t) + CI_R(t) - CE_R(t) - CF_R(t) - NG_R(t) - NC_R(t))dt + CS(t_0)$	
$CF_R(t)$	Crops to feed rate	Tonnes P.a ⁻¹	$P_7 \times CS(t)$	P_7 : Crops flow coefficient to feed stock
$NC_R(t)$	Net crops flow rate	Tonnes P.a ⁻¹	$P_9 \times CS(t)$	P_9 : Net crops flow coefficient
$CFO_R(t)$	Crops flow rate to food	Tonnes P.a ⁻¹	$P_{10} \times NCS(t)$	P_{10} : Crops flow coefficient to food
$FSH_R(t)$	Fish flow rate	Tonnes P.a ⁻¹	$NFSH_R(t) - FSHL_R(t)$	
$FSHP_R(t)$	Fish production rate	Tonnes P.a ⁻¹	$P_{11} \times k_8$	P_{11} : Fish production variable
$NFSH_R(t)$	Net fish catch rate	Tonnes P.a ⁻¹	$FSH_R(t) + FSHP_R(t) - FSHE_R(t)$	
$APP_R(t)$	Animal food production rate	Tonnes P.a ⁻¹	P_{12}	P_{12} : Production variable for animal food
P_{12}	Production variable for animal food	Tonnes P.a ⁻¹	$\sum_{i=1}^5 P_{12(i)} \times k_{12(i)}$	$P_{12(i)}$: Production rate of animal food i $k_{12(i)}$: P content in animal food i
$FAS(t)$	Food additives stock	Tonnes P	$\int_0^t (FAP_R(t) + FAI_R(t) - FAE_R(t) - FAC_R(t))dt + FAS(t_0)$	
Imports				
$MI_R(t)$	Mined P import rate	Tonnes P.a ⁻¹	$I_0 \times k_6$	I_0 : Mined P import variable

Variable	Term	Unit	Equation	Coefficients and variables
$PI_R(t)$	Phosphoric acid import rate	Tonnes P.a ⁻¹	$I_1 \times k_1$	I_1 : Phosphoric acid import variable k_1 : P content in phosphoric acid
$IPI_R(t)$	Iron ores import rate	Tonnes P.a ⁻¹	$I_2 \times k_5$	I_2 : Import variable for iron ores
$FI_R(t)$	Fertilizers import rate	Tonnes P.a ⁻¹	$I_3 \times k_2$	I_3 : Import variable for fertilizers
$FEI_R(t)$	Feed import rate	Tonnes P.a ⁻¹	$I_4 \times k_6$	I_4 : Import variable for feed k_6 : P content in Feed
$CI_R(t)$	Crops import rate	Tonnes P.a ⁻¹	$I_5 \times k_7$	I_5 : Import variable for crops
$DI_R(t)$	Detergents import rate	Tonnes P.a ⁻¹	$I_7 \times k_4$	I_7 : Import variable for STPP
$FAI_R(t)$	Food additives import rate	Tonnes P.a ⁻¹	$I_6 \times k_2$	I_6 : Import variable for food additives k_2 : P content in food additives
$FSHI_R(t)$	Fish import rate	Tonnes P.a ⁻¹	$I_8 \times k_9$	I_8 : Fish import variable
$MI_R(t)$	Mined P import rate	Tonnes P.a ⁻¹	$I_0 \times k_0$	I_0 : Mined P import variable
$LI_R(t)$	Livestock import rate	Tonnes P.a ⁻¹	$I_9 \times k_9$	I_9 : Import variable for livestock k_9 : P content in livestock
Exports				
$ME_R(t)$	Mined P export rate	Tonnes P.a ⁻¹	$E_0 \times k_0$	E_0 : Mined P export variable
$PE_R(t)$	Phosphoric acid export rate	Tonnes P.a ⁻¹	$E_1 \times k_1$	E_1 : Phosphoric acid export variable
$IPE_R(t)$	Iron ores export rate	Tonnes P.a ⁻¹	$E_2 \times k_5$	E_2 : Export variable for iron ores
$FE_R(t)$	Fertilizers export rate	Tonnes P.a ⁻¹	$E_3 \times k_2$	E_3 : Export variable for fertilizers
$FEE_R(t)$	Feed export rate	Tonnes P.a ⁻¹	$E_4 \times k_6$	E_4 : Export variable for feed
$CE_R(t)$	Crops export rate	Tonnes P.a ⁻¹	$E_5 \times k_7$	E_5 : Export variable for crops
$FAE_R(t)$	Food additives export rate	Tonnes P.a ⁻¹	$E_6 \times k_2$	E_6 : Export variable for food additives k_2 : P content in food additives
$DE_R(t)$	Detergents export rate	Tonnes P.a ⁻¹	$E_7 \times k_4$	E_7 : Export variable for detergents
$FSHE_R(t)$	Fish export rate	Tonnes P.a ⁻¹	$E_8 \times k_9$	E_8 : Export variable for fish
$LE_R(t)$	Livestock export rate	Tonnes P.a ⁻¹	$E_9 \times k_9$	E_9 : Export variable for livestock
III. Consumption				
$LS(t)$	Livestock	Tonnes P	$\int_{t_0}^t (FEG_R(t) - AFP_R(t) - FC_R(t) - AM_R(t))dt + LS(t_0)$	

Variable	Term	Unit	Equation	Coefficients and variables
$AFS(t)$	Animal food stock	Tonnes P	$\int_0^t (AFR_R(t) - AFC_R(t) - AFL_R(t))dt + AFS(t_0)$	
$FEC_R(t)$	Feed consumption rate	Tonnes P.a ⁻¹	$C_0 \times FAS(t)$	C_0 : Feed consumption coefficient
$FOC_R(t)$	Food consumption rate	Tonnes P.a ⁻¹	$VFD_R(t) + AFD_R(t)$	
$VFD_R(t)$	Vegetable food Demand rate	Tonnes P.a ⁻¹	$C_2 \times P(t)$	C_2 : Demand of vegetable food coefficient per one person $P(t)$: EU population
$AFD_R(t)$	Animal food demand rate	Tonnes P.a ⁻¹	$C_3 \times P(t)$	C_3 : Demand of animal food coefficient per one person
$DC_R(t)$	Detergent consumption rate	Tonnes P.a ⁻¹	$C_4 \times DS(t)$	C_4 : Detergent consumption coefficient
$FOS(t)$	Food Stock	Tonnes P	$\int_0^t (CFO_R(t) + FAC_R(t) + FOI_R(t) + AFC_R(t) + FSH_R(t) - FL_R(t) - FOC_R(t) - FOE_R(t) - FW_R(t))dt + FOS(t_0)$	
$FAC_R(t)$	Food additives consumption rate	Tonnes P.a ⁻¹	$C_4 \times FAS(t)$	C_4 : Food additives consumption coefficient
$NCS(t)$	Net crops stock	Tonnes P	$\int_0^t (NCR(t) - CL_R(t) - CFO_R(t))dt + NCS(t_0)$	$NCS(t)$
IV. Waste generation				
$AW_R(t)$	Animal waste rate	Tonnes P.a ⁻¹	W_0	W_0 : Waste flow variable from animals
$HER(t)$	Human excreta flow rate	Tonnes P.a ⁻¹	$W_1 \times FOC_R(t)$	W_1 : Waste flow coefficient from human bodies
$SW_S(t)$	Solid wastes stock	Tonnes	$\int_0^t (AW_R(t) + FW_R(t) - NSW_R(t) - TSW_R(t))dt + SW_S(t_0)$	
$NSW_R(t)$	non treated solid waste rate	Tonnes P.a ⁻¹	$W_2 \times SWS(t)$	W_2 : Non-treated solid waste flow coefficient
$TSW_R(t)$	treated solid waste flow rate	Tonnes P.a ⁻¹	$W_3 \times SWS(t)$	W_3 : Solid waste treating coefficient
$TSW(t)$	Treated solid waste stock	Tonnes P.a ⁻¹	$\int_0^t (TSW_R(t) - SWL_R(t) - SWR_R(t))dt + TSW(t_0)$	
$SWL_R(t)$	solid waste landfilling rate	Tonnes P.a ⁻¹	$W_4 \times TSW(t)$	W_4 : Solid waste landfilling coefficient
$WWS(t)$	Wastewater stock	Tonnes P	$\int_0^t (HE_R(t) + DC_R(t) - UWW_R(t) - CWWR_R(t))dt + WWS(t_0)$	

Variable	Term	Unit	Equation	Coefficients and variables
$CWW_R(t)$	wastewater collecting rate	Tonnes P _a ⁻¹	$W_5 \times WW_S(t)$	W_5 : Wastewater collecting coefficient
$CWW(t)$	collected wastewater	Tonnes P	$\int_{t_0}^t (CWW_R(t) - NWW_R(t) - TWW_R(t))dt + CWW(t_0)$	
$FW_R(t)$	Food waste generation rate	Tonnes P _a ⁻¹	$W_7 \times P(t) \times k_{10}$	k_{10} : P content in food wastes W_7 : Waste generated per capita
$TWW_R(t)$	wastewater treating rate	Tonnes P _a ⁻¹	$W_6 \times CWW(t)$	W_6 : Wastewater treating coefficient
$TWW(t)$	treated wastewater stock	Tonnes P	$\int_{t_0}^t (TWW_R(t) - WR_R(t) - SL_R(t) - SO_R(t))dt + TWW(t_0)$	
$SL_R(t)$	sludge landfilling rate	Tonnes P _a ⁻¹	$W_7 \times TWW(t)$	W_7 : Sludge landfilling coefficient
$SO_R(t)$	Sludge flow rate for other uses	Tonnes P _a ⁻¹	$W_8 \times TWW(t)$	W_8 : Sludge use for other uses coefficient
V. Recycling				
$MR_R(t)$	Manure recycling rate	Tonnes P _a ⁻¹	$\sum_{i=1}^n R_{0(i)} \times k_{5,10(i)}$	$R_{0(i)}$: manure from animal i $k_{5,10}$: P content in manure from animal i
$SWR_R(t)$	Solid waste recycling rate	Tonnes P _a ⁻¹	$R_1 \times TSW(t)$	R_1 : recycling coefficient for wastewater
$WWR_R(t)$	Wastewater recycling rate	Tonnes P _a ⁻¹	$R_2 \times TWW(t)$	R_2 : recycling coefficient for solid waste
$TR_R(t)$	Total recycling rate	Tonnes P _a ⁻¹	$MR_R(t) + SWR_R(t) + WWR_R(t)$	
VI. Loss				
$ML_R(t)$	Mining loss rate	Tonnes P _a ⁻¹	$M_R(t) \times L_9$	L_9 : P loss coefficient from mining rate
$PL_R(t)$	Phosphorus mining, beneficiating and processing loss rate	Tonnes P _a ⁻¹	$BL_R(t) + PL_R(t) + ML_R(t)$	
$BL_R(t)$	Beneficiation loss rate	Tonnes P _a ⁻¹	$L_9 \times MS(t)$	L_9 : Loss coefficient during the beneficiation process
$PL_R(t)$	Processing waste rate	Tonnes P _a ⁻¹	$L_1 \times BS(t)$	L_1 : Loss coefficient during processing to phosphoric acid
$PR_R(t)$	Phosphorus runoff rate	Tonnes P _a ⁻¹	$L_2 \times A \times T$	L_2 : Loss coefficient from fertilizers due to runoff of P A : Arable land % of total land in EU T : Total land in EU
$FL_R(t)$	Food loss rate	Tonnes P _a ⁻¹	$L_3 \times FOS(t)$	L_3 : Food loss coefficient
$AFR_R(t)$	Animal food loss rate	Tonnes P _a ⁻¹	$L_4 \times AFS(t)$	L_4 : Animal food loss coefficient
$CLR_R(t)$	Crops loss rate	Tonnes P _a ⁻¹	$L_5 \times CS(t)$	L_5 : Crops loss coefficient

Variable	Term	Unit	Equation	Coefficients and variables
$FSHL_R(t)$	Fish loss rate	Tonnes P _a ⁻¹	$L_6 \times NFSH_R(t)$	L_6 : Fish loss coefficient
$NWW_R(t)$	Non treated wastewater rate	Tonnes P _a ⁻¹	$L_8 \times CWW(t)$	L_8 : Non treated wastewater coefficient
$DWW_R(t)$	Uncollected wastewater rate	Tonnes P _a ⁻¹	$L_7 \times WWS(t)$	L_7 : Uncollected wastewater coefficient
$TL_R(t)$	Total loss rate	Tonnes P _a ⁻¹	$PMI_R(t) + PR_R(t) + FL_R(t) + AFL_R(t) + C L_R(t) + FSH L_R(t) + NWW_R(t) + DWW_R(t)$	

4 **Table S2**

5 Data sources for all inputs of the proposed model.

Input	Description	Unit	Value range	Value in this study	Data sources
k_0	P content in phosphate rock	%	13.5	13.5	[61]
k_1	P content in phosphonic acid	%	-	31.6	Mathematical calculation
k_2	P content in P_2O_5	%	-	43.6	Mathematical calculation
k_3	P content in food additives	%	-	19.7	Mathematical calculations: Avg P in potassium, calcium, magnesium and ammonium phosphates
k_4	P content in STPP	%	25	25	Mathematical calculation
k_5	P content in iron ores	%	1.112	1.112	[51]
k_6	P content in Feed	%	Avg: 0.003	0.003	[42]
$k_{7(1)}$	P content in Rice	Tonnes P-(t crop) ⁻¹	0.0041	0.0041	[42]
$k_{7(2)}$	P content in Wheat	Tonnes P-(t crop) ⁻¹	0.0051	0.0051	[42]
$k_{7(3)}$	P content in Barley	Tonnes P-(t crop) ⁻¹	0.004	0.004	[42]
$k_{7(4)}$	P content in Sorghum	Tonnes P-(t crop) ⁻¹	0.0051	0.0051	[42]
$k_{7(5)}$	P content in Millet	Tonnes P-(t crop) ⁻¹	0.0048	0.0048	[42]
$k_{7(6)}$	P content in Rye	Tonnes P-(t crop) ⁻¹	0.0045	0.0045	[42]
$k_{7(7)}$	P content in Rye	Tonnes P-(t crop) ⁻¹	0.0052	0.0052	[42]
$k_{7(8)}$	P content in Oat	Tonnes P-(t crop) ⁻¹	0.006	0.006	[42]

Input	Description	Unit	Value range	Value in this study	Data sources
$k_{7(9)}$	P content in Potato	Tonnes P-(t crop) ⁻¹	0.0008	0.0008	[42]
$k_{7(10)}$	P content in sweet potato	Tonnes P-(t crop) ⁻¹	0.001	0.001	[42]
$k_{7(11)}$	P content in Soybean	Tonnes P-(t crop) ⁻¹	0.0002	0.0002	[42]
$k_{7(12)}$	P content in Rapeseed	Tonnes P-(t crop) ⁻¹	0.007	0.007	[42]
$k_{7(13)}$	P content in Olive	Tonnes P-(t crop) ⁻¹	0.0007	0.0007	[42]
$k_{7(14)}$	P content in Sugar beet	Tonnes P-(t crop) ⁻¹	0.0007	0.0007	[42]
$k_{7(15)}$	P content in Sugar cane	Tonnes P-(t crop) ⁻¹	0.0003	0.0003	[42]
$k_{7(16)}$	P content in seed cotton	Tonnes P-(t crop) ⁻¹	0.016	0.016	[42]
$k_{7(17)}$	P content in Fruit	Tonnes P-(t crop) ⁻¹	0.0008	0.0008	[42]
$k_{7(18)}$	P content in vegetable and melon	Tonnes P-(t crop) ⁻¹	0.0007	0.0007	[42]
k_8	P content in fish	%	Avg: 0.26	0.26	FAO. Save food: global initiative on food loss and waste reduction (2018) Accessed (03 February 2018). Retrieved from http://www.fao.org/save-food/resources/keyfindings/en/
k_9	P content in Livestock	Tonnes P-(t livestock) ⁻¹	Avg: 0.002242	0.002242	Inorganic feed phosphates (IFP) (2018) Phosphorus: a virtual source of animal nutrition. Accessed (19 January 2018). Retrieved from http://www.feedphosphates.org/index.php/guides/11-guides/11-phosphorus-as-a-vital-source-of-animal-nutrition
k_{10}	P content in Food waste	g P-(kg waste) ⁻¹	4	4	[35]
$k_{11(1)}$	P content in cattle manure	%	0.074	0.074	FAO. manure a source of nutrients (2018) Accessed (01 February 2018) Retrieved from

Input	Description	Unit	Value range	Value in this study	Data sources
$k_{11(2)}$	P content in Sheep manure	%	0.014	0.014	http://www.fao.org/docrep/008/a0013e/a0013e08.htm FAO, manure a source of nutrients (2018) Accessed (01 February 2018) Retrieved from http://www.fao.org/docrep/008/a0013e/a0013e08.htm
$k_{11(3)}$	P content in poultry manure	%	0.2	0.2	FAO, manure a source of nutrients (2018) Accessed (01 February 2018) Retrieved from http://www.fao.org/docrep/008/a0013e/a0013e08.htm
$k_{11(4)}$	P content in Horse manure	%	0.13	0.13	FAO, manure a source of nutrients (2018) Accessed (01 February 2018) Retrieved from http://www.fao.org/docrep/008/a0013e/a0013e08.htm
$k_{11(5)}$	P content in swine manure	%	0.21	0.21	FAO, manure a source of nutrients (2018) Accessed (01 February 2018) Retrieved from http://www.fao.org/docrep/008/a0013e/a0013e08.htm
M_0	Mining variable	Tonnes P rock $\cdot a^{-1}$	(658347-956564)	2000-2015: (658347-956564) 2016-2050: Model estimations	British Geological Survey (BGS) (2017) Production of phosphate rock in Europe from 2000 to 2010. Accessed (01 February 2018) Retrieved from http://www.bgs.ac.uk/mineralsUK/statistics/wms.cfc?method=ListResults&dateType=Production&commodity=112&dateFrom=2000&dateTo=2010&country=270&agreeToTsAndCs=agreed British Geological Survey (BGS) (2017) Production of phosphate rock in Europe from 2011 to 2016. Accessed (01 February 2018) Retrieved from http://www.bgs.ac.uk/mineralsUK/statistics/wms.cfc?method=ListResults&dateType=Production&commodity=112&dateFrom=2011&dateTo=2016&country=270&agreeToTsAndCs=agreed
M_1	Beneficiating coefficient	Dimensionless	0.84	0.84	Mathematical calculation
M_2	processing coefficient	Dimensionless	0.87	0.87	Mathematical calculation
P_0	Production variable for fertilizers	Tonnes P ₂ O ₅ $\cdot a^{-1}$	(11022187-1396866)	2000-2015: (11022187-1396866)	Food and Agriculture Organization of the United Nations (Faostat), (2017) fertilizers by nutrient. Accessed (10 January 2018) Retrieved

Input	Description	Unit	Value range	Value in this study	Data sources
P_4	Production variable of phosphates from iron ores	Tonnes Iron ores.a ⁻¹	(24000000-40000000)	2000-2015: (24000000-40000000) 2016-2050: Model estimations	J.FIRST&sortC=ASC-- J.FIRST&rsip=&cSip=&rDCh=&cDCh=&rDM=true&DM=true&footnes=false&empty=false&wai=false&time_mode=ROLLING&lang=en Eurostat (2017) Sold production, exports and imports by PRODCOM list (NACE Rev. 2) – annual data. Accessed (25 January 2018), Retrieved from http://appsso.eurostat.ec.europa.eu/mui/show.do?query=BOOKMARK_D_S:06634I_QID_47D72158_UID_3F17EB0&layout=INDICATORS.C.X.0:DECL.Y.0:PRCODE.B.Z.0:PERIOD.L.Z.1&Selection=DS:06634I:PERIOD:200952:DS:06634I:PRCODE:07101000:&rankName1=PRCODE_1_0_-1_2&rankName2=DECL_1_0_1_0_1_2&rankName3=INDICATORS_1_0_1_0&rankName4=PERIOD_1_0_1_2&sort=ASC-- J.FIRST&sortC=ASC-- J.FIRST&rsip=&cSip=&rDCh=&cDCh=&rDM=true&DM=true&footnes=false&empty=false&wai=false&time_mode=ROLLING&lang=en
P_5	Production variable for crops		(806550-1867480)	Distribution: Weibull Shape: 6.117 Scale: 149531×10^6 Number of data points = 56 AD: 0.603 Corresponding p-value = 0.117 Sample mean = 1384791 Sample St.Dev = 277512 Minimum = 806550 Maximum = 1867480 (Table A3)	Food and Agriculture Organization of the United Nations (2017), Crops. Accessed (10 January 2018), Retrieved from http://www.fao.org/FoodandAgricultureOrganizationoftheUnitedNations (Eoastat)/en/#/data/OC
P_6	Grass cutting coefficient	Dimensionless	0.3	0.3	[42]
P_7	Crops flow coefficient to feed stock	Dimensionless	0.35	0.35	[42]
P_8	Deposition variable	Tonnes P.a ⁻¹	44000	44000	[24]
P_9	Net crops flow coefficient	Dimensionless	-	0.65	Mathematical calculation
P_{10}	Crops flow coefficient to food	Dimensionless	-	0.77	Mathematical calculation
P_{11}	Fish production variable	Tonnes P.a ⁻¹	(4419600-5496100)	2000-2015: (4419600-5496100)	Eurostat, fishery statistics (2017)

Input	Description	Unit	Value range	Value in this study	Data sources
P_{12}	Animal food production variable	Tonnes P_2O_5 -a ⁻¹	(187030-205052)	2016-2050: Model estimation Distribution:3-Parameter Weibull Shape: 1.117 Scale:6430.896 Number of data points = 36 AD: 0.485 Corresponding P value = 0.236 Sample mean = 192954 Sample St Dev = 5440.52 Minimum = 187030 Maximum = 205052 (Table A3)	Eurostat (2017). Fishery statistics. Accessed (16 January 2018). Retrieved from http://ec.europa.eu/eurostat/statistics-explained/images/002/Fisheries_statistics_in_detail_Version_6_EI-update.xlsx Food and Agriculture Organization of the United Nations (2018). The composition of fish. Accessed (13 January 2018). Retrieved from http://www.fao.org/wairdocs/iau/x5916e/x5916e01.htm
I_0	Mined P import variable	Tonnes P rock-a ⁻¹	(3899015-10047349)	2000-2015: (3899015-10047349) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for phosphate rock. Accessed (26 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?xyppm=114719125101411112113111
I_1	Phosphoric acid import variable	Tonnes phosphoric acid-a ⁻¹	(1477616-1883884)	2000-2015: (1477616-1883884) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for phosphoric acid. Accessed (23 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?xyppm=114719128092016112113111
I_2	Import variable for iron ores	Tonnes iron ores-a ⁻¹	(80071312-116460431)	2000-2015: (80071312-116460431) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for iron ores and concentrates. Accessed (05 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?xyppm=114719126011411112113111
I_3	Import variable for fertilizers	Tonnes P_2O_5 -a ⁻¹	(1738698-3207357)	2000-2015: (1738698-3207357) 2016-2050: Model estimation	Food and Agriculture Organization of the United Nations (Faostat). (2017) fertilizers by nutrient. Accessed (10 January 2018). Retrieved from http://www.fao.org/food/agriculture-organization-of-the-united-nations/faostat/en/#data/RFN
I_4	Import variable for feed	Tonnes feed-a ⁻¹	(62377364-93474426)	2000-2015: (62377364-93474426)	Trademap (2017). EU trade statistics for food additives. Accessed (23 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?xyppm=1147191283

Input	Description	Unit	Value range	Value in this study	Data sources
I_5	Import variable for crops	Tonnes crops a^{-1}	(23277940-33813545)	2016-2050: Model estimation 2000-2015: (23277940-33813545) 2016-2050: Model estimation	539161122113111 Trademap (2017). EU trade statistics for fruits. Accessed (16 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?nyvpm=1147191081121122113111 Trademap (2017). EU trade statistics for cereals. Accessed (16 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?nyvpm=1147191081121122113111 Trademap (2017). EU trade statistics for vegetables. Accessed (16 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?nyvpm=1147191071121122113111
I_6	Import variable for food additives	Tonnes polyphosphates a^{-1}	(117118-177543)	2000-2015: (117118-177543) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for food additives. Accessed (23 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?nyvpm=1147191283539161122113111
I_7	Import variable for STPP	Tonnes STPP a^{-1}	(124089-331307)	2000-2015: (124089-331307) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for Sodium triphosphates (STPP). Accessed (20 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?nyvpm=1147191283531161122113111
I_8	Fish import variable	Tonnes fish a^{-1}	(5513302-7306503)	2000-2015: (5513302-7306503) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for Fish products. Accessed (20 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?nyvpm=1147191031211122113111
I_9	Import variable for livestock	Tonnes livestock a^{-1}	(10822728-1109420)	2000-2015: (10822728-1109420) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for animal products. Accessed (10 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?nyvpm=1147191031211122113111
E_0	Mined P export variable	Tonnes P rock a^{-1}	(9001613-5633637)	2000-2015: (9001613-5633637) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for phosphate rock. Accessed (26 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?nyvpm=1147191251121122113111

Input	Description	Unit	Value range	Value in this study	Data sources
E_1	Phosphoric acid export variable	T phosphoric acid·a ⁻¹	(82660-291817)	2000-2015: (82660-291817) 2016-2050: Model estimation	01411211311 Trademap (2017). EU trade statistics for phosphoric acid. Accessed (23 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?hvpm=11471912809201611211311
E_2	Export variable for iron ores	Tonnes iron ores·a ⁻¹	(1392921-2476034)	2000-2015: (1392921-2476034) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for iron ores and concentrates. Accessed (05 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?hvpm=1147191211311
E_3	Export variable for fertilizers	Tonnes P ₂ O ₅ ·a ⁻¹	(1241785-2043913)	2000-2015: (58070-12627) 2016-2050: Model estimation	Food and Agriculture Organization of the United Nations (FAO). (2017) fertilizers by nutrient. Accessed (10 January 2018). Retrieved from http://www.fao.org/food/agriculture-organization-of-the-united-nations/faostat/en/#data/RFN
E_4	Export variable for feed	Tonnes feed·a ⁻¹	(48292445-102619157)	2000-2015: (48292445-102619157) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for food additives. Accessed (23 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?hvpm=11471912835391611211311
E_5	Export variable for crops	Tonnes crops·a ⁻¹	(15679095-23643751)	2000-2015: (15679095-23643751) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for fruits. Accessed (16 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?hvpm=1147191051211211311
E_6	Export variable for food additives	Tonnes polyphosphates·a ⁻¹	(113350-159050)	2000-2015: (113350-159050)	Trademap (2017). EU trade statistics for cereals. Accessed (16 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?hvpm=1147191101211211311 Trademap (2017). EU trade statistics for vegetables. Accessed (16 January 2018). Retrieved from https://trademap.org/Country_SelProduct_TS.aspx?hvpm=1147191071211211311

Input	Description	Unit	Value range	Value in this study	Data sources
				2016-2050: Model estimation	https://trademap.org/Country_Sel/Product_TS.aspx?hvpm=114719128353916112113111
E_7	Export variable for detergents	Tonnes STPP·a ⁻¹	(82372-283966)	2000-2015: (82372-283966) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for Sodium triphosphates (STPP). Accessed (20 January 2018), Retrieved from https://trademap.org/Country_Sel/Product_TS.aspx?hvpm=114719128353116112113111
E_8	Export variable for fish	Tonnes fish·a ⁻¹	(3071709-4068776)	2000-2015: (3071709-4068776) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for Fish products. Accessed (20 January 2018), Retrieved from https://trademap.org/Country_Sel/Product_TS.aspx?hvpm=114719103121112113111
E_9	Export variable for livestock	Tonnes Livestock·a ⁻¹	(14806405-15051620)	2000-2015: (14806405-15051620) 2016-2050: Model estimation	Trademap (2017). EU trade statistics for animal products. Accessed (10 January 2018), Retrieved from https://trademap.org/Country_Sel/Product_TS.aspx?hvpm=114719101121112113111
C_0	Feed consumption coefficient	Dimensionless	1	1	[42]
C_1	Detergent consumption coefficient	Dimensionless	1	1	[42]
C_2	Demand of vegetable food coefficient per one person	kg P·capita·a ⁻¹	0.3	0.3	[31]
C_3	Demand of animal food coefficient per one person	kg P·capita·a ⁻¹	0.6	0.6	[31]
C_4	Food additives consumption coefficient	Dimensionless	1	1	[42]
W_0	Waste flow variable from animals	Tonnes P·a ⁻¹	(58070-12627)	2000-2015: (58070-12627) 2016-2050: Model estimation	Eurostat (2017) generation of waste by waste category, hazardousness and NACE Rev. 2 activity. Accessed (08 February 2018), Retrieved from http://appsso.eurostat.ec.europa.eu/nui/submitViewTableAction.do
W_1	Waste flow coefficient from human bodies	Dimensionless	1	1	[42]

Input	Description	Unit	Value range	Value in this study	Data sources
W_2	Non-treated solid waste flow coefficient	Dimensionless	0.03	0.03	Eurostat (2017) generation of waste by waste category, hazardousness and NACE Rev. 2 activity. Accessed (08 February 2018). Retrieved from http://appsso.eurostat.ec.europa.eu/mui/submitViewTableAction.do
W_3	Solid waste treating coefficient	Dimensionless	0.97	0.97	Eurostat (2017). Treatment of waste by waste category. Accessed (10 February 2018). Retrieved from http://appsso.eurostat.ec.europa.eu/mui/submitViewTableAction.do
W_4	Solid waste landfilling coefficient	Dimensionless	0.56	0.56	Eurostat (2017) generation of waste by waste category, hazardousness and NACE Rev. 2 activity. Accessed (08 February 2018). Retrieved from http://appsso.eurostat.ec.europa.eu/mui/submitViewTableAction.do
W_5	Wastewater collecting coefficient	Dimensionless	0.93	0.93	OECD (2017), waste water treatment. Accessed (03 February 2018) Retrieved from https://data.oecd.org/water/waste-water-treatment.htm
W_6	Wastewater treating coefficient	Dimensionless	0.79	0.79	OECD (2017), waste water treatment. Accessed (03 February 2018) Retrieved from https://data.oecd.org/water/waste-water-treatment.htm
W_7	Food waste generated per capita	kg-capita ⁻¹	173	173	[74]
W_8	Sludge use for other uses coefficient	Dimensionless	0.22	0.22	Eurostat (2017), Sewage sludge production and disposal from urban wastewater (in dry substance). Accessed (04 January 2018) Retrieved from http://ec.europa.eu/eurostat/tgm/table.do?tab=table&plugin=1&code=ten00030&language=en
$R_{0(1)}$	Manure amount from cattle	T-head-a ⁻¹	(8772652-95859633)	2000-2015: (8772652-95859633) 2016-2050: Model estimations	Faostat, manure applied to soils (2018) Accessed (01 February 2018) Retrieved from http://www.fao.org/docrep/008/a0013e/a0013e08.htm
$R_{0(2)}$	Manure amount from sheep	T-head-a ⁻¹	(109659619-127235994)	2000-2015: (109659619-127235994) 2016-2050: Model estimations	Faostat, manure applied to soils (2018) Accessed (01 February 2018) Retrieved from http://www.fao.org/docrep/008/a0013e/a0013e08.htm
$R_{0(3)}$	Manure amount from poultry	T-head-a ⁻¹	(747348400-889690008)	2000-2015: (747348400-889690008) 2016-2050: Model estimations	Faostat, manure applied to soils (2018) Accessed (01 February 2018) Retrieved from

Input	Description	Unit	Value range	Value in this study	Data sources
$R_{0(4)}$	Manure amount from horse	T·head·a ⁻¹	(3451176-4113186)	2000-2015: (3451176-4113186) 2016-2050: Model estimations	http://www.fao.org/docrep/008/a0013e/a0013e08.htm Faostat, manure applied to soils (2018) Accessed (01 February 2018) Retrieved from http://www.fao.org/docrep/008/a0013e/a0013e08.htm
$R_{0(5)}$	Manure amount from swine	T·head·a ⁻¹	(146858481-162668840)	2000-2015: (146858481-162668840) 2016-2050: Model estimations	Faostat, manure applied to soils (2018) Accessed (01 February 2018) Retrieved from http://www.fao.org/docrep/008/a0013e/a0013e08.htm
R_1	Recycling coefficient for wastewater	Dimensionless	0.53	0.53	Eurostat (2017), Sewage sludge production and disposal from urban wastewater (in dry substance). Accessed (04 January 2018). Retrieved from http://ec.europa.eu/eurostat/tgm/refreshTableAction.do?tab=table&plugin=1&pcode=te000308&language=en
R_2	Recycling coefficient for solid waste	Dimensionless	0.44	0.44	Eurostat (2017), Treatment of waste by waste category. Accessed (10 February 2018). Retrieved from http://appsso.eurostat.ec.europa.eu/multi/submitViewTableAction.do
L_0	Beneficiating loss coefficient	Dimensionless	0.16	0.16	[61]
L_1	Loss coefficient during processing to phosphoric acid	Dimensionless	0.13	0.13	[61]
L_2	Loss coefficient from fertilizers due to runoff of P	kg P·ha ⁻¹	Avg: 2.515	2.515	[S2]
L_3	Food loss coefficient	Dimensionless	0.103	0.103	FAO, Save food: global initiative on food loss and waste reduction (2018) Accessed (03 February 2018). Retrieved from http://www.fao.org/save-food/resources/keyfindings/en/
L_4	Animal food loss coefficient	Dimensionless	0.13	0.13	FAO, Save food: global initiative on food loss and waste reduction (2018) Accessed (03 February 2018). Retrieved from http://www.fao.org/save-food/resources/keyfindings/en/

Input	Description	Unit	Value range	Value in this study	Data sources
L_5	Crops loss coefficient	Dimensionless	0.23	0.23	http://www.fao.org/save-food/resources/keyfindings/en/ FAO. Save food: global initiative on food loss and waste reduction (2018) Accessed (03 February 2018). Retrieved from http://www.fao.org/save-food/resources/keyfindings/en/
L_6	Fish loss coefficient	Dimensionless	0.235	0.235	FAO. Save food: global initiative on food loss and waste reduction (2018) Accessed (03 February 2018). Retrieved from http://www.fao.org/save-food/resources/keyfindings/en/
L_7	Uncollected wastewater coefficient	Dimensionless	0.07	0.07	OECD (2017), waste water treatment. Accessed (03 February 2018). Retrieved from https://data.oecd.org/water/waste-water-treatment.htm
L_8	Non treated wastewater coefficient	Dimensionless	0.21	0.21	OECD (2017), waste water treatment. Accessed (03 February 2018). Retrieved from https://data.oecd.org/water/waste-water-treatment.htm
L_9	P loss coefficient from mining rate	Dimensionless	0.18	0.18	[61]
A	Arable land % of total land in EU	%	avg: 26	26	The world bank (2017), Arable land % of land area in EU (sq. km) in EU. Accessed (04 February 2018). Retrieved from https://data.worldbank.org/indicator/AG.LND.ARBL.ZS?end=2015&locations=EU&start=2001
T	Total land in EU	Sq km	(4238886-4241383)	2000-2015: (4238886-4241383) 2016-2050: Model estimation	The world bank (2017), Land area (sq. km) in EU. Accessed (04 February 2018). Retrieved from https://data.worldbank.org/indicator/AG.LND.TOTL.K2?end=2016&locations=EU&start=2001
$P(t)$	EU population	capita · a ⁻¹	(491841900-509564400)	2000-2050: (491841900-509564400)	United Nations (2017), Population division, world population prospects – Total population, Both sexes. Accessed (07 February 2018). Retrieved from https://esa.un.org/unpd/wpp/DVD/Files/J_Indicators%20(Standard)/EXCEL_FILES/1_Population/WPP2017_POP_F01_T1_TOTAL_POPULATIION_BOTH_SEXES.XLSX

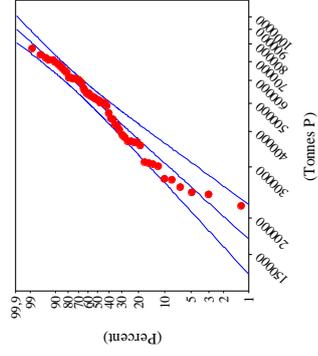
6 **Table S3**

7 **Distribution fitting and data details**

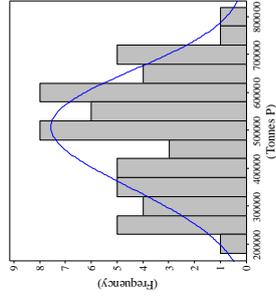
Variables distribution summary

Feed production rate
 Distribution: Weibull
 Shape: 3.892
 Scale: 549083×10^5
 Threshold: 1.113
 Number of data points = 56
 AD: 0.472
 Corresponding p-value = 0.24
 Sample mean = 495517
 Sample St Dev = 147657
 Minimum = 220195
 Maximum = 777078

Probability Plot of Feed production
 Weibull - 95% CI



Histogram of Feed production
 Weibull

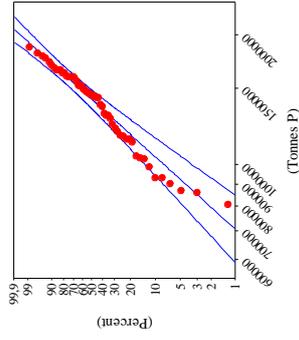


Crop production rate

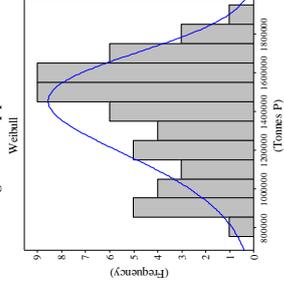
Distribution: Weibull
Shape: 6.117
Scale: 149531×10^6
Number of data points = 56
AD: 0.603
Corresponding p-value = 0.117
Sample mean = 1384791
Sample St Dev = 277512
Minimum = 806550
Maximum = 1867480

Probability Plot of Crop production

Weibull - 95% CI



Histogram of Crop production

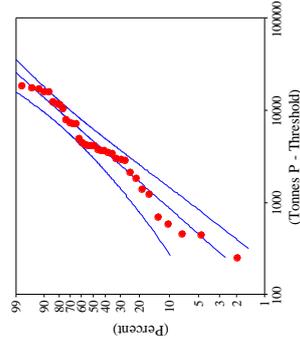


Animal production rate

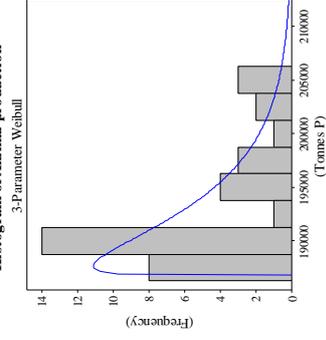
Distribution: 3-Parameter Weibull
Shape: 1.117
Scale: 6430.896
Number of data points = 36
AD: 0.485
Corresponding p-value = 0.236
Sample mean = 192954
Sample St Dev = 5440.52
Minimum = 187030
Maximum = 210562

Probability Plot of Animal production

3-Parameter Weibull - 95% CI



Histogram of Animal production



- 10 [S1] Y. Jin, T. Jiang, Y. Yang, Q. Li, G. Li, Y. Guo, Removal of phosphorus from iron ores by chemical leaching, *J. Cent. South Univ. Technol.* 13 (2006) 673–
11 677.
- 12 [S2] S. Fortune, J. Lu, T.M. Addiscott, P.C. Brookes, Assessment of phosphorus leaching losses from arable land, *Plant Soil*. 269 (2005) 99–108.

13

Publication II

Rahimpour Golroudbary, S., El Wali, M., and Kraslawski, A.
**Environmental sustainability of phosphorus recycling from wastewater, manure and
solid wastes**

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Environmental sustainability of phosphorus recycling from wastewater, manure and solid wastes

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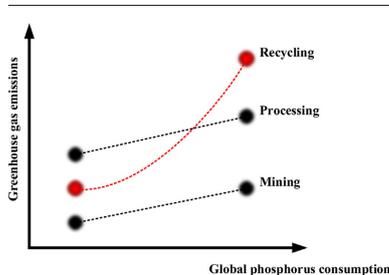
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HIGHLIGHTS

- Dynamic model for analyzing energy consumption and GHG emissions of phosphorus flow
- Assessment of possible amount of phosphorus to be recovered from wastes
- Assessment of environmental sustainability of phosphorous recycling (2000–2050)
- Impact of current and future phosphorus consumption on the amount of GHG emissions

GRAPHICAL ABSTRACT



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ABSTRACT

Phosphorus (P) is an important critical material essential for crops cultivation and animal husbandry. Effective phosphorous recycling is considered one of the most significant factors in alleviating its criticality. However, despite the importance of phosphorous recycling, its sustainability is not studied extensively. This paper aims to answer the question if recycling of phosphorus is an environmentally sustainable option. To address this problem, two issues are analyzed in this paper: energy consumption and greenhouse gas (GHG) emissions in phosphorous recycling. The analysis was performed by simulating mass and energy flows in the global phosphorus supply chain (from mining to recycling) in order to understand and analyze its environmental impact in 2000–2050. The results of simulation show that around 82% of recycled phosphorous originates from manure. Moreover, the calculations indicate that about 70% of total GHG emissions from phosphorous recycling is caused by wastewater processing. In addition, the results show that phosphorous obtained from recycled wastewater constitutes only 2% of the whole amount recovered in the recycling process. Therefore, the obtained results show a clear need for a detailed analysis of the sustainability of phosphorous recycling processes. Moreover, the analysis of scenarios of phosphorus consumption indicates that GHG emissions increase slowly in the mining phase and grow exponentially in the recycling stage. The main finding of this paper contradicts the general opinion about environmental friendliness of recycling. It shows that phosphorus recycling is not a sustainable solution in a longer perspective.

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1. Introduction

Supply of phosphorus (P) and its current consumption pattern is not sustainable in a long term (Childers et al., 2011; Daneshgar et al., 2018b). The major areas of concern associated with sustainability of phosphorus were its scarcity (George et al., 2016), need for recycling (Morse et al., 1998), environmental pollution (Daneshgar et al., 2018a) and call for new sustainable policies and strategic framework (Cordell et al., 2011). Therefore, recycling or recovery of phosphorus from waste streams has been proposed as a possible approach to handle the issue of its sustainable use (EU Commission, 2017; Withers et al., 2015b, 2015a).

The initial concept of criticality originates from the concerns about the availability of raw materials. The general understanding of the criticality of resources is based on their scarcity and high demand (Calvo et al., 2017; Frenzel et al., 2017). It is commonly acknowledged that the criticality of materials is determined by several aspects such as supply risk, economic importance and vulnerability to supply restriction (El Wali et al., 2018; Rahimpour Golroudbary et al., 2019). However, we lack a generally accepted definition of critical materials due to the differences in methodologies applied for criticality assessment (Frenzel et al., 2017; Jin et al., 2016).

Phosphorus (P) has been identified as an important critical material according to many assessments (EU Commission, 2017; U.S. Geological Survey, 2018; Scholz and Wellmer, 2013). Phosphorus is a chemical element which is primarily obtained from the phosphate rock. Sustainable supply of this element is essential for plants and animals, in particular for the security of food supply (Diallo et al., 2015). For over half a century, phosphorus has been one of the non-substitutable resources in food production (Jacobs et al., 2017) and about 50% of food production is based on the use of mineral phosphate rock (Scholz and Wellmer, 2013, 2015a).

The amount of mined phosphate rock ore depends on several factors, e.g. technological developments, exploration efforts, demand, price level (Scholz and Wellmer, 2013). A limited number of countries - Morocco and Western Sahara, China, Algeria, Syria, Russia, and South Africa - control 88% of the world phosphate rock reserves (Chen and Graedel, 2016). Globally, more than 90% of the anthropogenic input to the phosphorus life cycle is used for agro-food production including animal feed. The rest is applied as food and feed additives as well as other industrial phosphates (Scholz and Wellmer, 2015a). There is no danger of imminent shortages of phosphorus despite its limited resources (Scholz and Wellmer, 2015b). However, due to the already mentioned significant role of phosphorus in the global food chain, its global scarcity may have huge impact on the future food security (Cordell and Neset, 2014). In 2015, the global production of phosphate rock increased due to strong growth in global phosphorus consumption (Scholz and Wellmer, 2015a) and most of it was reported in the Middle East and South America (U.S. Geological Survey, 2018).

Phosphorus is not only a case of particular concern in terms of resource management, but its mining could also be potentially disturbing to the environment (Rowe et al., 2016; Scholz and Wellmer, 2016). The policy measures adopted within the framework of the circular economy (increased recycling rates and waste reduction of critical raw materials) should mitigate not only future potential supply risks of these materials, but also the environmental impact associated with their life cycle (Elia et al., 2017; EU Commission, 2015). From this perspective, there is a global trend towards improved recovery of phosphorus. It has been demonstrated that a global co-operation for recycling and reuse of phosphorus in waste streams is urgently required (Dawson and Hilton, 2011; Elser and Bennett, 2011). Therefore, recycling of phosphorus needs to be considered as an integral part of phosphorus management policies. Otherwise, considerable fraction of phosphorus existing in the waste streams will be permanently lost.

On the other hand, environmental performance at each stage, e.g. mining, processing and production or recycling is one of the important

criteria in the assessment of overall sustainability of phosphorus supply chain. The impact of energy consumption on the sustainability of a supply chain is well-known (Azadeh and Arani, 2016). It is manifested by the depletion of non-renewable energy resources and greenhouse gas (GHG) emissions. It is worth to mention that the rapid growth of agricultural production has major impact on development of mining and recycling of phosphorus (Wu et al., 2017). The scale of the phosphorus supply chain motivates the attempts to assess its environmental impact. One of its elements is the analysis of phosphorus supply chain aimed at quantitative assessment of energy consumption and GHG emissions.

In recent years, several studies have offered a quantitative analysis of phosphorus flow. Some of them introduced structural models at various scales, e.g. regional (Chowdhury et al., 2016; Theobald et al., 2016), national (Cooper and Carliell-Marquet, 2013; Li et al., 2015), continental (Jedelhauser and Binder, 2015; Matsubae and Webeck, 2019; Ott and Rechberger, 2012), and global (Chen and Graedel, 2016; Y. Liu et al., 2008; Van Vuuren et al., 2010). In this paper, we introduce a dynamic model to analyze environmental impact of phosphorus supply chain at global scale. The main objective of this study is to answer the question whether recycling of phosphorus is an environmentally sustainable option viable in a longer perspective.

2. Model of phosphorus global supply chain

The need for a systematic analysis of phosphorus supply chain has been presented in different studies, e.g. Chen and Graedel (2016); Kleinman et al. (2015); Cordell (2013); Neset et al. (2016); and Van Vuuren et al. (2010). This paper aims at determining energy consumption and GHG emission at different stages of phosphorus supply chain using the system dynamics modeling (Forrester, 1997). The analysis presented in this paper covers a 50-year time horizon. The reason for this time interval analysis is a need to explore future global environmental impact of phosphorus mining, processing and recycling. Also, we consider significant challenges of phosphorus supply in the near future. For example, it is predicted that we will face 50–100% increase of phosphorus demand in 2050 (Cordell et al., 2009; EFMA, 2000; Steen, 1998). It will be triggered by the growth of global demand for food (up to 70% by 2050) and a changing diet (e.g. growing interest in meat and dairy-rich diet, which requires phosphorus intensive food production) (Fraiture, 2007). In the case of phosphorus, several approaches have been applied to analyze dynamic interactions among various components of the system under investigation, e.g. phosphorus mass flow (Modin-Edman et al., 2007), solid waste management (Kollikkathara et al., 2010), decoupled aquaponics system (Goddek et al., 2016), phosphorus flows in food and waste chains (Treadwell et al., 2018), and the impact of recycling improvement on phosphorus life cycle (El Wali et al., 2018).

Fig. 1 gives an overview of the proposed model. The system consists of three sub-systems: material flow, energy consumption and GHG emission. The material flow sub-system is composed of the following modules: mining, beneficiation, processing, production, consumption, waste generation, and recycling. The energy consumption sub-system includes energy consumed in mining, processing and production as well as recycling. The GHG emission sub-system is primarily related to the energy consumption and therefore the structure of both subsystems is identical. The details of the structure of a dynamic model are given in Appendix (Figs. A1 and A2) as well as notation and the used data are presented in Appendix (Tables A1 and A2). Fig. A1 (a) represents the mass flow across the different stages of phosphorus supply chain starting from mining and ending at the post-consumption stages. Fig. A1 (b) shows mass flow of phosphorus across the waste streams where material goes to recycling, loss, landfill or other applications. Fig. A2 shows the relationship between energy consumption and GHG emissions in mining stage of phosphorus supply chain, as an example. The sources of data, used as an input into the calculations of material flow, energy consumption and GHG emissions, are presented in Fig. 2.

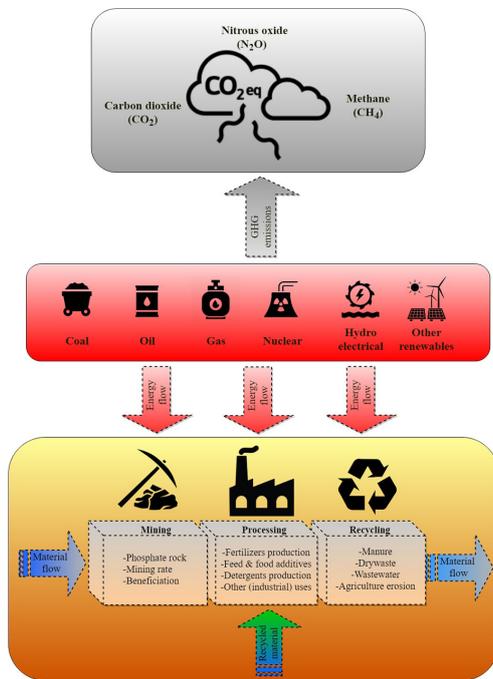


Fig. 1. Conceptual model of phosphorus global supply chain.

2.1. Material flow of phosphorus model

We can distinguish three main steps in the phosphorus supply chain. The first step is the industrial stage, where phosphate ores are mined, and next processed applying the beneficiation process (Chen and Graedel, 2016; Koppelaar and Weikard, 2013; Scholz and Wellmer, 2015a). The second step corresponds to the production stage, where phosphorus is present in various streams, e.g. agricultural fertilizers, feed and food additives, laundry detergents and other industrial applications (Belboom et al., 2015; Mottet et al., 2017; Noya et al., 2017; Van Hoof et al., 2017). The final step corresponds to the dissipation and the recycling of phosphorus (Chen and Graedel, 2016; Fowdar et al., 2017; Harris et al., 2017; Hobbs et al., 2017; Leon and Kohyama, 2017; Maguire and Fulweiler, 2017; Ortiz-Reyes and Anex, 2018). In recycling, other materials such as nitrite associated with phosphorus can be recovered in parallel. However, the analysis of their flows is beyond the scope of this model.

Fertilizer industry is the major user of phosphoric acid (Belboom et al., 2015). Fertilization of soil, aiming at the increase of crop yield, is done thanks to phosphorus originating from fertilizers and recycling streams. In this work, the production of the following crops was considered: rice, wheat, barley, sorghum, millet, rye, oats, potatoes, sweet potatoes, cassava, soybeans, beans, peas, rapeseed, olive, sugar beet, sugar cane, seed cotton, vegetable, and fruits. The produced crops are used as feed and food (Mottet et al., 2017; Noya et al., 2017). The feed is consumed by the livestock. In the presented model, we considered the feeding of the following animals: sheep, goats, horses, cattle, buffaloes, mules, pigs, ducks, chickens, geese, and turkeys.

There are losses of phosphorus along its supply chain. Major amounts of phosphorus are released into the marine systems. It takes place in mining, beneficiation and processing of phosphorus (Chen and Graedel, 2016). In addition, a significant amount of phosphorus enters water resources through soil in the process of agricultural production. A substantial amount of phosphorus is removed from the soil through crops harvesting. The rest of phosphorus remains in the soil as a surplus and is retained in it. The loss of phosphorus occurs due to various phenomena; e.g. soil loss, leaching, runoff and erosion (Fowdar et al., 2017; Harris et al., 2017; Ortiz-Reyes and Anex, 2018). Another source of phosphorus in the marine system is the effluents containing detergents, industrial wastewater and animal production waste (Chen and Graedel, 2016; Hobbs et al., 2017; Maguire and Fulweiler, 2017).

Moreover, the human factor has twofold impact on the phosphorus loss to inland waters. First, the increasing population accelerates the leakage of phosphorus through land-water interface towards the coastal zones (Withers et al., 2014). The second factor is urbanization (Matsubae and Webeck, 2019).

Currently, recycling is considered as one of the best solutions to manage phosphorus management challenges. Recycling is the process of turning waste into usable materials. The recycling of phosphorus takes place mainly in agriculture. It is based on the use of various sources containing P, e.g. sludge from wastewater treatment (Shiu et al., 2017; Ye et al., 2017), livestock manure (Haase et al., 2017) and dry waste (Pearce and Chertow, 2017). Therefore, in this study, the main waste streams considered in the phosphorus supply chain are wastewater, manure and solid waste and, as a result, the recovery of phosphorus from those three streams is investigated.

The significance of phosphorus recovery from wastewater has been presented in many studies (Kumar and Pal, 2015; Musfique et al., 2015). They showed that wastewater treatment provides a good opportunity for phosphorus recovery. Recovering phosphorus from wastewater could considerably reduce eutrophication and create a supplementary source of fertilizers (Ye et al., 2017). The second main stream at the recycling stage corresponds to manure. According to the recent research, in Europe, manure contains around 2000 Kt/a of phosphorus, that is much higher than the amount of phosphorus in sewage and slaughter waste (Buckwell and Nadeu, 2016; Leip et al., 2014). As recycled manure goes directly to fields, most of phosphorus stays within agricultural applications. Manure considered in this study comes from donkeys, cattle, buffaloes, chickens, ducks, mules, sheep, goats, and pigs. In the third main recovery stream of phosphorus, the recycling of solid waste takes place in the composting process. An efficient management of solid waste is needed to limit and possibly decrease the environmental burden. Collected solid waste is immediately treated and next either recycled or used in other applications, e.g. landfilling (Behrooznia et al., 2018).

2.2. Energy consumption according to phosphorus model

During the mining of phosphate ores, energy consumption is mainly associated with the use of mining machines and equipment. That equipment includes draglines, pumps, pit cars, and equipment necessary for water treatment, pumping, beneficiation and transport of raw material to phosphate processing sites (Schroder et al., 2010).

In the production stage, the production of feed and fertilizers are analyzed separately. Energy demand in feed production varies between its different types. In this paper, the analyzed types of feed are hays, barley, maize (gluten meal, grains and silages), oats, salts and minerals, soybeans, and wheat. This study considers the direct energy inputs for feed production, which includes the energy used in feed processing and by the delivery machinery (Frorip et al., 2012). Energy demand for phosphate fertilizers production corresponds to the energy used by the slurry method (X. Zhang et al., 2017). The data for energy demand correspond to the production of two phosphate fertilizers (mono ammonium phosphates and di-ammonium phosphates). Those two types of fertilizers are the most commonly produced phosphate fertilizers in China.

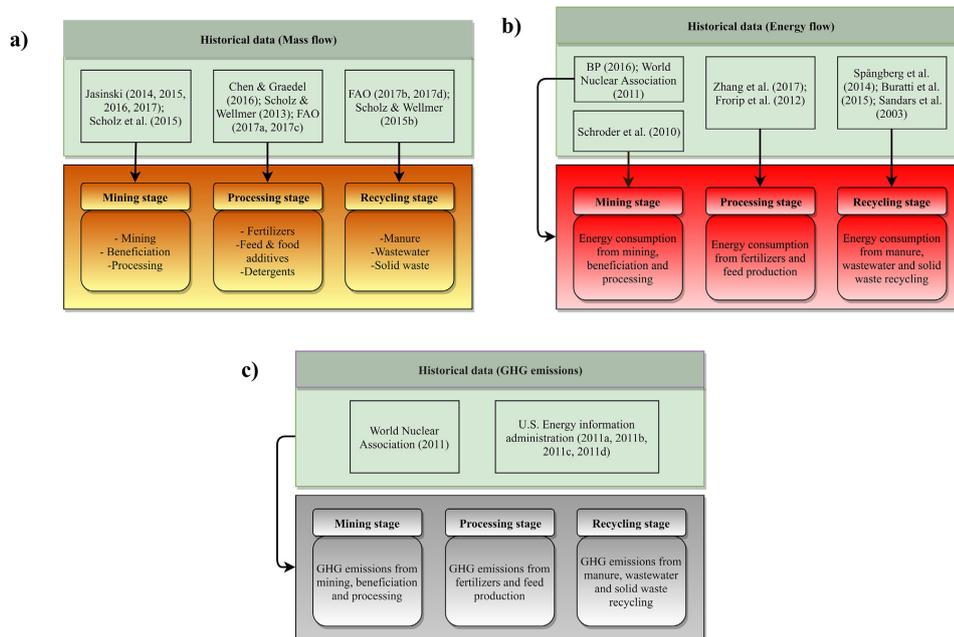


Fig. 2. Data sources of three stages of the phosphorus supply chain. a) Material flow; b) energy consumption; c) GHG emission.

In the recycling stage, our study is limited to the analysis of energy demand related to the recycling of phosphorus from waste streams coming from wastewater, food, animal, human excreta and households. In this work, the data presented by Sandars et al. (2003) are used as a basis for determining the energy requirements of manure recycling. The following sources of manure for recycling are analyzed here: animal origin, households, anaerobic digesters, storage lagoons, and land application of other residues.

The wet chemical approach and the thermo-chemical treatment are the two main technologies for P recovery from sludge produced from wastewater treatment plants (Appels et al., 2010). In wet chemical process, strong acids are added to extract P from the sludge phases. Thermo-chemical treatment refers to the addition of chloride additives to remove heavy metals from the sludge, thus facilitating the chemical removal of P (Ye et al., 2017). It is worth to mention that phosphorus recovery from wastewater is difficult due to different sludge compositions (Amann et al., 2018). The data given by Buratti et al. (2015) were used for determination of energy consumption in recycling of phosphorus from solid waste. There are mainly two treatment technologies of waste for P recovery, undifferentiated and source separated collections. In both cases, the treatment of waste and recovery of phosphorus is performed using aerobic biological facility. The undifferentiated collection is meant primarily for the landfilling of waste. In this paper, we consider the second option, source separated collection, in which treated waste is composted as fertilizers.

2.3. Greenhouse gas emissions according to phosphorus model

Carbon dioxide (CO₂), nitrous oxide (N₂O), and methane (CH₄) are the major greenhouse gases (GHG) (Brander, 2012). The emissions of these gases occur in different processes either directly (e.g. during agricultural activities) or indirectly (e.g. during the process of mining,

production and recycling) (Massé et al., 2011; Wood and Cowie, 2004). In this model, as mentioned above, the structure of GHG emissions sub-system is identical to the system of energy flow. Therefore, we analyzed the GHG emissions related to energy consumption in mining, production (e.g., fertilizer and feed), and recycling (e.g., manure, wastewater and dry waste).

The required data of the sub-model of GHG emissions originate from specialized reports (World Nuclear Association, 2011; US Energy Information Administration (2011a), Fig. 2 (b–c)). These data sources are used to estimate the share of each gaseous emission (CO₂, CH₄ or N₂O) and its sources (coal, nuclear, gas, oil, renewables).

2.4. Mathematical formulations

Dynamic modeling was used to simulate the phosphorus supply chain. The model was built using the concept presented in Fig. 1. Below, we present the main formulas used in calculating material, energy, and GHG flows. All equations derived from the main formulas and the details of the model are given in Appendix, Tables A1–A3.

There are two types of equations in the model, which represent the flow of mass, energy and GHG: stock equations (state equations) and flow equations (rate equations).

The stocks assumed in the material flows of the model are: mined and beneficiated phosphorus, phosphoric acid, detergents, fertilizers, feedstock of vegetable and animal food, crops, food, livestock, food waste, animal waste, dry waste, wastewater stock, and organic fertilizers. The mass stock equations are given after Forrester (1997) as:

$$Stock(t) = \int_{t_0}^t [Inflow(t) - Outflow(t)] dt + Stock(t_0) \quad (1)$$

where t_0 is the initial and t is the final year considered; $Stock(t)$ is a mass accumulated in the system in the moment t of the period 2000–2050 due to influx $Inflow(t)$ and loss $Outflow(t)$.

$$Inflow(t) = f(Stock(t), V(t), P);$$

$$Outflow(t) = f(Stock(t), V(t), P) \tag{2}$$

where, $V(t)$ is an exogenous variable in time t , e.g. the benefiting rate depends on the amount of extracted phosphorus and the stock of globally mined phosphorus every year. P is a parameter considered in the system, e.g. benefiting coefficient, phosphorus content in P_2O_5 and P_2O_5 content of phosphate rock (All parameters are presented in Appendix, Table A2).

Every stage in the supply chain consumes energy obtainable from the different sources. The rate equation for energy is given as Eq. (3).

$$EC_m(t) = P_m(t) \times \sum_{n=1}^6 \sigma_{m,n} \tag{3}$$

where, $EC_m(t)$ is energy consumption of stage m in the moment t of the period 2000–2050; $m = 1,2,3$ represents stage of the phosphorus supply chain: mining, processing, and recycling; $n = 1,2,\dots,6$ corresponds to the type of energy obtainable from different energy sources: coal, nuclear, gas, oil, hydroelectricity and other renewables; $P_m(t)$ is the amount of phosphorus in stage m in the moment t of the period 2000–2050; $\sigma_{m,n}$ is the energy required per one ton of phosphorus flow in a given stage $m = 1,2,3$ (mining, processing, and recycling) from each energy source $n = 1,2,\dots,6$ (coal, nuclear, gas, oil, hydroelectricity and other renewables).

The GHG emissions are estimated based on energy consumption in the phosphorus supply chain. The rate equation of GHG flows is formulated as:

$$GHG_{x,m}(t) = EC_m(t) \times \sum_{n=1}^6 \delta_{x,n} \tag{4}$$

where, $GHG_{x,m}(t)$ represents greenhouse gas emissions of type $x = 1, 2, 3$ (CO_2, CH_4 or N_2O) in m stage of the supply chain, $m = 1,2,3$ (mining, processing, and recycling) in the moment t of the period 2000–2050; $EC_m(t)$ is the energy consumption in stage $m = 1,2,3$ (mining, processing, and recycling) in the moment t of the period 2000–2050; $\delta_{x,n}$ is the

GHG emitted as type $x = 1, 2, 3$ (CO_2, CH_4 or N_2O) per 1 J of energy consumed using the n resource, $n = 1,2,\dots,6$ (coal, nuclear, gas, oil, hydroelectricity and other renewables).

2.5. Validation of the model

Usually, in order to determine the validity of the model the obtained outputs are compared against experimental data and their statistical compliance is tested. The method proposed by Barlas (1996) was used for the validation of the proposed model. The variables such as fertilizer and feed production as well as manure recycling rates were used to validate the model. Differences between the results obtained from the model and experimental data of the above-mentioned variables on average amounted to 0.04, 1.44 and 1.90%, respectively. The results of validation and calculation of the error of the model are presented in Table 1.

3. Results and discussion

Simulation results are given in Fig. 3 (a–f). Energy consumption and emissions of GHG in mining, processing and recycling stages are illustrated in Fig. 3 (a–b), Fig. 3 (c–d), and Fig. 3 (e–f) respectively.

In the mining stage, energy consumption fluctuates dynamically and CO_2 emission reaches the highest level as shown in Fig. 3 (a–b). The main cause of the observed trend along the supply chain is the dynamics of phosphorus flow from mining to recycling. This phenomenon has been also observed by Chen and Graedel (2016). The trend of energy consumption and consequently GHG emissions in the mining stage is influenced by the continuous change in the market value of phosphate rock. Calculations in the mining stage show the annual mean of GHG emissions is estimated at around 70 million metric tons CO_2 equivalent (mt CO_2e) between 2000 and 2050.

The consumption of mined rock in different sectors has fluctuated significantly for the past 20 years. In the processing stage (Fig. 3 (c–d)) energy consumption also fluctuates continuously. These changes produce different GHG emissions at this stage. In 2013, the level of GHG emissions reached about 197 mt CO_2e as a result of the decrease in energy consumption. This decrease reflects changes in material production in the phosphorus supply chain. Global demand for phosphate fertilizers decreased in 2013 from 41.6 to 40.3 mt P_2O_5 – equivalent to 17.6 mt phosphorus – from which it increased to 41.3 million mt P_2O_5 in the following year 2014. Changes in demand for fertilizers caused a

Table 1
Calculation of the model error.

Year	Historical data			Simulation model results		
	Fertilizer production ^a	Feed production ^b	Manure recycling ^c	Fertilizer production	Feed production	Manure recycling
2000	N/A	1,301,851.60	13,304,913.32	–	1,452,755.5	14,126,384.05
2001	N/A	1,356,200.56	13,386,242.41	–	1,585,635.13	14,321,608.75
2002	16,087,280.14	1,347,503.36	13,540,544.05	20,196,138.11	1,414,358.00	14,111,058.49
2003	17,061,371.41	1,349,972.08	13,745,584.98	18,744,831.81	1,477,137.52	13,884,506.86
2004	17,986,839.42	1,457,628.76	13,985,807.53	18,709,318.15	1,417,280.63	14,114,006.80
2005	18,398,560.64	1,444,219.52	14,223,828.63	19,909,526.19	1,380,451.96	14,381,810.55
2006	18,107,386.42	1,424,900.04	14,434,801.65	22,554,161.72	1,378,671.73	13,980,106.01
2007	19,205,169.53	1,441,938.36	14,647,319.06	20,805,530.76	1,477,424.94	14,241,787.37
2008	20,150,567.45	1,557,968.28	14,774,087.36	20,067,328.42	1,347,941.07	14,403,686.46
2009	18,991,954.18	1,489,393.52	14,850,020.48	20,366,313.65	1,388,951.29	14,263,566.09
2010	21,752,206.61	1,515,894.32	14,884,172.53	18,438,580.86	1,451,976.45	13,976,649.00
2011	23,243,419.59	1,590,210.84	14,888,172.36	21,982,372.77	1,462,088.08	13,870,226.49
2012	22,944,776.89	1,582,593.92	15,038,770.34	20,096,501.86	1,353,806.82	14,172,936.77
2013	22,843,212.38	N/A	15,102,132.7	21,772,061.33	–	14,068,006.76
2014	23,270,725.43	N/A	15,260,869.27	16,295,006.91	–	14,041,772.72
Ave.	20,003,343.85	1,450,790.40	14,404,524.38	19,995,205.58	1,429,883.01	14,130,540.88
			Error	0.04%	1.44%	1.90%

^a Initial data based on FAOSTAT source are given in tons P_2O_5 (FAO, 2017a). Conversion factor to tons phosphorus assumes 0.436 (Scholz et al., 2014).
^b Data based on FAOSTAT data source for the production of crops used as feed commodities (FAO, 2017b). P content in crops is taken from Chen and Graedel (2016).
^c Data based on FAOSTAT source showing the amount of manure applied to soil (FAO, 2017c). P content in manure is based on FAO report on the environmental impact of manure (FAO, 2017d).

decrease in the processing stage of phosphorus supply chain in 2013, which significantly affected energy consumption and GHG emissions. The maximum amount of GHG emissions was estimated at up to 237 mt CO₂e in 2034. In another study by Cordell et al. (2009), the global peak of phosphorus production was estimated around 2030. The model shows an increasing trend in GHG emissions from 2014 to 2015. This reflects the increase in mining production in 2015. According to Scholz and Wellmer (2015a), the increase in the global mining production in 2015 was due to the growth in global consumption of phosphorus.

Calculation shows that the annual mean of GHG emissions from the processing stage is ca. 220 mt CO₂e in the given period (2000–2050).

Considering the recycling stage (Fig. 3 (e–f)), Fig. 3 (e) shows the impact of the reduction of phosphorus production on the supply of phosphorus to the recycling stage. For example, the collapse of the Soviet Union (FSU) between 1989 and 1993, resulted in dramatic decrease of fertilizer demand in processing stage. Moreover, demand for phosphate fertilizers decreased in Western Europe and North America in 2000 (Cooper et al., 2011; Cordell et al., 2009) as well. The main reason was

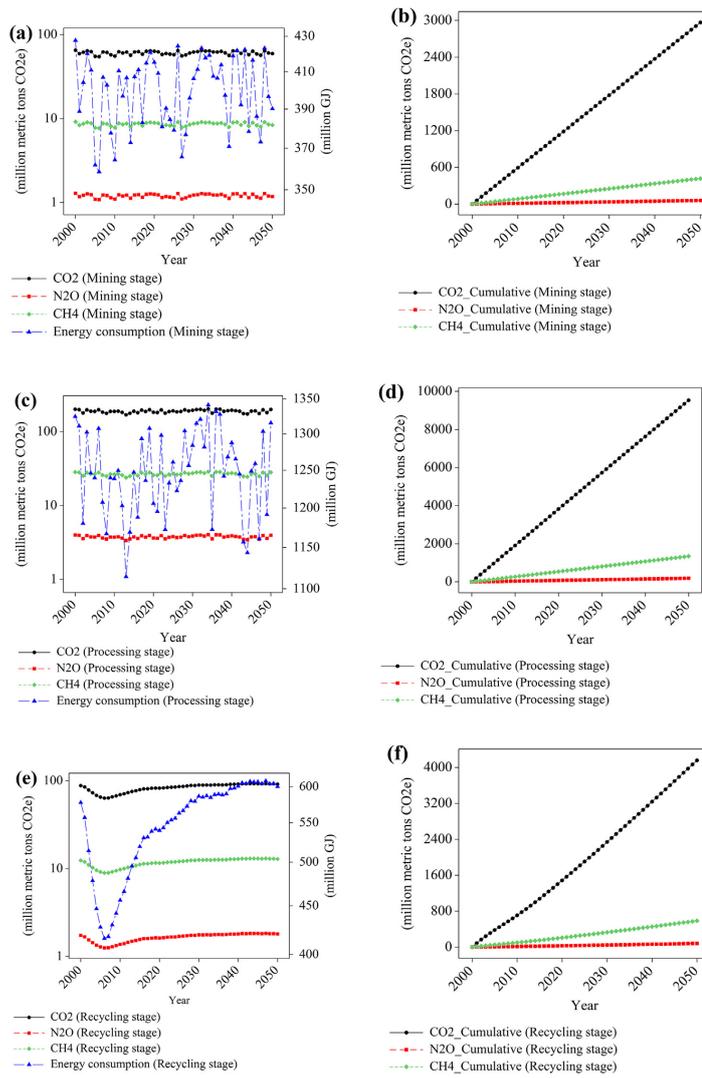


Fig. 3. Greenhouse gas emissions and energy consumption levels in the life cycle stages of phosphorus (2000–2050). (a): Mining stage (annual). (b): Mining stage (cumulative). (c): Processing stage (annual). (d): Processing stage (cumulative). (e): Recycling stage (annual). (f): Recycling stage (cumulative).

the increased awareness of soil saturation (i.e. after the decades of over-use, there was the sufficient phosphorous stock in the soil so that the application rates could be reduced). Furthermore, since 2000, the awareness of eutrophication has also reduced phosphate demand in the developed world aimed at the reduction of leakage to waterways (Scholz et al., 2014). These aspects are the main causes why phosphorus flow decreased at the beginning of the simulation period covered by this study. Restrictions on the use of phosphates in detergents affect wastewater treatment efficiency. However, the rising demand for phosphorus results in an upward energy consumption and, consequently, in the increased GHG emissions. The GHG emissions will reach a maximum of approximately 107 mt CO₂e in 2047. Calculation shows that the annual mean of GHG emissions from the recycling stage is around 97 mt CO₂e between 2000 and 2050. This result implies the impact of benefits resulting from systemic approach to the recycling stage, which shed light on the needs of other policies aimed at controlling phosphorus cycle to prevent environmental problems.

Results presented in Fig. 4 show that, assuming we use the current technology, the processing stage will be the major energy consumer and the biggest GHG emitter in the years 2000–2050. Interestingly, over the same period of time, phosphorus recycling is characterized by higher energy consumption and GHG emissions than mining. Therefore, mining or substitution of phosphorus still have to be considered as important alternatives to recycling if environmental sustainability is to be achieved in the years 2000–2050.

In the recycling stage, phosphorus recovery from two main waste streams including wastewater and manure is analyzed in detail. The processes of phosphorus recovery from wastewater assumed anaerobic digestion. In manure recycling, main processes are solid-liquid separation and anaerobic digestion as well as drying and small-scale incineration (Nättorp et al., 2019). Fig. 5 presents the relation between the amount of phosphorus recycled and CO₂e emitted when recycling phosphorus from both sources. Manure recycling provides around 82% of total recycled phosphorus from various sources while wastewater recycling provides only 2% (Fig. 5). At the same time, the amount of GHG emitted in wastewater recycling is about 70% of total CO₂e emissions in the recycling stage. The presented result shows in expediency of recycling phosphorus from wastewater if only the amount of the recycled phosphorus and GHG emissions would be taken into account. However, it is obvious that the removal of phosphorus from wastewater must be done because of a different reason. Excessive presence of phosphorus in water causes eutrophication, and eventually leads to the collapse of ecosystems (Xu et al., 2017; Liu et al., 2012; Chen and Graedel, 2016; Bradford-Hartke et al., 2015).

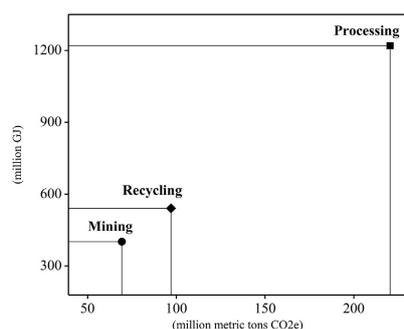


Fig. 4. Annual mean of greenhouse gas emissions and energy consumption in phosphorus life cycle in years 2000–2050.

4. Scenario of phosphorus consumption

Dynamic models are often used to study the behavior of a system when demand, supply, and other conditions are changing. The studies of scenarios developed for various situations are especially useful in this case. In the paper (El Wali et al., 2018), we analyzed several options for the improvement of phosphorus recycling. In this study, the proposed model is used to evaluate different scenarios of phosphorus consumption and its global environmental impact in the years 2000–2050. The objective is to explore the impact of future global phosphorus consumption on GHG emissions. Knowledge about this impact is essential for making decisions aimed at reducing GHG emissions.

Changes in phosphorus supply and demand resulting from various factors have been studied extensively (e.g., livestock production (Bouwman et al., 2013) together with demand for phosphate fertilizers, food supply chain (Cordell et al., 2009), and the effect of increasing world population (Shu et al., 2006)). One of the main challenges in phosphorus supply chain management results from increased production of food and fertilizers, e.g. the overall food production is estimated to increase by around 70% until 2050 (Fraiture, 2007). The problem consists in limited resources of phosphate rock as well as non-substitutive character of phosphorus (Ye et al., 2017; Edgerton, 2009). Therefore, phosphorus recovery is very important as a solution that may help manage its shortages in the future. On the other hand, an increase in the production of phosphorus requires more energy and generates more waste and GHG emission in all stages of its supply chain. Given the main objective of this paper, we analyze various scenarios of phosphorus consumption in order to assess its impact on GHG emissions at each stage of the supply chain until 2050. When building the scenarios we assumed that all parameters remain constant except the amount of phosphorus in production and recycling processes.

In this work, four situations have been studied: scenario A - current level of phosphorus consumption as well as B, C and D scenarios representing the cases of 10%, 30% and 40% increase in phosphorus consumption, respectively. The main reason why we have considered this value range, 10–40%, is the technological feasibility of economically justified phosphorus recycling. The probability of obtaining phosphorus from economically sound recycling reaches 40–50% in the next 12–15 years (El Wali et al., 2018; Scholz and Wellmer, 2018).

The results obtained in scenario B compared to those in scenario A represent, on average, 9% and 27% increase in GHG emissions from mining and recycling stages of phosphorus supply chain respectively, whereas emissions remain the same throughout the processing phase (Fig. 6 (a–b)). In scenario C, by increasing the consumption of phosphorus by 30% we induce exponential growth of environmental problems in the recycling stage (Fig. 6 (c)). Therefore, in this case, recycling should not be recommended as a method ensuring sustainable supply of phosphorus in a long-term perspective. Scenario C shows that GHG emissions from recycling increase by around 49% while the mining stage emits about 23% more than in scenario A. Exponential growth of environmental problems caused by phosphorus recycling intensifies in scenario D (Fig. 6 (d)). The analysis of the results demonstrates that the growth in global phosphorus consumption slowly increases environmental problems throughout the mining stage and exacerbates them in the recycling stage, which is going to be a major problem. This result highlights the need for a strategic decision on the development of technologies for phosphorus recycling and a clear need for careful planning of the recycling process.

The significance of phosphorus recycling is obvious from a material flow perspective. However, several important limiting factors such as environmental issues (energy consumption and GHG emissions) should be considered when analyzing future options of recycling. Previous studies focused on the environmental impact of P recovery technologies by comparing different alternatives, e.g. Sandars et al. (2003) for livestock manure, Buratti et al. (2015) for solid waste, and Spångberg et al. (2014), Amann et al. (2018) and Ye et al. (2017) for wastewater.

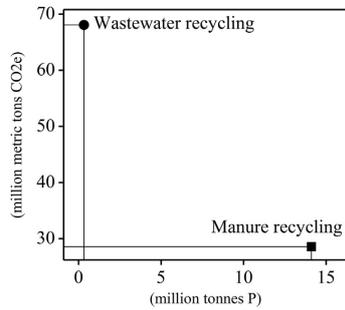


Fig. 5. Environmental issues of wastewater and manure recycling (2000–2050).

This paper presents a system dynamics model to analyze energy consumption and GHG emissions (CO₂, N₂O, CH₄ emissions) within the phosphorus supply chain over a 50-year time horizon. We examined three stages of the supply chain: mining, processing & production, and recycling. The main flows in the model were: at the mining stage, the extraction process of phosphate ores; in production, manufacturing fertilizers and animal feed; and in recycling, phosphorus recovery from manure, dry waste and wastewater.

The material flow analysis of this study shows that most of the recycled phosphorus comes from manure (e.g., around 14 million tons in 2013). This result supports previous research of the phosphorus supply chain. Scholz and Wellmer (2015a) estimated that the amount of manure used for soil fertilization ranged between 15 and

20 million tons P in 2011. Results obtained in this research show around 14.3 million tons P in the same year. Results of this paper also confirm previous findings indicating high energy demand and consequently large GHG emissions from the recycling stage of phosphorus supply chain. Results of this paper support findings highlighted by Amann et al. (2018) for the recovery of phosphorus from sewage sludge and Egle et al. (2016) for its recovery from wastewater and sludge treatment systems; as well as considerable GHG emissions from sewage sludge treatment noticed by Piippo et al. (2018).

5. Limitations of the study

Even though phosphorus is predominantly used in agricultural production, analysis of energy consumption for other uses was not considered (i.e. detergent production, other chemical uses) due to the lack of data.

6. Conclusions

Some important environmental issues such as high energy consumption and greenhouse gas emissions need to be addressed to ensure environmentally sustainable phosphorus supply chain. This article gives a comprehensive overview of various flows in phosphorus supply chain from the perspective of environmental sustainability.

In this paper, the obtained results show that the majority of recycled phosphorus comes from manure (around 82%). However, a detailed analysis of environmental issues shows that around 70% of GHG emissions in phosphorus life cycle originates from wastewater treatment process, which provides only around 2% of total phosphorus. This finding highlights the need for new strategies of recycling management as well as a need for the improvement of recycling technologies, especially

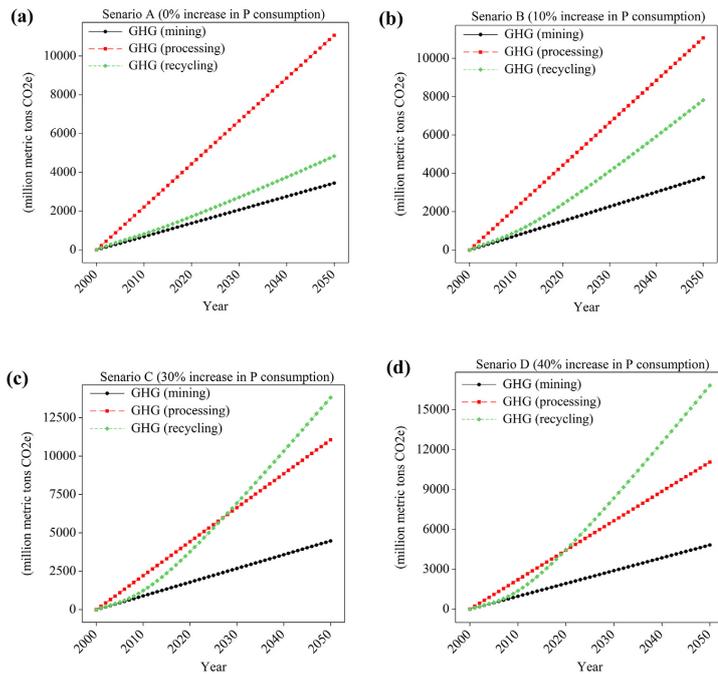


Fig. 6. Changes of greenhouse gas emissions from phosphorus supply chain in different scenarios (2000–2050).

for the recovery of phosphorus from wastewater. Any technology development for phosphorus recovery should take into account the environmental impact of the process and not just consider technical feasibility of recycling.

Moreover, the analysis of different scenarios shows that the increase in global phosphorus consumption slowly increases environmental problems in mining, which intensify in the recycling stage. It creates a major problem in a longer perspective, which, from environmental perspective, produces unsustainable phosphorus supply chain. Therefore, there is a clear need for a careful analysis of strategy of phosphorus recycling. Moreover, mining of phosphorus still has to be considered as an important alternative to recycling if environmental sustainability is to be achieved in the years 2000–2050.

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Competing financial interests

The authors declare no competing financial interests.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2019.03.439>.

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Appendix.

Table A1. Equations and calculation processes for the proposed phosphorus supply chain modeling.

Variable	Term	Unit	Equation	Coefficient
I. Mining				
$MS(t)$	Mined phosphorus stock	Tons P	$\int_{t_0}^t (M_R(t) - B_R(t) - BL_R(t))dt + MS(t_0)$	
$M_R(t)$	Mining rate	Tons P·year ⁻¹	$Pr * k_0 * k_1$	k_0 : phosphorus content in P_2O_5 k_1 : P_2O_5 content of Phosphate rock P_1 : Beneficiating coefficient
$P_R(t)$	Beneficiating rate	Tons P·year ⁻¹	$P_1 * MS(t)$	
$BS(t)$	Beneficiated phosphorus stock	Tons P	$\int_{t_0}^t (B_R(t) - PWL_R(t) - P_R(t))dt + MS(t_0)$	
$PS(t)$	Phosphoric acid stock	Tons P	$\int_{t_0}^t (P_R(t) - FEP_R(t) - FAP_R(t) - DP_R(t) - IP_R(t))dt + PS(t_0)$	
$P_R(t)$	Processing rate	Tons P·year ⁻¹	$P_2 * BS(t)$	P_2 : processing coefficient from primary material to phosphoric acid
II. Processing and Production				
$FEP_R(t)$	Fertilizer production rate	Tons P·year ⁻¹	$k_0 * P_3$	P_3 : production variable for fertilizers
$FAP_R(t)$	Feed production rate	Tons P·year ⁻¹	$P_4 * PS(t)$	P_4 : production variable for feed
$DP_R(t)$	Deiergents production rate	Tons P·year ⁻¹	$P_5 * PS(t)$	P_5 : production coefficient for deiergents
$DS(t)$	Deiergents stock	Tons P	$\int_{t_0}^t (DP_R(t) - DWW_R(t) - DC_R(t))dt + DS(t_0)$	
$IP_R(t)$	Industrial uses flow rate	Tons P·year ⁻¹	$P_6 * PS(t)$	P_6 : production coefficient for other industrial uses

Variable	Term	Unit	Equation	Coefficient
$FS(t)$	Fertilizers stock	Tons P	$\int_{t_0}^t (FEP_R(t) + TR_R(t) - CP_R(t) - PRL_R(t) - GC_R(t))dt + FS(t_0)$	
$CP_R(t)$	Crop production rate	Tons P·year ⁻¹	$P_7 * FS(t)$	P_7 : production coefficient of crops
$GC_R(t)$	Grass cut rate	Tons P·year ⁻¹	$P_8 * FS(t)$	P_8 : grass cutting coefficient
$FAS(t)$	Feed Stock	Tons P	$\int_{t_0}^t (FAP_R(t) + GC_R(t) + CF_R(t) - FEC_R(t))dt + FAS(t_0)$	
$CS(t)$	Crops Stock	Tons P	$\int_{t_0}^t (CF_R(t) + FOP1_R(t) - CF_R(t))dt + CS(t_0)$	
$CF_R(t)$	Crops to feed rate	Tons P·year ⁻¹	$P_9 * CS(t)$	P_9 : crops flow coefficient to feedstock
$FOS(t)$	Food Stock	Tons P	$\int_{t_0}^t (FOP1_R(t) + FOP2_R(t) - FOL_R(t) - FC_R(t))dt + FOS(t_0)$	
$FOP1_R(t)$	Vegetable food production rate	Tons P·year ⁻¹	$P_{10} * CS(t)$	P_{10} : Production coefficient of vegetable food
$FOP2_R(t)$	Animal food production rate	Tons P·year ⁻¹	$P_{11} * LS(t)$	P_{11} : production coefficient of animal food
III. Consumption				
$LS(t)$	Livestock	Tons P	$\int_{t_0}^t (FEC_R(t) - FOP2_R(t) - FC_R(t) - AWR_R(t))dt + LS(t_0)$	
$FC_R(t)$	Food consumption rate	Tons P·year ⁻¹	$VFD_R(t) + AFD_R(t)$	
$VFD_R(t)$	Vegetable food Demand rate	Tons P·year ⁻¹	$P_{12} * P(t)$	P_{12} : demand for vegetable food coefficient per one person
$AFD_R(t)$	Animal food demand rate	Tons P·year ⁻¹	$P_{13} * P(t)$	P_{13} : demand for animal food coefficient per one person

Variable	Term	Unit	Equation	Coefficient
$DC_R(t)$	Detergent consumption rate	Tons P·year ⁻¹	$P_{14} * DS(t)$	P_{14} : Detergent consumption coefficient
$FEC_R(t)$	Feed consumption rate	Tons P·year ⁻¹	$P_{15} * FAS(t)$	P_{15} : Feed consumption coefficient
$HS(t)$	Phosphorus from food consumption stock	Tons P	$\int_{t_0}^t (FC_R(t) - HL_R(t) - HE_R(t) - DW_R(t) - DMR_R(t))dt + HS(t_0)$	
IV. Waste generation				
$AW_R(t)$	Animal waste rate	Tons P·year ⁻¹	$W_0 * LS(t)$	W_0 : waste flow coefficient from animals
$HE_R(t)$	Human excreta flow rate	Tons P·year ⁻¹	$W_1 * HS(t)$	W_1 : waste flow coefficient from human bodies
$AWS(t)$	Animal waste stock	Tons P	$\int_{t_0}^t (AW_R(t) - AWL_R(t) - MR_R(t))dt + AWS(t_0)$	
$DW_R(t)$	Dry waste flow rate	Tons P·year ⁻¹	$W_2 * HS(t)$	W_2 : waste flow coefficient as dry waste from human bodies
$DWS(t)$	Dry waste stock	Tons P	$\int_{t_0}^t (DW_R(t) - DWL_R(t) - DWR_R(t))dt + DWS(t_0)$	
$DWL_R(t)$	Dry waste landfilling rate	Tons P·year ⁻¹	$W_3 * DWS(t)$	W_3 : landfilling coefficient of dry waste
$FECW_R(t)$	Feed consumption waste	Tons P·year ⁻¹	$P_{16} * FEC_R(t)$	P_{16} : Waste from feed consumption coefficient
$DWW_R(t)$	Detergent flow to wastewater	Tons P·year ⁻¹	$L8 * DS(t)$	L_8 : detergent flow to wastewater coefficient
$WWS(t)$	Wastewater stock	Tons P	$\int_{t_0}^t (HE_R(t) + DC_R(t) - WWT_R(t) - WWL_R(t))dt + WWS(t_0)$	
V. Recycling				

Variable	Term	Unit	Equation	Coefficient
$MR_R(t)$	Manure recycling rate	Tons P·year ⁻¹	$R0 * AWS(t)$	R_0 : recycling variable of manure
$DWR_R(t)$	Dry waste recycling rate	Tons P·year ⁻¹	$R1 * DWS(t)$	R_1 : recycling coefficient of dry waste
$WWR_R(t)$	Wastewater treating rate	Tons P·year ⁻¹	$R2 * WWS(t)$	R_2 : recycling coefficient of waste from wastewater
$OFS(t)$	Organic fertilizers stock	Tons P	$\int_t^{t_0} (MR_R(t) + DWR_R(t) + WWR_R(t) - OF_R(t)) dt + OFS(t_0)$	
$OF_R(t)$	Organic Fertilizers rate	Tons P·year ⁻¹	$DWR_R(t) + WWR_R(t) + MR_R(t)$	
$ROR_R(t)$	Recycling rate from agriculture runoff	Tons P·year ⁻¹	$R3 * PRL_R(t)$	R_3 : recycling coefficient of phosphorus from runoff of agriculture
$TR_R(t)$	Total recycling rate	Tons P·year ⁻¹	$ROR_R(t) + OF_R(t)$	
VI. Loss				
$PML_R(t)$	Phosphorus mining loss rate	Tons P·year ⁻¹	$BL_R(t) + PWL_R(t)$	
$BL_R(t)$	Beneficiation loss rate	Tons P·year ⁻¹	$L1 * MS(t)$	L_1 : loss coefficient during the beneficiation process
$PWL_R(t)$	Processing waste rate	Tons P·year ⁻¹	$L2 * BS(t)$	L_2 : loss coefficient during processing to phosphoric acid
$PRL_R(t)$	Phosphorus runoff rate	Tons P·year ⁻¹	$L3 * FS(t)$	L_3 : loss coefficient from fertilizers due to runoff of P
$FOL_R(t)$	Food loss rate	Tons P·year ⁻¹	$L4 * FOS(t)$	L_4 : loss coefficient due to food loss
$PAL_R(t)$	Animal waste loss rate	Tons P·year ⁻¹	$L5 * AWS(t)$	L_5 : loss coefficient from animal waste
$HL_R(t)$	Human waste loss rate	Tons P·year ⁻¹	$L6 * HS(t)$	L_6 : loss coefficient of human waste from human bodies

Variable	Term	Unit	Equation	Coefficient
$IL_R(t)$	Phosphorus loss rate from industrial uses	Tons P·year ⁻¹	$L_7 * PIL_R(t)$	L_7 : phosphorus loss coefficient from industrial uses
$DL_R(t)$	Phosphorus loss from detergent consumption	Tons P·year ⁻¹	$L_8 * DS(t)$	L_8 : phosphorus loss coefficient from detergent consumption
$PIL_R(t)$	Phosphorus industrial loss rate	Tons P·year ⁻¹	$L_9 * (IL_R(t) + DL_R(t))$	L_9 : loss coefficient of material from industrial
$PHL_R(t)$	Phosphorus loss rate to water from human waste	Tons P·year ⁻¹	$L_{10} * HLR(t)$	L_{10} : phosphorus loss coefficient from human waste to water
$WWL_R(t)$	Wastewater loss rate	Tons P·year ⁻¹	$L_{11} * WWS(t)$	L_{11} : loss coefficient of material from wastewater stock
$PL_R(t)$	Phosphorus loss rate	Tons P·year ⁻¹	$PMI_R(t) + PRI_R(t) + PIL_R(t) + FOL_R(t) + PAL_R(t) + PHL_R(t) + WWL_R(t)$	

VII. Energy Consumption

$EC_{mining}(t)$	Energy consumption in mining	Joules year ⁻¹	$P_{mining}(t) \times \sigma_{mining}$	$P_{mining}(t)$: flow of phosphorus (Tons P/year) in mining $\sigma_{mining}(t)$: Energy requirement for phosphate rock mining (Joules/Tons P)
$ECS_{mining}(t)$	Stock of energy consumption in mining	Joules year ⁻¹	$\int_{t_0}^t (EC_{mining}(t)) dt + EC_{mining}(t_0)$	
$EC_{Fertilizer}(t)$	Energy consumption in fertilizer	Joules year ⁻¹	$P_{fertilizer}(t) \times \sigma_{fertilizer}$	$P_{fertilizer}(t)$: flow of phosphorus (Tons P/year)

Variable	Term	Unit	Equation	Coefficient
	production			in fertilizer production
$EC_{S_{Fertilizer}}(t)$	Stock of energy consumption in fertilizer production	Joules year ⁻¹	$\int_{t_0}^t (EC_{Fertilizer}(t))dt + EC_{Fertilizer}(t_0)$	$\sigma_{Fertilizer}(t)$: Energy requirement for fertilizer production (Joules/Tons P)
$EC_{Feed}(t)$	Energy consumption in feed production	Joules year ⁻¹	$P_{Feed}(t) \times \sigma_{Feed}$	$P_{Feed}(t)$: flow of phosphorus (Tons P/year) in feed production σ_{Feed} : Energy requirement for feed production (Joules/Tons P)
$EC_{S_{Feed}}(t)$	Stock of energy consumption in feed production	Joules year ⁻¹	$\int_{t_0}^t (EC_{Feed}(t))dt + EC_{Feed}(t_0)$	
σ_{Feed}	Energy requirement for feed production	Joules/Tons P	$\frac{K_2}{K_3}$	K_2 : Energy required for feed production K_3 : phosphorus content in feed
$EC_{Dry\ waste}(t)$	Energy consumption in dry waste recycling	Joules year ⁻¹	$P_{drywaste}(t) \times \sigma_{drywaste}$	$P_{drywaste}(t)$: flow of phosphorus (Tons P/year) in dry waste recycling $\sigma_{drywaste}$: Energy requirement for dry waste recycling (Joules/Tons P)

Variable	Term	Unit	Equation	Coefficient
$EC_{Dry\ waste}(t)$	Stock of energy consumption in dry waste recycling	Joules year ⁻¹	$\int_{t_0}^t (EC_{Dry\ waste}(t)) dt + EC_{Dry\ waste}(t_0)$	
$EC_{Manure}(t)$	Energy consumption in manure recycling	Joules year ⁻¹	$P_{manure}(t) \times \sigma_{manure}$	$P_{manure}(t)$: flow of phosphorus (Tons P/year) in manure recycling σ_{manure} : Energy requirement for manure recycling (Joules/Tons P)
$EC_{Manure}(t)$	Energy consumption in manure recycling	Joules year ⁻¹	$\int_{t_0}^t (EC_{Manure}(t)) dt + EC_{Manure}(t_0)$	
$EC_{Wastewater}(t)$	Energy consumption in wastewater recycling	Joules year ⁻¹	$P_{wastewater}(t) \times \sigma_{wastewater}$	$P_{wastewater}(t)$: flow of phosphorus (Tons P/year) in wastewater recycling $\sigma_{wastewater}$: Energy requirement for wastewater recycling (Joules/Tons P)
$EC_{Wastewater}(t)$	Energy consumption in wastewater recycling	Joules year ⁻¹	$\int_{t_0}^t (EC_{Wastewater}(t)) dt + EC_{Wastewater}(t_0)$	

VIII. GHG Emissions

$CO_2\ mining(t)$	Carbon dioxide emissions from energy consumption in mining	Tons CO ₂ e year ⁻¹	$EC_{mining}(t) \times \left(\sum_{n=1}^6 \delta_{CO_2,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{CO_2,n}$: Tons CO ₂ -e Carbon dioxide emissions per one
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Variable	Term	Unit	Equation	Coefficient
$N_2O_{mining}(t)$	Nitrous oxide emissions from energy consumption in mining	Tons CO_2e year ⁻¹	$E_{C,mining}(t) \times \left(\sum_{n=1}^6 \delta_{N_2O,n} \times \beta_n \right)$	joules of Energy type (n) β_n : ratio of Energy type (n) used in mining n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{N_2O,n}(t)$: Tons CO2-e Nitrous Oxide emissions per one joules of Energy type (n)
$CH_4_{mining}(t)$	Methane emissions from energy consumption in mining	Tons CO_2e year ⁻¹	$E_{C,mining}(t) \times \left(\sum_{n=1}^6 \delta_{CH_4,n} \times \beta_n \right)$	β_n : ratio of Energy type (n) used in mining n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{CH_4,n}$: Tons CO2-e Methane emissions per one joules of Energy type (n)
$GHG_{mining}(t)$	GHG emissions from energy consumption in mining	Tons CO_2e year ⁻¹	$CO_2_{mining}(t) + N_2O_{mining}(t) + CH_4_{mining}(t)$	β_n : ratio of Energy type (n) used in mining
$CO_2(S)_{mining}(t)$	Cumulative carbon dioxide emissions stock from energy	Tons CO_2e	$\int_{t_0}^t (CO_2_{mining}(t)) dt + CO_2(S)_{mining}(t_0)$	

Variable	Term	Unit	Equation	Coefficient
$N_2O(S)_{mining}(t)$	consumption in mining Cumulative nitrous oxide emissions stock from energy consumption in mining	Tons CO _{2e}	$\int_{t_0}^t (N_2O_{mining}(t)) dt + N_2O(S)_{mining}(t_0)$	
$CH_4(S)_{mining}(t)$	Cumulative methane emissions stock from energy consumption in mining	Tons CO _{2e}	$\int_{t_0}^t (CH_4_{mining}(t)) dt + CH_4(S)_{mining}(t_0)$	
$GHG(S)_{mining}(t)$	Cumulative GHG emissions stock from energy consumption in mining	Tons CO _{2e}	$CO_2(S)_{mining}(t) + N_2O(S)_{mining}(t) + CH_4(S)_{mining}(t)$	
$CO_2Fertilizer(t)$	Carbon dioxide emissions from energy consumption in fertilizer production	Tons year ⁻¹	$E_{Fertilizer}(t) \times \left(\sum_{n=1}^6 \delta_{CO_2,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{CO_2,n}$: Tons CO ₂ -e Carbon dioxide emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in Fertilize production n : Type of energy where
$N_2OFertilizer(t)$	Nitrous oxide emissions from energy	Tons year ⁻¹	$E_{Fertilizer}(t) \times \left(\sum_{n=1}^6 \delta_{N_2O,n} \times \beta_n \right)$	

Variable	Term	Unit	Equation	Coefficient
	consumption in fertilizer production			$n = 1, 2, 3, 4, 5, 6;$ $\delta_{N_2O,n}$: Tons CO ₂ -e Nitrous Oxide emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in fertilizer production n : Type of energy where $n = 1, 2, 3, 4, 5, 6;$
$CH_4_{Fertilizer}(t)$	Methane emissions from energy consumption in fertilizer production	Tons year ⁻¹	$E_{C_{Fertilizer}}(t) \times \left(\sum_{n=1}^6 \delta_{CH_4,n} \times \beta_n \right)$	$\delta_{CH_4,n}$: Tons CO ₂ -e Methane emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in fertilizer production n : Type of energy where $n = 1, 2, 3, 4, 5, 6;$
$GHG_{Fertilizer}(t)$	GHG emissions from energy consumption in fertilizer production	Tons year ⁻¹	$CO_{2,Fertilizer}(t) + N_2O_{Fertilizer}(t) + CH_4_{Fertilizer}(t)$	
$CO_2_{Feed}(t)$	Carbon dioxide emissions from energy consumption in feed production	Tons year ⁻¹	$E_{C_{Feed}}(t) \times \left(\sum_{n=1}^6 \delta_{CO_2,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6;$ $\delta_{CO_2,n}$: Tons CO ₂ -e Carbon dioxide emissions per one joules of Energy type (n) β_n : ratio of Energy type (n)

Variable	Term	Unit	Equation	Coefficient
$N_2O_{Feed}(t)$	Nitrous oxide emissions from energy consumption in feed production	Tons year ⁻¹	$EC_{Feed}(t) \times \left(\sum_{n=1}^6 \delta_{N_2O,n} \times \beta_n \right)$	used in Feed production n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{N_2O,n}$: Tons CO ₂ -e Nitrous Oxide emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in Feed production
$CH_4_{Feed}(t)$	Methane emissions from energy consumption in feed production	Tons year ⁻¹	$EC_{Feed}(t) \times \left(\sum_{n=1}^6 \delta_{CH_4,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{CH_4,n}$: Tons CO ₂ -e Methane emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in Feed production
$GHG_{Feed}(t)$	GHG emissions from energy consumption in feed production	Tons year ⁻¹	$CO_2_{Feed}(t) + N_2O_{Feed} + CH_4_{Feed}$	
$CO_2_{Production}(t)$	Carbon dioxide emissions from energy consumption in production	Tons year ⁻¹	$CO_2_{Fertilizer}(t) + CO_2_{Feed}(t)$	
$N_2O_{Production}(t)$	Nitrous oxide emissions from energy	Tons year ⁻¹	$N_2O_{Fertilizer}(t) + N_2O_{Feed}(t)$	

Variable	Term	Unit	Equation	Coefficient
CH_4 Production (t)	consumption in production Methane emissions from energy consumption in production Cumulative carbon dioxide emissions stock from energy	Tons year ⁻¹ CO _{2e}	$CH_4_{Fertilizer}(t) + CH_4_{Feed}(t)$	
$CO_2(S)$ production (t)	stock from energy consumption in production Cumulative nitrous oxide emissions stock from energy	Tons CO _{2e}	$\int_{t_0}^t (CO_2_{production}(t)) dt + CO_2(S)_{production}(t_0)$	
$N_2O(S)$ production (t)	stock from energy consumption in production Cumulative nitrous oxide emissions stock from energy	Tons CO _{2e}	$\int_{t_0}^t (N_2O_{production}(t)) dt + N_2O(S)_{production}(t_0)$	
$CH_4(S)$ production (t)	stock from energy consumption in production Cumulative methane emissions stock from energy	Tons CO _{2e}	$\int_{t_0}^t (CH_4_{production}(t)) dt + CH_4(S)_{production}(t_0)$	
$GHG(S)$ Production (t)	stock from energy consumption in production Cumulative GHG emissions stock from energy	Tons CO _{2e}	$CO_2(S)_{production}(t) + N_2O(S)_{production}(t) + CH_4(S)_{production}(t)$	
CO_2 Dry waste (t)	Carbon dioxide emissions from	Tons year ⁻¹ CO _{2e}	$E_{C_{drywaste}}(t) \times (\sum_{n=1}^6 \delta_{CO_2,n} \times \beta_n)$	n: Type of energy where

Variable	Term	Unit	Equation	Coefficient
	energy consumption in dry waste recycling			$n = 1, 2, 3, 4, 5, 6;$
				$\delta_{CO_2,n}$: Tons CO ₂ -e Carbon dioxide emissions per one joules of Energy type (n)
				β_n : ratio of Energy type (n) used in dry waste recycling
				n : Type of energy where
				$n = 1, 2, 3, 4, 5, 6;$
$N_2O_{Dry\ waste}(t)$	Nitrous oxide emissions from energy consumption in dry waste recycling	Tons year ⁻¹	$E_{C_{Dry\ waste}}(t) \times \left(\sum_{n=1}^6 \delta_{N_2O,n} \times \beta_n \right)$	$\delta_{N_2O,n}$: Tons CO ₂ -e Nitrous Oxide emissions per one joules of Energy type (n)
				β_n : ratio of Energy type (n) used in dry waste recycling
				n : Type of energy where
				$n = 1, 2, 3, 4, 5, 6;$
$CH_4_{Dry\ waste}(t)$	Methane emissions from energy consumption in dry waste recycling	Tons year ⁻¹	$E_{C_{Dry\ waste}}(t) \times \left(\sum_{n=1}^6 \delta_{CH_4,n} \times \beta_n \right)$	$\delta_{CH_4,n}$: Tons CO ₂ -e Methane emissions per one joules of Energy type (n)
				β_n : ratio of Energy type (n) used in dry waste recycling
				n : Type of energy where
				$n = 1, 2, 3, 4, 5, 6;$
$GHG_{Dry\ waste}(t)$	GHG emissions from energy consumption in dry waste recycling	Tons year ⁻¹	$CO_{2,Dry\ waste}(t) + N_2O_{Dry\ waste}(t) + CH_4_{Dry\ waste}(t)$	β_n : ratio of Energy type (n) used in dry waste recycling

Variable	Term	Unit	Equation	Coefficient
$CO_2\text{ Manure}(t)$	recycling Carbon dioxide emissions from energy consumption in manure recycling	Tons year ⁻¹ CO ₂ e	$E_{C\text{manure}}(t) \times \left(\sum_{n=1}^6 \delta_{CO_2,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{CO_2,n}$: Tons CO ₂ -e Carbon dioxide emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in manure recycling
$N_2O\text{ Manure}(t)$	Nitrous oxide emissions from energy consumption in manure recycling	Tons year ⁻¹ CO ₂ e	$E_{C\text{manure}}(t) \times \left(\sum_{n=1}^6 \delta_{N_2O,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{N_2O,n}$: Tons CO ₂ -e Nitrous Oxide emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in manure recycling
$CH_4\text{ Manure}(t)$	Methane emissions from energy consumption in manure recycling	Tons year ⁻¹ CO ₂ e	$E_{C\text{manure}}(t) \times \left(\sum_{n=1}^6 \delta_{CH_4,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{CH_4,n}$: Tons CO ₂ -e Methane emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in manure recycling
$GHG\text{ Manure}(t)$	GHG emissions from	Tons year ⁻¹ CO ₂ e	$CO_2\text{ Manure}(t) + N_2O\text{ Manure}(t) + CH_4\text{ Manure}(t)$	β_n : ratio of Energy type (n) used in manure recycling

Variable	Term	Unit	Equation	Coefficient
$CO_{2,wastewater}(t)$	energy consumption in manure recycling Carbon dioxide emissions from energy consumption in wastewater recycling	Tons CO_{2e} year ⁻¹	$E_{C,wastewater}(t) \times \left(\sum_{n=1}^6 \delta_{CO_2,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{CO_2,n}$: Tons CO ₂ -e Carbon dioxide emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in wastewater recycling
$N_2O_{wastewater}(t)$	Nitrous oxide emissions from energy consumption in wastewater recycling	Tons CO_{2e} year ⁻¹	$E_{C,wastewater}(t) \times \left(\sum_{n=1}^6 \delta_{N_2O,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{N_2O,n}$: Tons CO ₂ -e Nitrous Oxide emissions per one joules of Energy type (n) β_n : ratio of Energy type (n) used in wastewater recycling
$CH_{4,wastewater}(t)$	Methane emissions from energy consumption in wastewater recycling	Tons CO_{2e} year ⁻¹	$E_{C,wastewater}(t) \times \left(\sum_{n=1}^6 \delta_{CH_4,n} \times \beta_n \right)$	n : Type of energy where $n = 1, 2, 3, 4, 5, 6$; $\delta_{CH_4,n}$: Tons CO ₂ -e Methane emissions per one joules of Energy type (n)

Variable	Term	Unit	Equation	Coefficient
$GHG_{wastewater}(t)$	GHG emissions from energy consumption in wastewater recycling	Tons CO_2e year ⁻¹	$CO_2_{wastewater}(t) + N_2O_{wastewater}(t) + CH_4_{wastewater}(t)$	β_{w} : ratio of Energy type (n) used in wastewater recycling
$CO_2\ Recycling(t)$	Carbon dioxide emissions from energy consumption in recycling	Tons CO_2e year ⁻¹	$CO_2_{Dry\ waste}(t) + CO_2_{Manure}(t) + CO_2_{wastewater}(t)$	
$N_2O\ Recycling(t)$	Nitrous oxide emissions from energy consumption in recycling	Tons CO_2e year ⁻¹	$N_2O_{Dry\ waste}(t) + N_2O_{Manure}(t) + N_2O_{wastewater}(t)$	
$CH_4\ Recycling(t)$	Methane emissions from energy consumption in recycling	Tons CO_2e year ⁻¹	$CH_4_{Dry\ waste}(t) + CH_4_{Manure}(t) + CH_4_{wastewater}(t)$	
$GHG_{Recycling}(t)$	GHG emissions from energy consumption in recycling	Tons CO_2e year ⁻¹	$GHG_{wastewater}(t) + GHG_{Manure}(t) + GHG_{Dry\ waste}(t)$	
$CO_2(S)_{recycling}(t)$	Cumulative carbon dioxide emissions stock from energy consumption in recycling	Tons CO_2e	$\int_{t_0}^t (CO_2_{recycling}(t)) dt + CO_2(S)_{recycling}(t_0)$	

Variable	Term	Unit	Equation	Coefficient
$N_2O(S)_{recycling}(t)$	recycling Cumulative nitrous oxide emissions stock from energy consumption in recycling	Tons CO _{2e}	$\int_{t_0}^t (N_2O_{recycling}(t)) dt + N_2O(S)_{recycling}(t_0)$	
$CH_4(S)_{recycling}(t)$	Cumulative methane emissions stock from energy consumption in recycling	Tons CO _{2e}	$\int_{t_0}^t (CH_4_{recycling}(t)) dt + CH_4(S)_{recycling}(t_0)$	
$GHG(S)_{recycling}(t)$	Cumulative GHG emissions stock from energy consumption in recycling	Tons CO _{2e}	$CO_2(S)_{recycling}(t) + N_2O(S)_{recycling}(t) + CH_4(S)_{recycling}(t)$	
$\delta_{CO_2,n}$	Tons CO _{2-e} Carbon dioxide emissions per one joules of Energy type (n)	Tons CO _{2e} joules ⁻¹	$GHG_n \times \gamma_{CO_2}$	GHG_n : GHG emissions from energy type (n)
$\delta_{N_2O,n}$	Tons CO _{2-e} Nitrous Oxide emissions per one joules of Energy type (n)	Tons CO _{2e} joules ⁻¹	$GHG_n \times \gamma_{N_2O}$	γ_{CO_2} : Carbon dioxide share in GHG emissions GHG_n : GHG emissions from energy type (n)
$\delta_{CH_4,n}$	Tons CO _{2-e} Methane emissions per	Tons CO _{2e} joules ⁻¹	$GHG_n \times \gamma_{CH_4}$	γ_{N_2O} : Nitrous Oxide share in GHG emissions GHG_n : GHG emissions from energy type (n)

Variable	Term	Unit	Equation	Coefficient
	one joule of Energy type (n)			γ_{CH_4} : Methane share in GHG emissions
Table A2. Data sources for all parameters of the proposed model.				
Parameter	Description	Unit	Value range	Value in this study
Pt	Phosphate rock mining (mining rate)	Tons phosphate rock gross	2.17e+008-2.64e+008	2.17e+008-2.64e+008
K ₀	P content in P ₂ O ₅	Dimensionless	0.436	0.436
K ₁	P ₂ O ₅ content in Phosphate rock	%	30	30
P ₁	Beneficiating coefficient	Dimensionless	-	0.84
L ₁	loss coefficient during the beneficiation process	Dimensionless	0.16	0.16
P ₂	processing coefficient from primary material to phosphoric acid	Dimensionless	-	0.875
L ₂	loss coefficient during the processing to phosphoric acid	Dimensionless	0.125	0.125
P ₃	Production variable for fertilizers	Tons P2O5	36.846,725-53.299,875	36.846,725-53.299,875
P ₄	Production variable for feed	Million tons P	1.12696-1.58259	Distribution: 3-Parameter Weibull Shape: 1.653 Scale: 0.23 Threshold: 1.113 Number of data points = 25 AD: 0.266 Corresponding p-value > 0.151 Sample mean = 1.31810 Sample St Dev = 0.126806 Minimum = 1.12696 Maximum = 1.58259
P ₅	Production coefficient for detergents	Dimensionless	0.03	(Supplementary Table A3) 0.03
P ₆	Production coefficient for other industrial uses	Dimensionless	0.08	(Scholz and Wellmer, 2013) (Scholz et al., 2015)

Parameter	Description	Unit	Value range	Value in this study	Data sources
P ₇	Production coefficient for crops	Dimensionless	0.3	0.3	(Chen and Graedel, 2016)
P ₈	Grass cutting coefficient	Dimensionless	0.32	0.32	(Chen and Graedel, 2016)
L ₃	Loss coefficient from fertilizers due to runoff of P	Dimensionless	0.38	0.38	(Chen and Graedel, 2016)
P ₉	crops flow coefficient to feedstock	Dimensionless	0.3	0.3	(Chen and Graedel, 2016)
P ₁₀	production coefficient of vegetable food	Dimensionless	0.7	0.7	(Chen and Graedel, 2016)
W ₀	waste flow coefficient from animals	Dimensionless	0.92	0.92	(Chen and Graedel, 2016)
P ₁₁	production coefficient of animal food	Dimensionless	0.08	0.08	(Chen and Graedel, 2016)
P ₁₂	demand of vegetable food coefficient per one person	Tons P/ capita year ⁻¹	0.0003	0.0003	(Cordell et al., 2009)
P ₁₃	demand of animal food coefficient per one person	Tons P/ capita year ⁻¹	0.0006	0.0006	(Cordell et al., 2009)
P ₁₄	Detergent consumption coefficient	Dimensionless	0.7	0.7	(Chen and Graedel, 2016)
P ₁₅	Feed consumption	Dimensionless	1	1	(Scholz and Wellmer, 2015a)
L ₅	loss coefficient from animal waste	Dimensionless	0.1	0.1	(Scholz and Wellmer, 2015a)
R ₀	Recycling variable of manure	Million tons P year ⁻¹	9,79895-15,260869	Distribution: Weibull Shape: 9,99 Scale:13,35 Number of data points = 54 AD: 0,375 Corresponding p-value > 0,250 Sample mean = 12,8374 Sample St.Dev = 1,39695 Minimum = 9,79895 Maximum = 15,2609	(FAO, 2017c)
R ₁	recycling coefficient of dry waste	Dimensionless	0.67	(Supplementary Table A3) 0.67	(Scholz and Wellmer, 2015a)
R ₂	recycling coefficient of waste from wastewater	Dimensionless	0.71	0.71	(Scholz and Wellmer, 2015a)
R ₃	recycling coefficient from	Dimensionless	0.25	0.25	(Chen and Graedel, 2016)

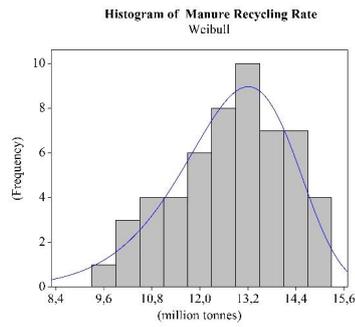
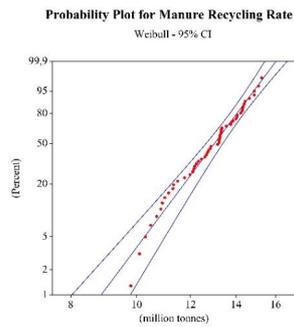
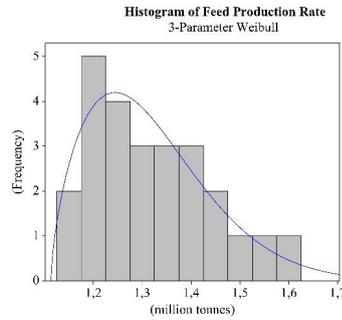
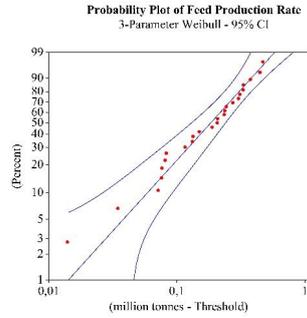
Parameter	Description	Unit	Value range	Value in this study	Data sources
L ₆	phosphorus runoff from agriculture	Dimensionless	0.12	0.12	(Scholz and Wellmer, 2015a)
L ₇	loss coefficient of human waste from human bodies	Dimensionless	0.08	0.08	(Chen and Graedel, 2016)
L ₈	phosphorus loss coefficient from industrial uses	Dimensionless	0.25	0.25	(Scholz and Wellmer, 2015a)
L ₁₀	phosphorus loss coefficient from detergent consumption	Dimensionless	0.029	0.029	(Chen and Graedel, 2016)
L ₉	phosphorus loss coefficient from human waste to water	Dimensionless	0.017	0.017	(Chen and Graedel, 2016)
W ₁	loss coefficient of material from industrial	Dimensionless	0.5	0.5	(Scholz and Wellmer, 2015a)
W ₂	waste flow coefficient from human bodies	Dimensionless	0.38	0.38	(Scholz and Wellmer, 2015a)
W ₃	waste flow coefficient as dry landfilling coefficient of dry waste	Dimensionless	0.33	0.33	(Scholz and Wellmer, 2015a)
R ₂	recycling coefficient of dry waste	Dimensionless	0.67	0.67	(Scholz and Wellmer, 2015a)
R ₃	recycling coefficient of material flow from wastewater stock	Dimensionless	0.71	0.71	(Scholz and Wellmer, 2015a)
L ₁₁	loss coefficient of material flow from wastewater stock	Dimensionless	0.29	0.29	(Scholz and Wellmer, 2015a)
GHG ₁	GHG emissions from Coal energy	Tons CO _{2e} GW ⁻¹	Low: 756 High: 1,310 Mean: 888	888	(World Nuclear Association, 2011)
GHG ₂	GHG emissions from Gas energy	Tons CO _{2e} GW ⁻¹	Low: 362 High: 891 Mean: 499	499	(World Nuclear Association, 2011)
GHG ₃	GHG emissions from Oil energy	Tons CO _{2e} GW ⁻¹	Low: 547 High: 935 Mean: 733	733	(World Nuclear Association, 2011)
GHG ₄	GHG emissions from Nuclear energy	Tons CO _{2e} GW ⁻¹	Low: 2 High: 130 Mean: 29	29	(World Nuclear Association, 2011)
GHG ₅	GHG emissions from Hydroelectric energy	Tons CO _{2e} GW ⁻¹	Low: 2 High: 237	26	(World Nuclear Association, 2011)

Parameter	Description	Unit	Value range	Value in this study	Data sources
GHG_6	GHG emissions from Other renewables energy	Tons CO ₂ e GWh ⁻¹	Mean: 26 Low: 6 High: 124 Mean: 26	26	(World Nuclear Association, 2011)
β_1	Ratio of coal energy used in energy consumption	Dimensionless	0.3	0.3	(Petroleum, 2016)
β_2	Ratio of gas energy used in energy consumption	Dimensionless	0.24	0.24	(Petroleum, 2016)
β_3	Ratio of oil energy used in energy consumption	Dimensionless	0.33	0.33	(Petroleum, 2016)
β_4	Ratio of nuclear energy used in energy consumption	Dimensionless	0.04	0.04	(Petroleum, 2016)
β_5	Ratio of hydroelectric energy used in energy consumption	Dimensionless	0.07	0.07	(Petroleum, 2016)
β_6	Ratio of other renewable energy used in energy consumption	Dimensionless	0.02	0.02	(Petroleum, 2016)
σ_{mining}	Energy requirement for phosphate rock mining	MJ/Kg P	12	12	(Schroder et al., 2010)
$\sigma_{fertilizer}$	Energy requirement for fertilizer production	kWh/Tons P	1,691.328	1,691.328	(F. Zhang et al., 2017)
K_2	Energy for Feed production	MJ/Kg ingredients	Average: 3.792	3.792	(Frorip et al., 2012)
K_3	Total phosphorus in Feed	g P/Kg	Average: 9.2975	9.2975	(Inorganic Feed Phosphates (IFP), 2018) (Sandars et al., 2003)
σ_{manure}	Energy requirement for manure recycling	MJ/11.3 Kg P	126.1	126.1	(Spångberg et al., 2014)
$\sigma_{wastewater}$	Energy requirement for wastewater recycling	MJ/0.15 Kg P	179	179	(Buratti et al., 2015)
$\sigma_{drywaste}$	Energy requirement for dry waste recycling	kWh/T waste	14.95	14.95	(U.S. Energy information administration, 2011b)
γ_{CO2}	Carbon dioxide share in GHG emissions	Dimensionless	0.862	0.862	(U.S. Energy information administration, 2011c)
γ_{N2O}	Nitrous oxide share in GHG emissions	Dimensionless	0.017	0.017	(U.S. Energy information administration, 2011d)
γ_{CH4}	Methane share in GHG emissions	Dimensionless	0.121	0.121	(U.S. Energy information administration, 2011d)

Table A3. Distribution fitting and data details.

Variables distribution summary

Feed production rate
 Distribution: 3-Parameter Weibull
 Shape: 1.653
 Scale: 0.23
 Thresh: 1.113
 Number of data points = 25
 AD: 0.266
 Corresponding p-value > 0.151
 Sample mean = 1.31810
 Sample St Dev = 0.126806
 Minimum = 1.12696
 Maximum = 1.58259
Manure recycling rate
 Distribution: Weibull
 Shape: 9.99
 Scale: 13.35
 Number of data points = 54
 AD: 0.375
 Corresponding p-value > 0.250
 Sample mean = 12.8374
 Sample St Dev = 1.39695
 Minimum = 9.79895
 Maximum = 15.2609



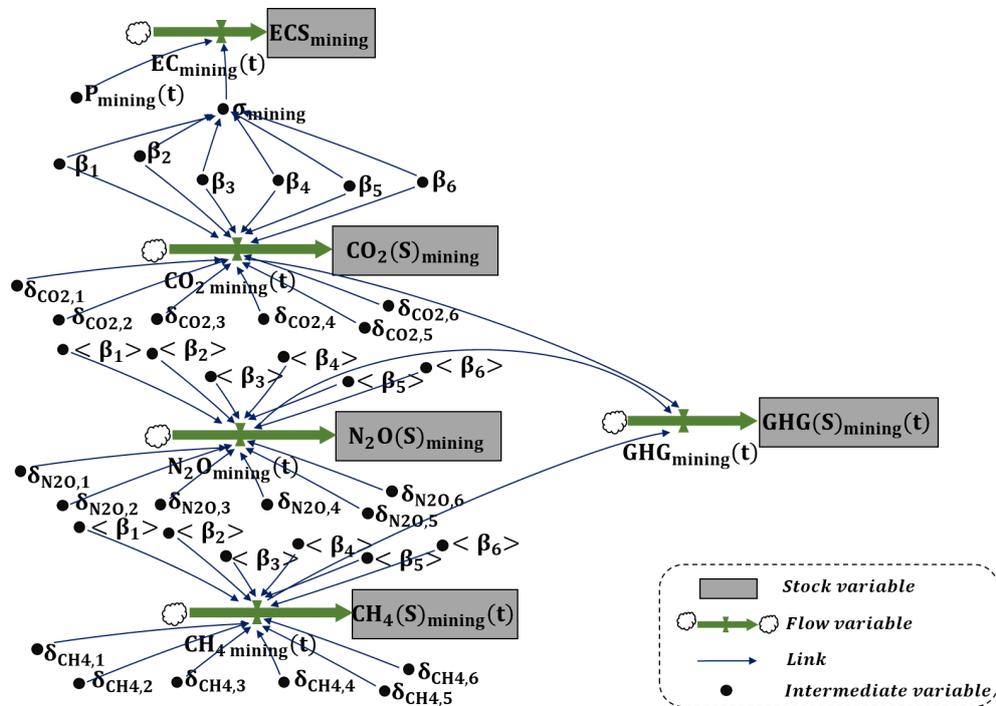


Figure A2. Model structure of energy consumption and GHG emissions for mining stage as an example.

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Article

Environmental Sustainability of Niobium Recycling: The Case of the Automotive Industry

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Abstract: The recycling of scrap is one of the common approaches aiming at reduction of mining-based production of critical metals and mitigation of their supply risk as well as processing-related environmental impact. The number of currently available end-of-life vehicles (ELVs) indicates—significant potential for critical metals recycling, especially niobium (Nb). Therefore, the quantification of environmental impact of niobium recovery starts to be an important issue in assessment of sustainability of large-scale recycling processes. In this paper, we assess energy consumption and greenhouse gas (GHG) emissions in individual stages of niobium supply chain in the automotive industry over the period 2010–2050. The different stages including mining, production and recycling are analyzed using dynamic simulation. The results show the majority of the consumed energy (45% of energy demand in niobium supply chain) is used in the primary production stage. This stage also contributes to 72% of total gas emissions of supply chain over the period 2010–2050. Mining of niobium consumes up to 36% of energy and generates ca. 21% of GHG emissions. While, in recycling stage, the secondary production of niobium requires 19% of supply chain energy and generates 7% of gas emissions. The detailed calculations show that recycling of niobium could save around 133–161 m GJ energy between 2010 and 2050. The recycling would also contribute to the reduction of 44–53 mt CO₂-eq in the same period. It shows around 18% reduction of annual emissions between 2010 and 2050 thanks to reuse of niobium in secondary production rather than primary production.

Keywords: critical materials; niobium; recycling; energy consumption; greenhouse gas emissions; dynamic model

1. Introduction

Niobium (Nb) is an essential element for production of steel and superalloys, superconductors, electronic components, medical implants, etc. The estimated resources of niobium are substantial and they are sufficient to meet the worldwide demand in the foreseeable future [1]. However, the oligopoly nature of the niobium market and the lack of substitutes create its supply risk [2–5]. It is worth to mention that the niobium demand has increased dramatically over the past decade, particularly as an element of microalloys in high strength and stainless steels used in the automotive industry [6].

The global market of niobium grew annually by 10% between 2000 and 2010 [7]. In view of the rapid increase in primary and secondary niobium production over the last 15 years [8], the production

rate will peak in 2025 with recycling rate at around 60% [9]. Therefore, the most significant increase in demand for niobium will take place in the coming years.

In the production of steel for the automotive industry, Nb is used in the form of ferroniobium. Ferroniobium represents the most significant fraction of the world's demand of niobium—around 87% [1]. The growing use of ferroniobium in the automotive industry is driven by a trend to reduce the weight of the vehicles [10–12].

The end-of-life vehicles (ELVs) have become a major waste stream [13]. Therefore, maximisation of recyclability is one of the dominant trends in car manufacturing. It can contribute considerably to the reduction of the wastes but also constitutes a significant source of raw materials. For example, in 2010, the total number of ELVs was around 40 million; mainly in Germany, Italy, France, UK, Spain, USA, Canada, Brazil, Japan, China, Korea and Australia [14]. On average concentration of Nb in steel alloy is low and generally lower than 0.1 wt% [1]. However, a considerable number of ELVs makes this source of niobium quite important. In 2010, based on the available global number of ELVs, the amount of niobium in high-strength steel (HSS) alloy used in passenger cars (average weight 1.5 tonnes) could be estimated at around 36,000 tonnes compared to 49,100 tonnes of niobium mined globally [15]. The annual global production of niobium alloyed steel was estimated at about 50 million tonnes [16]. According to the estimates provided by the automotive industry for 2020, the steel content in a typical car will increase from 54% to 64% [17].

The decision makers need to be aware of possible environmental effects at each stage of the supply chain when analysing the use of virgin ores or recycled materials [18]. In the automotive industry, recycling strategies for critical materials in ELVs are poorly understood due to the complexity of the material flows [19] as well as environmental and economic implications of critical materials recycling [20].

In recent decades, supply chain research has attempted to address the issues of environmental impact and social sustainability [21–23]. Moreover, new avenues of research have been opened to aim at developing holistic perspectives of sustainability of supply chain. From this perspective, researchers have started investigating the available options for reaching both higher economic growth and lower GHG emissions [24,25]. In this case, the effect of energy consumption on sustainability and its impact on GHG emissions are very important issues relevant in all stages of the supply chain [26,27]. However, the question remains as to what levels of energy consumption and GHG emissions at each stage of the niobium supply chain would ensure environmental sustainability. Therefore, the main objective of this paper is to present a dynamic model of the niobium supply chain to investigate energy consumption and GHG emissions at each stage including mining, production and recycling.

2. Dynamic Model of Niobium Supply Chain

The assessment of specific modifications of supply chain needs a systems approach. It consists of the detailed analysis of dynamics of all stages of the supply chain [12]. Therefore, the methodology of system dynamics introduced by Forrester [28] is used in this study. The system dynamics methodology has been used in many applications, both in social sciences and engineering. In the case of critical materials, system dynamics models have been used in different sectors for such materials as indium [29], platinum group metals [30], rare earth elements [31], uranium [32], lithium [33], phosphorus [34] and niobium [35].

The model presented in this paper considers the complex interrelationships between mining and processing, production and recycling stages of niobium global supply chain. In production stage, we consider niobium manufacturing in the automotive industry. The structural model is built and simulated by identifying the key variables and their interactions in the different stages of niobium life cycle. The conceptual model is presented in Figure 1, where niobium flow starts with the mining stage and next the interrelations with other stages are considered. Each stage of the model consists of three main layers (submodels), including material and energy flow as well as GHG emissions. The first submodel, material flow, is composed of the following modules: mining,

extraction, processing, production, consumption, collection and recycling. The energy consumption submodel takes into account energy consumed in mining, production and recycling. The GHG emissions submodel is primarily related to the energy consumption and therefore the structure of both submodels are identical. There was analyzed flow of niobium in the different processes, e.g., pyrochlore ore extraction, ferroniobium production, standard grade ferroniobium production, HSS steel production for automobile industry, ELVs collection process and recycling of HSS steel. Therefore, there were assessed energy consumption and GHG emissions of those processes. Below, we present the analysis of the particular stages in detail.

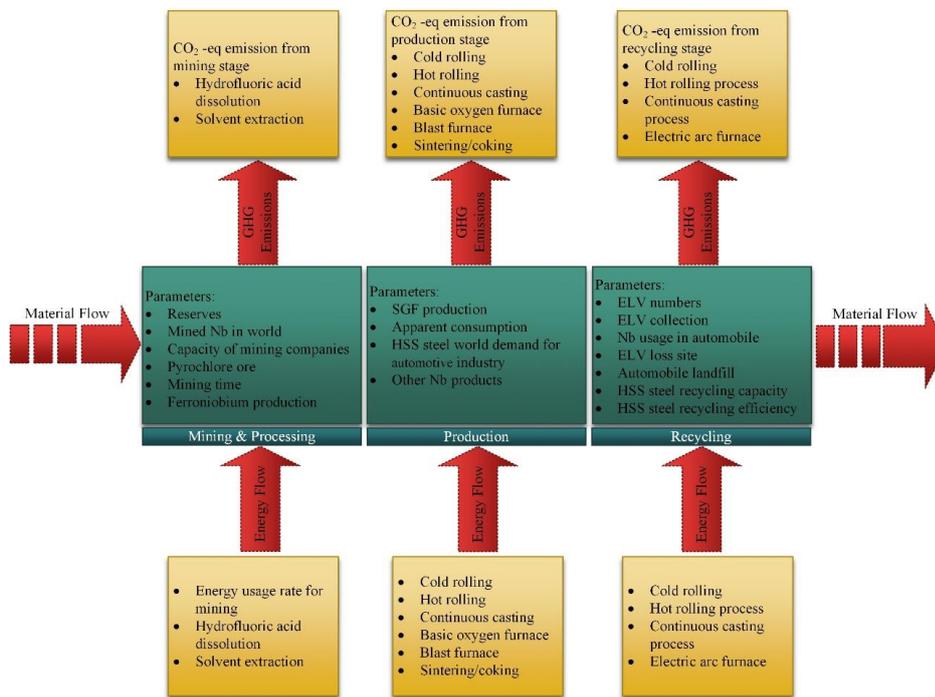


Figure 1. Conceptual model of niobium global supply chain (application in automotive market).

System Definition and Model Description

Figure 2 shows the simulation model. In system dynamics modeling, state variables change continuously over time. System dynamics model is a complex system incorporating three types of variables: (i) stock (or level) variable that is a reservoir of a given resource (also called state variable), (ii) flow variable that adjusts the level of stock through inbound and outbound flows, and (iii) intermediate variable (auxiliary) consisting of functions of stocks (and constants or exogenous inputs) [36]. It is important to note that every variable in system dynamics model is calculated at each time step. For example, in this work, we consider time step equal to one year as the used input data are based on yearly reports. In this study, data are collected from different sources for time horizon 1950–2017 (Appendix, Table A1). In Table 1, we present all variables and parameters.

Table 1. Description of variables and parameters of the niobium simulation model.

Notation	Term
Nb _{M-R} (t)	The world production of mineral concentrates (niobium content)
EM (t)	Extracted material from primary production of niobium
SGF _{P-R} (t)	Standard grade ferroniobium production rate
OP _{P-R} (t)	The rate of niobium flow in other products
SGF (t)	Standard grade ferroniobium stock
Nb _{P-R} (t)	The rate of niobium flow in the production stage (through primary production of high strength alloy steels in automobile industry)
T _{W-ELV} (t)	Total weight of car collected as ELV
T _{S-ELV} (t)	Total high strength alloy steels used in ELV
Nb _{ELV} (t)	The amount of niobium available in ELV
S _{Nb-ELV} (t)	The stock of available niobium from collected ELVs
Nb _{R-S-R} (t)	Recyclable niobium from high strength alloy steels in ELVs
W _{Landfill-R} (t)	Niobium in the scrap recycling process from the automobile loss
Nb _{R-R} (t)	The rate of niobium in the recycling stage
S _R (t)	The stock of recycled materials
Nb _{MP-Brazil} (t)	World production of mineral concentrates (niobium content) by Brazil
Nb _{MP-Canada} (t)	World production of mineral concentrates (niobium content) by Canada
Nb _{MP-Other} (t)	World production of mineral concentrates (niobium content) by other countries
Y _{Nb}	Yearly world production of mineral concentrates (niobium content) by each country
SGF _{Coeff}	Percentage of global niobium production used to produce ferroniobium applied in high strength alloy steels
OP _{Coeff}	Percentage of global niobium production used in manufacture of niobium alloys, niobium chemicals and carbides, high purity ferroniobium, and other niobium metal products
δ ₁	Energy usage through hydrofluoric acid dissolution process
δ ₂	Energy usage through solvent extraction process
γ ₁	The greenhouse gas emitted through hydrofluoric acid dissolution process
γ ₂	The greenhouse gas emitted through solvent extraction process
P _{Nb}	Nb grade in HSS ferroniobium applied in automobile
P _S	Steel in automobile
W _{Car}	Weight of car
ELV _S	ELVs number in different countries/state including European Union, Germany, Italy, France, England, Spain, Russian Federation, USA, Canada, Brazil, Japan, China, Korea, and Australia
AL _T	Automobile average lifetime (average vehicle age)
α ₁	The amount of energy required in cold rolling process
α ₂	The amount of energy required in hot rolling process
α ₃	The amount of energy required in continuous casting process
α ₄	The amount of energy required in basic oxygen furnace process
α ₅	The amount of energy required in blast furnace process
α ₆	The amount of energy required in sintering/coking process
β ₁	The greenhouse gas emitted in cold rolling process
β ₂	The greenhouse gas emitted in hot rolling process
β ₃	The greenhouse gas emitted in continuous casting process
β ₄	The greenhouse gas emitted in basic oxygen furnace process
β ₅	The greenhouse gas emitted in blast furnace process
β ₆	The greenhouse gas emitted in sintering/coking process
AR _R	Automobile recycling efficiency
SR _E	Scrap recycling efficiency
ρ ₁	The amount of energy required in cold rolling process for secondary production
ρ ₂	The amount of energy required in hot rolling process for secondary production
ρ ₃	The amount of energy required in continuous casting process for secondary production
ρ ₄	The amount of energy required in electric arc furnace process for secondary production
λ ₁	The greenhouse gas emitted in cold rolling process for secondary production
λ ₂	The greenhouse gas emitted in hot rolling process for secondary production
λ ₃	The greenhouse gas emitted in continuous casting process for secondary production
λ ₄	The greenhouse gas emitted in electric arc furnace process for secondary production

The general mathematical formulation of stocks and flows according to Forrester [28] is as follows:

$$Stock(t) = \int_{t_0}^t [Inflow(t) - Outflow(t)]dt + Stock(t_0) \tag{1}$$

where t_0 is the initial and t is the final year considered; $Stock(t)$ is a mass accumulated in the system at the moment t of the period 2010–2050 due to influx $Inflow(t)$ and loss $Outflow(t)$.

$$nflow(t) = f(Stock(t), V(t), P); Outflow(t) = f(Stock(t), V(t), P) \tag{2}$$

where, $V(t)$ is an auxiliary variable in time t . For example, in the niobium model, the rate of niobium mining expressed as a flow, depends on the amount of extracted pyrochlore ore and world production of niobium. P represents input parameters such as amount of niobium produced by each supplier or capacity of producers (all input parameters are given in Appendix, Table A1). Table 2 presents all equations corresponding to the different types of variables given in Figure 2.

Table 2. Equations and calculation processes for different type of variables of niobium simulation model.

Variable	Equation	Type (Tonnes)
$Nb_{M-R}(t)$	$Nb_{MP_{Brazil}}(t) + Nb_{MP_{Canada}}(t) + Nb_{MP_{Other}}(t)$	Flow
$EM(t)$	$\int_{t_0}^t (Nb_{M-R}(t) - SGFP_{P-R}(t) - Nb_{OPP-R}(t))dt + EM(t_0)$	Stock
$SGFP_{P-R}(t)$	$EM(t) \times SGFCoeff$	Flow
$OPP_{P-R}(t)$	$EM(t) \times OPCoeff$	Flow
$SGF(t)$	$\int_{t_0}^t (Nb_{P-R}(t) - SGFP_{P-R}(t))dt + SGF(t_0)$	Stock
$Nb_{P-R}(t)$	$\min(P_{Nb} \times T_{S_ELV}(t), SGF)$	Flow
$T_{W-ELV}(t)$	$W_{Car} \times ELV_{number}(t)$	Auxiliary
$T_{S-ELV}(t)$	$T_{W-ELV}(t) \times P_S$	Auxiliary
$Nb_{ELV}(t)$	$\text{Delay}(T_{S_ELV}(t) \times P_{Nb}, AL_T, 0)^1$	Flow
$S_{Nb-ELV}(t)$	$\int_{t_0}^t (Nb_{ELV}(t) - Nb_{RS-R}(t) - W_{Landfill-R}(t))dt + S_{Nb-ELV}(t_0)$	Stock
$Nb_{RS-R}(t)$	$S_{Nb_ELV}(t) \times AR_R$	Flow
$W_{Landfill-R}(t)$	$S_{Nb_ELV}(t) \times SR_E$	Flow
$Nb_{R-R}(t)$	$Nb_{RS-R}(t) + W_{Landfill-R}(t)$	Auxiliary
$S_R(t)$	$\int_{t_0}^t (Nb_{RS-R}(t) + W_{Landfill-R}(t))dt + S_R(t_0)$	Stock

¹ Delay (Input, Delay Time, Initial Value).

Next, we present the structure of the simulation models at the different stages and the variables needed for the analysis of environmental sustainability of niobium supply chain.

Mining and Processing Stage

At present, the main source of the niobium is pyrochlore ore (containing both tantalum and niobium). Niobium exists as Nb_2O_5 in other ores, e.g., columbite, tantalite, microlite, tapiolite, ixiolite, wodginite, loparite, lueshite, latrappite, and euxenite [1]. In our model, we consider mining of pyrochlore due to its economic importance.

The initial flow of this model consists of the global production of mineral concentrates as well as the reserves of niobium. In the model, we take into account only the existing mines and their capacity reported by the United States Geological Survey (USGS) [5]. The supply sources of niobium are not diversified and the market is dominated by one leading supplier, Brazil, which is the source of more than 89% of world niobium production. Canada follows Brazil with around 10% of world production [2,37–39]. Three world leaders of ferroniobium production are two Brazilian companies, Companhia Brasileira de Metalurgia e Mineração (CBMM) and Mineração Catalão de Goiás, and Canadian enterprise IAMGOLD Corp (Niobec Mine). These companies collectively account for the majority of world production of niobium. Two main processes considered in niobium mining and processing stages are hydrofluoric acid dissolution and solvent extraction.

Energy consumption in the mining stage is assessed as follows:

$$E_{M_R}(t) = Nb_{M_R}(t) \times \sum_{n=1}^2 \sigma_n \quad (3)$$

where, $E_{M_R}(t)$ is energy consumption in the mining stage in the year $t = 2000, 2001, \dots, 2050$. $Nb_{M_R}(t)$ is the amount of material in the mining flow in the year $t = 2000, 2001, \dots, 2050$. σ_n is the energy (gigajoule) required per one tonne of niobium in the mining stage through each process $n = 1, 2$ which corresponds to the amount of energy required in hydrofluoric acid dissolution and solvent extraction.

The GHG emissions from the mining stage are estimated based on energy consumption in hydrofluoric acid dissolution and solvent extraction. They are assessed as follows:

$$GHG_{M_R}(t) = E_{M_R}(t) \times \sum_{n=1}^2 \gamma_n \quad (4)$$

where, $GHG_{M_R}(t)$ represents greenhouse gas emissions (CO₂-eq) for the mining stage in the year $t = 2000, 2001, \dots, 2050$. $E_{M_R}(t)$ is the energy consumption in the mining stage in the year $t = 2000, 2001, \dots, 2050$. γ_n is the greenhouse gas emitted from each process $n = 1, 2$ (hydrofluoric acid dissolution and solvent extraction).

Production Stage

Niobium is used in several different forms, such as standard grade ferroniobium (SGF)—primarily used in high-strength and stainless steels; vacuum grade ferroniobium (VGF)—used in superalloys production; niobium metal and alloys—used in superconductors production and niobium chemicals—used in special ceramics and chemical processes [1]. The majority of niobium is used in the form of SGF for HSS steels production, which accounts for around 90% of total niobium consumption [40]. Therefore, in the production stage, we assumed SGF production flow as the main input. Considering the significance of HSS steels in the automotive industry explained in the introduction, the presented model in this stage is focused on estimation of energy consumption and GHG emissions of using niobium in the HSS steels applied in the automotive industry. Typical passenger cars are considered in the model based on their weight and percentage of used HSS steel [14,41].

The different methods are used in production of HSS steel. In 2005, in the global steel industry, basic oxygen steelmaking furnaces (BOFs) accounted for approximately 65% of world steel production—China has the highest share of BOFs steel production; electric arc furnaces (EAFs) accounted for approximately 32%—the USA have the highest share of EAFs in steel production; open hearth furnaces (OHFs) production accounted for the remaining 3%—Ukraine has the highest OHFs steel production [42,43]. Therefore, in the present study, we assume BOFs as the main process of primary production of HSS steels.

In the production stage, six processes including cold rolling, hot rolling, continuous casting, basic oxygen furnace, blast furnace, and sintering/coking are considered. In the second submodel, energy consumption in the production stage is estimated as follows:

$$E_{P_R}(t) = Nb_{P_R}(t) \times \sum_{n=1}^6 \alpha_n \quad (5)$$

where, $E_{P_R}(t)$ is energy consumption in the production stage in the year $t = 2000, 2001, \dots, 2050$. $Nb_{P_R}(t)$ is the amount of material in the production flow in the year $t = 2000, 2001, \dots, 2050$. α_n is the energy (gigajoule) required per one tonne of niobium in the production stage through each process $n = 1, 2, \dots, 6$ which corresponds to six main processes mentioned above.

The GHG emissions from the production stage are estimated based on the energy consumption in cold rolling, hot rolling, continuous casting, basic oxygen furnace, blast furnace, and sintering/coking. In the third submodel, they are assessed as follows:

$$GHG_{P_R}(t) = E_{P_R}(t) \times \sum_{n=1}^6 \beta_n \quad (6)$$

where, $GHG_{P_R}(t)$ represents greenhouse gas emissions (CO₂-eq) for the production stage in the year $t = 2000, 2001, \dots, 2050$. $E_{P_R}(t)$ is energy consumption in the production stage in the year $t = 2000, 2001, \dots, 2050$. β_n is the greenhouse gas emitted from each process $n = 1, 2, \dots, 6$ (cold rolling, hot rolling, continuous casting, basic oxygen furnace, blast furnace, and sintering/coking).

Recycling Stage

Based on the amount of steel obtained from ELVs, the auto steel recycling rate was estimated to grow from 85% in 2007 to 95% in 2050 [44]. The scrap in ELVs is classified into one of three classes of remeltable; the same type of material (recycling), other material types (cascading) or loss to landfill [12]. After collection, the scrap is processed into a physical form and chemical composition enabling its use in steel mills. The scrap is melted in BOF or EAF. In the recycling of high-strength low-alloy steel, about 0.05% of niobium will be probably oxidised to the slag phase and lost during recycling to BOF or EAF [1,45]. We assume that EAF is the main process in this stage of the model [44].

In the recycling stage, four processes including cold rolling, hot rolling, continuous casting, and electric arc furnace are considered. Energy consumption in the recycling stage is estimated as follows:

$$E_{R_R}(t) = Nb_{R_R}(t) \times \sum_{n=1}^4 \rho_n \quad (7)$$

where, $E_{R_R}(t)$ is energy consumption in the recycling stage in the year $t = 2000, 2001, \dots, 2050$. $Nb_{R_R}(t)$ is the amount of material in recycling flow in the year $t = 2000, 2001, \dots, 2050$. ρ_n is energy (gigajoule) required per one tonne of niobium flow in the recycling stage through each process $n = 1, 2, \dots, 4$ which corresponds to the amount of energy required in cold rolling, hot rolling, continuous casting, and electric arc furnace.

The GHG emissions from the recycling stage are estimated based on energy consumption in cold rolling, hot rolling, continuous casting, and electric arc furnace. They are assessed as follows:

$$GHG_{R_R}(t) = E_{R_R}(t) \times \sum_{n=1}^4 \lambda_n \quad (8)$$

where, $GHG_{R_R}(t)$ represents greenhouse gas emissions (CO₂-eq) for the recycling stage in the year $t = 2000, 2001, \dots, 2050$. $E_{R_R}(t)$ is energy consumption in the recycling stage in the year $t = 2000, 2001, \dots, 2050$. λ_n is the greenhouse gas emitted from each process $n = 1, 2, \dots, 4$ (cold rolling, hot rolling, continuous casting, and electric arc furnace).

3. Validation of the Model

Validation determines whether a simulation model is an accurate representation of the actual system [46]. Validation consists in quantifying the accuracy of the model by comparison of numerical outputs of the model with experimental data [47].

The method proposed by Barlas [48] was used for validation of the proposed model. According to the model, the annual mean of niobium mined globally is 51,831 tonnes in years 2005–2015. This estimation compared to the global average amount of niobium mined annually (i.e., 47,855 tonnes for 2005–2015, based on USGS data sources) shows that the error of the model is 3%.

The amount of niobium needed for the primary production of HSS steels in the automotive industry was adjusted based on the demand of passenger cars market. Average lifetime of a car is assumed to be 16 years [49,50]. There were several reasons to adopt the long term perspective (2000–2050) in the simulation. One of the most important causes is a need to contain the delay mechanisms of the system. Moreover, one of the objectives of this study is to assess, in the longer perspective, the effect of number of ELVs on environmental sustainability of niobium supply chain. The base year of the analysis is 2010 due to the availability of data on ELVs numbers in the different countries [14] as well as obtainable historical data in the referenced period [12].

4. Results and Discussion

It is evident that environmental requirements may directly change recycling processes. The balance of energy consumption and GHG emissions from the supply chain helps to determine the environmental sustainability of recycling and the level of investment in recycling. To assess the environmental aspect of niobium recycling, the factors such as energy consumption, GHG emissions, and material flows were quantified and evaluated for all stages of the supply chain. Next, we present the simulation results along with a brief analysis of environmental assessment at different stages of niobium life cycle.

The correlation between energy consumption and GHG emissions is clearly visible in all stages of niobium supply chain. Figure 3a shows that in the mining stage, assuming energy used in hydrofluoric acid dissolution and solvent extraction, the annual mean of energy consumption from the extraction of niobium in years 2010–2050 is around 2.5 million gigajoules (m GJ). Considering six main processes (cold rolling, hot rolling, continuous casting, basic oxygen furnace, blast furnace, sintering/coking) the annual mean of energy used in years 2010–2050 in the production of HSS steels for the automotive industry is around 3 m GJ. In the production stage, energy consumption oscillates due to the dynamics of niobium flow caused by HSS steels demand. Considering energy required in cold rolling, hot rolling, continuous casting, and electric arc furnace processes, in the recycling stage of HSS steels from ELVs, the annual mean level decreases to around 0.3 m GJ of energy. Figure 3b shows that cumulative energy used in years 2010–2050 increases from 17 to 115 m GJ in mining, from 20 to 144 m GJ in production stage and from 0.7 to 11 m GJ, in recycling. It shows that the lowest average energy consumption takes place in recycling stage of niobium supply chain and both production and mining are the most energy consuming processes.

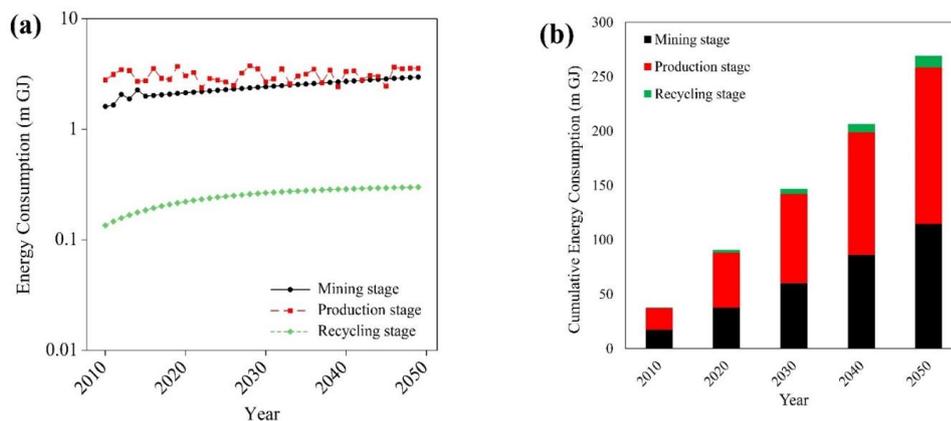


Figure 3. Energy consumption levels in different stages of niobium supply chain (2010–2050). (a) Annual energy consumption in different stages. (b) Cumulative energy consumption in different stages.

Figure 4 shows total GHG emissions from each stage of niobium lifecycle, focusing on Nb use in HSS steels for the automotive industry. Figure 4a shows that in the mining stage the annual mean of GHG emissions from the extraction of niobium in years 2010–2050 is more than 0.4 million tonnes (mt) CO₂-eq. The annual mean of GHG emissions in the production stage of HSS steels for the automotive industry is around 1 mt CO₂-eq in the recycling stage of HSS steels containing niobium from ELVs, the annual mean level decreases to less than 0.08 mt CO₂-eq of emissions. It shows the lowest average GHG emissions for all stages of niobium supply chain. Figure 4b illustrates the increase in cumulative GHG emissions in years 2010–2050. The increase is from 3 to 17 mt CO₂-eq in mining, from 7 to 47 mt CO₂-eq in production stage and from 0.3 to 3 mt CO₂-eq in recycling. As shown in Figure 4a,b, the amount of GHG emitted in production stage is much higher than in the other ones.

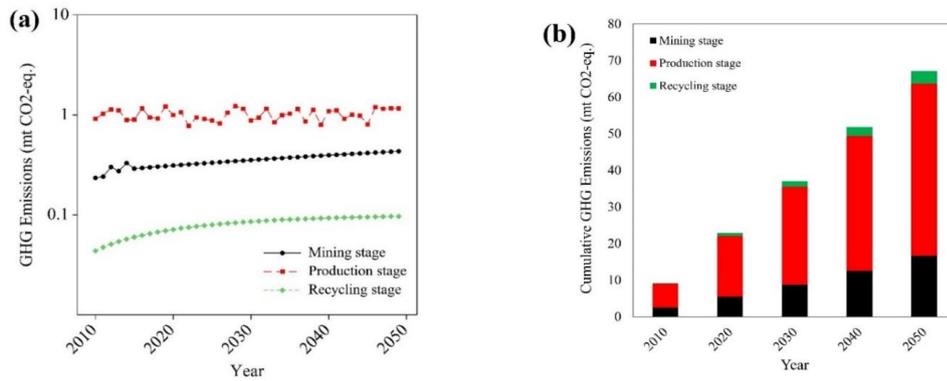


Figure 4. Greenhouse gas (GHG) emissions in different stages of niobium supply chain (2010–2050). (a) Annual GHG emissions in different stages. (b) Cumulative GHG emissions in different stages.

In the production and recycling stages, the number of ELVs is a primary variable affecting the energy demand and GHG emissions. The details are shown in Table 3. The analysis is limited to the countries with more than 10 million units of automobiles and ELVs in 2010 [14]. The obtained results of simulation show that the highest energy consumption and GHG emissions are accounted for USA, followed by EU, China and Japan, Table 3.

Table 3. Contribution of each country to the increase of energy consumption and GHG emissions in the each stage of the niobium life cycle in the automotive industry (2010–2050).

Country	Production Stage		Recycling Stage	
	Energy Consumption (m GJ)	GHG Emission (mt CO ₂ -eq.)	Energy Consumption (m GJ)	GHG Emission (mt CO ₂ -eq.)
European Union (EU-27)	24.2	7.9	1.9	0.6
Germany	1.5	0.5	0.1	0.0
Italy	5.0	1.6	0.4	0.1
France	4.9	1.6	0.4	0.1
England	3.6	1.2	0.3	0.1
Spain	2.6	0.8	0.2	0.1
USA	37.1	12.1	2.8	0.9
Canada	3.7	1.2	0.3	0.1
Brazil	3.1	1.0	0.2	0.1
Japan	9.2	3.0	0.7	0.2
China	10.8	3.5	0.8	0.3
Korea	2.1	0.7	0.2	0.1
Australia	1.5	0.5	0.1	0.0
Global	124.3	40.6	10.0	3.2

As mentioned in the introduction, previous studies show the growing demand for niobium steel production in the automotive industry. The increase in the share of steel produced from scrap is estimated to be 50–80% until 2050 as shown in several studies [51,52]. In addition, some works also identified the global growth of ELVs numbers and its impact on GHG emissions [12,53]. Due to the previously mentioned significance of a lightweighting strategy by using HSS steel in the automotive industry [11], the likelihood of the growth of niobium demand is quite high [54]. To address this issue, we used a scenario-based approach to identify the impact of the increase of ELVs, and in consequence global demand for niobium, on GHG emissions.

The statistical analysis for predicting the possible growth of numbers of automobiles and ELVs has been presented in other studies [12,50]. They assessed the total car stock from 1950 to 2050 by considering two main variables: Population and car ownership. The results show a possible increase of the number of cars and ELVs by 50–60%. In our work, we assumed the highest value of ELVs growth rate to be 50%. Three scenarios were considered for the growth rate of ELVs to show the changes in cumulative energy consumption and GHG emissions from primary and secondary production of HSS containing niobium in the automotive industry; scenario A, 10%; scenario B, 25%; and scenario C, 50% compared to the baseline scenario (number of ELVs in 2010).

Based on the data available for the reference year (2010), we carried out the analysis assuming the main sources of ELVs are the following countries: Australia, Korea, China, Japan, Brazil, Canada, the USA, Spain, England, France, Italy, Germany and the European Union (EU-27). Figure 5 shows that for the case of 50% increase in the number of ELVs, the estimated GHG emissions from primary production of HSS steels containing niobium will increase by 20% in 2050. While this increase in secondary production would be between 16% and 24% in 2050. Table 4 provides the details of the reduction of energy consumption and GHG emissions for each country by using niobium in secondary production of HSS steel in the automotive industry from 2010 to 2050. Globally, around 133–161 m GJ energy could be saved by using Nb in secondary production of HSS steel in the automotive industry. It also contributes to 44–53 mt CO₂-eq, mainly in the USA, followed by the EU, China and Japan.

Figure 6a shows cumulative energy use in mining stage increases from 17 m GJ to 115 m GJ between 2010 and 2050. In production stage, it increases from 20 m GJ to 144 m GJ and in recycling stage, it increases from 0.6 m GJ to 11 m GJ in the same period. These results prove the importance of recycling for saving energy over a long-term period. On average, energy demand in recycling is around 19% of total energy consumption of niobium supply chain. While, production and mining stage requires about 45% and 36%, respectively. The results in Figure 6b show that cumulative global GHG emissions in the mining stage increase from 2.5 mt CO₂-eq in 2010 to 17 mt CO₂-eq in 2050. It is, on average, four times more than GHG emissions in recycling in any scenario. Globally, GHG emissions in the recycling of HSS steel containing niobium will increase from around 0.2–0.3 mt CO₂-eq in 2010 to around 3–5 mt CO₂-eq in 2050 corresponding to 10–50% increase in ELVs number. It shows around 18% reduction of annual emissions between 2010 and 2050 thanks to the reuse of niobium in secondary production rather than primary one. Considering 50% increase in demand, the estimated GHG emissions in the production stage will increase to the highest level of all stages. On average, the production stage accounts for 72% of total emissions followed by mining with 21% and recycling accounted for 7%.

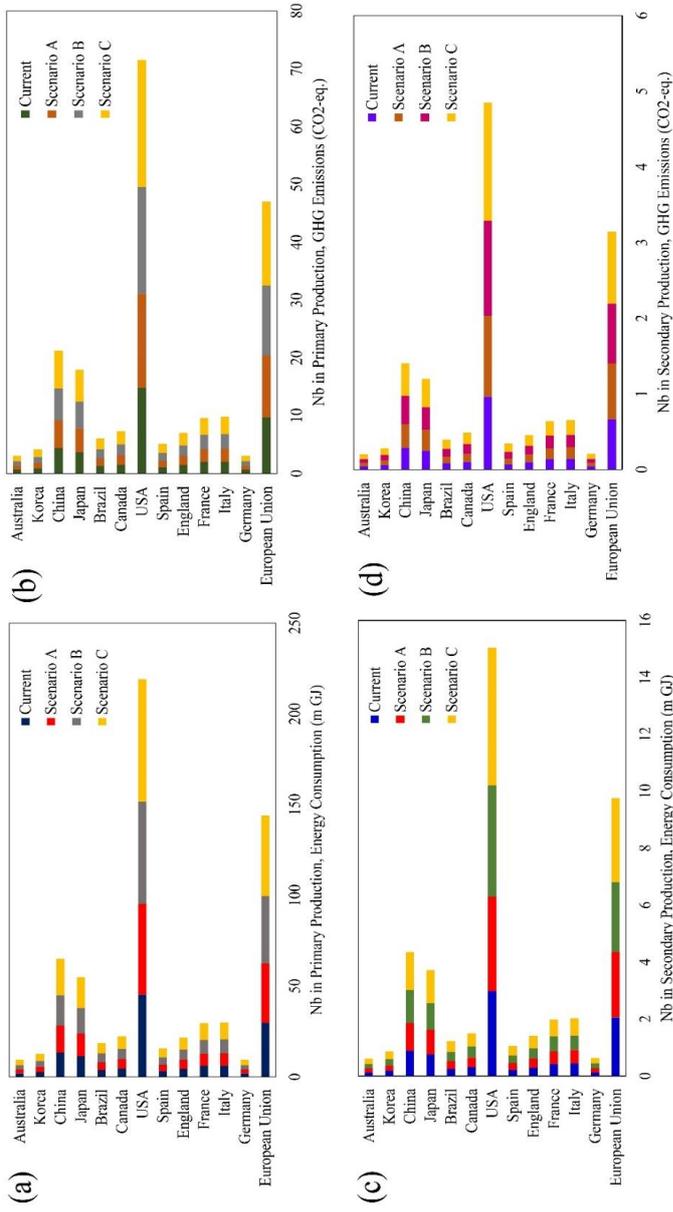


Figure 5. Cumulative energy consumption and greenhouse gas emissions from primary and secondary production of high-strength steel (HSS) containing niobium in automotive markets in the different countries (2010–2050). **(a)** Energy consumption in production stage. **(b)** GHG emissions in production stage. **(c)** Energy consumption in recycling stage. **(d)** GHG emissions in recycling stage.

Table 4. Reduction of energy consumption and greenhouse gas emissions, applying Nb in secondary production of HSS steel in the automotive industry (2010–2050).

	Scenario	European Union											Global		
		European Union	Germany	Italy	France	England	Spain	USA	Canada	Brazil	Japan	China		Korea	Australia
Energy Consumption (m GJ)	Current	27.7	1.8	5.8	5.7	4.2	3.0	42.4	4.3	3.6	10.6	12.5	2.5	1.8	133.3
	A	30.4	2.0	6.3	6.2	4.6	3.3	46.5	4.7	4.0	11.6	13.8	2.7	2.0	145.6
	B	34.7	2.2	7.2	7.1	5.2	3.8	52.6	5.4	4.5	13.2	15.6	3.1	2.3	155.6
	C	41.5	2.7	8.7	8.5	6.2	62.6	6.5	5.4	15.8	18.8	3.7	2.7	161.4	
GHG Emissions (mt CO ₂ -eq.)	Current	9.1	0.6	1.9	1.9	1.4	1.0	13.9	1.4	1.2	3.5	4.1	0.8	0.6	43.6
	A	9.9	0.6	2.1	2.0	1.5	1.1	15.2	1.6	1.3	3.8	4.5	0.9	0.6	47.6
	B	11.3	0.7	2.4	2.3	1.7	1.2	17.2	1.8	1.5	4.3	5.1	1.0	0.7	50.9
	C	13.6	0.9	2.8	2.8	2.0	20.5	2.1	1.8	5.2	6.1	1.2	0.9	52.7	

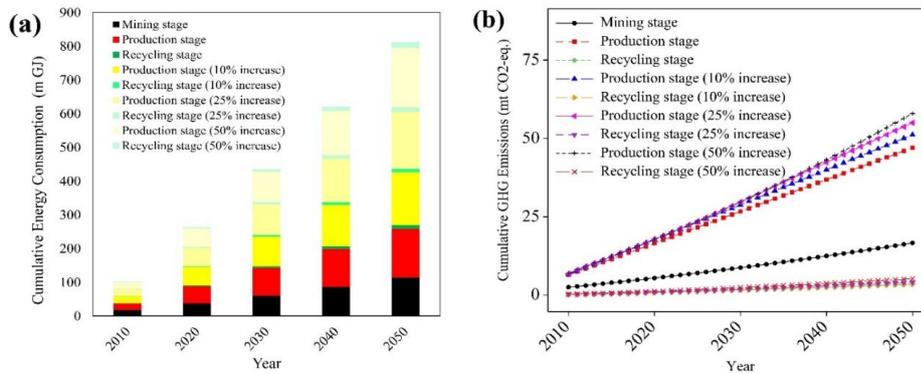


Figure 6. Global energy consumption and greenhouse gas (GHG) emissions in different stages of niobium supply chain (2010–2050). (a) Cumulative energy consumption in different scenarios. (b) Cumulative GHG emissions in different scenarios.

This study presents a holistic view of the supply chain of niobium. The analysis carried out in previous publications had not offered insight into energy consumption and GHG emissions in the different stages of niobium supply chain. Moreover, the previous research has been focused on material flow in mining, production or recycling without showing the dynamics of their mutual relations. It means the previous research considered only one stage of supply chain focusing on supply and demand of material or assessment of various technologies.

The environmental requirements will force countries to introduce new laws and drive the automotive industry towards more sustainable use of resources. In this context, the presented study should be very useful in delivering long-term estimates of environmental performance of niobium supply chain. The results presented in this paper show how niobium recycling can lead to the reduction of annual and cumulative emissions and save energy consumption in a long-term perspective.

5. Limitations of the Study

One of the limitations of this study is a lack of tantalum analysis, which is an important by-product in the niobium value chain. This limitation results from restricting of the presented model to the use of niobium in HSS steel for the automotive industry. Another limitation of this study is the lack of the economic assessment of technologies for primary and secondary production of niobium.

6. Conclusions

This article presents an initial attempt to use dynamic simulation model of niobium supply chain. The dynamic model allows for comprehensive description and analysis of the niobium supply chain in a long-term perspective considering the environmental protection policies related to the reduction of energy consumption and mitigation of emissions. The results show energy demand and GHG emissions in different stages of niobium life cycle—mining, production and recycling.

The results indicate that mining requires 36% of total energy demand and is responsible for 21% of total emissions in niobium supply chain over the period 2010–2050. The production stage consumes around 45% of energy and contributes to 72% of total emissions. The energy used in the recycling constitutes about 19% of total energy demand and generates only 7% of total emission in niobium supply chain. This result highlights the potential benefit of recycling in saving energy and reducing emissions. Globally, the recycling of niobium could save around 133–161 m GJ energy between 2010 and 2050. It also could lead to the reduction of emissions by 44–53 mt CO₂-eq in the same period. The analysis of the results in different countries shows that the highest impact would be observed

in the USA, EU, China and Japan. It shows a possibility to reduce annual emissions by 18% in years 2010–2050 thanks to the reuse of niobium in secondary production rather than primary one.

It should be noted that due to the ever-changing dynamics of material flows, further research should update the findings of this study in the near future by considering the newest technologies emerging in every stage of the supply chain.

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Appendix A

This appendix contains a list of parameters, description, their units, initial values of the parameters, time and sources of data.

Table A1. Data sources for all inputs of the proposed model.

Variable/Parameter	Description	Unit	Value Range	Time	Data Sources
N _{bMP-Brazil} (t)	World production of mineral concentrates (niobium content) by Brazil	Tonnes	21,800–101,022	2000–2015	US Geological Survey (USGS). Available at: https://minerals.usgs.gov/minerals/pubs/commodity/niobium/
N _{bMP-Canada} (t)	World production of mineral concentrates (niobium content) by Canada	Tonnes	2280–5774	2000–2015	US Geological Survey (USGS). Available at: https://minerals.usgs.gov/minerals/pubs/commodity/niobium/
N _{bMP-Other} (t)	World production of mineral concentrates (niobium content) by other countries	Tonnes	89–853	2000–2015	US Geological Survey (USGS). Available at: https://minerals.usgs.gov/minerals/pubs/commodity/niobium/
R _{Brazil} (t)	Reserves in Brazil	Tonnes	3,300,000–4,100,000	1996–2017	US Geological Survey (USGS). Available at: https://minerals.usgs.gov/minerals/pubs/commodity/niobium/niobimcs96.pdf https://minerals.usgs.gov/minerals/pubs/commodity/niobium/mcs-2017-niobi.pdf
R _{Canada} (t)	Reserves in Canada	Tonnes	140,000–200,000	1996–2017	US Geological Survey (USGS). Available at: https://minerals.usgs.gov/minerals/pubs/commodity/niobium/niobimcs96.pdf https://minerals.usgs.gov/minerals/pubs/commodity/niobium/mcs-2017-niobi.pdf
C _{1-Brazil} (t)	One of the leading niobium ore and concentrate producers: Companhia Brasileira de Metalurgia e Mineração (CBMM) in Brazil	Tonnes	19,500–150,000	1991–2016	US Geological Survey (USGS). Available at: https://minerals.usgs.gov/minerals/pubs/commodity/niobium/230494.pdf https://minerals.usgs.gov/minerals/pubs/commodity/niobium/myb1-2014-niobi.pdf
C _{Canada} (t)	One of the leading niobium ore and concentrate producers: IAMGOLD Corporation (Niobec Mine) in Canada	Tonnes	3300–5480	1994–2014	US Geological Survey (USGS). Available at: https://minerals.usgs.gov/minerals/pubs/commodity/niobium/230494.pdf https://minerals.usgs.gov/minerals/pubs/commodity/niobium/myb1-2014-niobi.pdf

Table A1. *Cont.*

Variable/Parameter	Description	Unit	Value Range	Time	Data Sources
$C_{2-brazil}$ (t)	One of the leading niobium ore and concentrate producers: Mineração Catalão de Goiás in Brazil	Tonnes	3550–4700	1995–2014	US Geological Survey (USGS). Available at: https://minerals.usgs.gov/minerals/pubs/commodity/niobium/230495.pdf https://minerals.usgs.gov/minerals/pubs/commodity/niobium/myb1-2014-niobi.pdf
SGF_{Coef}	Percentage of global niobium production used to produce ferriobium used in high strength low alloy steels	%	0.89	2011	British Geological Survey's Centre for Sustainable Mineral Development MineralsUK. Mineral Profiles. Niobium and Tantalum. Available at: http://www.bgs.ac.uk/downloads/start.cfm?id=2033
OP_{Coef}	Percentage of global niobium production used in manufacture of niobium alloys, niobium chemicals and carbides, high purity ferriobium, and other niobium metal products	%	0.11	2011	British Geological Survey's Centre for Sustainable Mineral Development MineralsUK. Mineral Profiles. Niobium and Tantalum. Available at: http://www.bgs.ac.uk/downloads/start.cfm?id=2033
δ_1	Energy usage through hydrofluoric acid dissolution process	GJ (tonne ore) ⁻¹	2	2003	National Institute of Materials Science (estimation of CO ₂ emission and energy consumption in extraction of metals) http://www.nims.go.jp/genso/0ej0070000039eq-att/0ej0070000039j5.pdf
δ_2	Energy usage through solvent extraction process	GJ (tonne ore) ⁻¹	31.4	2003	National Institute of Materials Science (estimation of CO ₂ emission and energy consumption in extraction of metals) http://www.nims.go.jp/genso/0ej0070000039eq-att/0ej0070000039j5.pdf
γ_1	CO ₂ emission through hydrofluoric acid dissolution process	CO ₂ -eq.	1.8	2003	National Institute of Materials Science (estimation of CO ₂ emission and energy consumption in extraction of metals) http://www.nims.go.jp/genso/0ej0070000039eq-att/0ej0070000039j5.pdf

Table A1. *Cont.*

Variable/Parameter	Description	Unit	Value Range	Time	Data Sources
γ_2	CO ₂ emission through solvent extraction process	CO ₂ -eq.	4.6	2003	National Institute of Materials Science (estimation of CO ₂ emission and energy consumption in extraction of metals) http://www.nims.go.jp/genso/0ej0070000039eq-att/0ej0070000039j5.pdf [1,55]
P_{Nb}	Nb grade in HSS ferromiobium applied in automobiles	%	0.04–0.08, 0.1	2011–2017	PROMETIA, Factsheet available at: http://prometia.eu/wp-content/uploads/2014/02/NIOBIUM-TANTALUM-v02.pdf [56]
P_S	Steel in Automobile	%	61.7	2012	[12]
W_{Car}	Weight of Car	Tonne	1.11361–1.49131	1950–2010	[12]
ELV _{number} (t)	End-of-life vehicles (ELV) including countries: European Union, Germany, Italy, France, England, Spain, Russian Federation, USA, Canada, Brazil, Japan, China, Korea, and Australia	Units year ⁻¹	European Union:7,823,211 Germany:500,193 Italy:1,610,137 France:1,583,283 England:1,157,438 Spain:839,637 USA:12,000,000 Canada:1,200,000 Brazil:1,000,000 Japan:2,960,000 China:3,506,000 Korea:684,000 Australia:500,000 Global total:40,176,051	2010	[14]
AL_T	Automobile Average Life Time (Average Vehicle Age)	Year	16	2007–2014	[12,49,50,57]
α_1	The amount of energy required in cold rolling process	GJ tonne ⁻¹	1.63–1.935	1999–2012	[12,58]

Table A1. *Cont.*

Variable/Parameter	Description	Unit	Value Range	Time	Data Sources
α_2	The amount of energy required in hot rolling process	GJ tonne ⁻¹	1.7–1.88	1999–2012	[12,58–60]
α_3	The amount of energy required in continuous casting process	GJ tonne ⁻¹	0.076	1999–2012	[12]
α_4	The amount of energy required in basic oxygen furnace process	GJ tonne ⁻¹	0.4	1999–2012	[12,59]
α_5	The amount of energy required in blast furnace process	GJ tonne ⁻¹	12.3–16	1999–2012	[12,59,60]
α_6	The amount of energy required in sintering/coking process	GJ tonne ⁻¹	43.8	1999–2012	[12,61]
β_1	The greenhouse gas emitted in cold rolling process	Tonne CO ₂ -eq.	0.008	2013	[12,60]
β_2	The greenhouse gas emitted in hot rolling process	Tonne CO ₂ -eq.	0.082	2013	[12,60]
β_3	The greenhouse gas emitted in continuous casting process	Tonne CO ₂ -eq.	0	2013	[12,60]
β_4	The greenhouse gas emitted in basic oxygen furnace process	Tonne CO ₂ -eq.	0.09	1999–2012	[12,61,62]
β_5	The greenhouse gas emitted in blast furnace process	Tonne CO ₂ -eq.	1.22–1.46	1999–2012	[12,60–62]
β_6	The greenhouse gas emitted in sintering/coking process	Tonne CO ₂ -eq.	0.43	1999–2012	[12]
ARR	Automobile Recycling Rate	%	85	1998–2013	US Geological Survey (USGS), Flow Studies for Recycling Metal Commodities in the United States. Available at: https://pubs.usgs.gov/circ/2004/1196am/c1196a-m_v2.pdf ISRI. Available at: http://www.isri.org/docs/default-source/recycling-industry/fact-sheet---iron-and-steel.pdf?sfvrsn=16

Table A1. *Cont.*

Variable/Parameter	Description	Unit	Value Range	Time	Data Sources
SR _E	Automobile Scrap recycling efficiency	%	50	1998	US Geological Survey (USGS). Flow Studies for Recycling Metal Commodities in the United States. Available at: https://pubs.usgs.gov/circ/2004/1196am/c1196a-m_v2.pdf
ρ_1	The amount of energy required in cold rolling process for secondary production	GJ tonne ⁻¹	1.63–1.935	1999–2012	[12,58]
ρ_2	The amount of energy required in hot rolling process for secondary production	GJ tonne ⁻¹	1.7–1.88	1999–2012	[12,58–60]
ρ_3	The amount of energy required in continuous casting process for secondary production	GJ tonne ⁻¹	0.076	1999–2012	[12]
ρ_4	The amount of energy required in electric arc furnace process for secondary production	GJ tonne ⁻¹	2.5–2.8	1999–2012	[12,58–60]
λ_1	The greenhouse gas emitted in cold rolling process for secondary production	Tonne CO ₂ -eq.	0.008	2013	[60]
λ_2	The greenhouse gas emitted in hot rolling process for secondary production	Tonne CO ₂ -eq.	0.082	2013	[60]
λ_3	The greenhouse gas emitted in continuous casting process for secondary production	Tonne CO ₂ -eq.	0	2013	[12,60]
λ_4	The greenhouse gas emitted in electric arc furnace process for secondary production	Tonne CO ₂ -eq.	0.06–0.09	2006–2011	[42,63]

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Publication IV

Rahimpour Golroudbary, S., Calisaya-Azpilcueta, D. and Kraslawski, A.
**The Life Cycle of Energy Consumption and Greenhouse Gas Emissions from Critical
Minerals Recycling: Case of Lithium-ion Batteries**

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The Life Cycle of Energy Consumption and Greenhouse Gas Emissions from Critical Minerals Recycling: Case of Lithium-ion Batteries

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Abstract

Sometimes the applications of lithium-ion batteries (LIBs) are labeled as “zero emissions”. However, the emissions generated in the procurement and production stage of supply chain is not considered. Battery production is one of the main contributors to emitting greenhouse gas (GHG) emissions through electric vehicle (EV) manufacturing. In this case, recycling of LIBs is recommended to reduce energy consumption and mitigate GHG emissions as well as result in considerable natural resource saving compared to landfill. Also, accelerating production of LIBs in the line of clean-energy technologies has led to a sharply increasing criticality of minerals such as lithium (Li), cobalt (Co) and manganese (Mn). The spent LIBs could consider the secondary source of these minerals. The environmental sustainable way of recovering critical minerals from this waste is very important. Therefore, the primary aim of this paper is to answer the question if recycling of LIBs to recover the mentioned critical minerals is an environmentally sustainable option. To address this question, two aspects are analyzed: energy consumption and GHG emissions. These aspects were analyzed through a dynamic simulation model based on the principles of the system dynamics methodology. We provide an environmental analysis of recycling of critical minerals from spent LIBs including LMO, lithium manganese oxide; LCO, lithium cobalt oxide; LFP, lithium iron phosphate; NMC, lithium nickel manganese cobalt oxide; and LiNCA, Lithium nickel cobalt aluminum oxide. The results show that recycling of LIBs helps to prevent the shortage of critical minerals from a mass flow perspective. However, from an environmental perspective, the current technology is not recommended to recover lithium from LIBs which leads 38–45% more consumption of energy and 16–20% higher air emissions than its primary production.

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Keywords: Critical minerals; recycling; energy consumption; greenhouse gas emissions; dynamic model; Lithium-ion batteries

1. Introduction

Lithium-ion batteries (LIBs) are increasingly attractive due to their application in electric vehicles (EV) to reduce greenhouse gas (GHG) emissions and decrease dependence on oil. LIBs consider the best technology for sustainable transport in the current battery technologies for a variety of reasons such as high energy and power per unit battery as well as they are lighter and smaller than other relevant battery types [1]. Their

use is expected to double from 2014 to 2020 [2]. The main application of LIBs is projected for plug-in and full-battery EV. LIBs are composed of cathode, anode, binder, electrolyte and battery manage system. Most of the cathodes have critical minerals in their composition such as lithium (Li), cobalt (Co), and manganese (Mn) [3]. The importance of the elements is variable for different type of batteries. For example, around 35% of lithium production and 48% of cobalt production are consumed in the batteries manufacturing. Even though

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manganese application in batteries is not high, it is an essential element in the production of cathodes such as lithium manganese oxide (LMO) and lithium nickel manganese cobalt oxide (NMC). In this study, we consider different types of LIBs including LMO, NMC, lithium cobalt oxide (LCO), lithium iron phosphate (LFP), and lithium nickel cobalt aluminum oxide (LiNCA).

While the importance of the clean energy continues growing, this could cause adverse consequences including unsustainable consumption and criticality of metals such as Li, Co and Mn. Considering unsustainable consumption of metals, significant pressure is imposed on the supply side of Li [4]. The shortage of Li is predicted between 2021 and 2023 if Li is not recycled [5]. A range of studies has analyzed the impact of the penetration of batteries on future Li availability as the main driver in the production of LIBs [6,7]. It is predicted that Li required for the production of LIBs will face a severe shortage in the foreseeable future [8]. Given the criticality of the mentioned metals, they have been cataloged as critical minerals/materials in a variety of assessments by different methods [3,9]. In this case, recycling of spent LIBs is important as it can recover these valuable metals. This recycling is mainly crucial from the economic and environmental perspectives [1]. Moreover, there is an essential need for the improvement of the waste management systems internationally and the development of new recycling systems for Li [4]. Several problems in Li recycling, particularly from LIBs, were discussed in other research. For example, an increase in the lifetime of automotive leads the amount of material in the stock in use increases with a small flow of end of life LIBs per year [10]. Also, the complex chemical interaction between different materials requires high effort economic and energy consumption [11].

On the other hand, from an environmental perspective, the recycling of LIBs is important in order to avoid the hazardous results from heavy metals included the post-consumer products. Therefore, the significance behind Li recycling comes from not only to conserve resources but to avoid environmental burdens [12]. Sometimes the applications of batteries are labeled as “zero emissions”. However, according to Dunn [13], the emissions generated in the production stage of the supply chain is not considered. Battery production is one of the main contributors to emitting GHG through EV manufacturing [14]. Therefore, it is of crucial importance for a supply chain to have a detailed analysis of the life cycle of energy consumption and GHG emissions from mining to recycling.

Keeping in mind that ecological issues are primary drivers to select between either virgin ores or recycled materials [15], decision-makers need to be aware of possible undesirable environmental effects at each stage of the supply chain. For example, the rapid growth of demand for LIBs for the transportation and industrial application has led to increasing concerns about the environmental impact of their production [16]. Environmental issues could affect the supply security of materials due to potential environmental regulations and strong certification requirements [17].

Moreover, the reduction of environmental issues such as energy consumption and GHG emissions through supply chain helps industries to get a significant competitive advantage and

better public image [18]. However, we should consider that the reduction of emissions in a given stage of the supply chain may prove detrimental by increasing emissions in the other stages, e.g. through the utilization of more emission intensive materials [19]. The necessity of assessment of specific modifications through the supply chain in a longer perspective needs a systems approach. It consists of the detailed analysis of dynamics through all stages of the supply chain.

The numerous aspects of Li life cycle have already been studied in the recent years [20]. However, a comprehensive picture of the life cycle of energy consumption and GHG emissions from different critical minerals and their change over time has not yet been provided. Several research carried out an environmental assessment of different types of LIBs. Mainly, they have a product perspective and focus only on one stage of the supply chain, e.g. on production stage [21] or recycling stage [1,22]. The lack of adequate systemic environmental analysis for critical minerals recycling enhances the risk of environmental damage created by their supply chain. This indicates that a more comprehensive understanding of the global minerals cycle is necessary.

Therefore, the main objective of this paper is to present a dynamic model of the lithium supply chain to investigate energy consumption and GHG emissions at each stage including mining, production and recycling. In addition, it analyzes how much recovery of critical minerals including lithium, cobalt and manganese from spent LIBs may contribute to save energy and mitigate emission.

2. Materials and Methods

2.1. Data collection

A group of data sources was taken into account to run the model. In the mining stage, the impact to the environment was considered from the brine and ore processes. In this stage, the United States Geological Survey (USGS) [23], the Center for Energy Economics (CEE) [24] and the National Institute of Materials Science (NIMS) [25] as the primary data sources were used in the model. In the production stage, the impact to the environment was assumed from two steps including the cathode production and the assembling of batteries. In this stage, the Deutsche bank [26] and the GREET-2017 software were used as the main data sources. In the recycling stage, the impact to the environment was considered from two metallurgical processes including pyrometallurgy and hydrometallurgy. In this stage, the Karlsruher Institut für Technologie (KIT) [27] and the report from the European Union Commission [28] were used as the main data sources.

2.2. Dynamic model of lithium supply chain

The need for systematic analysis of Li supply chain has been presented by previous research [29]. The dynamic of Li material flows were discussed by Ziemann et al. (2018) [30] and Sverdrup (2016) [31]. Therefore, system dynamics methodology introduced by Forrester in the 1960s [32] is used in this study. System dynamics modeling is useful to show trends of variables in the given system as a decision support

tool [33]. System dynamics has been applied to various policy for natural resources and environmental problems of the supply chain. For example, in the case of critical materials, system dynamics models have been used in different sectors and cases including materials such as indium [34], platinum group metal [35], uranium [36], niobium [37] and tantalum [38] and phosphorus [39]. Considering lithium, a few studies used system dynamics method to examine dynamic interactions among various components of lithium supply chain, e.g. Sverdrup (2016) [31] analyzed market price and available extractable amounts of lithium globally and Idjisi et al. [40] assessed the issues of recycling lithium battery. The presented models in earlier publications lack the environmental feedback structure expected from a system dynamics model.

2.3. The system under study

The system considered in this paper focuses on the complex inter-relationships between mining & processing, production and recycling stages of lithium global supply chain applied in LIBs. Figure 1 shows the basic construction of our model through the causal loop diagram. Causal loop diagram assists as initial drafts of causal hypotheses throughout the model development. A causal link shows how the elements in the system influence each other. The positive (+) or negative (-) signs next to the arrowhead indicate whether the connected elements change in the same or opposite direction. As it is shown in Figure 1, there are four balancing feedback loops presented by *B* sign. A balancing loop displays a goal-seeking behavior. The first balancing loop includes mining of critical minerals, processing of brine or ore, manufacturing of LIBs, waste generation, collection of wastes, recycling of used LIBs and recovery of critical minerals. The second balancing loop includes processing of brine or ore, manufacturing of LIBs, demand from market and demand from industry. The third balancing loop includes manufacturing of LIBs and demand from market. Finally, the fourth balancing loop includes processing of brine or ore and demand from industry.

By identifying the key variables and their interactions throughout the different life cycle stages of Li, a primary structural model is built and simulated. The system under study incorporates the following activities: mining, processing, manufacturing, product use, waste management including a collection of used products and recycling. Particularly, production of LIBs for different industries such as EV, energy storage system, traditional batteries, and two wheels electronic vehicle is analyzed in the model to quantify energy consumption and GHG emissions.

Based on the system under study, each stage of the model consists of three main layers, including material and energy flow as well as GHG emissions. The material flow is composed of the following modules: suppliers, mining, processing, battery manufacturing, battery use, battery collection, battery recycling and critical minerals recovery. The energy consumption includes energy consumed in different stages: mining & processing, production and recycling. The GHG emissions is primarily related to the energy consumption and therefore the structure of both subsystems is identical. Since all the lithium extracted in the world is not used in batteries, the

share of the material flow that goes to the production of LIBs is calculated from the mining and processing stages. Then, in the production stage, different type of batteries including LMO, LCO, LFP, NMC and LiNCA were considered. The manufacturing part is divided into the cathode production and the assembly of the battery. In the final stage, the collected batteries and their recycling processes are considered. In this stage, the amount of the critical minerals contained in the batteries and the percentage of their recovery are considered.

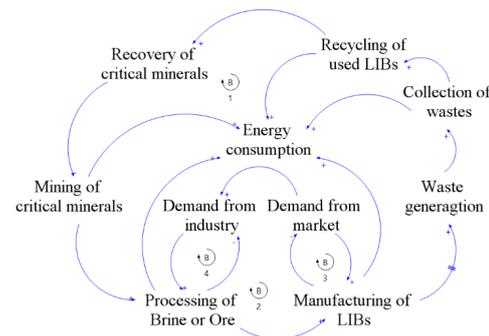


Fig. 1. Causal loop diagram for critical minerals recovery from LIBs market.

2.4. Mathematical formulation

The dynamic behavior of the lithium supply chain is implemented by a set of mathematical equations. Here we present the main formulas used in calculating material and energy consumption flow as well as GHG emissions.

According to the principles of system dynamics simulation, the model includes three types of elements [41]. The first element is a stock (or level) variable that is a reservoir of a given resource. The dynamic behavior of the level variables are given by a time integral of the net inflows minus the net outflows. The second element is a flow variable that adjusts the level of stock through inbound and outbound flows. The third element is an intermediate variable (auxiliary) consists of functions of stocks (and constants or exogenous inputs).

The general mathematical formulation of stocks according to Forrester [32] is as follows:

$$Stock(t) = \int_{t_0}^t [Inflow(t) - Outflow(t)]dt + Stock(t_0) \quad (1)$$

Where t_0 is the initial and t is the final year considered; $Stock(t)$ is a mass accumulated in the system at the year t of the period 2015-2025 due to influx $Inflow(t)$ and loss $Outflow(t)$.

$$Inflow(t) = f(Stock(t), V(t), P);$$

$$Outflow(t) = f(Stock(t), V(t), P) \quad (2)$$

Where, $V(t)$ is an auxiliary variable in time t . For example, in proposed model, the brine process as flow depends on the amount of extracted brine and the stock of global supply of lithium. P represents input parameters.

Energy consumption in the different processes is assessed as follows:

$$E_f(t) = M_f(t) \times \sum_1^n \sigma_n \quad (3)$$

Where, $E_f(t)$ is the flow of energy consumption in different processes such as mining, processing, production and recycling in the year $t = 2015, 2016, \dots, 2025$. $M_f(t)$ is the amount of material in the flow in the year t . σ_n is the energy (gigajoule) required per one tonne of LCE through each process from n type of energy sources ($n = 1, 2, \dots, 8$). Different types of energy sources includes non fossil fuel, coal fuel, natural gas fuel, fossil fuel, petroleum fuel, renewable, nuclear and biomass.

The GHG emissions are estimated based on energy consumption in each process. They are assessed as follows:

$$GHG_f(t) = E_f(t) \times \sum_1^n \gamma_n \quad (4)$$

Where, $GHG_f(t)$ represents the flow of greenhouse gas emissions ($\text{CO}_2\text{-eq}$) for each process in the year $t = 2015, 2016, \dots, 2025$. $E_f(t)$ is the flow of energy consumption in different processes such as mining, processing, production and recycling in the year t . γ_n is the greenhouse gas emitted from each process by consuming different type of energy sources $n = 1, 2, \dots, 8$ (non fossil fuel, coal fuel, natural gas fuel, fossil fuel, petroleum fuel, renewable, nuclear and biomass).

3. Results and discussion

The balance of energy consumption and GHG emissions from the supply chain policies help to determine the environmental sustainability of recycling and the level of investment in recycling. To assess the environmental aspect of critical minerals recycling from LIBs, the factors such as energy consumption, GHG emissions, and material flows were quantified and evaluated for all stages of the supply chain. Next, we present the simulation results along with a brief discussion for environmental assessment of different stages.

Figure 2 shows the results correspond to the mining stage. Annual mass flow of lithium through processing of brine and ore as well as the amount of $\text{CO}_2\text{-eq}$ emitted in each year (2015-2025) are presented in Figure 2 (a). The demand for lithium encourages mining to increase the amount of lithium extracted. This means that the impact generated by mining on the environment increases proportionally, as long as the same technologies are considered for the exploitation of lithium. The results show that the production of Li applying in battery industries is increasing, which reaches more than 450,000 tonnes LCE in 2025. As a consequence, the cumulative energy consumption and GHG emissions through the mining stage are increasing from 2015 to 2025, as shown in Figure 2 (b,c). Due to the unit operations needed in the ore process to extract the lithium, this process consumes more energy and emit more $\text{CO}_2\text{-eq}$ in compare to brine process.

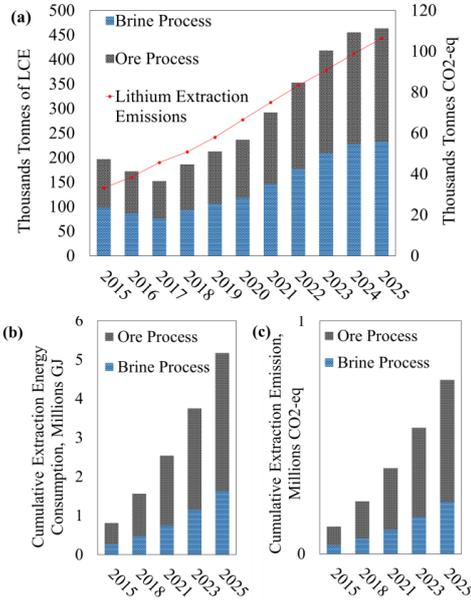


Fig. 2. Environmental impact of mining stage from 2015 to 2025. (a) Annual; (b) Cumulative energy consumption; (c) Cumulative emissions.

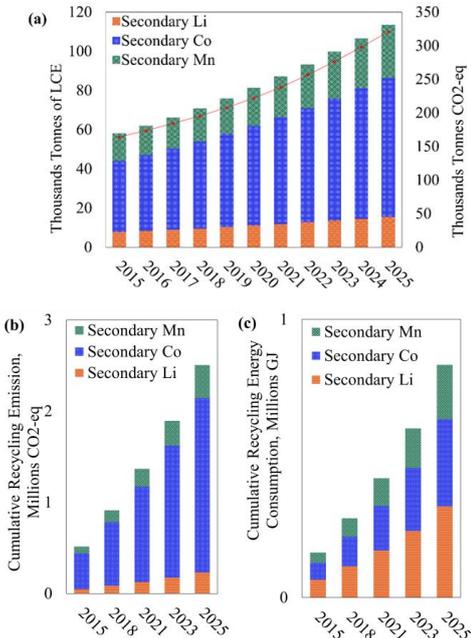


Fig. 3. Environmental impact of production stage from 2015 to 2025. (a) Annual; (b) Cumulative energy consumption; (c) Cumulative emissions.

The second stage of the model corresponds to the production of LIBs. Due to the different impact of process on the environment, we analyze the production of cathodes and assembling of the batteries, separately. In the production of cathodes, five different LIBs are considered: LMO, LCO, LFP, NCM and LiNCA. Figure 3 shows the results regarding the LIBs production. As shown in Figure 3 (a), the most produced cathodes are in order as follows: NCM, LCO, LFP, LiNCA and LMO. In Figure 3 (b,c), the cumulative of energy consumption and GHG emissions are presented from 2015 to 2025. Regarding energy consumption, the cathode production that requires more energy is LCO (around 7.8 million (m) GJ in 2025). LiNCA requires the less energy among cathodes, with a cumulative value around 0.8 m GJ. Regarding GHG emissions, the cathode production that emits more CO₂-eq is NMC, with a cumulative value of around 0.44 million tonnes (mt) CO₂-eq. LMO production emits less emission, around 0.1 mt CO₂-eq.

The third stage of the model corresponds to recycling of LIBs. Physical process is usually used to obtain different streams of waste materials. Then, in chemical process a metallurgical method is applied to recover materials or separate impurities from a specific waste stream [20]. Possible processes and technologies for the recycling of LIBs were discussed by a range of studies [12]. In recycling of LIBs, pyrometallurgical process is the main technology that is used at near commercial scale following by hydrometallurgical process [42]. The results from this stage of the model help us to have a better analysis by comparison between LIBs primary production and secondary production. Since cathodes contain diverse materials and their environmental issues would be different, in this step our focus is on using LCE in processes. Figure 4 shows the recycling stage to analyze environmental issues from different critical minerals: Li, Co and Mn. Figure 4 (a) shows Co is the element which is more recovered from LIBs. In terms of energy consumption, considering the amount of recovered materials, Co recovery consumes less energy compared to other materials. However, the cumulative energy consumption level in ten years for Co and Li recovery are very close (Figure 4 (b)). The highest level of GHG emissions generated by the recycling processes corresponds to Co in the given period (Figure 4 (c)).

The detailed calculations by comparison of different stages show that energy required for recovery Li is 38-45% higher than primary production. Consequently, the increase in pollution would be 16-20%.

Environmental decisions at different levels and scales, especially in the presence of dynamic and continuously changing conditions, significantly affect the operation of supply chain of materials over their lifespan. The results of this paper support the main finding of research by Sonoc et al. [43] which proposed a high recovery of Li from LIBs is essential to ensure the sustainability of the EV market. Comparing the results of this study with previous research, a discrepancy of the results obtained in the energy consumption in the assembling of batteries could be because of different approaches of study. For example, Majeau-Bettez et al.[44] and Notter et al.[14] developed a study of the energy consumption of the assembling of batteries with different level of detail. In the case of recycling, there are not many studies that have

discussed the energy consumption and emissions from a longer perspective.

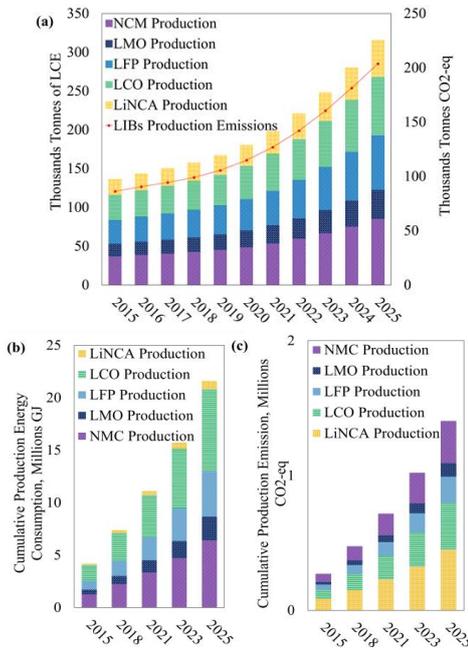


Fig. 4. Environmental impact of recycling stage from 2015 to 2025. (a) Annual; (b) Cumulative emissions; (c) Cumulative energy consumption.

4. Conclusion

The presented dynamic model of lithium supply chain for critical minerals recovery from LIBs allows comprehensive environmental analysis of energy consumption and greenhouse gas emissions of all stages of material life cycle. The increasing environmental consciousness will force states to introduce new laws, which will drive the industry decisions to a new competitive environment. In this case, our model may be used to obtain long-term estimates on system environmental performance for specific policies.

The results of this study indicate that recycling of LIBs helps to prevent the shortage of critical minerals from a mass flow perspective. However, from an environmental perspective, the current technology is not recommended to recover lithium from LIBs which leads 38-45% more consumption of energy and 16-20% higher air emissions than primary production. The obtained results indicate a clear need for a careful analysis of the recycling processes and technology. The sustainability of the recycling processes could not be taken for granted as it could vary depending on the materials.

The impact of environmental policies and environmental awareness is certainly not exhausted. Even if we limit our attention to some issues examined here, there remain quite a few unanswered questions that deserve further investigation.

An interesting topic for future research is to determine the relationship between the critical material compositions of lithium cathodes with the impact on environment through each stage of supply chain.

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