Interactive comment on "Volatile organic compounds over Eastern Himalaya, India: temporal variation and source characterization using Positive Matrix Factorization" *by* C. Sarkar et al.

Anonymous Referee #1

Received and published: 10 January 2015

This study presents measurements of 18 VOCs in Darjeeling, Himalaya and uses PMF to apportion the sources among diesel, gasoline, biomass burning, solvents and so forth. Unfortunately I have many concerns with this paper (analytical methods need more description; precision and accuracy need to be stated; BTEX does not include p-xylene, yet comparisons were made with other cities; paper needs editing for grammar; too much self-referencing; often too many significant figures; etc.). A major scientific concern is that the reported CCl_4 concentrations are not plausible in today's atmosphere (they have not been as low as reported here since the 1940s), but this wasn't

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recognized by the authors. The second major concern is apparent plagiarism (five examples in the introduction, after which I stopped checking). Due to numerous concerns about this paper, unfortunately I must recommend that it be rejected. Detailed comments follow.

P32135 L8: Use fewer significant figures (four is too many especially with such large error bars). Same comment elsewhere in the paper.

P32135 L14: "related mainly to tourists". Is this speculation or is there evidence for this? What percentage of vehicle emissions, biomass and coal burning, etc. are due to tourists rather than the local population? For example it is surprising that coal burning and solvent use would be mainly related to tourists.

P32135 L20: Please add: "Of the measured compounds, diesel exhaust was also found..." Many compounds that impact ozone formation were not measured, so the conclusions are limited to the 18 species presented here.

P32136 L4: Guo isn't really the right reference for this. Use a primary reference.

P32136 L7-10: "to form SOA by nucleation and condensation with a significant aerosol yield and thus they influence gas phase pollutants directly and particle-phase pollutants indirectly." This text is very similar to that in Saxena and Ghosh (2012), which directly quotes Brocco et al. (1997) and Odum (1997): "contribute to SOA formation by nucleation and condensation ... with a significant aerosol yield and therefore, aromatic VOCs influence gas phase pollutants directly and particle-phase pollutants indirectly." At very least the Brocco and Odum sources should be cited.

P32136 L19: This statement isn't correct. VOC sources in Asia are highly complex have been attributed to many sources, not just vehicular emissions. The single self-reference to a study of urban petrol centres in India (Srivastava et al., 2005a) is confusing because it does not capture the complexity of sources throughout Asia.

General comment: There is a lot of self-referencing in this paper, including 10 refer-

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ences to papers led by Srivastava.

P32137 L9-11: I am concerned about plagiarism in this paper. Lines 9-11 read: "Such studies over Himalayan region are of paramount interest as the ecology of the Himalaya is under serious threat from various forms of pollutants (Bostrom, 2002)." This is identical to a recent paper by Adak et al. (2014) on aerosols (many of the same co-authors): "Further, the study of aerosol over Himalayan region is of paramount interest as the ecology of the Himalaya is under serious threat from various forms of pollutants (Bostrom, 2002)." You can also find the same sentence in Sharma et al. (2011) (different co-authors): "Further this study in the Himalayan region of Kullu-Manali is of particular interest as the ecology of the Himalaya is under serious threat from various forms of pollutants (Bostrom 2002)." The sentence should have been changed and also had some attribution to Sharma et al.

P32137 L11-14: The next sentence is also plagiarized. It reads: "The increase in the loading of atmospheric pollutants over the Himalaya is a matter of concern, since most of the glaciers in the region have been retreating since 1850 (Mayewski et al., 1979) with increasing melting rates." This is identical to Adak et al. (2014): "The increase in the loading of atmospheric aerosols over the Himalaya is a matter of concern, since most of the glaciers in the region have been retreating since 1850 (Mayewski et al., 1979) with increasing melting rates." Also similar to Chatterjee et al. (2010) except the topic is changed from aerosols to atmospheric pollutants: "The transport of optically-active aerosol to the higher Himalayas is a matter of concern, since most of the glaciers in the region have been retreating since 1850 (5) with increasing melting rates."

P32137 L14-17: Same with the next sentence: "The rising anthropogenic interferences for rapid urbanization and development in the Himalaya not only affect the immediate landscape environment, but also the atmospheric environment which is becoming an increasing concern (Momin et al., 1999)." This is also identical to Adak et al. (2014): "The rising anthropogenic interferences for rapid urbanization and development in the Himalaya not only affect the immediate landscape environment, but also the atmo-

spheric environment which is becoming an increasing concern (Momin et al., 1999)."

P32137 L17-21: Same with the next sentence: "The anthropogenic activities such as increasing vehicular traffic due to increased tourism-related activities, biomass burning and fuel wood burning for cooking and heating are the causes of concern for most of the Himalayan high altitude hill stations in India which apparently look like pollution-free regions as situated far away from the Indian mega-cities." This is also identical to Adak et al. (2014): "The anthropogenic activities such as increasing vehicular traffic due to increased tourism-related activities, biomass burning and fuel wood burning for cooking and heating are the causes of concern for most of the Himalayan high altitude hill stations in India which apparently look like pollution-free regions as situated far away from the Indian mega-cities." I am stopping looking for other examples at this point, but much of the introduction is essentially identical to a previous paper by many of the same co-authors in 2014, which in turn used some of the same sentences as earlier manuscripts.

P32138 L16: A map still needs to be shown here (this paper needs to stand alone). You could even use Figure 3.

P32139 L2: Rainfall amount has too many significant figures.

P32139 L9: What was the sample integration time? Is it 12 hours?

P32139 L10: Two samples collected once a week for a year should give about 100 samples . . . why are there only 90?

P32139 L11: What tests have you done to ensure that the custom-made glass sampling tube is appropriate for VOC measurements? Have you done sensitivity tests to see whether VOCs might interact with the surface or whether concentrations in the tubes change with time? How soon after sample collection was the analysis done?

P32139 L20: Are the CPCB references accessible to the general reader?

P32140 L1: More information needs to be given about the quality of the measurements.

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What is the detection limit? What is the precision? What is the accuracy? How has this been demonstrated? I have not heard of the German company; are they synced to internationally recognized calibration scales? Some reference or website needs to be provided for this company.

P32140 L2: 'For estimation of the target compounds external five point calibration curve was prepared in triplicate using VOC mix 20 by Dr. Ehrenstorfer GmbH, Germany.' As discussed in the comment below, the CCl4 concentrations presented here are not plausible in today's atmosphere. Therefore the five point calibration curve in triplicate appears to have been ineffective for CCl4, leading to concern about the quality of the remaining VOC measurements.

Table 1 gives a mean CCl4 concentration of 0.18 ug/m3. The molecular weight of CCl4 is 153.82 g/mol, so this translates to about 30 ppt. CCl4 is long-lived and very well mixed in the atmosphere, with global concentrations in 2011-2012 of about 85 ppt, without much interhemispheric difference (http://cdiac.ornl.gov/oceans/new_atmCFC.html). CCl4 concentrations have not been as low as 30 ppt since the 1940s, and there is nowhere on Earth today where you can find a reading this low. This is also not a question of altitude (the measurements from this paper were taken at an altitude of 2200 m) as demonstrated by aircraft measurements of CCl4 throughout the troposphere. I am concerned both at the quality of the CCl4 measurements (and therefore of the other VOC measurements), and that the authors did not recognize that this concentration is not possible in today's atmosphere.

P32140 L20: Of the xylenes is appears that only m-xylene and o-xylene are measured (not p-xylene). How does this therefore affect comparisons with other cities (Table 4), given that one of the components of BTEX is missing?

P32142 L11: What evidence is there that VOC emissions remained comparable during pre- and post-monsoon?

P32143 L1 and elsewhere: Given that meteorology is being used to explain much of the

seasonality in concentrations, it would help to show plots of weekly temperature and insolation, so the reader can see how this relates to seasonal variability. Likewise it would help to show the individual data points with time, rather than seasonal averages, to see if the correlations make sense.

P32143 L25: Please provide a more in-depth statistical analysis, rather than 'greater than 1.0'.

Section 4.3: While they may be correct, the arguments in this section all seem speculative. Please provide more concrete arguments.

P32144 L25 and throughout: There are too many significant figures. Make sure the significant figures match the error bar and the measurement precision/accuracy. It seems unlikely to be able to measure these numbers to within 0.1 ug/m3.

P32145 L1: Because the error bars on the concentrations are large, also use error bars for the factors of 7 and 1.5.

P32146 L10-24: This is introductory/methods material, not results.

P32147 L26: All readers might not be familiar with PMF. Describe the 'decay adjustment problem' in more detail. On P32148 L3 describe 'stable Q values'.

P32148 L7: Provide some literature that shows the typical B/T ratio in gasoline exhaust and gasoline evaporation. How does it compare to the B/T ratio measured here?

P32149 L17: Do back-trajectories confirm any history of travel over lowland townships and cities?

P32149 L23: Provide more detail of the 'various environments'.

P32150 L15-17: General comment: This study provides a limited range of VOCs, so concluding that the major source of VOCs in Darjeeling is vehicular exhaust only applies to these 18 VOCs. A wider range of compounds would have given different results. Please discuss this further.

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P32151 L9-11: Again, this conclusion is based on only 18 VOCs, plus does not factor in the role NOx might play.

P32152 L25: Too many significant figures.

Table 1: There are too many significant figures in Table 1: 2304.38 is not credible. Determine the correct number of significant figures based on the precision of the measurements.

Table 1: The reader needs to be told the DL in order to know what BDL is.

Table 2: Too many significant figures here as well.

Figure 4 needs labeling to show which data points correspond to which VOCs.

Figure 4: The number of significant figures needs to be reduced to a more reasonable number.

Figure 5: After the discussion in the introduction about vehicles and biomass burning, I'm surprised that biomass burning isn't one of the factors (different from coal burning). For example benzene is a general combustion tracer. Please discuss.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 32133, 2014.