Atmospheric Gas Imaging with Off-beam Backscattered Light

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Executive summary

We have developed a new framework for analysing and interpreting measurements of backscattered light arriving at the detector from oblique directions. Although this 'off-beam' component of the optical measurement is commonly regarded as backscatter noise because its trajectory cannot be uniquely resolved, we have shown how this can be effectively treated as a useful signal in developing a new, Lidar-like modality capable of tomographic imaging of atmospheric gases. Our technology combines the operational protocols of Differential Absorption Lidar (DIAL), but captures and exploits both on-beam and off-beam photon returns. To process these data, our novel algorithmic framework, fuses optical scattering and gas dispersion models, enabling - *for the first time* - to extract absorption information from multiply scattered light. In turn, this new capability facilitates profoundly faster acquisition times and accurate 3D quantitative imaging of gas concentrations even in highly scattering conditions. Ultimately, this advantage overcomes the performance degradation of conventional DIAL in the presence of significant scatter. The next steps towards commercialization involve the design and characterisation of a dual, 'on-beam' & 'off-beam', sensor development, and a subsequent controlled experimental validation. The financial resources and strategic advice of by industry experts will catalyse and expedite the deployment and impact of our technology. We anticipate that Hamamatsu's Pilot Line service and Gas Analysis expertise, will help us demonstrate the concept and scope out a broader range of applications where off-beam light interpretation can also lead to innovative or enhanced solutions and new products. This endeavour aligns to global efforts in improving environmental monitoring and address pressing issues related to air pollution, and the effective management of industrial emissions.

1 Innovation

1.1 Measuring Beyond Single Scattering

DIAL systems are considered to be the gold-standard in mapping gas concentrations by measuring the differential attenuation of backscattered light. Short laser pulses at two wavelengths are fired into a gas plume from different directions, and the backscatter photon-counts are recorded on the detector only within a narrow field of view (nFOV) aligned to the direction of the outgoing light. This measurement, attributed to optical absorption, yields data consisting exclusively of single-scattered photons, having travelled forth and back, through the plume, on an almost straight trajectory. Whilst the nFOV measurement is optimal for ranging applications, where light typically bounces off opaque surfaces in a very directed way, when dealing with soft scatterers like gas plumes, the measurement's efficacy is significantly impacted by the plume's optical thickness, which can cause the measured intensity to become highly non-homogeneous. We argue that measuring from a nFOV as well as a wider FOV (wFOV) becomes more advantageous or even necessary for the imaging gas concentrations as the plumes become more optically thick.

Optically thin gas plumes When light propagates through nearly transparent gas plumes, there is just enough scattering to cause a small fraction of the photons to bounce backwards from various points along a straight optical trajectory. By measuring the photon returns in a nFOV centred on this trajectory alongside their time of flight (ToF) which allows to specify how far they have reached insight the plume, and hence associate absorption measurements to gas concentration profiles. In the absence of any scattering, transmission and detection must take place in opposite ends of the trajectory, or retrofit reflectors can be utilised, however both alternatives are impractical to deploy in an adhoc basis. In effect, in optically thin media photon counts are expectedly *very small* and dominated by single scattering events, for which nFOV measurement are both suitable and sufficient, as they contain most of the informative absorption signal while at the same time blocking the adversarial impact of ambient illumination. This setting is graphically illustrated in figure 1.

Optically thick gas plumes In the alternative case where gases are dispersed in optically thick, environments the above situation changes considerably, as multiple scattering dominates, sending a significant portion of the photons 'off-beam', i.e. outside the straight trajectory. In effect, only a tiny fraction (much smaller than the optically thin case) of the backscattered photons would return to the detector's nFOV having scattered just once, rendering the 'conventional' nFOV measurement practically unusable. By contrast, as illustrated in figure 2, the photon count at the wFOV will be significantly larger, as more light reaches the detector from off-beam directions having gone through non-straight trajectories, bestowing the wFOV measurement with a workable signal to noise ratio (SNR) that is enough

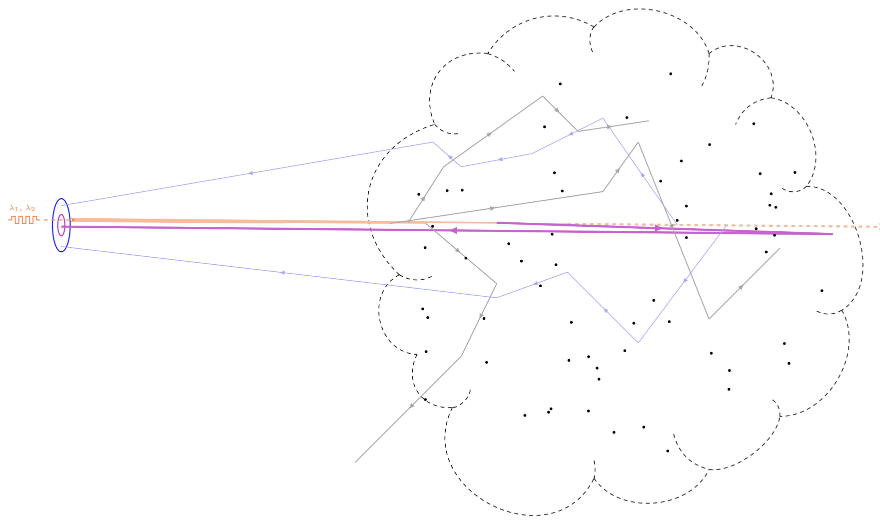

Figure 1: Photon pulses at wavelengths λ_1, λ_2 fired into an optically thin plume. The predominant component of the small amount of backscattered light ends up at the narrow field of view part of the detector (magenta circle), having scattered just once (magenta beams), whilst most of the light propagates through.

> to outweigh the effects of ambient illumination on which the wFOV measurement is prone to. Extracting the necessary information from these wFOV measurements however is far from obvious, as they do not adhere to straight trajectory paths, and hence a redesign of the data processing cycle becomes imperative.

Having a principled, physics-rooted framework to process the wFOV data in conjunction with the Beer-Lampert law that governs the nFOV intensity measurements, our approach addresses the intrinsic shortcomings of the nFOV-exclusive technologies. Starting with DIAL, but potentially transferring the main ideas to other gas analysis modalities like the Tunable Diode Laser Absorption Spectroscopy (TDLAS) and the Differential Optical Absorption Spectroscopy (DOAS) in the future, we hope that our pioneering results will find their place in measurement practice and new advanced products.

1.2 Image reconstruction

In Differential Absorption Lidar (DIAL) applications, accurately retrieving 3D gas concentration fields relies on measuring the small absorption differences between on- and off-resonant laser pulses. When absorption is weak, as is often the case, the signal-to-noise ratio (SNR) becomes a critical limiting factor as

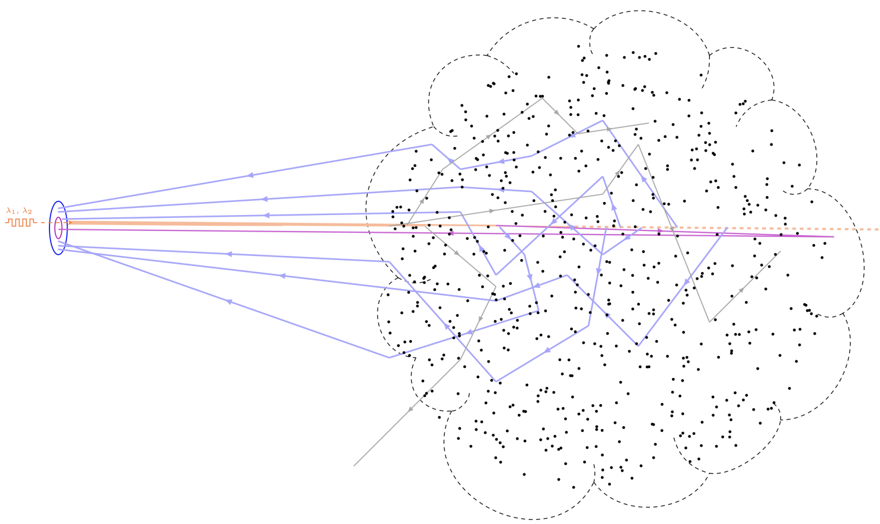

Figure 2: Photon pulses at wavelengths λ_1, λ_2 fired into an optically thick plume. In this environment multiple scattering is dominant within the plume, hence the single scattered photons (magenta beams) returning at the narrow field of the detector (magenta circle) reduce considerably. On the other hand, some of the multiply scattered photons (blue beams) can be recorded at the detector's wide field of view (blue circle). Notice, the intensity of the narrow field of view photons significantly is less than those in figure 1.

it increases statistical uncertainty in the inversion process. We thus propose to augment a classical narrow field-of-view Lidar system with additional wide FOV data.

From an image reconstruction point of view, the nFOV data is beneficial for reducing background light and improving spatial precision, but it can exacerbate the low SNR problem by restricting the amount of backscattered light collected. Wide FOVs on the other hand collect more light, i.e. we will observe a higher photon count, but not only does this also include a significant amount of ambient light (that carries no information about the plume whatsoever), due to the complexity of multiple scattering it is not possible to determine photon paths from time-of-flight (ToF) binning at the detector alone and each measured photon, even if actively released, carries less information.

To fully exploit the available data and mitigate the loss of information due to the more complex nature of radiative transfer, it is essential to integrate the inversion process with an atmospheric dispersion model. The dispersion model provides physically plausible constraints that help stabilise the solution and provably compensates for the lack of photon-path information that is inherent to wide FOV measurements. Instead of relying purely on the direct Lidar measurements, the inversion leverages knowledge of how the gas plume evolves due to advective transport and diffusion, allowing for more robust estimation even in cases where the raw absorption signal is weak. Additionally, the coupling provides us with an intuitive and interpretable model reduction that makes the inversion process, which involves multiple solutions of the Radiative-Transfer-Equation (RTE), computationally tractable on low-cost consumer grade hardware.

2 Relevance and impact

We have developed a novel technology that significantly advances optical remote emission sensing, surpassing the inherent limitations of conventional Differential Absorption Lidar (DIAL) and other single scattering reliant technologies, in environments characterised by high scattering coefficients. This enhanced capability is crucial for accurate and reliable atmospheric monitoring in densely populated urban areas, which frequently experience substantial concentrations of scattering particulates such as soot and dust. Furthermore, its applicability extends across a broader geographic scale, offering a vital tool for atmospheric characterization in regions like the Middle East and North Africa, where the frequent occurrence of significant sand and dust storms presents a persistent challenge for conventional remote sensing techniques.

Beyond air quality monitoring, the bespoke combination of rapid data acquisition and the high-speed imaging our methodology offers, makes it suitable for emergency chemical hazard assessment. The capacity for rapid remote detection and plume tracking is imperative for effectively guiding first responders and mitigating potential risks. Consequently, we anticipate a valuable contribution to safeguarding public health and security through the deployment of this technology.

3 Feasibility

An important aspect of our approach, adding to its overall appeal, is the simplicity of the hardware modifications needed on existing DIAL instrumentation. Yet, despite this evocative similarity, as shown in 3, our bespoke data processing approach is a game changer for optical remote sensing. Following a rigorous mathematical approach, firmly attached to practical instrumentation capabilities we make a departure from the conventional ToF formalism as a 'model' for interpreting optical backscattered measurements, to consider data bouncing from and within a dispersion process (plume). We also hope that we can steer some interest from the optical instrumentation community once we manage to convenience them that for the appropriate choice of model, off-beam light returns should not necessarily be treated as optical noise.

Having established the relevant theory of our approach in [2] our priority is now on exploring its potential impact and practical performance in real conditions. The pending technical challenge is the development of the nFOV + wFOV sensor and its integration within a DIAL system to perform a proof of concept experiment. The feasibility study will involve a controlled release of a non-toxic absorbing gas like Methane or Carbon dioxide along with a scattering agent like smoke in an open space so that optical measurements can be captured from a reasonable distance. Capturing both FOV data will allow a direct comparison of our method compared to standard DIAL imaging, in terms of spatial resolution and errors in the estimated concentrations. This experiment will also provide valuable information on the true impact of background illumination interference and the robustness of our approach to various forms of noise and model discrepancies.

As we have already tested and published [1] our algorithms based on simulation and synthetic data at realistic or even harsh conditions in terms of ambient noise intensities, sensor detection rates and gas plumes of low concentration in the absorbing gases, we have a reasonable amount of confidence that we can implement the intended experiments and process the data in around a six month period, on our own computational resources.

4 Quality of methods

We outline a few metrics based on which, we feel, the quality of our application could be assessed

Novelty/Innovation Our recent theory [1, 2] challenges the prevailing view that multiply scattered photons are effectively "noise" that must be removed before it reaches the detector, or ultimately filtered from the data, as one should strive to measure only the single scattered light from a very

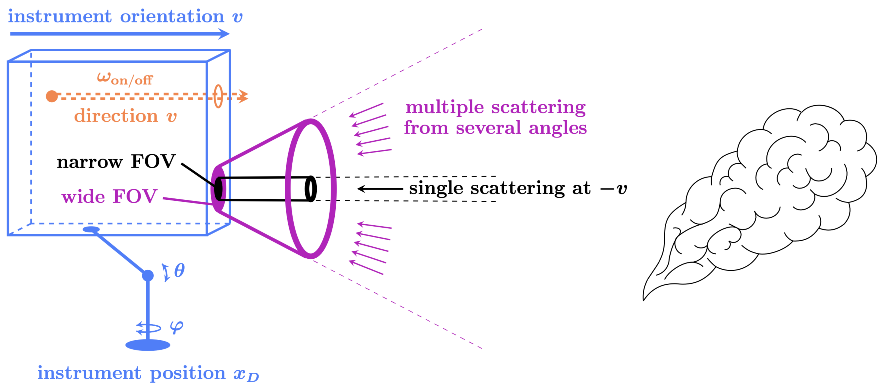

Figure 3: A schematic illustration of the concept instrument, incorporating a laser source emitting photon pulses at two frequencies $\omega_{\mathrm{on/off}}$ like a DIAL, and the dual field of view sensor. The platform on which the source and detector are located, rotates around two angles to acquire time resolves photon count returns, across a cone of directions v.

narrow field of view. To the best of our knowledge, no form of optical remote sensing the utilises wide fields of view exists outside [2, 1], and hence there is untapped synergistic potential across several optical modalities.

Scientific foundation & reliability Published in a top-tier journal on the mathematics of imaging, and defended in a PhD thesis, we demonstrate that under conditions of significant scattering, a nFOv and wFOV measurement provides additional information to make the gas concentration estimate more accurate. Our hypothesis has been proven and sufficiently explained, so although non-conventional, we trust our approach to be reliable as it is grounded in physics (radiative transfer and dispersion), as well as principled, analysed statistical algorithms for data processing.

Robustness We provide clear explanations and justifications on the circumstances where the wFOV is informative (e.g. scattering environments). Our algorithms have been tested across a range of instrumentation and optical noise levels, as indeed detector sensitivities and gas concentration levels. Regarding the sensor technology, likely InGaAs, is well established and analysed in the realm of gas sensing applications, so some optical engineering design is what is essentially needed to realise and integrate the dual measurement.

Practicality The dual measurements can be collected seamlessly using the DIAL acquisition protocol, albeit with a modified sensor. Data acquisition does not require retrofit reflectors, which makes it agile to deploy. In its current version, our algorithm can process the data offline using standard level compute power over a few minutes, without the need of high performance computing.

References

[1] R. Lung and N. Polydorides, “Imaging of atmospheric dispersion processes with differential absorption Lidar”, SIAM Imaging Sciences, Vol. 17, Iss. 3, 2024. *Available online*.

[2] R. Lung, “Atmospheric imaging with differential absorption Lidar off-beam returns”, PhD Thesis, University of Edinburgh, 2024. *Available online*.