



Doctoral Thesis

### Novel approaches to enhance regional nutrients management and monitoring applied to the Austrian phosphorus case study

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by

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### Abstract

Austria, like the rest of Europe and most of other world countries, relies heavily on imports to satisfy the high demand of phosphorus (P) of its food production system. P is obtained from phosphate rock, which is a finite and non-renewable resource confined in a handful of countries. Owing to the great economic importance of P and to future supply risk, in the year 2014 the European Commission included phosphate rock in its List of Critical Raw Materials. The domestic use of P in Austria and in the rest of Europe presents several inefficiencies and losses. Further, P is not only a fundamental resource but also a harmful pollutant in water bodies, where its excessive concentration can cause sever eutrophication problems.

Research is needed to understand P flows and stocks through anthropogenic and natural compartments in order to identify where actions can be taken to increase use efficiency, to reduce losses and emissions and thus to lower import dependency. Material Flow Analysis (MFA) constitutes a suitable approach for such investigations and has been widely applied to describe mostly static P budgets at diverse geographical scales. Temporal dynamics have been however mostly neglected, although they can provide relevant information for the design of management strategies and for the monitoring of their performance.

This Thesis presents a multiyear MFA model that represents the Austrian P budget from 1990 to 2011. The systematic analysis of this model reveals that even in this relatively short and stable period of time P flows and stocks have undergone significant changes. Relevant trends in management performance are identified as well as the impact of apparently unrelated regulatory or economic developments on P flows. Further, the investigation in detail of temporal patterns has led to findings highly relevant in the field of water quality and emissions to water bodies. It was found that the decline of P point discharges in the 1990s did immediately translate into lower concentrations at low flow conditions in the Danube, but not into lower loads. The hypothesis put forward in this Thesis and supported by the analyses is that only after the large flood in the year 2002 intensively scoured the river bed and depleted the in-stream P stock accumulated during years of net retention, the reduced availability of dissolved P primarily stemming from point discharges was reflected in lower loads. It is therefore necessary to consider more thoroughly the interweaving effects of anthropogenic measures and hydrological processes in large rivers, in order to properly interpret and assess the performance of environmental management.

The MFA model of the Austrian P budget should not only be applied to inform policy makers on the status quo and on the existing problems, but also as a basis to provide a solid assessment of possible solutions and improvements. A wide range of different actions is available to optimize the system. The detailed system provided by the MFA model allows the comparison of the relative contribution of the measures, which helps in setting priorities. The results show that both recycling from waste streams and reduction of consumption hold similar potentials to decrease import dependency, although the latter are affected by higher uncertainty and more complex challenges. If the whole set of actions were implemented and a maximum effort of P governance were undertaken, Austria could fully replace the use of mineral fertilizers, reduce emissions to water bodies by 28% and its import dependency by 89%. These shall be considered maximum achievable targets, but are not necessarily optimal, since this assessment still does not include costs and other environmental aspects, which need to be included in future research.

Moreover, this work has characterized data quality through a set of indicators, has assessed how different aspects of data quality have improved or changed over time, has shown to which extent the consideration of uncertainty influences the ability of monitoring flows and stocks and has contributed to prioritize the improvement of data collection schemes.

In conclusion, this Thesis presents the feasibility and the manifold benefits provided by approaching regional nutrients management based on multiyear MFA. If public authorities integrated such an accounting scheme into their routine statistics and reporting, it could assist them in the design of governance strategies and target systems, in the understanding of feedbacks and trade-offs between measures, in the monitoring of their effectiveness, in getting feedbacks on data quality and on prioritizing data collection.

### Kurzfassung

Wie viele andere Länder Europas und der Welt ist Österreich in hohem Maß auf Importe angewiesen, um den hohen Phosphor bedarf des Lebensmittelproduktionssystems zu stillen. Als Rohstoff wird Phosphor (P) aus begrenzten und nicht-erneuerbaren Phosphaterzen gewonnen, deren Produktion auf wenige Länder beschränkt ist. Aufgrund der großen wirtschaftlichen Bedeutung bzw. des zukünftigen Versorgungsrisikos hat die Europäische Kommission im Jahre 2014 Phosphaterz als kritischen Rohstoff definiert. Der P-Gebrauch weist in Österreich und in Europa mehrere Ineffizienzen und Verluste auf. Zudem ist P nicht nur eine essentielle Ressource, sondern auch ein Schadstoff in Wasserkörpern, wo hohe P-Konzentrationen zu Eutrophierung führen können.

Forschungsaktivitäten sind erforderlich, um P-Flüße und -Lager durch natürliche und anthropogene Prozesse zu verstehen. Deren Ziel ist es, Maßnahmen zu identifizieren, die zur effizienteren Ressourcennutzung bzw. zu niedrigeren Emissionen und Verlusten und zur geringeren Importabhängigkeit führen können. Die Materialflussanalyse (MFA) ist eine geeignete Methodik für derartige Untersuchungen. Sie wurde häufig angewendet um meist statische P-Haushalte auf verschiedenen geographischen Ebenen zu darzustellen. Zeitliche Veränderungen wurden jedoch häufig vernachlässigt, obwohl sie relevante Informationen im Hinblick auf die Entwicklung von Managementstrategien und auf die Leistungskontrolle liefern können.

In dieser Dissertation wird ein mehrjähriges MFA-Modell entwickelt und untersucht, das dem österreichischen P-Haushalt von 1990 bis 2011 entspricht. Die systematische Analyse dieses Modells zeigt, dass sogar in diesem vergleichsweise kurzen und stabilen Zeitraum P-Flüße und –Lager erheblich geändert wurden.Relevante Trends in Bezug auf Managementleistungen bzw. die Auswirkung von scheinbar nicht zusammenhängenden gesetzlichen und wirtschaftlichen Entwicklungen auf P-Flüße wurden identifiziert. Außerdem hat die detaillierte Untersuchung der zeitlichen Muster zu Ergebnissen geführt, die sehr relevant im Bereich Wassergüte sind – speziell angesichts der Emissionen in die Gewässer. Es wurde festgestellt, dass sich die Verminderung von P-Emissionen über Punktquellen in den 1990ern in geringeren Konzentrationen bei niedrigen Durchflussraten in der Donau umgehend widerspiegelte, aber nicht in geringeren Frachten. Die in dieser Dissertation aufgestellte und durch die Analyse gestützte Hypothese lautet: nur nachdem das große Hochwasser im Jahre 2002 die Gewässersohle intensiv aufgerissen hatte und nachdem es das durch mehrjährige Retention akkumulierte P-Lager abgebaut hatte, spiegelte sich die geringere Verfügbarkeit vom gelösten Paus Punktquellen in verminderten Frachten wider. Es ist daher erforderlich, die verflöchtenen Auswirkungen von anthropogenen Maßnahmen und hydrologischen Prozessen in großen Flüßen zu betrachten, um die Leistung des Umweltmanagements angemessen zu interpretieren.

Das MFA-Modell des österreichischen P-Haushalts soll nicht nur angewendet werden, um die Entscheidunsträger über den Status quo und die bestehenden Probleme zu informieren, sondern es soll auch als Grundlagefürdie Begutachtung verschiedener Lösungsansätze dienen.Verschiedene Massnahmen sind vorhanden, um das System zu optimieren. Das detaillierte System des MFA-Modells ermöglicht den Vergleich von den relativen Beiträgen der verschiedenen Massnahmen, was behilflich bei der Festlegung von Prioritäten ist. Die Ergebnisse zeigen, dass Recycling von Abfallströmen und Verbrauchsminderung ein ähnliches Potential zur Senkung der Importabhängigkeit aufweisen, obwohl letzteres von höherer Unsichereit und von komplexeren Herausforderungen betroffen ist. Wenn sämtliche Maßnahmen umgesetzt würden und maximale Anstrengung unternommen würde, könnte Österreich P-Mineraldünger vollständig ersetzen bzw. die Emissionen um 28% und die Importabhängigkeit um 89% reduzieren. Solche Ziele sollten als erreichbare Maximumwerte und nicht unbedingt als Optima angesehen werden, da diese Bewertung weder Kosten noch weitere Umweltaspekte berücksichtigt, die in weiteren Forschungsarbeiten erfasst werden müssen.

Ferner wurde die Datenqualität durch ein Indikatorenset charakterisiert. Es wurde dargestellt, wie verschiedene Aspekte der Datenqualität sich im Laufe der Zeit geändert haben, und wie die Berücksichtigung der Unsichereit unsere Fähigkeit stark beeinflusst Flüße und Lager zu überwachen. Das hat auch dazu beigetragen, die Verbesserung der Datenerhebung zu priorisieren.

Abschließend stellt diese Dissertation die Machbarkeit und die mehrfachen Vorteile dar, die ein regionales auf mehrjärige MFA gestütztes Nährstoffmanagement anbiet. Die Integrierung derartiger Materialbuchführung in die reguläre Statistiken würde deshalb öffentliche Behörden unterstützen Governance-Strategien zu entwickeln, Zielsysteme zu definieren, Feedbacks und Gegenleistungen zwischen Maßnahmen zu verstehen, Managementleistungen zu beobachten bzw. Datenerhebung zu priorisieren.

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# Chapter 1 Introduction

Phosphorus (P) is an essential and non substitutable element for sustaining life on earth. Notwithstanding its fundamental role as nutrient, during the last decades P has been mainly treated as a pollutant, since it is widely acknowledged that its release into water bodies is a main trigger for eutrophication. Therefore, the efforts of both research and policy towards a better management of P have been driven by the aim of protecting water systems. This is exemplified for instance by two EU COST Actions, both focused on P in relation to water systems: Action 832 "Quantifying the Agricultural Contribution to Eutrophication" and Action 869 "Mitigation options for nutrient reduction in surface water and groundwaters". Decades of efforts via research, regulation, subsidies and technological improvements have lead to a decline of the impact that nutrients exert on water systems, but there are still relevant unsolved problems and open research questions [Sharpley et al., 2015].

In addition to eutrophication, P is also associated with other environmental impacts. Phosphate rocks typically contain several hazardous elements, the most common being cadmium (Cd), arsenic (As), chromium (Cr), lead (Pb), selenium (Se), mercury (Hg), uranium (U) and vanadium (V) [Van Kauwenbergh, 2009]. Although they are processed and purified during the production of fertilizers, soils all over the world are actually being exposed to contamination through the application of fertilizers. Nziguheba and Smolders [2008], among others, have shown a correlation between metal concentrations in fertilizers applied in Europe and their P concentration, which supports the hypothesis that metals in fertilizers primarily stem from phosphate rocks. Another aspect of this problem is that, due to the content of hazardous elements, mining of phosphate rocks generates large amounts of highly polluted waste, called phosphogypsum. According to rough estimations, each tonne of phosphate processed from phosphate rock generates five tonnes of phosphogypsum, which in some locations shows not only high heavy metal content, but also high radiation levels [Cordell et al., 2009]. This activity is thus a source of potentially high localized environmental degradation.

In parallel to addressing P as pollutant, in more recent years there has been a renewed interest for P as resource, given that high levels of P inputs are required to ensure long-term food and energy security. The biogeochemical P cycle is so slow that phosphate rock can be considered a non-renewable resource and since its deposits are heavily concentrated in a handful of countries, the EU has recently included it in the revised list of Critical Raw Materials on the grounds of high supply risk [EC, 2014b].

Given this situation, there is a need to better understand the human influence on the P cycle and of the efficiency of P use, in order to identify problems and potentials for improvement. A suitable approach is Material flow analysis (MFA), which is a standardized input-output system analysis methodology for the systematic investigation of material flows into, within and out of a given system and its associated material stocks [Brunner and Rechberger, 2004]. It has been widely applied in resource and waste management and for the assessment of several P budgets at different scales throughout the world, as shown by the reviews of Cordell et al. [2012] and Chowdhury et al. [2014].

A common trait of most P budgets is that they typically consist of static snapshots of a specific or average year. They do not take into account change in time, which might reveal further insights on the dynamics and the drivers within the system and thus be of decisive importance to select the most appropriate and effective measures. The assumption that a one-year P budget is a robust and sufficient basis for further studies or for decision making might be valid in some cases, but not in others. This criticism is supported by studies which have, to some extent, investigated the change in time of P use and have shown significant changes, trends and dynamics, such as Li et al. [2012], Ma et al. [2012], Senthilkumar et al. [2012], Lamprecht et al. [2011], and Neset et al. [2008]. Therefore, one of the conclusions of the review by Chowdhury et al. [2014] is that the lack of consideration of time and dynamics is a major research gap in the study of P budgets.

This Thesis addresses in depth the importance and the implications of temporal variability, dynamics and interweaving mechanisms in view of regional phosphorus management and monitoring. This is done both from a system perspective and with a higher resolution focus on the water bodies compartment. The role played by data and uncertainty in such context is also thoroughly investigated, with the ultimate objective being the prioritization of data collection schemes. Further, the suitability and the added values of MFA as basis for the design of optimization strategies are tested.

Temporal variability is the core topic of Chapter 2, which contains the paper by Zoboli et al. [2015a] in the Journal of Industrial Ecology. This work presents a multiyear Austrian P budget, which is built on the highly detailed model developed by Egle et al. [2014b] and which covers the period 1990-2011. The time series composed of 22 one-year budgets is systematically analyzed to investigate if and how the system has changed in a period of relative socio-economic stability. Another issue thoroughly addressed in the paper concerns the characterization of data quality, the quantification of uncertainty and their implications for the monitoring of P management strategies. For that, a novel approach is applied, which was recently developed by Laner et al. [2015]. Further, this case-study is used to test the hypothesis that multiyear MFA can contribute to the improvement of the model itself.

The MFA model representing the Austrian P budget includes also the hydrosphere, where the Danube river plays a major role, in that it drains more than 96% of the total

national territory [ICPDR, 2014a]. Through the analysis of the time series, a dramatic and enduring decline of total phosphorus (TP) concentration and TP loads was detected in the Danube after the year 2002. Although it is well known that floods cause peak transport loads in rivers due to elevated sediment P mobilization, which explains the high load peak in 2002 (year characterized by a century flood [Zessner et al., 2005]), a long-lasting decline afterwards was not expected. This raised the following questions: can large floods exert a long-term effect on P concentration and loads in rivers? Is the major decline of P emissions from point sources in the 1990s reflected in the transported loads? How do the effects of these strong anthropogenic and nature driven changes interweave in determining the actual transported loads? Does the in-stream P stock play a relevant role? These questions, which are of fundamental importance to correctly interpret monitoring data and the performance of management strategies in reducing P emissions to water bodies, are addressed in Chapter 3, where the publication of Zoboli et al. [2015b] in Science of the Total Environment is presented.

After the specific focus of Chapter 3 on P emissions to water bodies and on the interweaving mechanisms taking place in large rivers, Chapter 4 recovers the systemic perspective by concentrating once again on the whole Austrian P budget. This chapter presents the work contained in the paper submitted by Zoboli et al. [submitted] to Science of the Total Environment. Whereas the approach of Chapter 2 is the use of MFA to describe, quantify and analyze P flows and stocks with the intent of identifying losses, inefficiencies and relevant trends, in Chapter 4 MFA serves as basis for the optimization of the system. Schoumans et al. [2015] and Withers et al. [2015] have recently reviewed and put forward several different actions that could be taken to achieve P stewardship in Europe. However, it is necessary to assess the applicability and quantify the actual potential that such actions have in different regions. In Austria Thaler et al. [2015], Egle et al. [2014a] and Zessner et al. [2013] have addressed some specific actions, namely the shift to a healthy and balanced diet, the recovery and recycling of P from sewage sludge and the reduction of diffuse emissions from agriculture. An exhaustive comparative assessment, accompanied by a systemic approach, that can properly support decision makers in designing a national governance strategy, is still lacking. The questions addressed in this chapter are: which measures are applicable to the Austrian case-study and which limitations exist? What would be the relative contribution of each of them on the overall national performance? How uncertain is this assessment and which data should be given priority when improving data collection schemes? How would the Austrian P budget look like if all actions were implemented? In other words, what is the maximum that Austria could achieve in the context of P stewardship?

### Chapter 2

# Added values of time series in Material Flow Analysis. The Austrian phosphorus budget from 1990 to 2011

#### 2.1 Abstract

Material flow analysis is a tool that is increasingly used as a foundation for resource management and environmental protection. This tool is primarily applied in a static manner to individual years, ignoring the impact of time on the material budgets. In this study, a detailed multiyear model of the Austrian phosphorus budget covering the period 1990–2011 was built to investigate its behavior over time and test the hypothesis that a multiyear approach can also contribute to the improvement of static budgets. Further, a novel method was applied to investigate the quality and characteristics of the data and quantify the uncertainty. The degree of change between the budgets was assessed and showed that approximately half of the flows have changed significantly and, at times, abruptly since 1990, but it is not possible to distinguish unequivocally between constant and moderately changing flows given their uncertainty. The study reveals that the phosphorus transported in waste flows has increased more rapidly than its recovery, which accounted for 55% to 60% of the total waste phosphorus in 1990 and only 40%in 2011. The loss ratio in landfills and cement kilns has oscillated in the range of 40%to 50%. From a methodological point of view, the multiyear approach has broadened the conceptual model of the budget, making it more suitable as a basis for material accounting and monitoring. Moreover, the analysis of the data reconciliation process over a long period of time proved to be a useful tool for identifying systematic errors in the model.

#### 2.2 Introduction

#### 2.2.1 Importance and challenges of phosphorus

The role played by phosphorus (P) in modern societies is essential and irreplaceable. Not only is it often a limiting factor for growth and a fundamental component of fertilizers, it also finds manifold industrial applications, such as in car engine lubricants, pesticides, rechargeable batteries, and flame-retardants [Schipper, 2014]. P is exclusively obtained from mineral deposits, and its future availability is currently plagued by uncertainty. The figures for phosphate rock reserves and resources as well as the models applied to estimate their peak production and lifespan are still highly debated, with predictions ranging from few decades to hundreds of years [Cordell et al., 2009, Van Kauwenbergh, 2010, Van Vuuren et al., 2010, Cooper et al., 2011, Cordell and White, 2011, Sverdrup and Ragnarsdottir, 2011, Vaccari and Strigul, 2011, Ott and Rechberger, 2012, Edixhoven et al., 2013, Koppelaar and Weikard, 2013, Mohr, 2013, Scholz and Wellmer, 2013]. Notwithstanding the disputed scarcity concern, there are other important rationales to pursuing a more efficient and sustainable P management. The most pressing issues are water body eutrophication, accumulation of heavy metals in agricultural soils through the application of mineral fertilizers, and geopolitical and equity risks associated with the concentration of mineral deposits in a handful of countries [Condron et al., 2013, Ulrich et al., 2013a,b]. Based on the supply risk, the European Commission has recently added phosphate rock to the revised list of Critical Raw Materials [EC, 2014b].

#### 2.2.2 One-year and historical phosphorus MFA

As shown by Ulrich and Schnug [2013], since 2008 multi-stakeholder platforms have been formed and researchers have intensified their effort in tackling the challenges related to P. Material flow analysis (MFA) has played a key role in assessing flows and stocks and identifying hotspots of poor management and recycling potential, as manifested by two recent reviews of P MFAs carried out at different scales throughout the world [Cordell et al., 2012, Chowdhury et al., 2014]. Until recently, these studies have primarily focused on static 1-year budgets (balanced models of material flows and stocks obtained through MFA methodology), and the review carried out by [Chowdhury et al., 2014] identified the paucity of multi-year analyses as a major knowledge gap. The few efforts put forth in this direction have led to significant outcomes.

Li et al. [2012] analyzed the urban P metabolism through food consumption in China from 1986 to 2006 and identified the relationships among urban P inflow and population growth, dietary changes, and velocity of urban area expansion. Furthermore, their research showed how the rising urban per capita income has altered the urban P stock and the P outflow to surrounding non-urban ecosystems. Ma et al. [2012] conducted a P MFA for all of China from 1984 to 2008, which not only showed a significant increase in P consumption and waste, an increasing (though fluctuating) P ore extraction, and a decline in P recycling rates but also highlighted the correlation between these changes and specific socioeconomic factors. Although European countries have not undergone socioeconomic changes as drastic as those of China in recent decades, Senthilkumar et al. [2012] revealed that France halved the use of mineral fertilizers in agriculture from 1990 to 2006, while maintaining the same levels of agricultural production and losses to the environment. Neset et al. [2008] analyzed the flow of P in food consumption and production for a city in Sweden in the period 1870-2000 and found an increase in P reaching consumers, due both to population growth and to a shift toward a diet richer in meat and dairy products. Their study also analyzed the related outflow of P in waste and wastewater, revealing a steep decline in the ratio between reuse and losses. Lamprecht et al. [2011] quantified the high impact that the Bovine Spongiform Encephalopathy crisis exerted on the P cycle in Switzerland by banning the reuse of P-rich meat and bone meal as animal feed.

These examples prove that multiyear MFAs can provide insights with important implications for environmental, resource, and waste management. In economy-wide accounting and analysis, multi-year schemes are already widespread to the extent that several countries have incorporated them into their statistical information systems [Fischer-Kowalski et al., 2011]. As suggested by Chowdhury et al. [2014], however, the main obstacle that might dissuade most researchers from conducting multi-year studies in the specific field of phosphorus is the lack of data. In this respect, Austria offers a favorable case study due to the existence of a detailed budget representing an average year between 2004 and 2008 [Egle et al., 2014b] and large data availability for the past.

This study presents a multiyear analysis of the Austrian P budget covering the period from 1990 to 2011. The first aim of the work is to identify and to assess the extent of the temporal changes occurred in the system during the last two decades. It differs from previous multiyear P MFAs due to the very high level of detail of the model, which allows for an unprecedented systematic assessment. One of the main novelties lies in the characterization of data quality and uncertainty, and in the quantification and discussion of the implications of the latter for the assessment of temporal changes. The second goal of this contribution is to test the hypothesis that a multiyear MFA can lead to a better understanding of the data quality and to the improvement of the model itself, which in turn would benefit the 1-year static budgets. Therefore, although investigating the changing traits of P flows and stocks is relevant per se, this work tests novel methodological approaches that could prove useful in the study of other materials.

#### 2.2.3 Data uncertainty

Although the uncertainty of the data has been increasingly recognized as an essential aspect in MFA studies, in reality it is still often ignored or only qualitatively discussed [Laner et al., 2014, Rechberger et al., 2014]. As presented in the review by Laner et al. [2014], there exist different approaches that can be grouped as follows: a) qualitative and semi-quantitative methods, which reflect the authors' confidence in the results without formally assessing the quality of the data; b) methods based on data classification, used to evaluate the data quality without applying rigorous mathematical procedures to propagate uncertainties in the model; and c) statistical methods, which do propagate uncertainties in the model. This study applies a novel approach developed by Laner

et al. [2015], which builds on the idea proposed by Hedbrant and Sörme [2001] of quantifying the material flow uncertainty combining data classification and exponential-type uncertainty characterization functions, and on the scheme introduced by Weidema and Wesnæs [1996] to assess the quality of life cycle inventory data. This novel approach was selected for different reasons. In the first place it is based on a characterization of the data quality through the use of different indicators, which allows describing the characteristics of the data and identifying specific problems. Second, the quantification of the uncertainty follows a procedure designed to ensure its consistency within the model. Moreover, it is based on coefficients of variation, making the results directly utilizable for the MFA software STAN applied to balance the model in this study. Last, this method was chosen in order to provide a well documented and transparent characterization of the uncertainty.

#### 2.3 Materials and Methods

#### 2.3.1 Model and data

This work follows the methodology of MFA described by Brunner and Rechberger [2004] and addresses the flows of P on the national level. According to the classification suggested by van der Voet [2002], this work belongs to the accounting category, which represents the most suitable modeling approach for keeping track of flows and stocks in order to enable policy makers to detect trends and to assess the performance of implemented measures, and also for identifying missing or inaccurate data. The design consists of a multiyear static model, which simultaneously balances 22 time nodes (yearly snapshots). The 22 time nodes are independent, in that data-based values for the flows are introduced as input to the system for each one of the studied years. The only elements that are not independent among time nodes are stocks, for which only the initial value for the year 1990 is introduced as input, whereas thereafter their total amount is obtained by calculating the stock changes in each individual year based on closed mass balances. The reason behind this choice is that only poor information is available to calculate the total value of stocks, so that it is preferable to carry out such estimations only as a starting point and let the rest be based on the flows, for which better data is available. Although stocks are not entirely time independent, this approach differs substantially from MFA dynamic modeling, which includes time as a modeling parameter to explore the behavior of stocks and flows in time. Dynamic modeling is mostly applied for the analysis of accumulated stocks of metals and other persistent toxics and to predict future scenarios with respect to waste flows and emissions [van der Voet, 2002, Müller et al., 2014]. The multiyear static model presented here does not capture the dynamics between stocks and future flows and therefore does not aim at predicting future scenarios. It is instead designed to perform a descriptive, detailed and data-based analysis of the system over time.

The work of Egle et al. [2014b] is used as a foundation to conceptually describe the national P budget. Given that the model structure is identical for the whole time series,

some modifications had to be made to the existing model. A flow of meat and bone meal used as animal feed was added, because it represented a very important use before the ban introduced in the year 2001. Further, the recent expansion of the bioenergy sector justifies this process being highlighted separately. In contrast, the results of Egle et al. [2014b] revealed that the role of the chemical industry in Austria is not so significant that it requires a separate process; therefore, it was lumped within the *Industry* process, which also encompasses the trade and production of food, feed, raw phosphates, and fertilizers. The adjusted model is shown in Figure 2.1 and is structured in nine processes that, with the exception of *Crop farming* and *Water bodies*, are described by sub-systems, which are available in the Appendix A. In total, the system is composed of 56 processes, eight stocks, 122 flows, and two transfer coefficients.

With respect to the input values, a detailed description of the equations and data sources used in the calculations is available in the Appendix B. The majority of the flows were obtained by multiplying goods (defined as substances or mixtures of substances that have economic values assigned by markets, according to Brunner and Rechberger [2004]) with their P concentration. For a subset of flows, however, data on the P flows (P mass per time) were directly available given the abundance of research and statistical accounting already developed for this substance. Examples of this occurrence are wastewater, sewage sludge and effluents, mineral fertilizers, and P flows in the environment, namely atmospheric deposition and emissions to water bodies from agricultural and forestry soils. For the goods and the direct P flows, all of the compartments of the system are characterized by large data availability, but they do show important differences with respect to the frequency and level of detail. For the production, import, export, and consumption of food and animal feed products, highly detailed and yearly data are available in the supply balance sheets. National statistics and official sector reports also provide further detailed and yearly data for the processes Animal husbandry, Crop farming, and *Forestry*; for food consumption, and for the import/export of chemicals. With respect to the *Bioenergy* process, it is necessary to make a distinction between its sub-processes. Biofuels production is highly regulated and documented in official yearly reports, although attention is mostly focused on the biofuels themselves and not as much on the materials used for their production or on their by-products, which is the relevant information for the P budget. The data on biogas or biomass plants, in contrast, offer more information on input materials and by-products, but are not available with the same regularity. In compliance with the European Directive 91/271/EEC on urban wastewater treatment, official reports (published with a frequency that recently increased to biannual) provide data on the P loads in and out of municipal treatment plants as well as information on the average P removal rate and the connection rate to the sewage system. Direct data on the specific contribution of households and industries to the total P load are not available, and therefore, these quantities were estimated based on information on the average P load per capita. The most important source for Waste management is represented by the national waste management plans, which contain information on the volumes of wastes generated, their main origins, management, and disposal. Such plans are not a satisfactory source for this study, because they are published only every five years and also because they often present a high level of aggregation, especially for the management and disposal of waste flows. This information could be partially complemented with official reports that provide greater detail on individual processes or specific waste flows, certain of which also supply multi-year information. This notwithstanding, the *Waste management* process, which plays a crucial role in the P budget, does not possess the same level of information as the production and import-export sectors. The estimation of the P flows in the environment had to rely largely on modeling despite fairly abundant data availability, owing to the high complexity of the processes involved.

In addition to the partial lack of data, a multiyear MFA faces another problem, namely, the consistency of sources over time, which can further reduce the data availability in comparison to a static 1-year budget. For the current study, an example is the trade of mineral fertilizers for which FAOSTAT had to be discarded as a possible source because its accounting methodology was modified in 2002, giving rise to two incompatible datasets.

The data corresponding to P concentration are largely available and primarily obtained from peer-reviewed literature and scientific reports, although this information was often derived from studies carried out in different countries and distinct years. To a lesser extent than the data regarding the goods, the P concentration also shows a decreasing level of information from production and consumption to waste management. Nonetheless, throughout the entire system, this information is usually more detailed than the existing categories of the material flows would require.

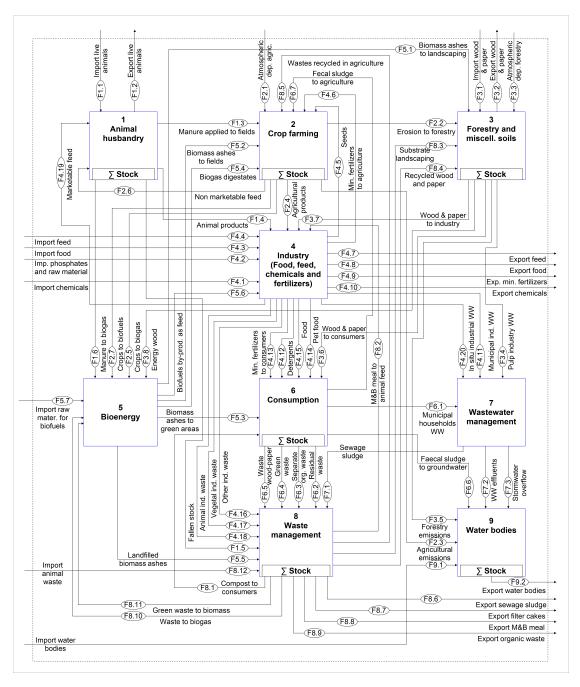


Figure 2.1: Qualitative MFA model of the Austrian P budget.

#### 2.3.2 Uncertainty characterization

The uncertainty of material flow data was characterized using an approach developed by Laner et al. [2015], based on the work of Weidema and Wesnæs [1996] and Hedbrant and Sörme [2001]. It is a stepwise procedure that starts with the assessment of how representative the data at hand are with respect to the actual values of interest. Similarly to the pedigree matrix proposed by Weidema and Wesnæs [1996], this assessment relies on a set of quality indicators and on the definition of criteria to select evaluation scores. The indicator *Reliability* refers to the documentation of the data and it reflects the transparency in regard to the sampling, collection, and verification procedures. Completeness describes how complete the available data are in comparison to the actual values of interest. Temporal correlation and Geographical correlation take into account to which extent the available information diverges from the datum of interest, from a temporal and geographical perspective, respectively. The indicator Further correlation reflects deviations other than temporal or geographical, such as parameters of a specific type of technology or process. With respect to the original set of indicators suggested by Laner et al. [2015], Composition was added in this work to describe the level of information on the composition of the goods, which is of crucial importance for the correct selection of the P concentration, especially in case of heterogeneous or aggregated flows. For each indicator four evaluation scores are available, where 1 represents the best level and 4 the worst. The indicators and the criteria defined to assign the evaluation scores are reported in Table 2.1. Whereas the indicators *Reliability* and *Completeness* play a decisive role for any type of data, the relative importance of the other indicators changes from case to case; therefore, their contribution to the final uncertainty also considers the level of sensitivity of the data, defined as the degree of influence of the deviation (e.g., temporal or geographical) from the existing datum to the value of interest. Three levels of sensitivity are defined: *Highly sensitive*, *Sensitive*, and *Not sensitive* for which a deviation of the datum is highly relevant, relevant, or not relevant, respectively. The evaluation scores of the data quality indicators are directly translated into coefficients of variation (CV) through continuous exponential-type functions, which also allow the use of intermediate scores. As shown by Laner et al. [2015], the selection of the functions is critical, in that it largely determines the uncertainty ranges. The reason for choosing exponential-type curves is the fact that these have been the mostly used functions in the past, among others by Hedbrant and Sörme [2001], on which this approach builds. Another important decision lies in the selection of the parameters that describe the functions. These need to be determined by the modelers, in order to ensure their plausibility according to the specificities of each case study. In this work the mathematical functions illustrated in the Appendix C were used, which produce the CVs presented in Table 2.2. These values are well in line with the uncertainty levels applied in other phosphorus MFA studies, included within the ranges 1.5%-50% [Klinglmair et al., submitted], 7%-51% [van Dijk et al., 2015], 10%-100% [Egle et al., 2014b], 5%-100% [Ott and Rechberger, 2012], 5%-50% [Neset et al., 2008], 0%-50% [Antikainen et al., 2005]. Given that each of the indicators represents an independent aspect of the overall data uncertainty and under the assumption of normality, the coefficient of variation  $CV_a$  that represents the total

uncertainty of the input data is calculated through equation 2.1, where  $CV_r$ ,  $CV_c$ ,  $CV_{co}$ ,  $CV_t$ ,  $CV_g$ , and  $CV_f$  represent the coefficients of variation of the indicators *Reliability*, *Completeness, Composition, Temporal correlation, Geographical correlation*, and *Further correlation*, respectively.

$$CV_{a,i} = \sqrt{CV_{r,i}^2 + CV_{c,i}^2 + CV_{co,i}^2 + CV_{t,i}^2 + CV_{g,i}^2 + CV_{f,i}^2}$$
(2.1)

If no data are available and the input values are determined through expert judgments, the uncertainty is directly assessed through the indicator *Expert judgment*.

α	Score: 1	Score: 2	Score: 3	Score: 4
on ess	Methodology of data generation is well documented and consistent (e.g. Standard documentation - Meta information of National Statistics; laboratory analytical methods)	Methodology of data generation is described but not fully transparent	Methodology is not described but principle of data generation is clear	Methodology of data generation is unknown (data presented without any meta-information)
ų	Complete acquisition of data (no extrapolation; for aggregated flows data available for all goods)	Partially fragmented data (minor need for extrapolation; for aggregated flows data available for majority of goods)	Fragmented data (considerable need for extrapolation; for aggregated flows data available for minority of goods)	Highly fragmented data (major need for extrapolation; for aggregated flows data available for less than one third of the goods)
	Value is expressed in detailed categories (adequate to select correct P concentration for each category) or no categories exist (single/uniform P concentration)	Value is expressed in large categories	Value is only partially expressed in categories	No information on the composition is available (no basis to select appropriate P concentration)
	Value relates to the correct year	Deviation of 1-5 years	Deviation of 6-10 years	Deviation of more than 10 years
Geographical Value relation	Value relates to the studied region	Value relates to comparable region/economy/society	Value relates to less comparable region/economy/society	Socio-economically different region
Value rela Further correlation product, the	Value relates to the same product, the same technology, etc.	Value relates to similar technology, product, etc.	Values deviates from technology/product/of interest, but still acceptable	Value deviates strongly from technology of interest; correlation unknown
Expert judgment Formal staten $\epsilon$	Formal statement from qualified expert	Robustly based estimation	Weakly based estimation	Speculation or crude assumption

Score	Sensitivity	1	<b>2</b>	3	4
Reliability	-	4%	10%	22%	50%
Completeness	-	0%	10%	22%	50%
Composition Temporal correlation Geographical correlation Further correlation	Highly sensitive Sensitive Not sensitive	0% 0% 0%	$10\% \\ 5\% \\ 2\%$	22% 11% 4%	50% 22% 8%
Expert judgment	-	10%	20%	40%	80%

Table 2.2: Coefficients of variation (%) for quality indicators, according to score and sensitivity level (where it applies).

*Note*: "Temporal correlation," "Geographical correlation," "Further correlation", and "Composition" are four independent indicators. They all have the same set of coefficients of variation that are determined by the three sensitivity levels (in the second column). That is why the coefficients of variation are aligned to the rows of sensitivity levels and not to the rows of the four indicators.

This method is partially exposed to subjectivity in two aspects, namely, in the assignment of evaluation scores for the quality indicators and in the choice of the functions that translate the scores into CVs. With the exception of *Temporal correlation*, for which scores are defined through a precise number of years, for the other indicators the choice of scores 1 (best) and 4 (worst) are straightforward, whereas the scores in between are not univocal and are meant to allow the modelers to make use of their own experience and express also subtle differences. With respect to the mathematical functions, the choice is not unique and, in this type of studies, it is very hard or typically impossible to validate them owing to lack of empirical data. Therefore, any comparison of the uncertainties here calculated with uncertainties estimated in other studies shall be carried out with prudence. This notwithstanding, the approach has the advantage of providing the authors with a scheme that, thanks to the use of continuous functions, ensures consistency of uncertainty estimates within the model. Besides the general importance that internal consistency plays in any MFA study due to its potential effects on the data reconciliation process and on the identification of the most critical data, it is of particular relevance in this work, where the flows, their temporal change and their uncertainties are subject to a systematic assessment and comparison. In addition, the structured uncertainty characterization provides the readers with a documentation of the rationales and the calculations underlying the uncertainties of the MFA results, which makes it reproducible.

A more detailed description of the method and of its positioning in the context of other existing approaches is available in Laner et al. [2015].

#### 2.3.3 Analysis of the degree of change of the budget

To assess and quantify if and to what extent the system has changed in the 22 years examined in the study, the degree of temporal change for the 122 flows and the eight stock change rates (which can be handled as flows for this purpose) was analyzed and

categorized. For each year and each flow, the percentage change was calculated with respect to 1990, the first year of the time series, which was taken as a reference baseline. The presence of null values provokes disproportionate results because the year in which a flow ceases to exist produces a change of -100%, whereas the calculated change when a new flow appears is excessively high. Therefore, in the latter case, an artificial change of 100% was introduced to equally consider the appearance and disappearance of flows. Based on the outcomes, the flows and stock rates were divided into three categories: constant (if no change has occurred), moderately changing and extremely changing (according to the degree of temporal change). To equally treat increases and decreases, the threshold between the two latter categories was set at the factor 2 that corresponds to a positive change of 100% and to a negative change of -50% (Fig. 2.2a). The following step of this analysis (Figure 2.2b) was designed to investigate whether and to which extent the consideration of the uncertainty would affect the ability of actually detecting the temporal changes. For this purpose, different tolerance levels were defined:  $\pm 5\%$ ,  $\pm 10\%$ ,  $\pm 15\%$ , and  $\pm 20\%$ , these being the ranges within which the temporal changes were ignored (i.e. flows were considered to remain constant). Further, two additional tolerance levels were applied to take into account the specific uncertainty of each flow:  $\pm 1$  standard deviation and  $\pm 2$  standard deviations. In this case, the variation of a flow was considered as an actual change only if the respective 68.3% and 95.5% data intervals of the normal distributions characterizing each flow did not overlap.

To evaluate whether the flows have changed gradually or suddenly, the same analysis was carried out by calculating the change of each year with respect to the previous year.

The stocks were excluded from the analysis, partially because their total value is much higher than the contribution of flows of single or even multiple years and partially because of their large initial uncertainty.

#### 2.3.4 Analysis of the reconciliation process

The model was balanced using the software STAN [Cencic and Rechberger, 2008]. The assumption underlying the algorithm used by STAN is that the parameters are normally distributed independent random variables and that uncertain values are expressed using the mean and the standard deviation. Based on this, the software applies Gaussian error propagation and data reconciliation to alter the mean values and calculate the final uncertainty. Because the standard deviation is used as a weighting factor during the reconciliation process, values with higher uncertainty are more heavily reconciled than values with lower uncertainty. The mathematical details of the calculation can be found in Fellner et al. [2011].

The effect of the reconciliation process on the time series was analyzed to assess whether the flows were randomly altered or if the input value was regularly increased or decreased, thus suggesting the presence of a systematic error. If the difference between the input and reconciled values was less than 5%, it was also considered random and therefore not significant for this analysis. Further, the reconciliation of the whole system was also calculated to assess whether there are differences in this respect between the years under study. It was assessed by summing up the percentage variation of the

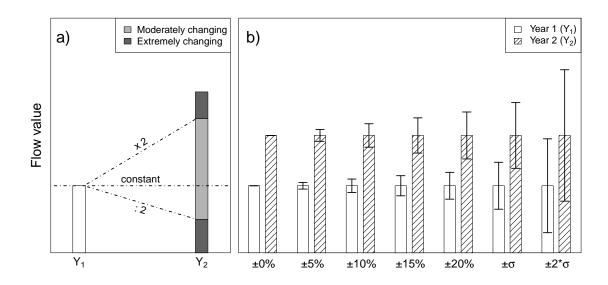


Figure 2.2: Illustration of the analysis of change in the flows over time: a) categories of the degree of temporal change; b) applied tolerance levels to explore impact of different uncertainty levels on capability of detecting the changes. In this example the indicated change would be rated as moderate for the first five pairs of columns and as constant for the last two ones (overlapping tolerance levels).

input values of all flows for each year, subsequently normalized with respect to the reconciliation in 1990 to compare the different years. This comparison should also take into account the degree of over-determination of the model, that is to say the difference between number of equations and number of unknown variables, but in this case it was not necessary, because the model has the same degree of over-determination for each year.

#### 2.4 Results and discussion

#### 2.4.1 Quality of the dataset

The approach applied in this work for the characterization of data quality is based on a set of indicators (Table 2.1) and on the selection of evaluation scores (1-4) for each of them. With the aim of assessing the overall quality of the system, the arithmetical mean of the scores assigned to all flows for each indicator and year was calculated and the outcomes are presented in Figure 2.3. Out of the 106 flows for which input values could be quantified, more than 80% were calculated by multiplying goods with their P concentration, whereas for the remainder, the P flows were directly available. Therefore, this evaluation was carried out separately for these types of data. The *Reliability* (Fig. 2.3a), an expression of the level of documentation of the data, was notably good for the P concentration and for the directly available P flows in the past due to a long

tradition of measurements of the P content in several goods. The reliability for the goods was, and still is, poorer but has improved over time, especially in the production and trade statistics, due to adopted international guidelines and procedures. The lowest level of documentation and transparency was detected for the *Consumption* and *Waste* management processes. As depicted in Figure 2.3b, incompleteness of the data is not an issue for this study, although a slight quality decrease began near the year 2003 together with the increasing role of the *Bioenergy* process for which important information is still incomplete. With respect to the *Composition* indicator, which evaluates the level of detail and categorization of the data, the quality is quite high and has slightly improved over time, although with certain unexpected oscillations on the goods level. As far as it concerns the *Temporal correlation* (Fig. 2.3d), for directly available P flows and for P concentration, the average deviation from the datum to the year of interest oscillates around 6 years, whereas for the goods, it has considerably decreased from 10 to less than 5 years. In this respect, the increase after 2009 could be misleading and should not be interpreted as a general decrease of the frequency, because at the time of this study, certain important reports and statistics had not yet been published for the years 2010 and 2011. The geographical deviation (Fig. 2.3e) from the study area affects only the P concentration because the information is often obtained from international literature, although generally from countries similar to Austria. The indicator Further correlation describes all other types of deviations from the actual datum of interest, for example, market year versus calendar year or live weight versus carcass weight. As shown in Figure 2.3f, such deviations are present in the study, although they do not represent a significant problem. From this evaluation, it can be deduced that the data available for the Austrian national P budget are of generally high quality (average low scores), although it is important to remember that this analysis uses average scores and that data problems for certain specific flows still exist. Moreover, a fraction (however small) of the information required for the calculation of the flows is not available and must be estimated; its corresponding uncertainty was directly quantified through the indicator Expert judgment and is therefore not represented in Figure 2.3, which illustrates the quality of the actually available data. The relative uncertainties estimated for all the flows and time series are available in the Appendix C.

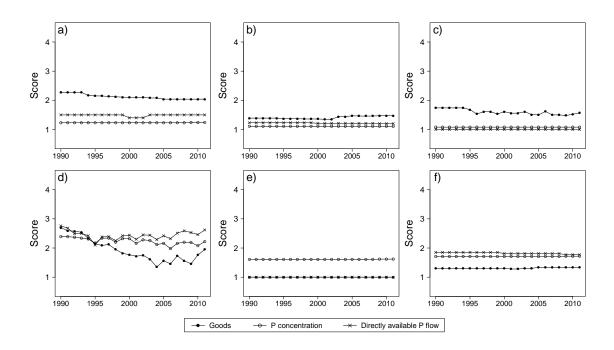


Figure 2.3: Average scores of the six quality indicators calculated for the goods (mass per time), P concentration, and directly available P flows (P mass per time): a) reliability, b) completeness; c) composition; d) temporal correlation; e) geographical correlation; f) further correlation.

#### 2.4.2 Degree of temporal change of the budget

Figure 2.4a shows the outcomes of the analysis of the degree of change of the budget with respect to the reference year 1990. The results are partly sensitive to the applied tolerance levels. If tolerance levels between 0% and  $\pm 5\%$  are applied, the analysis indicates that one third of the flows and stock change rates changed moderately, and two thirds were affected by an extreme variation, whereas with ranges from  $\pm 10\%$  to  $\pm 20\%$ , the fraction of moderately changing flows and stock rates gradually decreases until 15%. The specific standard deviation shows outcomes very similar to the  $\pm 20\%$  range, whereas the level of twice the standard deviation decreases both the extreme and moderate fractions to 50% and 5%, respectively. In conclusion, the analysis reveals that half of the flows and stock change rates changed substantially, with certain flows that appeared or disappeared and others that at least doubled or halved their initial value. For the other half, in contrast, it is not possible to ultimately conclude which fraction remained constant or changed moderately from this analysis because of the high uncertainty of the results. The second component of this analysis instead (Fig. 2.4b), explores to what extent the flows and stock change rates changed from a given year to the following one to provide an overview of whether the changes took place gradually or rather abruptly. This analysis predictably and reasonably suggests that a large proportion of the flows were affected by gradual and moderate changes, but between 24% and 33% of the flows (depending on the considered tolerance level) recorded at least one extreme variation, indicating the noteworthy presence of substantial and sudden changes. The outcomes also highlight the difficulty of detecting smaller annual changes when uncertainty ranges are applied. This situation poses a problem in view of P management monitoring.

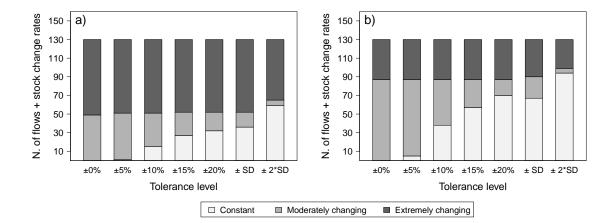


Figure 2.4: Degree of temporal change of 122 flows and 8 stock change rates: a) categorization according to the change with respect to the reference year 1990; b) categorization according to annual change. Results are shown for different tolerance levels (uncertainty thresholds used to determine whether temporal changes can actually be detected or not). The y-axis indicates the number of flows and stock change rates in each category.

Figures 2.5 and 2.6 show a comparison between the budgets in the year 1990 and in 2011. In the latter, the flows are colored according to the change of the entire time series from 1990 and are based on the standard deviation as the tolerance level. This figure shows that the majority of the extreme changes are connected to industrial trade, sewage sludge, meat and bone meal, and the *Bioenergy* process.

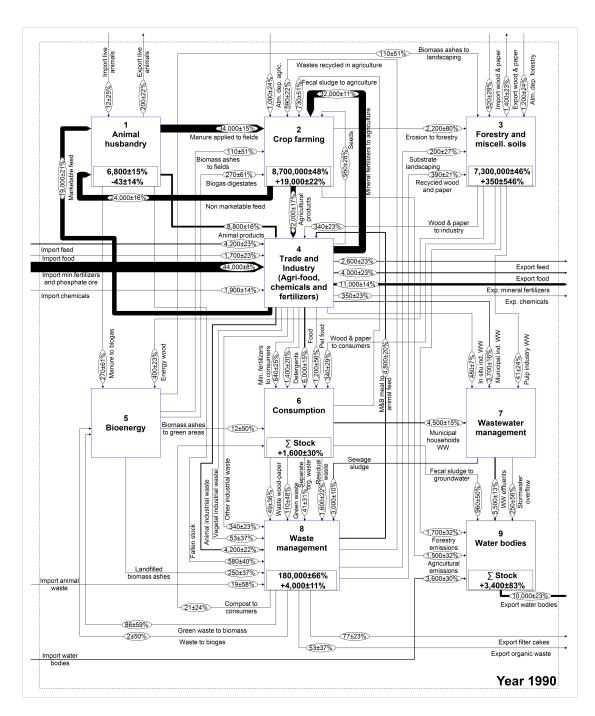


Figure 2.5: Austrian P budget representing the year 1990. Units for flows and stocks are t/y and t, respectively.

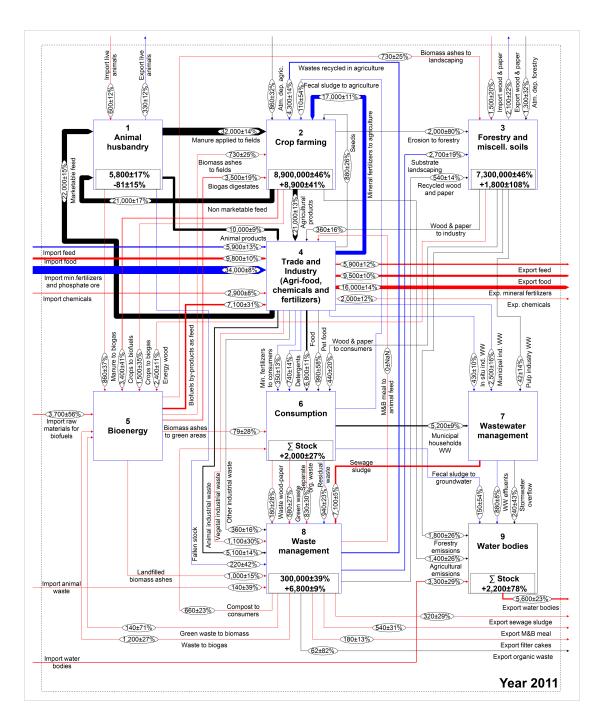


Figure 2.6: Austrian P budget representing the year 2011. Units for flows and stocks are t/y and t, respectively. The colors represent the change calculated over the entire time series with respect to 1990 (Fig. 2.5), taking SD as the tolerance level: (black) constant, (blue) moderately changing, (red) extremely changing.

#### 2.4.3 Long-term data reconciliation

The analysis of the data reconciliation process on the time series reveals that less than 10% of the flows were systematically reconciled, that is to say they were always altered in the same direction. This result indicates the presence of systematic errors, however small. The *Composting* sub-process, which is a subset of the *Waste management* compartment, offers a clear example. As shown in Figure 2.7, over the entire period under study, the input flows were reduced, and the output flows were increased by the reconciliation process. Two potential sources of error lie in the fact that the data on the composted sewage sludge are often aggregated together with data on sludge handled by other mechanical-biological treatments, and that the P content of the compost products is quite heterogeneous. Owing to the lack of further information, it is not possible to identify the actual cause of the problem, but at least the systematic inconsistency was detected and could be addressed in the future if more accurate data are made available. The analysis of the reconciliation process was carried out not only at the end of the study but was also included as an additional step within the usual iterative procedure behind MFA, as described by Laner et al. [2014]. This action has aided in identifying errors, which could be solved by either gathering more detailed data or correcting estimations, thus leading to an improved database for the budget.

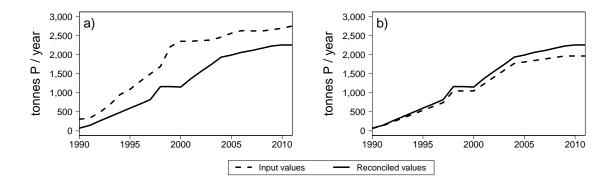


Figure 2.7: Comparison between input and reconciled values of the Composting subprocess: a) sum of input flows; b) sum of output flows.

With respect to the entire system, the results (available in the Appendix D) show that the impact of the reconciliation on the input values has decreased in time due to a general improvement of the data quality. Notwithstanding this general improvement, the impact of the reconciliation presents fluctuations in the time series, showing that the quality of a static MFA is sensitive to the choice of the year for which it is carried out.

#### 2.4.4 Selected time series relevant to sustainable management

As previously shown by Egle et al. [2014b] and in line with the trend in western European countries [Schröder et al., 2011], the use of mineral fertilizers was remarkably

reduced over the last 20 years. In addition, in the same period, the Austrian livestock population has constantly decreased; therefore, the accumulation of P in agricultural soils was further reduced. Nonetheless, the surplus accumulated in the past decades was so high that no impact on crop production or emissions to water bodies was visible, which is in agreement with the findings of Senthilkumar et al. [2012] for France.

The results show that the average daily intake of P of the Austrian population has remained practically constant with an average value of 1.6 gP/cap\*d, in agreement with the findings of Welch et al. [2009] for Germany and of Elmadfa [2009] for Central and Eastern Europe, which does exceed the values recommended by DGE et al. [2003]. The actual intake can diverge considerably from food availability, depending on the degree of loss of edible food and nutrients through the consumption process. This study reveals that the available P that is wasted was already approximately 30% in 1990 and increased up to 40% by 2011. What has improved in this respect is the recovered fraction of the waste P from the *Consumption* process, which has increased from approximately 20% in 1990 to circa 60% in 2011, primarily owing to the enhanced separate collection of organic waste. To a lesser degree, the improvement was also impacted by more widespread home composting, which is not an indicator of better management per se given the potential accumulation of unnecessary P in gardens and green areas, as discussed in Egle et al. [2014b]. An important aspect that has emerged from this study with respect to food P is the trend of rapidly increasing import, from  $1.700 \pm 23\%$  tP in 1990 to  $9.800 \pm 10\%$  tP in 2011. This amount is relevant in terms of material accounting and environmental and resource management because it poses the question of how to measure, control, or even take into account the mineral P consumption and the pollution taking place in other countries.

Whereas biomass thermal plants have a longer tradition in Austria, the biogas and biofuels sectors (with the exception of few rural and small plants) have recently emerged in the years 2002-2003 and were characterized by notably quick expansion with a consumption of  $1,200 \pm 32\%$  tP contained in crops in the year 2004, which grew to 8,600  $\pm 30\%$  tP in 2011. Further, they currently handle approximately 1,000 tP/y of industrial organic waste and circa 800 tP/y of manure. Biogas, bioethanol, and biodiesel contain practically no P. Therefore, a component returns to the agricultural fields under the form of biogas digestates, whereas the remainder ends up in biofuels by-products, which are typically used as animal feed. In the United States, where biofuels production is rapidly growing, this new stream of P-rich animal feed is creating concerns because it could undermine efforts towards feed management practices designed to reduce the P load in manure [Simpson et al., 2008]. This study estimates that 7,100  $\pm 31\%$  tP were contained in the stream of these by-products in 2011, which is noteworthy if compared with the total P in animal feed consumption of  $42,800 \pm 12\%$  tP. Nevertheless, the only datum available for this study was the generation of dried distiller's grains, that is to say bioethanol by-products, whereas the estimations of the biodiesel by-products and their final use relied on indirect information and assumptions.

The total input of P into the *Waste management* process, the time series of which is illustrated in Figure 2.8a, encompasses the entire generation of solid waste from house-

holds and similar sources, industry and green areas, fallen stock from Animal husbandry, total sewage sludge generated in wastewater treatment plants, disposed ashes generated in biomass thermal plants, and imported waste. Items that are not included are home-composted kitchen and green residues and organic by-products of the food industry directly re-used for the production of animal feed, which are both separately accounted as internal flows of the processes Consumption and Industry. The total amount of P transported in waste flows has constantly and significantly increased from 1990 to 2011. Population growth only partially explains this trend because the waste P expressed per capita was augmented from 1.2-1.5 kg P in 1990 to 2-2.2 kg P in 2011. The main determinant was sewage sludge, which increased from 3,000  $\pm 11\%$  tP in 1990 to 7,100  $\pm 6\%$  tP in 2011 due to population growth, incremented connection rates, and particularly, highly enhanced P removal in treatment plants. A less crucial but still important role was played by organic industrial waste, expressed by the growing food industry as well as disposed biomass ashes, which are indicative of the development of the bioenergy sector.

Figure 2.8b illustrates the fraction of the input wastes that are recovered, namely, meat and bone meal used as animal feed, materials (sewage sludge, compost and meat and bone meal) applied on agricultural fields, recycled wood and paper, compost applied in gardens and green areas, wastes valorized in biogas plants, and green residues used in biomass thermal plants. The fraction of waste used in landscaping activities is excluded because it does not necessarily imply a useful recovery of P, as well as the exported fraction for which the final management or disposal information is not always known. In the 1990s, approximately 5,000 tP in meat and bone meal per year were recovered as animal feed, but in 2001, this use was banned, which explains the drastic drop in Figure 8b. The total amount of recovered P showed a rapid increase after 2001, primarily due to the application of processed animal fat and meal on agricultural fields. Nevertheless, the ratio of total recovery with respect to the total input of waste has decreased from 55-60% in 1990 to approximately 40% in 2011, showing that in terms of P, waste generation has grown faster than recycling. A large fraction of the total waste P, approximately 40-45% on average, has been lost in cement kilns and landfills, with oscillations ranging between 35% and 55% in different years. In addition to the minor contribution of sewage sludge, the P is mostly lost in cement kilns through the use of meat and bone meal as a secondary fuel; therefore, this flow was almost null until 2001, when it suddenly reached notably high values of approximately 4,000 tP/y, which was gradually reduced to approximately 3,000 tP/y in more recent years (Fig. 2.8c). The amount of P deposited in landfills has not shown important variations in the period under study and has remained in the range of 4,000-6,000 tP/y, as illustrated in Figure 2.8d. Nevertheless, the material streams transporting it have varied considerably. In 1990, the P was mainly landfilled through direct disposal of sewage sludge, residual waste, and ashes of municipal waste incineration and sludge co-incineration, but in 2011, the main flows in order of importance were ashes of municipal waste incineration, sludge co-incineration, and thermal biomass plants, followed by sludge and municipal waste stabilized through mechanical-biological processes. The time series in Figure 2.8, which are depicted together with the uncertainty, show the differences in the level of information behind different flows within the Waste management process. The total input (Fig. 2.8a) presents less uncertainty than the recovery (Fig. 2.8b) and the loss in cement kilns (Fig. 2.8c) because the report for the generation of waste is more detailed in general than for its management. With respect to the loss in landfills (Fig. 2.8d), the uncertainty is significantly lower and decreasing in time because the disposal of organic waste and sewage sludge has been specifically regulated and monitored.

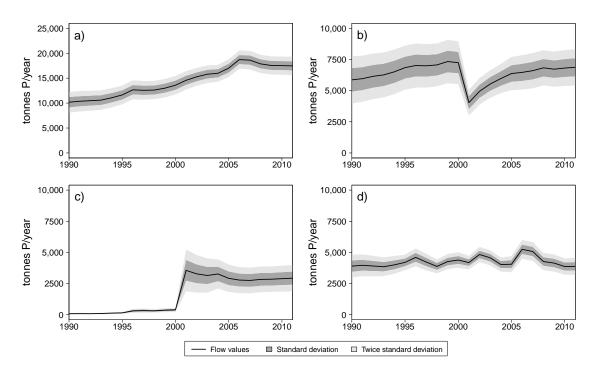


Figure 2.8: Time series of flows and stock change rates reproduced with their specific uncertainty: a) total P input in the *Waste management* process; b) total P recovery within the *Waste management* process; c) total P used in cement kilns; d) total disposal of P in landfills.

# 2.5 Conclusions

Even over a relatively short and stable period of 22 years, the national P budget of Austria has undergone significant and partially abrupt changes that have affected different compartments of the system. In the last decade, the bioenergy sector has gained increasing importance and it now affects large flows of raw materials, waste, and by-products. This rapid growth was however not accompanied by a sound monitoring of the associated flows, which shall be implemented because such flows could negatively affect the P balance in agriculture, especially in animal husbandry activities. With respect to P recovery, although in household wastes it has considerably increased from 20% in 1990 to 60% in 2011, in total wastes it has declined from 55-60% to approximately 40%. In

this context, the sudden and large loss of P contained in the meat and bone meal and diverted since 2001 to cement kilns, as a consequence of the Bovine Spongiform Encephalopathy crisis, represents a clear example of the impact that apparently unrelated modifications can exert on the P budget.

If the uncertainty of the results is considered in the analysis of temporal changes, extreme changes over the entire period are still clearly visible, but for half of the flows it is difficult to determine whether they remained constant or if moderate changes took place. The study also shows that the ability to detect annual variations is very sensitive to the uncertainty of the dataset and this is highly relevant for the monitoring of the performance of future measures or policies.

From a methodological point of view, this study has improved the model, in that it has aided the detection of systematic problems and of inconsistencies in the data sets, which could have not been revealed by a 1-year static study.

This exercise has demonstrated that it is feasible to carry out a time series of a detailed national budget, it has identified temporal developments with relevant implications for environmental, resources and waste management, and it has shown manifold methodological benefits and added values. Given all the aforesaid, this study suggests the need and usefulness for national authorities to set up similar resource accounting schemes, which could provide different services. First, they could identify and detect negative trends of flows, stocks, efficiencies and recovery rates. Second, they could monitor the effectiveness of regulations and directives. Last, they could aid in optimizing the national data collection, by identifying the specific contribution of different areas to the uncertainty of the system.

# Chapter 3

# Impact of reduced anthropogenic emissions and century flood on the phosphorus stock, concentrations and loads in the Upper Danube

# 3.1 Abstract

Patterns of changes in the concentration of total and soluble reactive phosphorus (TP, SRP) and suspended sediments at different flow levels from 1991 to 2013 in the Austrian Danube are statistically analyzed and related to point and diffuse emissions, as well as to extreme hydrological events. Annual loads are calculated with three methods and their development in time is examined taking into consideration total emissions and hydrological conditions. The reduction of point discharges achieved during the 1990s was well translated into decreasing TP and SRP baseflow concentrations during the same period, but it did not induce any change in the concentrations at higher flow levels nor in the annual transport of TP loads. A sharp and long-lasting decline in TP concentration, affecting all flow levels, took place after a major flood in 2002. It was still visible during another major flood in 2013, which recorded lower TP concentrations than its predecessor. Such decline could not be linked to changes in point or diffuse emissions. This suggests that, as a result of the flood, the river system experienced a significant depletion of its in-stream phosphorus stock and a reduced mobilization of TP rich sediments afterwards. This hypothesis is corroborated by the decoupling of peak phosphorus loads from peak maximum discharges after 2002. These results are highly relevant for the design of monitoring schemes and for the correct interpretation of water quality data in terms of assessing the performance of environmental management measures.

# 3.2 Introduction

The Danube is the second largest river in Europe and is responsible for almost 60% of the freshwater and for the majority of sediments and nutrients entering the Black Sea [Maksimovic and Makropoulos, 2002, Schreiber et al., 2005]. Consequently its elevated transport of phosphorus was identified as one of the main causes of the severe eutrophication that affected the sea during the 1980s and early 1990s [Kroiss et al., 2006]. The control of phosphorus pollution in the Danube is therefore of primary importance, both to sustain the ecological health of the river itself, and to reduce the loads transported downstream.

Austria accounts for 10% of the total area of the Danube Basin, which drains more than 96% of its territory [ICPDR, 2014a]. In the last 30 years the country has undertaken several efforts to reduce phosphorus emissions. In the 1980s the use of phosphates in detergents was dramatically reduced [Behrendt et al., 2005] and in the 1990s the Austrian edict BGBl. Nr. 180/1991, later replaced by the BGBl. Nr. 210/1996, introduced the mandatory removal of phosphorus in wastewater treatment plants (WWTP). Moreover, the Agri-Environmental Programme ÖPUL was launched in the year 2000 to address diffuse nutrient losses [BMLFUW, 2000]. In addition to anthropogenic changes, the Upper Danube Basin was also exposed to extreme hydrological conditions. After a succession of flood-poor decades, with the exception of a minor event in 1991, the Austrian Danube was hit first in August 2002 by a flood that, due to its extension and duration, was termed a "century flood", and then in June 2013 by one of the largest floods to have taken place in the last two centuries [Blöschl et al., 2013]. In 2003, on the contrary, the whole Basin experienced a pronounced drought, with below-average rainfalls and above-average temperatures. The relative precipitation recorded in Austria in that year, for example, corresponded to 74% of the long-term annual average [ICPDR, 2014c].

At the beginning of the 1990s an extensive monitoring network was set up by two independent agencies, the Austrian Federal Ministry of Agriculture, Forestry, Environment and Water Management (BMLFUW) and the International Commission for the Protection of the Danube River (ICPDR). It is thus feasible to examine in detail this period of combined anthropogenic changes and extreme hydrological regimes, which represent an exceptional opportunity for exploring interweaving causalities in a large river.

The link between improved wastewater phosphorus removal and decline in phosphorus concentration in European rivers has been demonstrated and quantified in previous studies [Neal et al., 2010, Raike et al., 2003]. Conversely, investigations dealing with episodic flood events, total emissions, and their relationship have focused on riverine loads (mass time<sup>-1</sup>), rather than on concentrations (mass volume<sup>-1</sup>). Behrendt and Opitz [2000] observed, across 100 basins in Europe, a considerable discrepancy between total emissions and measured transport loads, with the latter being significantly lower. They found that the difference increased as a function of the specific runoff of the basins. Similar findings were reported by Zessner and Kroiss [1999] for the Upper Danube. The reason for such discrepancies lies in the retention process, which has been increasingly recognized as a relevant mechanism to be further investigated and included in river basin models [de Klein and Koelmans, 2011, Venohr et al., 2011]. The retention of phosphorus takes place essentially through deposition and algae growth. The stock generated is then exposed to remobilization during flood events, when peaks of phosphorus are transported downstream and exported to river banks and flooded areas [Zessner et al., 2005, House et al., 1997, Dorioz and Ferhi, 1994, Johnson et al., 1976]. According to Stamm et al. [2014], this depletion is replaced by the deposition of particulate phosphorus during the recessional part of the storms, which represents a renewed internal source of baseflow phosphorus. Although this might hold true for small episodic events, the impact of a major flood could be more intense and exert a more profound impact on both the instream stock and the concentration, with consequences not only for the peak transport of phosphorus to downstream standing water bodies and to the lateral river system, but also for the river ecology itself.

In this study, time series of water quality data of the Upper Danube are examined to identify patterns of change in phosphorus concentration, and to link them to anthropogenically driven changes and to extreme hydrological conditions. The aim of the work is to assess the performance and effectiveness of environmental management strategies, and to investigate the short-term and long-term impact of large episodic events on the in-stream phosphorus concentration and on the stock of the river system.

To gain a deeper insight into the drivers of shifts in concentration, not only is total phosphorus (TP) analyzed, but also soluble reactive phosphorus (SRP, equivalent to orthophosphate) and suspended sediments (SS). SRP contributes to identifying the impact of point emissions, because it is typically the prevalent phosphorus species in WWTP effluents [Jarvie et al., 2006]. SS provides further information regarding diffuse pathways, because particulate-bound phosphorus is the predominant species transported by storm-dependent agricultural runoff and erosion processes [Withers and Jarvie, 2008].

# 3.3 Materials and methods

### 3.3.1 Data sets

The Danube was analyzed at its entrance into Austrian territory from Germany (Inflow) and at its exit from Austria (Outflow) (Fig.3.1). This enables the phosphorus contribution within the Austrian catchment to be determined.

The study used a collection of different data sources, namely the  $H_2O$  database created and maintained since 1991 by BMLFUW [BMLFUW, 2014c], and two ICPDR databases, one obtained through a first campaign performed from 1992 to 1998 (Bucharest Declaration data set) and the other through the Transnational Monitoring Network (TNMN) launched in 1996 [ICPDR, 2014b]. In addition, for the Outflow a specific sample collected during the flood of August 2002 [Zessner et al., 2005] and a data set of semi-continuous measurements for the entire year 2013 [BMLFUW, 2013a] were available (Tables 3.1 and 3.2). Since 1991 the monitoring of surface and groundwater water quality in Austria has been regulated by federal legislation (BGBL Nr. 338/1991 re-

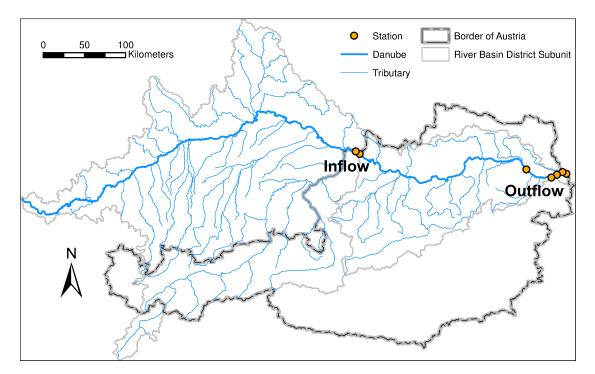


Figure 3.1: Map of the Upper Danube Basin and of the monitoring stations located at the Austrian Inflow and Outflow.

placed by BGBl. II Nr. 479/2006) that specifies the standard analytical procedures to be followed for each parameter. With respect to total phosphorus and phosphorus compounds, at the beginning of the 1990s analyses were required to comply with the Standard ÖNORM M 6237:1986, which was then replaced by ISO 6878:1998 and later revised by ISO 6878:2004. These standards all maintained the spectrometric determination using ammonium molybdate as basis. Although accredited laboratories are entitled to apply different methods, they are required to prove their equivalence to the standard procedures. These protocols were applied by all data sources considered here. This ensures a substantial consistency of the analytical methods throughout the studied time period, although some minor variations cannot be entirely excluded. In addition, the analyses of each sample have been run in triplicate in accordance with the aforementioned protocols, with the exception of the data set of Zessner et al. [2005], which relies on single tests.

### **3.3.2** Data sets subdivision in time periods and flow intervals

Phosphorus concentration and species fractionation can vary largely as a function of flow levels due to differing primary pathways and driving natural processes. TP concentration generally rises at increasing flow levels due to the higher transport of particulate-bound phosphorus in more turbulent conditions [Zessner and Kroiss, 1999]. On the contrary,

Station	Station code	Distance from mouth [km]	Sampling period	Sampling frequency
$H_2O$ - Jochenstein	FW40607017	2204	Jul. 1991 - June 1995	monthly
$H_2O$ - Obernzell	FW40607037	2210	Feb. 1996 - Dec. 2006	monthly
ICPDR Bucharest - Jochenstein	D2130	2204	Jan. 1992 - Feb. 1998	monthly
ICPDR TNMN - Jochenstein Austria	AT1	2204	Jan. 1996 - Dec. 2011	monthly (1996-2006) 3 per month (2007-2011)
ICPDR TNMN - Jochenstein Germany	D02	2204	Jan. 1996 - Dec. 2011	bi-weekly

Table 3.1: Data sets employed for the Inflow of the Danube into Austria.

Table 3.2: Data sets employed for the Outflow of the Danube from Austria.

Station	Station code	Distance from mouth [km]	Sampling period	Sampling frequency
$H_2O$ - Wolfsthal	FW31000027	1874	July 1991 - Dec. 2002	monthly (1991-2000); bi-weekly (2001, 2002)
$H_2O$ - Deutsch Altenburg	FW31000017	1887	July 1991 - May 1995	monthly
$H_2O$ - Wildungsmauer	FW31000187	1895	Jan. 1996 - Dec. 2012	monthly
ICPDR Bucharest - Wolfsthal	D1840	1873	Jan. 1992 - Feb. 1994	monthly
ICPDR TNMN - Wolfsthal	AT4	1874	Jan. 1996 - Dec. 2005	monthly (1996-1998); bi-weekly (1999-2005)
ICPDR TNMN - Hainburg	AT6	1879	Jan. 2006 - Dec. 2011	bi-weekly
Zessner et al. [2005]	-	1930	Aug. 2002	2-8 per day
BMLFUW - Wolfsthal	-	1879	Jan. 2013 - Dec. 2013	bi-hourly

SRP concentration is expected to be higher at baseflow conditions, because its primary pathway is the quasi-constant WWTP effluent discharge, which becomes more heavily diluted at higher flows [Jarvie et al., 1997]. To preserve the information contained in these differences, the flow levels were subdivided into intervals, following the flow duration curve scheme suggested by EPA [2007]. The flows were categorized according to the percentage of exceedance, as follows: *High flows* (0-10%), *Moist conditions* (10-40%), *Mid-range flows* (40-60%), *Dry conditions* (60-90%), and *Low flows* (90-100%). The availability of data during the two flood events in 2002 and 2013 at the Outflow also enables the investigation of the differences and changes in time in high flow conditions, which are especially important for the transport of phosphorus. Thus, the interval *High flows*, *Very high flows*, and *Extremely high flows*, 10,570 m<sup>3</sup> s<sup>-1</sup> being the maximum daily discharge measured within the time series. The calculation of the cumulative frequency was based on two data sets of daily mean discharges measured at the same locations,

covering the periods 1985-2011 and 1977-2013 for the Inflow and Outflow, respectively [BMLFUW, 2014b]. The flow intervals obtained following this procedure are shown in Table 3.3.

Location	Low flows	Dry conditions	Mid-range flows	Moist conditions	High flows	Very high flows	Extremely high flows	
Inflow	< 800	800-1,100	$1,\!100\text{-}1,\!400$	1,400-2,100	> 2,100	-	-	
Outflow	< 1,100	1,100-1,600	1,600-2,000	2,000-3,000	3,000-5,500	5,500-8,000	> 8,000	

Table 3.3: Flow intervals  $(m^3 s^{-1})$  for the Inflow and the Outflow.

Studies have shown that low frequency sampling can lead to large errors in the calculation of riverine phosphorus loads [Johnes, 2007, Cassidy and Jordan, 2011]. Similar findings were presented by Skarbøvik et al. [2012] with regard to estimating the mean concentration of suspended sediments, which could also be extended to particulate contaminants like phosphorus. Thus, the fact that the available data sets mostly rely on monthly or bi-weekly measurements impedes a reliable analysis of rapid inter-annual variations. This study focuses instead on longer-term changes, for which purpose the time series was subdivided into periods, based on the rationales described in Table 3.4.

Period	Time range	Rationale				
T1	Jan 1991 - Dec 1995	Starting phase of the implementation of the regulation on wastewater phosphorus				
		removal				
<b>T2</b>	Jan 1996 - Aug 2002	Mature phase of implementation of the regulation on wastewater phosphorus removal; flood in August 2002				
<b>T</b> 3	Sep 2002 - Dec 2007	Short-term effects of 2002 flood				
<b>T</b> 4	Jan 2008 - Dec 2012	Long-term effects of 2002 flood				
$\mathbf{T5}$	Jan - Dec 2013	Flood in June 2013; semi-continous data				

Table 3.4: Time periods and rationale for their categorization.

### 3.3.3 Consistency and combination of the data sets

The data sets were statistically compared, firstly to verify the consistency among different stations and monitoring agencies, and secondly to determine the appropriateness of their combination. The K-sample Anderson-Darling test [Scholz and Stephens, 1987] was chosen to test whether at each flow interval and time period the different samples can be considered to belong to the same distribution. It was applied to the three flow

ranges with highest data availability, namely Dry conditions, Mid-range flows, and Moist conditions, with the assumption that they be representative for the whole flow duration curve. For the Outflow and the period T2, the samples within High flows were also tested to include the data set of Zessner et al. [2005]. The standard statistical significance levels associated to p values were adopted: significant (p < 0.05), very significant (p < 0.01), and highly significant (p < 0.001).

As shown in Tables 3.5 and 3.6, the data sets generally present a good level of consistency (i.e. p values greater than 0.05). Therefore, none of the data sets was dismissed and all were merged to obtain the final samples shown in Tables 3.7 and 3.8. This notwithstanding, the analyses of the study were reiterated alternately removing the data sets that presented some discrepancies, in order to assess their actual impact on the results and on the conclusions.

Table 3.5: Results of the K-sample Anderson-Darling test applied to the TP data sets available for the Inflow;  $H_0$ : samples are drawn from the same distribution. Statistically significant differences are indicated in bold and sample size in parentheses.

Period	Data set A Data set B		Dry conditions	Mid-range flows	Moist conditions
T1	$H_2O$ - Jochenstein	ICPDR Buch Joch.	p > 0.05 (10 - 7)	p > 0.05 (10 - 5)	p > 0.05 (12 - 7)
	$\rm H_2O$ - Obernzell	ICPDR Buch Joch.	p > 0.05 (16 - 8)	p > 0.05 (21 - 7)	p > 0.05 (26 - 5)
	$\rm H_2O$ - Obernzell	ICPDR TNMN - Joch. A01	p > 0.05 (16 - 19)	p > 0.05 (21 - 22)	p > 0.05 (26 - 20)
T2	$H_2O$ - Obernzell	ICPDR TNMN - Joch. D02	p > 0.05 (16 - 30)	p > 0.05 (21 - 40)	p > 0.05 (26 - 52)
	ICPDR Buch Joch.	ICPDR TNMN - Joch. A01	p > 0.05 (8 - 19)	p > 0.05 (7 - 22)	p > 0.05 (5 - 20)
	ICPDR Buch Joch.	ICPDR TNMN - Joch. D02	p > 0.05 (8 - 30)	p > 0.05 (7 - 40)	p > 0.05 (5 - 52)
	ICPDR TNMN - Joch. A01	ICPDR TNMN - Joch. D02	p > 0.05 (19 - 30)	${f p} < {f 0.05}\ (22$ - 40)	p > 0.05 (20 - 52)
	$H_2O$ - Obernzell	ICPDR TNMN - Joch. A01	p > 0.05 (13 - 27)	<b>p</b> < <b>0.001</b> (8 - 19)	p > 0.05 (15 - 24)
Т3	$\rm H_2O$ - Obernzell	ICPDR TNMN - Joch. D02	p > 0.05 (13 - 26)	${f p} < {f 0.05}\ (8$ - 5)	${f p} < {f 0.01}\ (15$ - 6)
	ICPDR TNMN - Joch. A01	ICPDR TNMN - Joch. D02	p > 0.05 (27 - 26)	p > 0.05 (19 - 5)	p > 0.05 (24 - 6)
Τ4	ICPDR TNMN - Joch. A01	ICPDR TNMN - Joch. D02	p > 0.05 (48 - 47)	p > 0.05 (25 - 25)	p > 0.05 (37 - 37)

Table 3.6: Results of the K-sample Anderson-Darling test applied to the TP data sets
available for the Outflow; $H_0$ : samples are drawn from the same distribution. Statisti-
cally significant differences are indicated in <b>bold</b> and sample size in parentheses.

Period	Data set A	Data set B	Dry conditions	Mid-range flows	Moist conditions	High flows
	ICPDR Buch Wolfsthal	$\rm H_2O$ - Wolfsthal	<b>p</b> < <b>0.01</b> (10 - 16)	p > 0.05 (5 - 6)	p > 0.05 (9 - 13)	-
T1	ICPDR Buch Wolfsthal	$H_2O$ - Deutsch-Altenburg	<b>p</b> < <b>0.001</b> (10 - 10)	p > 0.05 (5 - 6)	p > 0.05 (9 - 11)	-
	$\rm H_2O$ - Wolfsthal	$H_2O$ - Deutsch-Altenburg	p > 0.05 (16 - 10)	p > 0.05 (6 - 6)	p > 0.05 (13 - 11)	-
	$H_2O$ - Wolfsthal	ICPDR TNMN - Wolfsthal	p > 0.05 (29 - 31)	p > 0.05 (33 - 32)	p > 0.05 (44 - 46)	p > 0.05 (14 - 16)
	$H_2O$ - Wolfsthal	$\rm H_2O$ - Wildungsmauer	p > 0.05 (29 - 20)	p > 0.05 (33 - 23)	p > 0.05 (44 - 19)	p > 0.05 (14 - 7)
T2	ICPDR TNMN - Wolfsthal	$\rm H_2O$ - Wildungsmauer	p > 0.05 (31 - 20)	p > 0.05 (32 - 23)	p > 0.05 (46 - 19)	p > 0.05 (16 - 7)
	$\rm H_2O$ - Wolfsthal	Zessner et al. [2005]	-	-	-	p > 0.05 (14 - 7)
	ICPDR TNMN - Wolfsthal	Zessner et al. [2005]	-	-	-	p > 0.05 (16 - 7)
	$\rm H_2O$ - Wildungsmauer	Zessner et al. [2005]	-	-	-	p > 0.05 (7 - 7)
	$H_2O$ - Wolfsthal	ICPDR TNMN - Wolfsthal	-	-	p > 0.05 (4 - 24)	-
	$\rm H_2O$ - Wolfsthal	$\rm H_2O$ - Wildungsmauer	-	-	p > 0.05 (4 - 17)	-
Т3	ICPDR TNMN - Wolfsthal	$\rm H_2O$ - Wildungsmauer	p > 0.05 (26 - 17)	<b>p</b> < <b>0.05</b> (14 - 13)	p > 0.05 (24 - 17)	-
	$\rm H_2O$ - Wolfsthal	ICPDR TNMN - Hainburg	-	-	p > 0.05 (4 - 14)	-
	ICPDR TNMN - Wolfsthal	ICPDR TNMN - Hainburg	p > 0.05 (26 - 10)	p < 0.05 (14 - 10)	p > 0.05 (24 - 14)	-
	$\rm H_2O$ - Wildungsmauer	ICPDR TNMN - Hainburg	p > 0.05 (17 - 10)	p > 0.05 (13 - 10)	p > 0.05 (17 - 14)	-
T4	$\rm H_2O$ - Wildungsmauer	ICPDR TNMN - Hainburg	p > 0.05 (25 - 32)	p > 0.05 (13 - 21)	p > 0.05 (16 - 25)	-

Period	Determi- nand	Low flows	Dry cond.	Mid- range flows	Moist cond.	High flows
	TP	14	17	15	19	5
T1	SRP	8	14	12	14	4
	$\mathbf{SS}$	14	17	15	19	5
	TP	40	73	90	103	50
T2	SRP	40	73	90	103	50
	$\mathbf{SS}$	40	73	90	103	50
	TP	93	66	32	45	10
T3	SRP	93	66	32	45	10
	$\mathbf{SS}$	72	52	24	39	9
	TP	46	95	50	74	34
$\mathbf{T4}$	SRP	46	95	50	74	34
	$\mathbf{SS}$	22	47	25	37	16

Table 3.7: Number of paired instantaneous flow and TP, SRP, and SS values for the Inflow, after merging the data sets.

Table 3.8: Number of paired instantaneous flow and TP, SRP, and SS values for the Outflow, after merging the data sets.

Period	Determi- nand	Low flows	Dry cond.	Mid- range flows	Moist cond.	High flows	Very high flows	Extr. high flows
	TP	10	36	17	33	9	0	0
T1	SRP	7	33	15	29	8	0	0
	$\mathbf{SS}$	8	36	17	33	9	0	0
	TP	13	80	88	110	44	20	13
T2	SRP	13	80	88	110	44	19	13
	$\mathbf{SS}$	13	80	88	110	44	19	13
	TP	29	53	39	59	20	3	0
$\mathbf{T3}$	SRP	29	48	36	55	20	3	0
	$\mathbf{SS}$	29	53	39	59	20	3	0
	TP	11	57	34	41	13	0	0
$\mathbf{T4}$	SRP	10	47	28	35	12	0	0
	$\mathbf{SS}$	11	57	34	41	13	0	0
	TP	0	771	1178	1119	601	46	48
$\mathbf{T5}$	$\operatorname{SRP}$	0	852	1301	1175	603	46	48
	$\mathbf{SS}$	-	-	-	-	-	-	-

### 3.3.4 Analysis of change in time

The combined data sets (Tables 3.7 and 3.8) were statistically analyzed to examine the behavior in time of the TP, SRP and SS mean concentrations. The null hypothesis  $H_0$  was that the mean concentrations calculated at every flow range did not change significantly from each period to the following one. It is therefore a step trend hypothesis, which is tested through a *t*-test for the significance of the difference between the means of two independent samples [Hirsch et al., 1991]. In view of the diverse sample size and possibly unequal variance, the Welch's *t*-test was selected [Welch, 1947].

### 3.3.5 Load calculation

The calculation of annual TP loads was carried out with three different methods: M1 - Linear, M2 - ICPDR, and M3 - Flow intervals.

### Method M1 - Linear

The first calculation procedure (Eq.3.1) is a widely used method, and was applied by Littlewood and Marsh [2005] and by Johnes [2007] as the reference to test other methodologies.

$$L = \frac{K \sum_{i=1}^{n} (C_i Q_i)}{\sum_{i=1}^{n} (Q_i)} \cdot \bar{Q_r}$$
(3.1)

Annual loads L are calculated as the product of sampled instantaneous concentration  $C_i$  and discharge  $Q_i$  (n is the number of samples in a year), divided by the sum of sampled discharges and multiplied by the average annual discharge  $\bar{Q}_r$ , K being a factor to account for measurement units and the duration of the period.

This method has a considerable shortcoming due to the underlying assumption of a linear relationship between load and discharge. As this is usually depicted by an exponential function, Equation 3.1 will tend to deliver overestimations when data collected at high flow conditions or during storm events are included, as demonstrated by Cassidy and Jordan [2011].

#### Method M2 - ICPDR

Equation 3.2 shows the calculation procedure officially selected and applied by ICPDR [ICPDR, 2000].

$$C_m = \frac{\sum_{i=1}^m (C_i Q_i)}{\sum_{i=1}^m Q_i} \qquad L_m = K C_m Q_m \qquad L = \sum_{m=1}^{12} L_m \tag{3.2}$$

The method is based on the calculation of monthly loads  $L_m$ , which are obtained as the product of average monthly discharge  $Q_m$  and average monthly concentration  $C_m$  (m is the number of samples per month), K being a factor to account for measurement units and the number of days in each month.  $C_m$  is the product of the measured concentrations and discharge values, divided by the sum of sampled discharges. In comparison to M1 - *Linear*, this procedure adds the inclusion of seasonality. Therefore, it indirectly takes into account, to a certain degree, the relationship between TP concentration and discharge.

### Method M3 - Flow intervals

A specific calculation procedure was developed on the one hand to appropriately consider the relationship between TP concentration and discharge, and on the other hand to examine the impact that the shifts in concentration exerted on the total riverine transport. As shown in Equation 3.3, the annual loads L are obtained as the sum of daily loads, calculated as the product of each daily discharge and the mean concentration of the respective flow interval  $(Q_l, Q_d, Q_m, Q_{mo}, Q_h, Q_v, Q_e)$  being daily discharge values and  $\mu_l, \mu_d, \mu_m, \mu_{mo}, \mu_h, \mu_v, \mu_e$  mean concentrations at Low flows, Dry conditions, Midrange flows, Moist conditions, High flows, Very high flows, and Extremely high flows, respectively), where K is a factor to account for measurement units. For each year, the mean concentrations of the corresponding time period are applied.

$$L = K\left(\sum_{l,d,m,mo,h,v,e=0}^{n} (Q_{l}\mu_{l}) + (Q_{d}\mu_{d}) + (Q_{m}\mu_{m}) + (Q_{mo}\mu_{mo}) + Q_{h}\mu_{h}\right) + (Q_{v}\mu_{v}) + (Q_{e}\mu_{e})) \quad (3.3)$$

The results of the three methods are compared to the loads that ICPDR has calculated, based on M2 - *ICPDR* and using exclusively the TNMN data sets [ICPDR, 2000, 2001, 2002, 2003, 2004, 2005, 2006, 2007, 2008, 2009, 2010, 2011].

### **3.4** Results and discussion

### 3.4.1 Shifts in phosphorus concentration

### Reduction of point emissions during the 1990s

The analysis at the Inflow shows that, from the first to the second half of the 1990s, a highly significant decline of the mean TP concentration took place at *Low flows*, from 0.10 to 0.07 mg L<sup>-1</sup>, whereas at other flow ranges no variation was found (Fig.3.2a). At the Outflow the change was more pronounced, with a clear reduction of the TP concentration at *Low flows* (from 0.12 to 0.08 mg L<sup>-1</sup>), *Dry conditions* (from 0.10 to 0.09 mg L<sup>-1</sup>), and *Mid-range flows* (from 0.13 to 0.08 mg L<sup>-1</sup>) (Fig. 3.2b). The analysis of SRP depicts similar patterns. At the Inflow the calculated mean SRP concentrations at *Low flows* and *Dry conditions* both decreased from 0.05 mg L<sup>-1</sup> to 0.03 mg L<sup>-1</sup>, but this was found to be statistically significant only for the latter, whereas for *Low flows* the samples were too small and displayed too much variability (Fig. 3.3a). At the Outflow the decline was larger and broader, with a reduction at *Low flows* from 0.09 to 0.04 mg L<sup>-1</sup>, at *Dry conditions* from 0.06 to 0.03 mg L<sup>-1</sup>, at *Mid-range flows* from 0.05 to 0.03 mg L<sup>-1</sup>, and at *Moist conditions* from 0.04 to 0.03 mg L<sup>-1</sup> (Fig. 3.3b).

Such shifts reflect the improvements achieved during this period in the efficiency of phosphorus removal in WWTP. Austrian WWTP gradually increased their average transfer of phosphorus contained in receiving wastewater to sewage sludge from 50% in 1995 to 82% in 2001 [BMLFUW, 1996, 2008]. In the first half of the 1990s, German WWTP achieved the target of maximum 1 mg  $L^{-1}$  TP in the effluent, equivalent to an approximately 80% removal rate [TMLNU, 2009], which explains the delayed decrease of TP and SRP concentrations at the Outflow when compared to the Inflow. The link between the diminished TP concentration and the reduced point emissions is supported by the fact that the decline was detected only at low up to mid-flow conditions, whereas at higher flows it remained constant or even increased, since quasi-constant effluent discharge from WWTP becomes less diluted in drier conditions. This causality is evidenced by the reduction of SRP, a typically dominant phosphorus species in WWTP effluents.

The decline of TP and SRP concentrations in low flow conditions, as a result of the reduced phosphorus load in WWTP effluents, is in accordance with the findings of Neal et al. [2010], which showed a consistent decrease of SRP in the River Thames, induced by the augmented phosphorus removal in WWTP and mostly visible at baseflow conditions. Although the observed trend was similar, the range of variation was very different, since the SRP concentration in the Thames started from 1.6 mg L<sup>-1</sup> in 1997 and reached approximately 0.4 mg L<sup>-1</sup> in 2006, still 10 times higher than in the Upper Danube. The reasons for these large differences may include the higher population density in the catchment of the River Thames and the still relatively low phosphorus removal rate in the UK (approximately 57% in the year 2009) [Cooper and Carliell-Marquet, 2013].

#### Consequences of the major flood of August 2002

The analysis shows that, after the major flood of August 2002, a significant and longlasting decline in the TP concentration occurred. The mean TP concentration at the Inflow varied from 0.07 mg  $L^{-1}$  at Low flows to 0.13 mg  $L^{-1}$  at High flows before the flood, but ranged from 0.06 mg  $L^{-1}$  to 0.08 mg  $L^{-1}$  afterwards (Fig. 3.2a). At the Outflow this effect was even more visible. The mean TP concentrations reached very high values of 0.23, 0.54 and 1.25 mg  $L^{-1}$  at High flows, Very high flows and Extremely high flows, respectively. After the flood, the range at all flow levels was reduced to 0.05-0.08 mg  $L^{-1}$  (Fig. 3.2b). The decline was not only sharp, but also enduring. At both locations the T4 period (2008-2012) was characterized by only slight and inconsistent increases, found to be significant only at *Moist conditions* for the Inflow (Fig. 3.2a) and at *High* flows for the Outflow (Fig. 3.2b). Furthermore, the analysis of the semi-continuous data set collected at the Outflow in 2013 shows that the TP concentration persisted at the same low levels at Dry conditions and Mid-range flows (Fig. 3.2b). It increased at Moist conditions and High flows, but never reaching the higher values measured before and during the 2002 flood. The repetition of the analyses alternately excluding the data sets that had presented problems of inconsistencies led to the same results, meaning that the few detected discrepancies were offset by the extent of the changes in time. The only effect on the results was a slightly lower statistical significance caused by the reduced size of the samples.

The observed shift in TP concentration cannot be explained through a reduction of point emissions, because in 2002 both Germany and Austria had already achieved a very high removal rate of phosphorus from wastewater -more than 80%-, leaving scope for only minor further improvements. This is confirmed by the behavior of the mean SRP concentration, which at both locations remained almost constant in the range 0.03-0.04 mg L<sup>-1</sup> at all flow intervals (Fig. 3.3a and 3.3b).

On the other hand, the analysis of the SS (Fig. 3.4a and 3.4b) does not provide any evidence that, following the flood, the Danube experienced a reduced turbidity and transport of sediments, which could have explained the decrease of TP concentration, especially at high flow conditions. Therefore, it was the phosphorus content of the sediments that declined. The very high amount of phosphorus accumulated through decades of fertilization in Austrian agricultural soils [Egle et al., 2014b] excludes the possibility that the phosphorus content of eroded sediments decreased significantly. According to Behrendt et al. [2005], diffuse phosphorus emissions in the Danube river system remained relatively constant during the last decades of the 20<sup>th</sup> century. With respect to the early 2000s, Zessner et al. [2011b] carried out a detailed study aimed at assessing the impact of the agri-environmental program ÖPUL, launched in 2000, on nutrient emissions in Upper Austria. The most important measures implemented were the use of winter cover crops and the strip-till practice, both aimed at preventing soil loss. Given these measures can decrease emissions up to 50% - 70% and that they were applied respectively to 20%and 10% of available arable land, it was estimated that their implementation led to a total decrease of approximately 10% of the phosphorus emissions. However, this 10%reduction regards exclusively the emissions associated with agricultural erosion, and this in turn accounts for approximately 40% of total diffuse phosphorus emissions (the rest being mainly split among urban runoff, natural erosion and groundwater pathways). In view of the general implementation status of the OPUL program, these results are also applicable to the rest of Austria. Therefore, the contribution of the changes in diffuse emissions to determining such a steep decline of the in-stream TP concentration can be regarded as unimportant.

As a result, the only convincing explanation for the sharp and enduring decline of TP concentration in the Danube after the flood is a reduced mobilization of phosphorus within the river, resulting from a combined effect of the strong event and lower point emissions. It can be hypothesized that the flood intensely scoured the river bed, removing the pool of phosphorus primarily embedded in algae mass. Although algae represent a minor fraction of the total stream sediments, their average phosphorus content is considerably higher than that of soil particles, the first being approximately 1-2% [Borchardt and Azad, 1968, Benedini and Tsakiris, 2013] and the latter ranging between 0.02-1.14% [Koljonen and Darnley, 1994, Noll et al., 2009]. This explains the strong reduction of TP concentration without significant changes in the total SS. As a result of point discharges reduction, the lower availability of soluble and easily available phosphorus hindered rapid algae growth, delaying the regeneration of the internal stock, which clarifies the long duration of the TP decline. These mechanisms are presented schematically in Figure 3.5.

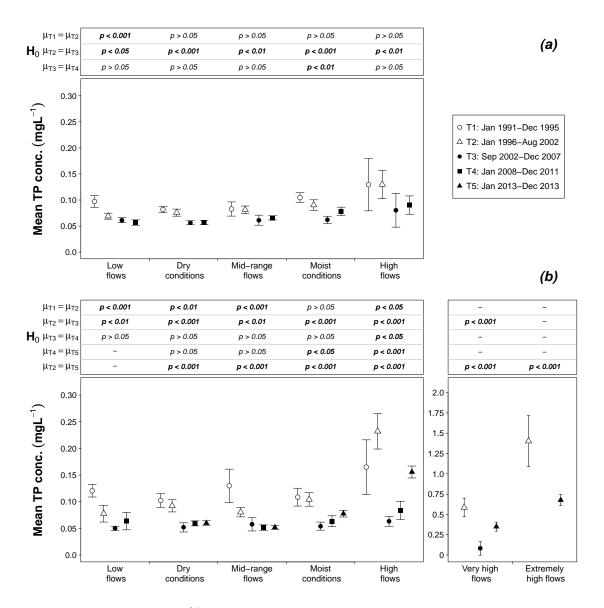


Figure 3.2: Mean and 95% Confidence Interval of the total phosphorus concentration at each flow interval and time period at: a) the Inflow and b) the Outflow; significance level of the difference between mean values  $\mu$  among time periods (Welch's *t*-test) are also shown. Statistically significant differences are indicated in bold.

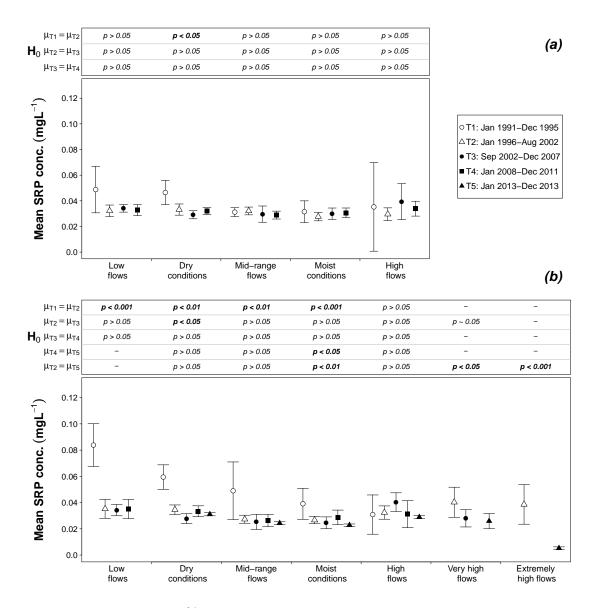


Figure 3.3: Mean and 95% Confidence Interval of the soluble reactive phosphorus concentration at each flow interval and time period at: a) the Inflow and b) the Outflow; significance level of the difference between mean values  $\mu$  among time periods (Welch's *t*-test) are also shown. Statistically significant differences are indicated in bold.

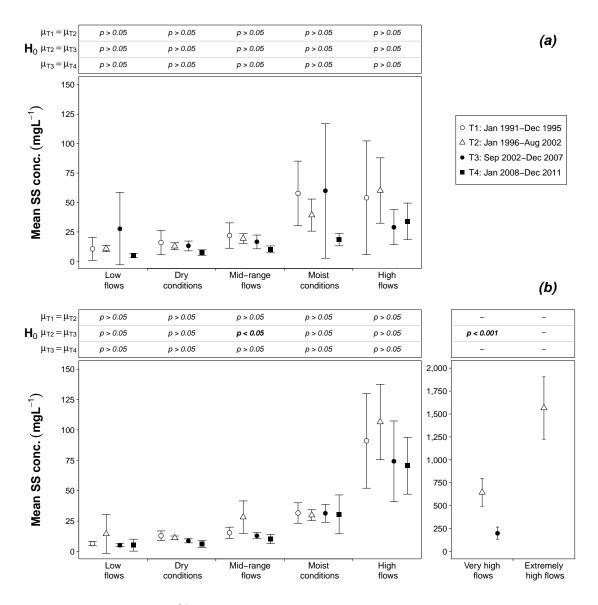


Figure 3.4: Mean and 95% Confidence Interval of the suspended sediments concentration at each flow interval and time period at: a) the Inflow and b) the Outflow; significance level of the difference between mean values  $\mu$  among time periods (Welch's *t*-test) are also shown. Statistically significant differences are indicated in bold.

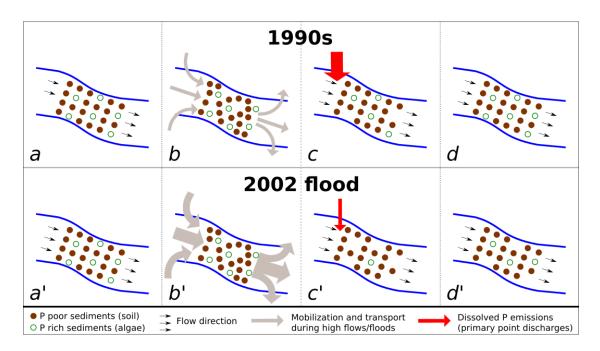
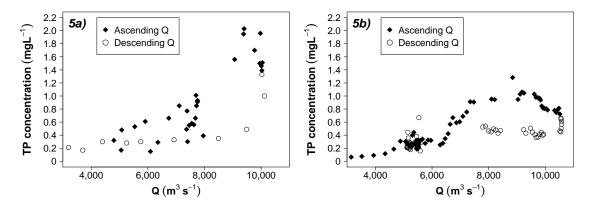


Figure 3.5: Schematic illustration of the main processes affecting the phosphorus instream pool during the observed time series: a) initial pool during the 1990s; b) transport and mobilization during high flows and minor flood events; c) small depletion of phosphorus rich sediments and high levels of dissolved phosphorus emissions; d) quick recovery of in-stream pool due to high availability of soluble reactive phosphorus for algae growth; a') initial pool before 2002 flood; b') transport and mobilization during 2002 flood; c') strong depletion of in-stream phosphorus rich sediments and low dissolved phosphorus emissions; d') slow recovery of in-stream pool due to low availability of soluble reactive phosphorus for algae growth.

### Comparison of major floods in 2002 and 2013

Figure 3.6 presents the behavior of the TP concentration at the Outflow during the August 2002 and June 2013 floods. In both events the TP concentration at a given discharge was higher during the rising limb of the hydrograph and lower during the falling one. This pattern, well known as clockwise hysteresis effect [Bowes et al., 2005, House and Warwick, 1998, McDiffett et al., 1989], proves the high levels of mobilization and short-term depletion effects that floods exert on the in-stream phosphorus pool. Furthermore, at *Extremely high flows* the mean TP concentration measured in 2013 (0.7 mg L<sup>-1</sup>) was considerably lower than it was in 2002 (1.4 mg L<sup>-1</sup>), despite the fact that the maximum daily discharge of 10,570 m<sup>3</sup> s<sup>-1</sup> reached during the 2013 event was much higher than the 10,116 m<sup>3</sup> s<sup>-1</sup> recorded in 2002. This supports the hypothesis that there was an enduring decline in the internal phosphorus stock in the river. This is only a hypothesis, which should be tested through further research. In this respect, the recent flood of 2013 provides an ideal opportunity for further investigations into the river to



compare the consequences of the two events.

Figure 3.6: TP concentration measured at the Outflow at ascending and descending discharges during: 5a) August 2002 flood and 5b) June 2013 flood.

# 3.4.2 Annual P loads

### Inflow

If compared to the large variations found by Johnes [2007] and Cassidy and Jordan [2011] in their study of different calculation methods and sampling frequencies, the annual TP loads calculated here present a substantial consistency. This is primarily due to the combination of data sets, with a consequent increase of the samples size, and also to the fact that the Danube is a large river with less pronounced flow dynamics than the smaller streams investigated in the aforementioned studies. Nevertheless, a few notable discrepancies are also detected. As depicted in Figure 3.7, the results obtained with methods M2 - ICPDR and M3 - Flow intervals are highly consistent, whereas M1 -Linear presents larger fluctuations within the period T2 (Jan. 1996 - Flood 2002). All methods capture the episodic event of 1995 well, but only M1 - Linear calculates a spike load in correspondence of the flood of 1999, and none of them delivers a higher load in 2002. This confirms that regular, low frequency monitoring is not a sufficient basis on which to calculate loads in years with important flood events. To obtain an indication of the extent of these underestimations, M3 - Flow intervals was also applied with a correction to take into account the high phosphorus concentrations at flood events. The high flow levels were split into *High flows*, Very high flows, and Extremely high flows, following the same procedure as for the Outflow. The mean concentrations applied for these intervals in each period were obtained by adapting the values calculated for the Outflow, according to the relationship between the mean concentrations at *Mid-range* flows at the two locations. Although this calculation is prone to large uncertainties, its outcomes are highly consistent with all methods for the year 1995 and with M1 - Linear in 1999, and in addition they present the expected higher load in 2002.

Despite the abovementioned discrepancies, all results depict the same trend, composed of relatively high and fluctuating loads until 2002, followed by a sudden fall in 2003 and by a stable low level thereafter. The calculations published by ICPDR since 2000 show a similar pattern, with two remarkable exceptions in the years 2005 and 2006, for which no plausible explanation could be found.

As shown in Figure 3.8, until the year 2002, there was a strong correspondence between peaks of annual TP load and peaks of maximum daily discharge. Afterwards maximum discharges as high or even higher than pre-2002 were recorded, but were no longer coupled with high TP loads. The mean annual discharge after 2002 was slightly lower than in the period 1995-2002, but not lower than in the time prior to 1995. It can be concluded that the low level of annual TP loads after 2002 was not caused by an alteration of the hydrological regime, but instead by reduced mobilization of phosphorus in the river. This supports the hypothesis that a depletion of the internal stock occurred as a result of the flood and that its delayed regeneration was a consequence of the reduced discharges of soluble phosphorus.

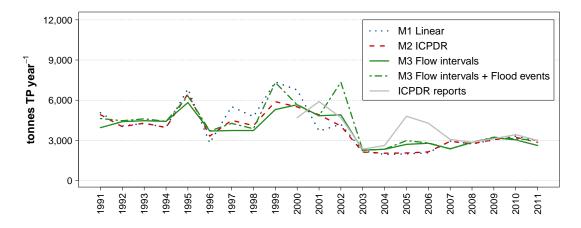


Figure 3.7: Time series of annual TP loads at the Inflow, obtained with 3 different methods and compared to the calculations published by ICPDR.

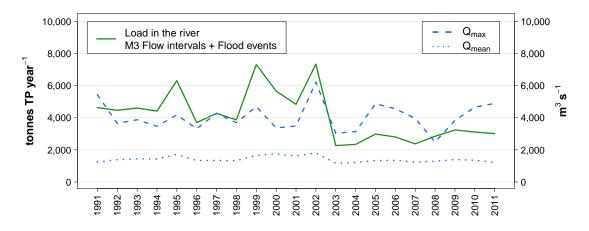


Figure 3.8: Time series of annual TP river loads, maximum daily discharge and mean daily discharge at the Inflow.

### Outflow

As shown in Fig. 3.9, during the period T2, M1 - Linear and M2 - ICPDR show more pronounced fluctuations than M3 - Flow intervals, because the latter applies average mean concentration over the whole period, whereas the other methods only consider the concentrations measured in each year. The largest discrepancy affects the load transported in 2002. Whereas M2 - ICPDR and M3 - Flow intervals lead to the same result, the outcome of M1 - Linear is twice as high, because it includes a stratified data set combined with the linear approximation of the algorithm.

The results altogether present the same trend that was observed for the Inflow, namely high and fluctuating loads until 2002, followed by a collapse in 2003 and a long-lasting low level thereafter. The year 2013 presents a renewed high load, expected due to the major flood in June, but still lower than the loads in 1999 and 2002. These results are consistent with the calculations published by ICPDR, with exception of 2002, for which ICPDR estimated a much lower load, because it did not include the data collected during the flood event.

As observed for the Inflow, the patterns of annual riverine TP loads and of maximum discharges were clearly decoupled after 2002 (Fig. 3.10). In 2009 the two curves show again a simultaneous peak, but whereas the maximum discharge is even higher than the ones recorded before 2002, the TP load is still considerably lower. The same consideration holds true, although to a smaller degree, for the comparison between 2002 and 2013.

In Figure 3.10 the developments of riverine TP loads are further compared to total phosphorus emissions from the German and Austrian catchments. These were estimated using the results and background information of Zessner et al. [2011a] as a basis. Their detailed calculation of the emissions for the average period 2001-2006 was adjusted on a yearly scale in consideration of the development of point emissions in Germany [TMLNU,

2009] and Austria [BMLFUW, 1996, 2008, 2012]. Therefore, they reflect the decreasing trend of point emissions during the 1990s. However, they fail to capture the fluctuations of diffuse emissions linked to extreme hydrological conditions, in that the average value for the period 2001-2006 was kept constant for the whole time series, except for a 10% increment of the emissions related to erosion processes before the launch of the agrienvironmental program ÖPUL in 2000 [Zessner et al., 2011b].

With this limitation in mind, the comparison of the two time series still offers an interesting insight on the mechanisms and drivers regulating the behavior of phosphorus in the river. For the 1990s decade, two observations can be made. Firstly, despite the reduction of point emissions and the decline of TP and SRP baseflow concentrations, there was no decreasing trend of riverine loads. Secondly, the loads were considerably lower than the total input from the catchments, which means that, provided that the discrepancy is not due to an overestimation of the emissions, large amounts of phosphorus were retained every year by the river. In 1999 and 2002, years characterized by flood events, the peaks of TP loads were either equal to or exceeded the emissions. Nonetheless, these peaks did not transport the pool retained during the previous decade downstream, which was instead presumably exported to river banks and flooded areas. After 2002 the retention process resumed its major role, with riverine loads notably lower than total emissions, until 2013, when it was offset once more by the mobilization exerted by a large flood.

This examination highlights and confirms the relevance of the retention process, which creates a large pool of phosphorus distributed among flooded areas, river banks and bed sediments, which is only partially transported downstream during episodic storm events. Moreover, it brings more evidence of the important role played by the mobilizable in-stream phosphorus stock. Its presence during the 1990s hampered the direct translation of the reduction of point discharges and TP and SRP baseflow concentrations into a decline of riverine TP loads. Reversely, after the 2002 flood, the depletion of such reservoir led to long-term low riverine loads, despite the occurrence of moderate floods. This can be further illustrated by a simple calculation. Since point emissions represent the primary pathway of phosphorus at baseflow conditions, it can be expected that their reduction would cause a decline in the loads transported at these flow levels. The average yearly TP point emissions within the period T1 (Jan. 1991 - Dec. 1995) T2 (Jan.1996 - Aug.2002), T3 (Sep.2002 - Dec.2007), and T4 (Jan.2008 - Dec.2012) were around 7,000, 3,100, 2,000, and 1,700 t TP, respectively. For each of these periods three yearly loads are calculated, assuming a constant mean value of Low-flows (950  $\text{m}^3 \text{ s}^{-1}$ ), Dry conditions  $(1,300 \text{ m}^3 \text{ s}^{-1})$  and Mid-range flows  $(1,750 \text{ m}^3 \text{ s}^{-1})$ , multiplied by the mean TP concentration at the corresponding flow interval and time period. The mean of these loads is then calculated, obtaining the following average values for the periods T1, T2, T3, and T4: 5,000 t, 3,500 t, 2,300 t, and 2,400 t respectively. The comparison of these loads with the point emissions shows an expected decline in both, but not to the same extent, which indicates that considerable retention took place in the period T1, most likely in the form of algae growth. Moreover, both baseflow loads in the periods T1 and T2 are substantially lower than the actual loads calculated including all flow

levels; in T3 this difference reaches its minimum, and in T4 they diverge again. This further shows that the mobilization of particulate phosphorus at high flow levels played a major role in determining the total loads during the 1990s, whilst after the 2002 flood its contribution dramatically declined and only slowly started to regain its relevance.

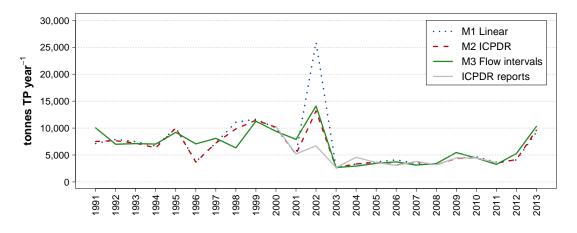


Figure 3.9: Time series of annual TP loads at the Outflow, obtained with 3 different methods and compared to the calculations published by ICPDR.

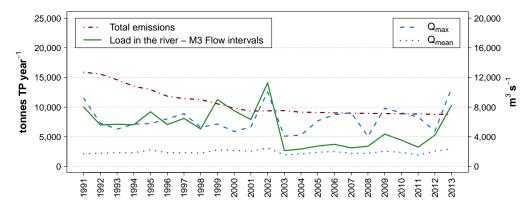


Figure 3.10: Time series of annual TP river loads, TP emissions, maximum daily discharge and mean daily discharge at the Outflow.

# 3.5 Conclusions

The first and rather predictable outcome of this study is that the concentration of TP and SRP in the Upper Danube decreased substantially at baseflow conditions during the 1990s, thanks to the efforts undertaken by Germany and Austria to reduce point emissions.

The time series analysis, however, has revealed an even more pronounced and unexpected decline of the TP concentration following the major flood in August 2002. Interestingly, this reduction was very significant even in high flow conditions, and lasted for a long period of time. Such decline was also reflected by the transported TP loads, despite the fact that mean and maximum discharges presented almost no variation before and after the 2002 flood. The validity of these results is strengthened by the fact that the two locations in the study showed very similar and consistent patterns, both in terms of concentrations and loads.

These findings bring new evidence of the significant impact that flood events can exert not only on the episodic transport of riverine loads, but also on the in-stream phosphorus stock and its propensity to mobilization. The existence of a mobilizable pool of phosphorus in the river, built up through the sedimentation of phosphorus rich algae mass, sustained during the 1990s a high level of annual riverine loads, neutralizing the efforts of reduction of point discharges. It was only after the August 2002 flood removed the richest fraction of this pool, that such efforts were translated into lower long-term loads, in that they slowed down the growth of algae and therefore delayed the regeneration of the stock.

It is thus crucial to better understand and to consider more thoroughly the role that retention, floods and in-stream stocks play in altering the phosphorus mobilization in rivers, either by interfering or amplifying the anthropogenically induced changes. This is highly relevant both for the improvement of river basin models and for the correct interpretation of water quality data in relation to the assessment of environmental management measures.

This study also highlights the usefulness of analyzing shifts in phosphorus concentration as a function of flow level, which offers insight into the different drivers and processes.

Lastly, it flags the value of interrogating long-term, low frequency water quality data sets that, as argued by Burt et al. [2011], should not be dismissed in favor of more recent semi-continuous monitoring, but considered instead as a complementary and valuable source of information to investigate long-term patterns and mechanisms.

# Chapter 4

# Enhancing phosphorus governance in Austria: potential, priorities and limitations

# 4.1 Abstract

Protecting water bodies from eutrophication, ensuring long-term food security and shifting to a circular economy represent compelling objectives to phosphorus governance strategies. This study determines how and to which extent the management of phosphorus in Austria can be optimized. A detailed national model, obtained for the year 2013 through Material Flow Analysis, represents the reference system. Applicability and limitations are discussed for a range of measures aimed at reducing consumption, increasing recycling, and lowering emissions. The potential effect of each field of action on the reference system is quantified and compared using three indicators: Import dependency, Mineral fertilizers consumption and Emissions to water bodies. Further, the uncertainty of this assessment is characterized and priorities for the upgrade of material accounting schemes are identified. Moreover, all the gains that can be obtained through the measures discussed in the article are integrated in the reference system to generate an ideal target model. The fact that this is characterized by an extremely low import dependency of 0.23 kgP cap<sup>-1</sup>y<sup>-1</sup> (2.2 kgP cap<sup>-1</sup>y<sup>-1</sup> in 2013) and by a 28% decline of emissions to water bodies indicates that governance in Austria offers a large scope for phosphorus stewardship. This case study shows the added value of using Material Flow Analysis as a basis to design sound governance strategies, in that the systemic approach inherent to it allows performing a proper comparative assessment of different measures, identifying priorities, and visualizing the target system.

# 4.2 Introduction

The need for enhancing phosphorus (P) governance in most countries is driven by two major goals: protecting surface waters from eutrophication and ensuring future food and energy security under scenarios of uncertain supply (Withers et al, 2015). The latter is particularly true for European countries, as confirmed by the recent inclusion of phosphate rock into the revised list of Critical Raw Materials [EC, 2014b]. Further, the management of P-rich flows can strategically serve as role-model within the ambitious framework of a circular economy that shall enable economic growth by minimizing waste, use of raw materials and environmental damage [EC, 2014a].

The first step essential to achieve these objectives is the profound understanding of the status quo, which allows identifying losses, inefficiencies and processes that require action. This was done in Austria by Egle et al. [2014b], who performed a detailed Material Flow Analysis (MFA) study at the national level for an average year within 2004-2008. Zoboli et al. [2015a] carried forward this work, by shedding light on the dynamics of the system, on the temporal development of management performances and on the uncertainties affecting the results.

Once the system has been thoroughly analyzed, it is necessary to move forward and to assess how it can be optimized. A number of studies have investigated in detail specific issues in Austria, namely the potential of P recycling from wastewater and sewage sludge [Egle et al., 2014a], the potential impact of changing dietary habits on nutrients flows [Thaler et al., 2015], and the performance of the national agri-environmental ÖPUL programme in reducing diffuse nutrients emissions [Zessner et al., 2013]. Nevertheless, the holistic assessment of the optimization options, that provides decision makers with a comprehensive perspective and with the capability of setting priorities, is still missing.

Schoumans et al. [2015] and Withers et al. [2015] have reviewed and put forward a number of management strategies to address P-related challenges, with a particular focus on the European context. The objective of this work is to contextualize such strategies in a specific case study, with the intent to optimize it, by addressing the following questions: to which extent are these measures applicable to the Austrian P system and which limitations exist? Since knowing the relative contribution of the measures to the overall P governance can be very helpful in setting priorities, the potential effect of each of them is quantified using three indicators of the national performance. Further, given the crucial role of data for the accurate assessment of the status quo and for the monitoring of future management, present material accounting schemes are discussed and priorities for improvement are identified. The last objective of this study is the modification of the reference system, based on the assessment of the optimization potential, to produce a target model.

# 4.3 Materials and methods

### 4.3.1 Reference year – losses and inefficiencies

This study is based on the Austrian MFA of P, for which detailed information regarding model structure, flows calculation and data sources is provided in Zoboli et al. [2015a]. For the present work, the time series of P budgets has been extended to the years 2012 and 2013 in order to analyze a more updated system and the most recent year, 2013, is chosen as reference (Fig. 4.1). <sup>1</sup>

The detailed system overview provided by the MFA model unveils the weak points where actions can be taken to improve national P governance. A major loss of P, equal to approximately 6,300 tP y<sup>-1</sup>, is occurring via sequestration in landfills and cement kilns. Other 2,600 tP y<sup>-1</sup> are exported through P-rich waste flows, instead of following a route for domestic recycling. In addition to this,  $2,000 \text{ tP y}^{-1}$  are contained in material employed as substrate for landscaping activities and are therefore lost as productive resource. The budget also shows an inefficient accumulation on private and public green areas, for a total amount of ca. 2,000 tP y<sup>-1</sup>. Thanks to a major reduction of P mineral fertilizers use in agriculture and to the decline of the total livestock and consequently of the manure loads, the P surplus accumulation on agricultural soils has largely decreased over the past two decades. Nevertheless, it is still positive, with a surplus equal to  $2,300 \text{ tP y}^{-1}$  (0.78 kgP ha<sup>-1</sup>y<sup>-1</sup>). Although surplus is usually defined as total inputs minus agricultural outputs, in this work losses to the environment are also included, so that the estimated optimal balance can also be sustained in the long-term. The average dietary intake of 1.6 gP cap<sup>-1</sup>d<sup>-1</sup> is too high and exceeds the nutritional recommendations of the German Nutrition Society [DGE et al., 2003]. As concerns emissions to surface water, point discharges have dramatically declined thanks to the enhanced P removal from wastewater, but diffuse emissions are still considerable (3,600 tP y<sup>-1</sup>). Their further decline is imperative, both to meet the target levels of P loads reaching the Black Sea [ICPDR, 2015] and to achieve the good ecological status defined by the Water Framework Directive in the streams that still fail due to the exceedance of orthophosphate values [BMLFUW, 2013b].

<sup>&</sup>lt;sup>1</sup>Atmospheric deposition flows have been removed, because in reality they do not constitute external imports into the system (but rather mobilization of internal stocks) and therefore they may lead to the underestimation of the input necessary to maintain in the long-term a P balance in agricultural soils.

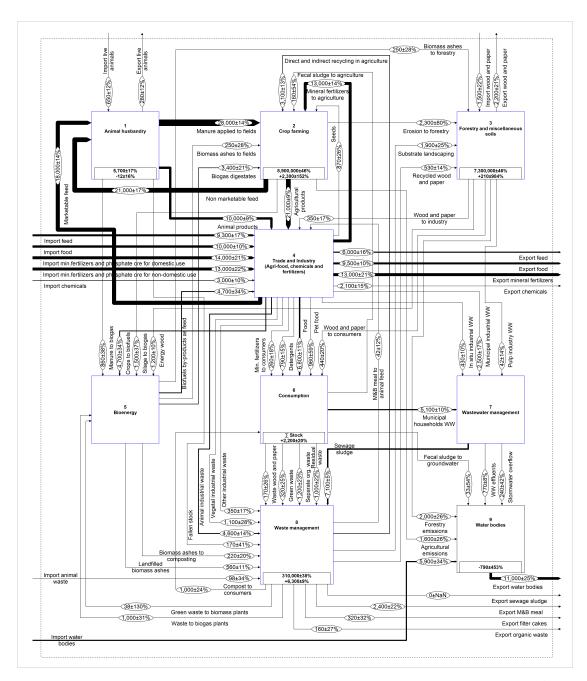


Figure 4.1: Austrian phosphorus budget for the reference year 2013 (unit:  $tP y^{-1}$ ).

# 4.3.2 Fields of action

Based on the analysis of the reference year and on the identification of losses and inefficiencies, specific fields of action are proposed, the potential improvement with respect to 2013 is estimated and barriers and opportunities are discussed. They are grouped into three categories: 1. Increase of P recovery and recycling; 2. Reduction of P demand and consumption; 3. Reduction of emissions to water bodies. Furthermore, the uncertainty affecting this assessment is discussed and priorities for the improvement of data quality are pointed out.

### 4.3.3 Indicators

The relative contribution of each field of action on the overall national P governance is quantified through three indicators: *Import dependency*, *Mineral fertilizers consumption* and *Emissions to water bodies*.

Import dependency, as the name suggests, measures how dependent the country is from imported P and was first put forward by Cooper and Carliell-Marquet [2013], with the name "Net imports". Reasons for being dependent from imports lie in excessive resource demand, inefficient production chains as well as in losses via sequestration in final sinks and via export of valuable waste materials. It is calculated as net import of P in economic goods. It does not include import/export via surface waters, because it is meant to reflect the economic dependency of the country from imported P.

In *Import dependency*, P embedded in all material flows is considered in an aggregated form. Nonetheless, it is also worth distinguishing between mineral fertilizers and other goods. Therefore, the specific contribution to *Mineral fertilizers consumption* is also assessed.

There are two main reasons to evaluate separately the effect on *Emissions to water bodies*. In the first place, this indicator addresses the goal of protecting water bodies from P seen as pollutant, whereas the other two regard P as a critical resource. Secondly, diffuse emissions to water bodies are largely dependent on stocks of P accumulated in soils during decades and on hydrological factors, whilst *Import dependency* and *Mineral fertilizers consumption* are almost not influenced by natural variability nor by past legacy.

### 4.3.4 Uncertainty of the assessment

The comparative assessment of the different fields of action via the three indicators is also accompanied by a qualitative characterization of its uncertainty. If the MFA flows related to a given field of action have less than 10% uncertainty and there are no significant gaps of data that hinder a proper estimation of the scope for improvement, the overall uncertainty is defined as *Low*. If MFA flows are affected by more than 50% uncertainty and/or the estimation of the potential is associated with major data gaps or with complex system feedbacks, the overall uncertainty is depicted as *High*. *Moderate* describes intermediate cases.

### 4.3.5 System optimization

Based on the assessed potential of the selected fields of action, the reference system is modified with the goal of minimizing import dependency, use of mineral fertilizers and emissions to water bodies.

The proper design of a national strategy should be based on a multi-criteria optimization that also considers costs, social acceptance and other environmental impacts in order to account for trade-offs between different objectives and for possible rebound effects. Such a complex approach lies beyond the scope of this article and will be addressed in future research. The focus here is on one-dimensional optimization, as the first necessary step to understand what could be achieved and which limitations exist in the given system.

# 4.4 Results

## 4.4.1 Increase of P recovery and recycling

### Increase of P recycling from meat and bone meal

The use and handling of animal by-products is strictly regulated on the basis of three risk categories [EC, 2011]. Out of the 86,100 t of meat and bone meal produced in Austria in 2013, half was regarded as low risk Cat.3, whereas the other half belonged to high (Cat.1) and medium (Cat.2) risk categories [BMG, 2013]. 27% of the total meat and bone meal found direct application as agricultural fertilizer and 1% was employed for pet food production. Another 19% was incinerated, whilst the rest (53%) was exported to be incinerated, employed as fertilizer and for feed production abroad [BMLFUW, 2014a]. Export has dramatically increased over the past years, at the expense of domestic incineration and fertilizer use.

The national recycling rate could be improved in different ways. In the first place, there are 20,000 t of Cat.3 meal (approximately 1,000 tP  $y^{-1}$ ) that could be applied as organic fertilizers, either directly or indirectly, instead of being exported. A second approach would be the P recycling from the ashes that stem from the incineration of meat and bone meal. This would imply ceasing its usage as secondary fuel in cement kilns, in favor of its mono-incineration or co-incineration with other P–rich materials such as sewage sludge. Since more than 85% of P can be recovered from the ashes, approximately 2,000 tP y<sup>-1</sup> could be recovered every year, provided that the whole amount of Cat.1 and Cat.2 followed this route. Another relevant aspect worth being discussed is the fact that the meal of Cat.1 and Cat.2 mostly (about 85%) derives from by-products belonging to the latter [BMLFUW, 2014a]. This, however, can also be allocated to composting and anaerobic digestion if combined with adequate pre-treatment. This use is exclusively allowed if these materials never got in contact with Cat.1; therefore, the current handling and transport practices should be substantially modified. Approximately 1,800 tP y<sup>-1</sup> could be recovered through this change.

An important source of uncertainty affecting these results concerns the P concentration in meat and bone meal, which can vary between 3% and 6.6% [Garcia and Rosentrater, 2008]. The estimates assume an average and homogeneous P concentration of 5.2% (resulting from the mass balance), but it would be useful to perform a specific survey to assess more exactly the recycling potential of the different categories and the

specific flows.

### Increase of P recycling from sewage sludge

In 2013 Austria generated 240,000 t (dry matter) of municipal sewage sludge that contained approximately 6,600 tP [BMLFUW, 2014a,d]. Only 16% was directly applied in agriculture and 10% indirectly after composting; 49% was incinerated, 22% used in landscaping activities and 3% landfilled after biomechanical stabilization [BMLFUW, 2014a]. A considerably higher share of direct or indirect (via composting) agricultural application in the future is not realistic, since it is banned or partially restricted in most federal states.

Egle et al. [2014a] performed a comprehensive assessment of the technologies currently developed or under development to recover P from wastewater and sewage sludge. By combining their outcomes with the specific traits of the national wastewater treatment infrastructure, they estimated an actual recovery potential ranging from 0.16 to  $0.81 \text{ kgP cap}^{-1}\text{y}^{-1}$ , i.e. from 1,300 to 6,700 tP y<sup>-1</sup>, depending on the type of technology and related investment effort. If the calculations of the report are adjusted to the updated values of total P load in municipal sludge, which slightly decreased over the past years, the maximum recovery potential with respect to the year 2013 corresponds to 0.74 kgP cap<sup>-1</sup>y<sup>-1</sup>, i.e. 6,200 tP y<sup>-1</sup>.

Two main sources of uncertainty could be identified. The first one concerns the current use of sludge: only agricultural application, incineration and landfill disposal are indicated separately in the bi-annual reporting of sludge management, whereas the rest is aggregated under the category "Others". Further, incineration should be more precise, at least by distinguishing between mono-incineration and co-incineration. The second important source of uncertainty stems from the technological performance of P recovery, in terms of product quality, costs, energy and chemicals consumption. In order to reduce these uncertainties, there is a strong need for full-scale demonstration plants. Nevertheless, as summarized in the review by Kabbe et al. [2015], the primary obstacles to P recycling from sewage sludge are no longer technological, but are rather linked to the lack of legal framework and secure market for recycled products. Therefore, the priority for national governance should be to ensure a level playing field for all fertilizers.

As for industrial sewage sludge, P recycling may represent an interesting business opportunity for a handful of industries, e.g. in the agri-food or paper sectors, but from a national perspective this P flow is too low to exert a significant effect on the three indicators considered here.

### Increase of P recycling from compost

In the year 2013, approximately 3,000 tP were contained in compost products (including composted sewage sludge). According to a governmental survey [BMLFUW, 2005], one third of this compost was applied in agriculture, another third was used for private and public gardening, and the rest was employed in landscaping activities. The last one of these applications needs large volumes of material but does not necessarily make use

of the nutrients contained in it. Therefore, compost could be replaced with other less valuable materials, by making 1,000 tP y<sup>-1</sup> available for agriculture, provided that it complies with the quality requirements regulated by the Compost Ordinance, [BML-FUW, 2001]. Moreover, on the ground of the considerable surplus being accumulated on private and public green areas, it can be argued that also the large use of compost in gardening activities may not be optimal from a national P governance perspective and that it shall be rather employed in agriculture.

This field of action is affected by a significant lack of public data. The last published data on compost production refers to 2009 [BMLFUW, 2011a], the last study assessing the share of different uses of compost products corresponds dates back to 2005 [BMLFUW, 2005], and the range of P concentration given in these reports is very broad (0.4-1.3 P2O5% of dry matter). Therefore, both to better assess the potential optimization of P recycling through this route and to assess the future performance, a more structured scheme of data collection is required.

### Increase of P recycling from digestates

In 2013, approximately 3,400 tP were returned to agricultural fields via anaerobic digestates. These were generated in 380 biogas plants of primarily small-medium size highly distributed on the whole territory [Energie-Control Austria, 2014]. Therefore, their current use as agricultural amendment does not pose problems of local concentration and surplus accumulation and does not present further potential for resource use optimization.

Nonetheless, the experiments performed by Stutter [2015] indicate that anaerobic digestates show a potentially high risk of P leaching; it is thus essential that strategies to minimize such risk are implemented, such as limiting application to periods of crop growth.

As reported by the review of Möller and Müller [2012], anaerobic digestates exhibit a wide range of P concentration (0.6-1.7 % of dry matter) due to the high variability of the feedstock composition. The current estimate relies on information on the use of raw materials collected through specific surveys based on voluntary participation. There is thus a lack of structured national monitoring of material and nutrients flows in this sector.

### Increase of P recycling from biomass ashes

The utilization of ligneous and herbaceous biomass for heat and electricity production has increased substantially in Austria in the past years. In 2013, this industry generated 127,000 t of wood and straw ashes [BMLFUW, 2014a] that given their average composition [Obernberger and Supancic, 2009] contained approximately 1,200 tP. They are partially applied in agriculture and forestry. In addition, the Compost Ordinance [BMLFUW, 2001] allows and regulates their use as additive in composting. However, ca. 44% are still being landfilled. A target of 100% recycling is not realistic, on the one hand because the heavy metals content of a fraction of these ashes impedes their direct use in agriculture and composting and on the other hand because their relative low P content hinders the utilization of P recovery technologies. The heavy metals rich fraction consists mainly of fine fly ashes, which represent approximately 2-15% of the total weight [Obernberger and Supancic, 2009]. Hence, it can be roughly estimated that 400 tP could be additionally recycled in agriculture or forestry. At the beginning of 2011, new and updated national guidelines were published with the aim of fostering, regulating and monitoring the application of these ashes as fertilizers [BMLFUW, 2011b]. Given the above, the basis for an enhanced recycling has been already set.

These new guidelines also prescribe the establishment of registers for the application of ashes in agriculture and forestry, which shall considerably improve the currently available information on this usage.

### Increase of P recycling from manure

Austria is neither characterized by a segregation of animal husbandry and crop systems nor is its livestock production as intensive as it is in other countries. It can thus be reasonably assumed that manure application is not provoking nutrient unbalances. Nevertheless, best management practices and precision farming techniques are not yet fully put into practice and still offer scope to reduce mineral fertilizers consumption, but a major lack of information hinders its quantification.

This flow is also affected by another significant data issue. Given the outstanding relevance of manure loads for the proper assessment of agricultural nutrient balances and for accurate modeling of emissions to water bodies, Velthof et al. [2015] carried out a comprehensive study commissioned by Eurostat on excretion factors of livestock across Europe. Although their review primarily focused on nitrogen, their conclusions can also be applied to P and indicate a pressing need for harmonization of methods and data.

### Improvement of municipal and industrial organic waste collection and management

The separate collection and recycling of the organic fraction of municipal solid waste (MSW) has increased substantially over the past 20 years, but approximately 1,000 tP are still contained in residual waste and are therefore lost via incineration or disposal in landfills. Austria already meets the Waste Framework Directive [EU, 2008] target of 50% overall recycling of MSW by 2020 and it also complies with the Landfill Directive [EU, 1999], by currently landfilling less than 3% of biodegradable MSW. The national organic recycling rate is 33% and it is characterized by strong regional differences [EEA, 2013]. If a 50% target were set on a national level, 300 tP y<sup>-1</sup> could be additionally recovered.

In 2013, approximately 1,400,000 t of organic by-products and waste were generated by the food processing and retail industry. To a large extent, they were either employed for feedstuff production or recycled via composting and anaerobic digestion. Nevertheless, a relatively small fraction of 160 tP embedded in dairy by-products was exported. These results are affected by considerable uncertainties. One first issue concerns the P content in MSW: the most recent measurements in Austria date back to the study by Skutan and Brunner [2006], so for more recent years this value needs to be adjusted as a function of the changing organic fraction. Another relevant source of uncertainty regards the current usage of industrial by-products, for which information on management routes is either highly aggregated or lacks of clear links to the by-products categories. Hence, the modest potential assessed here might actually be higher.

This field of action does not end at optimizing the management of generated waste, but it should also address its prevention, which is why its title does not focus on recycling only, but on improvement in general. The Federal Waste Management Plan 2011 includes a Waste Prevention Programme [BMLFUW, 2011a]. Out of the 5 areas that are prioritized, two address household and food waste. The Programme has not set yet concrete targets, but one of its primary objectives is identifying the potential for reduction, which due to lack of sufficient information is left out of the present analysis.

### 4.4.2 Reduction of P demand and consumption

### Achievement of a balanced and healthy diet

Thaler et al. [2015] investigated the potential impact that a shift towards a healthy and balanced diet in Austria would exert on the nutrients fluxes at national scale, compared to the reference period 2001-2006. They estimated that aligning the dietary habits with the recommendations of the German Nutrition Society and the World Health Organization (60% less meat and dairy products, 13% more corn, rice and potatoes and 63% more fruit and vegetables) would imply a 5-6% decline of P diffuse emissions to rivers and a 20% reduction of P net imports, which would largely affect feedstuff and other biomass flows and only to a smaller extent mineral fertilizers.

As shown by the scenarios investigated within the study, such estimations are sensitive to the structure of the agricultural system that would accompany such dietary shift (e.g. share of organic farming, share of self-sufficiency versus international trade and intensity of land use for energy crops). Furthermore, the authors have not analyzed the socio-economic implications of their scenarios. As concluded by Westhoek et al. [2014], cutting meat and dairy products consumption in Europe to meet dietary recommendations would have a major economic impact on livestock farmers and on other actors involved in the related supply-chain, but it would also bring major health benefits, 40% reduction of nitrogen emissions, 25-40% decline of greenhouse gas emissions and 23% per capita reduction of cropland employed for food production. This field of action can therefore play a very important and strategic role, but its applicability and effectiveness are not as straightforward as they are for other measures.

### Increase of the use efficiency in crop farming

As most other western countries, Austria has followed during decades the strategy of fertilizing in excess, with the consequent accumulation of a large P stock in the soils.

As argued e.g. by Withers et al. [2014] and Schoumans et al. [2015], there is sufficient scientific evidence as well as compelling resource, economic, and environmental reasons to shift from an insurance strategy of oversupply to a more efficient and targeted crop fertilization. Schröder et al. [2011], in their review of measures aimed at improving P use efficiency in agriculture, put forward as "low hanging fruits", i.e. measures that could be rapidly implemented and pay for themselves, the adjustment of inputs to outputs and the revision of fertilizer recommendations.

Thanks to the declining trend of mineral fertilizers consumption, in Austria many progresses have been made in balancing inputs and outputs. For the year 2013, a surplus of 2,300 tP y<sup>-1</sup> (0.78 kgP ha<sup>-1</sup>y<sup>-1</sup>) was estimated. That year however was extraordinary, in that it was characterized by an extended drought and consequently by low production levels. In 2011 and 2012, a greater agricultural output was achieved, which resulted in less surplus, equal to 1,800 and 500 tP y<sup>-1</sup> (0.61 and 0.17 kgP ha<sup>-1</sup>y<sup>-1</sup>), respectively. The achievement of a perfect balance may not be realistic, but the positive surplus values show that there is still room for further reduction of mineral fertilizers consumption. A revision of the fertilizer recommendations might be an effective mean of strengthening and maintaining the declining trend.

However, in order to sustain in the long-term high levels of crops production with reduced fertilizer application rates, more sophisticated approaches might be required, e.g. methods to make use of soil P legacy, maintenance of soil quality, or breeding of crops with lower P requirements [Schröder et al., 2011, Withers et al., 2014]. Furthermore, targeted fertilization could contribute to decrease P emissions through surface runoff, which account for ca. 6% of the total P emissions to water bodies [Schilling et al., 2011], and in the long term also to the emissions through other pathways (mainly erosion), since the legacy P in the soil would gradually be reduced.

Unlike other countries, such as France for example [Senthilkumar et al., 2011], Austria does not present strong spatial differences due to geographical segregation of crop and livestock systems; therefore national aggregated soil balances do not mask local opposite unbalances and are appropriate for future assessments.

As far as it concerns data, priorities should be set on the harmonization, validation and update of excretion factors of livestock (see chapter *Increase of P recycling from manure*) and of the P concentration in agricultural products. As shown by a French survey [COMIFER, 2007], the P content in several types of crop and fodder has considerably changed from the 1990s.

#### Optimization of P content in animal feed

Total P content in feedstuff often exceeds animal requirements in order to overcome limited P digestibility in plant and mixed feed. Alternative ways to achieve a higher efficiency exist however, e.g. feeding in accordance with growth phases, lowering phytate-P content, adding phytase enzymes or using more digestible feedstuff [Schoumans et al., 2014]. For the farmers, this could imply savings but also higher costs, depending on the required storage and feeding equipment and on the fluctuating market price of feedstuff ingredients. Austria has currently no scheme of nutrients monitoring at farm scale. This renders difficult to properly assess the current state of implementation of such techniques. According to the studies of Kebreab et al. [2012] and Maguire et al. [2005], dietary total P could be reduced up to 20% without affecting livestock performance. Such a high theoretical potential would per se justify an effort in increasing data availability. In addition, dietary P reduction would directly translate into a decline of P content in excretions. On the one hand, this would lead to lower P emissions wherever inappropriate application of manure causes losses to water bodies, although such effect is not quantifiable in this study. On the other hand, in case of efficient use of manure and neutral agricultural balance, the reduction of P in manure would have to be compensated through more fertilization. Given the existence of agricultural surplus, this impact is not visible in 2013. Nevertheless, these issues are highly relevant for sustaining the system in the long-term and they provide additional reasons to enhance data collection schemes.

#### Reduction of P use in detergents

Since the 1990s, Austria has met the target of 100% "phosphate-free" laundry detergents, by means of voluntary commitments [EC, 2004]. About 850 tP y<sup>-1</sup> are still consumed via automatic dishwasher detergents (Wind, 2007), but they will soon decline too, thanks to the EU Regulation N<sup>o</sup> 259/2012 that has recently introduced specific restrictions (as of January 2017).

#### Reduction of P use in other industrial processes

Withers et al. [2015] mention the use of ortho-phosphates dosing for controlling lead in drinking waters, but this does not apply to the Austrian case, since raw water used for public supply is practically lead free [Jung and Heiss, 2007].

With respect to other industrial uses, Schipper [2014] lists a number of applications of P, namely for car engines, pesticides, rechargeable batteries and flammable retardants. Unfortunately, the only available data concerns pesticides, for which Egle et al. [2014b] estimated a very small flow of about 15 tP y<sup>-1</sup>. Due to the lack of data, it is thus not possible to assess any real potential for optimization nor to monitor future changes.

#### Reduction of surplus accumulation in private and public green areas

The national MFA has revealed the presence of P accumulation on public and private green areas at a rate of 2,000 tP y<sup>-1</sup>. This is not a negligible stock accumulation, since it corresponds to 15% of the annual consumption of mineral fertilizers in agriculture.

Nevertheless, this result is affected by large uncertainties that could be considerably reduced through better information on home-composting and on sales of compost products to privates, which together account for 80% of total inputs.

#### 4.4.3 Reduction of emissions to water bodies

#### **Reduction of point discharges**

The connection to municipal wastewater treatment plants (WWTPs) is 94.5% [BML-FUW, 2014d]. A 100% connection rate is unrealistic, given the rural and dispersed character of the remaining settlements. The wastewater of the non-connected 5.5% of inhabitants is handled either via domestic treatment plants or via septic tanks and the resulting sludge is either transported to municipal WWTPs or applied on agricultural fields. Therefore, the connection rate to municipal WWTPs does not offer further room for improvement, neither to reduce emission to water bodies nor to increase P recovery.

The current total P removal rate from municipal wastewater is 90% [BMLFUW, 2014d]. The P load still contained in the effluents is approximately 700 t. The 90% removal rate represents the national average, with specific performances varying within the range 86-96%. Therefore, a further improvement of up to 400 tP y<sup>-1</sup> is still achievable.

Driven by the requirements of the Urban Wastewater Directive [EU, 1991] concerning the reporting of the disposal of urban wastewater and sludge by the end of 2005, the data quality regarding the performance of municipal WWTPs has considerably increased in terms of its harmonization, frequency and completeness. As far as it concerns industries with in-situ wastewater treatment, however, only large plants from selected sectors must comply with a legal reporting obligation. From a national perspective, the total load of P emitted by industries is very low (ca. 70 tP y<sup>-1</sup>), but it can exert a considerable local impact on vulnerable surface waters and therefore in specific situations an improvement of these data should be considered.

#### Reduction of erosion from agricultural soils

The interest on P total national emissions is driven by the ultimate goal of reducing the load reaching the Black Sea. According to the draft River Basin Management Plan 2015 elaborated by the International Commission for the Protection of the Danube River [ICPDR, 2015], the loads of total phosphorus (TP) have considerably decreased, but they still exceed by about 20% the reference target set at the levels of the 1960s. In the considered period 2009-2012, the specific contribution of the Austrian territory per surface unit was among the lowest of the whole Danube basin, which reflects the success of the efforts addressing point discharges.

Erosion, however, still constitutes about 42% of the total Austrian emissions. Through the empirical nutrient emission model MONERIS, Schilling et al. [2011] estimated that, in the period 2001-2006, agricultural soils were responsible for 43% of the TP transported through erosion, whereas natural land accounted for 57% of it, which is primarily due to the high nutrients export from glaciers [Zessner et al., 2011c]. Given that the current average value of soil loss in Austria is 3.4 t ha<sup>-1</sup>y<sup>-1</sup> [Baumgarten et al., 2011] and that a level of 1 t ha<sup>-1</sup>y<sup>-1</sup> is generally considered as tolerable and often unavoidable [Eurostat, 2015], a reduction by 70% could be realistically achieved. Such a contraction of soil losses in agriculture would contribute to the decline of total TP emissions by 13% and would also decrease proportionally the need for fertilizers. Despite the modest potential of this field of action on the national scale, its relevance must not be underestimated. In 2012, 20% of the monitored surface waters could not achieve the good ecological status defined by the EU Water Framework Directive, owing in 75% of the cases to exceedance of orthophosphate limits [BMLFUW, 2013b]. Moreover, although erosion is responsible for one third of total P emissions nationwide, it constitutes the dominating pathway in several catchments and especially in those where orthophosphate limits are exceeded.

It is also worth mentioning that this field of action can be extremely cost-effective if its implementation is well designed. As shown by Kovacs et al. [2012] and by Zessner et al. [2013], focusing on phosphorus emission hotspots can remarkably help prioritizing interventions and investments, given that the application of best management practices on less than 10% of the total area would be sufficient to achieve a considerable improvement of water quality.

The quantification of erosion is inherently uncertain, since it has to rely on models for which direct validation is not available. The primary source of uncertainty in such models, pointed out by Sharpley et al. [2015] as major challenge and research need, lies in retention processes and in the long-term accumulation and transport of legacy P.

#### 4.4.4 Comparative assessment of all fields of action

Table 4.1 presents the potential gain that can be achieved through each field of action, expressed as percentage of the indicators values in the reference year 2013. Absolute values and rationales underlying these results are given in the Appendix E.

The group of measures aimed at enhancing P recycling holds a noteworthy scope to lower *Import dependency* and *Consumption of mineral fertilizers*, with meat and bone meal, sewage sludge and compost representing the most relevant flows, whereas other waste materials can contribute only marginally. The combined recycling from all waste flows could replace more than 75% of the present mineral fertilizer use. If compared for instance with the findings of Senthilkumar et al. [2014], who estimated for France a substitution potential of merely 21%, this high value makes evident the extraordinary potential of P recycling in Austria. From a lumped national perspective, these measures do not alter *Emissions to water bodies*, although specific local impacts can be relevant, as it was discussed in previous chapters.

The scope to optimize national P governance through a decline in P consumption is of a similar order of magnitude, with the most relevant actions to lower *Import dependency* being the ones that address diet and P content in feedstuff, followed by use efficiency in crop farming and surplus on green areas. *Consumption of mineral fertilizers* can be reduced by addressing use efficiency in crop farming and accumulation on green areas. The shift to a balanced diet and the reduction of P in detergents are the only measures that can contribute to the decline of *Emissions to water bodies*, which can be further reduced through actions on point discharges and erosion from agricultural soils by respectively 10% and 13%.

The assessment of the group of measures aimed at reducing P consumption is affected by larger uncertainty, except for P use in detergents, which is regulated by law. This owes partly to a more severe lack of data and partly to the complex system feedbacks they depend on. For the same reason, also the efficacy (defined as the actual power to produce a desired result or effect) and the quantification of the costs involved can be considered significantly more uncertain for this group of measures.

A crucial aspect that generally needs to be considered when prioritizing actions within a national strategy is the time frame. For the measures put forward in this article, it can be reasonably argued that the effects would appear almost immediately after their implementation, except for the decline of diffuse emissions to surface water which are largely dependent on legacy P. As shown by the analysis of the time series of the Austrian P budget [Zoboli et al., 2015a], once clear and effective decisions are made, P flows can change dramatically even from a year to the next. Nevertheless, what may lead to substantial disparities is the time required for the decision process and for the design of the implementation methods.

ment, expressed through three indicators. Percentage values	
Table 4.1: Relative effect of the fields of action on the national P management,	indicate the estimated improvement with respect to the reference year 2013.

Field of action	Scope for reduction of <i>Import</i> <i>dependency</i>	Scope for reduction of <i>Mineral</i> <i>fertilizers</i> consumption	Scope for reduction of <i>Emissions to</i> <i>water bodies</i>	Uncertainty	Main data gaps	Main challenges
Increase of P recycling from meat and bone meal	16%	23%	I	Moderate	P concentration	Legal framework and market uncertainties for recovered fertilizers
Increase of P recycling from sewage sludge	23%	32%	I	Moderate	Performance and product quality for new recovery technologies	Legal framework and market uncertainties for recovered fertilizers
Increase of P recycling from compost	11%	15%	I	High	Current use shares; P content	Regulation/coordination of sales in large number of composting plants
Increase of P recycling from digestates	I	I	I	Low	Feedstock amounts and composition	Large number and heterogeneity of biogas plants
Increase of P recycling from biomass ashes	2%	3%	I	Moderate	Current recycling rate Ash quality	Lack of economic incentives that offset logistical costs
Increase of P recycling from manure	I	I	I	High	Livestock excretion factors Use efficiency of manure as fertilizer	Enhancement of agricultural advice services
Improvement of municipal and industrial organic waste collection and management	2%	3%	I	Moderate	P concentration in MSW Current use of ind. Food waste prevention potential	Resistance of households and similar establishments to further increase separate collection Increase of logistical effort and costs for the municipalities
Achievement of a balanced and healthy diet	20%	ı	5-6%	${ m High}$	Complexity of system feedbacks	Resistance to behavioral change Opposition of meat producers
Increase of the use efficiency in crop farming	8%	11%	ı	Moderate	Livestock excretion factors P concentration in crops	Enhancement of agricultural advice services
Optimization of P content in animal feed	20%	I	I	High	Current state of optimization; complexity of system feedbacks	Enhancement of agricultural advice services
Reduction of P use in detergents	4%	I	2%	Low	I	I
Reduction of P use in other industrial processes	I	I	I	$\operatorname{High}$	Material flows in industrial applications	Substitutability of P
Reduction of surplus accumulation in private and public green areas	11%	15%	I	High	Home compositing Sales of compost to privates	Resistance to behavioral change Coordination of large numbers of people
Reduction of point discharges	I	I	10%	Low	Loads and performances of in situ industrial WWTPs	Higher Fe levels in sewage sludge would pose a problem for several P recovery technologies
Reduction of erosion from agricultural soils	12%	17%	13%	High	Retention processes; long-term behavior of "legacy" P	Implementation at large scale; Identification of hotspots
Indicator value in 2013	$18,600 \text{ tP } y^{-1}$ 2.2 kgP cap <sup>-1</sup> y <sup>-1</sup>	$13,200 \text{ tP } y^{-1}$ 1.6  kgP $ ext{cap}^{-1} y^{-1}$	$4,600 \text{ tP } y^{-1}$ 0.54  kgP $ ext{cap}^{-1}y^{-1}$			

#### 4.4.5 Optimized system

Figure 4.2 displays how the national system would look like if all the measures discussed in this article were fully implemented. Flows that have changed are marked in green, but not the ones that have been slightly altered by the reconciliation process, in order to highlight significant changes only.<sup>2</sup>

Mineral fertilizers consumption is null. This might not necessarily represent the optimal target, but it manifests the extremely high potential held by the system to lower and replace the use of mineral fertilizers without affecting agricultural production. Surplus accumulation does not take place in any soil compartment. However, there is still a partially unavoidable loss of P in landfills that will mainly depend on the efficiency of the applied P recovery processes. *Import dependency* measures 2,000 tP y<sup>-1</sup> (0.23 kgP cap<sup>-1</sup>y<sup>-1</sup>) and *Emissions to water bodies* 3,300 tP y<sup>-1</sup> (0.39 kgP cap<sup>-1</sup>y<sup>-1</sup>).

The design of the optimized system is a fundamental complement to the comparative assessment shown in Table 1, since from the latter alone misleading conclusions may be drawn. Although the sum of all estimated potentials exceeds 100%, it does not mean that Austria could turn into a net P exporter, owing to the existence of trade-offs among the different fields of action. Achieving a balanced and healthy diet, for instance, would imply a strong contraction of meat production and consequently a decline of the recycling potential from meat and bone meal.

Taking into account all imports and exports, instead of only trade of economic goods as done within the indicator *Import dependency*, results in Austria actually being a net P exporter. This is due to the large internal mobilization of P taking place in natural soils, which is partly exported via water bodies and which partly enters the economic system via wood and paper.

This model must not be considered a fully optimized system, but rather a visual representation of the maximum that can be achieved to reduce import dependency, consumption of mineral fertilizers and emissions to water bodies. For a proper optimization of the system, a multi-criteria study, which takes into account factors likely to show relevant trade-offs with P stewardship, such as costs and other environmental impacts, is required.

As indicated in table 4.1, the actual implementation of the measures discussed in this article faces several different obstacles and challenges. Partial goals may however be realistically achieved in the short-mid term. The sewage sludge produced by the municipal WWTP of Vienna is already being mono-incinerated, which eases and lowers considerably the financial burden of introducing P recovery. Following this example, the other large municipal WWTPs (over 100,000 PE) could also implement this route for their sludge management. Further, it is likely that struvite recovery will be introduced in the WWTPs that meet basic requirements, given the manifold operational benefits that it offers. The recycling of meat and bone meal of Cat.3 does not require any investment

<sup>&</sup>lt;sup>2</sup>The year 2013 was characterized by a century flood, which caused extremely high P loads and net P mobilization in the Dabube river. For the visualization of an ideal target system, a more typical situation of water bodies is shown, with average import and export P loads corresponding to the period 2003-2012 (extracted from Zoboli et al. [2015b]) and positive P retention.

nor legal changes and it involves only a very small number of processing plants. Further, the material of Cat.1 and Cat.2 could be incinerated in the facilities employed for sewage sludge, allowing the recovery from P-rich ashes. New regulations for the enhancement of biomass ashes recycling and for the reduction of P content in detergents are already in force and their effects can be expected in the short term. As for the reduction of emissions to water bodies, a 95% removal rate of P from wastewater can be achieved without major infrastructural or operational changes and a decline of diffuse emissions through erosion from agricultural soils by 10% can be expected as result of the agrienvironmental programme ÖPUL [Zessner et al., 2013]. Therefore, with a moderate effort, in the short-mid term *Import dependency* could be reduced from 2.2 to 1.4 kgP cap<sup>-1</sup>y<sup>-1</sup>, *Mineral fertilizers consumption* from 1.6 to 0.87 kgP cap<sup>-1</sup>y<sup>-1</sup> and *Emissions to water bodies* from 0.54 to 0.47 kgP cap<sup>-1</sup>y<sup>-1</sup>.

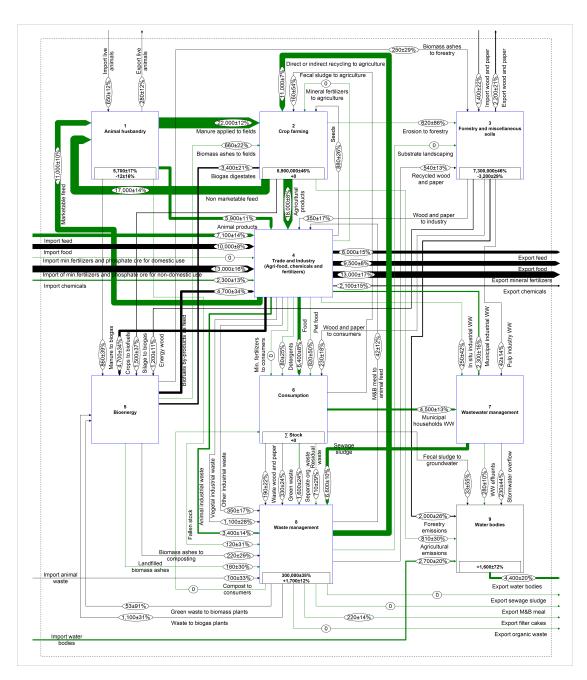


Figure 4.2: Optimized Austrian phosphorus budget based on the reference year 2013 (unit: tP y<sup>-1</sup>). Objectives for the optimization: reduction of import dependency, consumption of mineral fertilizers and emissions to water bodies. Flows that have changed are marked in green.

#### 4.5 Conclusions

There are several strategies to achieve phosphorus (P) stewardship, but their applicability and actual potential is highly dependent on the specific context. In Austria, recycling from waste streams and reduction of consumption offer an optimization scope of a similar order of magnitude, although the latter is affected by larger uncertainty and more difficult obstacles. If all measures selected in this study were implemented, the domestic use of P mineral fertilizers could be fully replaced, the import dependency could be reduced from 2.2 to 0.23 kgP cap<sup>-1</sup>y<sup>-1</sup> and emissions to water bodies would decline from 0.54 to 0.39 kgP cap<sup>-1</sup>y<sup>-1</sup>, with respect to the reference year 2013.

The systemic approach of Material Flow Analysis (MFA) has allowed quantifying the relative effect of each field of action on the national performance measured with different indicators, and thus to perform a proper comparative assessment. Further, it has made possible the generation and visualization of a target system, obtained through the integration of all potential gains in the reference model. The resulting concise though exhaustive overview can be very useful to support decision makers in designing national governance strategies and setting priorities, as well as to assist domain experts in fitting their work into a broader context.

As next step, this study needs to be complemented with the analysis of the different costs involved in implementing each field of action. Such assessment is subject of ongoing research.

# Chapter 5 Conclusions

This Thesis presents a deep and multifaceted insight into phosphorus (P) flows and stocks in Austria. It has been shown that, even in a relatively stable socio-economic period of time, they have undergone dramatic and sometimes sudden changes in several different anthropogenic and natural compartments. By applying the systemic approach provided by Material Flow Analysis (MFA), it was possible to identify the impact on P flows and stocks exerted by regulations or economic developments that were not directly aimed at nutrients management. Two clear examples of it are the ban of meat and bone meal usage as feedstuff ingredient in 2001 and the rapid development of the bioenergy sector around 2003-2004.

Further, the detailed analysis of the time series has also revealed unexpected temporal patterns of total phosphorus (TP) loads in the Danube river, which have conducted to new research questions and ultimately to relevant findings in the field of water quality and emissions control. It was found that the large decline of P emissions through point discharges that took place in the 1990s did translate into lower TP concentrations at low flow conditions in the river, but not in lower transported loads. Only after the century flood in 2002 and the partial depletion of the in-stream P stock, which was accumulated through years of retention in sediments and algae biomass, the decline of point emissions was actually reflected into lower loads. The lack of understanding of these interweaving anthropogenic and hydrological mechanisms would hinder the correct interpretation of patterns of concentrations and loads in large rivers and the correct assessment of the performance of environmental management measures.

Besides these added values, such a multiyear scheme has proven appropriate and useful to directly monitor the national performance of nutrients management. This is exemplified by the identification of a declining trend of total P recovery and recycling from waste streams, which indicates the lack of consideration of P in the design of waste management.

It has therefore been demonstrated that it is feasible to perform national multiyear accounting of P flows and stocks and that public authorities would gain several benefits if they implemented such material accounting in their routine statistics and reporting schemes. As this Thesis has also shown, thanks to its systemic approach, such multiyear accounting can be used not only to monitor specific actions or compartments of the system, but also the overall national performance with respect to P stewardship.

Thanks to the deep understanding of the system provided by the MFA model, it was possible to assess the potential held by a set of different measures and actions aimed at reducing P import dependency, consumption of mineral fertilizers and emissions to water bodies. It was found that measures that address consumption and demand and end-ofpipe actions that on the contrary focus on recycling hold a similar potential, although the first group is affected by larger uncertainty and by more complex implementation challenges. If the maximum possible effort were undertaken, Austria could dramatically decrease its P import dependency, completely replace mineral fertilizers and also achieve a considerable decline of emissions to water bodies. This means that the design of an appropriate P governance strategy in this country offers a broad scope to achieve an ambitious level of P stewardship.

Thanks to the system perspective, it was possible to perform a comparative assessment of the relative contribution of each action, which can be very useful to support decision makers in setting priorities. Further, working with MFA allowed the design of an optimized system that can also support decision makers in visualizing the target system to be achieved and in understanding the feedbacks and interactions between different compartments.

Last, this Thesis has investigated in depth data quality and the role played by uncertainty in view of material accounting and monitoring. In general, the quality declines throughout the P cycle in the anthroposphere, with extremes being the standardized monthly reporting of agricultural production and the complete lack of data for a number of flows in the waste management sector or in the recently developed bioenergy industry. It was shown how the characterization of data quality through a set of indicators can provide relevant feedback to data producers, because it can help them prioritize the efforts needed to enhance data collection schemes. In this respect, another source of feedback to data producers stems from the analysis of the multiyear MFA model, given that the behavior of the reconciliation process over time can contribute to the identification of systematic problems, as was exemplified in this Thesis by the case of the Composting process.

Understanding data quality alone is not sufficient. In this work, it was shown that the rigorous consideration of uncertainty alters the capability of actually detecting temporal changes, especially if the aim is the assessment of annual or short-term variations. The quantification of the uncertainty performed in this study is still exposed to a considerable degree of subjectivity. Nevertheless, the approach applied here has the advantage of ensuring internal consistency within the system and this should be a crucial aspect to be maintained in any future methodological development.

To summarize, this Thesis has developed and tested novel methodological approaches based on MFA, aimed at enhancing and monitoring regional nutrients management. Such methods applied to the Austrian P budget have proven that they can provide relevant findings and several benefits to the public authorities in charge of designing governance strategies and of monitoring their performance.

The accounting schemes proposed in this work should be implemented and maintained by national or international statistics and by public agencies. The scientific community can nevertheless still contribute greatly to the further development of these methodologies and to the deeper understanding of the systems involved. This Thesis, in particular, has pointed out new research questions that shall be addressed in the future.

The long-term effects exerted by large floods on the P in-stream stocks in large rivers should be integrated in the models that estimate nutrients emissions and river loads, but this is still very challenging, owing to the high complexity of retention and mobilization processes.

Some compartments of the Austrian P budget are affected by high uncertainty and this partially undermines the assessment of the optimization potentials. Therefore, further efforts should be undertaken to gather more data and information and to deepen the understanding of the system. A notable example is the relatively large net P accumulation on parks and green gardens, for which only poor information is available.

Further, the optimization performed in this Thesis constitutes only a first step towards a full optimization, which needs to consider also the costs involved in each different action as well as other social or environmental goals that may show relevant trade-offs with the objectives set in this work.

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### Appendix A

## Sub-systems of the MFA model

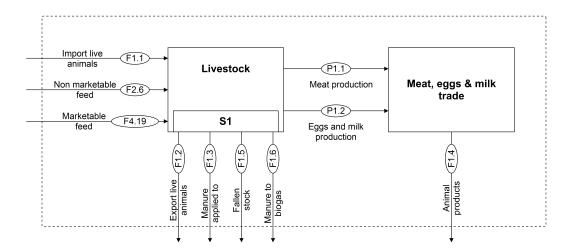


Figure A.1: MFA model of the Animal husbandry subsystem.

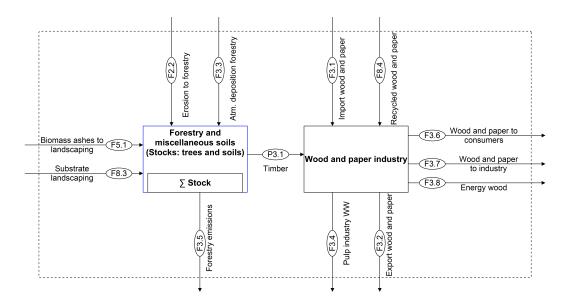


Figure A.2: MFA model of the Forestry and miscellaneous soils subsystem.

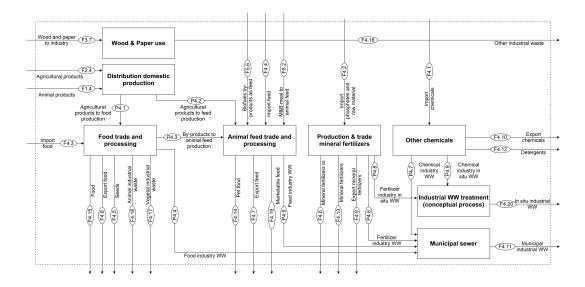


Figure A.3: MFA model of the *Industry* subsystem.

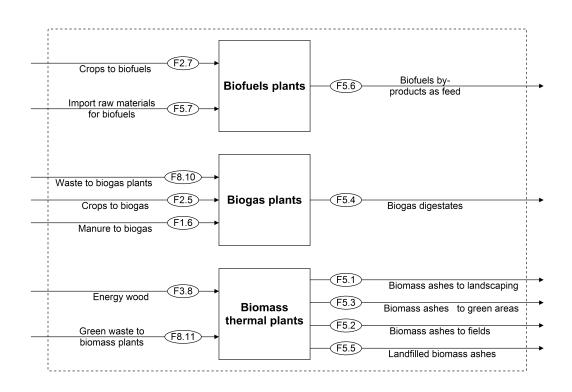


Figure A.4: MFA model of the *Bioenergy* subsystem.

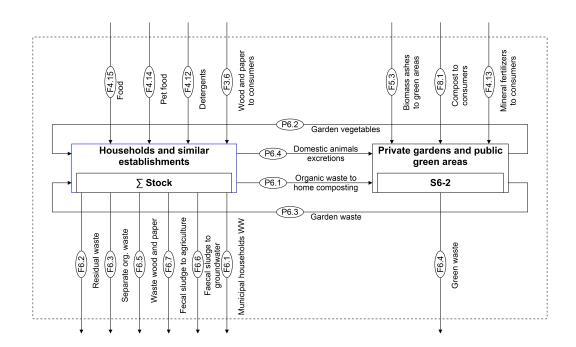


Figure A.5: MFA model of the *Consumption* subsystem.

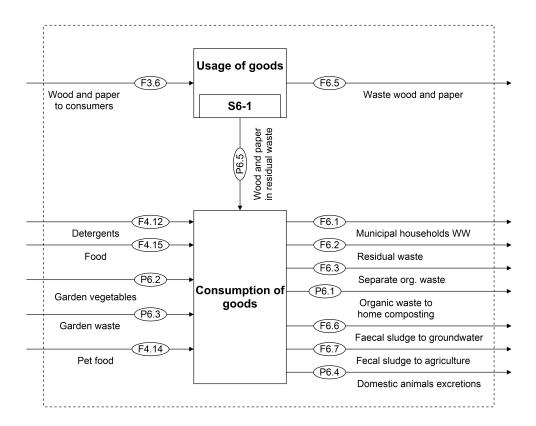


Figure A.6: MFA model of the *Households and similar establishments* second level subsystem.

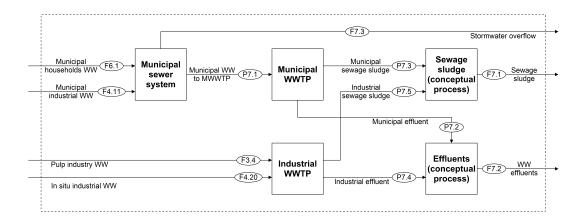


Figure A.7: MFA model of the Wastewater management subsystem.

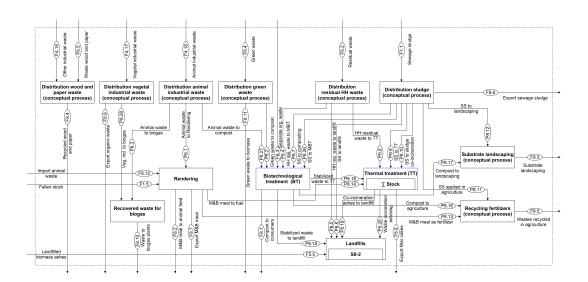


Figure A.8: MFA model of the *Waste management* subsystem.

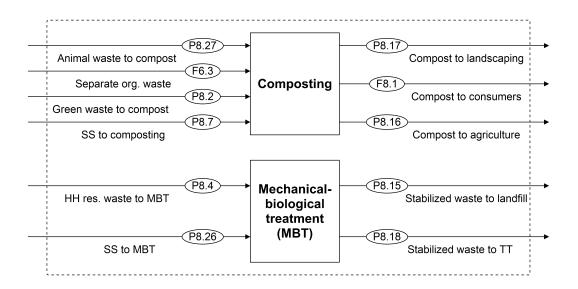


Figure A.9: MFA model of the *Biotechnological treatment* (BT) second level subsystem.

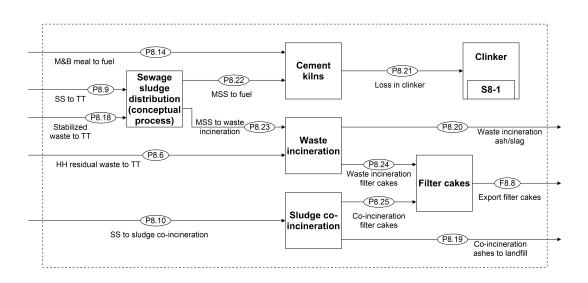


Figure A.10: MFA model of the *Thermal treatment* (TT) second level subsystem.

Appendix B

# Description of flows, stocks, transfer coefficients, equations for their calculation and data sources

Flow N.	Flow N. Flow name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
F1.1	Import live animals	Amount of P in imported live animals	Yearly imported animal carcass weight x Amount of P in imported live animals PC	Statistik Austria, 1	UBA,4 Glenck et al. 1995 LFL, 2013 Sibbesen and Runge-Metzger,1995
F1.2	Export live animals	Amount of P in exported live animals	Yearly imported animal carcass weight x conversion factor of carcass-live weight x PC	Statistik Austria, 1	UBA,4 Glenck et al. 1995 LFL, 2013 Sibbesen and Runge-Metzger,1995
F1.3	Manure applied to fields	Amount of P in manure applied on agricultural fields	(Yearly n° of animals x specific P excretion) - (yearly manure treated in biogas plants x PC)	BMLFUW, 1 E-Control UBA,4 Resch et al. 2004 Jyväskylä Innovation Oy,2009 Pötsch, 2004	Kroiss et al., 1998 BMLFUW, 4,5
F1.4	Animal products	Amount of P in domestic production of meat, milk and eggs	STAN – principle of mass balance		
F1.5	Fallen stock	Amount of P in animals that died for reasons other than slaughtering	Yearly fallen animal live weight x PC	BMG,1,2 UBA,4 BMLFUW,2	UBA,4 Glenck et al. 1995 LFL, 2013 Sibbesen and Runge-Metzger,1995
F1.6	Manure to biogas	Amount of P in manure digested in biogas plants	Yearly manure x PC	E-Control UBA,4 Resch et al. 2004 Jyväskylä innovation Oy,2009 Pötsch, 2004	Kroiss et al., 1998 BMLFUW, 4, 5
F2.1	Atm. deposition agric.	Amount of P transported onto agricultural soils through wet and dry atmospheric deposition	Yearly n° of agricultural hectares x P atmospheric deposition per hectare	BMLFUW,1	Glenck et al., 1995
F2.2	Erosion to forestry	Amount of P eroded from agricultural fields and transported to forestry and miscellaneous soils	Estimation		
F2.3	Agricultural emissions	Amount of P transported from agricultural fields to water bodies	MONERIS model results for 2001-2006, yearly scaled through yearly precipitation factor	Zessner et al., 2011	

Table B.1: Description of flows, equations and data sources for their calculation.

Flow N.	Flow N. Flow name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
F2.4	Agricultural products	Amount of P in crops supplied to food- animal feed industry	(Yearly crops production x PC) – (Yearly use of energy crops x PC)	Statistik Austria,4-17	LFL, 2013 Kroiss et al., 1998
F2.5	Crops to biogas	Amount of P in energy crops used in biogas plants	Yearly consumption of energy crops by biogas plants x PC	E-Control Resch et al. 2004 Jyväskylä Innovation Oy,2009 Pötsch, 2004	LFL, 2013 Kroiss et al., 1998
F2.6	Non marketable feed	Amount of P in non marketable animal feed	Yearly available fodder x PC	BMLFUW,1 Statistik Austria,19	COMIFER,2006 LFL, 2013 Kroiss et al., 1998
F2.7	Crops to biofuels	Amount of P in inland produced crops used for biofuels production	(Yearly consumption of energy crops for biofuels production x PC) x domestic fraction	UBA,5 BMLFUW,1	COMIFER,2006 European Standard EN14214/EN15487 Kroiss et al., 1998 Hein and Leemans,2012 LFL,2013 Rutkowski,1971 ORNL
F3.1	Import wood and paper	Amount of P in imported wood and paper	Yearly imported wood and paper x PC	Statistik Austria, 18	Kroiss et al., 1998 Binder et al., 2009 Antikainen et al., 2004
F3.2	Export wood and paper	Amount of P in exported wood and paper	Yearly exported wood and paper x PC	Statistik Austria, 18	Kroiss et al., 1998 Binder et al., 2009 Antikainen et al., 2004
F3.3	Atm. deposition forestry	Amount of P transported onto forestry and miscellaneous soils through wet and hectares x P atmospheric deposition per dry atmospheric deposition	Yearly n° of forestry and miscellaneous hectares x P atmospheric deposition per hectare	BMLFUW,1	Glenck et al., 1995
F3.4	Pulp industry WW	Amount of P in wastewater of the pulp industry which is treated in-situ	Readily available data	ICPDR,1 Kroiss et al.,1998	
F3.5	Forestry emissions	Amount of P transported from forestry and miscellaneous soils to water bodies	MONERIS model results for 2001-2006, yearly scaled through yearly precipitation factor	Zessner et al., 2011	
F3.6	Wood and paper to consumers	Amount of P in wood an paper consumed by households and similar establishments	(Household wood consumption x PC) + (Total paper consumption x PC x fraction consumed by households)	Austrian Energy Agency Austropapier	Kroiss et al., 1998 Binder et al., 2009 Antikainen et al., 2004
F3.7	Wood & paper to industry	Amount of P in wood and paper consumed by the industrial sector	(Industrial wood consumption x PC) + (Total paper consumption x PC x fraction consumed by industriess)	Austrian Energy Agency Austropapier	Kroiss et al., 1998 Binder et al., 2009 Antikainen et al., 2004
F3.8	Energy wood	Amount of P in energy wood consumed in biomass plants	Energy wood consumption x PC	BMLFUW,1 Austrian Energy Agency	Kroiss et al., 1998 Binder et al., 2009 Antikainen et al., 2004

Flow N.	Flow N. Flow name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
F4.1	Import chemicals	Amount of P in imported chemical products	(Yearly imported selected chemical products x PC) – (yearly phosphoric acids used by fertilizer industry x PC)	Statistik Austria, 18	Calculations based on molar mass of compounds
F4.2	Import min.fertilizers and phosphate ore	Amount of P in imported phosphate ore and phosphate mineral fertilizers	Yearly imported mineral phosphate products + yearly imported phosphate ore and intermediate products (estimated as difference between domestic production and import of products)	IFA	
F4.3	Import food	Amount of P in imported food	Yearly imported food x PC	Statistik Austria,1-18	LFL, 2013 Kroiss et al., 1998
F4.4	Import feed	Amount of P in imported animal feed	Yearly imported animal feed x PC	Statistik Austria,18 BMLFUW,1	LFL, 2013 Binder et al., 2009
F4.5	Seeds	Amount of P in seeds	Yearly crop seeds sales x PC	Statistik Austria,4,7-9 BMLFUW,7	White,2012
F4.6	Mineral fertilizers to agriculture	Amount of P in phosphate fertilizers applied in agriculture	Total yearly domestic consumption of phosphate mineral fertilizers x proportion sold for agricultural use	IFA BMLFUW,1	Heinzlmaier, 2010 Strasser,2010
F4.7	Export feed	Amount of P in exported animal feed	Yearly exported animal feed x PC	Statistik Austria, 18 BMLFUW ,1	LFL, 2013 Kroiss et al., 1998 Binder et al., 2009
F <b>4</b> .8	Export food	Amount of P in exported food	Yearly exported food x PC	Statistik Austria,1-18	LFL, 2013 Kroiss et al., 1998
F4.9	Export mineral fertilizers	Amount of P in exported phosphate fertilizers	Yearly exported phosphate mineral fertilizers	IFA	
F4.10	Export chemicals	Amount of P in exported chemical products	Yearly exported selected chemical products x PC	Statistik Austria, 18	Calculations based on molar mass of compounds
F4.11	Municipal ind. WW	Industrial contribution to P load in municipal wastewater	Total yearly P load in municipal wastewater – P load in households municipal wastewater	Statistik Austria, 22 BMLFUW, 8, 9	Lindtner and Zessner, 2003 Posch, 1999
F4.12	Detergents	Amount of P in detergents used in households and similar establishments	Yearly n° of inhabitants x specific P detergents consumption	Binder et al.,2009 de Madariaga et al.,2007 Statistik Austria,22	
F <b>4</b> .13	Min. fertilizers to consumers	Amount of P in phosphate fertilizers applied in private gardens and urban green areas	Yearly total domestic consumption of phosphate mineral fertilizers x proportion sold for uses other than agriculture	IFA BMLFUW,1	Heinzlmaier, 2010 Strasser,2010
F4.14	Pet food	Amount of P consumed through pet food	Yearly n° of cats and dogs x average P consumption per animal	IEMT	Kalmykova, 2012
F4.15	Food	Amount of P in food consumed by population	Yearly available food supply x PC	Statistik Austria,1-17 BMLFUW,6	LFL, 2013 Kroiss et al., 1998
F <b>4</b> .16	Other industrial waste	Amount of P in wood and paper waste generated by industrial sector	Yearly industrial wood and paper waste x PC	BMLFUW,2	Kroiss et al., 1998 Binder et al., 2009 Antikainen et al., 2004

#### B. Description of MFA inputs, calculation and data sources

Flow N.	Flow name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
F4.17	Vegetal industrial waste	Amount of P in vegetal wastes and by- products generated by the industry	(Yearly industrial waste of vegetal origin x PC) – (yearly industrial waste of vegetal origin directly reused for animal feed production x PC)	BMLFUW, 2 Statistik Austria	LFL, 2013 Kroiss et al., 1998 Binder et al., 2009 Klages et al., 2009
F <b>4.</b> 18	Animal industrial waste	Amount of P in animal wastes and by-	(Yearly industrial waste of animal origin x PC) – (yearly industrial waste of animal origin directly reused for animal feed production x PC)	BMLFUW,2 UBA,4	Binder et al., 2009 Hoppenheidt et al. 2008 Kroiss et al., 1998 Lamprecht et al., 2011 LFL, 2013
F4.19	Marketable feed	Amount of P in marketable animal feed consumption	Yearly available marketable animal feed x PC	BMLFUW,1 Statistik Austria,19	LFL, 2013 Kroiss et al., 1998 Binder et al., 2009
F4.20	In situ ind. WW	Amount of P in industrial wastewater treated in situ	Readily available data	BMLEUW,8,9 Kroiss et al.,1998 ICPDR,1	
F5.1	Biomass ashes to landscaping	Amount of P in biomass ashes applied in landscaping activities	(Yearly total biomass ashes – yearly landfilled biomass ashes) x proportion applied in landscaping activities x PC	Austrian Energy Agency UBA,2,8	Obernberger and Supancic, 2009
F5.2	Biomass ashes to fields	Amount of P in biomass ashes applied on agricultural fields	<u> </u>	Austrian Energy Agency UBA,2,8	Obernberger and Supancic, 2009
F5.3	Biomass ashes to green areas	Amount of P in biomass ashes applied on private gardens and public green areas	(Yearly total biomass ashes – yearly landfilled biomass ashes) x proportion applied in private gardens and public green areas x PC	Austrian Energy Agency UBA,2,8	Obernberger and Supancic, 2009
F5.4	Biogas digestates	Amount of P in digestates generated by biogas plants	STAN – Principle of mass conservation		
F5.5	Landfilled biomass ashes	Amount of P in biomass ashes disposed of in landfills	Yearly landfilled biomass ashes x PC	Austrian Energy Agency UBA,2,8	Obernberger and Supancic, 2009
F5.6	Biofuels by-products as feed	Amount of P in by-products generated by the biofuel industry	(Yearly generation of bioethanol by- products x PC) + (yearly production of biodiesel x ratio by-product/product x PC)	UBA,5 BMLFUW,1	COMIFER,2006 European Standard EN14214/EN15487 Kroiss et al., 1998 Hein and Leemans,2012 LFL,2013 Rutkowski,1971 Simpson et al.,2008 ORNL
F5.7	Import raw materials for biofuels	Amount of P in imported raw materials for biofuel production	(Yearly consumption of energy crops for biofuels production x PC) x imported fraction	UBA,5 BMLFUW,1	COMIFER,2006 European Standard EN14214/EN15487 Kroiss et al., 1998 Hein and Leemans,2012 LFL,2013 Rutkowski,1971 ORNL

Flow N.	Flow N. Flow name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
F6.1	Municipal households WW	Households municipal wastewater	(Yearly n° of inhabitants connected to sewers x yearly specific P load per capita in wastewater) + (Yearly fecal sludge treated in MWWTP x PC)	Statistik Austria,22 BMLFUW,8,9	Posch, 1999 Lindtner and Zessner, 2003 Kroiss et al., 2008
F6.2	Residual waste	Amount of P in residual waste from households and similar establishments	Yearly household residual waste x PC (PC yearly modified according to proportion of organic fraction)	BMLFUW,2	Glenck et al., 1995 Skutan and Brunner, 2006
F6.3	Separate org. waste	Amount of P in separately collected organic waste from households and similar establishments	Yearly separately collected organic waste x PC	BMLFUW,2	EPEA, 2008 Glenck et al., 1995 Sokka et al., 2004
F6.4	Green waste	Amount of P in separately collected green waste from private gardens and public green areas	Yearly separately collected green waste x PC	BMLFUW,2	Binder et al., 2009 Kroiss et al., 1998
F6.5	Waste wood and paper	Amount of P in separately collected wood and paper waste from households and similar establishments	Yearly separately collected wood and paper waste x PC	BMLFUW,2	Antikainen et al., 200 <b>4</b> Binder et al., 2009 Kroiss et al., 1998
F6.6	Fecal sludge to groundwater	Amount of P in fecal sludge discharged in the underground and groundwater	Readily available data as of 1992, yearly scaled according to connection rate of the population to the sewer system and the fraction of fecal sludge treated in MWWTP	BMLFUW,8,9 Kroiss et al., 1998	
F6.7	Fecal sludge to agriculture	Amount of P in fecal sludge directly applied on agricultural fields	Readily available data as of 1992, yearly scaled according to connection rate of the population to the sewer system and the fraction of fecal sludge treated in MWWTP	BMLFUW,8,9 Kroiss et al., 1998	
F7.1	Sewage sludge	Amount of P contained in municipal and inductrial sewage sludge	STAN – principle of mass conservation		
F7.2	WW effluents	Amount of P contained in effluent of municipal and industrial WWTP	STAN – principle of mass conservation		
F7.3	Stormwater overflow	Amount of P contained in stormwater overflow	3% of P load of municipal wastewater	BMLFUW,8,9 Fenz, 2002	
F8.1	Compost to consumers	Amount of P in compost applied on private gardens and public green areas	Total compost production x PC x fraction used in private gardens and public green areas	BMLFUW,2,3	BMLFUW,3
F8.2	M&B meal to animal feed	Amount of P in meat and bone meal used as animal feed	Meat and bone meal reused as animal feed x PC	BMLFUW,2 UBA,4 BGBI. I Nr. 143/2000	Binder et al., 2009 Kroiss et al., 1998 Lamprecht et al., 2011 Klock and Taber, 1996
F8.3	Substrate landscaping	Amount of P in wastes recovered for substrate landscaping	STAN – principle of mass conservation		
F8.4	Recycled wood and paper	Amount of P in wood and paper waste that is recycled	STAN – principle of mass conservation		
F8.5	Wastes recycled in agriculture	Amount of P in wastes and by-products applied on agricultural fields	STAN – principle of mass conservation		

B. Description of MFA inputs, calculation and data sources

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BMEUM         BMEUM <th< td=""><td>F8.6</td><td>Export sewage sludge</td><td></td><td>Total yearly P in sewage sludge x yearly exported fraction</td><td>BMLFUW,1,8,9 UBA,1,6,7</td><td></td></th<>	F8.6	Export sewage sludge		Total yearly P in sewage sludge x yearly exported fraction	BMLFUW,1,8,9 UBA,1,6,7	
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Master to blogges plants         Amount of P in industrial wastes and bring appendix         TAN - principle of mass conservation         Molecular transmission         Molecular transmis	F8.9	Export organic waste	Amount of P in exported organic waste	Yearly exported organic waste x PC	BMLFUW,2 UBA,4	Binder et al., 2009 Klages et al., 2009 Kroiss et al., 1998 LFL, 2013
Green waste to biomass plants         Amount of P in green waste used in plants         Verity green aloo of biomass thermal plants         BM.FUW, 2           Import animal waste         Amount of P in imported animal waste x PC         BM.FUW, 2         BM.FUW, 2           Import animal waste         Amount of P in imported animal waste x PC         BM.FUW, 10         BM.FUW, 10           Import animal waste         Amount of P in imported animal waste x PC         BM.FUW, 10         BM.FUW, 10           Import water bodies         Amount of P in imported animal waste x PC         BM.FUW, 10         BM.FUW, 10           Import water bodies         Amount of P in imported animal waste x PC         BM.FUW, 10         BM.FUW, 10           Import water bodies         Amount of P in imported animal waste x PC         BM.FUW, 10         BM.FUW, 10           Import water bodies         Amount of P in infores at their entrance         Meet flow and monthly P concentration         BM.FUW, 10           Meet production         Amount of P in domestic meet         Yearly wast flow and monthly P concentration         BM.FUW, 10           Meet production         Amount of P in domestic meet         Yearly imber extraction x PC         BM.FUW, 10           Meet production         Amount of P in domestic meet         Yearly imber extraction x PC         BM.FUW, 10           Meet production         Amount of P in domestic	F8.10	Waste to biogas plants	Amount of P in industrial wastes and by- products treated in biogas plant	STAN – principle of mass conservation		
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Import water bodiesAmount of P in rivers at their entranceModeling of P yearly load based on dailyBMLEUW, 10Export water bodiesAmount of P in rivers at their exit fromwater flow and monthly P concentrationBMLEUW, 10Export water bodiesAmount of P in rivers at their exit fromModeling of P yearly load based on dailyBMLEUW, 10Meat productionAmount of P in domestic meatVearly meat production x PCStatistik Austria.1Eggs and milk productionAmount of P in domestic meatVearly meat production x PCStatistik Austria.1ImberAmount of P in domestic meatVearly trans to concentrationBMLEUW, 10Eggs and milk productionAmount of P in domestic eggs and milk production x PCStatistik Austria.1BMLEUW, 10ImberAmount of P in domestic eggs and milk production x PCBMLEUW, 10MontelyW, 11ImberAmount of P in domestic enges and milk production x PCBMLEUW, 10BMLEUW, 10ImberAmount of P in domestic enges and milk production x PCBMLEUW, 10BMLEUW, 10ImberAmount of P in domestic enges and milk production x PCBMLEUW, 11BMLEUW, 11ImberAmount of P in domestic enges and milk production x PCBMLEUW, 11BMLEUW, 11ImberAmount of P in domestic enges allocatedStatistik Austria.2.1BMLEUW, 11ImberAmount of P in domestic enges allocatedStatistic Austria.2.1BMLEUW, 11By-products to animal feedAmount of P in food inducty veased for animal feedBMLEUW, 11BMLEUW, 11By-products to animal fe	F8.12	Import animal waste	Amount of P in imported animal waste	Yearly imported animal waste x PC	BMLFUW,2 UBA,4	Hoppenheidt et al.,2000 Kroiss et al., 1998 Lamprecht et al., 011 LFL,2013
Export water bodiesAmount of P in rivers at their exit from MatriaModeling of P yearly load based on daily White WW, 10BMLEUW, 10Meat productionAmount of P in domestic meat productionVearly weat production x PC + (Yearly milk production x PC) + (Yearly mount of P in domestic cross allocated mount of P in domestic cross allocated productionTAN - principle of mass conservationStatistik Austria, 2.3 mount of P in domestic cross allocated mount of P in domestic cross allocated productionTAN - principle of mass conservationStatistik Austria, 2.3 mold of mount of P in domestic cross allocated mount of P in domestic cross allocated productionTAN - principle of mass conservationStatistik Austria, 2.3 mold of mount of P in domestic cross allocated mold of mass conservationModeling p in the food mold of mount of P in domestic cross allocated mount of P in domestic cross allocated 	F9.1	Import water bodies		Modeling of P yearly load based on daily water flow and monthly P concentration		BMLFUW, 11 ICPDR,2
Meat productionAmount of P in domestic meat productionYearly meat production x PCStatistik Austria,1 BMLFUW,1Eggs and milk productionAmount of P in domestic eggs and milk production x PC)Yearly eggs production x PC) + (Yearly 	F9.2	Export water bodies		Modeling of P yearly load based on daily water flow and monthly P concentration		BMLFUW, 11 ICPDR,2
Eggs and milk productionAmount of P in domestic eggs and milk(vearly eggs production x PC) + (Yearly milk production x PC)Statistik Austria,2,3 BMIFUW,1TimberAmount of P in domestic timber productionYearly timber extraction x PC)Statistik Austria,21 	P1.1	Meat production	Amount of P in domestic meat production	Yearly meat production x PC	Statistik Austria,1 BMLFUW,1	Kroiss et al., 1998 LFL, 2013
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Products to food productionAmount of P in domestic crops allocatedSTAN - principle of mass conservationMount of P in domestic crops allocatedSTAN - principle of mass conservationMount of P in domestic crops allocatedSTAN - principle of mass conservationMount of P in domestic crops allocatedSTAN - principle of mass conservationMount of P in domestic crops allocatedSTAN - principle of mass conservationMount of P in domestic crops allocatedSTAN - principle of mass conservationMount of P in domestic crops allocatedSTAN - principle of mass conservationMount of P in domestic crops allocatedMount of D in domestic cro	P3.1	Timber	Amount of P in domestic timber production	Yearly timber extraction x PC	Statistik Austria, 21 BMLFUW,1 Waldinventur	Kroiss et al., 1998 Antikainen et al., 2004
Products to feed production         Amount of P in domestic crops allocated         STAN – principle of mass conservation         Amount of P in domestic crops allocated         STAN – principle of mass conservation         Statistic Mustria, 20         Must	P4.1	Products to food production	Amount of P in domestic crops allocated to food production	STAN – principle of mass conservation		
By-products to animal feed products directly reused for animal feed production         Amount of P in food industry by- production         Yearly organic by-products of the food industry directly reused for animal feed production         Statistik Austria,20           By production         production         production x PC         UBA,4           Amount of P in food industry wastewater connected to municipal sewer         production x PC         BMLFUW,8,9           Food WW         connected to municipal sewer         in municipal wastewater treated in municipal sewer         BMLFUW,8,9	P4.2	Products to feed production	Amount of P in domestic crops allocated to marketable animal feed production	STAN – principle of mass conservation		
Food WW         Amount of P in food industry wastewater connected to municipal sewer         P load in industrial wastewater treated in municipal WWTP x fraction connected to municipal sewer	P4.3	By-products to animal feed production	Amount of P in food industry by- products directly reused for animal feed production	Yearly organic by-products of the food industry directly reused for animal feed production x PC	Statistik Austria, 20 UBA,4	LFL, 2013 Kroiss et al., 1998 Binder et al., 2009
	P4.4	Food WW	Amount of P in food industry wastewater connected to municipal sewer		BMLFUW,8,9 Kroiss et al.,1998 ICPDR,1	

Flow N.	. Flow name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
P4.5	Animal feed WW	Amount of P in feed industry wastewater connected to municipal sewer	P load in industrial wastewater treated in municipal WWTP x fraction corresponding to feed industry	BMLFUW,8,9 Kroiss et al.,1998 ICPDR,1	
P4.6	Fertilizer WW	Amount of P in fertilizer industry wastewater connected to municipal sewer	P load in industrial wastewater treated in municipal WWTP x fraction corresponding to fertilizer industry	BMLFUW,8,9 Kroiss et al.,1998 ICPDR,1	
P4.7	Chemical WW	Amount of P in chemical industry wastewater connected to municipal sewer	P load in industrial wastewater treated in municipal WWTP x fraction corresponding to chemical industry	BMLFUW,8,9 Kroiss et al.,1998 ICPDR,1	
P4.8	Fertilizer industry in situ WW	Amount of P in fertilizer industry wastewater treated in situ	P load in wastewater of chemical and fertilizer industry treated in situ x fraction corresponding to fertilizer industry	BMLFUW,8,9 Kroiss et al.,1998 ICPDR,1	
P4.9	Chemical industry in situ WW	Amount of P in chemical industry wastewater treated in situ	P load in wastewater of chemical and fertilizer industry treated in situ x fraction corresponding to chemical industry	BMLFUW,8,9 Kroiss et al.,1998 ICPDR,1	
P6.1	Organic waste to home composting	Amount of P in organic waste from households and similar establishments, handled through home compositing	Separately collected household organic waste x PC	BMLFUW,2	EPEA, 2008 Glenck et al., 1995 Sokka et al., 2004
P6.2	Garden vegetables	Amount of P in vegetables grown in private gardens	Yearly garden vegetables production x PC	Statistik Austria,12	COMIFER,2006 Kroiss et al., 1998 LFL,2013
P6.3	Garden waste	Amount of P in garden residues collected together with household waste	Yearly separately collected household organic waste x fraction composed of garden waste x PC	BMLFUW,2	Binder et al., 2009 Kroiss et al., 1998
P6.4	Domestic animals excretions	Amount of P in domestic animals excretions, ending up on gardens and public green areas	Estimation		
P6.5	Wood & paper in residual waste	Amount of P in wood and paper fraction of household residual waste	Yearly household residual waste x fraction of wood and paper x PC	BMLFUW,2	Kroiss et al., 1998 Binder et al., 2009 Antikainen et al., 2004
P7.1	Municipal WW to MWWTP	Amount of P in municipal wastewater treated in WWTP	Readily available data	BMLFUW,8,9	
P7.2	Municipal effluent	Amount of P in effluents of municipal WWTP	Readily available data	BMLFUW,8,9	
P7.3	Municipal sewage sludge	Amount of P in sewage sludge generated Input – output in municipal WWTP (both by municipal WWTP	Input – output in municipal WWTP (both readily available data)	n BMLFUW, 2,8,9 UBA,1,6,7	
P7.4	Industrial effluent	Amount of P in effluents of industrial in situ WWTP	Readily available data	BMLFUW,8,9	
P7.5	Industrial sewage sludge	Amount of P in sewage sludge generated Input – output in industrial in situ WWTP (both readily available data)	lnput – output in industrial in situ WWTP (both readily available data)	BMLFUW,8,9 Kroiss et al., 1998 UBA,1	

B. Description of MFA inputs, calculation and data sources

Flow N.	Flow name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
P8.1	Animal waste to Rendering	Amount of P in animal waste and by- products handled through the rendering process	Yearly animal waste and by-products handled through rendering x PC	BMLFUW,2 UBA,4	Lamprecht, 2011 Kroiss, 1998 EU, 2001
P8.2	Green waste to compost	Amount of P in green wastes handled through compositing process	Yearly separately collected green wastes handled through composting x PC	BMLFUW,2,3	BMLFUW,3 Binder et al., 2009 Kroiss et al., 1998
P8.3	Animal waste to biogas	Amount of P in animal waste handled in biogas plants	Yearly animal waste and by-products used in biogas plants x PC	E-Control UBA,4 Resch et al. 2004 Jyväskylä Innovation Oy,2009 Pötsch, 2004	Lamprecht, 2011 Kroiss, 1998 EU, 2001
P8.4	HH res. waste to MBT	Amount of P in household residual waste handled through mechanical-biological treatment	Yearly household residual waste x PC (PC yearly modified according to proportion of organic fraction) x fraction stabilized through mechanical-biological treatment	BMLFUW,2 UBA,3	Glenck et al., 1995 Skutan and Brunner, 2006
P8.5	HH res. waste to landfill	Amount of P in landfilled household residual waste	Yearly household residual waste x PC (PC yearly modified according to proportion of organic fraction) x landfilled fraction	BMLFUW, 2	Glenck et al., 1995 Skutan and Brunner, 2006
P8.6	HH res. waste to $\Pi$	Amount of P in incinerated household residual waste handled	Yearly household residual waste x PC (PC yearly modified according to proportion of organic fraction) x incinerated fraction	BMLFUW, 2	Glenck et al., 1995 Skutan and Brunner, 2006
P8.7	SS to composting	Amount of P in composted sewage sludge	Total yearly P in sewage sludge x yearly composted fraction	BMLFUW,2,3,8,9 UBA,1,6,7	
P.8.8	SS to landfill	Amount of P in landfilled sewage sludge	Total yearly P in sewage sludge x yearly landfilled fraction	BMLFUW,2,8,9 UBA,1,6,7	
P.8.9	SS to TT	Amount of P in incinerated sewage sludge	Total yearly P in sewage sludge x yearly incinerated fraction	BMLFUW,2,8,9 UBA,1,6,7	
P8.10	SS to co-incineration	Amount of P in sewage sludge incinerated in specific sludge co- incineration plants	Total yearly P in sewage sludge x yearly fraction incinerated in sludge co- incineration plants	BMLFUW,2,8,9 UBA,1,6,7	
P8.11	SS applied in agriculture	Amount of P in sewage sludge directly applied on agricultural fields	Total yearly P in sewage sludge x yearly fraction directly applied in agriculture	BMLFUW,2,8,9 UBA,1,6,7	
P8.12	SS to landscaping	Amount of P in sewage sludge applied in landscaping activities	Total yearly P in sewage sludge x yearly fraction applied in landscaping activities	BMLFUW,2,8,9 UBA,1,6,7	
P8.13	M&B meal as fertilizer	Amount of P in meat and bone meal applied on agricultural fields	Yearly meat and bone meal applied on agricultural fields x PC	BMLFUW,2 UBA,4	Binder et al., 2009 Kroiss et al., 1998 Lamprecht et al., 2011 Klock and Taber, 1996

Flow N.	Flow name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
P8.14	M&B meal to fuel	Amount of P in meat and bone meal used as fuel in cement kilns	Yearly meat and bone meal used as fuel in cement kilns x PC	BMLFUW,2 UBA,4	Binder et al., 2009 Kroiss et al., 1998 Lamprecht et al., 2011 Klock and Taber, 1996
P8.15	Stabilized waste to landfill	Amount of P in waste stabilized through mechanical-biological treatment and then landfilled	Yearly waste stabilized through mechanical-biological treatment and then landfilled x PC	BMLFUW,2 UBA,3	Estimation
P8.16	Compost to agriculture	duct applied lds	Total comp appl	BMLFUW,2,3	BMLFUW,3
P8.17	Compost to landscaping	Amount of P in compost product applied Total compost production x PC x fraction in landscaping activities	Total compost production x PC x fraction applied in landscaping activities	BMLFUW,2,3	BMLFUW,3
P8.18	Stabilized waste to TT	Amount of P in waste stabilized through mechanical-biological treatment and then incinerated	Yearly waste stabilized through mechanical-biological treatment and then incinerated x PC	BMLFUW UBA,3	Estimation
P8.19	Ash co-inc. to landfill	Amount of P in landfilled ashes of sludge co-incineration	STAN – transfer coefficient		
P8.20	Ash/slag to landfill	Amount of P in landfilled ashes of municipal waste incineration	STAN – transfer coefficient		
P8.21	Loss in clinker	Amount of total P lost in cement kilns	STAN – principle of mass conservation		
P8.22	SS to fuel	Amount of P in sewage sludge used as fuel in cement kilns	Total yearly P in sewage sludge x yearly fraction used as fuel in cement kilns	BMLFUW,2,8,9 UBA,1,6,7	
P8.23	SS to waste incineration	Amount of P in sewage sludge incinerated in municipal waste incineration	Total yearly P in sewage sludge x yearly fraction incinerated with municipal waste	BMLFUW,2,8,9 UBA,1,6, <b>7</b>	
P8.24	Filter cakes Inc.	Amount of P in filter cakes generated by municipal waste incineration	STAN – transfer coefficient		
P8.25	Filter cakes Co-inc.	Amount of P in filter cakes generated by sludge co-incineration	STAN – transfer coefficient		
P8.26	SS to MBT	Amount of P in sewage sludge stabilized through mechanical-biological process	Total yearly P in sewage sludge x yearly fraction stabilized through mechanical- biological treatment	BMLFUW,2,8,9 UBA,1,3,6,7	
P8.27	Animal waste to compost	Amount of P in composted waste and by- products of animal waste and by- products of animal origin	Yearly composted animal waste and by- products x PC	BMLFUW,2,3 UBA,4	BMLFUW,3
P8.28	Veg. ind. to biogas	Amount of P in organic industrial waste of vegetal source treated in biogas plants	Industrial waste of vegetal source used by biogas plants x PC	E-Control Resch et al. 2004 Jyväskylä Innovation Oy,2009 Põtsch, 2004	LFL, 2013 Kroiss et al., 1998 Binder et al., 2009

Stock N.	Stock N. Stock name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
<b>S1</b>	Livestock	Amount of P in the livestock at the beginning of the year	Yearly livestock x PC	BMLFUW,1	Kroiss et al., 1998 LFL, 2013
S2	Agricultural fields	Amount of P in the agicultural fields at the beginning of the year	Estimation		
S3	Trees and soil	Amount of P in the trees, forestry and miscellaneous soils at the beginning of the year	Estimation		
S6-1	Stock in buildings and furniture	Amount of P in the buildings and furniture at the beginning of the year			
S6-2	Private gardens & public green areas	Amount of P in the soils o private gardens and public green areas at the beginning of the year	Estimation		
S8-1	Clinker	Amount of P lost in the cement kilns at the beginning of the year	Estimation of loss until beginning of 1990		
S8-2	Landfills	Amount of P in the landfills at the beginning of the year			
6S	Water bodies	Amount of P in the water bodies at the beginning of the year			

Table B.2: Description of stocks and transfer coefficients, equations and data sources for their calculation.

Stock N.	Stock N. Stock change rate name	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
<b>S1</b>	Livestock	Yearly change of P in the livestock at the beginning of the year	(Yearly livestock x PC) - (Previous year livestock x PC)	BMLFUW,1	Kroiss et al., 1998 LFL, 2013
S2	Agricultural fields	Yearly change of P in the agicultural fields at the beginning of the year	STAN – principle of mass conservation		
S3	Trees and soil	Yearly change of P in the trees, forestry and miscellaneous soils at the beginning of the year	STAN – principle of mass conservation		
S6-1	Stock in buildings and furniture	Yearly change of P in the buildings and furniture at the beginning of the year	STAN – principle of mass conservation		
S6-2	Private gardens & public green areas	Yearly change of P in the soils o private gardens and public green areas at the beginning of the year	STAN – principle of mass conservation		
S8-1	Clinker	Yearly change of P lost in the cement kilns at the beginning of the year	STAN – principle of mass conservation		
S8-2	Landfills	Yearly change of P in the landfills at the beginning of the year	STAN – principle of mass conservation		
6S	Water bodies	Yearly change of P in the water bodies at the beginning of the year	STAN – principle of mass conservation		

Process N.	rocess Process name (transfer N. coefficient)	Description	Calculation	Data sources – goods and readily available flows	Data sources – P concentrations and other calculation factors
P30	Waste incineration	Transfer of phosphorus to ashes during waste incineration		Morf et al., 2005	
P29	Sludge co-incineration	Transfer of phosphorus to ashes during sludge co-incineration		UBA,1	

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- $^{11}$  Supply balance sheet for vegetable fats and oils as of 1994/1995
- $^{12}$  Supply balance sheet for vegetables as of 1994/1995
- $^{13}$  Supply balance sheet for wine as of 1994/1995
- $^{14}$  Supply balance sheet for beer as of 1994/1995
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### Characterization of data uncertainty

The CV values presented in Table 2.2 in the manuscript are dependent only on two parameters for each evaluation score (1 to 4). The parameter values for the different sensitivity levels are defined as follows:  $a_{not \ sensitive} = 1.0$ ,  $a_{sensitive} = 2.6$ ,  $a_{highly \ sensitive} = 4.5$ ,  $b_{not \ sensitive} = 0.71$ ,  $b_{highly \ sensitive} = 0.8$ .

For score = 1 : CV = 0

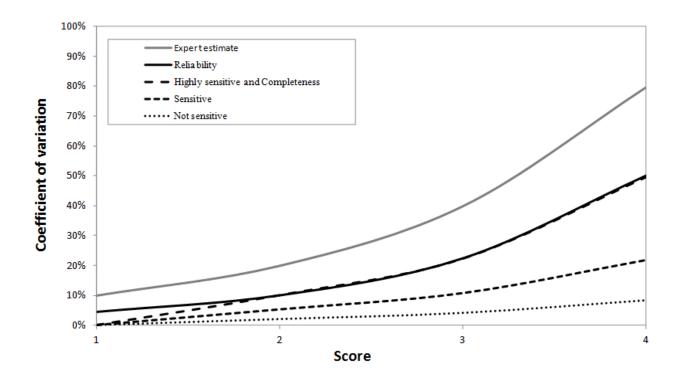
For score= ]1,4] : CV = a \* e<sup>(b \* (score-1))</sup>

The CVs for *Reliability* and *Expert Judgment* are determined using a slightly adapted function (Equation below), because the best score should not result in zero uncertainty. The parameter values used for *Reliability* are a = 0.02 and b = 0.805, whereas for *Expert Judgment* a = 0.05 and b = 0.692.

For score= ]1,4] : CV = a \* e<sup>b</sup> \* score-1

The resulting functions are depicted below in Figure C.1.

Figure C.1: Continuous functions that reflect the relationships between indicator scores and coefficients of variation.



tainty of the input data for the flows. Empty lines indicate that no input values were calculated	ted directly by the software STAN, according to the principle of mass conservation.
Table C.1: Relative uncertainty of the input	and that they were calculated directly by the

1.1         Deprind the annek;         25         35 <th>N N</th> <th>Flow N. Flow name</th> <th>1990</th> <th>1991</th> <th>1992</th> <th>1993</th> <th>1994</th> <th>1995</th> <th>1996 1</th> <th>1997 19</th> <th>1998 1999</th> <th>99 2000</th> <th>0 2001</th> <th>1 2002</th> <th>2 2003</th> <th>3 2004</th> <th>2005</th> <th>2006</th> <th>2007</th> <th>2008</th> <th>2009 2</th> <th>2010 2</th> <th>2011</th>	N N	Flow N. Flow name	1990	1991	1992	1993	1994	1995	1996 1	1997 19	1998 1999	99 2000	0 2001	1 2002	2 2003	3 2004	2005	2006	2007	2008	2009 2	2010 2	2011
Deportise match;         Zys         Zys <thzys< th="">         Zys         <thzys< th=""></thzys<></thzys<>	1	Import live animals	22%	20%	19%	18%	_	_	_			_						12%	12%	12%	12%	12%	12%
Memore relation         Ty         Ty <thty< th="">         Ty         Ty</thty<>	~	Export live animals	22%	20%	19%	18%	12%	_		_	_				_		12%	12%	12%	12%	12%	12%	12%
Annol         Annonnol         Annol         Annol		Manure applied to fields	27%	27%	27%	27%	27%				_						27%	27%	27%	27%	27%	27%	27%
Inderection:         Giale	4	Animal products																					
Amme u biggs:         Cits	6	Fallen stock	40%	41%	41%	42%	43%	44%	-		-	-	-	_			3996	3996	40%	40%	3996	40%	41%
Image         Total         Total <th< td=""><th>6</th><td>Manure to biogas</td><td>61%</td><td>61%</td><td>61%</td><td><b>35%</b></td><td>203</td><td>47%</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td>37%</td><td>37%</td><td>3636</td><td>35%</td><td>35%</td><td>37%</td><td>37%</td></th<>	6	Manure to biogas	61%	61%	61%	<b>35%</b>	203	47%									37%	37%	3636	35%	35%	37%	37%
Freedom         Bost	_	Atm. deposition agric.	24%	24%	24%	24%	24%	24%			-			-	-		27%	28%	30%	32%	32%	32%	32%
Metrication         338 <th< td=""><th>2</th><td>Erosion to forestry</td><td>80%</td><td>808</td><td>80%</td><td>80%</td><td>80%</td><td>80%</td><td></td><td></td><td></td><td></td><td>-</td><td>-</td><td></td><td></td><td>808</td><td>808</td><td>808</td><td>808</td><td>80%</td><td>80%</td><td>808</td></th<>	2	Erosion to forestry	80%	808	80%	80%	80%	80%					-	-			808	808	808	808	80%	80%	808
Approximation         Costs	<b>"</b>	Agricultural emissions	32%	32%	30%	28%	27%	-		-				-	-		23%	23%	24%	24%	-		26%
Corpor b bloges         Corpor bloges	4	Agricultural products	20%	20%	20%	20%	17%		-		-				$\vdash$		14%	14%	14%	14%	14%		14%
Nonmetheche/ered         138	۵.	Crops to biogas	22%	35%	55%	499%	43%	-	-	-	+	-	-	⊢	-	$\vdash$	35%	34%	339%	339%	-	-	35%
Corper behinder         12%		Non marketable feed	18%	18%	18%	18%	18%	18%	-	_	-			-			18%	18%	18%	18%	18%	18%	18%
Interfactional paperial containable and and paperial containable and paperi containable and paperial containable and paperial containable a	~	Crops to biofuels	12%	12%	12%	12%	12%	-	-	-			-	-			-	41%	41%	-	-	-	41%
Dependention         27%         7%         27%         2%		Import wood and paper	27%	27%	27%	26%	25%										20%	20%	21%	20%	20%	20%	20%
Ann deportient function         216		Export wood and paper	27%	27%	27%	26%	25%		_	_	-		_	_				25%	25%		-	25%	25%
New         Description         23% <th< td=""><th>_</th><td>Atm. deposition forestry</td><td>24%</td><td>24%</td><td>24%</td><td>24%</td><td>24%</td><td></td><td></td><td></td><td></td><td></td><td></td><td>_</td><td></td><td></td><td></td><td>28%</td><td>30%</td><td>32%</td><td>32%</td><td>32%</td><td>32%</td></th<>	_	Atm. deposition forestry	24%	24%	24%	24%	24%							_				28%	30%	32%	32%	32%	32%
icrostry emissions         23%	_	Pulp industry WW	23%	23%	23%	2396	20%											ŝ	10%	11%	12%		14%
Wood and paper to constantes         336         376         376         276         276         216         216         276		Forestry emissions	32%	32%	30%	28%	27%			_				_				23%	24%	24%	24%	25%	26%
Nood & paper to inductivy         30%         30%         30%         30%         30%         30%         30%         30%         30%         30%         30%         30%         30%         30%         30%         20% <th></th> <td>Wood and paper to consumers</td> <td>30%</td> <td>30%</td> <td>30%</td> <td>30%</td> <td>30%</td> <td></td> <td></td> <td></td> <td></td> <td></td> <td></td> <td></td> <td>_</td> <td></td> <td></td> <td>20%</td> <td>21%</td> <td></td> <td></td> <td>2096</td> <td>20%</td>		Wood and paper to consumers	30%	30%	30%	30%	30%								_			20%	21%			2096	20%
Image: begin		Wood & paper to industry	3006	30%	30%	30%	30%		_	_			_				20%	20%	21%	20%	20%	2096	20%
Import chemicals         Zes         Zis         Zis <thzis< th="">         Zis         <thzis< th=""></thzis<></thzis<>		Energy wood	29%	29%	29%	2996	28%	-	-	_	-	-	_	-	-	-	17%	18%	18%	-	17%	_	17%
Import min fertilizers and phocphate ore         2% <th>_</th> <td>Import chemicals</td> <td>25%</td> <td>22%</td> <td>21%</td> <td>1996</td> <td>18%</td> <td></td> <td></td> <td></td> <td></td> <td></td> <td></td> <td>_</td> <td></td> <td>_</td> <td>15%</td> <td>15%</td> <td>15%</td> <td>_</td> <td>15%</td> <td>_</td> <td>15%</td>	_	Import chemicals	25%	22%	21%	1996	18%							_		_	15%	15%	15%	_	15%	_	15%
Import food         23%         21%         20%         13% <th< td=""><th></th><td>Import min fertilizers and phosphate ore</td><td>22%</td><td>22<b>%</b></td><td>25%</td><td>25%</td><td>22% 22</td><td></td><td></td><td>_</td><td></td><td></td><td></td><td></td><td></td><td></td><td>× X</td><td>25%</td><td>25%</td><td>ж Х</td><td>2% 2%</td><td></td><td>25%</td></th<>		Import min fertilizers and phosphate ore	22%	22 <b>%</b>	25%	25%	22% 22			_							× X	25%	25%	ж Х	2% 2%		25%
Import feed         238 <th< td=""><th>_</th><td>Import food</td><td>23%</td><td>21%</td><td>20%</td><td>1996</td><td>13%</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td></td><td>11%</td><td>11%</td><td></td><td>10%</td><td></td><td>j0%</td></th<>	_	Import food	23%	21%	20%	1996	13%											11%	11%		10%		j0%
Seeds         ZMS         ZMS <thzms< th=""> <thzms< td="" th<=""><th>4</th><td>Import feed</td><td>23%</td><td>23%</td><td>23%</td><td>23%</td><td>24%</td><td>_</td><td>-</td><td>-</td><td>-</td><td>_</td><td>-</td><td>-</td><td>_</td><td>_</td><td>-</td><td>12%</td><td>12%</td><td>12%</td><td>12%</td><td>12%</td><td>12%</td></thzms<></thzms<>	4	Import feed	23%	23%	23%	23%	24%	_	-	-	-	_	-	-	_	_	-	12%	12%	12%	12%	12%	12%
Momenal fertilizers to agriculture         11%         <	5	Seeds	28%	27%	27%	27%	26%											26%	26%	26%	26%	_	26%
Image: Construction         Z3%         Z3% <thz3%< th="">         Z3%         <thz3%< th=""></thz3%<></thz3%<>		Mineral fertilizers to agriculture	11%	11%	11%	11%	11%	_	_	_	_			_				11%	11%	_		11%	11%
Report food         23%         21%         13% <th< td=""><th>~</th><td>Export feed</td><td>23%</td><td>23%</td><td>23%</td><td>23%</td><td>24%</td><td></td><td></td><td></td><td></td><td></td><td></td><td>_</td><td></td><td></td><td>12%</td><td>12%</td><td>12%</td><td>_</td><td></td><td>_</td><td>12%</td></th<>	~	Export feed	23%	23%	23%	23%	24%							_			12%	12%	12%	_		_	12%
Coport mineral fertilices         14% <th></th> <td>Export food</td> <td>23%</td> <td>21%</td> <td>20%</td> <td>19%</td> <td>13%</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>11%</td> <td>11%</td> <td>11%</td> <td>-</td> <td>-</td> <td>-</td> <td><b>1</b>0</td>		Export food	23%	21%	20%	19%	13%	-	-	-	-	-	-	-	-	-	11%	11%	11%	-	-	-	<b>1</b> 0
Coport chemicals         22%         20%         13%         17%         12%		Export mineral fertilizers	14%	14%	14%	14%	14%	_	-	_	-		-	_	_	_	14%	14%	14%	14%	14%	_	14%
Image: Interview         21%         22%         22%         23%	9	Export chemicals	22%	20%	18%	17%	15%	-		-	-		-	-		-	12%	12%	12%	-	-	12%	12%
Intermediate         Detergents         Z3%         Z3% <thz3%< th="">         Z3%         Z3%</thz3%<>	쾨	Municipal ind. WW	21%	22%	22%	22%	734K	-	-	-	-	-	-	-	-	_	16%	16%	17%	17%	-	-	20%
Mn. fertilizers to concumers         Z54	2	Detergents	23%	22%	22%	22%	21%	-	-	-	-	-	-	-	-	-	×	89%	8%	36	10%	-	13%
Per food         Edity	В	Min. fertilizers to consumers	25%	25%	25%	25%	25%	25%	-	_	-	-	_	-	-	_	15%	14%	13%	13%	12%	11%	12%
Food         20%         20%         20%         20%         1%         <	14	Pet food	64%	64%	64%	64%	64%	_	_		_		_	_	_		58%	57%	57%	_	57%	_	57%
Other industrial waste         36%         36%         36%         36%         36%         36%         26%         27%         26%	띩	Food	20%	20%	20%	20%	20%	_	-	-	_	-	_	-	-	_	16%	15%	15%	-	15%	15%	36
Vegetal inductrial waste         43%         42%         43%         42%         42%         42%         42%         42%         42%         42%         42%         42%         42%         42%         42%         43%         43%         33% <th>я</th> <td>Other industrial waste</td> <td>3696</td> <td>3698</td> <td>36%</td> <td>3698</td> <td>3698</td> <td>36%</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>-</td> <td>_</td> <td>26%</td> <td>26%</td> <td>26%</td> <td>26%</td> <td>-</td> <td>26%</td> <td>26%</td>	я	Other industrial waste	3696	3698	36%	3698	3698	36%	-	-	-	-	-	-	-	_	26%	26%	26%	26%	-	26%	26%
Animal inductival waste         69%         69%         64%         60%         64%         44%         43%         43%         41%         75%         31%         32%         33%	ы	Vegetal industrial waste	43%	42%	43%	43%	42%	44%	_	_	-	-	-	-	-	_	3698	3698	3696	36%	36%	3696	3698
Marterable feed         59%         57%         47%         43%         37%	18	Animal industrial weste	69%	69%	69%	6996	64%	60%	_	_	_	_	_	_	_	_	41%	41%	42%	42%	41%	42%	43%
In struind WW 10% 9% 8% 7% 4% 7% 8% 8% 9% 10% 12% 13% 13% 11% 10% 10%	อ	Marketable feed	28 26	23%	47%	43%	368 26	37%	_	_	_	_	_	-	_	_		19%	19%	۶,			31%
	8	In situ ind. WW	10%	ŝ	836	88	×	4%	_	_	_	-	_	_	_	_	<b>10%</b>	10%	10%	10%	11%	12%	12%

Flow	Flow N. Flow name	1990	1991	1992	1993	1994	1995	1996	1997 1	1998 19	1999 2000	00 2001	1 2002	2 2003	3 2004	2005	2006	2007	2008	2009	2010	2011
<b>B</b> 4	Biomass a shes to landscaping	9095	<b>%9</b> 5	<b>X9</b> 5		<b>X0</b> 2	44%												<b>X</b> 2X			27%
B2	Biomass ashes to fields	805	<b>X9</b> 5	<b>3</b> 65	-	¥05	44%	40%	-	-		-	-	-	-	-	-	27%	25%	27%	25%	27%
БЗ	Biomass ashes to green areas	9095	2695	36 <u>9</u> 5	395 76	<u>8</u>	44%	40%	37%	34% 3	32% 30%	202	⊢	6 27%	828	5 27%	25%	27%	25%	27%	25%	27%
F5.4																						
BS	Landfilled biomass ashes	52%	52%	52%	52%	44%	38%	339%	29%	25% 2	23% 20%	96 18%	K 17%	6 16%	K 12%	12%	12%	12%	12%	12%	12%	16%
B.6	<b>Biofuels by-products as feed</b>	15%	15%	15%	15%	15%	15%	15%	15%		15% 15%	96 15%	K 23%	6 52%	K 52%	52%	52%	52%	52%	52%	52%	52%
В.7	Import raw materials for biofuels	12%	12%	12%	12%	12%	12%	12%	12%	12% 1	12% 12%	% 12%	K 21%	60%	K 60%	808	60%	60%	60%	60%	60%	60%
H6.1	Municipal households WW	21%	22%	22%	22%	23%	23%	24%	25%	21% 2	20% 20%	96 1996	K 19%	9061	K 18%	316%	16%	17%	17%	18%	19%	20%
H6.2	Residual waste	339%	339%	339%	339%	339%	339%	27%	 7857	20% 20%	30% 34%	96 32%	K 31%	6 29%	K 228%	28%	25%	28%	28%	200	31%	32%
H6.3	Separate org. waste	27%	27%	27%	27%	27%	27%	27%	27%	27% 2	27% 27%	% 27%	20%	20%	K 27%	ж Х	27%	×	27%	76%	28%	<b>3</b> 87
H6.4	Green waste	52%	53%	52%	53%	54%	27%	27%		30%	31% 32%	96 3196	808	9667	K 27%	2000	308	20%	27%	27%	20%	20%
H6.5	Waste wood and paper	3698	3696	3698	3696	3696	36%	3698	3628	31% 2	29% 26%	% 26%	-	6 228%	K 26%	228%	26%	28%	26%	26%	28%	28%
H6.6	Fecal sludge to groundwater	51%	51%	20%	51%	51%	51%	51%	51%	51% 5	52% 52%	96 5396	K 54%	6 55%	K 55%	55%	55%	55%	55%	55%	35%	55%
H6.7	Fecal sludge to agriculture	51%	51%	98 98	51%	51%	51%	51%	51%	51%	52% 52%	36 23 23 23 23 23 23 23 24	828	6 55%	K 55%	55%	55%	25%	55%	55%	25%	35%
F7.1	Sewage sludge																					
F7.2	WW effluents																					
F13	Stormwater overflow	56%	<b>369</b> 5	49%	44%	30%	35%	3396	31%	29%	28% 27%	% 26%	K 27%	6 27%	K 27%	28%	29%	31%	33%	36%	3996	44%
F8.1	Compost to consumers	26%	24%	22%	21%	20%	19%	1996	1896	18% 1	18% 17%	36 18%	8 13%	8.1139%	K 19%	20%	21%	22%	24%	26%	28%	28%
F8.2	M&B meal to animal feed	3682	3236	30%	30%	3662	3682	28%	2896	28% 2	27% 27%	% 12%	K 12%	9571 9	K 12%	3621 3	12%	12%	%ZT	12%	12%	12%
F8.3	Substrate landscaping																					
F8.4	Recycled wood and paper																					
F8.5	Wastes recycled in agriculture																					
<b>F</b> 8.6	Export sewage shudge	59%	58%	51%	45%	40%	35%	34%	32%		29% 28%			6 25%	K 25%	5 24%	24%	24%	27%	27%	27%	29%
F8.7	Eqort M&B meal	268 268	89 59	ŝ	89 26	ŝ	45%	<b>40%</b>	37%	34%	32% 31%	% 30%	285 298	2836	K 26%	26%	26%	28%	ž	30%	31%	32%
F8.8	Export filter cakes																					
<b>F</b> 83	Export organic was te	83%	83%	83%	83%	83%	83%	83%	83%	83% 8	83% 83%	9K 839K	K 83%	83%	K 47%	5 47%	47%	%£8	83%	47%	83%	83%
F8.10	Waste to biogas plants																					
<b>F8.11</b>	Green waste to biomass plants	81%	81%	81%	81%	81%	81%	81%	81%	81% 8.	81% 81%	% 81%	<b>81%</b>	6 81%	K 81%	5 81%	81%	81%	81%	81%	81%	81%
F8.12	Import animal waste	60%	60%	60%	60%	54%	49%	45%	42%	40% 3	38% 37%	369K	K 35%	6 35%	K 33%	33%	33%	35%	35%	36%	37%	38%
<b>19</b> .1	Import water bodies	30%	30%	30%		30%	30%	30%		_	29% 29%	_		6 29%	_	20%	-	29%	29%	20%	29%	20%
<b>F</b> J.2	Export water bodies	23%	23%	23%	23%	23%	23%	23%				96 2396		6 23%		5 23%		23%	23%	23%	23%	23%
P11	Meat production	22%	20%	۶,	18%	29 20	15%	15%		-	15% 15%	-	-	6 14%	_	-	_	13% 1	13%	12%	12%	12%
P12	Eggs and milk production	13%	13%	13%	13%	13%	13%	13%	13% 261	-	3961 3961	9K 139K	-	a 13%6	K 12%	12%	12%	11%	11%	11%	11%	11%
P3.1	Timber	31%	31%	<b>%</b>	20%	70X	20%	20%	31%	31% 3	31% 29%	% 70%	2087 9	6 31%	K 3196	31%	31%	<b>X6</b> X	20%	20%	31%	31%
P4.1	Products to food production													_								
P4.2	Products to feed production																					
БЩ.3	By-products to animal feed production	61%	<b>35%</b>	95 20	46%	43%	41%	3996	3686	37% 3	36% 36%	9K 269K	K 26%	6 26%	K 26%	26%	26%	26%	26%	26%	26%	3698
P4.4	Food WW	2695	265 265	36 26	395 205	265 26	268	20%	X.	20 20 20 20	56% 56%	3695 7695	892	9 208	805 2008	208	25 26	36 36	20%	26 <u>8</u>	269K	200
P4.5	Animal feed WW	56%	2698	20%	20%	2695	56%	20%	2005	_	_	_	_	-	_	_	_	2695	56%	26%	2008	2698
P4.6	Fertilizer WW	80%	808	808	808	808	80%	80%	_	-	-	_	_	-	_	-	_	808	80%	80%	80%	80%
P4.7	Chemical WW	80%	808	80%	80%	80%	80%	80%	80%	80%	80% 80%	80%	K 80%	80%	K 80%	80%	808	80%	80%	80%	80%	80%
P4.8	Fertilizer industry in situ WW	40%	37%	35%	+	30%	28%	30%	-	-	-	-	+	+	-	+	+	43%	43%	43%	43%	43%
6M3	Chemical industry in situ WW	40%	37%	32%	32%	30%	<b>%</b>	ЖÖ	32%	35% 3	37% 40%	9K 439K	43%	4396	439%	43%	43%	43%	43%	43%	43%	43%

Flow N.	Flow N. Flow name	1990	1991	1992	1993	1994	1995	1996 1	1997 19	1998 1999	99 2000	0 2001	2002	2003	2004	2005	2006	2007	2008	2009 2	2010 2	2011
P6.1	Organic waste to home compositing	3696	34%	32%	31%	30%	30%	28%	30%	30% 28%	30%	30%	30%	30K	78 <b>%</b>	30%	<b>78%</b>	3006	30%	28%	3006	3006
P6.2	Garden vegetables	2096	<b>199</b>	19%	<b>3</b> 61	18%	18%	18%	18% 15	18% 18%	96 1896	1896	18%	18%	18%	18%	18%	18%	18%	18%	18%	18%
P6.3	Garden waste	32%	3696	35%	3696	37%	35%	368	36%	37% 38%	8 37%	3696	34%	33%	31%	32%	32%	31%	<b>%</b>	362	NOR NOR	30%
P6.4	Domestic animals excretions	808	808	80%	808	808	<b>%0%</b>			808 808			¥08	¥08	<b>%0</b> 8	80%	808	808	<b>%0</b> %	_	808	80%
P6.5	Wood & paper in residual was te	3995	3996	38%	38%	38%	37%	34%	32% 34	30% 28%	% 27%	27%	27%	27%	26%	27%	27%	27%	26%	26%	27%	<b>X</b> 42
T'Ld	Municipal WW to MWWTP	<b>3</b> 62	20%	18%	17%	16%	368	13%	13% 8	<b>%ET %8</b>	<b>%E 13%</b>		10%	11%	10%	30T	968	360T	30%	300%	8%	8%
P7.2	Municipal effluent	21%	18%	16%	14%	13%	<b>3</b> 6	13%	13% 8	89% 139%	<b>%</b> 13%		10%	<b>%</b> 11	10%	10%	868	10%	10%	10%	88	<b>%</b>
P7.3	Municipal semage sludge	23%	20%	18%	17%	16%	88	13%	13% 8	8% 13%	<b>%</b> 13%		10%	311%	10%	10%	8%	10%	10%	10%	8%	10%
P7.4	Industrial effluent	10%	366	8%	8%	ĸ	4%	×.	83%	866 968	X 10%	12%	13%	15%	17%	20%	3226	3672	323%	22%	. %77	22%
P7.5	Industrial sewage sludge	10%	<b>3</b> 6	8%	968	ĸ	4%	ž	8 %8	966 968	960T 9	9621 9	13%	15%	17%	3602	%ZZ	3622	%77	322%	: %77	322%
P8.1	Animal was te to Rendering	70%	7667	28%	78X	27%	27%		2776 2	27% 27	_				27%	27%	27%	27%	27%	27%	27%	27%
P8.2	Green waste to compost	40%	37%	34%	32%	31%	30%	2994		26% 26%		28%	29%	30%	31%	32%	34%	37%	40%	$\vdash$		2690
P8.3	Animal was te to biogas	3962	3995	39%	3996	<b>363</b>	37%	3656	329K	35% 34%	3436		33%	Neg Mage Mage Mage Mage Mage Mage Mage Ma	33%	33%	33%	33%	33%	33%	New Sector	33%
P8.4	HH res. waste to MBT	47%	47%	47%	47%	47%	474	40%	40%	39% 36%	37%	3696	3698	3698	34%	34%	40%	34%	34%	34%	New Year	3696
P8.5	HH res. waste to landfill	47%	47%	47%	47%	47%	47%	40%	40% 35	39% 36%	94 37%	3696	36%	3696	34%	34%	40%	34%	34%	34%	3294	36%
P8.6	HH res. waste to TT	47%	47%	47%	47%	47%	47%	40%	40%	39% 36%	97. 17. 17.	3696	3698	3698	34%	34%	40%	34%	34%	34%		3696
P8.7	SS to composting	31%	29%	28%	27%	27%	25%	27%	26% 24	24% 26%	% 26%	6 24%	25%	26%	26%	26%	25%	25%	25%	24%	24%	25%
P8.8	SS to landfil	24%	21%	20%	<b>3836</b>	17%	13%		17% 1	13% 16%	369T 363K	13%	14%	15%	14%	15%	<b>33%</b>	15%	14%	4%	4%	4%
P8.9	Science	24%	21%	20%	18%	17%	13%	17%	17% 1	15% 17%	% 17%	9,621 3,9%	14%	18%	17%	18%	16%	18%	18%	17%	17%	18%
P8.10	SS to co-incineration	308	28%	25%	22%	<b>36</b> 5	15%	 17	16% 1	12% 15%	<b>%</b> 14%	10%	10%	311%	10%	10%	8%	10%	12%	12%		13%
P8.11	SS applied in agriculture	24%	21%	20%	18%	17%	13%	17%	17% 1	13% 16%	<b>%</b> 16%	13%	14%	15%	14%	15%	13%	15%	14%	14%	13%	15%
P8.12	SS to landscaping	32%	29%	29%	29%	29%	29%	32%	34% E	35% 40%	96 45%	4996	26%	56%	2696	56%	55%	56%	56%	26%	25%	56%
P8.13	M&B meal as fertilizer	41%	41%	41%	41%	41%	41%	41%	41% 41	41% 41%	% 41%	52%	30%	30%	29%	28%	28%	28%	28%	28%	28%	28%
P8.14	M&B meal to fuel	41%	41%	41%	41%	41%	41%	41%	41% 43	41% 41%	% 22%	34%	34%	34%	27%	27%	27%	27%	27%	27%	27%	28%
P8.15	Stabilized waste to landfill	76%	76%	76%	71%	68%	65%	63%	_	60% 59%	<b>%</b> 59%	_	58%	_	57%	57%	58%	58%	59%	_	_	61%
P8.16	Compost to agriculture	26%	24%	22%	21%	20%	19%	19%	18% 18	18% 18%	% 17%	6 18%	18%	18%	19%	20%	21%	22%	24%	26%	28%	28%
71.8q	Compost to landscaping	26%	24%	22%	21%	20%	<b>19%</b>	19%	18% 18	3834 1894	% 17%	18%	18%	18%	<b>76</b> 1	20%	21%	%ZZ	24%	26%	28%	28%
P8.18	Stabilized waste to TT	58%	58%	58%	52%	47%	43%	40%	37% 3	35% 34%	<b>%</b> 33%	52%	32%	30%	30%	30%	32%	32%	33%	34%	3294	37%
P8.19	Ash co-inc. to landfill																					
P8.20	Ash/slag to landfill																					
P8.21	Loss in clinker																					
P8.22	MSS to fuel	32%	30%	29%	28%	27%	25%	27%	27% 22	_	_		18%	17%	16%	16%	14%	15%	14%	15%	14%	16%
P8.23	MSS to waste incineration	32%	30%	29%	78%	27%	22%		27%	23% 23%	<b>%</b>	18%	18%	ľ.	1 <b>69</b>	16 <b>%</b>	14%	15%	1 <b>4%</b>	15%	14%	169
P8.24	Filter cakes Inc.																					
P8.25	Filter cakes Co-inc.																					
P8.26	SS to MBT	<b>36</b> 5	283	57%	<b>8</b> 92	<b>45</b> %	39%	34	34%	30% 30%	28 28 8	_	27	2°K	28 <b>%</b>	24%	26%	27%	78%	NA NA	308	33%
P8.27	Animal was te to compost	<b>36</b> 8	3996	39%	39%	3836	37%	_	_	-	_	_	-	_	33%	33%	33%	33%	33%	_	_	33%
P8.28	Veg. ind. to biogas	24%	24%	24%	24%	24%	24%	24%	24% 24	24% 24%	<b>% 24%</b>	6 24%	24%	31%	42%	<b>29%</b>	59%	2096	29%	2005	20%	20%

of the input data for the stocks, stock change rates and transfer coe
indicate that no input values were calculated and that they were calculated directly by the software STAN, according to the
principle of mass conservation.

Stock N.	Stock N. Stock name	1990	1991	1992	1993 1	994 1	995 1	1 966	1 266	1991 1992 1993 1994 1995 1996 1997 1998 1999 2000 2001 2002 2003 2004 2005 2006 2007 2008 2009 2010 2011	99 200	00 200	1 200	2003	2004	2005	2006	2007	2008	2009	2010	2011
S1	Livestock	14.5%			-	-	-	-	-	-												
S2	Agricultural fields	51.0%																				
S3	Trees and soil	30.6%																				
S6-1	Stock in buildings and furniture																					
S6-2	Private gardens & public green areas	60.4%																				
S8-1	Clinker	30.9%																				
S8-2	Landfills	70.3%																				
S9	Water bodies																					

Stock N.	tock N. Stock change rate name	1990	1991	1992	1993	1994	1995 1	1 966	1 766	1993  1994  1995  1996  1997  1998  1999  2000  2001  2002  2003  2004  2005  2006  2007  2008  2010	99 20	00 200	11 200	200	3 2004	2005	2006	2007	2008	2009	2010	2011
S1	Livestock	3621	3621	15%	15%	3651		15%	15%	15% 1	3651 3651	361 15%	% TE%	%5T %	K 15%	965T	3621	72%	3621	<b>%</b> 5T	3621	15%
S2	Agricultural fields																					
S	Trees and soil																					
S6-1	Stock in buildings and furniture																					
S6-2	Private gardens & public green areas																					
S8-1	Clinker																					
S8-2	Landfills																					
<mark>S9</mark>	Water bodies																					

ier coefficient)	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	010	110
	366	366	968	968	¥69 ¥42	6	89	969	265	26	<b>%</b> S	<b>3</b> 65	4%	4%	4%	<b>%</b>	26	36	5%	969	869	8
	366	366	366 366	366	366	36	366	366	366	368	8%	ř	969	969	969	×.	<b>3</b> 45	<b>3</b> 5	26	4%	266	×.

Appendix D

## Degree of impact of the reconciliation on the whole system

The degree of impact of the reconciliation process on the whole system is calculated through the equation D.1,  $\overline{D}$  being the mean relative deviation of the reconciled from the entered values, m the total number of flows,  $x_n$  sthe entered value for flow n, and  $\widehat{x_n}$ the reconciled value for flow n. The normalized result is displayed in Figure D.1.

$$\bar{D} = \frac{\sum_{n=1}^{m} |\frac{x_n - \hat{x_n}|}{x_n}| \cdot 100}{m}$$
(D.1)

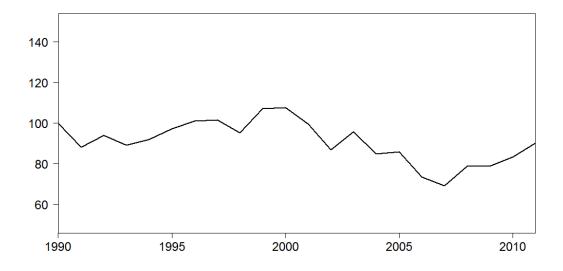


Figure D.1: Degree of the impact of the reconciliation on the whole system from 1990 to 2011, normalized with respect to 1990.

Appendix E

Absolute values and rationales for the calculation of the scope for improvement of different fields of action to enhance P governance in Austria

Values indicate the estimated improvement	
Table E.1: Relative impact of the fields of action on the <i>Import dependency</i> indicator.	with respect to the reference year 2013 (18,600 tP $y^{-1}$ ; 2.2 kgP cap <sup>-1</sup> $y^{-1}$ ).

Field of action	Max. improvement [tP y <sup>-1</sup> ]	Max. improvement [kgP cap <sup>-1</sup> y <sup>-1</sup> ]	Rationale
Increase of P recycling from meat and bone meal	3,000	0.35	Direct agricultural application of all Cat.3 and full recycling from mono-incineration ashes of Cat.1 and Cat.2.
Increase of P recycling from sewage sludge	4,200	0.49	Max. P recovery and recycling of sludge not currently applied in agriculture or composted.
Increase of P recycling from compost	2,000	0.24	Agricultural usage of compost currently employed for landscaping and gardening.
Increase of P recycling from digestates	ı	ı	No scope for further improvement.
Increase of P recycling from biomass ashes	400	0.05	Agricultural usage of safe fraction of ashes.
Increase of P recycling from manure	1	ı	Potential exists, but impossible to quantify due to lack of information on status quo.
Improvement of municipal and industrial organic waste collection and management	460	0.05	Recycling of 50% of MSW and of whole industrial organic waste.
Achievement of a balanced and healthy diet	3,700	0.44	Reduction of net imports of food, feedstuff and other biomass.
Increase of the use efficiency in crop farming	1,500	0.18	Perfect balance achieved; reference is the average surplus of the period 2011-2013.
Optimization of P content in animal feed	3,700	0.43	20% reduction of P content in feedstuff assuming that it affects exclusively imports.
Reduction of P use in detergents	765	0.09	90% reduction of P currently contained in dishwasher detergents.
Reduction of P use in other industrial processes	I	ı	No data available.
Reduction of surplus accumulation in private and public green areas	2,000	0.24	Input currently causing surplus fully used to replace mineral fertilizers in agriculture.
Reduction of point discharges	ı	ı	No effect on net imports, unless combined with recycling from sewage sludge.
Reduction of erosion from agricultural soils	2,200	0.26	70% reduction of soil losses translates proportionally in less need for fertilizers.

Field of action	Max. improvement [tP y <sup>-1</sup> ]	Max. improvement [kgP cap <sup>-1</sup> y <sup>-1</sup> ]	Rationale
Increase of P recycling from meat and bone	3 000	0.35	Direct agricultural application of all Cat.3 and full recycling
meal	0,000	0.00	from mono-incineration ashes of Cat.1 and Cat.2.
	006 4	070	Max. P recovery and recycling of sludge not currently
Increase of P recycling from sewage sludge	4,200	0.49	applied in agriculture or composted.
	000 6	760	Agricultural usage of compost currently employed for
increase of r recycling from composi-	2,000	0.24	landscaping and gardening.
Increase of P recycling from digestates	I	I	No scope for further improvement.
Increase of P recycling from biomass ashes	400	0.05	Agricultural usage of safe fraction of ashes.
			Potential exists, but impossible to quantify due to lack of
Increase of P recycling from manure	I	I	information on status quo.
Improvement of municipal and industrial	160	р 100	Recycling of $50\%$ of MSW and of whole industrial organic
organic waste collection and management	400	CU.U	waste.
			Reduction of net imports of food, feedstuff and other
Achievement of a balanced and nearing diet	I	I	biomass. No effect on the consumption of mineral fertilizers.
	1 600	0.1.0	Perfect balance achieved; reference is the average surplus of
increase of the use enciency in crop farming	1,300	01.0	the period 2011-2013.
			No visible effect on use of fertilizers due to current surplus
Optimization of F content in annual reed	1	I	levels.
Reduction of P use in detergents	1	I	No effect on the consumption of mineral fertilizers.
Reduction of P use in other industrial processes	I	I	No data available.
Reduction of surplus accumulation in private	2,000	0.24	Input currently causing surplus fully used to replace mineral
and public green areas			tertilizers in agriculture.
Roduction of noint discharmes	,		No effect on the consumption of mineral fertilizers, unless
Iveruction of point discretizinges	I	I	combined with recycling from sewage sludge.
Reduction of erosion from earioultured coils		0.96 0	70% reduction of soil losses translates proportionally in less

### E. Comparative assessment of fields of action

Field of action	Max. improvement [tP $y^{-1}$ ]	Max. improvement [kgP cap <sup>-1</sup> y <sup>-1</sup> ]	Rationale
Increase of P recycling from meat and bone meal			No effect on the emissions to water bodies.
Increase of P recycling from sewage sludge			No effect on the emissions to water bodies.
Increase of P recycling from compost	1		No effect on the emissions to water bodies.
Increase of P recycling from digestates	I	I	No effect on the emissions to water bodies at national level, but local relevance for vulnerable streams.
Increase of P recycling from biomass ashes	I		No effect on the emissions to water bodies.
Increase of P recycling from manure	I	I	No effect on the emissions to water bodies at national level, but local relevance for vulnerable streams.
Improvement of municipal and industrial organic waste collection and management	I	1	No effect on the emissions to water bodies.
Achievement of a balanced and healthy diet	250	0.03	Reduction of diffuse emissions.
Increase of the use efficiency in crop farming	I	1	No effect on the emissions to water bodies at national level, but local relevance for vulnerable streams.
Optimization of P content in animal feed	I		No quantifiable effect at national level, but local relevance for vulnerable streams.
Reduction of P use in detergents	80	0.01	90% reduction of P currently contained in dishwasher detergents and 90% removal rate from wastewater.
Reduction of P use in other industrial processes	I	I	No data available.
Reduction of surplus accumulation in private and public green areas	I	1	No effect on the emissions to water bodies at national level, but local relevance for vulnerable streams.
Reduction of point discharges	440	0.05	Raise of the national average of the removal rate performance in municipal and industrial WWTPs to the present highest level (96%).
Reduction of erosion from agricultural soils	580	0.07	Erosion on all arable land reduced by $70\%$ .

 ${\bf E.}\ Comparative\ assessment\ of\ fields\ of\ action$ 

# Appendix F Authorship

Chapter 2 of this Thesis is based on the publication "Added values of Material Flow Analysis. The Austrian phosphorus budget from 1990 to 2011" by Ottavia Zoboli, David Laner, Matthias Zessner and Helmut Rechberger. Journal of Industrial Ecology (2015). The contribution of Ottavia Zoboli to this paper was:

- Literature review
- Concept development
- Data mining
- Creation of the multiyear MFA
- Analysis and evaluation of the results
- Elaboration of graphs and figures
- Paper writing

Chapter 3 of this Thesis is based on the publication "Impact of reduced anthropogenic emissions and century flood on the phosphorus stock, concentrations and loads in the Upper Danube" by Ottavia Zoboli, Alberto Viglione, Helmut Rechberger and Matthias Zessner. Science of the Total Environment (2015) Vol. 518-519, pp. 117-129. The contribution of Ottavia Zoboli to this paper was:

- Literature review
- Concept development
- Data mining
- Statistical analysis and evaluation of the results
- Elaboration of graphs and figures

• Paper writing

Chapter 4 of this Thesis is based on the publication "Enhancing phosphorus governance in Austria: potential, priorities and limitations" by Ottavia Zoboli, Matthias Zessner and Helmut Rechberger. Science of the Total Environment (submitted). The contribution of Ottavia Zoboli to this paper was:

- Literature review
- Concept development
- Data mining
- Analysis and evaluation of the results
- Design and interpretation of the target system
- Elaboration of graphs and figures
- Paper writing