

Abstract

In situ measurements of atmospheric ozone (O_3) are performed routinely from many research aircraft platforms. The most common technique depends on the strong absorption of ultraviolet (UV) light by ozone. As atmospheric science advances to the widespread use of unmanned aircraft systems (UASs), there is an increasing requirement for minimizing instrument space, weight, and power while maintaining instrument accuracy, precision and time response. The design and use of a new, dual-beam, polarized, UV photometer instrument for in situ O_3 measurements is described. The instrument has a fast sampling rate (2 Hz), high accuracy (3%), and precision (1.1×10^{10} O_3 molecules cm^{-3}). The size (36 l), weight (18 kg), and power (50–200 W) make the instrument suitable for many UAS and other airborne platforms. Inlet and exhaust configurations are also described for ambient sampling in the troposphere and lower stratosphere (1000–50 mb) that optimize the sample flow rate to increase time response while minimizing loss of precision due to induced turbulence in the sample cell. In-flight and laboratory intercomparisons with existing O_3 instruments show that measurement accuracy is maintained in flight.

1 Introduction

Ozone is a key atmospheric trace gas that is produced and transported in both the troposphere and stratosphere and is present in mixing ratios ranging from parts per billion to parts per million. Ozone in the stratosphere protects the biosphere from harmful UV radiation from the sun and ozone in the free troposphere plays a major role in atmospheric oxidation. In the lower troposphere, ozone is a pollutant that is harmful to plants and ecosystems (e.g., Ashmore, 2005) and has negative impacts on human health (e.g., Mudway and Kelly, 2000).

A number of different techniques are commonly used for in situ O_3 measurements in environmental research, including: UV absorption (Bowman and Horak, 1974; Maier

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is an increased need for airborne instruments that are lighter, smaller, and require less power to operate, while maintaining the performance specifications of larger contemporary instruments.

We describe a new ozone UV photometer, designated here as NOAA-2 O₃, specifically built for use on high- and low-altitude research aircraft. The new photometer was successfully operated on board two research aircraft, the NASA Global Hawk UAS and the NASA WB-57F high-altitude aircraft. The new instrument represents an example of how size, weight, and power can be reduced without loss of instrument performance. The following sections describe the instrument and sampling inlet designs along with performance results from laboratory and flight testing. The instrument performance tests include intercomparisons with a previously built NOAA ozone instrument, described by Proffitt and McLaughlin (1983) and designated here as NOAA-1 O₃. This instrument has flown on hundreds of flights on board the NASA ER-2 and other high-altitude research aircraft since 1985.

2 Instrument description

2.1 General description

The NOAA-2 O₃ photometer design illustrated in Fig. 1 is a derivative of the dual-beam, unpolarized, UV absorption technique described by Proffitt and McLaughlin (1983). Briefly, ambient and O₃-scrubbed airflows are alternately directed into two identical absorption cells. 253.7 nm UV light from a mercury lamp is split into two beams that are each directed into one of the absorption cells. Since O₃ strongly absorbs photons at 253.7 nm, the UV beam passing through the cell containing ambient ozone is attenuated more than the beam passing through the cell containing O₃-scrubbed air. Knowing the O₃ absorption cross section (σ) and the absorption path length (L), the O₃ partial pressure (p_{O_3}) can be derived using Beer's law. For a given optical system, the instrument precision generally increases with increasing L . For photometers designed

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for airborne measurements, L is typically 40 cm or less, and is limited in part by the allowable instrument overall dimensions.

2.2 Optical configuration

In the new photometer design, a novel optical system was developed that folds the UV beam inside the absorption cells and, thereby, doubles the absorption path length for a given physical cell length. An optical-isolator type configuration is utilized to fold the UV beam inside the absorption cells (see Fig. 1), giving an effective 60 cm absorption length with a 30 cm cell. The cell length in the NOAA-1 O₃ instrument is 40 cm. In this configuration, the unpolarized output of a mercury lamp (see Sect. 2.3) is collimated by a lens and passes through a 254 nm bandpass filter before being vertically polarized by a polarizing beam splitter (PBS1). The resulting polarized beam is then split into two beams using a non-polarizing polka-dot beam splitter (BS) and mirror (M1) combination, with half of the light entering each of two absorption cells unimpeded through a polarizing beam splitter (PBS2 and PBS3, respectively). On the distal end of each cell, the light is reflected and the polarization rotated by 90° using a precision quarter-wave plate and mirror combination (WP1, M2 and WP2, M3). After the return pass through the absorption cells, the now horizontally polarized light is reflected upward into a silicon photodiode by entrance polarizing beam splitter (PBS2 and PBS3).

A reduced cell length reduces the required physical size of the instrument for a given precision. In addition, the shorter cell of this double-pass design has the advantage of a shorter sample-air residence time inside the cells, which increases the allowable sampling rate for a given sample flow. Furthermore, the folded optical path allows an optimally compact design by locating all the significant optical and electronic components on only one end of the cell. The new instrument specifications are listed in Table 1. A photo of the instrument without its flight enclosure is shown in Fig. 2.

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2.3 UV lamp

The NOAA-1 O₃ instrument uses a Pen-Ray mercury lamp (UVP, LLC Upland, CA). In the NOAA-2 O₃ instrument, linearly polarizing the UV beam from the lamp reduces by one half the UV flux available in each absorption cell. As a consequence of the use of polarized beams and an increase in L (60 cm vs. 40 cm), the Pen-Ray lamp does not provide sufficient photon flux to achieve a similar detection limit. Instead, the new instrument uses a capillary mercury lamp (Jelight Company, Inc., Irvine, CA, Part No. 3CPG-1). An aperture ($d \sim 0.5$ mm) blocks stray light, creating an effective UV point source. With a custom power supply, this lamp consumes approximately 6 W of power and provides a useable photon flux over 1 order of magnitude greater than the Pen-Ray lamp.

2.4 Sample flow control

The NOAA-2 O₃ instrument does not use active pumping to pull sample air through the absorption cells. Instead, ram pressure (about 20–40 hPa) at the sampling inlet opening created by the aircraft movement at 100–200 ms⁻¹ pushes ambient air through the instrument from the inlet opening to the exhaust opening. As shown in Fig. 1, the inlet flow is split with half passing through an O₃ scrubber unit and half remaining unscrubbed. Three-way valves (V1 and V2) serve to alternate the scrubbed and unscrubbed flows in each absorption cell every 10 s.

The flows exiting the absorption cells are merged into a common exhaust line. A volumetric flow meter (Pneumotachograph Type 0, Phipps and Bird Fleisch, Richmond, VA) is used for measuring the total merged flow downstream of the absorption cells. Flow regulation is achieved using a custom butterfly vane valve (V3) with an ID of 9.53 mm based on the design by Gao et al. (1999).

For an instrument with an emphasis on high precision and fast time response, careful flow control is very important. Sufficient flow is needed to ensure the time required to completely flush the absorption cell volume is the same or smaller than the time

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differential across the instrument. The advantage of the configuration in the top panel of Fig. 3 is that debris such as dirt, insects or hydrometeors with sufficient inertia are prevented from turning the corner and entering the sample line to the instrument. This keeps the instrument cell windows and walls clean, thus maintaining instrument performance. The disadvantage of this configuration is that it is more prone to create turbulence in the sample line and cells, which leads to a slight degradation of instrument precision.

2.6 Catalytic ozone scrubber

As shown in Fig. 1, the sample flow is split into two, and one of the flows passes through the O₃ scrubber unit before entering one of the absorption cells. The unit shown in Fig. 4 was specifically designed and constructed to catalytically destroy O₃ in the flow while not causing a significant pressure drop compared a sample line of the same length. A large pressure drop can lead to unbalanced flows in the two absorption cells and therefore degraded instrument time resolution. As shown, the sample air stream entering the scrubber is expanded and slowed to increase reaction time. Brass membranes containing a variety of holes are located at the entrance and exit to ensure near plug flow of the sample air inside the scrubber. Catalytic reactions occur on a total of 20 manganese oxide screens, taken from commercial catalysts (www.thermoscientific.com, Part #14697) that are stacked uniformly in the middle of the scrubber volume. The exposed screen diameter is 51 mm. The scrubber unit was tested by passing air with 6 ppm O₃ through it at 14 LPM at pressures down to 50 mb. The output of the scrubber was sampled with the NOAA-2 O₃ photometer which showed no measurable O₃ (<0.5 ppb) for several hours of operation.

2.7 Temperature control

O₃ artifacts can also be caused by temperature gradients along or between the absorption cells. To ensure the best performance, the entire volume of the instrument

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is temperature controlled. As shown in Fig. 2, two 50 W heaters are mounted on aluminum heat exchangers placed directly in front of two of three circulation fans inside the instrument flight enclosure (not shown). The circulation warms and cools components to achieve a uniform operating temperature. Temperature controllers using sensors placed upstream of the fans control enclosure air temperatures to 300 ± 1 K, or $\pm 0.3\%$. A 1 mm thick aluminum enclosure serves as a thermal barrier from ambient temperatures that vary depending on the location of the instrument. When the instrument is used in unheated and unpressurized aircraft payload locations, a 3 mm thick Nomex insulation felt lines the inside surfaces of the enclosure as additional thermal insulation. Sample air is heated before entering the absorption cells as it flows through an 80 cm segment of Teflon[®] tubing (11.4 mm OD) located inside the enclosure on top of instrument in Fig. 2.

2.8 Pressure sensor

Absorption cell pressure is needed to calculate the O₃ mixing ratio from O₃ partial pressure. During a flight, the cell pressure fluctuates in response to changes in ambient pressure. An accurate measurement of the cell pressure is therefore critical for accurate and precise O₃ mixing ratio data. The pressure sensor chosen must be independent of ambient pressure in the payload location. An Esterline 78005001A (Esterline Advanced Sensors, Farnborough, England) vibrating-cylinder pressure sensor was used based on its excellent stability, accuracy, precision, temperature compensation, and the lack of sensitivity to ambient pressure changes. Frequent intercomparisons with NOAA laboratory standards showed that its measurements were stable and accurate over 2 yr to approximately ± 0.5 hPa, or $< \pm 1\%$.

3 Instrument performance

3.1 Precision

Since the NOAA-2 O₃ instrument measures the O₃ molecular density, the instrument precision is expressed in units of molecules cm⁻³. The intrinsic instrument precision is determined by noise in the optical system, signal digitizer, and pressure and temperature sensors. The intrinsic precision was determined with data recorded with no sample flow through the instrument as illustrated in Fig. 5. As shown, the typical value of the intrinsic precision is about 5×10^9 molecule cm⁻³.

With a folded light path, the O₃ partial pressure measurement is twice as sensitive to sample flow turbulence as a single-path configuration. To optimize instrument performance, flow-induced noise was recorded as a function of pressure and sample flow. Figure 6 shows a typical measurement at a fixed flow rate of 20 LPM. Based on these measurements, the flow controller was programmed to control the sample flow in flight at 27 LPM for cell pressure <200 hPa, 15 LPM with $200 \leq$ cell pressure <500 hPa, and 10 LPM with cell pressure \geq 500 hPa. At these flow rates, the flow-induced noise is always smaller than the intrinsic precision measured with no flow. Note that at flow rates of 15 and 10 LPM the minimum O₃ measurement time resolution lengthens to approximately 1 s and 2 s, respectively.

For atmospheric sampling, additional noise generally arises from pressure fluctuations related to the sample inlet configuration and aircraft electronic interference. An upper limit of in-flight precision can be determined by examining measurement statistics in 250 s flight segments with low O₃ variability. As an example, data collected on-board the NASA Global Hawk UAS with the simple forward-facing inlet (Fig. 2, lower panel), an ambient pressure of 110 hPa and a sample flow near 27 LPM yielded an in-flight 1- σ precision of approximately 1.1×10^{10} molecule cm⁻³ or approximately twice the intrinsic precision.

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The precision can be greatly affected by the inlet configuration. On board the Global Hawk UAS with the inlet configuration using the side-facing Teflon[®]-lined tube (Fig. 2, upper panel), an ambient pressure of 80 hPa and a sample flow near 25 LPM yielded an in-flight 1- σ precision of approximately 1.9×10^{10} molecule cm^{-3} or approximately four times the intrinsic precision. The additional noise is attributed to small sub-Hertz oscillations in the O₃ partial pressure values.

3.2 Laboratory accuracy

Since the NOAA-2 O₃ instrument is based on the same principles described by Proffitt and McLaughlin (1983), their detailed error analysis can be directly applied here. Briefly, the largest error sources are absorption length, O₃ absorption cross section at 253.7 nm, pressure and temperature of sample air inside absorption cells, and O₃ loss to the wall material. In our new instrument, a high quality pressure sensor and good overall temperature control ensure the temperature and pressure errors are well less than 1 %. Temperatures measured at several positions inside the instrument vary within ± 1 K of 300 K, or about $\pm 0.4\%$, during flight. All Teflon[®] wetted surface construction and fast sample flow prevent measurable ambient O₃ loss inside the instrument. Error in absorption cell length determination is also less than 1 % (3 mm). Overall accuracy is $\pm 3\%$, the same as given by Proffitt and McLaughlin (1983) without wall loss.

As a further assessment of accuracy, the NOAA-2 O₃ instrument was intercompared in the laboratory with the NOAA-1 O₃ instrument and a commercial primary standard (Model 49i-PS, Thermo Electron Corporation (TECO), Franklin, MA). All of these instruments are based on the same UV absorption technique. The TECO O₃ instrument has an accuracy specification of ± 1 – 2% . It is treated at NOAA as a primary standard using sample flows that are restricted to dry synthetic air. The laboratory setup for intercomparing two instruments is shown in Fig. 7. The typical set of results for the NOAA-2 O₃ and the TECO instruments in Fig. 8 shows that the instruments agree well with each other with the correlation slope being 1.01 over a wide cell-pressure range. A similar slope is found with another TECO instrument and with the NOAA-1 O₃

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instrument, indicating that the NOAA-2 O₃ has a systematic positive bias of 1 % with respect to the TECO primary reference measurement. Although the cause of this bias is unknown, the NOAA-2 O₃ atmospheric measurements are adjusted downward by 1 % to be consistent with the other three instruments.

In conducting the intercomparison, care was taken to ensure that instrument leaks did not influence the outcome. Since many parts such as the flow switching valves and absorption cells are made of Teflon[®], leaks are always a potential threat to the O₃ measurements. Both NOAA-1 and NOAA-2 O₃ instruments are designed for aircraft operation in pressurized or unpressurized payload areas. In the latter, leaks are generally not an issue whereas large leaks in a pressurized environment could potentially alter the O₃ measurements. We note that any leak of air with less O₃ than in the sample air occurring into one of the absorption cells or further upstream usually causes a negative bias in the measurement. Leaks further downstream do not affect the measurements. To ensure no leaks are large enough to affect the measurements, we routinely run the NOAA-1 and NOAA-2 O₃ instruments at various sample pressures using the laboratory setup shown in Fig. 7. As shown, the sample pressure (P_1) for the TECO O₃ is always at a pressure slightly above ambient pressure. Partial control of P_1 is provided by the all-Teflon[®] valve, V_1 . An above-ambient value of P_1 ensures that no leaks can occur into the TECO instrument and cause errors in the measurements. The pressure in the NOAA instrument being intercompared is controlled by another valve, V_2 , and a pump. If there is no significant leak in the NOAA instruments, then the results should be independent of P_2 , as demonstrated in Fig. 8.

3.3 Artifact response

A negative artifact has been observed under some conditions when the sample humidity abruptly changes from wet to dry (Meyer et al., 1991; Kleindienst et al., 1993; Wilson and Birks, 2006). This artifact appears to be related to water on the cell walls and optical elements, affecting the transmission of the unfocused UV beam. When the

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humidity is reduced abruptly, water degassing from the catalyst causes an imbalance in cell humidity and, thereby, an artifact response. This degassing and artifact response will in general be strongly dependent on composition and surface area of the catalyst material (and possibly cell optical components). For the catalyst and optical components used in both NOAA O₃ instruments, this artifact is small. For example, when O₃-scrubbed ambient air (~1 % water) is replaced with dry synthetic air (<10 ppm water), both instruments show a negative bias of <10 ppb at 820 hPa that drops to ~2 ppb after 6 min and continuously decreases to ~0.2 ppb over 30 min. This artifact is mostly pressure independent and, therefore, is relatively more important at lower absorption cell pressures and lower O₃ values. An important feature of the NOAA instruments is that the water artifact diminishes when O₃ is present in the dry air after the humidity change in the sample flow. Specifically, when the O₃ density level is above about 130 ppb at ~125 hPa, the artifact is completely unobservable. This feature, together with the relatively fast decay of the artifact, means that on an aircraft ascending quickly from near sea level to the upper troposphere or flying out of a high humidity in a cloud, the NOAA instruments will likely not show a significant O₃ artifact.

3.4 Field intercomparison

In March and April 2011, the NOAA-1 and NOAA-2 O₃ instruments flew together on board the NASA WB-57F aircraft based in Houston, TX as part of the NASA Mid-latitude Airborne Cirrus Properties Experiment (MACPEX) mission. The aircraft extensively sampled the UT/LS at altitudes up to 18 km, with O₃ mixing ratios reaching values of about 1 ppm. The two instruments agreed very well during all flights with the aforementioned –1 % adjustment to the NOAA-2 O₃ data. Data from a typical flight are shown in Fig. 9. As shown, the agreement between the two instruments is well within 1 %. The NOAA-2 O₃ data shown in Fig. 9 are 2-point averages, matching the 1 s data rate of the NOAA-1 O₃. The intercomparison of precisions is not particularly meaningful due to different inlet configurations. As it is, the NOAA-2 O₃ has a slightly better precision with the 2-point average.

4 Conclusions

A new in situ instrument for atmospheric O₃ mixing ratio measurements, NOAA-2 O₃, was developed and tested in the laboratory and field. The instrument builds upon the UV-absorption technique used by the long-standing NOAA-1 O₃ instrument, which has flown hundreds of flights in the UT/LS. The NOAA-2 O₃ instrument departs from earlier designs by employing polarized UV beams in the absorption cells in order to fold the optical path, which leads to a reduction in the physical cell length required for a given precision. A capillary Hg lamp was used to increase the available UV intensity. Laboratory tests with an O₃ primary standard instrument confirm an accuracy of better than 2%. Flight tests on the NASA WB-57F and Global Hawk UAS platforms showed that laboratory precision and accuracy are maintained in flight. In comparison to the NOAA-1 O₃ instrument, the NOAA-2 O₃ instrument has an improved sampling rate (×2), a better intrinsic precision (by 25%), reduced weight (by 30%) and reduced payload volume (by 43%), while maintaining a similar accuracy (< ±3%) and power consumption. The sampling rate of 2 Hz of the NOAA-2 O₃ corresponds to a horizontal resolution of 100–200 m and a vertical resolution of 5 m or less, respectively, at typical research speeds. These features represent a substantial improvement in the feasibility and desirability of integrating the NOAA-2 O₃ instrument on board UAS platforms, which generally have less payload capacity, as well as on manned aircraft platforms.

Acknowledgements. This research was supported by the NOAA Atmospheric Composition and Climate Program, the NASA Radiation Sciences Program, and the NASA Upper Atmosphere Research Program. We thank K. K. Kelly for insightful discussions.

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Table 1. Specifications of the NOAA O₃ instruments.

Parameter	New NOAA-2 O ₃ instrument	NOAA-1 O ₃ instrument
Sampling rate	2 Hz	1 Hz
Accuracy	3%	3%
Precision with sample flow	1.1×10^{10} O ₃ molecules cm ⁻³	1.5×10^{10} molecules cm ⁻³
Size	48 cm × 34 cm × 22 cm	76 cm × 46 cm × 18 cm
Weight	18 kg	26 kg
Power (28 VDC)	~ 200 W (peak)*; ~ 50 W (normal)	~ 200 W (peak)*; ~ 50 W (normal)

* A large portion of power is used for achieving (peak) and maintaining (normal) an adequate internal temperature during an aircraft flight in the UT/LS. The normal power consumption depends strongly on the instrument ambient temperature conditions.

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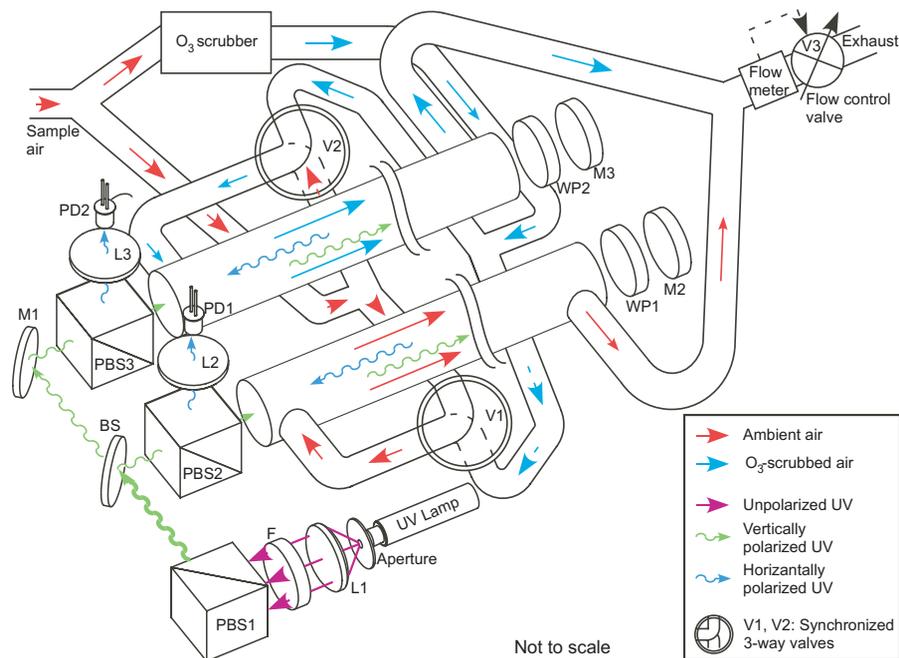


Fig. 1. Schematic of the new NOAA-2 O₃ instrument showing the configuration of the key optical and air flow components: Ln: lens; F: UV filter; PBSn: polarizing beam splitter; BS: polka-dot 50–50 beam splitter; Mn: mirror; WPN: quarter-wave plate; PDn: silicon photodiode detector; V1 and V2: synchronized 3-way flow-control valves; V3: flow-control valve. Schematic is not to scale. The legend indicates how the polarizations of the UV beams, and scrubbed and unscrubbed sample air are illustrated.

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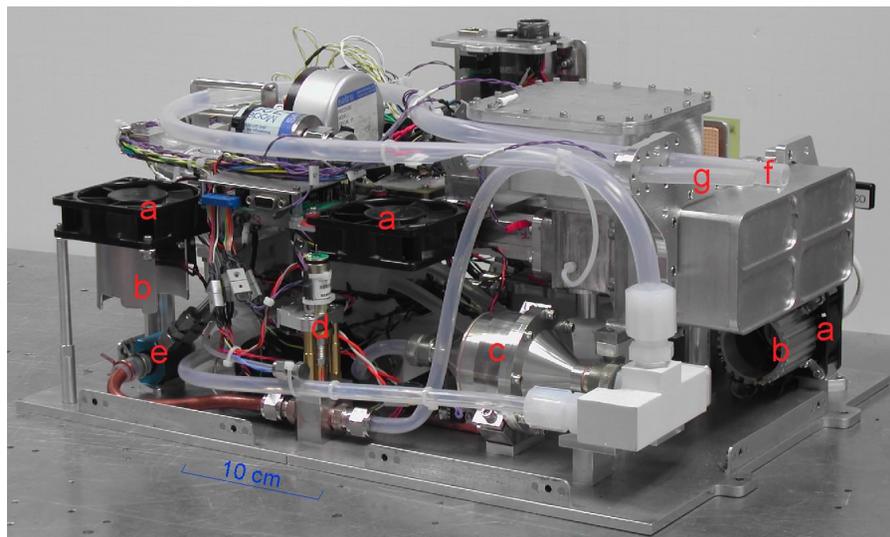


Fig. 2. A photo of the NOAA-2 O₃ instrument without its enclosure showing a: fans; b: heaters; c: catalytic scrubber; d: flow control valve (V3); e: sample flow sensor; f: sample line; g: exhaust line.

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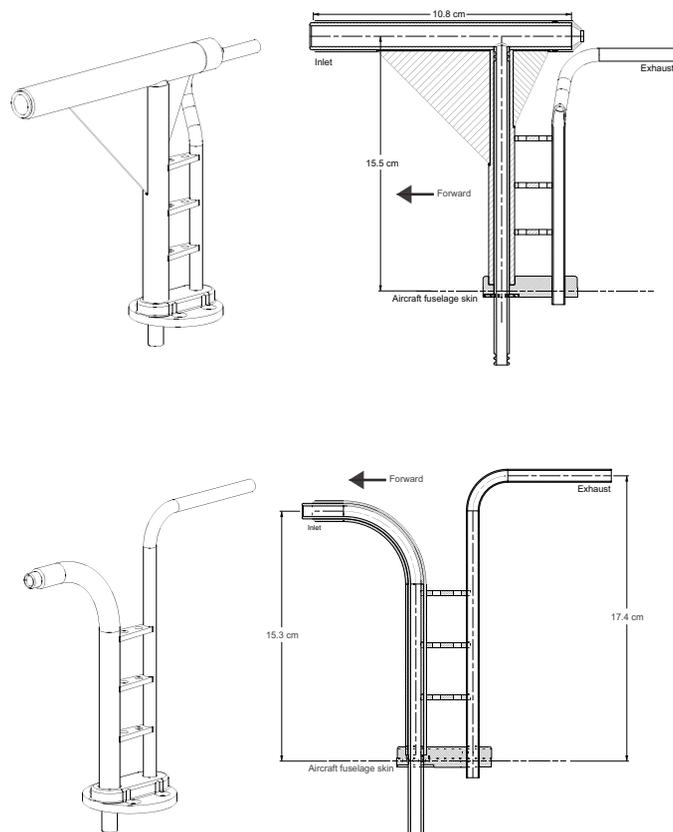


Fig. 3. Two types of NOAA-2 O₃ inlets tested on board the NASA WB-57F aircraft. Upper panel: side-facing inlet with a tubular scoop. Lower panel: simple forward-facing inlet. Drawing is shown to scale.

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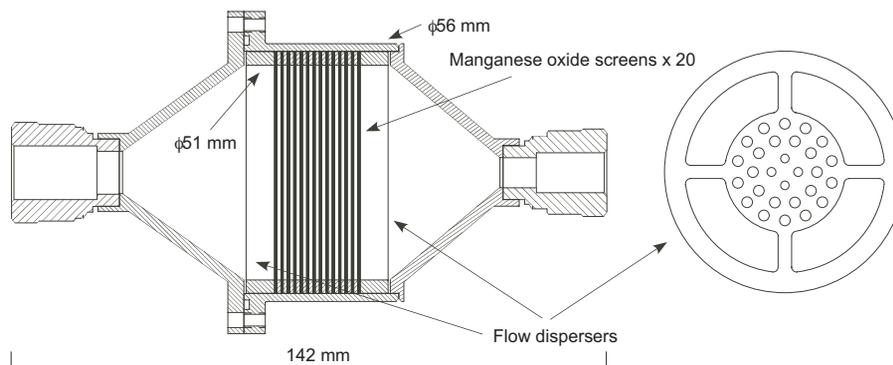


Fig. 4. Cross-sectional view of the catalytic O₃ scrubber (left) and flow disperser (right). The exposed manganese oxide screen diameter is 51 mm. The schematic is shown to scale.

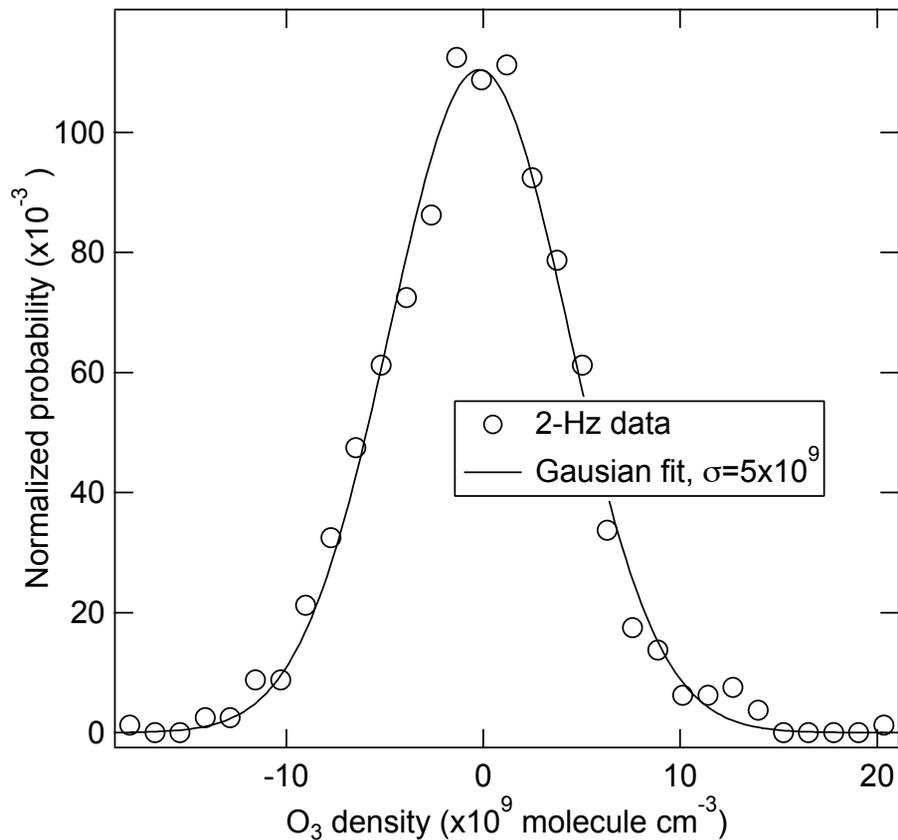


Fig. 5. A typical determination of the intrinsic instrument precision of NOAA-2 O₃ instrument. O₃ measurements recorded for a 1000-s period at 2 Hz without sample flow were used to form a probability distribution function (PDF). The PDF integral is normalized to 1. As shown, the distribution of the data is very close to a perfect Gaussian, indicating that the measurement noise is random. The Gaussian fit gives a precision (2 Hz, 1 σ) of 5×10^9 molecule cm⁻³.

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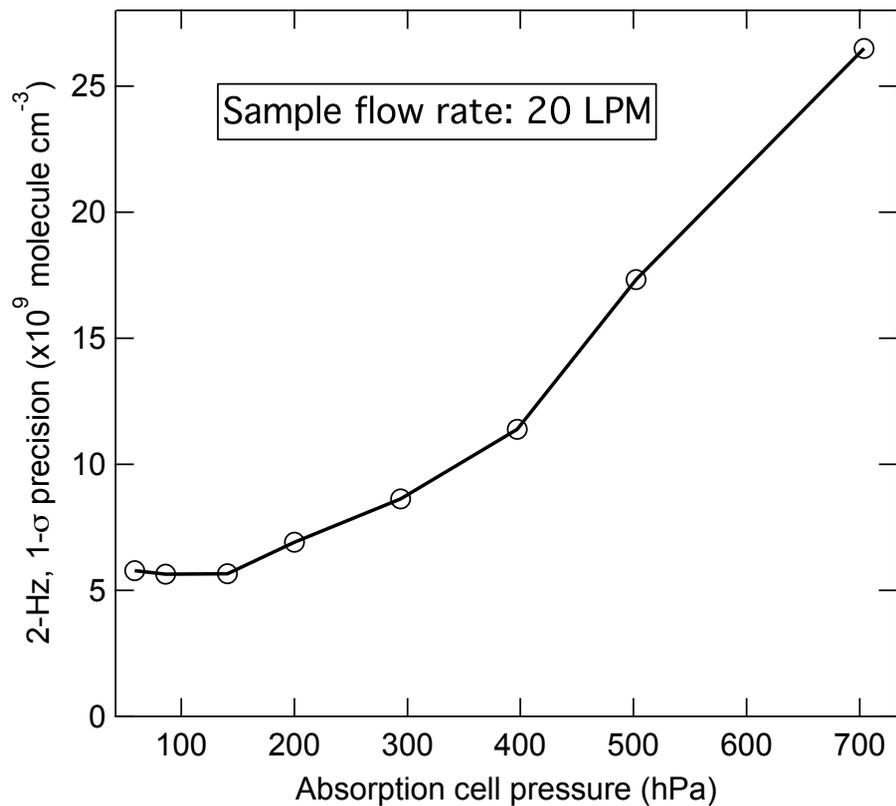


Fig. 6. NOAA-2 O₃ instrument precision (2 Hz, 1σ) as a function of absorption cell pressure at a fixed sample volume flow of 20 LPM. The intrinsic instrument precision (no flow) is 5 × 10⁹ molecule cm⁻³.

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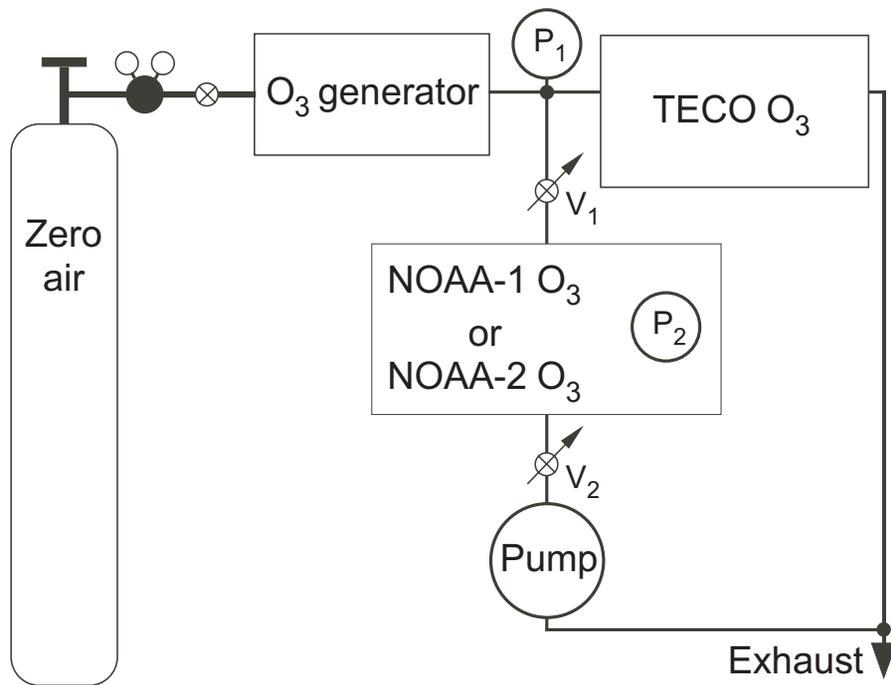


Fig. 7. Laboratory setup for the O₃ instrument intercomparison. Gas lines between the O₃ generator and O₃ instruments and valve V_1 are made of PFA Teflon[®]. V_1 is used to ensure that pressure P_1 is always above laboratory ambient pressure. V_2 is used to control the pressure inside the NOAA-1 or NOAA-2 O₃ instruments undergoing an intercomparison with the TECO Model 49i-PS primary standard instrument.

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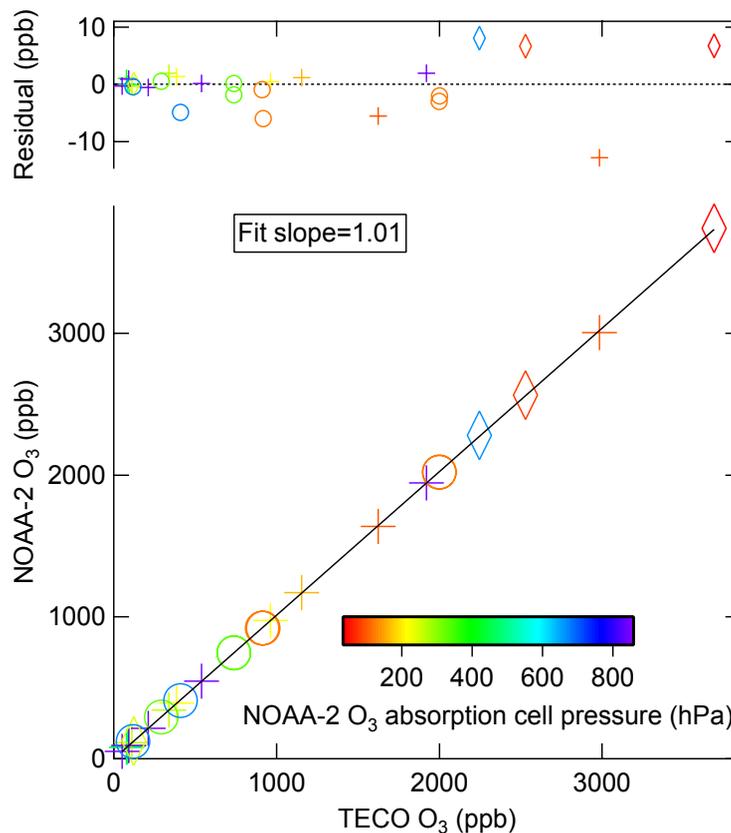


Fig. 8. Intercomparison between the NOAA-2 O₃ and the TECO O₃ primary standard instrument over a range of cell pressures in the NOAA-2 O₃ instrument. Each data point represents a 40-s average of O₃ mixing ratio in units of parts per billion (ppb). The slope of the linear best fit through the origin is 1.01 ± 0.01 . The fit residuals are shown in the top panel.

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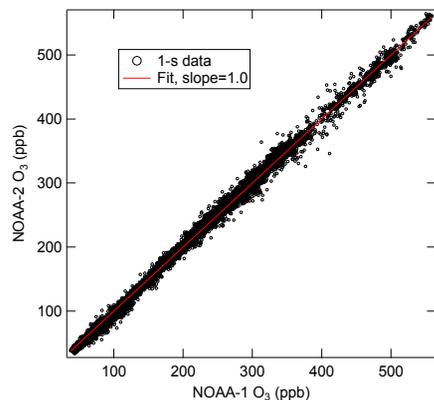
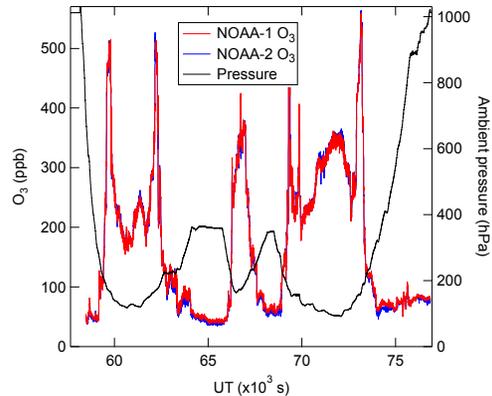


Fig. 9. In-flight intercomparison between the NOAA-1 O₃ and NOAA-2 O₃ instruments while operating onboard the NASA WB-57F high altitude research aircraft on 11 April 2011 in Houston, TX. Top panel: time series of O₃ mixing ratio data over the approximately the 5 h flight along with ambient pressure. The NOAA-2 O₃ data points are averaged to 1 s to match the time resolution of the NOAA-1 O₃ instrument. Bottom panel: correlation of the two in-flight datasets (1 s data) showing a slope of the linear best fit through the origin of 1.00 ± 0.01 .