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Precipitation trend increases the contribution of dry reduced nitrogen deposition

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Given the leveling off in oxidized nitrogen emissions around the world, the atmospheric deposition of reduced nitrogen ($NH_x = NH_3 + NH_4^+$) has become progressively critical, especially dry deposition, which presents great threats to plant growth. A combination of historical deposition data of measured wet NH_x and modeled dry NH_x in China suggests that dry NH_x deposition has been increasing substantially (4.50% yr^{-1}, $p < 0.05$) since 1980. Here, chemical transport model (WRF-EMEP) results indicate that variation in NH_3 emissions is not a dominant factor resulting in the continually increasing trends of dry NH_x deposition, while climate change-induced trends in precipitation patterns with less frequent light rain and more frequent consecutive rain events (with ≥2 consecutive rainy days) contribute to the increase in dry NH_x deposition. This will continue to shift NH_x deposition from wet to dry form at a rate of 0.12 and 0.23% yr^{-1} ($p < 0.05$) for the period of 2030–2100 in China under the RCP4.5 and RCP8.5 scenarios, respectively. Further analysis for North America and Europe demonstrates results similar to China, with a consistent increase in the contribution of dry NH_x deposition driven by changing precipitation patterns from ~30% to ~35%. Our findings, therefore, uncover the change of precipitation patterns has an increasing influence on the shifting of NH_x deposition from wet to dry form in the Northern Hemisphere and highlight the need to shift from total NH_x deposition-based control strategies to more stringent NH_3 emission controls targeting dry NH_x deposition in order to mitigate the potential negative ecological impacts.

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INTRODUCTION

Nitrogen deposition, an important component of the global nitrogen cycle, has increased twofold over the past 100 years globally[1] and by 60% from 1980 to 2010 in China[2], as a result of reactive nitrogen emissions associated with rapid expansion in agriculture and increased fossil fuel combustion[3]. These reactive nitrogen species include reduced ($NH_x = NH_3 + NH_4^+$) and oxidized ($NO_y = NO_x$ ($NO + NO_2$) + its oxidation products) compounds. Given the continual reductions in NO_x emissions, NH_x deposition has become increasingly important, with its contribution currently reaching 60–85% of the total nitrogen deposition in the United States and Europe[4–6]. Enhanced NH_x deposition is favorable for crop production and plant growth in nitrogen-limited regions[7], whereas excess NH_x deposition causes greater damage to nitrogen-sensitive ecosystems, including global biodiversity loss and eutrophication, compared with NO_y deposition[8].

The removal of NH_x from the atmosphere can occur by dry or wet processes[1,9,10]. Wet deposition occurs mainly through scavenging by rainfall, sleet, fog, snow, and hail[2,11], while dry deposition occurs by vertical turbulent transfer to the earth's surface[8,9]. Dry NH_x deposition (often dominated by gaseous NH_3) is a continuous process that drives plant species composition change and reduces species cover and diversity much faster, and inflicts greater threats to plant growth compared to the same unit of wet NH_x deposition[12,13]. Previous studies have characterized the spatiotemporal variation and ecological effects of NH_x deposition; however, the main focus of these studies has been on wet NH_x deposition because it is relatively easy to measure in existing networks[2,11], whereas long-term and large-scale dry NH_x deposition has been less studied due to the technical difficulty in directly measuring and monitoring dry deposition. It has been reported that dry NH_x deposition has been a critical component of total NH_x deposition in recent years[14,15]. Based on a combination of historical measurement-based wet NH_x deposition and model-based dry NH_x deposition data, Yu et al.[16] suggested that there has been a shift in the relative contributions of wet and dry deposition to total NH_x deposition in China from 1980 to 2015; wet deposition previously dominated total NH_x deposition, but the modeled ratio of dry to wet NH_x deposition ($R_{NHx(dry/wet)}$) gradually rose from 1980 to 2015, to the point where wet and dry deposition now have an approximately equal contribution to total NH_x deposition (Fig. 1). Thus, there is an urgent need to better understand why dry NH_x deposition and $R_{NHx(dry/wet)}$ have increased in China during the past decades according to the modeled deposition, especially when NH_3 emissions have remained stable since 2005[16].

The major factors that control the variation of NH_x deposition include emissions, meteorology, and land cover[9,17,18]. Chemical transport models (CTMs) have been widely used to investigate the

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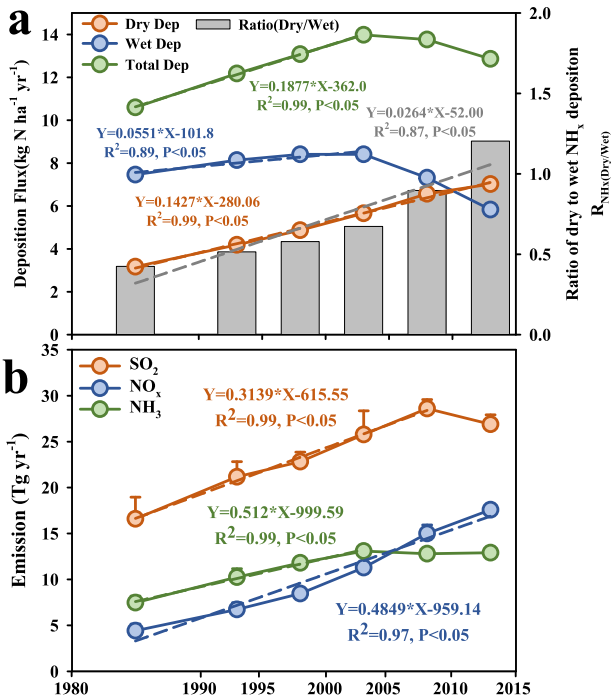

Fig. 1 Temporal trends in NH_x deposition and precursor emissions over China from 1980 to 2015. a Deposition flux (kg N ha^{-1} yr^{-1}) and the ratio of dry to wet NH_x deposition ($R_{NHx(dry/wet)}$). **b** Anthropogenic emissions of SO_2, NO_x, and NH_3 (Tg yr^{-1}) (data obtained from Yu et al.[16]); circles represent the average modeled dry deposition, measured wet deposition, total deposition (dry + wet), and emissions for a given time period; the dashed lines are a linear fit to the points; gray bars are $R_{NHx(dry/wet)}$ and are plotted on the right-hand axis; error bars indicate standard deviation (s.d.).

relative impact of these factors on nitrogen deposition. The results suggest that the long-term trends of NH_x deposition depend to a large extent on emissions and meteorology on regional and global scales, while land use and land cover are important for the spatial distribution of NH_x deposition[19–22]. Specifically, variations in meteorological variables (e.g., precipitation, surface air temperature, wind speed, solar radiation, and atmospheric stability) driven by global climate change are closely related to the partitioning of dry and wet NH_x deposition, although they play only a minor role (less than 20%) in controlling the long-term trends of total nitrogen deposition[19–22]. Precipitation, in particular, controls the partitioning of dry and wet NH_x deposition due to its role in the wet scavenging process. Regardless of the importance of precipitation amount in the partitioning of dry and wet NH_x deposition[18,23], regional and global annual precipitation amounts have not shown a statistically significant trend in the past decades[24] and accordingly cannot explain the decadal-scale variation of the modeled partitioning of dry and wet NH_x deposition. Precipitation patterns (e.g., occurrence, intensity, and duration), however, have changed significantly around the world. Specifically, extreme and consecutive rain events have intensified in the past 100 years[25,26] and are projected to intensify further in the future around the world[27,28]. The change in the precipitation patterns can be mainly attributed to the increased moisture in the atmosphere due to global warming and persistent low pressure due to the slowing of atmospheric circulation[29,30]. Nevertheless, the influence of changing precipitation patterns on the partitioning of dry and wet NH_x deposition remains poorly understood[19,20,23,31–33].

In this work, we address the following questions: how do the variations in climate change-induced precipitation patterns contribute to the continual increase in dry NH_x deposition and $R_{NHx(dry/wet)}$ in China[15] ? Is the importance of climate change-induced precipitation patterns for the partitioning of dry and wet NH_x deposition specific to China, or is it a ubiquitous phenomenon that also occurs in other regions worldwide? We first establish a linear emission-deposition response relationship based on CTM (WRF-EMEP) simulations to estimate the contribution of precursor emission variation to the change in dry NH_x deposition and $R_{NHx(dry/wet)}$. The change in dry NH_x deposition and $R_{NHx(dry/wet)}$, which cannot be explained by precursor emissions, suggests the effect of the interdecadal variability of precipitation patterns driven by climate change. We further explore the potential impact of future climate change on the partitioning of dry and wet NH_x deposition by relying on prediction equations established through historical data and predicted data under two future climate change scenarios, representative concentration pathway (RCP) 4.5 and 8.5, for the period of 2030–2100. Finally, we broaden our analysis to include the United States (USA) and Europe, the other two hotspots of NH_x deposition, to assess the universality of the increasing importance of dry NH_x deposition. The purpose of this study is to advance our understanding of the partitioning of dry and wet NH_x deposition driven by global climate change to date and in the future, which will have important implications for assessing ecological impacts and formulating policy to reduce NH_3 emissions in the face of climate change.

RESULTS

Historical perspective of NH_x deposition

Historical NH_x deposition data based on a combination of measured wet NH_x deposition and modeled dry NH_x deposition[15] (see "Methods" section for detailed information regarding data sources and methodology) for the period of 1980–2015 indicate that the total NH_x deposition in China increased considerably at the rate of 0.188 kg N ha^{-1} yr^{-2} (1.77% yr^{-1}, $p < 0.05$) before 2005 and showed a slight decreasing trend (0.112 kg N ha^{-1} yr^{-2}, 0.80% yr^{-1}) after 2005 (Fig. 1). On average, the total NH_x deposition was estimated at 12.7 ± 1.1 kg N ha^{-1} yr^{-1} in China from 1980 to 2015. In general, the variation in total NH_x deposition was consistent and in line with the trends of NH_3 emissions, with a significant positive correlation observed between them ($r = 0.95$, $p < 0.05$).

With respect to the forms of NH_x deposition, variation in wet NH_x deposition was consistent with total NH_x deposition, but the growth rate for wet NH_x deposition (0.055 kg N ha^{-1} yr^{-2}, 0.74% yr^{-1}) was lower than that of total NH_x deposition before 2005, while the corresponding rate of decrease (0.258 kg N ha^{-1} yr^{-2}, 3.07% yr^{-1}) was relatively higher after 2005. In contrast, modeled dry NH_x deposition was on the rise during the study period, at the rate of 0.143 kg N ha^{-1} yr^{-2} (4.50% yr^{-1}, $p < 0.05$), leading to an upward trend for modeled $R_{NHx(dry/wet)}$ (ratio of dry to wet NH_x deposition) at the rate of 0.026 per year (6.21% yr^{-1}, $p < 0.05$). These results indicate a shift in the relative contributions of wet and dry deposition to total NH_x deposition, with modeled $R_{NHx(dry/wet)}$ increasing from 0.42 (1980s) to 1.20 (2010s), implying that China has gone from having wet deposition as the dominant contributor to total NH_x deposition to having approximately equal dry and wet NH_x deposition for the last two decades.

Variation in deposition velocity is not responsible for the partitioning of NH_x deposition

The dry deposition flux is calculated as the product of ambient concentration and vertical deposition velocity (V_d)[5] (see detailed information in Supplementary Note 1); ambient concentration depends heavily on precursor emissions while V_d relies heavily on meteorological conditions, surface characteristics, and the

pollution regime[34–36]. The parameterization of gaseous V_d utilizes a resistance approach analogous to Ohm's law in electrical circuits[34–36] (see detailed information in Supplementary Note 1). The resistance includes aerodynamic resistance (R_a), quasi-laminar sublayer resistance (R_b) and canopy resistance (R_c), among which, R_c is generally the most dynamic and difficult to estimate[37–40]. Furthermore, R_c is dominant in the deposition process as it is typically the largest in magnitude among the three resistances[40]. Non-stomatal resistance (R_{ns}) is one of the most important terms of R_c for gaseous NH_3 V_d since non-stomatal uptake is the dominant pathway for highly water-soluble gaseous NH_3[37,38]. R_{ns}, is mostly influenced by surface temperature, relative humidity, and the presence of other trace substances (i.e., SO_2 and NH_3)[38,39]. According to the work of Simpson et al.[39], R_{ns} was calculated in this study (see Supplementary Eqs. (3 and 4)). Stomatal resistance, R_{ns}, did not change significantly during the study period (Supplementary Fig. 1), although surface temperature (Supplementary Fig. 2) and precursor emissions (Fig. 1b) changed substantially in the past few decades. Further evidence provided by the WRF-EMEP model also revealed that the annual average V_d for gaseous NH_3 and particulate NH_4^+ had no obvious change between 2010 and 2017 (Supplementary Fig. 3, see detailed information in Supplementary Note 5). The results suggest that V_d affected by anthropogenic emissions and meteorology was not responsible for the partitioning of dry and wet NH_x deposition.

Variation in emissions is not a dominant factor affecting the partitioning of NH_x deposition

Among the precursor emissions, NH_3 emissions are the primary driver determining the magnitude and trends of NH_x deposition. On the other hand, emissions of the acidic gases SO_2 and NO_x can affect the equilibrium between gaseous NH_3 and particulate NH_4^+ through the variation of aerosol acidity. The variation in aerosol acidity can, in turn, influence the partitioning of dry and wet NH_x deposition[11,14]. In order to quantify the contribution of variations in precursor emissions to the change in modeled dry NH_x deposition and $R_{NHx(dry/wet)}$, we first establish a linear emission–deposition response relationship based on CTM (WRF-EMEP) simulations (see "Methods" section for detailed information). As a result, we estimated that the changes in precursor emission amounts during 1985–2015, with values of +5.4 (+73% yr^{-1}), +10.3 (+62% yr^{-1}), and +13.2 (298% yr^{-1}) Tg yr^{-1} for NH_3, SO_2, and NO_x, respectively, resulted in changes of +1.00 kg N ha^{-1} yr^{-1} and +0.03 for the variation in modeled dry NH_x deposition (ΔDry) and modeled $R_{NHx(dry/wet)}$ (ΔRatio), respectively. To summarize, changes in precursor emissions explained 26% of the increase in modeled dry NH_x deposition and 4% of the increase in modeled $R_{NHx(dry/wet)}$, with an overall variation of +3.85 kg N ha^{-1} yr^{-1} and +0.78 for ΔDry and ΔRatio, respectively, suggesting that variation in anthropogenic emissions is not a dominant factor affecting the partition of dry and wet NH_x deposition from the perspective of long-term trends. This finding is also confirmed by the spatial analysis of NH_x deposition (Supplementary Fig. 4) and anthropogenic emissions (Supplementary Fig. 5), which corroborate the finding from the temporal analysis that variation in anthropogenic emission is not a dominant factor affecting the partition of dry and wet NH_x deposition (Supplementary Note 3).

Effect of climate change-induced precipitation patterns on the partitioning of NH_x deposition

It is widely accepted that precipitation amount is the most important meteorological factor affecting the magnitude of NH_x deposition flux[18,41]. However, there has been no significant variation in the long-term annual accumulated precipitation amount in China during the past few decades, with an average value of 1009–1047 mm yr^{-1} (Supplementary Fig. 6a). In addition, there has been no statistically significant correlation between precipitation amount and modeled dry NH_x deposition, modeled $R_{NHx(dry/wet)}$, suggesting that interdecadal variability in precipitation amount is not sufficient to explain the upward trends in modeled dry NH_x deposition and $R_{NHx(dry/wet)}$. However, precipitation patterns (e.g., intensity and duration) have changed significantly around the world despite stable precipitation amounts. In order to characterize rain intensity and rain event duration quantitatively, rain intensity was classified as light rain (0.1–10 mm d^{-1}) and moderate rain or above (≥10 mm d^{-1}). Based on rainfall duration, a rain event was classified as a consecutive rain event (with two or more consecutive rainy days) or a single rain event.

As shown in Supplementary Fig. 6b, c, the frequency of light rain decreased significantly while upward trends were observed for consecutive rain events in China during 1985–2015 under the effect of climate change. Moreover, the frequency of light rain was negatively correlated with modeled dry NH_x deposition ($r = -0.73$) and modeled $R_{NHx(dry/wet)}$ ($r = -0.90$), and the occurrence of consecutive rain events was positively correlated with modeled dry NH_x deposition ($r = 0.55$) and modeled $R_{NHx(dry/wet)}$ ($r = 0.70$), indicating that rain intensity and rain event duration may be responsible for the upward trends in modeled dry NH_x deposition and $R_{NHx(dry/wet)}$ in China.

Previous studies have revealed that owing to global climate change, variation in precipitation patterns plays a more important role than the amount of rain in the removal of air pollutants[42,43]. In terms of rain intensity, it has been theoretically and empirically proven that pollutant concentration in rainwater has a negative correlation with rainfall intensity and that light rain is the most effective at removing water-soluble pollutants through below-cloud scavenging processes[44–49]. Wang et al.[50] further demonstrated that aerosol wet scavenging on a global scale is predominantly constrained by light rain and that the magnitude of scavenging varies approximately exponentially with precipitation intensity. To verify the relationship between the wet removal of NH_4^+ and rain intensity, the concentration of NH_4^+ in rainfall and the rain intensity was analyzed with data across China, the USA, and Europe. Results showed an exponential relationship between the two parameters, with r-squared values above 0.80 and p-values less than 0.001 (Supplementary Fig. 7, see Supplementary Note 2.1 for data sources), providing additional evidence that light rain has a stronger washout capacity for NH_4^+ globally. With respect to rain event duration, the washout amount of NH_4^+ from the atmosphere by consecutive rain events is less than that by an equivalent amount of inconsecutive rain events. This is because as a rain event continues, less NH_x content is removed from the atmosphere due to less available NH_3 in the atmosphere. NH_3 is less available because of suppressed NH_3 emissions during the rainfall process and less accumulated NH_3 due to washout in the previous time period[51]. As an example, the dynamics of NH_4^+ concentrations recorded during two consecutive rain events on April 11th, 2009, in Guiyang, China, revealed that NH_4^+ was scavenged quickly at the start of each precipitation event, followed by a relatively low level of scavenging until the end of the event[41] (Supplementary Fig. 8).

In order to characterize the precipitation pattern-induced effect on the below-cloud wet removal of NH_x content with the co-influence of rain intensity and rain event duration, we have defined a new indicator, the precipitation scavenging index (PSI), in this study. The PSI is calculated as follows:

$$\mathrm{PSI} = \frac{\mathrm{LR}}{\mathrm{CR}} \tag{1}$$

Where LR represents the frequency of days with light rain each year, and CR represents the occurrence of consecutive rain events each year. PSI is, therefore, a dimensionless indicator. A lower value of PSI suggests that less frequent but increasingly intense

rainfall events are less efficient at "cleaning" the atmosphere of gaseous NH_3 and particulate NH_4^+. This implies that more NH_x remains in the atmosphere for dry deposition by turbulent diffusion to vegetation and other surfaces. This indicator, despite not having a specific physical meaning, provides a simple way to reflect the negative correlation between LR and dry NH_x deposition and $R_{NHx(dry/wet)}$, as well as the positive correlation between CR and dry NH_x deposition and $R_{NHx(dry/wet)}$, and comprehensively represents the contrasting effect of LR and CR on the below-cloud wet removal of NH_x.

An overall negative trend was found for PSI in China during 1980–2015 (Fig. 2). The negative correlations were found between PSI and modeled dry NH_x deposition ($r = -0.79$) as well as modeled $R_{NHx(dry/wet)}$ ($r = -0.63$), indicating that variation in climate change-induced precipitation patterns favor a shift in NH_x deposition from the wet to dry form. Spatial analysis of NH_x deposition (Supplementary Fig. 4) and precipitation patterns (Supplementary Fig. 9) in China further confirmed that changing precipitation patterns contributed to the increase in dry NH_x deposition in China (Supplementary Note 3). Our finding is supported by studies conducted in the USA, which also revealed that most of the increases in ambient NH_3 concentration are a manifestation of climate change[52,53].

Implications for future partitioning of NH_x deposition

As global climate change is accelerating, partitioning of dry and wet NH_x deposition driven by precipitation pattern changes is expected to continue in the future. The results from the two future global climate scenarios (RCP4.5 and RCP8.5) show that less frequent light rain and more consecutive rain events are expected for the period of 2030–2100 (Supplementary Fig. 6b, c). Therefore, the PSI will continue to decline in the future (Fig. 2), implying that more NH_x remains in the atmosphere and can subsequently be removed from the atmosphere via dry deposition processes. Further reductions in anthropogenic SO_2 and NO_x emissions, and a steady increase in NH_3 emissions, are projected from 2030 to 2100 under these global climate scenarios (Supplementary Fig. 10). Hence, the projected $R_{NHx(dry/wet)}$, as a function of the emission index (EI) and PSI (see detailed information in Supplementary Note 6), will continue to increase between 2030 and 2100 at a rate of 0.007 per year (0.59% yr^{-1}, $p < 0.05$) and 0.010 per year (1.13% yr^{-1}, $p < 0.05$) for the RCP4.5 and RCP8.5 scenarios, respectively (Fig. 3a). As a result, modeled dry NH_x deposition shows increasing trends at a rate of 0.011 kg N ha^{-1} yr^{-2} (0.16% yr^{-1}, $p < 0.05$) for RCP4.5 and 0.020 kg N ha^{-1} yr^{-2} (0.30% yr^{-1}, $p < 0.05$) for RCP8.5, respectively (Fig. 3b). Accordingly, based on the projected dry NH_x deposition and $R_{NHx(dry/wet)}$,

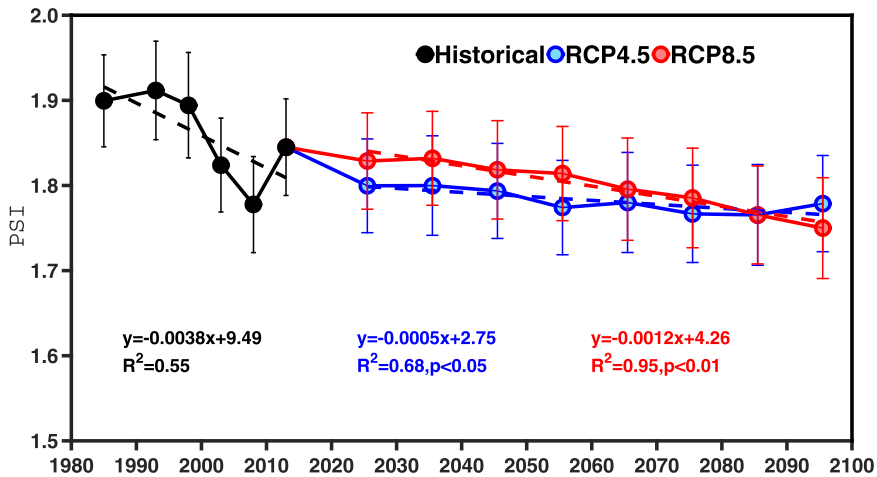

Fig. 2 Long-term trends of the historical (1980–2015) and predicted (2030–2100) precipitation scavenging index (PSI) in China. Predicted PSI was calculated by using the meteorological parameters from the two global future climate change scenarios RCP 4.5 and RCP 8.5. Error bars indicate 1/3 standard deviation (s.d.).

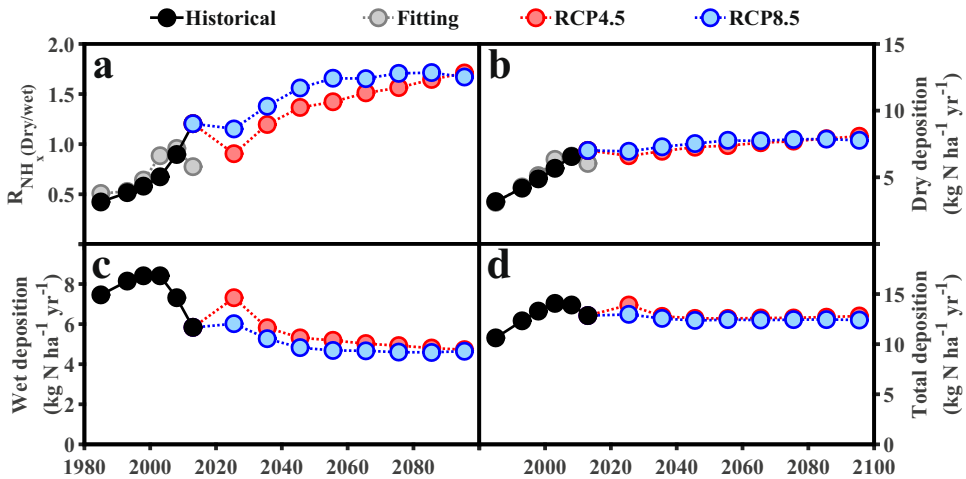

Fig. 3 Temporal variations of NH_x deposition and $R_{NHx(dry/wet)}$ in China over the historical (1980–2015) and future period (2030–2100). a $R_{NHx(dry/wet)}$; **b** dry deposition; **c** wet deposition; **d** total deposition.

we extrapolated that wet NH_x deposition shows a continuing downward trend (Fig. 3c), while the total NH_x deposition remains stable since NH_3 emissions remain constant (Fig. 3d). Even though the total NH_x deposition remains almost stable at around 12–13 kg N ha^{-1} yr^{-1} in the future, the threats to natural ecosystems from NH_x deposition are expected to increase in the future since the projected increase in dry NH_x deposition could lead to greater damage to plant growth as compared to a similar amount of wet NH_x deposition[12,13]. Meanwhile, the shifting of the partitioning of NH_x deposition will also affect the air quality[16,54,55]. This is because the shifting of the partitioning of NH_x deposition from the particulate phase to the gaseous phase would decrease particle pollution to some extent[54]. However, the shifting of the partitioning of NH_x deposition would increase the lifetime of NH_x, and this can facilitate long-range transportation of NH_x, which may affect other regions[55].

DISCUSSION

In light of the importance of dry NH_x deposition to the nitrogen cycle and the associated ecological effects, we broaden our analysis to historical NH_x deposition data in the USA and Europe, which are recognized as additional hotspots of NH_x deposition (see more details in Supplementary Note 2.1 and Supplementary Note 4). Consistent upward trends for modeled $R_{NHx(dry/wet)}$ also occur in the USA and Europe, irrespective of changes in total, wet, and dry deposition of NH_x (Supplementary Fig. 11). Specifically, modeled $R_{NHx(dry/wet)}$ increased at a rate of 0.010 per year (2.05% yr^{-1}, $p < 0.05$) in the USA and 0.005 per year (1.19% yr^{-1}, $p < 0.05$) in Europe. Although wet deposition remained the dominant mechanism for NH_x removal in the USA and Europe, the contribution of modeled dry NH_x deposition to total NH_x deposition increased in the past two decades, from ~30% to ~35% for both the USA and Europe. Similar to the estimation in China, we extrapolate that precipitation pattern variations driven by climate change offset the effect of stable or decreasing NH_3 emissions and increase the contribution of modeled dry NH_x deposition to total NH_x deposition in the USA and Europe (see detailed information in Supplementary Note 4).

The results from the two additional continents further suggest the widespread and growing importance of dry NH_x deposition driven by precipitation patterns resulting from global climate change. This is in contrast with previous studies that suggest relatively little influence of climate change, which may partly be because they only considered changes in precipitation amount, whereas we show that precipitation patterns (intensity and duration) play a far more important role in the shifting of NH_x deposition partitioning[19–22]. Figure 4 schematically illustrates how NH_x deposition partitioning shifts under the influence of climate change-induced variations in precipitation patterns. In view of the

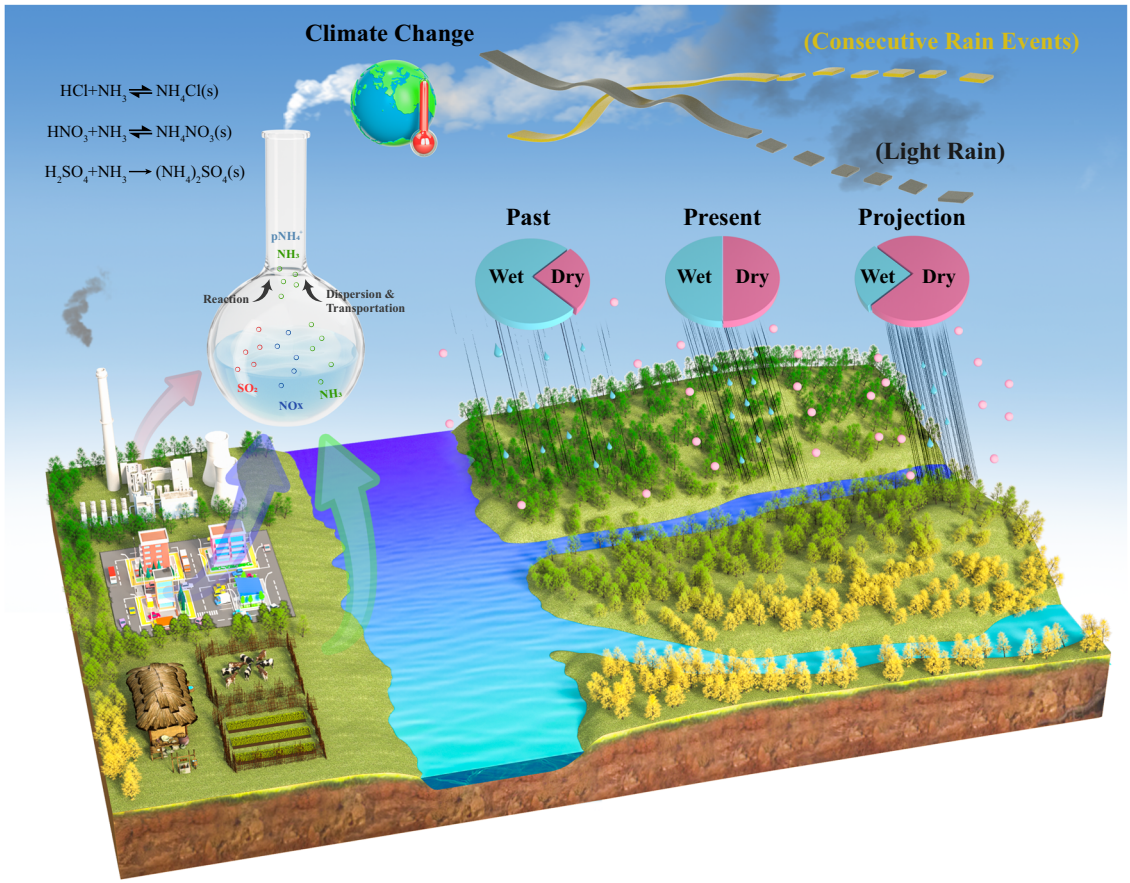

Fig. 4 Schematic representation of the shift in NH_x deposition partitioning driven by climate change-induced variations in precipitation patterns. Atmospheric reduced nitrogen (NH_x) is emitted in the form of NH_3 to a large extent by agricultural activities (nitrogen fertilizer application and livestock manure) and then deposited back to the earth's surface via dry and wet removal pathways. NH_3 emissions, in combination with emissions of the acidic gases SO_2 and NO_x, can influence the partitioning of dry and wet NH_x deposition by affecting the equilibrium between gaseous NH_3 and particulate NH_4^+[11,18]. More importantly, as a consequence of climate change, global trends in precipitation patterns, in particular trends toward less frequent light rain and more frequent consecutive rain events, shift the partitioning of the dry and wet deposited NH_x fractions. Specifically, this leads to a decrease in wet deposition and an increase in dry deposition, leading to negative ecological consequences since the continued increasing dry NH_x deposition causes greater damage to plant growth as compared to the same unit of wet NH_x deposition[12,13].

acceleration of global climate change, less frequent light rain and more frequent consecutive rain events are expected at present and in the coming decades. In particular, extreme rain events are projected to become more frequent in the future, which will further strengthen the shifting of NH_x deposition from the wet to the dry form. Consequently, relatively less NH_x content will be removed from the atmosphere via wet deposition, resulting in a continued increase in the contribution of dry NH_x deposition, which will have a series of side effects on natural ecosystems and human health and air quality[12,54,56]. More importantly, other extreme weather events (e.g., drought and heatwave) due to global climate change will weaken the growth of plants and increase their susceptibility to increased dry NH_x deposition in the future[57].

Note that changes in NH_x deposition can reflect the tendency of total nitrogen deposition to a certain extent, although we focus only on the partitioning of dry and wet NH_x deposition in this study without analyzing NO_y deposition. For example, the increasing importance of NH_x deposition compared with NO_y deposition can be inferred based on the combination of historically measured wet nitrogen deposition and modeled dry nitrogen deposition data shown in Yu et al.[16]. Looking to the future, NH_x deposition will dominate total nitrogen deposition after 2030, as NH_3 emissions are expected to surpass NO_x emissions at that time (Supplementary Fig. 10), suggesting that the role of NH_x deposition in nitrogen deposition will become more prominent, especially with the continued strict control of NO_x emissions. Lamarque et al.[58] also reported that nitrogen deposition was projected to increase over certain regions around the world by the end of the 21st century, especially in Asia, owing to the projected increase in NH_3 emissions. Consequently, NH_3 emission control is still the most effective and fundamental way to reduce NH_x deposition and to curb its damage to natural ecosystems and human health, although NH_3 emission abatement cannot fully control NH_x deposition as previous studies documented that per-unit reduction in NH_3 emissions led to only a 60–80% mitigation of NH_x deposition[10,59]. Nevertheless, the analysis in the USA and Europe also highlights that the expected benefits from NH_3 emission reductions on dry NH_x deposition are diminished by the effects of climate change. Hence, more stringent NH_3 emission controls are required to achieve the desired reductions in NH_x deposition levels in the future[20]. A significant proportion of the NH_3 emissions in China can be attributed to its agricultural sources, and improving agricultural nitrogen management (e.g., reduced fertilizer application, covered manure storage, and implementation of the deep application of fertilizers during growing seasons) could be the most effective approach to reduce NH_3 emissions from agricultural sources in China to date and in the future[54,60,61].

This study prompts our re-examination of the significance of dry NH_x deposition in the global nitrogen cycle and ecosystem assessment and poses an urgent need to mitigate NH_x deposition in the face of global climate change. Nevertheless, this study is still subject to four major uncertainties and limitations (see more information in Supplementary Note 7): (1) Long-term trends of dry NH_x deposition are modeled results, which entail a higher degree of uncertainty than the observed wet NH_x deposition[11,62,63]. Deposition velocity (V_d) is the dominant contributor to the uncertainty of dry deposition simulations and could potentially affect the estimation of dry NH_x deposition flux[34–36]. However, year-specific instead of time-varying V_d was used to estimate long-term trends of dry NH_x deposition in this study, which could introduce additional uncertainties. (2) The historical NH_3 emissions data were calculated based on bottom-up activity data and static emissions factors, which did not consider the effect of interannual meteorological variability[64–67]. Whereas NH_3 emissions were extremely weather/climate-sensitive[68], the influence of climate change-induced precipitation patterns and other meteorological factors (e.g., temperature and wind speed) are supposed to affect the emission processes of NH_3, which might also introduce additional uncertainties. (3) The land/atmosphere exchange of gaseous NH_3 is, in fact, bi-directional and controlled by a range of environmental factors[69–71]. Nonetheless, owing to the difficulty of obtaining detailed input parameters and empirical tuning at a large scale, NH_3 bi-directional exchange process has been incorporated into only a few of the regional- and global-scale CTMs (i.e., CMAQ, CAM_X, GEM-MACH, and LOTOS-EUROS)[35,36,72,73]. Therefore, the lack of consideration of bi-directional parameterization schemes simultaneously affects the estimation of NH_3 emissions and NH_x deposition[74–76] and introduces additional uncertainties. (4) Apart from precipitation patterns, global climate change can also affect NH_x deposition partitioning through other aspects such as surface temperature, solar radiation, relative humidity, soil temperature, and natural surface properties[68,77,78]. However, the complex interactions of multiple meteorological parameters, as opposed to their individual effects on NH_x deposition partitioning, make it difficult to isolate and quantify the relative importance of these meteorological parameters. Hence, the quantification of global climate change-induced NH_x deposition partitioning is still subject to large uncertainty and remains a great challenge for the whole community. Development of climate-dependent models for better quantification of the dynamic relationship between NH_3 emissions, climate/meteorology, and deposition under global environmental change in the future is therefore needed[77].

METHODS

Historical dry and wet NH_x deposition data in China

Both the annual modeled dry NH_x deposition and measured wet NH_x deposition data in China for the period of 1980–2015 were obtained from Yu et al.[16], which is the most comprehensive and current data on long-term NH_x deposition available in China. General information regarding the sources of the NH_x deposition data collected in China has been summarized in Supplementary Table 1. Briefly, observed wet NH_x deposition data were obtained from four sources, including (1) peer-reviewed published papers screened by Yu et al.[16], (2) the Acid Deposition Monitoring Network in East Asia (https://www.eanet.asia/, last access: 3 January 2023), (3) the National Ecosystem Research Network of China (http://www.cnern.ac.cn/index.action, last access: 3 January 2023), and (4) the Nationwide Nitrogen Deposition Monitoring Network established by China Agricultural University[72], with a total of 956 monitoring sites.

Modeled dry NH_x deposition fluxes were obtained from inferential methods, which combine ambient concentrations and dry deposition velocities (see Supplementary Note 1 for the parameterization of dry NH_3 deposition)[5,79,80]. The average land use specific V_d collected from previous studies was used to calculate annual dry NH_x deposition (Supplementary Fig. 12, see Extended Data Table 4 in Yu et al.[16]). Specifically, modeled V_d for 9 land use types was derived from Xu et al.[11], who applied the GEOS-Chem model coupled with the scheme of Wesely[81] and Zhang et al.[82] to calculate V_d for gaseous NH_3 and particulate NH_4^+, respectively. Modeled V_d in the rest of 5 land use types (i.e., desert, tundra, water, wetland, and ice) was estimated by using similar parameterization schemes used as in Xu et al.[11] but with different CTMs (CMAQ, RegAMDS)[83–88]. It is worth noting that the modeled V_d in the 14 land use types derived from Yu et al.[16] was originally based on a unidirectional method, except for Su et al.[83], who used a bi-directional NH_3 exchange method to calculate V_d for gaseous NH_3 over deserts.

Yu et al.[16] averaged the annual NH_x deposition during 1980–2015 for 6 periods, including 1980–1990, 1991–1995, 1996–2000, 2001–2005, 2006–2010, and 2011–2015[15]. Hence, we

also averaged precursor emissions for the corresponding six periods (see Supplementary Note 2.3 for detailed information regarding emissions data). The data in each period were fitted with the middle year of the corresponding period in order to obtain the linear regressions. Subsequently, the slope of the linear regression was used to represent the average annual rate of change. The relative annual rate of change was then calculated by normalizing the annual rate of change with the value for the first time period in each region.

Quantification of the contribution of emissions to NH_x deposition partitioning

The response relationship between NH_x deposition and precursor emissions was developed based on the WRF-EMEP model (the Weather Research and Forecasting model coupled with the European Monitoring and Evaluation Program)[32]. Model configuration and evaluation are presented in Supplementary Note 5. A series of sensitivity studies were conducted to quantify the response of NH_x deposition to reductions in SO_2, NO_x, and NH_3 separately, as well as $SO_2 + NO_x + NH_3$ together. The reduction values for individual emissions were set to be 10%, 30%, 50%, 70%, and 90%, respectively. Sensitivity results show that the response of regional NH_x deposition to individual emission reductions is nearly linear. This is consistent with the study by Civerolo et al.[19], which also revealed that the response of global nitrogen deposition to emissions was nearly linear based on six different tropospheric chemistry models. Furthermore, sensitivity results also revealed that the effect of NH_3 emission variation is far more important than that of SO_2 and NO_x emissions, which has also been found by previous studies[74,89]. Last but not least, the response to the co-reduction of $SO_2 + NO_x + NH_3$ was nearly equal to the sum of the individual reductions of SO_2, NO_x, and NH_3. Therefore, the total variations in dry NH_x deposition (ΔDry) and $R_{NHx(dry/wet)}$ (ΔRatio) caused by precursor emissions during 1980–2015 can be obtained using Eq. (2):

$$\Delta\text{Dry or }\Delta\text{Ratio} = a_1 \times \Delta SO_2 + a_2 \times \Delta NO_x + a_3 \times \Delta NH_3 \quad (2)$$

Where a_1, a_2, and a_3 are constants, indicating the variations in dry NH_x deposition and $R_{NHx(dry/wet)}$ caused by per-unit reduction in SO_2, NO_x, and NH_3 emissions, respectively, as listed in Supplementary Table 2. ΔSO_2, ΔNO_x, and ΔNH_3 are the changes in SO_2, NO_x, and NH_3 emissions between 1980 and 2015, respectively.

DATA AVAILABILITY

All data used in this study are available in the main text or the supplemental materials. Links to download the NH_x data (i.e., ambient concentration, precipitation concentration, and deposition flux) can be found in Supplementary Table 1. Links to download historical and predicted anthropogenic emissions data and meteorological data are provided in the Supplementary Note 2.

CODE AVAILABILITY

We performed simulations using WRF v3.9.1 coupled with the EMEP rv4.17 model, which are freely available at https://www2.mmm.ucar.edu/wrf/users/ (last access: 3 January 2023) and https://github.com/metno/emep-ctm (last access: 3 January 2023), respectively.

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AUTHOR CONTRIBUTIONS

W.H.C., S.G.J., X.M.W., and M.S. conceived the study and led the analysis. W.H.L. helped with precipitation data processing. B.Q.Z., W.W.W., and J.Y.M. helped with emissions and ammonium deposition data processing. M.C. helped with the EMEP model simulation. C.F., A.G., P.F.Y., X.J.L., G.R.Y., and G.C. provided important views on the study. All co-authors contributed to improving the analysis and interpretation.

COMPETING INTERESTS

The authors declare no competing interests s as defined by Nature Research, or other interests that might be perceived to influence the interpretation of the article.

ADDITIONAL INFORMATION

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