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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Impacts of seasonal and regional variability in biogenic VOC emissions on surface ozone in the Pearl River Delta region, China" *by* S. Situ et al.

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The authors would like to thank Referee #3 for their detailed comments and particular remarks. We give response to each comment below, and the manuscript has been revised accordingly.

The manuscript addresses a very relevant question and rather large task, describing emission measurements at a particular site, simulating the air quality of rather complex region as well as carrying out some sensitivity analyses. I appreciate this effort very much although the necessary multitude of assumptions makes the paper difficult to

digest. Overall it is well written although sometimes I would appreciate a more stringent style. The greatest problem I see is that the measurements and simulations at the site scale hardly match but are still used to justify the application on the regional scale. Despite being judged as 'reasonalbe', micrometeorological conditions are considerable biased towards more radiation (more than 30% off) and higher wind speed (100% off). Peak monoterpenes emissions are 2-fold the size of the measurements and peak isoprene emission 15-fold. Even compared to separate isoprene concentrations the model is hardly touching the measurement at the lower range. Nevertheless, the simulations are described as 'agrees well' in both cases.

Respond: Thanks very much. We have revised the clerical errors.

While the meteorological conditions are at least covered in the sensitivity analyzes, the emissions factor uncertainty is probably not. If the arithmetic mean of 0.7 ppm (0.1-1.3) emission is sensible indicator for the simulated isoprene emission, at least the 7-fold uncertainty range has to be investigated. In fact, using the average OH concentration rather than the maximun observed for calculation, and/or using a LAI of 4 as indicated in Zhang et al. 2006 for evergreen deciduous forests in this region, would probably lead to even higher estimates of isoprene concentrations.

Respond: We used an LAI value of 3, which we got from the papers describing the Dinghu site. We also checked the monthly LAI in the dynamic MODIS product of LAI, and the MODIS LAI indicated the LAI varied from 3 to 5 at this site with a value of 3 in November 2010. So, we used the LAI value of 3 when we ran MEGAN at this site. To the OH concentration, it is better to use the OH concentration observed simultaneously, but there are few studies on the OH concentration over the PRD region and the paper Lu et al. (2012) didn't report the average OH concentration. Limited to the data we have, we used the maximum observed reported by Lu et al (2012). to convert the emission flux, which is representative of the midday value in the same way as Guenther et al. (1999).



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Overall, the decision is if the measurements are judged as trustworthy and representative – in this case the emission factors should be reduced – or if the model assumptions are judged as more reasonable - in that case a clear statement is required, the problem of using point sources for evaluation of area integrated model results can be discussed, and the description of the measurements could be shortened.

Respond: Thank you very much. This is the initial measurement on the BVOC emission flux and the observed isoprene is much lower than the model predicted in fall, but we expect the model to work better in summer. This result indicates that our knowledge of seasonal variation is limited, at least at a site including obvious dry and wet seasons, and we don't have enough observations to tell us whether this low emission is occasional or normal at this site. We still used the BVOC emissions estimated by MEGAN to drive the chem model and estimate the impacts of BVOCs on surface ozone, but we have shortened the description of the measurements in this manuscript and made the statement clearer. Moreover, considering the uncertainties of BVOC emissions estimated by MEGAN, sensitivity experiments were also conducted to assess the sensitivity of surface ozone to MEGAN drivers and the results were presented in the Section 4.6 in this manuscript.

Some particular remarks:

(1) P6730, L15: delete 'significantly'

Response: Thank you. We have deleted 'significantly'.

(2) P6730, L17 (and discussion): Why are some cases more affected in summer and others in autumn? I think that the explanation should be a main result here.

Response: Thank you. The surface ozone in the downwind area is most sensitive to the change of BVOC emissions. But the downwind areas change in autumn and in summer, because the PRD region has southerly or southwesterly prevailing wind directions during the summer and northerly or northeasterly prevailing wind directions



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in the autumn. That is why some cities are more affected in summer and others in autumn. We have added the explanation in Section 4.4 in the manuscript.

(3) P6731, L17ff: rather old references (see e.g. Laothawornkitkul et al., 2009)

Response: Thank you. We have updated the references.

(4) P6732, L4: For eucalypt emissions, Winters et al., 2009 would be a more appropriate ref.

Response: Thank you. We have updated the references.

(5) P6735, L18: biogenic sources for anthropogenic emissions

Response: Thank you. We have revised it. The sections on anthropogenic emissions include power plant, industry, mobile and residential sources.

(6) P6738, L14/15: repetition

Response: We have deleted it.

(7) P6742, L20ff: Barkley et al. reported isoprene shutdown prior to the dry season. However, in I don't expect such an impact at the measurement site here. Nevertheless, is it possible that a winter-downregulation occurred since the coldest month is January?

Response: Thank you. As other studies mentioned, cool temperature leads to a down regulation of isoprene emission (Petron et al., 2001) and isoprene emission is low during drought condition (Pegoraro et al., 2004). It was cold and dry season in the PRD region when the REA measurement was conducted, and the temperature and humidity data observed at Dinghu site indicates the temperature and humidity kept decreasing during the REA sampling period. So, drought and cold are assumed to be the reason for the low isoprene emission, and the MEGAN emissions algorithms may not account for it very well.

(8) P6743, L1ff: MEGAN uses 24 hour as well as 240 hour means of temperature and



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radiation to account for seasonal developments. However, it is assumed that this is still dampening the speed of adjustment (Grote et al. 2010).

Response: Thank you very much. We have included it as one of the possible reasons for the overestimated isoprene emissions flux by MEGAN.

(9) P6744, L20ff: This is a true but very general statement. I would think it is okay for the introduction but in discussion or conclusion it should be used only in context with a specific example.

Response: Thank you. We have revised it.

(10) P6746, L1: limited

Response: Thank you. We have revised it.

(11) P6748, L10ff: The exercise to run the model at a different season is particularly problematic if the statement is kept that the seasonality in MEGAN is not appropriate.

Response: From the comparison of modeling and measurement in summer in Section 4.1.2, we can see the performance of MEGAN is reasonable in summer in the PRD region. We wanted to see the impacts of seasonal variability of BVOC emissions on surface ozone in the PRD region, because the PRD region has southerly or south-westerly prevailing wind directions during the summer and northerly or northeasterly prevailing wind directions in the autumn. So, the pattern of impacts of BVOC emissions on surface ozone in summer is different from that in autumn.

(12) P6750, L22ff: Please note that the ranges that can be found in the literature also include 4 (Steward et al. 2003) and 5 (Simpson et al. 1999) fold uncertainties.

Response: Thank you. We have included these two studies in the manuscript.

(13) P6752, L1ff: This conclusion is only another summary except for the last paragraph. What about the need for more evaluation measurements and better representations of processes? What can be derived for decision making e.g. is the consideration



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of BVOC emissions important for air quality studies? Under which circumstances?

Respond: Thank you. The need for more evaluation measurements was included in Section 4.1.2 and the last paragraph in the conclusion section. The consideration of BVOCs importance for air quality studies has been added in the conclusion section.

(14) Fig. 2: note the difference units

Response: Thank you. We have revised it. In the latest version of the manuscript, all the emission fluxes are in unit of kg km-2 hr-1.

(15) Fig. 3: I think the reference is to Fig. 1, not 2

Response: Thank you. We have revised it.

(16) Fig. 4: note that this is simulated

Response: We explained the Fig.4 in Section 4.1.3, Section 4.3 and Section 4.5.

References mentioned

(1) Grote, R., Keenan, T., Lavoir, A.-V., Staudt, M., 2010. Process-based modelling of seasonality and drought stress in isoprenoid emission models. Biogeosciences 7, 257-274.

(2) Laothawornkitkul, J., Taylor, J.E., Paul, N.D., Hewitt, C.N., 2009. Biogenic volatile organic compounds in the Earth system. New Phytologist 183, 27-51.

(3) Simpson, D., Winiwarter, W., Börjesson, G., Cinderby, S., Ferreiro, A., Guenther, A., Hewitt, C.N., Janson, R., Khalil, M.A.K., Owen, S., Pierce, T.E., Puxbaum, H., Shearer,M., Skiba, U., Steinbrecher, R., Tarrason, L., Öquist, M.G., 1999. Inventorying emissions from nature in Europe. Journal of Geophysical Research 104, 8113-8152.

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(5) Zhang, Lei-Ming; Yu, Gui-Rui; Sun, Xiao-Min; Wen, Xue-Fa; Ren, Chuan-You; Fu, Yu-Ling; Li, Qing-Kang; Li, Zheng-Quan; Liu, Yun-Fen; Guan, De-Xin; Yan, Jun-Hua 2006. Seasonal variations of ecosystem apparent quantum yield ($\alpha$) and maximum photosynthesis rate (Pmax) of different forest ecosystems in China, Agricultural and Forest Meteorology, Volume 137, Issues 3–4, Pages 176-187

(6) Winters, A.J., Adams, M.A., Bleby, T.M., Rennenberg, H., Steigner, D., Steinbrecher,R., Kreuzwieser, J., 2009. Emissions of isoprene, monoterpene and short-chained carbonyl compounds from Eucalyptus spp. in southern Australia. Atmospheric Environment 43, 3035-3043.

Response: Many thanks for these references mentioned. We have read all of them and some of them have been cited in this manuscript, including:

(1) Grote, R., Keenan, T., Lavoir, A.-V., Staudt, M., 2010. Process-based modelling of seasonality and drought stress in isoprenoid emission models. Biogeosciences 7, 257-274.

(2) Laothawornkitkul, J., Taylor, J.E., Paul, N.D., Hewitt, C.N., 2009. Biogenic volatile organic compounds in the Earth system. New Phytologist 183, 27-51.

(3) Winters, A.J., Adams, M.A., Bleby, T.M., Rennenberg, H., Steigner, D., Steinbrecher,R., Kreuzwieser, J., 2009. Emissions of isoprene, monoterpene and short-chained carbonyl compounds from Eucalyptus spp. in southern Australia. Atmospheric Environment 43, 3035-3043.

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