Appendix

Table 1 presents the sources used to estimate nutrient flows in Catalonia's agro-ecosystems, and the text that follows provides full details about each numbered nutrient flow.

1. Rainfall

Rainfall brings small amounts of elemental nutrients with it. To estimate these inputs, or "wet deposition," we looked for values from a low-polluted nearby watercourse. In Catalonia, La Castanya valley in Barcelona province is probably the most well-known sampling site for ecologists. Rodrigo (1998) measured wet deposition of $N\text{-}NO_3^-$, $N\text{-}NH_4^-$, $P\text{-}PO_4^{3-}$ and K^+ as 27.9 (±2.3), 31.9 (±3.4), 1.03 (±0.12) and 3.45 (±0.42) µeq/L during one year (1995-96). We corrected these values with the average annual precipitation of each agro-ecoregion (Agencia Estatal de Meteorologia en España 2014). Holland et al. (1999) attribute pre-industrial nitrogen deposition levels of 0.43 $kg \cdot ha^{-1} \cdot y^{-1}$ (0.07-0.60) and 0.67 $kg \cdot ha^{-1} \cdot y^{-1}$ (0.19-1.12) to Mediterranean scrubland and xenomorphic forest/woodland respectively. However, they did not take into account pre-industrial cropland areas, but only potential natural vegetation, which could explain why their values are so low.

2. Free-fixation

Free-fixation by bacteria in soils require anaerobic conditions, which in turn requires wet conditions. Hence, for hot, summer dry areas such as Mediterranean Spain, we estimate 1-5 $kg \cdot ha^{-1}$ (Loomis et al. 2011), a lower value compared to the 5-10 $kg \cdot ha^{-}$ found in wet areas like the United Kingdom (Goulding 1990).

3.Symbiotic fixation

Symbiotic association between legume plant roots and bacteria from the *Rhizobium* genus fixes atmospheric nitrogen into plant-useable form. Taking into account the N fixed in aerial parts of the plants (38%) and their roots (60%), plus the share of the nitrogen fixed in the plant that would have settled into the soil (rhizodeposition of 18%), we multiplied the total N content of legume plant production by 1.14 (Garcia-Ruiz et al. 2012).

4. Irrigation

Irrigation water also delivers elemental nutrients to cropland. Nutrient concentrations in current unpolluted streams nearby the study area approximate the amount of nutrients dissolved in irrigation water (Garcia-Ruiz et al., 2012). Lacking data of unpolluted streams, we used average data over 10 years from springs in unpolluted areas of each province available from the ACA[1] online database. The N, P and $K \cdot L^{-1}$ values of 2.0, 0.05 and 2.0 mg respectively were similar to those employed by Garcia-Ruiz et al. (2012).

Unlike other parts of Catalonia, on the plains of Lleida province there was irrigation on 26% of the olive groves and 31% of vineyards. When estimating soil nutrients added by irrigation water, the annalysis accounts for these irrigated lands. However, the sources did not specify respective yields, so it was impossible to distinguish productivity between irrigated and non-irrigated crops.

5. Seeds

Seeds are a mix of genetic material and small amounts of nutrient reserves that feed emerging seedlings. These reserves can be considered recycled nutrients and were very important in pre-industrial systems (Chorley 1981). For all cereals, legumes, and potatoes sown

[1] *Agència Catalana de l'Aigua*, is a public entity considered the hydraulic authority in Catalonia.

by splitting the tuber, we used the seeding rates per hecatare and the composition described in Soroa (1953).

6. Miscellaneous Organic Fertilizers

The report of JCA (1921) lacks garbage data for Barcelona and Lleida. At the end of the nineteenth century the production of garbage for the city of Barcelona was estimated as the same for Paris, i.e. 1 $L \cdot cap^{-1} \cdot day^{-1}$ (García-Faria 1893). However, the heterogeneity and the uncertainty of the management and production of garbage makes it difficult to estimate it for 1920. Given all the uncertainties around garbage and pomace collection and use, together with the fact that this kind of fertilizer corresponded to only a minor share in comparison with the others described in this section, this analysis uses garbage data directly as described in JCA (1921). One discrepancy concerns oil and winery pomaces reported in JCA (1921), as the provinces with the highest production did not seem to use them as fertilizer, and JCA (1923) described Girona as apparently using more wine pomace than it produced. In some cases, farmers may have prefered to feed pomace to livestock, rather than use if for a fertilizer. In other cases, pomaces were used to make distillates. The excess in Girona is clearly an error when compared to values reported in the literature for similar processes (Cabrera 1995; Daneo 1921). Consequently, we adjusted the value to the one reported in JCA (1923).

7. Humanure

Both the numbers for humanure and livestock manure appear inconsistent in JCA (1921). For humanure, we calculated the collection potential according to the three main settlement patterns (urban, village, dispersed farm) and their associated main disposal system types. For the biggest urban area, Barcelona city, we assumed two thirds of humanure was released to the sea, with one third stored in cesspits and so potentially collected (García-Faria 1893). To differentiate between rural and urban we followed the criteria of the Spanish *Instituto Nacional de Estadística*, which considers settlements with a population higher than 2000 inhabitants as urban. We did not account for urine, due to the difficulties of collecting it and the high nitrogen losses due to ammonia volatilization. Production of human feces is between 135-270 $g \cdot day^{-1} \cdot cap^{-1}$ fresh weight without urine (Gotaas 1956). Based on the number of inhabitants per settlement (Esteve-Palós 2003), the average of potentially collected human feces was 37.8±12.3 kt.

Table A1. Human feces (fresh weight without urine) potentially collected in Catalan cities.

Province	**Average potentially human feces collected (t)**
Barcelona	37,827±12,270
Girona	11,330±3,114
Lleida	10,523±3,288
Tarragona	11,418±3,197

Source: see table 1.

8. Manure

The number of livestock reported in censuses (see Table 3) are multiplied by stabling coefficients reported by Cascón (1918). According to the production average of each livestock type (Table 4), the potential manure applied was 4.5, 11.0, 1.2, 6.3 and 0.9 t/cropland area in Barcelona, Girona, Lleida Plains, Pyrenees and Tarragona respectively. The average composition from all livestock types together was consistent with those that Gotaas (1956) reported as average for stable manure in a fresh state: 70-80% moisture, 0.3-1.9% N, 0.1-0.6% P_2O_5 and 0.3-

1.2% of K_2O. These numbers also match with those adopted by Gallego (1986): 0.62%N, 0.27% P_2O_5 and 0.63% of K_2O.

9. Synthetic fertilizers

Fraud in the composition of synthetic and mineral fertilizers was rather common in Spain at the beginning of the twentieth century. Legislation to counter passed in 1900, but still buyers knew that purchasing synthetic fertilizers outside of farmers' unions or associations exposed them to fraud (Sanz 2005). This situation made synthetic fertilizers fall into disrepute among farmers (García-Luzón 1922). In spite of these facts, and due to lack of data, we did not take into account the differences of richness between adulterated or pure forms of synthetic fertilizers. The composition of pure forms can be found in a series of authors (López-Mateo 1922; García-Luzón 1922; Soroa 1953), unsurprisingly matching among them and also with the composition that Gallego (1986) uses in his article, which depends on data from Aguirre-Andrés (1971).

10. Harvests in Lleida

Data in the yearbook of 1922 (JCA 1923) are at the provincial level, an administrative unit too aggregated to match with the argo-ecoregions in Figure 1. To adapt it we had to disaggregate the province of Lleida by *Partido Judicial*, and split it into the plains zone (Balaguer, Borges Blanques, Cervera and Lleida) and the Pyrenees (Seu d'Urgell, Solsona, Sort, Tremp and Viella). The *Partido Judicial* was the next smaller administrative division in Spain at that time, and Spanish statistical reports frequently used it as a unit of analysis. The first cropland map for Spain was not published until 1933 (DGA 1933), and it had low resolution and was not georeferenced. Notwithstanding, it provided valuable information when we estimated the share of annual crops between the plains and the Pyrenees (65% and 35% respectively) in Lleida. An additional irrigation report (JCA 1916) accounts the area irrigated by the channels of Urgell, Aragón, Cataluña and Pinyana, which were the most important waterways from the Segre River to the plains of Lleida; together they covered 90% of the irrigated land in Lleida province. With this information, we could estimate the location of irrigated land.

JCA (1923) disaggregated some of Lleida's crops (tomatoes, peppers, plums, almonds, olive trees and vines) by *Partido Judicial*. Forages rotated with other crops and were more common in the plains than in the mountains. They were mostly consumed within the province, except alfalfa, which locals sold to Barcelona or Tarragona to feed livestock used in industry (JCA 1914). Consequently, we estimated there was a larger share of forage crops in the plains than in the Pyrenees (82% and 18% respectively) depending on the percentages reported in DGA (1933). The livestock census of 1917 (JCA 1920) reported data to calculate total cropland together with total pastures and forests per *Partido Judicial.* According to this source, 84% of meadow, forest and scrub land of Lleida was in the Pyrenees. Crosschecking these results gave consistent values.

11. Nitrogen losses

The "Guidelines of the Intergovernmental Panel on Climate Change" (IPCC, 2006) quantify the N emissions linked to agriculture as nitrous oxide (N_2O), a greenhouse gas. Not all N emissions are directly in the form of N_2O. IPCC (2006) also accounts for other forms of nitrogen, such as ammonia (NH_3), nitrates (NO_3^-), and nitrites (NO_2^-), which eventually may become N_2O through reduction or oxidation. NH_3 emissions are through volatilization, NO_3^- and NO_2^- through leaching. Nitrification is the aerobic microbial oxidation of ammonium (NH_4^+) into nitrate, while denitrification is the anaerobic microbial reduction of nitrate to nitrogen gas (N_2), both processes producing N_2O eventually.

Nitrogen emissions in agriculture follow two processes: through storage, which resulted in the loss of almost 50% of the N in manure; and through the management of agricultural soils, which included application of other fertilizers, tillage, and irrigation. To calculate losses during storage we separated dung from straw (since the main losses occured from dung). Therefore, we estimated the amount of straw used and its composition, correcting the amount of straw used per livestock type from Cascón (1918) to match the available straw (JCA 1923) and the NPK values for crop type in Soroa (1953). These considerations apply to humanure as well. Author (2015: 116-125) provides an extended discussion of the remaining details of the application of the IPCC guidelines to the Catalan case study.

While we applied the N storage losses only to manure and humanure, which could be almost half of their N content, management losses also included those caused by the application of each type of fertilizer plus cropland tillage. The emissions due to land management, storage and crop management emissions per cropland area are consistent with broad global averages that Smil (1999) calculated for denitrification, leaching and volatilization from fertilizers, around 18-29.6 kg N·ha^{-1} altogether. The main difference is that he gave more importance to leaching losses (10-15 kg N·ha^{-1}) than here, due to the aridity of some regions in our case study.

12. Flows not included

Finally, there were two geological nutrient flows described by Garcia-Ruiz et al. (2012) that we could not include: additions from soil formation and nutrient losses from erosion of surface soil horizons. Although georeferenced data on bedrock and soil type could be found in

ICGC *(Institut Cartogràfic i Geològic de Catalunya*)[2], the reconstruction of a detailed map to allocate them to each land use in 1920 for all of Catalonia was beyond this study's scope. This omission, however, would not significantly affect the balances, as weathering of the bedrock has low intensity in dry Mediterranean regions (Garcia-Ruiz et al. 2012). Moreover, due to the terracing of vineyards, erosion should be lower than that reported for olive groves in the southern Iberian Peninsula (Infante-Amate et al. 2013; Vanwalleghem et al. 2011).

[2] http://www.igc.cat