### S1. N outputs and crop yields for the Ebro crops considered in this study. Mean value yield observed in the 18 provinces and their standard deviations (see the specific comments below).

<table>
<thead>
<tr>
<th>Type</th>
<th>Crop</th>
<th>Yield ± SD Rainfed cultures (Tn/ha)</th>
<th>Yield ± SD Irrigated cultures (Tn/ha)</th>
<th>Yield ± SD Greenhouse cultures (Tn/ha)</th>
<th>N output (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cereal</td>
<td>Wheat</td>
<td>3.4 ± 1.3</td>
<td>4.7 ± 2.1</td>
<td></td>
<td>1.81</td>
</tr>
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<td>3.2 ± 1.2</td>
<td>4.5 ± 2.0</td>
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<td>2.5 ± 1.1</td>
<td>4.0 ± 2.4</td>
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<td>3.3 ± 1.7</td>
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<td>4.0 ± 2.2</td>
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<td>6.0 ± 3.1</td>
<td></td>
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<td>Legumes</td>
<td>Chickpea</td>
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<td>1.4 ± 0.8</td>
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<td>Legumes</td>
<td>Pea</td>
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<td>Legumes</td>
<td>Lupin</td>
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<td>1.2 ± 0.3</td>
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<td>2.0 ± 1.0</td>
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<td>2.00</td>
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<td>9.0 ± 3.4</td>
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<td>Table grape</td>
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<td>10.1 ± 5.7</td>
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<td>0.34</td>
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<td>Vine grape</td>
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<td>2.0 ± 0.8</td>
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<td>Oil olive</td>
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<td>Arpicot</td>
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<td>Cherry</td>
<td>3.1 ± 1.1</td>
<td>5.8 ± 3.1</td>
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<td>Type</td>
<td>Crop</td>
<td>Yield ± SD Rainfed cultures (Tn/ha)</td>
<td>Yield ± SD Irrigated cultures (Tn/ha)</td>
<td>Yield ± SD Greenhouse cultures (Tn/ha)</td>
<td>N output (%)</td>
</tr>
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<td>-----------------</td>
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<td>7.2 ± 4.1</td>
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<td>7.8 ± 1.8</td>
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<td>0.30</td>
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<td>0.57</td>
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<td>0.57</td>
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<td>3.0 ± 2.7</td>
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<td>Lettuce</td>
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<td>31.3 ± 19.6</td>
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<td>Vegetables</td>
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<td>33.8 ± 8.3</td>
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<td>Type</td>
<td>Crop</td>
<td>Yield ± SD</td>
<td>Yield ± SD</td>
<td>Yield ± SD</td>
<td>N output (%)</td>
</tr>
<tr>
<td>------------</td>
<td>-------------------------------------</td>
<td>------------</td>
<td>------------</td>
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<tr>
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<td>Rainfed cultures (Tn/ha)</td>
<td>Irrigated cultures (Tn/ha)</td>
<td>Greenhouse cultures (Tn/ha)</td>
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<td>Fodder</td>
<td>Whinter cereals (Forage)</td>
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<td>22.9 ± 13.0</td>
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<tr>
<td>Fodder</td>
<td>Maize (Forage)</td>
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<td>Sorghum (Forage)</td>
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<td>55.1 ± 10.3</td>
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<td>Vicia sativa (Forage)</td>
<td>17.1 ± 8.6</td>
<td>26.6 ± 13.7</td>
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<td>Fodder</td>
<td>Hedysarum coronarium</td>
<td>7.0 ± 1.6</td>
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<td></td>
<td>0.45</td>
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<td>Fodder</td>
<td>Other legume (Forage)</td>
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<td>19.6 ± 9.7</td>
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<td>21.5 ± 12.1</td>
<td>33.2 ± 16.9</td>
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<td>0.23</td>
</tr>
<tr>
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<td>Beet (Forage)</td>
<td>25.3 ± 15.2</td>
<td>41.5 ± 21.2</td>
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</tr>
<tr>
<td>Fodder</td>
<td>Cabbage (Forage)</td>
<td>16.0 ± 8.7</td>
<td>32.3 ± 15.7</td>
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<td>0.23</td>
</tr>
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<td>Fodder</td>
<td>Avena fatua</td>
<td>28.2 ± 17.4</td>
<td>51.2 ± 20.5</td>
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<td>0.21</td>
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<tr>
<td>Fodder</td>
<td>Other gramineae (Forage)</td>
<td>11.8 ± 4.3</td>
<td>25.5 ± 9.3</td>
<td></td>
<td>0.21</td>
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<tr>
<td>Fodder</td>
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<td>27.9 ± 8.2</td>
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<td>Onobrychis viciifolia</td>
<td>12.9 ± 5.5</td>
<td>24.7 ± 5.5</td>
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<tr>
<td>Pasture</td>
<td>Cultivated grassland</td>
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</table>

Notes:

Yield corresponds to the averaged harvested yield of each crop in the 18 provinces present in the Ebro River Basin. Data is split into rainfed, irrigated and greenhouse cultures. The information was provided by the Spanish Ministry of Agriculture (http://www.mapa.es/). To create the spatialized output map, we used the information on yields for each corresponding province. SD: Standard Deviation among the provinces, showing the spatial variability within the Ebro river basin.

N outputs in harvested straw have also been calculated for Cereals and Legumes (0.35% and 0.75%, respectively).

In the permanent crops, 100% of trunks, 50% of roots and 50% of leaves (if perennial) have been considered as outputs.

Yields in fodder crops are expressed as humid weight.

Yields in pastures are expressed as dry-weight. N output in natural pastures is lower than that in cultivated grassland because not all the production is finally harvested (a value of 60% consumption has been considered).

N output (%) expressed the N content in the harvested biomass. This information is based on Urbano (2002) and, when necessary, it has been completed with MMARM (2010). To calculate the actual N output, the following relationship was used: N output (kg N/ha) = crop yield (kg/ha) * (N output (%)/100).
S2. N fixation equation

Biological N\textsubscript{2} fixation by legumes is a difficult term to be accurately assessed. It is of current practice to use general figures by crop, which can however overestimate N fixation in low productive crops and underestimate it in high-yield crops. We developed a formula that relates total N fixation by a legume crop to crop yield, includes non-harvested residues and underground biomass, and takes into account the fact that, in the period prior to nodulation, N is obtained by legumes from mineral nitrogen present in the soil, while only after nodulation is achieved, N is progressively assimilated from N\textsubscript{2} fixation. The relationship is the following:

\[ N \text{ fix} \text{ (kg N/ha/yr)} = \alpha \times N_{\text{yield}} - A \]

where \( N_{\text{yield}} \) is the harvested biomass expressed in N content (kg N/ha/yr); \( \alpha \) is a coefficient expressing the ratio of total biomass produced with respect to harvested biomass (a typical value of \( \alpha \) is 1.4, Carlsson and Huss-Daniel 2003) and A is the amount of N taken up by the legume crop from the soil mineral N pool prior to nodulation. We approximated the latter term as the amount of mineral fertilizers applied to the legume crop (between 12 and 100 kg N/ha depending on the crop).

S3. Provinces statistics and CORINE CLC assemblages

First of all, we gathered all the information on crop yields and crop surfaces for the year 2000 provided by the Spanish Ministry of Agriculture (http://www.mapa.es/). This information is given by province. Secondly, we assigned to each crop from the Spanish agricultural statistics its corresponding category in CLC, e.g. barley, triticale and wheat were assigned to CLC rainfed herbaceous category (code 21100). Third, knowing the proportion of each type of crop within each type of CLC category per province, and taking into account their related N inputs and outputs, we obtained the characteristic N fluxes of each CLC category in each different province. Based on this, we created the map in Fig. 2 and Suppl. Mat. 5 where each CLC polygon contains the precise information on N inputs and outputs that correspond to the real crop proportion of the province where it is located. Once we have such map with precise spatial information on N inputs and outputs, we overlay either the TUs layer or the sub-catchments layer and we calculate the N budgets within each different polygon (polygons may correspond to a specific TU or to a particular sub-catchment). Finally, we use the relationship between retention and proportion of reservoirs and channels obtained with data from the 21 sub-catchments to assess the retention in each TU (see Table 3).

S4. Additional maps

Spatialized N inputs of synthetic fertilizers in the Ebro River Basin.
Spatialized N inputs by biological nitrogen fixation (BNF) in the crops of the Ebro River Basin.

Spatialized N inputs of manure in the crop lands of the Ebro River Basin.

Spatialized N inputs by deposition of reduced N forms in of the Ebro River Basin. From EMEP database for 2000 (http://www.emep.int/)
Spatialized N inputs by deposition of oxidized N forms in the Ebro River Basin. From EMEP database for 2000 (http://www.emep.int/)

Spatialized N inputs by point sources at municipal level in the Ebro River Basin.

S5. Point source estimations

To estimate inputs from the population we used data on population density for 2001 from the Spanish National Institute of Statistics (http://www.ine.es) and the inventory of wastewater treatment plants (WWTP) provided by the CHE (http://www.oph.chebro.es), which includes the type of depuration technique performed, the capacity and the period of activity. The population was considered to emit 5 kgN/inhab/y, based on diet statistics by FAO.

First of all, we applied a generic reduction of N emissions to the data set comprising all the population without WWTP services and to the indirect industrial emissions. This generic reduction rate was applied in order to simplify subsequent calculations for population with WWTP, a reduction in N emission depending on the type of treatment was applied. This reduction varied from 15% for basic treatments to 60% for some techniques with nutrient reduction. A 0% reduction rate was applied to inhabitants connected to collection systems but without WWTP and we assumed 50% reduction for people not connected to any collection system. All these population emissions were spatialized coupling an Excel sheet to a GIS layer with municipalities and calculating the emission per area for every municipality polygon.
Indirect industrial effluents were included and spatialized in WWTP surpluses. Direct industrial effluents for 2001 were completed with data from the European Pollutant Release and Transfer Register (www.prtr-es.es). Finally, these emissions from PRTR were added to the GIS layer with municipality emissions and the total emission per area for each municipality polygon was calculated.

S6. Sub-catchments information

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<th>Ebro/Tributary</th>
<th>X Coordinate</th>
<th>Y Coordinate</th>
<th>Channel Index (km/km²)</th>
<th>Drainage reservoirs (%)</th>
<th>N Retention (%)</th>
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