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J. Le Noë[a,*], G. Billen[a], F. Esculier[a,b], J. Garnier[a]

[a] Milieux Environnementaux, Transferts et Interactions dans les hydrosystèmes et les Sols (METIS, UMR7619) – CNRS, Sorbonne Université- UPMC, EPHE, Case courrier 105, 4 place Jussieu, 75005 Paris, France

[b] Laboratoire Eau, Environnement et Systèmes Urbains (LEESU, UMR MA-102) AgroParisTech, École des Ponts ParisTech (ENPC), Université Paris-Est Marne-la-Vallée (UPEMLV) & Université Paris-Est Créteil Val-de-Marne (UPEC), 6-8 avenue Blaise Pascal, 77455 Champs sur Marne cedex 2, France

A B S T R A C T

We present a quantitative description of the N and P flows characterizing the agro-food system metabolism of 33 agricultural regions in France and their time evolution since the middle of the 19th century. The data were interpreted in terms of connection between crop production, livestock breeding, human nutrition and trade of agricultural goods, and were linked to their historical background. Until the early 20th century, the integrated crop and livestock farming model dominated everywhere, and the slow increase in crop production was only possible because of an increase in livestock density. Specialized cash crop farming systems appeared in the central Paris basin only in the first half of the 20th century together with the increase in the use of industrial fertilizers. Only after WWll, under the pressure of strong interventionist policies, did specialization of French territories lead to five types of systems, favoring their openness and integration into the international market, with harmful environmental impacts. The 1980s were marked by a policy shift towards more liberalism, which reinforced specialization. However, greater environmental concern stabilized or decreased nutrient losses, while maintaining largely open biogeochemical cycles.

1. Introduction

Today, agriculture is largely responsible for disturbing the nitrogen (N) and phosphorus (P) cycles, which Steffen et al. (2015) pointed out as one of the nine earth-system limits. These perturbations are mainly due to the shift from an organic agriculture closely coupling livestock and vegetal production to an industrial agriculture based on mineral fertilization and specialization that has separated crop and livestock farming. The transition from organic fertilization toward mineral-based fertilization was initially made possible by two historical discoveries: (i) the existence of phosphate deposits, in the form of guano in Peru first, and as phosphorus rock in Florida and Morocco thereafter (Clark and Foster, 2009; Pluvinage, 1912) and (ii) the invention of the Haber-Bosh process in 1913 (Smil, 2001). By the end of the 19th century, these new farming techniques responded to the problem of soil exhaustion, which had already raised concerns among scientists (e.g., Liebig) as early as 1840 (Foster, 2000). It also made it possible to better integrate agriculture into the market economy as yields increased with these new means of fertilization, while the need for closing the nutrient loop locally became less stringent. In many places, this implied the progressive specialization of agro-food systems over the course of the 20th century in order to remain competitive and maximize profits (Mazoyer and Roudart, 1998). In return, decoupling animal and vegetal production resulted in ever-increasing trade flows of agricultural products (Lassaletta et al., 2014a). The consequences for N and P cycles were a complete opening at the global scale (Smil, 2000; Galloway and Cowling, 2002) leading to the eutrophication of water streams (Carpenter, 2005), an increase in NH_3 emissions, which have dangerous side effects for both ecosystems and human health (Bouwman et al., 1997), and emission of N_2O, a powerful greenhouse gas (Bouwman, 1996). Furthermore, the opening of the P cycle also raised the issue of its exhaustion over the more or less long term (Elser and Bennett, 2011) given that it is a limited and nonrenewable resource over the human time scale. As an indispensable element to plant growth, restricted accessibility to P is likely to jeopardize the capacity of agro-food systems to produce enough food for a growing world population, which may generate geopolitical conflicts (Cordell et al., 2009).

In this context, a number of recent studies have focused on the socioecological metabolism of agro-food systems, applying material, nutrients or energy flow analysis approaches from local to global scales (Güldner and Krausmann, 2017; Guzmàn et al., 2017; Garnier et al., 2015, 2016; Soto et al., 2016; Billen et al., 2012). Analyses of

* Corresponding author.
E-mail address: julia.le_noe@upmc.fr (J. Le Noë).

socioecological metabolism conducted within a long-term perspective can shed light on the interrelation between human and natural history in the evolution of agro-ecosystems. Material and energy flows embedded in the functioning of agro-food systems are shaped by human activities of extraction, processing and consumption of biomass, but are also mediated by complex and dynamic natural processes such as ammonia volatilization, nitrate leaching and P sorption. Therefore, as stated by Gizicki-Neudlinger et al. (2017), agricultural systems, perhaps more than any other human activity, need to be considered as "historical phenomen[a] at the intersection of economy and ecology, as a hybrid between nature and culture."

However, to our knowledge few studies within this perspective have examined the case of France, with the exception of the recent article of Harchaoui and Chatzimpiros (2017), which analyzed the transformation of livestock breeding and its consequences over land use and agricultural trade in France over the 1961–2010 period. Yet, the national scale adopted in their study is blind to regional disparities and to the structural link between livestock breeding, arable land and grassland production. The case of France is noteworthy because this country is currently one of the main agricultural powers in Europe, the first exporter of cereals in 2014 (FAO, 2017), although the modernization of French agriculture had long lagged behind other Western European countries such as the United Kingdom, The Netherlands, Belgium and Germany (Ruttan, 1978). The late modernization of agriculture in France has been studied in great depth by historians (Jollivet, 2007; Duby and Wallon, 1993; Muller, 1984; Ruttan, 1978; Duby and Wallon, 1977) who highlighted the political and economic reasons for this delay. Nevertheless, the consequences of the particular evolution of French agriculture in terms of N and P cycles and environmental and agronomic performance are still lacking in the analysis. Therefore, the present research explored the co-evolution of the different patterns of agro-food systems in terms of N and P cycles in their historical context, applying the Generalized Representation of Agro-Food System (GRAFS) as developed by Billen et al. (2014) and Le Noë et al. (2017).

Studying the trajectories of agro-food systems from the perspective of N and P flows is an original prism for such analysis because at the same time it provides information on their degree of openness, their agronomic and environmental performance in terms of N and P use efficiency (NUE and PUE, i.e. the fraction of all N and P inputs that is effectively harvested), and the N and P balances. What are the trajectories that have led to the current agro-food systems at the regional scale? What were the main drivers of these trajectories? What are the environmental consequences? What are the legacies from the previous history on the current state of the system? Answering these questions contributes to responding to the general challenge for future sustainable agricultural production because (i) lessons can be drawn from the past given that traditional agricultural systems relied mainly on nutrient recycling at the farm and regional scale and (ii) designing future patterns of production will require accounting for the effect of agriculture development based on soil fertility, particularly P.

We therefore studied the period from 1852 to 2014. The mid-19th century corresponds to the very beginning of mineral fertilization in France (Boulaine, 2006) and also to the beginning of the Napoleonian Second Empire and the documentation of regular and detailed agricultural statistics. In this paper, some concrete findings regarding the evolution of agro-food system for three typical and contrasted French regions are first analyzed in order to capture the main characteristic of these systems. Based on this first analysis, a typology is then developed in order to objectify main types of agro-food system and to further systematize the analysis of regional trajectories. Finally, based on this typology, we investigate the evolution of agro-food systems toward the different types identified, the consequences of these evolutions in terms of N and P fluxes and some interpretations regarding the drivers of these evolutions are proposed based on literature review.

2. Concepts of the method, data collection and hypothesis

2.1. Concepts of the GRAFS approach

The GRAFS approach (Generalized Representation of Agro-Food Systems, Billen et al., 2014) is a generic biogeochemical approach for describing the agro-food system of a given territory, from the farm (Garnier et al., 2016) to the global scale (Lassaletta et al., 2016), by quantifying nutrient fluxes between cropland, permanent grassland, livestock, humans and the natural environment. From the GRAFS perspective, cropping systems are considered to convert nutrient inputs (such as fertilizers, manure, symbiotic N fixation, and atmospheric deposition) into harvestable vegetal products. Crop products can be used (i) to meet the vegetal protein requirements of human nutrition, (ii) to be exported or (iii) to feed livestock, to the extent that they are not fed by grazing on permanent and semi-natural grassland or by imported feedstuffs. The difference between nutrient inputs and outputs informs on the nutrient balance of either crop or grassland soils. More detailed information is provided in Supplementary material SM 1.

The efficiency of feed and grazing conversion into consumable animal proteins (meat, milk and eggs) determines the amount of excreted N, a part of which can be recycled into cropland as manure, making cropland fertility partly dependent on transfers from permanent grassland. The fishery and aquaculture sector also contributes, providing animal proteins for human nutrition, but is considered in our analysis as a separate system for the sake of simplicity. Nutrient losses to the environment occur at each stage of the chain, particularly through leaching and volatilization from cropland, loss from animal excreta and waste from food processing and, finally, from human excretion. Storage in the soil pool is also a possible fate for nutrients.

The GRAFS representation enables to draw direct links between different aspects of the agro-food system, e.g., the link between livestock breeding, grassland areas, and forage crops and the link between fertilization of cropland and grassland and environmental nutrient losses. GRAFS also provides some key indicators for analyzing agro-food systems from both the environmental and agronomic perspectives. A full description of the GRAFS methodology has been provided by Le Noë et al. (2017). Hereafter, we only provide a synthetic description of the main hypotheses and data sources to establish GRAFS for past and current times. A more detailed description of the method is provided in SM1. In this study, we applied the approach to 33 regions defined by Le Noë et al. (2016) for 22 dates from 1852 to 2014.

2.2. Data collection for past and present

2.2.1. Human population and consumption

Total, urban and rural population figures at the "département" (Nuts 3) level are provided by the French National Institute for Statistics and Economic Studies (INSEE, 2017) for the census years, every 5 years from 1852 to 2014, interpolated to the years of the survey. The changes in the human diet in France over the last two centuries were investigated from different data sources and archives (Philippe, 1961; Toutain, 1971; Barles, 2007; AFSSA, 2009; Chatzimpiros, 2011; INSEE, 2017). The conversion coefficients used to translate consumption of each food item into N and P were taken from several databases including CIQUAL (https://pro.anses.fr/tableciqual/), Diet Grail (http://dietgrail.com) and USDA (http://ndb.nal.usda.gov). These coefficients were provided in detail by Le Noë et al (2017).

2.2.2. Livestock metabolism

Animal production and livestock numbers for different age classes and animal categories were provided in carcass weight equivalent by annual agricultural statistics, available online at http://agreste.agriculture.gouv.fr/la-statistique-agricole/, starting in 1970. For earlier periods, data were obtained from nondigitized official registers (available from Gallica.bnf.fr). Meat, milk and egg production in terms of N and P were derived from these sources following the same procedure as described by Le Noë et al. (2017). We considered that there was no temporal evolution in the N and P contents of animal organs.

Total excretion in terms of N was estimated from numbers of livestock following a procedure similar to Le Noë et al. (2017), yet temporal evolution was considered.

Total ingestion in terms of N was taken as the sum of total production and excretion. However, part of the N excreted was considered lost by direct volatilization in the form of ammonia. Ammonia volatilization factors depended on manure management which was also documented.

For the sake of consistency between N and P fluxes, total P ingestion was calculated from the P/N ratio of the different types of ingested feed and from the estimated use of P feed additives after 1946. Subsequently, total excretion in terms of P was estimated as the difference between P ingestion and animal production in terms of P. More information regarding hypotheses and data compilation for livestock metabolism is specified in SM1.

2.2.3. Input to cropland and permanent grassland

Application rates of **N and P mineral fertilizers** to agricultural land were taken from Unifa (2016) (Union des Industries de la Fertilisation) at the "département" scale from 1970 to 2014. For the 1929–1965 period, the application rate of N and P fertilizers was provided at the "département" scale by annual agricultural statistics. Prior to 1929, fertilization data did not appear in annual agricultural statistics and were obtained at the national level from the compilation of various sources including Pluvinage (1912) and Duby and Wallon (1993) (see SM1 for further details).

To assess the distribution of total fertilizer use among crop- and grasslands, we used regional data available from special surveys on grassland fertilization carried out by Agreste (Enquêtes Pratiques) in 1958, 1982, 1998 and 2011. No mineral fertilization of grassland was considered prior to 1955 (Richard, 1951).

For **wet and dry N atmospheric deposition** (under reduced and oxidized forms), we used the values provided by EMEP (2006) at the scale of 50 × 50-km grids from 1981 to 2014, assuming even distribution at the regional scale. We calculated the N atmospheric deposition considering a variable background level of N deposition for the 1852–1981 period following the long-term trend established by Ruoho-Airola et al. (2012) for Rothamsted and adding the livestock contribution proportional to livestock density at each period (see SM1 for further details on calculation).

The rate of **atmospheric P deposition** was taken as 0.4 kg P ha^{-1} yr^{-1}, based on the measurements of Némery et al. (2005) and Némery and Garnier (2007) for the Seine Basin, ranging from 0.28 to 0.52 kg P ha^{-1} yr^{-1}. We assumed even distribution across landscapes and geographical areas and used the same rate of P deposition over the course of the whole period studied.

Symbiotic N_2 fixation over arable land was estimated according to the relationship linking N fixation to yields (Y) for grain and forage legumes (Anglade et al., 2015; Lassaletta et al., 2014b). For permanent grassland, we assumed legumes to be responsible for 25% of the total production. The same procedure was used across the entire period studied.

N and P inputs through seeds were estimated as a variable percentage of N and P in harvest based on different sources (Chamber of Agriculture of the Manche "département": http://www.chambre-agriculture-50.fr/cultures/cereales/semis-des-cereales/; Agreste, 2017; Toutain, 1971). All details on calculation are provided in SM 1.

N and P inputs to cropland or grassland via **animal manure** were calculated according to the same procedure as described by Le Noë et al. (2017). Briefly, the fate of N and P embedded in livestock excretion depends on the time spent by livestock outdoors (direct excretion) versus indoors and on the type of manure management for excrement collected indoors. For each type of animal and each region, the available amount of livestock excrement remaining after losses is allocated between grassland and cropland according to the following rules: (1) direct excretion during grazing is allocated to both temporary and permanent grassland pro rata to their respective surface area; (2) excretion indoor is allocated to cropland.

N and P inputs to cropland through recycling of **human excreta** was calculated based on an estimated rate of human waste recycling, differing for urban and rural populations, and varying regionally and throughout the whole period (Esculier et al., 2018) (see SM1 for further details in Supplementary material).

2.2.4. Output from cropland and permanent grassland

Crop- and grassland production was the mass harvested or grazed in wet weight or dry weight depending on the product, provided by annual agricultural statistics. The conversion of vegetal production from mass unit to ktN yr^{-1} and ktP yr^{-1} was based on coefficients for 36 different types of vegetal products gathered by Lassaletta et al. (2014a, Suppl Mat, compiling FAO data). With too few trials investigating possible historical evolution of mineral content of crops, we considered that N and P content in harvested or grazed products did not change over the course of the period investigated.

Hypotheses regarding the allocation of crop- and grassland production between the local human population, livestock production and export to other French or foreign regions to livestock are explained in SM1.

2.2.5. N and P balances in cropland and grassland

The N balance was defined as the sum of N inputs through mineral fertilizers, manure after NH_3 volatilization, symbiotic fixation by legumes, sludge, seeds and atmospheric deposition, minus N output through crop harvesting. The N balance on arable land represents a potential loss to the environment, either as N_2, N_2O and NO emissions or as N leaching (Benoit et al., 2015); part of the N balance can also be stored in the SOM pool. By contrast, below a threshold of about 100 kg N ha^{-1} yr^{-1}, the N balance in grassland is mostly stored or denitrified instead of being leached to groundwater (Billen et al., 2013).

Similarly, the annual soil P balance was calculated considering P inputs through mineral fertilizers, manure, sludge, seeds, and atmospheric deposition, and P output through harvest. Contrary to N, P leaching is generally low because phosphate anions are strongly sorbed onto soil particles and tend to precipitate with Ca^{2+} cations. Most of the time erosion is the dominant loss process (Kronvang et al., 2007). Therefore, in a first approximation, we considered positive P balances to indicate potential P accumulation in soils (with possible subsequent P losses through erosion), while negative P balances revealed P removal from the soil pool (Garnier et al., 2015).

2.2.6. Feed trade

For the 1981–2014 period, the net import of animal feed was obtained from the complete matrix of animal feed exchanged between the 33 French agricultural regions from the analysis of the SitraM database on commodity transport of the French Ministry of the Environment, using the methodology of Silvestre et al. (2015). For the 1961–1981 period, we used the data on feed import at the national scale provided by the FAO (2017). We assumed that the distribution of imported feed from abroad between the 33 French agricultural regions during this latter period followed the same pattern as in 1981, the earlier date documented in SitraM. Before 1961, we considered imported feed to be insignificant, based on the very low volume of imported feed reported by the FAO (2017) at the beginning of the 1960s.

2.3. Uncertainty

Some assumptions in the GRAFS approach, such as those ranking of priorities for allocating nutrients flows between grassland, arable land, livestock and human population, might be a source of bias in our results. It is rather difficult to quantitatively assess this kind of uncertainty, which is part of the construction of the approach itself.

By contrast, operational uncertainties in the data and parameters used can be quantitatively assessed, as described in Le Noë et al. (2017). We used the Monte Carlo method to generate random samples of values for each primary data and each parameter, considering their own level of uncertainty, which can differ over the time period covered by the study. The model's intermediate variables (such as vegetal or animal production in N and P) and outputs (such as N or P balances) were computed in accordance to the Monte Carlo simulation of the primary data (Loucks et al., 2005). We thus generated a distribution of the main variables and outputs of the model by bootstrapping the Monte Carlo simulation with replacements (1000 replicates). The uncertainty for each variable and parameter was given by the standard error of the mean of the 1000 replicates.

3. Results and discussion

3.1. Regional features revealed by the GRAFS approach

3.1.1. Changes in the overall metabolism of three typical regions

Fig. 1 shows the temporal evolution of N flows in agro-food systems for three typical but contrasted French regions: Picardy, Brittany and Loire Amont. In 1852, all three regions were characterized by self-sufficiency for crop, grass and livestock production, which was just enough to fulfil the local human demand. The only exogenous inputs to arable land and grassland were symbiotic N fixation and atmospheric deposition, and total inputs were close to balance with N output through harvest. Consequently N balance over arable land was very low (5 ± 7, 1 ± 5 and 1 ± 3 kg N ha^{-1} yr^{-1} for Picardy, Loire Amont and Brittany, respectively), while the P balance was almost balanced in Brittany and Loire Amont but slightly negative over arable land in Picardy (-2 ± 1 kg P ha^{-1} yr^{-1}), indicating that soil P was being mined at a time when P enrichment of soil was low, which challenged the sustainability of the agro-food system. Mining in soil fertility around the mid-19th century has also been reported in other studies (Güldner and Krausmann, 2017).

The cases of Picardy and Loire Amont, regions that already had a significant excess of crop and livestock production with respect to their local livestock and human population requirements, would suggest that these products already had the status of a commodity. This hypothesis would be particularly relevant for Picardy, where the additional production was likely exported to the nearby Paris food market, leading to a negative P balance. Brittany showed complete self-subsistence since no products had to be imported or exported. Nevertheless, although some differences can be observed, the overall similarity between agro-food systems is striking.

Over one century later, the situation had drastically changed. The GRAFS of the three regions considered in 1970 (Fig. 1) showed contrasted production patterns, although the common features of the evolution of these agro-food systems can be found in the increased recourse to both N and P mineral fertilizers and the overall intensification of internal and external nutrient flows. This happened concomitantly with a sharp decrease in the contribution of symbiotic N fixation to overall inputs (Fig. 2). This stressed out the preeminent role that legume crops had in the previous periods, when they provided both direct fertilization to cropland and indirect fertilization through manure input by fed livestock. In **Picardy**, arable crop production doubled together with the stagnation of livestock density and the local human population, generating an increased crop surplus with more than 80% of the production being available for exportation. Simultaneously, N and P balance over arable land rose significantly, reaching 57 ± 8 kg N ha^{-1} yr^{-1} and 46 ± 1 kg P ha^{-1} yr^{-1}, respectively. The apparent specialization in crop production in Picardy was accompanied by a significant decrease of NUE and PUE from 85 ± 13% and 160 ± 29% in 1852 to 66 ± 4% and 27 ± 2% in 1970, respectively. In **Brittany**, the complementarity between livestock, arable land and grassland continued to be effective since animal excretion and N symbiotic fixation still made up more than 60 ± 3% of arable land fertilization, while livestock were mainly fed from arable land and grassland. However, imported feed was already needed to meet the requirement of increasing livestock. The agro-food system therefore turned from a complete self-sustaining regime in 1852 to a clear system of interdependence for both vegetal and animal production. The consequences of these changes in terms of environmental and agronomic performance was an increase in N and P balances over arable land, reaching 29 ± 11 kg N ha^{-1} yr^{-1} and 37 ± 3 kg P ha^{-1} yr^{-1} and the concomitant drop in NUE and PUE from 96 ± 16% and 118 ± 26% in 1852 to 81 ± 7% and 35 ± 3% in 1970, respectively. The case of **Loire Amont** in 1970 showed much smaller changes compared to the situation of 1852. The recycling of N within the agro-food system continued to make up most arable land and grassland fertilization together with N symbiotic fixation, while livestock production was still self-reliant. Paradoxically, this region showed relatively low environmental and agronomic performance since N and P balances over arable land reached 77 ± 10 kg N ha^{-1} $yr^{-1}$and 39 ± 3 kg P ha^{-1} yr^{-1}, while NUE and PUE decreased from 89 ± 15% and 118 ± 35% in 1852 to 49 ± 4% and 21 ± 2% in 1970, respectively.

Finally, the GRAFS for 2010 confirmed the trends observed in the 1970s, namely the increased specialization in crop and livestock farming for Picardy and Brittany, respectively, and the continued integrated crop and livestock farming in Loire Amont. In Picardy, grassland was less than 15 ± 0.4% of the utilized agricultural area (UAA) and N mineral fertilizer contributed 73 ± 2% to total fertilization of arable land. The case of Brittany is particularly striking in that the agro-food system was almost entirely dedicated to livestock production, with 77 ± 3% of the arable production used for livestock ingestion and more than 55 ± 3% of animal feed being imported. Moreover, although Loire Amont was still characterized by apparently integrated crop and livestock farming, the livestock production was henceforth dependent on imported feed.

Overall, the evolution of these agro-food systems can be analyzed as three aspects of the same process: the progressive expansion of a "metabolic rift" (Foster, 2013) mostly captured, in the present study, through the rising openness of N and P cycles with increasing dependence on external inputs and a growing share of the production being exported.

3.1.2. Historical legacy of P

Long-term accumulation (or depletion) of P in arable land, calculated by summing up annual P balances of cropland over the whole period covered, revealed both similar and contrasted patterns in Picardy, Loire Amont and Brittany (Fig. 3). In all cases the late 19th and early 20th centuries were characterized by the continuous mining of P reserves, indicating that inputs could not compensate for output. This trend was clearly reversed after WWII when a substantial P stock had accumulated until the 1990s. Yet for the recent period, due to a large reduction in mineral fertilizers, Picardy again showed a net mining of the cumulated P balance, while it plateaued for Loire Amont. By contrast, Brittany continued to accumulate P in arable land. Again, this particularity can be explained by the huge amount of P inputs through animal excretion due to the concentration of livestock breeding in the region. For the case of Brittany, P inputs through manure originated, to a large extent, from feed import from South America (Le Noë et al., 2018). Our estimations of P balances over arable land for these three regions were in accordance with two other studies on France, which also indicated a sharp rise of P in arable land at the turn of the 1960s (Ringeval et al., 2014) and their relative stagnation in the last two decades (Senthilkumar et al., 2012). Our results highlighted that, according to their specific trajectories, French regions have constituted their own P legacy, deriving from three main sources: (i) a transfer of fertility from the local permanent grassland, (ii) the importation of

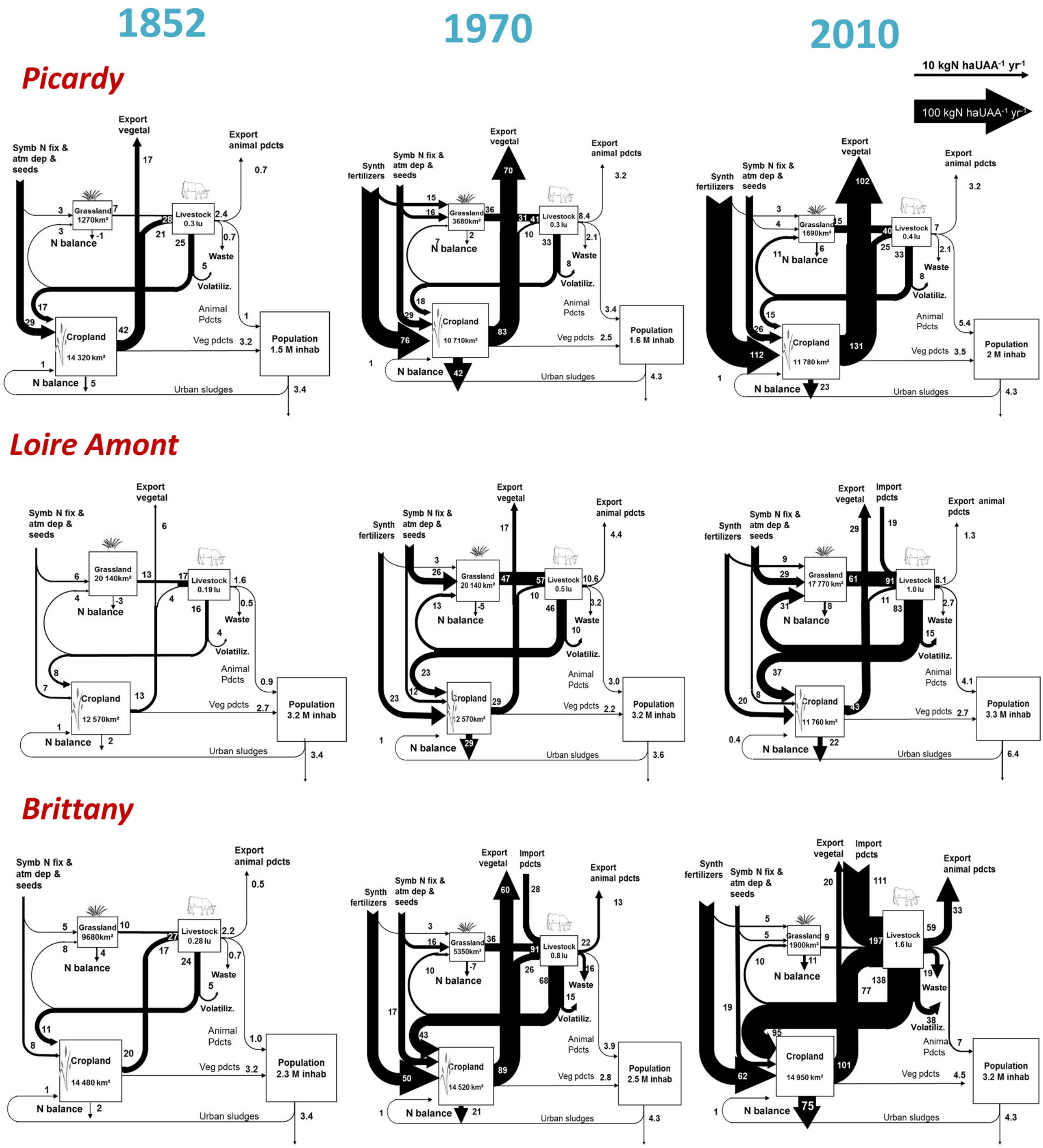

Fig. 1. GRAFS-based N flows for Picardy, Loire-Amont and Brittany in 1852, 1970 and 2010. Fluxes of animal and vegetal import/export relate to trade fluxes of agricultural production with other French regions or foreign countries. Fluxes of Symbiotic N fixation and atmospheric deposition derive from the atmosphere which can be considered as the "surrounding environment". Fluxes of synthetic fertilizer mostly originate from industrial sector, mostly outside of the region and can be seen as external fluxes. N balance represents a potential loss to the hydrospheric or atmospheric surrounding environment through lixiviation and gaseous emission, although part of the N balance remains in the soil of the region.

mineral fertilizers mainly from foreign mining resources and (iii) a transfer of fertility from arable land of feed-exporting regions. In the future, although anthropogenic P accumulated in agricultural soil should contribute to sustaining plant growth for some years (Ringeval et al., 2014), a possible shortage of P (Cordell et al., 2009) could threaten the ecological maintenance of agricultural soil. Regions that benefited from high P fertilization rates in the past will thus take advantage of their past legacy in the future, whereas regions that have historically exported nutrients could suffer from the unpaid cost of ecological soil degradation. Such historically unequal exchanges have been defined as ecological debt by several authors (Roberts and Parks, 2009; Clark and Foster, 2009).

3.2. Typology of the different kinds of agro-food systems

The GRAFS approach could provide as many monographs as regions considered. To systemize the analysis of our results and to obtain an objective assessment of the trajectories of the 33 French regions, we

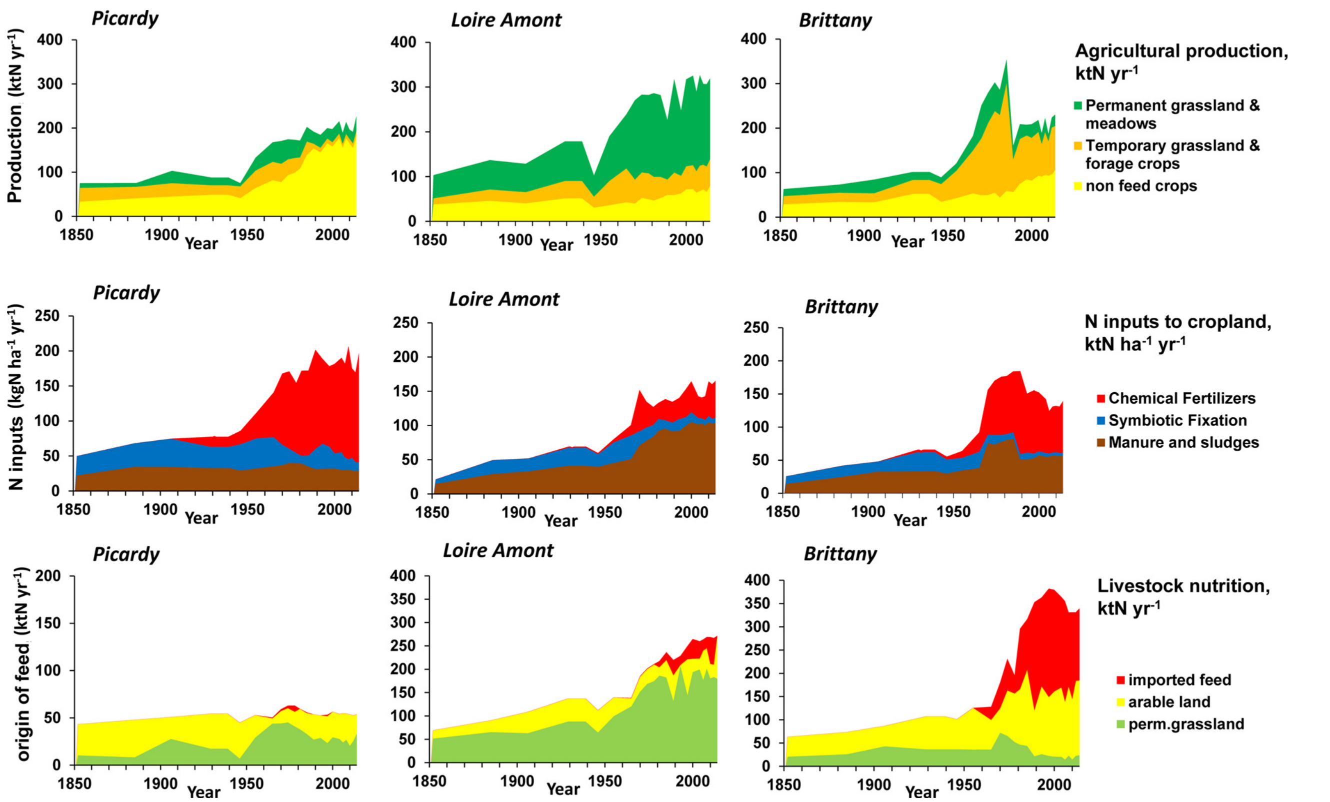

Fig. 2. Features of the agricultural systems of Picardy, Loire Amont and Brittany over time. a. Agricultural production; b. type of N fertilization; c. origin of livestock feed.

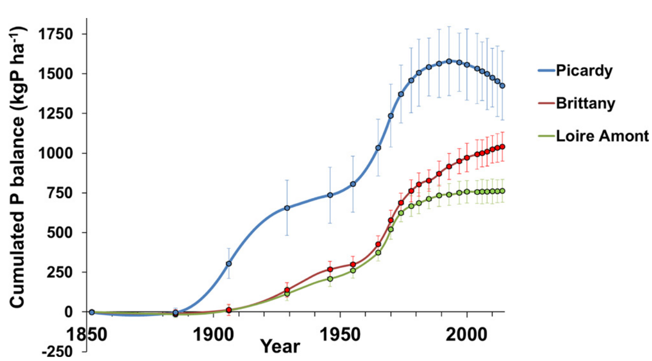

Fig. 3. Cumulated P balance over arable land in the course of the 1852–2014 period in Brittany, Loire Amont and Picardy. Error bars provide uncertainties as calculated by the Monte Carlo analysis.

chose here to aggregate these regions according to similarities in their production pattern. This typology corresponds to an algorithm that filters various results provided by the GRAFS analysis and subsequently defines each region as belonging to an agro-food system type. The choice of criteria and thresholds is necessarily somewhat arbitrary, minimized by a thorough analysis of the raw results. The analysis of the three typical trajectories of agro-food systems (Fig. 1) indicated that specialization and intensification were the two main changes captured through the GRAFS approach. Such a typology can help characterize the structure of an agro-food system based on the trajectories' degree of specialization, openness or self-reliance and inter-connections between crop, grassland and livestock breeding.

For this purpose, we defined biogeochemical criteria to check the level of interaction between arable land, livestock and grassland, based on nutrient fluxes or flux ratios. The ratio between arable crop production and livestock excretion was used as an indicator of the dominance of crop production over livestock breeding. The share of manure in arable land fertilization and the proportion of local arable production in livestock feeding were both used as indicators of the connection between crop and livestock farming. The proportion of grass in livestock feeding was taken as an indicator of the connection between grassland production and livestock production. Finally, the livestock density and the share of imported feed in livestock feeding were considered as indicative of the specialization in livestock production and the dependence of livestock production on feed supply. The typology was established based on N production and fertilization data, but the subsequent analysis can also account for P management in each of the different types of region defined. The decision tree accordingly represents the differentiation logic, criteria and thresholds used (Fig. 4).

The agro-food system types can be literally defined as follows: (i) "Arable crop" systems are characterized by the dominance of arable crop production over livestock breeding; (ii) "Intensive livestock farming" systems are characterized by high livestock density and dependence on feed import. However, the connection between arable and livestock production exists in this system through the high proportion of manure in arable land fertilization. By contrast, (iii) "Extensive integrated crop and livestock farming" systems are defined by the connection between livestock, grass and crop production through the double criteria of a large share of grass in livestock feeding and a significant contribution of manure to arable land fertilization. Systems defined as (iv) "Intensive integrated crop and livestock farming" are characterized by the connection between livestock and arable production through either the significant contribution of manure in arable land fertilization and/or the high proportion of local arable production in livestock feeding. However, the share of grass in livestock feeding is of lesser importance compared to systems defined as extensive integrated crops and livestock farming. Lastly, (v) "Intensive crops-extensive livestock farming" systems are characterized by the connection between livestock and grassland production owing to the significant proportion of grass in livestock feeding, while the link between livestock and arable land production is weak, since manure plays only a minor role in

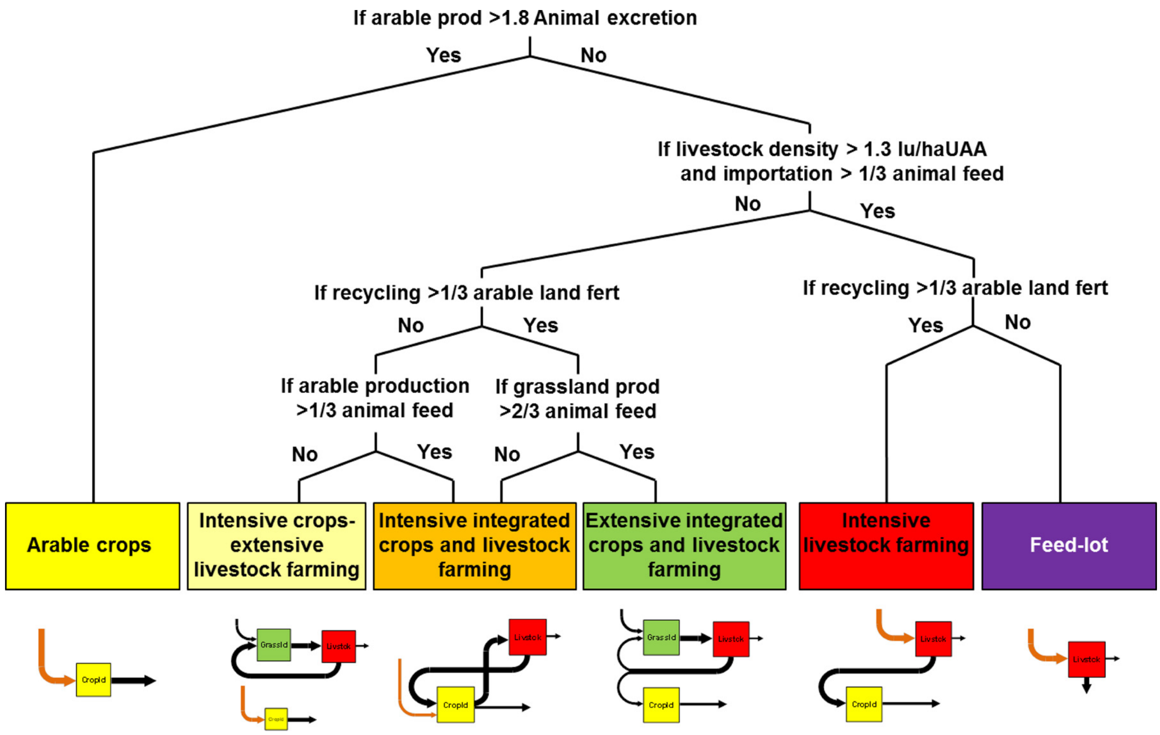

Fig. 4. Decision tree representing the elaboration of a typology of agricultural regions in France.

arable land fertilization and local arable land production contributes only marginally to livestock feeding. Consequently, arable land is mainly fertilized by mineral fertilizers, which justify the first term "intensive crops" for this type of system.

All criteria gathered regions with a homogeneous production pattern; however, the internal variability among regions belonging to the same type was sometimes concealed. For instance, in 1929, the average yield of arable land, a parameter that was not included in our typology, reached 50 ± 15 and 54 ± 9 kg N ha^{-1} yr^{-1} in regions of extensive and intensive integrated crops and livestock farming, respectively. In 1989, the average yield of arable land of the different regions was 130 ± 15 kg N ha^{-1} yr^{-1} for Arable crops, 97 ± 23 kg N ha^{-1} yr^{-1}, and 97 ± 21 kg N ha^{-1} yr^{-1} for Extensive and Intensive integrated crops and livestock farming, and 81 ± 38 kg N ha^{-1} yr^{-1} for Intensive crops-extensive livestock farming. The variability of individual regional values around the mean emphasized the need for a dual approach: at the regional level to describe specific trajectories and at the national level to analyze the overall evolution of production patterns toward specialization or continued integrated crops and livestock. In the next section we did not discuss the variability within each type of region but provided uncertainty values as calculated by the Monte Carlo procedure (see Section 2.3).

3.3. Trajectories of agro-food systems

In view of the above, we can now trace the evolution the agro-food system of the 33 regions in France toward specialization or continued integrated crop and livestock farming (Figs. 5a–d and Figure 6). Putting the changes captured within their political and economic context should allow us to identify the drivers of these changes together with their impact on N and P fluxes in agro-food systems. In the following, N and P fluxes are therefore analyzed under their dual, social and material, dimension.

3.3.1. 1852–1906: the dominance of integrated crop-livestock farming

Integrated crop and livestock farming was the rule everywhere during the late 19th century (Fig. 6), because fertilization mainly relied on manure recycling and symbiotic N fixation. Therefore, the progressive intensification of production with increasing yields occurring during this period (Fig. 5b) was only driven by an increase of livestock density with the increase in forage crops and grassland. The agricultural system in this period therefore fully obeyed the well-established paradigm that crop production can only be improved by increasing manure resources (Krausmann, 2004). At the farm scale, diversification of production was also the best suited way to cope with pests, disease and weather and ensure the subsistence of the farm while multiplying possible outlets. As long as technical possibilities remained limited, integrating crop and livestock was inherent in farming systems (Antoine and Herment, 2016). The visible shift from intensive integrated crop and livestock farming to extensive integrated crops and livestock farming systems between 1852 and 1906 (Fig. 6) also reflects the increase in grassland area as a corollary of the growing importance of livestock breeding activities. The integrated crop and livestock farming model was also well adapted to respond to a growing demand for animal protein whose distribution became physically possible with the development and extension of the railway network (Freycinet plan in 1878). Consequently, animal production, which had long been a "necessary evil" (cf. Lavoisier 1791), started to become a marketable product. The expansion of the railway in the late 19th century also engendered the emergence of a national market for food products by opening up isolated rural regions. As a consequence, price differentials between regions lessened: for instance, the maximum difference among "départements" in the price of wheat shifted from 70% in the mid-19th century to less than 10% in 1913 (Duby and Wallon, 1993).

The slow increase in productivity of French agriculture, essentially driven by an increase in manure production rather than by the recourse to industrial fertilizers, contrasted with the development of agriculture in England and Germany during this period (Duby and Wallon, 1993; Muller, 1984). Duby and Wallon (1977) interpreted this delay in French agriculture to result from the structure of agriculture in France at these times that was well adapted to the needs of a national economy that matched the place France held in the world economy. Until the end of WWII the economic power of France in global capitalism arose mostly from its financial role as an international banking power. According to

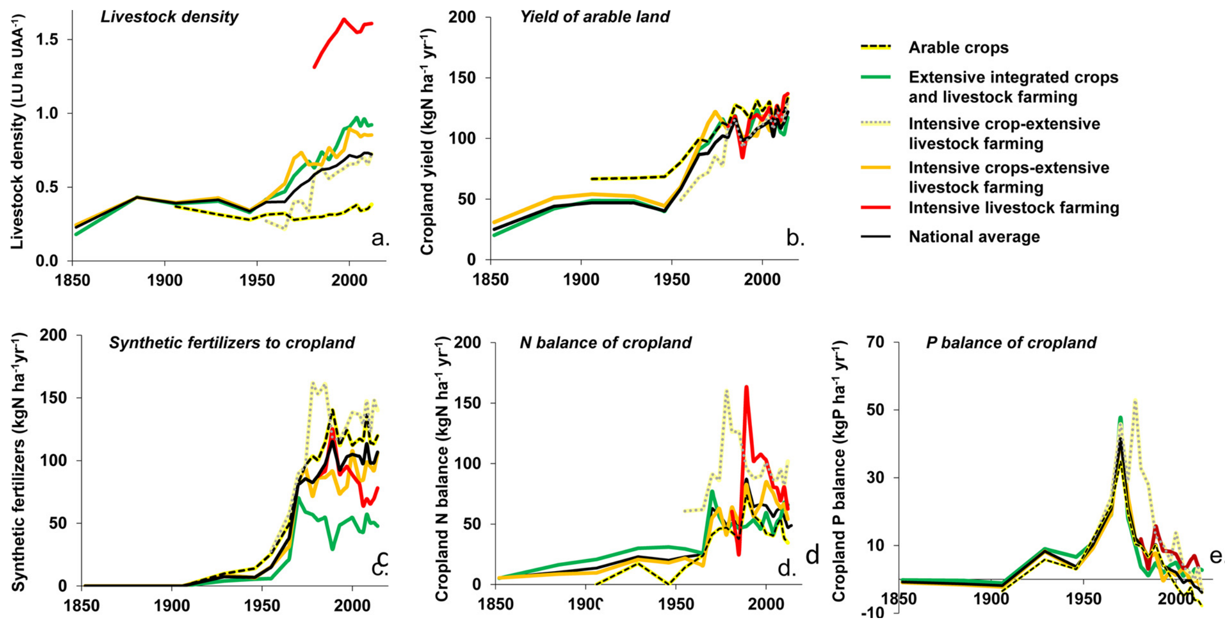

Fig. 5. Evolution in the five different types of regions considered for France over the 1852–2014 period. a. Livestock density (LU haUAA^{-1}); b. yield of arable land (kgN ha^{-1} yr^{-1}); c. rate of N synthetic fertilizer application over arable cropland (kgN ha^{-1} yr^{-1}); d. N balance of arable land (kgN ha^{-1} yr^{-1}); e. P balance of arable land (kgP ha^{-1} yr^{-1}).

Duby and Wallon (1993), with their savings, peasants contributed almost one-third of the surplus money that could be appropriated through the banking channel and invested in the industrial sector and, above all, invested abroad as loans. For this reason, there was no political will in France to enhance labor productivity by mechanization or other forms of modernization, since this would have resulted in an increased rural exodus and a shift of peasant savings to investments. The protectionist measures implemented by the French Minister of Agriculture Jules Méline between 1884 and 1892 and until the beginning of WWI offered a sort of artificial prosperity to farmers, who were ensured outlets for their production without improving labor productivity through mechanical and chemical modernization; instead, famers could fulfil their dream of purchasing land (Duby and Wallon, 1993). The limited rate of fertilization induced by this slow modernization resulted in very high NUE and PUE, a low N balance and a slightly negative P balance (Fig. 5c and d), indicating that the intensification of agricultural production led to loss of soil fertility, with only quite limited inputs of exogenous fertilizers.

The results could provide the impression of uniformity in the agricultural production pattern in the 1852–1906 period. However, some areas within a region, such as the Rennes Basin in Brittany, were already in a process of specialization in the sense that their local production was already dedicated to exportation (Cocaud, 2016), while others remained behind this movement. In total, the perspective adopted in this study revealed that, despite a possible patchwork of farming, the overall regional structure of agro-food systems led to similar biogeochemical patterns.

3.3.2. 1906–1946: timid beginnings of crop specialization

For the same reasons as those explained in section 3.3.1, and in spite of the major crises of WWI and 1929, the inter-war period continued to be characterized by the near homogeneity of integrated crop–livestock farming. Self-subsistence was still perceived as the best protection against poverty and integrated crops and livestock farming thus remained the best-suited system. This assumption can be confirmed by the figures of on-farm consumption in 1938: 30% of the pork meat, 30% of the dairy products, 40% of the eggs, 20% of the wine and 10% of the wheat harvest were consumed on the farm (Duby and Wallon, 1977). Nevertheless, the very beginning of regional specialization in crop production around the Ile-de-France region was already visible (Fig. 6). Following Von Thünen's theory, the specialization in cash crop production in this area should be attributed to the closeness of Paris; however, other factors such as proximity to the Seine River or the establishment of a complex commercial network also explain the geography of specialization (Antoine and Herment, 2016).

During this period, N mineral fertilization clearly rose in regions specialized in arable crops, shifting from almost zero at the end of WWI to 9.9 ± 0.9 kg N ha^{-1} yr^{-1} in 1929 and then 14 ± 1.2 kg N ha^{-1} yr^{-1} in 1946. By contrast, in regions of integrated crops and livestock farming, N mineral fertilization rose more slowly. Between 1929 and 1946, the rates of N mineral fertilizer over arable land shifted from 4 ± 0.4 kg N ha^{-1} yr^{-1} and 8.5 ± 0.8 kg N ha^{-1} yr^{-1} to 5.5 ± 0.6 kg N ha^{-1} yr^{-1} and 6.7 ± 0.7 kg N ha^{-1} yr^{-1} in regions of extensive and intensive integrated crops and livestock farming, respectively. Regarding P, rates of mineral fertilization increased considerably between 1906 and 1929 but then slightly decreased between 1929 and 1946, probably due to the economic difficulties arising from the economic crisis and WWII (8.7 ± 0.7, 8.5 ± 0.7 and 9.2 ± 0.8 kg P ha^{-1} yr^{-1} in 1929 and 7.5 ± 0.6, 7.1 ± 0.6 and 6.3 ± 0.5 kg P ha^{-1} yr^{-1} in 1946 for arable crops, extensive and intensive integrated crops and livestock farming systems, respectively). Although it was limited, the progress in the use of mineral fertilizers during the inter-war periods has been mainly attributed to the popularization of new technical possibilities provided by industry and national syndicates for mineral fertilizers, such as the propaganda created in 1920 (Duby and Wallon, 1977). Furthermore, modernization was triggered after WWI resulting from the losses of millions of peasants killed in combat in WWI; increasing the productivity of agricultural labor was therefore a necessity. However, the role of the state remained limited to the creation of the national Grain Board in 1936 by the Front Populaire, allowing farmers to be involved in the fixation of wheat prices. In the absence of national policy, the investment in new technical equipment remained limited and more than two-thirds of peasant debts involved land acquisition.

In spite of the development of mineral fertilizer, arable crop yields stagnated, although disparities between the different types of agro-food systems showed better performance in regions of arable crop with average yields over arable land of 67 ± 4, 47 ± 3 and 51 ± 3 kg N

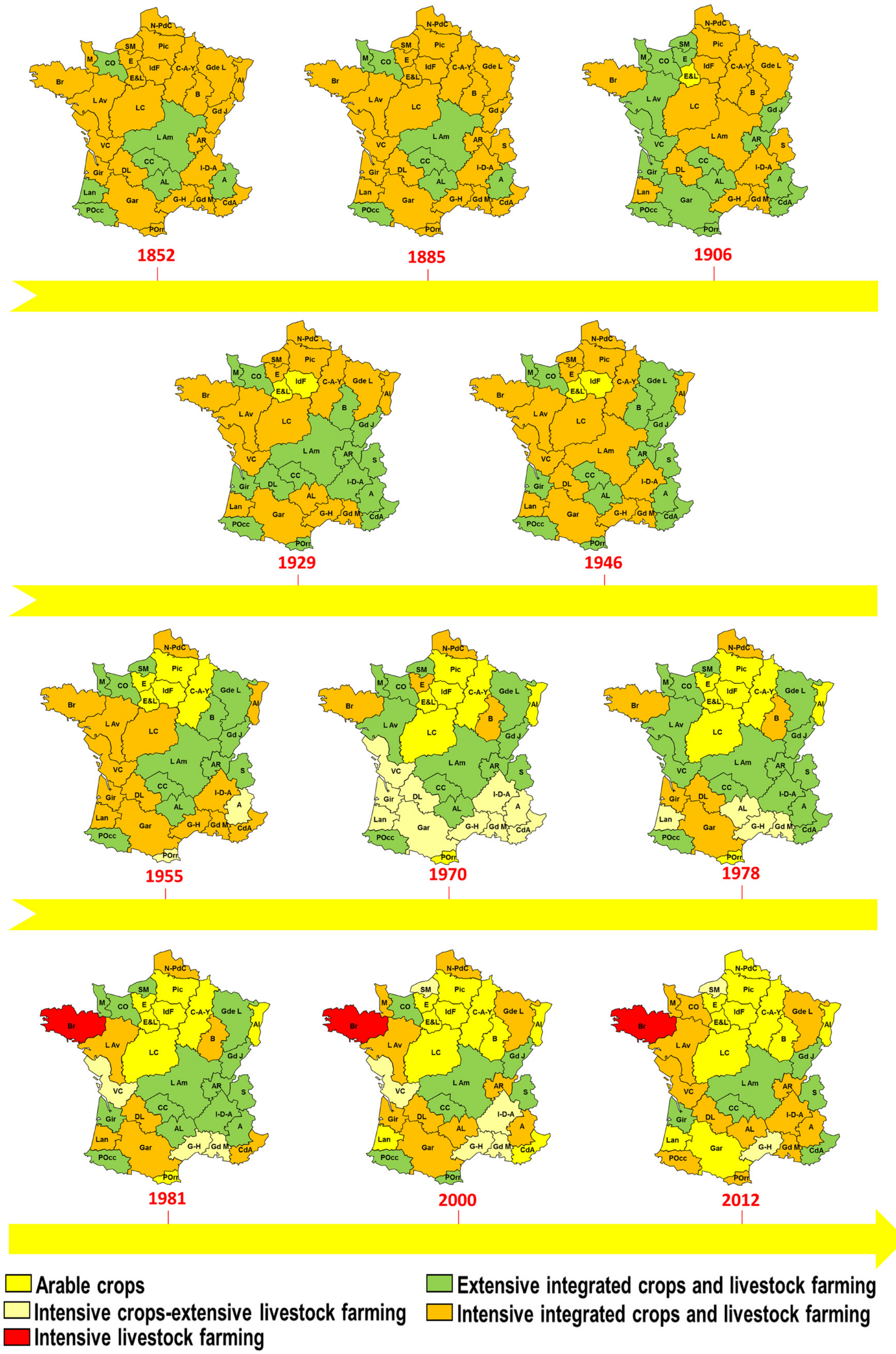

Fig. 6. Chronology of the different types of agricultural regions in France over the 1852–2014 period. A: Alpes; Al: Alsace; AL: Aveyron-Lozère; AR: Ain-Rhône; B: Bourgogne; Br: Bretagne; C-A-Y: Champagne-Ardennes-Yonne; CC: Cantal-Corrèze; CdA: Côte d'Azur; CO: Calvados-Orne; DL: Dordogne-Lot; E: Eure; E&L: Eure-et-Loire; Gar: Garonne; Gd J: Grand Jura; Gd M: Grand Marseille; Gde L: Grande Lorraine; G-H: Gard-Hérault; Gir: Gironde; I-D-A: Isère-Drôme-Ardèche; IdF: Ile de France; L Am: Loire Amont; L Av: Loire Aval; Lan: Landes; LC: Loire Centrale; M: Manche; N-PdC: Nord Pas-de-Calais; Pic: Picardie; Pocc: Pyrénées Occidentales; POr: Pyrénées Orientales; S: Savoie; VC: Vendée-Charentes.

ha^{-1} yr^{-1} for arable crops, extensive and intensive integrated crops and livestock farming systems, respectively (Fig. 5a). The combination of stagnant yield and little progress in terms of N fertilization resulted in a progressive increase in N balance over arable land, but this remained quite low (Fig. 5c). Similarly, the slight decline in P mineral fertilization between 1929 and 1946 led to reduced, but still positive, P balances for cropland in all types of regions.

Interestingly, regions that specialized were also among those identified by Bonnin et al. (2014) as presenting the greatest loss of wheat genetic diversity, promoted by intensification and integration of crop production to market. However, their study showed that other regions had already been concerned by losses in wheat genetic diversity since 1912, mainly the Paris basin and the northwest quadrant. It is therefore possible that regions such as Picardie, Eure and Champagne-Ardennes-Yonne were already on the path toward arable crop intensification, as will be confirmed in the next section.

3.3.3. 1946–1978: the expansion of crop specialization and the great acceleration

The three decades following WWII were marked by the expansion of arable crop system around the Paris Basin and the development of a new type of agro-food system defined in our typology as intensive crops-extensive livestock farming. The use of synthetic fertilizer started to increase from the end of WWII and exploded at the beginning of the 1960s, generating high N and P balances over arable land (Fig. 5c and d) and grassland. This trend was not specific to France: similar patterns also occurred in other European countries (Bouwman et al., 2017). The degree of openness of N and P cycles strongly depended on the type of agro-food system. Regions of arable crops exported most of their vegetal production and relied almost completely on N and P mineral fertilizers to compensate for export through harvest. Regions of intensive crops-extensive livestock farming relied mainly on N and P mineral fertilizer for arable land production but retained significant livestock farming where animals were mainly fed on permanent grassland. In this type of region, the complementarity between arable land, grassland and livestock production was giving way to a disconnection of animal and crop production. Along with the expansion of agricultural specialization, regions defined as extensive and intensive integrated crop and livestock farming still existed, with strongly interlinked N and P flows through arable land, grassland and livestock production, but were characterized by an intensification of both arable land and livestock production (Fig. 5a, b) and an increased use of synthetic fertilizer, particularly P. Therefore, although specialization only concerned crop production, intensification of arable production was occurring everywhere with increased yield. Densification of livestock production was also characteristic of this period, except for regions specialized in crop production (Fig. 5b). As a consequence, N and P balances rose sharply from the beginning of the 1960s in all types of region, resulting in a rapid decrease in NUE and PUE.

These far-reaching changes in the intensity and nature of N and P fluxes in the different types of agricultural systems in French regions have to be linked to their historical background. In the post-WWII context, France was completely devastated and almost everything had to be rebuilt. Consequently, France had to live on credit and its commercial balance was strongly budget-deficit. By comparison with previous periods, the economic situation of France in the world economy had completely changed (Duby and Wallon, 1977). A voluntarist policy was thus implemented to find a new acceptable place for the French economy within Europe, implying an enormous increase in agricultural production, in order not only to meet domestic demand, but also to become a net exporter on the international agricultural market (Jollivet, 2007). The Monnet Plan (1945) followed by the Marshall Plan (1947) thus explicitly aimed at improving labor productivity in agriculture in order to export food products, accelerate the rural exodus and free labor for employment in industry, as well as making agriculture an important outlet for the mineral and machine industry (Duby and Wallon, 1977). To reach that goal, the French government implemented a strong policy of popularization and education in the countryside and developed new land ownership legislation that favored the development of the medium-sized farm, eliminating the smallest and least competitive landowners while avoiding the concentration of land in the hands of non-farmers, which would have led to the development of wage-earning in the agricultural sector (Duby and Wallon, 1977). The implementation of the European common market in 1957 fostered the modernization of agriculture by protecting internal European markets with custom barriers. Later on, the implementation of the Common Agricultural Policy beginning in 1962 ensured that farms adopted modernization to remain viable by providing them outlets for food products (Bureau and Thoyer, 2014).

Therefore, modernization of agriculture in France was stimulated through propaganda and economic incentives but was constrained by laws restricting and regulating access to land, reinforcing the competition between farmers who had to modernize to survive. This fierce competition combined with modernization is probably the main factor explaining the expansion of crop specialization during the 1946–1981 period (Jollivet, 2007). Machines, fertilization and herbicides were first designed for arable land to facilitate crop specialization. Furthermore, cereals producers were the main beneficiaries of the French government and European subsidies through price setting of wheat and export subsidies (Bureau and Thoyer, 2014). Therefore, the great acceleration in crop specialization, the increasing openness of N and P fluxes in agro-food systems and the dependency on external inputs were driven by economic interests and by the political commitment on the part of the French government and the Common Agriculture Policy.

3.3.4. 1978–2014: livestock specialization and the rational use of fertilizers

The most recent period from 1978 until the present was characterized by the continuous development of specialized arable crop systems together with the emergence of a new type of agro-food system, i.e., "intensive livestock farming" (see Fig. 6). At the same time, regions defined as intensive crops-extensive livestock farming declined, either as a result of a lower rate of mineral fertilizer, which gave greater weight to manure recycling on arable land and grassland and therefore led to the conversion of this type of region into an integrated crop and livestock farming system, or because they turned into specialized arable crop system. The 1980s marked the beginning of intensive livestock farming in the west of France. Poultry and pig breeding were the most emblematic of this new form of industrial breeding, which were no more than factories converting vegetal proteins into animal proteins (Duby and Wallon, 1977). The consequences in terms of N and P fluxes were an increased dependence upon feed import and a structural excess of N and P inputs through manure to arable land and grassland, generating high N and P balances, although moderate mineral fertilization has reduced these balances over the two last decades (Fig. 5d).

Indeed, this period also showed a deep inversion in the trends of mineral fertilization. At the same time, arable yields kept rising, although at a slower rate than in the previous period. As a consequence, the N and P balances on arable land steadily decreased. In regions of arable crops, P balances have even become increasingly negative over the two last decades. As pointed out in a previous study (Le Noë et al., 2018), this could probably be achieved without loss in crop yield due to the huge soil P legacy built up between 1960 and 1990. This decreasing trend is taking place in all types of regions and started by the end of the 1970s. It coincided with the second oil crisis, which generated a peak in energy prices and therefore in the price of N and P fertilizers (Mew, 2016). With the 2007–2008 food crisis, phosphate rock and fertilizer prices increased by 700% in 14 months (Cordell et al., 2009), probably contributing to reinforcing the decline in P mineral fertilizer use. After the 2000 s the decline in N and P mineral fertilizer use might also be attributed to the implementation of agro-environmental measures introduced at the European level through a series of successive European directives gathered within the Water Framework Directive in 2000

(Bureau and Thoyer, 2014). The consequences of these changes were the increase of NUE and PUE in every type of region except intensive livestock farming regions.

Summarizing the changes during this period, regional specialization strengthened while N and P inputs through mineral fertilizers decreased but the ever-growing dependence for both inputs and outlets, increased the openness of N and P cycles. On the economic side, policies progressively shifted from interventionism and support to production to give way to greater liberalism by lowering and sometimes eliminating customs barriers. The Common Agricultural Policy separated subsidies from the volume of production, which instead became indexed on the surface area farmed and herd size. The aim was to incite farmers to adapt to international competition (Bureau and Thoyer, 2014) which subsequently reinforced these specialized production patterns and fully integrated agriculture into the market economy.

4. Conclusion

The GRAFS approach was implemented herein to study the long-term evolution of agricultural production patterns in the regions of France for the first time. The associated changes in N and P flows and balances over arable land and grassland were analyzed. In the mid-19th century, the production patterns of regional agricultural systems were quite homogeneous. Integrated crop and livestock farming systems were the rule everywhere and manure recycling was the main source of agricultural land fertilization. However, at the beginning of the 20th century, the sustainability of this farming system began to be jeopardized by the mining of soil nutrient stocks, especially P. Over the course of the 20th century, this situation shifted toward more contrasted production patterns. This became particularly true after the Second World War: modernization of agriculture contributed to the emergence of specialized cereal cropping systems in the most fertile plains of the North of France, exporting a large fraction of their cereal production. This trend was characterized by a decreased connection to livestock. The significant rupture after 1946, which accentuated the openness of regional agro-food system, was promoted by the increased recourse to mineral fertilizers and the subsequent separation of crop and animal production in regions on a path to specialization. The consequences for N and P cycles varied, depending on the specific trajectory of each region. Overall, regions of intensive cropping revealed the highest degree of openness through crop exportation, while the region of intensive livestock farming was characterized by a growing reliance on feed importation and over-fertilization of arable land through manure inputs. However, all regions intensified their production during the second half of the 20th century by increasing rates of mineral fertilizer inputs. As a result, arable yield rose, as did the N and P balances. The rise of fertilizer prices together with the implementation of agro-environmental measures over the last two decades resulted in the lowering of N and P balances, the latter being even negative in regions specialized in crop production. However, as long as the agro-food system remains specialized, possible changes through improved fertilization will be limited by the inefficiency of nutrient recycling at the regional level.

This study demonstrates the value of adopting a long-term perspective to articulate a vision of how production pattern changes and environmental and agronomic performances are related. The analysis of these changes in light of their political and economic contexts, based on the existing literature, showed that changes in production patterns as well as their consequences on N and P flows and agro-environmental performances should be mostly attributed to external factors related to political choices and modes of insertion within the market economy, rather than to mechanisms endogenous to agricultural production itself. Material flow analysis was shown to be a way to capture both the biophysical processes governing agricultural production and its impact on natural resources and the environment. At the same time, analyzing social, economic and political processes, based on the existing literature, allowed determining the controlling factor of how these processes evolve. A holistic approach linking the materiality of agricultural production to its political and economic logic was therefore indispensable to understand past changes for a vision of possible transformations of agro-food systems in the future. More generally, the present study stressed out the artificiality of the distinction between "natural" and "anthropic" fluxes and the need for coupling quantitative and qualitative approaches. This, in turn, implies to reconsider the duality between nature and culture around which the human and social sciences on the one hand and the natural sciences on the other hand, are divided. Hence, this study should also be an incentive for further interdisciplinary collaboration at least in the fields of ecology, agronomy and environmental sustainability.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.agee.2018.06.006.

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