Comments from Reviewer 1:

General comments:

This manuscript explores projected simulations of atmospheric oxidized nitrogen deposition to the region of East Asia and adjacent waters of the northwest Pacific Ocean, using the ACCMIP model ensemble, under various scenarios of climate and emissions changes. The study is well constructed and the presentation of the manuscript is clear, logical and thorough. The conclusions of the work (i.e. evolving influences of emission change and changes in precipitation on northwest Pacific marginal seas primary productivity) are interesting and concisely communicated, together with an indication of the limitations of the study. I recommend publication in Atmospheric Chemistry and Physics, with only minor typographic and presentational amendments.

Response: We thank the reviewer for the positive and constructive comments concerning our article. Please see the detailed responses to the specific comments below.

Specific comments:

Line 147. change to “some models”

Response: This has been revised.

Line 296. insert “deposition” after “wet NOy” ?

Response: This has been revised.

Line 307. “change are”, not “change is”.

Response: This has been revised.

Line 422: change to “who reported that nitrogen”?

Response: This has been revised.

Line 434: insert “from NOy” after “PP” ?

Response: This has been revised.

Lines 424 - 447. It may be worth noting in this paragraph that the projected changes in PP from NOy discussed here take no account of potential limitations on PP due to scarcity of other nutrient species

Response: We thank the reviewer for the suggestion and this has been addressed in the revised manuscript (last paragraph of Section 6).

Figures 1 – 6 and 9: These are all good figures, containing a lot of interesting information. However, because they contain a lot of panels, it can be difficult to read the rather small lettering identifying each individual scenario. For the final version of the manuscript I would suggest having a single row of column headings (Spring, Summer, etc.) at the top of each figure and moving the row identifications (e.g. TRMM, GPCP, ACCMIP for Fig. 1) to a single column of labels beside each row.

Response: Thank you for the suggestion for improving the legibility of figures. This has been revised following your suggestion. Figures 1 – 6 and 9 now have a single row of column headings and a single column of labels.

Comments from Reviewer 2:

General comments:

Zhang et al., used the ensemble model outputs from ACCMIP to study the future spatial distributions of total NOy deposition, including wet and dry NOy. They discussed that under the future reductions of anthropogenic emissions, the fractions of the ship emissions, as well as lightning emissions will have relatively important role in contributing the NOy deposition. The authors also estimated the marine primary production form the future NOy depositions. The manuscript is well-written and designed. I suggest to be accepted by ACP with minor revisions.

Response: We thank the reviewer for the constructive comments to help us further improve the manuscript. Please see the detailed responses to the specific comments below.

Specific comments:

In the abstract, I suggest the authors to add the marine primary production projected in the future, as this could be one innovation distinguished from other studies. I will suggest move the sentences from line 61 to line 63 before the discussion of ship and lightning emissions.

Response: We thank the reviewer for the suggestion to highlight the finding. The projected future changes of the marine primary production under RCP scenarios have been addressed in the revised abstract. For the sentences from line 61 to 63, it refers to ship emission. The original manuscript did not specifically point this out and this has been clarified in the revised manuscript.

In section 3, I suggest the authors also add the model evaluations for the NOy deposition in East Asia sine Larmarque et al.,2013 focused on wet NO3 only.

Response: It would be good to add the evaluation for the NO_y deposition. Unfortunately, the observations of EANET does not contain NO_y deposition. Therefore, the precipitation in East Asia was evaluated as it has a dominant influence on wet NO_y deposition.

Also in reporting the future NOy changes under the four scenarios (RCP4.5, RCP8.5, Em2000Cl2030, Em2000Cl20100), I would suggest the authors to add tables listing the standard deviations, considering the multi-model and multi-year averages.

Response: As the reviewer suggested, the standard deviation of regional future NO_y changes over BYE areas for each season (Table S2 for dry NOy and Table S3 for wet NO_y) has been added.

P3 line 73-74: Split up these references so that they are associated with the specific impacts being discussed, rather than all placed at the end of the sentence.

Response: This has been revised following your suggestion.

P3 line 95: Should HNO4 also included in the NOy species?

Response: Right, this has been added in the revised manuscript.

P4 line 107-108: I feel the reference to the ship emissions are out of nowhere. I know the authors discuss heavily on the contributions of future lightning and ship emissions on NOy deposition, but I do not think the authors did a very good job in summarizing the current literature on ship emissions. Instead, line 336-341 should be moved up to the introduction.

Response: We thank the reviewer for bringing up the important point. We have revised the manuscript by moving up line 336-341 to the introduction, and rephrased the sentences as well.

P6 line 173: captured to captures

Response: This has been revised.

In section 5: add a table discussing the emission changes in 2030s and 2100 from the ship and lightning from ensemble models, since the authors were arguing the these two emission sources will have import influence for NOy deposition in the future.

Response: We have added a table (Table S4) summarizing the multi-model mean ship and lightning NO_x emission over Yellow Sea and East China Sea under all scenarios studied. The corresponding discussion has been added in last two paragraphs of section 5, paragraph 7,8.

In Fig 2: Adjust the vertical color bar to cover the plots on second to fifth rows. In row 1, add the region names BS, YS, and ES into the top left plot.

Response: This has been revised. Region names BS, YS and ES have been added into the top left plot with pink colors for names and boxes. The vertical color bar has been adjusted, and this applied to Fig. 3 as well.

In Fig 7&8: I will suggest to change "eminox" for ship to "emisnox"

Response: This has been revised.

Comments from Reviewer 3:

General comments:

In this work, the authors used the multimodel results from the ACCMIP study to investigate the projected changes in dry and wet deposition of oxidized nitrogen compounds (NOy) in 2030–2039 and 2100–2109. This builds on the work of Lamarque et al. (2013), who used the same model results to examine changes in mean nitrogen and sulfur deposition. An important contribution over the past work is the examination of the separate impacts of emission and climate changes on NOy deposition, as opposed to the combined effects, and how the change in NOy deposition could affect the primary productivity (PP) of the eastern China seas. However, unlike the past work, they did not examine the contributions and changes in deposition of reduced nitrogen compounds and are thus missing a significant fraction of the reactive nitrogen deposition budgets. I think this is a missed opportunity to make an important contribution to our understanding of how reactive nitrogen may change in the coming decades. Ammonia emissions have different spatial and temporal patterns from NOx emissions and are likely to increase or remain relatively constant in the coming decades. Therefore, the changes in total inorganic reactive nitrogen deposition and its causes could be quite different compared to NOy deposition and the resulting impacts on PP of China seas. While I do encourage the authors to include reduced nitrogen in their analyses at some level, I think the current work makes enough of a contribution to our understanding of these issues to warrant publication.

Response: We thank the reviewer for the constructive and insight comments. We agree that adding the analysis of reduced nitrogen is meaningful and interesting to elucidate the impact of ammonia emissions. However, the analysis of reduced nitrogen will likely form a separate study and therefore leave this part of analysis in future study. For now, we mainly focus on the oxidized reactive nitrogen deposition. Please see the detailed responses to the specific comments below.

Specific comments:

The figures are small and the text difficult to read. I suggest that column and row headings be added to the tables of maps to allow the reader to more easily follow what is being presented.

Response: We thank the reviewer for the suggestion to improve the quality of the figures. We have modified the texts in the busy figures including Figs. 1-6 and Fig. 9.

Lines 108–109. How do the shipping emissions compare to the other NOx emissions in East Asia?

Response: The contribution of shipping NOx emission has been added in the revised introduction, and they account for nearly 9% of total NO_x emissions in East Asia.

Lines 123–125. It would be interesting to know the areas where the deposition had to increase to compensate for the decreased deposition over the tropics and northern hemisphere midlatitudes.

Response: Discussion here has been revised in order to make it clearer. The deposition over ocean increases, compensating for the decreased deposition over the tropics and Northern hemisphere midlatitudes land areas (due to the decrease of large-scale precipitation) through the transport effect (Line 128-132 in the revised manuscript).

Section 3. Evaluation of the ACCMIP results. Some discussion of the variability in the results across the different models would be very interesting, which is alluded to in the first sentence of section 4.

Response: In the part for evaluation of ACCMIP precipitation, discussion of the inter-model variability has been added in the revised manuscript (before section 4) and Fig. S2 in the supporting information has also been added.

In Figure 5, what does each data point represent? If it’s the changes for a grid cell in each region, then it would be informative to color the data points by region, so that the reader can see the differences in each region.

Response: Yes, each data point represents a grid cell in the BYE areas. We have adjusted the color of points in Fig. 5 with red points indicating grids in BS, blue points for grids in YS and black points for grids in ES.

In Figures 7 and 8, it is not clear what role the changes in shipping and lightning emissions play in the changes in their relative contributions to NOy deposition. Therefore, please add a discussion on the changes in shipping and lightning emissions in the future scenarios examined in this work.

Response: We have revised Fig. 7,8 (add a line for total NO_y deposition) and added a table (Table S4) summarizing the shipping and lightning emissions in historical period and the future scenarios. The corresponding discussion have been added in the revised manuscript section 5, paragraph 7,8.

Lines 421–423. The Zhang et al. and Qi et al. studies examined the total inorganic nitrogen deposition and not just the NOy fraction. Please clarify this in the manuscript and adjust the comparisons as needed.

Response: Thank you for the suggestion. The difference of species used has been clarified in the revised manuscript.

It is not completely clear how the results in Figure 9 were generated. However, since by equation 6.2 PPnoy is proportional to NOy deposition, I would think that the percent changes in PP due to changes in NOy deposition should be the same as the percent change in total NOy deposition. If this is not the case, then please provide a more thorough discussion on how Figure 9 was calculated. If true, then I suggest that Figure 9 be replaced with the percent change in total NOy deposition, which could then include the changes over land. Then, note in the discussion that the percent changes in PP in the eastern China seas are the same as the change in total NOy deposition.

Response: The reviewer is correct that the percent changes in PP due to changes in NOy deposition is the same as the percent change in total NOy deposition. Therefore, based on the reviewer's suggestion, we redrew Fig. 9 with rows 2 – 7 representing change of total NO_y deposition (i.e., cover both land and ocean areas). In addition, the first row remains the PPnoy over the ocean to illustrate the spatial distribution of PP as the base case.

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. Marine primary production from both dry and wet NO_y deposition increases by 19-34% in 2030s and decreases by 34-63% in 2100s over the East China Sea. 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, the seasonal mean contribution of ship emission to 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.~~ human health (~~Doney et al., 2007; Butchart et al., 2010; Stevens et al., 2015;~~ Galloway et al., 2008)~~.~~, biodiversity (Butchart et al., 2010), primary production (Doney et al., 2007; Stevens et al., 2015), etc. 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, HNO_4 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. For NO_x emissions over the oceans, shipping emission is the dominant contributor (Dalsøren et al., 2009; Eyring et al., 2010). Lauer et al. (2007) discussed the significant impact of shipping emissions on aerosols such as aerosol nitrate burden, implying potentially subsequent influence on nitrogen deposition. Fan et al. (2016) concluded that 85% of ship emissions took place within 200km of the coastlines~~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.~~, indicating a stronger influence of ship emissions on coastal seas than remote areas. 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, accounting for nearly 9% of total NO_x emissions in East Asia and 16.5% of global shipping NO_x emissions. In ACCMIP, the NO_x emission over the ocean mainly comes from the shipping, with much smaller amount from aircraft as well as lightning, since lightning NO_x is concentrated in the tropical land areas (Price et al., 1997).

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 land areas of tropics and Northern Hemisphere ~~midlatitudes with reduction in~~mid-latitude due to the decrease of large scale precipitation~~.~~, subsequently enhancing the increase of wet deposition over ~~land.~~the ocean through the transport effect. 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 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~~models 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

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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~~captures 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. To further illustrate the uncertainties among different models, the standard deviation of seasonal mean precipitation across all ACCMIP models over East Asia is shown in Fig. S1, within 1-2 mm/day over Chinese coastal seas.

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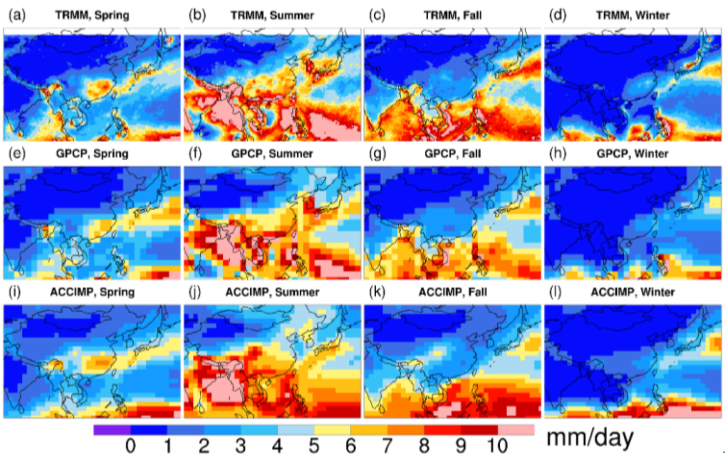


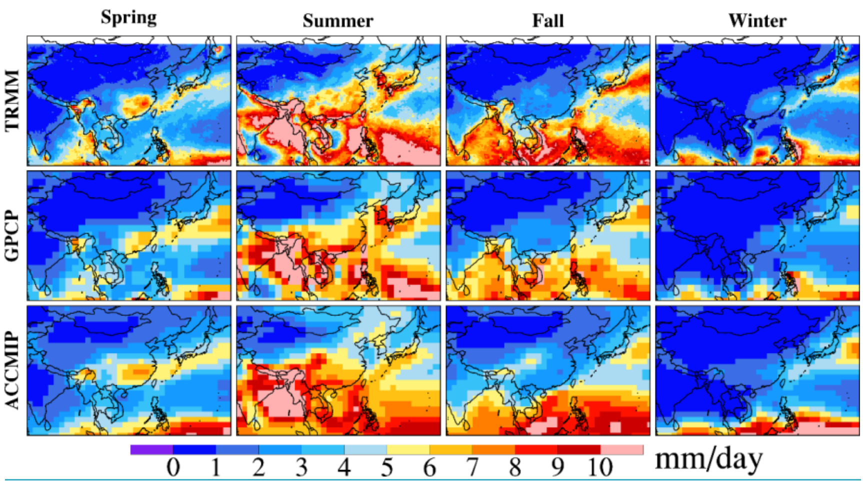


Fig. 1. Evaluation of seasonal mean precipitation during 2001-2010: ACCMIP multi-model ensemble mean vs. TRMM and GPCP

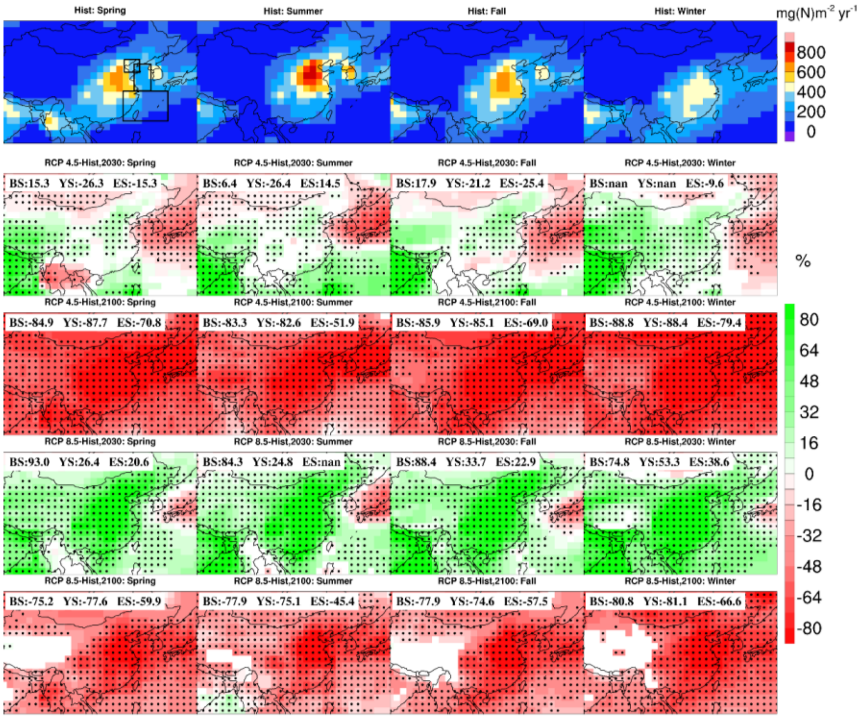
4. Future changes of NO_y deposition in East Asia

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.

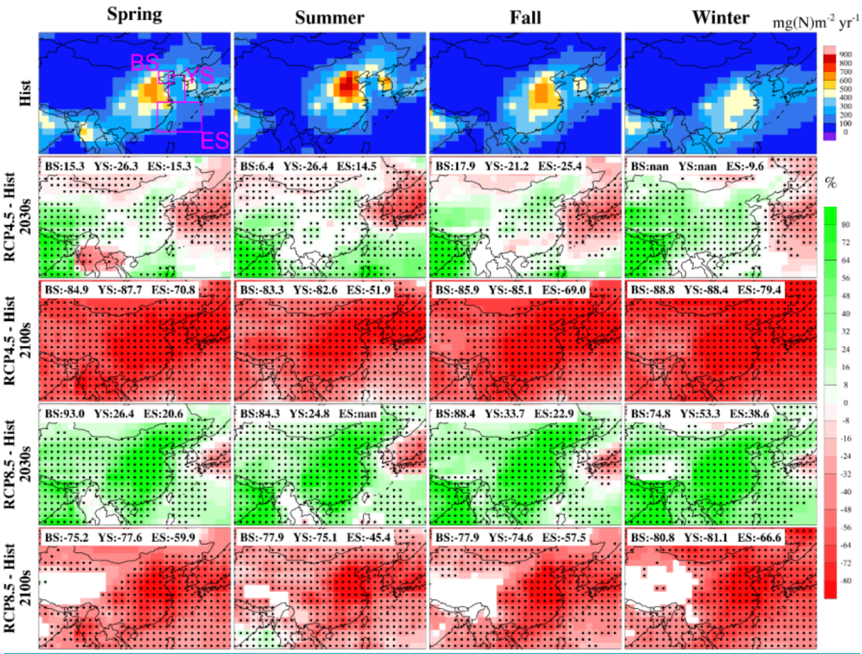
The seasonal mean distribution of dry NO_y deposition over East Asia areas for historical (2001-2010) and projected future changes under RCPs scenarios (RCP 4.5 and RCP 8.5) during the two periods of 2030 and 2100 are shown in Fig. 2. The four seasons defined in this study are spring (March to May), summer (June to August), fall (September to November) and winter (December to February). Regional mean changes over BYE areas are shown in each panel, calculated from multi-model mean results. The corresponding standard deviations of multiple models are shown in Table S2.

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~~S2 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~~S2).



Hist: Spring
Hist: Summer
Hist: Fall
Hist: Winter
mg(N)m-2 yr-1
800
600
400
200
0
RCP 4.5-Hist,2030: Spring
RCP 4.5-Hist,2030: Summer
RCP 4.5-Hist,2030: Fall
RCP 4.5-Hist,2030: Winter
BS:15.3 YS:-26.3 ES:-15.3
BS:6.4 YS:-26.4 ES:14.5
BS:17.9 YS:-21.2 ES:-25.4
BS:nan YS:nan ES:-9.6
%
RCP 4.5-Hist,2100: Spring
RCP 4.5-Hist,2100: Summer
RCP 4.5-Hist,2100: Fall
RCP 4.5-Hist,2100: Winter
BS:-84.9 YS:-87.7 ES:-70.8
BS:-83.3 YS:-82.6 ES:-51.9
BS:-85.9 YS:-85.1 ES:-69.0
BS:-88.8 YS:-88.4 ES:-79.4
RCP 8.5-Hist,2030: Spring
RCP 8.5-Hist,2030: Summer
RCP 8.5-Hist,2030: Fall
RCP 8.5-Hist,2030: Winter
BS:93.0 YS:26.4 ES:20.6
BS:84.3 YS:24.8 ES:nan
BS:88.4 YS:33.7 ES:22.9
BS:74.8 YS:53.3 ES:38.6
RCP 8.5-Hist,2100: Spring
RCP 8.5-Hist,2100: Summer
RCP 8.5-Hist,2100: Fall
RCP 8.5-Hist,2100: Winter
BS:-75.2 YS:-77.6 ES:-59.9
BS:-77.9 YS:-75.1 ES:-45.4
BS:-77.9 YS:-74.6 ES:-57.5
BS:-80.8 YS:-81.1 ES:-66.6
80
64
48
32
16
0
-16
-32
-48
-64
-80



Spring
Summer
Fall
Winter
mg(N)m-2 yr-1
Hist
BS
YS
ES
RCP4.5 - Hist 2030s
BS:15.3 YS:-26.3 ES:-15.3
BS:6.4 YS:-26.4 ES:14.5
BS:17.9 YS:-21.2 ES:-25.4
BS:nan YS:nan ES:-9.6
%
RCP4.5 - Hist 2100s
BS:-84.9 YS:-87.7 ES:-70.8
BS:-83.3 YS:-82.6 ES:-51.9
BS:-85.9 YS:-85.1 ES:-69.0
BS:-88.8 YS:-88.4 ES:-79.4
RCP8.5 - Hist 2030s
BS:93.0 YS:26.4 ES:20.6
BS:84.3 YS:24.8 ES:nan
BS:88.4 YS:33.7 ES:22.9
BS:74.8 YS:53.3 ES:38.6
RCP8.5 - Hist 2100s
BS:-75.2 YS:-77.6 ES:-59.9
BS:-77.9 YS:-75.1 ES:-45.4
BS:-77.9 YS:-74.6 ES:-57.5
BS:-80.8 YS:-81.1 ES:-66.6

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 (α=0.05). Regions of Bohai Sea (BS), Yellow Sea (YS) and East China Sea (ES) are marked by the ~~black~~pink rectangles in the top left panel, with mean changes shown on the top left of each panel starting from the second row. Only grids with significant change in the ocean areas are calculated. 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~~2), with standard deviation of wet deposition shown in Table S3. 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.

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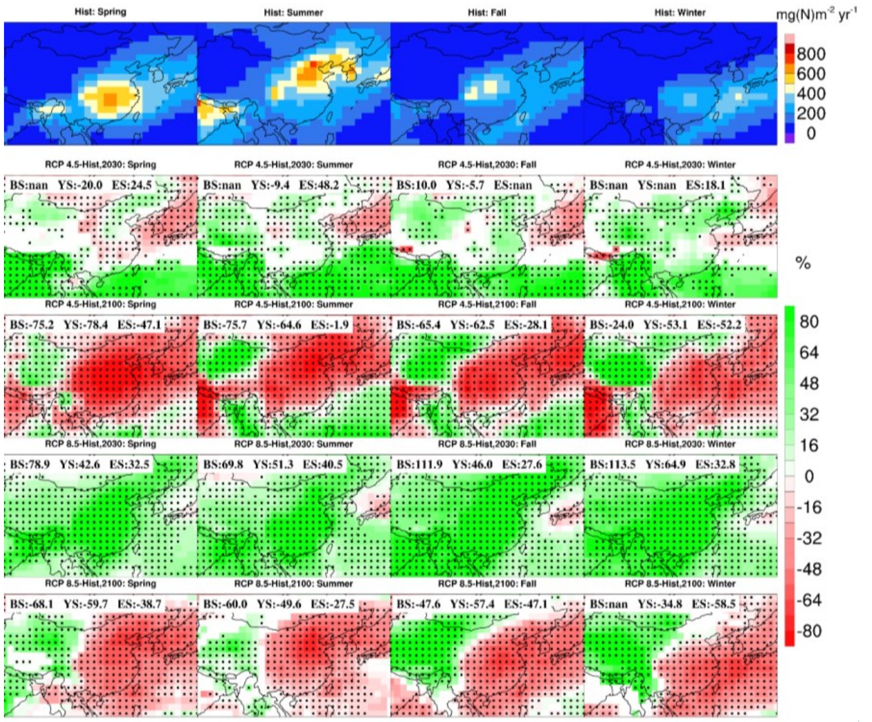
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Hist: Summer
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mg(N)m-2 yr-1
800
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RCP 4.5-Hist,2030: Spring
RCP 4.5-Hist,2030: Summer
RCP 4.5-Hist,2030: Fall
RCP 4.5-Hist,2030: Winter
BS:nan YS:-20.0 ES:24.5
BS:nan YS:-9.4 ES:48.2
BS:10.0 YS:-5.7 ES:nan
BS:nan YS:nan ES:18.1
%
RCP 4.5-Hist,2100: Spring
RCP 4.5-Hist,2100: Summer
RCP 4.5-Hist,2100: Fall
RCP 4.5-Hist,2100: Winter
BS:-75.2 YS:-78.4 ES:-47.1
BS:-75.7 YS:-64.6 ES:-1.9
BS:-65.4 YS:-62.5 ES:-28.1
BS:-24.0 YS:-53.1 ES:-52.2
RCP 8.5-Hist,2030: Spring
RCP 8.5-Hist,2030: Summer
RCP 8.5-Hist,2030: Fall
RCP 8.5-Hist,2030: Winter
BS:78.9 YS:42.6 ES:32.5
BS:69.8 YS:51.3 ES:40.5
BS:111.9 YS:46.0 ES:27.6
BS:113.5 YS:64.9 ES:32.8
RCP 8.5-Hist,2100: Spring
RCP 8.5-Hist,2100: Summer
RCP 8.5-Hist,2100: Fall
RCP 8.5-Hist,2100: Winter
BS:-68.1 YS:-59.7 ES:-38.7
BS:-60.0 YS:-49.6 ES:-27.5
BS:-47.6 YS:-57.4 ES:-47.1
BS:nan YS:-34.8 ES:-58.5
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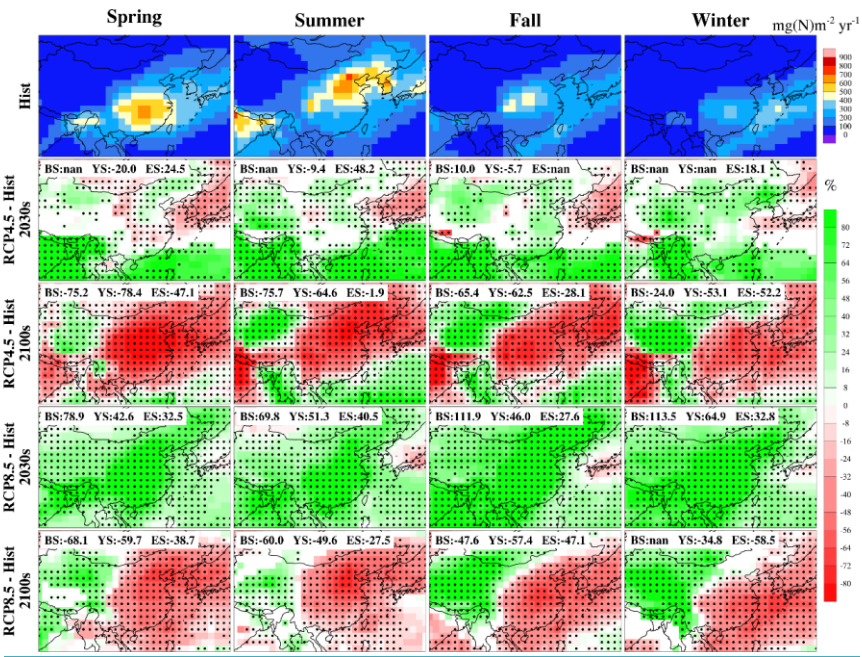
Spring
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Fall
Winter
mg(N)m-2 yr-1
Hist
RCP4.5 - Hist 2030s
RCP4.5 - Hist 2100s
RCP8.5 - Hist 2030s
RCP8.5 - Hist 2100s
BS:nan YS:-20.0 ES:24.5
BS:nan YS:-9.4 ES:48.2
BS:10.0 YS:-5.7 ES:nan
BS:nan YS:nan ES:18.1
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BS:-60.0 YS:-49.6 ES:-27.5
BS:-47.6 YS:-57.4 ES:-47.1
BS:nan YS:-34.8 ES:-58.5
%

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.

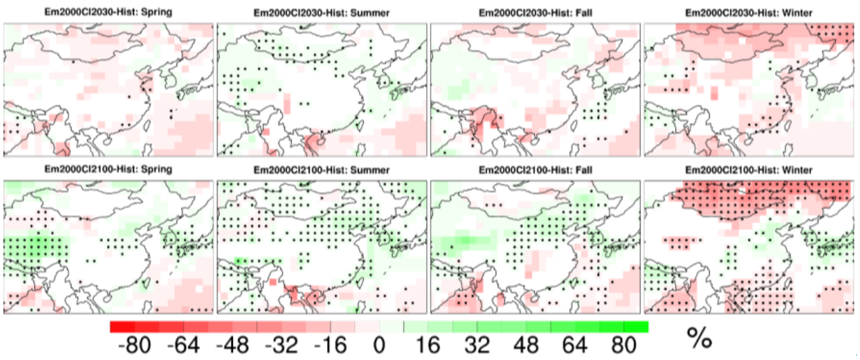
Climate change alone has negligible contributions to the dry NO_y deposition changes, as shown in Fig. 4. Generally, calculation of dry deposition flux in chemical models follows equation 5.1, where F is vertical dry deposition flux, C is concentration of specific gas or particle and v_d is the dry deposition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

$$\sout{F = v_d C} \quad F = -v_d C \qquad (5.1)$$

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,

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

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~~2), indicating statistically insignificant change of r_a under a warmer climate.



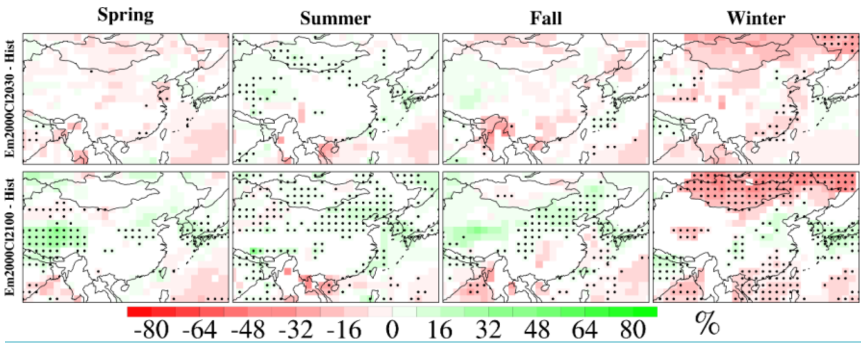


Fig. 4. Spatial distribution of mean seasonal dry NO_y deposition change over East Asia under experimental scenarios of ACCMIP (Em2000Cl2030 and Em2000Cl2100) relative to historical period (2001-2010). The distribution of mean seasonal dry NO_y deposition under historical period is shown in Fig. 2 (top 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).

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 deposition and precipitation in winter under Em2000Cl2100 over East China Sea which will be discussed in detail next.

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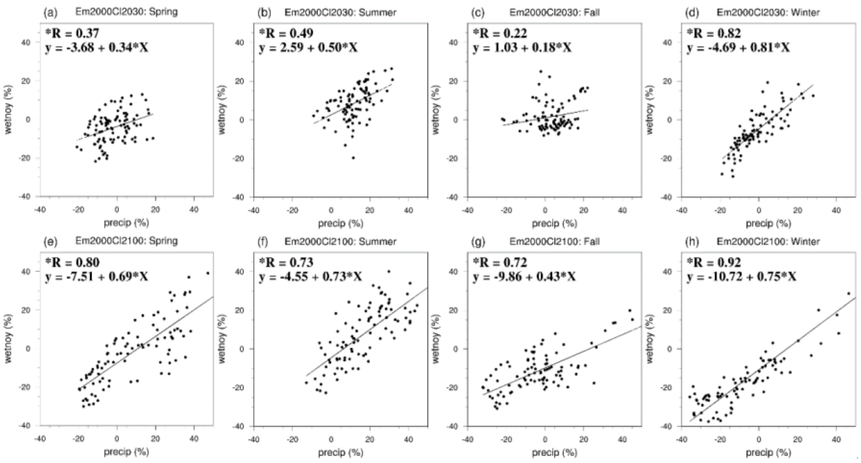


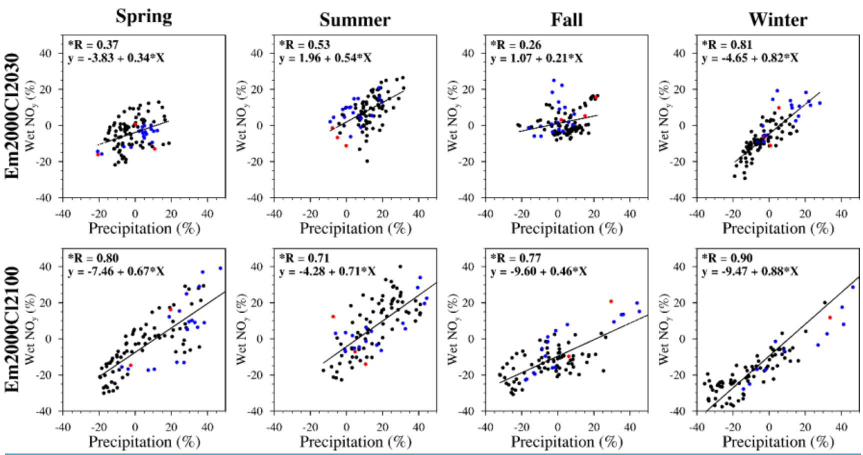
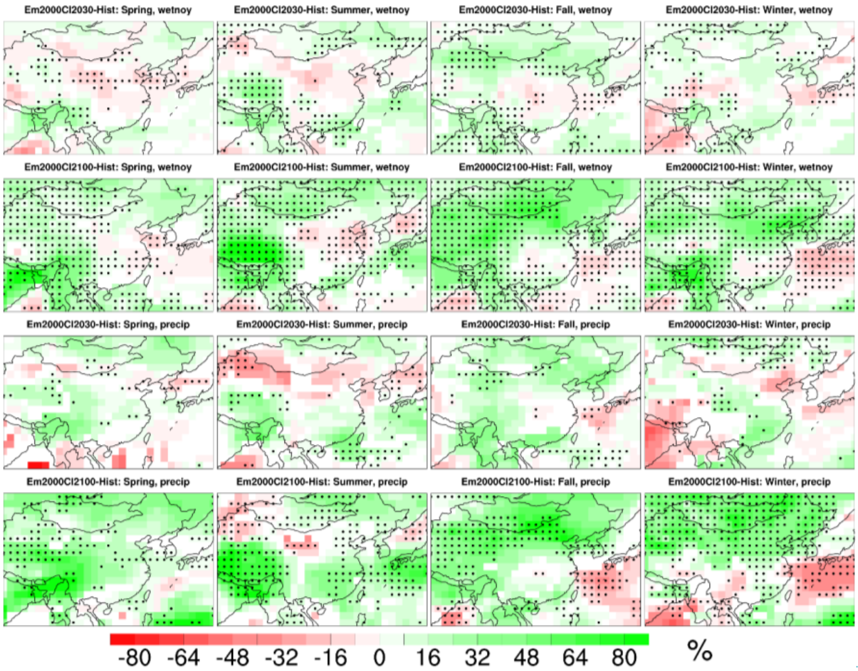


Fig. 5. Comparison between precipitation and wet NO_y deposition changes under the experimental scenarios of ACCMIP (Em2000Cl2030 and Em2000Cl2100) relative to historical period (2001-2010) over Bohai Sea (red points), Yellow Sea (blue points) and East China Sea (black points). An r-test (α=0.05) is 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~~are mostly insignificant (first row) and correspond well with the insignificant 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 21st 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.



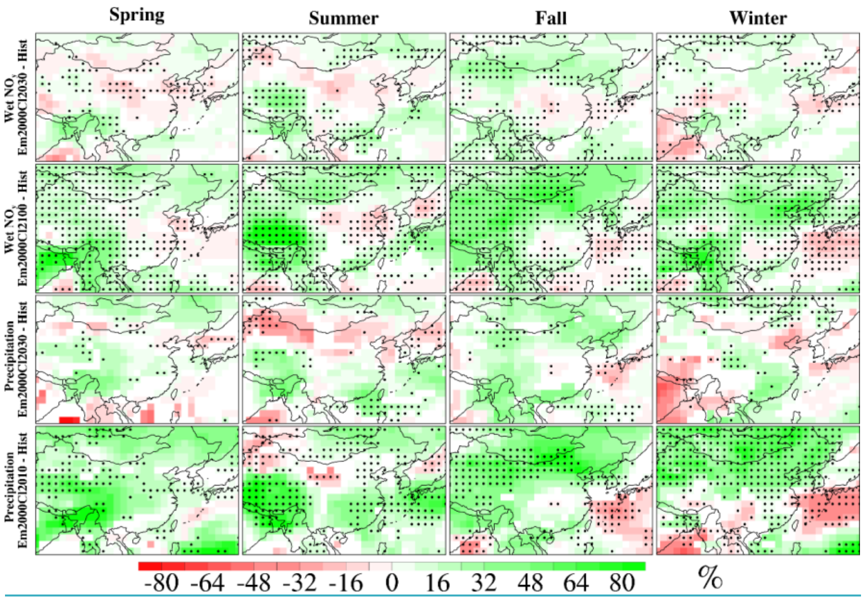


Fig. 6. Spatial distribution of mean seasonal wet NO_y deposition change and precipitation change under EM2000Cl2030 and Em2000Cl2100 relative to historical period (2001-2010). The panels are drawn and arranged in the same manner as Fig. 2.

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~~For a particular region, the NO_y deposition can be considered as the contribution of both local NO_x emission such as shipping and lightning as well as the transport such as from the East Asia continent. Therefore, we calculate the multi-model seasonal mean NO_y deposition and NO_x emissions from shipping and lightning. As was documented~~. 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, indicating the strong effect of ship emission over Chinese coastal seas. 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. ~~. Thus, ship emission may contribute significantly to emissions in the Chinese coastal seas. The~~Summary of shipping and lightning NO_x emission over Yellow Sea and East China Sea under historical, RCP and other scenarios is listed in Table S4. Overall, lightning emission is much smaller than shipping emission. In future, seasonal mean shipping emission increases in 2030s in particular under RCP 8.5 and decreases in 2100s under both RCP scenarios, whereas the changes of lightning emission are small except in summer with mean increase of 73% over Yellow Sea and East China Sea. Based on NO_x emission and NO_y deposition, 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~~First, total NO_y deposition was shown as the green dash line, with all values consistent with the spatial distributions in Figs. 2-4. By the end of this century (2100), total NOy deposition decrease substantially under both RCP 4.5 and RCP 8.5, whereas in 2030s, total deposition shows dramatic increase in RCP 8.5, but moderate change in RCP 4.5 (slight decrease in Yellow sea and increase in East China sea). 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 and Fig. 3 (third and fifth rows), due primarily to emission reduction. Second, ~~with~~albeit the decrease of shipping emission in 2100s, ~~larger emission reduction over land (e.g., eastern China) compared to ocean (Fig. S1),~~ the contribution of ship emission to total NOy deposition ~~over the seas may become larger in future~~increases substantially under both RCP 4.5 and 8.5 over Yellow Sea and East China Sea (black color bars in Figs. 7,8), due primarily to the larger emission reduction over land (e.g., eastern China) compared to ocean (Fig. S2). For instance, over the historical period, the seasonal contribution of ship emission to total NO_y ~~depostion~~deposition 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~~.~~ (orange bars in Figs. 7,8). In particular, due to the reduction in anthropogenic emissions over land and ship emissions in ~~2100~~2100s under RCP 4.5 and RCP 8.5~~,~~ (Fig. S2) along with the increase of lightning NO_x emissions over Chinese coastal seas (Table S4), 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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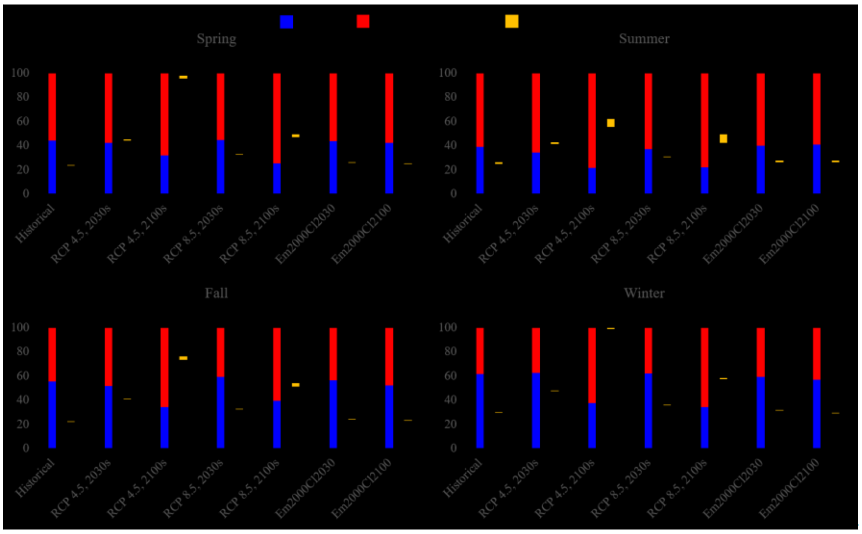

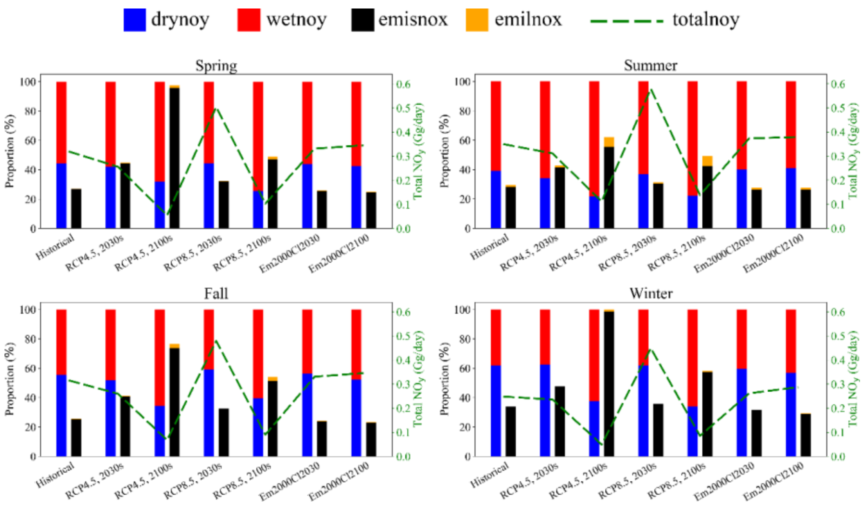

Fig. 7. Stacked bars of seasonal ratio of NO_y deposition from wet (wetnoy) and dry (drynoy)

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deposition and NO_x emissions from ship (~~eminox~~emisnox) 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~~emisnox (black) and emilnox (orange). A green dash line representing total NO_y deposition is added for each panel with y-axis on the right.

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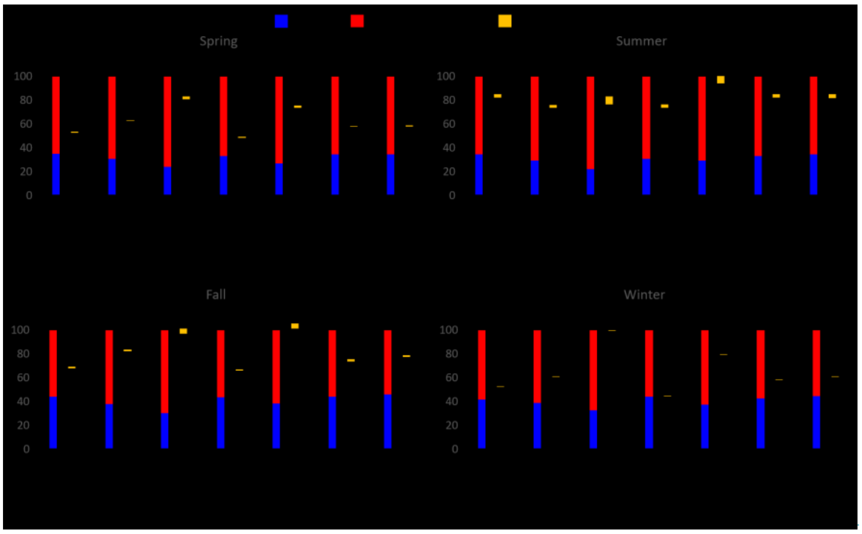


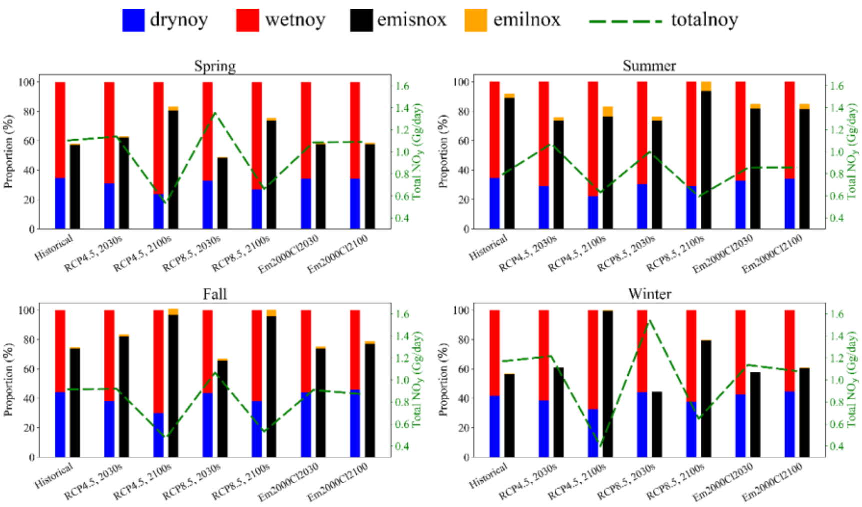


Fig. 8 Same as Fig. 7 except for East China Sea.

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.

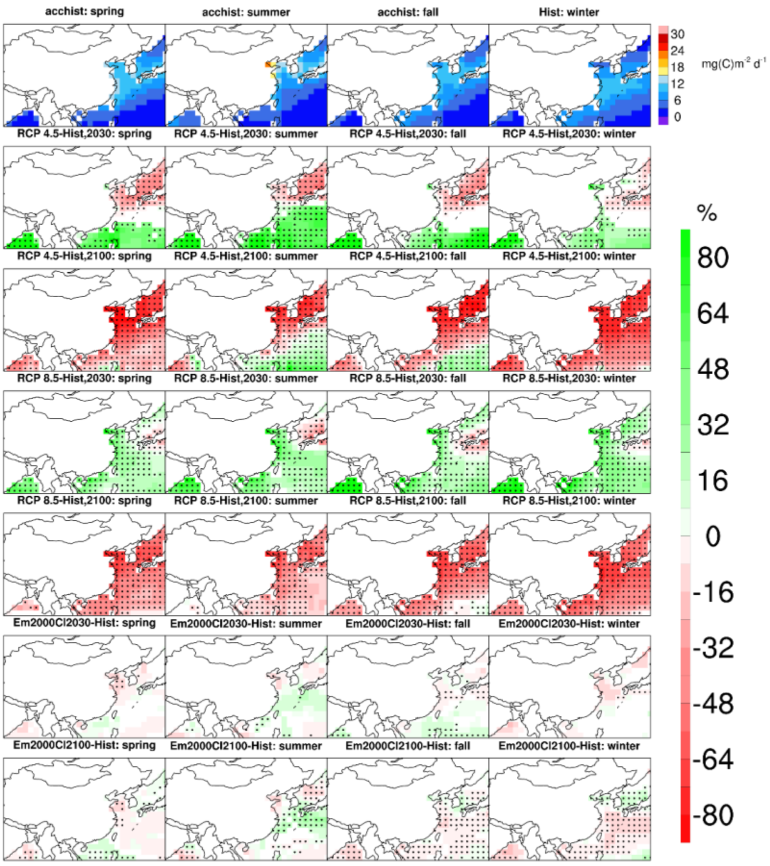
$$C:N:P=106:16:1 \quad C:N:P=106:16:1 \tag{6.1}$$

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

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, albeit of slightly smaller due to the consideration of oxidized nitrogen only in our study, with a previous study reported bystudies. For instance, Qi et al. (2013), which indicated a contribution of 0.3~6.7% to PP from thetotal dissolved nitrogen deposition over the Yellow Sea from July 2005 to March 2006, and Zhang et al. (2010), who found that total inorganic nitrogen deposition accounted for 1.1~3.9% of PP over East China Sea in 2004.

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 global decrease (up to 30%) 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.It should be noticed that panels from the second row in Fig. 9 refer to the percentage changes of total NO_y deposition. According to the Redfield ratio, PP from NO_y is proportional to total NO_y deposition, yielding the same percentage change between PP and NOy. Therefore, the values in the ocean (Fig. 9) also represent the changes of primary production resulted from NO_y deposition. Note that PP in the first row of 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. Moreover, as the Redfield ratio is used to estimate PP from NO_y under all scenarios, potential influence of changes in other nutrients (e.g., carbon and phosphorus) under RCP scenarios and experimental scenarios is not considered. Under RCP scenarios, consistent with the change patterns of total NO_y deposition (not shown), PP from NO_y 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 increase or decrease under RCP 4.5 (second row in Fig. 9). The large increase of NO_y in the near future suggests the

Formatted: Indent: First line: 1.5 ch 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.



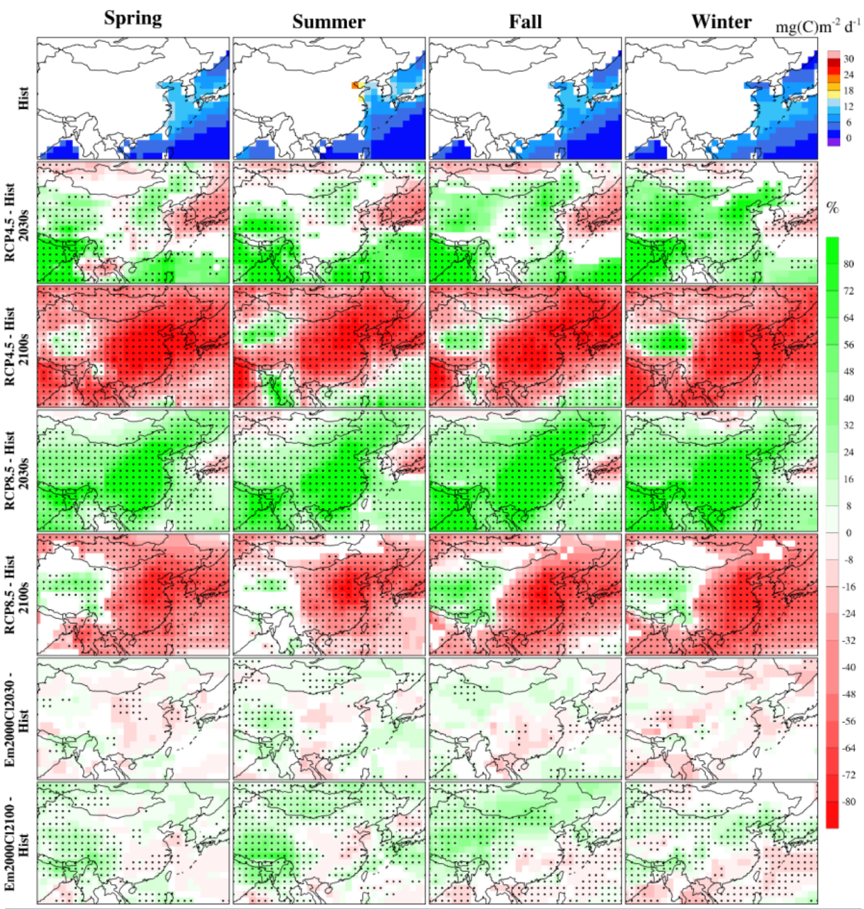


Fig. 9. ~~Spatial~~First row is spatial distribution of ~~the percentage change of~~ marine primary production ~~generated~~resulted from NO_y deposition over ~~east~~East Asia ~~for all scenarios used~~ in ~~this study~~historical periods. Areas over land are blank because the Redfield ratio ~~used~~ is only ~~application~~applied to the ocean areas. The spatial distributions from the second row refer to the percentage change of total NO_y deposition for all RCP scenarios and other scenarios used in this study. Values in the ocean areas can be seen as changes of PP from NO_y based on the definition of the Redfield ratio. 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.

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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.

Acknowledgement. This research was supported by grants from the National Key Project of MOST (2017YFC1404101), Shandong Provincial Natural Science Foundation, China (ZR2017MD026) and National Natural Science Foundation of China (41705124, 41822505 and 91544110). PNNL is operated for DOE by Battelle Memorial Institute under contract DE-AC05-76RL01830.

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