Responses to Referee 1

This is an interesting paper that presents important data and should be published. I have a number of suggestions for improvement, however. Most of my comments are minor.

Thank you very much for your significant and useful comments on the paper "Measurement report: Method for evaluating CO_2 emissions from a cement plant using atmosphere $\delta(O_2/N_2)$ and CO_2 measurements and its implication for future detection of CO_2 capture signals" by Ishidoya et al. We have revised the manuscript, considering your comments and suggestions. Details of our revision are as follows. The line numbers denote those of the revised manuscript.

Don't think you need "Measurement report" in the title

We understand your suggestion, however, I wrote the phrase following the Editor's comment.

The title doesn't sound quite right to me. I think "by atmosphere O2/N2 and CO2 measurements" should be "using atmosphere O2/N2 and CO2 measurements"

Title: We have modified the title, as suggested.

The O2/N2 and CO2 are referred to as "amount fractions", I think it should be either "mole fractions" or "molar fractions".

We recognize "mole fractions" you suggested is more familiar with our research field, however, I have used the phrase following the Editor's comment.

In some places you use "analyses" and I think it should be "analysis". If you're referring to one study, you use the word "analysis." But when you're referring to multiple studies, you use the term "analyses." Also, you use "analyzed" and I think this should be "analysed".

We have changed "analyses" to "analysis" and "analyzed" to "analysed" throughout the paper, following your comments.

Need to be careful when using the words "significantly" & "significant" as this usually implies some sort of statistical test with a p-value. In some places maybe change it to "substantially"

We have changed "significantly/significant" to "substantially" at some places, as suggested.

Lines 16-18: "The simulated CO2 amount fractions were converted to O2 amount fractions by using the respective OR values for each of the incorporated CO2 fluxes, and then simulated OR values were calculated from the calculated O2 and CO2 amount fractions." Rephrase this sentence as it is difficult to follow, its sounds like you used OR values to calculate O2 and then used O2 to calculate OR values

Lines 16-18: The sentences have been rewritten as "The simulated CO_2 amount fractions were converted to O_2 amount fractions by using the respective ER values of 1.1, 1.4, and 0 for the terrestrial biospheric activities, fossil fuel combustion, and cement production. Thus obtained O_2 and CO_2 amount fractions changes were used to derive simulated ER for comparison with the observed ER.".

Line 30: Should probably also include here reference for Pickers et al., 2022, Science Advances
Lines 31-32: We cited Pickers et al. (2022) here, as suggested.

Lines 32-34: These ratios are typically very stable/ tend not to vary very much. The analysis you do later on is only possible because these ratios are so reliable so need to say so somewhere. Also, mention here that 1.4 is global average for fossil fuels, as you use the 1.4 ratio later on.
Lines 37-38: The sentence "The ERs are typically very stable, and the global average ER for fossil fuels is about 1.4 (e.g. Keeling and Manning, 2014)" has been added, considering your suggestion.

Lines 34-36: "Therefore, atmospheric O2 and CO2 fluxes due to terrestrial biospheric activities and fossil fuel combustion (excluding cement production) vary in opposite phase." This sentence needs to be reworded, as respiration in the terrestrial biosphere takes in O2 and releases CO2 which is the same phase as fossil fuel combustion.
Lines 34-36: The sentence has been rewritten as "Therefore, atmospheric O_2 amount fraction varies in opposite phase with CO_2 amount fraction, owing to terrestrial biospheric activities and fossil fuel combustion" to avoid confusion.

Line 39: There is a full stop at the end of the equation which I don't think should be there.
Lines 40: Since our past paper published in ACP, there are some cases we used a full stop at the end of the equation. So, we leave it as it is, but we will revise it if the editorial support team also instructs us to change the format.

Line 43: I think "global fossil CO2 emissions" should be changed to "global fossil fuel CO2 emissions".
Lines 44: The words "global fossil CO_2 emissions" has been changed to "global fossil fuel CO_2 emissions", as suggested.

Line 70: It says that you have been continuously making measurements since 2017. That implies that the measurements are still ongoing. But if that is the case, why are you only using measurements until 19th November 2018? If you have more data for 2019 to 2022 you should include this, as at the

moment you only have 16 months of measurements, and more measurements to base the conclusions on would make the study better.

As you pointed out, the measurements are still ongoing now. However, we consider the data presented in this paper are enough to discuss an effect of cement production. Also, due to our manpower constraint, we cannot calculate the atmospheric CO_2 amount fraction using the fine-scale 3-D atmospheric transport model (AIST-MM) longer time period than the seven months presented in the revised manuscript. Therefore, we leave the observational data period as they are in the revised manuscript.

Methods: In the first paragraph of the methods section, say that the measurement site is on the coast.

Lines 72-73: The words has been modified as "Atmospheric $\delta(O_2/N_2)$ and CO_2 amount fractions have been observed continuously at a coastal station Ryori (RYO: 39° 2′ N, 141° 49′ E, 260 m a.s.l.; Fig. 1)….", to say that the measurement site is on the coast.

Line 86: 1/0.2094 is 4.78 to 2 decimal places and 4.8 to 1 decimal place. I'm not expecting you to change it for this article, because it will make a tiny difference, but in the future, you may want to think about whether you really want to round it to 1 decimal place. I think lots of studies instead of dividing by 4.8, will multiply by 0.2094. This is a wider problem within the O2 community, but we should probably try and have O2 datasets produced using the same method, so different groups can be compared.

Lines 90-91: We understand your concern. In this study, we converted changes in $\delta(O_2/N_2)$ to those in O_2 amount fraction by multiplying by 0.2094 in actual. So, we have rewritten the sentence as "Therefore, observed relative changes in $\delta(O_2/N_2)$ were converted to those in O_2 amount fraction by multiplying by 0.2094 μmol mol^{-1} $(per\ meg)^{-1}$".

Line 96: Should probably say something like, "gaps in the data due to routine calibrations, maintenance and technical issues". I can see from Figure 2 that there is a gap at the end of August/ beginning of September 2017. Might also want to say that there are X number of data points or what percentage of the time period has data.

Lines 109-111: The sentences "It should be noted that gaps in the data seen at the end of August to beginning of September 2017 are due to maintenance and technical issues other than routine calibrations described above. The number of $\delta(O_2/N_2)$ (and CO_2 amount fraction) data points shown in Fig. 2 is 9221" have been added, as suggested.

Line 97: I think you need more detail about the measurement system, instead to referring the readers to another article. Add in a sentence or two summarizing the measurement system. Then you can say "for more information see Ishidoya et al. (2017)".

Lines 92-108: We have added some sentences to describe the measurement procedures in more detail, as suggested.

Line 108: You talk about the CO2 calibration scale but not the O2 scale. In Figure 2 the O2 data is 0 to -300 per meg so you can't be using the Scripps scale. Need to add information about the O2 calibration scale.

Lines 92-95: The sentences have been added to add information about the O_2 calibration scale as "In this study, $\delta(O_2/N_2)$ of each air sample was measured with a paramagnetic analyzer using high- and low-span standard air of which $\delta(O_2/N_2)$ had been measured against our primary standard air (Cylinder No. CRC00045; AIST-scale) using a mass spectrometer (Thermo Scientific Delta-V) (Ishidoya and Murayama, 2014). The scale based on the primary standard air is our original scale, called as "EMRI/AIST scale" in Aoki et al. (2021)".

Line 120: Should probably explain what "clinker" is.

Lines 135-137: To explain what "clinker" is, the sentences have been rewritten as "The CO_2 emissions from the cement plant were estimated from the clinker production capacity of the Ofunato plant in 2018 (Japan Cement Association 2020). The clinker is a solid material produced in the cement manufacture as an intermediary product of Portland cement, mainly consisting of CaO, SiO_2, Al_2O_3 and Fe_2O_3".

Results and discussion: There is only just over 1 year of measurements so need to be careful about the wording when talking about the seasonal cycle. In order to properly investigate seasonal cycles at least a few years of data are needed. So try rephrasing to something like "over one year of measurements CO showed a seasonal cycle with" or "in 2017/2018 the seasonal cycle was", etc.

Lines 189-191: The sentence has been modified as "Over one year of measurements CO amount fractions also showed a seasonal cycle with a summertime minimum that is attributed to the air mass around Japan: in winter the air mass is of continental origin and in summer it is of maritime origin", following your suggestion.

Lines 132-135: It says that O2 exchange is faster than CO2 but I'd actually put the timescales in here, O2 is about a month and CO2 is about a year.

Lines 180-183: The sentence has been modified as "In contrast, the atmospheric O_2 variation (μmol mol^{-1}) due to the air–sea exchange of O_2 is much larger than that of CO_2 on timescales shorter than 1

year because of the difference in their equilibration times between the atmosphere and the surface ocean: the equilibration time for O_2 is about a month and CO_2 is about a year because of the carbonate dissociation effect on the air–sea exchange of CO_2 (Keeling et al., 1993)", following your suggestion.

Line 145: Change "1-week average" to "1-week rolling average".
Line 193: The words "1-week average" have been changed to "1-week rolling average".

Line 153: I think you need to explain more clearly what the ratios tell you. 1.05 to 2.00 indicates terrestrial or fossil fuel. Anything lower than this indicates cement as the 0 ratio mixes with the surrounding air that has already been influenced by terrestrial or fossil fuels pulling down the ratio.
Lines 192-209: Considering your comments and another referee's comments, we have expanded the discussion by adding new figures (Fig. 3b, c). Please confirm the revised manuscript.

Lines 157-160: Why did this study choose these particular 5 months to focus on? Does this mean that not every month has evidence of cement production, or these were the months where the evidence was largest? And if so why do you think this is, there was less cement production taking place at the plant then, or air was coming from the direction of the plant less often?
Every month has evidence of cement production, so that we have recognized that we should carry out calculations by the AIST-MM throughout the observation period. In the revised manuscript, we have added the calculations for January and April 2018. However, due to our manpower constraint, we cannot calculate longer time period than the 7 months.

Lines 157-159: "In October 2017, short-term variations in observed CO2 and d(O2/N2) were opposite in phase, and the amplitudes of some CO2 variations were larger than those of the corresponding d(O2/N2) variations. This result suggests an effect of cement production." I think these two sentences don't join together properly. CO2 & O2/N2 opposite in phase doesn't suggest cement production, CO2 increasing and O2/N2 staying the same would suggest cement production.
Lines 222-226: The sentences have been rewritten to make the meaning clearer as "In October 2017, short-term variations in observed CO_2 and $\delta(O_2/N_2)$ were opposite in phase, and the amplitudes (in $\mu mol\ mol^{-1}$) of some CO_2 variations were larger than those of the corresponding $\delta(O_2/N_2)$ variations (Fig. 5). If the short-term variations driven by terrestrial biospheric activities and the consumption of gas, liquid, and solid fuels, then the amplitudes of CO_2 should be smaller than those of the $\delta(O_2/N_2)$. Therefore, this result suggests an effect of cement production superimposes on fossil fuel combustion and/or terrestrial biosperic activities".

Line 168: Change “land biospheric” to “terrestrial biospheric” as that is what you have used everywhere else.

Line 230: The words “land biospheric” have been changed to “terrestrial biospheric”, as suggested.

Lines 191-192: Used “however” twice in two sentences.

Lines 164-168: We have modified the sentences to use “however” only one time.

Line 208: Change “This means CO2 presumably as well” to “"This means CO2 is presumably released as well".

Line 247: The words “This means CO_2 presumably as well” have been changed to “This means CO_2 is presumably released as well”, as you pointed out.

Line 213: “CO2cement”.

Line 252: The word “$CO_{2cement}$” has been changed to “$y(CO_2, \text{cement})$”.

Summary: Articles usually include something about “next steps”, how the research could be developed in the future.

Also say something about the limitations of the study. Although the limitations can go in the results and discussion section if you think it will fit better there

Lines 319-325: Following sentences have been added to show limitations of the study and suggest next step. “As a remaining topic, we point out the fact that detail variations in the CO_2 amount fraction were not reproduced by the AIST-MM enough. This is due to insufficiency of spatial resolution of the AIST-MM at least partly, to reproduce air transport from a point source such as the cement plant in the present study. Therefore, as a next step, we should use higher-resolution atmospheric transport model to improve an agreement between the observed and simulated CO_2 amount fractions. It is also needed to develop more accurate method to extract $y(CO_2^*)$ due only to cement production especially for the period air-sea O_2 flux is substantial. Such improvement will make it possible to estimate amounts of CO_2 capture and/or CO_2 leak around the observation site from an inversion analysis using the higher-resolution atmospheric transport model”.

Line 284: In the acknowledgments change “observation” to “observations”.

Line 359: The word “observation” has been changed to “observations”, as suggested.

Lots of Figures: Figure 4 and Figure 5 are actually 5 figures each, (a-e) for each of the months. I think this is probably too many figures. Could you try combining them in some way, or choose an example month and move the others to the supplement.

We have chosen an example month and moved the others to the supplement, following your comments.

In Figure 2 and the top panels of the Figure 4's CO2 is in units of μmol mol-1. Isn't this just ppm units, that is what most people are more familiar with?
We recognize "ppm" you suggested is more familiar with our research field, however, I have used the unit following the Editor's comment.

Figure 2 Caption: Change "1-week average" to "1-week rolling average"
Figure 2 Caption: The words "1-week average" have been changed to "1-week rolling average".

Figure 2 Caption: Add something about how the CO2 & O2 y-axes are scaled to be visually comparable or the O2 y-axis is 5 times larger than the CO2 y-axis or something like that.
Figure 2 Caption: The sentence "$\delta(O_2/N_2)$ and CO_2 y-axes are scaled to be visually comparable" has been added, as suggested.

Figure 2: Could you add another panel for the Oxidative Ratio. I know we can see it for some of the individual months but I'm curious to see it for the whole time period.
Figure 3(b): The figure to show the Oxidative Ratio for the whole period has been added as Fig 3b.

The supplement doesn't include any of the model output or the CO measurements.
The CO amount fraction data can be found at the WDCGG. Model outputs of CO_2 amount fraction have been added to the supplement.