

First we would like to thank the reviewers for their valuable comments and effort to improve the manuscript. We have addressed all comments as can be seen in the enclosed list. The original reviewer comments are shown in italics, while our responses are shown in bold text.

Reviewer #1:

Dear Editor,

Please find attached my remarks on manuscript "Comparison of different real time VOC measurement techniques in a ponderosa pine forest. Kaser et al. Atmos. Chem. Phys. Discuss., 12, 1–34, 2012 version 4. The manuscript has been improved since version 1. This manuscript presents interesting results in an organized way. It covers important issues on the measurements of VOCs by proton transfer reaction mass spectrometry supported with chromatographic analyses.

In order to improve the manuscript, find my remarks and comments below.

Page 5, line 2: "co-located measurement": Please add a few words on the horizontal distribution of the measurement towers.

All our VOC measurements were conducted on the same tower. Co-located measurements refer to measurements on the same tower. For clarity this can be added in the revised manuscript.

Page 6, line 4: This paper clearly is a VOC measurement intercomparison. Please include a reference of an accompanying paper to read more on the atmospheric interpretation of these data.

The quality of the measured data is assured in this comparison study. So far part of this data has been used in some studies (DiGangi et al. 2011, 2012, Kim et al. 2012). Several other studies addressing the atmospheric interpretation of this compared dataset are in preparation and this comparison study will be very valuable for those. There will be another manuscript Kaser et al. dealing with eddy covariant flux measurements at the same site.

Page 6, line 6: Does "summer of 2010" refer to the period of August 10 to August 21?

"summer of 2010" refers to the whole measurement campaign BEACHON-ROCS that took place from end of July to the beginning of September 2010. For the comparison study the period of August 10 to August 21 was chosen, as this was a time frame where all compared instruments were measuring simultaneously. This is clarified in the revised manuscript.

Page 6, line 20: Please provide details on the atmospheric conditions (precipitation, history of the air mass,... for the data under discussion here (10/08 - 21/08) or include a reference that covers these topics.

Details on atmospheric conditions such as wind direction and history of the air mass are presented in Fry et al. (2012) for the same measurement site during the BEACHON ROMBAS field campaign in July/August 2011. The synoptic wind patterns were similar during the BEACHON ROCS study in August 2010. Every night winds are from the south and southwest (drainage flows down the valley). During the day wind shifts either

to the west, which usually brings clean air or originates from an upslope flow from the east (NE to SE), which typically brings pollution from the Front Range.

Typically days were sunny in the morning and some clouds build up in the afternoon. Most of the days were dry with a small amount of precipitation on August 19th (DOY 231). Shortly before the comparison period the site experienced a more significant precipitation event on August 9th (DOY 221). Details on the atmospheric conditions as well as a reference describing the meteorology at this site in detail will be added to the revised manuscript.

Page 7, line 8: Was all tubing of equal length for the gradient system?

Each inlet of the gradient system was of similar length, about 35m each. Only data from the upper most gradient inlet line system, for which we stated the length in the manuscript, is used and therefore no more information was added to the updated manuscript.

Page 7, line 9: comma misuse: "A second (eddy-covariance, EC) inlet was used."

This is changed in the revised manuscript.

Page 7: please, state to flow through all inlet lines. Has the residence time in the tubing been calculated and taken into account for the instrumental intercomparison?

The flow through the gradient inlet line (35m long, 1/4" OD) was 3.5 SLPM, the flow through the EC inlet line (35m long, 3/8" OD) was ~20 SLPM and the inlet line used by FILIF (35m long, 3/8" ID) was 80 SLPM. This results in residence times of 7, 4.5 and 2 s, respectively. The delay due to the residence time in the lines is very short compared to the measurement times of several minutes and was therefore not taken into account. The flow values through the different inlet lines are added in the revised manuscript.

Page 8, line 18: limits of detection (LOD) were determined from the 2 sigma. Why not 3 sigma? LOD refers to the lowest quantity of a substance that can be distinguished from the absence of that substance thus the background signal. The background signals were scarcely recorded for some of the instruments used in this study for a thorough background evaluation and interpolation over the course of time. Please comment.

We decided to be consistent with earlier publications in the PTR-MS community and therefore used 2 sigma of the standard deviation of the background system. This enables an easier comparison to already published papers in this field.

Page 11, line 23: please, add a short description on how the LOD has been calculated for the TOGA as the 2 sigma-rule applied to determine the LOD of the PTR-(TOF)-MS cannot be not apply to chromatographic methods in the same manner.

TOGA limits of detection are defined as a signal to noise (S/N) = 5 for chromatographic peaks. This description is added to the revised manuscript.

Page 13, line 20: please, describe how the whole air samples were obtained. Have the canisters been evacuated and then filled up to atmospheric pressure? Or filled with overpressure?

This two questions were already described in the paper under review (see page 13 line 13: The 2-liter evacuated stainless steel canisters were filled to approx. 30 psi.)

How have they been stored (temperature controlled)?

Canisters were stored in the main operations building prior to shipping and were not temperature controlled during shipment. Moreover, with our 20+ years of experience with these particular canisters (2 liter electro-polished stainless steel from the University of California, Irvine) and shipping them all over the world, temperature control during shipping and storage is not necessary nor has it proven to be problematic. There are an array of publications from the UCI (Rowland/Blake Group) and Sive groups demonstrating this, so we refer the reviewers to the literature if there are further questions regarding the storage of the canister. Worth noting, the canisters were shipped via FedEx to the University of New Hampshire where they were analyzed using a multi-column multi-detector GC system (described below).

how much time was there between sampling and analysis?

All canisters were analyzed within two months of sample collection. In Russo et al. (2010b), we have quantitatively shown that samples can be stored for 3+ months (for most gases) without compromising the sample integrity. Canister storing time is added to the revised manuscript.

Which instrument has been used to analyze them? Include the calibration procedure.

Details regarding the sample analyses and calibrations are outlined in Sive et al. (2005, 2007), Zhou et al. (2005, 2008), White et al. (2008, 2009, 2010), Russo et al. (2010 a, b), Ambrose et al. (2007, 2010), Talbot et al. (2005, 2011) in addition to others. We referred to several of these papers in the manuscript for the specific details of the analytical procedures. We added all the above mentioned publications to the reference list at the end of this answer to the reviewer comments which are directly related to the canister sampling in this manuscript. Additionally we here give a summary of the analytical details regarding the sample analysis and calibration procedures.

The canister samples were analyzed in the laboratory at UNH for C₂-C₁₀ nonmethane hydrocarbons, C₁-C₅ alkyl nitrates, C₁-C₂ halocarbons, several OVOCs, and select sulfur compounds. A three gas chromatograph system equipped with two flame ionization detectors (FID), two electron capture detectors (ECD), and a mass spectrometer (MS) was used for analysis of each 1500 cc (STP) sample aliquot. A PLOT-FID, OV-1701-ECD, VF-1ms -FID, OV-624-MS, and a CP-PoraBond-Q/Restek XTI-5-ECD column-detector combinations were used to separate and quantify a wide range of VOCs. The MS was operated in electron impact mode with single ion monitoring for measuring OVOCs and sulfur compounds, as well as duplicate measurements of several halocarbons and NMHCs. A 1500 cc aliquot from one of two working standards was

assayed every ninth analysis. The measurement precision for each of the halocarbons, hydrocarbons and alkyl nitrates, sulfur compounds and OVOCs ranged from 0.1–12%.

In order to ensure that the VOC mixing ratios in samples analyzed at different times are comparable, whole air and synthetic standards were routinely analyzed and calibration scales were cross referenced and validated. When conducting the canister sample analysis, one of two whole air standards was analyzed after every eighth sample in order to monitor changes in detector sensitivity and measurement precision and to quickly detect any analytical problems. The whole air working standards employed for this work have mixing ratios representative of clean free tropospheric air and suburban air, thus bracketing the low and high ranges for these measurements. Moreover, linearity studies are regularly conducted to evaluate the detector response over the observed mixing ratio ranges for all classes of compounds. The working standards were part of the larger network of whole air standards maintained by B. Sive at UNH as part of the AIRMAP program. In total, there are currently ten high pressure cylinders, six 36-liter electropolished low-pressure pontoons (~350 psi), and three 34-liter electropolished high pressure pontoons (~900 psi) containing whole air standards that have been filled and calibrated by UCI (D. Blake) and B. Sive. We estimate the upper limit of the absolute accuracy of the calibrated standards to be on the order of ± 1 –5% for the gases reported here. In addition to the whole air standards, calibrations are conducted using five different high-pressure cylinders containing synthetic blends of selected NMHCs, OVOCs, and halocarbons at the ppbv level (Apel-Riemer Environmental, Inc.). The absolute accuracy for all of the gases in the synthetic standards is ± 5 %.

Page 14, line 4: I'm wondering why the correlation coefficient R, which is measures for the strength and the direction of a linear relationship between two variables, has been chosen over the coefficient of determination R² which represents the percent of the data that is the closest to the line of best fit.

We appreciate this comment of the reviewer. As in the case of a linear fit the correlation coefficient R and the coefficient of determination of R² are closely related we justify the use of R.

Also we decided to be consistent to earlier intercomparison studies (e.g. Apel et al. 2008) where also R is used as a measure of the fit.

Page 14, line 22: please, provide details on the diurnal and nocturnal boundary layer conditions (depth, sunrise, sunset, humidity, clouds, . . .) or refer to an accompanying paper that discusses these issues for this field study.

During the BEACHON ROCS measurement campaign in August 2010 sunrise was at approximately at 6 AM and sunset around 6 PM which was determined by measured PAR values. The change of sign in the temperature difference measured at 25 and 2 meters above ground can give an indication on the stability of the atmosphere. Unstable conditions were observed during the day between 8 AM and 4 PM and stable conditions during the night. Relative humidity ranged from 10-20% during the day and 70-90% during the night. Typically days were sunny in the morning and some clouds build up in the afternoon. This details were added to the description of the field site. A reference

describing the opposite diurnal pattern of MBO and MT at this site related to the boundary layer height is added to this part of the text to strengthen our argument on why monoterpene concentrations are high during the night and low during the day and opposite for MBO concentrations.

Page 17, line 8: It is known in the literature that the concentration of isoprene and other reactive compounds are not stable in canisters which have been stored.

While we appreciate the reviewers concern about isoprene and other reactive compounds stability in canisters, this is not the case. Even gases such as MBO can be quantified if proper care is taken – see below.

The canisters used in this study from UCI are superior to other canister on the market – we have shown this for an array of different compound classes and gases. Moreover, the Sive, Apel and UCI laboratories have all spent an extraordinary amount of time and effort making sure that when data from these canisters are generated, they are both quantitatively and qualitatively accurate. We have done numerous studies demonstrating that ozone does not react with isoprene and other gases in canisters when a sample is collected and gases like isoprene (and other alkenes, including monoterpenes) are stable. We do acknowledge that the long-term stability of MBO in ambient air samples can be problematic, careful characterization of the canisters and a rapid turnaround time for the analysis results in reliable measurements. We refer the reviewer to the list of published papers on the canisters regarding isoprene stability in addition to the other compounds.. Worth noting, E. Apel headed up the Nonmethane Hydrocarbon Intercomparison Experiment (NOMHICE). Moreover, papers by the UCI group (D. Blake, leader) in addition to many others demonstrate that canister measurements, when properly conducted, result in very high quality measurements. In summary, the methods of E. Apel and B. Sive have been rigorously tested through both formal and informal measurement intercomparison experiments and their analytical procedures have consistently yielded excellent quantitative results.

Page 17, line 13: The PTRMS has been measuring the gradient in the forest at high time resolution compared to the TOGA but is not used in the discussion here. That's odd. Furthermore, with an EC system on site a very good filter can be made to subdivide the data set into periods during for which advection and convection dominated. Please comment.

Gradient measurements of the sum of 232-MBO and isoprene obtained by PTRMS showed that concentrations peaked at 12 m (middle of canopy) decreasing by 20% towards the ground. These measurements can however not exclude the presence of local isoprene sources. Overall we believe that we can not fully answer the question whether there are any local isoprene sources and propose that selective eddy covariance measurements of isoprene and more plant cuvette measurements are needed. While eddy covariance measurements can be used to determine the stability of the atmosphere as pointed out by the reviewer, we don't think the dataset is sufficiently constrained to tease out advection terms, since this would have required a horizontal advection study with concentrations measurements performed upwind.

Page 17, line 18: as OH concentrations are not under discussion here, please refer to the paper that describes these measurements for this study. Then, mention here the concentration range for OH and O3 measured during this study and perhaps add a reference which includes the calculation of the photochemical age.

OH concentration measurements are in detail described by Kim et al. (2012). OH ranged from 1-6 10^6 molecules cm^{-3} and Ozone from 5-70 ppbv (study average of 34 ppbv). Photochemical age was calculated as described by Apel et al. (2002). We have included this in the revised manuscript.

Page 29 Table 1: to complete the table, I suggest including the measurement frequency and the frequency of the background analyses.

Two rows are added to Table 1 including time average for the comparison study for each instrument as well as details on the frequency of background measurements.

Reviewer #2:

Authors compare field measurements data between different instruments and show good agreements for some compounds. Comparisons between on-site GCMS and PTR-TOF-MS and between PTR-MS and PTR-TOF-MS are interesting. I would like to ask authors to consider comments below.

Some papers show, in PTR-based systems, that relative abundances of molecular ion and major fragment ion are affected by E/N and humidity. Compounds MAC and cymene produce significant amount of fragment ions. Sensitivity of molecular ions of the compounds measured in the study is also not constant across humidity, in particular at lower E/N (<110 Td). Authors need to check difference in mixing ratio of water vapor between calibration and field air. As humidity affects relative abundances of molecular ion and major fragment ion, this also may cause error in determining the concentration.

The PTR-(TOF)-MS instruments were calibrated on site using a VOC gas standard diluted with air through a catalytic converter. The air used was ambient air (containing ambient humidity) and therefore the calibrations are representative for our measurements. MAC and cymene are addressed more in detail in the answers below.

Method section.

1 What was the range of humidity (or mixing ratio of water vapor) in the field air and various calibration air? As it affects PTR-MS sensitivity and abundance of fragmentation, it should be mentioned in manuscript.

Absolute humidity ranged from 10-25 mmol/mol. during the comparison period. The range covered by our calibrations using ambient air humidity as described above was 14-25 mmol/mol. The sensitivity variation within this humidity range was smaller than 4% and therefore clearly within our stated accuracy (15%). We justify therefore the use of an average sensitivity over the whole humidity range. Some text is added to the revised manuscript describing the observed humidity dependent calibrations.

2 How did the authors determine PTR sensitivities for MAC? MAC is not included in the compounds used for calibration.

During this study a gas standard containing only MVK was available for calibration. As shown by Liu et al. (2012) sensitivities for MVK and MAC are very similar. Furthermore Zhao and Zhang (2004) calculated that the reaction rate coefficients for both compounds are very similar. Therefore we justify to use the same sensitivity for MAC as calibrated for MVK. A sentence clarifying this will be added to the revised manuscript.

Result and discussion.

3 Isoprene. Some data were far from 1:1 line. Some are quite higher in PTR-MS and others higher in PTR-TOF-MS (Fig. 2). Authors should mention the difference and address the reason.

Comparing online measurements in a real forest (strong source of BVOC emissions) is especially complicated if the instruments do not have exactly the same “sampling time” or as in the case of PTR-(TOF)-MS one instrument is measuring disjunct (PTR-MS) and the other continuous (PTR-TOF-MS). This averaging/interpolating problems become especially evident for VOCs originating from a strong emission source close to the inlet line as is the case for MBO + isoprene above the Ponderosa Pine forest during daytime (under high turbulence conditions). The three measurement points (marked in blue in the updated figure in the updated manuscript) are such cases where interpolation between PTR-MS and PTR-TOF-MS datasets works poorly as MBO + isoprene shows highly fluctuating concentrations within less than minutes. We chose to mark those measurement points in blue in the updated figure to visualize the issue of interpolating data with different time stamps but decided to omit them in the calculation of the correlation coefficient. This results in a change from R=0.77 to R=0.84. Figure 2a and 3, table 2 and the text is updated accordingly.

4 Monoterpene. line 12-13 “due to the uncertainties in the calibration factors used for the remaining monoterpenes.” Tell us the detail please. Reaction rate constant or relative abundance of m/z 81+137?

This sentence refers to TOGA and not to PTR-MS. It is therefore not related to m/z 81 + 137 but to the fact that not all monoterpenes were calibrated by TOGA which leads to some uncertainties in the results for monoterpenes from TOGA. This is clarified in the revised version of the manuscript.

5 MAC and MVK. Does MAC produce a fragment ion of m/z 43? The relative abundances of molecular ion and the fragment ion are affected by humidity. The abundance of the molecular ion was higher under higher humidity conditions and lower E/N value. How much degree are PTR sensitivities for molecular ions of the 2 compounds affected by humidity? Humidity difference between calibration and field air and between individual measurements may cause significant error. If the author monitored the molecular ion only, the concentration of MAC+MVK might have been overestimated.

The PTR-TOF-MS used in this study was coupled right after this measurement campaign to a GC system (Langebner et al. in preparation). When using the same instrument settings no fragment of MAC at m/z 43 was observed, which should clearly arrive at the same retention time as the MAC molecular ion. Therefore we can exclude this possible overestimation. As described above calibrations using ambient humidity were conducted. Sensitivities of MVK used also for MAC changed only slightly (<2%) with varying humidity which is much less than our given instrument accuracy of 15%.

The comment regarding the lower E/N would need to be addressed only for PTR-MS, which was not monitoring m/z 71 and was therefore not available in this comparison study.

6 Toluene (Cymene). Same suggestion is given for cymene. Humidity difference between calibration and measurement air and between individual measurements may cause error in determining contribution of m/z 93 originated from cymene. The 2 E/N values give different relative abundance of m/z 93 ion.

The change in fragmentation pattern of cymene is only slightly humidity dependent shown by Tani et al. (2004). The fraction of the m/z 93 fragment of cymene at 124Td changed from 0.67 (RH 98%) to 0.71 (RH 28%). Our calibration for cymene was conducted at RH 60% that is between the two values reported from Tani et al. (2004). As even at these extreme values of RH change the fragmentation only slightly we justify the use of our calibration for all different humidity's. A sentence describing this is added to the updated manuscript. Again only measurements from PTR-TOF-MS and not PTR-MS (as m/z 135 was not monitored by PTR-MS) are available. Therefore the difference coming from different E/N values cannot (does not need) to be addressed.

7 Conclusion. Line 7 "At this site were MBO and monoterpene emissions are dominating" The word "were" should be replaced by "where"?

This is changed in the revised manuscript.

List of references for this answer to the reviewer's comments:

Ambrose, J. L., K. Haase, R. S. Russo, Y. Zhou, M. L. White, E. K. Frinak, H. R. Mayne, R. Talbot, and B. C. Sive: An Intercomparison of GC-FID and PTR-MS Toluene Measurements Under Conditions of Enhanced Monoterpenes Loading, *Atmos. Meas. Tech.*, 3, 959-980, 2010.

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