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Dear Editor,

In the current version of our manuscript we have addressed the comments and technical corrections suggested by the reviewers. Please find hereafter the list of main changes and the replies to the reviewers' comments. I hope that you will find the present version of the paper suitable for publication in *Atmospheric Chemistry and Physics*. We would like to thank you again for your consideration.

Yours sincerely,

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List of main changes

- To address Reviewer #3 comments, a new Figure (Figure 4) has been included in the revised manuscript displaying the spatial distribution of the correlation coefficient between the emissions and the main meteorological drivers of the emissions. This figure is discussed in the first paragraph of Section 3 on "Variability and trends in isoprene emissions". Furthermore, Figure 5 (previously Figure 4) now includes the trend in the soil moisture activity factor between 1979-2012 based on ERA-Interim data. This is discussed in Section 3.
- We followed the very good suggestion of Reviewer#3 and included a new section (Section 6) on the comparison of bottom-up inventories against available field measurements. More specifically, we have compared the measured isoprene fluxes of the OP3 field campaign (Langford et al. 2010) with calculated emissions from the S0 and S4 simulations (new Figure 11). Furthermore, we have compared our inventory with isoprene flux measurements from a tropical rubber tree plantation in Yunnan (Baker et al. 2005). The comparisons are discussed in Section 6.
- The abstract has been slightly modified to include a sentence on the correlation between the Oceanic Niño Index and isoprene fluxes, on the comparisons with field measurements, and on the soil moisture as a main cause for interannual emission changes.
- We included a new paragraph in Section 3 (penultimate paragraph) addressing the possible role of LAI interannual variability in the isoprene emissions (reply to Reviewer's#3 comment). We also corrected the last paragraph of Section 3 to account for Reviewer's#1 remark.

On behalf of all authors, I would like to thank the reviewers for their positive evaluation of the manuscript and for the useful comments and suggestions. Below we address the raised concerns. The reviewers' comments are *italicized*.

Reviewer #1

The authors present isoprene emission model output from five simulated cases, with each case beyond the first progressively adding additional corrections to the land-use, emission factor, and radiation inputs of the base case. Each of these corrections are explained and justified in detail, and the results of the final case are then compared to top-down isoprene emission estimates from satellite HCHO column measurements, showing good agreement in most areas within the examined domain. This work represents an important contribution towards the evaluation and improvement of existing land-use and emissions datasets, thoroughly outlining the major factors influencing isoprene emissions in Asia, as well as the recent history of land-use and climatic changes in the area. The paper is generally clear and well-organized, and offers conclusions with clear importance to the atmospheric chemistry community, while highlighting the need for further research into emission measurement campaigns. Word choice, punctuation, and phrasing are occasionally awkward to my ear (and I have listed suggested fixes below), but never unintelligibly so. Overall this is a well-written paper with important contributions to future climate and air-quality modeling efforts.

General questions:

- 1. Though the time series is short, there appears to be a decreasing trend in the difference between the bottom-up and top-down emissions estimates, as shown in Table 3. Is this an artifact caused by abnormally high and low discrepancies in 2007 and 2012, respectively, or is there any reason to believe that this could be indicative of some underlying trend in the error of either the top-down or bottom-up estimate?
 - There is indeed a fairly continuously decreasing trend in the difference between top-down and bottom-up estimates over China as well as over the entire domain. Over Indonesia, India, Malaysia and Myanmar, the difference between bottom-up and top-down estimates remains approximately constant, and over Indochina, the year 2012 appears to be an outlier. The reasons for these (un)changing differences are unclear, but they do indeed indicate that the errors in either the top-down or the bottom-up estimates are not always constant in time.
- 2. The negative trend in isoprene emissions attributable to CO₂ is identified as approximately 0.15% yr-1, but is neglected in the analysis because this is "small compared to the trends associated with climate change." However, in the conclusion the primary cause for increasing emissions in Asia is identified as surface warming, with a net impact on isoprene emissions of around 0.2% yr-1. How can a negligible driver can have an impact only 25% smaller in magnitude than a primary one?

The referee is correct. We modified the paragraph as follows:

"Note that the increasing trend in CO_2 concentrations might have a significant impact on isoprene emission (Arneth et al., 2007). Using the simple parameterization proposed by Heald et al. (2009), based on the observed long-term response of isoprene to CO_2 changes for aspen trees, the CO_2 increase between 1979 (337 ppmv) and 2012 (394ppmv) is estimated

to induce a decrease of ca. 5% in the isoprene emission, corresponding to a negative trend of 0.15%/yr. Although significant compared to the trends associated with climate change, it has been ignored here due to its high uncertainty, and because it is not a significant driver of interannual variability."

Suggested changes:

- 1. Page 29553, lines 3-6: "Finally, a decreasing trend in the top-down Chinese emissions inferred after 2007, is in line with the cooling episode recorded in China after that year, thus suggesting that the satellite HCHO columns are able to capture climate-induced changes in emissions." -¿ "Finally, a decreasing trend in the inferred top-down Chinese emissions since 2007 is in line with recorded local cooling, thus suggesting that the satellite HCHO columns are able to capture climate-induced changes in emissions."
- 2. Page 29553, line 11: Remove comma after "conditions".
- 3. Page 29553, line 15: Change "as measured e.g. by" to "as estimated by indicators such as"
- 4. Page 29554, line 1: Remove comma after "uncertainties"
- 5. Page 29556, line 3: Add "and" after final comma.
- 6. Page 29566, line 14: Remove comma before "because of"
- 7. Page 29566, line 14: Remove comma after "reduced".
- 8. Page 29567, lines 1-4: I recommend completely reworking this sentence for clarity.
- 9. Page 29567, lines 4-6: "Moreover, the emissions are also reduced, by about 25% in Indonesia, due to the higher cropland fraction in S1 than in S0. Furthermore, the emissions are strongly reduced in the S2 scenario, by a factor of 2-3 on average, as already discussed in Sect. 4." replace by "The emissions are further reduced by approximately 25% in Indonesia due to the higher cropland fraction in S1 compared to S0, with additional reduction by a factor of approximately 2-3 in the S2 scenario as discussed in Sect. 4."
- 10. Page 29567, line 12: Replace "dominated" with "driven"?
- 11. Page 29567, line 14: Replace "all simulations, however" with "all simulations. However", and remove final comma before "due".
- 12. Page 29568, line 16: Remove space in "MetOp- A"
- 13. Page 29570, line 4: "comforting the strong emission reduction derived in the S4 simulation, compared to the much higher fluxes of the standard S0 scenario." replace by "supporting the strong emission reductions of the S4 simulation over the much higher fluxes of the standard S0 scenario."
- 14. Page 29571, line 2: Remove both uses of "by".
- 15. Fig. 4: It appears that the bottom right panel has a misplaced % sign. Should it be in parentheses at the end, as in the top panel?

All suggested changes have been introduced in the revised manuscript.

Reviewer #2

The paper shows the impact of the weaker emissions from a rain tropical forest, expanding oil palm with the higher emissions, and more realistic downward solar radiation, on estimations of isoprene emissions in Asia. The estimated bottom-up emissions were also verified by satellite-based emissions. The methods seem sound and the most up-to-date. The results are very useful for the related researchers. The paper is well-organized and the interpretation seems so clear that a reader can easily understand the contents. In the text, the reviewer could almost find the reasons for some questions, which he/she would like to ask about the methods and the estimations/results. But he/she still has one question: Why are isoprene emissions larger in S2 than in S3 in Malaysia (Fig. 9)? The results are different from those in Indonesia, although the reasons are found in Page 29564, L28-Page29565, 29565. This means original standard emission factor and/or fraction of oil palm are in S2 more than that in S3?

Emissions from oil palms in Indonesia were not considered in the MEGAN distribution of emission factors, and therefore, accounting for these emissions in S3 leads to a net emission increase in Indonesia. In Malaysia, however, MEGAN already accounted for the presence of oil palm. This is now clarified in Section 2.2 "Description of the simulations" with the following text:

"Note that, over Malaysia, the S0 simulation already accounts for the presence of oil palm, as reflected by increased basal emission rates (3-4 mg m $^{-2}$ h $^{-1}$) for the cropland PFT over this country. In S3, the basal emission rate of croplands (excluding oil palm) over Malaysia was set to 0.8 mg m $^{-2}$ h $^{-1}$, comparable to values found over Indonesia in MEGAN."

The emissions are slightly lower in S3 than in S2, because the very high emission rates in the new oil palm PFT are more than compensated by the lower emission rate in the "other cropland" PFT. This is now mentioned in the fourth paragraph of Section 4 "Isoprene fluxes across S0-S4 simulations".

Minor comments:

• Page 29555, Lines 17-20: There is a similar report of weaker isoprene emission from a rain tropical forest canopy in the Malay Peninsula: Saito, T., Yokouchi, Y., Yoshiko Kosugi, Y., Tani, M., Philip, E., Okuda, T.: Methyl chloride and isoprene emissions from tropical rain forest in Southeast Asia, Geophys. Res. Lett., 35, L19812, doi: 10.1029/2008GL035241, 2008. This also supports your results.

We thank the reviewer for bringing this reference to our attention. The paper is cited and briefly discussed.

• Page 29563, Lines 8-21: The content (i.e., the relationship between Isoprene emission in Asia and ONI) seems a little bit abrupt, and it should also be stated in the introduction's last paragraph, in advance.

We have now added a sentence in the end of the first paragraph of the abstract. "The isoprene flux anomaly over the whole domain and studied period is found to be strongly correlated with the Oceanic Niño Index (r = 0.73), with positive (negative) anomalies related to El Niño (La Niña) years."

• Page 29566, Line 6-8, "negative trend": Here, it is better to state that the negative trend is due to replacement of cropland with tree plantations, as mentioned in page 29554, lines 14-16.

The sentence now reads: "This trend is strongly reinforced when adopting the land use changes of S1 scenario (0.7%/yr), due to the replacement of cropland with tree plantations in China between 1979 and 2005 (Fig. 1).

Reviewer #3

Stavrakou et al. improve upon earlier studies of biogenic isoprene emissions from Asia, a region undergoing dramatic land-use and climate changes in recent decades, by applying the MEGAN emissions model coupled to a canopy vegetation model. Applying base conditions (meteorology, static vegetation map, and emission factors), they calculate the spatial and temporal changes in isoprene emissions over the period 1979- 2012. Starting from their base simulation, the authors perform four additional simulations to correct for previously identified biases using observation-derived emission factors, and the observed trends in land-use and solar radiation. They find that their best bottom-up isoprene emission trends and distributions are consistent with those derived using GOME-2 formaldehyde (HCHO) columns.

The information provided in this paper is valuable to both the emissions and air quality communities as it clearly highlights that biogenic isoprene emissions in Asia are changing significantly in response to environmental changes and this will have important consequences for regional air quality. The study highlights the improvement in modeled estimates of biogenic emission in Asia when constraints from limited groundbased observations are included -enhanced observational network in that region will lead to better-informed emission models. Overall the paper is well-written with valuable information for emission and air pollution modeling. There is some choppy organization (see general comments below) which can be easily addressed. The paper is appropriate for publication in ACP after minor corrections have been made.

General Comment:

1. Before discussing the trends in Asian isoprene emissions, the authors should evaluate the isoprene fluxes from their base simulation against observations in Asia, preferably ground-based. This will convince the readers that the current model setup suffers from similar biases as noted by Langford et al. (2010) and that the corrections applied in subsequent simulations indeed improve upon the base simulation. Thus, it would be helpful to evaluate the base isoprene fluxes for a particular time period (e.g. 2005 following the discussion in section 4/figure 7) against measurements from OP3 and measurements from other regions wherever available. For example, observational estimates of isoprene fluxes from other parts of Asia are available (Bai et al., 2004; Geron et al., 2006; Varshney and Singh, 2003; Singh et al., 2007; Singh et al., 2008).

We followed this very good suggestion and included comparisons of our inventory results against two campaigns in Asia (Section 6): the OP3 campaign (Langford et al., 2010) and measurements at a tropical plantation in Yunnan, China (Baker et al., 2005). Note that most studies mentioned by the reviewer (Geron et al., 2006; Varshney and Singh, 2003; Singh et al., 2007; Singh et al., 2008) report enclosure measurements of emission factors on a dry weight basis (typically in $\mu g g^{-1} h^{-1}$) for specific plant species, and not above canopy emission rates with which we could evaluate our inventory. The study of Bai et al. (2004) reports the same emission rate measurements as the study of Baker et al. (2005).

We added the following text discussing the comparisons with ground-based measurements:

"We evaluate the inventory against the tower measurements of the OP3 (Oxidant and Particle Photochemical Processes above a South-East Asian Rainforest) project (Hewitt et al., 2010) at the Bukit Atur station in the Danum Valley region of Sabah, Malaysia (4.98°N, 117.84°E). The measurements were carried out over two four week periods with phase 1 (OP3-I) taking place during the months of April and May 2008 and phase 2 (OP3-III) between June and July 2008. The comparison of calculated fluxes with the measurements (Fig. 11) confirms the strong overestimation of the basal emission rate for tropical forests in Southeast Asia (Langford et al., 2010). The average simulated fluxes in S0 are overestimated by factors of about 7 and 5 during the wet (phase 1) and the dry season (phase 2), respectively. These larger factors compared to the factor of 4.1 inferred by Langford et al. (2010) are due to the larger average BER (10 mg m⁻² h⁻¹) at the location of Bukit Atur in our simulation S0 compared to the basal emission rate of 6.6 mg m⁻² h⁻¹ used by Langford et al. (2010). Adopting the latter BER value in our simulations would lead to overestimation factors of 4.5 and 3.3 during the two phases, in excellent agreement with Langford et al. (2010). The factor of \sim 2 higher emissions during the dry season compared to the wet season are only partly explained by the dependece of emissions on temperature and radiation in the MEGAN model, since the average temperature and radiation levels were only moderately higher during phase 2 compared to phase 1 (by 0.5 K and 25%, respectively). Changes in phenology are therefore the most likely cause for the higher apparent basal emission rate during the dry season compared with the wet season, by a factor of about 1.4 according to the model simulations.

In contrast with the seasonal variation discussed above, the emissions of isoprene (also monoterpenes) measured by eddy covariance above a tropical plantation of rubber trees (Hevea brasiliensis) in the Xishuangbanna Gardens (21.92°N, 101.27°E), Yunnan, South China, exhibited much higher values in the wet season than in the dry season (Baker et al., 2005). The average daytime isoprene emissions were found to be 1 and $0.15~\mathrm{mgC~m^{-2}~s^{-1}}$ during the wet season (July 2002) and the dry season (February-March 2003), respectively, whereas our MEGAN-based inventory (S0) predicts higher emissions during the dry season (0.64 mgC m⁻² s⁻¹) than in the wet season (0.44 mgC m⁻² s⁻¹), due to generally lower cloudiness and higher temperatures and radiation levels during the dry season. As discussed by Baker et al. (2005), the lower dry season fluxes of monoterpenes result from the drought deciduous nature of the main monoterpene emitter, Hevea brasiliensis, which is however a low isoprene emitter. The very low dry season isoprene fluxes were very probably caused by extreme water stress conditions (Baker et al., 2005). The soil moisture activity factor is however equal to unity at all times in 2002/2003, based on the MEGAN parameterization (Guenther et al., 2006) using the ECMWF wilting point and soil moisture fields. In fact, the severe drought at the site in February/March 2003 is not recorded in the ERA-Interim data which does not even show lower soil moisture content values in this period compared to other months of the year. The soil moisture activity factor is very uncertain, as it has been found to be very dependent on the choice of the soil moisture and wilting point database (Müller et al., 2008, Marais et al., 2012, Tawfik et al., 2012, Sindelarova et al., 2014): for example, the reduction in global annual emissions due to this activity factor ranges between 7% (Guenther et al., 2006) and 50% (Sindelarova et al., 2014)."

2. In terms of the organization, section 4 should be moved before section 3. Section 6.1 belongs as a sub-section in Section 2.

Section 4 includes discussion of the isoprene emissions per country across the simulations S0-S4, and, to our opinion, should not be placed before Section 3 where the results of the S0-S4 simulations are first presented. Section 7.1 of the revised version describes the inversion set-up in a global model. By moving this in Section 2 there is a risk of confusion between bottom-up (Section 2) and the top-down (Section 7.1) simulation setups. We therefore preferred to keep the current organization of the sections.

Specific Comments:

1. Abstract, line 8-13: The authors should mention that they incorporated these factors (changes in land-use, solar radiation etc.) to correct for the biases identified in previous studies and also to account for deficiencies in meteorological inputs that have important implications for simulating trends and variability in isoprene emissions in this region.

We inserted the following text: "In order to remedy for known biases identified in previous studies, and to improve the simulation of interannual variability and trends in emissions, this study incorporates..."

2. Line 18-19: The authors attribute the variability and trends in emissions to changes in temperature and solar radiation here while including soil moisture as one of the main drivers in the conclusions (page 29572, line 1). I would suggest being consistent in the abstract and the main text. Also see comment 14 below.

This is entirely correct. The text now reads "Changes in temperature, solar radiation are the major drivers of the interannual variability and trend in the emissions, except over semi-arid areas such as Northwestern China, Pakistan and Kazakhstan, where soil moisture is by far the main cause for interannual emission changes."

3. Line 19-22: Remind the readers that the trend discussed here is from the base simulation that does not include the additional factors considered in sensitivity simulations.

We changed the sentence to "In our base simulation, an annual positive flux trend of 0.2% and 0.52% through the entire period is found in Asia and China, respectively, related to positive trend in temperature and solar radiation."

4. Page 29554, Line 10: A reference is needed for "...since crops are known to be weaker isoprene emitters than the forests they substitute."

A reference is added here (Guenther et al. 2006).

5. Page 29554, Line 15: For better clarity, the sentence should be revised to: "Crops in China are being converted to tree plantations (e.g....) for economic reasons, resulting in..."

Changed as suggested.

6. Page 29555, Line 16: The sentence "Their estimation is uncertain, as it relies..." needs to be rephrased for clarity.

Changed to "Their estimation is uncertain due to possible errors in the emission capacities for both natural forests and managed landscapes."

7. Page 29556, Line 19: Remove "literature"

Removed.

8. Page 29558, Line 13: The word "realized" reads awkward here. Replace with perhaps "accomplished"

Replaced by "achieved".

9. Page 29559, Lines 9-10: What level of uncertainty is introduced in the calculated emission trends with the use of climatological mean MODIS LAIs prior to 2002, particularly, since emission flux rate is a function of LAI (equation 2)? Are the same LAIs used for all simulations S0 through S4?

The same LAIs are used for all simulations. Additional calculations were performed to determine the role of LAI variability on the emissions. The following text has been added in Section 3:

"In order to assess the possible role of LAI interannual variability on isoprene emissions, we compare the trends in our base (S0) emissions between 2002 and 2012 (i.e. using LAI varying from year to year) with emission trends calculated using climatological LAI during the same period. The emission trends are almost unaffected over the whole domain (e.g. -0.221 and 0.237%/year using variable or climatological LAI, respectively) or in Southeast Asia (e.g. 0.148 and 0.160%/year over Indonesia). Larger changes are found over more arid areas such as Western India and Northwestern China (e.g. 0.796 and 0.439%/year over India). Noting that these regions are characterized by low LAI values (typically <1.5) for which the MODIS-based estimations are expected to be very uncertain, we conclude that the impact of LAI interannual variability is generally either small or uncertain."

10. Page 29560, 2nd paragraph: It would be helpful to provide the basal emission factors for oil palm trees.

Here we use the parameterization of Mizstal et al.(2011) based on the average canopy temperature (T_c) and a basal emission rate of 22.8 mg m⁻²h⁻¹, as now clearly stated in the text.

11. Page 29561-29562: Discussion of Figure 4 should be presented before the discussion of Figure 5. Further, in relation to Figure 5, it would be helpful to provide a quantitative estimate of the dominant drivers of fluxes either in the Asian domain or in China by performing statistical correlation of fluxes with temperature, radiation, and soil moisture (since emissions are dependent on these time-varying factors). Similarly, a pattern correlation of the trends in Figure 4 is needed to quantitatively substantiate statements like "the increasing trend in emission is due to increases in the soil moisture activity factor, most likely reflecting positive trends in soil moisture..."

We included a new figure (Fig. 4) displaying the spatial distribution correlation coefficient between the emissions and the main meteorological drivers of the emissions. Furthermore, Fig. 5 (previously Fig. 4) now includes also the trend in the soil moisture activity factor. Those figures are presented as follows:

"Figure 4 displays the distribution of the correlation coefficient between annual isoprene emissions in the S0 simulation and the main meteorological drivers of the emission, namely air temperature, above-canopy radiation and the soil moisture stress activity factor. Unsurprisingly, the calculated emissions are strongly correlated with PAR levels over most non-arid regions, and especially over forested areas. This is a consequence of both the direct effect of PAR on emissions (Eq. 2) and the indirect effect through the temperature activity factor (Eq. 4) and the dependence of leaf temperature on solar radiation (Müller et al., 2008). Compared to PAR, air temperature is less well correlated with the emissions in many regions, in part because it is leaf temperature, not air temperature, which drives the temperature activity factor. Furthermore, at extratropical latitudes, the emissions are likely better correlated with summertime temperature than with annual temperature, since most of the annual emissions take place during the summer. Over arid and semi-arid regions, soil moisture is clearly the main meteorological driver of interannual variability. Negative correlation coefficients between the emissions and the soil moisture activity factor over e.g. Eastern China result from the correlation of soil moisture with cloudiness which is itself anticorrelated with PAR. The same effect also explains the negative correlations between emission and PAR over arid areas.

The spatial distribution of the 1979-2012 trends in isoprene emissions (as estimated by the standard S0 simulation) is generally well explained by the distribution of trends in temperature, radiation and the soil moisture activity factor (Fig. 5). Over non-arid areas, temperature and radiation dominate the behaviour of the resulting flux. Over arid and semi-arid regions (Kazakhstan, Pakistan, Western India and Northwestern China), however, positive trends in emissions are primarily caused by increasing trends in soil moisture in those areas. "

12. Page 29563, Lines 8-21: As the other reviewers note, the discussion of the relationship between emissions and ONI is abrupt and needs a preface in Section 1. Also, it is not clear how the isoprene flux anomaly is calculated? Is this anomaly with respect to the mean of 1979-2012?

We included the following sentence in the abstract: "The isoprene flux anomaly over the whole domain and studied period is found to be strongly correlated with the Oceanic Niño Index (r=0.73), with positive (negative) anomalies related to El Niño (La Niña) years." The reviewer's guess is correct: the isoprene flux anomaly is calculated with respect to the 1979-2012 mean. This is now added in Section 3.

13. Page 29564, Line 25: Although the authors discuss S3 results for 2005, Figure 7 does not show fluxes for S3. This oversight should be corrected.

The S4 simulation combines the effects of solar radiation updates in India, China and Japan, and oil palm plantations in Indonesia and Malaysia (as explained in the caption). These updates are independent, since they apply to different countries. The results of the S4 simulation are therefore identical to those of S3 over Indonesia and Malaysia. A clarification is now added in the figure caption (now Figure 8).

14. Page 29566, Section 5: To me, this section is an extension of the discussion of variability and trends in isoprene emission in the S0 simulation (section 2). Perhaps the authors could combine the two to make the text and figures more concise. As an example, the black line in Figure 9 for China is the same as in Figure 5 (second panel from the top), although it appears

that the calculated trends are different (why is it 0.42%/yr in Fig 9 versus 0.52%/yr in Fig 5). Additionally, all panels in Figure 9 do not show fluxes for the 5 simulations (S0-S4). Is there a reason for showing selective simulations for each country? If so, it should be stated clearly, although I would recommend showing all simulations to be consistent as otherwise this would be akin to cherry-picking to support conclusions.

The emission trends over China in Figure 9 (0.42%/yr) and Figure 5 (0.52%/yr) in the S0 simulation are different because they apply to different periods: 1979-2005 in the first case, 1979-2012 in the second as shown in the horizontal axis of the plots. The reason for the shorter period in Figure 9 is explained by the lack of solar radiation data beyond 2005. This is mentioned in the first paragraph of Section 5.

The reviewer is right about the fact that panels of Figure 9 do not include all S0-S4 runs. The reason is that the updates in S0-S4 do not concern all countries. For example, in China, the results of S3 and S4 simulations are identical because oil palm plantations are implemented only in Indonesia and Malaysia. Similarly, S4 simulation over Malaysia and Indonesia is identical as S3 since solar radiation correction concerns only Japan, China, and India. A clarification is now included in the figure caption.

15. Page 29571, line 24: replace "builts" with "builds"

Corrected.

16. Page 29572, line 1: I suggest that the authors perform a quantitative analysis of the role of soil moisture in driving variability and trend in Asian isoprene emissions to support their conclusion statement "Temperature, solar radiation, and soil moisture are the main drivers of interannual variability.

As discussed above, soil moisture is now clearly shown to be the main driver of emission variability over (semi-)arid regions, as seen in the new Figures 4 and 5(d).

17. Page 29572, line 15: The statement "...in better agreement with ground-based observations." needs to be supported by comparing the fluxes simulated in S4 (and S0) with ground-based observations.

See above (General comment 1). We have followed this suggestion and included comparisons with two measurement campaigns.

18. Figures 1, 2, 4, 7, 8, and 10: The labels on the label bar are too small to read. Please use bigger and darker font.

Done.

19. Figures: Please consider labeling panels as (a), (b), (c) and so on for figures that have greater than one panel.

Panels have been added in Figures 1, 2, and 4.

Reviewer #4

The paper presents an interesting evaluation of evolution of isoprene emissions in the last three decades over Asia using modeling driven by local land-use changes, climatology, in combination with top-down constraints from remote sensing and explicit consideration of recent field measurements at oil palm plantation and rainforest in Borneo. As field data of isoprene fluxes are extremely limited in Asia, satellite approaches seem to provide a robust approximation of the emissions. The top-down and bottom-up estimates seem to agree well overall and over specific subdomains including China, India, Borneo, Japan, etc. This paper can clearly be an important contribution attracting attention to Asia in terms of air quality and land-use effects, and gives the motivation for focusing more atmospheric measurements in that region.

Questions/comments:

1. Page 29554, Lines 7-11: "Moreover, Southeast Asia faced massive land use changes during the last decades, in particular deforestation and conversion of primary forests to croplands, leading to a decrease of isoprene fluxes, since crops are known to be weaker isoprene emitters than the forests they substitute".

Was the major expanding crop in Southeast Asia the oil palm? What fraction of croplands constitute oil palm plantations in Malaysia and Indonesia? As oil palm has an extremely high isoprene emission potential (e.g. Fowler et al., 2011), the statement could be true, if oil palm was excluded from the crops, but it is not clear in the text if this exclusion was made.

Indeed the oil palm plantation is the most important expanding crop in Indonesia and Malaysia. In Indonesia (Sumatra, Kalimantan, and Papua) oil palm plantation area has almost doubled between 2000 and 2010. In the same period, intensive agriculture has increased by 46%, whereas the area of agroforest and other plantations has decreased by about 9%. In 2010 oil palms contributed 23% to the total cropland area, and accounted for approximately 5% of the total surface of Indonesia, as described in the study by Gunarso et al. (2013) based on LandSat satellite data.

Oil palm plantation area has increased in Malaysia by 51% in 2000-2010, whereas agroforest and other plantations decreased by 15% and intensive agriculture remained almost constant. About half of the Malaysian cropland area was covered by oil palms in 2010. The oil palm area in 2010 accounted for 15% of the total area (Gunarso et al. 2013).

For clarity the text now reads: "Moreover, Southeast Asia faced massive land use changes during the last decades, in particular deforestation and conversion of primary forests to croplands, leading often to a decrease of isoprene fluxes, since crops are generally known to be weaker isoprene emitters than the forests they substitute (Guenther et al. 2006)."

The article Fowler et al. (2011) is now cited in the introduction.

Oil palms are excluded from the crops (see also next comment).

Gunarso, P., M. E. Hartoyo, F. Agus, and T. J. Killeen, Oil palm and land use change in Indonesia, Malaysia and Papua New Guinea, Reports from the Technical Panels of the 2nd Greenhouse Gas Working Group of the Roundtable on Sustainable Palm Oil (RSPO), www.rspo.org, November 2013.

2. P29560 L.5 Treating oil palm as a separate PFT makes sense because it is a very specific crop type with orders of magnitude higher emissions capacities than most other crops. Was

the expansion in oil palm area reflected in this PFT as the expansion in crop areas or how were crop and oil palm PFTs separated and used in the model?

The separation of oil palm from crop was made by simply subtracting the oil palm fractional area from the total crop area obtained from Ramankutty and Foley (1999).

3. P29556 L21 Does the emission factor represent here the net flux above the canopy? If so consider adding this information.

Yes. In Section 2.1 the sentence now reads "More specifically, the net flux rate above the canopy is given by ...".

4. P29568 L21 "HCHO columns and their error characterization are available at the TEMIS website (http://h2co.aeronomie.be)". I could not find any information on the error characterization at this link. It might be helpful to a reader to briefly describe the uncertainties here and if they are the same/different for the regions studied.

The reviewer is correct. Indeed, this information is not available on the website itself but it is included in the data files. For clarity we have added the sentence "A detailed error characterization of the columns is given in De Smedt et al. (2012)."

5. P29552. L21 "The impact of oil palm expansion in Indonesia and Malaysia is to enhance the trends over that region, e.g. from 1.17% to 1.5% in 1979-2005 in Malaysia". By comparing S2 and S3 for Malaysia (Table 2 and Fig. 9) it seems that this enhancement is very small. In fact the emissions seem very slightly lower in S3 than S2. Is it because the model is so insensitive to the oil palm expansion vs more extensive rainforest, or is there perhaps an issue with the crop/oil-palm PFT or crop/oil-palm emission factor? See also next comment.

Emissions from oil palms in Indonesia were not considered in the MEGAN distribution of emission factors, and therefore, accounting for these emissions in S3 leads to a net emission increase in Indonesia. In Malaysia, however, MEGAN already accounted for the presence of oil palm. This is now clarified in Section 2.2 "Description of the simulations" with the following text:

"Note that, over Malaysia, the S0 simulation already accounts for the presence of oil palm, as reflected by increased basal emission rates (3-4 mg m $^{-2}$ h $^{-1}$) for the cropland PFT over this country. In S3, the basal emission rate of croplands (excluding oil palm) over Malaysia was set to 0.8 mg m $^{-2}$ h $^{-1}$, comparable to values found over Indonesia in MEGAN."

The emissions are slightly lower in S3 than in S2, because the very high emission rates in the new oil palm PFT are more than compensated by the lower emission rate in the "other cropland" PFT. This is now mentioned in the fourth paragraph of Section 4 "Isoprene fluxes across S0-S4 simulations".

6. P29564 L28: "While Indonesian emissions are increased, no significant change is found for Malaysia, where oil palm plantations were already considered as a major crop in the MEGAN distribution of emission factors." The MEGAN paper (Guenther et al., 2006, P.3189) assigns different emission factor to crops where oil palm is dominant and different emission factor to the remaining crop areas, so I assume the former emission factor or the emission factor from Misztal et al., 2011 was applied to the Malaysian region. If so, it is still somewhat

surprising that S3 did not increase significantly the emissions between 1979 and 2005 (Fig. 9), for example as implied by the trends in Fig. 2.

The Misztal et al.(2011) algorithm has been applied in all oil palm plantations, not just in Malaysia. The relatively small change in the trend between S2 and S3 is simply due to the the fact that oil palms represent still a minor fraction of the total vegetated area over Malaysia.

7. Some changes in the landcover-driving variables are attributed to a likely cause of significant changes in isoprene emissions (e.g., effects from the dimming due to aerosol or brightening due to reduction in clouds). This is very interesting over China where upward trend was observed in the base simulation and was further enhanced in S4 possibly by solar brightening in the isoprene-rich part of China. Could these changes in isoprene emissions result in a feedback on aerosol formation from isoprene oxidation and possibly induce the changes in the opposite direction?

This is an interesting remark. The feedback of isoprene emissions on aerosol abundances and properties are however still poorly understood and quantified.