

Interactive comment on “Isoprene emissions in Africa inferred from OMI observations of formaldehyde columns” by E. A. Marais et al.

Anonymous Referee #2

Received and published: 16 March 2012

Marais et al. present an enhanced method to derive biogenic isoprene emissions from satellite HCHO columns and apply this method to the African continent. They present new approaches to isolate satellite retrievals affected by biomass burning sources or anthropogenic emission of HCHO. They further attempt to account for the current unknowns in isoprene oxidation chemistry by using two different oxidation mechanisms, and for the effects of different chemical regimes with respect to NO_x by using satellite NO_2 retrievals. The paper is very well written, and tackles the difficult and error-prone problem of estimating isoprene emissions from satellite HCHO columns in a thorough and methodical fashion. The method presented by Marais et al. appears to offer improvements over previous methods, especially for Africa. Further, a top-down estimate of isoprene emissions which specifically considers the African continent and accounts for its particular characteristics helps further our knowledge of the emis-

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sions of this atmospherically-important compound from a region believed to contribute globally-significant emissions. I recommend publication in ACP subject to addressing the following comments.

Specific comments.

1) As is acknowledged by the authors in the conclusion, one of the biggest impediments to retrieving isoprene emissions from satellite HCHO columns is the current problems in understanding the oxidation of isoprene under low- $[\text{NO}_x]$. An attempt to address this issue in this work is made by using both the standard MCM isoprene oxidation scheme, and a modified scheme by Paulot et al. (2009) which produces more OH during isoprene oxidation. The more radical (with respect to OH) scheme of Peeters et al. is discounted based on recent experimental results. However, I would suggest that this consideration of a single possible modification of the isoprene oxidation scheme is overly optimistic, and barely scratches the surface of this complex parameter space. Recent studies (Stone et al., 2011; Archibald et al., 2010) have shown that the amount of additional OH produced by the Paulot et al. scheme is nowhere near enough to explain the field measurements of OH. Thus, whilst it may represent a step forward, this scheme alone is clearly not the answer to the problems in understanding isoprene oxidation. In Fig. 4, Marais et al. show the potential modification to the amount and timescale of HCHO yields due to the use of the Paulot et al. scheme relative to the standard one. The modifications at low and very low $[\text{NO}_x]$ shown in Fig. 4 are substantial, and perhaps indicative of the magnitude of changes that may be induced by modifying the isoprene oxidation scheme. However, there is no reason to believe that further future revelations regarding isoprene oxidation will not result in larger differences in HCHO yields compared to the standard scheme at low-mid $[\text{NO}_x]$ than the Paulot et al. scheme does. Given the current unknowns I think the authors have made an reasonable effort to address this problem. However, I would contend that the statements made on pg. 7495, line 23 and pg. 7499, line 2 that the errors from smearing are larger than those from the chemical scheme cannot be justified given the current

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poor understanding of isoprene oxidation. I suggest that the authors note this, and mention that true isoprene oxidation may differ significantly from both of the schemes used here.

2) Following on from the previous point, on pg. 7491, line 26 it is suggested that the negligible smearing of HCHO (at high wind speeds) in the AMMA observations may be a result of the relatively high $[\text{NO}_x]$ conditions, whilst the much larger smearing across Central Africa (at low wind speeds) is due to more moderate $[\text{NO}_x]$ levels. However the reported NO_x mixing ratios are similar (360 pptv NO_x for AMMA and ~ 400 pptv NO_2 for Central Africa). Thus based on Fig. 8, I would disagree that the AMMA observations are close to the threshold between high and low $[\text{NO}_x]$ conditions (which appears to occur at ~ 1 ppbv), as stated on pg. 7494, line 25, and instead suggest that the NO_x regimes in AMMA and Central Africa are similar (as also suggested in Fig. 6), and these differences in smearing are evidence of our poor understanding of isoprene oxidation chemistry (based on both satellite and MEGAN estimates of isoprene emissions in Fig. 9, the AMMA and Central Africa transects both show substantially different isoprene emissions, and therefore likely the influence of isoprene on the overall atmospheric chemistry of the two regions differs). I think the text should be changed to reflect this.

3) Potentially how substantial is the isoprene emission from the excluded grid squares? Perhaps a first assessment could be done by summing up the bottom-up emissions from MEGAN for these grid squares?

4) Large numbers of grid-square-months are removed due to the effect of smearing (Section 4.2). However, these removed HCHO values are indicative of emissions in their source grid square. As I understand it, this lost fraction of HCHO is implicitly accounted for by assuming that GEOS-CHEM correctly simulates the real smearing (and therefore loss of HCHO from a grid square via transport) and thus calculates the right ratio of Ω_{HCHO} to E_{ISOP} . I think this assumption should be spelled out to the reader.

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Minor comments.

Pg. 7477, line 16. This final sentence is too strong given the uncertainties in the top-down method. I suggest replacing with, "Our results suggest a large overestimate of central African rainforest emissions in the state-of-the-science MEGAN inventory."

Pg. 7477, line 22. Guenther et al. (2006) only report isoprene emission estimates. A further reference is needed to justify this 50% figure, or else use the 44% value from Guenther et al. (1995).

Pg. 7481, line 27. "...only a few hours, whereas..."

Pg. 7481, line 27. I think this sentence refers to oxygenated VOCs emitted by biomass burning? In which case this should be made more explicit to avoid apparently contradicting the point made on pg. 7478, line 25 regarding isoprene dominating HCHO production from NMVOCs.

Pg. 7490, lines 18, 20, 23, 25 and pg. 7492, line 15. As pressure increases with decreasing height it would be more explicit to say "below the 900 hPa level", etc.

Pg. 7491, line 8. "...there is no significant smearing of these species and provides..."

Pg. 7491, line 17. This sentence assumes we are confident of HCHO formation pathways and yields from the isoprene oxidation scheme. See main comment above.

Pg. 7492, line 27. Please provide boundary layer mixing ratio equivalents of these NO₂ columns to aid the reader.

Fig. 7. "MEGAN" should be in the key with the other variables, rather than floating. Presumably the arrow shows wind direction, in which case this should be stated.

Fig. 9. Two extra panels showing OMI + and - the calculated error would be very informative for the comparison with MEGAN, particularly as this error varies with NO_x regime.

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