Responses to Reviewers on "Impacts of historical climate and land cover changes on tropospheric ozone air quality and public health in East Asia over 1980–2010" by Y. Fu and A. P. K. Tai. (MS No.: acp-2015-249)

We would like to thank the reviewers for the thoughtful and insightful comments. The manuscript has been revised accordingly, and our point-by-point responses are provided below. The reviewers' comments are *italicized*, and our new/modified text is highlighted in **bold** below, and highlighted in blue in the manuscript.

Response to Anonymous Referee #1

On the whole this is an interesting and scientifically valuable paper that is worthy of publication in ACP. It is clearly structured, well written, appropriately illustrated and is commendably concise. The introduction is particularly clear and well written. I have highlighted a few points below that need to be addressed, but these are relatively minor in nature.

General Comments:
My principle concern regards the robustness of the results. The study actually addresses the difference between two five-year periods, and it is not clear how well this represents the changes over 1980-2010. How variable is the surface ozone within each 5-year period, and how does this variability compare with the difference between the periods? Are the spatial distributions and magnitudes of the changes robust if 3-or 4-year periods are selected from within each 5-year run to compare? Two specific years are selected for LAI (1982 and 2010), but how representative are these years of their respective 5-year periods? A brief exploration and discussion of these issues is needed to convince the reader that the results described are solid and robust.

We have examined the interannual variations of surface ozone concentration within each 5-year period based on the simulations CTRL and simulations COMB. Mean absolute deviation (MAD) values are used to quantify the interannual variations, which are shown in Fig. A. We now include this in the main text as follow:

(Sect. 6, P12, L383-389): "**We also examine the interannual variations of surface ozone concentration within each 5-year period based on the simulations CTRL and COMB, which are quantified using the mean absolute deviation (MAD) (Supplement Fig. S5). We find that the interannual variations vary within the range of 0.2-3.0 ppbv across East Asia. Therefore, in comparison with such variations, the changes in surface ozone induced by climate and LCLU changes in this study are shown to be significant.**"

We also check the differences in ozone by comparing the results from selected 3-year periods with those changes obtained from this study. As shown in Fig. B, the spatial distribution and the magnitude of the changes derived from 3-year simulations are in accord with the results presented in our manuscript (Figure 4), indicating that the results in this study are reasonable and robust.

To examine the impacts of LCLU changes on air quality, we use MODIS-derived land cover dataset with the classification scheme of IGBP as a basis for producing the LCLU between 1980 and 2010. LAIs in year 1982 and 2010 are chosen to represent land cover change because the satellite-based LAIs from Liu et al. (2012) are not available for year 1980 and early 1981. We now also include a brief discussion in the main text as follows:

(Sect. 2.2, P7, L226-L229; P8, L232-235): "… satellite data with a resolution of half month and 8 km (Liu et al., 2012). **To represent land cover change, LAIs in year 1982 and 2010 are chosen in this study because the satellite-based LAI datasets are not available for the year 1980 and early 1981, and LAIs from these years are consistent with the average over each 5-year simulation period.** Monthly mean LAIs are then averaged… **The impact of interannual variations of vegetation density within the 5-year period is not explicitly included in this study, but such impact on ozone is shown to be relatively small (less than 0.5 ppbv) (Fu and Liao, 2012)**."

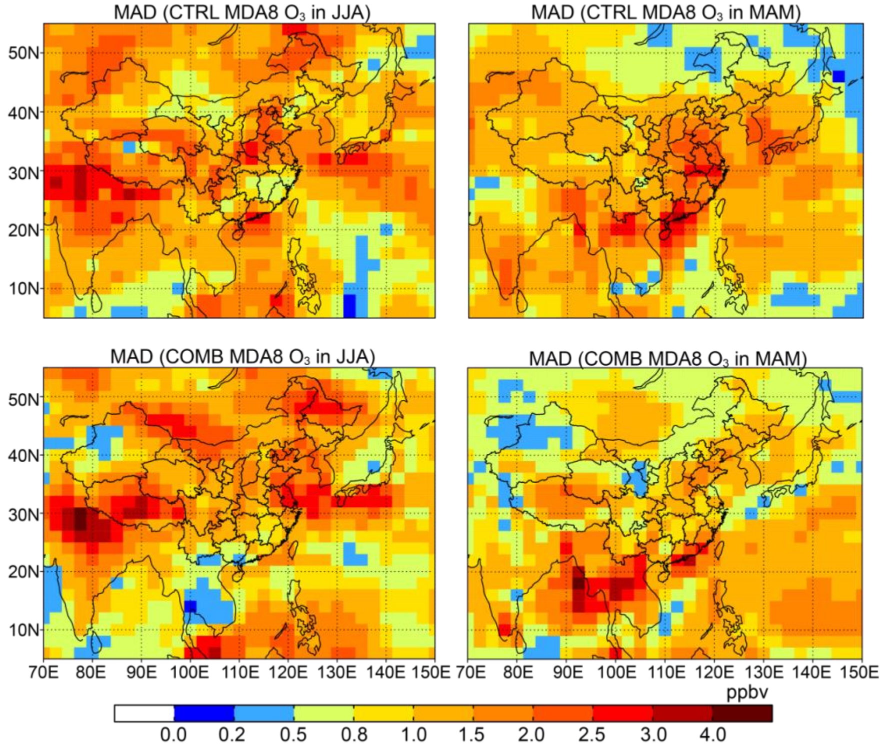

Fig. A (also Supplement Fig. S5). Mean absolute deviation (MAD) of surface ozone in JJA and MAM from the simulations CTRL (2007-2011) and the simulations COMB (1981-1985).

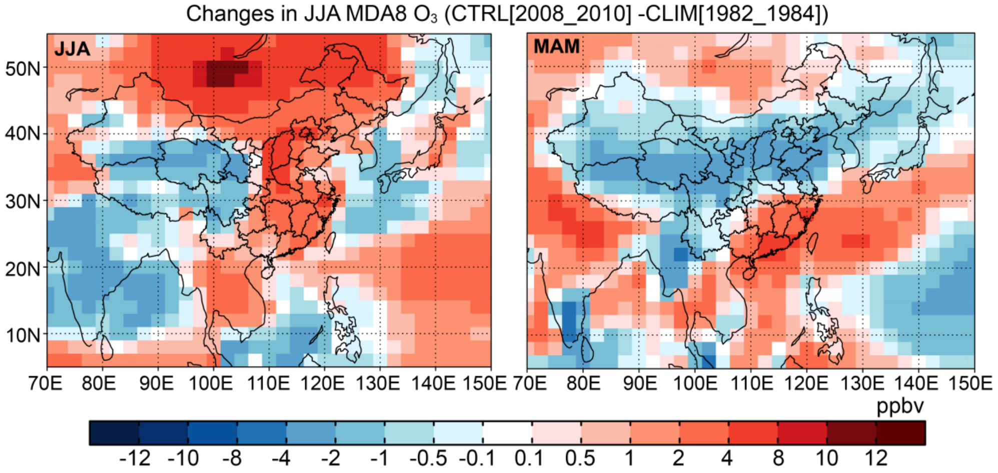

Fig. B. Changes in surface ozone concentration in JJA and MAM as a result of climate change alone between the 3-year periods 1982-1984 and 2008-2010.

The climate-driven ozone changes presented in Fig 4 appear very large (although they seem consistent with the large temperature and humidity changes identified). Is this just is a consequence of the short periods considered? Is so, then the difference between these periods does not represent longer-term climate changes realistically. How do the climate changes compare with other assessments over this part of the world?

The meteorological parameters used to drive the simulations are from Modern Era Retrospective-analysis for Research and Applications (MERRA), which are produced by the NASA Goddard Earth Observing System Data Assimilation System Version 5 (GEOS-5), focuses on historical analysis of the hydrological cycle on a broad range of timescales and covers the modern satellite era from 1979 to present.

As an example, we evaluate the MERRA reanalyzed surface air temperature in JJA by comparison with the observed temperature from weather stations in China (http://cdc.nmic.cn/dataSetLogger.do?changeFlag=dataLogger#) and NECP/NCAR reanalysis for years 1981-1985 and 2007-2011. The comparisons show that the MERRA surface temperatures agree fairly well with the observations from weather stations in China (Fig. C). We also compare the changes in 5-year average and 10-year average surface air temperature in JJA between the two periods (Fig. D). The distribution of the changes in JJA temperature using 5-year average are similar to those using 10-year average temperature, despite the slightly high bias in 5-year average case in some regions. The magnitudes of the temperature changes are broadly consistent with that of the observed temperature especially for most of the eastern half of China, despite regional discrepancies around Shangdong province and in many parts of western China (which are not the major ozone pollution regions).

We now include these information in Sect. 2.1 (P5, L152-155): "**Comparisons of MERRA surface temperature (including its changes) with surface weather stations in China and NCEP/NCAR reanalysis show good agreement especially for most of the eastern half of China, reflecting a robust multidecadal trends.**"

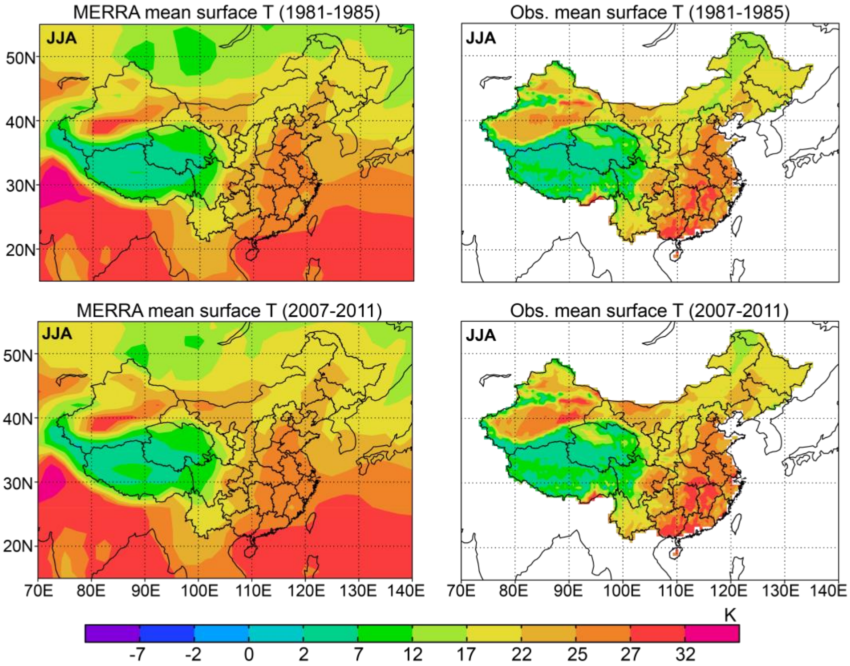

Fig. C. MERRA reanalyzed and observed surface air temperature in JJA for years 1981-1985 and 2007-2010 over China.

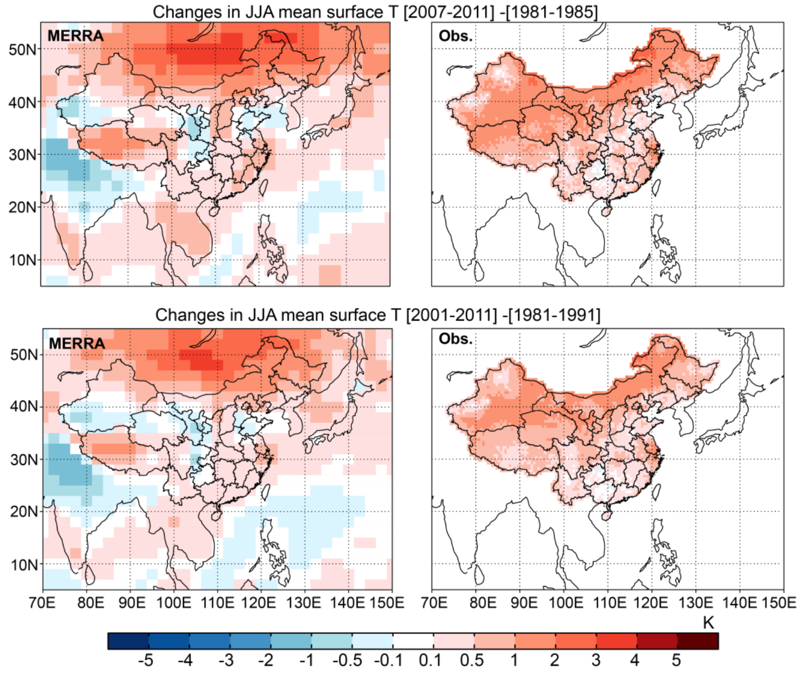

Fig. D. Changes in MERRA reanalyzed and observed surface air temperature in JJA over China between 1981-1985 and 2007-2011(upper panel), and between 1981-1991 and 2001-2011(lower panel).

Substantial additional information is provided in the supplement, and some of this information could usefully be included in the main text of the paper. A few additional summarizing comments could be added to Section 5 of the paper to help interpret the effects of climate changes, for example.

The changes in relative humidity and planetary boundary layer (PBL) as a result of the changes in climate alone ([CTRL]–[SIM_CLIM]) were shown in the supplementary materials, but now we would show it in the main text in Figure. 4 (e) - (h). The caption of Figure 4 is revised to “Changes in (a) surface maximum daily 8-hour average ozone concentration (MDA8 O_3) in summer (JJA); (b) surface MDA8 O_3 in spring (MAM); (c) mean JJA temperature; (d) mean MAM temperature; **(e) mean JJA relative humidity; (f) mean MAM relative humidity; (g) mean JJA planetary boundary layer (PBL); and (h) mean MAM PBL** driven by 1980-2010 changes in climate alone ([CTRL] – [SIM_CLIM]). Values are differences between the five-year averages over the present-day and historical periods.” (P23, L750-752; P27)

We also add the following discussion in Sect. 5 (P11, L358-363): “**We further investigate the impact of individual meteorological variable on surface ozone by comparing the results from [CTRL_2010] with the sensitivity simulations [SIM_TMP] and [SIM_RH] (Supplement Sect. S4). Both the temperature-driven or relative humidity-driven ozone changes are consistent with the large temperature and humidity changes identified, indicating their significant roles in ozone formation and destruction.**”

Specific Comments:

p.14116, l.14: What emissions were used for 1985, and what measures were taken to ensure that they were consistent with the 2005 inventory described? Also, how was methane treated in these studies: consistent with the climate period, the anthropogenic emissions, or fixed?

As suggested by the reviewer, we now include these information in Sect. 2.1 (P6, L166-175): “In this study, anthropogenic emissions of **SO_2, NO_x, and NH_3** in Asia are taken from Streets et al. (2003; 2006) and are scaled to 2005 levels. **To quantify the impact of anthropogenic emission changes, emissions for SO_2, NO_x in Asia are then scaled to 1985 levels. The scaling factors for SO_2 and NO_x are based on economic data and energy statistics as described by van Donkelaar et al. (2008). Emission for NH_3 is scaled to 1980 level by a ratio derived from historical changes between 1980 and 2003 in the Regional Emission Inventory in Asia (REAS) (Ohara et al., 2007). Methane concentrations used are fixed throughout the troposphere to annual zonal mean values in four latitudinal bands and is not determined by emission inventory.**”

p.14117, l.28: What measures were taken to ensure the self-consistency of the PFT definitions across the period? Different classification of PFTs into the limited MEGAN and Wesely categories may lead to inconsistencies if the sources of PFT data differ.

The concerns are addressed in the supplementary materials, including an explicit conversion scheme between PFTs from different datasets. To clarify, we now include such information in the main text as follows:

(Sect. 2.2, P7, L201-L221): “To examine the impacts of historical changes in land cover and land use (LCLU) on air quality, we derive model-specific land cover inputs for East Asia **between 1980 and 2010** using the Moderate Resolution Imaging Spectroradiometer (MODIS) land cover product (MCD12Q1) **with the scheme of International Geosphere-Biosphere Program (IGBP) as the baseline, which has 17 land cover types** including 13 vegetation classes and 4 non-vegetated land types. **To ensure the self-consistency of the PFTs across the**

period, we assume that the definition (vegetation composition) for each PFT remains unchanged. To obtain the land cover types used in the model, we first combine the MODIS-IGBP in year 2010 with the Koppen main climate classes following Steinkamp and Lawrence (2011). A new land cover map MODIS-IGBP-Koppen in year 2010 with 23 land cover types is developed, which is required in simulating soil NO_x emission. The distribution of LCLU types in 2010 are shown in Supplement Fig. S1. **The method we use to reconstruct LCLU in 1980 is similar to that of Liu and Tian (2010), and is based on the MODIS-IGBP-Koppen LCLU in year 2005 (derived similarly as with 2010) as base year and applies appropriate ratios to scale up/down the 2005 data, with the sum of fractional coverages of all land types including bareland of each grid cell always constrained to unity (see Supplement Sect. S1 for details).** For biogenic VOC emissions, we merge the 23 PFTs into the 5 PFTs used by MEGAN (broadleaf trees, needleleaf trees, shrubs, crops and grasses). **The details for the merging scheme are shown in Supplement Table S2. For calculating dry deposition, the model uses the Olson land map with 74 land types. Hence, we assign an Olson land type to each of the 23 land types in MODIS-IGBP-Koppen that matches the best (Supplement Table S3).**"

p.14119, l.27: Changes in agricultural practices are suggested here. Do you have any suggestions for what these might be? The changes in seasonality affect the seasonality of ozone, so some interpretation here would be particularly valuable.

We now mention the likely agricultural practices reported by some of previous studies (S. Liu et al., 2010; Sangram, 2012; Hou et al., 2015) in Sect. 3.

(P9, L283-287) Sect.3: "......changes in agricultural practices **such as the earlier end of spring harvest season in semiarid drylands of India (Sangram, 2012), the clearance of forests and brushes before crop and timber production through fire burning in Southeast Asia (S. Liu et al., 2010), and structural adjustments of agriculture in eastern China (Hou et al., 2015).**

p.14120, l.9: If the region is VOC-limited, then increased isoprene emissions should increase ozone. The explanation here needs to be clearer.

(P9, L297; P10, L298-301) We have revised the sentence as "**Much of China east of ~100 °E is in a high-NO_x, VOC-limited regime. For example, in much of central China and Japan, enhanced isoprene emission should increase ozone production, but the decreases in ozone in those regions indicate that enhanced isoprene emission might play a smaller role in affecting ozone than enhanced dry deposition, which decreases ozone.**"

p.14123, Section 7: This section is rather too brief given that the health impacts are highlighted in the title of the manuscript. The section would benefit from a brief description of how the health effects were calculated (this can be taken from the Supplement).

We have added more description of the approach that we used to calculate the health effects in the main text.

(Sect.7, P13, L411-422): "**Because there are very limited studies reporting long-term ozone-related mortality in East Asia, we apply epidemiological concentration-response functions (CRFs) from American Cancer Society (ACS)**

in this study following the methods of Anenberg et al. (2010) and Silva et al. (2013). The estimates of excess ozone-related respiratory mortality (ΔM, in 1000 deaths per year per squared km) for all adults aged 30 and above are calculated by

$$\Delta M = y_0\left(1 - e^{-\beta\Delta X}\right)P$$

where y_0 represents the baseline mortality rate (deaths per thousand people per year), β is a concentration-response factor, ΔX represents the differences in ozone concentration in terms of April-September 6-month averaged of 1-h daily maximum ozone concentration (Jerrett et al., 2009), and P is the exposed population (people per squared km). Please see Supplement Sect. S6 for details."

Minor Comments:

l.14113, l.6: Add "with" after "albeit"

(P3, L65) Added.

l.14118, l.12: It is not necessary to list the five simulations here, as they are already listed in the table and are described adequately in the following sentences.

(P8, L238-239) Deleted as suggested.

l.14122, l.5: parenthetic alternatives should be avoided, please remove "(reduced)" and "(lower)" and/or rephrase the sentence.

(P11, L365; P12, L366) Deleted as suggested.

l.14130, l.9: The Raquel reference lists many of the authors first names in place of their surnames! (This also needs correcting in the Supplement).

(P13, L415, P19, L658-663; Supplement P7, L99; P13) Revised.

l.14135, Figs 2 and 3: panel (c) has units of cm/s, should these be mm/s? Panel (d) needs an area unit, are these per grid square?

(P25, P26) Due to an oversight during plotting, the wrong units of "10^{-2} m/s" in Figs 2 and 3 panels (c) were published. The correct units are "10^{-2} cm/s", and the correct units are shown in Fig. 2 (c) and Fig. 3 (c). The units in Fig. 2 and 3 panel (d) are revised to "Gg/grid/yr" as suggested.

p.14139, Fig 6 incorrectly duplicates Fig 5; this has already been corrected.

(P29) Corrected, and thanks.

Response to Anonymous Referee #2

The paper is clear and well written and details an original study, with very interesting results and keys for discussion. I have several, generally minor, comments, mostly with the objective to clarify some aspects of the methodology or the limitations associated with the results presented in this study, which I warmly recommend for publication in ACP.

General comments:
The term "historical", used in several places of the paper, especially in the title, is misleading, since it usually refers to long-term changes, suggesting that long-term simulations (in this case simulations from 1980 to 2010) are performed, which is not the case here. I would recommend to change the text accordingly, in the title but also in the abstract and other parts of the manuscript, to make it clearer that changes BETWEEN 1980 and 2010 are investigated, and not OVER the whole period.

> (P1, L1-2) They are changed throughout the manuscript, and now the title is changed to "**Impacts of climate and land cover changes on tropospheric ozone air quality and public health in East Asia between 1980 and 2010**".

Section 2.1, NO_x emission calculation: when estimating NO_x emissions from soils, was a change in fertilizer use and quantity actually considered between 1980 and 2010? If not, how was the consistency between crop location, where most of the fertilizers would be used, and NO_x distribution, insured when changing the vegetation distribution? This could affect strongly emission levels, and therefore affect ozone concentrations as well, which should be discussed in the text and especially in the conclusion-discussion section.

> We assume the reservoir of nitrogen associated with manure and chemical fertilizer remains unchanged between 1980 and 2010 by using the fixed inventory for fertilizer and manure emissions from Potter et al. (2010), because it is very difficult to resolve the change in manure and chemical fertilizer with limited information in crop-specific harvested area and fertilizer statistics.
>
> We agree with the reviewer that changing vegetation distribution would affect the fertilizer use and quantity, and neglecting such changes between 1980 and 2010 could influence soil NO_x emission and therefore ozone in NO_x-limited regions. We revise the description of soil NO_x emission scheme in Sect. 2.1. We also add discussions in relation to the uncertainty induced by the unchanged chemical fertilizer and manure in Sect. 8.
>
> (P6, L185-190) Sect. 2.1: "Soil NO_x emission follows Yienger and Levy (1995), with updates from Hudman et al. (2012). It considers biome-specific emission factors, **a continuous dependence on temperature and soil moisture**, the latest gridded inventory for fertilizer and manure emissions, the timing and distribution of nitrogen fertilizer based on satellite-derived seasonality, **modified length and strength of pulsed nitrogen emissions, and fertilization effect of nitrogen deposition to natural soils.**"
>
> (P15, L492-496) Sect. 8: "**Our study also does not account for the changes in manure and chemical fertilizer associated with changes in LCLU and agriculture practices (Potter et al., 2010), which could affect soil NO_x emission and ozone concentration, though such effects are expected to be relatively**

minor given the VOC-limited regions prevalent in most of China."

Section 2.1, dry deposition scheme: Resistances are used in the Wesely scheme to calculate dry deposition of chemical gases over surfaces. Resistances related to vegetation (stomatal, cuticular, mesophyll) can be significantly variable from one plant species to another. Was the change in those resistances values considered in the model when changing the vegetation distribution and if not, could the authors precise the limitation they would expect, as dry deposition is shown in this study to be a key driver of ozone change?

As the reviewer pointed out, surface resistance for calculating dry deposition is associated with land types. The resistance values for 11 land types are fixed when we change the vegetation distribution between 1980 and 2010, because the vegetation composition for each land type is considered unchanged in this study (see above). In this case, changes in dry deposition result from changes in vegetation distribution and density only. As suggested by the reviewer, we now state this limitation in Sect. 8.

(P16, L509-513) Sect. 8: "**In this study we assume the vegetation composition for each vegetation type and the resistance values for each dry deposition land type remain unchanged between 1980 and 2010. How compositional changes in each PFT in response to future environmental changes will affect air quality definitely warrants further investigation.**"

Values for LAI are derived from satellite observations, and therefore do not integrate the variation from one vegetation types to another. However, the distribution of LAI between high and low emitters of biogenic VOCs could have an impact on the eventual emissions. Could the author clarify and give a bit more details on how the LAI was considered in the model (was only one LAI actually indeed considered for the whole grid or was a species-distribution taken into account) and the possible uncertainties related?

MEGAN does not treat LAI in each grid as a uniform quantity but assumes that foliage covers only the part of the grid containing vegetation (referred to as LAI_v). LAI_v is then used for calculating the biogenic VOC emissions in model. As the reviewer said, different PFTs can have different LAI values and spatiotemporal distribution. However, MEGAN as embedded in GEOS-Chem only resolves the grid average LAI but not PFT-specific LAI. We also compare the LAI used in this study and the weighted average LAI from PFT-specific LAIs simulated by LPJ Dynamic Global Vegetation Model (DGVM), and the comparisons show that the changes pattern of LAI in this study generally agree with those changes from PFT-specific LAIs. It is difficult to quantify the uncertainties in biogenic VOC emissions using gridded LAI vs. PFT-specific LAI. Guenther et al. (2006) suggested that the estimates of isoprene emission using MODIS LAI are generally ~20% lower than that using the other LAI datasets (e.g., from AVHRR or simulated from dynamic vegetation models). To clarify, we have revised the following sentences in Sect.2.1, Sect. 2.2, and Sect. 3.

(P6, L178-183) Sect.2.1: "Emissions of VOC species **in each grid cell**, including isoprene, monoterpenes, methyl butenol, sesquiterpenes, acetone and various alkenes, are simulated as a function of **canopy-scale emission factors** modulated by environmental activity factors to account for changing temperature, light, leaf age and LAI. **The gridded canopy-scale emission factors are determined by the**

weighted average of PFT-specific emission factors and PFT fraction in each grid."

(P8, L232) Sec.2.2: "Monthly mean LAIs are then averaged over the fraction of land area covered by vegetation in the model grid cell following the approach of Guenther et al. (2006) and Müller et al (2008), which are then **used in the calculation of biogenic VOC emissions.**"

(P9, L273-276) Sect. 3: "**The pattern of satellite-derived LAI changes used in this study generally agrees with the changes derived from PFT-specific LAIs simulated by these vegetation models between 1980 and 2010.**"

Section 2.1, page 14116, line 22-24: Please explain "but as implemented by Barkley et al. (2011)". What does this imply specifically for the model integration and BVOC emission calculations or the model generally? Are GEOS-Chem and MEGAN coupled or is MEGAN actually embedded in GEOS-Chem, or running separately and calculated emissions therefore used as forcing?

(P6, L177-178) The MEGAN module is embedded in GEOS-Chem. To clarify, we have revised the sentence in Sect. 2.1:"Biogenic VOC emissions are computed by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1 (Guenther et al., 2006; 2012), **which is embedded in GEOS-Chem.**"

Section 3: Atmospheric CO_2, vegetation, and biogenic emissions: For a better understanding of possible impact of changes in atmospheric CO_2 concentrations on LAI, please specify the CO_2 levels in 1980 and 2010. Regarding BVOCs, changes in atmospheric CO_2 concentrations have been demonstrated to be potentially a strong driver of plant emission capacity in the case of isoprene (Possell et al. 2005 and Wilkinson et al. 2009 for instance), with plant capacity decreasing when atmospheric CO2 increases. Has this inhibition effect been considered in this study when calculating isoprene emissions and if not, what would be the related uncertainty? This really need to be addressed in this section, and discussed in the conclusion as well.

The effect of CO_2 inhibition on isoprene emission is not considered in this study. According to the records given by the NOAA Earth System Research Laboratory Global Monitoring Division (http://www.esrl.noaa.gov/gmd/ccgg/trends), the globally averaged marine surface annual mean CO_2 levels for the periods 1981-1985 and 2007-2011 are 342.5 ppmv and 386.6 ppmv, respectively. Here for example, we apply the empirical CO_2-isoprene relationship of Possell and Hewitt (2011) to estimate the changes in isoprene emission over China between the simulations CTRL and simulations COMB. Without the CO_2 effect, climate and LCLU change enhances isoprene emission by 14% in China. While the inclusion of CO_2 effect, climate and LCLU change leads to a 3% enhancement in isoprene emission over China. We now discuss this uncertainty in Sect. 8.

(P15, L497-501; P16, L502-503) Sect. 8: "**Previous studies have indicated that ambient CO_2 level could affect isoprene emission and thus the air quality (Possell et al., 2005, 2011; Wilkinson et al., 2009), but this effect is not considered here. Tai et al. (2013) suggested that the inclusion of CO_2 inhibition would generally reduce the sensitivity of surface ozone to climate and natural vegetation where isoprene emission is important. However, experimental data for CO_2-isoprene relationship at sub-ambient CO_2 levels characteristic of the past are generally scarce and not consistent enough to buttress inclusion for**

our model period.”

Section 4, lines 8-18, BVOC emissions and crops: The fact that cropland expansion is estimated to benefit to public health, through changes in BVOC emissions, is strongly dependent on emission factors prescribed. Results could be significantly different in a biofuel-type scenario for instance, for which high emitters (oil palm) can be selected. This is a strong limitation of BVOC emission estimates, and of their potential role in the atmospheric chemical composition change, that is not discussed in this study, and for which some elements should really be added in the text, and in the conclusion discussion section as well.

Agree. We have added discussions in Sect.4. and Sect. 8

(P10, L313-322) Sect. 4: “**Our results indicate that the land use change such as cropland expansion in some regions could be beneficial for ozone air quality through reducing biogenic emissions, since crops are generally low-emitting species. However, such effects may be complicated by that some economic biofuel crops such as oil palms are high isoprene emitters, and large-scale replacement of nature vegetation with these crops is expected to increase biogenic emissions (Kesselmeier et al., 1999; Guenther et al., 2006; Wiedinmyer et al., 2006), and thereby enhancing ozone depending on the region. Although such replacement is not characteristic of the history and the regions focused in this study, future work concerning ozone-crop interactions should definitely consider the effects of different crop types.**”

(P15, L487-492) Sect. 8: “**Likewise, cropland expansion is shown to affect ozone but the sign of effect also depends on the relative importance of dry deposition vs. biogenic emissions. In addition, the replacement of natural vegetation with high isoprene-emitting species such as some biofuel crops may further complicate the effects, and the implications for air quality need to be considered in future studies especially for tropical East and Southeast Asia.**”

Specific comments:

Abstract, page 14112, lines 23-24: add “on” in “is more dependent on dry deposition than ON isoprene emissions”

(P2, L52) Added.

Introduction, page 14113, lines 1-2: change “public health concerns facing us today” to “public health concerns that we have to face today”

(P3, L62) Revised.

Introduction, page 14113, line 15: remove “’s ” in “Earth’s climate”

(P3, L74) Changed to “climate”.

Figures: For quicker and clearer analysis of the figures, please add titles on the plots, on top of having them described in the legend, as done in the figure 1 for instance, increasing the font size for better reading.

We have revised the figures following the reviewer’s suggestions. Thanks.

Figure legends: when dry deposition is illustrated I understand it is related to ozone, please add the information in the legend.

(P23, L739, L744; P25, L773, P26 L780) Revised as suggested.