Supplementary Information 1 - Data used for substance flow analyses to calculate input, output and internal flows.

Table S1: Nutrient contents based on fresh weight of an item.

Group	Item	%N	%P	Case study	Source
Cereal	Maize grain	1.47	0.32	Huantai	1
	Maize straw	0.87	0.13	Huantai	1
	Rice grain	1.3	0.36	Lake Tai	2, 3
	Rice straw	0.91	0.1	Lake Tai	2, 3
	Wheat grain	2.25	0.41	Lake Tai	2, 3
	Wheat grain	2.16	0.37	Huantai	1
	Wheat straw	0.62	0.1	Lake Tai	2, 3
	Wheat straw	0.62	0.07	Huantai	1
Fruit and vegetables	Tomato crop	0.31	0.031	Yangling	4
	Tomato residue	1.83	0.71	Yangling	5
	Cowpea crop	1.21	0.16	Yangling	6
	Cowpea residue	2.02	0.48	Yangling	5
	Cucumber crop	0.21	0.044	Yangling	4
	Cucumber residue	2.75	0.69	Yangling	5
	Strawberry crop	0.11	0.02	Yangling	6
	Strawberry residue	1.64	0.86	Yangling	5
	Muskmelon crop	0.09	0.015	Yangling	6
	Muskmelon residue	3.34	0.81	Yangling	5
	Water melon crop	0.25	0.039	Yangling	4
	Water melon residue	2.47	0.46	Yangling	5
Input	Irrigation water	0.183	0.007	Lake Tai	7
	Irrigation water	0.96	0.05	Yangling	8, 9
	Mixed commercial manure	2.33	1.34	Lake Tai	7
	Monopotassium phosphate		22.1	Lake Tai	7
	Urea	46.4		Lake Tai	7

[1]NATESC, 1999; [2]Ma et al., 2011; [3]Ma et al., 2008; [4]Gao et al., 2012; [5]Zhou (personal communication); [6]http://nutritiondata.self.com; [7]Lai (personal communication); [8]Yuan et al., 2010; [9] Zhang and Fang, 2006.

Table S2: Inputs and outputs in Lake Tai.

Item	Type	Amount	Unit
Urea on wheat	Input	190	Kg product/ha
Monopotassium phosphate on wheat	Input	140	Kg product/ha
Atmospheric deposition	Input	27.9	Kg N/ha
Biological fixation	Input	30	Kg N/ha
Mixed commercial manure (pig and chicken) on wheat	Input	1000	Kg manure/ha
Urea on rice	Input	540	Kg product/ha
Monopotassium phosphate on rice	Input	145	Kg product/ha
Mixed commercial manure (pig and chicken) on rice	Input	900	Kg manure/ha
Irrigation water	Input	2680000	L water/ha
Irrigation water N	Input	1.83	Mg N/L
Irrigation water P	Input	0.073	Mg P/L
Seed rate wheat	Input	150	Kg seeds/ha
Seed rate rice	Input	135	Kg seeds/ha
Residue wheat	SC[a]	8.1	T residue/ha
Residue rice	SC[a]	7.6	T residue/ha
Yield of wheat	Output	7.2	t grain/ha
Yield of rice	Output	9	t grain/ha

[a]SC: System cycling, cereal residue is incorporated into the field

Table S3: Data for cropland within Huantai county based on a 5 year average (2007 – 2011)

Item	Type	Amount	Unit
Land area	Cropland	23723	ha
N	Input	503	Kg N/ha
P	Input	82	Kg P/ha
Irrigation water	Input	14	Kg N/ha
Total N from Seeds	Input	3	Kg N/ha
Total P from Seeds	Input	1	Kg P/ha
Wheat	Output	178598	T grain
Maize	Output	217185	T grain
Proportion of wheat straw returned	SC[a]	90	%
Proportion of maize straw returned	SC[a]	86	%
Wheat: straw/grain mass ratio		1.04	
Maize: straw/grain mass ratio		0.91	

[a] SC: System Cycling

Table S4: Average inputs to greenhouses in Yangling and total area.

Item	Type	Amount	Unit
Greenhouse area		2.4192	Ha
Fertiliser application – N	Input	836.1	Kg N/ha
Fertiliser application – P	Input	309.8	Kg P/ha
Manure application – N	Input	653	Kg N/ha
Manure application – P	Input	228.7	Kg P/ha
Amount of irrigation water	Input	5954702	L water/ha
Irrigation water –Nitrate-N only	Input	9.6[a]	Mg N/L
Irrigation water –P	Input	0.5[b]	Mg P /L

[a]Yuan et al., 2010; [b]Zhang et al., 2006.

Table S5: Greenhouse outputs.

Vegetable	Area cropped in ha[a]	Amount of residue in t DM/ha[b]	Vegetable yield in t fresh weight/ha[b]
Tomatoes	2.8896	5.54	108.16
Cowpea	0.1344	6.91	30.21
Cucumber	0.7392	1.93	88.93
Strawberry	0.4032	5.23	11.25
Muskmelon	0.0672	1.51	22.50
Watermelon	0.6048	3.33	37.50

[a] this considers the area available for both seasons, meaning that it is twice the area available in one season

[b] yields are given for one crop in one season, because the effect of double cropping is already considered in the area under cultivation for an individual crop.

Table S6: Input of nutrients in kg/ha via transplanted seedlings (Wim Voogt, personal communication) and derived average input into greenhouses of Yangling.

Vegetable	N in kg/ha	P in kg/ha	Area occupied in %
Tomatoes	7.14	0.78	60
Cucumber	5.04	0.62	15
Average nutrient input[a]	6.3	0.72	

[a] the nutrient values for cucumber were used for the remaining 25% of the cultivated area of the greenhouses, because over 60% of this area are within the same family.

Table S7: Nitrogen deposition and biological N fixation rates for the case studies in which no experimental measurements were available.

Case study	N deposition rates in kg N/ha	N fixation rates in kg N/ha
Huantai	76[1]	5[3]
Yangling	4[2]	5[3]

[1]He et al., 2007; [2]Liang et al., 2014; [3]Bouwman et al., 2005.

Calculation of atmospheric losses

For P, it was assumed that no gaseous losses occur. Empirical models with a series of factors were used for the calculation of losses of ammonia (NH_3) (Bouwman et al., 2002) and the nitrogenous greenhouse gases, nitrous oxide (N_2O) and nitric oxide (NO) (Stehfest and Bouwman, 2006). The amount of nitrogen gas (N_2) lost was estimated via the ratio of N_2 to N_2O produced during denitrification using the freely available spreadsheet model SimDen (http://agro.au.dk/en/research/sektioner/soil-fertility/fpv/simden/).

Table S8 provides an overview of the factor class used in the published functions for each of the five case study catchments.

Table S8: Factor classes for the calculation of atmospheric N losses according to Bouwman et al., (2002); Stehfest and Bouwman, (2006) and SimDen.

	Factor	Lake Tai	Huantai	Yangling
NH_4	**Fertiliser type**	Urea, manure	Urea, compound fertiliser	Ammonium phosphates, urea, compound fertiliser, manure
	Croptype	Other crop, Wetland rice	Other crop	Other crop
	pH	5.5-7.3	7.3 - 8.5	7.3 – 8.5
	CEC	16 – 24	16 - 24	24-32
	Climate[a]	Temperate	Temperate	Temperate
	Application method	Broadcast or incorporated then flooded	Broadcast	Incorporated or applied in solution[b]
N_2O	**SOC**	1 – 3	1 - 3	1-3
	pH	5.5 - 7.3	7.3 - 8.5	7.3 – 8.5
	Texture	Medium	Medium	Medium
	Climate[a]	Temperate continental	Temperate continental	Temperate continental
	crop type	Cereals Wetland rice	Cereals	Other crop
NO	**Soil N content**	0.05 - 0.2	0.05-0.2	>0.2
	Climate[a]	Temperate continental	Temperate continental	Temperate continental
N_2	**Soil type**	Sandy loam (26% clay)	Clay loam	Clay loam
	SOM/precipitation	High	High	High

[a]see

Table S9 for an explanation of climate thresholds; [b]all manure is incorporated along with 15% of the fertiliser, 85% of the fertiliser is applied in solution

Table S9: Thermal climate classification units taken from Fischer et al., (1996).

Division	Subdivision	Characteristics
Tropics		monthly T_{mean}> 18 °C
Subtropics	Summer rainfall	monthly T_{mean}> 5 °C and at least one month T_{mean}<18 °C rainfall mainly in summer
	Winter rainfall	As above, but rainfall mainly in winter
Temperate	Oceanic	4 or more months T_{mean}> 10 °C and at least one month T_{mean}< 5 °C Difference in T_{mean} between warmest and coldest month □ 20 °C
	Continental	As above, but difference in T_{mean} between warmest and coldest month >20 °C
Boreal	Oceanic	Less than 4 months with T_{mean}> 10 °C and at least one month T_{mean}< 5 °C Difference in T_{mean} between warmest and coldest month □ 20 °C
	Continental	As above, but difference in T_{mean} between warmest and coldest month >20 °C
Polar/Arctic		Monthly T_{mean}< 10 °C

Aqueous losses – Erosion, runoff and leaching

Losses via runoff and leaching have been determined using the empirical model developed for N (Velthof et al., 2009), which has been extended to include erosion by Ma et al. (2010). The algorithms for the calculation of runoff and erosion are considered to be the same for N and P, which are related to fertiliser application rates and soil nutrient content, respectively. However, leaching of P is expected to be much lower compared to N. Therefore, the literature factor of 0.1 kg P/ha/year, as reported by Némery et al., (2005), is used throughout for P loss via leaching.

The approach of Velthof et al. (2009) requires information regarding ranges of slope, land use, soil type, soil and rooting depth, clay and carbon content, temperature and precipitation surplus that are reasonably widely available. The precipitation surplus was assumed to be at the lowest precipitation surplus applied. Table 10 reports the factor classes applied to each case study. It was assumed that runoff and erosion was zero for the greenhouses in the Yangling case study, because the soil is contained within the greenhouse walls.

Table 10: Factor classes used for the calculation of aqueous losses based on Velthof et al. (2009) and Ma et al. (2010).

	Factor	Lake Tai	Huantai	Yangling
Leaching	**Soil type**	Loamy (26% clay)	Loamy	Loamy
	Land use	Other	Other	Other
	Minimum of other factors	Precipitation < 50mm	Precipitation < 50mm	Precipitation < 50mm
Runoff	**Slope in %**	0-8	0-8	
	Land use	Other	Other	
	Minimum of other factors	Precipitation < 50mm	Precipitation < 50mm	
Erosion	**Slope in %**	0-8	0-8	
	Precipitation surplus	< 50mm	< 50mm	
	Minimum of other factors	Clay content 18 - 34%	Clay content 18 - 34%	

Note: the grey shading represents the assumption that no losses occur. In this case, runoff and erosion are assumed to be zero for the greenhouses as the soil is contained within the greenhouse walls.

Input, output, internal flows and losses

Table S11: Nutrient flow in N/P kg/ha.

	Huantai		**Lake Tai**		**Yangling**	
	N	**P**	**N**	**P**	**N**	**P**
Fertiliser	503	82	337	63	836	310
Manure	0	0	44	25	653	229
Atmospheric deposition	76	0	28	0	4	0
N fixation	15	0	30	0	5	0
Irrigation	14	0.2	5	0.2	57	3
Seed	3	1	5	1	6.3	0.7
Residue returned	106	14	119	19	0	0
Total input	**717**	**97.2**	**568**	**108.2**	**1561.3**	**542.7**
Crop uptake	418	73	397	81	685	128
Soil balance	299	24.2	171	27.2	876.3	414.7
Crop grain output	297	57	278	62	504	58
Residue exported	15	2	0	0	181	70
NH_3	71		30		216	
N_2O	6		3		32	
NO	4		1		305	
N_2	42		19		258	
Total atmospheric losses	**123**		**53**		**811**	
Leaching	79	0.1	25	0.1	125	0.1
Runoff	13	2	10	2	0	0
Erosion	50	1.6	89	36	0	0
Total aqueous losses	**142**	**3.7**	**124**	**38.1**	**125**	**0.1**

References

Bouwman, A.F., Boumans, L.J.M., Batjes, N.H., 2002. Estimation of global NH3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. Glob. Biogeochem. Cycles 16, 2.

Bouwman, A.F., Van Drecht, G., Van der Hoek, K.W., 2005. Global and regional surface nitrogen balances in intensive agricultural production systems for the period 1970-2030. Pedosphere 15, 137-155.

Fischer, G., dePauw, E., van Velthuizen, H., Nachtergaele, F., Antoine, J., 1998. A provisional world climate resources inventory based on the length-of-growing-period concept.

Fischer G., de Pauw E., van Velthuizen H.T., Nachtergaele F.O., Antoine, J., 1996. A provisional world climatic resource inventory based on the length of growing period concept, in: Batjes, N.H., Kauffman, J.H., Spaargaren, O.C. (Eds.) Proceedings of a workshop on National Soil Reference Collections and Databases (NASREC). Vol. 3. Papers and Country Reports. ISRIC. Wageningen, The Netherlands. pp. 30-43.

Gao, J.J., Bai, X.L., Zhou, B., Zhou, J.B., Chen, Z.J., 2012. Soil nutrient content and nutrient balances in newly-built solar greenhouses in northern China. Nutr. Cycl. Agroecosystems 94, 63-72.

He, C-E., Liu, X., Fangmeier, A., Zhang, F., 2007. Quantifying the total airborne nitrogen input into agroecosystems in the North China Plain. Agric. Ecosyst. Environ. 121, 395-400.

Liang, T., Tong, Y., Lin, W., Qiao, L., Liu, X., Bai, S., Yang, X., 2014. Spatial-temporal variability of dry and wet deposition of atmospheric nitrogen in different ecological regions of Shaanxi. Acta Ecologica Sinica 34, 3.

Ma, L., Ma, W.Q., Velthof, G.L., Wang, F.H., Qin, W., Zhang, F.S., Oenema, O., 2010. Modeling nutrient flows in the food chain of China. J. Environ. Qual. 39, 1279-1289.

Ma, W., Li, J., Ma, L., Wang, F., Sisák, I., Cushman, G., Zhang, F., 2008. Nitrogen flow and use efficiency in production and utilization of wheat, rice, and maize in China. Agric. Syst. 99, 53-63.

Ma, W., Ma, L., Li, J., Wang, F., Sisák, I., Zhang, F., 2011. Phosphorus flows and use efficiencies in production and consumption of wheat, rice, and maize in China. Chemosphere 84, 814-821.

NATESC, 1999. National Agro-tech Extension & Service Center, Nutrients database for organic fertilizers in China. China Agriculture Press, Beijing, China.

Némery, J., Garnier, J., Morel, C., 2005. Phosphorus budget in the Marne Watershed (France): urban vs. diffuse sources, dissolved vs. particulate forms. Biogeochemistry 72, 35-66.

Stehfest, E., Bouwman, L., 2006. N2O and NO emission from agricultural fields and soils under natural vegetation: summarizing available measurement data and modelling of global annual emissions. Nutr. Cycl. Agroecosystems 74, 207-228.

Velthof, G.L., Oudendag, D., Witzke, H.P., Asman, W.A.H., Klimont, Z., Oenema, O., 2009. Integrated Assessment of Nitrogen Losses from Agriculture in EU-27 using MITERRA-EUROPE. J. Environ. Qual. 38, 402.

Yuan, L-J., Ju, X-T., Zhang, L-J., Wang, J., Yang, Z-X., 2010. NPK accumulation in greenhouse soil and its effect on groundwater. Chin. J. Eco-Agric. 18, 14-19.

Zhang M., Fang L., 2006. Accumulation and transport of nutrients in agricultural sandy soils. Journal of Soil and Water Conservation, 20, 2, 46-49.

Zhang, M-K., Zhou, C., Fang, L.P., 2006. Phosphorus saturation degree of soils on vegetable and its effects on soil phosphorus release potential and water quality. Plant Nutr. Fertil. Sci. 12, 544-548.