

1 **A Phosphorous Flow Analysis in Spain**

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15 **Abstract**

16 Phosphorus (P) is a vital macronutrient required to improve the agricultural yields but its excessive
17 use as a fertilizer has resulted in pollution of water bodies leading to eutrophication. With no reserves
18 of phosphorus source in Spain, increased dependence on phosphorus in agriculture have not only
19 increased dependence on imports but also has raised concerns on its future availability as a
20 resource. A Phosphorous Flow Analysis (PFA) was conducted for Spain for the year 2012 focusing
21 on the food production and consumption systems. The results obtained were finally compared with
22 PFA at both country level and continent level (EU-27). To quantify food and non-food flows systems,

23 country specific data were considered. The sectors covered were crop production (CP), animal
24 production (AP), food processing (FP), non-food production (NF) and consumption (HC). The findings
25 reveal that a total of 325 kt P was imported by Spain in 2012; 66% of which was accumulated in
26 markets stock of food and feed, fertilizers and non-food (91 kt P) while 33% was lost to the
27 environment through land-fill, losses to water bodies, land accumulation and incineration. The largest
28 proportion of losses is associated with water bodies (44.7 kt P) followed by agriculture and land
29 accumulation (42.1 kt P). Wastewater treatment plants (WWTPs) received around 79.5 kt P within
30 wastewater, with 60% being removed in sewage sludge. The 31.7 kt P discharged within final effluent
31 represented the 71% of the total losses to water bodies. Around 69% of the sewage sludge was
32 recycled to agriculture and 27% was sent directly to landfill including the ashes from incineration. Net
33 accumulation was 1.84 kg P/cap which was similar to values reported for the EU-27 average (2.5 kg
34 P/cap).

35 **Keywords:** Substance flow analysis; MFA; secondary resources; waste management

36

37 **1. Introduction**

38 The United Nations growth forecast for world population of about 9.7 billion by 2050 (e.g., an
39 increase of 33% over the next 35 years) will undoubtedly represents an increase in demand for
40 fertilizers. This is also based on the forecast made by Food and Agriculture Organization (FAO) for a
41 1.8% annual increase in crop production by 2050 (FAO, 2015). Thus, the key to the short- and long-
42 term stability of the food chain is to close the cycles of the main resources, which are necessary to
43 support them (e.g., nutrients as phosphorus (P) and nitrogen (N)). P is an essential element to
44 sustain life since it is part of the crucial biological processes, such as reproduction (e.g., DNA), body

45 structures (e.g., bones) and energy supply in the form of ATP (Oelkers and Valsami-Jones, 2008)
46 and it cannot be substituted. It also plays a vital role in enhancing soil fertility, agricultural productivity
47 and therefore, on the global food security.

48 Although there is an increase in the global demand, the low diversity of the phosphate rock deposits
49 and the fact that it is considered a non-renewable resource makes P a critical raw material for a
50 nation dependent on agriculture (Sattari et al., 2012). The availability of the phosphate rock reserves
51 is restricted to five major countries (e.g., China, USA, Russia, Morocco and Sahara) and this global
52 imbalance underlies serious challenges at world scale (Childers et al., 2011, Jasinski, 2014, Cooper
53 et al., 2011). Past estimates suggested a time span of 50-100 years for the complete depletion of
54 these primary P reserves (De Haes et al., 2009; Vaccari and Strigul, 2011; Cordell et al., 2010,
55 Cordell et al., 2009a), but current estimates are relying on 300–400 years depending on the
56 dynamics of demand and supply under current extraction rates (Reijnders, 2014; Scholz and Wellmer
57 2013; Jasinski, 2014). There is also the probability that the price of P increases in the future with
58 increasing demand and depletion as well as the increase of the cost of production and this may affect
59 the affordability and may also escalate the price of food. This scenario could further create
60 geopolitical tensions which could in turn make it difficult for dependent nations to procure P from the
61 main producers.

62 Due to the facts and projections as stated above, the EU has included, in 2014, the P-rock in the
63 critical materials list (EC, 2014a). Thus, nations and regions having no P reserves or having low
64 deposits (e.g., EU with only very few P-deposits in Finland (de Ridder et al., 2012; Reijnders, 2014))
65 need to initiate a change in the cycle of this critical resource to reduce their dependence on primary
66 sources. The probable solution to the P-challenge has been directed to its sustainable management
67 consisting of its efficient use including higher and better recycling. Such potential solutions can be

68 identified by P flow analysis (PFA) studies that provide an insight into how humans have used P, and
69 how P has been transfer to the environment on different spatial scales (Chowdhury et al., 2014). The
70 PFA can also provide knowledge to identify in-sustainable uses for more sustainable P use (Cordell
71 et al., 2012). Various PFA's have been conducted at a global level (Smil, 2000; Liu et al., 2008;
72 Cordell et al., 2009a) and also at a continent level like the EU (Richards and Dawson, 2008; Ott and
73 Rechberger, 2012) and Africa (Cordell et al., 2009a). PFA's were also conducted on specific
74 countries like Australia (Cordell et al., 2010), China (Li et al., 2012), Japan (Matsubae et al., 2011;
75 Mishima et al., 2009), Austria (Egle et al., 2014; Seyhan, 2009), Belgium (Coppens et al., 2013),
76 Denmark (Klinglmair et al., 2015), Finland (Antikainen et al., 2005, 2008; Saikku et al., 2007), France
77 (Senthilkumar et al., 2012a,b), Germany (Gethke, 2012), Netherlands (de Buck et al., 2012; Smit et
78 al., 2010), Norway (Hamilton et al., 2015), Sweden (Linderholm et al., 2012a), Switzerland (Binder et
79 al., 2009; Lamprecht et al., 2011), Turkey (Seyhan, 2006; 2009), and United Kingdom (Cooper and
80 Carliell-Marquet, 2013). Such studies have given a qualitative as well as a quantitative description of
81 P flows which in turn have been used to forecast future P requirement and usage, recovery and
82 reuse options so as to secure both food as well as P availability.

83 The results of these analyses have some common findings despite the differences in terms of the
84 territory covered or the purpose of the study. It can be highlighted that some of these countries are
85 net phosphorous importers even those that are net food exporter (e.g., Australia). Substantial losses
86 and inefficiencies have been identified in the P cycles due to the low recycling rates for several P
87 flows with the largest losses within the systems to water and soil accumulations. Some studies
88 highlighted a considerable unexploited potential for improvement. Those efforts should be focused on
89 P removal and recovery at WWTP, as well as the developing more effective methods for recycling
90 bulky wastes such as animal manure, food waste and especially municipal sewage sludge, which

91 could potentially substitute a significant part of the total applied mineral P fertilizers. However,
92 resource recycling and, thereby, reducing P fertilizer use appeared to be less promising than
93 scenarios based on reduced food waste or redesigned agricultural systems.

94 Spain, like other European countries, has no P-rock mineral deposits and therefore depends on
95 imported mineral phosphate fertilizers to support Spanish fertilizer industries and agriculture.
96 Therefore, the food security in Spain is at present highly dependent on a secure and affordable
97 supply of mineral fertilizers derived from imported phosphate rock. Based on this context, a PFA
98 analysis of Spain has been conducted focusing mainly on the agriculture, food and fertilizers
99 production and consumption system. Since these are the flows where most of the phosphorus
100 transfer occurs, but including flows from industrial processes and waste management processes
101 which interact with P system. The flow diagram and system boundaries used in the PFA analysis
102 were based on a global perspective, which includes the food system (food consumption–production–
103 waste chain), as well as non-food flows (detergent, fertilizer and forestry industries), in addition to
104 obtaining quantitative information on imports, exports, major areas of loss or accumulation. The
105 objective behind the PFA analysis is to identify the areas requiring the primary resources of P and
106 simultaneously to identify available secondary resources. An analysis of the potential contribution of
107 secondary P resources associated with urban wastewaters, identified by the EU as one of the target
108 contribution against the P scarcity, was carried out and results compared to other PFAs as reported
109 worldwide. The study has extracted data on P cycles, industrial and agricultural uses to develop the
110 mass flow analysis using databases available for public consultation. For data not available,
111 published data from mass flow analysis on other country or region was taken into account. This PFA
112 on a global perspective will be especially relevant for the proposed EU approaches: circular economy
113 (EC, 2011, 2014d) and bio-based economy (EC, 2012).

114 **2. Methodology**

115 Material Flow Analysis (MFA), has been used to examine resources such as minerals, water or energy at
116 a wide range of geographical scales (from global to local) (Cordell and Neset 2014). The method was
117 established in the field of Industrial Ecology to aid environmental management for assessing the
118 'metabolism' of human (anthroposphere) or technical (technosphere) systems. The method is based on
119 two scientific approaches: i) mass balance, which enables a systematic assessment and tracking of the
120 flow of materials (e.g., P) between various processes, as well as the imports to and exports from the
121 system (Cooper and Carliell-Marquet, 2013) and finally ii) the system analysis. An important concept
122 regarding mass balance under the MFA is related to the conservation of mass in which a material is
123 transformed during its flow but cannot be destroyed (Baccini and Brunner, 2012).

124 At the global level P flows have been quantified along the whole P use chain (Cordell et al., 2009a).
125 This quantification was then used for: i) predicting the future P use so as to ensure long term global
126 food demand (Cordell et al., 2009b), ii) developing a system so as to recover and reuse P in order to
127 ensure global P security (Cordell et al., 2011, 2013) iii) identifying synergies for a sustainable future
128 based on global P scarcity (Neset et al., 2013) and finally, iv) developing a framework to assess the
129 vulnerability of national and regional food systems towards the multiple-dimensional stressors of P
130 scarcity (Cordell and Neset, 2014).

131 **2.1 System description and boundaries**

132 The phosphorous flow analysis covers the state of Spain, organized in 19 autonomous regions and
133 including Baleares and Canarias islands. The P flows were focused, primarily, on agriculture and the
134 food production systems since both accounts for more than 50% of all P uses (MAPAMA, 2016a), but
135 also has considered other industrial uses such as phosphorous in fertilizers, detergents, beverages

136 and food. Contrary to other EU countries where sewage sludge is incinerated, this scenario in Spain
137 has a marginal contribution. The flows associated with the recovery of P from waste waters and the
138 application of sewage sludge to agriculture has also been considered in the analysis.

139 **2.1.1 Data collection**

140 The quantification of the P flow network required the use of several data sources, including official
141 statistical databases, surveys and interviews; and published reports. The statistical data mainly
142 contained the amounts of P substances such as P-containing products, sown crop areas, chemical
143 fertilizers being applied to field, crop harvests, milk production, the number of livestock and human
144 population. Data concerning the amount of material flows were collected from European and Spanish
145 MINETAD databases: FAOStat (FAO, 2009), Eurostat (EUROSTAT, 2016), Spanish Ministry of
146 Agriculture, Food, Fishing and Environment (MAPAMA) (MAPAMA, 2016 a, b, c, d), Spanish Ministry
147 of Energy, Tourism and Digital Agenda (MINETAD) (MINETAD, 2016), Spanish Agency for
148 Consumer Affairs, Food Safety and Nutrition (AESON) (AESON, 2016) and National Institute of
149 Statistics (INE) (INE, 2016). When data were not available, the mass balance principle was applied
150 and missing flows were estimated. It is worth to mention that uncertainty was also considered,
151 especially those associated with the variation of P concentrations in goods or the mass flow
152 variability. In the present study, P flows were quantified by multiplying the material flows with their
153 respective P contents (described as an amount of P_2O_5) and were expressed in kt P /y.

154 The base year of 2012 was chosen as it represented the most recent year for which an almost
155 complete dataset could be gathered, and because it was also the year that an extended review on
156 sewage sludge database was carried out (MAPAMA, 2016 e, f). When data for 2012 was not
157 available, data from 2013 was taken to represent the value. Similar approaches have also been used

158 in other PFA in Finland (Antikainen et al., 2005), Japan (Matsubae et al., 2011), Australia (Cordell et
159 al., 2010), United States of America (Suh and Yee 2011), France (Senthilkumar et al., 2014), United
160 Kingdom (Cooper and Carliell-Marquet, 2013) or Denmark (Klinglmair et al., 2015). However, it should
161 be noted that this is a static model and that annual variations may be significant and comparison
162 between years would be very valuable, which would be part of a continuous exercise when more data
163 becomes available.

164 **2.1.2. Software platform for modelling**

165 The PFA follows the reference guides (Graedel and Allenby, 2010) and the main studies developed
166 for country/region levels described in the introduction. Quantitative modelling of P flow in such
167 complex system is usually done with the help of different mathematical and statistical tools (STELLA,
168 POWERSIM, VENSIM, STAN (Vienna University of Technology, 2012), and MATLAB/Simulink®. In
169 this study, the STAN code was used. STAN tool builds a graphical model with predefined
170 components (processes, flows, system boundary, text fields) where it is entered or import known data
171 (mass flows and stocks, volume flows and stocks, concentrations, transfer coefficients) for different
172 layers (good, substance, energy) and periods to calculate unknown quantities. The code includes the
173 possibility to consider data uncertainties. The algorithm uses mathematical and statistical tools such
174 as data reconciliation, error propagation and gross error detection.

175 **2.2. Data management and uncertainty assessment**

176 One of the main concerns of MFA is the identification of the potential errors and uncertainty of
177 results. This study has also employed cross-checking of original results through the use of alternative
178 calculations, comparing estimates to values obtained from the literature, and deriving mass balance
179 estimates where possible (Senthilkumar et al., 2012b). In some cases, various estimates have been

180 used for the same point, so there have been averages of these flows, which are shown in Table A1
 181 (Supplementary Material). The approach was useful to confirm the results by highlighting erroneous
 182 results. The double counting was also revised and avoided, for instance imported crops could be counted
 183 twice under both crops and under the imported animal feed. To avoid this, commodities with a potential
 184 risk of double counting were first identified. The use for each commodity was then determined and
 185 categorized into a single flow, ensuring that it was removed from other flows. The information collected
 186 shows the dispersion level of the results that have been used to obtain the average values.

187 Confidence range for PFA were obtained by using the HS approach developed by Hedbrant and Sörme,
 188 (2001) and widely used in different PFA studies (Seyhan, 2009; Cooper and Carliell-Marquet, 2013,
 189 Danius, 2002, Antikainen et al., 2005 and Asmala and Saikku, 2010). The methodology used has been
 190 described with more details by Cooper and Carliell-Marquet (2013). Uncertainty levels are assigned to
 191 various data sources and then applying an interval to each level. During situations when a series of
 192 calculations were used in order to make estimates for the same data points, a confidence interval was
 193 developed for each separate calculation as per the methodology described by Antikainen et al. (2005),
 194 and finally, an average confidence interval was taken for the overall data points. The details of the
 195 defined intervals used and examples are summarized within Table 1.

Table 1. Uncertainty intervals used on the PFA in Spain.

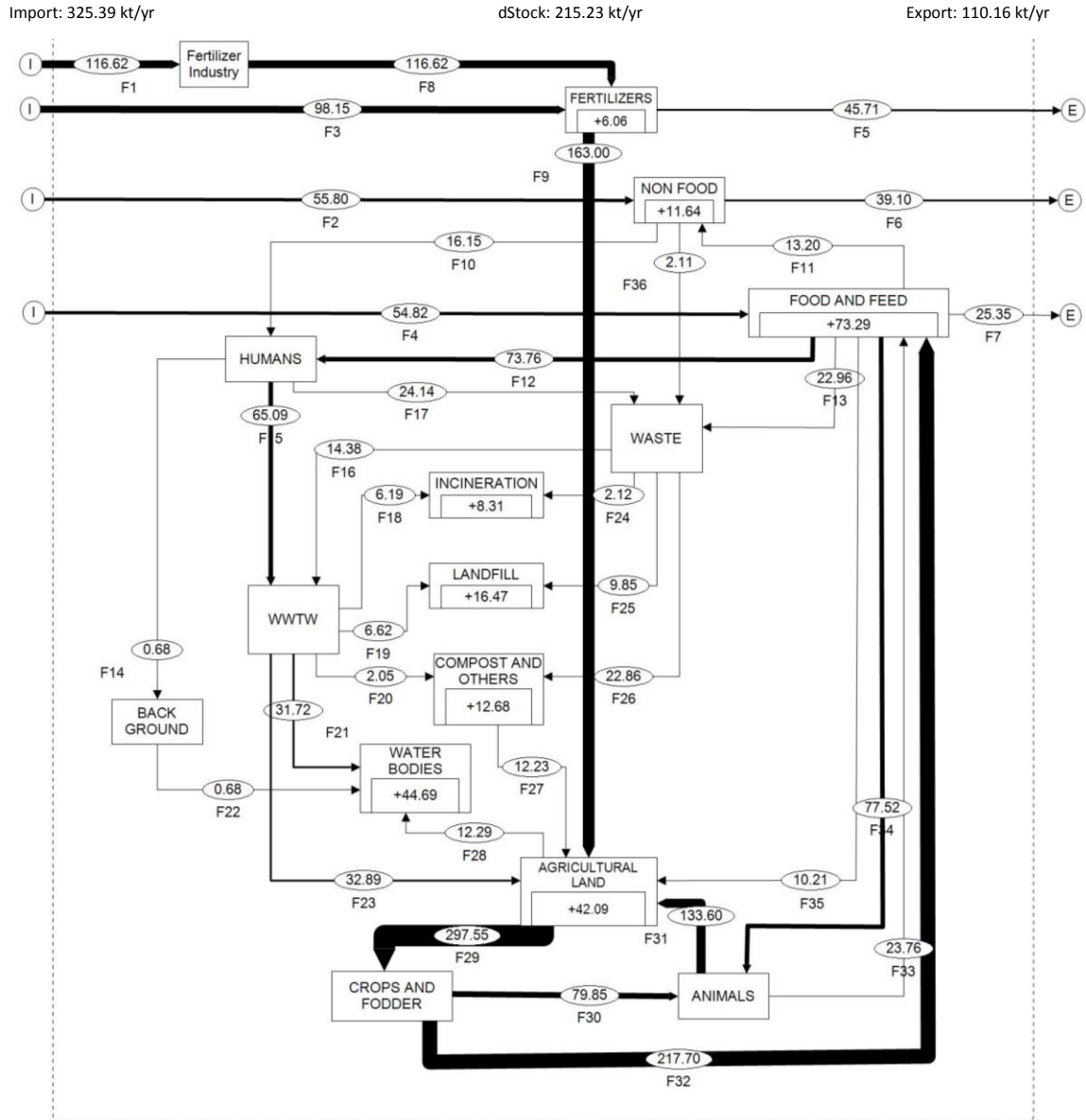
Level	Interval	Source of information	Example
1	*/1,05	Official statistics at Spain scale.	Spain population estimate
2	*/1,1	Official Statistics at Spain scale. Value from literature.	Area of land, crop yields, animal numbers from MAPAMA or INE. P contents from literature
3	*/1,2	Official statistics scaled up to Spain scale. Values from literature.	Detergent consumption scaled up to Spain scale. Specific values from other studies in literature.

4	*1,33	Values from literature without reference or methodology. Significantly scaled results or other assumptions.	Quoted values or own assumptions of P content. Assumptions about waste disposal or use of animal wastes
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196 The final results as incorporated into the STAN program are presented in Figure 1 and Table A1
197 (Supplementary Material), along with the 95% confidence limits.

198 **3. Results and discussion.**

199 This section presents the results obtained for the PFA in Spain, with the objective of determining
200 areas of inefficient use of P resource, identifying the main losses and accumulations, and calculating
201 the dependence of Spain on imports of phosphorus. Figure 1 shows the results of the PFA using the
202 STAN code, adopting a layout similar to that produced by Senthilkumar et al. (2012b). As shown in
203 Figure 1, while the boxes represent the main processes and stocks, the connecting arrows represent
204 the main flows. The flows are presented in the Sankey format, in which the width of the arrow is
205 shown proportional to the size of each flow. The quantity of each flow (expressed in kt P/y)
206 accompanied by the uncertainty, is included in the circle along each arrow, the description and name
207 of each P-flow are summarized in Table 2.



208

P flow Spain, 2012

Figure 1. PFA results for food production and consumption systems in Spain. Diagram plotted using the STAN program. All values are expressed in kt P/y. Calculations, assumptions and uncertainties for each flow value are summarized in Table A1 (Supplementary Material).

Table 2. Definitions and description of P flows used for the PFA in Spain.

Number	Flow Name	Description
F1	Rock import	Imported P rock
F2	Non-food import	Imported P in non-food commodities
F3	Fertilizer import	Imported P fertilizer
F4	Food and feed import	Imported P in food and feed
F5	Fertilizer export	Exported P fertilizer
F6	Non-food export	Exported P in non-food commodities
F7	Food and feed export	Exported P in food and feed
F8	Fertilizer production	Produced P in fertilizers
F9	Fertilizer application	Applied P fertilizer to agricultural land
F10	Human non-food consumption	P consumed as detergents
F11	Non-food commodities	P within food type commodities that have uses other than food, feed or seed (Biodiesel).
F12	Human food consumption	P consumed in domestic food market
F13	Food and feed processing waste	P loss during manufacturing food
F14	Other disposal for human excreta	P within human excreta which is not disposed of to sewers
F15	Human excreta to Wastewater Treatment works (WwTw)	Human P waste to treatment facilities
F16	Waste to sewers	P within human waste to sewers
F17	Human waste	P within human waste food
F18	Sewage sludge to incineration	P within sewage sludge
F19	Sewage sludge to landfill	P within sewage sludge
F20	Sewage sludge to composting or other disposal	P within sewage sludge
F21	WwTW final effluent	P within WwTW effluent discharged to water bodies
F22	Background losses to water	P within background losses to water bodies
F23	Sewage sludge to agriculture	P within sewage sludge
F24	Waste for incineration	P within waste for incineration
F25	Waste for landfill	P within waste for landfill

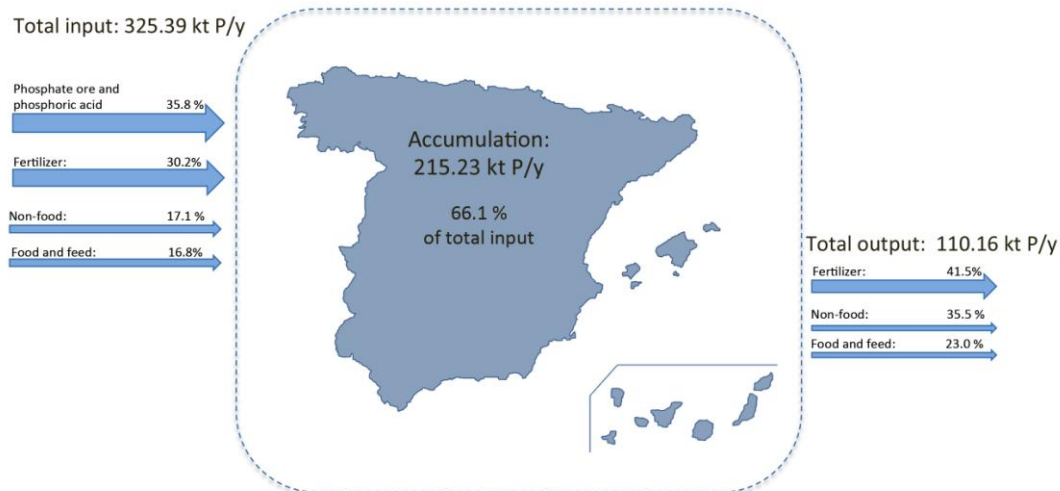
F26	Waste for composting or other disposal	P within waste for composting or other disposal
F27	Non-farm manures	P within non-farm manures
F28	Agricultural losses to water	P lost to water bodies from agricultural land
F29	Crop uptake	P removed from agricultural land in crops and grasses
F30	Animal grazing	P taken up during animal grazing
F31	Animal manure	P in animal manure applied to agricultural land
F32	Crop products	P within crop products
F33	Animal products	P within animal products
F34	Animal feed	P within animal feed
F35	Seed and planting material	P within seed and planting material
F36	Non-food processing waste	P loss during manufacturing non-food

209

210 **3.1 Main P flows in Spain**

211 Spain imports a total of 325.39 kt P /y (Figure 2), mostly from phosphate ore, phosphoric acid and
 212 fertilizers but also as food, animal feed and non-food commodities such as detergents. Total exports
 213 amount to 110.16 kt P /y resulting in a net import of around 215.23 kt P /y. The net import represents an
 214 annual accumulation of P within the territory of Spain.

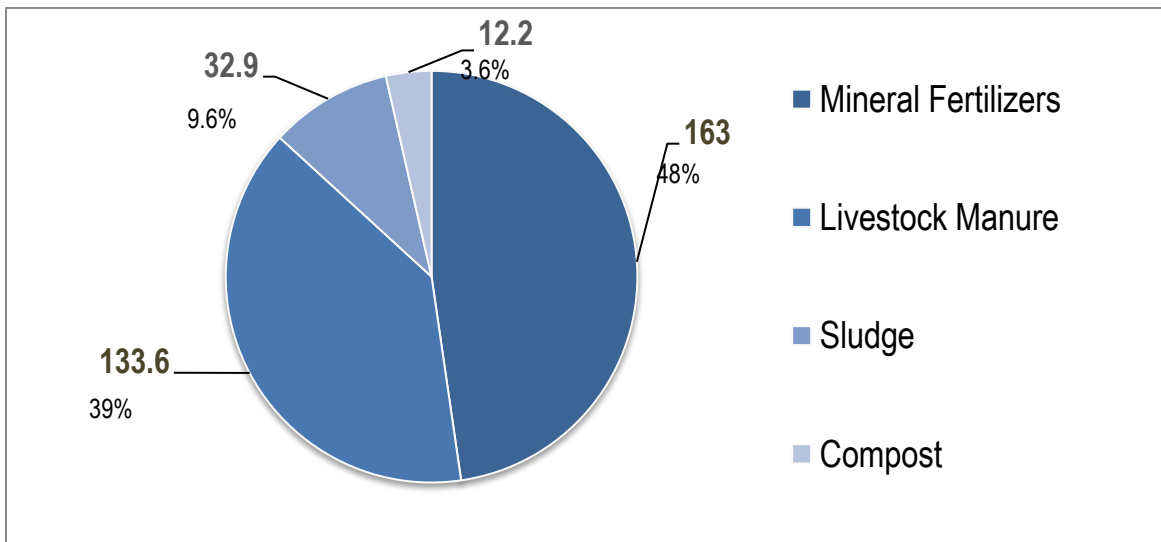
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216
217

Figure 2. Spain imports, exports and accumulation of P in 2012 according to PFA, values expressed in kt P/y.

218 The total P-flow input to agriculture is estimated at around 341.70 kt P/y, in the form of mineral fertilizers
 219 (48%), animal manure (39%), urban sewage sludge (10%), compost and others (3%) as it is shown in
 220 Figure 3. The outputs in the form of crops and grasses are 217.7 and 79.85 kt P/y, respectively,
 221 accounting for a total of 297.55 kt P/y and an average of 21.65 kg P/ha. The efficiency of the agricultural
 222 system in converting P inputs within fertilizers and manures (341.7 kt P/y) into outputs as crops and
 223 grasses (297.55 kt P/y) is around 85%. The difference between total inputs and outputs suggests an
 224 annual surplus of 54.38 kg P/y. Agricultural losses to water are estimated to be 12.29 kt P/y, showing an
 225 annual accumulation of 42.09 kt P/y onto agricultural land.



226

Figure 3. Distribution of P input sources (kt P/y) and their proportion in agriculture according to PFA in Spain.

227 For European PFA, flows and input–output balances have been focused mainly on agricultural
 228 balances, taking primarily agricultural flows related to crop and animal production into account
 229 (Csatho and Radimsky, 2009; 2012; Grizzetti et al., 2007; OECD, 2013; Richards and Dawson,
 230 2008; Sibbesen and Runge-Metzger, 1995; Tunney et al., 2003). Compared to these agricultural P
 231 analyses, results show that large P quantities are lost in industrial, consumption and waste
 232 management routes (e.g., wastewater and biodegradable solid wastes). These are important output
 233 flows from the system, in addition to the P not used or lost in the agricultural sector by accumulation
 234 in soils and leaching/runoff/erosion processes

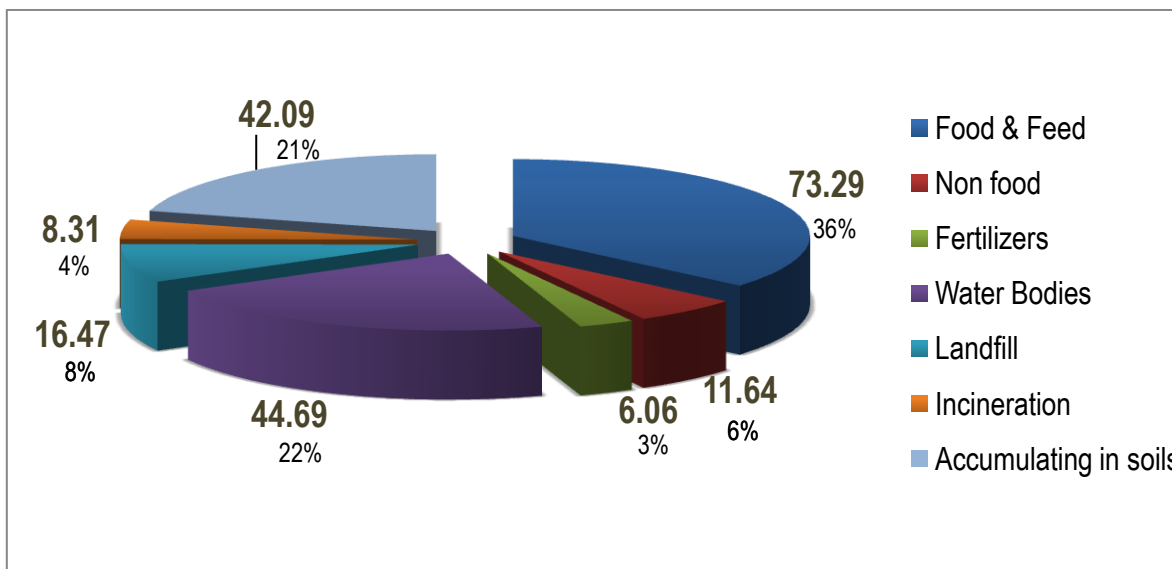
235 The animal farming stock remains constant, the P input to animals consisted of P within grasses during
 236 animal grazing (79.85 kt P/y) and P within animal feed (77.52 kt P/y), giving a total input of 157.37 kt P/y.
 237 The main outputs from animals are manure and animal products, which are estimated to contain 133.6
 238 and 23.76 kt P/y, respectively. The efficiency of the animals system converting P inputs into animal

239 products is only around 15%, thus, indicating that 85% of the inputs become manure.

240 The food and feed system receives inputs from agriculture as crops and animal products, as well as
241 imported food and feed materials. The total inputs are 296.28 kt P/y and the output is 223 kt P/y. The
242 difference between both flows suggests an annual stock of 73.28 kt P/y, and this indicates its
243 accumulation within this system. This accumulation may be due to the partial information that justifies the
244 output of P towards other processes; it may also be related to the generation of food stock as cereals,
245 due to market needs or interests. Animal feed was estimated at 77.52 kt P/y and it is the largest output
246 followed by human food consumption with 73.76 kt P/y. It is estimated that the Spanish system
247 generates around 22.96 kt P/y as waste, giving an efficiency of 92 %.

248 **3.2 P accumulation and losses**

249 P in Spain is accumulated in two different ways; on one hand P accumulated in market stocks of food
250 and feed, fertilizers and non-food, accounting for a total of 90.99 kt P/y, and on the other hand P lost to
251 the environment through incineration, landfill, losses to water bodies and land accumulation, which
252 accounts to a total of 111.56 kt P/y. The largest proportion of losses to environment is due to the losses
253 to water bodies, accounting 44.69 kt P/y, followed by agricultural land accumulation with 42.09 kt P/y as it
254 is shown in Figure 4. The main contributor of P losses to water bodies is the WWTPs treated effluents,
255 estimated in 31.72 kt P/y, which represents around 71 % of total losses to natural water bodies.



256

Figure 4. P accumulation and losses according to PFA in Spain.

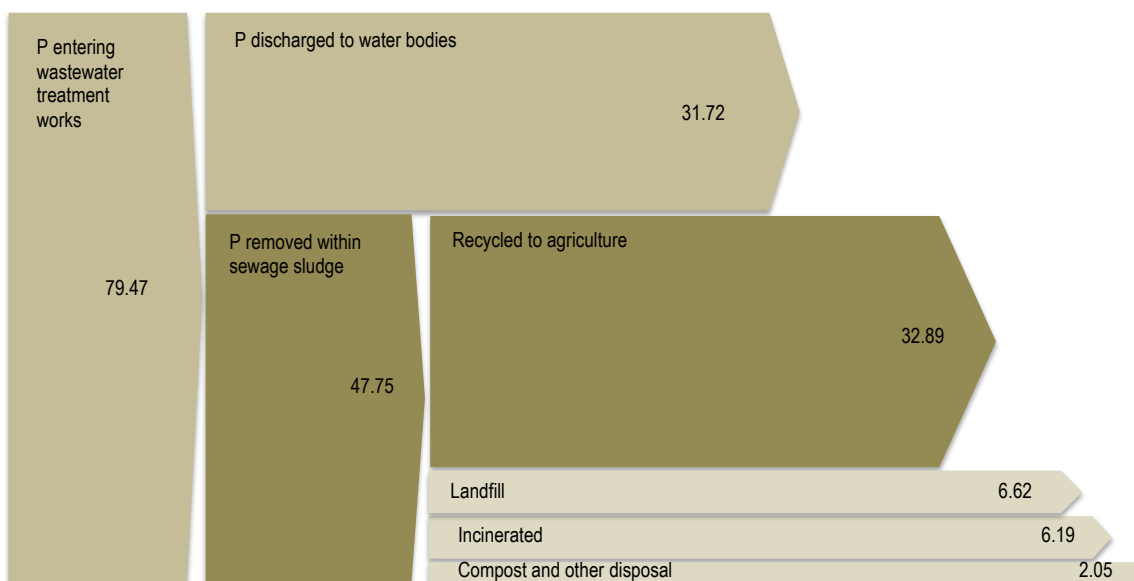
257 The P accumulation in the soils is estimated to be around 42.09 kt P/y, which is equivalent to 12 % of the
 258 total input to agricultural land, or equivalent to 26% the amount of P within mineral fertilizer uses. The
 259 loss of P through landfill was estimated around 24.78 kt P/y. The largest proportion of this amount is
 260 within urban organic waste landfilled, which is estimated to contain up to 16.47 kt P/y. Sewage sludge
 261 and waste incineration ash is assumed to be disposed also to landfill with 6.19 and 2.12 kt P/y
 262 respectively.

263

264 3.3 P flows affecting environmental compartments.

265 P is an essential resource and should also be considered as a pollutant. As agricultural fertilizers
 266 enhance crop growth, increased nutrient loading to aquatic ecosystems promotes eutrophication
 267 leading to an abundance of algae and aquatic plants, thereby causing a detrimental impact on the
 268 local ecosystem. Freshwaters are generally more responsive to P increases than other nutrients;

269 hence P is considered the main driver of eutrophication in lakes, rivers and estuaries of Spain. P
 270 enters water bodies through point sources, such as final effluent discharges from WwTw (more than
 271 2900 in Spain (MAPAMA, 2016f)) and through non-point or diffuse sources, such as soil erosion from
 272 agricultural fields or surface runoff of fertilizers and inappropriate use of animal manures (MAPAMA,
 273 2016g). The water treatment industry receives P loads within wastewaters up to 79.47 kt P/y, which is
 274 either discharged to natural water bodies or removed after transfer to sewage sludge. It is estimated that
 275 around 31.72 kt P/y is discharged (Figure 5) into the treated effluent, indicating a P removal efficiency of
 276 around 61 %.



277

278 Figure 5. Phosphorous flows through Spanish wastewater treatment works (WwTw) kt P/y according to
 279 PFA.

280 Approximately 69 % of the 47,75kt P/y removed within sewage sludge is recycled in agriculture uses and
 281 27 % is directed to landfill and composting and 4 % is sent to other uses. The P flow recycled to
 282 agriculture via the treated sewage sludge represents up to 10 % of the total P input to agricultural uses.

283 Accordingly, phosphorus management options in Spain should increase the efficiency and reduce the
284 losses with a special focus centered on the reduction of the P discharges to water bodies (31.72 kt
285 P/y). Such improvements could help to control both the environmental and resource scarcity issues
286 that are associated with the phosphorus challenge (García Albacete et al., 2012; Garrido-Baserba et
287 al., 2015). On a regional level, P balance studies have focused on the river basin levels that are
288 related to the P losses to surface waters and on the identification of eutrophication risk (Buzás, 1999;
289 De Wit and Behrendt, 1999; Delgado and Scalenghe, 2008; Torrent et al., 2007; Ulen et al., 2007). In
290 general, studies have been limited to quantifying the inputs and outputs of a system, but they have
291 not been able to emphasize the nature of flow of the nutrients (N and P) through the system. In
292 contrast, PFA studies focus on P flows throughout entire society and take different sub-sectors and
293 internal flows into consideration.

294 It should be highlighted that Spain is a net P importer providing a weak position in terms of
295 sustainability due to the risk of supply of this raw material and in consequence its agriculture and
296 economic system dependence on external markets. The current P cycle is inefficient compared with
297 other European and non-European countries with large quantities lost in industrial, consumption,
298 waste management routes and P accumulation in soils. It is well known that the increasing long-term
299 phosphorus demand would likely require demand management measures to reduce business-as-
300 usual demand by two-thirds, and the remaining third could be met through a high recovery rate of P
301 from human excreta, manure, food waste and mining waste (Cordell et al., 2011). The results indicate
302 that P cycle in Spain has a huge potential for improvement by defining more effective methods of
303 resource recycling and by promoting policies aimed at designing a sustainable P cycle considering
304 other global environmental and social challenges, including: climate change, energy supply, water
305 scarcity, land-use changes, population growth and urbanization trends.

306 **3.4 Assessment of PFA of Spain with other countries.**

307 Cooper and Carliell-Marquet (2013) and Li et al. (2016) have used indicators of new sustainable
 308 phosphorus practices that could be useful for monitoring changes over time, defining and comparing
 309 performance across countries. The main indicators that have been identified in the literature (fertilizer
 310 application, mineral fertilizer application, human consumption, wastewater flow and total input and out-
 311 put person flows) are summarized in Table 3.

Table 3. Definition of P flow indicators (Cooper and Carliell-Marquet, 2013) and (Li et al., 2016).

Indicator	Definition	Units
Fertilizer application	P fertilizer application on agricultural land	kg/ha
Mineral fertilizer application	Ratio p mineral fertilizer and total P fertilizer applied on agricultural land	%
Human consumption	P utilization from food and non-food commodities	kg/cap
WWTP effluent	Average P discharged in wastewater treatment plant	kg/cap
Total input	Average total input to the country per person	kg/cap
Total output	Average total output from the country per person	kg/cap
Exported food	Average P export through food per person	kg/cap
Landfill	Average P being landfilled in the country	kg/cap
Manure application	Animal manure applied to agriculture land	kg/ha
Agriculture loss	P loss from agriculture land to streams	kg/ha
Land accumulation	P accumulated in agricultural land	kg/ha
Soil in	P input to agricultural land	kg/ha
Soil out	P output from agricultural land	kg/ha
Agricultural efficiency	Ratio P input and output in agriculture	%
P recovery	P recovered from sewage in WWTP	%

312 The PFA results have been reported using total quantities of P (in kt P) and were further normalized in
 313 terms of population (kg P/cap), land area (kg P/ha) or efficiency estimates (%). Using these ratios it is
 314 possible to compare between countries, territories or cities with different population sizes and agricultural
 315 areas. A selection of the main published PFA from Japan, USA, Oceania (Australia and New Zealand)
 316 and Europe (France, Finland, United Kingdom, Denmark) and a study from the EU27 are summarized in
 317 Table 4.

Table 4. A comparison of P flows and selected indicators for different countries and regions.

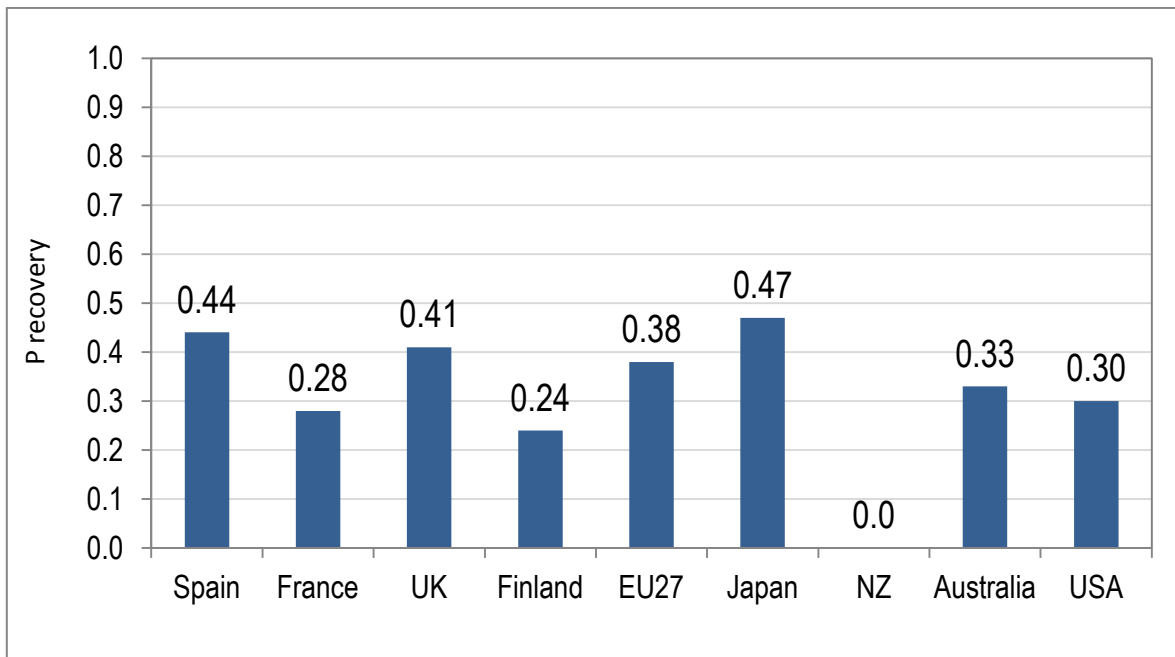
Indicator	Units	Spain	France	UK	Finland	EU27	Japan	NZ	Australia	USA
		a	b	c	d	e	f	g	h	i
Fertilizer application	kg/ha	22.89	10.00	4.00	14.90	6.75	87.00	18.00	1.00	4.00
Mineral fertilizer application	%	0.52	0.37	0.27	0.61	0.42	0.73	0.57	0.43	0.81
Human consumption	kg/cap	1.56	1.80	0.80	1.00	1.54	0.80	2.30	2.10	1.20
WWTP effluent	kg/cap	1.68	0.70	0.40	0.10	0.24	0.10	0.90	0.50	2.60
Total input	kg/cap	6.88	7.50	2.20	41.20	4.87	5.70	54.70	25.90	25.30
Total output	kg/cap	2.33	2.70	0.40	3.10	2.99	0.00	20.30	6.90	17.60
Exported food	kg/cap	0.54	2.10	0.30	0.50	0.49	0.00	10.40	5.20	1.30
Landfill	kg/cap	0.35	0.80	0.30	0.50	1.56	0.70	8.60	0.60	1.60
Manure application	kg/ha	9.72	10.60	9.70	8.60	9.17	31.60	13.20	1.20	1.00
Agriculture loss	kg/ha	0.89	1.60	0.80	1.10	0.44	8.60	4.70	0.10	2.00
Land accumulation	kg/ha	3.06	5.80	2.20	12.70	4.85	81.10	85.60	1.70	1.60
Soil in	kg/ha	25.60	26.70	15.60	24.90	17.45	118.80	31.40	2.20	7.80
Soil out	kg/ha	21.65	20.90	13.40	12.20	12.16	37.00	24.00	0.50	6.10
Agricultural efficiency	%	0.85	0.70	0.80	0.50	0.70	0.30	0.70	0.10	0.80
P recovery	%	0.44	0.28	0.41	0.24	0.38	0.47	0.00	0.33	0.30

Year	2012	2002-06	2009	1995-99	2005	2005	2012-14	2001	2007
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a: this study, b: (Senthilkumar et al., 2014), c: (Cooper and Cariell-Marquet, 2013), d: (Antikainen et al., 2005), e: (Ott and Rechberger, 2012), f: (Matsubae et al., 2011), g: (Li et al., 2016), h: (Cordell et al., 2010), i: (Suh and Yee 2011).

318 The comparison as shown in Table 4 has the limitation of covering a time frame from 2001 to 2014,
319 which makes it difficult to draw conclusions; however some trend can be identified. PFA studies
320 specifically showed significant differences between countries and also between regions within
321 countries as discussed qualitatively by Schröder et al. (2011) and compared by Jedelhauser and
322 Binder (2015). The European PFA studies differ in data, methodology and outcomes. In any case,
323 national PFA studies provide positive inputs as they use national parameters, specific knowledge and
324 assumptions to quantify the flows, as well as country specific aspects such as agricultural system
325 types, food habits, and waste management policies.

326 A comparison of P recovery efficiency in Spain vis-à-vis other countries (data taken from Table 4) is
327 graphically represented in Figure 6. It is evident that regardless of the country studied, the P recovery
328 efficiency is below 50% with New Zealand showing a zero recovery. This indicator provides an
329 appropriate guide to identify the use of the main secondary source of P. In the case of Spain, this
330 parameter has reported 44% (0.44) above the average of EU27 (0.38) and UK with 0.41 and far away
331 from France with 0.28. This is mainly due to the different legislation within the EU27 member states
332 regarding the agricultural use of sludge from WWTPs.

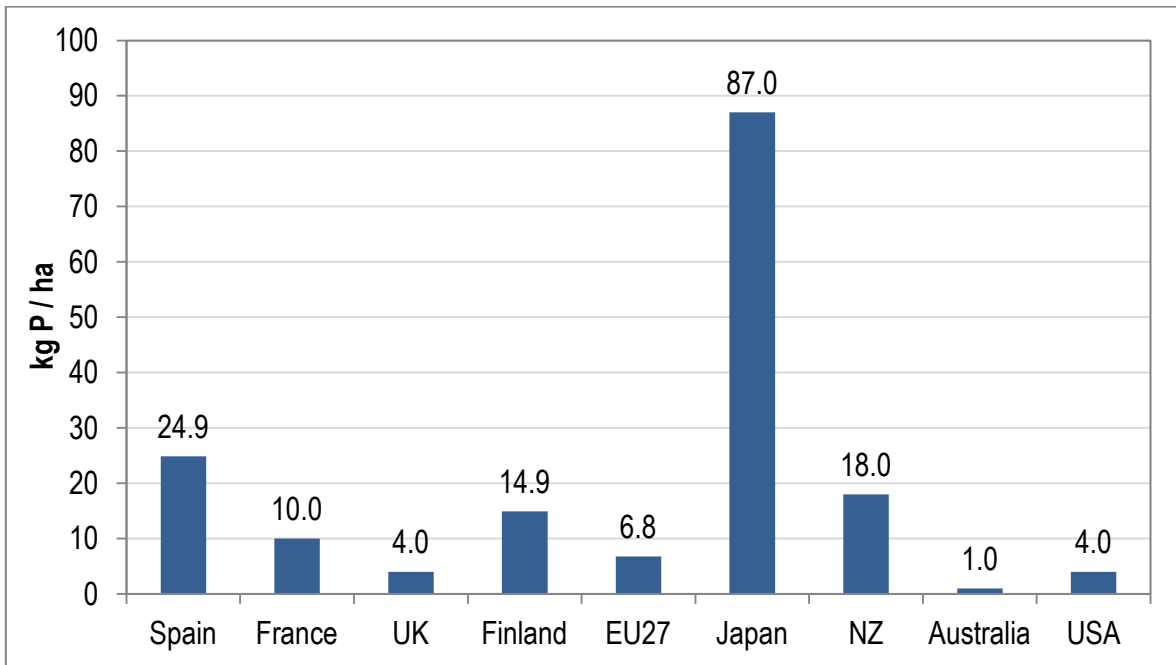


333
 334 Figure 6. Comparison of P recovery efficiency of Spain with model countries and regions (source data
 335 Table 4).

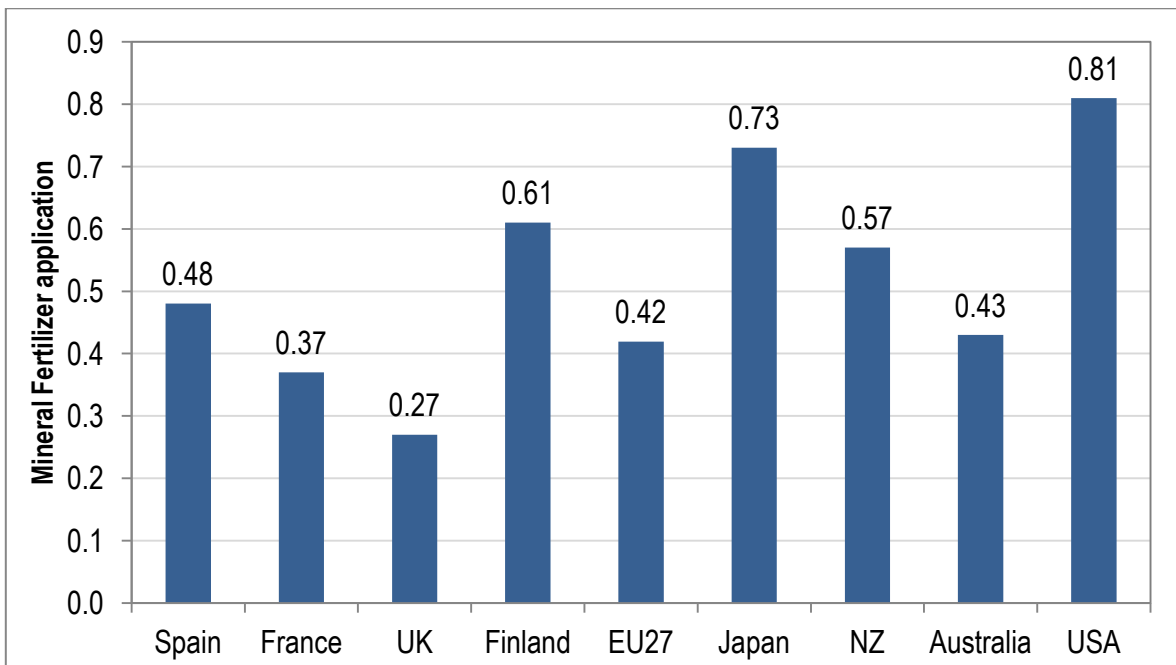
336 Figure A2 provides the country wise annual sludge potential from municipal WwTw in kg dry matter per
 337 hectare of utilized agricultural area and hence, it represents the theoretical recycling potential of sewage
 338 sludge in agriculture. The actual utilization is given in percentage of the total annual amount. For most
 339 member states corresponding data are available for 2008 or 2009. It can be seen that in Portugal, Spain
 340 and UK, highest proportion of sludge have been applied to agriculture which justifies its higher recovery
 341 rate than average in EU27. In the case of France and Finland, 50% and 25% of sludge has been applied
 342 to agriculture which is also reflected in the lower average ratios obtained. Country wise (EU member
 343 states) heterogeneity is observed in Figure A3 regarding the disposal of sludge from the WwTws.
 344 Basically, there are only five possible destinations for sludge, but each member state distributes sludge
 345 in different proportions as evident from Figure A3. Regarding the destination of the sludge, it is clear that
 346 there are three major trends depending on the sludge disposal.

347 The country wise use of fertilizers in terms of the application ratios (kg/ha) and the ratio of applied

348 mineral fertilizer to the total applied fertilizer are shown in Figure 7. Fertilizer uses indicate an overdosing
 349 trend in Spain up to 4 times the average for EU27 and only approximate the values reported for New
 350 Zealand and far away for the highest values reported for Japan.



351

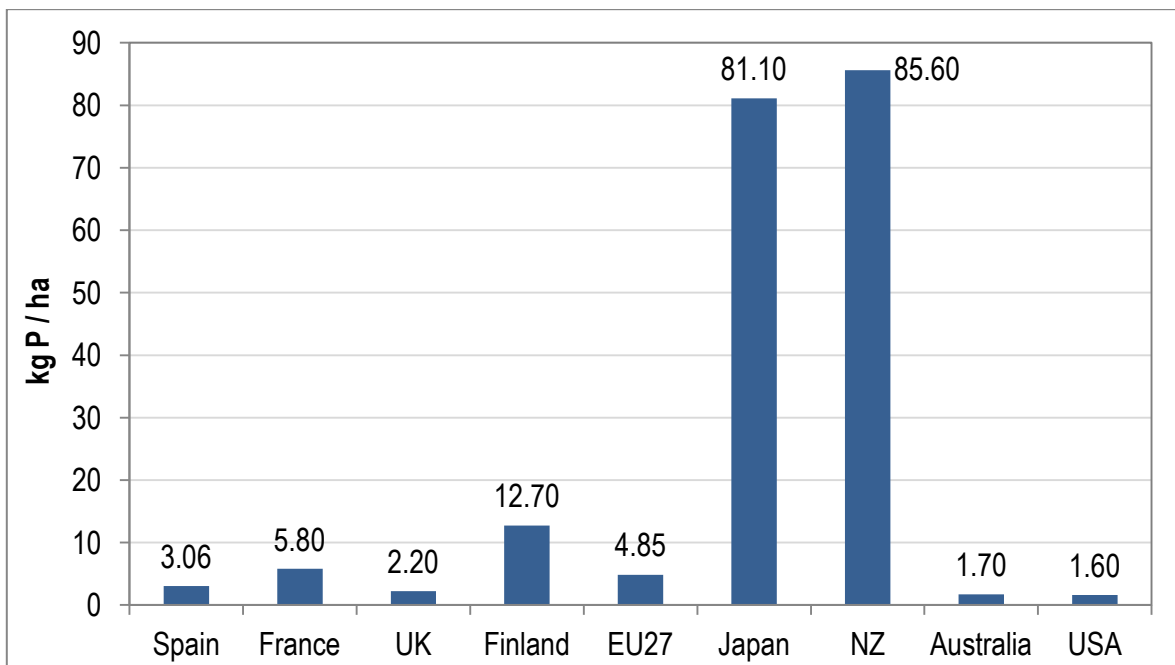


352

353 Figure 7. Comparison of a) fertilizer application (kg/ha) and b) mineral/total fertilizer application of Spain
354 with model countries and regions (source data Table 4).

355 While making a comparison in the case of the ratio of mineral to the total fertilizers, the performance of
356 Spain has approached the average value of the EU27, and is below the highest values reported for
357 countries where the use of sewage sludge in agriculture is reduced option as are the cases of Japan
358 USA or Finland in Europe (Meyer et al., 2015; Nelson and Janke, 2007).

359 The comparative data of the P accumulation on land with respect to different countries (as depicted in
360 Table 4) is shown graphically in Figure 8. In this sense, the performance of Spain is below the EU27
361 average and lower than those reported for France or Finland. The availability of P to the plants is highly
362 dependent on the properties of the soil, especially on the content of calcium, iron and aluminium rich
363 minerals. Although previously seen higher doses are applied, those values should be corrected by the
364 crops productivity by hectare.



365

366 Figure 8. Comparison of P land accumulation in Spain with model countries and regions (source data
367 Table 4).

Ott and Rechberger (2012) found that the annual net per capita P use in the EU-15 was 4.7 kg, of which only 1.2 kg reached the consumer but after use, only a small level of recycling (0.77 kg P/cap/y) was reported. Large fractions of the surplus P accumulated in agricultural soils (2.9 kg P/cap/year) were either sequestered to landfills (1.4 kg P/cap/y) or were emitted to the hydrosphere (0.55 kg P/cap/y). The authors therefore proposed to promote optimization of P use as fertilizer, collection and recycling of P-rich wastes, to increase the connection of households to sewer systems, and to implement a tertiary wastewater treatment for the recovery/removal of P from the WwTw so as to reduce P rock imports significantly.

368 **3.5 Identification of alternative resources for P: application of circular economy opportunities to** 369 **the phosphorus cycle**

370 As has been pointed out by the European Sustainable Phosphorus Platform (ESPP, 2015), phosphorus
371 use in EU-27 was characterized by “5 Ls”: a) large dependency on (primary) imports, b) long-term
372 accumulation in agricultural soils, especially in west European countries, c) leaky losses throughout
373 entire society, especially emissions to the environment and sequestered waste, d) little recycling with the
374 exception of manure, and sewage sludge and e) low use efficiencies, because of aforementioned issues,
375 providing ample opportunities for improvement. Applying the strategy established by the circular
376 economy to the P cycle it is possible to reduce the demand for primary phosphorus reserves by
377 managing to cover it with secondary sources as described by Cordell et al. (2011); EC (2014), and Liu et
378 al. (2008).

379 One of the most valued utilities from the analyses of the cycle of phosphorus is to identify and to quantify

380 across flows, the primary, secondary and tertiary sources presented in circular economy model. By
 381 means of these analyses we can determine the previous losses of the system, and to take necessary
 382 steps to reduce such losses or re-use them as secondary sources. The results can be used to determine
 383 the performances of the different processes presents in P cycle so as to assign potential improvements
 384 and reductions in consumption, and finally detect possible tertiary sources. Primary, secondary and
 385 recommended actions to promote tertiary sources are identified and collected according to Spain PFA,
 386 EU27 (Ohtake and Okano, 2015) and Japan (Van Dijk et al., 2016) in Table 5.

Table 5. P primary, secondary and recommended actions to promote tertiary sources from cycles of Spain, EU27 and Japan.

Country /Region	Primary sources		Secondary sources		Recommended actions to promote tertiary sources
	Flow	kt P/y	Flow	kt P/y	
Spain	1.Phosphate rock and Mineral fertilizers	214.77 54.82	1.Sewage sludge 2.WWTP effluent 3.Organic waste	47.75 31.72	1.Improve the ratio of removal of P in WWTP that do not have tertiary treatment 2.Improve agricultural production performance 3.Waste less food, reducing food consumption and waste generation
	2.Food an feed import	55.80	4.Agricultural losses to water	34.83	
	3.Nonfood import			12.29	
EU27	1.Phosphate rock and Mineral fertilizers	1389.43 778	1.Sewage sludge 2.WWTP effluent 3.Organic waste	394.27 128.89	1.Improve the ratio of removal of P in WwTP that do not have tertiary treatment 1.Improve agricultural production performance
	2.Food an feed import	215	4.Agricultural losses to water	255.1	

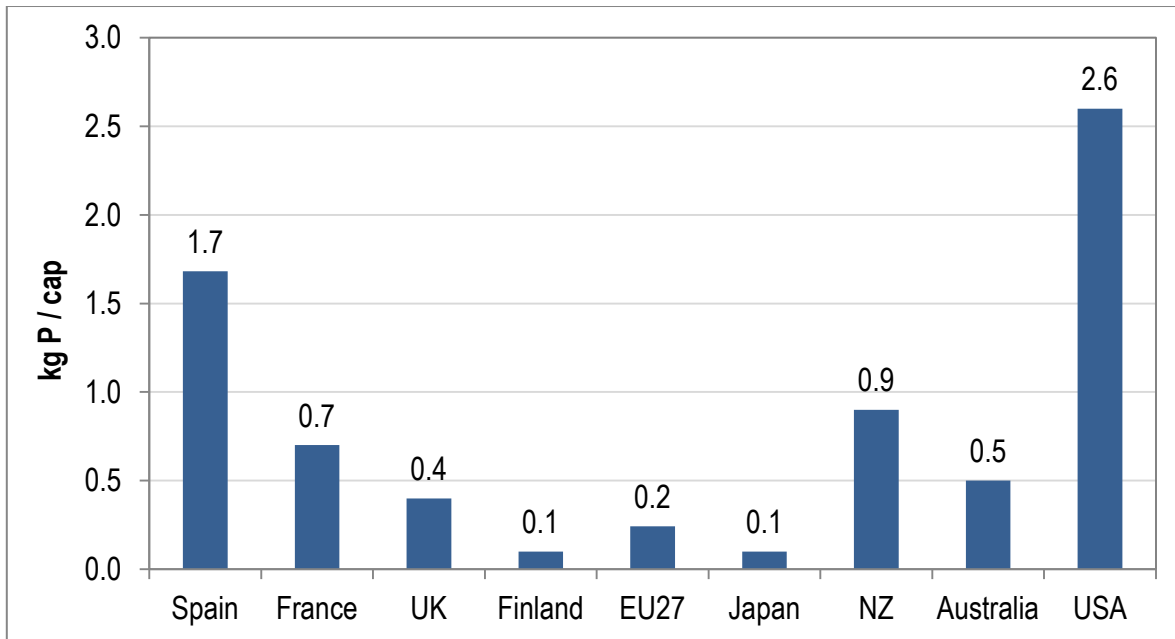
	3.Nonfood import		5.Food processing waste	84.49	2.Waste less food, reducing food consumption and waste generation
				389.95	
Japan	1.Phosphate rock, phosphorus compounds, yellow phosphorus	274	1.Sewage sludge	39.6	1.Improve the ratio of removal of P in WWTP that do not have tertiary treatment
	2.Mineral fertilizers		2.WWTP effluent	16.1	2.Improve agricultural production performance
	3.Food an feed import	163.1	3.Organic waste		3.Waste less food, reducing food consumption and waste generation
	4.Others minerals resources	274	4.Agricultural losses to water	44.1	
			5.Slag	31.5	
				104	

387 WWTPs are one of the most recurrent secondary sources as identified in Table 5 among all P cycles
388 analyzed and reviewed in this paper and on which more work is being done because of its potential as a
389 secondary source of P.

390 Secondary sources such as effluent from WWTPs also have shown a high magnitude, but this
391 magnitude is due to the large volume of treated water containing a very low concentration of P, about 2-
392 10 mg/L P. Consequently, efforts to reduce concentration of P in plants that already have a tertiary
393 treatment is nowadays an expensive management option. Furthermore, the volume of sludge generated
394 is less than the treated water, while the P concentration is up to 5 orders of magnitude greater than that
395 of water, about 19237 mg P/Kg (MAPAMA, 2016).

396 A country wise comparison of P losses to WwTw (Figure 9) reveals that losses through WwTw
397 discharges to water receiving bodies of Spain is far away from the values reported by Japan, up to 8
398 times higher than that of EU27 average, and only lower than USA (Figure 9). In the case of Spain with a

399 high costal line and a high presence of large cities close to coastal areas discharge limits to sea are not
400 so stringent. In the case of EU27, some countries as EU are pursuing levels of P below 1 mgP/L in the
401 same direction reported values for USA indicate that less stringent regulations have been applied
402 although some cities and states are reporting efforts to set-up, limits below 0.5 mgP/L.



403

404 Figure 9. Comparison of P losses through WWTP discharges to water receiving bodies of Spain with
405 model countries and regions (source data Table 4).

406 This result clearly indicates the need to improve the average P removal performance of the wastewater
407 treatment and the need for implementing tertiary treatment in plants having only secondary treatment, or
408 to increase the capacity of the operating treatment plants that have already implemented the tertiary
409 treatment but are saturated. In the case of Spain, the annual flow of P from wastewater treatment plant
410 sludge is 47.75 kt. The generation of this sludge is located near urban centers and is proportional to the
411 capacity of the WwTw, which is expressed in population equivalent (P.E.). In Spain there are about 2500
412 wastewater treatment plants, which in 2009 stripped a load of 68777 P.E. of a total of 2320 urban

413 agglomerations (National Plan for Water Quality: Sanitation and Treatment 2007-2015). It should be
414 pointed out that other alternatives can be explored to recover P from sewage sludge. For example the
415 implementation of MAP technology of crystallization, an emerging technology that is under development
416 throughout Europe, especially in Netherlands, Germany, Belgium and United Kingdom, but is having a
417 limited development in Spain. Only recently, the first crystallization MAP plant has been installed on a
418 WwTw in Madrid.

419 It is worth to mentioning the case of Japan that has essentially no reserves of P rock, but sufficient
420 amounts of secondary P resources has been identified such as food waste, animal manure, sewage
421 sludge, and steelmaking slag. Additionally, P recycling has been implemented in several WwTw and
422 black water treatment plants. Approximately 2000 WwTw and 1000 black water treatment plants are
423 operating in Japan, their operation has been considered as the beginning of the P sustainability
424 initiative. It is expected in the next decade the recovery of high-grade phosphoric acid and, therefore,
425 high-purity elemental P ($P_4(s)$) will be required for food safety and quality control of industrial
426 products. To accomplish P recycling, it is necessary to develop P refinery technology, which enables
427 the production of high-purity phosphoric acid and elemental P from a wide variety of secondary
428 resources.

429 **Conclusions**

430 In this study, P flows were analyzed and quantified in detail for the first time for the country of Spain
431 in accordance with the PFA results were compared with the European Union (EU-27) and were
432 additionally compared with significant countries in different continents. The results show how P is
433 used, reused and lost in Spain and the results further indicate the effective options for more
434 sustainable P management. The study also compared flows and P use efficiencies between sectors

435 and countries. In a vital first step towards creating such a system, the PFA has mapped and
436 quantified the relevant stocks and flows, allowing to identify specific measures to be implemented
437 against specific losses and areas of inefficient use of resource (forestry, agriculture, human
438 consumption, waste and wastewater management and urban soils).

439 The results from this analysis suggest focusing on P removal and recovery from WwTw (in both water
440 and sludge streams), as well as the developing of more effective methods for recycling industrial and
441 farming wastes such as animal manure, sewage sludge and food waste and sewage sludge in order
442 to reduce soil accumulations and replace imported fertilizers. This study has demonstrated the need
443 for an integrated assessment of P flows between sectors (such as fertilizer, food and sanitation sectors)
444 at the national/regional scale in order to reduce losses and increase the efficiency, recovery and reuse of
445 P within the food system. Understanding the key P inflows and outflows related to a particular
446 geographical scale is vital to identify the priority areas of P management for that scale. Conducting
447 additional PFAs at smaller scales (Spanish autonomous regions or metropolitan areas) may be
448 necessary in order to develop more specific measures, such as regional recycling strategies. Future
449 problems as it could be the increasing use of urban sewage sludge as fuel co-substitution option in
450 cement industry should be addressed from the perspective of the new EU regulations on P.

451

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