Responses to Dr. Carter

This paper discusses issues regarding processing of emissions of volatile organic compounds (VOCs) for air quality models in use different chemical mechanisms. Although the focus of this paper is on the emissions inventory for Asia, the issues they bring up are applicable for emissions and modeling everywhere. The speciation profiles used to determine model species for representing VOCs are important to model predictions of the effects of the VOCs on air quality, and they have significant uncertainties. They are also processed in inconsistent and undocumented manners when used to derive model-ready inputs for different chemical mechanisms, introducing additional uncertainties in the model predictions. In this work the authors examine and update a number speciation profiles representing important VOC sources in Asia, derive the model species for the profiles in a consistent manner for representative mechanisms, and show that the updates have significant effects on emissions input and are likely to have significant effects on model predictions. The effects of the particular updates used in this work may not be as great when examining emissions in the United States or Europe, but I suspect that there are similar issues with other profiles that would affect modeling in these regions. The issues discussed in this paper are something that modelers, and those that use model outputs, should better understand and appreciate. I believe that this paper makes an important contribution in this regard.

The paper is generally clearly written and informative, and I have only a few questions, comments or suggestions. These are given below.

Response: We appreciate the insightful and constructive comments from Dr. Carter. We deal with individual comments as below.

The high contributions of glyoxal and methyl glyoxal are surprising to this reviewer, who more used to U.S. emissions inventories. They give a reasonably good discussion of what caused this increase. But the large decrease for acetylene, which Figure 6 indicates was previously one of the two most important VOCs in terms of mass emissions, is also significant. The discussion suggests that it is due to the same changes that caused the large increases in the glyoxals, but it looks like the decrease in acetylene mass is quite a bit greater than the increased mass for the glyoxals. It might be helpful if they specified specifically why acetylene is so high in the INTEX-B inventory compared to the updated inventory in this work. Figure 9 indicates that the acetylene contribution in INTEX-B is huge for all regions except for "Other East Asia".

Response: The decrease of acetylene emissions and increase of glyoxal emissions are mainly due to the change of residential biofuel combustion profile. As can be seen from Fig. 4, the profile used in the original INTEX-B inventory is taken from Tsai et al. (2003) (P2 and P8), which contains high mass fraction of acetylene (27.4% for P2 and 27.0% for P8, respectively) in contrast to the composite profile. This work estimated lower emissions for acetylene (-5.43 Tg compared to INTEX-B), but higher emissions for glyoxal (+0.862 Tg), xylene (+1.63 Tg), and “unknown” (+3.05 Tg).

Table S1 or perhaps a separate table in the Supplementary Materials should indicate which profiles were updated or modified for this work and perhaps have some sort of indication of whether the modifications were potentially significant (e.g., by giving their OFP’s or OVOC fractions). Or were all the profiles listed in Table S1 modified? Were the ones with only one profile listed not modified?

Response: We list the profiles used in the original INTEX-B inventory in Table S1 of the revised paper.

It would be helpful to people interested in this work if the updated profiles developed in this work were available in the supplementary materials or at least at a web site that is cited in this work. By this I mean profiles in terms of chemical compounds or Speciate 4 chemical categories, not in terms of model species. I couldn’t find them at the web site they gave, though maybe I didn’t look in the correct place.

Response: The composite profiles for each source used in this work are now available from the following URL: http://mic.greenresource.cn/intex-b2006. We add the link in the revised paper.

Simon et al (2010) is the only reference citation given regarding the SPECIATE database, which appears to be the primary profile database used in this work. However, the current version of SPECIATE is 4.3, which is dated later. Which SPECIATE version did they use in this work? Would using the latest version affect any of the results or have any profiles that would be better? The reference citation for the SPECIATE database should be given when it is first mentioned, which is not the case in this manuscript. That citation, and the SPECIATE version, should also be given in Table S1.

Response: The version of SPECIATE database used in this work is 4.2, which is added in Table S1 and main text of the revised paper. Using SPECIATE 4.3 would have limited effect on the conclusion of this work as few measurements from Asia were added. More reliable local profiles measured with standardized sampling and analyzing method will be very helpful to improve the quality of profiles in the future.

A number of chemical categories in the SPECIATE database do not have model species assigned to them by Carter (2013). How important were they to the total mass of emissions and the profiles in this work?

Response: Emissions of five species are omitted during the mapping process as shown in below. It can be seen that emissions of those species are small and have little effect on the final emissions. Norfarnesane e (2.05 Gg), Norpristane (1.35 Gg), COAL TAR (0.18 Gg), COPPER NAPHTHENATE (0.18 Gg), and PARAFFINS (C16-C34) (6.76 Gg).

Part of this work involves estimations of ozone formation potentials (OFP's) of the profiles and emissions sources, which is useful. However, many chemical categories used in SPECIATE and presumably the other profiles do not have MIR values assigned to them. How were these treated? In this regard, they state that they ignore OFP's of unidentified mass. Isn't this introducing a bias in the analysis, by assuming poorly characterized profiles are necessarily less reactive? But this is not a major issue if the objective is to see which of the identified compounds attribute to OFP's, as done on Figure 5.

Response: Species with no MIR values were ignored during the calculation of OFPs. We agree that this treatment will introduce uncertainties in evaluating the magnitude of OFPs. As pointed out by the referee, the objective of OFP comparison in this work is to compare the differences of identified compound. We added the statement in the revised manuscript.

It might be an interesting comparison to see a version of Figure 5 for the INTEX-B speciation inventory, given the differences shown in Figure 6.

Response: Agree. We added the comparison in the supporting information of the revised manuscript (Fig. S5), as we feel that the current manuscript is lengthy already. In the original INTEX-B inventory, ethane and xylene are still the largest two contributors to OFP, but with different absolute contributions (50 Tg-O_3 and 14 Tg-O_3 respectively).

The only comment I feel absolutely needs to be addressed concerns the need to indicate the SPECIATE version used. However, I hope they can consider the other suggestions when finalizing this paper.

Response: Thanks again for constructive comments. We believe that we have addressed all comments raised.

Responses to Reviewer #2

The paper titled, "Mapping Asian anthropogenic emissions of non-methane volatile organic compounds to multiple chemical mechanisms" provides a useful summary and details of a method of creating model ready speciated emissions for air quality models. The authors propose a framework for creating chemical mechanism specific model ready files. The paper overall is well written. Hopefully emission inventory and emission processing for other parts of the world can use this paper as a guide when developing speciated emissions. Just a couple of notes and comments that hopefully the authors can address.

Response: We thank Referee #2 for the encouraging comments.

(1) Please indicate the version of SPECIATE used for this paper, since many versions have been released by EPA.

Response: The version of SPECIATE database used in this work is 4.2, which is added in the main text and Table S1 of the revised paper.

(2) The abstract discusses an "improved" speciation framework for Asian emissions. The information used to create the model ready emissions (tables, cross-reference information, etc) should be made available in addition to the model ready data. The availability of the entire framework should be indicated in the abstract.

Response: The framework and data are now available from the following website: http://mic.greenresource.cn/intex-b2006. We also indicated this in the revised abstract.

(3) Page 32653, line 8. The discussion of SMOKE should note that the SMOKE can process emissions for any chemical mechanism for which the user provides the appropriate input. The user is not constrained by the input files provided with SMOKE and it is the user's responsibility to ensure correct chemical mechanism SMOKE inputs.

Response: Corrected in the revised manuscript.

(4) Page 32657, lines 2-5. Sentence beginning "For of the entire.." is awkwardly worded. Recommend: "For all of Asia …" or "For the entirety of Asia…".

Response: Changed.

(5) Page 32659, Line 23: The process of grouping and averaging profiles to make a composite can sometimes introduce additional error or uncertainty.

Response: We agree. We indicated this potential uncertainty in the revised manuscript.

(6) Page 32662, line 24-28; Include a table with the cross referencing of source sectors to spatial proxies (surrogates) even though it is very small. Contrast this with other countries that have a detailed cross referencing of proxies to source categories.

Response: We added a cross-referencing table of sectors to spatial surrogates in the revised manuscript (Table 4).

(7) Page 32662, line 7: Change Cater(2013) to Carter (2013).

Response: Corrected.

Responses to Reviewer #3

<General Comments>

NMVOC mapping is very important procedure to support atmospheric chemistry modeling of ozone and aerosol. This paper by Li et al. integrates many existing chemical speciation information and presents an undated method to convert total NMVOC emissions into model-ready emissions for regional and global chemical transport model (CTM) simulations.

The authors suggest an improved speciation framework of NMVOC in INTEX 2006 Asian inventory adopting an explicit assignment (mechanism-dependent species mapping) and applying updated profiles (composite profiles by median average of local, U.S. SPECIATE and literature profiles). The new mapping methodology is explicitly described and includes some benefits. They added more local source profiles than Zhang et al. (2009) to segregate INTEX 2006 NMVOC into the CTM chemical mechanism species. Using the averaged source profiles seems to be more reasonable approach than single profiles for chemical mapping in the subjected geographical region (Asia).

Overall, their subject regarding an improved mapping method and comprehensive source profiles for the anthropogenic NMVOC emissions are relevant to the scope of ACP. However, there are several thins need to be added/improved as in the following specific comment section, to make this manuscript to be publishable.

Response: We thank Referee #3 for the valuable comments on how to improve the article. We addressed the comments as below.

<Specific Comments>

(1) The speciation profiles, in their present format, are hard to be used by other researchers since they are too summarized or just presented as OFPs. The final speciation profiles developed in this work would better be explicitly presented using comprehensive tabulations, as in Andreae and Merlet (2001), because one of the major virtue of this work is to provide improved speciation information to the related science community.

Response: The tabulated composite profiles used in this work are available from the following website: http://mic.greenresource.cn/intex-b2006.

(2) A comprehensive table which explains mapping between author's chemical species and other major speciation schemes (e.g. SAPRC, GEOS-Chem, MOZART, CB05) would better be developed and presented, instead of individual tables such as Tables 2 and 3.

Response: The mapping tables between individual species and the six major mechanisms (SAPRC-99, SAPRC-07, CBIV, CB05, RADM2, and RACM) are available from Dr. Carter's website: http://www.engr.ucr.edu/~carter/emitdb/. Those tables are too large to be included in the manuscript (including thousands of species). In the Sect. 2.4 of the revised manuscript, we added a sentence to indicate the availability of those mapping tables.

(3) Even though the profile development procedures are described in the chapter 2.2, the reasons of selecting speciation profiles are still unclear. Since large volume local and international speciation profiles should be reviewed during composite profiles development, comprehensive evaluation of existing speciation profiles should be very beneficial to the readers.

Response: We further clarified the profile selection process in the revised manuscript. The profiles are selected with the following steps. We first searched candidate profiles from SPECIATE database and a variety of literatures for each source category. As the numbers of local-measured profiles are still very limited, we include all available "local" profiles from literatures as candidate profiles. For those sources which local profiles are available and we believe that there are significant differences between Asian and western countries due to different technologies and/or legislations, only local profiles are used (e.g. solvent use). For sources which similar technologies are used in Asian and western countries (e.g., boilers, vehicles), profiles from SPECIATE database are also included. We then identified the OVOC rich sources and corrected the incomplete profiles which missed OVOC fraction. The "composite" profile for each source was finally developed with the same weighting factor for each individual candidate profile.

We agree that a comprehensive evaluation of existing profiles would be very useful for understanding the uncertainties. However, it is very difficult to conduct such an evaluation because profiles in literatures are always measured in inconsistent and incomplete ways and many profiles in SPECIATE database are provided without detailed information. In this work, we conducted a sensitivity analysis for a few important source categories, i.e., residential biofuel combustion and on-road vehicles, to evaluate the impact of profile selection to emissions. It is shown that the composite profiles represent the average level of ozone formation for these sources. We expect this sensitivity analysis results can relief the concern on the issue of profile selection to some degree.

(4) As the author have described (Page 32666: 23-24) one of the main objective of their study is to develop model-ready anthropogenic NMVOC emission datasets for CTMs. They applied a new mapping method and developed model-ready emissions for 8 chemical mechanisms, such as CB-IV, CB05, SAPRC-99, SAPRC-07, RADM2, RACM2, GEOS-Chem, and MOZART-4. In the context of the aim of their study, I think that the results and discussions about the consistency of the new mapping method for these chemical mechanism species are important and should be included. I guess that many of the atmospheric chemistry modeling community members also want to see the new NMVOC mapping method can generate a certain level of consistent results for

multiple chemical mechanisms.

Response: The mapping tables we used in this work are developed by Dr. Carter (Carter 2013, http://www.engr.ucr.edu/~carter/emitdb/), who is the developer of SAPRC mechanisms and the leading scientist in this field. In these mapping tables, each individual organic compound is assigned with conversion factors to mechanism species according to its carbon bond (for CBIV and CB05) and chemical group (for (SAPRC-99, SAPRC-07, RADM2, and RACM2), hence it provides a consistent way of species mapping for different chemical mechanisms. To our knowledge, this is the most accurate chemical mapping approach in the community, which has been used in processing U.S. emission inventories. However, in previous Asian emission inventory studies, the interfaces between NMVOC emissions and CTMs are underdeveloped. In this work, we improved the species mapping framework for Asia by using the accurate mapping table from Carter (2013).

(5) To present the effect of the new mapping methodology for Asian anthropogenic NMVOC emissions, the author compared the OFP that calculated with the newly mapped INTEX 2006 NMVOC emissions by applying MIR scale values and the OFP with the previous emissions. The MIR approach assumes high NOx (or highly VOC sensitive) condition for ozone formation (Carter 1994). If some countries or regions in Asia are highly VOC-sensitive, for which the MIR based OFP may be somewhat useful to investigate the effect of the newly derived emissions of mechanism species on ozone formation. If some countries or regions in Asia are highly NOx-sensitive, for which the MIR-based OFP is not appropriate. The availabilities of VOC and NOx can be affected by meteorological factors. Therefore, the MIR based OFP calculation without any consideration of the geographical distribution of NOx and meteorology in the subjected geographical region cannot reasonably present the effect of the new NMVOC mapping methodology. My suggestion is that a comparison with at least a CTM (e.g., GEO-Scheme, CMAQ, etc.) simulation results (New – versus Previous-mapping) should be included in this paper to investigate the valid effect of the new NMVOC mapping method on ozone (or aerosol) prediction in Asia.

Response: We agree that comparing emissions in CTMs is a good way of evaluating emission inventories. However, we feel that it is beyond the scope of this manuscript for the following two reasons. First, the main objective of this paper is to provide an improved NMVOC speciation framework for Asian regions. As pointed by another referee, the methodology presented in this work can be used as guidelines in developing speciated emissions. We used the same mapping method for both datasets (defined as “INTEX-B” and “this work” in the manuscript), with the differences in profile development and selection. In this respect, we think that evaluating the impact of profile selection by using CTM is not important for this paper, given the fact that we have demonstrated the impact by comparing OFPs. Second, most emission inventory papers do not include such evaluation. The community usually rolls in the following way: emission inventory groups developed bottom-up inventories and published their data; modeling groups evaluated the inventory and provided feedback; then emission inventory developers improved their work based on the evaluation. Compiling emission inventory is a very time consuming work, and many emission inventory groups do not have the capacity to run models. Setting a bar of including CTM evaluation in emission inventory papers would be unfair to emission inventory developers.

(6) Since isoprene and terpenes are also very important precursor species of ozone and aerosol formation, these are considered as primary explicit organic or lumped organic species in most of chemical mechanism such as SAPRC 99/07, CB05, and so forth. I cannot find any presentation for these species in this paper. Are their emissions negligible because this paper only covers anthropogenic emissions?

Response: Yes, this work only addressed anthropogenic emissions in which isoprene and terpenes are negligible.

(7) Although authors clearly outlined their methods, some parts of their assumption have weaknesses (e.g., ozone forming potential (OFP) calculation). In addition, the results are somewhat insufficient to support their objective and derive coherent discussions. In the context of their subject, this paper should contain some results and discussions whether the new method can yield consistent emission mapping results for different chemical mechanisms. In addition, it is needed to add reasonable investigations and discussions about how much are the new mapping method and data effective for the CTM ozone and aerosol prediction.

Response: See responses above.

<Minor Comments>
(1) The authors need to clarify the versions of SPECIATE.

Response: The version of SPECIATE database used in this work is 4.2, which is added in the main text and Table S1 of the revised paper.

(2) Figure 10: need separate labels for figures (a) and (b).

Response: Corrected.

References:

Carter, W. P. L.: Development of an improved chemical speciation database for processing emissions of volatile organic compounds for air quality models, report available at: http://www.engr.ucr.edu/~carter/emitdb/, 2013.