Reply to the comments by reviewer #2

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Comments: This manuscript reports on a synthesis or meta-study of studies that report the effects of N additions on soil CH4 fluxes. The authors did a literature search using several sources, calculated a response variable, stratified results according to dose and biome, and conclude that low-level N addition increased CH_4 uptake in boreal forest while decreases occurred at high N addition levels and in all other biomes. A meta-study is only as good as the sources where the information comes from, and therefore studies used in such a meta-study should be generally accessible and peer-reviewed. Unfortunately, several of the studies used in this meta-study did not meet these criteria. I find it problematic that apart from ISI Web of Science, also Google Scholar and China National Knowledge Infrastructure are used to search for literature. To my knowledge, these two latter sources also list reports that are not peer-reviewed. I strongly suggest to ONLY use ISI as a literature source, because here only peer-reviewed sources are listed. Studies that are not peer reviewed should not be included in a synthesis. In your meta-study I suggest to exclude the following studies listed in the supplement: - two of the studies listed are a MSc thesis (Wang, 2012, Pan, 2013). Such a thesis is not considered peer-reviewed, please exclude them. -all studies that involved incubations, instead of field measurements. For example, the study by Chen et al. 2017 mentions that they did laboratory incubations. You even state in Page 3, line 16 that you only included studies that used closed static chamber technology. Apparently that is not true. There may be more studies with incubations, I did not check them all. - Please, exclude studies published in Chinese (or other non-English publications) with only an English abstract. I do not consider such studies as generally accessible. For example, the study by Hu et al., 2011, is only accessible in Chinese and there may be more in the list.

Reply: Thanks for your comments, which have helped us to improve the manuscript substantially. We fully agree that the quality of data sources is very important for our meta-analysis. In the revised manuscript, we have updated the database by a) excluding experimental results from theses without fully peer-review (Wang, 2012; Pan, 2013), b) excluding experimental results from laboratory incubations (only Chen et al. 2017), c) excluding an experimental study that didn't report values of variance/standardized error (Steudler et al., 1989), d) excluding results of experiments with plot area < $10 m^2$ or without buffer zones, and e) including more results of field experiments using urea additions. We further beg your understanding to include reports published in fully peer-reviewed Chinese journals because these publications are generally of good quality in science. Overall, we updated our database based on the criteria above and the current database includes results of 28 experiments across 22 sites (see Figure R1).

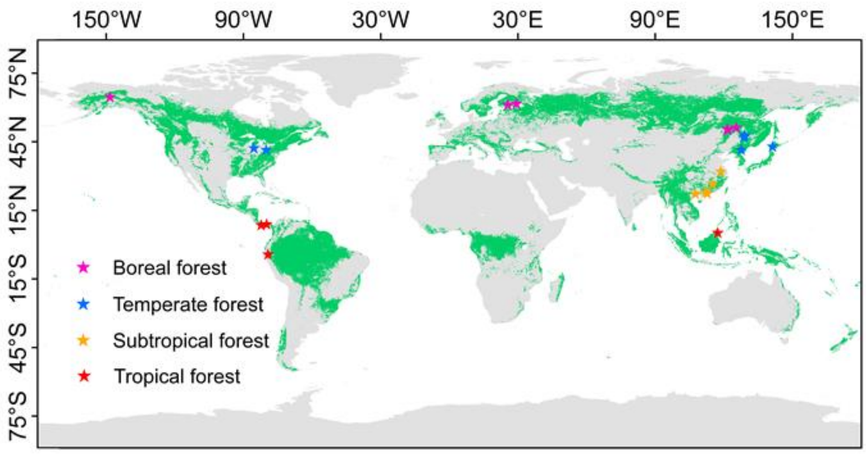

Figure R1. Geographical distribution of 28 forests receiving experimental nitrogen additions. Green shadows indicate the distribution of global forest.

Reference

Pan, D.R. 2013. Study on greenhouse gas emission for grassland soil below different forest soils under precipitation reduction and Nitrogen deposition in Shennongjia mountain. Gansu Agricultural University, Master thesis.

Steudler, P.A., Bowden, R.D., Melillo, J.M., Aber, J.D. 1989. Influence of nitrogen fertilization on methane uptake in temperate forest soils. Nature, 341(6240), 314-316.

Wang, R.N. 2012. Effects of simulated atmospheric nitrogen deposition on the exchange fluxes of greenhouse gases in the temperate forest soil. Beijing Forestry University, Master thesis

Comments: You use the 60 kg N ha^{-1} yr^{-1} as an arbitrary cut-off between 'low' and 'high' level N addition. Did you calculate also the background N-deposition in the N additions? For example, the study by Li et al., 2015, mentions that there is a background N deposition of more than 30 kg N ha^{-1} yr^{-1}, while the treatment is 40 kg ha^{-1}

yr^{-1}. Together this would be more than 60 kg N ha^{-1} yr^{-1} and the 40 kg treatment should be grouped as 'high' level N addition. I suspect that is not how you did this and it just illustrates how arbitrary the choice of 60 kg N ha-1 yr-1 is.

Reply: Thanks for your comments. We defined the threshold of N deposition versus high-level N addition 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 1~62.25 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 used 60 kg N ha^{-1} yr^{-1} as a threshold for a possible maximum of N deposition, in order to distinguish it from N fertilization with extremely high levels of N additions. In the revised manuscript, we have conducted a meta-analysis (Viechtbauer, 2010) by using background N deposition as a moderator. 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 results indicate that background N deposition has a significant influence on the effect of N additions ($p=0.0006$). However, a recent assessment of N deposition indicates that average N deposition varied significantly across global forest biomes, showing a trend subtropical forest (14.6 kg N ha^{-1} yr^{-1}) > temperate forest (7.3 kg N ha^{-1} yr^{-1}) and tropical forest (7.2 kg N ha^{-1} yr^{-1}) > boreal forest (1.2 kg N ha^{-1} yr^{-1}). Moreover, total N availability from mineralization, biological N fixation and N deposition generally shows a decrease from tropical forest to boreal forest (Cleveland et al., 2013). This hinders us to separating the effects of background N deposition/availability from the effect of forest biomes. We have revised the manuscript to avoid any misunderstanding.

Our reanalysis indicates that simulated N deposition (<60 kg N ha^{-1} yr^{-1}) 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. R2a). 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. R2b). If all N treatments are compiled, the overall effects are similar to those of high-level N additions. In summary, only temperate forest shows a significant response ratio to simulated N deposition, while high-level N additions result in significant response ratios across boreal, temperate and subtropical forests (Fig. R3). 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, our new findings have never been considered by existing studies at a global scale (Liu and Greaver, 2009; Aronson and Helliker, 2010).

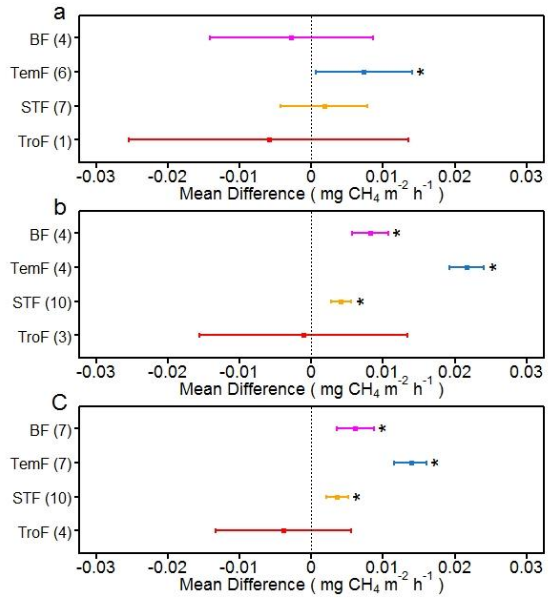

Figure R2. The mean difference (and 95% confidence intervals) of growing-season soil CH_4 flux to a) simulated N deposition (<60 kg N^{-1} yr^{-1}), b) high-level N addition (> 60 kg N^{-1} yr^{-1}) and c) all N treatments in boreal (BF), temperate (TemF), subtropical (STF), and tropical forest (TroF), respectively. The asterisk (*) indicates a significant effect ($p<0.05$).

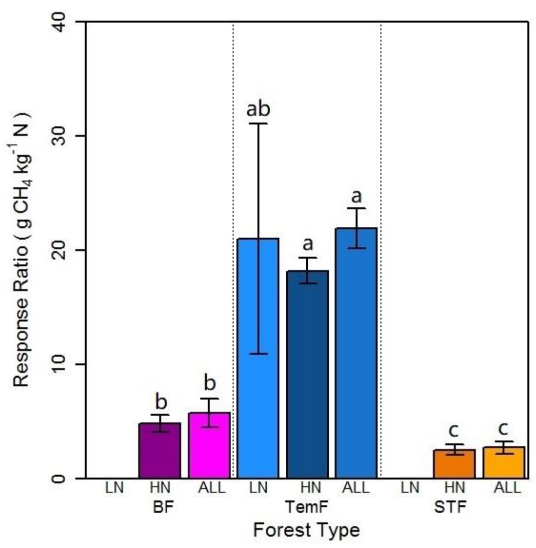

Figure R3. The response ratio of growing-season soil CH_4 flux to simulated N deposition (LN, <60 kg N^{-1} yr^{-1}), high-level N addition (HN, > 60 kg N^{-1} yr^{-1}) and all N treatments (ALL) in boreal (BF), temperate (TemF) and subtropical forest (STF), respectively. Response ratio of insignificant effect (N treatments in tropical forest and LN in boreal 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.

Cleveland, C. C., Houlton, B. Z., Smith, W. K., Marklein, A. R., Reed, S. C., Parton, W., et al. 2013. Patterns of new versus recycled primary production in the terrestrial biosphere. Proceedings of the National Academy of Sciences, 110(31), 12733-12737.

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.

Comments: You did not mention any other criteria for inclusion or exclusion of studies. However, I think you should define what you consider a sufficiently large plot and a sufficiently wide buffer zone between treatments. Also, were all studies having true replicates? This kind of important information on the quality of studies is completely ignored in your manuscript.

Reply: Thanks for your suggestions. In the revised manuscript, we have updated the database by a) excluding experimental results from theses without peer-reviews, b) excluding experimental results from laboratory incubations, c) excluding an experimental study that didn't report values of variance/standardized error (no information for replicates), d) excluding results of experiments with plot area < $10 m^2$ or without buffer zones. We have also recorded information of replicates, plot area, buffer zone, and variance/standard deviation in our updated database. In the revised method section, we have included more detailed information for the criteria of data collection.

Comments: I found it very adventurous that you excluded the only peer-reviewed study conducted in tropical ecosystems (Veldkamp et al., 2013) with the argument that urea as an organic N form has 'limited implications for the effects of N deposition', then cite Aronson and Helliker, (2010) as the source for this statement (page 3, line 20), and later lament that there are no studies conducted in tropical areas and then even extrapolate the results from subtropical forests to the tropics. -First of all, while urea is strictly spoken an organic N source, it is quickly hydrolysed ($NH_2CONH_2 + H_2O \rightarrow CO_2 + 2NH_3$) after application and the gaseous NH_3 reacts with water to form ammonium (NH_4^+). Only on soils with a high pH there are significant losses through volatilization. -Second, in the paper by Aronson & Helliker (2010), I did not find any statement that urea has limited implications for the effects of N deposition. In contrast, they also analysed studies with urea additions. They found no difference between Urea and other pure N fertilizers. They also concluded that 'any conclusions of the effects of specific N species relative to others must be highly qualified, as the form of N that results may be quite different from that added'. Therefore, to quote the Aronson & Helliker (2010) paper as the source why studies that add urea should be excluded is misleading. -Third, ignoring the only tropical study and later filling up the gap with studies from subtropical areas is very adventurous. Especially since the study conducted in the tropics did not following the hypothesized trend across biomes.

Reply: Thanks for your suggestions. We fully agree that it is not appropriate to exclude N addition experiments using urea in tropical forests. In the paper by Aronson and Helliker (2010), the authors showed that the effects of N fertilization on soil CH_4 uptake did vary significantly with N forms (Figure R4). Specifically, the effects of urea and ammonium nitrate showed slight overlap, although the statistical analysis indicated no significant difference (Figure R4). This result motivated us to exclude experiments with urea additions in our previous manuscript, because a) N deposition mainly occurs in inorganic N forms, and b) urea might be less capable to indicate the effects of N deposition. As suggested by you and other reviewers, we have updated our database by searching urea addition experiments in the literature in view of the fact that the effects of urea and ammonium nitrate were not significantly different. As a result, four experiments from tropical forest were included in our updated database (Veldkamp et al., 2013; Matson et al., 2016; Mori et al., 2017). Using a meta-analysis in R software with metaphor package (Viechtbauer, 2010), we show that simulated N deposition (<60 kg N^{-1} yr^{-1}) and high-level N addition (<60 kg N^{-1} yr^{-1}) both had no significant effect on soil CH_4 uptake (Figure R2a&b). Overall, no significant effect of N additions on soil CH_4 uptake was found in tropical forest (Figure R2c). We have revised the manuscript accordingly.

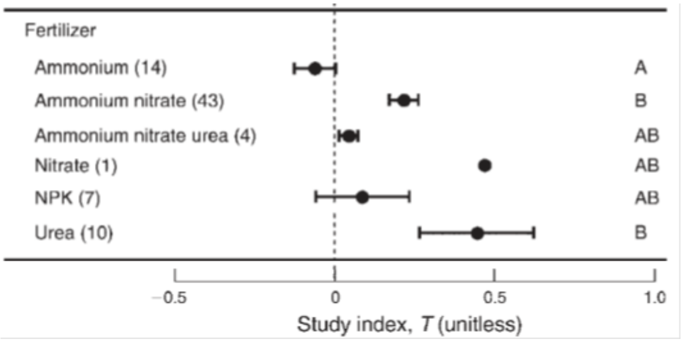

Figure R4. Effects of different N forms on soil CH_4 uptake (Aronson and Helliker, 2010)

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.

Matson, A. L. , Corre, M. D., Veldkamp, E. 2016. Canopy soil greenhouse gas dynamics in response to indirect fertilization across an elevation gradient of tropical montane forests. Biotropica, 49(2), 153-159.

Mori, T., Imai, N., Yokoyama, D., Mukai, M., Kitayama, K. 2017. Effects of selective logging and application of phosphorus and nitrogen on fluxes of CO_2, CH_4 and N_2O in lowland tropical rainforests of borneo. Journal of Tropical Forest Science, 248-256.

Veldkamp, E., Koehler, B., Corre, M. D. 2013. Indications of nitrogen-limited methane uptake in tropical forest soils. Biogeosciences, 10(8), 5367-5379.

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

Comments: The hypothesis to be tested (Page 2, line 21 and further) is weak and based on incomplete assumptions. You simply assume that N availability increases from boreal to tropical biomes, which is not true. If you read publications by Vitousek more carefully you will see that the main factor is not the biome but how heavily weathered soils are. More than half of the tropics is located on soils that are not heavily weathered (e.g. montane forests) and N availability is expected to be as low as other biomes where young, less weathered soils dominate.

Reply: Thanks for your comments. Nitrogen availability is internally driven by mineralization and externally from biological N fixation and N deposition. Recently, the role of N inputs from bedrock weathering is increasingly recognized, but this N source is only important when soil depth is shallow and N content is abundant in the bedrock. Previous studies have shown that the sum of N inputs from biological N fixation and N deposition increase from boreal forest to tropical forests (Cleveland et al., 1999&2013; Du and De Vries, 2018). Moreover, the rate of N mineralization also shows an increase from boreal forest to tropical forest (Cleveland et al., 2013; Deng et al., 2018). Overall, we think that the increase in N availability from boreal forest to tropical forest has been well recognized in literature.

Reference

Cleveland, C. C., Townsend, A. R., Schimel, D. S., Fisher, H., Howarth, R. W., Hedin, L. O., et al. 1999. Global patterns of terrestrial biological nitrogen (N_2) fixation in natural ecosystems. Global Biogeochemical Cycles, 13(2), 623-645.

Cleveland, C. C., Houlton, B. Z., Smith, W. K., Marklein, A. R., Reed, S. C., Parton, W., et al. 2013. Patterns of new versus recycled primary production in the terrestrial biosphere. Proceedings of the National Academy of Sciences, 110(31), 12733-12737.

Du, E., De Vries, W. 2018. Nitrogen-induced new net primary production and carbon sequestration in global forests, Environmental Pollution, 242, 1476–1487.

Deng, M., Liu, L., Jiang, L., Liu, W., Wang, X., Li, S., et al. 2018. Ecosystem scale trade-off in nitrogen acquisition pathways. Nature Ecology & Evolution, 2(11), 1724.

Comments: You group all forest ecosystems together and make no difference between natural forest and plantations or managed forests. Tree plantations typically have significant growth rates and are almost always N limited, also in tropical and subtropical conditions. Ignoring this may lead to wrong conclusions.

Reply: This is a good point. We have checked our database and include information of natural forest or plantations. We found that only 10 of 28 forests (1/7 boreal, 3/7 temperate and 6/10 subtropical) were plantations. This doesn't allow us to test the difference between plantations and natural forests for each forest biome due to uneven small sample size. As the database is the most updated from literature, our results represent the best knowledge we can derive at this stage. In the revised manuscript, we have discussed the uncertainties possibly due to the different N status of natural forest and plantations. We have also labelled natural forests and plantations in the revised summary table for the database.

Comments: In summary, this synthesis is poorly conducted. There are studies included that are not peer-reviewed and there were no quality criteria for the studies that were included. The hypothesis is weak and based on incomplete assumptions. The distinction between 'high level' and 'low level' N addition is arbitrary and the background N deposition is ignored. Finally, I could not find any objective reason why studies where urea was added were excluded. Given these weaknesses of this synthesis I strongly doubt the validity of the conclusions and I recommend not to publish this manuscript in Biogeosciences.

Reply: Thanks again for your helpful comments. First, we have updated the manuscript by a) excluding experimental results from master thesis, b) excluding experimental results from laboratory incubations, c) excluding an experimental study that didn't report values of variance/standardized error, d) excluding results of experiments with plot area < $10 m^2$ or without buffer zones, and e) including more results of field experiments using urea additions. More detailed information for data collection has also been included in the revised section on data and method. Second, we have included more detailed evidence from literature for the gradients of N availability across forest biomes, both internally driven by mineralization and externally from biological N fixation and N deposition. This underpins our hypothesis that the effect of N additions likely varies across forest biomes due to a shift in N availability. Third, the threshold of maximum N deposition versus high-level N addition is defined based on a global assessment of N deposition by the World Meteorological Organization (WMO) Global Atmosphere Watch programme (GAW), which shows a range of N deposition in various regions of the world, from 1~62.25 kg N ha^{-1} yr^{-1}. We thus used 60 kg N ha^{-1} yr^{-1} as a threshold for a possible maximum of N deposition, in order to distinguish it from N fertilization with extremely high levels of N additions. Finally, we have updated our database by including urea addition experiments and conducted a reanalysis using meta-analysis. Overall, we believe that the main concerns have been well addressed in the revised manuscript.