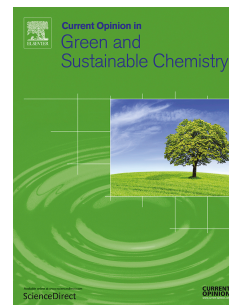


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Anthropogenic-driven perturbations on nitrogen cycles and interactions with climate changes

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Abstract

Anthropogenic activities have substantially perturbed the global nitrogen (N) cycle directly through enhancing reactive N (Nr) inputs and indirectly through climate and land-use change. However, the climatic impacts of the N cycle and its feedbacks on climate change remain very uncertain. In this review, we provide an overview of the dominant pathways by which anthropogenic Nr affects the climate system and summarize the available scientific assessments. We also review the latest progress on the responses of N cycle to changing climate to understand the potential for feedbacks between the N cycle and climate. With the urgent need to reduce Nr in the future to alleviate its negative environmental impacts, e.g. air pollution and eutrophication, we highlight the importance for bridging disciplines of atmospheric chemistry, ecology and climatology to improve the scientific understanding and develop co-benefits for both environmental protection and climate change mitigation.

Keywords: Anthropogenic nitrogen, Biogeochemistry-Climate interactions; Climate effects of anthropogenic nitrogen; Terrestrial carbon-nitrogen interactions; Atmospheric nitrogen chemistry; Ammonium and nitrate aerosols; Nitrogen cycles under climate changes

1. Introduction

Nitrogen (N) constitutes nearly 78% of the atmosphere, however nearly 99% of atmospheric N is gas-phase molecular N (N₂) (Galloway et al., 2003). Two processes in the natural environment, i.e. biological nitrogen fixation (BNF) and lightning, can break the triple bond of N₂ and thereby create reactive nitrogen (Nr). As one of the most important nutrients for living organisms supporting biosynthesis, growth, maintenance and propagation, Nr can be subjected to a vast number of chemical and biochemical reactions and functions through its importance in enzyme synthesis and biomolecules (Duce et al., 2008; Zaehle and Dalmonech, 2011). However, Nr entering marine and terrestrial ecosystems may also be exported out of the biosphere and emitted to the atmosphere, where it interferes with the radiative transfer in the atmosphere through tropospheric chemical reactions of greenhouse gases and aerosols. (Derwent et al., 2008; Shindell et al., 2009). As a result, anthropogenic Nr plays important roles in the global biogeochemical cycles (of carbon, nitrogen, phosphorus, etc.), further influencing environmental health (de Vries, 2021; Nieder and Benbi, 2022), ecosystem biodiversity (Bobbink et al., 2010), and climate changes (Erisman et al., 2011).

Fossil fuel combustion and N fixation technology (e.g. the Haber-Bosch reaction) have introduced large quantities of anthropogenic Nr to the Earth system since the beginning of the industrial revolution (around 1750). The scale of this perturbation has grown exponentially since the beginning of the industrial revolution and in current decades is estimated to be on par with or even exceed natural Nr generation through BNF and lightning (Fowler et al., 2013). Excessive Nr, on the one hand, substantially affects climate. For

instance, increased nitrous oxide (N_2O) concentrations and ammonium (NH_4^+) / nitrate (NO_3^-) aerosol loadings in the atmosphere could warm and cool the atmosphere through the greenhouse effect and solar-radiation diffusion, respectively (Thornhill et al., 2021b). The enhanced Nr inputs into ecosystems through N deposition, fertilizer and manure application may alleviate global warming by increasing ecosystem carbon sequestration (Lessmann et al., 2022; Schulte-Uebbing et al., 2022). Furthermore, short-lived nitrogen oxides (NO_x) play vital roles in atmospheric chemistry and non-linearly alter atmospheric greenhouse gases of ozone (O_3) (Lu et al., 2021a) and methane (CH_4) (Peng et al., 2022). On the other hand, climate change induced by anthropogenic greenhouse gases also substantially affects almost all of the natural N processes (Fowler et al., 2015; Greaver et al., 2016), with the possibility of causing feedbacks to climate. The net climate effects of anthropogenic Nr as well as the N climate feedbacks, although understood in principle, are challenging to quantify and therefore remain purely constrained.

The importance of interactions between N dynamics and climate has gained recognition in past decades (Altieri et al., 2021; Erisman et al., 2011; Fowler et al., 2015). Substantial efforts have been dedicated in the past years to improve the scientific understanding through field experiments, meta-data analysis, machine learning techniques and model simulations. In this review study, we aim to give an overview on previous studies, most of which have predominantly focused on specific N compounds or processes within the global N cycles, and integrate the latest key findings with respect to interactions of N cycles and climate change. As illustrated in Fig. 1, we will firstly overview the pathways that N cycles influence climate and identify the contributions led by direct anthropogenic Nr inputs (Sect. 2), then we will briefly demonstrate the recent discoveries of key N-cycle processes responses to changing climate as well as the anthropogenic perturbations (Sect. 3). Finally, we will conclude and outline future perspectives in Sect. 4.

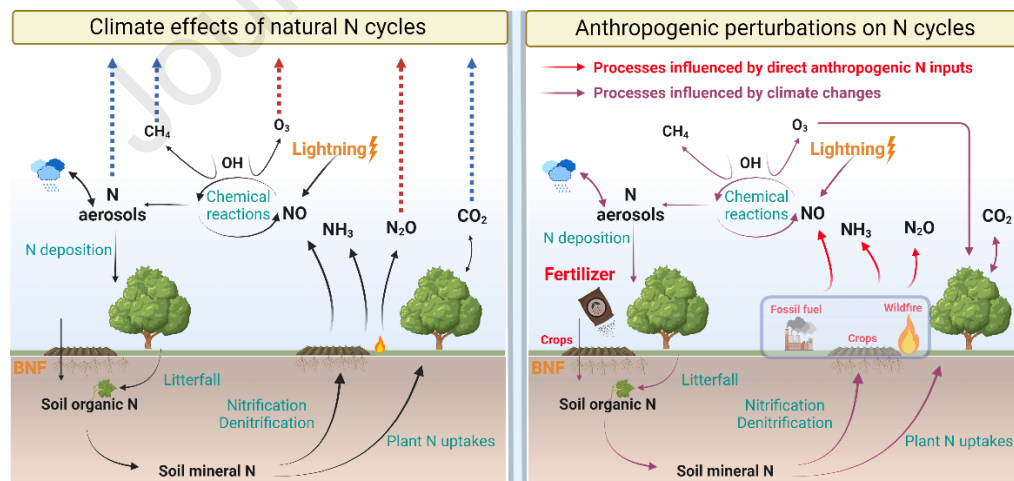


Figure 1. The climate effects of Nr compounds (left) also with anthropogenic-driven perturbations on N cycles (right). For each panel, the left part indicates the Nr inputs to the terrestrial ecosystem, while the right part indicates the Nr fluxes to the atmosphere. The blue or red dashed arrows on the top of the left panel indicate the cooling or warming effects of each component on climate, respectively.

2. Main climate-affecting N compounds

2.1 N₂O

N₂O is a long-lived greenhouse gas with a global warming potential that is about 273 times greater larger than that of CO₂ per unit mass over a period of 100 years. IPCC AR6 reported with high confidence that atmospheric N₂O mole fractions have risen from 270 ppb in 1750 to 331 ppb in 2019 and contributed to $+0.21 \pm 0.03 \text{ W m}^{-2}$ effective radiative forcing relative to the pre-industrial period (Canadell et al., 2021). Increases in agricultural fertilizer/manure application, N deposition and fossil fuel combustions were identified as primary drivers behind this increase of N₂O (Tian et al., 2019), but the exact attribution of anthropogenic contributions to the N₂O increases is still challenging. Anthropogenic sources during 2007-2016 were estimated to be 7.3 [4.2-11.4] Tg N yr⁻¹ among the global total 17.0 Tg N yr⁻¹ emissions (Tian et al., 2020). Limited but also challenging flux measurements (Barton et al., 2015; Liao et al., 2023; Wang et al., 2020), uncertain and dynamic spatiotemporal distributions of N fertilizer application and agricultural land uses (Lu and Tian, 2017), as well as imperfections in model representations (Feng et al., 2023) together are the main cause for this large range.

2.2 N-species aerosols

Aerosols remain in the troposphere for a timescale of days to weeks. Most compounds result in a cooling effect on climate as they scatter solar radiation and interact with cloud formation and lifetime. Ammonium (NH₄⁺) and nitrate (NO₃⁻) are the two dominant N-species aerosols. It is well-known that the major source of ammonia (NH₃), which is the primary precursor to generate NH₄⁺, is from agricultural activities (Hoesly et al., 2018), but significant gaps remain in quantifying global NH₃ emissions. Recent bottom-up estimates assessed global agricultural NH₃ emissions at about 58 Tg N yr⁻¹ in 2010 (Liu et al., 2022a), while the top-down inversion accounting for natural and other anthropogenic NH₃ sources suggested global emissions from 90 to 191 Tg N yr⁻¹ (Evangeliou et al., 2021; Luo et al., 2022). Some studies also showed current emission inventories tended to underestimate NH₃ emissions particularly in hotspot regions, such as Western Europe (Cao et al., 2022) and Eastern China (Kong et al., 2019). With uncertainties in NH₃ emissions, the effective radiative forcing (ERF) of NH₄⁺ in 2014 was estimated to be about -0.07 W m^{-2} based on ensemble means of earth system models (Thornhill et al., 2021b).

Emissions of nitrogen oxide (NO_x), which is the main precursor of NO₃⁻, are dominated by fossil fuel sources (Hoesly et al., 2018). Both bottom-up and top-down estimates showed consistently similar magnitudes and decadal trends in anthropogenic NO_x emissions (Ding et al., 2017; Jena et al., 2015; McDuffie et al., 2020). NO_x engages in numerous chemical reactions and significantly perturbs atmospheric hydroxyl radical (OH) concentrations (see below Sect 2.3). The non-linearity of atmospheric chemistry introduces uncertainties in the global burden, spatial distribution as well as the particle sizes of NO₃⁻, all of which are essential to quantify the NO₃⁻ climate effects. Therefore, estimates of the cooling effects of NO₃⁻ through solar diffusion varied from very small ($\sim -0.025 \text{ W m}^{-2}$) to more substantial effects (-0.14 W m^{-2}) (An et al., 2019; Bian et al., 2017; Hauglustaine et al., 2014; Thornhill et al., 2021b; Zaveri et al., 2021). The interactions between NO₃⁻ aerosols and cloud remained quite uncertain, with radiative forcing assessed by a limited number of studies ranging from -0.05 to -0.22 W m^{-2} (Bellouin et al., 2011; Lu et al., 2021c; Xu and Penner, 2012).

2.3 NO_x indirect effects through atmospheric chemical reactions

As the highly reactive gas, NO_x is involved in various chemical reactions with other greenhouse gases, in particular O₃ and CH₄. It thereby affects climate indirectly by changing the

lifetime and thus atmospheric burden of these gases. In general, increased atmospheric NO_x concentrations will lead to higher OH concentrations, further increasing O_3 concentrations (Lu et al., 2021a), while at the same time shortening CH_4 lifetime and thereby reducing CH_4 concentrations (Peng et al., 2022). A recent assessment based on an ensemble of Earth System models showed that NO_x -induced O_3 enhancement warmed the climate by $+0.2 \pm 0.07 \text{ W m}^{-2}$, while the reduced CH_4 lifetime led by NO_x contributed a cooling effect of about -0.2 W m^{-2} to -0.37 W m^{-2} until 2014 since pre-industrial (Thornhill et al., 2021b). The complexity of OH chemical dynamics and non-linear interactions among NO_x , volatile organic compounds and O_3 is the primary cause for the uncertainty in this quantification.

2.4 C-N interactions in terrestrial ecosystems

The majority of the N element in the terrestrial ecosystem constitutes plants and microbes in organic forms and plays crucial roles in enzyme-mediated processes. However, the available N for plants is normally in the forms of dissolved inorganic N, such as ammonium or nitrate ions, which is mineralized from soil organic N by microbial activities (Li et al., 2019). Meanwhile, soil microbes also regulate BNF levels, thereby influencing the overall N availability for plants (Aasfar et al., 2021). A portion of the mineralized inorganic N will be lost from the terrestrial ecosystem as gases phase through nitrification and denitrification (Feng et al., 2023), or as dissolved forms through leaching (Wang and Li, 2019).

Next to phosphorus, N in terrestrial ecosystems is one of the most important nutrients for plants and microbial organisms. It is essential for C assimilation, growth and maintenance and thereby tightly connected to the C cycle. The N limitation of biomass production is generally believed to be strong in natural temperate and boreal forests (Du et al., 2020), attenuating the expected increases in carbon storage due to CO_2 fertilization of plant photosynthesis under future climate change (Terrer et al., 2019). Anthropogenic-driven increases in N fertilizer application and atmospheric Nr deposition can increase productivity by lifting N limitation. Synthesis of available data from ecosystem manipulation experiments by meta-analysis or machine learning have revealed responses of terrestrial C sinks to N addition, ranging from $1 \text{ kg C (kg N)}^{-1}$ to $25 \text{ kg C (kg N)}^{-1}$ depending on different land cover types, amounts and types of N addition (Liu and Greaver, 2009; Liu et al., 2022b; Poulton et al., 2018; Schulte-Uebbing et al., 2022).

Terrestrial biosphere models (TBMs) integrate process understanding of biogeochemical cycles at large scales and is another useful tool to quantify the climate effects of N-induced increases in C sequestration (Zaehle and Dalmonech, 2011). For instance, Zaehle et al. (2011) attributed a cumulative cooling effect of about -0.1 W m^{-2} to Nr induced carbon storage in the terrestrial biosphere since pre-industrial times by applying a TBM to reproduce global observed trends of atmospheric CO_2 . In recent years, dynamic C-N coupling has been induced in an increasing number of TBMs (Davies-Barnard et al., 2020; Kou-Giesbrecht et al., 2023; Wieder et al., 2015; Zaehle et al., 2014). These models generally agree with earlier findings, but evaluation against independent benchmark highlights remaining uncertainty and a formidable challenge to further constrain model uncertainties to better quantify the N effects on terrestrial carbon sinks and the resulting climate impacts (Kou-Giesbrecht et al., 2023) (Fig. 2).

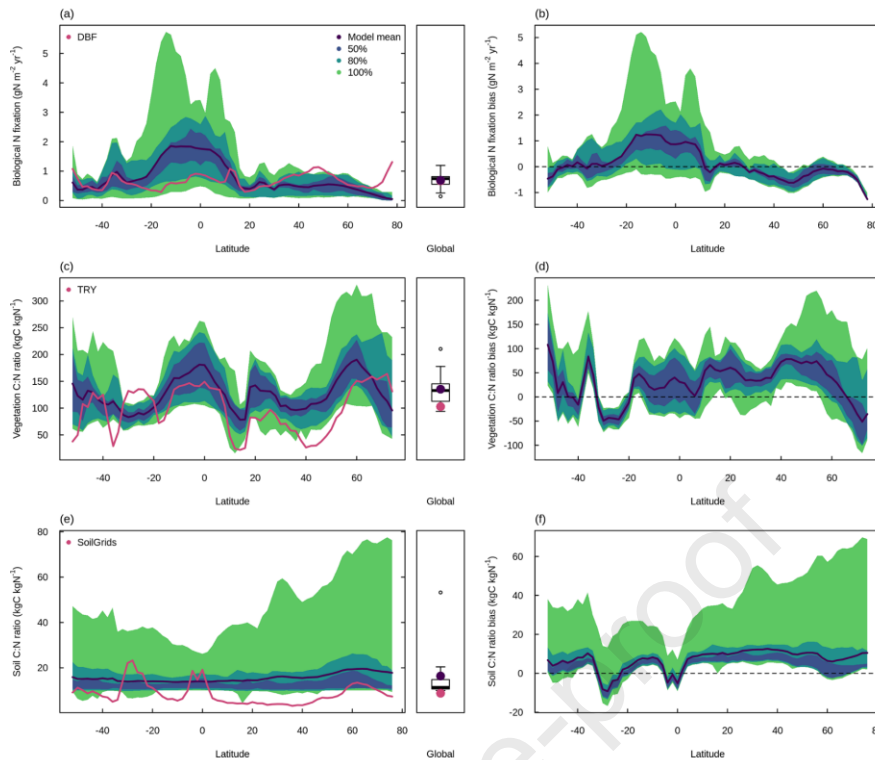


Figure 2. Latitudinal distributions and global means of BNF, vegetation C : N ratio, and soil C : N ratio simulated by the TRENDY-N ensemble (averaged across models over 1980–2021) in comparison to observations. Panels (a, c, e) show the latitudinal distribution of the mean and boxplots show the global mean. Panels (b, d, f) show the latitudinal distribution of the bias. Latitudinal distributions show the mean (black line) and the 50 %, 80 %, and 100% percentiles across models. Boxplots show the median, interquartile range (box), and 80% percentiles (whiskers) across models. Observation-based datasets are from Davies-Barnard and Friedlingstein (2020) for biological N fixation, the TRY plant trait database for vegetation C:N ratio, and SoilGrids for soil C:N ratio. This figure is from Kou-Giesbrecht et al. (2023).

2.5 Other Nr-related climate effects

There are additional, less well-quantified pathways of Nr known to affect climate, which are summarized here: (1) Nr-induced increases in aerosols and O_3 could influence terrestrial carbon sinks by reducing ratios of direct to diffuse photosynthetically active radiation (Mercado et al., 2009; Zhou et al., 2022) and by damaging leaf photosynthesis (Franz et al., 2018; Gong et al., 2021; Sitch et al., 2007), respectively. (2) Nr inputs into terrestrial ecosystems may reduce soil CH_4 uptake (Li et al., 2021b; Liu and Greaver, 2009) and thus increase atmospheric CH_4 . However, soil CH_4 uptake plays only a small role in the global CH_4 cycle and the magnitude of the Nr effects ranges from -60% to +10% (Xia et al., 2020). (3) Riverine and marine N_2O emissions and the C-N interactions might also be influential on climate. Yao et al. (2020) suggested that increased N inputs enhanced riverine emissions of N_2O from 70.4 ± 15.4 Gg N yr^{-1} in 1900 to 291.3 ± 58.6 Gg N yr^{-1} in 2016, while marine N deposition is estimated to contribute to present-day marine N_2O emissions by about 0.01-0.32 Tg N yr^{-1} (Canadell et al., 2021) and marine primary productivity by 0.3 Pg C yr^{-1} increases (Duce et al., 2008), respectively, but with neglectable changes in the resultant air-sea CO_2 fluxes (Yamamoto et al., 2022).

2.6 The net climate effects of anthropogenic Nr

Quantifying the net climate effects of anthropogenic Nr requires crossing different time-scales and disciplines among atmospheric chemistry, climate dynamics and ecological science. To date, this gap among different scientific communities and quantifications is typically bridge in a ‘puzzle-style’ framework, wherein climate effects (e.g. radiative forcing) or key parameters (e.g. terrestrial C responses to N addition; N₂O emission factors) from previous independent studies are combined to generate a ‘best estimate’ of anthropogenic Nr climate effects. Table 1 summarized the four most recent and comprehensive estimates to our knowledge, covering the U.S., Europe, China and globe, respectively. To improve our understanding of the Nr imprint of anthropogenic Nr on climate, it is important to (1) address remaining uncertainties in the underlying regional to global emission data-bases for all Nr species and improve consistency between emission categories; (2) integrate process-understanding to derive non-linear response functions of ecosystems to reduce the dependency of the assessments on linear approximations, (3) and appropriately deal with the evaluation of climate impact by accounting for regional and global short- as well as long-lived climate forcers.

Table 1. Summary of present-day climate effects of anthropogenic Nr. The uncertainty ranges of each estimate were indicated by standard deviation (\pm) or square brackets depending on each individual study.

	Erisman et al. (2011)	Pinder et al. (2012)	Butterbach-Bahl et al. (2011)	Shi et al. (2015)
Region	Global	U.S.	Europe	China
Metrics	Radiative forcing	GWP20 / GWP100	Radiative forcing*	GWP20 / GWP100
Units	W m ⁻²	Tg eCO ₂	W m ⁻²	Tg eCO ₂
N ₂ O	+0.16	+314 [+202, +428] / +291 [+180, +395]	0.017	345±19 / 311±17
Nr-related aerosols		-36.67 [-8.18, -103.5] / 0	-0.0165	-156±45 / -0.12±0.13
NO _x effects on CH ₄ lifetime	-0.38	-271 [-177.4, -385] / -7.19 [0, -15.64]	-0.0046	-349.8±155.6 / -13.2±7.6
NO _x effects on O ₃	+0.13		0.0029	
N-induced terrestrial carbon sinks	-0.2	-206.8 [-134.65, -292.4] / -155.02 [-98.96, -236.76]	-0.019 (excluding agricultural fertilization)	-49.9±54.7 / -86.2±39.7
Other effects	+0.05	[+66 +140]	0.0453	111±100 / 111±100
Net climate effects	-0.24 [-0.5 +0.2]	-200.47 [-299.6, +17.6] / +128.79 [+112.2, +317.1]	-0.0173	-100±414 / 322±163

*The values indicated the global radiative forcing induced by emissions in Europe.

3. Climate feedbacks due to perturbed N cycle

Besides direct anthropogenic Nr inputs, anthropogenic-driven climate change substantially alters the global N cycle, which in turn results in feedback effects on climate. Here, we provide a brief overview of key findings in the past years covering changes in ecosystem N cycles and atmospheric Nr-related chemistry and extrapolate the potential climate feedbacks.

3.1 Responses of ecosystem N cycling to climate changes

A number of processes in the terrestrial N cycle are substantially influenced by climate changes through the responses of enzyme activities, microbial species and strategy (Mattoo and Suman, 2023). Firstly, the biological nitrogen fixation, the major natural source of Nr, is responding to temperature with new emerging evidence to better quantify the optimal temperature for BNF and its thermal acclimation to growth temperature (Houlton et al., 2008; Bytnerowicz et al., 2022). Secondly, increased temperature accelerates the decay of organic material, which enhances inorganic N cycling in the soil and thereby enhance terrestrial carbon storage (Kou-Giesbrecht and Arora, 2023; Melillo et al., 2011). The intensification of inorganic N cycles can not only lead to a positive N₂O-warming feedback (Stocker et al., 2013), but also intensify the soil emissions of NH₃ (Shen et al., 2020) and NO_x (Romer et al., 2018). These processes may be particularly relevant in permafrost regions and peatlands, which are even more sensitive to global warming due to arctic amplification. Recent studies have demonstrated that the increases in mineralized N with permafrost thawing failed to enhance the N availability of plants due to increased N demands and higher N-gas loss (Kou et al., 2020; Lacroix et al., 2022; Ramm et al., 2022). Thirdly, the simultaneous changes in multiple environmental factors, e.g. elevated CO₂ concentrations, warming and changes in soil moisture, may enhance the strength of N limitation on terrestrial C uptake (Mason et al., 2022; Tu et al., 2022). Last but not least, land use changes and increased ecosystem disturbance (e.g. wildfire) may also deeply alter the whole ecosystem N cycle (Dove et al., 2020; Li et al., 2021a; Perez-Quezada et al., 2022). For example, the expansion of agricultural land may lead to higher N fertilizer application (Tian et al., 2022), while strategically intercropping of N-fixing crops (e.g. grain legumes) and cereals may alleviate such issue by improving N use efficiency (Jensen et al., 2020). Meanwhile, more frequent wildfire under the changing climate accelerates the turnover rates of terrestrial N into the atmosphere, generating varied Nr-related gases and particles and substantially feeding back to the climate (Fu et al., 2018; Ward et al., 2012). Overall, although specific components of the Nr related feedbacks have been studied, a comprehensive and quantitative overview of the likely effects on future climate-biogeochemical feedbacks, such as N availability for the carbon cycle, is still lacking (Canadell et al., 2021).

3.2 Responses of atmospheric Nr-related chemistry to climate changes

Higher temperatures in general accelerate reaction rates in atmospheric chemistry, but can also exert significant non-linear influences on individual reactions. Clear evidence supports the hypothesis that higher temperature could accelerate evaporation of ammonium nitrate and thereby reduce Nr-related aerosol loadings (Megaritis et al., 2013). Current earth system models in general reported positive sensitivities of lightning NO_x emissions to the warmer climate (Thornhill et al., 2021a), while insignificant or even negative sensitivities in other studies (Finney et al., 2018; Finney et al., 2016). The NO_x effects on CH₄ and O₃ with warming is strongly dependent on changes in global OH concentrations and distributions and therefore subject to significant uncertainties (Murray et al., 2021; Zhao et al., 2023).

4. Concluding remarks

In this review, we firstly summarized the known pathways that Nr impacts climate. Recent findings underscored the vital roles of the N cycles in affecting global climate, however, each

pathway was still associated with uncertainty. In particular, two major challenges of (1) improving estimates in global soil emissions of Nr gases (N_2O , NH_3 and NO_x), and (2) reducing uncertainties in atmospheric OH chemistry and associated NO_x effects on aerosols, O_3 and CH_4 substantially impede a more comprehensive understanding of the climatic effects of anthropogenic Nr. To fill this knowledge gap, on the one hand, long-term continuous observations of soil Nr-gas fluxes, especially for short-lived NH_3 and NO_x , and improved understandings in microbial dynamics with soil ammonification, nitrification and denitrification (e.g. Li et al., 2020) are both essential. On the other hand, new chemical mechanisms revealed from smog chambers (e.g. Chen et al., 2022), field campaigns (e.g. Cho et al., 2023) and the constraints by satellite retrievals (e.g. Pimlott et al., 2022; Zhang et al., 2018) could promote a better description in global OH chemistry. Last but not least, comprehensive Earth system models, which represent process-based terrestrial and marine N biogeochemical cycles, atmospheric chemical reactions, radiative transfer and climate dynamics, would be crucial to comprehensively reveal climate effects of anthropogenic Nr as well as the potential climate feedbacks through N cycles.

We further overviewed the latest progresses on the effects of anthropogenic perturbations on global N cycles. While the direct anthropogenic Nr addition into soil (e.g. via fertilizer application and N deposition) or atmosphere (e.g. via fossil fuel combustion) has been widely studied, the effects of indirect pathways, such as N processes influenced by changing climate, elevated CO_2 , land use changes, intensified wildfire and diverse agricultural management strategies, remained inadequately understood. In-depth manipulating experiments in crucial zones, including tropical rainforest (Lu et al., 2021b), permafrost (Voigt et al., 2020) and agricultural lands (Patil et al., 2010), with as many N-cycle variables are urgently required. On the other hand, some national or regional practices have indicated that improved human management on both forests and croplands holds promise in mitigating ecosystem disturbances (e.g. wildfire) (Tymstra et al., 2020) and increasing N use efficiency (You et al., 2023). Those practices could increase Nr sequestration in the terrestrial plants and soil, thereby alleviating the environmental pollution led by anthropogenic Nr. For instance, the extensively practiced crop rotation, in particular over South Asian, with more N-fixing crops is effective in reducing Nr loss and increasing crop yields with less fertilizer application (Kumar et al., 2020; Venkatesh et al., 2017). Nevertheless, it is still unknown how climate will respond to the efforts of reducing anthropogenic Nr. It is urgently required to bridge knowledge gaps among communities of agricultural nutrition, global biogeochemical science, atmospheric chemistry, and climate dynamics to achieve win-win in both environmental protection and climate change mitigation.

Conflict of interest

The authors declare no conflict of interest.

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Declaration of interests

☒ 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.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: