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Interactive comment on “Urban stress-induced biogenic VOC emissions impact secondary aerosol formation in Beijing” by A. Ghirardo et al.

Anonymous Referee #1

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Major comments

The current study presents some important and interesting results. The strengths are a detailed measurement campaign of biogenic emissions from trees planted in Beijing, some laboratory measurements of stress BVOCs and finally modeling efforts to understand their contribution to aerosol formation. While the measurements are solid, there are significant uncertainties in the modeling approach that need to be better acknowledged. Also, the presentation of the research needs to be improved in a number of areas. In my opinion, the important conclusion of the paper is given in Section 4.2, “The importance of measuring stress-induced BVOC emissions.” Finally, what is the relevance of phylogenetic modeling? How are these results relevant to this study? This should be removed. They might warrant publication in a separate paper.

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The title of the manuscript, “Urban stress-induced biogenic VOC emissions impact secondary aerosol formation in Beijing,” highlights a result from a very simple box model that has numerous assumptions. First, all the steps in the model need to be presented more clearly, from the emissions inventory to the secondary aerosol calculations. Much of this is described via words in the text. The methodology could be much clearer if equations were employed instead. Second, there are some critical variables in the model of aerosol formation: the chemical reaction rates, the height of the boundary layer, and the residence time. While there is some detail about the reaction rates, there are no references and little justification for the other two variables. While the simple model will use the same boundary layer height and residence time for anthro aerosols and therefore they do not affect the bio/anthro comparison, that’s not true about the rate constants and the anthro emission rates. Overall, there needs to be a detailed analysis of uncertainty. For example, the aerosol mass numbers in the abstract have three significant figures. This is certainly not justified by the large uncertainties in the model results.

In regards to just the emissions inventory, there should also be an error analysis. For example, what is the effect of the phenology corrections? What uncertainty do they introduce? In addition, there is almost no detail given about the tree inventory data. How was that collected? And again, equations should be used to give the detailed steps in the emissions model.

Some detailed questions about the stress experiment: The lab experiments are pulses of ozone, as opposed to the relatively constant ozone exposure in the field. Are there any previous studies that show the same pattern of stress BVOCs are observed in each case? Also, of the model plants, only one was a tree species. Three of the four don’t appear to be good analogs to the field plants. Considering the conclusion that the field plants are emitting stress BVOCs, another possibility is that these BVOCs were emitted because of handling and damage from being inserted into the cuvette, particularly the SQTs. Need to provide some assurance/evidence this was not the case.

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The quality of the manuscript needs to be improved. While some typos are in manuscript are inevitable, the list of minor corrections below is extensive. Also, there are lots of references to Supplementary material—too many for a paper that is already long. Finally, the quality of the manuscript varies. There are some sections with numerous grammatical errors and where the prose is not focused.

Minor comments:

23007, 13: “greening” instead of “greens.”

23007, 13-14: should state why ABVOCs are decreasing.

23007, 19-20: The units are confusing here. These are correct for aerosol mass, but the statement refers to emissions. Are you saying the emissions increase caused the stated biogenic SOA increase? This should be clearer. After reading the entire paper, I see these numbers are the result of a very simplified modeling approach. As mentioned in the major comments, these results should not be presented this way in the abstract.

23008, 5-9: The phrase “and ozone formation” should be moved to “altering ozone formation and the concentrations of hydroxyl radicals” since it also depends on NO_x.

23008: 11: add the mechanism which explains this: through altering the chemical lifetime of radiatively important gases.

23008, 12-13: “whereby” is not a good conjunction here, since the species specificity is not causing isoprene and monoterpenes to be dominant. “And” would be more appropriate.

23008, 22: My preference is to call linalool a monoterpenoid, not a monoterpene and to reserve the term monoterpene for C₁₀H₁₆ compounds. I understand you consistently use this convention, but realize it’s confusing to some.

23009, 16: What is “higher radiation” referring to? Solar radiation? I don’t think the urban heat island effect increases solar radiation. In any case, explain further and

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provide a citation.

23009, 18-20: You have already stated the relationship between BVOCs, NO_x and ozone formation.

23010, 14: The hyphen in “large-tree” should be removed. Large refers to the plantation program, not the trees. Table 1 says shrubs were also planted.

23010, 16: “Despite of” should just be “Despite”

Overall Materials and Methods section: the order of presentation is confusing, since you jump back and forth between the measurement and the modeling components of your study. You start by describing the trees to be measured, then turn to modeling, and finally return to the measurement of the trees. The order should flow more logically.

23011, 21: “does not” not “do not.” Also, “terpene emissions” instead of “terpene.”

23011, 24: “except for” instead of just “except”

23011, 26: “does not” instead of “do not”

23011, 27 “either” instead of “neither”

23013, 11: I don’t understand what the 76 min time resolution refers to. Is that the collection time for the cartridge? Also, the fumigation lasted 1-2 hours, and then it was back to clean air for the remaining 10 and 20 hours? The timing of these experiments should be stated more clearly. Also, state the rationale for the selected timings, and be explicit about what you were attempting to simulate.

23015, 24: Were these stainless steel or glass absorbent tubes?

23015, 15-16: refer to experiments, not figures, since the figures have not yet been introduced.

23015, 20-22: What was the average and maximum correction in percent that was produced by this algorithm?

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23015, 25 – 23016, 2: Is this assumption valid for all evergreen plants? What are the uncertainties associated with this assumption?

23016, 10-17: This description is not very clear and needs to be rewritten. In particular, I cannot follow the logic of (i). The methodology you used should be clear without needing to read the entire references. Perhaps some equations would make this section more clear? Step (iii) is somewhat clearer.

23016, 19: “tree number per species” should be “number of trees per species.” Also, some detail should be given about the tree inventory.

23016, 19: corrected by phenological development: is this the same SIM correction mentioned on the previous page? Reading further, this is described below. The order here is a bit confusing. Also, there should be more clarity, since phenological development is used to describe both the leaf-level emissions factor and also the leaf area/mass.

23016, 25: It's fine to use literature values for unmeasured trees. How did the measured values compare to literature values? Maybe this is presented in the results?

23017, 6: Should be “emissions were increased.”

23017, 26: This estimate of boundary layer height will have a big impact on the calculation. Also, there are covariances between temperature, pollution and boundary layer height that make using a single value problematic. There should be a reference for this. Overall, this is a very crude estimate of aerosol formation since it does not account for transport, residence times and chemistry.

23018, 6-12: Equations would make this clearer. This seems to be working backwards. You are starting with leaf-level estimates, and then you scale forward. Why is this necessary?

23018, 10-12: Need to acknowledge uncertainty in these estimates.

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23018, 25-28: Again, the 4 days is a very important assumption. Isn't the transport time much shorter than this? More information and references should be given to support this assumption.

23019, 1-8: Need more detail. Was this using the same concentration-lifetime assumption as above? How were concentration measurements converted to source strengths? Need to be more explicit here.

23020: 9-10: These values are lower than I expected for a heavily polluted city. Would these values have represented a violation of either US or European air quality standards?

23024, 1-4: Careful: your theoretical models has three components: threshold, dependence on severity, and large amounts. Your experiment has only demonstrated the first and last of these components.

23024, 12: Should be "and those that are grown"

23024, 12-14, Yes, but need to temper this statement, since overall stress BVOCs are a much lower percentage (14-15%, Table 2) due to the high cBVOC emission rates of the remaining third.

23025, 18 and following: this material would be more appropriate in the Introduction. And again, this phylogenetic perspective adds little to the paper.

23026, 12: I agree with this point, but you need to define these acronyms and give some references to the literature that describes these models.

23027, 1-3. I appreciate that you are acknowledging the potential systematic uncertainties in your results. But specific to cut branches, you are here acknowledging a potential bias, where on page 23011, lines 17-28, you have a lengthy defense of this procedure. You should harmonize your two discussions of this source of potential bias.

23028, 2: remove "species selection"

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23028, 3: previous experiences of development of what?

23028, 5-6: no, you made no assessment of ozone formation, so this statement is not correct. You only considered SOA formation.

23028, 6-22: First, this section does not belong in the discussion and is repetitious with material included elsewhere in the paper. Second, why bring up air quality and ozone formation now? This section reads as if were written for a purpose not in line with the rest of the paper. The final sentences just reiterate points that were made in the Results and don't provide much synthesis. This entire section is weak and not focused.

23028, 23-29: This is a return to material that is logical and well suited for the Discussion section.

23029, 3: Note the use of error bars for the literature estimate. Assessing and adding these to the current work will be necessary before publication.

23029, 13-16: I am getting confused by the logic of this paragraph. First, you point out that the biogenic fraction of SOA is small. Then you say that the anthro estimate is probably too high. Is the next section just getting back to your original point, or are you saying something new? This paragraph should be condensed and more focused. And then the next two sentences appear to seesaw back and forth. Just be clear: state your point, while acknowledging the small contribution.

23029: 20-29: This is a very nice comparison! Removing some of the weakly focused text in the Discussion will allow this to be highlighted.

23030, 2-3: This is not a conclusion of your research. You only briefly mentioned this, and it was from a source that you didn't create. It shouldn't start your Conclusion section.

23030, 6: Don't say pollution, since you only assessed particulate matter, and not gas-phase chemistry.

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23030, 6-9: Again, this is not a conclusion from your research.

23030, 12: “successfully” not “successful.”

23030, 10-20: Overall, a good paragraph for the conclusion, but remove the quotes from constitutive and stress-induced. Also, you should reword the final sentence, because again this is not the conclusion of your research. A more appropriate conclusion would be, ‘We conclude that “picking the right tree for urban greening” (Churkina et al., 2015) can potentially reduce the formation of pollution in megacities.’ You have not assessed the benefits of trees on air quality in your study.

Table 1: Make “Chinese Flowering Crabapple” have a consistent significant figures.

Table 2: The final four columns do not add anything to the analysis and can be easily calculated if someone was interested. They should be removed.

Figure 6: Between the interrupted vertical axes used in panels a and c and the mixed vertical axes that are used in panels a and b, the main points of this figure are obscured. Since cBVOC contribute so little, they should either be removed or allowed to have negligible bars. Or, the categories could be reduced to only c- and sBVOCs, to match panel c.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 23005, 2015.

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