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Dry climate aggravates riverine nitrogen pollution in Australia by water volume reduction

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Keywords: Nitrogen cycle; climate effects; surface water pollution; scenario prediction; policy

Abstract

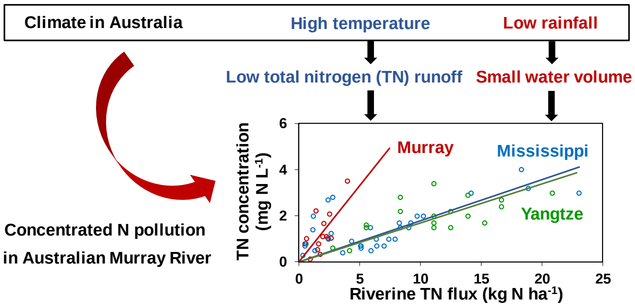
Freshwater is scarce resource and maintaining water quality is of great importance in dryland Australia. How water quality is affected by the dry climate and socio-economic influences in Australia remains widely unknown. Here, we find that agriculture activity dominates reactive nitrogen (N_r) emissions to water bodies. Such emissions not only contribute to deteriorating water quality in Southeastern Australia, but also harm marine ecosystems, including the Great Barrier Reef, a world natural heritage site. Dry and warm climate reduces the share of N_r emitted directly to water bodies, however, it increases the N_r concentration in surface water due to reduced water volume, leading to a threefold higher water N_r concentration compared to major rivers globally, e.g. in the USA or China. Business-as-usual socioeconomic development would increase the total N_r emitted to surface water by at least 43% in 2050, while effective mitigation measures could reduce N

runoff by about 27%. Advanced agricultural management strategies should be considered to reduce future environmental pressures due to N runoff in Australia.

Synopsis

This article reveals the concentration effect of dry climate on national aquatic nitrogen pollution, offering a reference of aquatic nitrogen control framework for global warm and dry regions.

Graphic for Table of Contents



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Introduction

To feed an increasingly wealthy population, human activities have more than tripled the nitrogen (N) input to terrestrial ecosystem compared to the pre-industrial era[1,2]. The anthropogenic perturbation of the N cycle results in a considerable amount of reactive nitrogen (N_r) losses into aquatic systems, including terrestrial surface water[3], groundwater, and marine water. Atmospheric N_r emissions could also affect surface water quality through dry and wet N deposition[4]. The safe operating space with regard to the planetary boundary of the natural N cycle has been exceeded by over two-fold[5]. Excessive N input to water bodies contributes to a range of environmental and social pressures, e.g. water eutrophication, marine ecosystem degradation, and health damage through unsafe drinking water[6-8]. To understand the underlying mechanisms of N losses to water bodies is crucial for global sustainable development goals (SDGs), especially on Ensure Availability and Sustainable Management of Water and Sanitation for All (SDG6)[9], and sustainable food production and overall environmental protection[10].

Aquatic N pollution is a topic of great concern in dryland Australia due to the shortage of water resources. About 40% of river basins show significant total nitrogen (TN, including nitrate-nitrogen (NO_3-N), nitrite-nitrogen (NO_2-N), ammoniacal-nitrogen (NH_4-N), and organic nitrogen) exceedances of national limit values[11], indicating prevalent water N pollution across Australia. Frequent algal blooms in Australian freshwaters caused by excessive N and phosphorus (P) cost about 140-185 million US dollars (USD) every year[12], and can further cascade to coastal and marine ecosystems[13], damaging the United Nations Educational, Scientific and Cultural Organization (UNESCO) natural heritage site of the Great Barrier Reef (GBR)[14]. Besides, climate also plays an important role in N loading and pollution. Large annual precipitation increases both N fluxes and water volumes[15-17], while high temperature promotes both N loading and instream N removal in rivers[17, 18]. Compared to other global regions, a dry and warm climate is unique to Australia, and how these factors affect water N pollution is not well understood. Meanwhile, as a world-leading producer and exporter of agricultural products, Australia has quintupled its crop production since 1960 with N fertilizer consumption increasing 36 times[19], and 71 million sheep and 26 million cattle are reared[20, 21]. Increasing our understanding about how agricultural activities, combined with the dry and warm climate, affect water quality is crucial for sustainable future development of Australia.

In this study, a comprehensive national N budget is compiled in order to quantify the effect of warm and dry climate on water quality. Previous studies mainly focused on empirical measurements of aquatic N_r releases from agriculture[22-24], neglecting emissions from other sectors such as forests, waste disposal and treatment, and atmospheric N deposition. In addition, the influence of a dry and warm climate has so far been rarely considered in previous N budget studies, leading to uncertainties when it comes to the level of water N pollution. In this paper, we conduct analyses on: (i) how the dry climate and intensive agriculture affect aquatic N_r emissions, as well as riverine N pollution in Australia; (ii) how terrestrial N runoff impacts the GBR marine ecosystem; and (iii) how riverine N runoff is projected to change under future climate change, which may enhance the dry and warm climate further, and socio-economic development.

Materials and Methods

Model and dataset. The study area for the assessment of N_r emissions covers the entire terrestrial territory of Australia, including inland surface water and groundwater bodies. We used the Coupled Human And Natural Systems (CHANS) model to evaluate annual N fluxes in Australia during the period 1961-2017. The whole country was divided into 14 subsystems, including cropland, grassland, feedlot, human, industry, aquaculture, forest, pets, urban green-land, solid waste, wastewater, atmosphere, surface water, and groundwater. Inputs, outputs, and the accumulation of N in each subsystem were calculated based on a mass balance approach. In the vertical direction, N deposition to land was considered as input to the system. Here we focused on two aquatic subsystems – surface water and groundwater - to identify N_r emissions from all other subsystems to water. A detailed description of the CHANS model can be found in Gu et al[25].

Data adopted in this study were divided into two parts: (i) information and activity data in Australia, including population, N fertilizer application, crop/livestock production, land use, energy consumption, etc., all derived from global statistics websites (FAO[26], IFA[19]) and the Australian Bureau of Statistics[27]; (ii) diverse parameters (e.g. N content in crops) and emission factors (EFs) for various sources, obtained from the literature and previous studies. Details can be found in Table S2-S5 in Supporting Information.

Emission distribution and validation. The spatial emission distribution of national TN runoff in Australia in 2017 was estimated as follows: We introduced the parameter "emission intensity[28] (kg N ha^{-1} yr^{-1})" to describe the rate of emissions across all regions of the country. Emissions originating from each subsystem were quantified for corresponding regions on the Australian land use map[29]. Accordingly, a N_r emission quantity was divided by the area of respective regions to calculate the emission intensity by sector. Emissions from grazing animals were allocated to pastures in 58 Natural Resource Management (NRM) regions in Australia. Emissions from all other sources were divided geographically in equal measure. The observations of annual average TN concentration in surface water were adopted from monitoring sites, set up by local Environment Protection Authority (EPA) cooperating with local governments and other monitoring reports[30-36]. The observation details, including sampling methods, periods and locations, can be found at EPA websites above, as well as in National Water Quality Management Strategy[37]. Hotspots of TN runoff were identified based on 58 NRM regions[38] in 8 states and territories, while water quality observations were identified based on 77 water regions, a secondary area divisions of 12 Australian drainage basins[38] (maps embedded in Figure 2b).

Terrestrial N loss to GBR has been analyzed. In this paper, the specific GBR catchment area was defined as the terrestrial river basins directly drain into the GBR lagoon[39]. Government estimates of TN runoff in the GBR catchment in Figure 2c were conducted by state of Queensland[40].

10,000 Monte Carlo simulations were executed to estimate the 99% confidence intervals of aquatic N emissions in Australia during the study period. For every emission item, Coefficients of Variation (CVs, %) were applied to data involved in the calculation (including activity data, parameters, and EFs), based on data origins and properties. Details can be found in Table S6-S7 in Supporting Information.

River comparisons. To explore the impact of climate on riverine N pollution in Australia, we compared the longest and most important river, Murray River in Australia, with other two national longest rivers, Mississippi River in the USA and Yangtze River in China. Runoff modulus was defined as the water runoff generated per unit area of a river basin in a year[41]. Emission per discharge referred to TN emissions divided by water volume discharge in a year. The riverine TN flux (kg N ha^{-1} yr^{-1}) was defined as the annual flux of N transported by the river to the ocean, divided by river basin area. The net anthropogenic nitrogen inputs (NANI, kg N ha^{-1} yr^{-1}) of watersheds included the use of synthetic N fertilizer, N fixation associated with agricultural crops, atmospheric deposition of oxidized N, and the net movement of N into or out of the region in human food and animal feeds[42]. The N flux and NANI for the Murray River basin were evaluated in this study. N fluxes for the Mississippi River basin were simulated using 2012 SPARROW Models conducted by U.S. Geological Survey (USGS)[43], and the NANI was adopted from McIsaac et al[44]. N fluxes for the Yangtze River basin originated from Wang et al.[45], while the NANI was derived from Wang et al[46]. The average of 154 global watersheds were summarized by Howarth et al[42]. The dots in Figure 4b present pollution patterns of small regions composing the river basins. For the Murray, Mississippi, and Yangtze rivers, the regions were defined as secondary river basins, states, and provinces, respectively. TN concentrations were derived from literature and previous studies[43, 47, 48].

Driving analysis and future trends. Driving analysis, defined as the correlation analysis revealing main driving factors of regional N runoff intensities, has been conducted with multiple linear regression methods using StataSE 15 software, to determine the contributory factors of TN emission intensity. Both independent and dependent variables were converted to averages for each NRM region. Five factors included disposable income per capita (DI), agriculture production value (AGRI), mean annual temperature (MAT), mean annual rainfall (MAR), and population density (PD), of which AGRI, MAT, and PD were found to have significant correlations with emission intensity. The analysis of variance was conducted with three significant factors and results are shown in Table 1.

Based on the regression analysis, future TN runoff intensities have been simulated at time steps of every five years for the period from 2020 to 2050. Five scenarios based on SSPs were modelled. The GDP, population, and temperature could be predicted for each SSP narrative[49]. The adoption rates for measures in the controlled emission scenario (CES) were assumed to increase steadily, from 0% in 2017 to 100% in 2050, at 14.3% for each five year time step.

The values of three contributory factors were calculated based on the results, and we assumed that agricultural production values would be proportional to GDP. The predictive average TN runoff intensity for each NRM region was distributed among the area based on the land use map, supposing that emissions from all sectors would increase evenly in the same multiple.

Cost and benefit. Emission abatement cost for each agricultural strategy in the CES were estimated separately. The unit cost of implementing riparian buffer strips in grazed watersheds was 6.4 USD kgN^{-1}, from official calculations[50]. The unit cost for implementing widespread use of controlled-release urea (CRU) on croplands was 35.9 USD kgN^{-1}, based on the predicted average price of N-permits trading in the GBR catchement[51]. For feedlot improvements, since the price of bran is lower than dry-rolled corn, implementation cost was assumed to be negligible. Societal benefits of N runoff reduction included environmental and health benefits[6], amounting to a total price of 18.9 USD kgN^{-1}.

Results and Discussion

Hotspots of N_r emissions to water. Australia emitted a total of 292 Gg N yr^{-1} to surface water and 972 Gg N yr^{-1} to groundwater in 2017. Compared with other countries, Australia has lower aquatic N_r emissions and a higher air/water emissions ratio (Table S1 in Supporting Information), mainly caused by the warm and dry climate and hence reduced rainfall. On the one hand, high temperature increases N_r emissions to air; on the other hand, low annual average rainfall (504 mm^{52}) and a low rate of river discharge largely limit loading and transport of N_r to aquatic systems[53] (Figure 1). Agriculture is the dominant emission sector for aquatic N_r, with livestock production contributing two thirds of the N runoff, and over 80% of N leaching. In 12 Australian drainage basins (Figure 2a), Murray-Darling ranked first with 87 Gg N yr^{-1} emissions to surface water, followed by Northeast Coast and Southwest Coast. The top three basins cover almost all of the areas of Wheat belt, Wheat/Sheep belt, and intensive grazing zones in Australia (Figure 2b)[20, 21, 54, 55]. With 26 billion USD production value annually, livestock and cropland industries in Australia account for only 1.9% of national Gross Domestic Product (GDP)[56, 57], but take up 95% of total aquatic N_r emission. The national water reform work program has been devoted to managing water allocation and trying to mitigate nutrients pollution from agriculture industry[58].

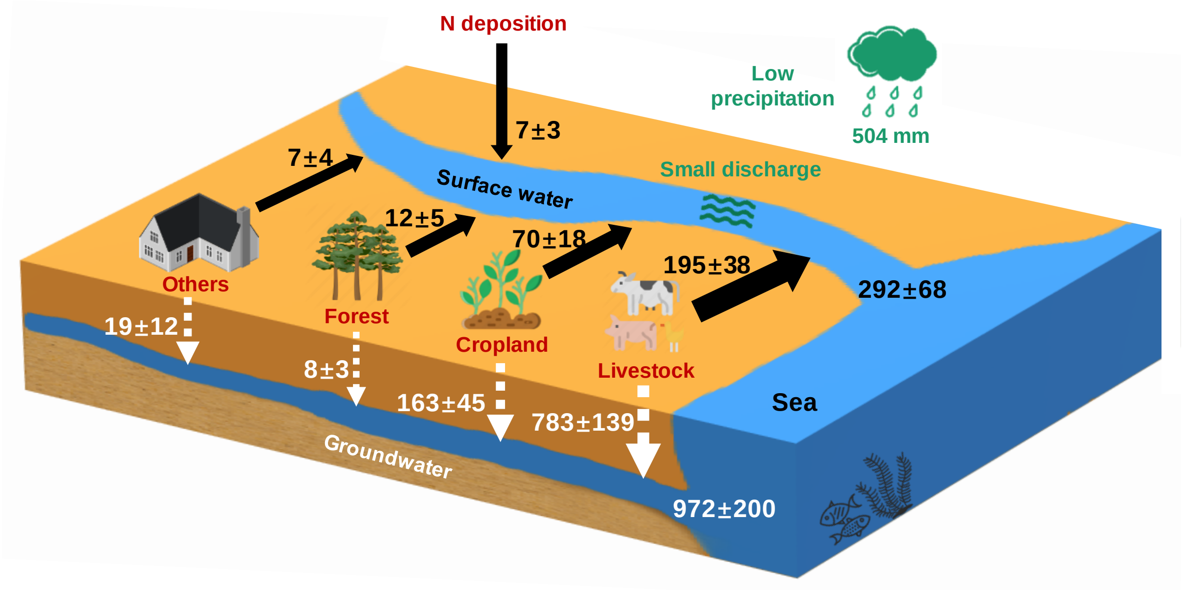

Figure 1. Aquatic N_r emission patterns in Australia in 2017. Units are Tg N yr^{-1}. The emission inventories include both natural and anthropogenic emissions. Agricultural sources dominated both surface water and groundwater N_r releases. The

low annual rainfall (504 mm) and small river discharge in Australia largely limit loading and transport of N_r within aquatic systems.

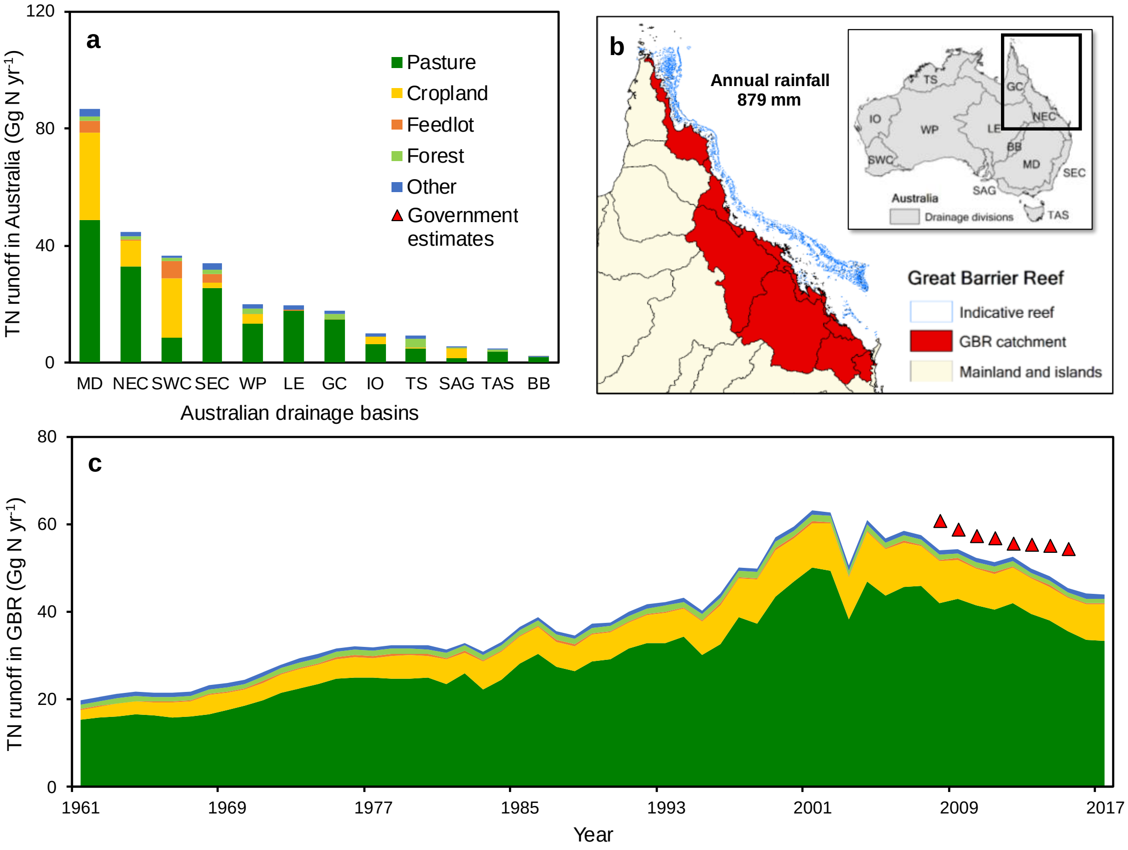

Figure 2. TN runoff patterns in Australia and Great Barrier Reef (GBR) catchment. a, TN runoff among 12 Australian drainage basins in 2017; **b**, distribution maps of drainage basins and GBR catchment; **c**, TN runoff trend in GBR catchment during 1961-2017. Legends of **c** are shown in **a**. GBR catchment area was defined as the river basins directly drain into the GBR lagoon. The government estimates of TN runoff in GBR catchment were conducted by state of Queensland[40]. MD: Murray-Darling; NEC: Northeast Coast; SWC: Southwest Coast; SEC: Southeast Coast; WP: Western Plateau; LE: Lake Eyre; GC: Gulf of Carpentaria; IO: Indian Ocean; TS: Timor Sea; SAG: South Australian Gulf; TAS: Tasmania; BB: Bulloo-Banncannia.

Figure 3a depicts the spatial distribution of TN runoff in 58 NRM regions across Australia in 2017. Pastures in Victoria were the most prominent hotspots, with an average TN emission intensity of more than 5 kg N ha^{-1}. Aquaculture, feedlot, and waste treatment sectors also had highly variable emission intensities (>50 kg N ha^{-1}), but were overall insignificant with small areas. Figure 3b shows the monitoring results of TN concentrations in surface water for 77 watersheds. The spatial emission patterns evaluated in this study agreed well with monitoring results (Figure 3c and 3d). Emission hotspots in Victoria were spatially consistent with high N pollution (TN>3.0 mg N L^{-1}) in south Murray-Darling and Southeast Coast basins in Australia. Both grazing sheep and cattle subject to high stocking rates in Victoria (0.45 head cattle and 1.73 head sheep per ha[20, 21]), leading to substantial N_r emissions to surface water. Pastures in Australia consume approximately 35% of total water supply, identifying extensive grazing and irrigation for the livestock industry as important causes of N loading of riverine systems[59].

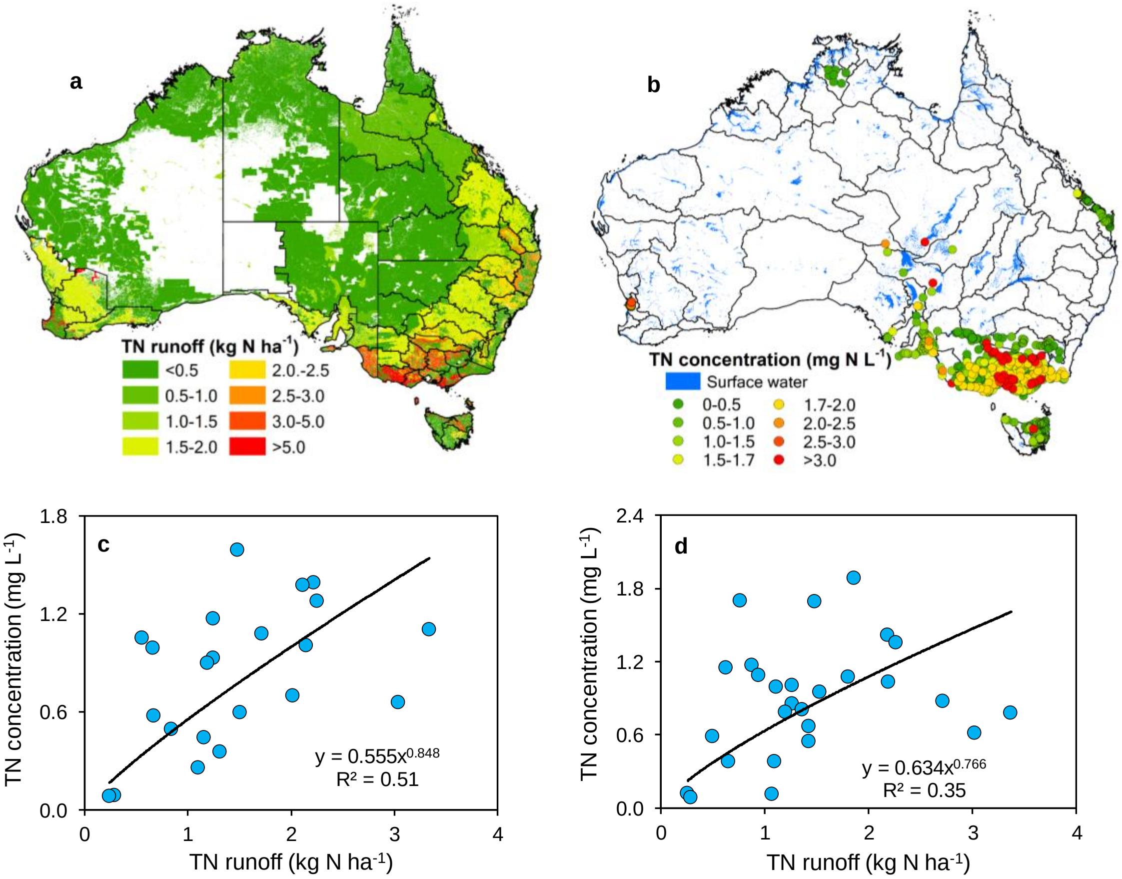

Figure 3. Spatial distribution of TN runoff and validation. **a**, spatial distribution of TN runoff in 58 Natural Resource Management (NRM) regions across Australia in 2017; **b**, the monitoring results of TN concentration in surface water in 77 water regions; **c**, correlations between TN runoff and TN concentration based on NRM regions; **d**, correlations

between TN runoff and TN concentration based on watersheds. TN runoff hotspots were mainly located on pasture lands of Victoria. The spatial emission patterns agreed well with monitoring results.

Dry climate aggravates N pollution. The Murray River is the longest and one of the most representative rivers in Australia, and the Murray-Darling Basin generates about 40% of Australia's total value of agricultural production[27]. Compared with similar global regions such as the Mississippi River in the USA and the Yangtze River in China, the Murray River has lower runoff modulus (2.3×10^{-5} m^3 s^{-1} ha^{-1}, Figure 4c), defined as the water runoff generated per unit area of a river basin in a unit time[41]. Statistical analysis suggests that a 1% change in mean annual rainfall would lead to a 2-3% change in mean annual riverine runoff[60]. However, in recent years, the amplification effect of rainfall on runoff change in the Murray River catchment were higher than most past periods and the long-term average, indicating that water runoff is decreasing while rainfall levels remain broadly constant[60], increasing the risk of droughts.

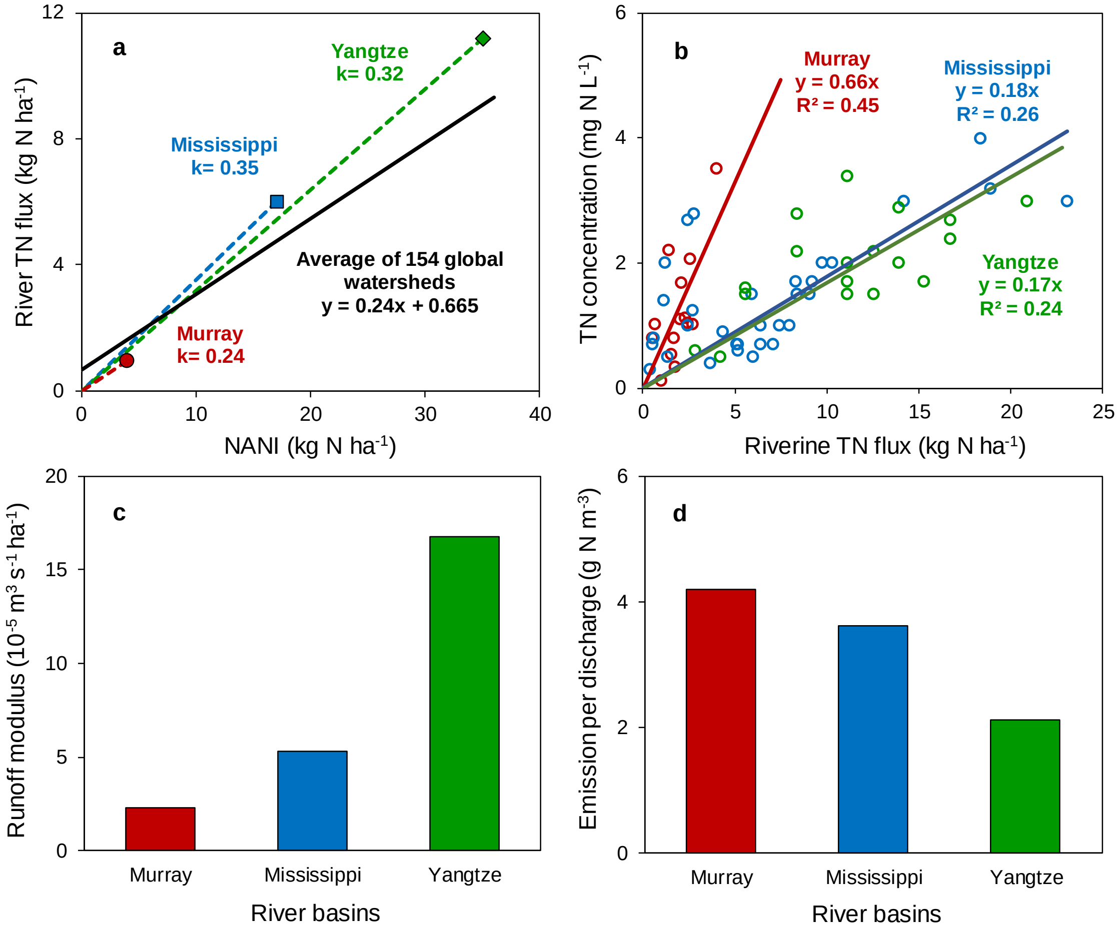

Figure 4. TN fluxes and N pollution in Murray, Mississippi, and Yangtze River. a, the relationship between river TN flux and net anthropogenic nitrogen inputs (NANI); **b**, N pollution was aggravated in Murray River compared to other rivers; **c**, low runoff modulus in Murray River; **d**, high emission per discharge in Murray River. The average of 154 global watersheds was gathered and analyzed by Howarth et al[42]. Dots in Figure 4b present pollution patterns of small regions composing the river basins. For Murray, Mississippi, and Yangtze River, the region was defined as secondary river basin, states, and provinces, respectively.

Annual water balance is a critical factor for loading and transport of TN within aquatic systems[53]. Low rainfall and water discharge lead to relatively small river TN fluxes. Annually, 102 Gg N is loaded into the Murray River, which represents only about 5% of comparable N loads in the Mississippi and Yangtze rivers[43, 45]. In the Murray river basin, only 24% of the net anthropogenic nitrogen input (NANI) is exported into the river, much lower than figures for the Mississippi (35%) and the Yangtze (32%) rivers, as well as the global average (Figure 4a). As a consequence of the low riverine TN emissions, a relatively large amount of N is retained, mostly emitted to the atmosphere through chemical and biological processes, e.g. ammonia vitalization[61], contributing to atmospheric N_r pollution.

Despite an overall low level of N_r emissions, the Murray River has the largest N_r emission per discharge (4.2 g N m^{-3}, Figure 4d) compared with other rivers and a three times larger TN concentration is observed, compared to the Mississippi and Yangtze rivers (Figure 4b). Despite the prevailing dry climate contributing to a reduction in N emissions, it aggravates N pollution of surface water through reducing the water volume. The warm and dry climate and high TN concentration in rivers provide prime conditions for blue green algae to thrive, leading to widespread water eutrophication[47]. Global warming could add further pressures on Australian inland water environments, indicating an urgent need for further research and policy measures to focus on the improvement of surface water quality under a changing climate[62].

Riverine N_r threats to the GBR. The GBR is one of the world's largest coral reef on Earth[13], and a unique UNESCO world heritage site, contributing 6.4 billion USD in value added to the Australian economy in 2015–16[63]. In this paper, the specific GBR catchment area was defined as the terrestrial river basins directly drain into the GBR lagoon[39] (Figure 2b). TN runoff in GBR catchment increased from 20 Gg N yr^{-1} in 1961 to 63 Gg N yr^{-1} in 2001, then fluctuated and decreased to 44 Gg N yr^{-1} in 2017 (Figure 2c). N_r pollution has been identified as the key threat to the GBR ecosystem[40, 64]. The growth of TN emissions in the previous four decades were a result of increasing fertilizer application and grazing livestock numbers[39]. The severe drought in 2003 largely reduced livestock numbers[65], especially sheep, thus reducing pasture TN emissions. Accordingly, the policy to protect the GBR – the Reef Water Quality Protection Plan – was launched in 2003[66]. Since then, TN releases to the GBR lagoon have been continually decreasing, a consequence of the improved management practices of all land-based activities, especially from agriculture, and nature conservation measures[67].

With 82% of the area occupied by agriculture[13], the GBR catchment contributes to a large extent to overall food production in Queensland. Pasture dominated the TN release, contributing 72-80% (Figure 2c). With 6.3

million cattle[20] and 31 million hectares of grazed land[68], livestock rearing reduced vegetation land cover on pastures and contributed to surface and sub-surface soil erosion, which constitute a main source of particulate nitrogen loading in rivers[40]. The Fitzroy river basin showed the largest TN emission intensity on pastures of all rivers in the GBR catchment, contributing to half of the grazing TN runoff. The Reef 2050 Water Quality Improvement Plan has conducted case studies for grazing emission hotspots[68], and the improvements require a combination of direct (e.g. gully stabilization) and indirect (e.g. cover and run-off management) techniques[40].

TN runoff from cropland in GBR catchment has increased from 2.3 Gg N yr^{-1} in 1961 to 11.4 Gg N yr^{-1} in 2004, and since decreased to 8.3 Gg N yr^{-1} in 2017 (Figure 2c). Sugarcane cropping is the dominant land use in cropping system among GBR catchment, with total area of 0.4 million hectares[68]. Reduction of fertilizer application rates to sugarcane could substantially decrease N runoff[23], and improved irrigation management could also effectively constrain aquatic N_r emissions while broadly maintaining yields[69].

Future projections of N runoff with climate change and economic development. Agricultural production value (AGRI), population density (PD) and mean annual temperature (MAT) are key determining factors for riverine N emissions (Table 1). This is consistent with the assumption that about 90% of TN runoff originates from agriculture, and the rest is mainly contributed from sources related to human activities. Therefore, more agriculture and people result in more N runoff to surface water. Meanwhile, MAT has a significant negative impact on N runoff ($p<10^{-4}$). High temperature increases atmospheric N_r emissions, such as NH_3, as well as water evaporation, which reduces TN runoff to surface water. On the basis of the analysis of these drivers, we conducted scenario simulations to predict TN runoff under climate change and different development pathways (Figure 5). Shared Socioeconomic Pathways (SSPs) represent the different challenges for mitigating and adapting to climate change. The five SSPs assessed include a variation of challenges to adaptation and mitigation, with the assumption of both being low (SSP1), both being high (SSP3), and variations of low/high (SSP5), respectively high/low (SSP4). Finally, a balanced scenario was included, reflecting both challenges considered ‘intermediate’ (SSP2)[49, 70].

Table 1. Variance analysis of factors contributing to TN runoff intensity (kg N ha^{-1}) among 58 Natural Resource Management (NRM) regions in Australia. AGRI: agriculture production value (billion USD); MAT: mean annual temperature (°C); PD: population density (person ha^{-1}). SS: sum of squares; df: degree of freedom; MS: mean square; F: F statistic; Significance F: the probability that the null hypothesis cannot be rejected; R^2: R-squared.

Source	SS	df	MS	F	Significance F	R^2
Model	14.63	3	4.88	22.72	1.26E-09	0.57
Residual	10.95	51	0.22			
Total	25.58					

	Coefficients	Standard error	t Stat	P-value	Lower 95%	Upper 95%
AGRI	0.85	0.14	5.93	2.81E-07	0.56	1.14
MAT	-0.071	0.016	-4.52	3.81E-05	-0.10	-0.039
PD	0.060	0.023	2.59	0.012	0.014	0.11
Intercept	1.82	0.32	5.71	6.16E-07	1.18	2.46

Historically, TN runoff increased from 224 Gg N yr^{-1} in 1961 to 336 Gg N yr^{-1} in 2001, then declined to 291 Gg N yr^{-1} in 2017. Agriculture dominated these emissions, and fluctuations were mainly a consequence of livestock number changes. In our scenario analysis, by 2050, the highest TN runoff in Australia (778 Gg N yr^{-1}) occurred under SSP5 with high fossil-fuel utilization, achieving the largest agriculture production value, population density, and temperature (Figure 5a). In this scenario, all of eastern Australia showed extremely high potential for N pollution in surface waters, with an emission intensity three times higher than that of 2017 (Figure 5f and Figure 3a). SSP1, SSP4, and SSP2 would lead to a TN runoff of 616, 590, and 542 Gg N yr^{-1} in 2050, respectively (Figure 5b, e, and c). Areas along the eastern seaboard of Australia were main hotspots, especially where agricultural lands are located, compared to 2017 where the hotspots were mainly concentration in south Victoria.

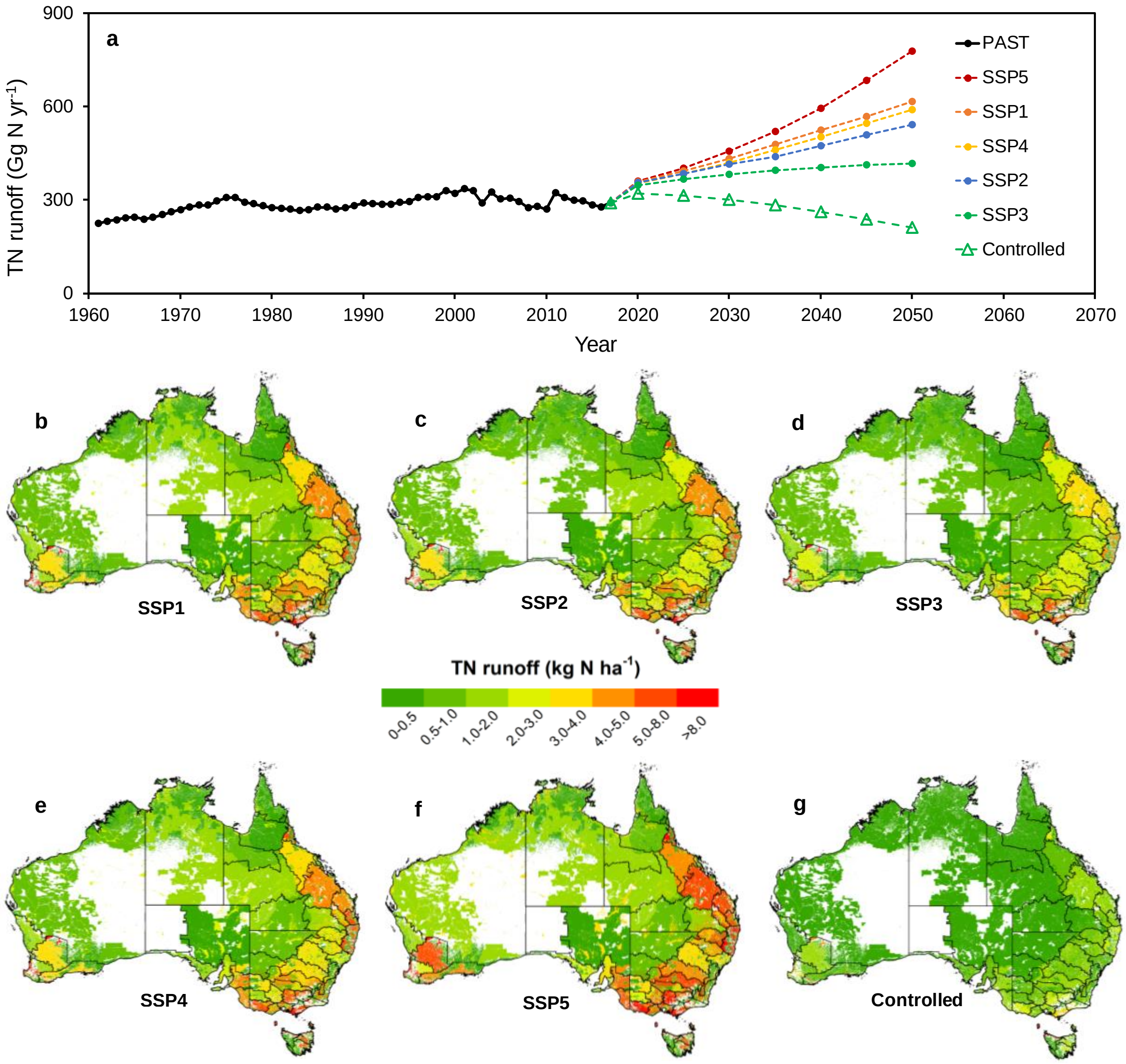

Figure 5. Scenario predictions of TN runoff patterns in Australia with Shared Socioeconomic Pathways (SSPs). a, TN runoff trends from 1961 to 2017, and predictions from 2020 to 2050. **b-f**, predicted distribution of TN runoff in 2050 for SSP1, SSP2, SSP3, SSP4, and SSP5. **g**, predicted distribution of TN runoff in 2050 for controlled emission scenario (CES).

In the SSP3 scenario, projected TN runoff declined to the lowest level of 416 Gg N yr^{-1} emitted to surface water in 2050, however still about 43% higher than emissions in 2017. Pastures in Victoria showed the highest emission intensities (Figure 5d). This scenario marked the lowest agriculture production value and population density, and the second highest temperature among all five SSPs (Table S8 in Supporting Information). Compared to the most sustainability-driven scenario (SSP1), SSP3 featured regional rivalry, focusing on national domestic issues, and de-prioritizing technological and economic developments[70]. In this scenario, additional efforts to foster technological development has the potential to reduce N_r losses to waters.

To evaluate technological effects, we further created a controlled emission scenario (CES) based on SSP3 (Figure 5g), which considered the improvements of agricultural managements, including implementation of prioritizing controlled-release urea (CRU) fertilizer[71], riparian buffer strips in grazed watersheds[71], and corn replaced with 30% bran in feedlots[72]. N runoff emissions from cropland, pasture, and feedlots under CES are projected to decrease by 60%, 54%, and 30% in 2050, respectively. A total of 211 Gg N yr^{-1} national runoff in 2050 is suggested, 27% lower than emissions in 2017, indicating the feasibility of TN runoff control in Australia with existing measures. The thorough implementation of those measures on water pollution control would provide an efficient way to improve the quality of drinking water in Australia, promoting the achievement of SDG6. The total cost of implementing the aforementioned abatement measures were estimated at approximately 3.0 billion USD. The net benefit indicates that this controlled scenario should be a feasible plan to balance environmental, economic and societal policy objectives for Australia in the future.

Population density and income growth would exacerbate N_r emissions to both atmosphere and hydrosphere in the future. Besides, as the mean annual temperature in Australia continually increases[52], climate change with dryer and warmer periods has the potential to increase N_r emissions to air over direct releases to water, while high temperature would worsen the water pollution. Any future increase in N runoff from land would both affect inland ecosystems, public health, and fresh water supply[62], as well as contribute to threats to marine ecosystems and the commercial use of marine resources[13]. Targeted strategies with a focus on emissions from agricultural sources[73, 74], like described in the CES, could be effective and an essential building block to constrain environmental N_r emissions. More efforts from government bodies such as the National Environment Protection Council to assess and maintain air quality, and cooperation with State and territory governments to provide information and tools to manage water resources would be essential for the effective implementation of such strategies.

Supporting Information. Table S1-Table S8.

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Author contributions. B.G. and Y.S. designed the study. Y.S. performed the research. Y.S. and X.Z. analyzed the data and interpreted the results. Y.S. and B.G. wrote the paper, S.R., D.C. and J.X. contributed to the discussion and revision of the paper.

Data availability. All data, associated protocols, and materials in this paper are publicly available. Data used to evaluate the N fluxes are divided into two parts. Information and activity data are all derived from global statistics websites (e.g. FAO, IFA) and the Australian Bureau of Statistics; diverse parameters and emission factors are obtained from the literature and previous studies, as mentioned in Method section and SI Appendix. All adopted data and resources, as well as the entire emission inventories are shown in SI Appendix. Detailed description of protocols to calculate these N fluxes in the CHANS model is available from https://person.zju.edu.cn/en/bjgu#930811.

Competing interests. The authors declare no competing interests.

Reference

(1) He, B.; Kanae, S.; Oki, T.; Hirabayashi, Y.; Yamashiki, Y.; Takara, K. Assessment of global nitrogen pollution in rivers using an integrated biogeochemical modeling framework. *Water Res.* **2011,** *45* (8), 2573-2586.

(2) Galloway, J. N.; Dentener, F. J.; Capone, D. G.; Boyer, E. W.; Howarth, R. W.; Seitzinger, S. P.; Asner, G. P.; Cleveland, C. C.; Green, P. A.; Holland, E. A.; Karl, D. M.; Michaels, A. F.; Porter, J. H.; Townsend, A. R.; V Osmarty, C. J. Nitrogen Cycles: Past, Present, and Future. *Biogeochemistry* **2004,** *70* (2), 153-226.

(3) Wang, M.; Houlton, B. Z.; Wang, S.; Ren, C.; van Grinsven, H. J. M.; Chen, D.; Xu, J.; Gu, B. Human-caused increases in reactive nitrogen burial in sediment of global lakes. *The Innovation* **2021,** *2* (4), 100158.

(4) Gao, Y.; Zhou, F.; Ciais, P.; Miao, C.; Yang, T.; Jia, Y.; Zhou, X.; Klaus, B.; Yang, T.; Yu, G. Human activities aggravate nitrogen-deposition pollution to inland water over China. *National Science Review* **2020,** *7* (2), 430-440.

(5) Steffen, W.; Richardson, K.; Rockstrom, J.; Cornell, S. E.; Fetzer, I.; Bennett, E. M.; Biggs, R.; Carpenter, S. R.; de Vries, W.; de Wit, C. A.; Folke, C.; Gerten, D.; Heinke, J.; Mace, G. M.; Persson, L. M.; Ramanathan, V.; Reyers, B.; Sorlin, S. Planetary boundaries: Guiding human development on a changing planet. *Science* **2015,** *347* (6223), 1259855-1259855.

(6) Sobota, D. J.; Compton, J. E.; McCrackin, M. L.; Singh, S.; Stockholms, U.; Naturvetenskapliga, F.; Stockholms, U. Ö. Cost of reactive nitrogen release from human activities to the environment in the United States. *Environ. Res. Lett.* **2015,** *10* (2), 25006.

(7) Dodds, W. K.; Smith, V. H. Nitrogen, phosphorus, and eutrophication in streams. *Inland Waters* **2016,** *6* (2), 155-164.

(8) Kaushal, S. S. Increased Salinization Decreases Safe Drinking Water. *Environ. Sci. Technol.* **2016,** *50* (6), 2765-2766.

(9) Sustainable Development Goals-Goal 6: Ensure access to water and sanitation for all. https://www.un.org/sustainabledevelopment/water-and-sanitation/ (accessed May 31, 2021).

(10) Gu, B.; Chen, D.; Yang, Y.; Vitousek, P.; Zhu, Y. Soil-Food-Environment-Health Nexus for Sustainable Development. *Research* **2021,** *2021*, 1-4.

(11) ABS *Australia's Environment: Issues and Trends, 2006*; Australia Bureau of Statistics: Canberra, Australia, 2006.

(12) Davis, J. R.; Koop, K. Eutrophication in Australian Rivers, Reservoirs and Estuaries – A Southern Hemisphere Perspective on the Science and its Implications. *Hydrobiologia* **2006,** *559* (1), 23-76.

(13) Australian Government-Department of Agriculture, Water and the Environment. https://www.environment.gov.au (accessed June 18, 2020).

(14) Convention Concerning the Protection of the World Cultural and Natural Heritage-Great Barrier Reef. https://whc.unesco.org/en/list/154/ (accessed June 1, 2021).

(15) Rice, J.; Westerhoff, P. High levels of endocrine pollutants in US streams during low flow due to insufficient wastewater dilution. *Nat. Geosci.* **2017,** *10* (8), 587-591.

(16) Farhadian, M.; Haddad, O. B.; Seifollahi-Aghmiuni, S.; Loáiciga, H. A. Assimilative Capacity and Flow Dilution for Water Quality Protection in Rivers. *Journal of Hazardous, Toxic, and Radioactive Waste* **2015,** *19* (2), 04014027.

(17) Ballard, T. C.; Sinha, E.; Michalak, A. M. Long-Term Changes in Precipitation and Temperature Have Already Impacted Nitrogen Loading. *Environ. Sci. Technol.* **2019,** *53* (9), 5080-5090.

(18) Donner, S. D. The influence of climate on in-stream removal of nitrogen. *Geophys. Res. Lett.* **2004,** *31* (20).

(19) IFASTAT Database. https://www.ifastat.org (accessed October 18, 2019).

(20) Cattle numbers – as at June 2018, MLA market information: MEAT & LIVESTOCK AUSTRALIA. https://www.mla.com.au/globalassets/mla-corporate/prices--markets/documents/trends--analysis/fast-facts--maps/cattle-numbers-map-2019-june-2018-1.pdf (accessed March 6, 2019).

(21) Sheep numbers – as at June 2018, MLA market information: MEAT & LIVESTOCK AUSTRALIA. https://www.mla.com.au/globalassets/mla-corporate/prices--markets/documents/trends--analysis/fast-facts--maps/sheep-numbers-map-2019-june-2018-1.pdf (accessed March 6, 2019).

(22) Thorburn, P. J.; Biggs, J. S.; Attard, S. J.; Kemei, J. Environmental impacts of irrigated sugarcane production: Nitrogen lost through runoff and leaching. *Agriculture, Ecosystems & Environment* **2011,** *144* (1), 1-12.

(23) Webster, A. J.; Bartley, R.; Armour, J. D.; Brodie, J. E.; Thorburn, P. J. Reducing dissolved inorganic nitrogen in surface runoff water from sugarcane production systems. *Mar. Pollut. Bull.* **2012,** *65* (4-9), 128-135.

(24) L. Burkitt, L. A review of nitrogen losses due to leaching and surface runoff under intensive pasture management in Australia. *Soil Res.* **2014,** *52*, 621-636.

(25) Gu, B.; Ge, Y.; Ren, Y.; Xu, B.; Luo, W.; Jiang, H.; Gu, B.; Chang, J. Atmospheric Reactive Nitrogen in China: Sources, Recent Trends, and Damage Costs. *Environ. Sci. Technol.* **2012,** *46* (17), 9420-9427.

(26) FAOSTAT Database. http://faostat.fao.org/home/E (accessed September 15, 2019).

(27) ABS statistics. https://www.abs.gov.au/browse?opendocument&ref=topBar (accessed October 18, 2018).

(28) Shibata, H.; Galloway, J. N.; Leach, A. M.; Cattaneo, L. R.; Cattell Noll, L.; Erisman; Gu, B.; Liang, X.; Hayashi, K.; Ma, L.; Dalgaard, T.; Graversgaard, M.; Chen, D.; Nansai, K.; Shindo, J.; Matsubae, K.; Oita, A.; Su, M.; Mishima, S.; Bleeker, A. Nitrogen footprints: Regional realities and options to reduce nitrogen loss to the environment. *AMBIO* **2017,** *46* (2), 129-142.

(29) Australian Government-Department of Agriculture and Water Resources. http://www.agriculture.gov.au (accessed January 18, 2020).

(30) Victoria-Water Monitoring. https://data.water.vic.gov.au/ (accessed September 15, 2020).

(31) Western Australia-Monitoring, evaluation and reporting - Catchment water quality. https://www.dpaw.wa.gov.au/management/swan-canning-riverpark/rivers-and-estuaries-science/monitoring-evaluation-and-reporting?showall=&start=2 (accessed September 15, 2020).

(32) Environment Protection Authority-New South Wales. https://www.epa.nsw.gov.au/ (accessed September 15, 2020).

(33) EPA Water Quality Data in South Australia: Creeks and Rivers. https://www.google.com/maps/d/viewer?mid=1dc7UEN99xcZNkbtZ-aVxL6Va3_I&ll=-32.75443183803141%2C138.24301149999997&z=5 (accessed September 15, 2020) (accessed September 15, 2020).

(34) Queensland-Water quality data and assessments. https://environment.des.qld.gov.au/management/water/quality-data-assessments (accessed September 15, 2020).

(35) Tasmania-State of Rivers Reports. https://dpipwe.tas.gov.au/water/water-monitoring-and-assessment/water-monitoring/surface-water-quality/state-of-rivers-reports (accessed September 15, 2020).

(36) Environment Protection Authority-Northern Territory. https://ntepa.nt.gov.au/ (accessed September 15, 2020).

(37) Australian and New Zealand Guidelines for Fresh and Marine Water Quality-Volume 1. https://www.epa.sa.gov.au/files/13808_anzecc_water_guidelines.pdf (accessed September 15, 2020).

(38) Australian Statistical Geography Standard (ASGS): Volume 3. https://www.abs.gov.au/AUSSTATS/abs@.nsf/Lookup/1270.0.55.003Explanatory%20Notes10July%202011?OpenDocument (accessed October 20, 2019).

(39) Gilbert, M.; Brodie, J. *Population and Major Land Use in the Great Barrier Reef Catchment Area: Spatial and Temporal Trends*; Great Barrier Reef Marine Park Authority: Townsville, Australia, 2001.

(40) Bartley, R.; Waters, D.; Turner, R.; Kroon, F.; Wilkinson, S.; Garzon-Garcia, A.; Kuhnert, P.; Lewis, S.; Smith, R.; Bainbridge, Z.; Olley, J.; Brooks, A.; Burton, J.; Brodie, J.; Waterhouse, J. *Scientific Consensus Statement 2017: A synthesis of the science of land-based water quality impacts on the Great Barrier Reef, Chapter 2: Sources of sediment, nutrients, pesticides and other pollutants to the Great Barrier Reef*; State of Queensland: Brisbane, Australia, 2017; pp 1-100.

(41) Wang, N.; Yao, Z.; Liu, W.; Lv, X.; Ma, M. Spatial Variabilities of Runoff Erosion and Different Underlying Surfaces in the Xihe River Basin. *Water-Sui.* **2019,** *11* (2), 352.

(42) Howarth, R.; Swaney, D.; Billen, G.; Garnier, J.; Hong, B.; Humborg, C.; Johnes, P.; Mörth, C.; Marino, R. Nitrogen fluxes from the landscape are controlled by net anthropogenic nitrogen inputs and by climate. *Front. Ecol. Environ.* **2012,** *10* (1), 37-43.

(43) 2012 SPARROW Models for the Midwest: Total Phosphorus, Total Nitrogen, Suspended Sediment, and Streamflow. https://sparrow.wim.usgs.gov/sparrow-midwest-2012/ (accessed July 18, 2020).

(44) McIsaac, G. F.; David, M. B.; Gertner, G. Z.; Goolsby, D. A. Nitrate flux in the Mississippi River. *Nature* **2001,** *414* (6860), 166-167.

(45) Wang, X.; Hao, F.; Cheng, H.; Yang, S.; Zhang, X.; Bu, Q. Estimating non-point source pollutant loads for the large-scale basin of the Yangtze River in China. *Environ. Earth Sci.* **2011,** *63* (5), 1079-1092.

(46) Wang, A.; Tang, L.; Yang, D.; Lei, H. Spatio-temporal variation of net anthropogenic nitrogen inputs in the upper Yangtze River basin from 1990 to 2012. *Science China Earth Sciences* **2016,** *59* (11), 2189-2201.

(47) Murray-Darling Basin Authority. https://www.mdba.gov.au/ (accessed June 10, 2020).

(48) Chen, J.; Gao, X.; He, D.; Xia, X. Nitrogen contamination in the Yangtze River system, China. *J. Hazard. Mater.* **2000,** *73* (2).

(49) International Institute for Applied Systems Analysis - SSP Scenario Database. https://iiasa.ac.at/web/home/research/researchPrograms/Energy/SSP_Scenario_Database.html (accessed October 1, 2020).

(50) Hall, M. *The Cost of Pollution: Supporting Cost-Effective Options Evaluation and Pollution Reduction*; The Urban Water Security Research Alliance: Brisbane, Australia, 2012.

(51) Smart, J.; Hasan, S.; Volders, A.; Curwen, G.; Fleming, C.; Burford, M. *A tradable permit scheme for cost-effective reduction of nitrogen runoff in the sugarcane catchments of the Great Barrier Reef*; Reef and Rainforest Research Centre: 2016.

(52) Bureau of Meteorology. http://www.bom.gov.au/ (accessed June 10, 2020).

(53) Baron, J. S.; Hall, E. K.; Nolan, B. T.; Finlay, J. C.; Bernhardt, E. S.; Harrison, J. A.; Chan, F.; Boyer, E. W. The interactive effects of excess reactive nitrogen and climate change on aquatic ecosystems and water resources of the United States. *Biogeochemistry* **2013,** *114* (1), 71-92.

(54) Australian Government-Department of Agriculture, Water and the Environment: Western Australia – Wheat Belt. https://www.agriculture.gov.au/abares/research-topics/aboutmyregion/wa-wheat-belt#regional-overview (accessed July 18, 2020).

(55) Davey, A.; Fisher, R. *Live Sheep Export Trade: Review of the Draft Regulation Impact Statement*; Animals Australia: Melbourne, Australia, 2020.

(56) World Bank Open Data. https://data.worldbank.org/ (accessed January 10, 2020).

(57) Australian Government-Department of Agriculture, Water and the Environment: Snapshot of Australian Agriculture 2020. https://www.agriculture.gov.au/abares/publications/insights/snapshot-of-australian-agriculture-2020#agricultural-production-is-growing (accessed June 18, 2020).

(58) Australian Government-Department of Agriculture and Water Resources: Council of Australian Governments water reform. https://www.agriculture.gov.au/water/policy/coag (accessed June 18, 2020).

(59) Yapp, G.; Munro, R.; Barson, M.; Chartres, C.; Hill, M.; Bernard, P. *Environmental Factors Affecting Australia's Livestock Industries*; Meat and Livestock Australia: Sydney, Australia, 2001; pp 1-122.

(60) Potter, N. J.; Chiew, F. H. S.; Frost, A. J.; Srikanthan, R.; McMahon, T. A.; Peel, M. C.; Austin, J. M. *Characterisation of Recent Rainfall and Runoff in the Murray-Darling Basin. A report to the Australian Government from the CSIRO Murray-Darling Basin Sustainable Yields Project*; CSIRO: Canberra, Australia, 2008.

(61) Swaney, D. P.; Hong, B.; Ti, C.; Howarth, R. W.; Humborg, C. Net anthropogenic nitrogen inputs to watersheds and riverine N export to coastal waters: a brief overview. *Curr. Opin. Env. Sust.* **2012,** *4* (2), 203-211.

(62) Argent, R. M. *Australia state of the environment 2016: inland water, independent report to the Australian Government Minister for the Environment and Energy*; Australian Government-Department of the Environment and Energy: Canberra, Australia, 2017.

(63) Australian Government-Great Barrier Reef Marine Park Authority. http://www.gbrmpa.gov.au (accessed July 10, 2020).

(64) Waterhouse, J.; Brodie, J.; Lewis, S.; Mitchell, A. Quantifying the sources of pollutants in the Great Barrier Reef catchments and the relative risk to reef ecosystems. *Mar. Pollut. Bull.* **2012,** *65* (4-9), 394-406.

(65) ABS *Year Book Australia, 2006*; Australia Bureau of Statistics: Canberra, Australia, 2006; pp 1-776.

(66) Kroon, F. J.; Thorburn, P.; Schaffelke, B.; Whitten, S. Towards protecting the Great Barrier Reef from land-based pollution. *Global Change Biol.* **2016,** *22* (6), 1985-2002.

(67) The State of Queensland and Commonwealth of Australia *Reef Water Quality Protection Plan: for catchments adjacent to the Great Barrier Reef World Heritage Area*; Queensland Department of Premier and Cabinet: Brisbane, Australia, 2003.

(68) Reef 2050 Water Quality Improvement Plan. https://www.reefplan.qld.gov.au/ (accessed June 10, 2020).

(69) Thorburn, P. J.; Biggs, J. S.; Attard, S. J.; Kemei, J. Environmental impacts of irrigated sugarcane production: Nitrogen lost through runoff and leaching. *Agriculture, Ecosystems & Environment* **2011,** *144* (1), 1-12.

(70) Riahi, K.; van Vuuren, D. P.; Kriegler, E.; Edmonds, J.; O Neill, B. C.; Fujimori, S.; Bauer, N.; Calvin, K.; Dellink, R.; Fricko, O.; Lutz, W.; Popp, A.; Cuaresma, J. C.; KC, S.; Leimbach, M.; Jiang, L.; Kram, T.; Rao, S.; Emmerling, J.; Ebi, K.; Hasegawa, T.; Havlik, P.; Humpenöder, F.; Da Silva, L. A.; Smith, S.; Stehfest, E.; Bosetti, V.; Eom, J.; Gernaat, D.; Masui, T.; Rogelj, J.; Strefler, J.; Drouet, L.; Krey, V.; Luderer, G.; Harmsen,

M.; Takahashi, K.; Baumstark, L.; Doelman, J. C.; Kainuma, M.; Klimont, Z.; Marangoni, G.; Lotze-Campen, H.; Obersteiner, M.; Tabeau, A.; Tavoni, M. The Shared Socioeconomic Pathways and their energy, land use, and greenhouse gas emissions implications: An overview. *Global Environmental Change* **2017,** *42*, 153-168.

(71) Li, P.; Lu, J.; Hou, W.; Pan, Y.; Wang, Y.; Khan, M. R.; Ren, T.; Cong, R.; Li, X. Reducing nitrogen losses through ammonia volatilization and surface runoff to improve apparent nitrogen recovery of double cropping of late rice using controlled release urea. *Environmental science and pollution research international* **2017,** *24* (12), 11722-11733.

(72) Erickson, G.; Klopfenstein, T. Nutritional and management methods to decrease nitrogen losses from beef feedlots. *J. Anim. Sci.* **2010,** *88* (suppl_13), E172-E180.

(73) Gu, B.; van Grinsven, H. J. M.; Lam, S. K.; Oenema, O.; Sutton, M. A.; Mosier, A.; Chen, D. A Credit System to Solve Agricultural Nitrogen Pollution. *The Innovation* **2021,** *2* (1), 1-2.

(74) Gu, B.; Zhang, L.; Van Dingenen, R.; Vieno, M.; Van Grinsven, H. J.; Zhang, X.; Zhang, S.; Chen, Y.; Wang, S.; Ren, C.; Rao, S.; Holland, M.; Winiwarter, W.; Chen, D.; Xu, J.; Sutton, M. A. Abating ammonia is more cost-effective than nitrogen oxides for mitigating PM2.5 air pollution. *Science* **2021,** *374* (6568), 758-762.