Reviewer 1

Summary:

The manuscript by Yoon et al. describes novel measurements using an ECC sensor modified for sensing SO2. The measurement principle is similar to that of the measurements of ozone using potassium iodide solutions.

The authors describe the efficiency of the ozone filter, which is needed to completely remove ozone from the sampled air. They also describe the importance of the dryer, which minimized the removal of SO2 by water in the filter. They show the results of several field campaigns, which indicate the validity of their approach.

The paper is an important contribution for vertical profiling of SO2 and I would recommend publication after some modifications.

Major comments:

Comment 1: The paper refers to Flynn and Morris, which I assume is a reference to a patent. For clarity for readers not as familiar with ECC sondes, I would recommend including a schematic of the ECC and how the bias current is applied.

Response: The authors agree that providing a schematic of the ECC and more clarity in how the bias current is applied would be beneficial to the manuscript. A schematic of the ECC is included in the manuscript as Figure S1 in the supplemental document (P1ln2) and provided below. The schematic helps to visualize the construction of the bias current. Updated text describing the bias current is part of the response to Comment 2.

P2ln6 in Supplemental Data "
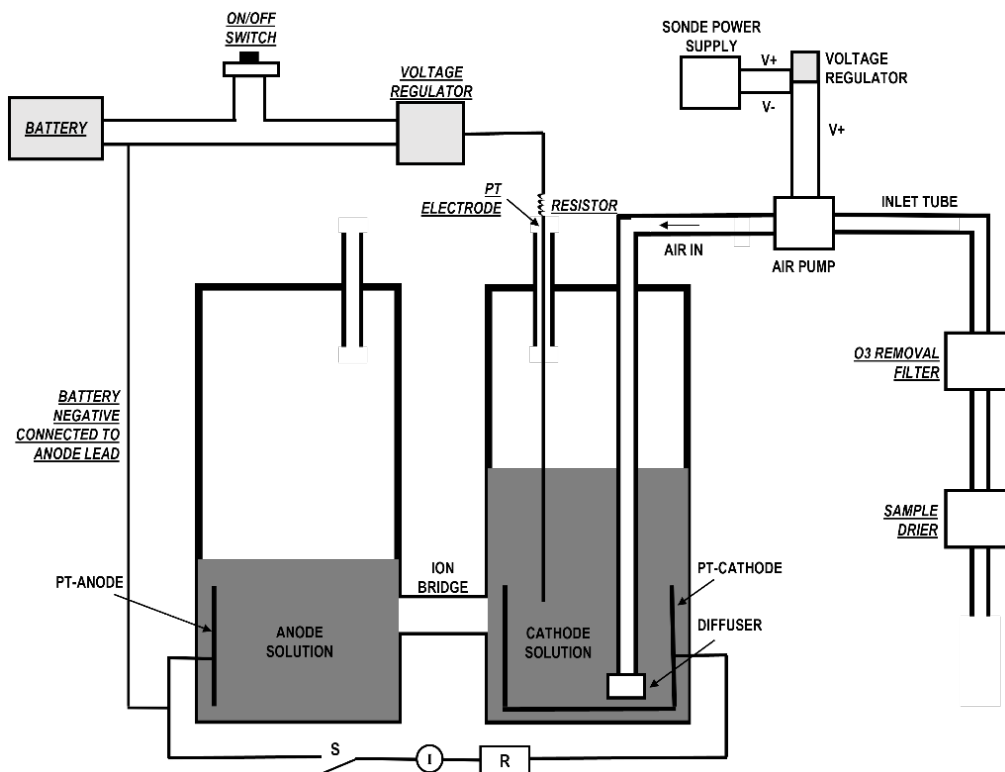
Figure S1: Schematic of the electronic concentration cell (ECC) for the SO$_2$ sonde. All italicized and underlined labels are items that were added to the traditional ECC for the SO$_2$ sonde.[1]"

Comment 2: In the context of this manuscript, the authors should explain the role of the magnitude of the bias current. The authors mention a few times that an instrument was configured for a specific measurement range. The magnitude of the bias current sets the upper detection limit, but this was not very clear. The authors should then point out, what the price is for selecting a high bias current. Figure 7d indicates that selecting a bias current that is too low can still lead to saturation of the sensor. How should the bias current be selected without a priori knowledge of the amount of SO2 that will be measured? Why does a high bias current increase the lower limit of detection?

Response:

The authors agree that a better description of the bias current is necessary including the impacts to the SO$_2$ sonde when the bias currents is too high vs too low. Updates are made to the manuscript and included below.

P5ln122 "The first version of the SO$_2$ system (SO$_2$ sonde v1.0) included the first two modifications: the bias current and an O$_3$ removal filter. The bias current sets the upper limit of detection (ULOD) for the SO$_2$ sonde and is set prior to measurement."

P6ln136 "The bias current is supplied by inserting into the cathode cell an additional platinum electrode powered by a 9V battery (Fig. S1) (Flynn and Morris, 2021). To maintain consistent power, the circuit

uses a 5V regulator. Varying the resistance allows for a range of bias currents to be introduced. The current version of the SO_2 sonde uses a fixed resistor which requires *a priori* knowledge of the desired SO_2 concentration range. The desired resistor is installed in series with the battery and the electrode."

P6ln152 "Examination of the SO_2 sonde data showed that noise was proportional to the measured signal, with 1-σ noise at approximately 0.2 – 0.3% of the measured signal. Because increases in the SO_2 concentrations result in decreases in the signal (i.e., lower cell currents), the magnitude of the applied bias current determines the saturation point (i.e., ULOD) of the SO_2 sonde; saturation occurs when the measured cell current drops to zero. Applying a higher bias current increases the ULOD but also increases noise and the LLOD. The reported LLODs of the bias currents are calculated as 3σ relative to the baseline signal when sampling zero air. During laboratory testing, the LLOD (3σ) was calculated for a range of applied bias currents (0.25 to 10.0 µA). The LLOD for the varying bias current of 0.25 to 10.0 µA ranged from approximately 0.002 to 0.084 µA, respectively. Results of calculated LLOD of a 0.25 µA biased current at varying altitudes (based on a density correction calculation) is included in Table S1. At the surface level, the LLOD of 20s averaged measurements is 0.17 ppbv. The final version of the SO_2 sonde (v1.1) requires the bias current to be selected prior to measurement. If the bias current is set too low, a measurement of larger than expected SO_2 concentrations can saturate the sensor while a bias current that is set too high will have higher LLOD due to the increase in noise. The applied magnitude of the bias current can be best determined based on known SO_2 sources including volcanic emissions, urban and/or industrial emissions."

Comment 3: The authors might also discuss what the uncertainties of their approach are. To what level of confidence can SO2 be measured using this approach, and what factors contribute to the measurement uncertainty?

Response: There are various factors that impact the SO_2 sonde measurement including signal to noise ratio of the applied biased current, SO_2 filter transmission efficiency, SO_2 sonde sensitivity, ambient air pressure, and other sonde parameters including pump speeds, pump flow rate, and time response. The authors have mentioned that building a database of these parameters will provide a quantitative measure of how these different factors impact the SO_2 sonde measurement. The sensitivity calculation is determined as "a regression analysis of the sonde's cell current to the SO_2 concentration measured by a SO_2 analyzer". The sonde-to-sonde variability of measured sensitivity takes into account some of these changing parameters and its impact on the SO_2 sonde measurement. The relative standard deviation of the sensitivity for SO_2 sonde v1.0 (37%) and SO_2 sonde v1.1 (12%). The subject of defining an uncertainty associated with the SO2 measurement will be pursued in future research, following the approach detailed in the GAW #268 for ozonesonde error budget given the similarity in the basic measurement technique.

Comment 4: Line 154: The proprietary O3 filter is not described in the reference. For this publication, the details of the selective O3 filter are essential. Figure 2 seems to indicate that the filter is not 100% efficient in removing ozone. (This Figure does not contain blue lines, even though the legend refers to them. What is meant here?)

Response: The mentioned reference is the patent for the SO_2 sonde system. The reviewer is correct that the patent does not include information on the construction of the O_3 removal filter, but the filter is proprietary, and authors are not willing to disclose the details in this manuscript. However, the authors have reported laboratory test results that provide evidence of the efficacy of the O_3 removal filter (Fig. 2). The test shows removal of approximately 500 ppbv of O_3 and minimal influence on SO_2 concentration

when the sample line is routed through an O$_3$ removal filter (gray). During these periods (gray) the O$_3$ removal filter destroyed O$_3$ to below the detection limit of the O$_3$ analyzer. The figure caption has also been updated.

Comment 5: Line 164ff: This sentence seems to contradict the statements just before and are not supported by the Figures shown. I understand that humidity is the limiting factor, which needs to be better discussed at this point. The supplemental Figure S1 is essential and needs to be included in the main text. It would be best if the authors had a Figure similar to Figure 2 using the dryer. In addition, it would be better to restructure the manuscript and discuss the RH dependence prior to the field deployments. I would suggest merging section 5 with the instrumental description, i.e. make it Section 3.4, then merging the two field deployment Sections into one single field deployment section.

Response: The authors believe the mentioned *"Line 164ff"* sentence is confusing to the reviewer. The *">99.9% efficiency"* is referring to the removal of O$_3$ and not the SO$_2$ transmission efficiency which was discussed in the previous sentence. The sentence has been updated in the manuscript and included below to make this statement clearer.

P7ln178 "Additional testing of the O$_3$ removal filter demonstrated that the filter removed approximately 1 ppm of O$_3$ at sea level with > 99.9% in O$_3$ removal efficiency, concentrations below the detection limit of the Thermo 49*i* O$_3$ monitor."

The authors agree that the discussion of the RH issues and addition of the sample dryer to the SO$_2$ sonde system needs to be discussed earlier in the manuscript. As advised, the discussion of the RH lab testing and sample dryer testing in section 5 has been moved to section 3.4 in the instrument description section. However, authors have kept the field deployment results/discussions into two sections: deployments with an (a) SO$_2$ sonde without and (b) with a sample dryer. This helps highlight the importance of all major components of the final SO$_2$ sonde version. This was also advised by Reviewer #2.

Minor comments:

Comment 6: The authors use the terms "bias current," "biased background current," and "background current" interchangeably. The authors probably just refer to the biasing of the cathode cell, i.e. this is just a "bias current" and not a background.

Response: The authors agree that providing various terms interchangeably can be confusing to the readers. All terms for the applied bias current mentioned as the "bias background current" and "background current" is now referred to as "bias current".

Comment 7: I would suggest defining the sensitivity of the sensor as how many uA of current are generated per incoming ppbv of SO2, which is the inverse of what the authors use. With that definition a higher sensitivity is better.

Response: The authors would like to keep the current sensitivity format as the concentration of SO$_2$ per uA of current generated.

Comment 8: This sensitivity of the sensor depends on some of the same factors as those of the ozone ECC, i.e. pump flow rate and conversion efficiency. In addition, the SO2 sonde has the transmission efficiency of the ozone filter. The chemical conversion efficiency in the cell is probably similar to that of the ozone reactions; and the flow rate is measured. The authors could point out that without dryer, calibrating the filter is essential; in fact, the comparisons they show are exactly that. In addition, this approach must assume that the filter humidity does not change much during the time of measurement. The authors could then strengthen the point, that using the dryer, calibrating the filter is less critical and the stability probably much better. Although this is in the text, the authors could highlight that the decreased need for calibration prior to launch is an essential feature of using the dryer. As suggested above, this could be discussed in a new Section 3.4.

Response: When the O_3 filter is exposed to water, SO_2 adsorbs onto the filter lowering the transmission efficiency (Fig 5). Authors have found that a sample dryer is necessary to produce stable SO_2 conversion efficiency when the filter is exposed to water vapor, even at consistent RH. Our standard operating procedure, with the sample dryer, is to calibrate the SO_2 sonde system to a separate SO_2 sonde analyzer prior to each measurement. The authors believe this pre-flight calibration is essential for making accurate ambient SO_2 measurements due to the novelty of the instrument and to account for potential sonde-to-sonde variability. The authors hope that once a sufficient database of the SO_2 sonde parameters (e.g., pump speeds and filter transmission efficiencies) are recorded, like the traditional ECC ozonesonde, these pre-flight calibrations will not be necessary. The manuscript has been updated to include the importance of pre-flight calibrations and confirm the necessity of the sample dryer for the SO_2 sonde system.

P8ln211 "The sample dryer, therefore, improved both the sensitivity and stability of the measurements observed. The addition of the sample dryer is necessary for providing accurate ambient SO_2 measurements. Pre-flight SO_2 sonde calibrations are also essential for making accurate ambient SO_2 measurements due to the novelty of the sonde system and also addressing potential sonde-to-sonde variability."

Comment 9: Line 266: I am not sure, whether the authors have shown the capability for sub ppbv detection limits. That would require a better uncertainty discussion. Figure 1 indicates that the chemistry exhibits some significant time response. This is not discussed.

Response: The authors have included laboratory test results showing SO_2 sondes (i.e., the biased current) capability for sub ppbv detection limits in the supplementary with reference in the manuscript. The LLOD was calculated as 3 times the standard deviation of the mean of the blank (i.e. zero air). The selected results were for a 0.25 µA biased current. Please refer below to the updated manuscript addressing this comment.

P1ln2 in Supplemental Data

"**Table S1: Results of lower limit of detection calculation (LLOD, 3σ) from laboratory testing of 0.25 µA biased current SO₂ sonde using dry zero air at various altitudes. Various altitudes replicated by controlling pressure of the SO₂ sonde system.**

Altitude, km	0.25 µA		
	LLOD, 1s (ppbv)	LLOD, 20s (ppbv)	ULOD (ppbv)

0	0.21	0.17	12
5	0.35	0.28	19
10	0.62	0.50	35
15	1.32	1.07	74
20	2.89	2.35	163
25	6.46	5.25	363
30	14.10	11.50	796

"

P6ln155 "Applying a higher bias current increases the ULOD but also increases noise and the LLOD. The reported LLODs of bias currents are calculated as 3σ relative to the baseline signal when sampling zero air. During laboratory testing, the LLOD (3σ) was calculated for a range of applied bias currents (0.25 to 10.0 µA). The LLOD for the varying bias current of 0.25 to 10.0 µA ranged from approximately 0.002 to 0.084 µA, respectively. Results of calculated LLOD of a 0.25 µA bias   current at varying replicated altitudes is included in Table S1. At the surface   , the LLOD of 20 s averaged measurements is 0.17 ppbv."

Comment 10: Unless there are additional differences, I would suggest that the authors refer to the SO2 sonde "without dryer" and "with dryer", rather than v1.0 and v1.1. If there are other important differences, then these should be explained.

Response: In the manuscript the distinction between discussing either version is mentioned frequently enough in both the instrumental description and field deployment sections that the authors believe it would be beneficial to have a convenient way (i.e., v1.0 and v1.1) to referring to the specific sonde version. The authors would like to leave the sondes as either v1.0 of v1.1 for the without and with dryer, respectively.

Comment 11: Section 3.2: It would be good to show a diagram of the sonde and of how the cathode cell is biased. It is not clear how the bias current is regulated. The text implies a fixed resistor between battery and electrode, which would make the current dependent on the battery voltage. The text also mentions a voltage regulator. Where is it used? Is the bias current through the cell regulated?

Response: The authors agree that including a schematic of the SO_2 sonde would be helpful in providing a better understanding of how the bias current is applied. The manuscript has also been updated to include clarity in how the bias is applied.

P6ln136 "The bias current is supplied by inserting into the cathode cell an additional platinum electrode powered by a 9V battery (Fig. S1) (Flynn and Morris, 2021). To maintain consistent power, the circuit uses a 5V regulator. Varying the resistance allows for a range of bias currents to be introduced. The current version of the SO_2 sonde uses a fixed resistor which requires *a priori* knowledge of the desired SO_2 concentration range. The desired resistor is installed in series with the battery and the electrode."

Comment 12: In Figure 1, phase F, the SO2 concentration exceeds that of the biased current. Can the authors explain, why the measured cell current does not go to zero? Does that indicate a baseline issue?

Response: The figure and corresponding table have been updated with baseline corrected measurements (i.e., SO_2 sonde, SO_2 thermo analyzer, O_3 sonde, and O_3 thermo analyzer). With the updated results, the measured ECC current in section F is approximately 0.06 µA and equivalent to 2.9 ppbv. This measurement is within the uncertainty range of at least 12% (uncertainty of SO_2 sonde v1.1). This laboratory test was included to highlight the overall success of SO_2 sonde's ability to measure a range of SO_2 contribution from a biased current (approx. 90 ppbv) while also observing minimal impacts of O_3 inclusion.

Comment 13: Line 231: I would say Figure 5 supports a variation of the transmission efficiency of 5%, not 1%.

Response: The "less than 1%" is referring to each test and not the variation amongst all tests. This is better clarified in the updated text which is referenced below.

P7ln199 "With the sample dryer in place, each of the laboratory SO_2 transmission efficiency (May 17-18 and 21, 2018) tests varied by an average of <1% across a range of 0-85% RH (Fig. 6)."

Comment 14: Line 278: Why does the reduction in pressure "significantly" affect the LLOD? Can the authors elaborate?

Response: With increasing altitudes the atmospheric pressure decreases. This decrease impacts sonde parameters (e.g., pump speed, flow rate, etc.), ultimately increasing uncertainty and LLOD. Reported laboratory tests (Table S1) show increasing LLOD when taking account altitude correction. However, for these Hawaii deployments no statistical test was conducted specifically for altitude vs. LLOD so the authors have agreed to exclude this comment.

Technical comments:

Comment 15: In the abstract, the authors refer to a standard deviation of the sensitivity in %. I would have expected the same units, i.e. ppbv/uA. What does this standard deviation refer to?

Response: The reported sensitivity values in the abstract are averages and reported as ppbv/uA. The relative standard deviation is the variability of these averages and reported as a percent. For better clarification we have updated the abstract as indicated below.

P1ln25 "Varying humidity levels affected the SO_2 sonde's sensitivity (avg = 84.6 ± 31.7 ppbv/µA, 1σ RSD = 37%) during initial field tests, which was resolved by adding a sample dryer upstream of the O_3 removal filter and pump inlet. This modification significantly reduced the variability and increased the sensitivity of the SO_2 measurements (avg = 47 ± 5.8 ppbv/µA, 1σ RSD = 12%)."

Comment 16: Line 36f: I assume the authors mean "cooling effect on the surface climate".

Response: The authors have updated text and is included below.

P2ln36 "Gaseous SO_2 can be converted to sulfate aerosols (Zhang et al., 2015), which are highly scattering, reduce visibility, and can have a cooling effect on the surface climate when injected into the stratosphere (Kiehl and Briegleb, 1993; Schmidt et al., 2010)."

Comment 17: Line 49: I assume they refer to "Small UAV". Large UAV, such as Global Hawk, could even measure stratospheric plumes.

Response: The authors will specify the size of the UAV.

Comment 18: Line 87: replace "but" with "which is"

Response: Correction has been made.

Comment 19: Equation 2 is not quite correct.

Response: Correction to equation has been made. Equation updated to:

P4ln92 "$I_2 + 2e^- \rightleftharpoons 2I^-$ (cathode)"

Comment 20: Line 92: Better use "pumped" instead of "diffused".

Response: Correction has been made.

Comment 21: Line 95: Better write "To rebalance the electrochemical potential of the cell ..."

Response: Correction has been made.

Comment 22: Line 99: Delete opening clause and move references to the end of this sentence.

Response: Correction has been made.

Comment 23: Equation 6 should probably only be 1 SO42- instead of 2 SO42-

Response: Correction has been made.

Comment 24: Lines 114 and 115: Do the authors mean averaging over 5 meter or 5 min?

Response: It was averaging over 5 min. We have made the correction of all "m" to be "min" when referring to minutes.

Comment 25: Line 123: The reference to Flynn and Morris (2020) only includes the title and no reference.

Response: The reference has been updated with correct bibliography format.

Comment 26: Lines 135f: Delete "to a signal of", to 90 ppbv add "of ozone". Lines 145f: Better "in a decrease in cell current, ..."

Response: Correction has been made.

Comment 27: Line 157: What do the authors mean by "stepwise dilution"?

Response: The authors have decided to exclude the term as it seems unnecessary.

Comment 28: Line 176: Change "escalated" to "increased"

Response: Correction has been made.

Comment 29: Line 214: Change "may" to "is likely to"

Response: Correction has been made.

Comment 30: Line 247: Was the campaign conducted in Ft. McMurray or Fr. MacKay? I would suggest removing one of the two names unless there was activity in both places.

Response: The campaign was conducted closer to Ft. MacKay. Correction has been updated.

Comment 31: Line 254: Although scientifically irrelevant, I would recommend adding that the LERZ eruption destroyed more than 700 homes in Puna, HI, and displaced thousands of residents.

Response: Section included in the sentence. Now reads:

P10ln271 "The active phase volcanic gas emissions resulted in localized evacuations in the Lower East Rift Zone (LERZ), destroying more than 700 homes and displacing thousands of residents, and resulting in poor air quality for much of the southern and western portions of the island (Tang et al., 2020)."

Comment 32: Lines 286ff: Why do the authors refer to HYSPLIT for the wind direction? Isn't it directly measured by the sondes?

Response: For these SO_2 sonde launch days in Hawaii, additional NOAA HYSPLIT back trajectory analysis was used to understand air mass transport. Instantaneous wind directions measured by the

radiosonde GPS may or may not be indicated of upwind source regions. For the referenced back trajectory analysis, the NOAA HYSPLIT back trajectory has been referenced in the text.

P11ln302 "Typical for the trade winds, NOAA HYSPLIT trajectories (Stein et al., 2015) showed the winds were out of the NE, consistent with the plume's transport from vents in the LERZ or the lava ocean entry points."

Comment 33: Figure 6: The caption to Figure 6 indicates 20 s averaging; the main text indicates 10 meter averaging. Which is it?

Response: The figure caption is correct. The SO_2 data is reporting at an averaging time of 20s. The 0 m averaging reference in the manuscript was an average of the altitude distance of these 20s averaging measurement. The manuscript has been updated to reflect correct averaging time.

Comment 34: The caption indicates the bias current and the lower limit of detection. How was the latter determined?

Response: The lower limit of detection is based on sonde noise when sampling zero air corrected for density. They are calculated as 3 times the standard deviation (3 σ) sampled zero air. Clarification of how the LLOD is calculated is included in the text as:

P6ln156 "The reported LLODs of the bias currents are calculated as 3σ relative to the baseline signal when sampling zero air."