Response to referee comment #1:

Yin et al present an emissions inventory for some BVOC across China, using the BVOC emissions model Megan and WRF-CLM. A number of previous studies have already presented emission inventories for China, as also mentioned by the authors. While their approach is somewhat different, I cannot get overly excited about the analysis as it is currently presented. It seems methodologically largely sound (but see my questions below); but apart from providing yet another BVOC emissions map there is little novelty in the study. Isoprene and MT emissions are largest from forests, most of the MT come from conifers; spatial patterns thus depend on weather as well as land cover, temporal patterns on weather and LAI; all of this has been found in many other studies before and the results are no surprise given the main BVOC algorithms that basically vary standard emission factors (which depend on the PFT) with weather.

Response: The manuscript was revised much according to three referees' comments.

The novelty in this study is that the BVOCs emission is estimated by including some PFT-specific physiological parameters. These parameters are derived from CLM4, but never considered in the previous BVOC estimation algorithms coupled in the weather forecasting models.

We found the improvements are important (more details could be found in the section 3.2). Firstly, the estimations by using leaf temperature in our study were about 20 % higher than those estimated based on air temperature as in the previous methods. Secondly, the separate treatments of sunlit and shaded leaves in this study, which affect within canopy solar radiation, lowered the estimations by a factor of 2 compared with estimates made by methods neglecting shaded canopy. Thirdly, in this study, leaf temperature and solar radiation were averaged over the past running time at each time step to estimate emission response to weather history. However, in the original code, this response was estimated based on fixed parameters. The improved representation in our study resulted in 50 % higher estimations than those based on fixed values.

The results were within a factor of 2 of most canopy-scale flux measurements and top-down isoprene inventories, indicating an overall good performance of the coupled model (section 4).

I would think that at least some simulation experiments that investigate emission changes historically and/or in future, in response to climate change, CO2 and/or land use change would be needed to warrant publication of the work. Given that the model system is in place this should not be too much effort and would make the paper some-what more interesting.

Response: The primary purpose of this study is to improve BVOC estimation in weather forecasting models by using physiological parameters provided by CLM4 land surface scheme. New discussions have been added in the section 3.2 (Page 7, Line 201) of the revised manuscript.

While I agree that high spatial resolution is an advantage when estimating BVOC emissions I do not follow the need to use a coupled WRF-CLM model version. This would have been needed if the authors wanted to do surface-atmosphere feedback experiments. But at the moment, all they do is to drive an BVOC emissions model with simulated high-resolution weather. This could have been done in offline experiments just as well, especially since the vegetation in this study is prescribed from MODIS.

Response: We reworded the Introduction section to illustrate the reason why we use CLM4 land surface scheme. The CLM4 scheme was used to provide real-time vegetation physiological factors through the parameterization of comprehensive ecological and biological processes for MEGAN. Related factors have been described in the Method section. The impacts of reasonable physiological parameter applications on estimates were discussed in section 3.2 of the revised manuscript.

Revisions: (Page 2, Line 44) "It has been demonstrated that BVOC emission rates are heavily affected by plant physiological factors and environmental conditions (Peñuelas and Staudt, 2010). Many biogenic emission models have been developed with a strong foundation in the physiological processes of a leaf (Guenther et al., 1991; Niinemets et al., 1999; Martin et al., 2000). The Model of Emissions of Gases and Aerosols from Nature (MEGAN) that estimates BVOC emission fluxes as basal emission rates modulated by emission activity factors has been intensively used for regional and global BVOC emission estimations (Guenther et al., 2006; Guenther et al., 2012). Process-based models linked BVOC production rate explicitly to leaf photosynthetic electron transport rate and election requirement for BVOC synthesis (Niinemets et al., 1999; Niinemets et al., 2002).

The MEGAN algorithms have been incorporated into Community Land Model (CLM), the terrestrial component of the earth climate system model, as one step toward integrating biogeochemical processes in the model. In the coupling of MEGAN and CLM, all the physical and biological variables required by BVOC estimation are determined by comprehensive ecological and physiological processes parameterized in CLM at each time step (Levis et al., 2003; Oleson et al., 2010; Lawrence et al., 2011). Process-based models are typically coupled within dynamic vegetation models that have a mechanistic model for leaf photosynthesis at their core (Arneth et al., 2007; Pacifico et al., 2011; Yue and Unger, 2015). In general, these coupled models are employed to investigate the long-term interactions and feedbacks between terrestrial vegetation and climate change with spin-up and simulation time from tens to thousands of years.

Instead of coupling detailed algorithms within the land surface parameterizations, a simplified version of MEGAN algorithm, the parameterized canopy emission activity (PCEEA) algorithm, has been coupled with weather and climate forecasting models as an independent module to generate online biogenic emission inventory for atmospheric chemistry simulation (Guenther et al., 2006; Sakulyanontvittaya et al., 2008;

Fu and Liao, 2012; Henrot et al., 2017). Instead of using a detailed canopy model to calculate leaf temperature and leaf-level photosynthetic photo flux density (PPFD), the PCEEA algorithm parameterizes the modification of these plant physiological variables on emission rates based on ambient temperature and canopy above solar radiation. Although leaf temperature is strongly related to ambient temperature, it is also affected by other physical and biological factors such as irradiation and evapotranspiration. Subin et al. (2011) indicated that the strong advection and boundary layer mixing during the day decoupled the air temperature from the vegetation temperature to a great extent, making daytime surface energy budget the primary controlling factors of vegetation temperature changes. Furthermore, due to the different morphological and physiological properties, relationships between air temperature and leaf temperature, and between canopy above PPFD and leaf-level PPFD, vary significantly among tree species. Since the PCEEA algorithm was based on standard MEGAN canopy model simulations for warm broadleaf forests, using the same equations for representations of other plant types leads to unpredictable uncertainties. Leaf temperature and PPFD averaged over the past 24 and 240 h are used in MEGAN algorithms to account for effects of medium-term weather history. However, the PCEEA algorithm obtains the past conditions from a prescribed climatological monthly mean dataset, which could be much different from the real meteorology (Zhao et al., 2016). Therefore, reasonable plant-specific physiological variables are needed to improve the BVOC estimation in weather models.

CLM version 4 (CLM4) was coupled and released with the Weather Research and Forecasting model (WRF), a mesoscale numerical model designed to simulate regional weather and climate, since version 3.5 as one of the land surface scheme options to better characterize land surface processes (Jin and Wen, 2012; Jin et al., 2010; Subin et al., 2011). Because MEGAN has been embedded within CLM as mentioned above, the coupling of WRF-CLM4-MEGAN allowed regional weather forecasting models to estimate BVOC emissions within a comprehensive ecological climatology framework. Besides improvements result from real-time plant physiological variables derived from land surface parameterizations, sub-grid vegetation compositions represented in CLM4 are also expected to provide a more reasonable estimation in view of the significant variability in basal emission ability among tree species. However, few studies employed the coupled mode to estimate regional BVOC emissions (Zhao et al., 2016)."

Lines 68-70: This statement is plain wrong. There are several approaches published in which authors have incorporated BVOC emission algorithms into ecosystem models that calculate also leaf gas exchange, and vegetation dynamics. See e.g. Unger et al., ACP 2013; Pacifico et al., ACP 2011, doi:10.5194/acp-11-4371-2011; Arneth et al., ACP 2007; Schurgers et al., Biogeosc. 2009. And there's presumably others. The *authors will need to do their homework more thoroughly. And since the authors didn't even use the dynamic vegetation module of CLM but prescribed land cover and LAI their claim that the study presented here is so much more 'accurate' (see line 79) than previous estimates seems overstated.*

<u>Response:</u> We reworded the Introduction section. Most studies have integrated BVOC emission algorithms into land surface and dynamic vegetation models to investigate the response of vegetation distributions and emissions to long-term climate change. The purpose of our study is to use physiological factors provided by CLM4 to improve estimates of regional BVOC emissions in weather and climate forecasting models. The significant effects of reasonable physiological parameter applications on estimates were investigated.

<u>Revisions:</u> (Page 2, Line 51) "The MEGAN algorithms have been incorporated into Community Land Model (CLM), the terrestrial component of the earth climate system model, as one step toward integrating biogeochemical processes in the model. In the coupling of MEGAN and CLM, all the physical and biological variables required by BVOC estimation are determined by comprehensive ecological and physiological processes parameterized in CLM at each time step (Levis et al., 2003; Oleson et al., 2010; Lawrence et al., 2011). Process-based models are typically coupled within dynamic vegetation models that have a mechanistic model for leaf photosynthesis at their core (Arneth et al., 2007; Pacifico et al., 2011; Yue and Unger, 2015). In general, these coupled models are employed to investigate the long-term interactions and feedbacks between terrestrial vegetation and climate change with spin-up and simulation time from tens to thousands of years."

(Page 13, Line 387) "This framework improved biogenic emission estimations by using reasonable PFT-specific physiological parameters derived from CLM4 land surface scheme to drive standard MEGAN algorithms. The simulated leaf temperature was typically higher than air temperature by 1~6 K in daytime and lower than ambient value by approximately 2 K during night. Using air temperature to parameterize BVOC emissions underestimated isoprene and monoterpene emissions in July by 23.9 % and 21.9 %, respectively. Because the sunlit fraction of broadleaf trees was typically lower than 0.3 in July, ignoring the difference in absorbed radiation of sunlit and shaded leaves overestimated emissions from broadleaf trees by a factor of 2.7. Assigning fixed values to variables that related to previous conditions made a similar estimation in January with using dynamic variables, while underestimated emissions in July by approximately 50 %. Due to the significant discrepancy caused by differences in driving variables, more reasonable parameter applications are important for accurately estimating biogenic emissions."

MEGAN defines canopy-level emission factors for multiple compounds. What is the *observational evidence, if any, for compounds such as myrcene, ocimene, sabinene etc? Where and what types of have canopy-scale measurements been made that would in fact support the value specified? And if such measurements are scarce/non-existent, what justifies their use in a large-scale inventory?*

Response: MEGAN defines emission factors based on about 300 studies. Although most studies measured emission rates of the sum of monoterpenes or of several abundant compounds such as α-pinene and β-pinene, a number of studies reported emission rates of myrcene, ocimene, sabinene based on enclosure or canopy-scale measurements, such as Geron et al. (2000); Harrison et al. (2001); Kim (2001); Dindorf et al. (2006); Holzke et al. (2006); Bai et al. (2012) etc.. Compared with studies based on enclosure method, much less studies offering emission rates of the above monoterpene species were based on canopy-scale methods. More research on these compounds are needed.

In our study, the observed isoprene emission factors in China, as well as those used in previous studies were used to determine isoprene emission factors. References were listed in the Supplement Table 1. Due to the scarce reports of local monoterpene emissions factor, emission factors of monoterpene species were roughly estimated based on global average emissions factors suggested in Guenther et al. (2012) and modified by the ratio of local to global isoprene emission factors. Emission factors is a significant source of uncertainty due to the difficulties in conducting large-scale and long-term measurements, which has been recognized in the Uncertainty section.

Revisions: (Page 4, Line 107) "PFT-specific EFs of isoprene were determined based on observations conducted in China and EF used in previous studies (as shown in Table S1). Due to the lack of detailed monoterpene EFs reports, the EFs of main monoterpene species were determined by scaling default MEGAN EFs with the ratio of local isoprene EF to default value presented in Guenther et al. (2012). Table 1 summarizes the EFs for each vegetation type used in this study."

Guenther et al. in their 2012 paper claim that they describe how leaf age fraction is estimated in section 2.4 of their paper, but section 2.4 has no mentioning of leaf age calculation at all (neither has any other section in their manuscript). The authors need to describe how leaf age (new, growing, mature and senescing categories) can be differentiated (realistically) from a remote sensing product such as MODIS.

Response: Accepted. Details of emission activity algorithms can be found in Guenther et al. (2006). The determination of leaf age fraction was described in the section 3.2.2 of their paper. The division of leaf age fraction is based on the change in LAI between the present time step and the previous time step. The MODIS 8-day LAI data was used in our study. Every 8 days used the same MODIS LAI database and LAI of the previous time step was set as the previous 8-day MODIS LAI data. We added the reference and brief descriptions in our Methods and Data section.

Revisions: (Page 4, Line 112) "The emission activity factor for each compound (γ_i) accounts for emission responses to solar radiation, leaf temperature, LAI, leaf age and soil moisture. The effects of variations in CO_2 concentration was neglected in this study because the simulation was performed for only one year and Heald et al. (2009) found that accounting for CO_2 inhibition has litter impact on predictions of present-day isoprene emission. Details of the algorithms could be found in Guenther et al. (2006) and Guenther et al. (2012)."

(Page 6, Line 167) "The same LAI data was used as current LAI (LAI_c) for 8 days and the past 8-day image was considered as LAI of the previous time step (LAI_p). The changes between LAI_c and LAI_p was used to determine leaf age (Guenther et al., 2006)."

Worden et al (ACP, 2019, https://doi.org/10.5194/acp-19-13569-2019) use MOPITT CO to infer isoprene emissions. While they don't present numbers for China, I wonder if it would not be worthwhile to approach these authors to find out if an emission map for China could be obtained in order to compare with the simulations shown here.

Response: Accepted. Worden et al. (2019) estimated biogenic CO flux based on MOPITT CO observations. They evaluated the seasonal and spatial patterns of the posterior CO against top-down estimates of isoprene fluxes based on OMI formaldehyde observations (Stavrakou et al., 2015; Bauwens et al., 2016). We evaluated our results against the top-down inventory based on OMI observations in the revised manuscript.

Revisions: (Page 12, Line 355) "Formaldehyde (HCHO), as a major intermediate product in the degradation of isoprene in the atmosphere, has been widely used as a proxy for estimates isoprene emissions. Fu et al. (2007) used a continuous 6-year record (1996–2001) of Global Ozone Monitoring Experiment (GOME) HCHO columns and made a top-down estimate for isoprene as 12.7 Tg yr^{-1} in China, which is comparable to our model outputs. However, Stavrakou et al. (2014) inferred isoprene emissions by inversion of GOME-2 HCHO columns and the satellite-derived emissions were found to decrease from 8.6 Tg in 2007 to 6.5 Tg in 2012, lower than emissions in this study by 33.7 %–76.9 %. Stavrakou et al. (2015) assessed the consistency of the top-down isoprene fluxes based on GOME-2 and OMI (Ozone Monitoring Instrument) HCHO observations. The isoprene emission from China in 2010 was estimated to be 5.9 Tg with GOM2-2 and 6.5 Tg with OMI data, with large discrepancies in southern China. Aside from the influence of difference meteorological conditions and land cover changes during the past years, the reliability of satellite-based constraints also needs to be improved because that the HCHO is affected by non-isoprene sources plus uncertainty and spatial smearing in isoprene-formaldehyde relationship (Fu et al., 2019)."

The section 'uncertainties' is somewhat thin. Nothing in it is wrong but it doesn't provide a lot of substance and the section reads a bit as an after-thought at the moment. Needs more concrete examples and/or even example simulations. And the section 'conclusions' is in fact merely a summary section, which could be removed, as the text contains a lot of repetition of what has been said elsewhere in the manuscript.

Response: We revised the Uncertainty and Conclusion section.

Revisions: (Page 12, Line 367) "The major areas of uncertainty include basal emission factors, land cover data (vegetation distribution and LAI) and physiological and environmental parameters.

The basal emission factors have been identified as the most important uncertainty source (Zheng et al., 2010; Situ et al., 2014; Wang et al., 2016). Local emission factors for isoprene reported by previous observations conducted in China were used in this study. Since measurements of the monoterpene emission factors are scarce, we calculated local emission factors based on the ratio of local isoprene emission factor to default emission factor in MEGAN literature. There are large uncertainties associated with the conversion approach. More in-situ observations on emission rates of different PFTs in China are required.

The conversion from IGBP land category to PFTs used in CLM4 resulted in uncertainty in vegetation map. For example, broadleaf deciduous trees in CLM4 are group into tropical, temperate and boreal tree categories, with specified canopy parameterizations. The climate zone separations were not considered in our study and all broadleaf deciduous trees in IGBP were grouped into broadleaf deciduous temperate tree category in CLM4. The conversion method may lead to uncertainties when parameterizing trees or shrubs in boreal and tropical area. Wang et al. (2018) adopted three LAI datasets to study the effect of the LAI input on BVOC emissions and indicated that the discrepancies between different LAI inputs do not obviously affect the estimates.

CLM4 parameterizes one layer of canopy, however, solar radiation is attenuated by foliage and leaf temperature varies among layers. A relatively simple representation of canopy is also a source of uncertainty. Guenther et al. (1995) found a less than 5 % difference in global annual isoprene emission estimated with one or five lays and no change in the estimations of other BVOC emissions, suggesting that BVOC emissions are relatively insensitive to the number layers. However, many studies indicated that the treatment of microclimatic factors such as light and leaf temperature within the canopy resulted in substantial difference in estimated emissions (Keenan et al., 2011)."

(Page 13, Line 387) "This study estimated the emission budgets and spatial-temporal patterns of BVOC in China in the year 2018 using the WRF-CLM4-MEGAN modeling system. This framework improved biogenic emission estimations by using reasonable PFT-specific physiological parameters derived from CLM4 land surface scheme to drive standard

MEGAN algorithms. The simulated leaf temperature was typically higher than air temperature by 1~6 K in daytime and lower than ambient value by approximately 2 K during night. Using air temperature to parameterize BVOC emissions underestimated isoprene and monoterpene emissions in July by 23.9 % and 21.9 %, respectively. Because the sunlit fraction of broadleaf trees was typically lower than 0.3 in July, ignoring the difference in absorbed radiation of sunlit and shaded leaves overestimated emissions from broadleaf trees by a factor of 2.7. Assigning fixed values to variables that related to previous conditions made a similar estimation in January with using dynamic variables, while underestimated emissions in July by approximately 50 %. Due to the significant discrepancy caused by differences in driving variables, more reasonable parameter applications are important for accurately estimating biogenic emissions. Using the CLM4-MEGAN framework, the annual emissions of BVOC in China was estimated to be 14.7 Tg C, with isoprene and monoterpenes accounting for 78.3 % and 21.7 % of the totals, respectively. The coupled model successfully reproduced the spatial and temporal patterns of BVOC emissions. Although past studies reported that emissions peaked in July, the highest emission was found in August in our research. This result was consistent with observations, indicating that the past-time leaf temperature and solar radiation must be considered in emission estimation. The predicted values in forest areas during wet seasons were within a factor of 2 of observed values, however, significant discrepancy was found in estimations under drought conditions. The predicted annual emission was within a factor of 2 of top-down estimates and at the upper end of values reported in previous modeling estimates. Comparisons indicated an overall good performance of the model during dominant BVOC emission seasons, but further efforts focusing on improving estimation of emissions in dry areas are still needed."

Scientific papers should be apolitical. Yet I note that Taiwan (Republic of China) is shown on the maps presented. Moreover, while not all of the islands in the South China Sea are shown on these maps (the Paracel islands and Pratas seem to be in-cluded) there are several occasions in the paper in which the authors state 'the small is-lands in the South China Sea are not included', implying that these should be counted. These islands are contested territory and the political status of Taiwan is also a non-consensual one. The authors write about emissions from China, and not emissions from the People's Republic of China, but in the day-to-day use of China the term is synonymous with PRC. I am concerned that misunderstanding might arise from this, and therefore suggest to restrict the reporting of numbers and maps to mainland China only.

Response: All the coauthors of this paper are Chinese researchers. From our perspective, Taiwan and the South China Sea are shown on the map to ensure that the paper is apolitical. Thank you very much for your understanding.

Line 36 – sounds as if global BVOC emissions are known to be 1150 Tg C a-1, which is not the case. There are huge uncertainties and no global observations. Revise.

Response: Accepted.

Revisions: (Page 2, Line 37) "Globally speaking, biogenic volatile organic compounds (BVOCs) emitted by terrestrial vegetation are estimated to be 500 ~ 1100 Tg C yr^{-1}, corresponding to about 90 % of the emission total (Guenther et al., 1995; Henrot et al., 2017)."

Line 50 '. . .that cannot be reproduced well in canopy environment models'. Unclear what is a canopy environment model.

Response: We reworded the Introduction section. Canopy environment models typically consist of radiative transfer and energy balance simulations to calculate solar radiation reflected and absorbed by the canopy, its transfer within the canopy, as well as leaf temperature of different layers of the canopy.

Lines 62, 66 and possibly elsewhere: hydrological cycle (not hydrologic cycle).

Response: Accepted.

Revisions: (Page 4, Line 99) "The CLM4 was coupled and released with WRF since version 3.5 as one of the land surface scheme options. CLM4 consists of components related to land biogeophysics, hydrological cycle, biogeochemistry, human dimensions and ecosystem dynamics."

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