The stocks and flows of nitrogen, phosphorus and potassium across a 30-year time series for agriculture in Huantai county, China

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Abstract

In order to improve the efficiency of nutrient use whilst also meeting projected changes in the demand for food within China, new nutrient management frameworks comprised of policy, practice and the means of delivering change are required. These frameworks should be underpinned by systemic analyses of the stocks and flows of nutrients within agricultural production. In this paper, a 30-year time series of the stocks and flows of nitrogen (N), phosphorus (P) and potassium (K) are reported for Huantai county, an exemplar area of intensive agricultural production in the North China Plain. Substance flow analyses were constructed for the major crop systems in the county across the period 1983-2014. On average across all production systems between 2010 and 2014, total annual nutrient inputs to agricultural land in Huantai county remained high at 18.1 kt N, 2.7 kt P and 7.8 kt K (696 kg N/ha; 104 kg P/ha; 300 kg K/ha). Whilst the application of inorganic fertiliser dominated these inputs, crop residues, atmospheric deposition and livestock manure represented significant, yet largely unrecognised, sources of nutrients, depending on the individual production system and the period of time. Whilst nutrient use efficiency (NUE) increased for N and P between 1983 and 2014, future improvements in NUE will require better alignment of nutrient inputs and crop demand. This is particularly true for high-value fruit and vegetable production, in which appropriate recognition of nutrient supply from sources such as manure and from soil reserves will be required to enhance NUE. Aligned with the structural organisation of the public agricultural extension service at county-scale in China, our analyses highlight key areas for the development of future agricultural policy and farm advice in order to rebalance the management of natural resources from a focus on production and growth towards the aims of efficiency and sustainability.

1 Introduction

China is the largest single consumer of inorganic fertilisers in the world, responsible for approximately 30% of annual global fertiliser use for each of the macronutrients: nitrogen (N), phosphate (P_2O_5 total nutrients) and potash (K_2O total nutrients) (FAOSTAT, 2014). The majority of China's demand for inorganic fertilisers is met by internal reserves or by synthesis, with the exception of potassium (K) for which China is heavily reliant on imports, to the extent that >15% of global imports of K entered China in 2014 (FAOSTAT, 2014). However, China is also recognised as a global hotspot of relatively low nutrient use efficiency within agricultural production (Foley et al., 2011; Vitousek et al., 2009). The high demand for inorganic fertilisers within China, coupled with inefficient nutrient use, exerts significant pressure on finite rock reserves (for K and phosphorus, P) and the global inorganic fertiliser markets that depend on these reserves. As high-quality rock reserves may diminish within the near future (Cordell and White, 2014; Wang et al., 2011), the pressure on fertiliser markets due to the demand exerted by China is likely to increase substantially. Further, the environmental costs associated with the production of inorganic fertilisers and with inefficient nutrient use within agriculture, including greenhouse gas emissions, degradation of soil, freshwater and marine ecosystems and declining air quality, are likely to grow and to be particularly pronounced within China (e.g. Chen *et al.*, 2014).

Responding to these challenges requires new frameworks comprised of policy, practice and the means of delivering change, in order to improve the efficiency of inorganic fertiliser use (Bellarby et al., 2015), whilst also meeting projected increases in the demand for food within China (Zhang et al., 2011). These frameworks should emerge from systemic understanding of the stocks and flows of nutrients within agriculture. In this context, substance flow analyses (SFAs) can be used to quantify the stocks and flows of a substance (in this case, individual nutrient elements) within a defined spatial unit and across different sectors within that spatial unit (Cooper and Carliell-Marquet, 2013; Senthilkumar et al., 2012). Previous SFAs within China have examined nutrient stocks and flows at country-level (Hou et al., 2013; Ma et al., 2010), at province-level (Ma et al., 2012; Sheldrick et al., 2003) and at the level of individual farm systems (Gao et al., 2012; Hartmann et al., 2014). These analyses reveal substantial regional differences in nutrient management within agriculture, largely reflecting differences between climatic regions and the resulting dominant production systems. In general terms, nutrient use efficiency (NUE) is greater in arable crop production systems than in vegetable and fruit production in China, whilst vegetable and fruit production demonstrate higher NUE than animal production systems (Ma et al., 2012). Enhancing NUE within animal husbandry in China is recognised as a particular challenge, due to increasing disconnection between concentrated animal production facilities and land to which animal manure can be returned (Bai et al., 2013; Chadwick et al., 2015).

However, the majority of SFAs to date have either examined only one nutrient element (usually N or P), or N and P in combination (Cooper and Carliell-Marquet, 2013; Ma et al., 2012; Senthilkumar et al., 2012). Little research has examined the third macronutrient, K, in combination with N and P (Sheldrick et al., 2003, Zhen et al., 2006), despite the fact that an imbalanced supply of the macronutrients N, P and K can adversely impact crop yield and decrease NUE (Dai et al., 2013). Further, the majority of SFAs have only focused on data from a single year, providing a snapshot of nutrient stocks and flows for a given spatial unit (Chowdhury et al., 2014). However, such snapshots do not capture longer-term trajectories of change in nutrient stocks and flows within a system, as driven by natural processes, such as variation in rainfall or temperature regimes, by management practices, such as crop rotations, by policies, such as variation in trade tariffs, farm input and fertiliser industry subsidies (Li et al., 2013; Sun et al., 2012), or by regulation, such as the ban on the burning of crop straw in China from 2008 (Miao et al., 2011). The use of longer time series of data to construct SFAs would help to avoid the risks associated with basing policy and practice on short-term analyses that may not accurately account for longer-term changes in nutrient management within a system (Sheldrick et al., 2003).

Our previous research suggests that the county-scale is a key spatial unit at which to consider the potential for change in nutrient management practices within China, particularly for largely rural counties in which the management of nutrients in agriculture is clearly important (Smith and Siciliano, 2015). The county-scale is especially relevant in China because of the corresponding structural organisation of the public agricultural extension service. Key decisions regarding agricultural policies and farm advice provision are made for county-wide execution by the County Agricultural Bureau, which has considerable autonomy with regard to such policies and advice (Bellarby et al., 2017; Smith and Siciliano, 2015). For example, the bureau is responsible for undertaking soil nutrient surveys and for the provision of fertiliser recommendations based on the resulting information. These recommendations are often applied county-wide, and form the basis for compound fertiliser formulations sourced from manufacturers for county-wide distribution. In the current paper, we report a county-level analysis of nutrient use within agricultural production systems in China, based on SFAs for the macronutrients N, P and K using a time series of data that spans 32 years from 1983 to 2014. The objectives of these analyses were: i) to quantify changes in N, P and K stocks and flows within individual production systems at county-scale in China over a 30-year timescale; ii) to interpret drivers of the observed changes in nutrient management over this timescale; and iii) to consider the ways in which analysis of historical patterns of nutrient use in agriculture can inform future policy and practice seeking more sustainable stewardship of N, P and K resources.

2 Materials and methods

2.1 System boundary and design of the substance flow analyses

Substance flow analyses were constructed for Huantai county in Shandong Province, China (Figure 1). Huantai county covers approximately 520 km^2 with a total farmed area of 354 km^2 (68%) in 1980. Agricultural production in the county is primarily an intensive rotational double cropping area of summer maize and winter wheat, typical of agriculture within the North China Plain (Ha et al., 2015). Crop production relies heavily on irrigation with groundwater (Chen et al., 2010; Liu et al., 2005). Other arable crops (cotton, peanut, potato, soybean and sweet potato), vegetable, fruit (apple, apricot, Chinese date, grape, hawthorn, peach and pear), as well as livestock, are produced in the county. In this paper, other arable crops, vegetable, fruit, and livestock are each considered as individual production systems (Figure 2). Approximately 250,000 of the county's 493,000 population are engaged in farming (Huantai Agricultural Bureau, 2014).

The SFA approach uses mass balance principles to systemically identify and quantify an element from source (here, input into one of the production systems within Huantai county), through internal stocks and flows within the defined system boundary, to the final managed or unmanaged outflow of an element across the system boundary (Cooper and Carliell-Marquet, 2013; Senthilkumar et al., 2012). Stocks and flows for the nutrients N, P and K were quantified for Huantai county on an annual basis from 1983 to 2014, incorporating multiple cropping cycles within a single year where relevant. The corresponding conceptual design for the SFA is reported in Figure 3, alongside data describing the average mass of nutrient elements over the most recent five years of our analyses (2010 – 2014). The SFAs were differentiated between four individual crop systems (cereal (incorporating wheat and maize), other arable crops, vegetable and fruit). Additionally, livestock production was included in order to estimate nutrient flows from the crop systems to the livestock system as feed, alongside flows from the livestock system to the crop systems as manure. Although livestock production occurs within Huantai county, the focus of the current paper is the major crop production systems. Detailed consideration of the individual stocks and flows of nutrient elements within livestock production in the county was beyond the scope of the research reported here.

2.2 Data sources and equations

2.2.1 Inputs, outputs and recycling of nutrients in agricultural production systems

Agricultural statistics collated and reported at county-level in China provide a consistent database and the foundation for constructing SFAs. Annual statistics for Huantai county have been published since 1980 by the Huantai Agricultural Bureau. Data from these yearbooks were used in the research reported here from 1983 onwards, because earlier data were not complete for all components of the SFAs. These yearbook data were supplemented by data and functions derived from the literature and by expert knowledge where necessary. We acknowledge the uncertainties which are unavoidable when constructing SFAs at this spatial and temporal scale, meaning that these uncertainties may introduce apparent fluctuations in nutrient stocks and flows that cannot clearly be attributed to changes in nutrient management practices. For example, these uncertainties include the application rates of fertiliser and manure as well as residue management practices, which were not available within statistical yearbooks (Table S1), but would have been valuable additions to the research reported here. However, data that was not available in the yearbooks have been carefully estimated based on interviews with local experts and farmers, and constitute the best information currently available with which to undertake the type of analyses reported here. An initial representation of the uncertainty associated with the individual components of the SFAs, alongside full details regarding the sources and the derivation of the data used in the SFAs, are given in Table S1.

The amount of crop residue returned to soil was derived from the straw to grain ratio (Peng et al., 2014, Table S3) which is then applied as crop input in the subsequent year. The exception was the first year, which used the crop production of the same year instead. The nutrient flows of all the arable systems were quantified using nutrient contents and straw/grain ratios (Tables S2 – S3). In the fruit system it is assumed that the "residue" incorporates the biomass increase though it should be noted that is likely an underestimate. In the livestock system, the nutrient flows were determined via the lifespan of livestock and the nutrient content of livestock outputs (Tables S4 and S5). The amount of livestock manure produced was calculated via livestock numbers and manure production rates per head (MOA, 2009, Table S5). The nutrient input via feed into the livestock system was calculated to balance all livestock outputs. All manure was assumed to be completely distributed between land under different forms of crop production, according to expert knowledge and local farming practices (e.g Huantai Agricultural Bureau, 2014). The actual mass of nutrient elements initially present in manure was reduced by a total of 50.8% N, 48.1% P and 43.3% K on the assumption that a given mass was lost during housing (Webb and Misselbrook, 2004) and during the storage of excreta, for example by ammonia volatilisation, based on Jia et al. (2014). Further, it was assumed that the amount of manure that was returned to land under fruit and vegetable did not exceed average nutrient application rates according to Chadwick et al. (2015). Given a surplus supply of manure in excess of these threshold values, the surplus manure was assumed to be exported out of Huantai county in order to avoid unrealistic manure application rates. It is recognised that surplus manure may be exported directly into water courses in other parts of China (Strokal et al., 2016). However, the SFAs reported here assumed that this was not the case in Huantai county, which is at least partly supported by strict environmental laws and low precipitation levels in the county resulting in low river flows that would render this option impossible.

2.2.2 Losses of nutrients to the environment across production system boundaries

2.2.2.1 Atmospheric losses

For P and K, it was assumed that no gaseous losses occurred. Empirical models were used to estimate losses of ammonia (NH_3) (Bouwman et al., 2002) and the nitrogenous greenhouse gases, nitrous oxide (N_2O) and nitric oxide (NO) (Stehfest and Bouwman, 2006). Di-nitrogen (N_2) emissions were estimated via the ratio of N_2 to N_2O produced during denitrification, using the spreadsheet model SimDen (Vinther, 2005). Table S9 provides an overview of the factor class used in the published functions. A slightly different approach was used for the calculation of N_2O and NO losses from the high nutrient-input systems (vegetable and fruit production), which were beyond the range of N application rates for the empirical functions developed by Stehfest and Bouwman (2006). In these cases, an emission factor (EF) of 0.96% has been specifically developed for lowland horticulture in China (Shepherd et al., 2015) and this EF was multiplied by the N application rate to estimate gaseous N losses as N_2O and NO for the relevant systems within Huantai county.

2.2.2.2 Aqueous losses – erosion, runoff and leaching

Nutrient export via soil erosion was not estimated because existing approaches rely on estimates of the total nutrient content within soil, which were not available for Huantai county. However, Huantai county is located in the extremely flat North China plain (land gradient ratios ranging between 1/800 and 1/3500, Liu et al., 2005), meaning that nutrient export via erosion is expected to be low or negligible, especially as open fields are also generally bunded (Wang et al., 2013b). Export of nutrients via runoff and leaching were determined using the empirical model developed for N (Velthof et al., 2009). This model requires widely available information regarding slope, land use, soil type, soil and rooting depth, soil clay content and precipitation surplus, in order to select a series of factor classes that ultimately determine a loss factor (Table S10). With respect to the precipitation surplus, this was assumed to be within the lowest factor class for Huantai county, because crops are irrigated, and in order to generate a conservative estimate of aqueous losses which have been suggested to be overestimated when applying this kind of empirical function to China (Ongley et al., 2010). The algorithms for the calculation of nutrient export via runoff were considered to be the same for N, P and K, which are related to fertiliser application rates. The leaching factor was multiplied by the nutrient surplus at the soil surface (nutrient surplus = total nutrient input – crop uptake), after NH_3 emissions were accounted for. However, the mobility of P and K in soil (and thus leaching) is lower compared to N (Lehmann and Schroth, 2003). Therefore, a leaching rate of 0.1 kg nutrient ha^{-1} $year^{-1}$, reported by Némery et al. (2005) for P, was assumed throughout for P and K.

2.2.3 Nutrient use efficiency

The concept of nutrient use efficiency (NUE) has been applied for many years to crop uptake in agricultural systems (e.g. Moll et al., 1982). However, there are multiple definitions of NUE, especially in regard to which nutrient inputs are considered. In the research reported here, NUE was calculated as described by Ma et al., (2012) for N, P and K for each individual production system in Huantai County based on the SFAs and using Equation 1:

$$\left(\frac{N,P\ or\ K_{product\ output}}{N,P\ or\ K_{external\ inputs}}\right) * 100 \qquad [1];$$

Here, N, P or $K_{\text{product output}}$ relates to marketable output, such as grain, and N, P or $K_{\text{external input}}$ includes all human and natural inputs, i.e. inorganic fertiliser, manure, atmospheric deposition, biological N fixation and nutrients introduced via crop seeds or seedlings and irrigation. Additionally, Ma et al., (2012) included human wastes and by-products from the food processing industry as well, which are not considered in this study.

2.2.4 Historical fertiliser recommendations

Fertiliser recommendations relating to wheat production in Huantai county for exemplar years were sourced from the Huantai Agricultural Bureau (2014). The county fertiliser recommendations were based on annual soil nutrient analysis, available fertiliser types and their nutrient contents, as well as the predicted crop yield and weather conditions for the forthcoming year (Huantai Agricultural Bureau, 1990). In the research reported here, these recommendations were compared to estimates of the mass of nutrients taken up by a crop and to farmer fertiliser application practice, based on the SFA results for the corresponding year.

3 Results

3.1 Summary of nutrient flows for all production systems during the period 2010-2014

Figure 3 reports annual nutrient flows for agricultural production in Huantai county, averaged for the period 2010 to 2014, providing a summary of total nutrient flows to, from and between individual production systems. Analysis of the 30-year time series for N, P and K is reported in subsequent sections.

The total average annual input of nutrients to agricultural soils within Huantai county between 2010 and 2014 was 18.1 kt N, 2.7 kt P and 7.8 kt K (696 kg N/ha; 104 kg P/ha; 300 kg K/ha). The majority of the overall input of nutrients was associated with inorganic fertilisers for N (67%) and P (81%). In contrast, fertiliser and returned crop residue contributed relatively similar proportions of the total K input (46% and 52%, respectively). Considering all production systems together, manure only contributed between 1.6% and 3.4% of the total input across N, P and K, because the input of manure is concentrated on a relatively small area of fruit and vegetable production within the county (Figure 2). The recycling of crop residue represented a larger input of nutrients to soil at county level (16% N, 15% P, 52% K) compared to the input via manure. For N, atmospheric deposition was also a more significant nutrient input (c.11% N) compared to manure (Figure 3). However, manure contributed more than 20% of the total P and K input, alongside around 19% of the total N input, to soil under fruit and vegetable production.

In absolute terms, the largest nutrient flows were observed in the wheat/maize and vegetable production systems in the county, driven by the large area of land under this form of production (for wheat/maize) or by the intensive use of nutrients to support production (vegetable). The proportion of nutrient input to a system that was subsequently taken up and incorporated into crop products varied between individual production systems, as reflected in the NUE data reported in Table 1 and discussed further in section 3.3. The balance term in the soil compartment of the SFA represents the proportion of the total nutrient input to soil, which is not taken up by crops. This mass of nutrients can either accumulate in the soil or be lost to the atmosphere or to receiving waters. Substantial accumulation of N, P and K in soil was observed under every form of production within Huantai county, although the absolute mass of nutrients that accumulated was particularly high under wheat/maize, vegetable and other arable crop production. The mass of nutrients accumulating within soil exceeded that in agricultural products for N, P and K under other arable crop and fruit production systems and, for P alone, under vegetable production. The losses of nutrients to the atmosphere or to receiving waters were at least 40% of the mass taken up by the different crops. In the extreme cases, losses of nutrients exceeded the mass taken up by crops by a factor of two for other arable crops and three for fruit (Figure 3).

The nutrients taken up by a crop are subsequently divided into fractions that are classed as product (e.g. the nutrient content of grain for wheat/maize), residue (nutrient content returned to the soil with crop residue) and waste (nutrient content within crop residue that is not returned to the soil). For "other arable crop", the waste nutrient content was approximately double the nutrient content within agricultural products themselves. However, the amount of waste nutrients in this production system was surpassed by the losses to the environment for N. The mass of nutrients returned to soil within residue was greatest in the wheat/maize production system, where 90% of residues are returned to soil. This is particularly apparent for K, where the return of residue was responsible for more than half of the total K input to soil. The wheat/maize and other arable crop systems also supply an input of nutrient elements to the livestock production system via feed. The livestock system was only differentiated between nutrients that are contained in livestock products (dairy, eggs and the whole animal) and nutrients that are contained in the excreta produced during the lifetime of the livestock. In the livestock sector, the amount of nutrients lost to the environment during housing and storage was greater than the sum of the total mass of nutrients (N, P and K) in livestock products and in manure returned to agricultural soils (Figure 3).

3.2 Long term trends in nutrient inflows and outflows at county-level

Total nutrient flows into and out of the soil surface across all production systems within Huantai county generally remained relatively stable or increased only gradually between 1983 and 1989 (Figure 4). However, inputs increased dramatically for N and P between 1989 and 1993 to reach maximum levels across the 30-year time series. The increase in K inputs was more prolonged, beginning in 1983 but not peaking until 1998, followed by a secondary increase in K inputs between 2009 and 2012. Outflows tended to mirror the increased inputs of nutrients between 1989 and 1993, although at a lower rate especially for P and K (Figure 4) in this period. After 1993, both inflows and outflows of N and P to the soil surface generally exhibited small decreases in absolute terms, with inflow and outflow of K remaining more constant. The outflow of each nutrient includes losses to the atmosphere and to receiving waters, which are particularly high for N, alongside the outflow of nutrient elements in agricultural products. Therefore, a positive net balance between inflows and outflows in Figure 4 indicates nutrient accumulation within the soils of the county, which is the case for N and, particularly, for P. The time series for K differs markedly compared to either N or P. A net deficit for K at the soil surface was observed between 1983 and 1993. Across all production systems, this deficit translated to approximately 8 kg ha^{-1} year^{-1} until 1989, after which the K deficit gradually decreased until inputs and outputs achieved an approximate balance from 1994 until around 2011, when a further increase in K inputs resulted in a net positive balance at the soil surface (Figure 4). Despite the positive soil K balance from around 2011 onwards, the overall soil K balance for the entire time series remains in deficit by 11.6 kg ha^{-1} when averaged across all production systems.

3.3 Time series for individual crop production systems in Huantai county

Due to substantial differences in the total area under production for individual crops in Huantai county, inputs and outputs of nutrients for each production system were normalised by area and are reported as kg nutrient ha^{-1} in Figures 5-8, allowing direct comparison between individual systems. Total nutrient inputs and NUE for each production system are reported as 5-year averages across the period 1983-2014 in Tables 1 and 2.

3.3.1 Wheat-maize production

In the wheat and maize production system, N and P application rates via inorganic fertiliser have fluctuated over the 30-year period, but have always remained by far the most significant source of both nutrient elements, with application rates consistently exceeding 400 kg N/ha and 50 kg P/ha. In comparison to inorganic fertiliser, other sources of N and P have remained relatively insignificant, although inputs of N and P via the recycling of crop residue, alongside N input via atmospheric deposition, have increased steadily between 1983 and 2014. For K, the input associated with recycling of crop residues grew in parallel with increasing inorganic fertiliser input, to the extent that each source contributed relatively equal masses of K to the total input to soil under wheat and maize production. The increase of residue returned to the soil occurred in two stages with wheat initially reaching a proportion of 90% being returned to the soil in 1995 followed by maize in 2008 (data not shown). Indeed, during the period 2007 to 2011 the input of K via inorganic fertiliser decreased in response to the increase of maize residue returned during that time, so that the return of crop residues to soil represented the most significant source of K to land under wheat and maize production. Manure application to wheat and maize always occurred at extremely low rates and finally decreased to zero after 1999, with vegetable production becoming the main recipient for manure generated in the county. The mass of nutrients that was estimated to be lost did not exhibit the same increase as observed for the input of nutrients during the period 1983-1993, particularly for P and K. Nutrient use efficiency for N and P increased substantially between the beginning and the end of the 30-year period, primarily as a result of increased output of nutrients within crop products rather than any substantial decrease in nutrient input. For K, NUE >100% was observed at the beginning of the 30-year period, reflecting greater offtake of K in agricultural products compared to the mass of K input to land under wheat and maize production. With increased K inputs in both inorganic fertiliser and crop residue from 1990 onwards, NUE decreased to below 100% and has remained relatively constant across the period 1990-2014. However, cropland is the only production system that still exhibits a negative soil accumulation for K (-995 kg ha^{-1}) across the whole time period.

Generally, the mass of inorganic fertilisers recommended by the Huantai Agricultural Bureau to be applied for wheat production has decreased since 1997, although there was a substantial increase in the recommended rate of K application comparing 1997 to 2004-2014 (Table 3). In 1997 and 2004, N input via inorganic fertiliser, as determined in the SFAs reported above, was within or above the recommended range. In contrast, in 2006 and 2014, fertiliser N input for Huantai county was below the recommended levels and was well matched to the combination of grain and straw uptake. Fertiliser P input was also within or below the recommended range across 1997-2014, being only slightly above combined grain and straw uptake in 2014 and 2006, but in excess of these outputs in 2004 and 1997. Other than for 1997, fertiliser inputs of K remained below recommended rates for wheat in Huantai county. For all years, fertiliser K inputs were below the combined uptake in wheat straw and grain, which was also the case for fertiliser recommendations although these recommendations were higher than recorded inputs in the SFAs (Table 3). In all years reported in Table 3, the application of N as inorganic fertiliser exceeded wheat grain output by factors between 1.3 and 1.6. The application of P as inorganic fertiliser was also at least 1.3 times the wheat grain output and reached a maximum of 1.9 times grain output. For K, recommended fertiliser application rates were at least twice the crop grain output.

3.3.2 Other arable crops (soybean, peanut, cotton, potato and sweet potato)

On other arable crops, inorganic fertiliser was also the main source of nutrients, with application rates that approach those for land under wheat/maize production, despite much lower output of nutrients in crop products for these other arable crops (Figure 6). The application rates for inorganic fertiliser fluctuated dramatically over the 30-year time series, ranging from <50 kg ha^{-1} to >400 kg ha^{-1} for N between 2003 and 2008, and from <40 kg ha^{-1} to approaching 100 kg ha^{-1} for P between 2006 and 2009. These variations in the input of inorganic fertiliser for N and P show no consistent trend over the 30-year period. The input of inorganic K fertiliser remained below 50 kg ha^{-1} until 2000, after which it increased rapidly to reach approximately 150 kg ha^{-1} in 2014. The mass of both N and P output from Huantai county in crop products has been approximately equal to the mass of each element lost to the environment between 1983-2014, with some periods in which the losses exceeded the output in crop products, including between 2006 and 2013 for N where losses exceeded crop output by a factor of up to 4.3. For K, the output in crop products has remained substantially above the mass lost to the environment throughout the 30-year period. Nutrient use efficiency for these other arable crops in Huantai county was extremely low (around 10%) for all nutrients in the period 2010 - 2014 (Table 2).

3.3.3 Vegetables

Vegetable production was associated with the highest nutrient input rates across all three elements throughout the 30-year time series (Table 2), which is at least partly justified by the relatively high nutrient output associated with vegetable products compared to other production systems in Huantai county (Figure 7). This is consistent with a relatively high NUE for vegetable production, certainly with respect to N and K, compared to other production systems (Table 2). The inorganic fertiliser application rates for N and P decreased gradually between 1983 and 2000 (Figure 7), before increasing dramatically between 2005 and 2010, reaching (for N) or even exceeding (for P) fertiliser application rates in 1983. The greatest proportion of the manure produced by livestock in the county has always been applied to land under vegetable production, which could reach levels of over 90% of the total of the total manure produced in the county, with the rest distributed between the other production systems (data not shown). It is reflected in the large proportion of the total nutrient input to land under vegetable production that is associated with manure, especially for P and K. The amount of manure applied has fluctuated with the livestock numbers within the county. However, a maximum threshold for N input via manure has been set beyond which excess manure is assumed to be exported from the county. This threshold has been met for most years for land under vegetable production (data not shown). The output of nutrients within vegetable products remained fairly constant between 1983 and approximately 2000, after which it almost doubled for N, P and K. Estimated losses of N to the environment from land under vegetable production exceeded losses from land under all other forms of production in the county, approaching 400 kg ha^{-1} both in the early and later stages of the 30-year time series. Substantial losses of P and K were also estimated from vegetable production, with only fruit production being associated with similar losses for these elements.

3.3.4 Fruit

Nutrient inputs to land under fruit production have followed similar patterns to vegetable production in Huantai county (Figure 8), reaching total input rates that are second only to land under vegetable production (Table 1). Because the mass of nutrients output in fruit products has remained relatively low, NUE for fruit production in Huantai county is also low, reaching a maximum of only 10% for N, 9% for P and 25% for K (Table 2). Manure has been a particularly important source of both P and K, and second only to inorganic fertiliser as a source of N, for fruit production. The estimated losses of N and P to the environment from land under fruit production have exceeded the mass of N and P output in fruit products for much of the period 1983-2014. However, between 1997 and 2000 there was a substantial increase (a factor of 13 across all elements) in the output within fruit production. At least for K, this resulted in the nutrient output in products exceeding the estimated losses to the environment from 1999 onwards.

4 Discussion

4.1 Nutrient use efficiency in Huantai county

When averaged across all production systems, NUE for both N and P increased by approximately 20% within Huantai county between 1983 and 2014 (Table 2), although the most pronounced increases in NUE occurred before the late 1990s, particularly for N. Because the definition of NUE in the research described above deliberately includes all nutrient inputs other than crop residue, absolute NUE is lower than has been reported for similar production systems when only the input of fertiliser is considered. For example, in a recent analysis of wheat-maize production in Huantai county, Zhang et al. (2017) reported a NUE for wheat that approached 83% in 2012. However, our underlying analyses are consistent with those of Zhang et al., with NUE for wheat in the period 2010-2014 exceeding 76% when only fertiliser input is considered (Table 3). The substantial reduction in NUE that is observed when additional sources of nutrients beyond inorganic fertiliser are considered highlights the importance of properly accounting for all inputs in nutrient management plans, if more sustainable agricultural production is to be realised.

Variation in NUE over time has also been examined at larger spatial scales across the whole of China. For example, Ma et al. (2012) examined NUE for N and P across 31 provinces in China for 1980 and 2005, highlighting a declining trend in N-NUE (40 to 33%) and P-NUE (65 to 37%) for Shandong province, within which Huantai county is located, consistent with the overall development across all of China (N-NUE: 32 to 26%; P-PUE: 59 – 36%). The contrasting trajectories for NUE at province level and at the level of Huantai county indicates that other counties in Shandong province are likely to have seen substantial reductions in NUE over the past 30 years, in contrast to the increase we report for Huantai county. This can simply be due to a different crop mix in different areas as was pointed out by Zhang et al., (2015). This highlights the likely heterogeneity of nutrient stocks and flows when considered at different spatial scales. In turn, this emphasises the importance of undertaking analyses of nutrient stocks and flows at spatial scales that are aligned with the structural organisation of bodies able to deliver change in agricultural policy and practice, specifically the county agricultural bureau in the research reported here.

Increasing NUE in Huantai country between 1983 and 2014 was primarily due to increases in nutrient output associated with higher crop yields, rather than decreases in N or P inputs. Substantial yield increases (69% for cereals, 43% for vegetables and 23% for fruit) have been observed globally during this time period, with China being no exception (FAOSTAT, 2014). The drivers of increased yields around the world are associated with the introduction of improved crop varieties, but also with changes in management practices such as fertiliser input and mechanisation (Hazell, 2009). Huantai is considered one of the most advanced counties in China with respect to the introduction of agricultural technology, including compound fertiliser formulations and mechanisation (Zhang et al., 2017; Huantai Agricultural Bureau, 1993). The introduction of higher-yielding varieties of wheat and maize in the 1990s and 2000s has been responsible for increases in NUE for land under this form of production within Huantai county, whilst local government support for the purchase of agricultural machinery has enhanced crop residue incorporation within soil and reduced excessive fertiliser application (Zhang et al., 2017). However, production systems in Huantai county other than wheat-maize have also seen distinct increases in yield due to changes in agricultural practices over the past 30 years that have enhanced NUE. For example, a substantial increase in yield was also associated with the switch from open-field to greenhouse vegetable production around 2000, resulting in higher nutrient masses associated with crop outputs that has persisted until the end of the time series in 2014.

Despite the increase in NUE for N and P between 1983 and 2014 when averaged across all crop production in Huantai county, considerable differences in NUE were observed between individual production systems, consistent with previous research (Miao et al., 2011). Particularly low NUE was observed for the production of other arable crops and fruit, where NUE remained ≤25 % across all the nutrients in the period 2010-2014. To our knowledge, no previous research has assessed the NUE associated with the production of other arable crops in China, such as cotton, peanut, soybean, potato and sweet potato. This group of crops as well as fruit was associated within the lowest NUE values for N, P and K in our analyses, indicating that further research would be helpful in order to better understand how NUE associated with these crops can be enhanced. Particularly, in regard to fruit a better estimation of the nutrient uptake associated with biomass increase would be desirable. Still, our data are consistent with other research in China that has shown low NUE in fruit and vegetable production, due to excessive inputs of inorganic fertiliser and manure (Gao et al., 2012; Lu et al., 2016). This partly reflects risk-aversion among farmers who are concerned not to reduce the yield of high-value fruit and vegetable crops, which is also reflected in fertiliser recommendations that often advise nutrient applications in excess of crop demand (Bellarby et al., 2017; Lu et al., 2016, 2014; Smith and Siciliano, 2015). Furthermore, manure is not widely recognised as a nutrient source in fruit and vegetable production, but rather as a soil improver (Bellarby et al., 2017). However, our analyses suggest that manure, in combination with crop residue, could account for a significant proportion of the demand for P and K exerted by vegetable production, as well as the entire N, P and K demand associated with fruit production, as has also been highlighted in some previous research (Gao et al., 2012; Lu et al., 2016). However, the actual contribution of manure to the crop nutrition will depend on the availability of these nutrients. The lack of accurate characterisation of the available nutrient content of manures is one of the significant barriers to the improved use of manure within agricultural production within China (Chadwick et al., 2015). This is aggravated by the widespread absence of the machinery required to apply manure to land (Ma et al., 2012).

The patterns of K use in crop production in Huantai county contrast strongly with those for N and P. Nutrient use efficiencies >100% for K in the period to approximately 1995 indicate that soil reserves of K were effectively mined during this time in order to support production. Prior to the wider introduction of chemical fertilisers in the mid 1960s (Miao et al., 2011), farmers in the county did not experience K limitation of crop production, because yields and therefore crop demands were lower and because manure was more heavily recycled to soils thereby supplying sufficient inputs of K (Meng et al., 2000). However, the increase in inorganic fertiliser use in China has generally reduced the input of organic materials to agricultural soils (Chadwick et al., 2015). Early use of inorganic fertilisers mainly involved the supply of N and P, meaning that crop demand exceeded K input and that net removal of K from soil reserves began. The depletion of soil K reserves in China was identified as a serious problem by the World Bank in the late 1990s, and one that could even lead to an irreversible soil degradation (Sheldrick et al., 2003). Compound fertilisers were gradually introduced for different crops to address this problem (Huantai Agricultural Bureau, 2014). After 1995 the increases in the inputs of K to soil by farmers in Huantai county have reversed the mining of soil K, resulting in NUE <100% and a net surplus of K at the soil surface (Figure 4). Important sources of K within the county extend beyond only inorganic fertiliser to include crop residues for the wheat-maize production system (Figure 5) and manure for the vegetable (Figure 7) and fruit production systems (Figure 8). These additional K inputs have been recognised by farmers in the county who have decreased fertiliser K input in wheat-maize systems in parallel with increasing incorporation of maize residue within soils (Figure 5). However, the input of K to the wheat/maize system has proven insufficient after 2005, leading to increased fertiliser K inputs in recent years (Figure 5). K input levels now exceed the immediate crop demand, which for the wheat maize system has still not bee sufficient to replenish the K mined in early years.

4.2 Nutrient balance at the soil surface in Huantai county

Despite the increases in NUE reported over the past 30 years for Huantai county, considerable surpluses of N, P and K have been observed across all production systems (with the exception of K in the wheat maize system) during this period. The N estimated to be accumulated in soil under the wheat maize system in this study (1.3 t ha^{-1}, derived from data in supplementary material) is consistent with a reported soil carbon stock increase of 12 t ha^{-1} in the same time period (Liao et al., 2014) assuming an approximate C:N ratio in soil of 1:10. A net surplus of nutrients at the soil surface may previously have been justified on the basis of needing to enhance the fertility of much agricultural land in China (Ju et al., 2004). However, this is not currently a requirement for large areas of land under agricultural production in the North China Plain. A continued surplus of nutrients at the soil surface has two potentially significant implications. Firstly, the surplus increases the risk of immediate export of nutrients to the environment and the adverse impacts associated with nutrient export from agricultural land. For example, the average export of N between 2010 and 2014 to the atmosphere or to receiving waters was at least 40% of the mass taken up by the different crops in Huantai county. In extreme cases, losses of N exceeded the mass taken up by crops by a factor of 1.8 for other arable crops and three for fruit. The environmental impact of excess nutrient leaching through the soil profile has already been documented locally for the concentration of nitrate in groundwater within Huantai county (Liu et al., 2005; Xue et al., 2015). More broadly, elevated nitrate levels in groundwater are a widespread problem in intensively farmed areas in China (OECD, 2007; World Bank, 2006), as in many other countries. Such pollution can impose significant economic costs in terms of water treatment requirements or, ultimately, the loss of available resources. Our estimates of losses are very conservative compared to other studies and our total N losses amount to only 77 kg ha^{-1} in 2005 in comparison to a total of 237 kg ha^{-1} losses of N reported by Ma et al., (2012). However, the total N losses were still at least 40% of applied fertiliser, which is consistent with other Chinese studies mentioned in Ma et al., (2012).

Secondly, even after accounting for losses of nutrient elements to the atmosphere or to water, a positive nutrient balance was observed at the soil surface across all nutrients and production systems (with K in wheat/maize being in exception to this), indicating that net accumulation of nutrients occurred within the agricultural soils of Huantai county (mean soil accumulation of N: 1743 kg ha^{-1}, P: 863 kg ha^{-1} and K: -872 kg ha^{-1}), which was confirmed indirectly via increased soil organic carbon levels (Liao et al., 2014) and elevated nitrate groundwater levels (Liu et al., 2005) in the same area but also directly by elevated soil nutrient levels across China (e.g. Chen et al., 2017; Yan et al., 2013). Increases in soil nutrient concentrations above optimum levels for crop production are known to increase the risk of diffuse water pollution from agriculture, a risk that may persist as a legacy of previous agricultural practices many decades after adjustments are made to the rate at which new nutrient inputs are applied to agricultural soils (Haygarth et al., 2014; Sharpley et al., 2013; Wang et al., 2013a).

4.3 Opportunities for future improvements in nutrient use efficiency in agriculture within Huantai county

Generally, the output of each nutrient element in crop products has stabilised since the mid-1990s in Huantai county (Figure 4). Therefore, further increases in NUE, leading to reductions in the adverse impacts associated with nutrient export from agricultural land, should focus on closer alignment between nutrient inputs and crop demand. The SFAs reported above highlight a number of key areas in which future agricultural policy and practice could try to deliver beneficial change in nutrient management. Firstly, there remain opportunities to optimise inorganic fertiliser applications within the county. Generally, research and advice is often more advanced for cereal crops in China, due to their major role in ensuring national food security (Dai et al., 2013) and the large areas of land under this form of cultivation. For example, a relatively large number of studies have examined nutrient management within wheat and maize production (e.g. Cai et al., 2002; Chen et al., 2014; Dai et al., 2013; Hartmann et al., 2014; Ju et al., 2009). This is reflected in fertiliser recommendations, which are generally well-matched to total crop output (Table 3). Relatively widespread mechanisation of fertiliser application to cereal crops in Huantai county has also helped to reduce the excessive input of inorganic fertiliser (Zhang et al., 2017). However, it is clear from our SFAs that parallel work still needs to be done in other production systems (fruit, vegetable and other arable crops) to reduce excessive applications of inorganic fertilisers. Whilst the area of land under these forms of production in Huantai county is much lower than for the wheat-maize rotation, the very high rates of inorganic fertiliser application to these areas of land, coupled with possible increases in the area of land under these forms of production as diets change within China (Huang et al., 2014; Yan et al., 2013), indicate that further attempts to limit excessive inorganic fertiliser applications to non-cereal crops is necessary. The priority to make these systems more sustainable is to drastically reduce the current chemical fertiliser input and take the contribution of nutrients from manure into account (Chen et al., 2017; Yan et al., 2013).

Secondly, there is clear need to appropriately account for additional sources of nutrients that are input to agricultural soils as part of future nutrient management strategies, across all nutrient elements and crop systems. This is illustrated through our observations of similar NUEs for both wheat-maize and vegetable production within Huantai county, despite the fact that inorganic fertiliser management is often deemed to be well-matched to wheat-maize demand within the county. Our observations regarding NUE partly reflect the substantial input of nutrients from sources other than inorganic fertiliser, in particular crop residues to wheat-maize systems and manure to vegetable systems. More generally, improvements in NUE could be generated by adjusting inorganic fertiliser applications to account for crop nutrient supply from: atmospheric N deposition; crop residue; manure and soil reserves. Clearly, there are significant challenges in the use and accurate accounting for nutrients within these sources. For example, the mineralisation of nutrients input to soil within crop residues or manure is critical for subsequent supply to crops, but the extent of mineralisation varies significantly depending on factors such as soil temperature, microbial community composition and soil nutrient status (Hartmann et al., 2014). Further, following the initial return of crop residue to soils, soil organic carbon levels will increase and may lead to the immobilisation of nutrients with the increase in organic matter (Liao et al., 2014), which in turn will decrease N_2O and NO emissions (Yao et al., 2017). Enhanced use of livestock manure will require further mechanisation of agriculture in China in order to support the distribution and application of manure to a more diverse range of production systems in ways that overcome current barriers, including labour and time costs associated with manure application (Hou et al., 2013) and the lack of characterisation of the nutrient content of manures (Chadwick et al., 2015). Currently, such barriers result in no manure application to cropland in Huantai county. More effective use of the substantial nutrient stocks within agricultural soils of Huantai county will require an effective soil sampling and analysis programme, coupled with an ability to modify inorganic fertiliser recommendations and the composition of compound fertilisers in ways that response to the spatial heterogeneity of soil nutrient supply (Sharpley et al., 2013). Despite these challenges, it is clear that a more integrated framework for nutrient management in Huantai county, accounting for all forms of nutrient supply, would help to deliver future increases in NUE. Examples of such integrated nutrient management frameworks exist in countries beyond China (e.g Defra, 2010) and could provide the basis for the development of a parallel framework that reflects the specific conditions within China. However, more importantly there is existing work in China on, for example, integrated soil-crop system management (Chen et al., 2014) or the use of a nutrient decision support system (Chuan et al., 2013) as well as more generic recommendations on nutrient and manure management (Bai et al., 2016; Chen et al., 2017; Yan et al., 2013).

4.4 Policy implications and conclusions

The results reported above demonstrate the value of analysing nutrient management practices over time, for different agricultural production systems and with varying spatial resolution. For intensive production systems and large areas representative of the North China Plain, use of inorganic fertilisers, manures and crop residues have been shown to remain inefficient in many cases and to present risks to the environment. National-scale SFAs are useful to identify aggregate trends and any need for policy reform at the highest level. However, the research reported here also demonstrates that drawing from data readily available for most counties in China, an SFA can provide important insights into nutrient management practices at more local scales to inform county-level administrators and technicians. More detail than was presented here can be drawn from the SFA depending on the requirements of county officials. Making use of this kind of data will also give individuals and organisations a means to monitor the impact of any policy and practice changes that may be implemented.

For policy in China, the headline message is the need to complete a shift from mobilisation of resources for production and growth, to management of resources for efficiency and sustainability. Improved approaches are needed in China to rebalance the importance of productivity with sustainable stewardship of farm inputs and natural resources (Smith et al., 2015). Evidence-based nutrient management strategies, farm advice and compound fertiliser formulations, all well-tailored to farming systems, farm characteristics and locations, can be seen as public goods (Bellarby et al., 2017), that will require coordinated development and delivery by the public extension system, research institutes, local government and input suppliers.

Use of multi-year monitoring and analyses will inform such strategies by reducing uncertainty and identifying the impacts of policies, trends or shocks. As agrarian structures and farming systems become more diversified and market-oriented in China, through further commercialisation and processes of land transfer (consolidation of land holdings through rental and transfer arrangements;

Smith and Siciliano, 2015), there is an increasing need for local solutions that reflect the heterogeneity of nutrient management requirements at sub-province, and even sub-county, scales. This need is illustrated, for example, by the contrasts in NUE between the wheat/maize and vegetable and fruit farming systems revealed for Huantai county in the SFAs reported above. As an increasingly commercialised farming sector responds to the changing patterns of food demand from a rapidly urbanising society, nutrient intensive crops and production systems will expand, particularly in peri-urban areas, and nutrient management plans and farm advice provision must become similarly dynamic and responsive. Integral to this, should be that localised and farming system-focused nutrient management plans better recognise and account for the crop nutrients exploitable in soil stocks, manures and crop residues. Soil nutrient stocks provide a key example of a localised resource that requires conservation and management for optimisation of productivity, sustainability and environmental protection. Achievement of the targeting and responsiveness suggested here will require further investment in capacities for monitoring, analyses and coordinated farmer advice provision. The latter will only be achieved through a variety of means, including print and digital media and on-farm trials and demonstrations (Smith et al., 2015).

Ideally there would be capacity to recognise and provide for soil nutrient status by farm or plot and how this varies depending on the historic input of nutrients. However, the number and fragmentation of farm holdings will remain at least a partial barrier to this for some time, although new remote sensing and other information technologies may increasingly find application. A successful knowledge transfer strategy is always associated with challenges even in less difficult areas (Rahn, 2013). Defining farmer types according to their farm system characteristics (e.g. farm size, land management practices, outputs) and social characteristics (e.g. age, income, education), and developing a tailored approach for each category may address the need for individual advice within current practical limitations. This is a task for county Agricultural Bureaus in China, and it is appropriate to continue to concentrate public resources for research and planning at county and sub-county levels, whilst evaluating alternative approaches, the most successful of which can then be adapted for wider implementation in China.

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6 References

Bai, Z.H., Ma, L., Oenema, O., Chen, Q., Zhang, F.S., 2013. Nitrogen and Phosphorus Use Efficiencies in Dairy Production in China. J. Environ. Qual. 42, 990. doi:10.2134/jeq2012.0464

Bai, Z., Ma, L., Jin, S., Ma, W., Velthof, G.L., Oenema, O., Liu, L., Chadwick, D., and Zhang, F. 2016. Nitrogen, Phosphorus, and Potassium Flows through the Manure Management Chain in China. Environ. Sci. Technol. 50, 13409–13418. doi: 10.1021/acs.est.6b03348

Bellarby, J., Siciliano, G., Smith, L.E.D., Xin, L., Zhou, J., Liu, K., Jie, L., Meng, F., Inman, A., Rahn, C., Surridge, B., Haygarth, P.M., 2017. Strategies for sustainable nutrient management: insights from a mixed natural and social science analysis of Chinese crop production systems. Environ. Dev. 21, 52–65. doi:10.1016/j.envdev.2016.10.008

Bellarby, J., Surridge, B., Haygarth, P.M., Lai, X., Zhang, G., Song, X., Zhou, J., Meng, F., Shen, J., Rahn, C., Burke, S., Smith, L., Siciliano, G., 2015. Inefficiency and environmental risks associated with nutrient use in agriculture within China and the UK.

Bouwman, A.F., Boumans, L.J.M., Batjes, N.H., 2002. Estimation of global NH_3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. Glob. Biogeochem. Cycles 16, 8–1. doi:10.1029/2000GB001389

Cai, G.X., Chen, D.L., Ding, H., Pacholski, A., Fan, X.H., Zhu, Z.L., 2002. Nitrogen losses from fertilizers applied to maize, wheat and rice in the North China Plain. Nutr. Cycl. Agroecosystems 63, 187–195. doi:10.1023/A:1021198724250

Chadwick, D., Wei, J., Yan'an, T., Guanghui, Y., Qirong, S., Qing, C., 2015. Improving manure nutrient management towards sustainable agricultural intensification in China. Agric. Ecosyst. Environ. 209, 34–46. doi:10.1016/j.agee.2015.03.025

Chen, S., Wu, W., Hu, K., Li, W., 2010. The effects of land use change and irrigation water resource on nitrate contamination in shallow groundwater at county scale. Ecol. Complex., Eco Summit 2007 Special Issue, Part Two 7, 131–138. doi:10.1016/j.ecocom.2010.03.003

Chen, S., Yan, Z., and Chen, Q. 2017. Estimating the potential to reduce potassium surplus in intensive vegetable fields of China. Nutr Cycl Agroecosyst 107, 265–277. doi: 10.1007/s10705-017-9835-0

Chen, X., Cui, Z., Fan, M., Vitousek, P., Zhao, M., Ma, W., Wang, Zhenlin, Zhang, Weijian, Yan, X., Yang, J., Deng, X., Gao, Q., Zhang, Q., Guo, S., Ren, J., Li, S., Ye, Y., Wang, Zhaohui, Huang, J., Tang, Q., Sun, Y., Peng, X., Zhang, J., He, M., Zhu, Y., Xue, J., Wang, G., Wu, Liang, An, N., Wu, Liangquan, Ma, L., Zhang, Weifeng, Zhang, F., 2014. Producing more grain with lower environmental costs. Nature advance online publication. doi:10.1038/nature13609

Chowdhury, R.B., Moore, G.A., Weatherley, A.J., Arora, M., 2014. A review of recent substance flow analyses of phosphorus to identify priority management areas at different geographical scales. Resour. Conserv. Recycl. 83, 213–228. doi:10.1016/j.resconrec.2013.10.014

Chuan, L., He, P., Pampolino, M.F., Johnston, A.M., Jin, J., Xu, X., Zhao, S., Qiu, S., and Zhou, W. 2013. Establishing a scientific basis for fertilizer recommendations for wheat in China: Yield response and agronomic efficiency. Field Crops Research 140, 1–8. doi: 10.1016/j.fcr.2012.09.020

Cooper, J., Carliell-Marquet, C., 2013. A substance flow analysis of phosphorus in the UK food production and consumption system. Resour. Conserv. Recycl. 74, 82–100. doi:10.1016/j.resconrec.2013.03.001

Cordell, D., White, S., 2014. Life's Bottleneck: Sustaining the World's Phosphorus for a Food Secure Future. Annu. Rev. Environ. Resour. 39, 161–188. doi:10.1146/annurev-environ-010213-113300

Dai, X., Ouyang, Z., Li, Y., Wang, H., 2013. Variation in Yield Gap Induced by Nitrogen, Phosphorus and Potassium Fertilizer in North China Plain. PLoS ONE 8, e82147. doi:10.1371/journal.pone.0082147

Defra, 2010. Fertiliser Manual (RB209) 8th Edition.

FAOSTAT, 2014. FAOSTAT.

Foley, J.A., Ramankutty, N., Brauman, K.A., Cassidy, E.S., Gerber, J.S., Johnston, M., Mueller, N.D., O'Connell, C., Ray, D.K., West, P.C., Balzer, C., Bennett, E.M., Carpenter, S.R., Hill, J., Monfreda, C., Polasky, S., Rockström, J., Sheehan, J., Siebert, S., Tilman, D., Zaks, D.P.M., 2011. Solutions for a cultivated planet. Nature 478, 337–342. doi:10.1038/nature10452

Gao, J.J., Bai, X.L., Zhou, B., Zhou, J.B., Chen, Z.J., 2012. Soil nutrient content and nutrient balances in newly-built solar greenhouses in northern China. Nutr. Cycl. Agroecosystems 94, 63–72. doi:10.1007/s10705-012-9526-9

Ha, N., Feike, T., Back, H., Xiao, H., Bahrs, E., 2015. The effect of simple nitrogen fertilizer recommendation strategies on product carbon footprint and gross margin of wheat and maize production in the North China Plain. J. Environ. Manage. 163, 146–154. doi:10.1016/j.jenvman.2015.08.014

Hartmann, T.E., Yue, S., Schulz, R., Chen, X., Zhang, F., Müller, T., 2014. Nitrogen dynamics, apparent mineralization and balance calculations in a maize – wheat double cropping system of the North China Plain. Field Crops Res. 160, 22–30. doi:10.1016/j.fcr.2014.02.014

Haygarth, P.M., Jarvie, H.P., Powers, S.M., Sharpley, A.N., Elser, J.J., Shen, J., Peterson, H.M., Chan, N.-I., Howden, N.J.K., Burt, T., Worrall, F., Zhang, F., Liu, X., 2014. Sustainable Phosphorus Management and the Need for a Long-Term Perspective: The Legacy Hypothesis. Environ. Sci. Technol. 48, 8417–8419. doi:10.1021/es502852s

Hazell, P.B.R., 2009. The Asian Green Revolution. IFPRI Discussion Paper.

Hou, Y., Ma, L., Gao, Z.L., Wang, F.H., Sims, J.T., Ma, W.Q., Zhang, F.S., 2013. The Driving Forces for Nitrogen and Phosphorus Flows in the Food Chain of China, 1980 to 2010. J. Environ. Qual. 42, 962. doi:10.2134/jeq2012.0489

Huang, J., Ridoutt, B.G., Zhang, H., Xu, C., and Chen, F. 2014. Water Footprint of Cereals and Vegetables for the Beijing Market. Journal of Industrial Ecology 18, 40–48. doi: 10.1111/jiec.12037

Huantai Agricultural Bureau, 2014. Huantai Yearbook of Agriculture.

Huantai Agricultural Bureau, 1993. Huantai Yearbook of Agriculture.

Huantai Agricultural Bureau, 1990. Annual wheat and maize production manual in Huantai.

Jia, W., Li, Y., Chen, Q., David, C., 2014. Analysis of nutrient resources in livestock manure excretion and its potential of fertilizers substitution in Beijing suburbs. Trans. Chin. Soc. Agric. Eng. 30, 156–167.

Ju, X., Liu, X., Zhang, F., Roelcke, M., 2004. Nitrogen Fertilization, Soil Nitrate Accumulation, and Policy Recommendations in Several Agricultural Regions of China. AMBIO J. Hum. Environ. 33, 300–305. doi:10.1579/0044-7447-33.6.300

Ju, X.-T., Xing, G.-X., Chen, X.-P., Zhang, S.-L., Zhang, L.-J., Liu, X.-J., Cui, Z.-L., Yin, B., Christie, P., Zhu, Z.-L., Zhang, F.-S., 2009. Reducing environmental risk by improving N management in intensive Chinese agricultural systems. Proc. Natl. Acad. Sci. 106, 3041–3046. doi:10.1073/pnas.0813417106

Lehmann, J., Schroth, G., 2003. Nutrient Leaching, in: Trees, Crops and Soil Fertility (Eds. G. Schroth and F.L. Sinclair), CAB International. CAB International.

Li, Y., Zhang, W., Ma, L., Huang, G., Oenema, O., Zhang, F., Dou, Z., 2013. An Analysis of China's Fertilizer Policies: Impacts on the Industry, Food Security, and the Environment. J. Environ. Qual. 42, 972–981. doi:10.2134/jeq2012.0465

Liao, Y., Wu, W.L., Meng, F.Q., Smith, P., Lal, R., 2014. Increase in soil organic carbon by agricultural intensification in northern China. Biogeosciences Discuss 11, 16497–16525. doi:10.5194/bgd-11-16497-2014

Liu, G.D., Wu, W.L., Zhang, J., 2005. Regional differentiation of non-point source pollution of agriculture-derived nitrate nitrogen in groundwater in northern China. Agric. Ecosyst. Environ. 107, 211–220. doi:10.1016/j.agee.2004.11.010

Lu, Y., Chen, Z., Kang, T., Zhang, X., Bellarby, J., Zhou, J., 2016. Land-use changes from arable crop to kiwi-orchard increased nutrient surpluses and accumulation in soils. Agric. Ecosyst. Environ. 223, 270–277. doi:10.1016/j.agee.2016.03.019

Lu, Y., Kang, T., Zhang, X., Gao, J., Chen, Z., Zhou, J., 2014. Evaluation of current fertilization status in kiwifruit orchards on the norther slope of Qinling Mountains: A case study of Yujiahe catchment, in Zhouzi County. Plant Nutr. Fertil. Sci. Chin.

Ma, L., Ma, W.Q., Velthof, G.L., Wang, F.H., Qin, W., Zhang, F.S., Oenema, O., 2010. Modeling nutrient flows in the food chain of China. J. Environ. Qual. 39, 1279–1289.

Ma, L., Velthof, G.L., Wang, F.H., Qin, W., Zhang, W.F., Liu, Z., Zhang, Y., Wei, J., Lesschen, J.P., Ma, W.Q., Oenema, O., Zhang, F.S., 2012. Nitrogen and phosphorus use efficiencies and losses in the food chain in China at regional scales in 1980 and 2005. Sci. Total Environ. 434, 51–61. doi:10.1016/j.scitotenv.2012.03.028

Meng, F., Wu, W., Xin, D., 2000. Changes of soil organic matter and nutrients and their relationship with crop yield in high yield farmland. (In Chinese). Plant Nutr. Fertil. Sci. 6, 370–374.

Miao, Y., Stewart, B.A., Zhang, F., 2011. Long-term experiments for sustainable nutrient management in China. A review. Agron. Sustain. Dev. 31, 397–414. doi:10.1051/agro/2010034

MOA, 2009. The handbook of pollution discharge parameters of livestock production for the 1st National Pollution Sources Investigation.

Moll, R.H., Kamprath, E.J., Jackson, W.A., 1982. Analysis and Interpretation of Factors Which Contribute to Efficiency of Nitrogen Utilization. Agron. J. 74, 562–564. doi:10.2134/agronj1982.00021962007400030037x

Némery, J., Garnier, J., Morel, C., 2005. Phosphorus budget in the Marne Watershed (France): urban vs. diffuse sources, dissolved vs. particulate forms. Biogeochemistry 72, 35–66. doi:10.1007/s10533-004-0078-1

OECD, 2007. Environmental Performance Reviews: China.

Ongley, E.D., Xiaolan, Z., Tao, Y., 2010. Current status of agricultural and rural non-point source Pollution assessment in China. Environ. Pollut. 158, 1159–1168. doi:10.1016/j.envpol.2009.10.047

Peng, C., Luo, H., Kong, J., 2014. Advance in estimation and utilisation of crop residues resources in China. Chinese Journal of Agricultural Resources and Regional Planning 35, 14–20.

Rahn, C., 2013. The challenges of knowledge transfer in the implementation of the Nitrate Directive in Proceedings of NUTRIHORT Nutrient management, innovative techniques and nutrient legislation in intensive horticulture for an improved water quality Gent 16 – 18 September 2013. https://iris.unito.it/retrieve/handle/2318/144228/338564/2013_NUTRIHORT_Nicola_2-8_Proceedings.pdf [WWW Document]. URL https://www.researchgate.net/publication/261595745_RAHN_C_2013_The_challenges_of_knowledge_transfer_in_the_implementation_of_the_Nitrate_Directive_in_Proceedings_of_NUTRIHORT_Nutrient_management_innovative_techniques_and_nutrient_legislation_in_intens (accessed 11.18.16).

Senthilkumar, K., Nesme, T., Mollier, A., Pellerin, S., 2012. Conceptual design and quantification of phosphorus flows and balances at the country scale: The case of France. Glob. Biogeochem. Cycles 26, GB2008. doi:10.1029/2011GB004102

Sharpley, A., Jarvie, H.P., Buda, A., May, L., Spears, B., Kleinman, P., 2013. Phosphorus Legacy: Overcoming the Effects of Past Management Practices to Mitigate Future Water Quality Impairment. J. Environ. Qual. 42, 1308. doi:10.2134/jeq2013.03.0098

Sheldrick, W.F., Syers, J.K., Lingard, J., 2003. Soil nutrient audits for China to estimate nutrient balances and output/input relationships. Agric. Ecosyst. Environ. 94, 341–354. doi:10.1016/S0167-8809(02)00038-5

Shepherd, A., Yan, X., Nayak, D., Newbold, J., Moran, D., Dhanoa, M.S., Goulding, K., Smith, P., Cardenas, L.M., 2015. Disaggregated N_2O emission factors in China based on cropping parameters create a robust approach to the IPCC Tier 2 methodology. Atmos. Environ. 122, 272–281. doi:10.1016/j.atmosenv.2015.09.054

Smith, L., Siciliano, G., Inman, A., Rahn, C., Bellarby, J., Surridge, B., Haygarth, P.M., Zhang, G., Li, J., Zhou, J., Meng, F., Burke, S., 2015. Delivering improved nutrient stwardship in China: the knowledge, attitudes and practices of farmers and advisers - Policy Brief No. 13.

Smith, L.E.D., Siciliano, G., 2015. A comprehensive review of constraints to improved management of fertilizers in China and mitigation of diffuse water pollution from agriculture. Agric. Ecosyst. Environ., Sustainable intensification of China's agriculture: the key role of nutrient management and climate change mitigation and adaptation 209, 15–25. doi:10.1016/j.agee.2015.02.016

Stehfest, E., Bouwman, L., 2006. N_2O and NO emission from agricultural fields and soils under natural vegetation: summarizing available measurement data and modeling of global annual emissions. Nutr. Cycl. Agroecosystems 74, 207–228. doi:10.1007/s10705-006-9000-7

Strokal, M., Ma, L., Bai, Z., Luan, S., Kroeze, C., Oenema, O., Velthof, G., Zhang, F., 2016. Alarming nutrient pollution of Chinese rivers as a result of agricultural transitions. Environ. Res. Lett. 11, 024014. 10.1088/1748-9326/11/2/024014

Sun, B., Zhang, L., Yang, L., Zhang, F., Norse, D., Zhu, Z., 2012. Agricultural non-point source pollution in China: causes and mitigation measures. Ambio 41, 370–379. doi:10.1007/s13280-012-0249-6

Velthof, G.L., Oudendag, D., Witzke, H.P., Asman, W.A.H., Klimont, Z., Oenema, O., 2009. Integrated Assessment of Nitrogen Losses from Agriculture in EU-27 using MITERRA-EUROPE. J. Environ. Qual. 38, 402. doi:10.2134/jeq2008.0108

Vinther, F.P. 2005. SimDen – A simple empirical model for quantification of N_2O emission and denitrification. Paper at: Manure - an agronomic and environmental challenge. NJF-seminar no. 372, Nils Holgerssongymnasiet, Skurup, Sweden. Abstract of oral presentation, 5-6 September 2005.

Vitousek, P.M., Naylor, R., Crews, T., David, M.B., Drinkwater, L.E., Holland, E., Johnes, P.J., Katzenberger, J., Martinelli, L.A., Matson, P.A., Nziguheba, G., Ojima, D., Palm, C.A., Robertson, G.P., Sanchez, P.A., Townsend, A.R., Zhang, F.S., 2009. Nutrient Imbalances in Agricultural Development. Science 324, 1519–1520. doi:10.1126/science.1170261

Wang, F., Sims, J.T., Ma, L., Ma, W., Dou, Z., Zhang, F., 2011. The phosphorus footprint of China's food chain: implications for food security, natural resource management, and environmental quality. J. Environ. Qual. 40, 1081–1089. doi:10.2134/jeq2010.0444

Wang, L., Butcher, A.S., Stuart, M.E., Gooddy, D.C., and Bloomfield, J.P. 2013a. The nitrate time bomb: a numerical way to investigate nitrate storage and lag time in the unsaturated zone. Environmental Geochemistry and Health *35*, 667–681. doi: 10.1007/s10653-013-9550-y

Wang, X., Ma, H.Y., Ni, X.M., 2013b. Water saving agriculture operation and management in well irrigation regions of North China Plain. China Rural Water Hydropower 7, 47–49.

Webb, J., and Misselbrook, T.H. 2004. A mass-flow model of ammonia emissions from UK livestock production. Atmospheric Environment 38, 2163–2176. 10.1016/j.atmosenv.2004.01.023

World Bank, 2006. China Water Quality Management: policy and institutional considerations.

Xue, D., Pang, F., Meng, F., Wang, Z., Wu, W., 2015. Decision-tree-model identification of nitrate pollution activities in groundwater: A combination of a dual isotope approach and chemical ions. J. Contam. Hydrol. 180, 25–33.

Yan, Z., Liu, P., Li, Y., Ma, L., Alva, A., Dou, Z., Chen, Q., and Zhang, F. 2013. Phosphorus in China's Intensive Vegetable Production Systems: Overfertilization, Soil Enrichment, and Environmental Implications. Journal of Environmental Quality 42, 982–989. doi: 10.2134/jeq2012.0463

Yao, Z., Yan, G., Zheng, X., Wang, R., Liu, C., and Butterbach-Bahl, K. 2017. Straw return reduces yield-scaled N2O plus NO emissions from annual winter wheat-based cropping systems in the North China Plain. Science of The Total Environment *590*, 174–185. doi: 10.1016/j.scitotenv.2017.02.194

Zhang, X., Bol, R., Rahn, C., Xiao, G., Meng, F., Wu, W. 2017. Agricultural sustainable intensification improved nitrogen use efficiency and maintained high crop yield during 1980-2014 in Northern China. Sci. Total Environ. 596–597, 61–68. doi: 10.1016/j.scitotenv.2017.04.064

Zhang, X., Davidson, E.A., Mauzerall, D.L., Searchinger, T.D., Dumas, P., Shen, Y. 2015. Managing nitrogen for sustainable development. Nature 528, 51–59. doi: 10.1038/nature15743

Zhang, F., Cui, Z., Fan, M., Zhang, W., Chen, X., Jiang, R., 2011. Integrated Soil–Crop System Management: Reducing Environmental Risk while Increasing Crop Productivity and Improving Nutrient Use Efficiency in China. J. Environ. Qual. 40, 1051. doi:10.2134/jeq2010.0292

Zhen, L., Zoebisch, M.A., Chen, G., and Feng, Z. 2006. Sustainability of farmers' soil fertility management practices: A case study in the North China Plain. Journal of Environmental Management 79, 409–419. doi: 10.1016/j.jenvman.2005.08.009

Table 1: Nutrient use efficiencies (%) for different time periods across 1983-2014.

Crop	N			P			K		
	1983 - 1987	1996 - 2000	2010 - 2014	1983 - 1987	1996 - 2000	2010 - 2014	1983 - 1987	1996 - 2000	2010 - 2014
Wheat/maize	34	52	52	50	56	72	178	52	60
Other arable	18	14	5	23	6	6	110	25	12
Vegetable	38	45	57	31	63	47	200	62	87
Fruit	6	10	10	5	9	7	25	19	23
Overall crops	**32**	**47**	**51**	**44**	**53**	**66**	**166**	**52**	**61**
Livestock	22	42	22	9	15	9	7	11	7

Table 2: Total nutrient input in kg ha^{-1} for different time periods across 1983-2014.

Crop	N			P			K		
	1983 - 1987	1996 - 2000	2010 - 2014	1983 - 1987	1996 - 2000	2010 - 2014	1983 - 1987	1996 - 2000	2010 - 2014
Wheat/maize	494	630	684	63	108	94	41	250	296
Other arable	250	220	379	28	66	70	30	67	159
Vegetable	1316	1057	1676	414	198	514	190	420	643
Fruit	639	745	1115	99	142	216	116	335	375
Overall crops	460	654	702	63	118	105	43	259	301

Table 3: Fertiliser recommendations (FR), estimated[a] fertiliser input (FI), actual grain output (GO) and straw output (SO) for wheat production in Huantai county for the respective year (kg ha^{-1}).

Year	Item	N	P	K
2014	FR	232 - 246	39 - 45	100
	FI	212	41	87
	GO	166	28	33
	SO	50	6	82
2006	FR	222 - 273	52	100
	FI	217	38	71
	GO	171	29	34
	SO	48	5	79
2004	FR	207	56 – 67	116
	FI	252	52	78
	GO	159	27	32
	SO	46	5	76
1997	FR	214.5 – 288	59	75
	FI	257	54	78
	GO	172	30	34
	SO	52	6	85

[a] wheat fertiliser input has been estimated by assuming that half of the fertiliser applied on the wheat/maize system is applied on wheat.

Figure 1: Location of Huantai County in Shandong province within east China.

Figure 2: Area of land under four major agricultural production systems and livestock units (LU) for Huantai county from 1983 - 2014. Livestock units were calculated according to information given on the Eurostat website (http://epp.eurostat.ec.europa.eu/statistics_explained/index.php/Glossary:LSU) using the following conversion factors to get LUs: cattle = 1, sheep = 0.1, pigs = 0.3, broiler = 0.007, layers = 0.014, other poultry = 0.03, rabbit = 0.02. Data is available for each year for all date series but only marked by symbols for the wheat/maize and other arable crop system.

Figure 3: Conceptual design and nutrient flows for Huantai county detailing 5 agricultural systems wheat/maize, other arable crops, vegetable, fruit and livestock. All values are 5 year averages in t/year for the years 2010 – 2014, N (bold), P (normal) and K (cursive). The contribution to the input from the different sources is provided as a bar chart representing their % contribution. The output is termed as product for crops and livestock.

Figure 4: Total inflow (fertiliser, seed, N fixation, air deposition, irrigation, straw) and outflow (straw, grain and losses) of the soil in Huantai county. Outflow is the sum of products and losses to the environment. NB different y-axes scales for individual elements.

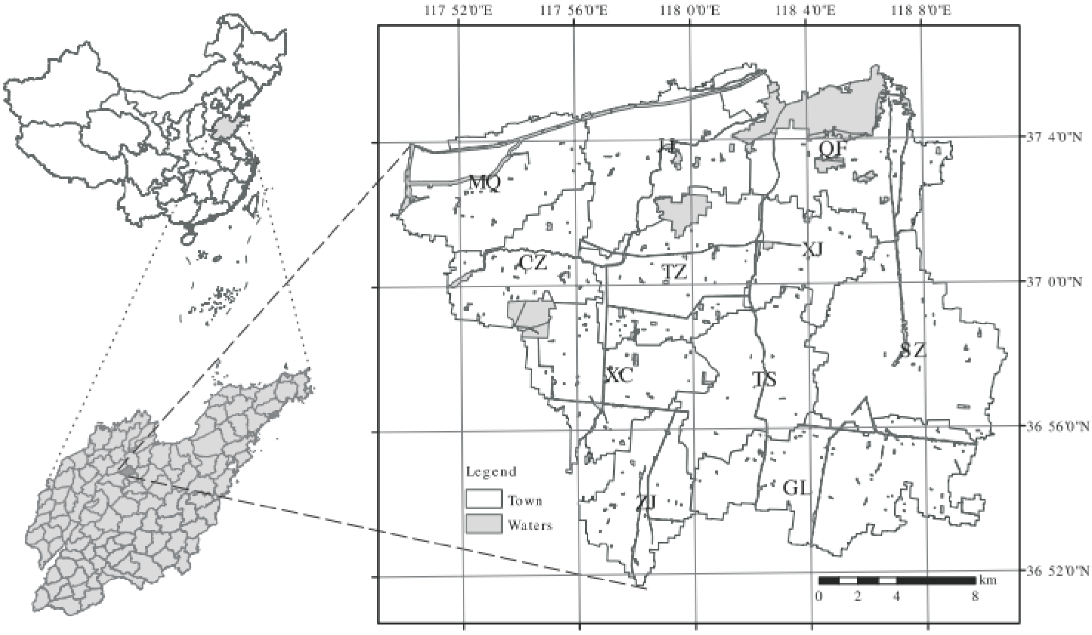
Figure 5: Nutrient flows of wheat/maize. All flows are presented separately here. NB different y-axes scales for individual elements.

Figure 6: Nutrient flows of other arable crop. All flows are presented separately here. NB different y-axes scales for individual elements.

Figure 7: Nutrient flows of vegetable. All flows are presented separately here. NB different y-axes scales for individual elements.

Figure 8: Nutrient flows of fruit. All flows are presented separately here. NB different y-axes scales for individual elements.

Figure 1.



117 52'0"E
117 56'0"E
118 0'0"E
118 4'0"E
118 8'0"E
37 4'0"N
37 0'0"N
36 56'0"N
36 52'0"N
MQ
JL
QF
CZ
TZ
XJ
XC
TS
SZ
GL
ZJ
Legend
Town
Waters
km
0 2 4 8

Figure 2.

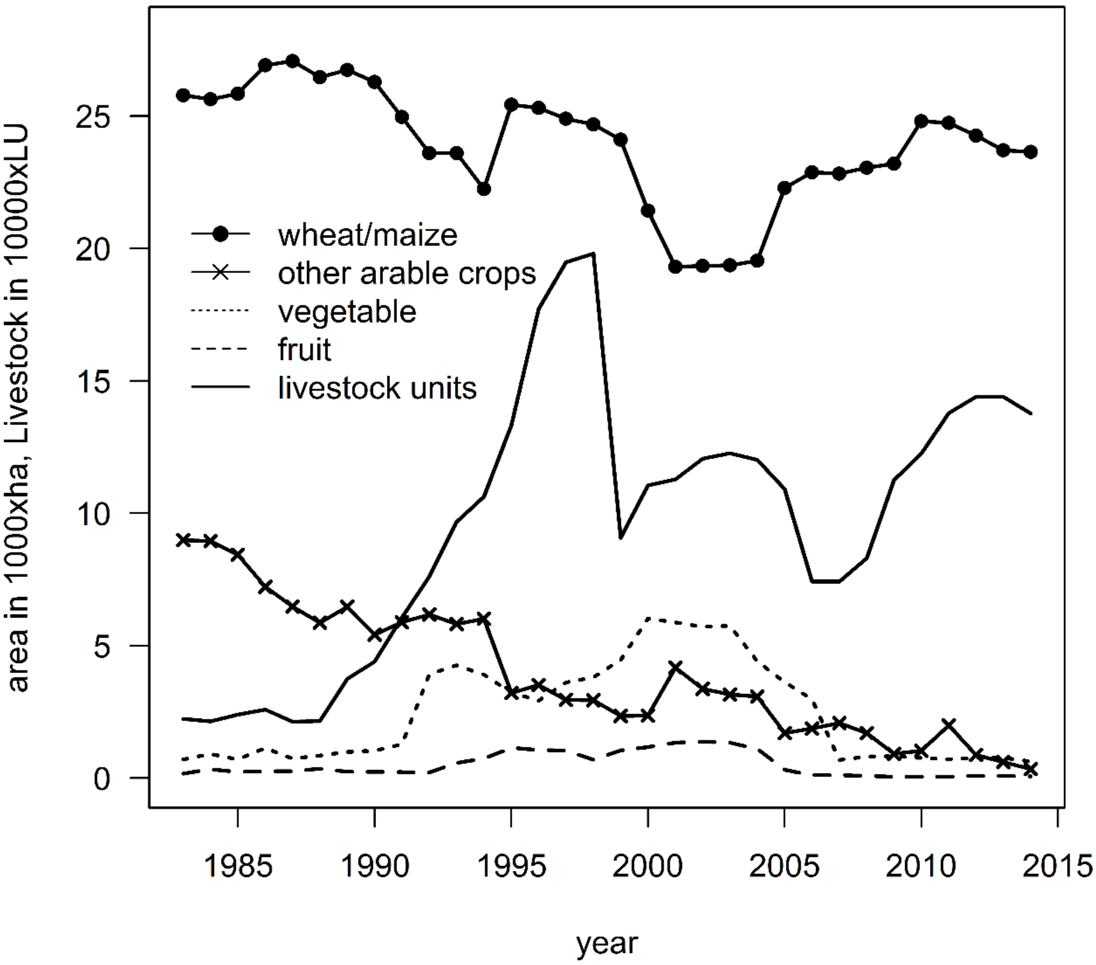
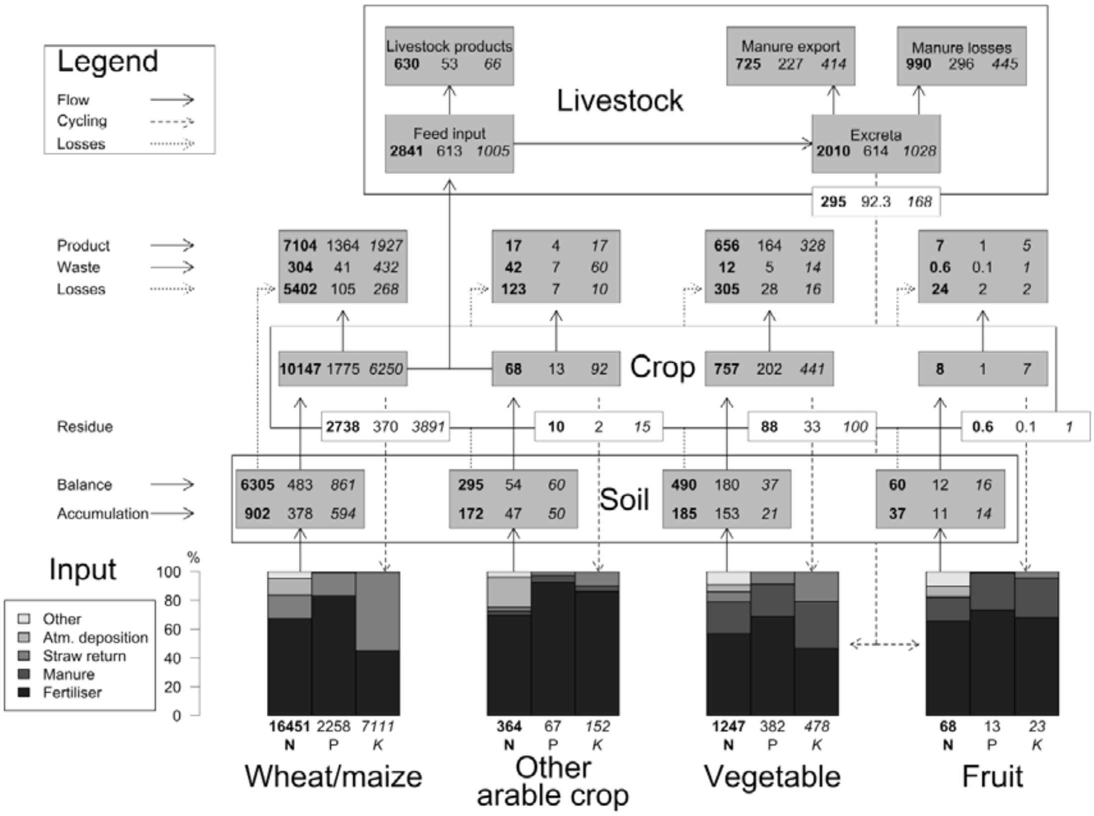

wheat/maize
other arable crops
vegetable
fruit
livestock units
area in 1000xha, Livestock in 10000xLU
year
0
5
10
15
20
25
1985
1990
1995
2000
2005
2010
2015

Figure 3.



Legend
Flow
Cycling
Losses
Livestock
Livestock products
630 53 66
Manure export
725 227 414
Manure losses
990 296 445
Feed input
2841 613 1005
Excreta
2010 614 1028
295 92.3 168
Product
Waste
Losses
7104 1364 1927
304 41 432
5402 105 268
17 4 17
42 7 60
123 7 10
656 164 328
12 5 14
305 28 16
7 1 5
0.6 0.1 1
24 2 2
Crop
10147 1775 6250
68 13 92
757 202 441
8 1 7
Residue
2738 370 3891
10 2 15
88 33 100
0.6 0.1 1
Balance
Accumulation
Soil
6305 483 861
902 378 594
295 54 60
172 47 50
490 180 37
185 153 21
60 12 16
37 11 14
Input
%
100
80
60
40
20
0
Other
Atm. deposition
Straw return
Manure
Fertiliser
16451 2258 7111
N P K
364 67 152
N P K
1247 382 478
N P K
68 13 23
N P K
Wheat/maize
Other arable crop
Vegetable
Fruit

Figure 4.

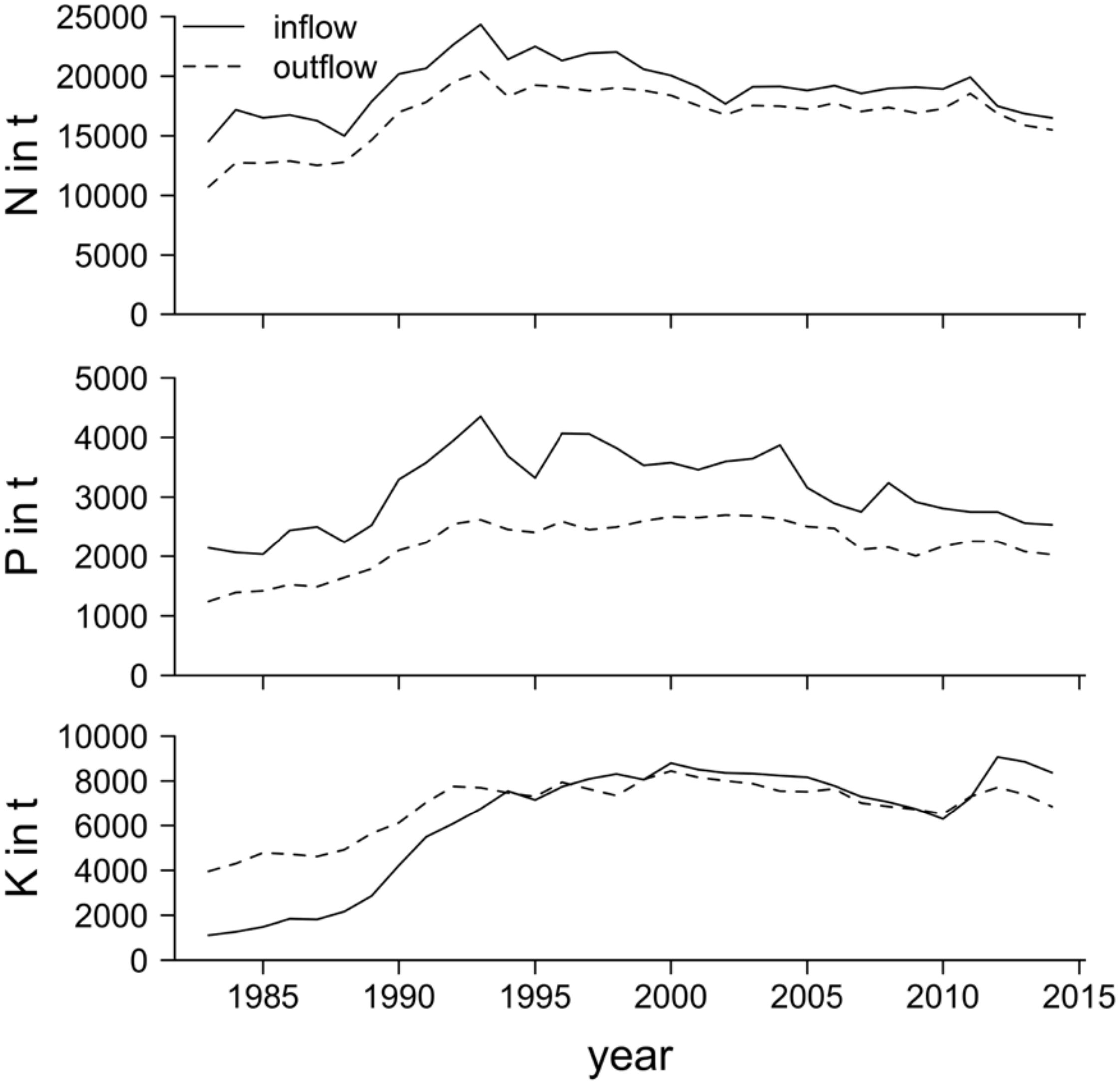
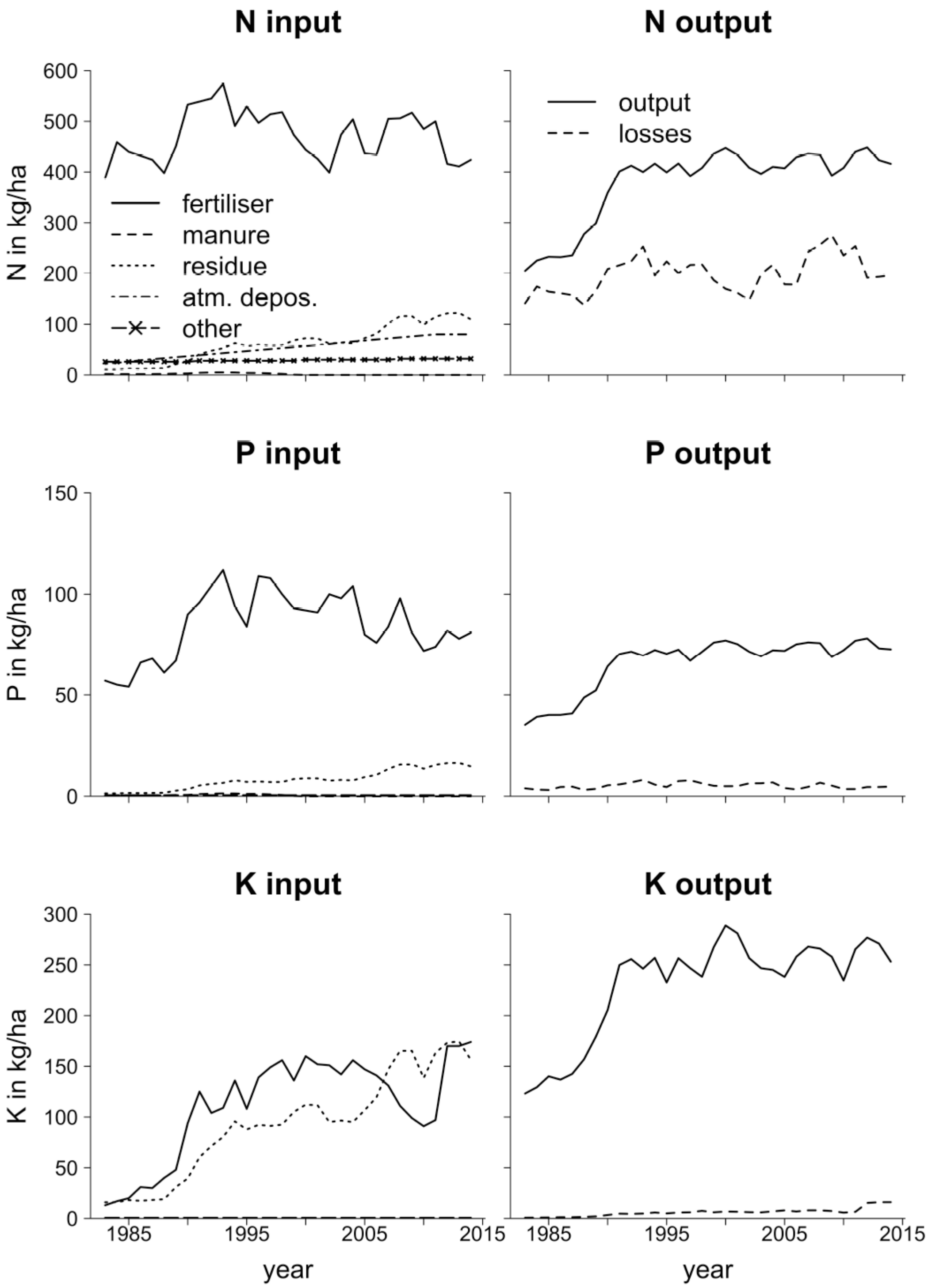

inflow
outflow
N in t
25000
20000
15000
10000
5000
0
P in t
5000
4000
3000
2000
1000
0
K in t
10000
8000
6000
4000
2000
0
1985
1990
1995
2000
2005
2010
2015
year

Figure 5.



N input
N output
P input
P output
K input
K output
N in kg/ha
P in kg/ha
K in kg/ha
fertiliser
manure
residue
atm. depos.
other
output
losses
year
year
0
100
200
300
400
500
600
0
50
100
150
0
50
100
150
200
250
300
1985
1995
2005
2015

Figure 6.

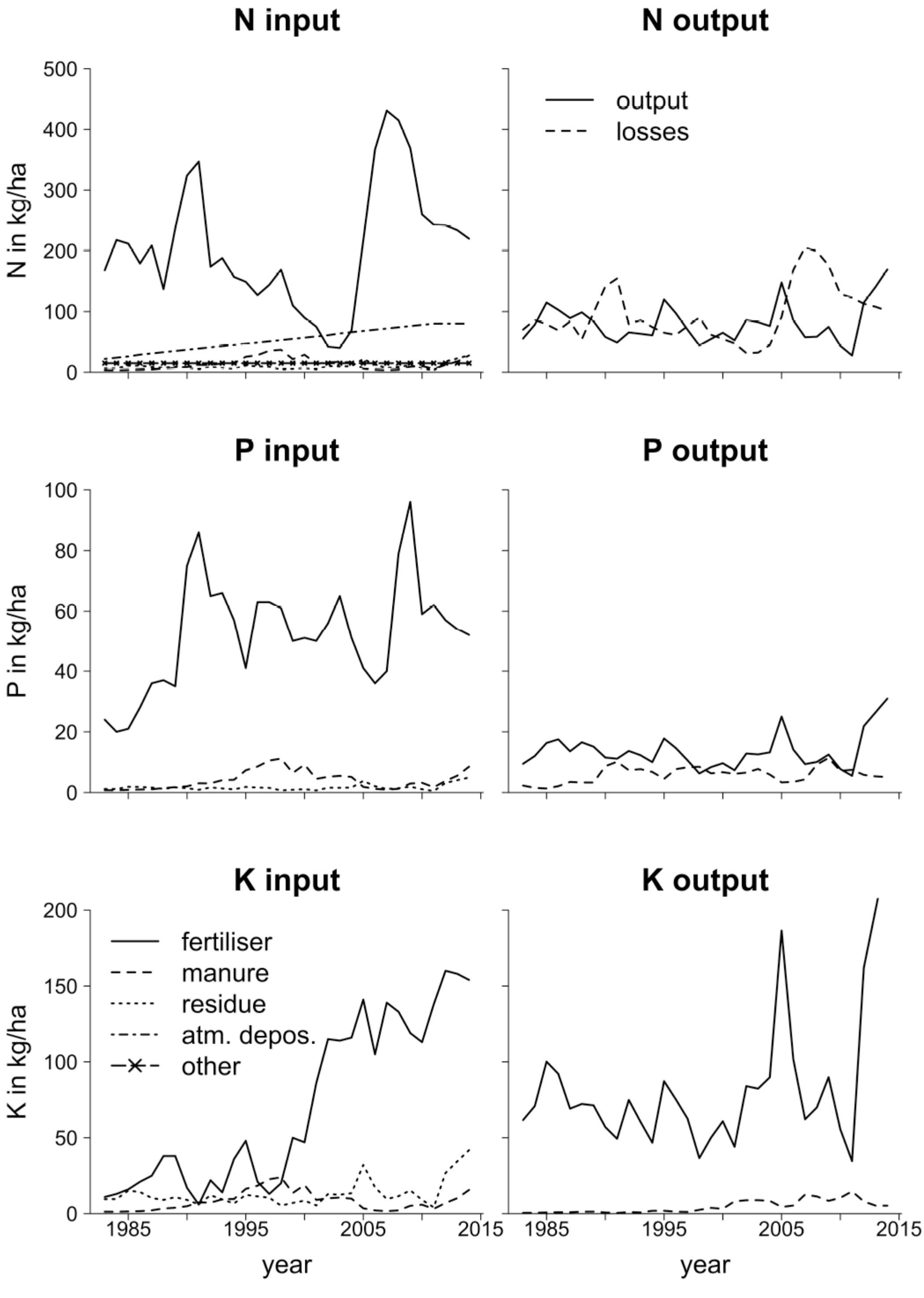

N input
N output
500
400
300
200
100
0
N in kg/ha
output
losses
P input
P output
100
80
60
40
20
0
P in kg/ha
K input
K output
200
150
100
50
0
K in kg/ha
fertiliser
manure
residue
atm. depos.
other
1985
1995
2005
2015
year

Figure 7.

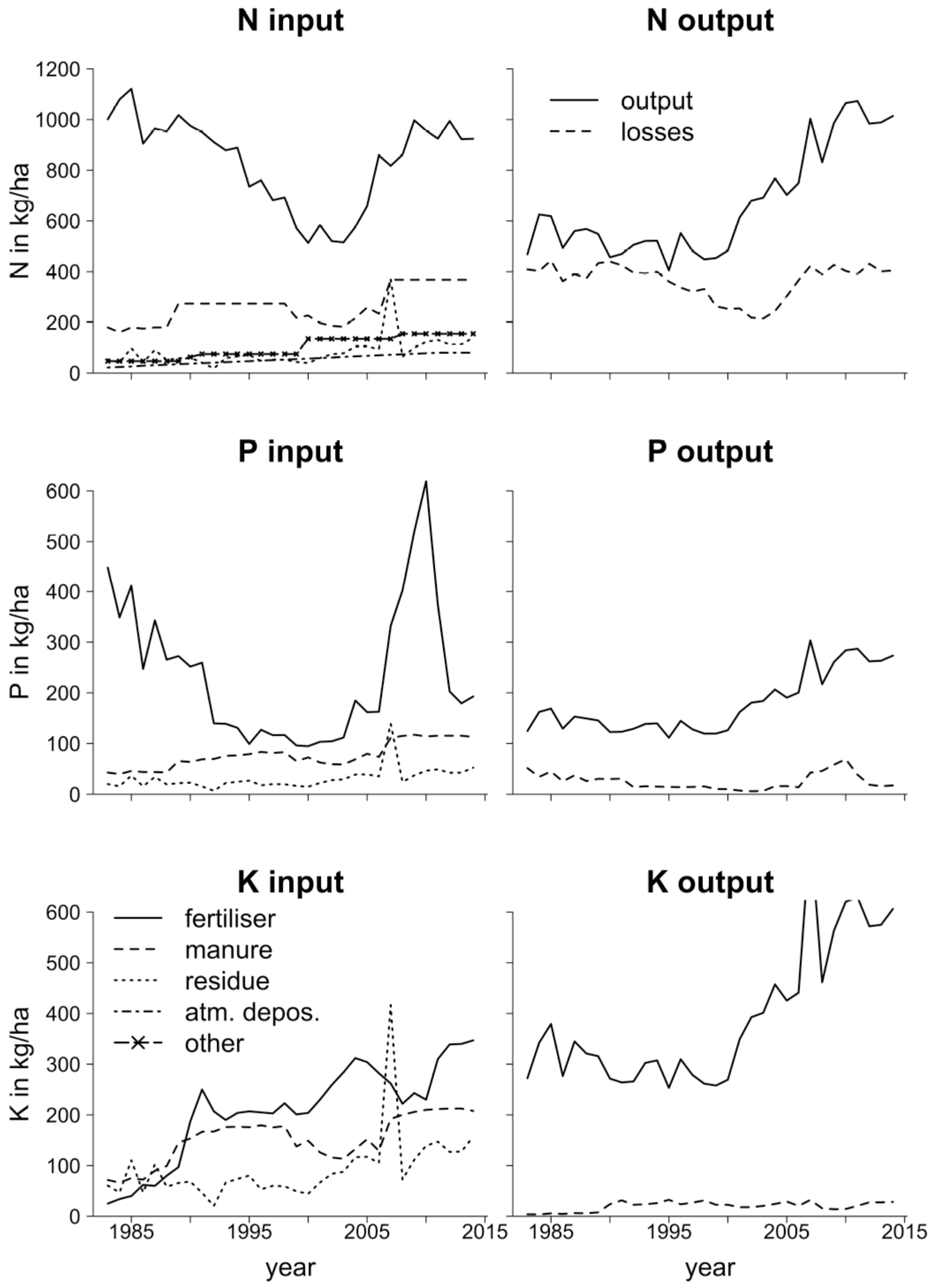
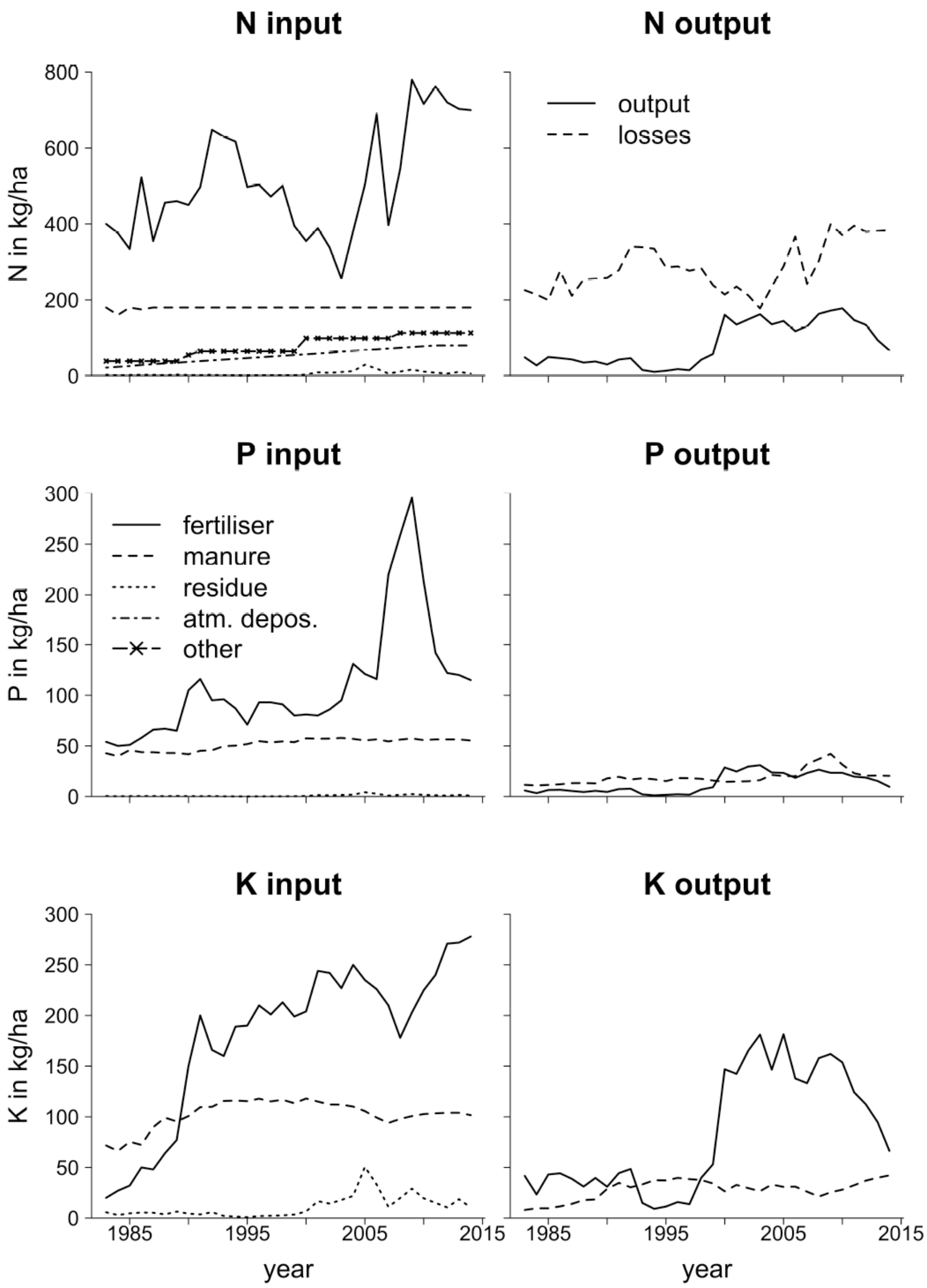

N input
N output
P input
P output
K input
K output
N in kg/ha
P in kg/ha
K in kg/ha
output
losses
fertiliser
manure
residue
atm. depos.
other
year
year
1985
1995
2005
2015

Figure 8.


N input
N output
P input
P output
K input
K output
N in kg/ha
P in kg/ha
K in kg/ha
800
600
400
200
0
300
250
200
150
100
50
output
losses
fertiliser
manure
residue
atm. depos.
other
1985
1995
2005
2015
year

The stocks and flows of nitrogen, phosphorus and potassium across a 30-year time series for agriculture in Huantai county, China – Supplementary Information

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Data and their sources for input and outputs

Table S1: Equations and data sources used to calculate nutrient flows in the analysis. "Other arable crops" are just referred to as "other" in this table for brevity.

Item No	Flow type	Item	Nutrient	Units, (calculations)	Data Source	Level of data certainty†
1	**Land area**	Wheat/maize, vegetable, fruits, other	NA	ha	a	***
	Input					
2	Fertiliser	Total county	N, P, K	T	a	***
3	Fertiliser	Wheat/maize	N, P, K	Kg/ha	a, b	**
4	Fertiliser	Fruit, vegetable, other	N, P, K	T (Item 2 – Item 3)	a	**
5	Fertiliser	Fruit and other	N, P	Kg/ha	a, b, c	*
6	Fertiliser	Fruit and vegetable	K	Kg/ha	a, b, c	*
7	Fertiliser	Vegetable	N, P	((Item 4 – (total input to other and fruit))/area of vegetable	a, b, c	*
8	Atmospheric deposition	Wheat/maize, other, vegetable, fruit	N	kg/ha (ranges from 16 – 80)	d, e	**
9	Irrigation water	Wheat/maize	N	In kg/ha (ranges from 8 – 14)	f, g	**
10	Irrigation water	Vegetable	N	between 1980 and 1990 4x, between 1990 and 2000 6x and between 2000 and 2014 10x the amount of irrigation for wheat/maize	f, g	**
11	Irrigation water	Fruit	N	between 1980 and 1990 3x, between 1990 and 2000 5x and between 2000 and 2014 7x the amount of irrigation for wheat/maize	f, g	**
12	Irrigation water	Other	N	No irrigation	f, g	**
13	Biological fixation	Wheat/maize, other, vegetable, fruit	N	15 kg/ha throughout	h	*
14	Seeds	Wheat/maize	N, P, K	Quantity of wheat and maize seed are 112.5 and 37.5 kg/ha, respectively.	b	***
	Output					
15	Crop uptake	Wheat, maize, vegetable, fruits, other		T of marketable product	a	***
16	Livestock uptake	Meat, milk, eggs		T of marketable product	a	***
17	Livestock uptake	Livestock head		Head of stock and head of livestock sold and slaughtered available	a	***

Level of data certainty†: * = low certainty, ** = medium certainty, *** = high certainty
[a] Huantai Agricultural Bureau, (2014), [b] Interview with local farmers and technicians, [c]Expert knowledge, [d]Zhang et al., (2006), [e]Liu *et al.*, (2013), [f]Liu and Wu, (2003), [g]Liu, (2016), [h]Zhu and Wen, (1992)

Table S2: Nutrient contents of all crops as percent air dried weight for wheat and maize and percent fresh weight for all other crops derived from NATESC, (1999).

System	Item	%N	%P	%K
Cereal	Maize grain	1.47	0.32	0.53
Cereal	Maize residue	0.87	0.13	1.11
Cereal	Wheat grain	2.16	0.37	0.43
Cereal	Wheat residue	0.62	0.07	1.02
Other	Cotton	1.67	0.42	1.92
Other	Cotton residue	1.1	0.2	1.7
Other	Peanut	3.7	0.215	0.65
Other	Peanut residue	1.3	0.13	0.8
Other	Potato	0.32	0.06	0.5
Other	Potato residue	0.99	0.09	0.67
Other	Soybean	5.696	0.465	1.503
Other	Soybean residue	1.2	0.17	1.2
Other	Sweet Potato	1.2	0.5	0.8
Other	Sweet Potato residue	1.2	0.3	1.33
Veg	Vegetable cop	0.6	0.15	0.3
Veg	Vegetable residue	0.4	0.15	0.45
Fruit	Apple	0.4	0.03	0.27
Fruit	Apricot	0.5	0.09	0.42
Fruit	Chinese date	0.6	0.09	0.58
Fruit	Fruit residue	0.7	0.10	1.20
Fruit	Grape	0.4	0.13	0.60
Fruit	Hawthorn	0.5	0.09	0.33
Fruit	Peach	0.58	0.09	0.45
Fruit	Pear	0.5	0.09	0.33

Table S3: Straw grain ratios of different crops to be applied on air dried weight for wheat and maize and fresh weight for all other crops as derived from Peng et al., (2014).

Crop	Residue to crop ratio
Maize	0.98
Wheat	1.06
Cotton	5
Peanut	1.2
Potato	1.2
Soybean	1.6
Sweet potato	1.2
Vegetables	0.225

Fruit	0.1

Table S4: Nutrient contents of fresh weight of livestock items.

Item	%N	%P	%K	Reference
Beef meat	3.15	0.17	0.22	1
Cow manure	0.39	0.08	0.23	2
Cattle manure	0.39	0.08	0.23	2
Dairy milk	0.48	0.07	0.11	1
Pig solid manure	0.55	0.24	0.29	2
Pig liquid manure	0.59	0.05	0.02	2
Pig meat	2.11	0.16	0.2	1
Duck egg	1.75	0.23	0.14	1
Duck manure	0.71	0.36	0.55	2
Duck meat	1.69	0.12	0.19	1
Broiler manure	0.9	0.41	0.73	2
Layer manure	1.03	0.41	0.72	2
Chicken egg	1.87	0.13	0.15	1,3
Chicken meat	2.04	0.156	0.251	1,3
Rabbit manure	0.87	0.3	0.65	2
Rabbit meat	3.15	0.29	0.37	1
Sheep manure	0.5	0.11	0.26	2
Sheep meat	2.74	0.15	0.23	1
Sheep milk	0.24	0.1	0.14	1

[1]Zhang, (2010); [2]NATESC, (1999); [3]Bai et al., (2013)

Table S5: Manure produced per animal and lifespan of livestock from MOA, (2009).

	kg manure per year	kg manure per day	Lifespan in days
Cow and cattle manure	8703		
Pig solid manure		2.44	180
Pig liquid manure		3.22	180
Duck manure		0.132	45
Broiler manure		0.1	42
Layer manure	42	0.12	500
Rabbit manure		0.159	90
Sheep manure	600		360

Recycling of nutrients

The amount of nutrients recycled back into the systems was partly based on the county yearbooks for crops (Table S7) with figures for wheat/maize different each year (Table S8). The amount of manure returned to soil is based on interviews with local farmers (Table S9).

Table S6: Percentage of residue returned to the field derived from Huantai Agricultural Bureau, (2014).

	% of residue returned
Cotton	20
Peanut	20
Potato	20
Soybean	20
Vegetable	88
Fruit	50

Table S7: Percentage of residue returned to the field, which were derived from Huantai Agricultural Bureau, (2014).

Year	% wheat straw returned to soil	% maize straw returned to soil
1983	20	17
1984	20	17
1985	20	17
1986	20	17
1987	20	17
1988	20	17
1989	30	21
1990	40	21
1991	50	24
1992	60	24
1993	70	25
1994	80	29
1995	90	27
1996	90	20
1997	90	16
1998	90	20
1999	90	27
2000	90	22
2001	90	23
2002	90	24
2003	90	30
2004	90	30
2005	90	50
2006	90	50
2007	90	70
2008	90	90
2009	90	90
2010	90	90
2011	90	90
2012	90	90
2013	90	90
2014	90	90

Table S8: Percentage of manure returned to the field, which were estimated based on interviews with local farmers.

Year	Vegetable	Fruit	Cropland	Other
1980	59.0	11.0	20.0	10.0
1981	61.3	9.7	19.0	10.0
1982	60.6	11.4	18.0	10.0
1983	58.8	14.2	17.0	10.0
1984	54.1	19.9	16.0	10.0
1985	56.2	18.8	15.0	10.0
1986	62.8	14.2	14.0	9.0
1987	58.0	21.0	13.0	8.0
1988	56.4	23.6	12.0	8.0
1989	65.9	16.1	11.0	7.0
1990	68.5	15.5	10.0	6.0
1991	72.3	12.7	9.0	6.0
1992	82.5	4.5	8.0	5.0
1993	77.6	10.4	7.0	5.0
1994	74.9	14.1	6.0	5.0
1995	67.2	23.8	5.0	4.0
1996	67.5	24.5	4.0	4.0
1997	72.3	20.7	3.0	4.0
1998	79.5	14.5	2.0	4.0
1999	76.9	18.1	1.0	4.0
2000	80.4	15.6	0.0	4.0
2001	78.2	17.8	0.0	4.0
2002	77.3	18.7	0.0	4.0
2003	77.9	18.1	0.0	4.0
2004	76.7	19.3	0.0	4.0
2005	91.2	7.8	0.0	1.0
2006	95.4	3.6	0.0	1.0
2007	85.2	13.0	0.0	1.0
2008	89.7	9.3	0.0	1.0
2009	93.4	5.6	0.0	1.0
2010	93.8	5.2	0.0	1.0
2011	92.1	6.9	0.0	1.0

Losses of nutrients

Table S9: Factor classes for the calculation of atmospheric N losses according to Bouwman et al., (2002); Stehfest and Bouwman, (2006) and Vinther (2005).

	Factor	Huantai	Factor used in equation	Equation
NH_4	**Fertiliser type**	Urea, compound fertiliser	0.14	Fertiliser application in t x 0.14
	Croptype	Other crop		
	pH	7.3 - 8.5		
	CEC	16 - 24		
	Climate[a]	Temperate		
	Application method	Broadcast		
N_2O	**SOC**	1 - 3	0.0038[b]	10^(total input in kg/ha * 0.0038 – 0.1088)
	pH	7.3 - 8.5		
	Texture	Medium		
	Climate[a]	Temperate continental		
	crop type	Cereals		
	N_2O background factor		- 0.1088	
NO	**Soil N content**	0.05-0.2	0.0061[b]	10^(total input in kg/ha * 0.0061 – 1.6942)
	Climate[a]	Temperate continental		
	NO background factor		- 1.6942	
N_2	**Soil type**	Clay loam	6.96	N_2O loss in t x 6.96
	SOM/precipitation	High		

[a]Classification according to Fischer et al., (1998); [b]only for cropland and other arable crops.

Table S10: Factor classes used for the calculation of aqueous losses based on Velthof et al., (2009) and Ma et al., (2010).

	Factor	Huantai	Factor used in equation
Leaching	**Soil type**	Loamy	0.1875
	Land use	Other	
	Minimum of other factors	Precipitation < 50mm	
Runoff	**Slope in %**	0-8	0.025
	Land use	Other	
	Minimum of other factors	Precipitation < 50mm	

Additional files

Table S12 lists all the files used in the calculation and production of figures. The R scripts have to be used in the order supplied as they build up on each other. Some explanations are provided within them.

Table S11: List of separately supplied files.

File name	Description
Originaldata19832014.csv	Collated original data
Parameters.csv	Parameters used in the calculation of derived figures
Loss_manure_parameters.csv	
Huantaibudget.txt	R script for N budget calculation
HuantaibudgetP.txt	R script for P budget calculation
HuantaibudgetK.txt	R script for K budget calculation
Huantai_budget_paper_figures_highres.txt	R script for plotting figures
Huantai_budget_average_for_SFA.txt	R script for making SFA figure in main manuscript
Huantai_budget_average_for_SFAmiddle.txt	R script for making SFA figure in supplemental material
Huantai_budget_average_for_SFAend.txt	R script for making SFA figure in supplemental material

Additional result figures

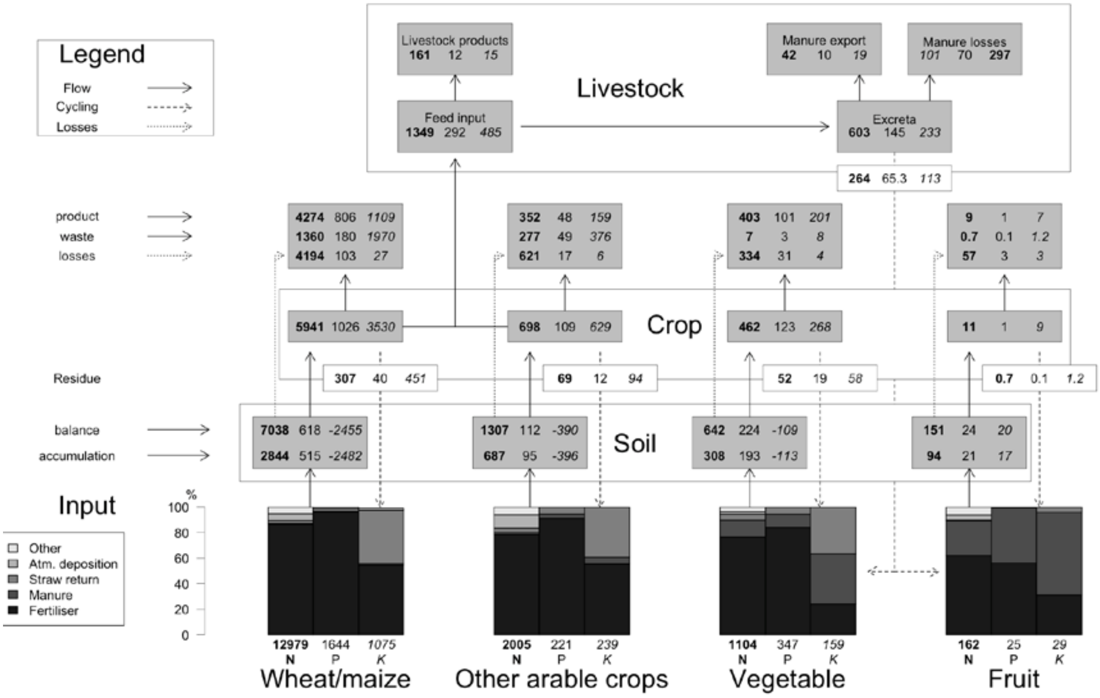

Figure 1: SFA of first 5 years

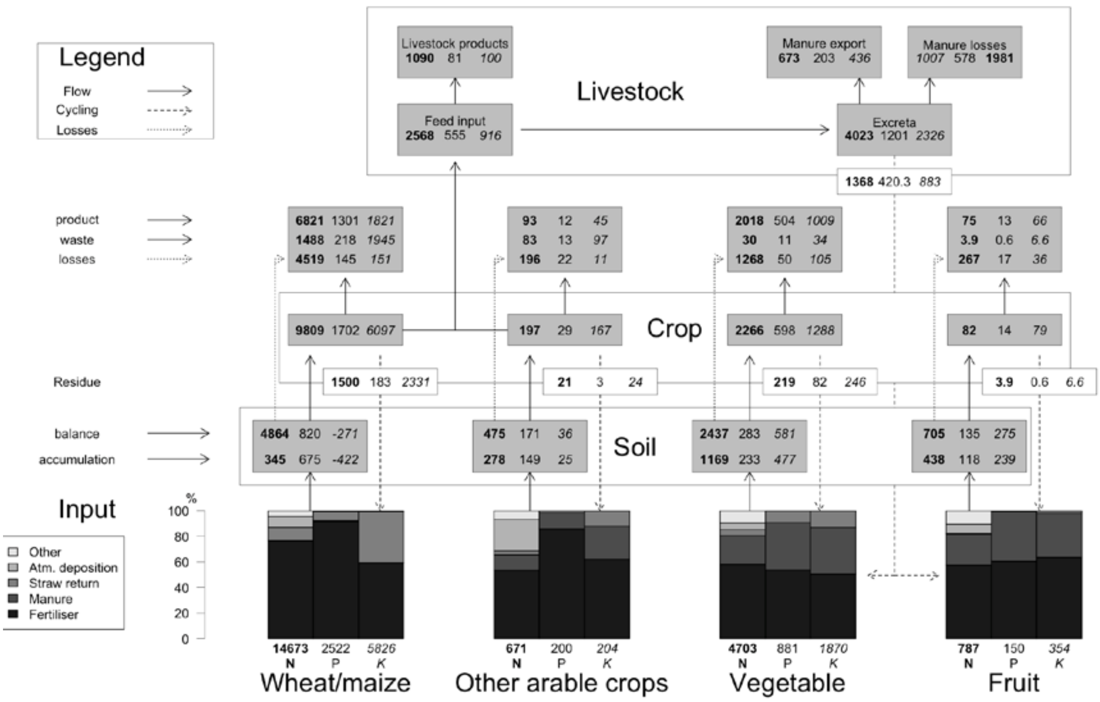

Figure 2: SFA between 1996 - 2001

References

Bai, Z.H., Ma, L., Oenema, O., Chen, Q., Zhang, F.S., 2013. Nitrogen and Phosphorus Use Efficiencies in Dairy Production in China. J. Environ. Qual. 42, 990. doi:10.2134/jeq2012.0464

Bouwman, A.F., Boumans, L.J.M., Batjes, N.H., 2002. Estimation of global NH_3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. Glob. Biogeochem. Cycles 16, 8–1. doi:10.1029/2000GB001389

Fischer, G., dePauw, E., van Velthuizen, H., Nachtergaele, F., Antoine, J., 1998. A provisional world climate resources inventory based on the length-of-growing-period concept.

Huantai Agricultural Bureau, 2014. Huantai Yearbook of Agriculture.

Liu, G., Wu, W., 2003. The dynamics of soil nitrate nitrogen leaching and contamination of the groundwater in high-yield farmland. Chin. J. Eco-Agric. 11, 91–93.

Liu, K., 2016. Sources of nitrogen in groundwater and influencing factors in intensive agricultural region of Norhtern Chine.

Liu, X., Zhang, Y., Han, W., Tang, A., Shen, J., Cui, Z., Vitousek, P., Erisman, J.W., Goulding, K., Christie, P., Fangmeier, A., Zhang, F., 2013. Enhanced nitrogen deposition over China. Nature 494, 459–462. doi:10.1038/nature11917

Ma, L., Ma, W.Q., Velthof, G.L., Wang, F.H., Qin, W., Zhang, F.S., Oenema, O., 2010. Modeling nutrient flows in the food chain of China. J. Environ. Qual. 39, 1279–1289.

MOA, 2009. The handbook of pollution discharge parameters of livestock production for the 1st National Pollution Sources Investigation.

NATESC, 1999. National Agro-tech Extension & Service Center, Nutrients database for organic fertilizers in China, Beijing, China Agriculture Press.

Peng, C., Luo, H., Kong, J., 2014. Advance in estimation and utilisation of crop residues resources in China. Chin. J. Agric. Resour. Reg. Plan. 35, 14–20.

Stehfest, E., Bouwman, L., 2006. N_2O and NO emission from agricultural fields and soils under natural vegetation: summarizing available measurement data and modeling of global annual emissions. Nutr. Cycl. Agroecosystems 74, 207–228. doi:10.1007/s10705-006-9000-7

Velthof, G.L., Oudendag, D., Witzke, H.P., Asman, W.A.H., Klimont, Z., Oenema, O., 2009. Integrated Assessment of Nitrogen Losses from Agriculture in EU-27 using MITERRA-EUROPE. J. Environ. Qual. 38, 402. doi:10.2134/jeq2008.0108

Vinther, F.P. 2005. SimDen – A simple empirical model for quantification of N_2O emission and denitrification. Paper at: Manure - an agronomic and environmental challenge. NJF-seminar no. 372, Nils Holgerssongymnasiet, Skurup, Sweden. Abstract of oral presentation, 5-6 September 2005.

Zhang, H., 2010. Nutrition parameters and feeding standards for animals (2nd edition). China Agriculture Press, Beijing.

Zhang, Y., Liu, X.., Zhang, F.S., Ju, X.T., Zou, G.Y., 2006. Spatial and temporal variation of atmospheric nitrogen deposition in North China Plain. Acta Ecol. Sin. 26, 1633–1639.

Zhu, Z., Wen, Q., 1992. Soil nitrogen in China. Jiangsu Press House, Nanjing.