Reply to the comments by reviewer #1

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Comment: I had the pleasure of reading your article entitled “Effects of nitrogen deposition on growing-season soil methane sink”. The manuscript is definitively a good effort to investigate the effect of nitrogen deposition on atmospheric CH_4 uptake by soils across forested ecosystems. This topic is very hot in the literature at the moment. The manuscript is very well written and easy to read and has a short but sufficient extent. That said, I found some fundamental flaws in three different aspects of the manuscripts: the assumptions you made by extrapolating your results, the datasets you employ and how the data was collected, and some problems with the general structure.

Reply: Thanks for your helpful comments. We have substantially improved the manuscript according to your suggestions. Please see more detailed information in our reply to your specific comments. We believe that the main concerns have been well addressed in the revised manuscript.

Comment: The definition and usage of the growing season extension is not well justified, nor correctly employed. Firstly, you never state why you are using the CH_4 uptake of the growing season and not for the whole year. Most of the papers you revise have data for more than a year. Also, the growing season varies greatly across and within ecosystems, thus cannot be set to a single value by biome. Growing season can be defined by multiple variables (temperature, precipitation, number of frozen days), thus the usage of a simple single value for each biome is simply not acceptable. As a result, there is no justification to consider the effect of nitrogen over the CH_4 uptake only during the growing season. Why not simply consider the whole year? If the growing season has importance for your analysis it is not reflected correctly in the manuscript and is never justified.

Reply: Our analysis focuses on the effects of N additions on growing-season soil CH_4 uptake because a) growing-season CH_4 uptake accounts for a major proportion of the annual CH_4 sink (Le Mer and Roger, 2001) and b) the reported data in literature are usually measured during the growing season. Based on comments by other reviewers, we have updated our database by a) excluding reports from theses without full peer-review, and

b) including results of field experiments using urea additions. The updated database includes experimental results of 28 forests across 22 sites (Fig.R1), but only 14 of the 28 experiments measured whole-year soil CH_4 uptake (0/7 for boreal forest, 0/7 for temperate forest, 10/10 for subtropical forest, 4/4 for tropical forests). Although some papers reported data for more than one year, they only measured soil CH_4 flux during the growing season of each year. When basing our results on measurements of annual soil CH_4 uptake, we are not able to conduct a statistical analysis for boreal and temperate forests. Due to the reasons discussed above, our analysis assesses the effects of N deposition on growing-season soil CH_4 uptake.

In our manuscript, mean growing season length was used to estimate the total growing-season soil CH_4 sinks and their response to N deposition for each biome. Generally, growing season can be defined by several approaches, including a) thresholds of multiple climatic variables, b) field monitoring of plant phenology and c) remote sensing of plant phenology. The estimated growing season length usually varies with different approaches. Moreover, we also understand that there are variations of growing season extension in a forest biome. Based on an assessment in northern hemisphere (Piao et al., 2007), growing season extension generally varies from 3 to 5 months in boreal forest and 6 to 8 months in temperate forest (compare Fig. R1 with Fig. R2). We thus used mean growing season length of 4 (mid May to mid Sep.) and 7 (Apr. to Oct.) months for boreal and temperate forest, respectively. In the revised manuscript, we have now also discussed the uncertainties of growing season length due to different approaches and the inner biome variations..

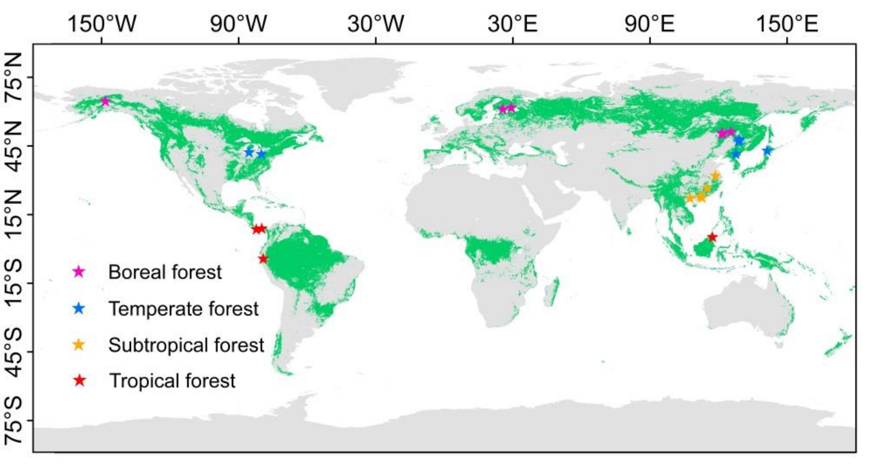

Figure R1. Geographical distribution of 22 forested sites with 28 forests receiving experimental nitrogen additions. Green shadows indicate the distribution of global forest.

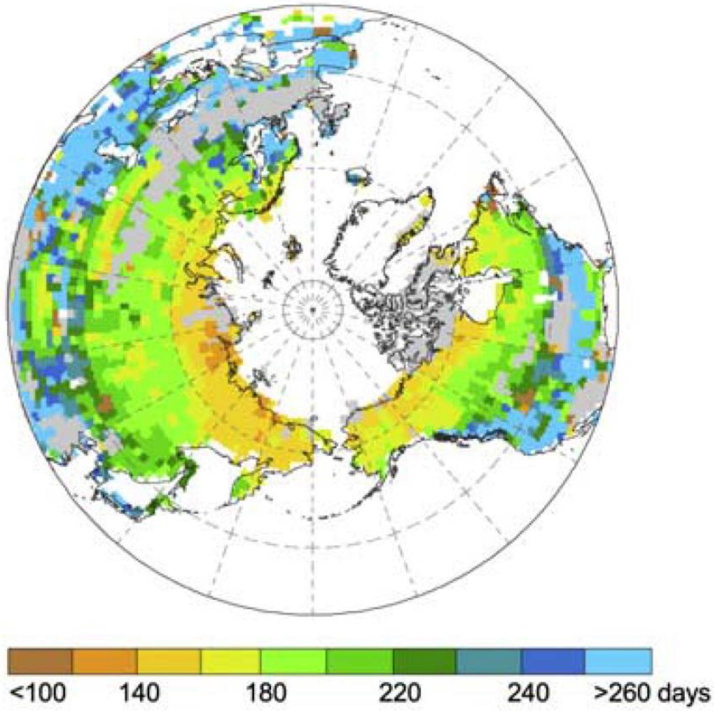

Figure R2. Growing season length of northern hemisphere (Piao et al., 2007)

Reference

Le Mer, J., Roger, P. 2001. Production, oxidation, emission and consumption of methane by soils: a review. European Journal of Soil Biology, 37(1), 25-50.

Piao, S., Friedlingstein, P., Ciais, P., Viovy, N., Demarty, J. 2007. Growing season extension and its impact on terrestrial carbon cycle in the Northern Hemisphere over the past 2 decades. Global Biogeochemical Cycles, 21(3): GB3018.

Comment: Secondly and possible the strongest criticism, your assumptions about the positive effect of nitrogen in the soil CH_4 sink in the boreal forest cannot be sustained with just four papers. The results are a wild extrapolation from studies that are not sufficient, nor analyzed correctly in your literature review. To be more precise: the work of Gulledge and Schimel (2000) found that nitrogen inhibit CH_4 consumption in boreal forest; Maljanen et al. (2006) found perform a factorial experiment with nitrogen and ashes and while nitrogen alone did increase the uptake it was not statistically different; additionally, ashes with nitrogen decrease the CH_4 uptake. Xu et al., 2014, found and increase in the uptake in the lowest N concentration (10 kg N ha^{-1} $year^{-1}$) but a negative effect with 20 and 40 kg N ha^{-1} $year^{-1}$ which are in the low N category you consider and do not be reflected in the results you obtained (why is the BF bar in figure 2 not going all the way to negative number based on this?). Finally, your fourth work Gao et al., 2013 is not available on google scholar. Based on this, the evidence to argue that boreal forest (the only ecosystem) presented an increased uptake due to nitrogen is not sustained at all. The other two ecosystems presented a decrease in all conditions, which is not novel.

Reply: Thanks for your helpful comments. We have realized that our previous analysis based on Student's t-test is not appropriate because it failed to consider the inter-study heterogeneity variance (τ^2) and the within-study variances (δ^2). This leads to inaccurate statistical speculations that are misleading, such as the results for boreal forest in our earlier manuscript. In the revised manuscript, we have tested the significance of N addition effect on soil CH_4 flux by conducting a meta-analysis in R software with the mixed effect model in metaphor package (Viechtbauer, 2010). We used a mean difference ($Flux_{treatment}$-$Flux_{control}$, the difference of mean growing-season soil CH_4 fluxes between the treatment plots and control plots) as the effect size to evaluate the effect of N additions. The heterogeneity variance (τ^2) across sites and the within-study variances (δ^2) were properly considered. If N additions result in a significant effect, we then estimated the response ratio of growing-season soil CH_4 flux based on the mean difference and the mean levels of N treatment.

Our reanalysis indicates that low-level N additions (<60 kg N ha^{-1} yr^{-1}), (being a range comparable to N deposition, only results in a significant decrease of soil CH_4 uptake in temperate forests, while no significant effects are found in boreal, subtropical and tropical forests (Fig. R3a). When receiving high-level N additions (>60 kg N ha^{-1} yr^{-1}), soil CH_4 uptake is significantly decreased in boreal, temperate and subtropical forests (Fig. R3b). If all N treatments are compiled, the overall effects are similar to those of high-level N additions.

The renewed analysis indeed leads to different results. In summary, only temperate forest shows a significant response ratio to low-level N addition, while high-level N fertilization results in significant response ratios across boreal, temperate and subtropical forests (Fig. R4). In view of the fact that N deposition is generally low in global forest ecosystems and rarely exceeding a maximum of 60 kg N ha^{-1} yr^{-1}(Vet et al., 2014; Schwede et al., 2018), our results imply that separating the effects of N deposition and high-level N fertilization is necessary to avoid overestimate the effects of global N deposition. However, this has never been considered by existing studies at a global scale (Liu and Greaver, 2009; Aronson and Helliker, 2010).

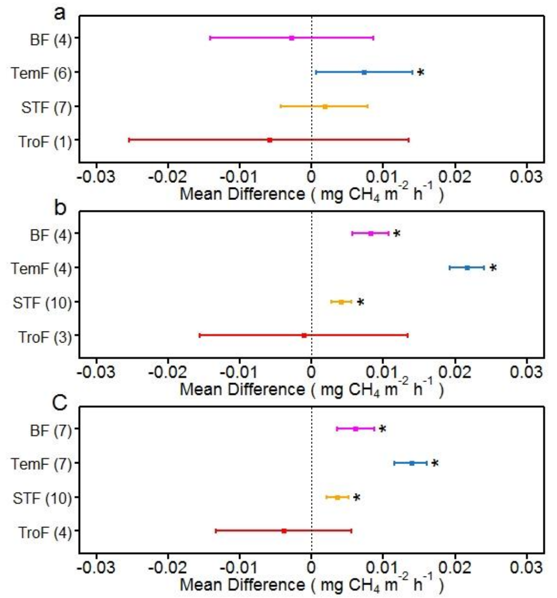

Figure R3. The mean difference (and 95% confidence intervals) of growing-season soil CH_4 flux to a) simulated low-level N addition (<60 kg N^{-1} yr^{-1}), b) high-level N fertilization (> 60 kg N^{-1} yr^{-1}) and c) all N treatments in boreal (BF), temperate (TemF), subtropical (STF), and tropical forest (TroF), respectively. The asterisk (*) indicates a significant effect ($p<0.05$).

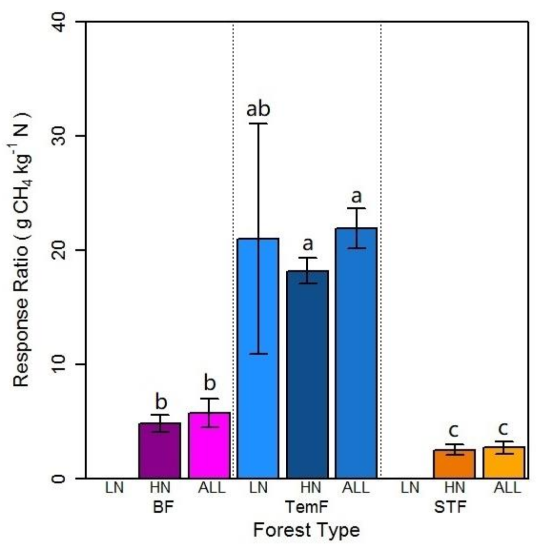

Figure R4. The response ratio of growing-season soil CH_4 flux to low-level N addition (LN, <60 kg N^{-1} yr^{-1}), high-level N fertilization (HN, > 60 kg N^{-1} yr^{-1}) and all N treatments (ALL) in boreal (BF), temperate (TemF) and subtropical forest (STF), respectively. Response ratio of insignificant effect (N treatments in tropical forest and LN in boreal and subtropical forest) is not shown.

Reference

Aronson, E.L., Helliker, B.R. 2010. Methane flux in non‐wetland soils in response to nitrogen addition: a meta-analysis. Ecology, 91(11), 3242-3251.

Liu, L.L., and Greaver, T.L. 2009. A review of nitrogen enrichment effects on three biogenic GHGs: the CO_2 sink may be largely offset by stimulated N_2O and CH_4 emission. Ecology Letters, 12, 1103–1117.

Schwede, D.B., Simpson, D., Tan, J., Fu, J., Dentener, F., Du, E., and De Vries, W.2018. Spatial variation of modelled total, dry and wet nitrogen deposition to forests at global scale. Environmental Pollution, 243, 1287-1301.

Vet, R., Artz, R.S., Carou, S., Shaw, M., Ro, C,U., Aas, W. et al. 2014.A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, Atmospheric Environment, 93, 3–100.

Viechtbauer, W. 2010. Conducting meta-analyses in R with the metafor package. Journal of Statistical Software, 36(3), 1-48.

Comment: Thirdly, your definition of the low and high N categories seems completely arbitrary and not justified based on literature. Why this threshold and not another?

Reply: We defined this threshold based on a global assessment of N deposition by the World Meteorological Organization (WMO) Global Atmosphere Watch programme (GAW) (Vet et al., 2014). This assessment shows a range of N deposition in various regions of the world, from 1~62 kg N ha^{-1} yr^{-1}. Specifically, the maximum level of N deposition occurs in eastern and southern China (Vet et al., 2014). We thus use 60 kg N ha^{-1} yr^{-1} as a threshold for maximum N deposition, in order to distinguish it from N fertilization with extremely high levels of N additions.

Reference

Vet, R., Artz, R.S., Carou, S., Shaw, M., Ro, C,U., Aas, W. et al. 2014.A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, Atmospheric Environment, 93, 3–100.

Comment: Finally, you are extrapolating data from 5-10 points, which are highly aggregated spatially to assume a biome-level behaviour, which is incorrect. In other literature reviews of the topic, the authors revise around 35 papers to propose general mechanisms that can be scaled. In other words, you are trying to extrapolate a pattern based on few observations, without the proposition of an underlying mechanisms to support the increase in spatial scale.

Reply: We fully agree that extrapolating data from limited sites could introduce large uncertainties. By updating our database and conducting a meta-analysis, we do a more precise analysis accounting for uncertainties in effects. Our results show that the effects of low-level N addition and high-level N fertilization can be significantly different in boreal and subtropical forest biomes. Low -level N addition, comparable to the range of N deposition, only results in a reduction of soil CH_4 sink in temperate forest. This finding implies that existing meta-analyses at a global scale have likely overestimated the effect of N deposition (Liu and Greaver, 2009; Aronson and Helliker, 2010), in view of the fact that N deposition is generally low in global forest ecosystems and rarely exceeding a maximum of 60 kg N ha^{-1} yr^{-1} (Vet et al., 2014; Schwede et al., 2018). To demonstrate the magnitude of the overestimation, we have now estimated and discussed the possible overestimation by comparing the scaling results based on the response ratios of low-level N addition and high-level N fertilization (Table R1; a global overestimation by approximately 20%). We have also mentioned that this kind of scaling has large uncertainty, but our purpose here is to demonstrate that separating the effects of low-level N addition (N deposition) and high-level N fertilization is necessary to avoid overestimate the effects of global N deposition.

Table R1. Estimated biome effect on soil CH_4 sinks in global forests based on the response ratios (RR) of low-level N addition (LN) and high-level N fertilization (HN).

Forest biome	Area (Million ha)	Mean N deposition (kg N ha^{-1} yr^{-1})	RR_{LN} g CH4 per kg N	RR_{HN} g CH4 per kg N	Biome $Effect_{LN}$ (10^{-3}Tg CH_4)	Biome $Effect_{HN}$ (10^{-3} Tg CH_4)
Boreal	1225	1.2	0	4.9 ±0.8	0	1.8 ±0.3
Temperate	673	7.3	21 ±10.1	18.2 ±1.1	43.2 ±21.8	37.5 ±2.3
Subtropical	320	14.6	0	2.6 ±0.4	0	12.2 ±1.9
Tropical	1798	7.2	0	0	0	0

Reference

Aronson, E.L., Helliker, B.R. 2010. Methane flux in non‐wetland soils in response to nitrogen addition: a meta-analysis. Ecology, 91(11), 3242-3251.

Liu, L.L., and Greaver, T.L. 2009. A review of nitrogen enrichment effects on three biogenic GHGs: the CO_2 sink may be largely offset by stimulated N_2O and CH_4 emission. Ecology Letters, 12, 1103–1117.

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Vet, R., Artz, R.S., Carou, S., Shaw, M., Ro, C,U., Aas, W. et al. 2014.A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, Atmospheric Environment, 93, 3–100.

Minor comments

Comment: There are some mistakes defining the sign for the CH_4 sink (both negative and positive signs are used along the paper). It needs to be consistent.

Reply: We have checked through the manuscript and used consistent signs for CH_4 sinks in the revised manuscript. Specifically, soil CH_4 sink/uptake is indicated by negative values. A positive value of the mean difference ($Flux_{treatment}$-$Flux_{control}$) and response ratio indicates a reduction of soil CH_4 sink/uptake. This has been clarified in the revised manuscript.

Comment: Page 2, line 27, you started to talk about China as a hot spot for N deposition with no previous justification and not using this particular region in the paper. If you are focusing on a global scale, you should either give more examples or eliminate the regional-level comparisons.

Reply: Based on a global assessment of N deposition by the World Meteorological Organization (WMO) Global Atmosphere Watch programme (GAW) (Vet et al., 2014), N deposition generally shows a global range of 1~62 kg N ha^{-1} yr^{-1}. Specifically, the maximum level of N deposition occurs in eastern and southern China (Vet et al., 2014). We thus went into a bit more details of N deposition status in China. We have revised the manuscript accordingly to avoid any misunderstanding.

Reference

Vet, R., Artz, R.S., Carou, S., Shaw, M., Ro, C,U., Aas, W. et al. 2014.A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus, Atmospheric Environment, 93, 3–100.

Comment: Page 6, line 25, the argument about the CO_2 equivalents to measure the N effect over soil CH4 sink is absolutely out of place. 1) You cannot predict the effect of N deposition in the next 100 years using current values, 2) you are using a GHG potential of 25 (should be 28), 3) you create very strong arguments with very little evidence and not sufficient data, 4) finally, CO_2-eq are not really used any longer, as the relationship of GHG to CO2 is not linear.

Reply: We generally agree with your points. In the revised manuscript, we have excluded the discussion based on CO_2 equivalents for CH_4 sink. Thanks again for the helpful comments.