## Supporting information

**Nitrogen budget and its environmental loading in an urban ecosystem with the rapid urbanization of China**

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## N budget model

**Cropland subsystem (CS).** The calculation equations used are as follows:

$$CS\_{IN}=CSIN\_{fer}+CSIN\_{dep}+CSIN\_{BNF}+CSIN\_{seed}+CSIN\_{man}+CSIN\_{str}$$

$$CS\_{OUT}=CSOUT\_{crop}+CSOUT\_{str}+CSIN\_{loss}$$

where CSIN and CSOUT are the total N input to and output from cropland; CSINfer is synthetic fertilizer application. Fertilizer N applied to cropland was directly obtained from the Zhengzhou Bureau of Statistics (2015) [1] using an N content of 30% for compound fertilizer. CSINBNF is the BNF, including symbiotic and non-symbiotic N fixation. Total BNF was estimated by the symbiotic N2 fixation rate of leguminous plants with sown area, and non-symbiotic N fixation rate for peanut and other crops with their sown area. Commonly used value of non-symbiotic N fixation rate is 105 kg N ha-1 year-1 for soybean, 112 kg N ha-1 year-1 for peanuts and 15 kg N ha-1 year-1 for other crops and vegetables [2]. CSINman is manure recycled to cropland from livestock. Manure input to croplands was obtained from the livestock and poultry subsystem. CSINdep is N deposition, which was estimated based on the per hectare deposition rate from a synthesis of measurements [3]. CSINstr is the straw recycled to cropland. CSINseed is N input to cropland from seed, which was calculated by multiplying the N concentration for each crop type, seeding amount by the sown area of that crop in ZZC (Zhengzhou City). Data on the area of various crops were extracted from Zhengzhou Bureau of Statistics (2015) [1]. The seeding amount and grain N concentration of various crops are shown in Table S1.

CSOUTcrop is crop production, which was estimated by total production [1] and N concentrations (see Table S2) for each crop type. CSOUTstr is straw production. The fate of straw produced included straw used as feed, fuel in rural areas and industrial materials, recycled to croplands, and burned in the field; the proportions of these utilization patterns were estimated by the synthesis of previous investigations (Table S2). CSOUTloss is Nr loss during crop production, including NH3 and NOx emissions, denitrification process, riverine runoff, and leaching to groundwater. In calculating N losses through NH3 volatilization, we used 10 kg N ha-1 as the conversion factor for the human and animal wastes, and1.5 kg N ha-1 as the conversion coefficients for fertilizers in upland [4]. N losses due to denitrification were calculated using data from Zhengzhou Statistical Yearbook (2015) [1]and the conversion factors (16 kg N ha-1) from upland fertilizer and manure N application [4].The N loss rate of fertilizer for runoff and leaching was 7.38% and 1.67%, respectively [3].

**Livestock and poultry subsystem (LS).** The calculation equations used are as follows:

$$LS\_{IN}=LSIN\_{str}+LSIN\_{fer}+LSIN\_{fish}$$

$$LS\_{OUT}=LSOUT\_{live}+LSOUT\_{manu}+LSOUT\_{loss}$$

where LSIN and LSOUT are the total N inputs to and outputs from the livestock and poultry subsystem; LSINstr is straw used as livestock and poultry feed from cropland subsystem; LSINfer is the urea input for straw ammonization to produce feed, which is about 1.5 times of LSINstr [3]. LSINfish is the fish powder supplied from aquaculture subsystem.

 LSOUTlive is livestock and poultry products, which was estimated based on the total production [1] and the N concentration of each livestock and poultry product (Table S3). LSOUTmanu is livestock and poultry excretion were recycled to cropland, which was calculated by the livestock and poultry raised each year and the excretion generated per animal, the N excretion rates are shown in Table S4. LSOUTloss is the N lost through riverine runoff, NH3 volatilization and denitrification, the lost ratios are 35%, 10%, and 16% in present study according to Gu (2011) [3].

**Aquaculture subsystem (AS).** The calculation equations used are as follows:

$$AS\_{IN}=ASIN\_{crop}+ASIN\_{fer}+ASIN\_{dep}$$

$$AS\_{OUT}=ASOUT\_{meat}+ASOUT\_{loss}$$

where ASIN and ASOUT are the total N input and output to AS. ASINcrop is crop that was used as aquaculture feed, which was calculated by the aquaculture production and the average feed conversion ratio [5]. ASINfer is fertilizer input to AS, which was estimated as 20% of the total feed use [6]. ASINdep is N deposition that was estimated based on the aquaculture area [1] and the N deposition rate [2].

ASOUTMeat is aquaculture production, which was estimated based on the total production and the N concentration of each aquaculture species (fish:3.0%, shrimp/crap: 2.9%, shellfish:2.1%, alage:3.7%, others:2.9%) [7]. ASOUTLoss is Nr loss during aquaculture production, including emission of NH3, N2 and N2O and riverine runoff, which was estimated as 15, 50, 1.25 and 10% of total N surplus, respectively [6].

**Industry subsystem (IS).** The calculation equations used are as follows:

$$IS\_{IN}=ISIN\_{HBNF}+ISIN\_{crop}+ISIN\_{str}+ISIN\_{LS}+ISIN\_{fuel}$$

$$IS\_{OUT}=ISOUT\_{fer}+ISOUT\_{indu}+ISOUT\_{loss}$$

where ISIN and ISOUT are the total N input and output to industry subsystem. ISIN HBNF is Haber-Bosch N fixation in factories, which was obtained from Zhengzhou Bureau of Statistics (2015) [1]; ISINcrop is agricultural product transferred to industry from cropland subsystem, ISINstr is straw transferred to industry for material production from cropland subsystem, and ISINLS is the livestock and poultry product transferred to industry, which can be obtained from cropland and livestock and poultry subsystems, respectively. IDINFuel is fossil fuel combustion during industrial production. It is generally considered to be that the input fossil fuels would be all emitted into atmosphere in the form of NOx-N via burning [3]. Therefore, we assume that the amount of input fossil fuel is equal to the amount of NOx emission.

IDOUTfer is Nr fertilizer production. IDOUindu is industrial products, N content of major industrial N products are showed in Table S5; IDOUTloss is the Nr loss during industrial production, including wastewater discharge, NOx emission, denitrification in industrial wastewater treatment plant. The amount of NOx emission can be obtained from the environmental quality bulletin of Zhengzhou city in 2014 (<http://www.zzepb.gov.cn/Article/Content/?id=32853>); the waste N generated, discharged and N removal rate from every product per unit of production can be estimated from the first nationwide pollution source survey of China (Table S6).

**Urban green-land subsystem (US).** The calculation equations used are as follows:

 $US\_{IN}=USIN\_{fer}+USIN\_{dep}+USIN\_{BNF}+USIN\_{exc}+USIN\_{liter}$

$$US\_{OUT}=USOUT\_{clip}+USIN\_{Loss}$$

where USIN and USOUT are the total N input and output to urban green land. USINfer is fertilizer applied to urban lawn, which was estimated based on the total green-land area [1] with a recommended N application rate of 300 kg N ha-1 yr-1 to the urban lawn [8]. USINdep is N deposition, the deposition rate refers to the cropland subsystem [3]. USINBNF is BNF in urban green land, the N fixed rate is 18 Kg ha-1 yr-1(Gu et al., 2009). USINexc is pet excretion recycled to urban lawn, which was obtained from pet subsystem. USINlite is the liter recycled back during lawn and tree clipping, which can be estimated based on the NPP of lawn and urban, annual pruning and litter collection [9].

USOUTclip is the N removal through lawn and tree clipping, which was estimated based on previous studies [9]. USOUTloss is Nr loss from green land, including NH3 volatilization, riverine runoff, denitrification, and leaking to groundwater, the N loss rates refers to the cropland subsystem since the urban green-land has similar characteristics with dry farmland [3].

**Human subsystem (HS).** The calculation equations used are as follows:

$$HS\_{IN}=HSIN\_{food}+HSIN\_{indu}+HSIN\_{str}+HSIN\_{fuel}$$

$$HS\_{OUT}=HSOUT\_{exc}+HSOUT\_{garb}+HSOUT\_{NOx}$$

where HSIN and HSOUT are the total N input and output to human subsystem; HSINfood is human food consumption, which can be obtained from Zhengzhou Bureau of Statistics (2015) [1] and the N contents in each of food was referred to the cropland, livestock and poultry, and aquaculture subsystems (Table S2, S3, S4). HSINindu is human industrial product consumption, the estimated method according to Gu et al. (2013) [10]. HSINstr is straw used as biofuel for cooking, which was obtained from cropland subsystem; HSINfuel is the NOx emission from domestic and traffic fossil fuel consumption. The NOx emission factors from residents were shown in Table S7, and the factors from transportation according to Zhang et al. (2008) [11]

HSOUTexc is human excretion, the human excretion utilization rate is shown in Table S8. HSOUTgarb is the food waste and industrial product abandoned, which was computed according to Li et al. (2001) [12]. HSOUTNOx is the NOx emission from fuel consumption, we assumed that the input N of fossil fuel will be totally emitted into atmosphere in the form of NOx-N.

**Pet subsystem (PS).**

The total N input to pet subsystem mainly is pet feed input. The numbers of dogs and cats were estimated based on the numbers of household in both rural and urban area [13]. The amount of N required per kilogram of weight per day for dog and cat are 0.56 and 0.88 g N [14]. The total output N is pet excretion, which is calculated according to Gu et al. (2009) [15].

**Wastewater treatment subsystem (WS).** The calculation equations used are as follows:

$$WS\_{IN}=WSIN\_{HS}+WSIN\_{LS}+WSIN\_{GT}$$

$$WS\_{OUT}=WSOUT\_{den}+WSOUT\_{lea}+WSOUT\_{river}+WSOUT\_{rec}$$

where WSIN and WSOUT are the total N input and output to wastewater treatment subsystem. WSINHS, WSINLS and WSINGT are the domestic wastewater, livestock excretion and landfill leachate in human, livestock and poultry and garbage treatment subsystems, respectively.

WSOUTdep is the denitrification in the wastewater treatment plant, accounting to about 30% of the total N input to WS [16]. WSOUTlea is the Nr leaching during the wastewater transferred to the wastewater treatment plant, accounting to about 9% of total N input; WSOUTriver and WSOUTrec are the Nr discharged and recycled after treated, the ratio of recycling is about 10%, the reminder will be discharged into surface water [3].

**Garbage treatment subsystem (GS).**

The input N of GS mainly include green waste sent to landfill from urban green land subsystem, garbage from human subsystem and pet excretion. The output N from GS mainly is the Nr contained in garbage released to groundwater, which was estimated according to Zhou et al. (2006) [17].

**References:**

[1] Zhengzhou Bureau of Statistics. Zhengzhou statistic yearbook. China Statistic Press,

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case study in Taizhou, China. Chin J Plant Ecol. 2010;6:651-660. (in Chinese)

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[17] Zhou H, Zhang H, Zheng X, et al. Research progress on treatment of municipal solid waste landfill leachate. Jiangsu Environmental Science and Technology. 2006;19(2):142-144. (in Chinese)

## List of Tables

**Table S1.** Coefficients for estimation of import of seeds

|  |  |  |  |
| --- | --- | --- | --- |
| N sources | Items | Seeding amount(kg ha-1)  | N concentration (%) |
| Import of seeds | Cereal | 30 | 1.78 |
| Soybean | 67.5 | 5.30 |
| Oil-bearing crops | 180 | 4.4 |

References:

[1] Xing G, Zhu Z. Regional nitrogen budgets for China and its major watersheds. Biogeochemistry. 2002;57:405-427.

**Table S2.** Nitrogen concentration in different crops and the ratio of straws/seeds

|  |  |
| --- | --- |
| Crops | N concentrations |
| Seeds | Straw | Straw/seed (leaf/stem) |
| Cereal | 1.53% | 0.64% | 1.2 |
| Soybean | 5.11% | 2.01% | 1.6 |
| Oil-bearing Crops | 3.73% | 0.84% | 1.7 |
| Vegetables | 0.50% | 0.50% | 0.5 |

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**Table S3.** Nitrogen concentration in livestock and poultry products

|  |  |  |  |
| --- | --- | --- | --- |
| Product | N concentrations (%) | Product | N concentrations (%) |
| Pork | 2.1 | Egg | 2.1 |
| Beef | 3.2 | Milk | 0.5 |
| Lamp | 3.0 | Goat’s milk | 0.2 |
| Rabbit | 3.2 | Honey | 0.1 |
| Chicken | 3.1 | Sheepskin | 8.4 |
| Duck | 2.5 | Wool | 12.2 |
| Goose | 2.9 |  |  |

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**Table S4.** Livestock and poultry nitrogen excretion rate

|  |  |  |  |
| --- | --- | --- | --- |
| Product | Rate (Kg N capita-1 yr-1) | Product | Rate (Kg N capita-1 yr-1) |
| Dairy cow | 69.1 | Rabbits | 0.45 |
| Draft cow | 35.5 | Ducks/Geese | 0.47 |
| Beef cattle | 45.87 | Chicken | 0.1 |
| Pig | 4.87 | Goat | 8.8 |

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[2] Wang F, Ma W, Dou Z, et al. The estimation of the production amount of animal manure and its environmental effect in China. China Environ Sci. 2006;26:614-617.

**Table S5** N contents of major N-containing industrial products

|  |  |  |  |
| --- | --- | --- | --- |
| Product | N content (%) | Product | N content (%) |
| Detergent | 0.5 | Leather | 12.6 |
| Drugs | 5.0 | Cotton | 3.0 |
| Dynamite | 18.0 | Bast | 0.8 |
| Nitrate | 22.0 | Sheepskin | 8.4 |
| Plastic | 0.5 | Silk | 3.4 |
| Synthetic dyes | 15.0 | Tobacco leaf | 1.5 |
| Synthetic fiber | 10.0 | Wood furniture | 0.3 |
| Synthetic rubber | 0.5 | Wool | 12.2 |

References:

[1] Gu B, Chang J, Min Y, et al. The role of industrial nitrogen in the global nitrogen biogeochemical cycle. Sci Rep. 2013;3(9):2579.

**Table S6** Waste N generated and discharged in industry subsystem

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Product | unit | Generation | Discharge | N removal |
| Synthetic ammonia | Kg N t product-1 | 1.27 | 0.88 | 0.31 |
| Chemical pesticides | Kg N t product-1 | 51.23 | 23.92 | 0.53 |
| Chemical drugs | Kg N t product-1 | 31.18 | 15.99 | 0.49 |
| cosmetics | Kg N t product-1 | 0.25 | 0.11 | 0.56 |
| dyestuff | Kg N t product-1 | 7.05 | 3.12 | 0.56 |
| Chemical fertilizer | Kg N t product-1 | 8.60 | 0.28 | 0.97 |
| Nitric acid | Kg N t product-1 | 0.25 | 0.25 | 0.00 |
| paint | Kg N t product-1 | 0.03 | 0.01 | 0.78 |
| pigment | Kg N t product-1 | 14.32 | 6.36 | 0.56 |
| soda | Kg N t product-1 | 1.13 | 1.06 | 0.06 |
| spice | Kg N t product-1 | 2.55 | 1.08 | 0.58 |
| Synthetic detergent | Kg N t product-1 | 0.01 | 0.01 | 0.45 |
| toothpaste | Kg N t product-1 | 0.03 | 0.03 | 0.05 |
| alcohol | Kg N 1000L product-1 | 2.22 | 1.34 | 0.39 |
| feed | Kg N t product-1 | 0.98 | 0.51 | 0.48 |
| Aquatic product | Kg N t product-1 | 33.06 | 15.95 | 0.52 |
| beer | Kg N 1000L product-1 | 0.99 | 0.26 | 0.74 |
| dairy | Kg N t product-1 | 0.97 | 0.17 | 0.82 |
| Egg processing | Kg N t product-1 | 3.13 | 1.35 | 0.57 |
| Fish oil | Kg N t product-1 | 1.02 | 0.37 | 0.64 |
| additive | Kg N t product-1 | 5.94 | 1.53 | 0.74 |
| wine | Kg N 1000L per product-1 | 0.09 | 0.06 | 0.26 |
| Ice cream | Kg N t product-1 | 0.10 | 0.03 | 0.73 |
| Liquor and spirits | Kg N 1000L per product-1 | 2.04 | 0.50 | 0.76 |
| Livestock slaughter | Kg N t product-1 | 1.46 | 0.72 | 0.51 |
| Meat products | Kg N t product-1 | 1.76 | 0.79 | 0.55 |
| Monosodium glutamate | Kg N t product-1 | 124.26 | 4.01 | 0.97 |
| Protein beverage | Kg N t product-1 | 0.08 | 0.05 | 0.44 |
| Bean products | Kg N t product-1 | 6.13 | 2.70 | 0.56 |
| Starch products | Kg N t product-1 | 1.88 | 1.11 | 0.41 |

References:

[1] CPSC (China Pollution Source Census). The first nation census on pollution sources: coefficient manual of industrial pollution produced and discharged. Beijing, Ministry of Environmental Protection of China Press, 2008.

**Table S7** NOx emission factors of fossil fuel combustion in Human Subsystem

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Sectors | coal | Gasoline | Diesel | Natural gas |
| Residents | 1.9 | 16.7 | 3.2 | 1.5 |
| Transportation | 2.3 | 7.4 | 16.5 | 0.6 |

References:

[1] Tian H, Hao J, Lu Y, Zhu T. Inventories and distribution characteristics of NOx emissions in China. China Environmental Science. 2001;21(6):493-497.

**Table S8** Excretion utilization in urban and rural areas

|  |  |
| --- | --- |
| Fate of excreta | Rate (%) |
| Urban sewage treatment | 66.0 |
| Urban excretion recycled | 10.0 |
| Urban excretion discharged | 24.0 |
| Rural sewage treatment | 6.60 |
| Rural excretion recycled | 58.2 |
| Rural excretion discharged | 35.3 |

References:

 [1] Gu B. Nitrogen cycle of coupled human and natural system- a case study of China (in Chinese), Zhejiang University: Hangzhou, China; 2011.