"Detection of formaldehyde emissions from an industrial zone in the Yangtze-River-Delta region of China using a proton transfer reaction ion-drift chemical ionization mass spectrometer" (**AMT-2016-194**) by Yan Ma et al..

Reply to the Comments by the Reviewer #1:

We are grateful to the reviewer for the helpful suggestions that have improved the manuscript substantially. Below we have included the review comments in italic followed by our responses. The manuscript has been revised accordingly.

> *1) The manuscript is well structured, easily comprehensible and well written. The characterization of the performance of the instrument needs to be more detailed (humidity dependence of background and LOD, uncertainty of quantification (precision and accuracy) bases on a range of humidities expected in the ambient sample) and fairly compared to other instruments and to characterizations of similar methods in the literature (e.g. as published in AMT). The statements drawn from the method comparison (PTR vs DNPH) need to be statistically substantiated.*
>
> *Even though the performance of this PTR instrument with regards to HCHO might not exceed those presented in the literature the authors are encouraged to present their instrument characterization in a quantitatively comparable way (and actually make the comparison). Based on a more thorough characterization they should be able to demonstrate that this instrument is suitable for the quantitative characterization of pollution episodes – the ambient trace gas data they present are in itself a valuable contribution.*

Response: As suggested by the reviewer we have revised the manuscript thoroughly. In addition, we have conducted additional calibrations at various humidity conditions to fully characterize the instrument. The detailed revisions can be found in the following responses to specific comments.

> *2) In the light of this manuscript focusing on a measurement technique for HCHO the introduction of and performance comparison with alternative methods (or identical methods but different instruments) is too superficial. E.g. Vlasenko et al. (2010) and Warneke et al. (2011) detail the performance of their PTR-MS approach to which this instrument could be compared. New LIF instruments (e.g. Cazorla et al. 2015) outperform instruments based on Hantzsch process, CIMS, or DNPH cartridges regarding the detection limit on short very short sampling periods (e.g. 10Hz). The performance of the method presented in this manuscript should be quantitatively compared.*

Response: As suggested by the reviewer, the following table of the performances of various HCHO detection techniques including this work has been added in L316.

Table 1. Performance inter-comparison of various HCHO measurement techniques.

Measurement Technique	Limit of Detection	Time Resolution	Precision	Reference
DNPH-HPLC	13.4 pptv	1 hr	2.9%	Fung and Grosjean (1981)
Long Path Length Fourier Transform Infrared Spectroscopy	6 ppbv	5 min	±4 ppbv	Tuazon et al. (1981)
Hantzsch-reaction/Fluorometric Flow Injection Analysis	0.1 μM[*]	80 s	5%	Dong and Dasgupta (1987)
Liquid-trapping/fluorescence detection	0.2 ppbv	80 s	5%	Kelly and Fortune (1994)
Laser-induced Fluorescence	36 pptv	1 s	20%	Cazorla et al. (2015)
Differential Optical Absorption Spectroscopy	4.5 ppbv	12~15 min	30%	Lawson et al. (1990)
Tunable Diode Laser Absorption Spectroscopy	0.25 ppbv	3 min	20%	Harris et al. (1989)
Quantum Cascade Laser Spectrometer	0.3 ppbv	1 min	20%	Herndon et al. (2007)
Ionicon PTR-MS	1~2 ppbv	5 s	20%	Karl et al. (2003)
	0.2~0.5 ppbv	5 s	25%	Inomata et al. (2008)
	78~95 pptv	5 s	< 24%[#]	Jobson and McCoskey (2010)
	0.1~0.3 ppbv	1 s	30%	Warneke et al. (2011)
PTR-ID-CIMS	0.9~2.4 ppbv	~10 s	<18%	This work[**]

[*] Aqueous Sample; [#] Based on the uncertainty of the instrument sensitivity; [**] averaged over 3 duty cycles.

> *3) L132: "The site was fairly away from local highways. . .". According to open street map highway G40 passes only a few km to the west of NUIST campus, and G205 is even closer (depending on the exact location of the measurement), which does not seem to be reflected by the text.*

Response: As suggested by the reviewer, we have provided more precise description of the highways around the observation site. The sentence has been revised as:

"The site was in the middle of two highways, i.e. G40 and G205, which were ~1.3 km to the west and ~1.5 km to the east of the site, respectively. From the aromatics measurement results, we found no substantial impacts on measured HCHO from traffic-related emissions, which will be detailed in the discussion section."

> *4) "L173+: Backgrounds. The authors state that m33 signals are the sum of protonated methanol and* $^{16}O^{17}O^{+}$ *as well as* O_2H^{+}*. They conclude that zeroing by means of redirecting the sample gas through a catalytical converter produces differential signals (m33ambient – m33cc) that are directly related to the density of protonated methanol and thus to the methanol sample concentration. This is only true if the other contributing m33 ion counts do not get altered by the zeroing procedure (e.g. by slightly changing humidity). The authors either need to demonstrate that the background contributors do not change due to the zeroing procedure or at least that humidity (thus m39/m21) in the*

> *drift tube does not change. This should be in the data. More importantly – since this manuscript focuses on HCHO – a similar demonstration needs to be presented for m31. A very strong humidity dependence of the m31 background signal is expected (see Fig 3c in Warneke et al., 2011).”*

Response: We agree with the reviewer that HCHO measurements by PTR-MS show strong humidity dependence and it is important to maintain the same humidity as the ambient humidity during background checks. To clearly demonstrate that the humidity in the drift tube did not change significantly during background checks, we have included a plot of the ratios of m19 (m21×500) to m37 (m39×250) on April 18 (an HCHO plume event) in Fig. 8b. It clearly shows that humidity did not change significantly during background checks. Also included in Fig. 8e is the time series of m31 raw data. The background signals of m31 appear to be independent of ambient RH. Since methanol has little humidity dependence, it is reasonable to assume m33 was not affected by the background checks. We also want to point out that there is a significant difference between this work and that of Warneke et al. (2011). Warneke et al. (2011) conducted an airborne measurement of HCHO. Their PTR-MS would experience rapid humidity and pressure changes when the flight altitude changed from ground level to the free troposphere as shown in their Fig 3c. The diurnal variation of m37/m19 in this work was about 0.2 to 0.24 and we did not observe significant background change, which is actually consistent with the results of Fig. 3c in Warneke et al. (2011).

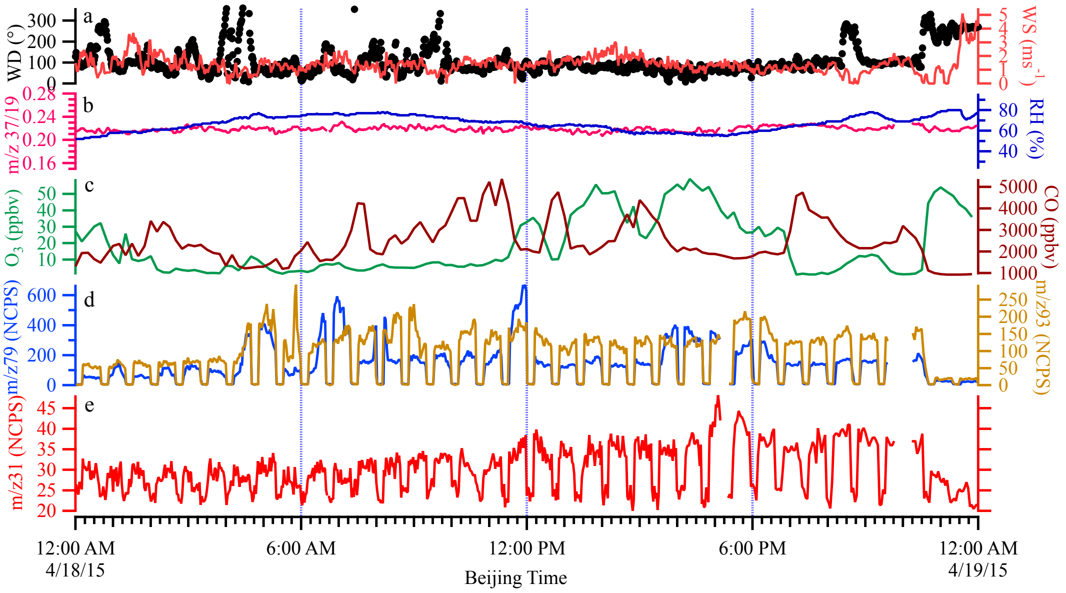


Figure 8. Time series of wind direction (WD) and wind speed (WS) (Panel a), m/z 37/19 ratio and RH (Panel b), O_3 and CO (Panel c), m/z 79 and m/z 93 (Panel d), and m/z 31 signal (Panel e) on April 18, 2015. The dips in panel d and e are the periods when background checks were made.

> *5) L197+: The QMS was set to sample for 2s at each of the 40+ mass channels, but then there are 2s in between the sampling of consecutive mass channels – does the quadrupole filter / HF generator really take 2s to tune from one channel to the next? Please, comment on this.*

Response: We apologize for the confusion. The actual voltages applied to the QMS can be set within a few milliseconds for any mass less than 1000 amu. However, we found that the slew time of the high-voltage power supply can cause unstable signals, leading to higher measurement uncertainty. Therefore, the QMS was programmed (Paw macro language, Extrel) to stop collection for 2 sec after reaching a new setting to account for the slew time and then continue to count signals of that mass (e.g. 30.9 - 31.1 amu in the case of HCHO) for 2 sec. The recorded data were averaged to counts per second. To avoid the confusion, we have added the following description in the manuscript (L212):

"The QMS was set to measure each mass for 2 s and then pause for 2 s after switching to a new setting to account for the slew time of the QMS power supply."

> *6) L251+: The use of N2 for dry calibrations will change ion mobilities and KE compared to air and thus will not produce comparable results.*

Response: We agree with the reviewer that the reduced ion mobility of hydronium ions will be a little different in pure nitrogen (N_2) than in pure air. However, since 78% of pure air is N_2 and the molecular mass of nitrogen (28 amu) is very close to oxygen (32 amu), we estimated this difference should be less than a few percent under the conditions of this work (Zheng et al., 2008). To fully address this potential issue, we have conducted additional calibrations using both pure N_2 and pure air as carrier gases. We found that sensitivity of the instrument varied about 4%. Since the data reported in this work were based on calibrations conducted using scrubbed ambient air, the data quality should not be affected. We have revised the uncertainty estimate with the new pure air based calibrations at various relative humidities. The following sentences are inserted into the manuscript (L304):

"To fully characterize the humidity effects on its performance, the PTR-ID-CIMS was also calibrated under various relative humidity conditions (1.0%, 17.5%, 29.0%, 47.0%, 62.5% and 81.5%) using pure air as the carrier gas. The relative humidity of the VOC standards was achieved by passing the carrier gas through a water bubbler similar to the one used in Zheng et al. (2015). The results are shown in Fig. 5. Based on three times the standard deviation of the background signals, the PTR-ID-CIMS detection limit (DL) of HCHO varied from 0.9 ppbv for dry condition to 2.4 ppbv for 81.5% RH at room temperature.

> *7) L270+: Calculating empirical standard deviations on sample sizes of N=3 may cause considerable uncertainties. What was the (apparent) calibration-to-calibration LOD variability?*

Response: We have conducted 3~4 calibrations at each RH setting (1.0%, 17.5%, 29.0%, 47.0%, 62.5% and 81.5%) and we found that the calibration-to-calibration LOD variability to be <16% with the minimum variation of 2.2% at 1.0% RH. The following sentence has been inserted into the manuscripts (L310):

"The variations of the instrument sensitivity (NCPS per ppbv) ranged from a few percent to 17%. We also determined the calibration-to-calibration DL variation at different RH levels to be less than 16% for RH < 81.5%."

8) L272+: Calibrations under conditions that are otherwise identical to those encountered during ambient measurements are part of the quantification process. Apart from the above mentioned problems with changing buffer gas compositions (N2 vs air) sensitivities of HCHO under completely dry conditions are much higher than under typical ambient conditions (see Fig 5 in Vlasenko et al. 2010 or Fig 3b in Warneke et al. 2011). A precision determined under dry conditions is meaningless for the performance of the instrument under ambient sampling conditions. Please, evaluate the precision based on the calibrations under varying humidity conditions and compare them with performance characterizations in the literature.

Response: We agree with the reviewer and accordingly we have conducted additional laboratory calibrations at various RH levels using pure air as the carrier gas. Figure 5 shows the calibration results with RH ranging from 1.0% to 81.5%. The variations of the instrument sensitivity (NCPS per ppbv) ranged from a few percent to 17%. We have inserted the following sentences into the manuscript (L304):

"To fully characterize the humidity effects on its performance, the PTR-ID-CIMS was also calibrated under various relative humidity conditions (1.0%, 17.5%, 29.0%, 47.0%, 62.5% and 81.5%) using pure air as the carrier gas. The relative humidity of the VOC standards was achieved by passing the carrier gas through a water bubbler similar to the one used in Zheng et al. (2015). The results are shown in Fig. 5. Based on three times the standard deviation of the background signals, the PTR-ID-CIMS detection limit (DL) of HCHO varied from 0.9 ppbv for dry condition to 2.4 ppbv for 81.5% RH at room temperature. The variations of the instrument sensitivity (NCPS per ppbv) ranged from a few percent to 17%. We also determined the calibration-to-calibration DL variation at different RH levels to be less than 16% for RH < 81.5%. Therefore, the instrument precision for HCHO measurements was estimated to be < 18%, including the 1% uncertainty associated with the HCHO standard concentrations."

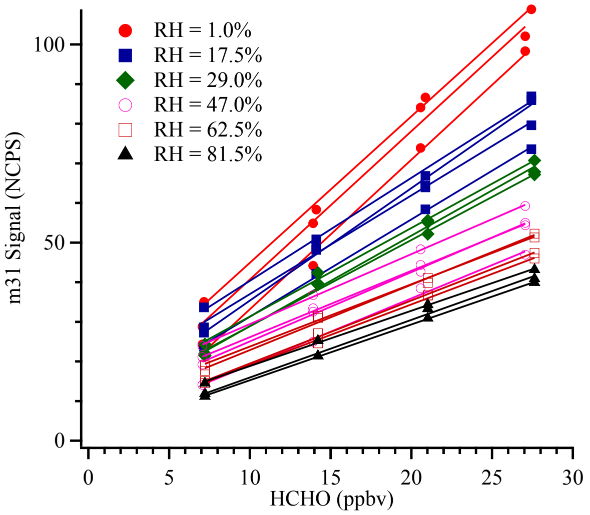


Figure 5. HCHO calibrations conducted at RH = 1% - 81.5%.

9）L287+: Please, substantiate (data or literature) that varying O3 concentrations (0-140ppbv; Fig 5) in a complex mix of organic trace gases and aerosol flowing through a *potassium iodide cartridge does not produce considerable carbonyl interferences. Results and Discussion:*

Response: We agree with the reviewer that high ozone level can be a potential interference for DNPH-HCHO measurements. The commercial KI cartridges were manufactured according to the US EPA TO-11A recommendations and have been demonstrated to be able to remove 125 ~ 200 ppbv O_3 for up to 100,000 ppbv-hours. Therefore, we believe 140 ppbv O_3 should not generate considerable carbonyls to cause interferences. The following sentences have been added to the end of this paragraph (L353):

"Note that a few high O_3 episodes were encountered during the measurement period with a maximum concentration of ~140 ppbv, which can potentially interfere with DNPH samples. However, as demonstrated by the US EPA, the KI cartridges can efficiently remove 125 ~ 200 ppbv O_3 from air samples for up to 100,000 ppbv-hours with a requirement of minimum moisture level of 10% RH (https://www3.epa.gov/ttnamti1/files/ambient/airtox/to-11ar.pdf, accessed October 2016). Given the relative high RH conditions during the campaign period, O_3 should not interfere with our DNPH-HCHO measurements."

10）L313+: This reviewer cannot discern wind conditions form the quiver plot in Fig 5. Please, replace the representation in a more suitable way (e.g. wind speed and direction time series).

Response: We have replaced the quiver plot with time series of wind direction and wind speed as suggested by the reviewer.

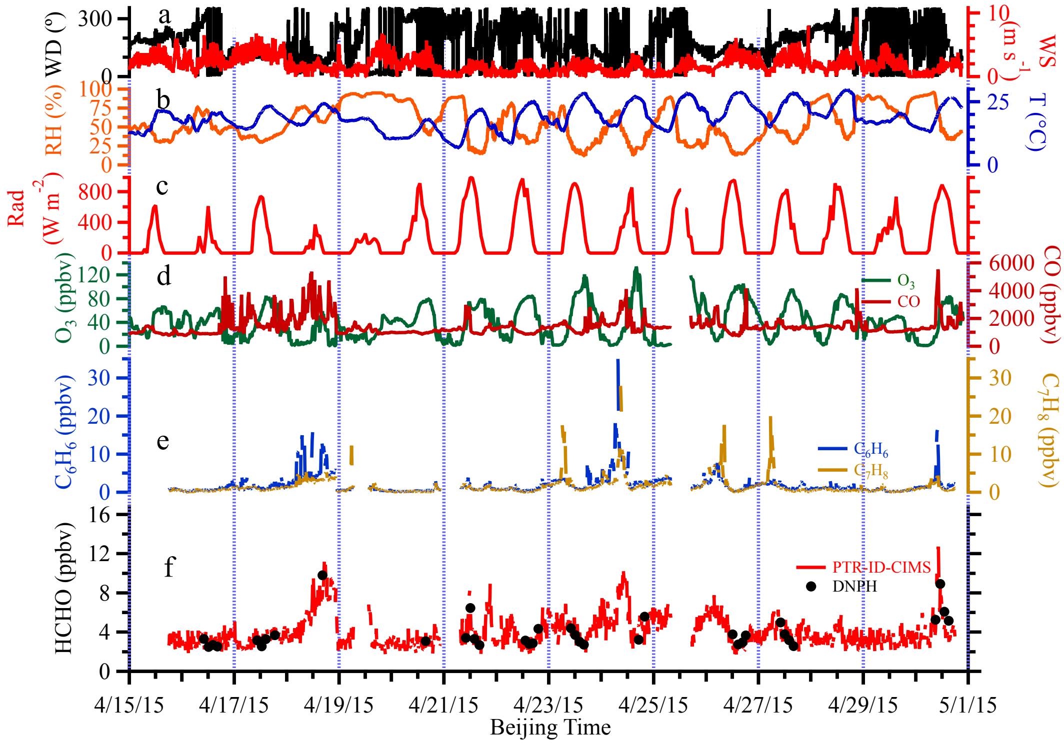


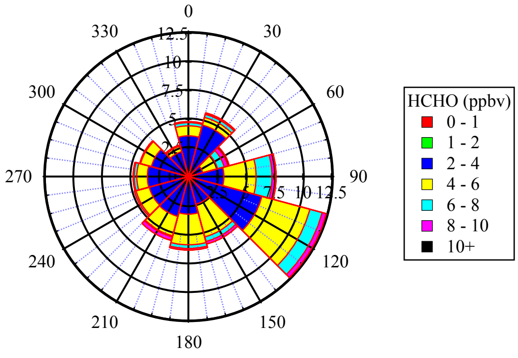
Figure 6. Time series of meteorological parameters (wind, RH, T, and solar radiation) (Panel a, b and c), trace gases (O_3 and CO) (Panel d), benzene (C_6H_6) and toluene (C_7H_8) (Panel e),

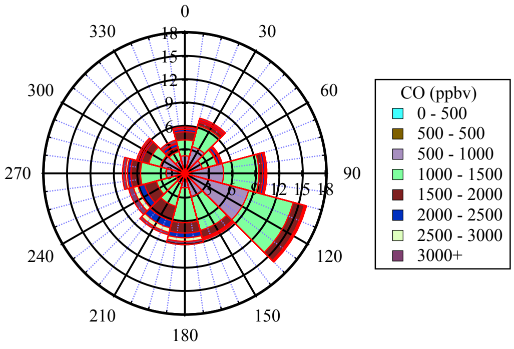
and HCHO (Panel f). The black dots in Panel e are DNPH cartridge measurements of HCHO.

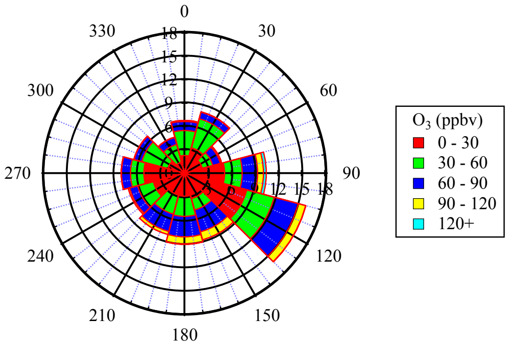
> *11）L328+: "In addition, all heavy pollution episodes . . . were associated with easterly wind or nearly calm conditions". Please, find a way of presenting the data so that the readers can convince themselves of this finding. I suggest a rose-plot of HCHO mixing ration distributions separated into e.g. 30deg wind sectors. Calm conditions (e.g. u<0.1 m/s) could be treated separately if needed. The same for CO, aromatics and O3.*

Response: As suggested by the reviewer, we have made rose-plots for HCHO, CO, O_3, and aromatics (see below). Clearly, all these pollutants were associated with southeasterly wind. We have revised the manuscript as following (L398):

"In addition, all heavy pollution episodes observed in this campaign were associated with easterly wind, which can be demonstrated by the rose-plots of HCHO, CO, O_3, and aromatics in Fig. 7."







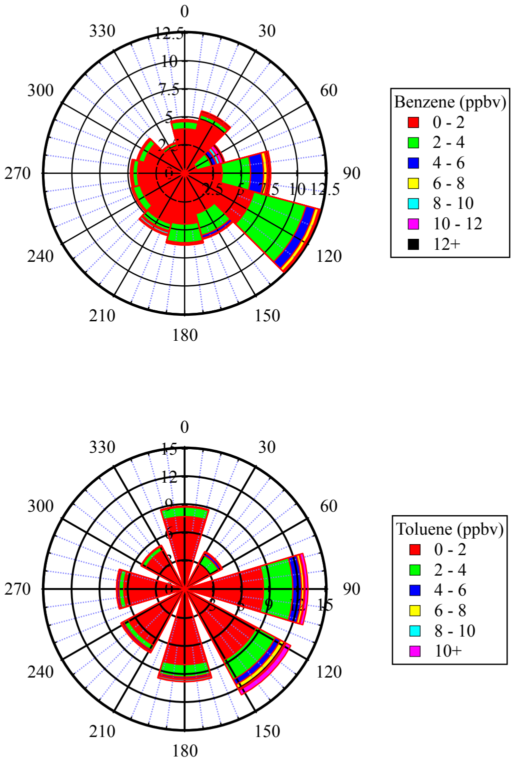


Figure 7. Rose-plots of HCHO, CO, benzene, toluene and O_3 The radius represents the percentage of each air pollutant within a certain mixing ratio range. The spread angle denotes the wind direction.

> *12）L335: Temporal variations of HCHO: The apparent proximity to significant point sources of pollutants leads to the expectation of strong variabilities of these pollutants. The limited duty cycle of the VOC measurements (~2s/180s) make it likely that the measurement does not sufficiently capture these variabilities. The observed covariances of HCHO with other VOCs are likely underestimated due to the lags time between the sequentially measured mass channels because these time lags may be large compared to the characteristic time scale of the VOC variabilities. This is detrimental to the multi-linear regression. Please, discuss how an improved measurement scheme (better duty cycle) and/or different mass spec technology (e.g. TOF-MS) would affect the multi-linear regression analysis as well as the LOD.*

Response: We agree with the reviewer that high time resolution data set can reveal more detailed variations of point source emissions. The industrial zone in Nanjing is a rather intensive point source in a regional scale as indicated by Fig. 1 of Zheng et al. (2015). It normally takes several hours for the plumes from the industrial zone to arrive at the site and thus it is reasonable to assume the plumes were well mixed. The VOC pollution episodes observed in this work typically lasted for more than a few hours, which were consistent with previous ammonia and amine measurements conducted at the same site (Zheng et al., 2015). The atmospheric lifetime of HCHO and aromatics are also on the order of at least a few hours. Therefore, we are confident that ~3 min time resolution should be sufficient to capture the variabilities of VOC plumes in this study. The time resolution can be substantially reduced by using a time-of-flight mass spectrometer (ToF-MS), which is capable of performing measurements at 1 Hz or better. Therefore, ToF-MS is more suitable for capturing highly variable point sources, such as automobile emissions during a tunnel experiment or flux measurements of trace gases (e.g., ammonia). We have inserted the following sentences into L408:

"On the other hand, the industrial zone in Nanjing is a rather intensive point source in a regional scale as indicated by Fig. 1 of Zheng et al. (2015a). It normally takes several hours for the plumes from the industrial zone to arrive at the site and thus it is reasonable to assume that the plumes are well mixed. The VOC pollution episodes observed in this work typically lasted for more than a few hours, which were consistent with previous ammonia and amine measurements conducted at the same site (Zheng et al., 2015a). The atmospheric lifetime of HCHO and aromatics are also on the order of at least a few hours. Therefore, we are confident that the ~3 min time resolution should be sufficient to capture the variabilities of VOC plumes in this study."

> *13）L342+: HCHO comparison: How was the linear fit calculated? A fair comparison between two methods should use orthogonal distance regression (ODR) rather than least square regression (LSR). The former appoints measurement uncertainties to the two datasets according to their performance tests (or a priori knowledge), the latter implicitly assumes that data plotted on the y-axis has uncertainties and the data plotted on the x-axis is precise.*

Response: We agree with the reviewer that the linear fit should be calculated using orthogonal distance regression (ODR) rather than least square regression (LSR). Indeed, ODR was used here to investigate the correlations between these two measurements. We have revised the sentences here (L450) as:

"Figure 9 shows a scatter plot of PTR-ID-CIMS versus DNPH with a slope of 0.81 and an intercept of 0.66 (R^2 = 0.80, based on orthogonal distance regression). Based on the laboratory calibrations conducted with both N_2 and air (at various levels of RH), we found that the uncertainty of PTR-ID-CIMS HCHO measurements was within 18% and that of he DNPH measurements was within 3.6%. Therefore, it is reasonable to believe that the observed discrepancy between these two data sets can be explained by the combined measurement uncertainties and the higher background level in the PTR-ID-CIMS measurements."

> *14）L348: Calculate confidence intervals for the fit parameters to test whether the slope, which differs from 1, and the bias are truly accounted for by the combined measurement uncertainties. If, under the consideration of the quantified measurement uncertainties, the slope and/or the bias are significantly different from 1 and 0, respectively, one must assume a systematic discrepancy. We won't know until the analysis is done (with measurement uncertainties quantified under ambient conditions; see comment to L272+ above).*

Response: According to the additional laboratory calibrations conducted with both N_2 and air (at various levels of RH), we found that the PTR-ID-CIMS detection limit of HCHO varied from 0.9 ppbv for dry condition to 2.4 ppbv for 81.5% RH at room temperature. The variations of the instrument sensitivity (NCPS per ppbv) ranged from a few percent to 17%. We also determined the calibration-to-calibration LOD variation at different RH levels to be less than 16% for RH < 81.5%. Accordingly, the uncertainty of PTR-ID-CIMS HCHO measurements was within 18% (including 1% uncertainty associated with the HCHO standard concentrations) and that of the DNPH measurements was within 3.6%. Therefore, it was reasonable to believe that the observed discrepancy between these two data sets can be explained by the combined measurement uncertainties. We have included the following sentences into L451:

"Based on the laboratory calibrations conducted with both N_2 and air (at various levels of RH), we found that the uncertainty of PTR-ID-CIMS HCHO measurements was within 18% and that of he DNPH measurements was within 3.6%. Therefore, it is reasonable to believe that the observed discrepancy between these two data sets can be explained by the combined measurement uncertainties and the higher background level in the PTR-ID-CIMS measurements."

> *15）L381+: ". . .the background level of HCHO was constrained to 1ppbv to represent the regional conditions." It is rather arbitrary to pick a background level from measurements performed 4 years earlier in a different season and at a different location. Did an unconstrained background level cause any of the parameters to turn negative? If so then non-negative matrix factorization might help to find the best solution. Or one could run a series of multiple-linear regressions over a range of (constrained) background levels and find the background level that maximizes R2. A different background level might result in a linear combination that explains more than 52% of the HCHO variability. However, insufficient capturing of the concentration variabilities due to limited duty cycle and insufficient capturing of covariances due to lag-times (see L335: Temporal variations of HCHO, above) as well as additional variability of HCHO may strongly limit the percentage of explained variability of HCHO. The source-apportionment needs to be discussed in this light.*

Response: As stated in the previous responses, we have demonstrated that the 3 min time resolution was sufficient to capture the VOCs variability in this work and thus will not affect the analysis results. Using unconstrained background level indeed can generate negative results. As suggested by the reviewer, we have performed a series of multiple-linear regressions with constrained background levels ranging from 0.5 to 1.7. The corresponding R^2 ranged from 0.43 to 0.53. In either case, the parameters of β_3 and β_4 in eq. E4 did not change substantially and remained the dominant terms. Therefore, the final conclusion of this work will not be affected significantly by choosing different background HCHO level. Even though the literature background values were ~4 year old due to the scarce availability of HCHO data sets in the region, this is the most reasonable estimate we can obtain. Therefore, we prefer 1.0 ppbv as the constrained background level. We have included the following statements into L511:

"To verify the usage of 1.00 ppbv as the background HCHO level, we also have performed a series of multiple-linear regressions with constrained background levels ranging from 0.5 to 1.7. The corresponding R^2 ranged from 0.43 to 0.53. In all cases, the parameters of β3 and β4 in eq. E4 did not change substantially and remained the dominant terms. Since 1.00 ppbv HCHO background level was supported by previous work (Wang et al., 2015), we thus preferred 1.00 ppbv as the constrained background level."

> *16）L394+: The linear regression between measured and modelled HCHO variability (also presented in (Fig 8) needs statistical treatment to allow for a quantitatively substantiated statement that measured and modelled HCHO variability agree reasonably (see comment L348 above).*

Response: We have performed a student t test on the inter-comparison data set and obtained a p value of 0.15, which is great than 0.05. We have revised the manuscript (L521) as:

"Based on a student t test, the measured and approximated HCHO values are not significantly different from each other (p value = 0.15) at a 0.05 significance level."

> *17) Fig 3: The fit curve of the water vapor concentration dependency of the HCHO sensitivity does not show a theoretically expected 4-5 fold change of the sensitivity (compare to Fig 5 in Vlasenko et al. 2010). A too narrow humidity range in the test might party be the reason that the 'true shape' is not captured by the fit. Please, comment on this. In any case, extrapolation of the humidity dependence of the HCHO sensitivity beyond the tested humidity range is not permissible and might limit the applicability of the calibrations to the ambient data set.*

Response: Figure 3 was based on calibrations conducted during the measurement period, which can best represent the actual performance of the PTR-ID-CIMS during the field operation. During laboratory calibrations with RH ranging from 1% to 81.5%, we found that the instrument sensitivity decreased by a factor of ~3 (see Fig. S2), which was significant less than the 4~5 fold change reported by the other work (Vlasenko et al., 2010). We believe the most likely reason is that the ion source of PTR-ID-CIMS used humidified pure nitrogen as carrier gas. The nitrogen flow rate (30.0 SCCM) was much higher than the water vapor flow rate used by a typical Ionicon PTR-MS and more importantly all of the humidified nitrogen were sucked into the drift tube instead of being pumped away in the case of PTR-MS. Consequently, the background water concentration in the drift tube of PTR-ID-CIMS was much higher than a PTR-MS. As indicated by Fig. 5 of Vlasenko et al. (2010), the instrument sensitivity will change substantially when the water content changes slightly from the completely dry condition. Therefore, the relatively high background water content in the PTR-ID-CIMS can lead to decreased sensitivity during "dry" calibrations, which can explain the much less variation in sensitivity observed in this work. We have inserted the following sentences into the manuscript (L264):

"In Fig. 3 much less variation in instrument sensitivity was observed in this work than the literature (Vlasenko et al., 2010). The most likely reason is that the ion source of the PTR-ID-CIMS used humidified pure nitrogen as carrier gas. The nitrogen flow rate (30.0

SCCM) was much higher than the water vapor flow rate used by a typical Ionicon PTR-MS and more importantly all of the humidified nitrogen were sucked into the drift tube instead of being pumped away in the case of PTR-MS. Consequently, the background water concentration in the drift tube of the PTR-ID-CIMS was significantly higher than a typical PTR-MS. As indicated by Fig. 5 of Vlasenko et al. (2010), the instrument sensitivity will change substantially when the water content changes slightly from the completely dry condition. Therefore, the relatively high background water content in the PTR-ID-CIMS can lead to decreased sensitivity during "dry" calibrations, which can explain the much less variation in sensitivity observed in this work."

> *18) Fig 5: Replace the quiver plot in the top panel by time series of wind speed and direction. Add vertical grid lines to improve comparability of the time series. Potentially add a rose-plot of HCHO sector distributions (as separate figure)*

Response: Figure 6 has been modified accordingly and a rose-plot of HCHO has been added into the manuscript (see the response Comment #11).

> *19) Fig 6: Plot also 1:1 line and confidence ranges of the linear fit to test slope and bias for statistical significance.*

Response: As suggested by the reviewer, we have added 1:1 line, confidence ranges of linear fit to Fig. 9

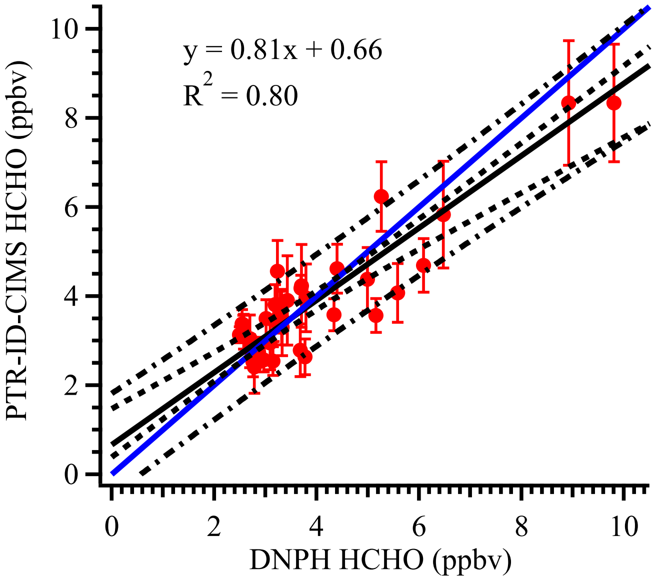


Figure 9. Linear correlation between PTR-ID-CIMS and DNPH measurements of HCHO. The blue trace represents the 1:1 guideline. The black solid line is the linear fit based on orthogonal distance regression. The dashed line and the dot-dashed line denote the 95% confidence interval band and the 95% prediction band, respectively. The error bars indicate one standard deviation of PTR-ID-CIMS data within the corresponding DNPH sample collection time.

> *20) Fig 8: Plot also 1:1 line and confidence ranges of the linear fit to test slope and bias*

for statistical significance.

Response: As suggested by the reviewer, we have added 1:1 line and confidence ranges of linear fit to Fig. 11.

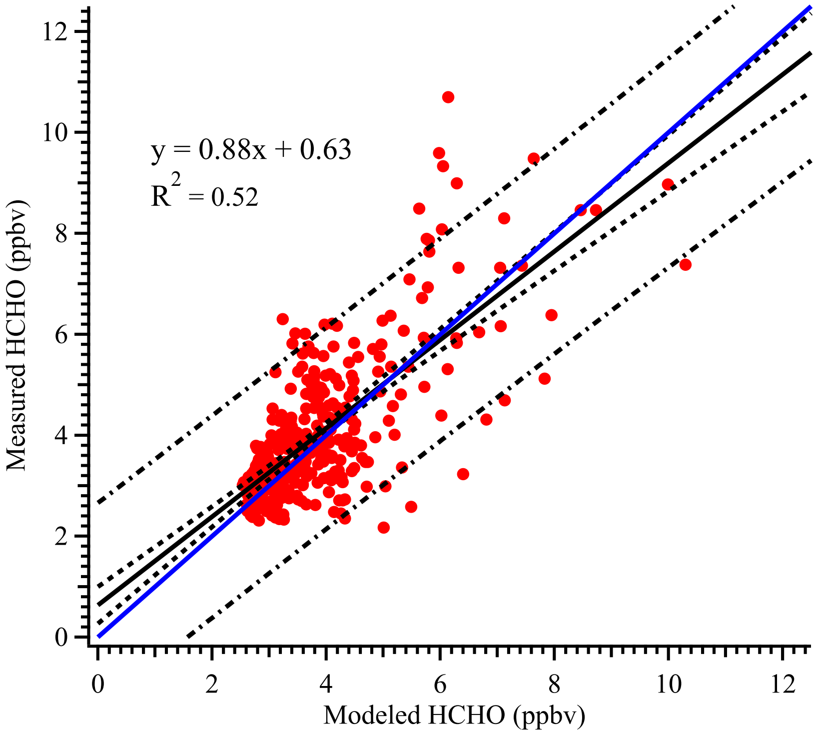


Figure 11. Linear correlation between the approximated and measured HCHO concentrations. Both data sets are synchronized into one-hour temporal resolutions. The blue trace represents the 1:1 guideline. The black solid line is the linear fit based on orthogonal distance regression. The dashed line and the dot-dashed line denote the 95% confidence interval band and the 95% prediction band, respectively.

21) L127: Please, add coordinates of the sampling location. Reproduce with permission or refer explicitly to Figure 1 in Zheng et al. (2015 a).

Response: As suggested by the reviewer, we have inserted the coordinates of the sampling location into the manuscript and the sentence has been modified as following (L134):

"Detailed description of the site (N32° 12' 20.8", E118° 42' 19.2") has been given in our previous work (please see Figure 1 in Zheng et al. (2015a) for details)"

22) L331: ". . .calm with light easterly wind." Was it calm (i.e. no wind) or were their light easterly winds?

Response: We have deleted "calm" and modified the sentence as (L437):

"From 10:00 AM of April 18, there was a persistent easterly wind with wind speed varying within a few meters per second. Meanwhile significant increase of HCHO was observed at the site."

References:

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