First, we would like to thank the reviewer for his/her valuable comments on the manuscript. We did our best to address all the comments and summarized the changes made to the revised manuscript below.

Response to referee #1:

>*Novelty: As this study describes a new instrument which is built based on an existing concept, you need to be careful to highlight strongly the new achievements of this presentation compared to previous work. In particular title and abstract should illustrate what is new, what is different to previous CRM instruments and publications, and what is the impact of the new findings.*

Title: The manuscript-title neither tells the reader anything about the newly build Mine Douai instrument nor it explains what is addressed by "experiment vs. modelling". May I suggest to formulate it more precisely? (E.g. "Detailed characterizations of the new Mine Douai Comparative Reactivity Method instrument via experiment and model")

The title has been modified in the revised manuscript and reads: "Detailed characterizations of the new Mines Douai Comparative Reactivity Method instrument via laboratory experiments and modeling "

> *Abstract: Until line 24 the reader finds nothing really new in the abstract. It would help to comprise this part of the abstract keeping all the interesting and important facts, on the one hand. And on the other hand, the new aspects and achievements of the presented study can be added, e.g. the new Mine Douai instrument, and the relative magnitude of the corrections (as examined in this paper). The second part of the abstract reads rather lengthy and may be shortened for the benefit of summarizing actual results. Similar to what is written in the introduction (p. 3808, l. 17-22) it could be more informative for the reader describing precisely what kind of model was used for what purpose. In general, I think the abstract should have information about the new instruments performance (LOD, uncertainty), about the relative importance of the examined and applied corrections (e.g. which correction has the greatest impact on the result? does it correct towards higher/lower values?), and about the results of the model compared to the experimental tests (e.g. what can you learn from the comparison?) What is the benefit of your study for other researchers and CRM users?*

The abstract has been rewritten following the recommendations of this reviewer. Details about the performances of the Mines Douai instrument (LOD, uncertainty), as well as the impact of each correction are now given in the abstract. The description of both mechanisms has also been shortened.

> *p. 3804, l.16: I find the description of this first correction not precise, or even confusing. It is not very clear, if you are talking about the humidity dependence of the PTR-MS sensitivity or the OH production inside the reactor. I believe that you mean the latter one. Please, could you modify the sentence accordingly?*

Indeed we are talking about the difference of humidity between wet zero air (C2 step) and ambient air (C3 step) that leads to different levels of OH inside the sampling reactor. The sentence has been modified as follows:
"a correction for a change in relative humidity between the measurement steps leading to different OH levels"

> *p. 3804, l. 18: HO2, NO: Abbreviations are commonly introduced for the first time by spelling out the entire word or phrase.*

HO$_2$ and NO have been introduced by their entire name in the revised manuscript, i.e. hydroperoxy radical and nitrogen monoxide respectively.

> *p. 3806, l. 4-6: Volatile organic compounds (VOC) are of particular interest for the OH chemistry not only because they are present in the atmosphere in large numbers, but because they are often highly reactive to OH. Please, add this important detail.*

The requested information has been added in the revised manuscript:
"Volatile Organic Compounds (VOCs) are of particular interest for the OH chemistry due to the presence of a large number of reactive species (10^4-10^5), emitted by natural and anthropogenic sources, or formed photochemically (Goldstein and Galbally, 2007)"

> *p. 3806, l. 9: ". . . orders of magnitudes lower than expected in the atmosphere."*

The requested modification has been made.

> *p. 3806, l. 28-29: There are many more references. Either list them all or insert "e.g."*

"e.g." has been inserted

> *p.3807, l. 4-5: Please, include the original publications for the different techniques here. It seems that the given reference is not published yet.*

The original publications have been cited in the revised manuscript: Kovacs and Brune (2001) and Sadanaga et al. (2004) for the total OH loss rate method and the pump-probe method, respectively.

> *p.3808, l. 10: Correction (iii) is not mentioned in the abstract. Please be consistent.*

This correction has also been listed in the abstract for consistency.

> *p. 3809, l. 6: Again, Hansen et al. (2015) is cited, even though the reference is not published yet. In this context either another publication that describes the CRM in detail may be mentioned or you present your manuscript as the first which discusses the MD-CRM in detail.*

The Hansen et al. publication is now available in AMTD (ref: Hansen, R. F., Blocquet, M., Schoemaecker, C., Léonardis, T., Locoge, N., Fittschen, C., Hanoune, B., Stevens, P. S., Sinha, V., and Dusanter, S.: Intercomparison of the comparative reactivity method (CRM) and pump-probe technique for measuring total OH reactivity in an urban environment, Atmos. Meas. Tech. Discuss., 8, 6119-6178, doi:10.5194/amtd-8-6119-2015, 2015.).

> *p. 3809, l. 8: Again, the abbreviation should be spelled out for the first time mentioned.*

The requested modification has been made.

> *p. 3810, l. 23: As far as I can see, this manuscript is published before Hansen et al. (2015). Please, modify the text accordingly.*

As mentioned above, the Hansen et al. publication is now available in AMTD.

> *p. 3811, l. 7: Please, be precise and list the entire list of chemicals or refer to Table S1 directly.*

We kept the same list but the reader is referred to Table S1 in the revised manuscript

> *p. 3813, l. 2-5: In the PTR-TOF-MS on m/z 19 the signal of primary ions and on m/z 37 the first water cluster are monitored. Due to their high abundance these two peaks are often saturated in such instruments and do not follow a linear behavior anymore. Could you please comment on your instrument's behavior? Since you use the m37/m19 ratio for correction, their accurate determinations seem to be critical for the OH reactivity measurement.*

We agree with the referee that with PTR-ToFMS, the signals at m/z 19 and m/z 37 are often saturated. To avoid this issue, we follow signals at m/z 21 and m/z 39, which correspond to $H_3^{18}O^+$ and the corresponding first water cluster. We then calculate the signals at m/z 19 and m/z 37 by multiplying the signals at m/z 21 and m/z 39 by 500 and 250, respectively. 500 corresponds to the isotopic ratio of ^{18}O compared to ^{16}O, and 250 the same ratio divided by two since the first water cluster at m/z 39 has one ^{18}O and one ^{16}O.

> *p. 3813, l. 15-19: When reading this paragraph for the first time, I wondered if you did the NO-sensitivity experiment only for zero OH reactivity or also for different OH reactivities introduced by different test-compounds? Later in your manuscript (p.3820, l.11-14), you mention that you did these tests. That is why I think that you should extend the description here and include a sentence about tests with various compounds and different levels of OH reactivities.*

A sentence describing the NO experiments performed with various gas standards at different OH reactivity values has been added in the revised manuscript:
"Similar experiments were conducted by adding ethane and isoprene together with NO in the sampling reactor, leading to OH reactivity values of 22.2 s^{-1} and 36.6 s^{-1}, respectively"

> *p. 3813, l. 25: Why did you not test for NO2 interference below 60 ppbV of NO2? Typical atmospheric levels are often lower. In your results it looks like the OH reactivity correction in this region (Figure 4, 0-60 ppbV NO2) is very sensitive to changes in NO2. It would be beneficial for the manuscript to include some measurements at these critical NO2 levels.*

For this test we used a mixture of NO_2 at 44.7 ppmv and it was difficult to lower the NO_2 mixing ratio below 60 ppbv inside the sampling reactor under the current operating conditions (total flow of approximately 380 sccm). Some experiments have been conducted under different operating conditions (total sampling flow rate of 510 sccm) which allowed us to reach NO_2 mixing ratios as low as 25 ppbv (see Figure R1). These results have not been shown in this publication since it corresponds to previous settings of the instrument. However, the figure below shows that the change in C3 seems to be linear with NO_2 below 60 ppbv and close to what is observed using operating conditions described in this publication. Therefore, we believe that the quadratic regression showed in Fig. 4 is suitable for NO_2 mixing ratios lower than 60 ppbv.
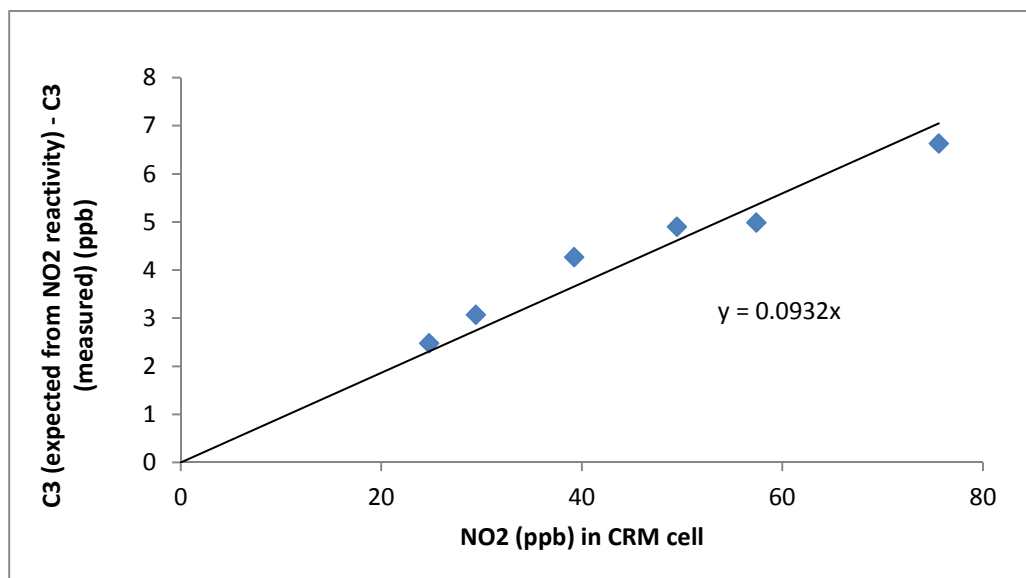
Figure R1: Laboratory experiments performed to investigate the NO_2 artifact using previous operating conditions for the CRM: change in C3 as a function of NO_2 inside the reactor.

> p. 3816, l. 11: typo: "extracted"

The requested modification has been made.

> p. 3815, l. 24 - p. 3816, l. 4 (Table S2): Could you please explain the choice of the reaction rate constants used in the simple model mechanism? Can you comment, in particular, on the slow rates attributed to the peroxy radical reactions (RO2+NO, RO2+RO2, RO2+HO2)? How much does the choice of these rate coefficients affect the agreement between simulation and measurement? Please, provide a reference for pyrrole+OH (Dillon et al., 2012).

The reaction rate constant for OH + Pyrrole corresponds to values published by Atkinson et al. (1984) and Dillon et al. (2012) as stated in section 2.1. These references have been added in the revised manuscript. Concerning the reaction rate constant chosen for the surrogate hydrocarbon, it has been arbitrary set to 5.0×10^{-12} cm^3 molecules^{-1} s^{-1}, which corresponds to the reaction of OH with C5-C6 alkanes or aromatics such as toluene. This has been indicated in the revised manuscript.
Concerning the peroxy radical reactions, the reaction rate constants are those for methylperoxy radical (CH_3O_2), as it has been mentioned in the original manuscript. Since, individual reaction rate constants are used in MCM, the impact of such a choice is within the differences observed between the results of both mechanisms, i.e. a negligible impact when simulating the deviation from pseudo first order conditions and a significant impact for simulating the NO artifact. The latter has already been discussed in the original manuscript (p3823, l. 12-24).

> p. 3816, l. 15-23: As stated in your manuscript, the model approach is a very crude one. The surrogate 2(5H)-Furanone might have a very different oxidation chemistry than pyrrole. Did you test your mechanism as well for other potential pyrrole surrogates? Are there significant differences? What advantage do you think you gain by using the MCM with a surrogate for pyrrole instead of using the simple mechanism? Can you use this model study to distill any significant result for the usage of CRM or the understanding of pyrrole chemistry?

We believe that using this surrogate is the best we could do since there is no information about the pyrrole chemistry in the literature.

To answer the referee's comment, we tested three other surrogates for pyrrole in MCM (n-pentane, 1-pentene, and benzene) keeping the reaction rate constants for Pyrrole (OH and O_3 reactions), and only forming the corresponding RO_2 of the surrogates to account for secondary chemistry. Simulations performed using these three surrogates for a standard addition of 38 ppbv of isoprene at a pyrrole-to-OH ratio of 1.4 lead to small relative differences in F ranging from 2 to 3%. These results are comparable to original simulations made using BZFUONE as a surrogate. However, simulations performed using these three surrogates for an addition of 60 ppbv of NO at a pyrrole-to-OH ratio of 1.4 lead to significant relative differences ranging from 6 to 21% when compared to original simulations using BZFUONE as a surrogate. These differences lie within the differences already observed between the simple mechanism and the MCM using BZFUONE as a surrogate.

Using MCM with a surrogate pyrrole instead of the simple mechanism brings valuable information since it allows including the secondary chemistry of the different gas standards. Moreover, the observation that the results from both mechanisms encompass the experimental results for the NO artifact reveals that the lack of RO_2 speciation and of secondary chemistry in the simple mechanism may be the cause for the disagreement observed between experiments and simulations. These results point out the need for a better understanding of the pyrrole chemistry and we, therefore, concluded in the original manuscript by recommending laboratory studies on pyrrole chemistry.

> *p. 3817, l. 8: typo: "Preliminary results of OH reactivity measurements. . ."*

The requested modification has been made.

> *p. 3817, l. 13-18: Could you specify what kind of inlet and flow you used for the measurements? Was it insolated/heated/flushed?*

We used two different high flow rate inlets (1 L min^{-1}) for the measurements of OH reactivity and VOCs, both of ¼ inch and made of Teflon with a length of approximately 5 m each. The VOC sampling line was heated at 50°C while the sampling line for the CRM was kept at ambient temperature. This has been added in the revised manuscript.

> *p. 3818, l. 4-13: Several points in this paragraph are unclear to me and should be clarified: You corrected the detected C2 pyrrole levels for the relative humidity which you determined via m37/m19 ratio. Did you do this for every single measurement point during C2? Or for the average of C2? Or for the interpolated C2 values that match the times of the C3 measurement points? Depending on your approach, I wonder if the noise of m/z 19 and m/z 37 may affect your corrected pyrrole levels? How do you account for rapidly changing humidity during C3 between two C2 measurements? Why did you chose to correct the C2 pyrrole measurements and not the C3 values?*

We corrected the 5-min averaged C2 values surrounding each C3 measurements. We then averaged two corrected C2 values surrounding one C3 and used it in Eq. 1. We found that uncertainties associated to the measurements of m/z 19 and m/z 37 are negligible (~0.05% of relative uncertainty) (see supplementary material S14) and do not affect the corrected pyrrole

levels. Finally, we chose to correct the C2 measurements based on the consideration that the RH value that is the most representative of the OH reactivity measurements is the value recorded during C3 (ambient air flowing inside the reactor). Based on this rational, the C2 measurements are not representative of the baseline that needs to be subtracted and the C2 values have to be corrected to match the RH observed during C3.

> p. 3818, l. 17: typo: "black"

The requested modification has been made.

> p. 3818, l. 18-20: Could you please give typical numbers for the amplitude of the correction? How much variability did you find in the pyrrole mixing ratios, how much is this in OH reactivity, and how big is the average relative correction of the OH reactivity data?

These details have been given in the revised manuscript at the end of section 5.1:
"(0.1 and 3.9 ppbv for the mean and the maximum variation of C2, respectively). The average correction was 5.2 ± 3.2 s^{-1} (1σ) for the whole field campaign.".

> p. 3822, l. 5-13: In this section it is very important to clearly define: What are "apparent ratios"? What has been measured? What was modeled? Could you please rephrase?

Apparent Pyrrole-to-OH ratios correspond to ratios calculated using eq. 2 (i.e. C1/(C1-C2)) and which have been defined as pyrrole-to-OH in previous studies (e.g. Sinha et al., 2008). However, this ratio does not correspond to the real ratio inside the reactor since the quantity C1-C2 is not the total mixing ratio of OH but the amount of OH reacting with pyrrole. In this paper the real pyrrole-to-OH ratio corresponds to the mixing ratio of pyrrole divided by the mixing ratio of OH set in the model to reproduce the C1-C2 modulation observed during laboratory experiments. This has been rephrased in the revised manuscript as follows:
"As already mentioned, the calculations do not lead to the real pyrrole-to-OH ratios (calculated from concentrations of pyrrole and OH used to initialize the model) but to apparent ratios since C1-C2 is not the total OH mixing ratio but the amount of OH reacting with pyrrole".

> p. 3822, l. 14-20: You describe in the SI how you derive experimentally the OH mixing ratios inside the glass reactor. I find this information very important and would encourage you to include it in the main manuscript. Additionally, I was wondering how did you determine the model OH mixing ratio?

The requested information has been added in the revised manuscript:
"Real mixing ratios of OH inside the reactor were also determined experimentally by introducing a large amount of Isoprene (3 ppmv) in the presence or absence of OH. OH mixing ratios were calculated from the consumption of isoprene and compared to levels set in the model to reproduce laboratory observations (see supplementary material, Fig. S4)".
Each simulation is initialized setting pyrrole, gas standards, and OH concentrations. The model OH mixing ratio is set at the beginning of the simulation to reproduce the apparent pyrrole-to-OH ratios observed during the experiments, as mentioned in the original manuscript (p3822, l. 14-15).

> p. 3824, l. 27: typo: "through"

The requested modification has been made.

> *p. 3826, l. 26: Are you talking about the "full range of reaction rate constants" of OH reactive compounds in your sample?*

We were talking about the full range of rate constants for the reactions of OH with volatile organic compounds found in the atmosphere. Since ethane is one of the less reactive VOC in the atmosphere (reaction rate constant: $2.4\ 10^{-13}\ cm^3$ molecules^{-1} s^{-1}) and isoprene is one of the most reactive (reaction rate constant: $1.0\ 10^{-10}\ cm^3$ molecules^{-1} s^{-1}), we recommend using these 2 compounds to derive the lower and upper bounds for the correction factor and to use an average of these 2 values for correcting ambient measurements.

> *p. 3827, l. 9-12: During your tests of the influence of the pyrrole-to-OH ratio on the correction, that you need to apply to account for pseudo-first order assumptions, you find that (as expected) the correction decreases for higher ratios. From your experiments you conclude that for higher ratios than 2.3 a correction factor smaller than unity was observed. However, "this tendency is only significant for one experiment". Since this observation is such a puzzling result, not easy to explain, the scatter of the data generally looks quite high (Fig. 7), and only one experiment has been carried out in this region of ratios, it seems logic to me to add a few more experiments in that region. The additional data will definitely tell you if you are converging to unity or some number below unity. Subsequently, you may try to explain the puzzle or find that the experiments actually show what was expected.*

Pyrrole-to-OH ratios observed during field experiments being in the range 1.6-2.2 (see colored area of Fig. 7 in the original manuscript), we did not perform additional experiments at ratios higher than 2.3. We agree with the referee that the decrease of the correction factor below unity is not clear since it is significant for one experiment only. The discussion about correction factors decreasing below unity has therefore been withdrawn and we now only comment the decrease of this correction with increasing pyrrole-to-OH ratios:
"A decrease of the correction factors with the pyrrole-to-OH ratio is consistent with a kinetic regime getting closer to pseudo first-order conditions (OH<<Pyrrole) and therefore to correction factors closer to one".

> *p. 3827, l. 27 (Figure 7): You conclude that the relationship between pyrrole-to-OH ratio and the correction factor (experimentally determined) is linear. However, theory expects a converging relationship (p. 3827, l.12). This can be seen as well by the model-runs presented in Figures S8-S11 or Figure 9. So, why do you use a linear regression rather than a polynomial one?*

A close look at the experimental data shows that no curvature is observed and a linear regression seems more suitable for the range of pyrrole-to-OH ratios observed in the laboratory, which encompasses the range of ratios encountered during field experiments. Using a polynomial fit would lead to a second-order monomial that wouldn't be statistically significant.

> *p. 3832, l. 19-27: It would be great if you could give numbers (absolute and relative) about how much is the impact of each of the previously discussed corrections on the measured total OH reactivity?*

Details about the absolute impact of each correction have been added in the revised manuscript:

"In urban environments such as the Dunkirk site, the correction for NO_x has the largest impact on OH reactivity measurements and is 9.2 ± 15.7 s^{-1} on average. The correction that has the second largest impact is due to not operating the instrument under pseudo-first-order conditions and is 8.5 ± 5.8 s^{-1} on average. The humidity correction is also significant and is 5.2 ± 3.2 s^{-1} on average due to fast changes in ambient RH (proximity of the sea). The correction for dilution is constant and leads to an increase of the measurements by a factor 1.23."

> Figure 1: For better visibility of the insert, please frame it.

The requested modification has been made.

> Figure 2: Please include error bars. Like this the reader can see how big is the variability for both the C2 signal (pyrrole mixing ratio) and the m37/m19 (tracer for humidity).

The error bars of C2 signals have been added in Figure 2. Error bars for m37/m19 ratio have not been added since they were too small (uncertainty of 0.5%).

> Figure 3: The labels and legends of the two lower graphs were too small to be readable on my print-out. Is the outlier point for the highest pyrrole-to-OH ratio the dry experiment? Was it included in the data to determine the fit?

Figure 3 has been modified to make it more readable. Indeed, the outlier point for the highest pyrrole-to-OH ratio corresponds to the dry experiment and was not included in the analysis as stated in the original manuscript (p3820, l. 7-11) and the original title of Fig. 3.

> Figure 4: Unfortunately, there are no experiments performed for NO2 mixing ratios between 0 and 60 ppbV. It would be great to include a couple of experiments in that region, since these are atmospherically relevant mixing ratios of NO2. Why are the green experiments (pyr/OH=2.0) so different from the other two experiments? These data points seem to be significantly lower.

See our previous answer concerning experiments performed for NO_2 mixing ratios below 60 ppbv.
We did not find any reason to explain why the experiment performed at a pyrrole-to-OH ratio of 2 seems lower than the two others (no obvious experimental issue during this experiment) and we therefore decided to use an average fit for this artifact.

> Please, check all your graphs to be consistent with the use of ppb or ppbV.

"ppbv" has been used for all graphs.

> Figure 5: The labels and legends of the two lower graphs were too small to be readable on my print-out. Also, the legend does not help to distinguish between the simple and the MCM simulations, because not the markers make the difference but the line appearance.

Figure 5 has been modified to make it more readable. The distinction between the two mechanisms is made with different marker colors and different line appearances. The legend for the markers also takes into account the different colors. The two mechanisms and the different line appearances are described in the caption.

> *Table S2: The given values for the bimolecular rate constants are missing a multiplication sign. Why is there nothing written behind the equal sign for RO2+RO2 and RO2+HO2? It would be important to highlight that both of these reactions are terminating steps. Could you explain your choice of rate constants in more detail, please?*

The multiplication signs have been added in the revised supplementary material. There is nothing written behind the equal sign for RO_2+RO_2 and RO_2+HO_2 since they are considered as termination pathways and no product is formed in the mechanism from these two reactions. A note has been added below the table to mention it. Please see our previous answer for selecting the rate constants.

> *Figure S4: You explain that you derived experimentally the OH mixing ratio inside the glass reactor by using isoprene as scavenger. Could you please be more precise on this description? Did you add isoprene instead or additionally to pyrrole to the reactor? Why is isoprene a better scavenger than pyrrole? They both have very similar reaction rate coefficients with OH. How did you observe a significant difference (larger than noise) between isoprene mixing ratios with and without OH, having in mind that isoprene is about 3 ppmV and OH about 50 ppbV? Maybe it would make it easier to understand if you showed the data of the experiment (e.g. isoprene and pyrrole as detected by the PTR-TOF-MS)? Did you try other scavengers? Did you get to the same result? How big is the effect of OH recycling (e.g. in the isoprene chemistry)*

Isoprene was added without pyrrole during these experiments. This information has been added in the revised supplements.
Isoprene is not a better scavenger than pyrrole, but pyrrole is very sticky on surfaces and we did not want to introduce large concentrations of this compound inside the CRM instrument and the PTR-ToFMS.
We agree that the differences in Isoprene mixing ratios with and without OH are close to the noise but they are still significantly higher (see the error bars representing the 1σ precision of this signal).
We did not try to use other scavengers and we cannot provide a direct answer about OH recycling in the isoprene chemistry. However, it is interesting to note that due to the large amount of HO_2 radicals in the CRM reactor, the loss of isoprene-derived peroxy radicals will likely be driven by RO_2+HO_2 reactions. As a consequence, OH recycling due to unimolecular reactions of isoprene-derived peroxy radicals is likely not significant. OH recycling from the reaction of hydroxy-containing RO_2 radicals with HO_2 has been studied by Dillon and Crowley (2008). Unfortunately, due to technical difficulties, the authors were not able to study isoprene-derived peroxy radicals. However, this study highlights that OH was not a major product for the reaction of other hydroxyl-containing RO_2 radicals with HO_2, with an upper limit for the OH yield of 5-6%. The impact of OH recycling during our isoprene experiments is therefore expected to be small.

> *Figure S6: Why don't you add the experiments for comparison to the model results (shown in Fig. S2)?*

Experiments shown in Fig. S2 are not shown in Fig. S6 for clarity. The main goal for these figures (S2 and S6) was to show the impact of gas standard additions on the NO artifact and not to compare experiments and modeling.

References

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