Response to reviewer 2

The authors thank the anonymous reviewer 2 for their comments and suggestions. We have carefully revised the manuscript, taking the comments into account. Our responses to the reviewer's comments are provided below. **All the line numbers in this document refer to the manuscript with highlighted changes (Latexdiff).**

General comments

The manuscript "Raman source using a hollow-core fiber for ozone monitoring in the lower troposphere" presents an innovative architecture for miniaturizing UV DIAL systems. The integration of a deuterium-filled hollow-core fiber (HCF) to generate the 289 nm Stokes wavelength via stimulated Raman scattering (SRS) is a highly compelling approach to reducing the size and pump-energy requirements of traditional free-space Raman cells. The initial atmospheric profiles demonstrating measurements between 400 m and 1700 m in an urban boundary layer show the technique's fundamental viability. However, there is a significant gap between the successful demonstration of a novel optical architecture and the paper's stated goal of creating an instrument optimized for "autonomous boundary-layer monitoring" and "routine operation". A true operational network asset requires robust, uninterrupted 24/7 profiling. The current manuscript lacks critical laboratory characterization regarding the long-term stability of the transmitter, specifically concerning potential fiber degradation and optomechanical drift. Addressing these stability concerns, alongside providing independent vertical validation, is necessary before the system can be evaluated as a candidate for autonomous network deployment.

We agree that the demonstration of a new transmitter architecture is distinct from establishing an instrument capable of routine autonomous monitoring. The original wording did not sufficiently distinguish these stages. We have revised the abstract, introduction, discussion of Fig. 6 and conclusions to clarify that DIABLO is currently a proof-of-concept prototype.

The objective of this study is to characterize a low-pulse-energy, deuterium-filled HCF Raman source, integrate it into an ozone DIAL configuration, and demonstrate its measurement capabilities through initial atmospheric observations. The profiles acquired between 400 and 1700 m provide a first assessment of these capabilities. Comparisons with in situ observations and CAMS estimates show how the retrieved concentration levels relate to available observations and model outputs, but do not independently establish their accuracy.

The revised conclusions identify the development of an industrial prototype as the next stage before considering network deployment. This requires extending the usable altitude range, establishing stable continuous operation, and improving and validating the retrieval processing. Extended tests will address laser lifetime, potential fiber ageing, gas retention, output-energy stability and sensitivity to temperature variations. Mechanical stability also requires assessment. Improved gas confinement will be investigated, while thermal regulation will be considered if the test results indicate that it is necessary.

Further development will also address acquisition and processing to target integration times of 15 min, together with instrumental and atmospheric corrections and their uncertainty propagation. Complementary aerosol lidar observations would help constrain aerosol interference. Independent, coincident vertical ozone measurements remain necessary to assess the retrieval accuracy.

Specific comments

State of the art

Regarding the characterization of existing systems, the authors state in the abstract that current UV DIAL ozone systems are typically "operated in campaign-based research configurations". This generalization should be revised, as it underrepresents the current operational capabilities of the community. For example, instruments within the Tropospheric Ozone Lidar Network (TOLNet), such as the Small Mobile Ozone Lidar (SMOL) platform, routinely execute autonomous, continuous 24/7 vertical profiling over multi-month deployments with minor servicing needed every several weeks rather than being limited to short-term research campaigns. Acknowledging this existing transition toward operational network architectures will provide a more accurate baseline for comparison.

We agree that the original wording underrepresented existing ozone lidar capabilities. In the abstract, we removed the statement that existing UV ozone DIAL systems are typically operated in campaign-based research configurations. The revised text identifies the objective of this study as the integration and initial atmospheric demonstration of a low-pulse-energy HCF Raman source and distinguishes this demonstration from readiness for continuous autonomous operation. **(Lines 71 to 90)**

In the introduction, we expanded the discussion of existing observations to include nighttime ozone observations (Sullivan et al., 2017), LISTOS measurements (Couillard et al., 2021), the Paris ACROSS campaign (Ancellet et al., 2024), and airborne measurements during TexAQS 2006 (Senff et al., 2010). We also reduced the accumulation of older references in this part. We now explicitly describe the autonomous capabilities of AMOLITE (Strawbridge et al., 2018) and SMOL (Chouza et al., 2025), acknowledging that extended, near-continuous ozone profiling has already been demonstrated.

Transmitter Stability

The most critical omission in the manuscript is the absence of a long-term stability test of the transmitter under continuous operation. It remains unclear whether the 289 nm output energy remains stable at 7 µJ after 24 or 48 hours of continuous pulsing, or if it degrades rapidly. The authors must include a continuous laboratory stability plot tracking the output pulse energy of both the 266 nm residual pump and the 289 nm Stokes beam over at least a 24- to 72-hour period.

We agree that extended monitoring of the output energies at both wavelengths is essential to assess transmitter stability and suitability for autonomous lidar operation. However, the present prototype is at an earlier stage of development. This study evaluates whether the HCF-based Raman source can support ozone DIAL measurements. The source was integrated into a DIAL prototype to demonstrate this capability and obtain an initial estimate of the achievable

measurement range. Long-term stability testing is therefore a necessary next step, beyond the scope of the present proof-of-concept demonstration.

To ensure consistent pressure and energies at both wavelengths, the fiber was refilled with deuterium to a pressure of 6.5 bar before each ozone measurement presented in the article. **(Lines 276 to 277)**

Optomechanical Drift and Coupling Stability

The authors note that coupling the 266 nm beam into the HCF is challenging due to a low numerical aperture (NA ~0.02) and tight angular acceptance, making the system "highly sensitive to alignment conditions". In an autonomous deployment, the system will be subjected to significant diurnal temperature swings and vibration during transportation and operations. Thermal expansion typically causes sub-micron drifts in optomechanical mounts, which could severely misalign the fiber coupling and destroy the Raman conversion efficiency. The authors should discuss how they expect to manage this issue.

We agree that coupling stability is a critical requirement for unattended operation. The present measurements do not establish the long-term stability of the coupling under varying environmental conditions. Tests under different temperature conditions are certainly needed to assess the sensitivity of the fiber coupling and Raman conversion efficiency to thermal drift. This will be done in the next phase of the project. Depending on the results, thermal regulation of the transmitter enclosure could be considered, which would be facilitated by the compact transmitter design. **(Line 377)**

The intended application is long-term deployment at fixed monitoring sites, with occasional transportation for installation or relocation. Therefore, resistance to frequent transportation is not a primary design requirement at this stage. Mechanical stability during operation remains important, and alignment will need to be checked after transportation.

Near-Field Blind Spot and Overlap Function

Although the instrument is designed for boundary-layer profiling, the lowest reported measurement altitude is 400 m. For urban air quality applications, capturing the lowest 400 m is arguably the most critical requirement for connecting boundary-layer dynamics to surface exposure. The authors state that this lower limit is constrained by incomplete overlap. However, given that the receiver has a relatively large full-angle field of view of 5 mrad and the transmitted beam divergence is estimated below 1 mrad, achieving full geometrical overlap should theoretically not be difficult at much shorter ranges. The authors should address whether there is an unusually large baseline separation between the transmitter and receiver causing this delayed overlap. If not, they should clarify if detector saturation or pulse pile-up at 1 kHz is the actual limiting factor and clearly outline the necessary receiver modifications required to profile this near-surface layer.

We are aware of the importance of extending the measurements towards the surface. The separation between the transmitter and receiver axes is approximately 12 cm. We agree that,

with perfectly aligned axes and well-collimated beams, the nominal receiver field of view would allow full geometrical overlap at substantially shorter ranges than 400 m. However, ozone DIAL retrievals depend on the variation of the ratio of the overlap functions at the two wavelengths with the range. Small differences in beam divergence, combined with residual transmitter–receiver misalignment, can therefore introduce a short-range retrieval bias.

Fig. 3(b) shows that the systematic error on the differential logarithmic slope near 250 m could be as large as 0.8 km^{-1} resulting in an error of 40 $\mu g\ m^{-3}$. on the ozone concentration at 250 m.

Such an error of 0.8 km^{-1} on the differential logarithmic slope could be related to differences of the overlap between the telescope and the laser beam at 266 and 289 nm.

$$\frac{\mathrm{d}}{\mathrm{dz}} \ln \left[\frac{F(z, \lambda_{on})}{F\left(z, \lambda_{off}\right)} \right]$$

Simulations of the overlap difference with a transmitter–receiver separation of 12 cm, different beam divergences between 0.4 and 2 mrad, and angular misalignments between 0.7 and 1.6 mrad have been made.

These simulations were used solely to assess the potential influence of differential overlap on the short-range retrievals. No overlap correction was applied to the ozone profiles presented in this study.

Laser divergence differences of the order of 0.5 mrad are indeed very likely and can be corrected if they are stable. Alignment accuracy between the laser and the telescope better than 1 mrad is also needed to avoid applying differential overlap correction and extend the ozone retrieval below 400 m.

These simulations demonstrate that differential overlap can produce a systematic bias of the magnitude required to explain the observed short-range slope distortion. We therefore retain 400 m as a conservative lower retrieval limit for the ozone measurements presented.

Photon-counting pile-up does not directly determine this limit because the near-range retrieval uses analog signals. This does not, however, exclude PMT or analog-channel non-linearity, which requires dedicated testing.

To extend the usable range, we will prioritize wavelength-resolved beam-divergence measurements, more precise collimation and transmitter–receiver alignment, and experimental characterization of the differential receiver response. A receiver configuration optimized for near-range collection will also be investigated.

The manuscript has been revised to clarify the beam-divergence estimates and overlap limitations **(lines 141 to 145)**, specify the transmitter–receiver separation **(line 149)**, and explain the conservative 400 m lower retrieval limit and the possible contribution of differential overlap **(lines 188 to 191)**. The use of analog signals at short range is described in **lines 193 to 200**, and the absence of overlap correction is stated in **line 191 to 192**. Planned improvements to the receiver, collimation and alignment are discussed in **lines 357 to 363**.

Validation with Ozonesondes

While the comparison with surface network data (such as the Paris 13ème and Tour Eiffel stations) and the CAMS model provides useful context, the manuscript lacks independent physical verification of the vertical structure. To properly validate the lidar's vertical profiling capabilities and confirm the observed boundary-layer ozone gradients, it would be highly beneficial to incorporate a couple of coincident ozonesonde launches during the measurement periods.

We agree that coincident ozonesonde measurements, or observations from another independently validated ozone lidar, would provide a stronger assessment of the retrieved vertical structure. No such observations are available for the measurement periods presented here. Consequently, the accuracy of the profiles and the observed vertical gradients cannot yet be independently established.

We have rewritten the discussion of Fig. 6 accordingly. The in situ observations are used to compare concentration levels and variability at different heights. Surface ozone exhibits stronger variability than the Eiffel Tower measurements at 300 m, illustrating why surface observations alone cannot characterize ozone throughout the lower troposphere. Except for the 19:00 UTC profile, the DIABLO concentrations near the lower retrieval limit are consistent with the concentration range measured at the Eiffel Tower.

CAMS is used only to compare the order of magnitude and vertical distribution of ozone concentrations with model estimates. We now explicitly identify the need of independent vertical ozone observations as a priority for future validation, both in the discussion of Fig. 6 **(lines 296 to 300 and 316 to 331)** and in the conclusions **(lines 343 to 349 and 383 to 384)**.

Gas Permeation Mechanism

The manuscript notes a deuterium pressure drop over a scale of days, which is currently attributed to permeation through the gas connections. While this pressure drop is not a primary concern for the overall viability of the technique, the authors should explore the physical mechanism more thoroughly and explain if thermal expansion and contraction as expected during typical field deployments and transportation could pose additional challenges.

We agree that the observed pressure decrease does not, by itself, establish permeation through the gas connections as the dominant mechanism. Possible contributions include leakage at the connections and seals, and permeation through the materials of the gas-containing assembly.

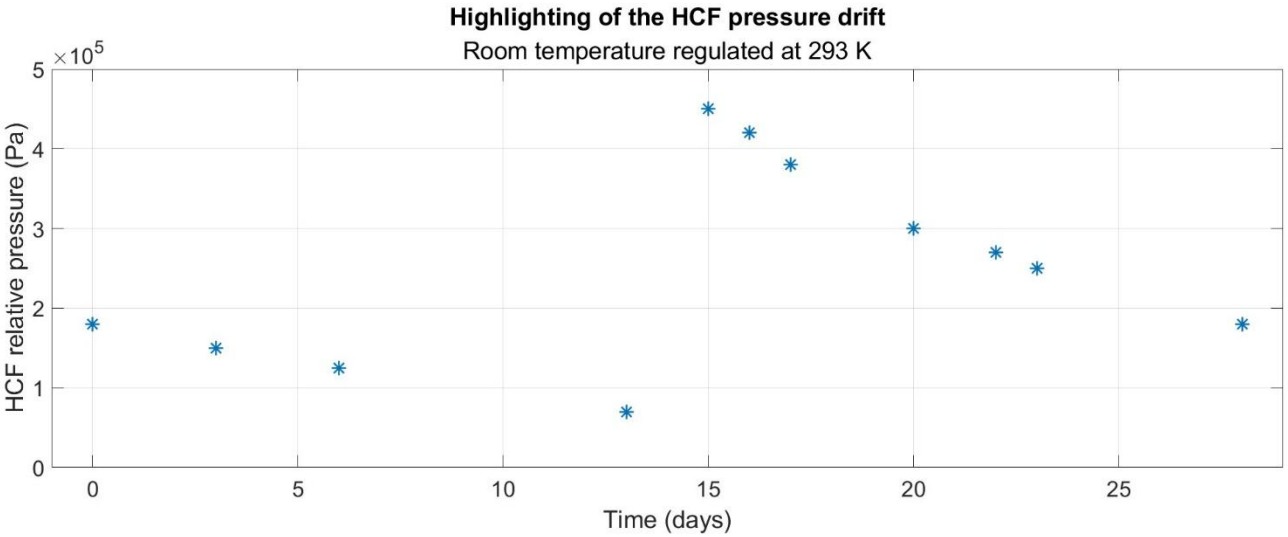


Figure R2.1. Pressure evolution in the gas-filled HCF assembly over a month. Room temperature was regulated at 293 K. The pressure increase on the 15th day corresponds to a gas refill.

Although continuous measurements of the output energies at 266 and 289 nm are not available, we have performed gas-pressure retention tests over several weeks. As shown in the Figure R2.1Figure R2.1. Pressure evolution in the gas-filled HCF assembly over a month. Room temperature was regulated at 293 K. The pressure increase on the 15th day corresponds to a gas refill., the pressure decay is faster at higher pressures and slows as the pressure decreases. This behavior is consistent with pressure-dependent gas loss. Following a refill, the pressure decreased from approximately 4.5 to 1.8 bar over thirteen days, under room-temperature regulation at 293 K. These observations characterize the timescale of pressure loss and provide information relevant to gas-refilling requirements. However, the actual refill interval depends on the minimum pressure required to maintain adequate output at both wavelengths.

Before each ozone measurement presented in the article, the fiber was refilled with deuterium to a pressure of 6.5 bar to ensure a consistent pressure.

Gas-retention tests performed by the fiber manufacturer, GLOphotonics, using argon showed very good gas confinement. However, these results cannot be directly extended to deuterium, since permeability depends on the gas species and material. In particular, molecular permeation of D_2 through silica has been experimentally demonstrated at temperatures encompassing ambient conditions (Shelby et al. 1977). Dedicated tests are therefore needed to distinguish permeation through the fiber from gas loss through connections and seals. If fiber permeation is confirmed as a significant contributor, one option would be to enclose the fiber in a sealed stainless-steel tube, potentially less than 1 cm in diameter, filled with deuterium at a pressure similar to that inside the fiber. Reducing the D_2 pressure difference across the fiber would reduce the driving force for outward permeation and could improve pressure stability.

Temperature may also modify material permeability, while differential thermal expansion and contraction can affect mechanical stresses and sealing performance at the connections. Transportation could introduce additional mechanical loading at these interfaces. If these effects cause deuterium loss, the resulting decrease in deuterium number density may also modify the Raman conversion efficiency.

Further characterizations are required to quantify the respective contributions of these mechanisms for this prototype. Improved gas confinement and thermal stabilization will be evaluated as measures to support long-term autonomous operation.

J. E. Shelby; Molecular diffusion and solubility of hydrogen isotopes in vitreous silica. *J. Appl. Phys.* 1 August 1977; 48 (8): 3387–3394. https://doi.org/10.1063/1.324180

Editorial comments

Section 1, Line 46: "...Network for the Detection for Atmospheric Composition Change..." – The correct name for NDACC is the Network for the Detection of Atmospheric Composition Change.

Section 2.2, Line 127: "...separed by 100 cm..." – Should be "separated".

Section 5, Line 271: "...(Chouza et al., 2025) Such developments..." – There is a missing period after the citation.

These errors have been corrected in the revised manuscript.