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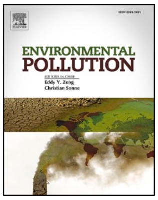

Nitrogen budgets in Japan from 2000 to 2015: Decreasing trend of nitrogen loss to the environment and the challenge to further reduce nitrogen waste☆

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ABSTRACT

The benefits of the artificial fixation of reactive nitrogen (Nr, nitrogen [N] compounds other than dinitrogen), in the form of N fertilizers and materials are huge, while at the same time posing substantial threats to human and ecosystem health by the release of Nr to the environment. To achieve sustainable N use, Nr loss to the environment must be reduced. An N-budget approach at the national level would allow us to fully grasp the whole picture of Nr loss to the environment through the quantification of important N flows in the country. In this study, the N budgets in Japan were estimated from 2000 to 2015 using available statistics, datasets, and literature. The net N inflow to Japanese human sectors in 2010 was 6180 Gg N yr^{-1} in total. With 420 Gg N yr^{-1} accumulating in human settlements, 5760 Gg N yr^{-1} was released from the human sector, of which 1960 Gg N yr^{-1} was lost to the environment as Nr (64% to air and 36% to waters), and the remainder assumed as dinitrogen. Nr loss decreased in both atmospheric emissions and loss to terrestrial water over time. The distinct reduction in the atmospheric emissions of nitrogen oxides from transportation, at −4.3% yr^{-1}, was attributed to both emission controls and a decrease in energy consumption. Reductions in runoff and leaching from land as well as the discharge of treated water were found, at −1.0% yr^{-1} for both. The aging of Japan's population coincided with the reductions in the per capita supply and consumption of food and energy. Future challenges for Japan lie in further reducing N waste and adapting its N flows in international trade to adopt more sustainable options considering the reduced demand due to the aging population.

1. Introduction

Artificial nitrogen (N) fixation, producing ammonia (NH_3) from dinitrogen (N_2), has become increasingly common since early 20th century (Erisman et al., 2008). Production in 2018 was reported to be 144,000 Gg N yr^{-1} (1 Gg N = 10^9 g of N) (USGS, 2021), more than double that of terrestrial biological N fixation (BNF) (Fowler et al., 2013). While artificially fixed N provides great benefits as fertilizers and materials, 80% of the fixed N is unintendedly lost to the environment through a multiplicity of N sources and forms (Sutton et al., 2019). The use of combustion to create energy and waste incineration also create nitrogen oxides (NO_X) (Galloway et al., 2002; Fowler et al., 2013).

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Reactive forms of N (N other than N_2), collectively called reactive N (Nr) (Galloway et al., 2002), intricately circulate among environmental media, known as the N cascade (Galloway et al., 2003). The various impacts of this widespread N pollution include global warming, stratospheric ozone depletion, air and water pollution, acidification, eutrophication, loss of biodiversity, and ecosystem changes (Sutton et al., 2011; Erisman et al., 2013).

Reducing N waste, i.e., N being released from human sectors to the environment, by reducing N input which is not utilized is necessary to mitigate N pollution (Sutton et al., 2019). Once used, N is eventually released to the environment also as N waste in the form of solid waste and wastewater. This study separated N waste into two types: direct N waste is the Nr lost to the environment from human sectors; and indirect N waste is the Nr created by various human sectors but turned back to N_2 by technologies to treat exhaust gas and wastewater (treatment) and microbial processes in land of human uses.

Severe air and water pollution due to rapid economic growth in the 1950s and 1960s in Japan prefaced the polices to control pollution and technological measures enacted since the 1970s (Barrett and Therivel, 1991) (see Supplementary Information for a historical summary). Two types of measures were enacted against N pollution: one is to reduce creation of Nr and the other is to convert the created Nr into N_2. The former is exemplified by emission controls of Nr for point and mobile sources, e.g., low NO_X combustion (Nishimura et al., 1997) and reducing surplus Nr input by nutrient management for non-point sources (Kumazawa, 2002). The latter is exemplified by denitration of exhaust gas of point and mobile sources, i.e., gas-phase reaction technologies to reduce NO_X to N_2 (Tezuka, 1988; Matsumoto, 1997) and denitrification in combination with nitrification for wastewater, i.e., microbial reaction technologies to convert nitrate and ammonium to N_2 (Yoshikura et al., 1999). After its economic heyday closed around 1990, Japan experienced long-term economic stagnation (Akram, 2019), and Japan became known as home to the world's oldest population (UN, 2019a). It is reasonable to assume that the decline in social activities implied by the aging population and the economic downturn might have impacted the N pollution status in Japan. Some countries have estimated their N budgets recently to grasp the status of N uses and N pollution, e.g., China (Gu et al., 2015), Canada (Clair et al., 2014), Denmark (Hutchings et al., 2014), New Zealand (Parfitt et al., 2012), and USA (Sabo et al., 2019), but not yet available for Japan except several studies focusing on Japanese food system (e.g., Shindo et al., 2009). Geupel et al. (2021) utilized N budgets to set the abatement target of N pollution in Germany. Estimating the N budgets for Japan is necessary to grasp the Nr flow from human sector to the environment (Nr loss) in the country over the entire N cascade.

The purposes of this study were firstly to estimate the N budgets in Japan from 2000 to 2015 including all human sectors and environmental media for both direct and indirect N waste, and then to identify the challenges involved in further reducing N waste in Japan under changing socioeconomic conditions. The N budgets directly provide information of anthropogenic sources of new Nr input to Japan, part of which eventually becomes N waste and then Nr lost to the environment. The environmental status in Japan was also discussed using the estimated N budgets and relevant indicators.

2. Materials and methods

2.1. Framework for estimating the N budgets

The system boundary, N pools, and N flows were defined to estimate the N budgets for Japan. Because Japan is an island country, its river systems are all internal. Its system boundary was therefore set as the area within the Japanese border including its territorial waters and the atmosphere above. One N pool corresponds to a single human sector or environmental medium considered as a subsystem in the system. A pool may have multiple sub-pools. The N flow is defined as the mass flow of N connecting two of the pools and sub-pools. The N flow can be expressed in terms of a specific N chemical species (e.g., NO_X emission) or total N (e.g., food supply). The N balance of a pool is defined as the difference between the total inflow (input) to and total outflow (output) from the pool. National N budgets consist of the N balance in all pools in the country. Also, the N flows due to international trade are covered in the estimates in order to allow closure of the N budgets in Japan.

The target period of this study was from 2000 to 2015 with the base year at 2010. A total of 14 pools were defined: energy and fuels, industry, cropland, livestock, grassland, fisheries, human settlements, solid waste, wastewater, atmosphere, forest, urban green, terrestrial water, and the coastal zone. The concept of industry was extended beyond chemical and heavy industries to all types of manufacturing, including the food and feed industries. Due to lack of quantitative information, particularly for groundwater, surface water and groundwater were aggregated into a pool as terrestrial water. The coastal zone was defined as the Japanese territorial waters: at approximately 0.43 million km^2, the coastal zone is larger than its land area (ca. 0.38 million km^2) (JCG, 2021). See Supplementary Information for details of pools and sub-pools.

Each N flow was quantified using relevant activity data and parameters to convert the mass flow to an N amount on an annual basis. The input and output for each pool was then calculated with the quantified annual N flows. A mass balance model to calculate all N flows in Japan for the whole system and each subsystem in the country was developed referring to the CHANS model for estimating the N budgets of China (Gu et al., 2012, 2015; Gu and Zhang, 2020), considering the large differences in N flows between Japan and China.

This study relied on the National Greenhouse Gas Inventory Report of Japan (NIR) and its datasets (NIES, 2021a; MOE, 2021a) for available N flow data. While the focus of the NIR is on nitrous oxide (N_2O) as a potent greenhouse gas, it also reports the activity data of other Nr related to N_2O emissions, e.g., fertilizer consumption and manure management. This study used the most recent NIR data from the viewpoint of consistency. For the many other N flows not considered by the NIR, the available primary and processed statistics and relevant literature were used to estimate their flows. For example, the food balance sheet of FAOSTAT (FAO, 2021) was used to calculate the flow of Nr in the food and animal feed. When neither statistic nor literature information were available, N flows were calculated using available activity data and parameters, such as N content and emission factors. The concrete method used to calculate each N flow is explained in the Supplementary Information.

2.2. Indicators to interpret N budgets

Two pressure indicators of Nr loss to the environment were employed in this study. One was the Trends in Loss of Reactive Nitrogen to the Environment (TLRNE), per capita Nr loss to the environment (Bleeker et al., 2013; BIP, 2021a). The other was the Trends in Nitrogen Deposition (TND), per area N deposition (Bleeker et al., 2011; BIP, 2021b). N use efficiency (NUE) was used as a performance indicator to evaluate the N flows related to food production. The following indicators were also used to support discussion on N budgets; the ratio of chemical fertilizer production to artificial N fixation, the per capita food N supply and consumption, the self-sufficiency ratios of food and animal feed, the per capita final energy consumption, the ratio of population aged 65 years or over, the old-age dependency ratio (= population aged 65 years or over/population aged between 20 and 64 years), and economic indicators such as the per capita gross domestic product (GDP), private final consumption, current balance, and trade balance. Air and water concentrations of Nr were used as state indicators of N pollution.

The TLRNE (kg N cap^{-1} yr^{-1}) was calculated by dividing the total Nr loss from human sectors by the population, and the TND (kg N ha^{-1} yr^{-1}) was calculated by dividing the total N deposition to land and surface water by their total area.

Using the N flows obtained by these calculations, NUE was evaluated for domestic crop, livestock, and fish production and for the entire food system in Japan. The crop production NUE was expressed as the ratio of produced crop N to total input N (= fertilizer N + manure N + BNF + N deposition + irrigation N), where the irrigation N denotes the Nr provided with irrigated water. While Lassaletta et al. (2014) reported the crop production NUE excluding the irrigation N, the crop production NUE excluding the irrigation N from the denominator was also calculated. The NUEs of the livestock and fish production were also expressed similarly by the ratio of product N to total input N (= feed N for livestock production; = feed N + BNF + N deposition at fish farming area for fish production). The NUE of the food system including international trade ($NUE_{foodsystem}$), was expressed as;

$$NUE_{foodsystem} = N_{food} / (N_{fert} + N_{manu} + N_{BNF} + N_{dep} + N_{import} + N_{irri}) \quad (1)$$

Where, N_{food}, N_{fert}, N_{manu}, N_{BNF}, N_{dep}, N_{import}, and N_{irri} are the supplied food N, fertilizer N, manure N, BNF, N deposition, net import N as all food and feed, and irrigation N, respectively. The original equation was introduced by Erisman et al. (2018), but with a term describing stock changes and without the term of irrigation N. In this study, the change in stock was treated as 0, and N_{irri} was added.

The ratio of the domestic chemical fertilizer production to the domestic artificial N fixation in Japan was calculated using the N flows obtained by this study. The ratio of global chemical fertilizer production (IFA, 2021) to global NH_3 production (USGS, 2021) was also calculated. The per capita food N supply and consumption in Japan was obtained from Hayashi et al. (2018). The self-sufficiency ratios for food (as calorie basis) and animal feed (as total digestible nutrients basis) were obtained from MAFF (2021). The per capita final energy consumption for industry, transportation, and residences was calculated using the final energy consumption (ANRE, 2021) and the population (SBJ, 2021). The ratio of population aged 65 years or over and the old-age dependency ratio were calculated using population data (SBJ, 2021). GDP, private final consumption, current balance, and trade balance were calculated using the economic data (COJ, 2021; MOF, 2021) and the population (SBJ, 2021).

In this study, NO_2 was considered an air pollutant, and nitrate and total N were considered water pollutants. Their annual mean concentrations were obtained as follows: atmospheric NO_2 nationwide (NIES, 2021b); nitrate in rivers, lakes, and coastal seas nationwide (MOE, 2021b); nitrate in the groundwater in the capital region (Tokyo Metropolitan Government, 2021; Kanagawa Prefecture, 2021; Saitama Prefecture, 2021; Chiba Prefecture, 2021; Ibaraki Prefecture, 2021); and total N in rivers, lakes, and coastal seas nationwide (MOE, 2021b). Monitoring stations or plots with complete data from 2000 to 2015 (from 2009 to 2015 for riverine total N) were extracted to grasp trends over the period. In the case of groundwater nitrate, all the available data were used since the general monitoring survey of groundwater was conducted at different wells on an annual basis.

3. Results

3.1. Sources of new Nr to Japan

Artificial N fixation and the net N inflow via international trade in Japan from 2000 to 2015 are shown in Fig. 1. The domestic NH_3 production showed a decreasing trend from 1410 Gg N yr^{-1} in 2000 to 791 Gg N yr^{-1} in 2015. The BNF in croplands and grasslands ranged between 144 and 156 Gg N yr^{-1}. The N inflow from the net import of fossil fuels, calculated for coal and crude oil, was ca. 3500 Gg N yr^{-1} larger than other flows and remained relatively flat from 2000 to 2015. A dependency was also found in other resources with a positive net import, particularly for crop products, 706–758 Gg N yr^{-1}.

The N budgets in Japan in 2010 are shown in Fig. 2 and Table 1. The total inflow and outflow of industry were estimated to be ca. 7750 Gg N yr^{-1}, half of which was attributed to fossil fuels used as energy sources, and 71% of the total inflow was imported via international trade. The total inflow and outflow of agriculture, as an aggregate of cropland, grassland, and livestock, were 1900 and 1730 Gg N yr^{-1}, respectively. Since human settlements received 1140 Gg N yr^{-1} and released 720 Gg N yr^{-1}, it was determined that 420 Gg N yr^{-1} was accumulated in the pool. Waste, as an aggregate of solid waste and wastewater, received 1230 Gg N yr^{-1}, of which 288 Gg N yr^{-1} was recycled as fuel, fertilizer, feed, and materials. The N flows in the human sector from 2000 to 2015 (Fig. S2) and discussion on uncertainty are shown in the Supplementary Information. All N flows were compiled as a Supplementary Dataset.

3.2. Nr loss to the environment in Japan

The Nr loss to the environment in Japan from 2000 to 2015 is shown in Fig. 3 by destination and source. Atmospheric NO_X emissions decreased for the study period ($R = -0.99$), which was attributed to the steadily decrease in emissions from transportation ($R = -0.99$). Decreasing trends were also shown in the Nr loss to terrestrial water ($R = -0.91$). For major sources, energy and fuels (combustion and transportation), agriculture (cropland and livestock), and waste (solid waste and wastewater) accounted for 38–40%, 34–36%, and 20–22%, respectively, of the total Nr loss.

Total N waste (direct and indirect N waste) and the Nr loss to the environment (direct N waste) in Japan in 2010 are shown in Fig. 4. The total N waste, estimated at 5760 Gg N yr^{-1}, consisted of the following: fuel combustion for energy use, at 3700 Gg N yr^{-1}; solid waste and wastewater, at 1230 Gg N yr^{-1}; and surplus N in food production, at 815 Gg N yr^{-1}. The thermal NO_X creation was not estimated due to lack of quantitative information. The estimated N budgets indicate that at least

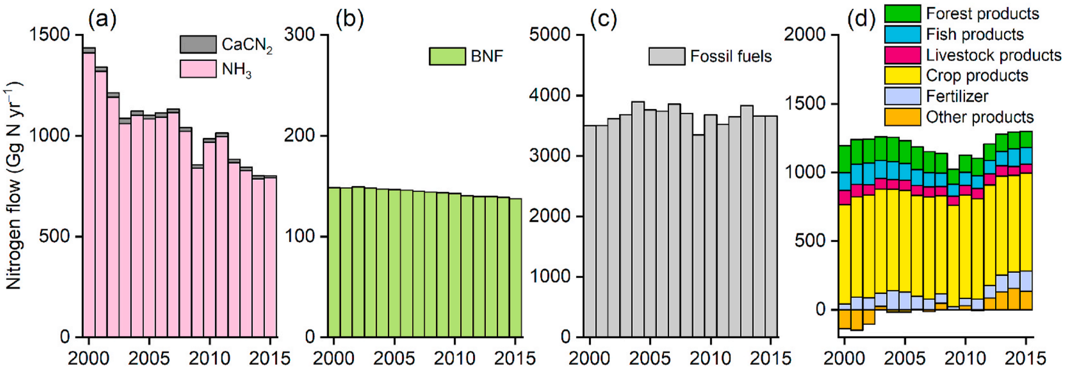


Fig. 1. Inflow of new reactive nitrogen to human sectors in Japan from 2000 to 2015. (a) Artificial fixation by the Haber–Bosch process for ammonia (NH_3) and the Frank–Caro process for calcium cyanamide ($CaCN_2$), (b) Biological nitrogen fixation at croplands and grasslands, (c) Net import of fossil fuels as coal and crude oil, and (d) Net import of other commodities. Negative value denotes net export.

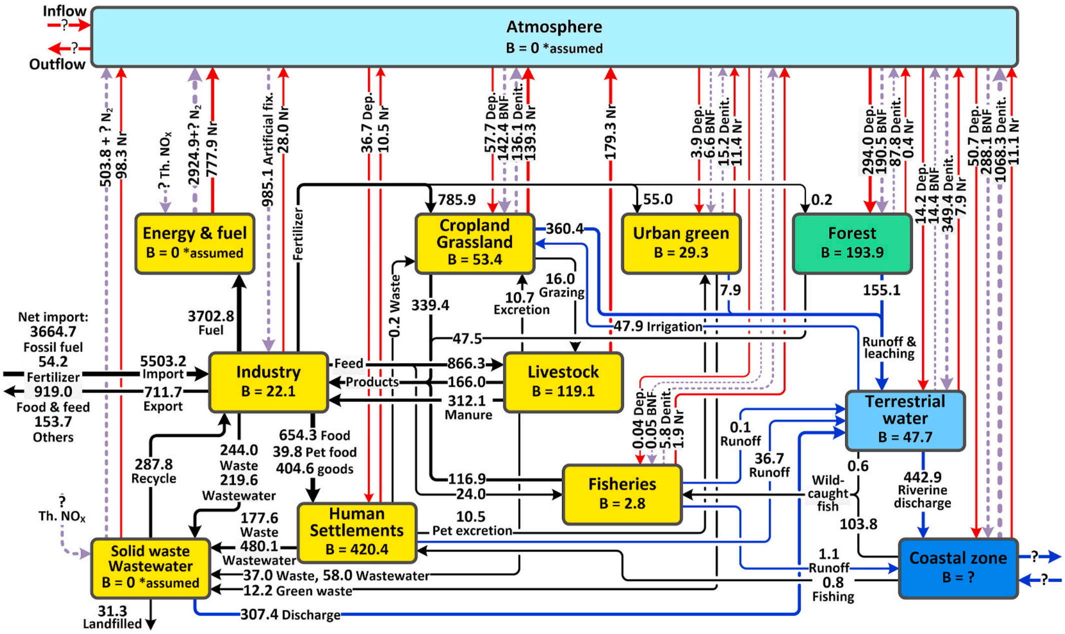


Fig. 2. Nitrogen (N) budgets in Japan in 2010 (unit: Gg N yr^{-1}). Black solid arrow, reactive N flow connected with human sectors; red solid arrow, reactive N flow via the atmosphere; purple dashed arrow, dinitrogen (N_2) flow via the atmosphere; blue solid arrow, reactive N flow with water; B, balance subtracting the total outflow from the total inflow. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Nitrogen inflow, outflow, and balance for each pool in Japan in 2010.

Pool	Inflow Gg N yr^{-1}	Outflow Gg N yr^{-1}	Balance Gg N yr^{-1}
Energy and fuels	3702.8	3702.8	0 (assumed)
Industry	7046.4	7024.3	22.1
Cropland	907.5	843.4	64.1
Livestock	882.3	763.2	119.1
Grassland	137.3	148.0	−10.7
Fisheries	128.6	125.8	2.8
Human settlements	1136.2	715.6	420.4
Solid waste	617.5	617.5	0 (assumed)
Wastewater	759.5	759.5	0 (assumed)
Forest	484.7	290.8	193.9
Urban green	76.0	46.7	29.3
Atmosphere	3191.0	3191.0	0 (assumed)
Terrestrial water	896.4	848.7	47.7
Coastal zone	782.8	?	?

3590 Gg N yr^{-1} of Nr in the human sectors was converted back N_2 via treatment of exhaust gas and wastewater (including complete denitrification at croplands). It was assumed that the remainder between the total N waste and the direct N waste was assigned as N_2 to close the N budgets. For wastewater treatment, the total input and discharge to water body in 2010 were 759 and 298 Gg N yr^{-1}, respectively, indicating the mean treatment efficiency of 61%. Also, atmospheric emissions accounted for 64% of the direct N waste, at 1960 Gg N yr^{-1}, and most of the remainder flowed into terrestrial water.

4. Discussion

4.1. Key features of Japanese N budgets

The total N supply as fossil fuels from 2000 to 2015, 3300–3880 Gg N yr^{-1} (Fig. 1) was the largest contributor to N waste as fuel NO_X due to combustion, whereas atmospheric NO_X emissions, including both thermal and fuel NO_X, were reduced to 722–925 Gg N yr^{-1} (Fig. 3). Controlled combustion technologies are known to reduce the creation of thermal NO_X (Nishimura et al., 1997). Atmospheric NO_X emissions are further reduced by technologies to convert NO_X into N_2 (collectively, denitration) (Tezuka, 1988; Matsumoto, 1997). Some technologies, like selective catalytic reduction, often use Nr as a reducer of NO_X. For example, NH_3 is used for point source like power plants (Nakajima and Hamada, 1996) and aqueous urea solution is used for mobile source like diesel vehicles (Hirata et al., 2005). Historically, the NOx emissions in Japan peaked around 1980, 1300 Gg N yr^{-1}, and have decreased steadily since around 1990 (Kurokawa and Ohara, 2020).

Import dependency is a feature of the Japanese N budgets, with a net import of 4330–5120 Gg N yr^{-1} (Fig. 1). A national N budgets approach does not count the Nr loss in other countries during the production of exports to the target country. It has been shown that Japan was the world largest net importer of N considering the Nr emissions associated with the production of exports to Japan, which was estimated to be 3300 Gg N yr^{-1} of net emissions outside Japan in 2010 (Oita et al., 2016a).

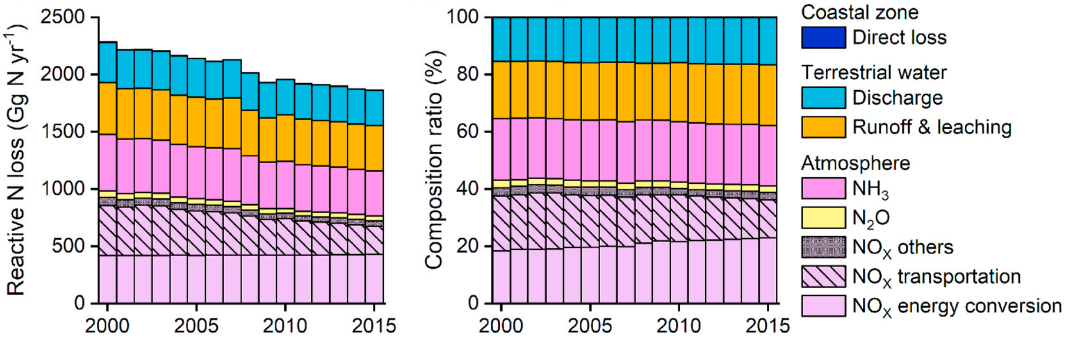
The total food supply in Japan from 2000 to 2015 ranged between 645 and 714 Gg N yr^{-1}, with a decreasing trend over time (Fig. S2). Since the total N input to Japanese food system ranged between 1840 and 2100 Gg N yr^{-1}, the input N not utilized for food, at 1190–1390 Gg N yr^{-1}, can be assumed to be N waste unless it was recycled. The consumed food N also eventually flows to solid waste and wastewater. As is the case in all countries, the food system in Japan is a major contributor to N waste.

The Japanese N budgets are compared with other countries with respect to the per capita food supply and Nr loss to the environment in Table 2. The per capita food supply in Japan in 2010 was lower than that in China, Denmark, and USA. The per capita Nr loss to air was lowest in China, highest in Canada, and was in the intermediate range in Japan. The per capita Nr loss to water was the lowest in Japan and the highest in Denmark, and the total loss to the environment was the lowest in Japan. The features particular to Japan were the small proportion of Nr loss to the total N waste (Section 3.2, Fig. 4) and that the per capita Nr loss was smaller than that in other countries.

4.2. Drivers of the changing Nr pressures and losses in Japan

The total N waste from the human sectors in Japan in 2010 was 5760

(a) Reactive N loss to the environment (by destination)



(b) Reactive N loss to the environment (by source)

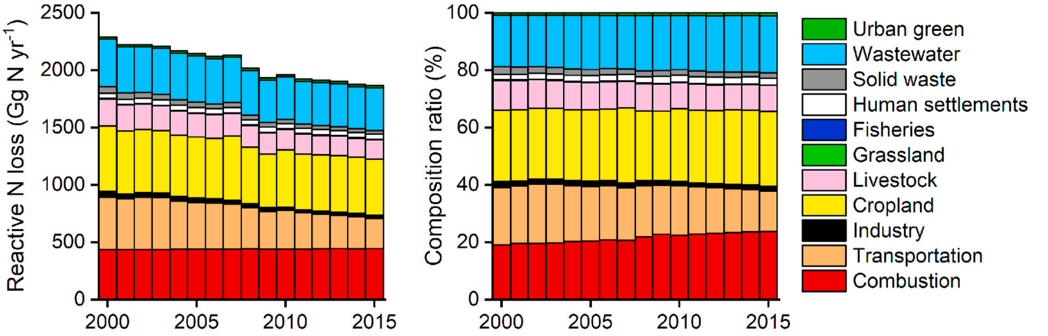


Fig. 3. Reactive nitrogen loss to the environment in Japan from 2000 to 2015. (a) By destination (atmosphere, terrestrial water, and coastal zone) (NO_X, nitrogen oxides; N_2O, nitrous oxide; and NH_3, ammonia) and (b) By source (human sectors).

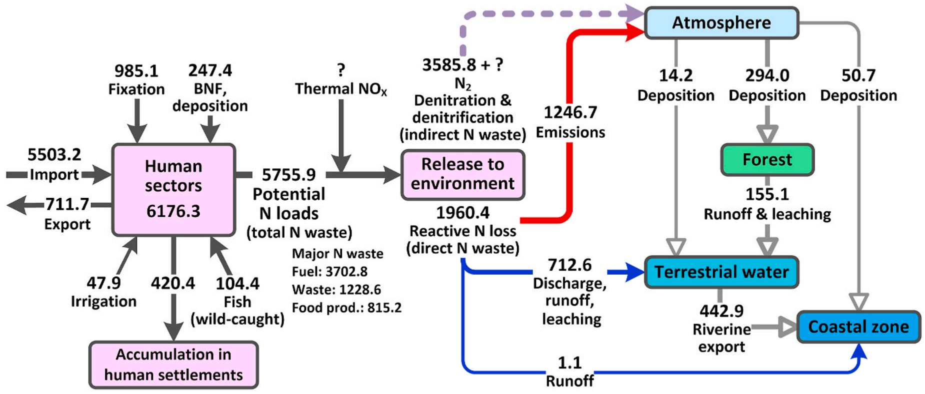


Fig. 4. Direct and indirect nitrogen (N) waste in Japan in 2010 (unit: Gg N yr^{-1}). Direct N waste, reactive N loss to the environment; indirect N waste, reactive N in human sectors converted to dinitrogen (N_2); black solid arrow, N flow in human sectors; red solid arrow, reactive N emissions from human sectors to the atmosphere; blue solid arrow, reactive N loss from human sectors to waters; purple dashed arrow, N_2 emissions from human sectors to the atmosphere; white solid arrow, N flow in the environment with anthropogenic and natural reactive N. Natural N flows were omitted here. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 2
Comparison of national nitrogen budgets for the domestic food supply and reactive nitrogen loss from human sectors to air and water (1 Tg N = 10^{12} g of N).

Country	Year	Population (million)	National total (Tg N yr^{-1}) and [Per capita] (kg N cap^{-1} yr^{-1})				Reference
			Domestic food supply	Reactive nitrogen loss from human sectors			
				Air	Water	Total	
Japan	2010	127.4	0.65 [5.1]	1.25 [9.8]	0.71 [5.6]	1.96 [15.4]	This study
Japan (food)	2005	127.0	0.67 [5.3]	–	–	–	Shindo et al. (2009)
China	2010	1341.3	7.6 [5.7]	6.7 [5.0]	18.1 [13.5]	43.8 [18.5]	Gu et al. (2015)
Canada	2007 ± 2	35	–	1.25 [35.7]	0.42 [12.0]	1.67 [47.7]	Clair et al. (2014)
Denmark	2010	5.6	0.035 [6.3]	0.14 [25.0]	0.16 [28.6]	0.30 [53.6]	Hutchings et al. (2014)
USA	2012	314.0	1.88 [6.0]	8.31 [26.5]	–	–	Sabo et al. (2019)

Gg N yr^{-1}, whereas the Nr loss to the environment was reduced to 1960 Gg N yr^{-1} (Section 3.2). The background factors which may have contributed to this are discussed using the various indicators shown in Fig. 5. The concrete values of the N budgets and the indicators in 2000, 2005, 2010, and 2015 are compiled in Table S2 (see Supplementary Information).

The TLRNE of Japan in 2008 estimated by Bleeker et al. (2013), 21.2 kg N cap^{-1} yr^{-1} was the second lowest among the OECD countries. The

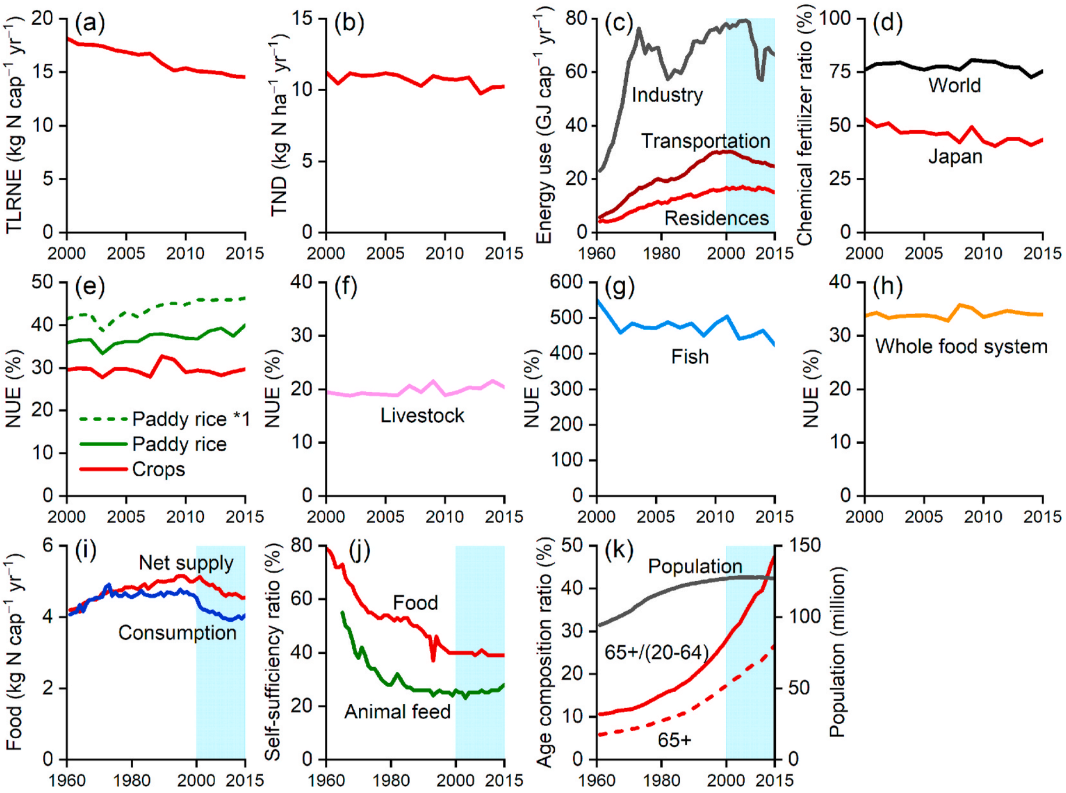


Fig. 5. Indicators related to nitrogen (N) budgets and human activities in Japan. (a) Trends in Reactive Nitrogen Loss to the Environment (TRNLE, per capita), (b) Trends in Nitrogen Deposition (TND, per area), (c) Per capita final energy consumption (residences, transportation, and industry), (d) Ratio of artificially fixed N used for chemical fertilizer, (e) Nitrogen use efficiency (NUE) of domestic crop production, additionally showing NUE of paddy rice (*1 NUE of paddy rice excluding irrigation N), (f) NUE of domestic livestock production, (g) NUE of domestic fish production, (h) NUE of the whole food system including international trade, (i) Per capita food supply and consumption (Hayashi et al., 2018), (j) Self-sufficiency ratio for food (calorie basis) and animal feed (total digestive nutrients basis), and (k) Ratio of population aged 65 years or over and old-age dependency ratio (65+ population/20–64 population).

TLRNE determined in this study was even lower, at 15.4 kg N cap^{-1} yr^{-1} in 2008, and was found to decrease over the period between 2000 and 2015 (Fig. 5). The large difference between the two studies is mainly ascribed to the different approaches whether conversion of Nr to N_2 was considered (this study) or not (Bleeker et al., 2013). Meanwhile, the Japanese N footprint including international trade ranged between 28 and 40 kg N cap^{-1} yr^{-1} (Shibata et al., 2014; Oita et al., 2016a). The low TLRNE of Japan can be attributed to the reduction in Nr loss to the environment by treatment (Section 3.2) and the low amount of domestic production due to the importing of food and feed (Section 4.1). The decrease in the TLRNE can mostly be attributed to the reduction in Nr loss, since little change was found in the Japanese population, the denominator of TLRNE, at 126.9–128.1 million during the period (SBJ, 2021).

As a pressure indicator, the TND indicates the potential impacts of N deposition on ecosystems (Section 4.4). The TND of Japan from 2000 to 2015 was in the range of 9.8–11.2 kg N ha^{-1} yr^{-1}, and decreased over time (Fig. 5). The nationwide N deposition was calculated using the datasets of seven models in the Coupled Model Intercomparison Project (CMIP6) (WCRP, 2021). Ban et al. (2016) reported a similar value of N deposition, 10 kg N ha^{-1} yr^{-1}, based on observations at remote sites in Japan from 2003 to 2012.

The per capita final energy consumption for transportation peaked around 2000 and then decreased, indicating a decrease in per capita activity (Fig. 5). This is likely one of the factors contributing to the decrease in NO_X emissions from transportation (Fig. 3). The final energy consumption in residences also decreased after the first decade of the 2000s, implying a further decrease in individual activity (Fig. 5). Despite this, the final per capita energy consumption in Japan from 2000 to 2015, at 115.9 GJ cap^{-1} yr^{-1} on average, was more than double the world mean of 51.3 GJ cap^{-1} yr^{-1}, as derived from the final energy consumption and global population (IEA, 2021; UN, 2019b). Japan was still one of the world's heavy energy users. Fossil fuels constituted the major N input to the Japanese budget (Fig. 1), of which according to our calculations only a small fraction was emitted to the atmosphere as Nr (Fig. 3) via combustion with abatement technologies reducing NO_X to N_2.

Little similarity was found between the supply and demand structure for N in Japan and the world. Approximately 80% of artificially fixed N was used for chemical fertilizer globally, that of Japan in 2000 was 53% and fell to over 40% after 2010 (Fig. 5). Besides being used to produce fertilizer, fixed N is used on a large scale in the production of synthetic fibers and plastics (Katagiri et al., 2018). The decreasing trend in NH_3 production (Fig. 1) is attributed to the decrease in the demand for chemical fertilizers (Fig. S2) and other raw materials (Katagiri et al., 2018).

The mean NUEs of domestic crop and livestock production were 29.5% and 19.7%, respectively, as shown in Fig. 5. The mean crop production NUE was lower than global mean value of 47% (Lassaletta et al., 2014). The NUE of paddy rice cropping, using most of the irrigated water (MLIT, 2014), was 37.0% on average. Using the equation in Lassaletta et al. (2014), which does not consider irrigation N, the value was 43.7%. Fish production had a very high NUE of 480% on average because as much as 89.1% of the supplied fish products were wild-caught fish. When international trade is considered, the whole food system NUE of Japan was 34.0% on average: this was as high as Sweden, UK, Portugal, and Italy among the thirteen European countries with the food system NUEs ranging between 10% and 40% in 2008 (Erisman et al., 2018). The relatively high NUE of Japanese food system was largely due to the high production NUE of fish products, and the assumption of a NUE of 100% for imported food and feed as no N loss occurred in Japan. This assumption makes sense to the purpose of this study considering the potential environmental impact to Japan; however, it would not make sense when considering footprint impacts to the world via Japanese imports.

The per capita food supply and consumption in Japan peaked in the late 1990s and then decreased as shown in Fig. 5. The discrepancy between food supply and consumption, i.e., consumer-level food loss (Liu et al., 2016; Hayashi et al., 2018), became larger in the years following 2000 despite the decreasing trends of food supply and consumption. This is reducible N waste.

The once-high self-sufficiency ratios of Japan in the 1960s decreased to ca. 40% and 26% for food and animal feed, respectively, in the years after 2000, as reflected in the import dependency of food and feed (Fig. 1).

Both the ratio of the population aged 65 years or over and the old-age dependency ratio in Japan have increased since 1961 and accelerated after 1990 (Fig. 5). Between 2000 and 2015, the ratio of the population aged 65 years or over increased from 17.4% to 26.6%, and the old-age dependency ratio increased from 27.9% to 47.5%. It is interpreted with the coincident decrease in per capita activity that the further aging of the population over time might result in further reductions in individual activity (Fig. 5).

The per capita GDP and private final consumption in Japan from 2000 to 2015 were relatively stable, at 4.03 ± 0.11 (SD) and 2.29 ± 0.04 million JPY $\mathrm{cap}^{-1}\ \mathrm{yr}^{-1}$, respectively (COJ, 2021). The per capita current balance and trade balance were one to two digits smaller than the GDP and the private final consumption (MOF, 2021). It is therefore reasonable to conclude that these economic indicators were at least apparently not associated with the reductions in individual activity (Fig. 5).

The decreasing trends of Nr loss to the environment after 2000 (Fig. 3) can be attributed to the following two factors: the effects of active emission controls and the decreasing activity numbers. The $-4.3\%\ \mathrm{yr}^{-1}$ rate of change from 2000 to 2015 (base year 2010) in transportation NO_X emissions (Fig. 3) was approximately three-fold larger than the $-1.5\%\ \mathrm{yr}^{-1}$ reduction in the transportation final energy consumption (Fig. 5). These figures suggest the two factors had quite an impact. In the case of emission controls, Wakamatsu et al. (2013) reported that the 2001 amendment to the Automobile NO_X Law led to reductions in atmospheric NO_2 concentrations in the following years. The rates of change for Nr loss to terrestrial water, the runoff and leaching from land of human uses and the discharge of treated wastewater were the same, at $-1.0\%\ \mathrm{yr}^{-1}$ (Fig. 3). The change rates for croplands, in terms of the chemical fertilizer input, organic fertilizer input, and crop production were -2.0%, -0.8%, and $-1.1\%\ \mathrm{yr}^{-1}$, respectively (Fig. S2). The reductions in both fertilizer input and crop production likely contributed to the reduction in runoff and leaching of N to terrestrial water (Fig. 3).

As shown in Fig. 5, the low NUEs of domestic crop and livestock production and the discrepancy between food supply and consumption indicate that there is further room for reducing N waste in the Japanese food system. That is, N waste can be reduced by improving the production NUE, reducing food waste, reducing overconsumption, and choosing food with a higher NUE (Hayashi et al., 2018; Oita et al., 2020). It is a fact that import dependency on food and feed reduces Nr loss due to domestic production. For example, the Japanese food N footprint increased by 9–10 kg N $\mathrm{cap}^{-1}\ \mathrm{yr}^{-1}$ assuming no imports (Shibata et al., 2014; Shindo and Yanagawa, 2017). It was suggested that the 2020 COVID-19 pandemic might have promoted local food production and consumption, in effect shortening the supply chains and reducing the risk of food insecurity (Laborde et al., 2020). In this case, a combination of adequate measures needs to ensure that N waste accompanied with the increased domestic food production does not increase. The high consumption of fish is a feature of the Japanese diet (Oita et al., 2016b, 2018; Hayashi et al., 2018). A high NUE is obtained by fish production due to wild-catch fish (Fig. 5), balancing with fishery resources is indispensable for sustainable consumption. It is important that aquaculture to compensate natural fishery resources is designed to reduce the N waste generated in production (Oita et al., 2016b).

4.3. Environmental Nr state in Japan

The annual mean concentrations of atmospheric NO_2 shown in Fig. 6 were lower than the environmental quality standard (EQS) for human health protection in Japan, i.e., 0.04–0.06 ppm or lower, with both for the median and mean values decreasing over time. This trend is consistent with that for NO_X emissions (Fig. 3). The achievement of EQS in 2010 was 100% at ambient air monitoring stations and 97.8% at roadside air monitoring stations (MOE, 2016).

The median values of the nitrate N (NO_3–N) and total N (T-N) concentrations in rivers, lakes, and coastal seas improved over the study period as shown in Fig. 6. The EQS of NO_3–N for human health protection in Japan is 10 mg N L^{-1} as the sum of NO_3–N and nitrite N: this was almost achieved in 2010, with 99.9% in rivers and 100% in lakes and coastal seas (MOE, 2011). The achieved EQS of T-N for environmental conservation in 2010 was very low for lakes, at 13.2%, and high for coastal seas, at 90.1% (MOE, 2011). Half of the Japanese lakes are categorized as eutrophic or mesotrophic (Kishimoto and Ichise, 2013). For lakes particularly classified as hypereutrophic (e.g., Lakes Kasumigaura, Teganuma, and Inbanuma), the main N sources were domestic wastewater and agriculture (Ibaraki Prefecture, 2017; Chiba Prefecture, 2017a, 2017b). While the NO_3–N concentrations in groundwater in the capital area decreased over time, they were higher than the surface water NO_3–N concentrations (Fig. 6). The achieved EQS of groundwater NO_3–N, same standard value as surface water, throughout Japan in 2010 was 95.7% in the general monitoring survey, 76.8% in the survey of the areas adjacent to polluted wells, and 52.8% in the regular monitoring survey of polluted wells (MOE, 2012). The main causes of groundwater pollution were primarily agriculture and secondarily inappropriate treatment of domestic wastewater (Yabusaki, 2010). Thus, despite the overall reduction in Nr loss to the environment (Fig. 3), N pollution was still an issue, particularly for lakes and groundwater (Sugimoto and Hirata, 2006; Tomiie et al., 2009; Matsuzaki et al., 2018).

4.4. Possible impacts of N pollution in Japan

Atmospheric NO_2 and water NO_3–N impact negatively on public health. Because the EQS was perfectly achieved (Section 4.3), the human health risk posed by NO_2 can be considered low in Japan. The EQS for surface water NO_3–N quality in Japan has also been well-achieved. The EQS of groundwater NO_3–N concentrations, however, was often violated particularly in and surrounding once polluted areas, which suggests that the long-term drinking of groundwater in these areas might be problematic (e.g., Amano et al., 2018).

The decreasing trend of Nr loss to terrestrial water (Fig. 3) is a good sign against the eutrophication problem. However, the T-N concentrations in many lakes and some enclosed coastal seas have exceeded the EQS for decades (MOE, 2011). By contrast, long-term total pollutant control might result in oligotrophication as has been reported in the Seto Inland Sea (Abo and Yamamoto, 2019). As eutrophication is also caused by excess phosphorous (Conley et al., 2009), water pollution control should combine an observation system of the N and phosphorous balance in waters and a scheme of flexible measures allowing adaptations due to changes in state (e.g., Tomita et al., 2016).

Atmospheric N deposition accelerates the N cycling in terrestrial ecosystems (Matson et al., 2002). The Japanese forested ecosystem near large urban areas has been subject to a large amount of N deposition and has exported nitrate to stream water as a result of N saturation (Orui and Mitchell, 1997; Ohte et al., 2001; Shibata et al., 2001; Yoh et al., 2001; Nishina et al., 2017). Concerns have been raised about the impact of increased N deposition with transboundary Nr transportation, particularly in the southwestern part of Japan (Morino et al., 2011; Chiwa et al.,

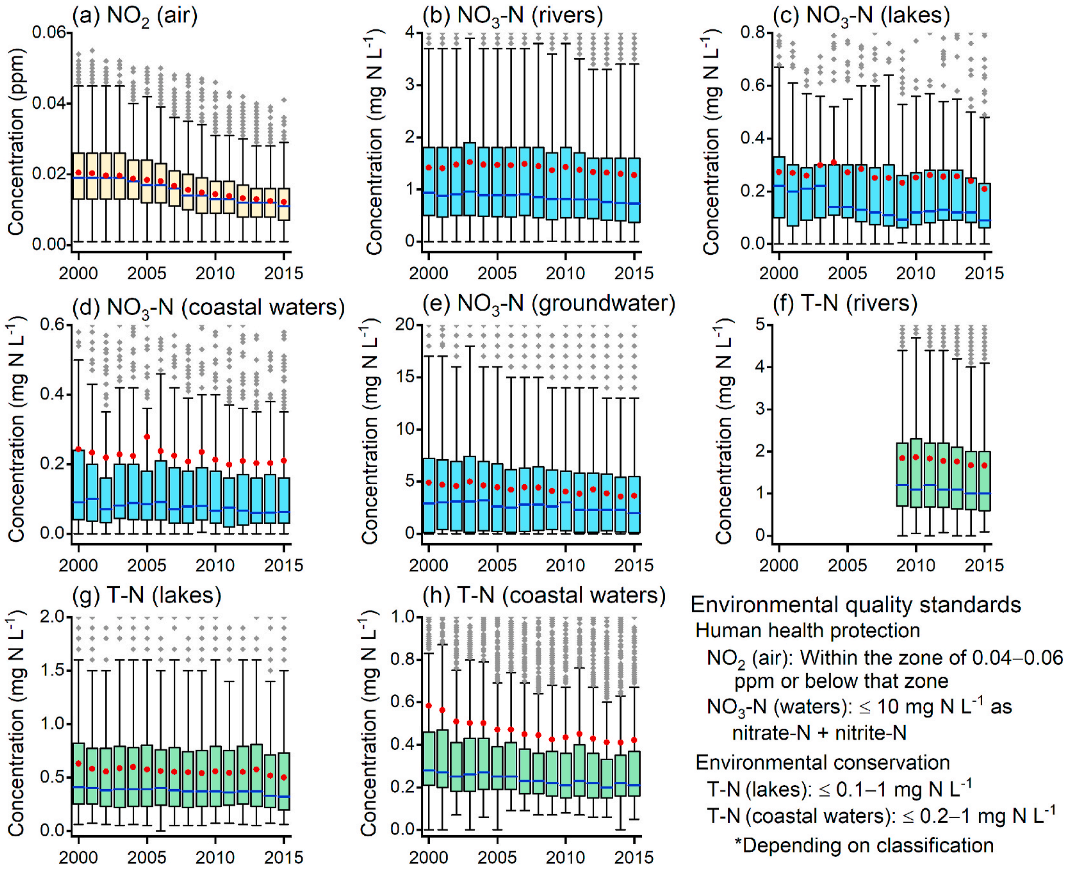


Fig. 6. Environmental concentrations of reactive nitrogen throughout Japan (other than groundwater) and in the capital area (groundwater). (a) Air, nitrogen dioxide (NO_2) ($n = 1385$), (b) Rivers, nitrate nitrogen (NO_3–N) ($n = 1744$), (c) Lakes, NO_3–N ($n = 218$), (d) Coastal seas, NO_3–N ($n = 486$), (e) Groundwater, NO_3–N ($n = 634$ to 984 depending on the year), (f) Rivers, total nitrogen (T–N) ($n = 3466$), (g) Lakes, T-N ($n = 391$), and (h) Coastal seas, T-N ($n = 1095$). Box, the range between 75% and 25% percentiles; blue line in box, the median; and red circle, the mean value. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2012; Sugimito and Tsuboi, 2017; Chiwa et al., 2019). The carbon dioxide (CO_2) fertilization effect on vegetation photosynthesis induced by elevating atmospheric CO_2 concentrations is downregulated by N limitations (de Vries et al., 2014; Wang et al., 2020). Increased carbon inflow to the forest ecosystem can impact the status of N limitations and saturation on a long-term basis (Groffman et al., 2018). The reduction in the TND in Japan from 2000 to 2015 (Fig. 5) suggests that such effects might be seen in Japanese forest in future. N deposition also causes biodiversity loss in terrestrial ecosystems due to the shift to N rich conditions (Bobbink et al., 2010). According to the critical loads of N deposition with respect to plant biodiversity (Bobbink et al., 2010), a global assessment was conducted using a threshold of 10 kg N ha^{-1} yr^{-1} (Bleeker et al., 2011). Despite reductions over time, the TND in Japan from 2000 to 2015 was around this threshold (Figs. 5), and 10 kg N ha^{-1} yr^{-1} was not necessarily high in Japan (Ban et al., 2016; Tan et al., 2018; Takahashi et al., 2020). Yu et al. (2014) indicated that Asian forests including Japan have a capacity of N uptake even under the N deposition of 10 kg N ha^{-1} yr^{-1}. Experiments to identify reliable thresholds of critical loads in Japanese terrestrial ecosystems are needed in conjunction with careful ecosystem monitoring.

4.5. *Towards estimating elaborated N budgets*

More quantitative information on the following items will improve estimations of the national N budgets.

1) NO_X creation and emissions with combustion: Identifying N fate in the fossil fuel supply and consumption is required because of the largest N flow in Japan (Fig. 1). Emission inventories provide details of post-treatment NO_X emissions, i.e., residual NO_X including both thermal and fuel NO_X. Detailed thermal NO_X creation data and the national mean denitration efficiency are necessary for more accurate estimates of the N budgets.
2) Uses of fixed N other than fertilizers: Half or more of the fixed N is destined for use in non-fertilizer applications in Japan (Fig. 5). In this study, only their upstream flows (inorganic and organic chemicals) were quantified because the downstream material flows were too complex to quantify without double counting in a material flow analysis. Both NH_3 and urea are also used as NO_X reducers (Section 4.1), while NH_3 as a fuel (Kobayashi et al., 2019) and as a hydrogen carrier (Guo and Chen, 2017) are still technologies currently being developed. A feasibility analysis of these uses considering possible Nr loss to the environment is needed.
3) Land-based N emissions to the atmosphere: In this study, data was obtained from the Japanese greenhouse gas inventory (NIES, 2021a; MOE, 2021a), the CHANS model (Gu et al., 2015), and other literature for emission factors of NH_3, N_2O, and N_2. Emission factors of NH_3 and N_2 which reflect the features of Japanese agriculture are limited. Although N_2 is harmless, N_2 flows are also necessary to close the N budgets. The most recent global N_2O budgets reported that agriculture is the greatest source of anthropogenic N_2O emissions, at 52% on average from 2007 to 2016, and that they are increasing significantly (Tian et al., 2020). In Japan, agriculture was the largest source of anthropogenic N_2O emissions, accounting for 44% in 2010, though the anthropogenic N_2O emissions have been decreasing since 2000 (NIES, 2021a). Therefore, a more elaboration of N_2O emission

factors reflecting the effects of measures would help providing more accurate estimates.

4) Contribution to secondary inorganic aerosols: According to the nationwide monitoring at ambient air stations in Japan in 2015, ammonium and nitrate accounted for 12% and 5%, respectively, of the total weight concentrations of fine particulate matters (PM2.5) (MOE, 2016). The EQS of PM2.5 for human health protection in Japan is 15 μg m^{-3} as the annual mean and 35 μg m^{-3} as the daily mean. The nationwide PM2.5 monitoring started in 2010. The achievement of the EQS at ambient air stations low at 32.4% in 2010 improved at 74.5% in 2015. Elucidation of the relationship between precursor emissions and PM2.5 concentrations is needed in addition to continuous atmospheric monitoring.
5) Atmospheric N input to land: The quantitative estimation of BNF involves a large degree of uncertainty (Fowler et al., 2013). Reliable factors of BNF rates for representative land uses are necessary for improving national N budgets estimation as well as numerical model simulation of N cycling. For atmospheric N deposition, the verification of model outputs to obtain the nationwide N deposition with observation data is important.
6) Hydrospheric N dynamics: Quantitative information of N flows in the groundwater and those between the surface water and groundwater is quite limited. Given the enormity of the challenge to achieve EQS for lakes and groundwater over the long-term (Section 4.3), elucidation of spatiotemporal N dynamics in these water bodies is especially important.

5. Conclusions

The N budgets in Japan from 2000 to 2015 indicated that Nr loss to the environment decreased over the period; however, the frequent violation of the water quality EQS particularly for lakes and groundwater was still a big issue in Japan. A large portion of the total N waste in human sectors was converted to N_2 via treatment. These treatment technologies require energy and resources to operate. In the challenge to achieve a more sustainable society, resource-consuming environmental measures should be replaced with more sustainable options, and further reductions in the total N waste are required by saving energy and resources, enhancing production NUE, recycling Nr such as organic fertilizer, reducing food waste and overconsumption, and choosing food with a higher NUE. A combination of these measures will result in a reduction in the cost for the energy and resources for treatment, reduce the demand for artificial N fixation, and eventually require less energy and resources for N fixation. Future decline and aging of population will result in lower gross human activity and perhaps reduce the capability to treat the N waste properly in a country as shown in the case of Japan.

The Japanese N budgets demonstrated the importance of Nr contained in traded goods for a country with active international trade. Japan's import dependency on food is a tradeoff between the advantages of reducing N waste by lower levels of domestic production and the disadvantages associated with food insecurity and accompanied Nr loss in countries providing these imports. The instability induced by the 2020 COVID-19 pandemic might have resulted in higher levels of domestic food production, meaning that N waste with food production will increase in the country. This presents the challenge of increasing the NUE of food production. The N waste associated with food should be reduced throughout the whole system. Using the Japanese N budgets and their deliverable indicators, such as NUE, it is possible to visualize the direct and indirect N waste in the N cascade in Japan. This provides a useful tool for evaluating the effects of policy and technological measures on N pollution control.

Credit author statement

Kentaro Hayashi: Conceptualization, Methodology, Investigation, Data curation, Visualization, Writing – original draft, Project administration. **Hideaki Shibata**: Methodology, Investigation, Data curation, Writing – review & editing. **Azusa Oita**: Methodology, Investigation, Data curation, Writing – review & editing. **Kazuya Nishina**: Data curation, Writing – review & editing. **Akihiko Ito**: Methodology, Investigation, Data curation. **Kiwamu Katagiri**: Methodology, Data curation. **Junko Shindo**: Methodology, Writing – review & editing. **Wilfried Winiwarter**: Writing – review & editing, Supervision, Project administration.

Main finding

Reactive nitrogen loss to the environment in Japan in 2010 was 1960 Gg N yr^{-1}, with decreasing trends in air and water emissions over time from 2000 to 2015.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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