

Response to Referee #2

1. Section 2.1: I recommend that you show a spatial map of changes in BVOCs over time? Li et al. (2020) shows only 2008-2018.

Response: Thank you for your valuable suggestion. In this study, the historical BVOC emissions were estimated using the same meteorology data to explore the influence of the interannual variability of vegetation biomass. So we add a spatial map of changes in BVOC emissions caused by vegetation biomass variability, as shown in Fig. 2. In addition, we add a new Section 3.1 “BVOC emissions” to describe the BVOC emissions in June 2018 and their interannual changes. The relative description of BVOC emissions in Section 2.1 of the original manuscript are moved here. In Section 3.1.2 “Sensitivity of the BVOC emissions to leaf biomass”, Figure 2 is added and the changes in BVOC emissions caused by vegetation biomass variability are discussed.

Line 195–204 in revised manuscript, "Due to the increased volume and production of vegetation, the total BVOC emissions increased by 58.66% at average rates of 96.64 Gg yr⁻¹, of which isoprene, monoterpene, sesquiterpene increased by 108.57%, 38.17%, and 33.35% at average rates of 11.10, 0.99, and 0.17 Gg yr⁻¹, respectively. Isoprene emissions increased more rapidly over the past 40 years, which is primarily due to the greater increase in the biomass of broadleaf trees, which have the highest isoprene emission rates. Monoterpene and sesquiterpene increased at a lower rate because the increase of leaf biomass of conifers is relatively small. Fig. 2 shows the spatial distribution of interannual variations in BVOC emissions caused by the changing leaf biomass. Since the needleleaf and broadleaf trees tend to have a higher emission potential than grass or crop (Guenther et al., 2012), their wide distribution and the substantial increase in biomass result in the largest interannual variability of BVOC emissions in the Great Khingan, Changbai Mountains, North China Plain, Central and Southern China, and Hainan Province. However, the emission of BVOCs in the northwest and southern coastal areas has decreased."

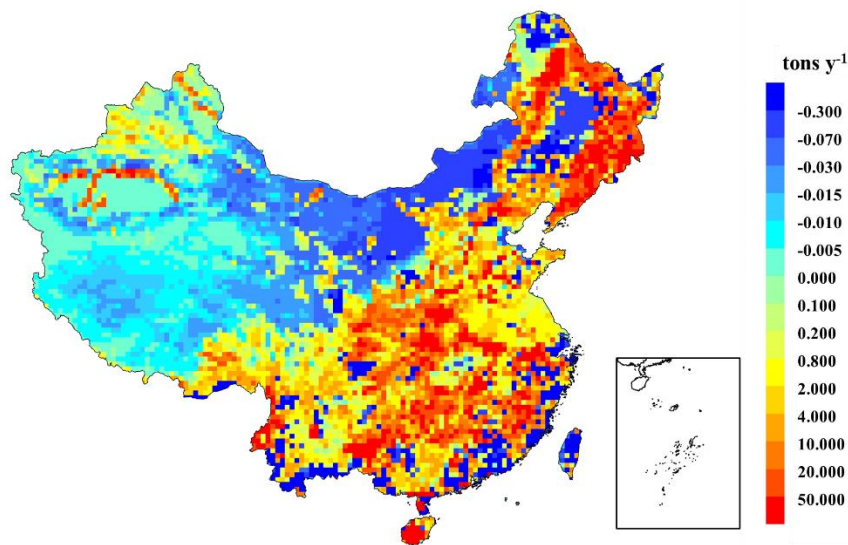


Fig. 2. Spatial distribution of interannual variations in BVOC emissions caused by leaf biomass changes.

References:

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The model of emissions of gases and aerosols from nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, *Geosci. Model Dev.*, 5, 1471–1492, <https://doi.org/10.5194/gmd-5-1471-2012>, 2012.

2. In the introduction you say that your study uses “more accurate BVOC emissions”. However, you don’t provide evidence of this. Satellite data of formaldehyde could help in the evaluation over the satellite era. Do a literature search as there are a number of studies that use HCHO data to constrain BVOC emissions. How will you validate your emissions before the satellite era (e.g., 1981–2004)?

Response: Thank you for your constructive comment and we are sorry for the unclear explanation for our “more accurate BVOC emissions”.

Firstly, we outstand the higher accuracy through comparing the method between our study and others in BVOC emission estimation. (1) **Determination of emission rate and leaf biomass.** In previous studies, traditional emission categories were used to determine isoprene

and monoterpene emission rates (Guenther et al., 1994; Klinger et al., 2002; Simpson et al., 1999; Wang et al., 2007). In this method, discrete emission categories (e.g., negligible, low, moderate, and high) were defined, which lacked theoretical evidence. And some studies utilized coarse emission intensity classifications. The determined emission rates typically differed between studies and had high uncertainty. In our study, we summarized a large number of observations from China and other countries to obtain more accurate basal emission rates by the theoretically effective statistical approach (Li et al., 2020). The statistical isoprene emission rates included seven categories by lowest, lower, low, moderate, high, higher, and highest. Monoterpene included six categories by lowest, lower, low, moderate, high, and higher. The accuracy of emission rates can be expected to be improved. For leaf biomass, the previous studies usually applied an average value for each vegetation class, such as broadleaf trees, needleleaf trees, crops, and grasses, without revealing their differences among regions and plant species (Klinger et al., 2002; Wang et al., 2007). In our study, the plant specific leaf biomasses were estimated based on the provincial or city-level statistic of vegetation volume and production using apportion models (Li and Xie, 2014). We obtained the gridded leaf biomass for the 23 vegetation species/types. (2) **Detailed vegetation classification.** Most studies on the BVOC emissions inventory in China typically include a coarse vegetation classification that is based on a less-detailed vegetation distribution (Gao et al., 2019; Klinger et al., 2002; Wang et al., 2007). And the MEGAN2.1 defined 15 vegetation types (Guenther et al., 2012). In our study, Vegetation Atlas of China (1:1,000,000), which had detailed vegetation distributions at a high horizontal resolution of about 250 m, was used to produce more detailed vegetation classification in Shandong Province, including 23 plant species/types (four broadleaf trees, five needleleaf trees, eight crop species, and six subtypes of shrub and grass).

In the revised manuscript, we add the explanation to show the higher accuracy of the determined emission factors and vegetation classification. Line 101–103, "Previous studies typically included a coarse vegetation classification that is based on a less-detailed vegetation distribution (Gao et al., 2019; Klinger et al., 2002; Wang et al., 2007). And the MEGAN2.1 defined 15 vegetation types by default (Guenther et al., 2012)." is added. Line 109–111, "In previous studies, traditional emission categories were used to determine emission rates (Guenther et al., 1994; Klinger et al., 2002; Simpson et al., 1999; Wang et al., 2007), which

usually utilized coarse categories and resulted in high uncertainty." is added. Line 113–115, "Previous studies usually applied an average value for each vegetation class, such as broadleaf trees, needleleaf trees, crops, and grasses, without revealing their differences among regions and plant species (Klinger et al., 2002; Wang et al., 2007)." is added.

Secondly, as you suggested, we add the validation of BVOC emissions by comparing the simulation with satellite observation of formaldehyde concentration and also the canopy-level emission flux measurements in China. The validation is conducted for estimation in 2018. The emissions in other years are not evaluated because the historical BVOC emissions are estimated using the same meteorology data to explore the influence of the interannual variability of vegetation biomass. They cannot display the real emissions for the years other than 2018. (1) Validation by comparing with canopy-level emission flux measurements. The canopy-level emission flux measurements in China were used for validation (Bai et al., 2015, 2016, 2017). The gridded BVOC emission estimated by MEGAN were extracted where locating the flux measurement sites. The comparison of model simulation and observation is shown in Fig. S1. The estimated of BVOC emissions are higher with an average mean bias of $1.11 \text{ mg m}^{-2} \text{ h}^{-1}$, mainly because of the differences in time between the simulation and measurements. But they are correlated by $r=0.84$, exhibiting good agreement in spatial variations. (2) Validation by comparing with satellite observation of formaldehyde concentration. We compared simulated isoprene emission with satellite-derived HCHO column concentration using the Ozone Monitoring Instrument (OMI) HCHO vertical column product. The monthly averaged OMI HCHO vertical column in June 2018 correlates with the model estimated results at a 99% confidence level.

In the revised manuscript, the above validations are added in Section 3.1.1. Meanwhile, a literature summarization that use HCHO data to constrain BVOC emissions is also added. Line 171–185, "The emission simulations were validated by using the measurements of BVOC emission flux and formaldehyde (HCHO) concentration. The flux measurements of BVOCs conducted in China were collected (Bai et al., 2015, 2016, 2017). The gridded BVOC emission estimated by MEGAN were extracted where the flux measurement sites were located to do the comparison (Fig. S1). The modeled fluxes of BVOCs in this study capture the spatial variability of observations better with a correlation coefficient of 0.84. But the estimation is higher than

measurement with an average mean bias of $1.11 \text{ mg m}^{-2} \text{ h}^{-1}$, mainly because of the differences in time between them. Isoprene is the main compound in BVOC species, accounting for nearly half of total BVOC emissions in China. It undergoes chemical and photochemical reactions in the atmosphere, and the oxidation product is mainly HCHO (Bai and Hao, 2018; Orlando et al., 2000). In forest areas and in summer, biogenic isoprene is the dominant source of HCHO, so satellite HCHO column concentration is widely used to constrain isoprene emissions (Opacka et al., 2021; Palmer et al., 2003; Stavrakou et al., 2018; Wang et al., 2021; Zhang et al., 2021). In this study, we used the HCHO vertical column detected by Ozone Monitoring Instrument (OMI) to validate the spatial variability of isoprene estimates. The monthly OMI HCHO data from the EU FP7 project QA4ECV product (Quality Assurance for Essential Climate Variables; <http://www.qa4ecv.eu>) was used in this study. The result of the statistical analysis with a confidence interval of 99% indicates that the monthly averaged OMI HCHO vertical column in June 2018 is significantly correlated to the model-estimated isoprene emissions." is added.

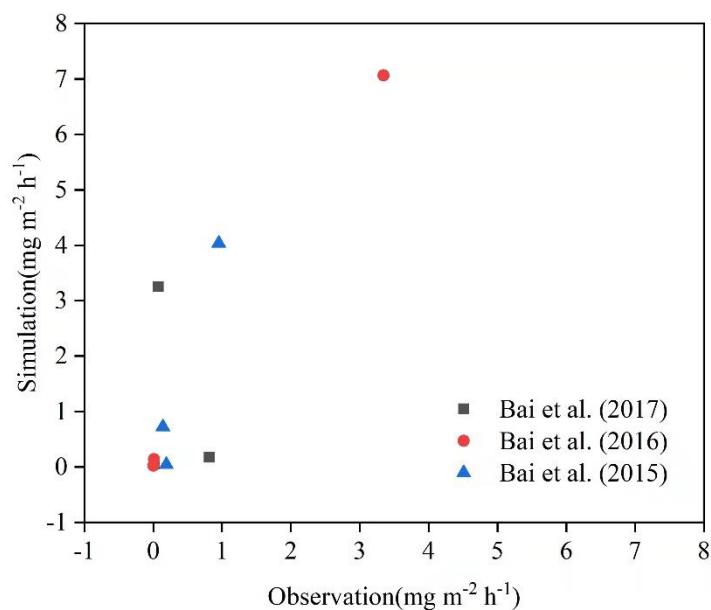


Fig. S1. Comparison of MEGAN model simulations with flux measurements in China.

References:

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Zhang, M., Zhao, C., Yang, Y., Du, Q., Shen, Y., Lin, S., Gu, D., Su, W., and Liu, C.: Modeling sensitivities of BVOCs to different versions of MEGAN emission schemes in WRF-Chem (v3.6) and its impacts over eastern China, *Geosci. Model Dev.*, 14, 6155–6175, <https://doi.org/10.5194/gmd-14-6155-2021>, 2021.

3. *Table 1: Why do you use “Year 2008” for your “HISTORY” run, but “Year 2018” for the rest of your simulations?*

Response: In the “HISTORY” simulation, we aimed to explore the impacts of interannual BVOC emission variations on O₃ and SOA formation caused by vegetation biomass variability during 1981–2018. In order to achieve this goal, we designed a scenario experiment by fixing meteorological data in one year and using the annual leaf biomass to drive BVOC estimates. The influences of annual meteorology on BVOC emissions and formation of secondary air pollutants were not considered. It is reported that vegetation change is the main driver of interannual variations of BVOC emissions (Li et al., 2020; Wang et al., 2021). The large-scale afforestation activities in recent years lead to the rapid increase of vegetation leaf biomass and therein BVOC emissions. In addition, the annual average temperature has a heating rate of 0.26 K per 10 years from 1951 to 2020 (Climate Change Center of China Meteorological Administration, 2021). Taking into account the long-term warming trend, we chose meteorological data of a mid-year 2008 over 1981–2018 as the constant input for historical simulation. The scenarios “BASE”, “BIO”, “ISOP”, “MTP”, “SQT”, and “ISOPRENOID” were designed to simulate the impacts of emissions of BVOCs, isoprene, monoterpenes, sesquiterpenes, and isoprenoids (total of isoprene, monoterpene and sesquiterpene emissions) in June 2018 on O₃ and SOA, respectively. So we used the meteorology of 2018 to drive MEGAN to estimate the emissions in 2018.

We add the explanation in the revised manuscript. Line 156–157, "For these simulations, the meteorology of 2018 was used to drive MEGAN to estimate biogenic emissions in June 2018." is added. Line 159–162, "For the meteorology, the fixing set of a mid-year 2008 over 1981–2018 were used for all the HISTORY simulations. To explore the impacts of interannual BVOC emission variations caused by vegetation biomass variability, influences of annual

meteorology on BVOC emissions and formation of secondary air pollutants were not considered." is added.

References:

Climate Change Center of China Meteorological Administration: China Blue Book of Climate Change (2021), Science Press, Beijing, China, 2021.

Li, L., Yang, W., Xie, S., and Wu, Y.: Estimations and uncertainty of biogenic volatile organic compound emission inventory in China for 2008–2018, *Sci. Total. Environ.*, 733, 139301, <https://doi.org/10.1016/j.scitotenv.2020.139301>, 2020.

Wang, H., Wu, Q. Z., Guenther, A. B., Yang, X. C., Wang, L. N., Xiao, T., Li, J., Feng, J. M., Xu, Q., and Cheng, H.: A long-term estimation of biogenic volatile organic compound (BVOC) emission in China from 2001–2016: the roles of land cover change and climate variability, *Atmos. Chem. Phys.*, 21, 4825–4848, <https://doi.org/10.5194/acp-21-4825-2021>, 2021.

*4. Line 176: Can you show a map of the simulated VOC to NO_x ratio to help you make your point. Also, there are a number of papers discussing the VOC to NO_x ratio over China, including as observed from space. For example: Jin, X., T. A. Holloway (2015). Spatial and temporal variability of ozone sensitivity over China observed from the Ozone Monitoring Instrument. *Journal of Geophysical Research Atmospheres*, 120(14), 7229–7246, doi: 10.1002/2015JD023250. Please do a literature search to see what they've concluded and if those conclusions are consistent with your study. It would also be helpful to show a spatial map of the simulated NO_x distributions.*

Response: Thank you very much for the constructive comment. In this study, we aim to explore the impact of interannual BVOC emission variations on O₃ and SOA caused by vegetation biomass variability. The impact of anthropogenic emissions was not considered. WRF-Chem model was run by fixing the anthropogenic emissions and meteorological data in a mid-year 2008 over 1981–2018. So we do not show the map of the simulated VOC to NO_x ratio and NO_x distribution, considering that there may be deviations with reality. However, we

do literature searches discussing the VOC to NO_x ratio over China to explain our results (Jin and Holloway, 2015; Lu et al., 2019; Lyu et al., 2016; Tan et al., 2018; Milford et al., 1989; Wang et al., 2008).

In the revised manuscript, we revise the relevant contents and made a more scientific explanation in line 218–235: "The spatial pattern of estimated MDA8 O₃ impacted by BVOC emissions differs from the spatial distribution of BVOC emissions mainly because of the variability of the nonlinear response relationship between O₃ formation and precursors. As the important precursors, VOCs and NO_x react in the presence of hydroxyl (OH) and hydroperoxyl (HO₂) radicals to create O₃. The O₃ formation is expected to be affected by the different levels of O₃ precursors in different land use functional areas. According to different VOCs/ NO_x ratio, O₃ formation regimes can be classified into VOC-limited (VOC-sensitive), transition, and NO_x-limited (NO_x-sensitive) regimes (Lu et al., 2019; Wang et al., 2008). From the spatial distribution of the BVOC effect (Fig. 3), the surface O₃ is sensitive to BVOC emissions in most regions in China which can furtherly indicate they are usually VOCs-limited. It confirms the conclusion made by Lu et al. (2019), Lyu et al. (2016), and Tan et al. (2018) that the VOCs-limited regime is dominant in southern China. Comparing with the spatial distribution of BVOC emissions (Fig. 1), the areas with high BVOC emissions usually have a higher contribution to O₃. Because the dense population leads to a large number of NO_x emitted by human activities, NO_x is saturated with the formation of O₃ which is more sensitive to VOC emissions. Therefore, the higher BVOC emissions usually cause greater contribution to O₃ in these areas. In the VOC-limited regime, the reduction of VOC emissions reduces the chemical production of organic radicals (RO₂), which in turn lead to decreased cycling with NO_x and consequently lower concentration of O₃ (Jin and Holloway, 2015; Milford et al., 1989). To decrease BVOC emissions by planting plants with low emission potential may contribute to O₃ pollution control. Notably, both northeastern and southern regions have the highest BVOC emissions, but their contributed O₃ differ much, which indicates that O₃ formation in the south is more sensitive to VOCs than in the northeast. Hainan province also has higher BVOC emissions but relatively lower contribution to O₃."

References:

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5. Line 192: *Can you mark (with boxes, for instance) the 5 regions on one of the maps, such as Figure 2?*

Response: Thank you for your valuable suggestion. In the revised manuscript, the five key regions are marked in Figures 4, 5, and 7.

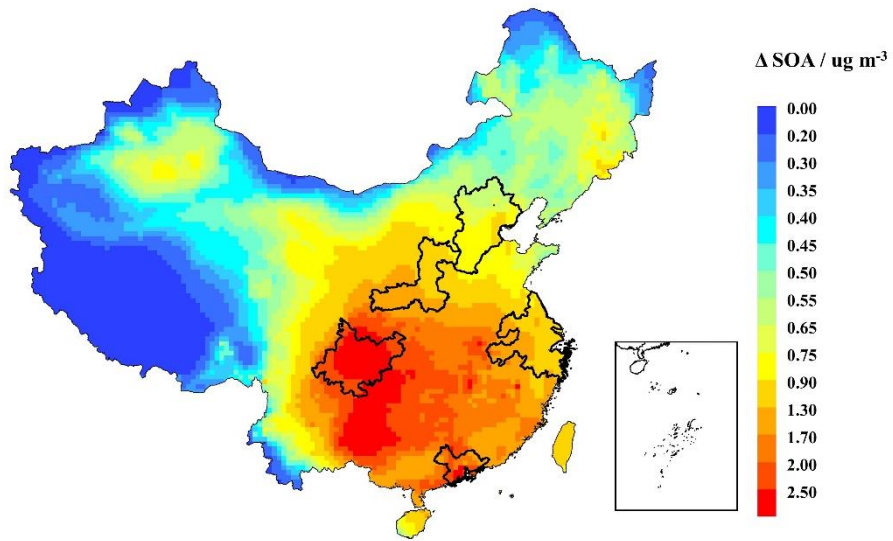


Fig. 4. Spatial variations in impact of BVOC emission on SOA concentration.

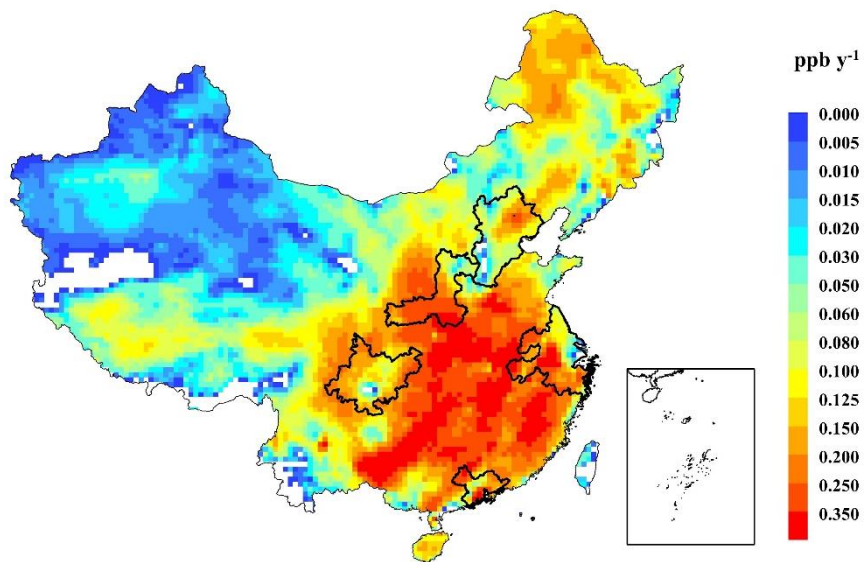


Fig. 5. Spatial distribution of interannual variations in O_3 simulated using annual BVOC emission factors.

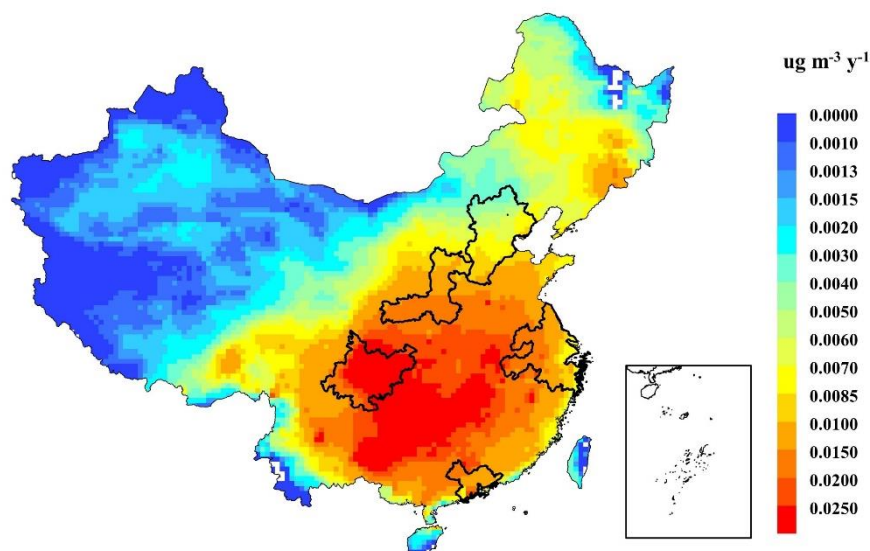


Fig. 7. Spatial distribution of interannual variations in SOA simulated using annual BVOC emission factors.

6. Line 245: “N3”? Do you mean NO₃?

Response: We appreciate your careful reading very much and are sorry for the error. In revised manuscript, line 288, it is revised to "NO₃".

7. Line 275: You don't show the BVOCs emission changes for your study in “HISTORY”. Why? A spatial map of the changes would be very helpful for the discussion.

Response: Thank you for your suggestion. In the revised manuscript, we add the detailed description of BVOC emission changes for the “HISTORY” simulation in **Section 3.1.2**. The spatial map of the changes is also added as Fig. 2. In addition, the reason for the changes is discussed. The revisions can refer to the **Response to Comment 1 of Reviewer 2**.

8. Section 3.3.1: The discussion would be facilitated by maps (e.g., in supplementary material) of vegetation changes, leaf biomass changes, emission factors, etc.

Response: Thank you for your suggestion. In the revised manuscript, we add a spatial map of leaf biomass changes over 1981–2018 as Fig. S2. The vegetation distribution in our

estimations was derived from the Vegetation Atlas of China (1:1,000,000), which provides a detailed vegetation distribution at a high resolution. It was produced based on nationwide vegetation surveys and research from the past four decades and was published in 2007. It is rarely updated, and it represents the average distribution of vegetation in China. Therefore, the same PFT distribution was used to estimate BVOC emissions for 1981–2018 in this study. Emission factors were extrapolated by the leaf-level emission rates and leaf biomass using the canopy environment model described in MGEAN2.1. Emission rates were constant by time. So the interannual change of emission factor can be explained by that of leaf biomass.

Line 187–194 in revised manuscript, we add descriptions for the changes of leaf biomass: "The leaf biomasses increased from 378.35×10^{12} g in 1981 to 1107.16×10^{12} g in 2018 at an average rate of 17.97×10^{12} g yr⁻¹. Among them, the forest and crop leaf biomass increased from 237.10×10^{12} to 518.38×10^{12} g and from 141.25×10^{12} to 588.79×10^{12} g, respectively, totally increasing by 192.63%. The spatial distribution of interannual variations in leaf biomass is presented in Fig. S2. The increase of leaf biomass is most significant in Great Khingan, Changbai Mountains, North China Plain, south and southwest China. This is mainly due to the increased stock of broadleaf and coniferous forests as a result of afforestation. Northern Qinghai-Tibet area and Northwest China have a relatively high grass cover rate but insignificant increase in leaf biomass of vegetation. It is because that the grass biomasses were the same over the historical simulations due to lacking of data."

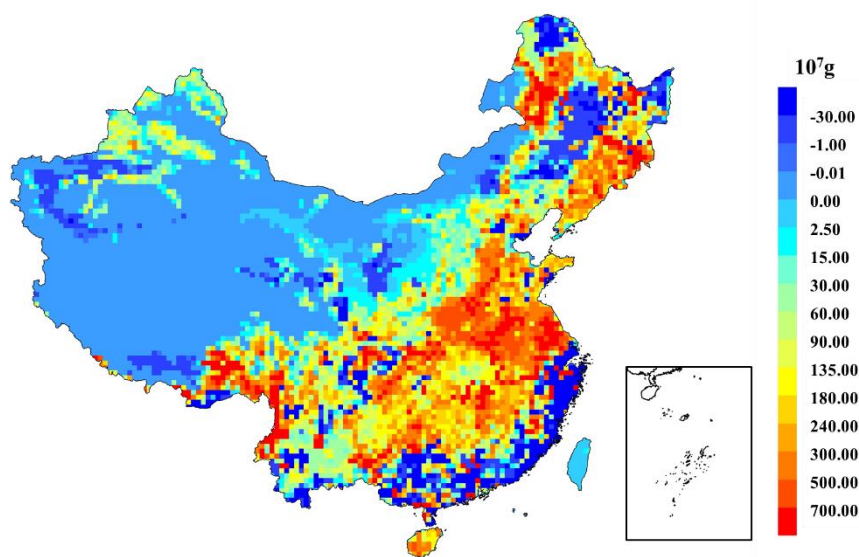


Fig. S2. Spatial distribution of interannual variations in leaf biomass.