We thank the anonymous referee for reading the paper carefully and providing thoughtful comments, many of which have resulted in changes to the revised version of the manuscript. We reply to each comment below in bold text.

Anonymous Referee #1

The authors report the application of a chemical titration system (IPI) for OH radicals, in order to determine the chemical zero of an OH LIF instrument. The system is similar to what has been reported by a different group (Mao et al., 2012) for a similar instrument. This system allows to identify interferences in the OH measurement by OH that is artificially produced inside the measurement cell. This OH would be attributed as ambient OH, if only the traditional modulation of the excitation wavelength is used, in order to determine the background signal of the instrument. It turns out that a substantial amount of the total fluorescence signal is caused by artificial OH. Therefore, this paper provides important information about measurements done with this particular instrument. Although the magnitude of this interference most likely strongly depends on the specific instrument and environment, in which OH measurements by LIF are performed, as pointed out by the authors, this report is also of great interest for other groups applying LIF for the detection of ambient OH.

This LIF instrument with the IPI system measured in three campaigns. The authors discuss in detail the technical part of the IPI system and how they determined ambient OH concentrations from the different signals they got from the LIF system, when they use the chemical titration scheme. They also report instrumental tests that were done during the measurement campaigns. A short discussion about the potential impact of the large background OH signal they observed in these campaigns, when they applied the IPI-system, on measurement during previous campaigns is given. All this is carefully done and clearly written in the manuscript.

The major criticism I have concerns what is not discussed in the manuscript. The manuscript is limited to what has been achieved during the three measurement campaigns. Most parts of the paper are like a technical description of the instrument which describes, how measurements were performed during the specific campaigns. This alone is important and makes certainly a paper that well fits within the scope of this journal. A major improvement of the instrument is described that is of great interest for the readership. However, I expected to read more about laboratory tests, comparison with a similar systems and literature, analysis of the dependence of the OH background signal on instrumental parameters and an analysis of potential reasons for this signal.

We are grateful for the comments but respectfully disagree with the reviewer on the point regarding the paper only containing descriptions of field data. A large suite of tests on the system are described in the manuscript. For reasons explained in the manuscript, i.e., the difficulty of supplying enough zero air to the IPI system, a large part of the characterization was completed whilst on a field using ambient air. At the same time, many tests were done with a stable source of OH radicals, making them equivalent to laboratory measurements. A follow-up paper has been submitted to PCCP, focusing on the nature of the interfering compound and its chemistry, which is based on laboratory studies, but we felt that this work was tangential to the description of using the IPI system to eliminate the interference in the OH FAGE measurements. In the revised paper we mention this and provide the reference.

This could include the following points:

General comments:

1. Application of the IPI-system with an OH source in laboratory tests, in order to determine the OH-loss in the IPI-system.

We agree that such tests would be beneficial, yet it is very difficult to produce a stable source of OH with such a large flow rate as required by the IPI system making such tests highly impractical. We instead completed these tests using ambient air in the way described in the manuscript and assumed a constant loss rate of the species causing the background OH. Within the practical limitations, we think that these tests were the most appropriate way of accounting for losses of OH and the atmospheric OH radical data intercomparison provide confidence that our assumptions are reasonable.

2. Analysis of the dependence of the interfering signal on instrumental parameters such as laser power or pressure in the low pressure region of the instrument.

The authors agree with the importance of these tests therefore we added a section in the results chapter that includes a discussion about the dependence of the interfering signal on instrumental parameters such as internal pressure, internal flushing and laser power. Please refer to Section 3.3 for this addition.

3. Tests of potential candidates which could be responsible for the observed background signal in laboratory experiments. Such tests would be also interesting even if no interference is found, in order to exclude potential interferences.

The HORUS instrument in use at the Max Planck Institute for Chemistry is based on the GTHOS (Ground Tropospheric Hydrogen Oxide Sensor) instrument described by Faloona et al. (2004). A large number of tests were performed on GTHOS and were presented in the publication by Ren et al. (2004). Some of these tests such as interference from SO_2, acetone and ozone and generation of OH caused by double pulsing were performed using HORUS and showed negligible effects at atmospheric conditions. Double pulsing is, in general, not a problem in the HORUS instrument as the residence time of an air parcel in the white cell is more than two times shorter than the time period between two consecutive laser pulses with a laser repetition rate of 3000 Hz. In agreement with this the interfering OH measured in ambient air showed no correlation with the product of laser power, ozone and water vapor concentrations. In addition, water vapor and ozone concentrations were comparatively similar during HUMPPA and HOPE campaigns while the laser power in the detection cell during HOPE was twice as much compared to HUMPPA but in contrast, the background OH was on average three times higher during HUMPPA than HOPE. The discussion about potential candidates has been added to Section 3.3.

4. Mao et al. report a significant removal of the interfering OH inside the measurement cell. They report a destruction of 80-94% of ambient OH in their inlet system when they add 1-3.3sccm C_3F_6 (residence time 100 ms). Here, the authors also remove 95% of the ambient OH within 2.5-4ms residence time in the IPI, so that the OH reactivity of the sampled air is most likely even larger than for the system described by Mao et al., when they add the OH scavenger. One

may suspect that also a significant part of the artificial OH inside the measurement cell would be removed in the HORUS system. This concerns also the question, where the artificial OH is produced as extensively discussed by Mao et al..

The general design of HORUS and GTHOS are very similar but not identical and from Mao et al. (2012) we believe there are important differences in how the atmospheric OH was scavenged by the two instruments. The two IPIs have a distinctive geometry and they were operated with different flows that will likely cause differences in the mixing efficiency. More in detailed, our approach when designing and building IPI was to work with a concentration of scavenger that would allow negligible scavenging in the low pressure section of the instrument and with a high flow to minimize losses of radicals on walls. By using the flows of dilution air and scavenger and the residence time of the air in IPI and in the instrument, it is possible to calculate the efficiency of the OH removal both at ambient and low pressure. Assuming perfect mixing between the sampled air and the scavenger, the propane concentration used allows for the removal of a negligible fraction, less than 2%, of the OH in the low pressure regime. As the calculation assumes perfect mixing, which might not be the case in our system, we tested the amount of internal scavenging by changing the propane flow between 5 and 35 sccm while having a constant and small concentration of atmospheric OH (night time period). Figure C1 shows how we have a negligible variation in the background OH signal, well within the precision of the instrument, even when increasing the propane concentration by a factor of 7. Therefore we can exclude any impact from internal OH scavenging on the determination of the atmospheric OH. In addition, when calculating the efficiency of the OH removal on GTHOS based on Mao et al. (2012), we determine that they were aiming for the total removal of OH at ambient pressure. Without knowing the exact residence time of the air inside their instrument it is impossible to calculate the amount of OH that would be scavenged inside. By assuming a residence time of 5 ms between the mercury lamp positioned just below the nozzle and the detection cell, we calculate that the concentration of scavenger they used would allow for the removal of more than 40% of their internally produced OH, as they observed.

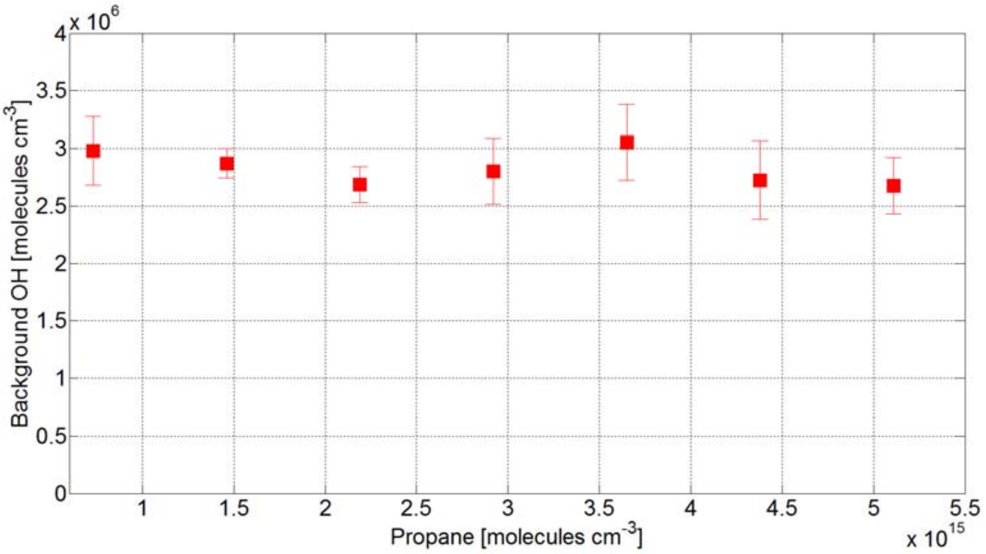


Figure C1. Background OH signal observed at a constant atmospheric OH concentration when injecting different concentration of propane.

Figure C1 and the relevant discussion were added to the paper.

5. The authors presented at the AGU meeting 2012 (cited by a different paper from the same group of authors, Hens et al., 2013) the hypothesis that Criegee radicals may cause artificial signals in the LIF instrument. This was also hypothesized by Mao et al. 2012, when they report similar effects. Mauldin et al. 2013 presented the same hypothesis for a background signal observed in the CIMS instrument applying a similar titration scheme as done here. However, no discussion or test of this hypothesis is found here. Do the authors reject this hypothesis? If so, why did they do so?

We do not reject the hypothesis that the OH formed inside the low pressure section of IPI-LIF-FAGE instrument is generated from some very reactive species formed during the ozonolysis of unsaturated compounds with Criegee intermediates being the most likely candidates. A preliminary result from tests on ozonolysis of some alkenes will be added to the paper in Section 3.3. As this paper focuses on the detection of atmospheric OH by removing possible interferences and on the description of the new tool designed in order to do so, we feel that a lengthy discussion on Criegee intermediate chemistry is out of scope. A detailed investigation of the role of Criegee intermediates in relation to the internally formed OH observed by IPI-LIF-FAGE has been done and is the main topic of a paper submitted to PCCP (reference provided).

6. The authors only discuss the interference signal for their OH detection cell. Is something similar also observed in the HO2 cell (e.g. if the cell is operated without the addition of NO)? Could it be that a similar interference is also present in the HO2 measurements (see also my comment regarding results from the HOx-Comp campaign below)? Is it possible that the magnitude increases with reaction time (distance between inlet nozzle and fluorescence detection) and/or addition of NO? What would be the consequences for the HO2 measurements?

Since Fuchs et al. (2013) we learned that the use of pure NO to convert HO_2 into OH in a LIF-FAGE instrument can cause, depending on the environment, an additional interfering HO_2 signal from the reaction between RO_2 and NO. This possible interference in our system is under investigation (Ernest et al., 2012) and the results will be published soon. During the HOPE campaign in the HO_2 cell measurements without the injection of NO were done hourly. The signal observed, after correction for the residual atmospheric OH signal detected in the HO_2 cell, was pretty constant at 1×10^6 molecules cm^{-3} (0.1 pptv) and showed no correlation with the background OH detected in the OH cell. The interfering signal is below the detection limit and smaller than the precision of our instrument (Hens et al., 2013) on the HO_2 data itself and therefore does not have any consequences on the HO_2 measurements.

The authors may have their reasons, why they limit themselves to the description of the application of the IPI-system in the three campaigns, but the paper would clearly benefit from a broader investigation of the nature of the background signal and the application of the IPI in laboratory tests. In my opinion, a discussion of the last three points are essential for this paper, because these questions arise from what has been reported in literature (see also my specific comments).

The investigation of the nature of the background signal comprises a rather large study in itself, and is being prepared as a separate publication. In the current paper we aim primarily to describe a method to eliminate the interference from FAGE OH measurements, by means of the

IPI system. A pure "laboratory" test of the IPI system, specifically, a test with a controlled OH flow, is hard to achieve in practice due to the specific nature of the IPI system that requires very high flow rates.

Specific comments:

1. p.820 l.5-7 and l.23: The authors should clearly state from the beginning (as they do later) that the background signal has only been observed in one other instrument and that it is not clear, if this applies to other LIF instruments.

Changed accordingly, please refer to the paper.

2. p.822/823: Although the instrument has been described in detail before, the authors may want to give some more details of the instrument such as laser power, distance between inlet and fluorescence detection (residence time of the air), so that the reader can clearly see the differences and similarities between this and other LIF instruments.

Additional information was added to Section 2.1.

3. p.823/824 Eq. 1-3, p.825 l.17, p.831 l.8-10: The authors assume in Eq. 1-3 that there is no removal of OH that is artificially produced in the measurement cell. As mentioned above Mao et al. experimentally found that this is not the case in their system for presumable less OH reactivity, when they add an OH scavenger. Why do the authors think that this does not apply for their instrument, although the general design of both instruments is the same as stated on p.822 l.19-22? How do they derive the removal efficiency of OH in the low pressure region given on p.831? Where do they think that the artificial OH is produced inside the system? How does this compare to what is reported by Mao et al.?

For the answer to this comment please refer to the answer given to point 4 of the general comments, i.e., figure C4 and related tests.

4. p.824 l.23: The numbers for the flow rate are not consistent with numbers in Table 1.

Changed accordingly.

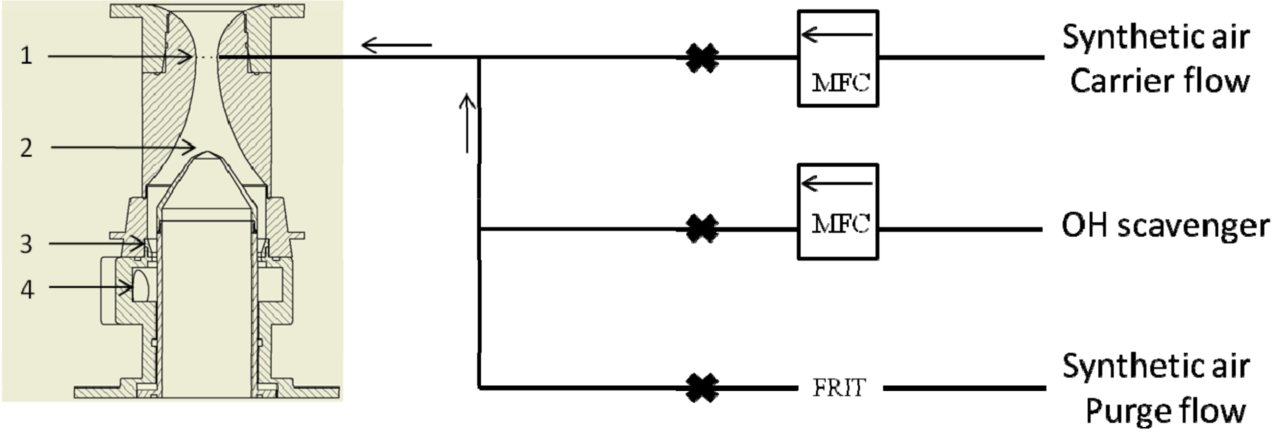
5. p.824 l.25: Could the authors add more details about the grid such as grid size, material, consequences for the sensitivity of the instrument? How do they know that the flow becomes more homogeneously for the complicated flow situation right downstream of the inlet orifice? Why is this important?

We feel that there was a misunderstanding about the position of the mesh and will rephrase the sentence in the paper and add additional information. The mesh is not located inside the low pressure segment of the instrument. It is located around the inlet inside IPI below the nozzle and before the connection to the blower pulling the air through IPI. We believe it does not change the sensitivity of the instrument as the sampling of the air happens before. As the mesh texture is very thin, and located above the blower connection, the resistance of the air through it is strong

enough to break any flow pattern that would be strongly pointing towards the direction of the pulling position of the blower, allowing a more homogenous flow and mixing.

6. p.825 l.24-29 and Fig. 3: It is not clear, what the purpose of the second synthetic air line with the frit is. I assume that this line is for the purge flow when switching to the measurement mode without the addition of the OH scavenger, but some explanation would help to avoid confusion. The position, where the excess flow leaves the IPI would be helpful to be shown in Fig. 3.

We have added some explanations about the second synthetic air line to avoid confusion and modified figure 3 adding a more specific label for the two different synthetic air lines as well as the position where the excess flow leaves IPI.



Updated figure 3 for the paper.

7. p. 827 Section 2.3: The measurement campaigns and instrumentation have been partly described in previous papers. However, concerning the specific topic of the comparison between the HORUS instrument and the CIMS instruments, more details of the experimental set up of both instruments could be given here, so that the reader is convinced that measurements of both instruments are comparable. This would include e.g. distance between instruments, heights of inlets, surrounding of instruments with respect to the question, if both instruments indeed sample the same OH concentration.

The specific points raised from the referee are now discussed and included in section 2.3.

8. p.827 l.26: Capital letters in “German Weather Service”.

Changed accordingly.

9. section 3.1: As mentioned above this would be the place where the reader would expect to read more about laboratory tests and not only about tests done in the field. All basic tests concerning the general operational parameters, proof of concept (such as titration efficiency of ambient OH), and OH loss in the IPI are given, but I would expect that this could be done with higher precision and accuracy in laboratory experiments instead of doing this under field conditions. I could imagine that especially the question of OH loss in the IPI would be much better determined in laboratory tests with an OH source. Tests in the field would certainly add

valuable information like loss of species which cause the interference and can show consistency with laboratory tests.

Please refer to the answer given to the first general comment.

10. p.828 l.15-18: Although the general procedure to estimate loss of species causing an OH signal is clear from the description given here, more information is required, in order to understand in detail what was done. How often was measured without the IPI? What was the reproducibility of the loss measurement? Did the authors observe any trend in the loss (during the day or over the campaign)? How long was measured without the IPI during one test? How was the loss exactly calculated? Did the authors interpolate signals between measurements before and after the IPI was removed? Are the large error bars caused by the variability of measurements during the time periods, over which measurements were averaged? Please define the exact meaning of the error bars. It would be helpful to see a figure showing single measurements and averages from an example, when the instrument sampled with and without the IPI showing, how the loss was determined.

Measurements without IPI were done on average every 4 days during HOPE 2012 campaign. The reproducibility of the loss measurements of atmospheric OH, at constant conditions, was similar to what observed in figure 4a for the total OH signal and it was taken into account for the accuracy of the HORUS instrument after the addition of IPI. No clear trend was observable during the campaign or over one day. IPI can be removed easily in less than 2 minutes and on average, we measured without IPI during one test for 30 minutes. Referring to equation E5 from the paper, for one IPI on and off test the numerator, the atmospheric OH signal affected by losses, is the result of the interpolation of the measured atmospheric OH signal immediately before and after the removal of IPI. In the denominator, the total OH measured when IPI was not on the top of the inlet is the average of the signal measured when IPI was not on the top of the inlet. The background OH measured with IPI on the top of the inlet is obtained from the interpolation of the measured background OH signal immediately before and after the removal of and it is divided by the loss of the background OH measured during night time tests. The error bars for every single point represent the variability of the data during a single test: the point is the average of a period of measurement without IPI on the top of the inlet and the error bars reflects the scatter of the data. As the measurement representing the losses of the total OH signal were done during daytime when the ratio atmospheric OH versus background OH was the highest, the large error bars reflect the large ambient variability of OH. Figure C2 is an example of one IPI on and off tests that includes the raw data and the average used to obtain the losses value for atmospheric OH. This more detailed description on how the losses on IPI were measured has been added to the paper.

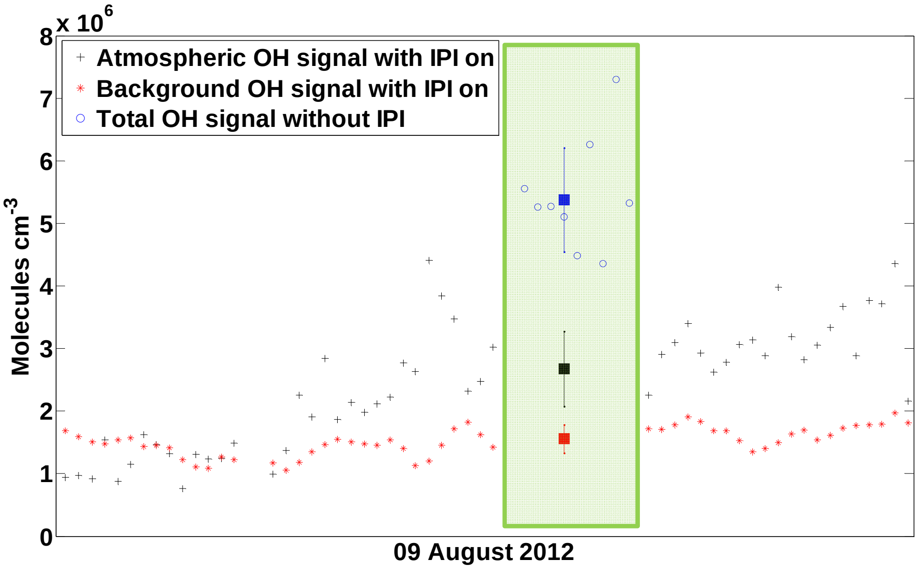


Figure C2. Example of an IPI on and off test during the HOPE 2012 campaign. The shaded area represent the test period when IPI was removed from the top of the inlet of the LIF-FAGE instrument. The blue circles represent the total OH signal measured by the instrument without IPI on the top of the inlet. The black crosses and the red stars represent, respectively, the atmospheric and the background OH signals measured with IPI on the top of the inlet. The square markers show the average of the total OH signal measured without IPI on the top of the inlet (blue) and the average of the interpolated signal before and after the test period for the atmospheric (red) and background (black) OH. The error bars represent the 1σ standard deviation of the data. By using equation E5 from the paper, this test results in an OH loss within IPI of 32%.

11. p.829: The authors assume that the 7% loss of equivalent OH signal observed during the night can be taken as general loss of the interfering species in their inlet system, so that they can use this number also to correct daytime observations. Is this really justified? This loss is only 5 times smaller than that for OH. Therefore, I would assume that the interfering species is also a reactive, short-lived species. The authors do not present any hint about the nature of the interfering species, so that I wonder, if the same mechanism necessarily applies during day and night. One hint that this could not be the case is that during the HOxCOMP campaign large differences between the HORUS instrument and the other instruments were only observed for nighttime conditions. In contrast, the background signal during the field campaigns reported here was much larger during daytime than during nighttime.

Hints to Criegee intermediates as the possible cause for the background OH in our system will be given. Therefore, as the ozonolysis of VOCs does not stop during daytime and these species are actually short lived, as assumed by the referee, when comparing the losses observed for the background OH with that calculated for the atmospheric OH we believe our assumption is valid.
If Criegee intermediates cause the background OH signal, we expect a variation with the available atmospheric mixture of VOCs species and ozone. The background OH signal is much larger during the day compared to the night for the HUMPPA campaign. During the HOPE campaign, conversely, there is not such a large difference between the day and the night (Figure

11 b of the paper) and there were several days in which the nighttime background OH was higher than the daytime background OH signal (Figure C3). The signal thus behaves differently in different environments. We believe that during the HOxCOMP campaign during nighttime we were affected from background OH and during day time its concentration was small enough as to not produce a significant OH interference (i.e. within the accuracy of the instrument).

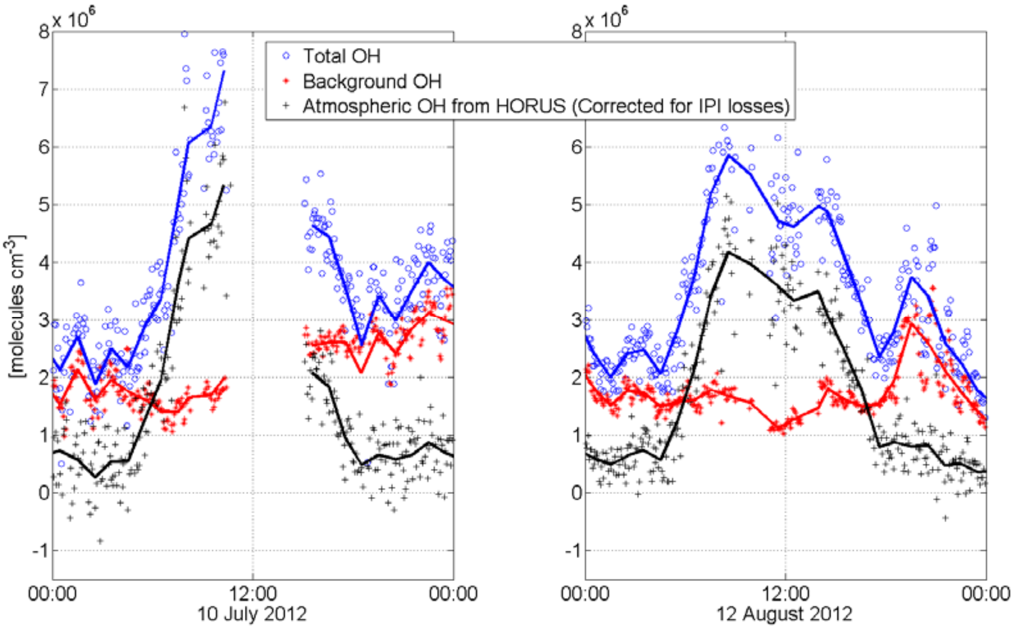


Figure C3. **OH signals measured by HORUS during the HOPE 2012 campaign. The blue circles represent the total OH signal measured in the absence of an OH scavenger. The red stars represent the background OH measured during the injection of an OH scavenger. The black crosses represent the atmospheric OH obtained by difference between total OH and background OH. The solid lines are 30 minute averages. Time is in UTC.**

12. p.831 l.14 and p.832 l.20: Figure 7 is mentioned before Figure 6.

Fixed.

13. p.832 l.23: Looking at the error bars, the authors may want to add the statement that they did not observe any significant difference. Once again, it is not clear, why the authors do not present a systematic investigation of the effect of OH production from propene ozonolysis in a laboratory study. I assume that they could determine the OH signal from propene ozonolysis for typical situations they experienced in the field by overflowing their inlet with synthetic air and ozone and doing the propene titration.

The statement has been added. As stated in the paper, propene is no longer used as scavenger in our system as for is formation of Criegee intermediates and OH after ozonolysis. As for the possible implications of its usage during the HUMPPA and DOMINO HOx campaign we believe that the test performed in the field (Figure 7) is valuable enough to indicate that we did not create a significant additional OH signal.

14. p.833 section 3.2: The good correlation between both instruments is quit convincing. However, the authors should give some more information about the CIMS measurements including a short statement about the major contributions to precision and accuracy of measurements. What are there differences between the two CIMS instruments used here? In Berresheim et al. 2000 and Mauldin et al. 2013 a similar titration system with propane is described as used for the LIF. Was this also applied here? If so, are the background measurements for the CIMS instrument similar to what is observed for the LIF? What does this mean for the nature of the background signal in the LIF measurements?

A statement about the major contributions to precision and accuracy of the CIMS measurements was added. Both the two CIMS are based on the instrument described by Berresheim et al. (2000). A propane titration was done in both instruments for the entire campaign as it is needed in order to properly measure the OH concentration in ambient air. One big difference between the two CIMS is that the one in use at the DWD has a very long inlet and a longer residence time of the air where SO_2 and propane are injected. We did compare the background OH measured from the different instruments but we did not observe many similarities. It should be underlined that CIMS and LIF-FAGE instruments are very different therefore it is unlikely that an interfering species will have a similar impact on both measurements. Moreover, as proposed by Berresheim et al. (2014), some additional production of what the CIMS measures as background OH might happen inside the inlet at ambient pressure making the intercomparison with the background OH measured by the LIF instrument difficult.

15. p.833 l.21-23 and Fig. 8: A similar figure is presented in a paper by the same group of authors (Fig. 6 in Hens et al. 2013). However, the error bars (LIF and CIMS) are different in both figures and therefore also the fit results are different. Please define the error bars in the correlation plots (e.g. 1sigma precision of data). Please add why the presentations are different or make them consistent, in order to avoid confusion.

The figure was changed and now it is consistent with the one shown in Hens et al. (2013).

16. p.834 l.10-18 and Fig. 11: The authors state that the offset is caused by observations of nighttime OH by LIF that is not observed by CIMS. This is surprising regarding the fact that in general the background signal was smaller during the HOPE campaign compared to the other campaigns. Is there any suspicion what the cause of this OH signal is in the LIF instrument? Is it possible that the CIMS instrument did not observe ambient nighttime OH or the instruments did not observe the same air mass? Does the regression indeed significantly changes, if only daytime measurements are included in the fit? I have the impression that the OH values measured by LIF are smaller than those measured by CIMS for higher OH values, so that the data set is deviating from a linear relationship. This may also cause the positive intercept in the regression analysis.

With our current knowledge, we do not have any reason to suspect that the nighttime OH measured by the LIF instrument is not atmospheric OH. The nighttime OH measured by HORUS is not a constant value: during some nights the two instruments agree, both showing an OH signal scattering around zero. Propane was used as scavenger both when HORUS measured zero atmospheric OH and when not and as far as we know, the instrument was run in the same conditions. No correlation between the atmospheric OH observed during night and the

background OH was observed. In addition, preliminary budget calculations suggest that as the OH reactivity measured was very small during nighttime, the production of OH via recycling of HO_2 through NO and from ozonolysis of VOCs could led to a concentration similar to what is detected by HORUS instrument. When considering only the day time data (between 7:30 and 18:30) still a slope of 0.9 ± 0.01 and a value of R^2 of 0.7 is observable with an offset of (3 ± 0.3) x 10^5 molecules cm^{-3} confirming the linear relationship between the two data sets.

17. p.835 l.1-10: It is not clear, if monoterpene concentrations were similar or different in the two campaigns. I would suggest to rephrase the sentences to avoid confusion. Is there any hint for differences in OVOC concentrations during both campaigns? Are there more differences in the conditions during the campaigns which may be related to the different contributions of the background signals to the total OH signal? Are there laboratory tests of potential interferences (as mentioned above)? Does any other measured quantity correlate with the magnitude of the background signal (like ozone, NOx, VOC species, laser power of the LIF instrument...)?

The sentence was rephrased. OVOC concentrations were not measured during the DOMINO campaign. During the HUMPPA campaign the averaged sum of the measured OVOC that included acetaldehyde, methacrolein, MVK, methanol, acetone, MEK and ethanol was about 7 ppbv and the average sum for the same OVOC measured during HOPE was about 9 ppbv. During both campaigns methanol and acetone account for most of the OVOC measured but during the HUMPPA campaign the average concentration of methacrolein and MEK were twice as high as during HOPE. In addition, the averaged value shown for the HUMPPA data is taken excluding the period during which "polluted air" from biomass burning was measured (Nölscher et al., 2012). It is therefore likely that the high concentration of methanol and acetone observed were due to the high oxidation capacity of the environment as the site was far from any anthropogenic activity while the similar concentrations of acetone and methanol observed during HOPE are more likely to be also caused by anthropogenic activity as the oxidation capacity of that environment was quite lower. Laser power does not correlate with the background OH. Other species or parameters that show some correlation with the background OH are ozone multiply by VOCs, acids and temperature and will be analised in detailed in a following publication.

18. p.837: The authors discuss the results from the HOxCOMP campaign regarding the now identified OH background signal. However, they do not try to use the observations during HOxCOMP, in order to discuss potential reasons for the unrealistic large background signal that was also observed during HOxCOMP for nighttime conditions. Especially the relatively simple experiments performed in the SAPHIR chamber (Schlosser et al. 2009) may give hints of the nature of the background signal. Also the fact that OH values measured for dark conditions were much larger for the HORUS instrument compared to other LIF instruments may contain valuable information as mentioned above. Larger signals of the HORUS instrument compared to other LIF instruments were also observed for HO2 measurements during dark conditions of certain SAPHIR experiments (Fuchs et al., 2010). Is it possible that also HO2 measurements are affected by the interference reported here exclusively for the detection of OH (see above)?

As answered previously (see answer to comment 6 of the general comments), from our measurement experience with IPI, we can exclude that the background OH, though having a high impact on the atmospheric OH signal, will have a detectable impact on HO_2.

19. p.838: The authors also discuss results from the GABRIEL campaign. They clearly state that they cannot exclude that part of the large OH concentration that could not be explained by traditional chemical models is caused by a background signal similar to that found for the campaigns reported here. One argument that an interference may have not been present during the GABRIEL campaign is that newly proposed isoprene degradation models predict more OH than traditional models. However, the authors do not mention that the theoretical study by da Silva et al. 2010 does not predict large additional OH. The experimental study by Crounse et al. 2011 and one recent study in a simulation chamber (Fuchs et al. 2013) do not show a production of additional OH precursors (HPALD) and additional OH from isoprene oxidation, respectively, which would be large enough to explain their observations during GABRIEL. One further argument is that a short-lived species may cause the interference which is only present very close to the emission source, so that airborne measurements are not affected. Is there any idea or speculation what this could be?

References as suggested by the referee have been added. About the cause of the interference, please refer to the answer given for comment 5 of the general comments.

20. Fig. 3, 7, 9, and 10: Please enlarge the size of symbols in the legends.

Fixed.

Anonymous Referee #2

The paper presents technical details and the characterisation of a newly constructed device allowing injecting a scavenger for atmospheric OH radicals prior to their measurement by FAGE technique, named IPI (Inlet Pre-Injector). The goal of such device is to detect a potential interference in the measurement of OH radicals by FAGE technique due to the generation of OH-radicals within the FAGE cell, and thus leading to an overestimation of the ambient concentration. The potential importance of such background OH has recently been demonstrated by Mao et al. using a similar device, and therefore the paper is certainly important for the community and within the scope of the present journal. Reviewer 1 has already given a very detailed report, and I agree in many points. In particular,

We thank the reviewer for taking the time to review the paper.

1. I would like to see a more detailed comparison with the technical details of the IPI used by Mao et al.: what are the major differences (improvements, I guess?) compared to Mao's device?

For the answer to this comment please refer to the answer given to comment 4 of the general comments from Referee #1.

2. Mao et al state that they do not observe any loss of radicals within the IPI while you observe around 30%: any comments?

As underlined before in the comments, the design and operation of "IPI" for the two LIF-FAGE instruments is very different and different flows patterns and mixing are likely to happen. It is therefore not possible for us to judge their results.

3. The description of how the OH loss within the IPI has been obtained (page 829) is too short and more details such as how often and how long the OH signal has been measured with and without IPI should be given. How simple is it to remove and put back the IPI?

For the answer to this comment please refer to the answer given to comment 10 of the specific comments of Referee #1.

4. As reviewer 1, I'm not so convinced that the background loss obtained at night can be used to correct the daytime data: that would assume that the mechanism of background OH signal is the same during night and day. Although, I'm aware that it is not possible to determine the background loss as long as its origin has not been identified.

For the answer to this comment please refer to the answer given to comment 11 of the specific comments of Referee #1.

5. Which brings me to the last point, also raised already by reviewer 1: There have been several presentations by the same group (also during a presentation at ACM 2012) that supposed that Criegee intermediates had been identified as being at least partially responsible for generation of a background OH signal within the FAGE. In the present paper there is no word about this hypothesis. Could you comment on that?

For the answer to this comment please refer to the answer given to comment 5 of the general comments of Referee #1. As mentioned before, the focus of this paper is the detection of atmospheric OH by removing possible interferences and on the description of the new tool designed in order to do so. A detailed investigation of the possible species causing the internally OH observed by IPI-LIF-FAGE has been done and is the main topic of a paper submitted PCCP which we will refer to.

In general, this paper being the first presentation of the IPI in a journal more focused on the measurement technique rather than on the chemistry behind, I also would have liked to see a more detailed characterization of the device in laboratory experiments prior to the description of the IPI in three field campaigns.

For the answer to this comment please refer to the answer given to comment 1 of the general comments of Referee #1.

Anonymous Referee #3

This manuscript describes a newly developed inlet pre-injector (IPI) coupled with an OH detecting instrument HORUS based on the laser-induced fluorescence technique designed with multiple optical path alignment. The IPI enables chemical modulation to correct for the instrumental background OH, which is indistinguishable from ambient OH with wavelength modulation. The loss of OH on the internal surface of the injector is unavoidable and the loss rate is taken into account when ambient OH concentrations are calculated. The performance of the IPI was demonstrated during the three field campaigns with different chemical characteristics, including cases where the interference from the background OH was dominant and other cases where the signal from the ambient OH was larger. The manuscript is well organized, within the scope of the AMT journal, and contains important information. However at certain points this manuscript needs clarification, especially on the design of the IPI system and on the determination of the factor F, which is used in Equation (3) but left unmentioned in section 3.1. I recommend publication after adequate revision.

We thank the reviewer for the time taken to read and comment on our manuscript.

Specific comments:

1. page 820, line 22. IPI instead of inlet-pre-injector.

Changed accordingly.

2. page 821, line 6. Replace "more than 90%" with "a large fraction."

Changed accordingly.

3. page 828. Have the authors performed fluid dynamics simulation on designing the IPI system to minimize the wall loss rate? Was the internal surface coated with halocarbon wax?

Fluid dynamic simulations were not performed when designing the IPI system but the elliptical shape was chosen based on Eisele et al. (1997) in order to have a small velocity of the air through the larger section of IPI and a high velocity where the inner diameter is smaller in order to reduce contact with walls. Here is where we inject the OH scavenger and we sample with our inlet in the middle of the air stream. The internal surface of IPI was not coated with halocarbon wax. Tests performed after coating the internal surface of the instrument inlet did show a decrease in the loss of OH on walls but the amount of OH lost changed with time and the change was not linear and therefore very difficult to monitor. We therefore preferred to have a, maybe higher, but constant OH loss.

4. page 828-830. Can the authors determine the loss rate of OH on the IPI surface using more stable and clean external source of OH (e.g., photolysis of water vapor with clean nitrogen carrier gas), in addition to the tests with ambient air as described here? I understand that the large volume flow will make the test difficult. But I also believe that the test, likely with lower background signal, should provide clearer determination of the loss rate currently estimated to be 34+/- 15%.

For the answer to this comment please refer to the answer given to comment 1 of the general comments of Referee #1.

5. page 830, line 3. The detection limit of the instrument should also be discussed in addition to the increased uncertainty.

A discussion about the detection limit of the instrument after the addition of IPI has been added.

6. page 831 line 10. Do the authors mean 0.1%?

We actually meant 2% and corrected the value in the paper.

7. page 831, line 10. So far the factor F in Equation (3) should have been determined by considering (1) loss of OH on the IPI internal surface and (2) scavenging efficiency. The authors should conclude which F value was used for correction.

A more detailed description of which F values was used has been added at the end of Section 3.1.

8. page 832. Can the Criegee intermediate, simultaneously produced via the ozonolysis of propene, influence the background OH level?

We had the same concern Referee #3 is raising, therefore, as explained in the paper, we stopped the use of alkenes as OH scavenger and decided to use an alkane. The results of the tests shown in figure 6 from the paper (now renamed 7 in the new version of the paper) copied below showed that for the conditions we were running IPI such as with an extremely short residence time between the injection of the scavenger and the inlet nozzle, there was not enough time to allow elevated production of Criegee intermediates to create a big additional artificial interference on the background OH signal.

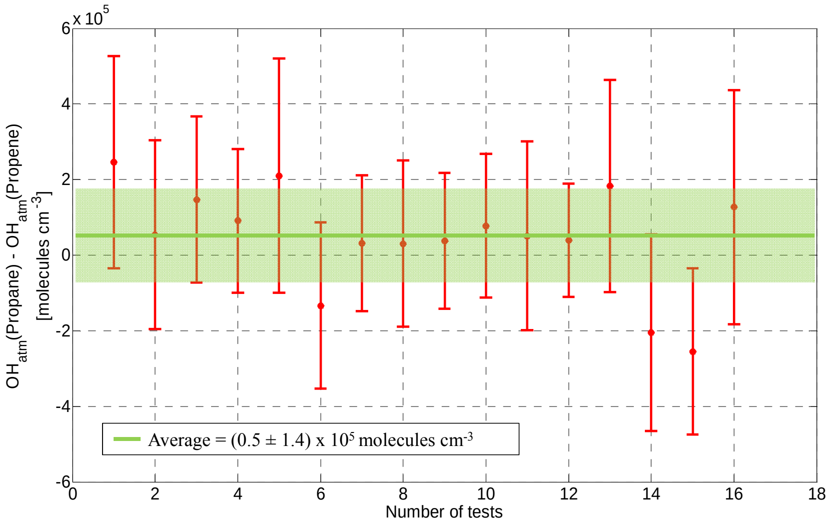


Figure 6. Difference between the atmospheric OH concentration determined with the use of propane as scavenger and the atmopspheric OH concentration determined with the use of propene. The points are one hour averages. The green line is the average value of 0.5 x 10^5 molecules cm^{-3} and the shaded area is the 1σ range.

9. pages 831 and 832. Text mentions Figure 7 earlier than Figure 6.

Fixed.

10. page 833, line 20. I did not understand "ageing detector".

During the HUMPPA campaign both MCP detectors changed in sensitivity decreasing over time.

11. page 834. What is the systematic uncertainty in the OH concentrations determined with CIMS?

The value for the systematic uncertainty in the OH concentration for the CIMS was added.

12. page 838, line 7. The authors should rather mention that Crounse et al. (2011) found that the proposed recycling was much slower than originally proposed.

The paragraph was rephrased accordingly also to account for comment 19 from the specific comments of Referee #1.

13. page 838, lines 19-23. It is not logically valid that agreement between observation and model indicates no interference.

The authors agree with the Referee and the sentence has been changed.

14. Have the authors tested if the background OH signal is always linearly dependent on the laser power?

Yes and we did not find a quadratic dependency on the laser power (please refers to Section 3.3 of the paper). The instrument was designed in order to maximise the speed of the air trough the detection cell to avoid the possibility of double pulsing. For additional information please refer to the answer to comment 3 of the general comments of Referee #1.

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