On behalf of the authors of the article, we would like to sincerely thank the reviewers for their valuable time, constructive comments, and thoughtful suggestions, which have helped us to significantly improve the quality and clarity of our manuscript. Please find below the answers to their questions. In the manuscript all the modifications that have been made respect to the original submitted version are highlighted in red.

RC1:

1. Although open path CEAS systems are relatively uncommon, the work should be justified more strongly with additional references at appropriate places. There are several statements in the paragraph beginning L30 where the reader would expect stronger evidence for the assertions being made and more references for the particular species being detected. A reference should be provided for Eq. (1). It would be helpful to note that measurements across atmospheric simulation chambers are a forerunner of the open path IBBCEAS systems.

The references were added to the article (line 29 and 178).

2. The structure of the work is a bit unusual, with a table of results comparing the work with others appearing after the objectives of the work. This does not make sense to me and would fit better in the results and discussion section. Moreover, the comparison with other work would be improved by adding the values in $(Hz)^{-1/2}$ as another column to Table 1 and defining the relationship to the effective pathlength.

The reviewer is right here. We moved Table 1 to the section results and discussion and added a definition of effective path length (Line 161: The effective path length used in the comparison is defined as $L_{eff} = LF/\pi$, where $F = \pi\sqrt{R}/(1 - R)$ is the cavity finesse). The values were already normalised by the square root of Hz.

3. Instrument details should be provided or clarified:

- The cavity length is missing, and the inner diameter of 16 cm is presumably incorrect.

The information about the cavity length (50cm) has now been added in the manuscript (line 74). Thanks for spotting the taping mistake on the inner diameter of the tube that is 16 mm and not cm. This was now corrected in the manuscript.

- It's not completely clear what the flowrate is to the two mirrors and cavity tube. Are they all 1.3 L/min?

This part has now been better explained (Line 90: A total flow of 4 L min−1 STP was used for flushing the two cavity mirrors (with ~1.3 L min−1 on each mirror) and the cavity tube with an inlet flow of ~1.3 L min−1 at the center of the tube.)

It is unclear how far the purge length extends beyond the mirrors, and the extent to which the flow of the purge gas into the optical cavity space shortens the effective sample pathlength. The assumption is that the velocity of the air flow through the box relative to the purge flow rate means the latter is insignificant — is there evidence to support this assumption?

The information about the purging extent has now been added in the manuscript (line 91: The mirror's holders are designed so that the purge length extends for ~1 cm beyond the mirrors) As mentioned in the manuscript, air circulation in the box is provided by a fan with a capacity of 600 L/min. This flow rate is not easy to measure without disrupting it due to the large inlet and outlet sections, but we were able to estimate it by placing a sheet of paper at the inlet and observing the effect of the suction on the paper. We are therefore fairly certain that the air flow rate in the box is well above 4 L/min.

4. There is a relatively big difference in going from a 4^{th} to 8^{th} order polynomial function for the baseline. It is unclear why such a higher level of parameterisation is required between indoor and outdoor measurements. Can the authors provide some insight into how this difference arises? Is it a temperature effect or sample flow effect or does it arise from particle extinction? How does the baseline fluctuate at some wavelength over time? Such insight would be helpful in considering the merits of the open path approach.

This is indeed a very good point. The high-order polynomial is due to the presence of particle extinction. Figure 3 shows the strong baseline shape for outdoor air. A high-order polynomial is required to fit this structure. Having now collected data from urban and marine areas, we can see that the baseline shape is not exactly the same. Therefore, we cannot easily attribute a specific extinction spectrum to aerosol particles. Unfortunately, this issue is more complex and would require further investigation. In this study, we limited our detection to target molecules, including the particle extinction in the baseline of the fit by using the large polynome.

This has now been detailed in the manuscript (from Line 109: For indoor measurement, a baseline described by a polynomial of order 4 was sufficient to provide a flat residual, while for outdoor measurement, the order was increased to 8. This is due to the strong broadband structure that appears on the baseline related to the extinction of light from aerosol particles. At present, it is difficult to assess the contribution of aerosol particles in a quantitative way due to the wide variety of particle types to be taken into account.)

5. Have other atmospheric absorbers been considered: methylglyoxal, ozone, water vapour?

Water absorption, glyoxal and ozone are fitted (this has now been detailed in the capture of Figure 3. CH_3COCHO absorption has not been included because its low spectral structure and low absorption cross-section, but it will be considered in future improvement of our system.

6. How many spectral elements were used in the analysis of NEAS? That is, what is *M*?

The number of pixels used for the fit is M = 800. This information was added to the article (line 116)

7. Fig 4 shows an Allan deviation that is distinctly different than that for the closed path system reported in Barbero et , 2020, showing both a slower decrease in the deviation and no minimum in the values. Can the authors put forward a reason for this behaviour? Where is the principle source of noise in this system and why does

the open path system not follow typical noise behaviour, even as the similar closed path system does?

The reviewer raises a very interesting question to which we have no absolute answer. We should start by saying that it is not easy to do this stability test with the open-path configuration. This would require putting the instrument inside a chamber flushed with a controlled air composition, but unfortunately, we did not have the facility in our laboratory, and we did not have the opportunity to conduct such a study so far, but we hope to be able to do it in the future. However, we decided to do this test taking advantage of a deployment of the instrument in a remote area where concentrations of NO2 and of the other absorbing molecules were very low (but still naturally varying). The aim was to estimate an optimum integration time for the measurement. We think that the deviation to the white noise trend as well as the flattering of the minimum absorption coefficient happening within less than 1 min is due to a combination of factors, and in particular: (i) to the lower stability of the system that was placed outside and exposed to meteorological changes; (ii) to the effect of the turbulence flow due to the open path configuration; and (iii) to the changes in atmospheric composition. This has now been detailed in the manuscript (line 120).

8. L146-9: The authors need to demonstrate that the difference in mean between the retrieved glyoxal concentrations in the open and close path configurations is statistically significant. It is unclear that a comparison of the two systems allows an inference to be made about the concentration of glyoxal in particles

The mean values of CHOCHO were reported in the manuscript with the error bars: -37.6 ± 108 and 388 ± 380 ppt for the CP and OP system respectively. Those values are therefore statistically different for us. What changes between the two setups that may create this offset is the presence of the particle filter at the inlet of the CP system. But the reviewer is right, we are not certain that this offset comes from the presence of glyoxal in aerosol particles. So we have adapted the sentence to make this conclusion more like a hypothesis (Line 138).

9. The agreement between open and closed path configurations is very good for indoor measurements, but more variable for the outdoor measurements. There are times when the deviation between the two systems is quite large (e.g., 4/10 and 8/10). Is this purely a cavity alignment issue or does it have some other cause? Why are the LED intensity and I0 so much noisier when switching to the outdoor measurements?

Examination of the data does not reveal any clear anomalies linked to a strong difference in particle extinction or spectral adjustment regarding the discrepancies observed on 4/10 and 8/10. Unfortunately, the data at our disposal do not enable us to explain these sporadic events. The fluctuation in I0 intensity is due to a change in the instrument's temperature. Although the aluminium plate on which the optics are mounted is temperature-stabilised, the cavity mirror supports are exposed to variations in air temperature (which is why we observe diurnal variability during outdoor measurements). The increased noise in LED intensity during outdoor experiments is likely due to the LED being exposed to the outside air. We do

not believe that this is a thermal effect either, since diurnal cyclicity is not evident and the LED is temperature-stabilised.
This part was commented in the manuscript in the lines 169-174: "...At the beginning of the experiment, the transmitted intensity was 5 % higher than during the instrument calibration. Furthermore, one can observe the sudden decrease by 2 % while installing the instrument outside, as well as the thermal effects on the mechanical stress of the instrument during the outdoor deployment, with an amplitude of the diurnal cycle of the spectrum intensity of 1-2%. From this data monitoring we could confirm that during the 10-days continuous outdoor measurements, the open-path instrument did not experience degradation due to a possible dirtiness of the cavity mirrors". Therefore no further information was added in the text.

10. How is the intensity drop in I_0 to a minimum, followed by a recovery, on 16/9 explained (Fig. 7)?

We should note that the intensity of this drop is 5%. The thermal effects observed during the outdoor experiments are not far off in amplitude. This could be due to mechanical instability or a loss of cavity finesse, followed by recovery; however, because of the shape of the I0 intensity signal, the second option seems more plausible.

11. Minor corrections:

We thank the reviewer for their very pertinent remarks. All technical comments were incorporated into the article.

RC2:

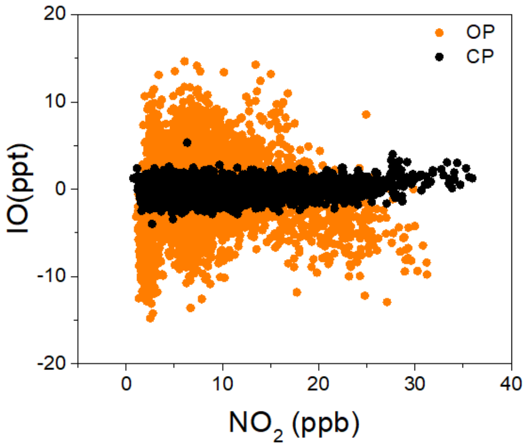
1. The results of optimal data acquisition time test used the minimum absorption coefficient in Figure 4. However, I would suggest presenting the results in mixing ratios for better comparison with previous studies

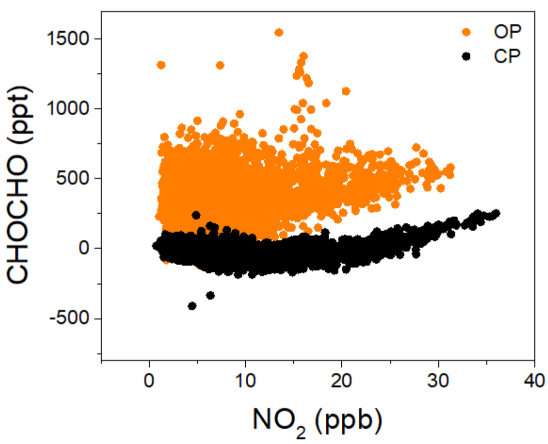
This was not done because we do not have the equipment to expose the open-cavity instrument to controlled and stable concentrations of the species under study. To produce Figure 4, we took advantage of a recent field campaign with low NO_2 concentrations to study the long-term stability of the measurements. This plot is useful for identifying the optimum averaging time for the data. We can see that, unlike the close-path configuration, there is no point in averaging the signal for longer than one minute in the open-path configuration.

2. For the detection of IO and glyoxal, the authors showed the measured results of these two species with NO2 obtained from an indoor and outdoor environment. However, these results are not convincing enough for me to demonstrate that the instrument is capable of measuring IO and glyoxal. First, the measured concentrations vary around zero and seem to exhibit as noises. Although it is possible in an indoor environment, these two species should show distinct diurnal pattern as they are both influenced by photochemical reactions. Second, while the glyoxal seem to be detected in an outdoor environment and reached up to around 500 pptv, there is no other evidence to exclude the influence of intensive NO2 absorption on IO and glyoxal detection as the absorption cross section and ambient concentration of NO2 is much larger than the other two species. This phenomenon

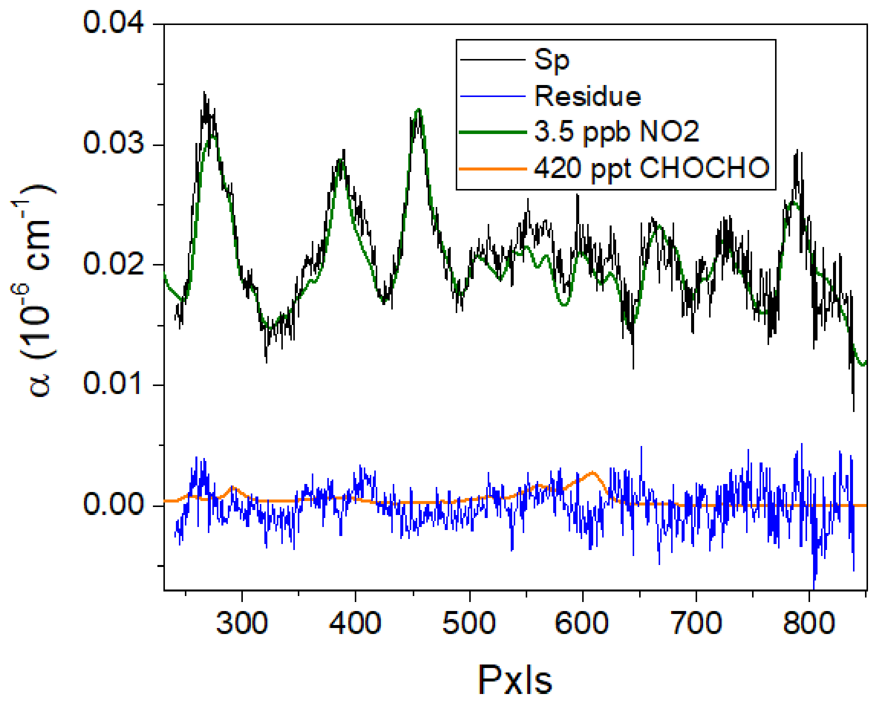
would systematically bias the measured concentration of glyoxal during spectrum fitting. In the meantime, the distinction effect of particles would aggravate this bias. The authors claimed that lower glyoxal concentration measured by CP system is due to the particulate glyoxal is therefore questionable. The detailed spectrum fitting results should be presented to validate glyoxal detection.

We thank the reviewer for this very interesting and pertinent question. This work derived from a previous work (Barbero et al 2020) where more details on the detection of CHOCHO and IO were provided. During the development phase we acquired data with relatively high levels of CHOCHO and IO to prove their detection, compare the spectra with the one available in the literature and produce the reference spectra used for the multi-component fit routine. This current work was limited to compare the systems (CP and OP) for indoor and outdoor measurements.
We report here the correlation plots of IO and CHOCHO with NO2 concentrations for outdoor measurements, since these are where the highest NO_2 concentrations are found. There are some minor effects at NO_2 concentrations above 20 ppb, but these remain within the acceptable range for the measurement precision. This proves that there are no important bias in the retrieving of the concentrations of CHOCHO and IO even with high concentrations of NO2.

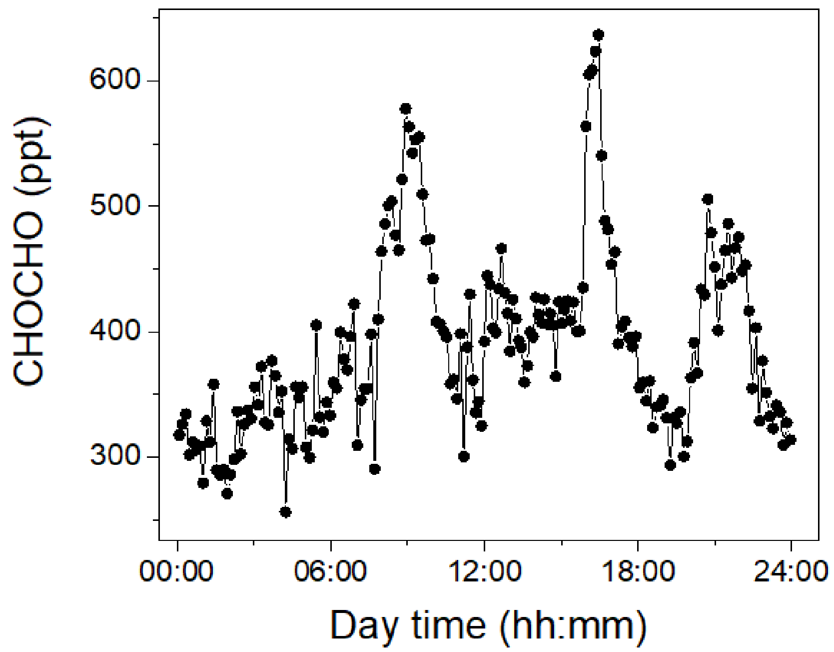




To further convince the reviewer on this point, we report here below an example of the spectrum registered with the open-path system on the 12/10/2023 at a time where NO2 concentration was low, to highlight the low but visible contribution of the CHOCHO absorption spectrum.



Regarding the diurnal cycles, the measurements were conducted in urban air in Grenoble, just outside our laboratory. Therefore, we are quite certain that we should not observe IO in this area and are not expecting to see its diurnal variability. Regarding CHOCHO, the reviewer is right; we did not examine the diurnal signal. We have now conducted this analysis by taking all the outdoor OP data and calculating the average diurnal variability. The graph below shows a diurnal cycle with an amplitude of about 100 ppt. We also observe an increase in CHOCHO during NO_2 peak events in the morning and evening due to urban traffic.



The above graphs will be included in the supplementary material.
We hope that the reviewer is convinced of the reliability of our measurements.

3. I found that the comparison of NO2 measured by CP and OP were not so convincing in outdoor environment especially during the period after Oct 8th. Is the discrepancy resulted from the influence of particles? Maybe the authors should provide some correction algorithm or uncertainty analysis to solve this problem.

There are some instances where there is a discrepancy in NO_2 measurements between CP and OP outdoor measurements (2, 3, 4 and 8/10). Unfortunately, we cannot explain these sporadic events from our data.

4. Technical comments:
    - The citations seem not be well presented in the manuscript. Please use the suitable format.

The format of the citations have been checked and corrected.

- Line 100: Please change “was maid to” to “was made to”.

Corrected

- Line 138: What is the meaning of “Form CHOCHO mean values” here?

The sentence has been rewritten as: “Mean values of -37.6 ± 108 and 388 ± 380 ppt for CHOCHO for the CP and OP system were respectively observed during outdoor measurements.”

- Suggest providing the mirror reflectivity of this instrument for better comparison with other studies.

The reflectivity curve was provided in our previous work (Barbero et al 2020). This information has been added in the manuscript (Line 74).

We thank the reviewer for their very pertinent remarks.