Associate Editor Decision: Publish subject to minor revisions (review by editor) (14 Jan 2020) by Roeland Van Malderen Comments to the Author: Dear authors, Thank you for taking most of the reviewer comments into account. However, some minor revisions and clarifications are still needed.

Especially the discussion of Fig 4 needs to be generalized. As also brought up by the first reviewer, this figure and its discussion only deals with one example of two ECCs, tested at three different weeks. However, in lines 380-381, you wrote that "a number of time-separated calibrations were conducted". So, how many of these experiments have been conducted? And why do you not present the means of those experiments (i.e. average values at 0, 5, 10, 15, 20, 25, 30 mPa of those experiments) at the different weeks, as in Fig. 3, instead of showing just one example? Is the shown example representative for the other experiments as well? Please comment. Furthermore, the discussion focuses on the calibration for 30 mPa, which is, an unrealistic high ozone amount for the stratosphere. Can you also be conclusive for the finding that "the 30 mPa response of the ECCs increases with the week (compared to the reference)" for other ozone partial pressures? Please be more specific and more general.

In this context, your suggestion in lines 404-405 that "On the other hand, the changes could simply be a normal evolution of typical performance behavior" only holds if the illustrated performance is consistent (i) for the different time-separated calibration tested ozonesondes and (ii) for different ozone partial pressure levels (not only for 30 mPa). The same argument holds for the conclusion of this test in the summary section, lines 491-493 ("Results from testing ECC cells over a period of three weeks, one test each week, showed the calibration changed, e.g. about 10 percent for 1.0 percent KI and about 4-5 percent for the 0.5 percent solutions."): it is not clear at all if this conclusion is valid only for the shown example and the numbers are true for all ozone partial pressure levels!

REPLY: We agree that the single example given in Figure 4 is insufficient to suggest ECC calibration increases weekly over a three-week period. We have reviewed the available sets (11) of 'three-weekly' calibrations and found some calibrations did increase but in the average of these data found small week-to-week changes for both the 1.0 and 0.5 percent KI solutions, but these were very small.

Because of the nebulous nature of these results we opted to remove Figure 4 and the applicable text.

Line 44: already mention in the abstract that this study only deals with Science Pump Corp. ECCs

REPLY: Because a one reviewer raised the question to be more specific as to the ECC manufacturer, we prefer for clarity, to maintain the present reference to SPC in the introduction.

Line 72: write out EnSci here.

REPLY: Environmental Science (EnSci) as been added to the text where it is first mentioned. See Section 2.1.

Lines 103-104: As asked by the second reviewer: use a more up-to-date reference for the accuracy of ozonesonde measurements here. E.g. the WMO GAW Report 201 (Smit & ASOPOS Panel 2014) gives an estimate (perhaps also Deshler et al., 2017; Thompson et al., BAMS, 2019).

REPLY: As suggested, text was changed indicating reference change.

Lines 119-130: provide the years at which the digital calibration benches were/are operationally used at Wallops, Payerne and Nairobi

REPLY: Text added showing Payerne since 1995, Nairobi since 2018, Wallops Island (development 2005-2008; operational 2009-2017-only used for preparation, no calibration).

Line 139: Write out TEI here.

REPLY: Added Thermo Environmental Instruments (TEI) where it first appears in Section 2.1

Lines 240-243: please check the order of filling the cells. If at Wallops, like you wrote it down, the anode cells are filled before the cathode cells, the SOPs are not followed, which is a major issue for the conclusions reached.

REPLY: Text has been corrected.

Lines 288-289: "Generally, the downward calibration experiences small differences from the upward calibration". In which sense? Consistent for all measurements? Related to the sensor response and consequently its memory of higher vs. lower ozone amounts?

REPLY: We have added text indicating that the measured partial pressure during the downward calibration is consistently higher for both 1.0 and 0.5 percent KI solutions. Apparantly, the ECC sensor retains the memory of experiencing high ozone concentration.

Lines 325-327: "The final background currents often were somewhat higher than background currents experienced with manual preparation, generally about 0.04 microns" Please specify which final background current you mean here (final background current obtained prior to balloon release??) and

give a possible reason why these background currents are higher with the automated procedure than with the manual procedure. Is this because of the linear calibration step in the automated procedure?

REPLY: Preparation with the digital bench obtains the final background current after experiencing the high ozone concentration of the calibration step (0-30 mPa). We expect that the residual memory of the ECC sensor is the reason for background currents higher than the manual. The text has been changed to reflect this effect. The manual value is obtained just prior to balloon release.

Section 3.2: you should mention in the beginning of this section that the 1.0% 1.0 B solution strength is the recommended one for Science Pump (e.g. Smit & ASOPOS, WMO GAW report 201, Deshler et al., 2017).

REPLY: Text added reflecting this.

Lines 365-367: Please repeat here the explanation given by Johnson et al. (2002) about the effect of different KI solution concentrations and the side effects from the buffers used, within the context of your 1.0%1.0B & 0.5%0.5B SPC comparison.

REPLY: Text has been added reflecting explaination from Johnson et al. (2002).

Line 436: The profiles were averageD

REPLY: Fixed. Thank you.

Lines 436-443: what about the total ozone normalization factors of the 12 dual flights? Which solution strengths are closer to the co-located Brewer/Dobson or satellite overpass total ozone measurements?

REPLY: Normalization is not done at Wallops Island. Dobson total ozone compares very well with the 0.5 percent KI solution. ECC 1.0 and 0.5 percent KI total ozone vs. Dobson total ozone has been added to the paper, e.g., the sample of 12 profil- mean DU for the ECC 1.0 % is 330.4 DU, ECC 0.5 % 308.3 DU, and 309.5 DU for the Dobson.

Lines 456-460: what about the total ozone normalization factors of the three dual flights? Which solution strengths are closer to the co-located Brewer/Dobson or satellite overpass total ozone measurements?

REPLY: Reference to dual-flights was removed. The authors considered the amount of information too slim.

Fig 04: I guess the units in these figures should be ranging from 0 to 30 mPa instead of 0 to 300?

REPLY: Figure 4 was removed from paper.

Fig 06: I guess the legend should show the 0.3% KI in red, instead of 0.5% KI. As in Fig. 3, you might also list the standard deviations

REPLY: Correction has been made and the std dev added. New figure added.

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7	An Automated Method for Preparing and Calibrating
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34 In contrast to the legacy manual method used to prepare, condition, and calibrate the Electrochemical 35 Concentration Cell (ECC) ozonesonde an automated digital calibration bench similar to one developed 36 37 by MeteoSwiss at Payerne, Switzerland was established at NASA's Wallops Flight Facility and provides reference measurements of the same ozone partial pressure as measured by the ECC. The 38 39 purpose of an automated system is to condition and calibrate ECC cells before launching on a balloon. 40 Operation of the digital calibration bench is simple and real-time graphs and summaries are available to 41 the operator; all information is archived. The parameters of interest include ozone partial pressure, 42 airflow, temperature, background current, response, and time (real and elapsed). ECC cells, prepared with 1.0 percent solution of potassium iodide (KI) and full buffer, show increasing partial pressure 43 44 values when compared to the reference as partial pressures increase. Differences of approximately 5-6 45 percent are noted at 20.0 mPa. Additional tests with different concentrations revealed the Science Pump Corp (SPC) 6A ECC with 0.5 percent KI solution and one-half buffer agreed closer to the reference 46 47 than the 1.0 percent cells. The information gained from the automated system allows a compilation of ECC cell characteristics, as well as calibrations. The digital calibration bench is recommended for ECC 48 49 studies as it conserves resources. 50

Abstract

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1. Introduction 52 53 54 Measurement disagreement between similar or identical instruments seems to be an historical problem. 55 Intercomparisons are generally conducted when new instruments are introduced and when operational changes or improved procedures become available. Such comparisons should be made under the same 56 57 environmental conditions and include a reference instrument as an aid for checking the accuracy and 58 reliability of the instruments. This would be ideal as a standard procedure. Unfortunately, balloon-borne ozone reference instruments are not usually available, mostly because they are too expensive for other 59 60 than occasional use or to expend on non-recoverable balloon packages. Ozonesonde pre-flight 61 calibrations are conducted, however these are basically single point calibrations made prior to its 62 release. An automated system designed to condition and calibrate the Electrochemical Concentration Cell (ECC) ozonesonde was fabricated at Wallops Flight Facility. The automated system can condition 63 the ECC prior to flight and, if desired, provide calibration over a wide range of ozone partial pressures. 64 This system, designated the digital calibration bench, enables consistent conditioning and calibration of 65 the ECC along with measurements of a reference value. In this paper the term ECC refers only to the 66 Science Pump Corp. (SPC) 6A ECC ozonesonde, although the automated system can accommodate the 67 68 Environmental Science (EnSci) ozonesonde as well. 69 70 There are a variety of ground-, aircraft-, satellite-, rocket-, and balloon-borne instruments available to 71 measure the vertical structure of atmospheric ozone and its total content. These instruments operate on different principles of measurement (Fishman et al, 2003; Kohmyr, 1969; Krueger, 1973; Holland et al, 72 73 1985; Hilsenrath et al, 1986; Sen et al, 1996). Although their spatial distribution is limited, balloonborne Electrochemical Concentration Cell (ECC) ozonesondes have had a key role as a source of truth 74 75 for the other instruments and for establishing algorithms necessary for the retrieval of satellite observations. Manual preparation of the ECC requires hands-on contact by an operator. 76 77 Reducing subjectivity is important and was considered serious enough to engage in the fabrication of 78 79 the automated system. The user is prompted throughout the calibration process while utilizing real-time graphs and summaries. The digital calibration bench provides consistent preparation procedures. ECC 80 81 measured ozone partial pressures vs. reference partial pressures are discussed and the results 82 corroborated with dual ECC comparisons at Wallops Island. During implementation of the digital 83 calibration bench, beta testing provided the dual ECC measurements used in this paper for demonstration purposes. Operational use at Wallops Island was intermittent and only provided a limited 84 number of ECC preparation records between 2009 and 2017, when bench components began to fail. 85

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Notwithstanding efforts to enhance ECC performance (Smit et al, 2004, 2007, 2014; Kerr et al, 1994; 89 90 Johnson et al, 2002; Torres, 1981) there remain uncertainties. The accuracy of the ECC is estimated at Deleted: Barnes (1982) and Barnes et al (1985) estimated t Deleted: s 91 5-10 percent and also varies with altitude (Deshler etr al, 2017; Smit and ASOPOS Panel, 2014), Deleted: pointed out that the accuracy 92 However, standardization of ozonesonde preparation methods has improved and better data quality Deleted: ed 93 control (Smit et al, 2014) and the homogenization of the ozone data (Deshler et al, 2017; Smit et al, Deleted: 94 2013) have raised the level of ozonesonde usefulness. Uncertainties also arise from poor compensation 95 for the loss of pump efficiency; erroneous background current; variable motor speed; solution loss from turbulent cathode cell bubbling; air flow temperature error and whether measured at the proper location; 96 97 and, the use of the appropriate potassium iodide (KI) concentration. Understanding the influence these 98 parameters have on the ozonesonde measurement capability is particularly important. The digital calibration bench is able to measure these parameters in a consistent way over a range of partial 99 100 pressures. 101 2 Digital Calibration Bench Description and Operational Procedure 102 103 104 2.1 Description 105 106 The computer-controlled preparation and calibration bench fabricated at NASA Wallops Flight Facility 107 borrows from the design of a bench developed by MeteoSwiss scientists B. A. Hoegger and G. Levrat 108 at Payerne, Switzerland. The MeteoSwiss digital calibration bench was first available in 1995 and Deleted: the 1990's continues to be used and is updated periodically. The MeteoSwiss and Wallops digital calibration 109 110 benches are functionally similar but are not identical in design. A comparable bench furnished by Deleted: also 111 MeteoSwiss to the meteorological station at Nairobi, Kenya has been in use since 2018. The Wallops Deleted: for a number of years 112 Island ozone site was interested in the digital bench because of its capability to provide precise and 113 repeatable preparation of ECC's, and its automated feature requires less interaction with the ECC then the manual preparation method. The Wallops Island digital bench was undergoing development 114 115 between 2005-2008 and used operationally only to prepare ECC's between 2009-2017. 116 117 Throughout the history of ECC ozonesonde performance, the concentration of the KI solution has been 118 questioned (Thornton and Niazy, 1982; Barnes et al, 1985; Johnson et al, 2002; Sterling et al, 2018). In 119 the late 1960's and early 1970's the recommendation to use 2.0 percent solution was unchallenged. In 120 the mid-1970's the concentration was changed to 1.5 percent, and in 1995 the KI solution was changed once more to 1.0 percent. Employing the Wallops digital calibration bench enables adjustment of the 121 datasets obtained with the different concentrations to be homogenized to improve the consistency of the 122 123 measurements of the long-term database. The digital calibration bench allows consistent, computercontrolled preparation of ECC instruments. The calibration bench accurately measures the ozone reaching the ECC cells while a Thermo Environmenmtal, Inc. (TEI) ozone generator provides the source of ozone at partial pressures between 0.0 and 30.0 mPa. A second TEI instrument accurately measures the ozone sent to the ECC, providing a reference value. Thus, performance comparisons are possible without expending costly instruments.

The Wallops digital calibration bench, shown in Fig. 1, consists of three major components: 1) mass flow meter to control air flow, 2) an ozone generator and analyzer (UV photometer), and 3) computer necessary to automate the timing of the programmed functions and process the data. Another important component, the glass manifold, enables the simultaneous distribution of the air flow to the ECC's and the UV photometer. The manifold also is a buffer maintaining constant air flow and inhibiting flow fluctuation. A graphical user interface controls the various input and output functions using an interface board and communications portal enabling synchronous communication protocols. A signal conditioning box allows connections to the ECC's analog signals that are conditioned with custom electronic components. Minor but necessary components include pressure and temperature sensors, and valves and solenoids to direct the flow of laboratory grade air. Calibration validity is accomplished by comparing the measured ECC ozone partial pressure against a reference partial pressure obtained with the UV photometer (TEI Analyzeer).

Fig. 2, from an unpublished technical note (Baldwin, private communication), illustrate the steps necessary to achieve a consistent calibration. By following the sequential flow diagram shown in Fig. 2, upper panel, the operator can better understand the sequence of tests. Each shape in the diagram is associated with a graphical window displayed on the monitor, as are notices that pop-up to instruct or direct the operator. The computer controlled digital bench follows the ECC preparation procedure in place at NASA Wallops Island at the time of the system's fabrication. Each ECC is recognized by its manufacturing date and serial number and includes the manufacturers test data. Changes to the steps are possible anytime through software reprogramming. The preparation sequence begins by verifying whether ECC cells are new or were previously conditioned. A different path is followed for either condition. New cells are flushed with high ozone prior to manually adding KI solution. Cells previously having had solution added skip over the high ozone step to determine the first background current. Following the first background check the remaining steps are completed. Other measurements accumulated with the digital bench include motor voltage, motor current, pump temperature, and linear calibration at seven levels (0.0-30.0 mPa). Program steps are displayed on the computer monitor with real-time information. All data are archived and backup files maintained.

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Fig. 2, lower panel, illustrates the functional diagram detailing the essential operation of the digital 170 171 calibration bench. Software control is shown in blue and air flow in green. Laboratory zero-grade dry 172 air or desiccated compressed air is introduced into the TEI ozone generator where a controlled amount 173 of ozone is produced. The ozone flows simultaneously to the ECC cells and to the TEI Model 49C 174 ozone analyzer. The analyzer contains the UV photometer that provides the reference partial pressure. 175 176 The digital bench reads the air flow from a Hasting mass-flow meter permitting a precise flow rate to be determined. The mass-flow is then converted to volume-flow by the conventional conversion formula. 177 178 The volume flow rate measurement was found to be comparable to the flow rate determined with the 179 volumetric bubble flow meter. The digital calibration bench uses the Hasting Mass-Flow Meter model ENALU with a HS500m transducer with a maximum mass-flow-range of 500 [scc/min].. In contrast, 180 the manual method uses a stop watch to estimate when 100 mL of air has flowed into a chamber. An 181 experienced operator, using a volumetric bubble flow meter is able to measure the time to less than 1 182 second. Tarasick et al (2016) points out that the operator uncertainty when reading the bubble flow 183 meter is about 0.1-0.3 percent. Further, the manual method requires that the effect of moisture from the 184 185 bubble flow meter's soap solution be accounted for; flow rates determined with the digital calibration 186 bench do not require a correction for moisture. Unfortunately, the calibration bench cannot determine 187 the pump efficiency correction (PEC); this is taken into account differently. For a number of years, the ECC's PEC was physically measured at Wallops Island using a specially adapted pressure chamber 188 189 (Torres, 1981). This system is no longer available. However, from its many years of use an extensive 190 number of measurements are available. A sample of 200 pressure chamber measurements were averaged to obtain a unique PEC that was adopted for use at Wallops Island. 191 192 After eliminating deficiencies and improving functionality the automated system was tested while 193 obtaining research data, primarily comparisons between different KI solution concentrations. 194 Calibration from 0.0 mPa to 30.0 mPa generally exceeds the nominal range of atmospheric ozone 195 partial pressure. Calibration steps are made in 5.0 mPa increments but larger or smaller increments are 196 197 possible with minimal software reprogramming. Differences between ECC and reference measurements, if seriously large, provide an alarm to possibly reject the ECC, or after further study the 198 199 differences between the ECC and reference calibration might be considered as a possible adjustment

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factor that would be applied to observational data.

2.2 Operational Procedure

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ECC preparation procedures at Wallops Island are carried out five to seven days prior to preparing the ECC for flight. The pump, anode and cathode cells, and Teflon tubing are flushed with high amounts of ozone to passivate their surfaces and is followed by flushing with zero-grade dry air followed by filling of the cells. The cells are stored until ready to be used.

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Operation of the automated system is simple, requiring only a few actions by the operator that include obtaining the first background current, air flow, 5 μ A or high ozone (170 nb) test, response test, second background current, linear calibration between 0.0 mPa and 30.0 mPa, and the final background current. As indicdated in Fig. 2, upper panel, two cells can be conditioned nearly simultaneously. i.e., the program alternates measurements between ECC's.

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The operator must first determine whether the cell being conditioned had already been filled with KI or never was filled. Whatever the status of the cell (wet or dry) the operator enters the identification information before proceeding. When a new, or a dry cell is to be processed the digital calibration bench initiates high ozone flushing. The program alerts the operator to turn on the high ozone lamp after which V3 of Fig. 2, lower panel, is switched to high ozone. The unit checks that ozone is flowing and after 30 minutes the program switches to zero air for 10 minutes and V3 switches back to the ozone generator. When completed, the operator is prompted by an instructional message on the monitor screen to fill the anode and cathode cells with the proper concentrations of potassium iodide (KI) solution, i.e., the cathode cell is filled first with 3 mL of 1.0 percent KI solution followed, after a 10 minute delay, by filling the anode cell with a saturated KI solution. The cells are stored until ready for further conditioning and calibration before being used to make an observation. Considering that the ECC cell had been filled earlier with solution the digital bench instruction bypasses the high ozone flushing. Ozonesonde identification is entered, as above. The operator, after fresh KI has been added to the cell, is prompted on the monitor screen to begin the first background current measurement. In either case, whether a dry cell for which flushing is complete, or a wet cell ready for calibration, the procedure starts with clicking the OK button displayed on the monitor screen. After 10 minutes of dry air the background current is recorded. The background current record contains the following information: date, time in 1-2 second intervals, motor current, supplied voltage, pump temperature, and cell current. As the measurement is being made identical information is displayed graphically on the monitor. Following the background test all further steps are automatic.

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Continuing to follow the steps outlined in Fig. 2, upper panel, the measurement of the air flow is accomplished on one ECC pump at a time by switching V1, shown in Fig. 2, lower panel, to the mass flow meter and at the same time V2 is switched to the glass manifold (ozone generator). When completed, V1 is switched back to the glass manifold and V2 is switched to the flow meter and the flow rate of the second cell is carried out. The air flow is output in sec/100 ml. The information stored includes: date, time in seconds at intervals of 7-8 seconds, mass flow meter temperature, atmospheric pressure, flow rate, and supply voltage.

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Measuring the response of the ECC to ozone decay requires setting the ozone generator to produce 17.0 mPa ozone partial pressure (approximately 5 uA). As ozone is produced the ozone level increases until the set level is reached. The elapsed time to reach this level is noted. The 17.0 mPa of ozone is the reference level used to initiate the response test. After recording 17.0 mPa of ozone for 10 minutes the ECC response check begins. To measure the response, the cells would have to be switched to zero air quicker than the cell responds. This is accomplished by switching both cells (assuming two cells are being calibrated) to the mass flow meter, the source of zero air. This is more efficient than setting the generator to zero and waiting for the manifold and residual ozone in the system to reach the zero level. Thus, V1 and V2 of Fig. 2, lower panel, are switched to the mass flow meter for immediate zero air and the program triggers a timer. The decreasing ozone is measured and recorded at five points used to reflect the cell response. As the ozone decays, measurements at 3-4 second intervals provide a detailed record of the response while also being displayed real-time on the monitor. From the detailed record the program selects five points (1, 2, 3, 5 and 10 minutes) successively that are used to calculate the response of ozone change that should be 80-90 percent lower than the reference of 17.0 mPa. V1 and V2 are switched back to the ozone generator and the next 10-min background current measurement begins. The response record contains the following: date, time in seconds, motor current, supply voltage, temperature, mass flow, cell current, and atmospheric pressure. Data are displayed on the monitor in real-time.

The ECC cells have been conditioned and are ready for the linear calibration. The 0.0 mPa to 30.0 mPa calibration is performed. Step changes begin with 0.0 mPa, followed by measurements at 5.0, 10.0, 15.0, 20.0, 25.0, and 30.0 mPa. Each step requires approximately 2-3 minutes to complete allowing time for the cell to respond to each ozone step change. The linear calibration includes the reference measurement made simultaneously with the ECC measurement. After the upward calibration reaches the 30.0-mPa level the calibration continues downward, to 0.0 mPa. The measurements are displayed on the monitor for the operators use and also sent to an Excel file. Generally, the downward calibration experiences small differences from the upward calibration. The available test data reveals that the downward calibrations are always higher than the upward calibrations. Between 5.0 mPa and 25.0 mPa the downward calibrations of the 1.0 percent KI solution are 0.8 mPa to 1.0 mPa higher than the upward calibration. The 0.5 percent solution downward calibration varies between 0.5 mPa and 0.9 mPa for the same partial pressures. Only the upward calibrations are used. Following the linear calibration, the final background current is obtained. This requires 10 minutes of zero grade dry air before making the measurement. The data are recorded in a summary file that contains the supply voltage, motor current, flow rate, pump temperature, response, and the background currents.

3 Digital Calibration Bench Practical Application

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Repetitive comparison operations can be carried out with the digital calibration bench as often as necessary. This could result in a potential cost saving as there would be no need to expend radiosondes,

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ECC's, and balloons. The testing with the digital calibration bench is limited to the ranges of pressures 297 and temperatures at sea level and would be an imprecise representation in the upper altitudes. 298 299 300 3.1 Digital Calibration Bench (General) 301 302 Quasi-simultaneous testing of two ECC's is possible, enabling comparisons of different concentrations 303 of KI solutions. Comparison of 2.0-, 1.5-, 1.0-, and 0.5- percent KI concentrations were carried out on 304 the digital bench demonstrating that agreement with the ozone reference value improved with lower 305 concentrations. In an earlier paper Johnson et al (2002), using SPC and EnSci ECC's demonstrated 306 similar changes occurred when testing various solution concentrations that also included varying amounts of buffer. Only the SPC 6A ECC's with 1.0 percent KI solution and full buffer (1.0%,1.0B) 307 308 and 0.5 percent KI solution and one-half buffer (0.5%,0.5B) concentrations are discussed here. 309 310 During the checkout of the digital calibration bench ECCs ondes were calibrated in pairs and included different KI solutions. Tests indicated the pressure and vacuum measurements were nominal, some 311 312 insignificant variation occurred but was not a cause for concern. Pump temperatures, controlled by the 313 room air temperature, varied 0.1°C to 0.2°C. Motor currents showed some variation, some measured 314 over 100 mA, suggesting a tight fit between the piston and cylinder. For example, one ECC motor current initially was 100 mA, a second measurement a week later the reading was 110 mA, a final 315 reading after running the motor for a short time was 96.5 mA. Flow rates fell within the range of 27 to 316 317 31 seconds per 100 ml, a range comparable to flow rates manually measured with a bubble flow meter. 318 Background currents were consistent. The lowest background current allowed by the digital bench is 319 0.0044 µA. The final background currents obtained with the digital bench often were somewhat higher 320 than background currents experienced with manual preparation, generally about 0.04 µA. Although 0.4 321 μA is relatively small it is possible the higher background current value results from the ECC's 322 residual memory following exposure to the high ozone concentration during the previous linear 323 calibration step. The final background currents, obtained manually immediately prior to an ECC Deleted: F Deleted: s 324 balloon release, were in the range between 0.01 and 0.02 μA. Finally, the response of all the cells was Deleted: was 325 good, falling within the required 80 percent decrease within less than one minute. Graphically checking Deleted: were 326 a small sample of high-resolution responses found some variation as the ozone decayed, Deleted: necessary Deleted: decreased 327 Deleted: to 0.0 mPa 328 3.2 Calibration and Potassium Iodide (KI) Solution Comparisons **Deleted:** The linear calibration (0.0-30.0 mPa), is useful for comparing different KI concentrations 329

As a practical example of the usefulness of the digital calibration bench is its capability to nearly

simultaneously obtain measurements from two ECC's, one prepared with (1.0%,1.0B) and the second

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341 with (0.5%,0.5B). The recommended KI solution strength to be used with the SPC 6A ECC's is 1.0 342 percent the with full buffer (Smit and ASOPOS PANEL, 2014). Conditioning of the ECC's followed 343 the steps given in Fig. 2, upper and lower panels. In the free stratosphere ozone partial pressures usually 344 range from 15.0 mPa to 20.0 mPa. Linear calibrations to 30.0 mPa are obtained, although a lower range 345 may be reprogramed. 346 347 Figure 3 is a graphical example of differences between the reference ozone measurement and the measurements of (1.0%,1.0B) and (0.5%,0.5B) KI concentrations. A sample of 18 digital bench 348 Deleted: Rather than showing the differences from a single measurements were averaged to provide a representative set of differences. The close proximity 349 Deleted: give 350 between the curves shown in the figure render the standard deviation lines too small, also they overlay Deleted: more Deleted: Standard deviations have been added to the 351 each other to some extent. The standard deviations have been added to the figure for greater clarity. The figure. 352 variations, although small, indicate greater variability with the (1.0%,1.0B) KI solution. Fig. 3 suggests Deleted: and Deleted: also that the two concentrations measured nearly identical amounts of ozone between 0.0 mPa and 8.0 mPa. 353 Deleted: text Both curves begin to separate and diverge above 8.0 mPa. The averaged data at 10.0 mPa indicate that 354 Deleted: exists 355 (1.0%, 1.0B) is 0.36 mPa, or 3.6 percent higher than the reference and (0.5%, 0.5B) is 0.04 mPa, or 0.4Deleted: nb percent higher; at 15.0 mPa the difference is 0.67 mPa, or 4.3 percent and 0.17 mPa or 1.1 percent Deleted: nb 356 357 higher, respectively; at 20.0 mPa the difference for (1.0%,1.0B) is 1.11 mPa, or 5.5 percent and 358 (0.5%,0.5B) is 0.48 nb or 2.4 percent higher, A check at the 30.0 mPa level indicated (1.0%,1.0B) was Deleted: , respectively 6.8 percent above the reference and (0.5%,0.5B) was 3.2 percent above. The ECC with (0.5%,0.5B) KI 359 concentration is closer to the reference than (1.0%,1.0B) KI. Both ECCs' partial pressure curves have a 360 slope greater than 1 trending toward higher amounts of ozone when compared to the reference value as 361 362 ozone partial pressure increases. It is clear that the (1.0%,1.0B) KI solution increases at a faster rate than the (0.5%.0.5B) solution. Johnson et al (2002) have explained the effect of different KI solution 363 364 concentrations as well as the side effects from the buffers used. Their study of the standard (1.0%, 1.0B) Deleted: in some detail 365 solution indicated the ECC can report higher ozone amounts, up to 5-7 percent under constant ozone 366 conditions and can also increase the ozone amount to higher values from the buffer reactions. Fig. 3 **Deleted:** The intent of the examples is merely illustrative of the advantage provided by the digital bench to examine 367 indicates that the 1.0 percent KI measurement is further from the reference than the 0.5 percent KI. The ECC behavior. Further, Deleted: while t 368 percentage difference between the two KI concentrations is virtually constant at 3.2 percent, or in terms of a ratio between the two solutions, 0.968. Referring to the SPC ozonesondes compared during 369 370 BESOS, Deshler et al (2017, Fig.5 and Table 2) indicate non-linearity between the (0.5%, 0.5B) and Deleted: s 371 (1.0%,1.0B) KI solutions and similar ratio values, 0.970/0.960. 372 373 The digital calibration bench turned out to be an ideal tool to obtain repeated ECC calibrations. The 374 digital bench can calibrate two ECC's nearly simultaneously reducing the need to expend costly dual-375 ECC balloons. A negative aspect, possibly, is that calibration at sea level cannot provide knowledge of Deleted: r

ECC behavior under upper altitude conditions. Eleven ECC pairs were calibrated over a period of three weeks. Two ECC's were prepared with (1.0%,1.0B) and (0.5%,0.5B) KI solutions. A number of time-separated calibrations were conducted with the expectation the resulting calibrations would be repeatable week-to-week. The cells were flushed and fresh KI solutions were used with each weekly test. Calibration over the full range, 0.0-30.0 mPa was carried out, Changes that might be due to improper preparation and conditioning procedures were not considered since, by definition, the digital bench is consistent in how ECC's are prepared. Consideration also must be given to the fact that the ECC sensor has a memory that may have an effect of inhibiting repeatability. The individual weekly calibrations showed varying results. Some calibrations showed an increase each week while other calibrations did not. An average of the data showed small increases week-to-week but these were too small to be significant. In essence no particular pattern was evident suggesting that calibrations on a week-to-week schedule would not be repeatable.

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To bring the ECC measurements into correspondence with the reference suggests that downward adjustment should be applied to each curve. When a large sample of similar digital bench measurements are obtained it should be possible to design a table of adjustments relative to ozone partial pressure that could be used to adjust ozonesonde measurements. However, since the calibrations are made at sea level such an adjustment table would not be able to account for the influence of upper atmospheric pressure and temperature. Nevertheless, any adjustment, seemingly, would be in the right direction and would aid in obtaining more representative ozone values.

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Although digital bench calibration comparisons are instructive, important comparisons have been made between ECC's and reference instruments using other methods. ECC measurement comparability have been quantified through in situ dual instrument comparisons (Kerr et al, 1995; Stubi et al, 2008; Witte et al, 2019), laboratory tests at the World Ozone Calibration facility at Jülich, Germany (Smit et al, 2004, 2007, 2014) and by occasional large balloon tests such as BOIC (Hilsenrath et al, 1986), STOIC (Kohmyr et al, 1995) and BESOS (Deshler et al, 2008). BESOS provided important performance information about the SPC 6A ECC and the EnSci ozonesondes. However, these complicated large balloon experiments that seem to occur every 10 years are expensive. The environmental chamber used in the Jülich tests (Smit et al, 2007) covers a full pressure range but is also expensive to use. The purpose here is to show a calibration method that is simple to use and provides calibrations that include useful reference values, and is complementary to other methods, such as employed in the Jülich Ozone Sonde Intercomparison Experiment (Smit et al, 2004; Smit et al, 2007).

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Deleted: only one three-week test is shown in Fig. 4a, b, c. The result shown is characteristic of similar calibrations performed over a similar number of weeks.

Deleted: but only the calibration at the 30.0 mPa partial pressure is discussed. During the first week, Fig. 4a, the (1.0%,1.0B) KI solution was approximately 2.1 mPa, or 7 percent higher than the corresponding reference value. The (0.5%,0.5B) KI solution was about 0.6-0.7 mPa or about 2 percent lower than the reference value. A second calibration one week later, designated week two in Figure 4b, showed the ECC with the (1.0%,1.0B) KI solution had moved further away from the reference, about 2.7-2.8 mPa or 9 percent higher (approximately 0.6-0.7 mPa higher than during week one), while the ECC with the (0.5%,0.5B) KI was now 1.2 mPa or 4 percent higher than the reference. A third calibration, week three in Fig. 4c, showed both ECC calibrations had moved again. The (1.0%,1.0B) KI calibration increased an additional 0.2 mPa and was now about 3.0 mPa, or 10 percent higher than the reference. The ECC with (0.5%,0.5B) KI increased an additional 0.1 mPa and now was 1.3 mPa, 4 percent higher than the reference value. Providing an explanation for the changes observed between week one and week three is difficult.

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In the 1998-2004 period the Wallops ozone station released a number of dual-ECC balloons, twelve pair successfully provided measurements to 30 km, and higher. The ECC's were attached about 35 meters below the balloon and each ECC separated a distance of 2 meters. Each pair was composed of an ECC with (1.0%,1.0B) and (0.5%,0.5B) KI solutions. The profiles were averaged, and are displayed in Fig. 4. It can be noted in the figure that the mean (0.5%,0.5B) solution reveals less ozone being measured than that of the (1.0%,1.0B) solution. Near the 65-70 hPa level the (0.5%,0.5B) ECC begins to report increasingly Jess ozone with increasing partial pressure than the (1.0%,1.0B) ECC. A similar feature was noted in Fig. 3 where the separation of the ECC's with different concentrations occur with increasing partial pressure. Fig. 4 shows the maximum ozone partial pressure level was about 14.0 mPa, near 22 hPa, where the (0.5%,0.5B) KI solution measured approximately 1.0 mPa, or 7 percent less ozone than the ECC with the (1.0%,1.0B) KI concentration. This difference is approximately 4 percent higher than the result given by the digital calibration bench results of Fig.3, where, at 15.0 mPa, the difference between the (1.0%,1.0B) KI and (0.5%,0.5B) KI is 3.2 percent. Dobson measurements of total ozone compared with total ozone derived from each of the ECC profiles used to the obtain the average profiles shown in Fig. 4 were, on average, in excellent agreement with (0.5%,0.5B). The total ozone difference between the Dobson (309.5 DU) and (1.0%,1.0B) (330.4 DU) is 20./9 DU; between the Dobson and (0.5%,0.5B) (308.3 DU) was 1.2 DU.

Given that the digital bench tests revealed the (0.5%,0.5B) KI solution is in close agreement with the reference measurement than the (1.0%,1.0B) solution suggested that a KI solution with a weaker concentration may, possibly, give closer agreement. A small number of dual ECC tests were carried out with a solution of 0.3 percent with one-third buffer (03%,0.3B). Six sets of ECC's were prepared for calibration. Each dual ECC test consisted of one ECC prepared with (1.0%,1.0B) KI solution and one with (0.3%,0.3B) KI solution. The digital bench comparison result disclosed the (1.0%,1.0B) result replicated the earlier results discussed above. As assumed, the lower concentration was nearly equal to, or slightly less than the reference. Average values and standard deviations derived from the six tests are shown in Fig. 5. Although the 0.3 percent solution might appear to be a better choice additional tests are necessary.

4 Summary

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509 510 The concept of an automated method with which to pre-flight condition and calibrate ECC ozonesondes was originally considered by MeteoSwiss scientists over 20 years ago. Drawing on their expertise, a facility designated as the digital calibration bench was fabricated at NASA Wallops Flight Facility between 2005-2008. The digital bench was put to use immediately to study ECC performance, conduct

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Deleted: To corroborate the bench results three balloonborne dual ECC sondes were flown, each with 1.0 and 0.3 percent KI solutions. Unhappily, the results were inconclusive: one flight showed (0.3%,0.3B) to be higher than (1.0%,1.0B), a second flight showed it to be lower, and the third flight showed (0.3%,0.3B) to be nearly the same value. ...

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comparisons of different KI concentrations, enabled ECC repeatability evaluation, as well as calibrating 532 the ECC over a range of partial pressures, including associated reference values. Tests conducted with 533 the digital bench were performed under identical environmental conditions. The digital bench 534 535 eliminates the expense and time associated with making similar tests in the atmosphere. 536 Early use of the digital bench was to calibrate ECC's, prepared with (1.0%,1.0B) KI solution, over a 537 538 range of partial pressures from 0.0 mPa to 30.0 mPa. Comparison between ECC's with (0.5%,0.5B) and (1.0%,1.0B) KI solution and simultaneously obtained reference values revealed the two KI solution 539 540 strengths were measuring more ozone than the reference. There was an increasing difference between 541 the ECC's and the reference as the partial pressure increased. For example, the ECC measurements slope upward to increasingly larger differences from the reference ozone measurements, i.e., increasing 542 543 from 4.3 percent higher partial pressure at 15.0 mPa (Fig. 3) to about 7 percent higher at 30.0 mPa. 544 Results from the digital bench also corroborate differences found between SPC 6A ECC'c flown on 545 dual-instrument flights at Wallops Island. The difference between ozonesondes at a pressure of 22 hPa 546 showed the (0.5%,0.5B) ECC to be about 1.0 mPa lower than the (1.0%,1.0B) ECC. 547 548 549 The digital calibration bench provides a capability to apply a variety of test functions whereby the valuable information gathered helps to better understand the ECC instrument. Evaluating SPC ECC 550 performance using an automated method diminishes the requirement for expensive comparison flights. 551 552 The tests performed, i.e., KI solution differences, calibrations over a time period, and dual-instrumented 553 balloon flights, were consistent, giving similar results. The tests described in this paper are simply examples of the utility of the digital bench. Furthermore, the digital calibration bench preparation 554 555 facility potentially could contribute to an understanding of separating ECC measurement variability 556 from atmospheric variability. Thus, the automated conditioning and calibration system provides valuable information, and as a useful tool should continue to be a valuable aid. 557 558 559 5 Data Availability 560 Data are available from the authors. 561 562 6 Author Contribution 563 The first author acquired and prepared the data for processing and the second author was instrumental in certifying the digital calibration bench was working properly. Both contributed equally to manuscript 564 565 preparation.

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Deleted: An instruments ability to repeat the same measurement is important, however, ozonesondes are used only one time. (There are exceptions when an occasional instrument is found and returned, but, unfortunately because of Wallops Island's coastal location nearly all sonde instruments fall into the Atlantic Ocean rendering them unfit to be reclaimed). The digital bench provided the opportunity to obtain repeatable calibrations of the ECC. Results from testing ECC cells over a period of three weeks, one test each week, showed the calibration changed, e.g., about 10 percent for 1.0 percent KI and 4-5 percent for the 0.5 percent solution.

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7 Competing Interests 581 582 The authors declare they have no conflict of interest. 583 584 585 8 Disclaimer 586 587 None 588 589 9 Acknowledgments 590 We acknowledge the successful use of the digital calibration bench to the skillful efforts of Gilbert Levrat (retired) of the MeteoSwiss site Payerne, Switzerland for his foresight in designing the original 591 592 bench and its simplicity. We are indebted to Tony Baldwin (retired) of NASA Wallops Flight Facility 593 for his electronic skill and programming expertise and to .E. T. Northam for assistance preparing the figures. We also appreciate the insightful suggestions given by the referees; they were instrumental in 594 helping us make the paper better. 595 596 597 10 References 598 Barnes, R. A., Bandy, A. R., and Torres, A. L.: Electrochemical Concentration Cell ozonesonde 599 600 accuracy and precision, J. Geophys. Res., Vol. 90, No. D5, 7881-7887, 1985. 601 602 Deshler, T., Mercer, J. L., Smit, H. G. J., Stubi, R., Levrat, G., Johnson, B. J., Oltmans, S. J., Kivi, R., Thompson, A. M., Witte, J., Davies, J., Schmidlin, F. J., Brothers, G., and Sasaki, T.: Atmospheric 603 604 comparison of electrochemical cell ozonesondes from different manufacturers, and with different 605 cathode solution strengths: The Balloon Experiment on Standards for Ozonesondes, J. Geophys. Res., 606 113, D04307, https://doi.org/10.1029/2007JD008975, 2008. 607 608 Deshler, T., Stubi, Rene, Schmidlin, Francis J., Mercer, Jennifer L., Smit, Herman G. J., Johnson, 609 Bryan J., Kivi, Rigel, and Nardi, Bruno,: Methods to homogenize electrochemical concentration cell 610 (ECC) ozonesonde measurements across changes in sensing solution concentration or ozonesonde manufacturer, Atmos. Meas. Tech., 10, 2021-2043, https://doi.org/10.5194/amt-10-2021-2017, 2017. 611 612 Fishman, J., Wozniak, A. E., and Creilson J. K.: Global distribution of tropospheric ozone from satellite 613 measurements using the empirically corrected tropospheric ozone residual technique: Identification of 614

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686	11 Figures	 Delete	d: ¶		
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688	Fig01. Illustration of the digital calibration bench showing operational configuration and mounting	 Delete	d: D		

691	position of two ECC ozonesondes. The major components include ozone generator and analyzer,		Deleted: instrumentation
692	computer, flow meter, and glass manifold.		Deleted: s
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694	Fig02. Digital calibration bench diagrams: a) sequential steps, and b) functional steps.		Deleted: showing
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696	Fig03. Simultaneous comparison of ECC ozonesondes prepared with (1.0%,1.0B) [blue] and		
697	(0.5%,0.5B) [red] KI solution concentrations. The reference curve is shown in black. Calibrations are		
698	made in 5.0 mPa steps from 0.0 mPa to 30.0 mPa.		
699			
700 701	Fig04. Average ozone profiles from 12 pairs of SPC 6A ECC ozonesondes indicating at the 22 hPa pressure level that the (0.5%,0.5B) ECCs' measured 0.7-0.8 mPa less ozone, approximately 5 percent		Deleted: Fig04. Calibrations of two ECC ozonesondes, one using 1.0 percent KI solution and the other 0.5 percent KI, over a three week period.
702	less, than the (1.0%,1.0B) ECCs'.	(/////	Deleted: 5
703			Deleted: a
704	Fig05. Digital calibration bench results between (1.0%,1.0B) solution, blue curve, and (0.3%,0.5B)	-	Deleted:
l 705	solution, red curve; the reference curve is shown in black.	/ ///	Deleted: ,
703	solution, red curve, the reference curve is shown in black.	1/1	Deleted: ,
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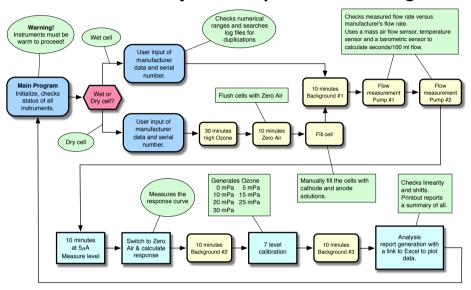
DIGITAL CALIBRATION BENCH

The system consists of a computer, mass flow meter, TEI 49C ozone generator, TEI 49C ozone analyzer, and incidental equipment.

The TEI generator and analyzer are calibrated each month using a primary standard 3-meter long-path photometer.

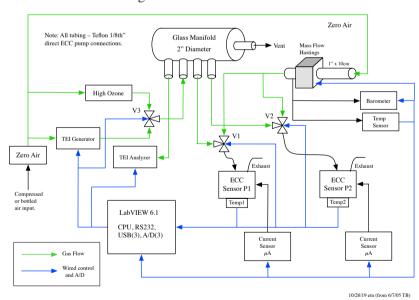
738 Fig 02.

ECC Calibration System Sequential Flow Diagram



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Functional Diagram Ozonesonde Calibration Test Bench



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