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Filling two needs with one deed: Potentials to simultaneously improve phosphorus and nitrogen management in Austria as an example for coupled resource management systems



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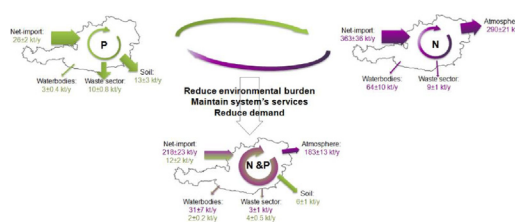
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HIGHLIGHTS

- Material flows of several substances can be simultaneously analyzed in a complex system.
- Coupled Material Flow Analyses reveal co-benefits and trade-offs between substances.
- The Austrian phosphorus and nitrogen systems are closely interrelated.
- Highest efficiency gains can be achieved by a combination of different measures.
- Potentials to increase resource efficiency are higher than for emission reduction.

GRAPHICAL ABSTRACT



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The tremendous increase in resource consumption over the past century and the environmental challenges it entails has spurred discussions for a shift from a linear to a circular resource use. However, to date most resource studies are restricted to one material or a single sector or process. In this work, a coupled material flow analysis taking the national phosphorus (P) and nitrogen (N) system of Austria as an example for two closely connected resource systems is conducted. Effects of different measures aimed at reducing P and/or N-demand, increasing recycling or reducing emissions to air and water are compared to a reference state (representing the actual situation in 2015). Changes in the mineral fertilizer demand of the system, P and N losses in the waste sector, water emissions of P and N, P soil accumulation and atmospheric N emissions are analyzed. Overall positive feedbacks between measures and between different goals of one measure always outweigh negative ones, which is why the highest efficiency gains (57±4%) can be achieved by a combination of all the 16 measures studied. Potentials for the reduction of mineral fertilizer demand are larger than for emission reduction though, confirming the past priority of environmental protection over resource protection. Although coupling significantly raises model complexity it can be shown that material flows of more than one substance can be simultaneously analyzed in a rather complex system. This may reveal interrelations, co-benefits and trade-offs between different resources that might have been omitted in a mono-substance analysis and thus improve judgment of sustainability and viability of different management strategies.

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1. Introduction

The transition of the world from an agricultural to an industrial society over the past century, which brought about a quadrupling of global population and a 5-fold increase in world GDP/cap, came at the expense of an explosion of global resource use. In 2005 around 60 Gt/yr of materials were extracted and used worldwide, an increase by a factor of eight compared to the beginning of the 20th century. This is linked to a number of problems, such as pressures on and damages to the environment, resource scarcity and distributional conflicts, and therefore poses a serious threat to global sustainability (Krausmann et al., 2009). Moreover, while the world has made progress on mitigating some of the other negative side-effects of industrialization, such as air pollution and acid rain caused by sulfur emissions (Stern, 2005) or the depletion of the Antarctic ozone layer (Hand, 2016), material consumption shows no signs of stabilization or reduction; to the contrary, growth rates during the first decade of the 21st century have been especially pronounced (Schaffartzik et al., 2014).

Among the resources that are of greatest concern in the future is phosphorus (P). Both phosphate rock and phosphorus are among the 27 critical raw materials listed by the EU, substances considered of high importance to the economy and of high risk associated with their supply (Bureau de Recherches Géologiques et Minières et al., 2017). The widespread application of industrial phosphorus fertilizer was crucial to the augmentation of agricultural yields during the green revolution after World War II (Borlaug, 1970) and continues to be an important factor in ensuring global food security. However, phosphate rock is a finite resource. Regardless of ongoing discussion about future availability and a potential peak in P production (e.g. Cordell and White, 2011; Scholz and Wellmer, 2013; Edixhoven et al., 2014), as reserves with pure and easily available P are depleted, extraction becomes more costly and quality of the mined product is declining. Furthermore, P that is currently economically feasible to extract is concentrated in a few countries only, with Morocco accounting for 80% of the known reserves (Jasinski, 2016). Most of the reserves are located in geopolitically unstable regions and some countries restrict extraction for strategic reasons, giving rise to strong price fluctuations on the global P markets (Cordell and White, 2015; Ridder et al., 2012). Despite these challenges, material flow analyses (MFA) such as conducted by Egle et al. (2014), Cooper and Carliell-Marquet (2013) or Senthilkumar et al. (2012) reveal that phosphorus management to date mainly follows a linear approach with high imports for fertilization purposes, the majority of which ends up in landfills, as waste export or is emitted to water bodies, where it can cause eutrophication.

However, scientific, political and industrial interest in recycling phosphorus from waste and wastewater and thus reducing import dependency has been rapidly growing over the past years. Several studies exploring improvement potentials in P management have been conducted (e.g. Hamilton et al., 2017; Klinglmair et al., 2017; Zoboli et al., 2016) and efforts to harmonize national MFAs to facilitate systematic comparison and transfer of lessons learned have been made (e.g. Jedelhauser and Binder, 2015; van Dijk et al., 2016). Policies like the German sewage sludge ordinance (Bundesregierung Deutschland, 2017) or the proposal for the EU Fertilizer Regulation revision (European Commission, 2016) set legal prerequisites to spur recovery and recycling and respective technologies have been developed (Egle et al., 2016). As efforts move from a theoretical to a more and more implementational stage, economic and environmental impacts of different measures and technologies are moving into focus as well (e.g. Egle et al., 2016; Ernst Basler + Partner AG, 2017; Jossa and Remy, 2015; Hanserud et al., 2017). Most of these analyses still view phosphorus management from a single-substance perspective though.

Meanwhile, an even more imminent problem in the near future may be the disruption of the global nitrogen (N) cycle. Unlike P,

ammonia (NH₃) fertilizer can be industrially produced from N₂ and H₂ in the Haber-Bosch process. However, this process is very energy-intensive; the production of N fertilizers accounts for 1.1% of global energy use and 0.93% of global greenhouse gas emissions (Dawson and Hilton, 2011; IFA, 2009). Human activities now convert more atmospheric nitrogen into reactive forms than all of the Earth's terrestrial processes combined and four times as much as is estimated to be tolerable for keeping the Earth's system in a stable environmental state (Rockström et al., 2009). Only a fraction of this effectively acts as a plant nutrient though; the remainder is lost to the environment, where it contributes to problems like air pollution (in the form of NO_x), stratospheric ozone depletion (as N₂O), terrestrial and aquatic acidification (NH_x, NO₃), eutrophication of ecosystems (NH_x, NO₃), groundwater pollution (NO₃) and climate change (NO_x, N₂O) (Galloway et al., 2002).

Given the close connection of the phosphorus and nitrogen cycle Rockström et al. (2009) consider them as a single domain in their planetary boundary concept. It therefore seems reasonable to also address inefficiencies in their management in a simultaneous way. However, MFAs of both P and N focus more on the flow patterns of each substance individually than on their interactions, irrespective of whether the analysis is conducted on a sectoral (e.g. Antikainen et al., 2005; Ma et al., 2013; Thaler et al., 2015) or regional (Coppens et al., 2016) basis. Building on existing work by Zoboli et al. (2016) who analyzed the improvement potentials of phosphorus management in Austria based on a detailed national MFA, here, the effects of measures in P management on the N system and vice versa will be studied. This should not only reveal co-benefits and conflicting goals between the two substances, but also, from a methodological viewpoint, foster understanding of whether and how a coupled, multi-substance material flow analysis can generate more robust and meaningful results compared to a mono-substance one. In this respect the coupled MFA of P and N should serve as a case study for coupled resource systems in general.

2. Materials and methods

2.1. Status quo model

Measures to improve Austrian P- and/or N-management were evaluated against the actual situation in 2015 ("status quo model"). To model this reference state, a coupled MFA for P and N, following the methodology described by Brunner and Rechberger (2017) and building on the Austrian MFA for P (Zoboli et al., 2015, 2016), was conducted. The freeware STAN was used (Cencic and Rechberger, 2012) to calculate mass balances for each process in the system, perform error propagation of initial data uncertainties and data reconciliation. The later is based on least squares regression and alters a priori input data so that initial contradictions in the mass balance are eliminated. The a priori uncertainty thereby serves as a weighting factor that determines the extent of adjustment for each data element.

Nine main sectors/processes, relevant for the national P and N management are depicted: Animal husbandry, crop farming, forestry, industry and trade, bioenergy, households and public establishments, wastewater management and waste management. Each of these processes is further described by one or more subsystems, to enable e.g. distinction between food-, timber- and chemical industries or between consumption and soil processes in private households and to show processes such as manure generation and handling in more detail. Exchanges of N and P between these processes in gaseous, liquid and solid form as well as import and exports across country borders are represented by flows of P and N in t/year. In the majority of cases data is present as mass flows of a good (a substances or mixtures of substances with an economic market value Brunner and Rechberger, 2017) reported in national statistic databases or governmental reports and their respective P and N concentrations (mostly

found in scientific literature). However, especially for gaseous and liquid flows, data is sometimes also directly reported as masses of N and P per time. In addition, transfer coefficients and relations are used which link data with linear equations. The rough concept of the model can be described as follows: P and N are imported into the system as fertilizer, food and feed and are distributed (partly after refinement) by the industry and sector among agriculture and consumers. Animal husbandry uses feed from industry and pastures to produce meat and dairy products; in addition manure is generated. Crop farming consumes mineral and organic fertilizers for food production, which (again via the industry and trade sector) is transmitted to consumers or exported. Waste and wastewater from all sectors are collected in the wastewater and waste management systems respectively. Wastewater is treated in wastewater treatment plants and effluents discharged to water bodies, while emissions from agricultural and natural soils reach these water bodies directly. Depending on its composition, waste is either incinerated, deposited in landfills or treated (e.g. by composting) to generate a recyclable product.

Although model structure, flow calculation and data sources largely correspond to the ones described in Zoboli et al. (2015), the extension of the model to nitrogen and the refined consideration of data uncertainty (see Section 2.4) required several changes. For instance, processes and flows to account for gaseous nitrogen fluxes during incineration processes, manure handling and microbial nitrogen fixation in soil, which were not relevant to the national P balance had to be added in the coupled model. Assumptions for the handling of organic waste from private households were revised according to the findings of Maier (2017) and differences in plant nutrient availability of different fertilizers included in the process “Crop farming” (see Table 1). The new stock “planetary boundary layer” was introduced, representing exchanges between the atmosphere and other sectors of the model. However, as the focus of the study is on the interaction between N and P, sectors and processes exclusively relevant to the N-system, such as combustion of fossil fuels for power generation and in vehicles, were left aside. Similarly, cross-border exchange of gases was not considered in the balance, due to high uncertainties in its quantification and minor relevance to the issue of this study. Therefore, the inputs to and outputs of the stock “planetary boundary layer” are incomplete, which is why it should not be considered part of the balanced system. Furthermore, a number of fluxes considered in an aggregated form by Zoboli et al. (2016), where split into their components in the present model. For instance, the flow “Crops” was split in a subsystem into 50 individual flows (wheat, potato, carrots, apples, etc.). This made it possible to enter P- and N-concentrations of the various flows into the model in the form they existed in the data sources without requiring intermediate calculation steps and thus reduced distortion during data reconciliation (see Section 2.4).

Overall, the changes described above significantly increased the complexity of the model. Total number of processes rose from 56 to 194, of stocks from eight to 15, of flows from 122 to 866 and

of processes with transfer coefficients from two to 55. Fig. 1 shows the structure of the adjusted system. Each of the processes depicted in the figure consists of one or more sub-systems, the structure of which is shown in the supplementary material S1.

The coupling of the phosphorus and nitrogen balance was achieved by introducing a goods-layer into the system, which represents the total mass of a stock or flow. Where applicable, a priori data (input data of the model prior to calculation) thus consists of the mass of a flow and the respective concentrations of P and N. For some processes, especially those involving flows of air and/or water, keeping mass balance on the goods layer would have required extensions of the model that were beyond the scope of the present work, whereas data on the mass flows of N and P was readily available. For these flows masses of N and P were therefore directly used as a priori data. In addition, depending on the data source, mass is alternatively stated as dry- or fresh matter. The goods layer should therefore only be regarded as an auxiliary layer and is partly excluded from the balancing process. (For 59% of the flows and 47% of the stocks and stock changes a priori data is available on the goods layer, for flows this number rises to 84% after calculation.) Linking the P and N layer takes place during data reconciliation, when masses and concentrations of flows are adjusted so that the mass balance for all processes is kept on both substance layers.

2.2. Efficiency improvement measures

On basis of management strategies put forward in Shibata et al. (2017), Withers et al. (2015) and Zoboli et al. (2016) 16 individual measures to make the Austrian P and N balance more efficient were identified. According to their main aim they can be grouped into measures directed at the increase of nutrient recovery and recycling, the reduction of demand and consumption and the reduction of emissions to the environment. This classification does not mean though that measures cannot fulfill more than one of these aims simultaneously.

It should be noted that the focus of this study was on the description and functioning of the system as a whole rather than on the development of readily applicable management strategy. Therefore the measures presented should neither be regarded as exhaustive nor are they devised in full detail.

Measures aimed at increasing nutrient recovery and recycling:

- Increased recycling of food industry waste:

In this measure it was assumed that food industry waste which is currently exported but would be suitable for composting (i.e. former foodstuff of animal origin, kitchen and food waste BMLFUW, 2017) is composted. The surplus of compost produced was assumed to be applied as agricultural fertilizer and substitute for mineral fertilizer.
- Increased recycling of biomass ashes:

Fine fly ashes from biomass plants contain high amounts of heavy metals, which impedes their use in agriculture and composting. Coarse ashes on the other hand could be largely used as P-fertilizer (Oberberger and Supanic, 2009). In this measure it was thus assumed that all coarse ashes from biomass plants are recycled, while the amounts of ashes applied in forestry and as additive in composting remain constant to the status quo and the rest is used in agriculture, where it substitutes for mineral fertilizer.
- Improved collection of organic household waste:

As centralized composting facilities are generally more effective and private gardens and public green areas are marked by a high nutrient surplus (Maier, 2017; Zoboli et al., 2016), home composting was fully replaced by separate collection of organic kitchen and garden waste in this measure. In addition, increased separation of organic material currently disposed of

Table 1

P and N plant availability of different fertilizer products as applied in the model. Values are based on Hamilton et al. (2017) and Syers et al. (2010) for P and BMLFUW (2006) and Gutser et al. (2005) for N.

	Plant available P [t/t P input]	Plant available N [t/t N input]
Mineral fertilizer	0.70	0.85
Manure	0.67	0.59
Sewage sludge	0.51	0.47
Compost	0.36	0.34
Meat and bone meal	0.28	0.68
Biomass ashes	0.46	0.57
Biomass digestates	0.49	0.55

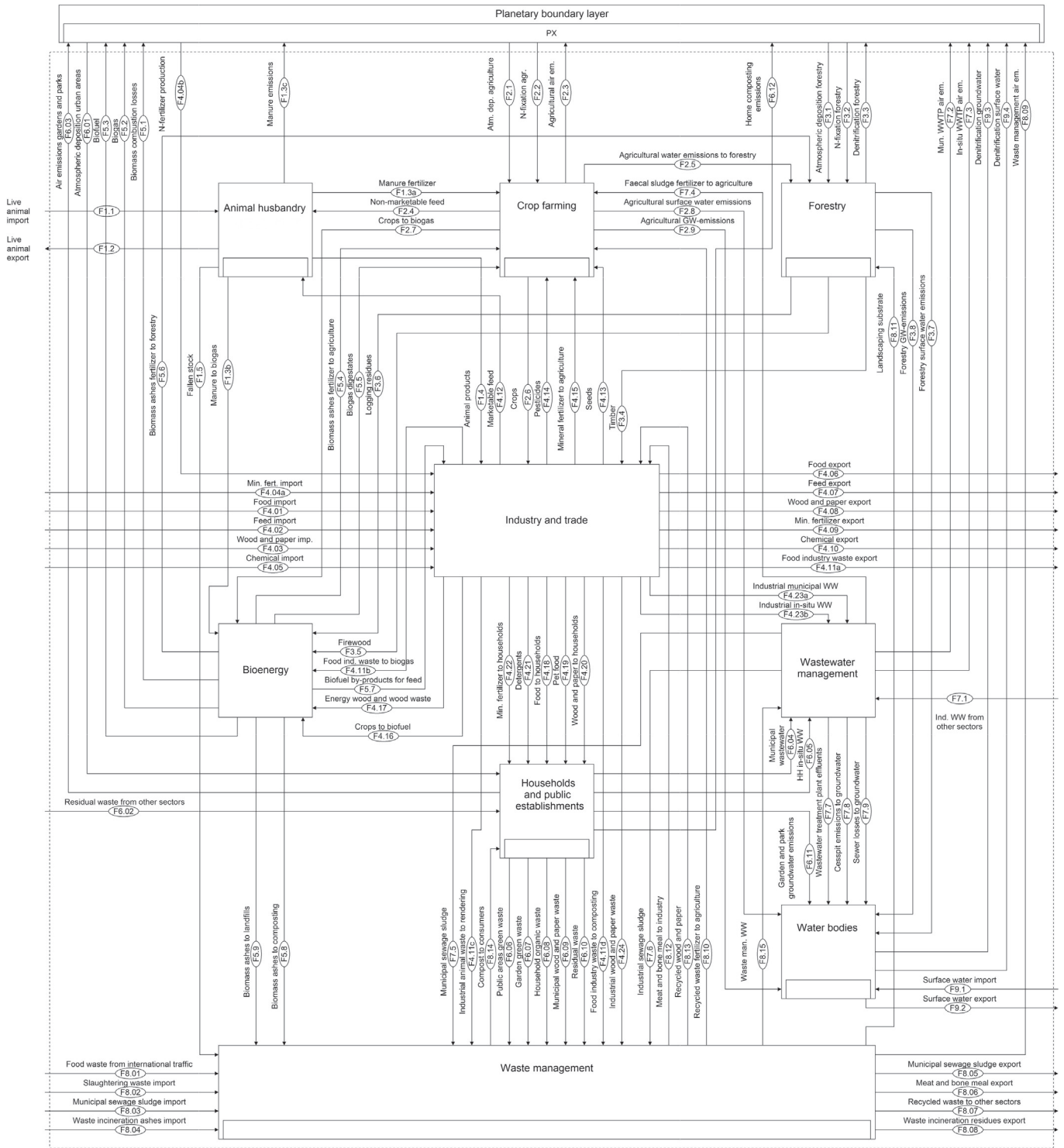


Fig. 1. Qualitative model of the Austrian P and N budget. Dashed line represents the system boundary, F-values in the ovals indicate the number assigned to each flow.

as residual waste was assumed. Implementing this measure would reduce N-inputs to private gardens and public green areas below plant requirements. The minimum additional amount of mineral fertilizer needed under the assumption that no N-losses to groundwater occur was therefore calculated. At the same time compost now available for agriculture substitutes for mineral fertilizer.

- Increased P-recovery from sewage sludge: In accordance with the national strategy for future sewage sludge management (BMLFUW, 2017) this measure anticipates a ban on composting and direct agricultural application of sewage sludge. Instead, it was assumed that all sewage sludge accrued is mono-incinerated and P-fertilizer recovered from the ashes. For P-recovery, LEACHPHOS® technology was

studied as an example due to its high recovery potential (70–80% of the ash input) and high plant availability of recovered P in the final product (Egle et al., 2016). LEACHPHOS® fertilizer is presumed to substitute for mineral P fertilizer.

- Increased nutrient recovery from meat and bone meal:

This measure requires separate collection of low risk (C3) and higher risk (C1 and C2) rendering material according to EU classification (European Commission, 2011, 2009). It was assumed that C3 material is directly used as fertilizer, except for a part equal to the amount in the status quo applied for feed production and other industrial purposes and that C1 and C2 material is mono-incinerated and P-fertilizer recovered from the ashes. For the later similar recovery potentials and P-plant availability as for P-recovery from sewage sludge ashes with LEACHPHOS® technology were assumed. It is again presumed that the recovered fertilizer substitutes for mineral P-fertilizer.

- Efficient use of compost:

In order to effectively make use of nutrients contained in it, all compost produced was assumed to be applied as fertilizer in this measure, while only less valuable material is used as landscaping substrate. Amounts going to private households and public institutions remain unchanged compared to the status quo so that all former landscaping substrate is used as substitute for mineral fertilizer in agriculture.

- Increased recycling of green waste:

As ashes from green waste co-incinerated with residual waste cannot be used as fertilizer due to contamination, in this measure green waste is entirely incinerated in biomass plants. It was assumed that the surplus compared to the status quo of thus created biomass ashes is applied in agriculture as a substitute for mineral fertilizer.

Measures aimed at reducing demand and consumption:

- Full application of P- and N-optimized feed for cattle, pig and poultry:

Following the Austrian guidelines of appropriate fertilizing (BMLFUW, 2006) optimized feeding practices in this measure involve a reduction of 20% of N and P content in feed for cattle and pigs and 26% for poultry. Several simplifications had to be made regarding this measure. First of all, due to lack of data, it was assumed that currently P- and N-optimized feeding plays a negligible role in animal husbandry. The effects of this measure may thus be overestimated. Furthermore, lower demand of P and N in feed were modeled as a mass reduction in production and import rates of all feed items equally, rather than as changes in concentrations or kind of feedstuff. Reduction of P and N content of feed are directly reflected in the nutrient content of manure, whereas P and N concentrations in meat and livestock stay constant.

- Reduction of P-concentration in detergents

Despite past reductions due to a ban of P in laundry cleaning products, Richards et al. (2015) still estimate a 90% reduction of detergent P load, if only non-P-based detergents for both laundry and dishwashing were used. This assumption was the basis of this measure and P concentrations of imported, exported and nationally used detergents reduced accordingly. Lower concentration of P in wastewater ultimately leads to lower P-input to agriculture with sewage sludge. As agricultural fields currently are generally over-fertilized, it was assumed that this reduction manifests itself in a reduction of water emissions and soil accumulation of P rather than in a higher demand for mineral fertilizer.

- Shift to a healthy, less meat-reliant diet:

Nutrition according to the recommendation of the Austrian Nutrition Report (Elmadfa et al., 2009) of the total population

was assumed in this measure. This corresponds to a reduction of meat consumption by two thirds and an increase in consumption of fruit, vegetables and carbon hydrates by a factor of 1.5. Percentage of food waste from industry and households was presumed to equal status quo. Changes in consumption of different food items are reflected in changes of their import and domestic production taking into account the respective ratio in the status quo. No changes in food export compared to the status quo were assumed. Changes in animal stock numbers, animal feed requirements, nutrient concentration in waste water, etc. arising from the assumptions described above, were considered as well. Reduction potentials for agricultural land demand, agricultural water emissions and fertilizer demand were taken from a study by Thaler et al. (2015), who take similar presumptions to the ones described above. It was assumed that land no longer used for agriculture is left to natural succession.

- Food waste reduction:

Reduction potentials of food waste from industry (ranging from 1% for meat to 45% for bakery products) and households (7% for separately collected waste and 57% for organics in residual waste) were based on estimations by Hietler and Pladerer (2017) and Pladerer et al. (2016). For changes in the system entailed by this waste reduction same assumptions as described in the measure “shift to a healthy, less meat-reliant diet” apply. However, in this case mineral fertilizer demand and agricultural emissions are not provided as a priori inputs, but calculated intrinsically in STAN.

Measures aimed at reducing environmental emissions:

- N-emission control during manure storage:

Covering liquid and solid manure during storage, for instance with peat, can reduce N emissions to the atmosphere by 80% (Rotz, 2004). Like for optimized animal feed it was assumed that this is not significantly practiced in the status quo. Lower storage losses lead to a higher N-content of manure applied to fields so that the demand for mineral N-fertilizer can be reduced.

- Exploitation of maximum fertilizer efficiency:

In this measure application losses of all types of fertilizers, including mineral fertilizers, manure, compost, etc. are reduced to a minimum and fertilizer inputs adjusted to actual plant needs. This can for instance be achieved by widespread application of the 4R Nutrient Stewardship principles – right fertilizer source at the right rate at the right time and in the right place (Reetz, 2016). Detailed analysis of individual actions especially with respect to nutrient legacy and interactions between plant, soil, water and nutrients were out of the scope of this study so that plant nutrient availability for all fertilizer products was merely shifted from average to upper boundaries of values found in literature (Hamilton et al., 2017; Syers et al., 2010; Gutser et al., 2005). An exception to this is manure: For P calculated plant nutrient availability in the status quo model after data reconciliation exceeds the values reported in literature, which is why no further increase was assumed. For N a maximum nutrient availability of 80% was applied (Ebertseder and Gutser, 2002). Fertilizer efficiency is, however, not only determined by application practice, but also depends on local factors such as soil and climatic conditions. Maximum fertilizer efficiencies are thus unlikely to be achievable for all regions and crops and effects of this measure probably overestimated. Plant availability of P in soil on the other hand tends to be underestimated in literature (Hanserud et al., 2016; Syers et al., 2010), so that potentials for mineral P fertilizer reduction may be underestimated. Savings in nutrient losses were assumed to reduce demand for mineral fertilizer.

- Agricultural erosion control:

Without looking at specific measures to implement this, it was assumed that erosion from agricultural land could be limited to a tolerable and often non-avoidable amount of 1 t/ha/year (Eurostat, 2015). Like the above, mineral fertilizer demand was reduced in line with the decreases in nutrient losses and further decrease of emissions following the reduced nutrient input taken into account.

- Increased P-removal efficiency of waste water treatment plants:

For this measure, it was estimated that P-removal efficiency of both municipal and industrial wastewater treatment plants could be raised to 96%. This corresponds to the efficiency currently reached in the Austrian Rhine river basin district (Überreiter et al., 2016). As for fertilizer efficiency, waste water treatment plant efficiency is dependent on local conditions. Implementing 96% P-removal may hence not be achievable on a national scale or at least require extensive structural changes. All P removed from the incoming wastewater is transferred to the sewage sludge. Once again, higher P-concentration of the part of sewage sludge used in agriculture enables substitution of mineral P-fertilizer.

- Increased N-removal efficiency of waste water treatment plants

The maximum achievable N-removal efficiency of municipal and industrial wastewater treatment plants was estimated with 90%, which is equivalent to the rate currently reached in plants with a size of 10,000–15,000 population equivalents (Überreiter et al., 2016). Contrary to P-removal, it is assumed that N transferred to sewage sludge remains equivalent to the status quo, while additionally removed N is emitted to the atmosphere. Considerations regarding the achievability of P-removal efficiency also apply for N.

In the agricultural sector differences in fertilizer efficiency were taken into account, whenever mineral fertilizer was substituted for by other products. With the exception of the measure “Exploitation of maximum fertilizer efficiency”, efficiency factors of each fertilizer were kept equivalent to the status quo, irrespective of changes in its mass, nutrient content or total nutrient input and output, which evidently constitutes a simplification of reality. Similarly, distribution of losses among soil, atmosphere, surface- and ground water was also kept equivalent to the status quo for all measures, except for “Agricultural erosion control”, where erosion control measures lead to a relative reduction of surface water emissions with respect to other losses.

For simplification, fertilizer efficiency was not taken into account in the forestry and household sectors. Instead changes in input and output of the processes “Forests and miscellaneous soils” and “Private gardens and public green areas” are reflected directly in a change of soil stock (P) and groundwater emissions (N). Especially for the measures “Improved collection of organic household waste”, “Efficient use of compost” and “Agricultural erosion control” that involve considerable reductions of P input into these processes, this simplification may lead to an overestimation of P water emissions in favor of lower soil accumulation of P.

In addition to individual measures a scenario, in which all of the 16 measures are combined, was analyzed as well. This made it possible to also consider feedback mechanisms between measures. For instance the measures “Full application of P- and N-optimized feed for cattle, pig and poultry” involves a decrease in field nutrient input with manure and thus may lead to higher requirements of mineral fertilizer, unless measures such as “Shift to a healthy, less meat-reliant diet” and “Food waste reduction”, that lead to a reduction of agricultural production and thus total fertilizer demand,

are implemented concomitantly. On the other hand applying the measure “Increased P-removal efficiency of waste water treatment plants” together with “Improved P-recovery from sewage sludge” would make both measures more efficient as the increased amount of P in sewage sludge due to better removal efficiency of waste water treatment plants is increasingly made available for agriculture by the recovery process.

Model structure for measure scenarios was essentially kept equivalent to the status quo; however, some measures required addition of new elements (like a process for the recovery of P from ashes for the measures “Improved P-recovery from sewage sludge” and “Improved nutrient recovery from meat and bone meal”) or redirection of flows (e.g. the flow “Green waste to incineration” from waste incineration plants to biomass plants in the measure “Increased recycling of green waste”). Moreover, due to differences in data sources, a priori input sometimes had to be inserted into the model as transfer coefficients rather than mass flows or vice versa. As for the status quo, these changes in the model structure are depicted in the supplementary material S1.

2.3. Evaluation indicators

Efficiency improvement potentials of each measure as well as of the combination of all measures with respect to the status quo were assessed using the following indicators:

- Demand of mineral P-fertilizer for domestic use
- Demand of mineral N-fertilizer for domestic use
- P accumulation in soil (including agricultural, urban as well as forested and natural soils)
- P losses in the waste sector (including accumulation in landfills, waste exports and P contained in cement and clinker after co-incineration of P containing waste as secondary fuel)
- N losses in the waste sector (including accumulation in landfills and waste exports)
- P emissions to ground- and surface water (including surface runoff and percolation from agricultural, urban as well as forested and natural soil and effluents from waste water treatment)
- N emissions to ground- and surface water (including surface runoff and percolation from agricultural, urban as well as forested and natural soil and effluents from waste water treatment; in order to avoid double accounting atmospheric N emissions from denitrification processes in ground- and surface water were subtracted)

Emissions occur almost exclusively in the form of NH₃; NH₄ emissions from waste water are negligible due to the well-developed secondary treatment stage (Überreiter et al., 2016).

- N emissions to atmosphere (including denitrification of agricultural, natural and urban soils, emissions during storage and spreading of fertilizer and emissions due to incineration, composting, rendering and waste water treatment processes)

No distinction between the different forms of N-emissions (N₂, NO_x, N₂O, NH₃) is made.

A detailed description of the flows used to compute each indicator can be found in the supplementary material S3. For each indicator x and measure y the efficiency improvement potential (EIP) was calculated as

$$EIP_{x,y} = 1 - \frac{\text{Indicatorvalue}_{x,y}}{\text{Indicatorvalue}_{x,\text{statusquo}}} \quad (1)$$

The total efficiency improvement potential (EIP_{tot}) of each measure was also stated as

$$EIP_{tot} = \frac{1}{8} * \sum_{y=1}^8 EIP_y \quad (2)$$

An EIP_{tot} of 1 thus signifies a situation, in which all losses and inefficiencies are completely eliminated; negative side-effects of measures exist, if indicator values < 0 occur.

2.4. Handling uncertainty

Initial uncertainty of data elements from literature was characterized using a method developed by Laner et al. (2015), combining qualitative data classification with exponential-type uncertainty characterization functions. As mentioned in Section 2.1, where possible, a priori input data to the STAN system was directly derived from the data sources and all necessary conversions from volume and concentration to mass and subsequent error propagation conducted within the software. However, in some cases, for instance, if concentrations provided in a data source referred to area rather than mass or volume, intermediate calculation steps were needed to create a priori input data. In these cases, Gaussian error propagation was used to compute a priori uncertainty. This is a main difference to the model of Zoboli et al. (2015), who equally applied the method of Laner et al. (2015) to define the uncertainty of masses and concentrations, irrespective of whether they were calculated or directly derived from literature. The new approach enables greater consistency in the initial qualitative data classification.

During data reconciliation in STAN values of uncertain data are altered in a way that contradictions in the balance disappear and uncertainty of the reconciled data is reduced (TU Wien, 2017). Great care was therefore paid to only use independent values as a priori input data (i.e. no a priori input value is used to calculate any other a priori input value), in order to avoid underestimation of final uncertainty after data reconciliation.

For the evaluation of the reconciliation process D-values specifying the degree by which a priori values are altered during data reconciliation with respect to a priori uncertainty are calculated as follows:

$$D = \frac{\sum \frac{|\text{reconciled value} - \text{a priori value}|}{\text{a priori uncertainty}}}{\text{No. of reconciled values}} \quad (3)$$

Measures are evaluated with respect to the status quo. Uncertainty of the data which changes with respect to the status quo is stated in the same way as described above. For flows, stocks, stock changes and transfer coefficients that are not affected by the measure on the other hand, reconciled values of the status quo are used as a priori input, assuming a very low uncertainty of 1% necessary to ensure model stability. In addition, only as much data from the status quo system was transferred to the measures scenario as is absolutely necessary for its calculation, keeping overdetermination to a minimum. The aim of both proceedings is to avoid altering of data elements that are not affected by the measure during reconciliation.

Uncertainty of evaluation indicators is derived with Gaussian error propagation from the uncertainty of model output data used to compute them. EIPs are considered significant, if their absolute value exceeds the uncertainty.

3. Results and discussion

3.1. The Austrian phosphorus and nitrogen system 2015

Modeling results confirm the close connection of the Austrian P- and N-systems. For both substances a linear management pattern

can be observed with high nutrient imports for food production and subsequent dissipation to various emissions and the waste sector (Figs. 2 & 3). However, there are differences in the predominant pathways of this dissipation: For P 50% of the net-import accumulates in agricultural and urban soils and another 37% is lost in the waste sector, whereas for N 80% of the net-imports are eventually emitted to the atmosphere (Fig. 4). A full list of all flows in the model and their respective quantities of N and P is provided in the supplementary material S2.

Table 2 gives an overview of the results of data reconciliation in STAN. The system has a low degree of overdetermination so that reconciliation was only possible for 40% of flow output data, 25% of TC output data and 84% and 86% of stock and stock delta output data respectively.

The average D-value (degree by which a priori values are altered during data reconciliation with respect to a priori uncertainty) of 4% could indicate high compatibility of a priori data from different sources; however, it may also be a result of the generally high uncertainty (34% on average for all a priori input data). Uncertainties of such magnitude are not uncommon in MFA: A priori mean uncertainty in the model of Zoboli et al. (2015) is 29.7%, while data reported by Cooper and Carliell-Marquet (2013) and Antikainen et al. (2005) on P flows in the UK- and P and N flows in the Finnish food production and consumption systems respectively both exhibit mean a priori uncertainties of 31%. Volatility of N during fertilization, manure management and waste water treatment, variability in soil leaching processes depending on soil type, slope and cultivation method as well as insufficient reporting of consumption and waste management processes were identified as the main sources of uncertainty, which matches the experiences gained in the present study. In particular increased tracking of flows of wastes and by-products, additional information on the amounts of fertilizer and compost consumed in private households and on nature and amounts of home composted waste, as well as improved methods to upscale soil leaching, erosion and N-emissions of a specific site or process to the country scale would be needed in order to reduce a priori uncertainty. The generation of such knowledge was, however, beyond the scope of the present study.

After calculation and data reconciliation, mean uncertainty of mass flows, stocks and stock changes of P and N rises to 125%. However, if uncertainty is scaled to flow mass, weighted mean reconciled uncertainty is reduced to 47% for P and 19% for N, which is again in line with the range commonly found in MFAs on P and N. The high mean uncertainty is hence caused by a small number of mass flows and stock changes with extraordinary high uncertainties, but small absolute values and was not deemed to restrict the overall outcome of the study. Furthermore, values, for which a priori input data was obtained from a difference of source data elements or which are computed in STAN from a difference of other flows in and out of the respective process, may exhibit uncertainties $> 100\%$ due to Gaussian error propagation. The uncertainty of a value C for which $C = A - B$ is calculated as:

$$\text{Uncertainty } C = \sqrt{\text{Uncertainty } A^2 + \text{Uncertainty } B^2} \quad (4)$$

Thus, if A and B have high uncertainties and their difference is small, the uncertainty of C will be larger than its value. This also explains why weighted mean reconciled uncertainty for P is considerably higher than for N: Soil stock deltas are calculated in STAN as the difference of input and output flows of the respective process and therefore disproportionately affected by the above-mentioned effect; moreover, they are relatively large in terms of mass. A main assumption of the N system is the absence of soil stocks and consequently uncertainties $> 100\%$ occur less often. Probabilistic MFA via

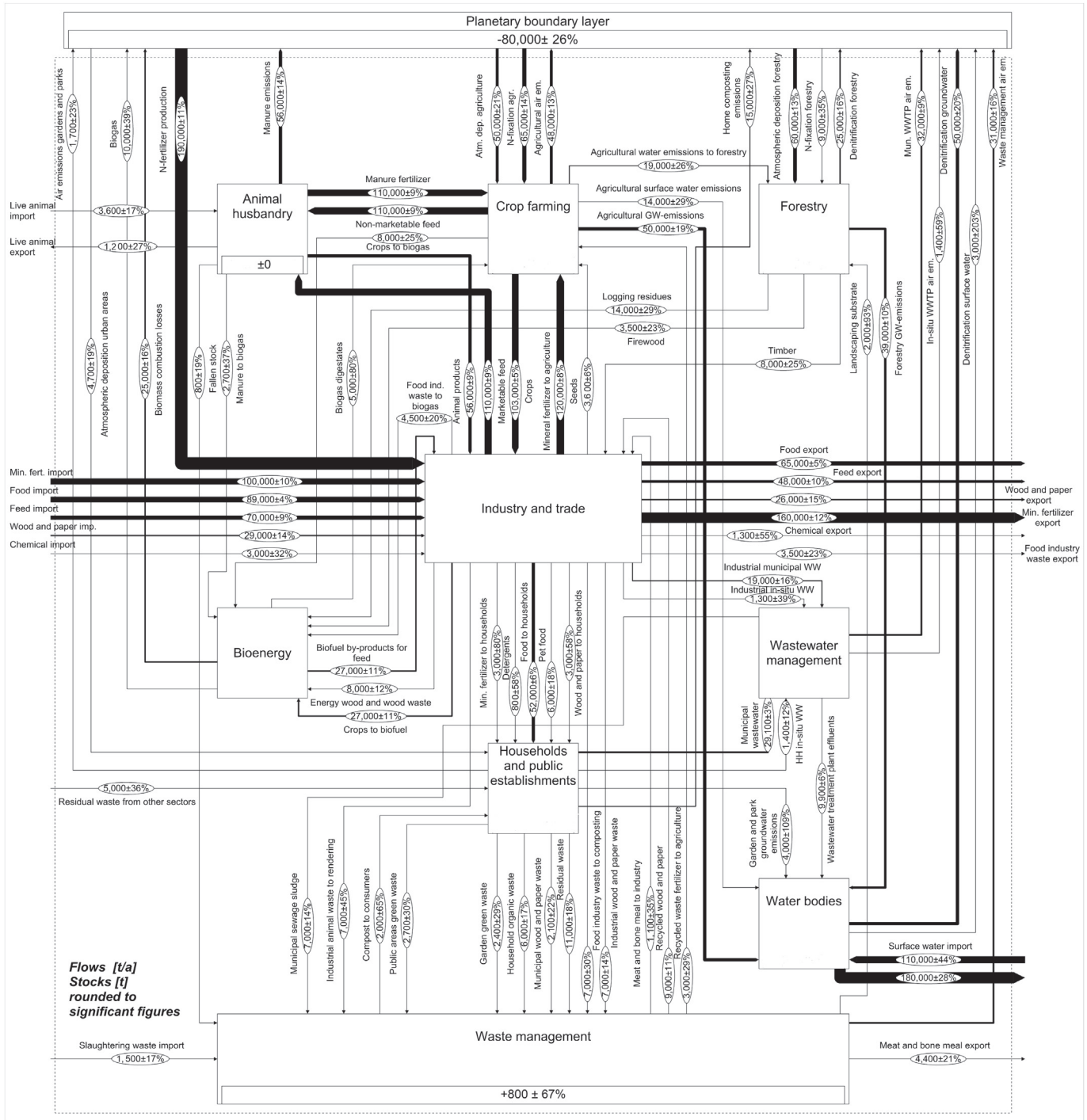


Fig. 3. Austrian nitrogen system for the reference year 2015. Only flows > 500 t N/year are shown.

indicator and in total. Positive feedbacks between measures hence clearly outweigh negative ones. Only water emissions of N may be equally well reduced by implementing the measure “Exploitation of maximum fertilizer efficiency” on its own as by the combination of measures, considering the respective uncertainty levels. (The improvement potentials for this measure may have been overestimated though, as mentioned in Section 2.2.)

Nevertheless, trade-offs between different goals within one measure exist. For example in the measure “Increased P-recovery from sewage sludge” sewage sludge that was formerly directly applied to agricultural land or composted is incinerated. The N present in the sewage sludge is lost to the air in the process, causing thus both an

increase in air emissions of N and in mineral N-fertilizer demand. The same is true for the measure “Increased nutrient recovery from meat and bone meal”; however, here the lack of N in meat and bone meal ashes is compensated by increased application rates of meat and bone meal that was formerly exported. As the fertilizer efficiency of meat and bone meal is lower than of mineral fertilizer, this measure is accompanied by higher losses of both P and N to water bodies and a higher soil P accumulation.

The highest negative impacts of this analysis occur for the mineral P-fertilizer demand in the measure “Full application of P- and N-optimized feed for cattle, pig and poultry”, where an increase of 11% compared to the status quo would be needed to compensate

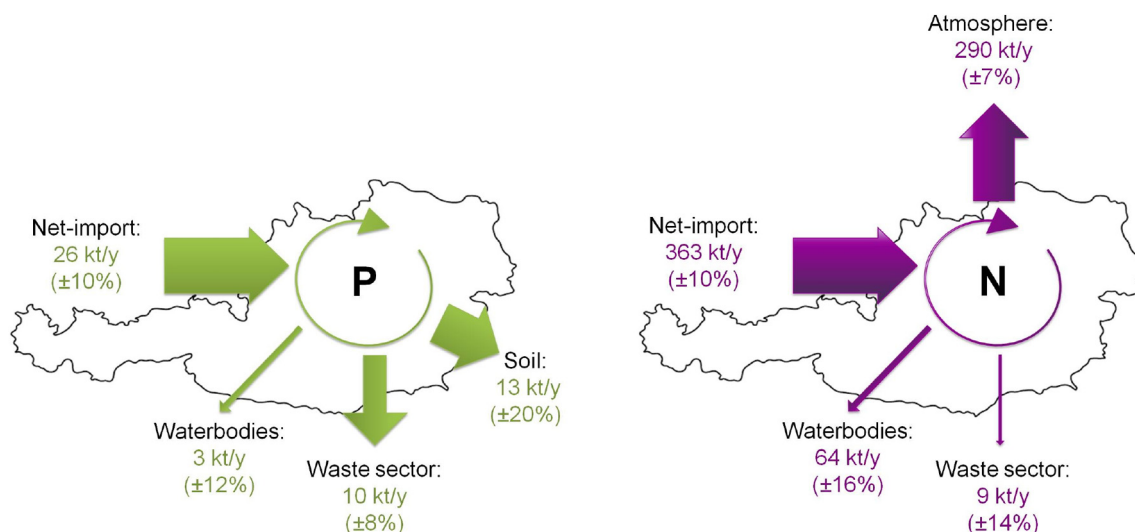


Fig. 4. Aggregated phosphorus and nitrogen balance for the reference year 2015.

for the lower P input with manure. For N, this effect is not visible, because contrary to P, the fertilizer efficiency of N is much lower than of mineral fertilizer, so that the reduction of N taken up by plants is smaller than the decrease in demand following the lower need for fodder production in this scenario. On the other hand, the low N-fertilizer efficiency of manure is the reason why N water emissions rise in the measure “N-emission control during manure storage”.

Similarly, the low P-fertilizer efficiency of compost causes P water emissions to rise in the measures “Improved collection of organic household waste” and “Efficient use of compost”. Water emissions of N from agriculture and P soil accumulation in agricultural soils also rise in these scenarios, however, as at the same time the amount of compost applied to private gardens and public green areas and forest and miscellaneous soils is reduced respectively, overall effects on these indicators remain positive. As mentioned in Section 2.2, results may be distorted by the simplified assumptions on fertilizer efficiency in the later two processes though. Lower fertilizer efficiency of biomass ashes compared to mineral P-fertilizer is also the reason, why water emissions and soil accumulation of P rise in the measures “Increased recycling of biomass ashes” and “Increased recycling of green waste”.

The measures “Increased P-removal efficiency of waste water treatment plants” and “Increased N-removal efficiency of waste water treatment plants” focus on reducing emissions to water bodies without further consideration of the fate of the removed nutrients. Consequently, these measures are marked by an increase of P losses in the waste sector as well as P soil accumulation and air emissions of N respectively.

In all cases negative impacts are however outweighed by positive effects in other indicators. Moreover, they can be mitigated by

combining for instance measures that would cause an increase in water emissions with measures aimed at improving fertilizer efficiency or by implementing measures that would reduce nutrient input to agriculture together with measures aimed at reducing the need for agricultural products, as can be seen in the combined measures scenario. In total, the annual amount of P and N making its way through the system can be significantly reduced: from 26 ± 2 kt/a to 12 ± 2 kt/a for P and 363 ± 36 kt/a to 219 ± 23 kt/a for N (Fig. 6) Note that Fig. 4 and Fig. 6 show the full MFA balances in aggregated form. The flow “Net-import” thus comprises not only P and N in the form of mineral fertilizer, but also with food, feed and other products, while the flow “Atmosphere” stands for the net-export of deposition and emissions of N. This is why relative reductions in these flows between Fig. 4 and Fig. 6 differ from the EIPs “Demand for mineral P/N-fertilizer for domestic use” and “N emissions to atmosphere” listed in Table 3.

In general, measures aimed at reducing demand for P and/or N score higher than those directed at emission reduction, as the later are often end-of-pipe solutions that tackle the system at points, where interaction with other sectors is limited. An exception is the measure “Exploitation of maximum fertilizer efficiency” because here the reduction of nutrient losses to water bodies is directly reflected in a lower fertilizer demand. The effect of measures directed at increased nutrient recycling largely depends on the mass of recyclable product possible to obtain and on its efficiency when used as fertilizer in agriculture. As mentioned in Section 2.2 EIPs of the measure “Exploitation of maximum fertilizer efficiency” are likely to have been overestimated. However, the focus of this study was not on the exact quantification of improvement potentials, but rather on interactions of the two studied substances and the behavior of the system as a whole. High effectiveness of fertilizer efficiency increases are in line with the findings of Abalos et al. (2016), Ahrens et al. (2010) and Ma et al. (2013).

Furthermore, what has already been observed by Zoboli et al. (2016) for phosphorus is also true for the present study: To date, we seem to be far more successful in environmental protection than in resource protection. This is reflected in the fact that improvement potentials for mineral fertilizer demand are about twice as high as for reduction of emissions to water bodies and air, both when looking at the combined measures scenario (69% lower fertilizer demand compared to 34% lower water emission for P and 84% lower fertilizer demand compared to 50% and 37% reductions in water and air emissions respectively for N) and average scores of individual measures

Table 2
Overview of data reconciliation results. D: degree by which a priori values are altered during data reconciliation with respect to a priori uncertainty.

	Reconciled data [% of output data]	D [%]	Mean a priori uncertainty [%]
Flows	40.3	3.8	17.6
Stocks	85.7	2.8	14.1
Stock deltas	84.0	3.8	2835.9*
Transfer coefficients	25.2	4.4	39.8
Overall	36.8	3.9	34.1

* Mean uncertainty > 100% is a result of Gaussian error propagation of a difference.

Table 3
EIPs of analyzed nutrient efficiency improvement measures. MFD: mineral fertilizer demand, LWS: losses in waste sector, WE: water emissions, SA: soil accumulation, AE: air emissions, WWTP: waste water treatment plants. All values rounded to significant digits. Values in brackets indicate uncertainty. Highest total and single-measure values for each EIP are marked in bold, for values in italics change is insignificant.

	MFD-P	LWS-P	WE-P	SA-P	MFD-N	LWS-N	WE-N	AE-N
Combined measures	0.69 (0.13)	0.59 (0.05)	0.34 (0.06)	0.56 (0.10)	0.84 (0.08)	0.63 (0.11)	0.50 (0.26)	0.37 (0.04)
Max. fertilizer efficiency	<i>0.15</i> <i>(0.20)</i>	<i>0.003</i> <i>(0.003)</i>	<i>0.08</i> <i>(0.09)</i>	<i>0.13</i> <i>(0.15)</i>	0.60 (0.09)	<i>0.0001</i> <i>(0.0009)</i>	0.34 (0.10)	0.190 (0.019)
Sewage sludge recycling	0.44 (0.06)	0.255 (0.023)	0.119 <i>(0.024)</i>	0.22 (0.04)	−0.008 <i>(0.004)</i>	0.0486 <i>(0.0009)</i>	0.0045 <i>(0.0021)</i>	−0.0059 <i>(0.0006)</i>
Meat and bone meal recycling	0.084 <i>(0.010)</i>	0.215 <i>(0.003)</i>	−0.037 <i>(0.004)</i>	−0.067 <i>(0.007)</i>	0.026 <i>(0.004)</i>	0.4833 (0.0005)	−0.0022 <i>(0.0021)</i>	−0.0030 <i>(0.0007)</i>
Healthy diet	−0.02 <i>(0.07)</i>	0.09 <i>(0.06)</i>	0.04 <i>(0.15)</i>	0.03 <i>(0.10)</i>	0.16 <i>(0.08)</i>	0.14 <i>(0.11)</i>	0.12 <i>(0.34)</i>	0.10 <i>(0.05)</i>
Food waste reduction	0.01 <i>(0.04)</i>	0.06 <i>(0.06)</i>	0.028 <i>(0.024)</i>	0.03 <i>(0.04)</i>	0.12 <i>(0.07)</i>	0.04 <i>(0.13)</i>	0.03 <i>(0.23)</i>	0.03 <i>(0.03)</i>
P-N-reduced animal feed	−0.11 <i>(0.07)</i>	0 <i>(0.03)</i>	0.02 <i>(0.03)</i>	0.039 <i>(0.023)</i>	0.16 <i>(0.08)</i>	0.0001 <i>(0.0009)</i>	0.10 <i>(0.04)</i>	0.089 <i>(0.018)</i>
Agricultural erosion control	0.05 <i>(0.06)</i>	0.0009 <i>(0.0014)</i>	0.073 <i>(0.024)</i>	0.03 <i>(0.05)</i>	0.05 <i>(0.09)</i>	0.0001 <i>(0.0009)</i>	0.04 <i>(0.15)</i>	0.011 <i>(0.017)</i>
P-reduced detergents	−0.0001 <i>(0.0018)</i>	0.057 <i>(0.012)</i>	0.1281 <i>(0.0018)</i>	0.044 <i>(0.010)</i>	0 <i>(0.003)</i>	0.0001 <i>(0.0009)</i>	0 <i>(0.004)</i>	0.0007 <i>(0.0014)</i>
Improved manure storage	−0.0001 <i>(0.0017)</i>	−0.0001 <i>(0.0009)</i>	0 <i>(0.0007)</i>	0 <i>(0.0009)</i>	0.169 <i>(0.009)</i>	0.0001 <i>(0.0009)</i>	−0.057 <i>(0.005)</i>	0.087 <i>(0.005)</i>
Improved org. waste coll.	0.088 <i>(0.006)</i>	0.023 <i>(0.015)</i>	−0.048 <i>(0.003)</i>	0.084 <i>(0.005)</i>	0.004 <i>(0.004)</i>	0 <i>(0.05)</i>	0.0067 <i>(0.0022)</i>	0.0004 <i>(0.0018)</i>
Efficient compost use	0.0689 <i>(0.0025)</i>	0.0003 <i>(0.0008)</i>	−0.0387 <i>(0.0008)</i>	0.0777 <i>(0.0008)</i>	0.010 <i>(0.004)</i>	0.0001 <i>(0.0009)</i>	0.0106 <i>(0.0020)</i>	0.0022 <i>(0.0006)</i>
Increased P-removal WWTP	0.009 <i>(0.003)</i>	−0.026 <i>(0.004)</i>	0.138 (0.023)	−0.0068 <i>(0.0013)</i>	0 <i>(0.003)</i>	0.0001 <i>(0.0009)</i>	0 <i>(0.004)</i>	0 <i>(0.0014)</i>
Improved N-removal WWTP	−0.0001 <i>(0.0018)</i>	−0.0001 <i>(0.0014)</i>	0 <i>(0.0008)</i>	0 <i>(0.0010)</i>	0 <i>(0.003)</i>	0.0001 <i>(0.0009)</i>	0.061 <i>(0.022)</i>	−0.014 <i>(0.005)</i>
Biomass ash recycling	0.007 <i>(0.003)</i>	0.0165 <i>(0.0011)</i>	−0.0024 <i>(0.0010)</i>	−0.0044 <i>(0.0009)</i>	0 <i>(0.003)</i>	0.0005 <i>(0.0009)</i>	0 <i>(0.004)</i>	0 <i>(0.0014)</i>
Food industry waste recycling	0 <i>(0.003)</i>	0.0007 <i>(0.0011)</i>	−0.0002 <i>(0.0010)</i>	−0.0003 <i>(0.0009)</i>	0 <i>(0.13)</i>	0.0030 <i>(0.0009)</i>	0 <i>(0.014)</i>	0 <i>(0.03)</i>
Green waste recycling	0.004 <i>(0.003)</i>	0.0083 <i>(0.0011)</i>	−0.0012 <i>(0.0010)</i>	−0.0022 <i>(0.0009)</i>	0 <i>(0.003)</i>	0 <i>(0.0009)</i>	0 <i>(0.0024)</i>	0 <i>(0.0012)</i>

(5% vs 3% for P, 8% vs 4%/3% for N). This is partly an effect of measures taken in the past. Municipal waste water treatment plant removal efficiencies for instance rose from 64% to 90% for P and from 51% to 82% for N since 1999 and are now considered close to their maximal achievable limits (Überreiter et al., 2016). Nevertheless, even slight further improvements of removal efficiencies to 90% for N and 96% for P, as assumed in the present study, could reduce water emissions by 6% and 14% respectively. Although smaller than the reduction potentials achievable for mineral fertilizer demand they thus should not be omitted.

For air emissions of N another factor comes into play as well. As mentioned in the introduction, N-emissions to the atmosphere can take various forms with very different consequences for the environment and human health. Current measures to reduce N-emissions to the atmosphere are predominately concerned with transforming reactive forms of N into N₂ prior to emission (e.g. Campos et al., 2016; Grosso et al., 2009). Indeed, N₂ can be considered as unproblematic from an environmental perspective; in terms of resource efficiency keeping reactive N in the system as long as possible would be preferable though, because it would reduce the need for energy intensive artificial conversion of N₂ into NH₃ in the Haber-Bosch process.

It has to be noted that the combined measures scenario only represents the best case in the context of the present analysis. Although interaction between different measures was taken into account, only a case in which all measures are fully implemented was studied. Even higher increases in efficiency improvement potentials may be reached by implementing measures only up to a level in which co-benefits with other measures are maximized and negative trade-offs minimized. Creating such a true optimization model could be the issue of further studies. Moreover, measures are specifically fitted to

the reference situation of 2015, which only represents a snapshot in time. While dynamic MFA has evolved into a frequently applied method for metals over the past 20 years (Müller et al., 2014) it has not yet been applied much to nutrients. Zoboli et al. (2015) and Keil et al. (2018) have conducted time-continuous studies on phosphorus flows in Austria and India, efforts that should be increased to gain better understanding of the dynamics in phosphorus resource management. The availability of an extensive data set on national phosphorus flows since 1990 from the work of Zoboli et al. (2015) provides an excellent basis for developing the present model into a dynamic MFA. A top-down approach, where stocks are derived from the difference of inflows and outflows (Müller et al., 2014), possibly using bottom-up estimates for calibration and verification as in Buchner et al. (2015), seems to be the most suitable approach. Furthermore, system dynamics models as in Treadwell et al. (2018) could be used to study effects of implementation time of the different management scenarios evaluated in the present study. Finally, results are highly dependent on the weights assigned to the different efficiency improvement indicators. In the present study, equal weights for all indicators were assumed; it could be argued though that, considering the small absolute amounts of N ending up in the waste sector (about half of this being exported meat and bone meal), relative reductions in this field should be valued less than for mineral fertilizer, where high total quantities are involved. In aquatic ecosystems on the other hand even comparatively small increases in nutrient inputs might cause ecological collapse, thus justifying higher weights assigned to this indicator. However, goal weighting is always a subjective process and highly dependent on the specific aim of the problem at hand. It was therefore not considered an issue of the present work.

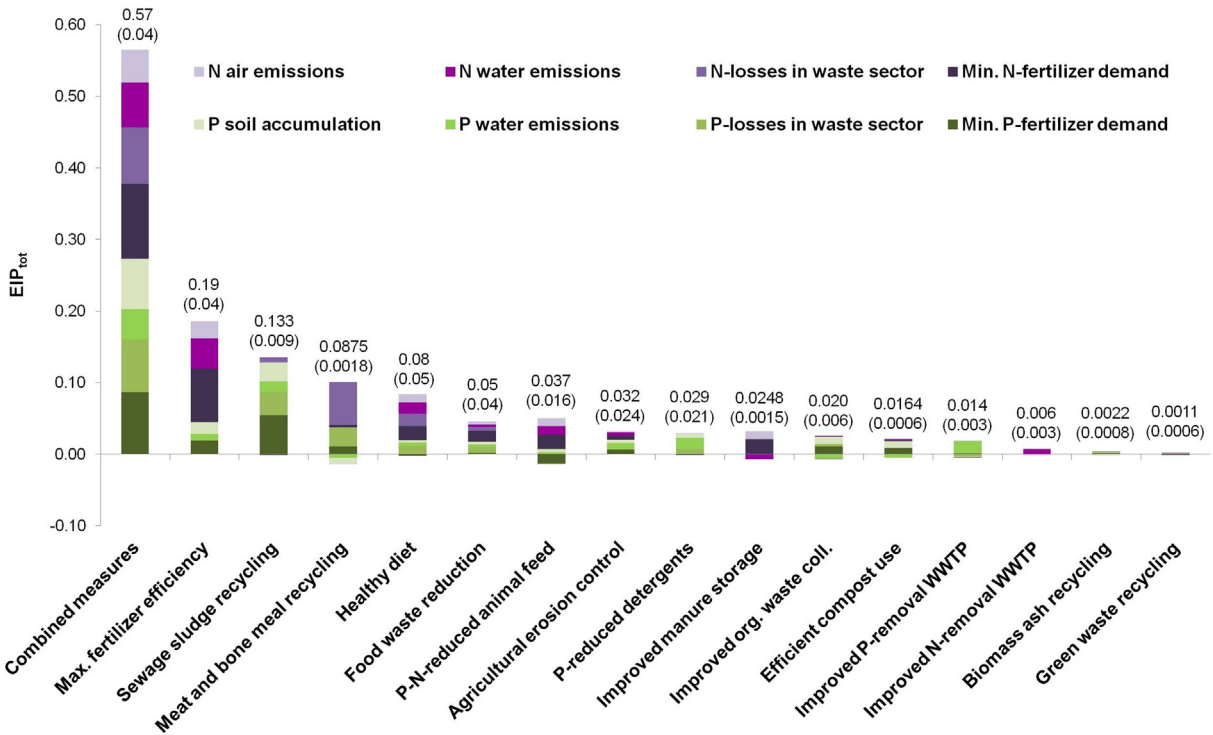


Fig. 5. EIP_{tot} of analyzed nutrient efficiency improvement measures. The measure “Increased recycling of food industry waste” was excluded from this depiction as it does not yield significant changes compared to the status quo.

4. Conclusion and outlook

The study highlights the close connection between the Austrian P and N systems and shows that measures aimed at improving P management are overall beneficial also for the N system and vice versa. Even where trade-offs exist, for instance where nutrient losses to water bodies increase due to a substitution of mineral fertilizers by organic fertilizers with lower nutrient efficiency, the negative effects are always outweighed by positive ones and can be compensated by combining different measures with each other. The best results in the context of the present study can thus be achieved by a simultaneous implementation of all the individual measures analyzed.

The optimal management strategy in terms of overall highest resource use efficiency can only be found by further analyses of the trade-offs and interactions between different flows, though. The experience gained in the present work may enable the inclusion of more substances that exhibit higher conflict potentials in management goals with P and/or N than the two nutrients show among each other into such an optimization model. For instance high concentrations of copper (Cu) have sometimes proven to be problematic in fertilizers derived from sewage sludge (Jossa and Remy, 2015). Especially for the measure “Full application of P- and N-optimized feed for cattle, pig and poultry”, which leads to an increase in mineral P-fertilizer demand, the rising contents of the toxic by-element

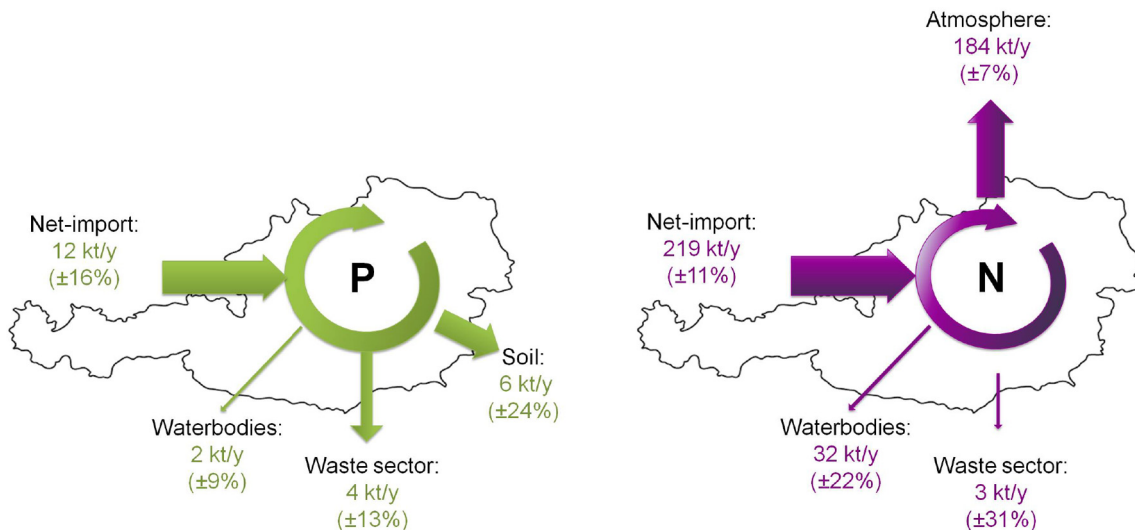


Fig. 6. Aggregated phosphorus and nitrogen balance for the combined measures scenario.

Cadmium (Cd) in rock phosphate as the quality of remaining reserves declines (Kratz et al., 2016), may be an additional concern. In order to identify strategies to handle conflicting goals extended statistical entropy (Sobaňka et al., 2014, 2012) can be used as an indicator. The advantage of this method is that it is both able to measure the overall resource efficiency of a system and allows for differentiation between different species of a substance, which is especially important to capture the effects of different forms of nitrogen emissions to water and atmosphere. Furthermore, methods of dynamic MFA could be applied to deepen knowledge of temporal trends and management dynamics.

The high level of uncertainty encountered in the present model, even though Austrian P management was among others chosen as a case study for its high level of reporting and generally good quality of available data compared to other substances (Zoboli et al., 2015), stresses the need to continue the ongoing research on dealing with uncertainty in MFA.

Nevertheless, it could be shown that although coupling significantly raises model complexity, material flows of more than one substance can be simultaneously analyzed in a rather complex system. Transition from mono-substance to multi-substance MFAs with the tools at hand (i.e. the MFA-software STAN) should thus be possible also for other resources. Confirming this was one of the main aims of the present study, as like for P and N, coupled MFA reveals interrelations, co-benefits and trade-offs of different resource systems that might have been omitted in a mono-substance analysis and thus improve judgment of sustainability and viability of different management strategies.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.05.177>.

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