

## 1 On the causes of trends in the seasonal amplitude of atmospheric CO<sub>2</sub>

2  
3 The observed increase in the seasonal amplitude of atmospheric CO<sub>2</sub> in the  
4 northern latitudes reveals major ecological changes that are not captured by terrestrial  
5 ecosystem models participating in the fifth phase of the Coupled Model Intercomparison  
6 Project (CMIP5) (Graven et al., 2013). Here, we used atmospheric CO<sub>2</sub> records from 26  
7 northern hemisphere stations with coverage longer than 15 years, and the LMDZ4  
8 atmospheric transport model prescribed with Net Ecosystem CO<sub>2</sub> Exchange (NEE) from  
9 an ensemble of nine terrestrial ecosystem models, to attribute change in the seasonal  
10 amplitude of atmospheric CO<sub>2</sub>. We found significant ( $P < 0.05$ ) increases in seasonal  
11 peak-to-trough CO<sub>2</sub> amplitude (AMP<sub>P-T</sub>) at 9 stations, and in trough-to-peak amplitude  
12 (AMP<sub>T-P</sub>) at 8 stations over the last three decades. Most of the stations with an  
13 increasing amplitude are located in Arctic and boreal regions ( $>50^{\circ}\text{N}$ ), consistent with  
14 previous observations that the amplitude increased faster at Barrow (Arctic) than at  
15 Mauna Loa (subtropics). The multi-model ensemble mean (MMEM) shows that the  
16 physiological response of ecosystems to rising CO<sub>2</sub> concentration (eCO<sub>2</sub>) and climate  
17 change are dominant drivers of the increase in AMP<sub>P-T</sub> and AMP<sub>T-P</sub> in the high latitudes.  
18 At the Barrow station, the observed increase of AMP<sub>P-T</sub> and AMP<sub>T-P</sub> over the last 33  
19 years is explained by eCO<sub>2</sub> (39% and 42%) almost equally than by climate change (32%  
20 and 35%). The increased carbon losses during carbon release period in response to  
21 eCO<sub>2</sub> are related to an enhancement of ecosystem respiration due to the eCO<sub>2</sub> caused  
22 increase in carbon storage during carbon uptake period. We also found smaller  
23 contributions of air-sea CO<sub>2</sub> fluxes (10% for AMP<sub>P-T</sub> and 11% for AMP<sub>T-P</sub>) and  
24 marginally significant impacts of change in land use (3% for AMP<sub>P-T</sub> and 4% for  
25 AMP<sub>T-P</sub>) at Barrow, highlighting the important role of these factors in regulating change  
26 in seasonal cycle of global carbon cycle, which has been generally ignored by previous  
27 studies.

## 1 **Introduction**

2 As an integrated signal of large scale ecological changes, the change in seasonal  
3 variations of atmospheric CO<sub>2</sub> concentration is an emerging property of the carbon cycle (e.g.  
4 Bacastow et al., 1985; Kohlmaier et al., 1989; Keeling et al., 1996; Randerson et al., 1997;  
5 Piao et al., 2008; Graven et al., 2013; Zeng et al., 2014; Gray et al., 2014; Barlow et al., 2015;  
6 Forkel *et al.*, 2016). The seasonal CO<sub>2</sub> amplitude (AMP) in the lower troposphere has  
7 increased by  $\approx 50\%$  north of 45°N since the 1960s (Graven et al., 2013), and this signal can  
8 only be explained by an increased seasonality of net biome productivity (NBP) in boreal and  
9 northern temperate ecosystems. Yet, there is no consensus on the major factors driving the  
10 increase of NBP and the quantitative contribution of other, smaller fluxes like fossil CO<sub>2</sub>  
11 emissions and air-sea exchange. On the one hand, Gray et al., (2014) and Zeng et al., (2014)  
12 suggested that agricultural improvements contributed to the increase of AMP by increasing  
13 the seasonal NBP uptake in cultivated lands, but there is a two-fold difference in the  
14 estimated contribution of this mechanism in the two studies (range 17%-45% of the  
15 increasing AMP). On the other hand, Randerson et al. (1997) and Forkel et al. (2016)  
16 showed that during the last three decades, most of the increase of amplitude took place at  
17 stations north of 55°N. In this view, agriculture improvement seems unlikely to be the only  
18 driving factor, because croplands are mainly in northern temperate latitudes (Foley et al.,  
19 2005). Using the LPJmL carbon cycle model with an improved phenology module coupled  
20 with the TM3 atmospheric transport model, Forkel et al. (2016) found that it is mainly the  
21 physiological response of northern plants to warming rather to increasing CO<sub>2</sub> that explains  
22 the trend of AMP over the last 20 years, which is partly inconsistent with the observation  
23 made by Graven et al. (2013) that AMP increased in the 1960s to the mid-1970s at a time  
24 when northern temperature slightly decreased, highlighting the need to search deeper in the  
25 attribution of the AMP trend.

26 In this paper, we investigate the AMP trend in the northern hemisphere over the last  
27 thirty years (1980-2012) using an ensemble of ecosystem models with different  
28 parameterizations of the effect of elevated CO<sub>2</sub> and climate change (TRENDYv2) (Sitch et  
29 al., 2015) with another transport model (LMDZ4) (Hourdin et al., 2013). We also separate  
30 the contribution of fossil fuel CO<sub>2</sub> emissions, air-sea fluxes as well as the effect of climate

1 change, rising CO<sub>2</sub> (eCO<sub>2</sub>), nitrogen deposition in some models and land use change on the  
2 trends in the seasonality of land ecosystem exchange. The contribution of atmospheric  
3 transport trends to AMP trends is further analyzed. Trends in seasonal atmospheric CO<sub>2</sub> from  
4 26 northern (north of 23°N) long-term atmospheric stations (>15 years in the period of  
5 1980-2012) of the NOAA-ERSL surface flask air-sampling network (Table S1 and Figure S1)  
6 are used.

7

## 8 **Results and Discussion**

9

### 10 **Observed CO<sub>2</sub> data**

11 The 26 northern (north of 23°N) atmospheric stations selected are shown in Figure S1  
12 and Table S1. According to the shape of detrended CO<sub>2</sub> seasonal cycle (Thoning et al., 1989,  
13 see methods) (Figure S2), we divided the amplitude into peak-to-trough (AMP<sub>P-T</sub>, defined as  
14 the difference between the peak value and the trough value of CO<sub>2</sub> seasonal cycle in a year)  
15 and trough-to-peak (AMP<sub>T-P</sub>, defined as the difference between the trough value of CO<sub>2</sub>  
16 seasonal cycle in a year and the peak value of the cycle in the next year). As shown in Figure  
17 1a, a significant ( $P < 0.05$ ) positive trend of AMP<sub>P-T</sub> ranging from 0.05 to 0.15 ppm yr<sup>-1</sup> is  
18 found at 9 stations, 8 of them being north of 50°N. The other stations do not show significant  
19 positive AMP<sub>P-T</sub> trends and 5 stations show negative trends (the latter being significant at  
20 only one station UUM). The significant increase in AMP<sub>P-T</sub> reflects mainly an increasing  
21 CO<sub>2</sub> drawdown (defined by the monthly net CO<sub>2</sub> concentration change) in June and July  
22 (Figure S3).

23 The trends in AMP<sub>T-P</sub> reported in Table S1 are similar to those of AMP<sub>P-T</sub>, logically  
24 expected because we remove a long-term mean trend of each CO<sub>2</sub> time series (Figure. 1b). In  
25 total, 7 out of 8 stations with a significant ( $P < 0.05$ ) increase of AMP<sub>T-P</sub> during 1980-2012  
26 are located north of 50°N. The months of September and October are those during which  
27 most of the negative trend of AMP<sub>T-P</sub> occurs at those sites (Figure S3). Overall, no sties  
28 show significant positive trend in AMP<sub>T-P</sub> during the study period.

29

### 30 **Terrestrial ecosystem output and simulation of CO<sub>2</sub> amplitude trends**

1 The Net Ecosystem CO<sub>2</sub> Exchange (NEE) from Eight dynamic global vegetation models  
2 (DGVMs) from TRENDYv2 (Le Quéré et al., 2014; Sitch et al., 2015) (Table S2) was  
3 prescribed to the atmospheric transport model (LMDZ4) (Hourdin et al., 2013, See Methods).  
4 Time-varying monthly NEE from each model under simulation S3 (driven by CO<sub>2</sub>, climate  
5 change and land-cover change) (Sitch et al., 2015), fossil fuel and cement emissions (Andres  
6 et al., 2011; Boden et al., 2016), and interannual air-sea fluxes (Buitenhuis et al., 2010) were  
7 prescribed to the global LMDZ4 transport model (Hourdin et al., 2013) with variable winds  
8 over the last 33 years. This is the T1 simulation (see Methods and Table 4), from which the  
9 modeled CO<sub>2</sub> concentration field was sampled at each station and analyzed for amplitude  
10 changes just like in the observed time series.

11 Most of T1 simulations results (except with the ISAM and JULES ecosystem models)  
12 produce a significant increase in AMP<sub>P-T</sub> at boreal (north of 50°N) stations (Figure 1a),  
13 though there are differences among models. In comparison with the observed average trend  
14 ( $0.094 \pm 0.033$  ppm yr<sup>-1</sup>) of AMP<sub>P-T</sub> at the 8 boreal stations with a significant increase in  
15 AMP<sub>P-T</sub>, three models show a larger AMP<sub>P-T</sub> positive trend (CLM4.5:  $0.105 \pm 0.046$  ppm yr<sup>-1</sup>;  
16 LPJ:  $0.101 \pm 0.053$  ppm yr<sup>-1</sup>; VISIT:  $0.101 \pm 0.059$  ppm yr<sup>-1</sup>). At the three boreal stations  
17 with no significant trend in observed AMP<sub>P-T</sub> (BAL, MHD, SHM in Figure 1a), the T1  
18 simulation results also correctly reproduce no trend (Figure 1a) except for ORCHIDEE at  
19 MHD and VISIT at SHM.

20 Similar to trends in AMP<sub>P-T</sub>, statistically significant increasing trough-to-peak CO<sub>2</sub>  
21 amplitude (AMP<sub>T-P</sub>) is found in the T1 simulation results (except again for ISAM and  
22 JULES), consistent with the observed trends. For the simulations with ISAM and JULES,  
23 temperate sites have more statistically significant increasing trends of AMP<sub>T-P</sub> than boreal  
24 and Arctic sites.

25 Overall, unlike previous studies that have shown a systematic underestimation of AMP  
26 trend by ecosystem models, namely the CMIP5 models (Taylor et al., 2012) and the  
27 MsTMIP models (Huntzinger et al., 2013; Wei et al., 2014) at high northern latitudes  
28 (Graven et al., 2013; Thomas et al., 2016), we found both underestimation and  
29 overestimation of AMP trends from the TRENDYv2 models (Figure S4). This phenomenon  
30 can be related to different climate forcing (between CMIP5 and other ensembles), partly

1 different terrestrial ecosystem models, and the transport simulation with a different transport  
2 model (LMDZ4 here instead of TM3 and ACTM in Graven et al. (2013) and TM3 in Thomas  
3 et al. (2016)).

#### 4 5 **Effects of different factors on the trend in AMP<sub>P-T</sub>**

6 In order to separate the contribution of different driving factors on the trend of AMP<sub>P-T</sub>,  
7 we performed transport simulations with Net Ecosystem CO<sub>2</sub> Exchange (NEE) changes  
8 caused by different factors from factorial runs of the TRENDYv2 models, respectively with  
9 variable CO<sub>2</sub> only (eCO<sub>2</sub>), variable CO<sub>2</sub> and climate, and variable CO<sub>2</sub>, climate and land  
10 cover change (Table S4, see Methods). To further separate the contribution of atmospheric  
11 transport trends to AMP trends, the LMDZ4 transport model was used with variable transport  
12 fields (Dee et al., 2011) but constant NEE, air-sea CO<sub>2</sub> flux and fossil fuel and cement  
13 emissions of the year 1979, so that trends in AMP from this simulation can be attributed to  
14 transport trends only.

15 The impact of climate change on NEE affecting AMP<sub>P-T</sub> trends estimated from  
16 multi-model ensemble mean (MMEM) varies among stations (Figure 2a). We find a positive  
17 trend of AMP induced by climate change at boreal atmospheric stations (8 of 11 stations  
18 north of 50°N (Figure. 2a and S5b). On average, climate change caused an enhancement of  
19  $0.015 \pm 0.025$  ppm yr<sup>-1</sup> in AMP<sub>P-T</sub> over boreal region (north of 50 °N) (Figure 3a), which is  
20 about 20% of the observed AMP<sub>P-T</sub> trend. It has been suggested that warmer temperature is  
21 associated with higher vegetation productivity of boreal ecosystems in spring through  
22 advancing vegetation green-up date (Keeling et al., 1996), although such positive effect may  
23 saturate (Piao et al., 2014; Fu et al., 2015). As shown in Figure S6a, for the BRW site (71 °N),  
24 the effect of climate change on AMP<sub>P-T</sub> is positive mainly during May and June.

25 By contrast at the temperate sites (in the band of 23-50 °N), the effect of climate change  
26 on the AMP<sub>P-T</sub> trends is mainly negative (10 of the 15 stations), although the impact is not  
27 statistically significant (except TAP significant at P<0.05 and ASK marginally significant  
28 at P<0.1). On average, climate change is modeled to cause a decrease of AMP<sub>P-T</sub> of  
29  $-0.012 \pm 0.040$  ppm yr<sup>-1</sup> at stations in the temperate band (Figure 3a). Analysis of NEE  
30 impacted by climate change (Trendy models S2 – S1 simulations) shows that climate change

1 alone caused a decrease in CO<sub>2</sub> uptake from April to August over western and central US, in  
2 Eastern Europe, northeast China and Mongolia (Fig. S7b), associated with declining soil  
3 moisture driven by rising temperature and decreasing precipitation in these regions (Sitch et  
4 al., 2015).

5 In the simulations of CO<sub>2</sub> with MEM, eCO<sub>2</sub> causes a statistically significant increase  
6 in AMP<sub>P-T</sub> at 10 of the 11 boreal stations (Figure 2a), and the magnitude of trend in AMP<sub>P-T</sub>  
7 driven by eCO<sub>2</sub> ( $0.036 \pm 0.005$  ppm yr<sup>-1</sup>) is about twice as large as that caused by climate  
8 change (Figure 3a). This larger effect of eCO<sub>2</sub> than climate change on AMP<sub>P-T</sub> trends in  
9 boreal zone is also present in simulations with individual ecosystem model NEE (Figure S5a  
10 and b). This result do not support previous findings by the study of Forkel et al. (2016)  
11 which suggest a more prominent signal of climate change than eCO<sub>2</sub> in the observed  
12 increase in AMP<sub>P-T</sub> in high northern latitudes. However, we agree that climate change rather  
13 than eCO<sub>2</sub> causes the latitudinal difference of trend in AMP<sub>P-T</sub>. As shown in Figure 3a, we  
14 found that the magnitude of eCO<sub>2</sub> effect to increase the trend of AMP in temperate regions  
15 ( $0.028 \pm 0.023$  ppm yr<sup>-1</sup>) is comparable to that in boreal regions, although less stations (9 of  
16 15) show statistically significant effect (Figure 2a). It should be noted that four TRENDY  
17 models (CLM4.5, ISAM, LPX and OCN) considered carbon-nitrogen interactions and  
18 nitrogen deposition, thus the signal of eCO<sub>2</sub> derived from these models includes the  
19 interactive effect of nitrogen deposition. Another simulation with nitrogen deposition using  
20 CLM4 model (Oleson et al., 2010; Mao et al., 2013) (see Methods) predicts that, however,  
21 the effect of nitrogen deposition on AMP<sub>P-T</sub> trend is not significant ( $P < 0.05$ ) at all sites  
22 (Figure S8a), while this result depends on individual model parameterizations (Galloway *et*  
23 *al.*, 2008). Further studies based on multiple models with C-N interactions are needed.

24 Both forest inventory data and model simulation show that afforestation and forest  
25 regrowth after abandonment of agriculture in northern ecosystems have an important role in  
26 the regional and global carbon balance (Pan et al., 2011; Houghton et al., 2015; FAO, 2015).  
27 As shown in Figure S7c, most TRENDYv2 DGVMs (except ISAM) predict that land use  
28 change enhance net carbon uptake during the period from April to August in Eastern Europe,  
29 China and central and eastern United States. Accordingly, a significant ( $P < 0.05$ ) or  
30 marginally significant ( $P < 0.10$ ) positive effect of LUC on the trend in AMP<sub>P-T</sub> is predicted

1 across 6 boreal sites and 3 northern temperate sites (Figure 2a and S5c), although the  
2 magnitude of signal is generally smaller than the effect of eCO<sub>2</sub> and climate change. Overall,  
3 the positive increase of AMP<sub>P-T</sub> attributed to land use change is similar between boreal  
4 region (0.007±0.009 ppm yr<sup>-1</sup>) and northern temperate region (0.004±0.008 ppm yr<sup>-1</sup>) (Figure  
5 3a), suggesting that the latitudinal difference in observed AMP<sub>P-T</sub> increase (0.07±0.05 ppm  
6 yr<sup>-1</sup> in boreal zone and 0.01±0.05 ppm yr<sup>-1</sup> in temperate zone) has little linkage with land use  
7 change. It should be noted that, however, there are still large uncertainties in estimating  
8 land-use-change effect on AMP<sub>P-T</sub> trend, primarily owing to processes of land use change  
9 and management not considered in some Trendy models (e.g., wood harvest, shifting  
10 cultivation, peat fires) (Table S3) and the lack of some critical processes (e.g., human  
11 settlement, erosion/redeposition, woody encroachment) in all models (Houghton et al.,  
12 2012).

13 Over the past thirty years, global CO<sub>2</sub> emissions from fossil fuel consumption have  
14 increased from 5.3 Pg C yr<sup>-1</sup> in 1980 to 9.7 Pg C yr<sup>-1</sup> in 2012 (Boden et al., 2016, Figure  
15 S9a). However, the pattern of change is not spatially uniform in northern hemisphere. A  
16 substantial significant increase in annual fossil fuel CO<sub>2</sub> emissions is found over northern  
17 temperate region, whereas a significant decline is found in boreal region (Figure S9a). This  
18 heterogeneity is also found in the period of April to August, during which AMP<sub>P-T</sub> is  
19 calculated for most northern temperate and boreal stations (Figure S9b). As a result, effect of  
20 changes in fossil fuel carbon emissions on the trend in AMP<sub>P-T</sub> is opposite between  
21 temperate and boreal sites, although most sites show non-significant trend in AMP<sub>P-T</sub> caused  
22 by trends of fossil CO<sub>2</sub> emissions. As shown in Figure 2a, a negative effect of fossil fuel  
23 emissions on AMP<sub>P-T</sub> trend is simulated at temperate sites (13 of 15 sites show negative  
24 trend with 3 significant sites and 1 marginally significant site) (Figure 2a), and a positive  
25 effect at most boreal sites (8 of 11 sites). The absolute value of the AMP<sub>P-T</sub> trend associated  
26 with fossil fuel emissions is generally larger at temperate (average of -0.013±0.022 ppm yr<sup>-1</sup>)  
27 compared to boreal sites (average of 0.003±0.007 ppm yr<sup>-1</sup>) (Figure 3a).

28 A recent study (Horton et al., 2015) demonstrated robust trends in sub-seasonal  
29 atmospheric circulation patterns over mid-latitude regions during 1979-2013, particularly in  
30 summer and autumn. Such changes in the large-scale atmospheric circulation may exert an

1 effect on the trend of CO<sub>2</sub> amplitude. As shown in Figure 2a, although only two stations  
2 (UUM and IZO) show statistically significant impact of transport trends on the trend of  
3 AMP<sub>P-T</sub>, the magnitude of transport caused AMP<sub>P-T</sub> trend is comparable or even larger than  
4 the effect of climate change and eCO<sub>2</sub> on NEE at some atmospheric stations, particularly in  
5 the temperate zone. For example, at UUM, the observed decrease in AMP<sub>P-T</sub> is primary  
6 caused by change in wind (Figure 2a). In terms of effects air-sea fluxes on the trend of  
7 AMP<sub>P-T</sub>, a weak contribution to AMP trends was simulated across most of sites except at  
8 BRW (0.010 ppm yr<sup>-1</sup>, P<0.05, 10% of the observed trend) and MBC (0.015 ppm yr<sup>-1</sup>, P<0.1,  
9 16% of the observed trend).

10 The mechanisms driving the trend in AMP<sub>P-T</sub> are here analyzed with observations at the  
11 Arctic station of BRW (71°N), the longest northern high latitude CO<sub>2</sub> record showing an  
12 increase of amplitude of 35% since 50 years, larger than at the Mauna Loa longest record  
13 located in the sub-tropics (e.g. Graven et al., 2013; Zeng et al., 2014; Gray et al., 2014;  
14 Barlow et al., 2015; Forkel *et al.*, 2016). Our transport simulations with multi-model  
15 ensemble mean (MMEM) NEE produce that AMP<sub>P-T</sub> at the BRW site significantly increased  
16 by about 0.095 ppm yr<sup>-1</sup> from 1980 to 2012, which is comparable with the observed trend of  
17 0.097 ppm yr<sup>-1</sup> (Figure 1a). eCO<sub>2</sub> is identified as the largest contributor of increasing AMP<sub>P-T</sub>  
18 with a trend of 0.039 ppm yr<sup>-1</sup> (40% of the observed trend, P<0.05), followed by climate  
19 change with a trend of 0.031 ppm yr<sup>-1</sup> (32% of the observed trend, P<0.05) (Figure S5a and  
20 b). The effect of ocean flux is of 0.010 ppm yr<sup>-1</sup> (10% of observed trend, P<0.05), and land  
21 use change has marginally significant contributions (0.003 ppm yr<sup>-1</sup> and 3% of observed  
22 trend, P<0.1) (Figure S5c and e). For other factors such as fossil fuel emissions and transport  
23 (Figure S5d and f), non-significant impacts on AMP<sub>P-T</sub> trend were produced.

24

### 25 **Effects of different factors on trend in AMP<sub>T-P</sub>**

26 We also assessed the effect of different factors on trend in AMP<sub>T-P</sub> with the same NEE  
27 and transport model simulation results (See Methods). In contrast to carbon uptake period,  
28 climate change accelerates carbon release from boreal ecosystems during the non-carbon  
29 uptake period. As shown in Figure 2b, an increasing AMP<sub>T-P</sub> (a negative trend in AMP<sub>T-P</sub>  
30 indicates a larger release) is simulated at 8 of the 11 boreal sites (1 site significant at P<0.05;



1 2 sites marginally significant at  $0.05 < P < 0.1$ ). In contrast, a decreasing  $AMP_{T-P}$  (shown with  
2 positive trend) is produced at 12 of 15 temperate sites (1 site significant at  $P < 0.05$ ; 1 site  
3 marginally significant at  $0.05 < P < 0.1$ ) (Figure 2b). It has been suggested that autumn  
4 warming enhance vegetation productivity through delaying vegetation senescence, as well as  
5 accelerate ecosystem respiration (Piao et al., 2008; Vesala et al., 2010). Therefore, the  
6 opposite effect of climate change on the trend of  $AMP_{T-P}$  in boreal region ( $-0.016 \pm 0.027$   
7  $\text{ppm yr}^{-1}$ ) and temperate region ( $0.011 \pm 0.040 \text{ ppm yr}^{-1}$ ) (Figure 3b) is probably due to  
8 different magnitude of the response of vegetation productivity (GPP) and ecosystem  
9 respiration (TER) to climate change. Indeed, the model results show that the rising  
10 temperature induced increase of TER is greater than that of GPP in high northern latitudes,  
11 whereas the increase of GPP is larger in temperate regions (Figure S10).

12 Atmospheric  $CO_2$  simulations from MMEM NEE produce an increasing  $AMP_{T-P}$  in  
13 response to  $eCO_2$  at 25 of 26 temperate and boreal sites (19 sites significant at  $P < 0.05$ ; two  
14 sites marginally significant at  $0.05 < P < 0.1$ , Figure 2b). NEE from 6 out of 8 terrestrial  
15 ecosystem models (except ISAM and JULES) also produces an enhancing  $AMP_{T-P}$  from  
16  $eCO_2$  (Figure S11a). This result indicates an acceleration of carbon release during the  
17 non-carbon uptake period as an indirect effect of the NEE response to  $eCO_2$ . This is due to  
18 the increment in carbon storage caused by the enhancement of net carbon uptake during the  
19 carbon uptake period under the effect of  $eCO_2$ , which stimulates ecosystem respiration  
20 during the non-carbon uptake period (Figure S12). Similar to the contribution of land use  
21 change to the trends of  $AMP_{P-T}$ , we also found statistically significant enlargement of  
22  $AMP_{T-P}$  in response to land use change at 9 of 26 sites (Figure 2b).

23 Similar to the effect on  $AMP_{P-T}$ , the contribution of fossil fuel  $CO_2$  emissions, air-sea  
24 fluxes and transport on the trend of  $AMP_{T-P}$  is only statistically significant at a minority of  
25 sites (only 1, 4 and 2 sites significant at  $P < 0.05$  for the effect of fossil fuel, air-sea fluxes and  
26 transport, respectively) (Figure 2b). However, the magnitude of signal induced by transport  
27 and fossil fuel emissions is generally remarkable over temperate region (Figure 3b), causing  
28 an average impact of  $-0.014 \pm 0.036 \text{ ppm yr}^{-1}$  and  $0.010 \pm 0.014 \text{ ppm yr}^{-1}$  in the trend of  
29  $AMP_{P-T}$ , respectively.

30 Overall, the observed significant enlargement of  $AMP_{T-P}$  at the BRW site ( $-0.090 \text{ ppm}$

1 yr<sup>-1</sup>) is mainly driven by eCO<sub>2</sub> (-0.038 ppm yr<sup>-1</sup> and 42% of observing trend, P<0.05),  
2 climate change (0.032 ppm yr<sup>-1</sup> and 35% of observing trend, P<0.05), ocean flux change  
3 (-0.010 ppm yr<sup>-1</sup> and 11% of observing trend, P<0.05) and land use change (-0.003 ppm yr<sup>-1</sup>  
4 and 4% of observing trend, P<0.05).

5

## 6 **Conclusion**

7 It is difficult to attribute trends in seasonal atmospheric CO<sub>2</sub> concentration both because  
8 of regionally different contributions, and because terrestrial ecosystem models lack  
9 mechanisms or did not report separately disturbance contributions and nitrogen deposition  
10 contributions. Unlike previous studies based on one model only (Zeng et al., 2014; Forkel et  
11 al., 2016), our results based on an ensemble of models to capture the amplitude trends suggest  
12 that rising atmospheric CO<sub>2</sub> concentration is the primary driver of enhancement of both  
13 AMP<sub>P-T</sub> and AMP<sub>T-P</sub>, although climate change plays a critical role and contributes largely to  
14 the latitudinal difference of AMP trend. In addition, the effect of other factors such as land  
15 use change, fossil fuel emissions, ocean flux, and transport on the trend in AMP<sub>P-T</sub> and  
16 AMP<sub>T-P</sub> is not statistically significant at most of stations, but still large enough to cancel out  
17 the effect of eCO<sub>2</sub> at some temperate sites where the observing seasonal CO<sub>2</sub> trends are small.  
18 However, the large uncertainties in the forcing data on land use change and fossil fuel  
19 emission at the moment do not allow an unequivocal statement on the contribution of these  
20 factors, and further studies based on spatially and temporally explicit historic data sets  
21 including land use and fossil fuel emission are needed. Finally, we found that rising  
22 atmospheric CO<sub>2</sub> concentration has opposite implication in the northern ecosystem carbon  
23 balance between carbon uptake period (trend of AMP<sub>P-T</sub>) and non-carbon uptake period (trend  
24 of AMP<sub>P-T</sub>), due to the lagged effects of increase in carbon storage during carbon uptake  
25 period on carbon cycle in non-carbon uptake period. Our results not only provide insights for  
26 large-scale field experiments, but also highlight the importance of understanding the carbon  
27 releasing processes during the non-growing season, which is critical for reliable projection of  
28 global carbon cycle, and thus, the future climate change.

29

## 30 **Methods**

1        **Atmospheric CO<sub>2</sub> concentration data.** Weekly atmospheric CO<sub>2</sub> concentration data  
2 were obtained from the Earth System Research Laboratory, National Oceanic and  
3 Atmospheric Administration (NOAA-ESRL) archive (Masarie et al., 2014) for the period of  
4 1980-2012. 26 northern temperate and boreal stations with observations longer than 15 years  
5 (Table S1) are included in our analyses, given the focus of our study is the long-term trend,  
6 which would not be robust without long-term observations. The seasonal curves of  
7 atmospheric CO<sub>2</sub> at each station were extracted by fitting the observation data with a function  
8 consisting of quadratic polynomial for the long-term trend, four-harmonics for the annual  
9 cycle, and a 80-days Full-Width Half-Maximum value (FWHM) averaging filter and a  
10 390-days FWHM averaging filter to further remove short term variations and remaining  
11 annual cycles still present in the residuals after the function fit (Thoning et al., 1989). The  
12 processing process is incorporated in the standard CO<sub>2</sub> data processing software (CCGCRV)  
13 developed by NOAA-ESRL (Thoning et al., 1989). From the seasonal curve of atmospheric  
14 CO<sub>2</sub>, we then obtained the amplitude and monthly concentration difference.

15        **Land-atmosphere CO<sub>2</sub> exchange.** An ensemble of eight dynamic global vegetation  
16 models (DGVMs) from TRENDYv2 was used to simulate monthly land-atmosphere CO<sub>2</sub>  
17 exchange (Net Biome Productivity, NBP) for the period 1979-2012. These models were  
18 coordinated to perform three simulations (S1, S2 and S3) following the TRENDYv2 protocol  
19 (Le Quéré *et al.*, 2013; Sitch *et al.*, 2015). In simulation S1, only atmospheric CO<sub>2</sub> was varied.  
20 In simulation S2, only atmospheric CO<sub>2</sub> and climate were varied. In simulation S3,  
21 atmospheric CO<sub>2</sub>, climate and land use were varied. The effects of rising atmospheric CO<sub>2</sub>,  
22 climate change and land use change on NBP can then be obtained from S1, the difference  
23 between S2 and S1, and the difference between S3 and S2, respectively. Among the eight  
24 TRENDY models, four models (CLM4.5, ISAM, LPX and OCN) considered carbon-nitrogen  
25 interactions and nitrogen deposition through simulation S1 to S3. All models used the same  
26 forcing datasets, of which global atmospheric CO<sub>2</sub> concentration was from the combination of  
27 ice core records and atmospheric observations (Keeling & Whorf, 2005 and update); historical  
28 climate fields were from CRU-NCEP dataset (<http://dods.extra.cea.fr/data/p529viov/cruncep/>);  
29 land use data was from the Hyde database (Hurtt et al., 2011). The effect of nitrogen  
30 deposition was derived from an additional simulation (S4) performed by CLM4 model

1 (Oleson *et al.*, 2010; Mao *et al.*, 2013) in which all the driving factors (atmospheric CO<sub>2</sub>,  
2 climate and land use) were kept constant at 1980 value except transient nitrogen deposition  
3 from 1980 through 2012 (Lamarque *et al.*, 2005). Detailed information of the nine DGVMs  
4 used in this study is listed in Table S2.

5 **Ocean-atmosphere CO<sub>2</sub> exchange.** A biogeochemical model PlankTOM5 combined  
6 with a global ocean general circulation model NEMO (NEMO-PlankTOM5) were used to  
7 simulate the physical, chemical and biological processes that affect the surface ocean CO<sub>2</sub>  
8 concentration and thus the ocean-atmosphere CO<sub>2</sub> exchange (Buitenhuis *et al.*, 2010; Le  
9 Quéré *et al.*, 2015). The PlankTOM5 model was forced by inputs of ions and compounds  
10 from river, sediment and dust (Cotrim da Cunha *et al.*, 2007; Aumont *et al.*, 2003). The  
11 NEMO model was driven by daily wind and precipitation from NCEP reanalysis (Kalnay *et*  
12 *al.*, 1996). Further details can be found in Buitenhuis *et al.* (2010).

13 **Fossil fuel CO<sub>2</sub> emission.** A gridded monthly time series of fossil fuel CO<sub>2</sub> emissions  
14 from CDIAC were constructed based on proportional-proxy approach (Andres *et al.*, 2011;  
15 Boden *et al.*, 2016). Firstly, available monthly fossil fuel consumption data from 21 countries  
16 was compiled, which account for about 80% of global total emissions. Then these data were  
17 used as a proxy for all remaining countries without monthly data based on countries'  
18 similarities in climates and economies (for few countries, geographic closeness was also  
19 considered). For some years without explicit monthly data, Monte Carlo methods were used  
20 to apply data in years with known monthly fractions to those missing-data years. Further  
21 details can be found in Andres *et al.* (2011).

22 **The atmospheric transport model.** We used LMDZ4, a global tracer transport model  
23 (Hourdin *et al.*, 2013) driven by the re-analysis 3-D atmospheric wind fields from the  
24 European Centre for Medium-Range Weather Forecasts (Dee *et al.*, 2011), to transform  
25 land-atmosphere CO<sub>2</sub> exchange, fossil fuel CO<sub>2</sub> emission and ocean-atmosphere CO<sub>2</sub>  
26 exchange into point estimates of CO<sub>2</sub> concentration at 26 sites. The model configuration we  
27 used has a horizontal spatial resolution of 3.75° longitude × 2.5° latitude with 19 vertical  
28 layers.

29 To separate the effects of changes in atmospheric CO<sub>2</sub> ('CO<sub>2</sub>'), climate ('CLIM'), land  
30 use ('LU'), fossil fuel ('FF'), ocean carbon flux ('Ocean') and atmospheric transport ('Wind')

1 on seasonal atmospheric CO<sub>2</sub> concentration change, we designed eight transport simulations  
2 (T1~T8, see Table S3). The first one (T1) used time-varying monthly land-atmosphere CO<sub>2</sub>  
3 exchange under scenario S3 (driven by rising CO<sub>2</sub>, climate change and land use change),  
4 fossil fuel CO<sub>2</sub> emission, and ocean-atmosphere CO<sub>2</sub> exchange coupled with LMDZ4  
5 transport model having variable winds, indicating the combined effects of ‘CO<sub>2</sub>’, ‘CLIM’,  
6 ‘LU’, ‘FF’, ‘Ocean’ and ‘Wind’. To assess the contribution of ‘Wind’, the LMDZ4 transport  
7 experiment was force by historical varying wind but constant land-atmosphere CO<sub>2</sub> exchange,  
8 fossil fuel CO<sub>2</sub> emission and ocean-atmosphere CO<sub>2</sub> exchange in 1979 (T6). Next, to  
9 investigate the single effect of ‘CO<sub>2</sub>’, ‘CLIM’ and ‘LU’, we utilized LMDZ4 model with  
10 varying winds to perform another three transport simulations (T2, T3 and T4, see Table 3) in  
11 which fossil fuel CO<sub>2</sub> emission and ocean-atmosphere CO<sub>2</sub> exchange were constant at 1979  
12 value but land-atmosphere CO<sub>2</sub> exchange was varying under three scenarios (S1, driven by  
13 CO<sub>2</sub>; S2, driven by CO<sub>2</sub> and CLIM; S3, driven by CO<sub>2</sub>, CLIM and LU). Consequently, the  
14 single effect of ‘CO<sub>2</sub>’ on seasonal CO<sub>2</sub> variation can be assessed by the difference between T2  
15 and T6, that of ‘CLIM’ from the difference between T3 and T2, and that of ‘LU’ from the  
16 difference between T4 and T3. In addition, we prescribed varying land-atmosphere CO<sub>2</sub>  
17 exchange from CLM4 model under scenario S4 (only nitrogen deposition varying), constant  
18 fossil fuel CO<sub>2</sub> emission and ocean-atmosphere CO<sub>2</sub> exchange to LMDZ4 model with  
19 constant winds (transport simulation T5) to obtain the effect of nitrogen deposition. Finally, to  
20 gain the single effect of ‘FF’ and ‘Ocean’ on CO<sub>2</sub> seasonal variation, we performed another  
21 two simulations in which only fossil fuel CO<sub>2</sub> emission or ocean-atmosphere CO<sub>2</sub> exchange  
22 was varying in addition to variable winds (T7 and T8). Thus the contribution of ‘FF’ can be  
23 calculated from the difference between T7 and T6, and that of ‘Ocean’ from the difference  
24 between T8 and T6.

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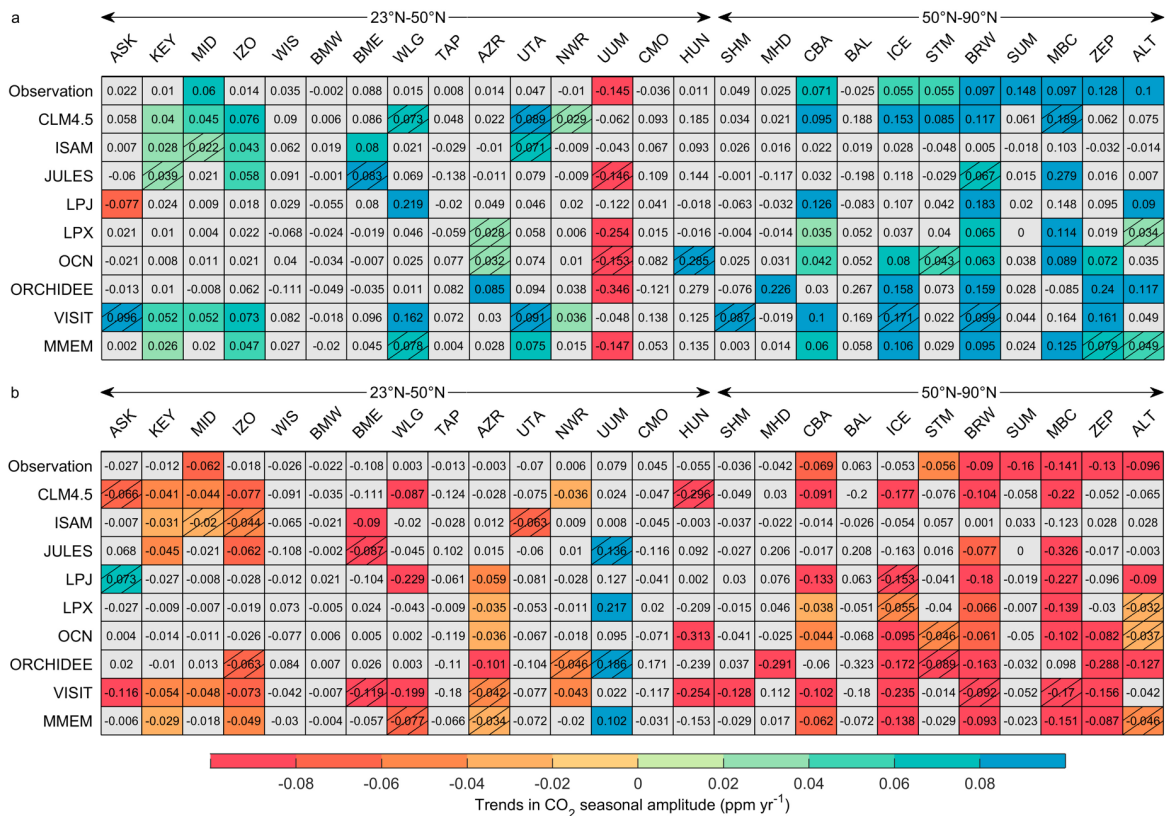
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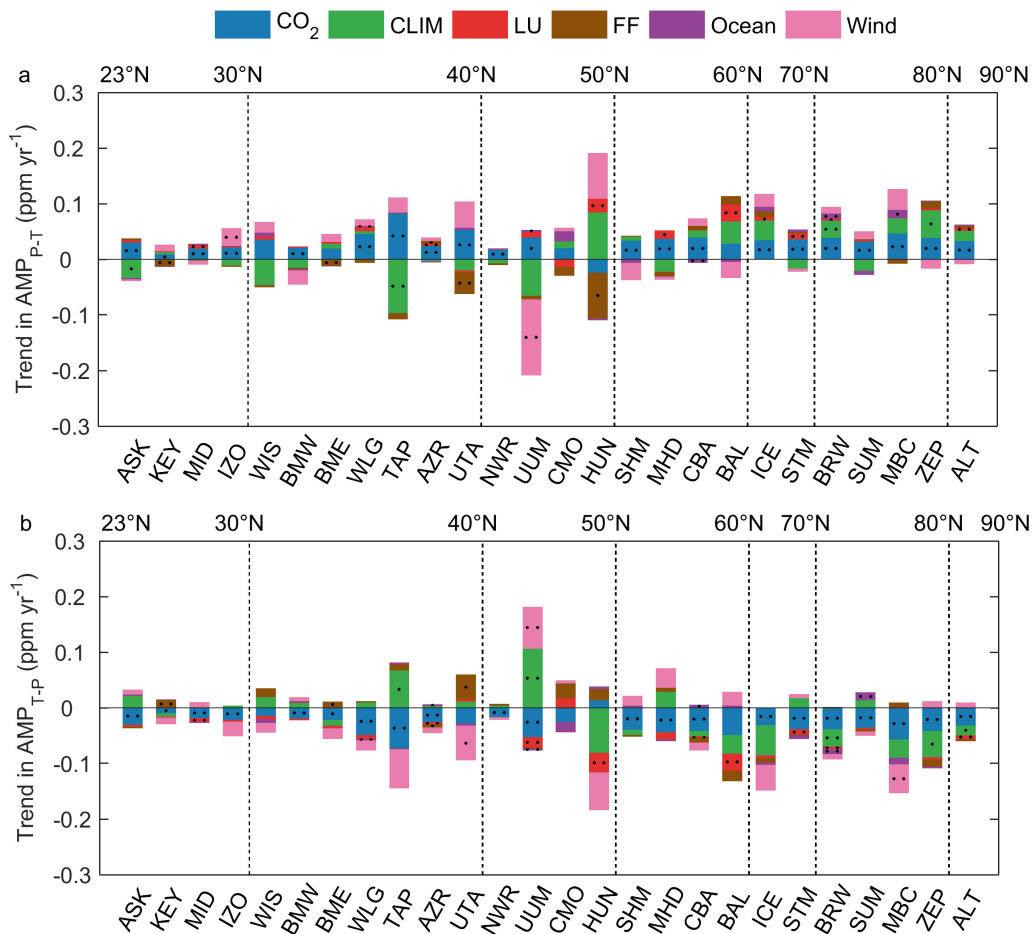
1 **Figure 1 Observed and modeled trends in CO<sub>2</sub> seasonal peak-to-trough amplitude**  
 2 **(AMP<sub>P-T</sub>) (a) and trough-to-peak amplitude (AMP<sub>T-P</sub>) (b).** Here we calculated the modeled  
 3 AMP<sub>P-T</sub>/ AMP<sub>T-P</sub> trends based on eight TRENDY models and multi-model ensemble mean  
 4 (MMEM) under T1 transport simulation (see methods). Shown on the top of the figure are the  
 5 abbreviated names of 26 atmospheric CO<sub>2</sub> concentration measurement sites in northern  
 6 temperate and boreal regions. We sort the sites according to their latitudes, from 23°N to 90°N.  
 7 Each row represents trends for different sites, while each column represents trends derived  
 8 from observation and model simulations at a certain site. Gray grids show insignificant trends  
 9 ( $P > 0.10$ ), while colored grids without slashes indicate statistically significant ( $P < 0.05$ ) and  
 10 those with slashes marginal significant ( $P < 0.10$ ). The number in each grid shows the value  
 11 of the trend. Site abbreviations are defined in Table S1.

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1 **Figure 2 Trends in CO<sub>2</sub> seasonal peak-to-trough amplitude (AMP<sub>P-T</sub>) (a) and**  
 2 **trough-to-peak amplitude (AMP<sub>T-P</sub>) (b) estimated by multi-model ensemble mean**  
 3 **(MEM) under different scenarios at 26 northern temperate and boreal sites. We**  
 4 present the results according to the latitudinal location of these sites. The single effect of  
 5 change in atmospheric CO<sub>2</sub> ('CO<sub>2</sub>'), climate ('CLIM'), land use ('LU'), fossil fuel ('FF'),  
 6 ocean-air carbon flux ('Ocean') and wind ('Wind') on CO<sub>2</sub> seasonal amplitudes was derived  
 7 from transport simulation (T2 - T6), (T3 - T2), (T4 - T3), (T7 - T6), (T8 - T6) and T6,  
 8 respectively (see Methods and Table S4). For each scenario, we denote those significant (P <  
 9 0.05) trends with two dots and marginal significant (P < 0.10) trends with a dot in the middle  
 10 of the bar.

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1 **Figure 3 Trends in CO<sub>2</sub> seasonal peak-to-trough amplitude (AMP<sub>P-T</sub>) (a) and**  
 2 **trough-to-peak amplitude (AMP<sub>T-P</sub>) (b) estimated by multi-model ensemble mean**  
 3 **(MMEM) under different scenarios, averaged over northern temperate region (23-50°N)**  
 4 **and boreal region (north of 50°N).** Model scenario simulations include change in  
 5 atmospheric CO<sub>2</sub> ('CO<sub>2</sub>'), climate ('CLIM'), land use ('LU'), fossil fuel ('FF'), ocean-air  
 6 carbon flux ('Ocean') and wind ('Wind'). Uncertainties are shown by error bars based on the  
 7 standard deviation of AMP trends across sites in each region.

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