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SAQN Scoping Study End of Project Report

Project Title	
Modular Relaxed Eddy Covariance sensor for Air Quality: MOREC-AQ	
Project Team	
Name	Role (PI / Co-I)
PI (to meet RCUK requirement)	Roderic Jones
Lead Co-I	Olalekan Popoola (OP)
Co-I	Thomas Wall (TW)
Co-I	Zaheer Nasar (ZN)
Proposed activities (copy from your project proposal)	

The project aims to develop a proof-of-concept for a cost-effective Modular Relaxed Eddy Covariance (MOREC-AQ) measurement approach to fluxes/source characterisation and a miniaturised cost-effective NH_3 instrument to incorporate into MOREC-AQ.
Our specific objectives include: (1) feasibility studies for a portable high-resolution NH_3 sensor through STFC; (2) design and characterisation of a prototype MOREC-AQ unit; (3) explore additional funding opportunities to further develop and optimise the prototype MOREC-AQ unit

We will achieve these objectives through

(1) modelling studies and proof-of-concept work for an ammonia instrument, building on experience from ongoing miniaturisation work and instrument development at RAL Space;

(2) designing a prototype MOREQ-AQ unit by leveraging existing knowledge from relaxed eddy covariance approach, but crucially incorporating low-cost air quality sensors;

(3) actively engaging with stakeholders with the prospect of seeking further funding opportunities.

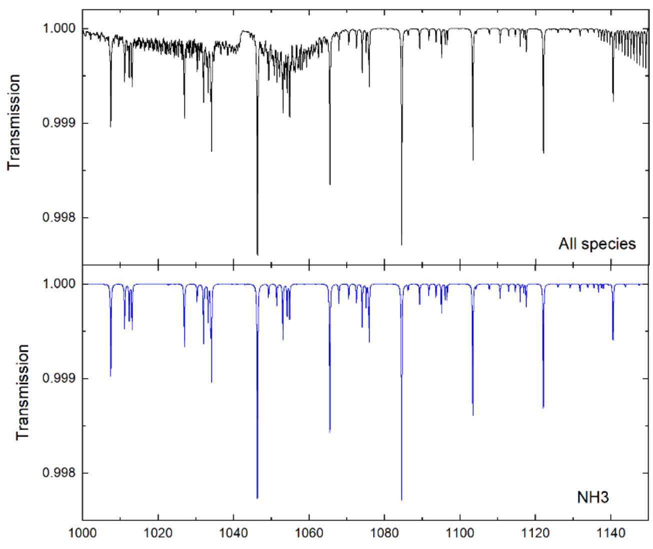
Please report on the activities completed in the project

(1) Feasibility studies for a portable high-resolution NH_3 sensor

The STFC component of this project was to investigate the feasibility of a laser-based spectrometer for detection of atmospheric ammonia (NH_3). Eddy covariance (EC) sensors often use closed systems for atmospheric detection. The gas samples are introduced into the sensor through an inlet. This does not work well for NH_3 as it is ‘sticky’: it can adhere to the inner surfaces of the sensor, which affects the concentration measurement, and can lead to aerosol production, which further perturbs the measurement. In order to avoid this problem, it is preferable to use an open system. We have investigated the feasibility of an open-path sensor for NH_3 detection that could be integrated into the MOREC-AQ system. This work has focussed on spectroscopic modelling to identify optimal wavelengths for NH_3 detection, as well as an investigation of how the detected signal can be improved by using a multi-pass cell (MPC).

The initial modelling found three main spectral regions for NH_3 detection, with the strongest being at a wavelength of round 10 μm. This is a spectral region of the mid-infrared (MIR) which the RAL Space Spectroscopy Group (SG) has experience of working in, and for which quantum cascade lasers (QCLs) are commercially available. The other strong regions were found to be at around 90 μm (where laser technology is not mature) and at around 6 μm. This latter spectral region could be used, given the available technology, but the molecular transitions are around 5 times weaker than in the 10 μm band.

With the basic spectral investigations complete, more detailed modelling was performed to locate the optimal wavelength for NH_3 detection. This involved modelling the detection method proposed for this new sensor: Chirped Laser Dispersion spectroscopy (CLaDS). This method measures the change in the phase of laser light as it interacts with molecular resonances. As opposed to traditional absorption spectroscopy, CLaDS is immune to fluctuations in the detected laser power, meaning that the measurements are robust against the effects of turbulent air movement, as well as dust and particulates passing through the laser beam. This method is thus well suited to an open system for flux measurements. The modelling work included a full description of the CLaDS detection mechanism, simulating the dispersion that would be measured in the envisioned sensor. The model included the effects of using a commercially available MPC to increase the laser path length to 30 m. This allows sensitive detection in a compact, open system.



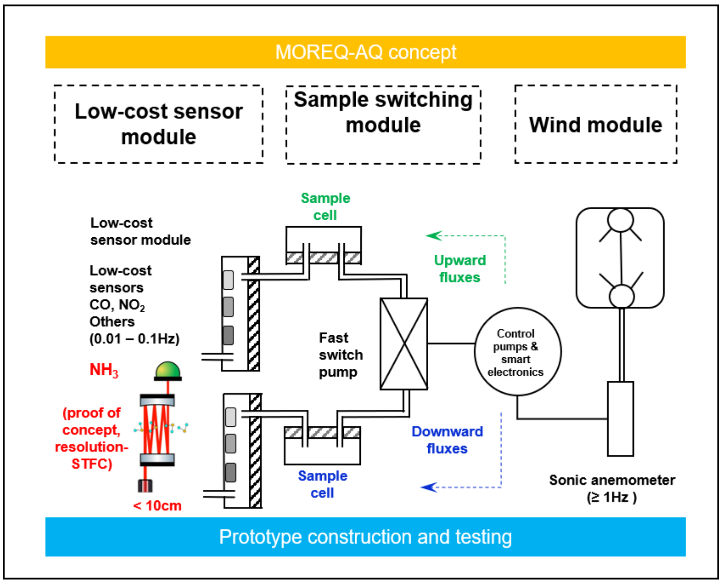
The dispersion modelling was first performed for NH_3 only, with a concentration of 10 ppb. This was used to explore the 10 μm band in detail, and to identify the strongest dispersion signals. Once this was complete a more realistic atmospheric model was performed that included all the main atmospheric species. This more complete modelling allowed the effects of interference from other species' spectral signatures to be investigated. Based on this modelling, optimal spectral regions were identified that included strong NH_3 dispersion signals which were isolated from those of other species. The model quantifies the 'sensitivity' of the dispersion signal in a way that includes the strength of the molecular transition as well as the effects of interferences from other atmospheric species. This analysis found a number of potential spectral regions which could be used for NH_3 detection in a future eddy covariance sensor.

Figure 1. an example of modelled transmission spectra. The top panel shows the transmission through a 30 m optical path inside an MPC, with all atmospheric species included. The lower panel shows the same spectrum, but this time only with NH3 included. The optimal molecular resonance in this spectral region is the one centred on 1084.6 cm^{-1}. This resonance leads to a strong feature that is well separated from interfering features from other atmospheric species. The phase dispersion modelling identified this as a good region for CLaDS detection, as well as the spectral region around 1103 cm^{-1}.

As well as the modelling work on NH_3, through discussions between the project partners, it became clear that it would also be interesting to investigate the possibility of detecting other atmospheric species. In particular, methane (CH_4) was identified as having the potential for future work. CH_4 is a major greenhouse gas, and it is very important to be able to measure CH_4 emissions. STFC already has expertise in CH_4 detection through previous instrument development work in the SG. Through discussions with the project partners, Thomas Wall (STFC) described the SG's previous CH_4 sensing work. In particular, the SG has experience of using mature telecoms technology for atmospheric CH_4 sensing. Discussions between the partners found that there is exciting potential to develop a CH_4 sensor based on relative low-cost near-infrared telecomms devices, and that this could be very well suited to relaxed eddy covariance, the form of eddy covariance that has been studied by the Cambridge partners in this project.

<u>(2) design and characterisation of a prototype MOREC-AQ unit</u>

The MOREC-AQ unit is based on relaxed eddy accumulation (RA), a micrometeorological technique used for flux measurements. This technique does not require fast time response instrumentation needed for classic eddy covariance. One of the novelties in our approach focuses on the exploitation of low-cost sensors (LCS) as the detector in place of high-grade instrumentation typically used in RA research. This will lower the cost and make the final unit more portable than the classic RA method. Another innovation is to make the design modular such that it can be applied for flux applications (using fast response vertical wind measurements) or source charaterisation when coupled to a 2-D horizontal wind device. The latter configuration can be used when trying to acquire data for source apportionment



studies. Schematics of the instrument layout are shown in Figure 2.

Figure 2. Schematics of prototype modular relaxed eddy instrumentation. The three modular units that comprise the design are shown in the figure.

One potential challenge of LCS in flux studies is related to the resolution of these devices. This effect was modelled by simulating a typical relaxed eddy covariance signal response from a 5 Hz high-precision device at BT tower London (data James Lee, York 2021). Based on this analysis, a typical minimal signal resolution required to resolve emission fluxes for NO_2 was in the range of 2-6 ppb (Figure 3 b-c). Real-world data from a low-cost sensor show that such resolution can be achieved for similar species (figure 4). This real-world signal show that LCS can resolve distinct signals with resolution on the order of 1-5 ppb, like the signal resolution simulated in Figure 3(b-c).

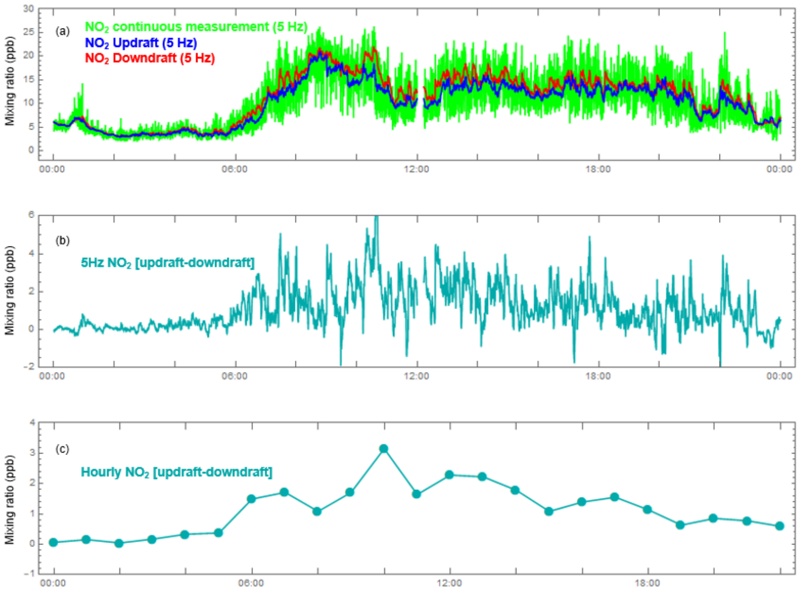


Figure 3. Example data for NO_2. (a) continuous real data from high grade reference instrument at 5Hz (green), simulated signal expected in updraft and downdraft component (blue and red respectively), (b) difference between the updraft and downdraft signals (a measure of flux), (c) hourly averages of the signal shown in (b).

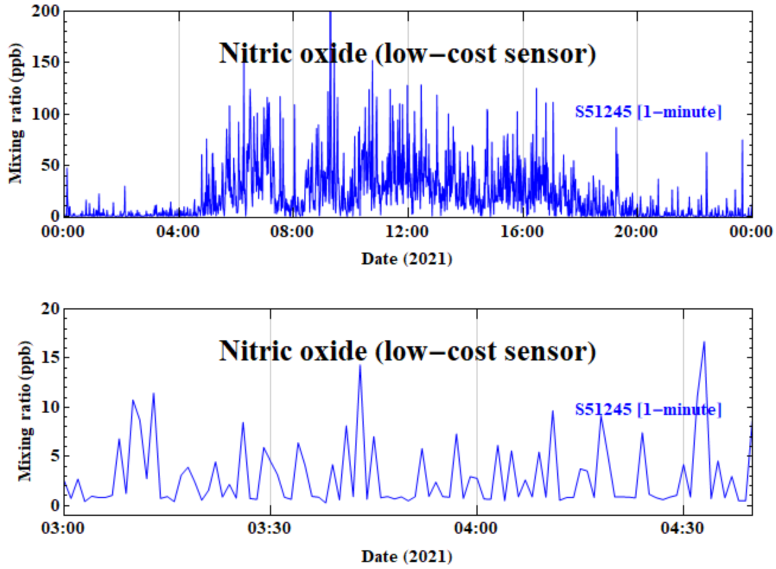


Figure 4. Sample data for real-world 1-minute NO data over a 24hr period (top) and zoomed in on the period 03:00 to 04:40, highlighting the magnitude of the early morning peak signals (bottom). Measurements are at ~4m above street level.

<u>(3) Exploring additional funding opportunities to further develop and optimise the prototype MOREC-AQ unit</u> We are actively engaging with stakeholders with the prospect of seeking further funding opportunities.
What are the next steps for this research? Will you be applying for further funding? What will you need to continue researching this topic?
We aspire to build and test an experimental prototype MOREC-AQ unit, translating the findings from this scoping study. Specifically focusing on critical testing of the three modules: wind, switching and detection module. We intend also to model additional atmospheric species like CH_4 through the STFC RAL Space Spectroscopy Group. We are actively seeking new funding mechanisms to pursues these activities.
Please outline the role of STFC in this project
The STFC RAL Space Spectroscopy Group (SG) contributed to this project through spectroscopic modelling using dedicated SG modelling and analysis software codes. The software was adapted to the MOREC-AQ project and used to ascertain optimal spectral regions for ammonia detection in a potential future eddy covariance instrument. Thomas Wall (SG) performed all the modelling. At the heart of this work was the forward model of CLaDS, a technique that has been developed for atmospheric sensing by the SG, and which is a key technology that the SG uses in many of its instruments. Furthermore, SG expertise in mid-IR lasers and optics contributed to the modelling work carried out in this project.
Please list a brief list of all outputs and impacts below. These may include papers, articles or blogs, presentations at events or conferences, meetings about future plans for the research. Please include links wherever possible
● The project was showcased in SAQN blog 2021. ● We aim to present some of the findings in network workshops including upcoming SAQN workshop in 2022.
Were there any unexpected outcomes as part of the project?
● Selected relevant gas species feasibility assessed, careful thought required for low-cost PM due to large error associated with estimating number density larger particles by OPC method ● In the current configuration, the NH_3 simulation looked at an open path system owing to surface deposition that might affect closed path sampling and sensing ● Current SG expertise in CH4 detection using telecomms components could be well suited to a future Relaxed Eddy Covariance sensor.
What are your plans to share the outcomes of this research with others? (Give details of any future meetings, conferences, papers or other dissemination planned)
Upcoming SAQN meeting (May 2022), and conference presentations.
Project Impact: What is the most significant output/impact from this project?
● Demonstrated the feasibility and applicability of cost-effective method for emission

flux research

- Shown the viability of cost-effective laser-based instrumentation for high-precision atmospheric observation
- Established collaborative activities between STFC, universities and other stakeholders in air quality research, including the UK Environment Agency.