Supplementary Material

Lassaletta et al. (2011) Biogeosciences

		Yield ± SD Rainfed cultures	Yield Irrigated	Yield ± SD Greenhouse cultures	N output
Туре	Crop	(Tn/ha)	cultures (Tn/ha)	(Tn/ha)	(%)
Cereal	Wheat	3.4 ± 1.3	4.7 ± 2.1		1.81
Cereal	Barley	3.2 ± 1.2	4.5 ± 2.0		1.51
Cereal	Oats	2.5 ± 1.1	4.0 ± 2.4		1.51
Cereal	Rye	2.3 ± 1.1	3.3 ± 1.7		1.51
Cereal	Titricale	2.8 ± 1.4	4.6 ± 1.8		1.58
Cereal	Maize	4.0 ± 2.2	8.2 ± 3.0		1.36
Cereal	Sorghum	1.9 ± 1.0	6.0 ± 3.1		1.66
Cereal	Rice		5.7 ± 1.4		1.48
Legumes	Beans	0.9 ± 0.5	1.8 ± 1.0		4.50
Legumes	Broad bean	1.3 ± 0.4	2.4 ± 1.3		4.00
Legumes	Lentil	0.8 ± 0.4	1.3 ± 0.7		3.40
Legumes	Chickpea	0.9 ± 0.4	1.4 ± 0.8		3.40
Legumes	Pea	1.5 ± 1.1	2.2 ± 1.2		3.80
Legumes	Vicia sativa	0.8 ± 0.4	1.5 ± 0.9		3.80
Legumes	Vicia ervilia	1.0 ± 0.7	1.8 ± 0.8		3.00
Legumes	Lupin	0.9 ± 0.3	1.2 ± 0.3		3.40
Roots	Potatoes	16.5 ± 8.6	29.9 ± 13.0		0.26
Industrial crops	Sugar beet	46.4 ± 18.4	67.9 ± 34.5		0.19
Industrial crops	Flax	1.0 ± 0.5	2.1 ± 1.2		3.29
Industrial crops	Hemp	1.6 ± 1.2	3.0 ± 1.7		3.29
Industrial crops	Sunflower	1.0 ± 0.5	2.2 ± 1.1		2.60
Industrial crops	Soy	1.1 ± 0.3	4.4 ± 1.0		6.00
Industrial crops	Rape	1.4 ± 0.8	2.0 ± 1.2		4.03
Industrial crops	Padrón Pepper	1.0 ± 0.2			0.26
Industrial crops	Saffron		0.0 ± 0.0		1.20
Industrial crops	Нор	1.4 ± 0.3			2.00
Industrial crops	Tobacco		2.5 ± 1.0		4.00
Permanent	Table grape	4.2 ± 2.5	9.0 ± 3.4		0.34
Permanent	Vine grape	5.8 ± 2.6	10.1 ± 5.7		0.34
Permanent	Table olive	0.9 ± 0.6	2.0 ± 0.8		0.81
Permanent	Oil olive	0.8 ± 0.7	1.8 ± 1.1		0.81
Permanent	Apple	7.4 ± 5.0	19.3 ± 9.1		0.15
Permanent	Pear	5.9 ± 3.3	15.6 ± 8.3		0.15
Permanent	Loquat	6.0 ± 1.4	10.1 ± 4.1		0.32
Permanent	Other pip	3.8 ± 2.1	7.9 ± 4.5		0.15
Permanent	Arpicot	5.7 ± 3.1	10.9 ± 6.5		0.22
Permanent	Cherry	3.1 ± 1.1	5.8 ± 3.1		0.39

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).

CORC SI(1)	Cont.	S1	(1)
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		Yield ± SD Rainfed	Yield Irrigated	Yield ± SD Greenhouse	N
Type	Crop	cultures (Tn/ha)	(Tn/ha)	cultures (Tn/ha)	N output (%)
Permanent	Peach	5.7 ± 3.6	16.2 ± 9.3	. ,	0.22
Permanent	Plum	3.8 ± 2.5	9.3 ± 6.6		0.22
Permanent	Fig	4.6 ± 2.4	7.2 ± 4.1		0.22
Permanent	Pomegranate	1.2 ± 0.3	5.7 ± 1.9		0.30
Permanent	Kiwi	0.8 ± 0.2	15.2 ± 10.3		0.30
Permanent	Other fleshy	7.8 ± 1.8	12.3 ± 5.2		0.30
Permanent	Almond	0.7 ± 0.6	1.8 ± 1.0		0.57
Permanent	Hazelnut	0.7 ± 0.4	1.0 ± 0.5		0.57
Permanent	Walnut	1.5 ± 1.4	3.0 ± 2.7		0.57
Permanent	Carob	1.9 ± 0.9	2.5 ± 0.6		0.57
Permanent	Orange		16.5 ± 7.6		0.44
Permanent	Bitter orange		7.0 ± 1.6		0.44
Permanent	Manderin		20.1 ± 8.2		0.44
Permanent	Lemon		15.1 ± 8.8		0.44
Permanent	Grapefruit		14.0 ± 3.3		0.44
Permanent	Avocado		11.5 ± 2.7		1.10
Vegetables	Cabage	17.1 ± 9.7	28.9 ± 9.8		0.63
Vegetables	Aspargus	2.9 ± 1.3	4.7 ± 2.6		0.30
Vegetables	Lettuce	16.3 ± 8.9	31.3 ± 19.6	55.7 ± 32.7	0.27
Vegetables	Escarole	13.3 ± 6.0	23.8 ± 10.4	34.3 ± 13.2	0.36
Vegetables	Espinach	8.8 ± 4.6	18.4 ± 8.8		0.27
Vegetables	Swiss chard	12.0 ± 6.0	25.6 ± 14.1	73.8 ± 36.8	0.45
Vegetables	Watermelon	9.6 ± 5.7	28.6 ± 14.8	35.5 ± 11.5	0.18
Vegetables	Melon	7.3 ± 3.8	22.4 ± 12.1	24.4 ± 8.0	0.21
Vegetables	Cucumber	15.3 ± 7.3	29.2 ± 15.3	76.5 ± 44.3	0.14
Vegetables	Courgette	12.7 ± 5.6	29.5 ± 17.1	59.6 ± 26.8	0.35
Vegetables	Aubergine	5.9 ± 2.3	26.0 ± 13.9	30.0 ± 7.1	0.52
Vegetables	Tomato	13.0 ± 8.2	38.0 ± 10.9	72.5 ± 39.0	0.22
Vegetables	Pepper	10.2 ± 5.1	17.9 ± 6.1	41.7 ± 23.4	0.26
Vegetables	Strawberry	4.7 ± 1.8	10.2 ± 5.4	25.1 ± 11.8	0.39
Vegetables	Artichoke	5.4 ± 1.7	13.8 ± 7.6		0.32
Vegetables	Cauliflower	15.3 ± 8.1	20.8 ± 8.8		0.69
Vegetables	Garlic	5.6 ± 3.4	10.7 ± 5.3		0.69
Vegetables	Onion	12.5 ± 6.8	33.8 ± 8.3		0.29
Vegetables	Carrot	15.2 ± 7.6	40.7 ± 26.0		0.49
Vegetables	Mushroom	1.3 ± 0.6			0.68
Vegetables	Green bean	6.4 ± 1.9	12.6 ± 4.9	26.3 ± 14.5	0.41
Vegetables	Green pea	4.8 ± 2.4	8.0 ± 4.1	13.0 ± 3.1	0.50
Vegetables	Green broad been	5.4 ± 2.7	9.0 ± 4.7		0.77

Cont. S1 (2)

		Yield ± SD Rainfed cultures	Yield Irrigated cultures	Yield ± SD Greenhouse cultures	N output
Туре	Crop	(Tn/ha)	(Tn/ha)	(Tn/ha)	(%)
Fodder	Whinter cereals (Forage)	13.4 ± 8.2	22.9 ± 13.0		0.19
Fodder	Maize (Forage)	34.4 ± 21.7	56.6 ± 26.2		0.20
Fodder	Sorghum (Forage)	16.5 ± 8.4	42.7 ± 20.2		0.20
Fodder	Alfalfa	24.7 ± 9.3	55.1 ± 10.3		0.56
Fodder	Vicia sativa (Forage)	17.1 ± 8.6	26.6 ± 13.7		0.45
Fodder	Hedysarum coronarium	7.0 ± 1.6			0.45
Fodder	Other legume (Forage)	12.5 ± 7.1	19.6 ± 9.7		0.45
Fodder	Turnip	21.5 ± 12.1	33.2 ± 16.9		0.23
Fodder	Beet (Forage)	25.3 ± 15.2	41.5 ± 21.2		0.15
Fodder	Cabbage (Forage)	16.0 ± 8.7	32.3 ± 15.7		0.23
Fodder	Avena fatua	28.2 ± 17.4	51.2 ± 20.5		0.21
Fodder	Other gramineae (Forage)	11.8 ± 4.3	25.5 ± 9.3		0.21
Fodder	Clover	19.5 ± 10.9	27.9 ± 8.2		0.45
Fodder	Onobrychis viciifolia	12.9 ± 5.5	24.7 ± 5.5		0.45
Pasture	Cultivated grassland	3.7 ± 1.2	6.2 ± 2.0		1.44
Pasture	Natural pasture	1.6 ± 0.5	6.2 ± 2.0		0.86

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 (<u>http://www.mapa.es/</u>). 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_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_2 fixation. The relationship is the following:

N fix (kg N/ha/yr) = α * Nyield – A

where Nyield is the harvested biomass expressed in N content (kg N/ha/yr); α is a coefficient expressing the ratio of total biomass produced with respect to harvested biomass (a typical value of α 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 (<u>http://www.mapa.es/</u>). 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 of the Ebro River Basin. From EMEP database for 2000 (http://www.emep.int/)



Spatialized N inputs by point sources at municipal level in of 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 (<u>http://www.oph.chebro.es</u>), 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 (<u>www.prtr-es.es</u>). 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.

EICA(nº				Channel Index	Drainage	N Retention
site)	Ebro/Tributary	X Coordinate	Y Coordinate	(km/km²)	reservoirs (%)	(%)
1	Ebro	503796.0	4726404.0	0.006	94.9	71
3	Tributary	586860.0	4692095.0	0.020	0.0	51
11	Ebro	670140.0	4619697.0	0.049	70.9	81
17	Tributary	779197.7	4602590.0	0.085	44.6	94
25	Tributary	785742.7	4594951.3	0.083	96.9	94
27	Ebro	796979.7	4524193.3	0.058	99.5	91
36	Tributary	539833.0	4685637.0	0.042	27.7	76
60	Tributary	642946.2	4641767.5	0.137	0.7	83
69	Tributary	599257.0	4738542.0	0.024	6.9	46
87	Tributary	654300.0	4623308.0	0.026	23.5	99
89	Tributary	679681.0	4615743.0	0.085	61.6	95
92	Tributary	468150.0	4738725.0	0.001	0.0	65
93	Tributary	463700.0	4730450.0	0.000	6.7	66
96	Tributary	816505.4	4634116.8	0.036	100.0	98
97	Tributary	797068.3	4642456.6	0.058	100.0	92
163	Ebro	799548.1	4566029.2	0.058	100.0	92
189	Tributary	501065.0	4721980.0	0.000	0.0	0.3
216	Tributary	677370.0	4613100.0	0.016	64.3	95
226	Tributary	757442.2	4618951.3	0.096	17.7	93
514	Tributary	460440.0	4760700.0	0.000	0.0	47
551	Tributary	719280.2	4669190.5	0.089	91.5	97

S6. Sub-catchments information