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# Impacts of climate change and emissions on atmospheric oxidized nitrogen deposition over East Asia

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**Abstract**

A multi-model ensemble of Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) simulations are used to study the atmospheric oxidized nitrogen ($NO_y$) deposition over East Asia under climate and emission changes projected for the future. Both dry and wet $NO_y$ deposition shows significant decreases in the 2100s under RCP 4.5 and RCP 8.5, primarily due to large anthropogenic emission reduction over both land and sea. However, in the near future of the 2030s, both dry and wet $NO_y$ deposition increases significantly due to continued increase in emissions. The individual effect of climate or emission changes on dry and wet $NO_y$ deposition is also investigated. The impact of climate change on dry $NO_y$ deposition is relatively minor, but the effect on wet deposition, primarily caused by changes in precipitation, is much higher. For example, over the East China Sea, wet $NO_y$ deposition increases significantly in summer due to climate change by the end of this century under RCP 8.5, which may subsequently enhance marine primary production. Over the coastal seas of China, as the transport of $NO_y$ from land becomes weaker due to the decrease of anthropogenic emissions, the effect of ship emission and lightning emission becomes more important. On average, seasonal mean total $NO_y$ deposition is projected to be enhanced by 24-48% and 3%-37% over Yellow Sea and East China Sea, respectively, by the end of this century. Therefore, continued control of both anthropogenic emission over land and ship emissions may reduce $NO_y$ deposition to the Chinese coastal seas.

Key words: ACCMIP, $NO_y$ deposition, RCP 8.5, ship emission

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## 1. Introduction

As a nutrient, nitrogen is essential to the terrestrial and marine ecosystems and plays vital roles in lives on earth from many perspectives such as biodiversity, primary production, etc. (Doney et al., 2007; Butchart et al., 2010; Stevens et al., 2015; Galloway et al., 2008). The oceans comprise the largest and most important ecosystems on Earth and atmospheric nitrogen deposition is an important pathway for delivering nutrients to the ocean (Duce et al., 2008).

The characteristics of atmospheric deposition have been widely studied around the world. The concentrations and fluxes of trace elements in atmospheric deposition are influenced by many factors such as rainfall amount, local emissions as well as the long-range transport of pollutants, etc. (Kim et al., 2000; Cong et al., 2010; Theodosi et al., 2010; Vuai and Tokuyama, 2011; Kim et al., 2012; Connan et al., 2013; Montoya-Mayor et al., 2013). Studies have shown significant changes of nitrogen deposition in the future under the influence of changes in both climate and emissions following the representative concentration pathways (RCPs) (Van Vuuren et al., 2011; Ellis et al., 2013; Lamarque et al., 2013a).

Since projections of future changes in nitrogen deposition from individual models are prone to specific model errors (Reichler and Kim, 2008; Shindell et al., 2013), multi-model ensembles in either climate (Gao et al., 2014; 2016) or chemistry (Lamarque et al., 2013a; 2013b) are important for identifying robust and non-robust changes projected by models. This study uses the nitrogen deposition from an ensemble of models that contributed to the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP; (Lamarque et al., 2013b)). The nitrogen deposition includes both the oxidized nitrogen deposition ($NO_y$, mainly including NO, $NO_2$, $NO_3^-$, $N_2O_5$, $HNO_3$ and organic nitrates) and reduced nitrogen ($NH_x$, mainly including $NH_3$, $NH_4^+$ and organic ammonium). Since the number of models with $NH_x$ in ACCMIP is less than 5, this study only focuses on the $NO_y$ (10 models or so) deposition, which mainly results from $NO_x$ emissions.

Due to the rapid economic development in China, $NO_x$ emission increase in the past (Wang et al., 2013) has led to an increase of nitrogen deposition. For example, Liu et al. (2013) found that nitrogen deposition over land in China increased from 13.2 kg/ha in 1980s to 21.1 kg $ha^{-1}$ in 2000s, with increase of 60%. In addition, the increased $NO_x$ emission may also enhance $NO_y$ deposition in Chinese coastal seas, due to the atmospheric and riverine transport of $NO_x$ (Luo et al., 2014). In particular, China has a long coastline of almost 18,000 km in length and over 300 million square kilometer sea areas, with high density population and industries in the coastal provinces. Liu et al. (2016) reported that the shipping $NO_X$ emissions in East Asia increased from 1.08 Tg in 2002 to 2.8 Tg in 2013.

Studies on the changes of nitrogen deposition under the influence of both climate and emission changes have been limited over East Asia. Using the old Special Report on Emissions Scenarios (SRES) such as A2, Lamarque et al. (2005) found large increases of nitrogen deposition over East Asia due to increased emissions, whereas the effect from climate change is much smaller and lacks consensus due to the small ensemble size. In 2100 nitrogen deposition changes due to changes in climate are much less than changes due to increased nitrogen emissions. In contrast, based on the new scenarios RCP 4.5 and 8.5, Lamarque et al. (2013a) found that the total $NO_y$ deposition (wet + dry; Fig. 5a in Lamarque et al. (2013a)) over East Asia was projected to decrease by the end of this century due to the combined effect of emissions and climate, but the changes are mainly triggered by the decrease of emissions. However, the individual effect of climate or emissions was not examined in that study. With the same dataset of ACCMIP, Allen et al. (2015) found that by keeping the emissions at current level, atmospheric aerosol shows significant changes with changing climate, i.e., aerosol wet deposition decreases over the tropics and Northern Hemisphere midlatitudes with reduction in precipitation over land. Climate change alone may modulate the changes in the deposition, particularly for wet deposition due to the response of precipitation to climate change. Hence, it is important to elucidate the influence of climate and emission changes on the dry and wet $NO_y$ deposition over East Asia using the multi-model

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ensemble ACCMIP results.

In what follows, we first discuss the capability of ACCMIP in capturing the deposition patterns, followed by the changes of dry and wet deposition in future under the combined effect of climate change and emissions. Lastly, we elucidate the individual effect from climate change or emissions.

## 2. Model description

In this study, about 10 models from ACCMIP are used, similar to Lamarque et al. (2013b). All the data are interpolated to a spatial resolution of 2° × 2° to facilitate analysis and comparison across models. To evaluate the impacts of climate and emission change as well as to isolate their individual effect, five cases of ACCMIP scenarios are used in this study, as listed in Table 1. The base case over the historical period covers the decade of 2000, mainly from 2001-2010. Two cases target the investigation of both climate and emission changes under future scenarios of RCP 4.5 and RCP 8.5, covering two periods in the decades of 2030 and 2100 (first column of Table 1). The remaining two cases are used to investigate the impact from climate change only in the 2030s and 2100s under RCP 8.5 by maintaining emission at the level of year 2000 (last column of Table 1). As different models have different simulation years, some model may not cover the entire decades of 2030 and 2100. Detailed simulation lengths for each model are listed in Table S1. In the ACCMIP dataset, the summation of all simulated oxidized nitrogen species is referred to as $NO_y$, which is the major focus of this study.

Table 1. Scenarios used in this study

| | Base | Changes in both climate and emissions | | Climate change only | |
|---|---|---|---|---|---|
| Scenarios | Historical | RCP 4.5 | RCP 8.5 | Em2000Cl2030 | Em2000Cl2100 |
| Period | 2000-2010 | 2030-2039 | 2030-2039 | 2030-2039 | 2100-2109 |
| | | 2100-2109 | 2100-2109 | | |

## 3. Evaluation of the ACCMIP results

The deposition results of ACCMIP have been extensively evaluated previously across land areas by comparing with three datasets including North American Deposition Program (NADP), European Monitoring and Evaluation Programme (EMEP) and Acid Deposition Monitoring Network in East Asia (EANET), and reasonable performance was demonstrated by the ACCMIP results (Lamarque et al., 2013a). There is a lack of deposition data over the ocean, making evaluation of the ACCMIP results across the oceans difficult. Recently, Baker et al. (2017) conducted an intensive evaluation of the ACCMIP multi-model mean based on a large number of dry $NO_y$ deposition samples, i.e., a total of 770 samples collected over the Pacific, showing comparable spatial distributions between observations and ACCMIP, such as a consistent northwest-southeast gradient with higher deposition flux closer to the coast (Fig. 12 in Baker et al. (2017)). In terms of wet deposition, considering the close relationship between wet deposition and precipitation (Kryza et al., 2012; Wałaszek et al., 2013), evaluation of precipitation is performed using the Tropical Rainfall Measuring Mission (TRMM; http://pmm.nasa.gov/trmm) and Global Precipitation Climatology Project (GPCP) v 2.3 (Adler et al., 2018) precipitation data. Fig. 1 shows a comparison of the annual mean precipitation over the historical period (2000-2010) among the ACCMIP multi-model ensemble mean and TRMM, which only covers 60°N-60°S, and GPCP. In general, the ACCMIP mean precipitation well captured the spatial variations of the observed precipitation from both TRMM and GPCP, with stronger precipitation in the southern part of Asia, particularly over the South China Sea and the Bay of Bengal, and lighter precipitation in northern China (i.e., Northwest China). In particular, the rain belt stretching from the east of Japan to the Philippines in summer is also well captured by ACCMIP.

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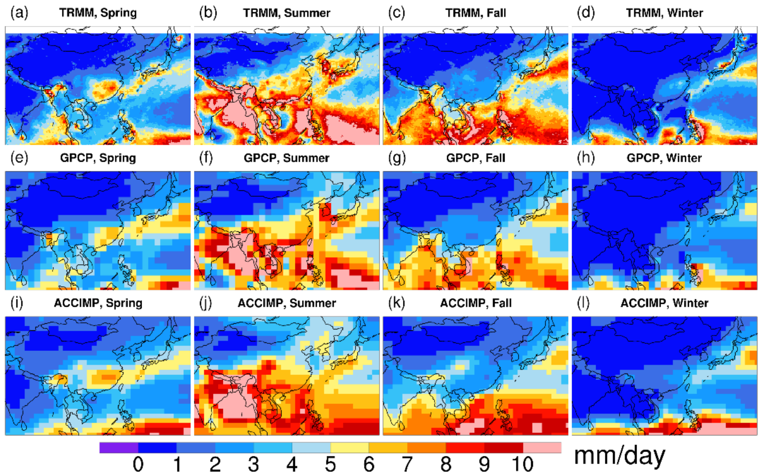


Fig. 1. Evaluation of seasonal mean precipitation during 2001-2010: ACCMIP multi-model ensemble mean vs. TRMM and GPCP

## 4. Future changes of $NO_y$ deposition in East Asia

Considering the uncertainty and variability among multiple ACCMIP results, all analyses, i.e., the future changes of deposition, are performed based on model agreement and statistical significance. Following our previous studies (Gao et al., 2014; 2015), results at a model grid cell are considered to have agreement if at least 70% of the ACCMIP models show the same sign of change as the ACCMIP multi-model ensemble mean. For models showing agreement with the ensemble mean, if more than half of the models show statistical significance at 95% level, then the ensemble mean change for that particular grid is considered to be statistically significant.

The seasonal mean distribution of dry $NO_y$ deposition over East Asia areas for historical (2001-2010) and projected future changes under RCPs scenarios (RCP 4.5 and RCP 8.5) during the two periods of 2030 and 2100 are shown in Fig. 2. The four seasons defined in this study are spring (March to May), summer (June to August), fall (September to November) and winter (December to February).

As anthropogenic activities play important roles in $NO_x$ emissions, high atmospheric dry nitrogen ($NO_y$) deposition values mainly cluster around areas with high population density and industrial activities in the historical periods (top row of Fig. 2), e.g., high values of $NO_y$ deposition can be seen in East China, Korea, Japan and their coastal seas. In this study, in addition to the land areas, we also focus on three coastal seas in East Asia (Bohai Sea, Yellow Sea and East China Sea from high latitude to low latitude, referred to as the BYE areas below), marked by the three black boxes in Fig. 2a. A gradient of decreasing $NO_y$ deposition is found (top row of Fig. 2) from eastern China to the coastal areas. Seasonal variations show that over mainland China, summer is the season with the highest dry and wet $NO_y$ deposition, with high dry deposition likely caused by the high deposition velocity (Zhang et al., 2017) and wet deposition due to larger precipitation in summer, consistent with previous studies (Liu et al., 2017; Zhang et al., 2017; Xu et al., 2018). Over the ocean such as Yellow Sea and East China Sea, the notably higher $NO_y$ deposition (first row of Fig. 2) is partly attributed to $NO_x$ emission transported from land to the coastal seas. In particular, the dry $NO_y$ deposition over the East China Sea is obviously higher in winter compared to summer, likely resulting from enhanced transport by the northwesterly winds during the winter monsoon (Ding, 1991).

Considering the projected future changes of $NO_y$ deposition, we show the distributions in the 2030s and 2100s under the RCP 4.5 and RCP 8.5 scenarios, representing near-term and long-term changes. Dry $NO_y$ deposition decreases remarkably in the 2100s under the RCP 4.5 and RCP 8.5 scenarios over East Asia, a result of large decrease in emissions (second column in Fig. S1 in the supporting information). In the 2030s, besides the decrease of dry deposition in Japan, Korea and the surrounding areas, RCP 8.5 shows a predominant increase of dry deposition (Fourth row in Fig. 2); in contrast, robust significant increases in western China and India are projected, with little or weak signals in eastern China in RCP 4.5, consistent with the emission change patterns (first column in Fig. S1).

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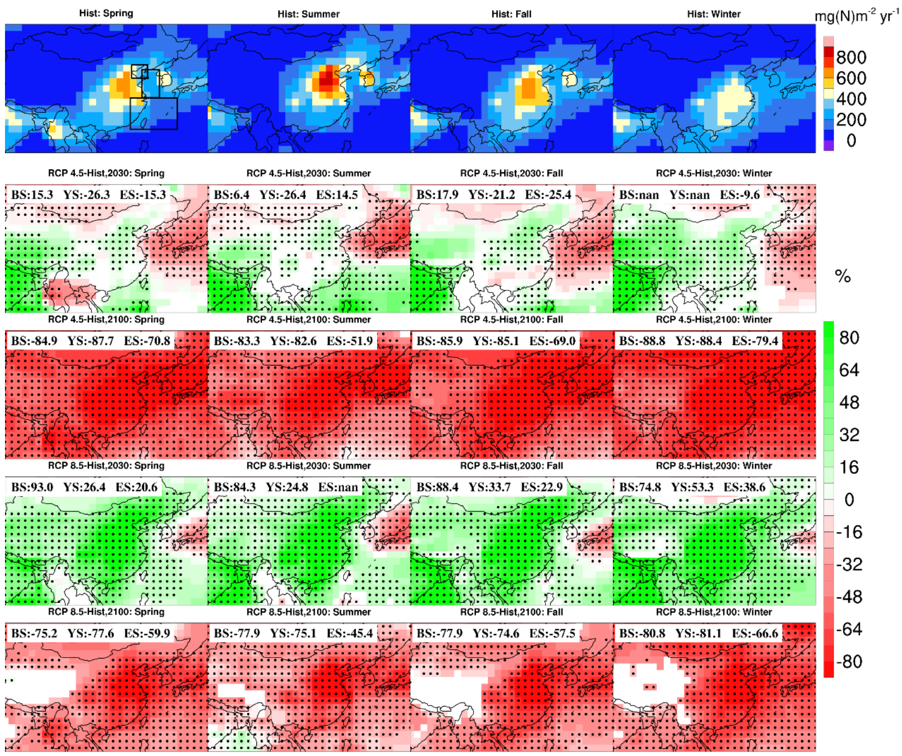


Fig. 2 Spatial distribution of mean seasonal dry $NO_y$ deposition over East Asia under historical (2001-2010; top row) as well as the future changes (second to fifth rows representing changes under RCP 4.5 2030, RCP 4.5 2100, RCP 8.5 2030 and RCP 8.5 2100 in relative to historical period). Only grids with multi-model agreement are shown (grids without model agreement are in white), and stippling marks areas with statistical significance ($\alpha$=0.05). Regions of Bohai Sea (BS), Yellow Sea (YS) and East China Sea (ES) are marked by the black rectangles in the top left panel, with mean changes shown on the top left of each panel starting from the second row. The mean change of a region is set to nan if the number of significant grids in this region is less than half of the area.

For wet $NO_y$ deposition, as discussed earlier, summer is the season with strongest deposition (first row of Fig. 3), primarily caused by the largest precipitation among the four seasons (Fig. 1). In the 2030s, changes of wet deposition (second and fourth rows of Fig. 3) are in general similar to the patterns of dry deposition changes (second and fourth rows of Fig. 2). However, in the 2100s, the patterns of wet deposition changes are different from those of dry depositions, with relatively clear east/west dipole features in particular under RCP 8.5. To elucidate what controls the dipole patterns, the individual effect of climate change and emissions is discussed in the next section.

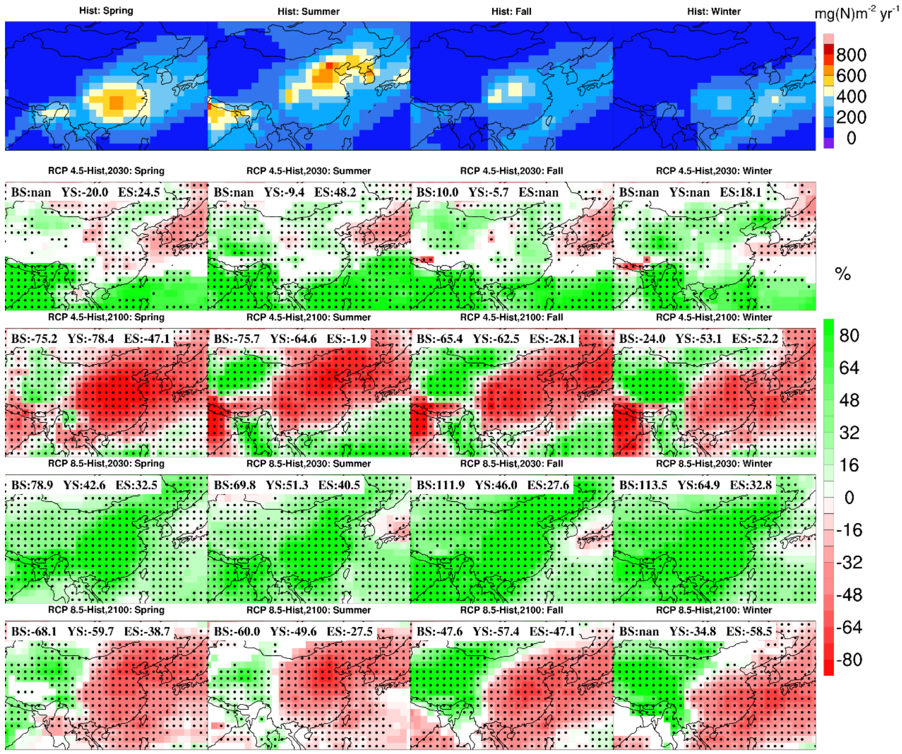


Fig. 3. Same as Fig. 2 except for wet $NO_y$ deposition.

## 5. The impact of climate change or emissions on $NO_y$ deposition

Two scenarios from ACCMIP are used in this study to isolate the influence of anthropogenic emissions and climate change on $NO_y$ deposition. The two scenarios are shown in Table 1, with emissions kept at the current level (the decade of 2000) but climate for the 2030s and 2100s under RCP 8.5 are compared.

Climate change alone has negligible contributions to the dry $NO_y$ deposition changes, as shown in Fig. 4. Generally, calculation of dry deposition flux in chemical models follows equation 5.1, where F is vertical dry deposition flux, C is concentration of specific gas or particle and $v_d$ is the dry deposition velocity.

$$F = -v_d C \tag{5.1}$$

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All models in ACCMIP calculated dry deposition velocity using the resistance approach (Lamarque et al., 2013b), which defines the inverse of dry deposition velocity as equation 5.2,

$$\frac{1}{v_d} = r_t = r_a + r_b + r_c \qquad (5.2)$$

where $r_t$ is the total resistance, $r_a$ is the aerodynamic resistance which is common to all gases, $r_b$ is the quasilaminar sublayer resistance and $r_c$ is the bulk surface resistance (Steinfeld, 1998). As $r_b$ depends on the molecular properties of the target substance and deposition surface and $r_c$ depends on the nature of surface (Steinfeld, 1998), they do not vary under climate change. As for $r_a$, it plays a significant role in transporting gases and particles from atmosphere to the receptor surface. $R_a$ is governed by atmospheric turbulent transport, mainly controlled by the wind shear as well as buoyancy (Erisman and Draaijers, 2003). Therefore, climate change affects dry deposition velocity for the gases or particles mainly through its modulation of $r_a$. As shown in Fig. 4, the changes of dry depositions from climate change alone are mostly negligible compared to the total changes from both climate change and emissions (Fig. 3), indicating statistically insignificant change of $r_a$ under a warmer climate.

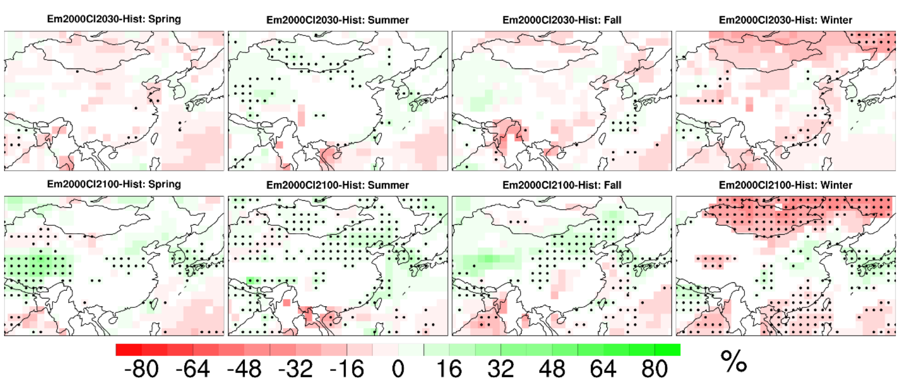


Fig. 4. Spatial distribution of mean seasonal dry $NO_y$ deposition change over East Asia under experimental scenarios of ACCMIP (Em2000Cl2030 and Em2000Cl2100) relative to historical period (2001-2010). The distribution of mean seasonal dry $NO_y$ deposition under historical period is shown in Fig. 2 (top row). Only grids with multi-model agreement are shown (grids without model agreement are in white), and among the grids with model agreement, stippling marks statistical significance (α=0.05).

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Considering the impact of climate conditions on $NO_y$ deposition, precipitation is an important factor and has been shown to positively correlate with wet $NO_y$ deposition (Kryza et al., 2012; Wałaszek et al., 2013). In order to further quantify the relationship between wet deposition and precipitation, we display in Fig. 5 the correlation between the changes of precipitation and wet $NO_y$ deposition over the BYE areas for the scenarios with fixed emissions. All correlations are positive and statistically significant. There is a larger inter-model spread of changes in Em2000Cl2100 compared to Em2000Cl2030, and the larger changes in precipitation and wet deposition allow a stronger correlation between them to emerge in the 2100s relative to the 2030s. Meanwhile, winter owns the highest correlation in both Em2000Cl2030 and Em2000Cl2100, partly related to the significant decrease of both wet $NO_y$ and precipitation in winter under Em2000Cl2100 over East China Sea which will be discussed in detail next.

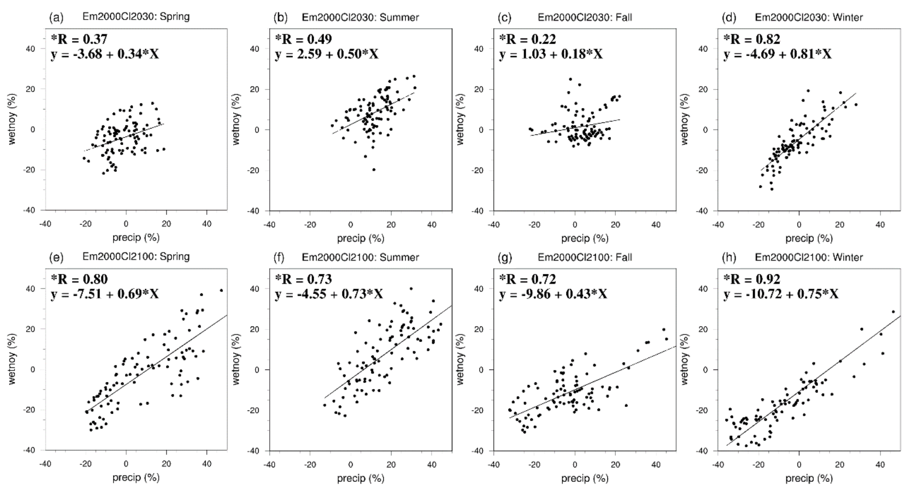


Fig. 5. Comparison between precipitation and wet $NO_y$ deposition changes under the experimental scenarios of ACCMIP (Em2000Cl2030 and Em2000Cl2100) relative to historical period (2001-2010) over Bohai Sea, Yellow Sea and East China Sea. An r-test (α=0.05) is performed in each panel for statistical significance and the star before "R" indicates statistical significance at 95% confidence level. Each point in this figure corresponds to the results from an individual model of ACCMIP.

As depicted in Fig. 6, the changes of wet deposition in the 2030s due to climate change is mostly insignificant (first row) and correspond well with the insignificant

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changes of precipitation (third row). Similarly, the patterns in the 2100s between the changes of wet deposition (second row) and precipitation (fourth row) are quite consistent. For example, in spring, summer and fall, a dominant increase in western China is projected (first three panels in the second and fourth rows), whereas in winter, a north and southeastern dipole feature is clearly seen. Over the East China Sea, wet $NO_y$ deposition increases significantly in summer (18%) and decreases significantly in winter (-13%), indicating a remarkable influence of climate change on the wet $NO_y$ deposition. The changes of precipitation are generally consistent with that reported in other studies. Both Chong-Hai and Ying (2012) and Wang and Chen (2014) show significant increase of precipitation except for eastern South China at the end of the 21[st] century under RCP 8.5. Comparing Fig. 6 with Fig. 3, it is clear that the dipole pattern of changes in wet deposition in the 2100s shown in Fig. 3 is primarily related to the large reduction of emission over eastern China.

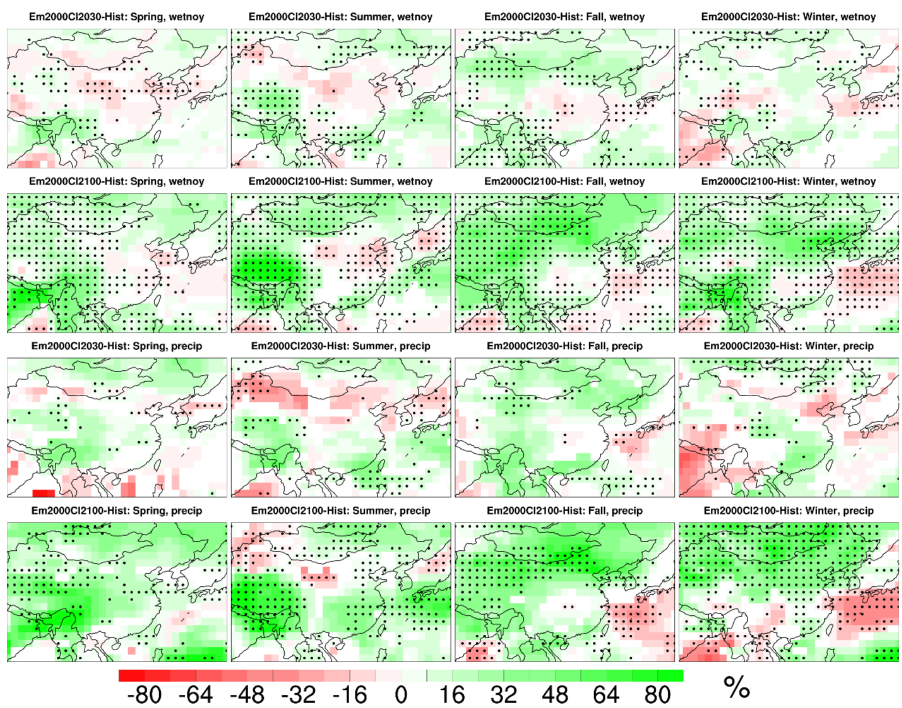


Fig. 6. Spatial distribution of mean seasonal wet $NO_y$ deposition change and precipitation change under EM2000Cl2030 and Em2000Cl2100 relative to historical period (2001-2010). The panels are drawn and arranged in the same manner as Fig. 2.

From the global perspective, the emission of nitrogen oxide is in general balanced by the $NO_y$ deposition as documented in Lamarque et al. (2013a) using the ACCMIP model results, although $NO_y$ deposition might be larger due to the downward transport from the stratosphere. Over the Chinese coastal seas, we calculate the multi-model seasonal mean $NO_y$ deposition and $NO_x$ emissions, mainly from ships and lightning. To avoid biases from spatial interpolation, calculation is performed based on the original model grid and regionally averaged for each model. Since the changes in Bohai are less significant in general, particularly in the near future (second row of Figs. 2,3), we only focus on the emission and deposition changes over Yellow Sea and East China Sea. The total $NO_x$ emissions over the oceans such as Yellow Sea and East China Sea can be mainly classified into two categories, ship and lightning emissions. As a dominant contributor of $NO_x$ emission in the ocean (Dalsøren et al., 2009; Eyring et al., 2010), Fan et al. (2016) concluded that 85% of ship emissions took place within 200km of the coastlines. As reported by Liu et al. (2016), ship emissions from East China Sea may account for a large percentage (31%) of the total ship emission in East Asia. Thus, ship emission may contribute significantly to emissions in the Chinese coastal seas. The percentage of dry and wet deposition, as well as the ratio of ship emission and lightning emission to the total $NO_y$ deposition are shown in Figs. 7 and 8. The ratio of ship emission and lightning emission to the total $NO_y$ deposition is used to characterize their contribution to $NO_y$ deposition with the assumption that all ship and lightning emission contribute to the $NO_y$ deposition, which can be considered an upper bound of their contribution.

A couple of features can be identified from Fig. 7 and Fig. 8. First, the percentage of dry deposition over Yellow Sea and East China Sea (third and fifth blue color bars from the left in each panel of Figs. 7, 8) decreases in the 2100s under RCP 4.5 and RCP 8.5, consistent with the patterns shown in Fig. 2 (third and fifth rows), due primarily to emission reduction. Second, with larger emission reduction over land (e.g., eastern China) compared to ocean (Fig. S1), the contribution of ship emission to total $NO_y$ deposition over the seas may become larger in future (black color bars in Figs. 7,8). For instance, over the historical period, the seasonal contribution of ship emission to total

$NO_y$ depostion is 22%-30% and 52%-82% for Yellow Sea and East China Sea, respectively; however, in 2100, it reaches 56%-99% (RCP 4.5) and 42-58% (RCP 8.5) for Yellow Sea, 81% to almost 100% (RCP 4.5) and 74% to almost 100% (RCP 8.5) for East China Sea, with mean seasonal increase of 24-48% and 3%-37% for Yellow Sea and East China Sea, respectively. Third, the contribution of lightning $NO_x$ in spring and winter is negligible, however, the contribution is nontrivial in summer and fall. In particular, due to the reduction in anthropogenic emissions and ship emissions in 2100 under RCP 4.5 and RCP 8.5, the contribution of lightning $NO_x$ becomes more obvious compared with the case without emission reduction. For example, in the summer of the 2100s, the contribution of lightning $NO_x$ increases from 1% to 7% (both RCP 4.5 and RCP 8.5) over Yellow Sea, 3% to 7% in RCP 4.5 and 6% in RCP 8.5 over East China Sea. In the fall of the 2100s, the contribution of lightning $NO_x$ increases from less than 1% to 3% (both RCP 4.5 and RCP 8.5) over Yellow Sea, and 1% to 4% (both RCP 4.5 and RCP 8.5) in East China Sea. These results illustrate a shift in the future towards enhanced impacts from ship and lightning emissions when anthropogenic emissions are largely controlled in the upwind land regions.

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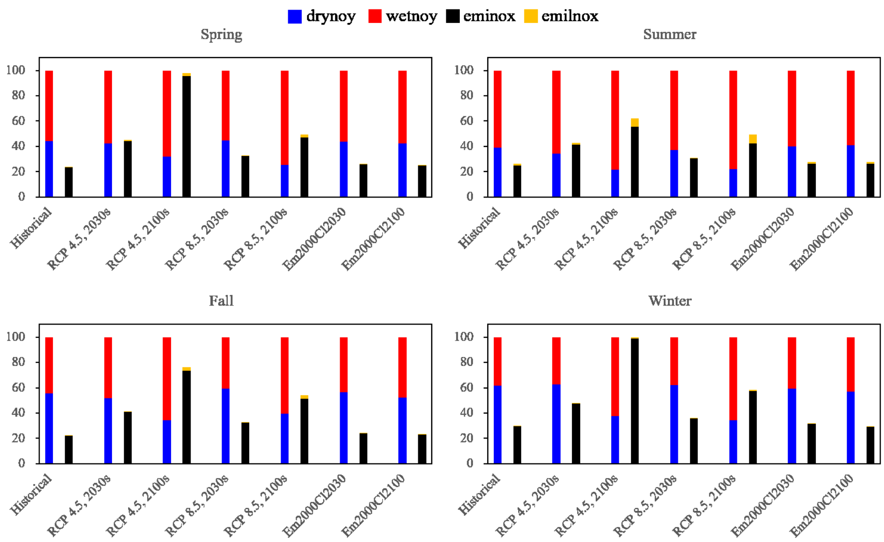


Fig. 7 Stacked bars of seasonal ratio of $NO_y$ deposition from wet (wetnoy) and dry (drynoy) deposition and $NO_x$ emissions from ship (eminox) and lightning (emilnox) to the total (wet + dry) $NO_y$ deposition in the historical and RCP scenarios over Yellow Sea. Two color bars are shown for each period with the left one representing dry $NO_y$ (blue) and wet $NO_y$ (red) deposition and the right one representing eminox (black) and emilnox (orange).

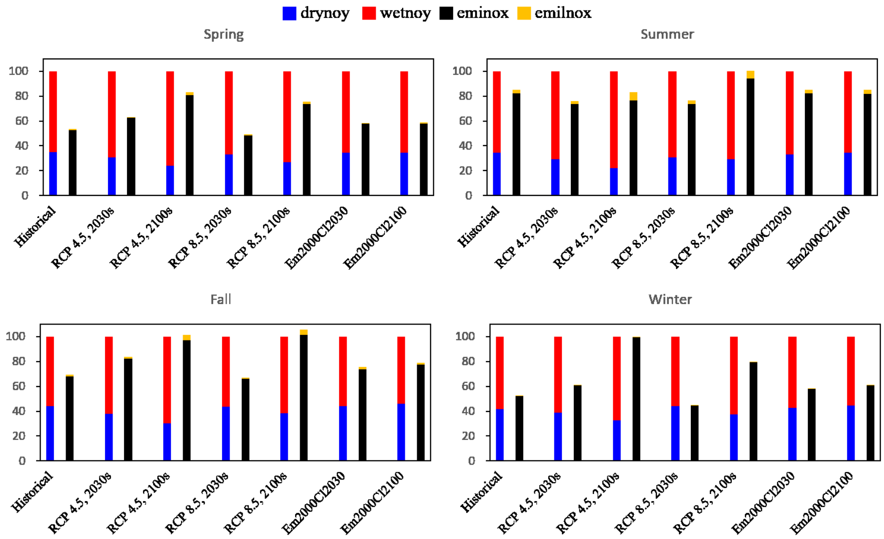


Fig. 8 Same as Fig. 7 except for East China Sea.

## 6. Marine primary production over the BYE areas and its future change

Generally, the Chinese coastal seas have rich nutrients and high total primary production (Gong et al., 2000; Son et al., 2005). Thus, these areas seldom lack nutrient but sometimes eutrophication is an environmental issue. For instance, a massive Ulva prolifera bloom occurred in June 2008 in the Yellow Sea and the harmful algal bloom caught a lot of attention. Hu et al. (2010) found that algal blooms occur in each summer of 2000-2009 in the Yellow Sea and East China Sea. Atmospheric deposition is an important source of nutrient for the marine ecosystem, and it can facilitate primary production (PP) in the ocean surface and contribute to the development of harmful algal blooms (Paerl and Hans, 1997; Paerl et al., 2002).

Several previous studies have investigated PP over the BYE areas and estimated the historical annual PPs to be 97gC m$^{-2}$ yr$^{-1}$, 236gC m$^{-2}$ yr$^{-1}$ and 145gC m$^{-2}$ yr$^{-1}$, respectively, for Bohai Sea, Yellow Sea and East China Sea (Guan et al., 2005; Gong et al., 2003). In this study, based on the assumption that all $NO_y$ deposited into surface ocean can be absorbed by phytoplankton, we estimate the model averaged PP from $NO_y$ deposition in the historical period over the BYE areas according to the Redfield ratio (Tett et al., 1985). The Redfield ratio refers to the ratios of carbon, nitrogen and phosphorus in phytoplankton listed in equation 6.1. Equation 6.2 are used to calculate PP generated from $NO_y$ deposition, where PPnoy represents the PP from $NO_y$ deposition and $NO_y$ represents total $NO_y$ (wet + dry) deposition.

$$C:N:P=106:16:1 \tag{6.1}$$

$$PP_{noy}=NO_y\times\frac{106}{16} \tag{6.2}$$

Results show that PP from historical $NO_y$ deposition is 5, 5.4 and 4.4 gC $m^{-2}$ $yr^{-1}$ over Bohai Sea, Yellow Sea and East China Sea, accounting for 5%, 2% and 3% of PP in those three seas, respectively. These values are consistent with a previous study reported by Qi et al. (2013), which indicated a contribution of 0.3~6.7% to PP from the nitrogen deposition over the Yellow Sea, and Zhang et al. (2010), who that nitrogen deposition accounted for 1.1~3.9% of PP over East China Sea.

Recently, several studies have evaluated the change of global primary production under future climate change (Steinacher et al., 2010; Koga et al., 2011; Laufkötter et al., 2015; Cabré et al., 2015). For instance, based on multi-model ensembles, Cabré et al. (2015) found a general decrease of total PP projected under RCP 8.5 by the end of this century. We calculate the seasonal PP from $NO_y$ using ACCMIP, with results shown in Fig. 9. Note that PP in Fig. 9 is the equivalent primary production converted from $NO_y$ deposited into the ocean through nutrients uptake by phytoplankton. Due to low sea surface temperature in winter, the conversion can hardly happen and the nutrients may remain until spring (Reay et al., 1999). Therefore, actual PP from $NO_y$ may shift from winter to spring instead. Under RCP scenarios, consistent with the change patterns of total $NO_y$ deposition (not shown), PP decreases significantly over the BYE areas in the 2100s, by 60~68% and 34~63% in the four seasons over the Yellow Sea and East China Sea, respectively, under RCP 8.5 (third and fifth rows in Fig. 9). However, in the 2030s, PP from $NO_y$ shows an increase over the BYE areas under RCP 8.5 (e.g., 32~53% in the Yellow Sea and 19~34% in East China Sea; fourth row in Fig. 9), with smaller

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increase or decrease under RCP 4.5 (second row in Fig. 9). The large increase of $NO_y$ in the near future suggests the increased risk of algal blooms if emission continues to increase, and the reduction in $NO_y$ in 2100 indicates the importance of emission reduction in the long-term. Without emission reduction, PP from $NO_y$ is projected to increase in 2100 during summer (last row in Fig. 9) over the East China Sea, consistent with the wet deposition pattern change depicted in Fig. 6, indicating that climate change increases eutrophication through enhancement of precipitation that increases wet deposition over this region. Hence our results illustrate the importance of reducing emissions on PP in the BYE areas in the future.

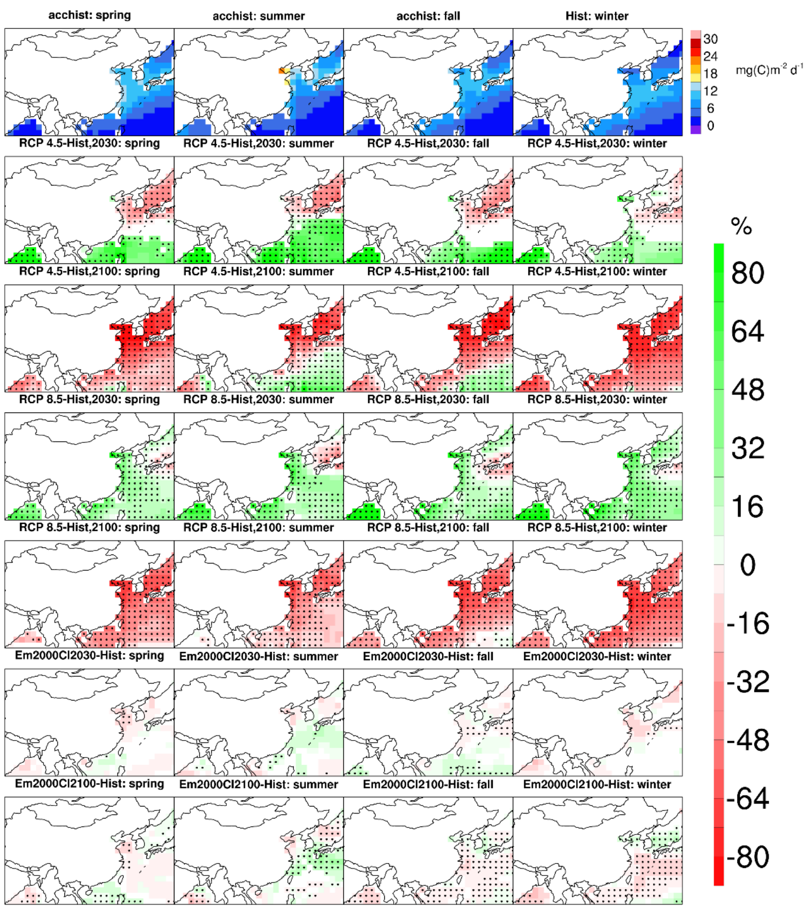


Fig. 9. Spatial distribution of the percentage change of primary production generated from $NO_y$ deposition over east Asia for all scenarios used in this study. Areas over land are blank because the

Redfield ratio used is only application to the ocean areas. From the second row, all distributions are percentage change compared to historical period and only values with agreement are shown. Values with statistical significance (α=0.05) are marked with a black dot.

## 7. Conclusions and discussions

Atmospheric $NO_y$ deposition over East Asia is analyzed to delineate the influence of climate and emission changes based on the ACCMIP multi-model ensemble. Under both RCP 4.5 and RCP 8.5 scenarios with combined effect of climate and emission changes, both dry and wet $NO_y$ deposition shows significant decreases in the 2100s, primarily as a result of large reduction in anthropogenic emissions. In the 2030s, both the dry and wet $NO_y$ deposition increases significantly, particularly under RCP 8.5, mainly because of enhanced emissions. The individual effect of climate change and emissions on the dry and wet $NO_y$ deposition is also identified, showing relatively minor impact of climate change on dry $NO_y$ deposition. In terms of wet deposition, the spatial patterns are in general consistent with those in the changes of precipitation, particularly at the end of this century. Take the East China Sea as an example, wet $NO_y$ deposition increases significantly in summer (18%) and decreases significantly in winter (-13%). While climate change alone generally increases wet deposition, reduction of emission has a dominant influence of reducing wet deposition over East China.

Over the Chinese coastal seas such as Yellow Sea and East China Sea, with decreasing transport of $NO_x$ from mainland China due to emission reduction, ship and lightning emissions from the ocean become the major source of $NO_y$ deposition, with mean seasonal increase of 24-48% and 3%-37% for Yellow Sea and East China Sea, respectively. Therefore, reducing ship emission in the Chinese coastal areas is a key factor to reduce nitrogen deposition in the future.

In the 2030s, PP from $NO_y$ shows increases over the BYE areas under RCP 8.5, suggesting the increased risk of algal blooms if emission such as from ships continues to increase in the near future (Liu et al., 2016). With climate change only, PP from $NO_y$

is projected to increase in 2100 during summer over the East China Sea, indicating a supportive role of climate change on eutrophication, and hence the importance of emission controls.

Although the ACCMIP multi-model ensemble has provided valuable information for projecting future changes in $NO_y$ deposition, the models used in ACCMIP have relatively coarse spatial resolution for resolving the complex meteorological and chemical processes. Dynamical downscaling may be applied in the future to further investigate the impact of climate and emission on nitrogen deposition over East Asia and the detailed processes involved. For analysis of marine primary production, we used a very simple approach that ignores biogeochemical processes in the ocean. An ocean biogeochemistry model will be useful to further quantify the effect of climate and emissions on PP.

**Competing interests.** The authors declare that they have no conflict of interest.

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