

# Ireland's Atmospheric Composition and Climate Change Network

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## ENVIRONMENTAL PROTECTION AGENCY

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- the contained use and controlled release of Genetically Modified Organisms (*GMOs*);
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- Office of Environmental Enforcement
- Office of Evidence and Assessment
- Office of Radiation Protection and Environmental Monitoring
- Office of Communications and Corporate Services

The EPA is assisted by an Advisory Committee of twelve members who meet regularly to discuss issues of concern and provide advice to the Board.

**EPA RESEARCH PROGRAMME 2014–2020**

# **Ireland's Atmospheric Composition and Climate Change Network**

**(2013-CCRP-FS.15)**

## **EPA Research Report**

Prepared for the Environmental Protection Agency

by

National University of Ireland Galway

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## **ACKNOWLEDGEMENTS**

This report is published as part of the EPA Research Programme 2014–2020. The EPA Research Programme is a Government of Ireland initiative funded by the Department of the Environment, Climate and Communications. It is administered by the Environmental Protection Agency, which has the statutory function of co-ordinating and promoting environmental research.

The authors would like to acknowledge the members of the project steering committee and reviewers, namely John McEntagart (Environmental Protection Agency), Margaret Ryan (Met Éireann) and Brett Kuyper (University of the Western Cape, Cape Town, South Africa).

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This report is based on research carried out/data from 2015 to 2018. More recent data may have become available since the research was completed.

The EPA Research Programme addresses the need for research in Ireland to inform policymakers and other stakeholders on a range of questions in relation to environmental protection. These reports are intended as contributions to the necessary debate on the protection of the environment.

**EPA RESEARCH PROGRAMME 2014–2020**  
Published by the Environmental Protection Agency, Ireland

ISBN: 978-1-84095-962-8

November 2020

Price: Free

Online version

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# Executive Summary

- The Atmospheric Composition and Climate Change network (AC3) is an established valuable national research and monitoring infrastructure that is being developed incrementally and which monitors greenhouse gases (GHGs) and short-lived climate forcers in line with best practice from both pan-European and global monitoring programmes. GHG measurements are undertaken under the umbrella of the ICOS (Integrated Carbon Observation System) pan-European monitoring infrastructure.
- Station infrastructure, instrumentation, visualisation and information technology infrastructure have all been significantly improved.
- Ancillary measurements are now well established at the monitoring sites (black carbon, particle number, aerosol composition, aerosol size distribution and ozone) and these measurements allow GHG data to be extremely well characterised in terms of differences between regional and local pollution.
- Significant redevelopment of the Carnsore Point site is currently under way. This involves a new laboratory facility and utilisation of a 60-metre tower for sampling. This will help to ensure more complete sampling of regional air masses.
- Data from the GHG network have recently been used to deduce high-resolution bottom-up estimates of Irish methane emissions for the year 2012. Data from this network will facilitate multiyear estimates for both methane and carbon dioxide emissions.
- An EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe) campaign characterising the performance of the source apportionment algorithm of the AE33 aethalometer has recently been undertaken and the data are currently under evaluation.
- Given both the national and international importance of climate change, it is critical to maintain a level of investment in infrastructure, analytical systems and associated complementary measurements to ensure that Ireland is at the forefront of this critical area.



# 1 Introduction

The Environmental Protection Agency (EPA) Atmospheric Composition and Climate Change network carries out measurements of the greenhouse gases (GHGs) CH<sub>4</sub> and CO<sub>2</sub>, under the preparatory phase of the ICOS (Integrated Carbon Observation System) programme, and short-lived climate forcers (SLCFs), as well as EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe) monitoring, at three Irish sites [Malin Head (Co. Donegal), Carnsore Point (Co. Wexford) and Mace Head (Co. Galway)], with a number of ancillary sites carrying out EMEP monitoring only (Oak Park, Co. Carlow, and Johnstown Castle, Co. Wexford). Characterisation of SLCFs includes black carbon (mass) measurement and aerosol characterisation [size distribution (scanning mobility particle sizer – SMPS), particle number (condensation particle counter – CPC), scattering (nephelometer), mass (tapered element oscillating microbalance – TEOM) and speciation (aerosol chemical speciation monitor – ACSM)].

Table 1.1 details the measurements being undertaken at the different sites.

## 1.1 EMEP

The Convention on Long-range Transboundary Air Pollution (CLRTAP), signed in 1979, is one of the central means for the protection of our environment. It establishes a broad framework for co-operative action on reducing the impact of air pollution and sets up a process for negotiating concrete measures to control emissions of air pollutants through legally binding protocols. In this process, the main objective of EMEP is to regularly provide governments and subsidiary bodies under the CLRTAP with qualified scientific information to support the development and further evaluation of international protocols on emission reductions negotiated within the Convention. Initially, EMEP focused on assessing the transboundary transport of acidification and eutrophication. The scope of the programme has since widened to address the

**Table 1.1. Location and height of the instrumentation deployed at the different sites**

Site	Location	Height	Instrumentation
Carnsore Point	52° 10' N, 6° 21' W	9 m	<ul style="list-style-type: none"> <li>• GHG measurements (CH<sub>4</sub> and CO<sub>2</sub>) (G1301 analyser)</li> <li>• ICOS-compliant meteorology</li> <li>• Black carbon (aethalometer – AE16 and AE33)</li> <li>• Ozone</li> <li>• CPC</li> <li>• PM<sub>10</sub> mass (TEOM)</li> <li>• Nephelometer</li> <li>• ACSM</li> <li>• PM<sub>10</sub> chemistry (HVS – high-volume sampler)</li> </ul>
Malin Head	52° 22' N, 7° 20' W	22 m	<ul style="list-style-type: none"> <li>• GHG measurements (CH<sub>4</sub> and CO<sub>2</sub>) (G1301 analyser)</li> <li>• G2401 analyser being tested</li> <li>• ICOS-compliant meteorology</li> <li>• Black carbon (aethalometer – AE33)</li> <li>• Ozone</li> <li>• CPC</li> <li>• PM<sub>10</sub> chemistry (HVS – high-volume sampler)</li> <li>• SMPS/FIDAS (Fine dust measurement device)</li> </ul>
Mace Head	53° 19' N, 9° 54' W	11 m	<ul style="list-style-type: none"> <li>• GHG measurements (CH<sub>4</sub> and CO<sub>2</sub>) (G1301 analyser)</li> <li>• Black carbon (aethalometer – AE33)</li> </ul>

PM<sub>10</sub>, particulate matter ≤ 10 µm.

formation of ground-level ozone and, more recently, emissions of persistent organic pollutants, heavy metals and particulate matter. The co-ordination and intercalibration of chemical air quality and precipitation measurements are carried out at the Chemical Coordinating Centre (CCC). New monitoring capacity for Ireland was established as part of the Irish contribution to EMEP monitoring under the CLRTAP. A full description of this measurement network, as it was originally configured, is provided by Leinert *et al.* (2008).<sup>1</sup> Although measurements of all parameters did not begin simultaneously, most measurements were fully operational by 2005. The aerosol measurements included atmospheric aerosol total number concentration ( $\text{cm}^{-3}$ ), black carbon mass loading ( $\text{ng m}^{-3}$ ), total particulate mass ( $\text{PM}_{10}$  – particulate matter  $\leq 10 \mu\text{m}$ ) and aerosol light-scattering coefficient, with daily high-volume bulk filter samples collected for  $\text{PM}_{10}$  chemical analysis. Precipitation samples were collected at three of the inland stations, Oak Park, Johnstown Castle and Glenveagh National Park, and some gas phase measurements were conducted for a time at Carnsore Point and Malin Head. This yielded a large data set of varied parameters from multiple measurement stations over about 5 years since the programme began, the analysis of which is the subject of this report. Statistical analyses of the data provide a good estimate of the variability in and background values of black carbon and aerosol total number concentration. Similar analyses of the  $\text{PM}_{10}$  aerosol chemical composition data obtained from the bulk filter samples yield values for some of the aerosol constituent species.

## 1.2 Climate Change

### 1.2.1 Background

Climate change is recognised as one of the most challenging problems facing humanity. The Intergovernmental Panel on Climate Change (IPCC, 2014) has stated that “warming of the climate system is unequivocal” and that the “human influence is clear”. The 2015 Paris Agreement established the global policy response to climate change. A key objective of that agreement is that global GHG emissions are

balanced with removals during the second half of this century. The national policy position is to reduce emissions of  $\text{CO}_2$ , the main GHG implicated in global warming, by 80% relative to 1990 emissions and to achieve neutrality for the agriculture and land use sector by 2050. Achievement of both the national and the global pathways will require implementation of a strong sectoral and cross-sectoral suite of policies, as well as increased understanding of emissions and removals by sinks and the processes by which the latter can be enhanced.

For developed countries, official data on GHG emissions and removals are provided annually through the national inventory process. National inventories are reported to relevant European Union (EU) and United Nations (UN) bodies. However, it is recognised that, while these inventories are robust, they have limitations, particularly in areas such as land use and agriculture. There has also been a push to develop independent analysis of emissions and removals using top-down analysis of observational data, such as those collected at Mace Head. A number of parties have reported top-down analysis along with official bottom-up data in their national inventory reports, e.g. the UK and Switzerland (UNFCCC, n.d.). These countries have found that top-down analysis helps to identify anomalies, such as overestimation of release of refrigerant use in the UK and the distribution of  $\text{CH}_4$  emission sources in Switzerland. This allows them to improve their inventories and consequently better target mitigation policies.

While these examples show the usefulness of top-down analysis, it is recognised that further development is needed to enable the potential of such analysis to be realised fully, particularly for complex gases such as  $\text{CO}_2$  and  $\text{N}_2\text{O}$ . Historically, these issues have been explored in a series of EU research projects, such as CarboEurope and NitroEurope. These research activities pointed to the need for a sustained and harmonised network of GHG observations across Europe that would provide data to support the analysis of GHG emissions and removals to complement and, in cases, supplement official GHG emissions data reported in national inventories to EU and UN bodies. The Global Climate Observing System

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<sup>1</sup> Associated data sets and digital information objects connected to this resource are available at the Secure Archive For Environmental Research Data (SAFER) managed by the EPA (<http://erc.epa.ie/safer/resource?id=7b484605-b039-102b-aa08-55a7497570d3>; accessed 5 October 2020).

(GCOS) implementation plan has also identified these data as essential climate variables. As a consequence, the Integrated Carbon Observation System Research Infrastructure (ICOS RI) was established in 2008 and included in the strategic European Strategy Forum on Research Infrastructures (ESFRI) Roadmap. This became an official European Research Infrastructure Consortium (ERIC) in 2015. The ICOS RI brings together the European high-quality national GHG measurement sites (atmospheric and ecosystems) in 12 European countries through an agreed management, co-ordination, calibration, analysis and support structure.

### 1.2.2 ICOS

The ICOS RI promotes fundamental understanding of the carbon cycle, GHG budgets and perturbations, and their underlying processes. To facilitate this, ICOS provides data from a high-precision, long-term network of stations that measure GHG fluxes from ecosystems and oceans and GHG concentrations in the atmosphere.

The ICOS RI consists of:

- observation sites that meet ICOS operational and instrumental standards;
- a calibration centre for the provision of reference standards for measured gases;
- thematic centres for atmospheric, ecosystem and oceanic observations;
- a data assimilation and processing centre for the management and storage of data;
- a headquarters hosted by Finland.

Figure 1.1 outlines the ICOS governance structure and Figure 1.2 shows data flows and common services.

Through UK, French and EU investments at Mace Head, Ireland has been at the forefront of top-down analysis of regional-scale emissions of GHGs. The EPA supported a pilot study of the potential of enhancing these analyses for industrial gases through complementary measurements at Carnsore Point. This study showed the potential for such a site to provide more accurate analysis of emissions from Ireland, the UK and other nearby European

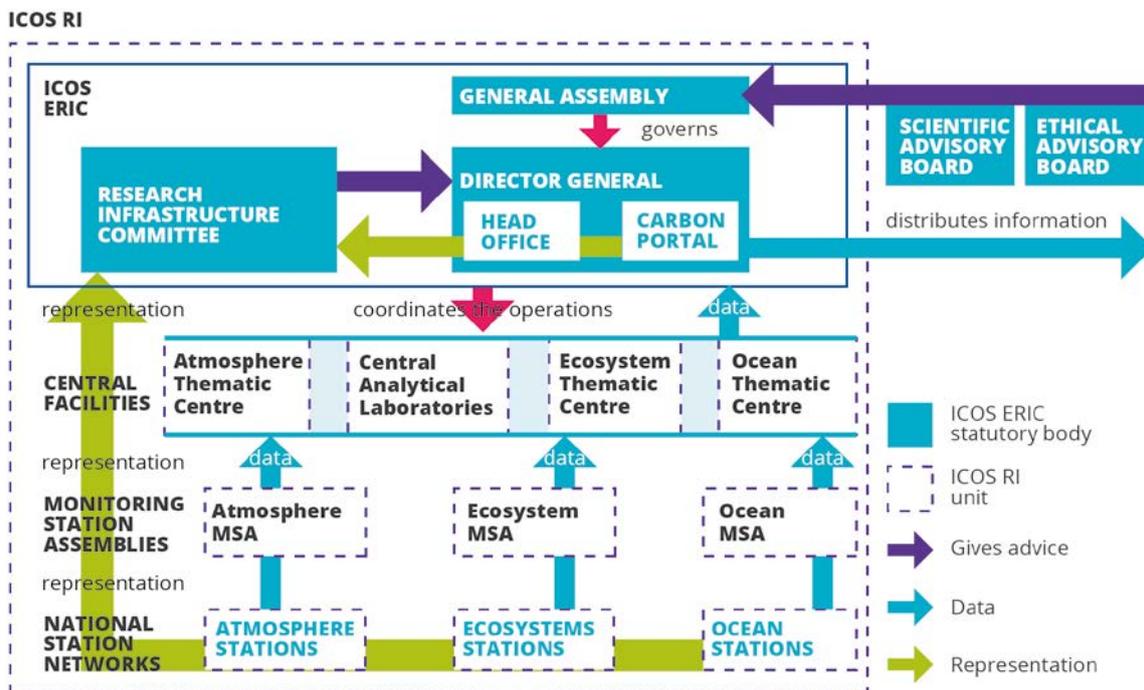
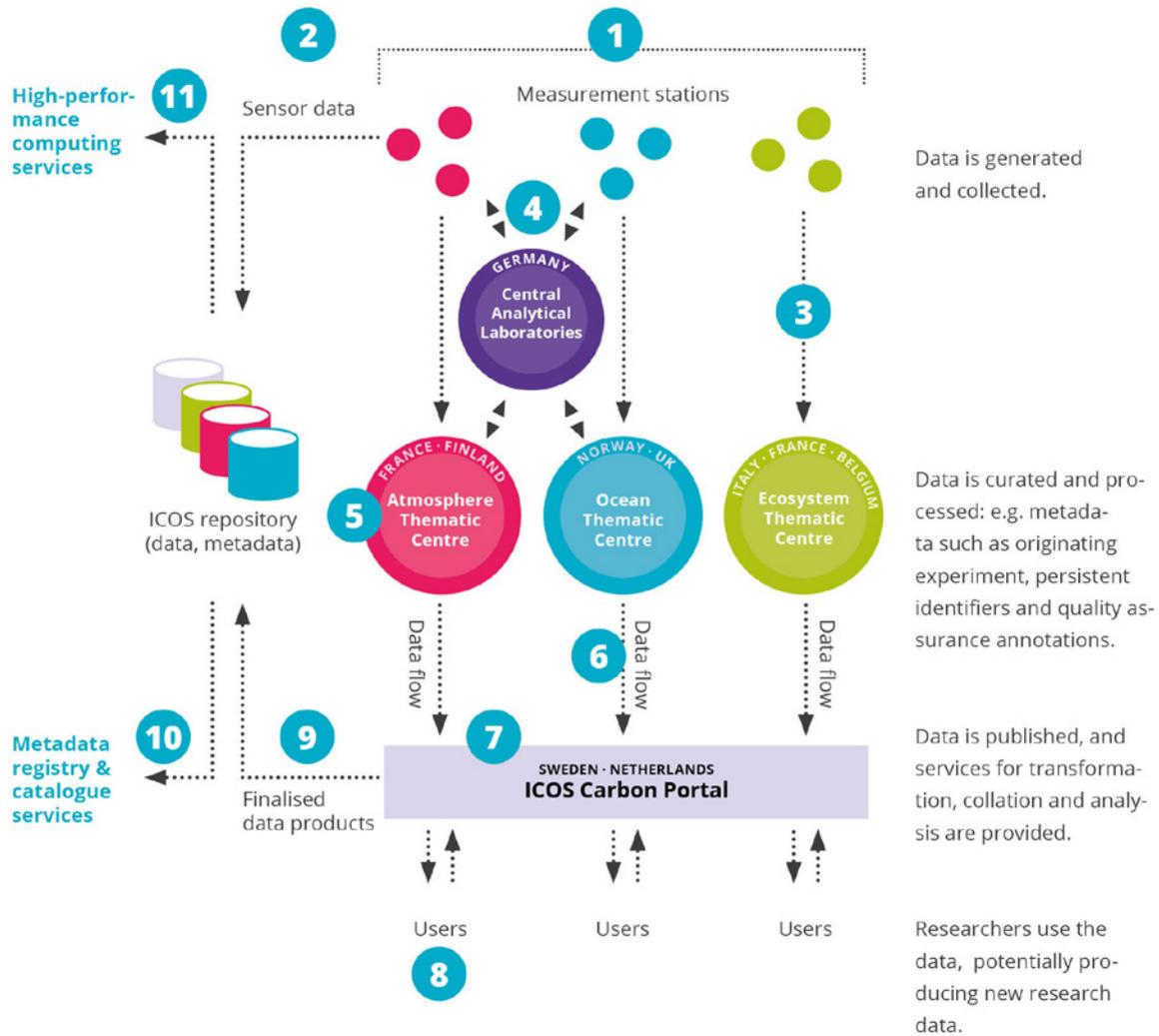


Figure 1.1. ICOS governance structure (<https://www.icos-cp.eu/about/organisation-governance/structure>). ATC, Atmospheric Thematic Centre; Atm, atmosphere; CRL Central Radiocarbon Laboratory; DG, Director-General; Eco, ecosystem; ETC, Ecosystem Thematic Centre; FCL, Flask and Calibration Laboratory; MSA, Monitoring Station Assembly; Oce, ocean; OTC, Ocean Thematic Centre; RI COM, Research Infrastructure Committee. ICOS data are licensed under the Creative Commons Attribution 4.0 International licence (<https://creativecommons.org/licenses/by/4.0/>). ICOS RI, licensed under CC4BY.



**Figure 1.2. ICOS data flows and common services** (<https://www.icos-cp.eu/data-services/data-collection/data-flow>). ICOS data are licensed under the Creative Commons Attribution 4.0 International licence (<https://creativecommons.org/licenses/by/4.0/>). ICOS RI, licensed under CC4BY.

regions. Subsequently, the EPA has supported the development of measurements of CO<sub>2</sub> and other GHGs at Carnsore Point and Malin Head, which, with Mace Head, effectively triangulate Ireland. Other groups, for example at Teagasc, Coford/Department of Agriculture, Food and the Marine, Bord na Móna and a number of universities, have established terrestrial flux measurements at ecosystem sites around Ireland. In combination, these provide a powerful observational network to develop the necessary models and tools to constrain emissions and removals from terrestrial systems. Because of its scale, location and geography, Ireland provides an excellent platform to develop and verify these models and tools. Doing so is necessary as key questions remain to be addressed if Ireland is to achieve its climate goals, such as whether terrestrial systems are a sink or a source for CO<sub>2</sub>

and what the impact of management systems and weather/climate volatility is on emissions/removals of GHGs. These questions and analyses are relevant at regional and global scales. However, because of its unique emissions profile and goal of neutrality for the agriculture and land use sectors, Ireland has a specific interest in using its natural advantages to address and resolve these issues, as far as is scientifically achievable. ICOS provides a platform for doing this. Joining ICOS would allow access to the systems and tools required to provide standard data sets that can be used for verification and analysis of official data reported and accounted for at national, EU and UN levels. This would also enable high-resolution analysis of emissions and trends at higher temporal and spatial scales and lead to improved estimates of Ireland's GHG emissions.

### **1.2.3 Scientific rationale**

The synergy between atmospheric concentration measurements and knowledge of local ecosystem fluxes has proved to be effective in reducing scientific uncertainties in carbon assessments. The ICOS RI will enable further enhanced analysis of carbon emissions and removals in a harmonised manner across Europe. ICOS will meet the data needs of carbon cycle and climate researchers, as well as those of the general public and all other relevant stakeholders. ICOS will serve as the backbone for users engaged in developing data assimilation models of GHG sources and sinks, namely reverse (or inverse) modelling, which allows the deduction of surface carbon flux patterns. This work is essential for the agriculture and land use sectors, where the application of international data and analysis may not be appropriate for Ireland and whose emissions dominate Ireland's emissions profile.

Consolidation of network data into an emissions verification system will be required. Accordingly, reporting systems need to be developed in parallel to inventory improvements to a level that captures current and future potential policy interventions. In addition to emissions inventories based on emission factors and activity data, emissions can be determined from atmospheric measurement data in conjunction with an atmospheric transport model that relates emissions to atmospheric concentrations by means of an inversion algorithm. The inversion algorithm adjusts the emissions used in the model to optimise the agreement between the observed and the simulated concentrations. Typically, the inversion involves the combination of observations of atmospheric trace gas concentrations and a priori knowledge of sources and sinks in conjunction with a chemical transport model. The costs here are for producing annual, seasonal and high-resolution maps of GHG emissions for Ireland.

Among other issues, ICOS can help to resolve fundamental issues relating to Ireland's GHG emissions:

- reduce uncertainties in analysis to an acceptable level to enable the nature and extent of the sources and sinks of GHGs in Ireland to be determined;
- assess how meteorological and other factors influence sinks on seasonal to decadal timescales and the interplay between these sinks and management systems;

- highlight geographical areas with high levels of uncertainty in bottom-up analysis of gases such as N<sub>2</sub>O and CH<sub>4</sub>;
- produce an independent integrated and comprehensive analysis of emissions and removals in Ireland in the context of a GHG neutrality goal for the agriculture sector.

In addition to investments by the UK, France and the USA in observational systems at Mace Head, the site is also of strategic interest to the European Space Agency and linked with Earth observations work in the EU Global Monitoring for Environment and Security (GMES)/Copernicus programme. This enables researchers in Ireland to participate in the most advanced global research programmes in this area and provides Ireland with unique opportunities to develop systems and tools for GHG emissions analysis. While these can have global uses, particularly in the areas of agriculture and land use, strategic and sustained investment is needed to provide observational data from strategic locations, and for key land use types/ecosystems, in a sustained manner, which are of high quality and comparable with international standards. It is also essential that analysis of these data is focused on issues in Ireland and on positioning groups in Ireland to avail of opportunities that will arise in this area.

### **1.2.4 Strategic and policy rationale**

The EPA hosted a meeting of ICOS stakeholders in June 2015, with the consensus outcome being that the provision of sustainable high-quality top-down analyses of key GHG emissions as operational products from analysis of atmospheric measurements is now a realistic goal. There is therefore a major opportunity to provide:

- independent verification of official inventories produced for the United Nations Framework Convention on Climate Change and EU; and
- a more detailed temporal and spatial analysis of emissions and removals.

The development of a pan-European GHG observational network under ICOS is central to these initiatives. The meeting concluded that it is in Ireland's interest to be part of these activities, i.e. that national research activities are linked to work in the UK and other European countries, either bilaterally or through

the Joint Programming Initiative and Horizon 2020 processes. The national observation network must therefore meet the various criteria and standards that are established for such observations. This may be readily accomplished and requires up to seven sites in Ireland to be designated as ICOS sites. The road map milestones required to advance Ireland's participation in the EU ICOS RI are outlined in the recommendations in this report.

ICOS requires participating countries to:

- establish and maintain nominated sites to the ICOS standard for an initial period of 5 years;
- contribute to the overall costs of the ICOS ERIC;
- participate in ICOS governance and development processes.

Participation in the ICOS ERIC therefore requires a commitment to capital investment in the establishment of the designated sites and to the ongoing operational costs. Ireland would have membership of the ICOS Governing Board and would be able to influence the development of ICOS. It would also enable involvement with the technical working groups. The costs therefore consist of the:

- costs of membership;
- costs of site establishment;
- operational and maintenance costs.

Membership costs are determined by the number and level of sites that are designated as ICOS sites.

## 2 Instrumentation and Experimental Protocols

### 2.1 Description of Current ICOS Instrumentation

#### 2.1.1 Background

Cavity ring-down spectroscopy (CRDS) is a laser-based technique developed in the 1980s and now well established as a highly sensitive absorption measurement technique. The CRDS technique can be simply described as follows. A laser source is used to excite a ring-down cavity, which consists of a low-loss optical resonator composed of at least two concave high reflectivity mirrors. As the injected laser pulse propagates back and forth between the mirrors, part of it is retransmitted at each pass outside the cavity. A photosensitive detector located behind one of the mirrors monitors the time decay of the laser pulse (known as the ring-down time). The ring-down time depends on the cavity loss but also on the presence of any absorbing species inside the cavity. Thus, the more absorber there is within the cavity, the shorter the ring-down time will be. The absorber concentrations are directly deduced from the measurement of the

characteristic decay time constant. In contrast to conventional absorption spectroscopy, CRDS offers two main advantages. First, because of the multiple reflections inside the optical cavity, CRDS works with 10–1000 times longer optical path lengths than traditional techniques. Second, as it is based on a decay time measurement, CRDS is not affected by the light source intensity fluctuations. Over the last decade, this technology has been developed for measurement of atmospheric trace gases such as CO<sub>2</sub>, CH<sub>4</sub>, NH<sub>3</sub> or CO (Majcherova *et al.*, 2005).

#### 2.1.2 Instrumentation

The Picarro G1301 analyser has been designed to continuously measure CO<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>O concentrations in air. The CO<sub>2</sub> concentrations are corrected based on the measurement of the water vapour content in the samples. The instrument consists of two modules, the data acquisition system (DAS) and the computer pump vacuum unit (CPVU) (Figure 2.1). The DAS module includes the optical

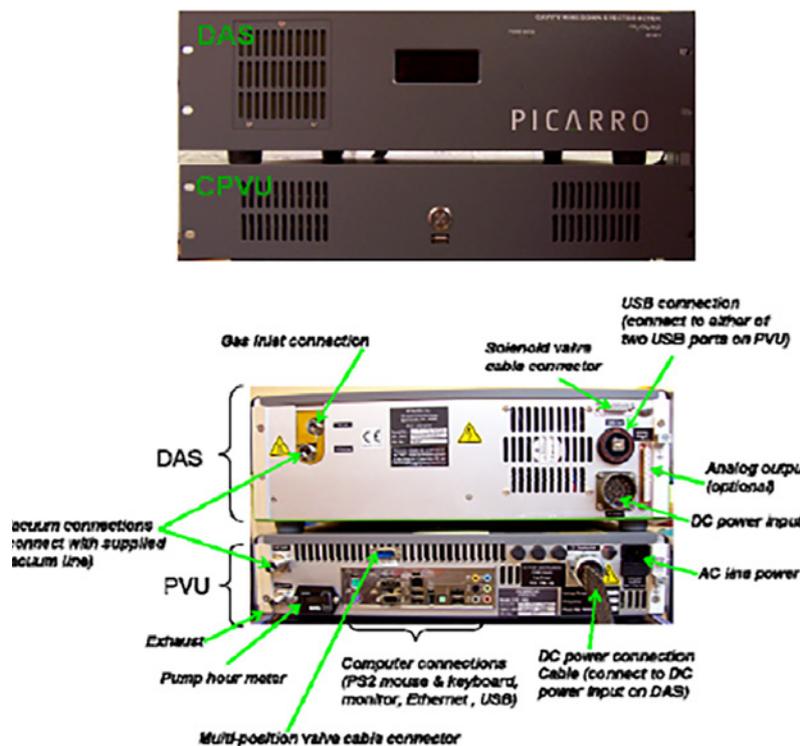


Figure 2.1. Front and back panels of the Picarro G1301 analyser.

bench and all of the electronics required for process regulation and data collection. The CPVU module contains a pump to suck the air into the optical cavity, different power supplies and a computer to run and store the measurements. The instrument is provided with six +12V power supplies that can be used to switch solenoid valves. A serial port is also included for the control of a multi-position valve. The data acquisition is fully automatic. Once the control computer is switched on, measurements are started. The CO<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>O concentrations are recorded every 5 seconds. The power consumption of the G1301 instrument is about 350W during normal operation.

Two telecom-grade distributed feedback laser diodes are used, one to measure a single <sup>12</sup>CO<sub>2</sub> spectral feature at 1602nm and the other to measure <sup>12</sup>CH<sub>4</sub> and H<sub>2</sub>O spectral features at 1651 nm (Figure 2.2). Light from the laser diodes is selected using an optical switch and is transported to a wavelength monitor, which allows a precise wavelength measurement

( $\pm 0.0001 \text{ cm}^{-1}$ ) over a range corresponding to greater than 150nm. The V-shape optical cavity is composed of three high-reflectivity mirrors situated within a 35-cm<sup>3</sup> vacuum chamber. Both pressure inside the cavity ( $140 \pm 0.2 \text{ torr}$  (0.2 bar)) and temperature ( $45 \pm 0.015^\circ\text{C}$ ) are precisely maintained, depending on the configuration of the instrument. When the frequency of the laser diode matches one of the optical cavity transmission resonances, a large intracavity build-up of energy occurs and a threshold circuit allows the laser to be switched off so that a ring-down decay time can be measured. Based on wavelength-scanned CRDS, the analyser continuously generates optical spectra of the gas sample within the cavity. By varying the laser diode emission, ring-down times are measured as a function of the laser wavelength. Each time the wavelength of the injected light matches an absorption feature of any absorber species in the cavity, the decay time increases proportionally to the species' concentration. Concentrations are thus deduced from the absorption line shapes (Figure 2.3).

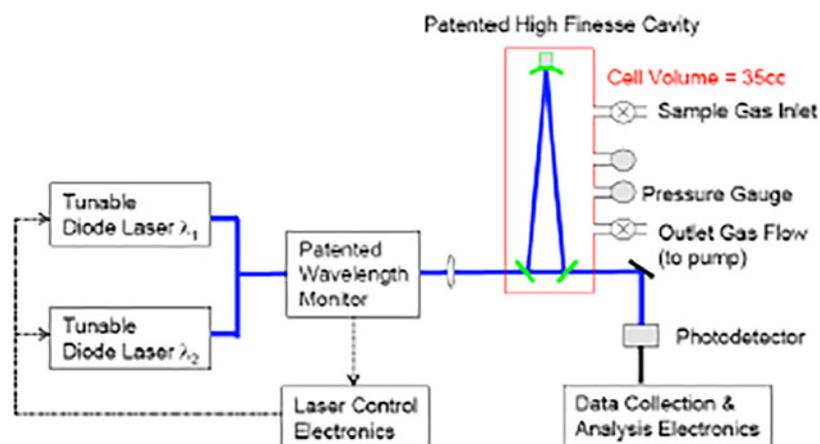


Figure 2.2. Schematic description of the Picarro G1301. Source: Tan *et al.* (2004). Reproduced with permission of Sze Tan.

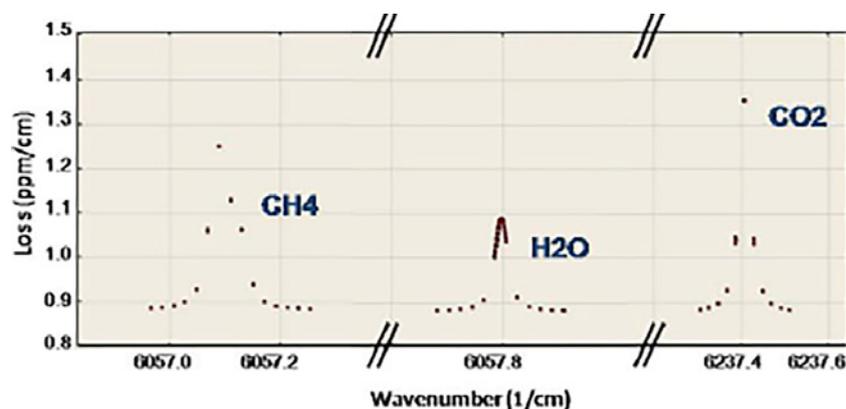


Figure 2.3. Absorption spectra for CH<sub>4</sub>, H<sub>2</sub>O and CO<sub>2</sub> as measured by the G1301 instrument.

The deployment of the Picarro G1301 analyser in the field required a connection scheme similar to that shown in Figure 2.4. The set-up consists of an air inlet combined with a primary pump to ensure a sufficient flush of air through the tubing line. Filters are used to prevent air particles from entering the analyser. Moreover, calibration cylinders with precisely known concentrations are needed for regular calibration of the instrument and an extra "target" cylinder is required for short-term quality control (the cylinder concentration is measured regularly by the instrument). To automatically select the gas sample to be measured, a multi-position valve controlled directly by the G1301 instrument has been added. To protect the instrument during power failures, the use of an uninterruptible power supply is recommended.

#### Air inlet

The air inlet is a stainless steel unit designed by the French company Dinamic. The air inlet is mounted on the roof of the laboratory (or on a mast at Mace Head) and connected to a 1/2" Dekabon tube (polyethylene-aluminium composite) that goes down inside the laboratory (Figure 2.5). The Dekabon tube (also called Synflex) is marked at Mace Head to distinguish it from the many other inlet lines.

#### Decanting bowl (Carnsore Point and Malin Head)

At Carnsore Point and Malin Head, where the length of the air inlet tubing is short (< 10 m), a decanting bowl is used to prevent liquid water entering the instrument. A decanting bowl is not used at Mace Head as the length of the air inlet tubing is so long (> 50 m) that it minimises the risk of water propagation. The decanting bowl consists of a 180-ml stainless steel/glass settle housing, with three 1/4" national pipe thread outlets (3S1NRGL2, EIF Filters). The bottom outlet can be used to drain the bowl when required (the top inlets are used for sample flow in and out). To automate the drain, a solenoid valve (B2DK1026, www.parker.com) was used that can be switched on directly from the Picarro G1301 instrument (Figure 2.6).

#### Filters and flow splitter

To minimise the response time of the instrument (time needed to see a change in ambient concentration), a primary pump (N86KT.18, KNF) is used that continuously flushes the inlet line at about 3 l minute<sup>-1</sup> (at Malin Head an ozone monitoring instrument equipped with a pump is used). The main air stream is split using a stainless steel T-piece (SS-400-3, Swagelok). One part is connected to a multi-position valve upstream from the G1301 instrument and the

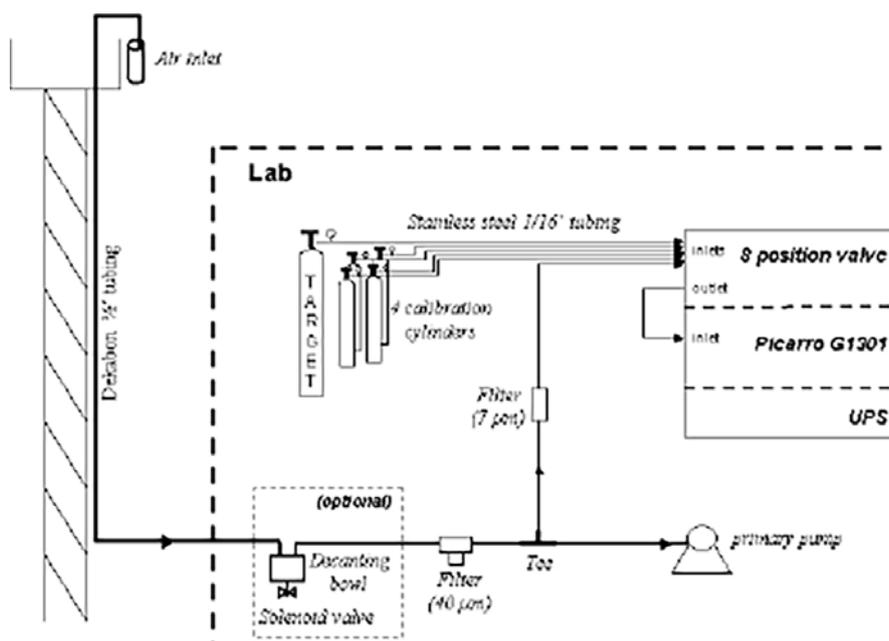


Figure 2.4. Schematic description of the experimental set-up. UPS, uninterruptible power source.



**Figure 2.5. Air inlet at Carnsore Point.**

other part is connected directly to the primary pump. At Mace Head the pump is located in a specific room used to house all instrument pumps and 40- $\mu\text{m}$  filters (SS-4TF-40, Swagelok) and 7- $\mu\text{m}$  filters (SS-4F-7, Swagelok) are used to protect the pump and the multi-position valve from dust. The only purpose of the 7- $\mu\text{m}$  filter is to improve the protection of the valve (Figure 2.7).

#### *Calibration and “target” cylinders*

Originally, four calibration cylinders (10l) were used to regularly calibrate the G1301 analyser, with one “target” (TGT) cylinder (40l) used to check the short-term stability of the instrument. These cylinders were purchased from Steininger. They were filled with dried synthetic air to reflect the ICOS specifications for  $\text{CO}_2$  and  $\text{CH}_4$  mixing ratios. The concentrations of  $\text{CO}_2$  and  $\text{CH}_4$  were precisely measured at the Laboratory for Sciences of Climate and Environment (LSCE, Gif sur Yvette, France) by the RAMCES (Réseau atmosphérique de mesure des composés à effet de serre) group against international reference scales (World Meteorological Organization X2004 for  $\text{CH}_4$  and WMO X2002 for  $\text{CO}_2$ ). The cylinders are used with two-stage pressure regulators from Scott Specialty Gases (05114CD14). Table 2.1 shows the initial network standards; future standards will be made at the ICOS Central Analytical Laboratories.

#### *Multi-position valve*

An eight-position valve (EMTCSD8UW, Valco, <https://www.vici.com/vval/vval.php>) is used to select the gas



**Figure 2.6. Decanting bowl and solenoid valve for automatic purge.**

sample to be measured by the G1301 instrument (Figure 2.8). The valve uses 1/16” Valco fittings and is protected from dust by 2- $\mu\text{m}$  filters (ZUFR1, or ZBUFR1 for the bulkhead version, Valco).

The multi-position valve can be controlled either by a digital input/output connector or by a serial port (see technical note 415, [www.vici.com/support/tn/tn415.pdf](http://www.vici.com/support/tn/tn415.pdf)). No serial port was available to control the Valco valve in the old versions of the Picarro G1301. It was necessary to use the +12V power supplies provided with the instrument to activate relays and then control the digital interface of the valve. Control of the valve is made easier by using the built-in serial port of later versions of the Picarro G1301 and the specific cable to connect directly to the valve (I-22697, ref: Valco). The valve port connection scheme is shown in Table 2.2.

#### *Maintenance*

##### **Leak checks (regularly)**

Leaks in the inlet line can have a critical effect on monitoring. They can interfere with the atmospheric signal causing data spikes. Leaks in the system can result in an unnecessary overconsumption of gas from the cylinders, generating additional expenses.

##### **Gas cylinder pressure control (regularly)**

It is important to regularly check (typically monthly) the pressure of the gas cylinders (Figure 2.9). The cylinders can be expensive and are the link between the instrument and the international  $\text{CO}_2$  and  $\text{CH}_4$  scales. Cylinder pressures are recorded in a logbook.

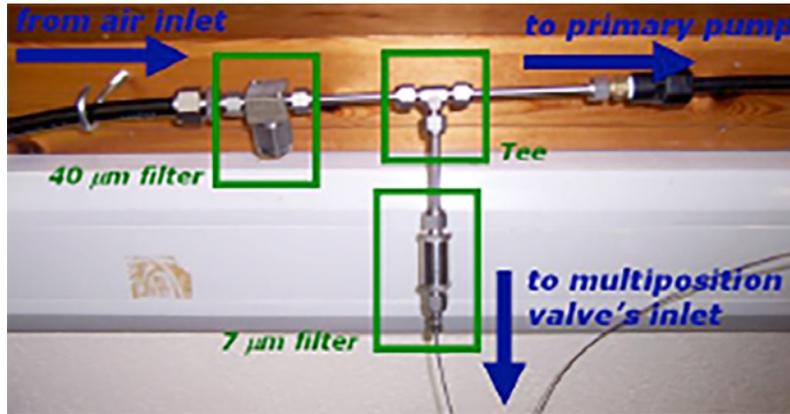


Figure 2.7. Flow splitter description.

Table 2.1. Initial calibration and target cylinder concentrations as measured by the LSCE RAMCES group

Cylinder ID	Analysis date at LSCE	Assigned CO <sub>2</sub> concentration (ppm)	Assigned CH <sub>4</sub> concentration (ppb)	Location
Cal#1 D590434	April 2009	342.02	1715.4	Mace Head
Cal#2 D590443	April 2009	372.15	1813.6	Mace Head
Cal#3 D590429	April 2009	401.66	2009.2	Mace Head
Cal#4 D590424	April 2009	462.21	2164.6	Mace Head
TGT D609286	April 2009	381.11	1889.3	Mace Head
Cal#1 D481376	October 2009	352.30	1689.3	Malin Head
Cal#2 D481365	October 2009	380.70	1799.1	Malin Head
Cal#3 D481420	October 2009	402.31	1992.3	Malin Head
Cal#4 D481387	October 2009	450.56	2190.7	Malin Head
TGT D489534	November 2009	390.96	1911.1	Malin Head
Cal#1 D481366	October 2009	351.02	1698.2	Carnsore Point
Cal#2 D481368	October 2009	381.76	1804.0	Carnsore Point
Cal#3 D481388	October 2009	402.27	1997.1	Carnsore Point
Cal#4 D481390	October 2009	449.83	2192.2	Carnsore Point
TGT D609208	November 2009	391.03	1925.2	Carnsore Point

ppb, parts per billion; ppm, parts per million.

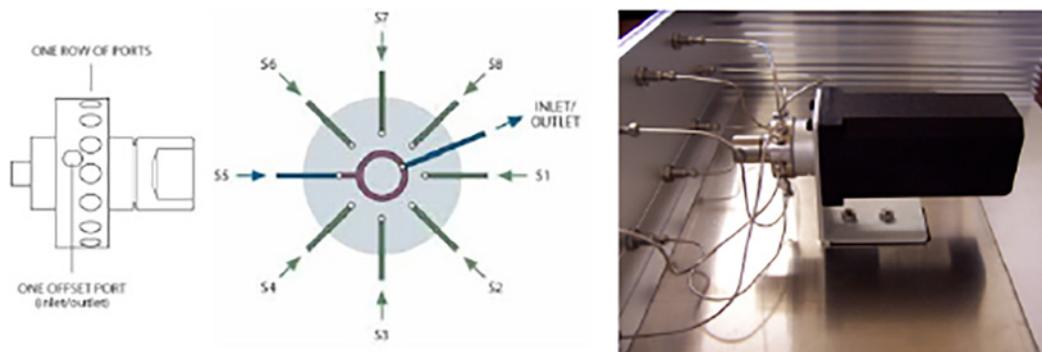
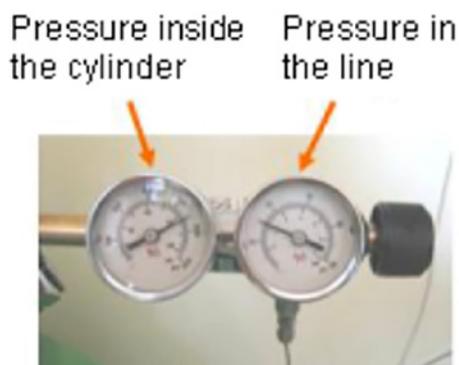


Figure 2.8. Valco valve and schematic description.

**Table 2.2. Connection of the Valco valve ports**

Valve port number	Gas sample
1	Ambient air
2	TGT tank
3 to 6	Calibration tanks #1–#4
7 and 8	Free

**Figure 2.9. Pressure regulator readings.**

Typically, a calibration episode will decrease the total cylinder pressure by 2 bar for a 100 ml minute<sup>-1</sup> flow rate and a measurement time of 180 minutes. For the “target” cylinder, a pressure drop of 0.1 bar for a measurement time of 30 minutes at a flow rate of 130 ml minute<sup>-1</sup> is expected. The lifetime of the calibration cylinders should be >5 years while the lifetime of the target cylinder is approximately 18 months.

Typically, gas cylinders are replaced when the pressure drops below 30 bar. This prevents contamination from surface effects. The expected lifetime of the “target” cylinder is shorter, about 1 year, if it is calibrated every 7 hours for 30 minutes. Once the critical pressure has been reached, the cylinders should be returned for refilling. In the future it is hoped that this will be carried out at the central ICOS laboratory in Jena, Germany, which would incur some costs.

#### Replacement of filters (twice a year)

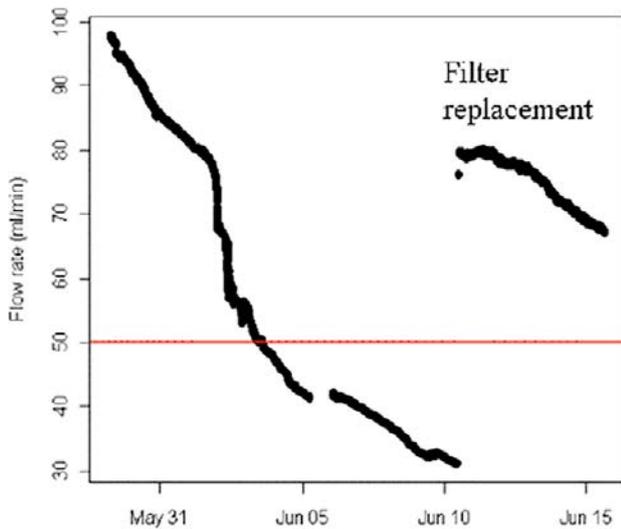
The 40- $\mu$ m, 7- $\mu$ m and 2- $\mu$ m filters are used to protect the pump and the multi-position valve from dust. The optical cavity of the G1301 instrument is also protected by an additional 0.5- $\mu$ m filter located inside the DAS unit. To determine when these filters need to be replaced (this depends on the environment) the

sample gas flow rate exiting the instrument should be assessed. The flow rate is expected to decrease when the filters become blocked. The flow rate can be measured directly at the rear of the instrument with the digital flowmeter, but the Outlet\_Proportional\_Valve variable (in the “UserLog.dat” files) can also be used as a proxy. This variable corresponds to the aperture of the downstream solenoid valve, which is used for pressure regulation inside the cavity. Its value is proportional to the gas flow rate exiting the cavity according to a formula that has to be established for each instrument. For instance, the following relationship holds for the Picarro CFADS-046 instrument at Mace Head: flow rate =  $3 \times 10^{-72}(\text{Outlet\_Proportional\_Valve})^{15.98}$ . By using such a relationship an approximation of the gas flow rate exiting the instrument can be determined and the saturation of the filters can be followed.

With new filters, the typical flow rate for the ambient air sample is around 100 ml minute<sup>-1</sup>. It is recommended that the filters are changed when the flow rate decreases to below 50 ml minute<sup>-1</sup> (shown by the red line in Figure 2.10). To know which filter has to be replaced, the filters should be disconnected one by one (beginning with the larger one) to see what effect each one has on the flow rate. There is no need to replace the entire filter unit, only the filter element inside (SS-4F-K4-40 and SS-4F-K4-7, Swagelok, and 2SR2-10, Valco). The instrument must be thoroughly checked for leaks once the filter elements have been replaced.

#### Valco valve cleaning and replacement (every year)

The Valco multi-position valve should be cleaned regularly, depending on the environment. Dirt and particles can accumulate in the rotor (the moving part of the valve) even if filters are used to protect it. This needs to be carefully examined at least once a year. It must be removed, inspected and replaced if



**Figure 2.10. Example of decreasing flow rate with saturation of the filters at Mace Head.**

necessary. This operation has to be performed with extreme care and requires a degree of technical expertise (see technical note 703, [www.vici.com/support/tn/tn703.pdf](http://www.vici.com/support/tn/tn703.pdf)).

#### Internal pump replacement (every 1.5 years)

The vacuum pump located in the CPVU of the instrument is rated for 10,000 hours, which seems to be realistic based on experience to date (Figure 2.11, marked 'F'). The pump should be replaced every 1.5 years; the meter at the rear of the CPVU indicates the number of accumulated hours of operation. The pumps used are manufactured by KNF but can be purchased only from Picarro (as a result of a commercial agreement between Picarro and KNF). We have recently moved to using external pumps because of the unreliability of the internal pumps. In addition to the external pumps being more reliable, access is easier, which provides better troubleshooting capabilities.

## 2.2 Aethalometer

The newly developed aethalometer model AE33 (Magee Scientific) follows the same basic measurement principle as older models. Aerosol particles are continually sampled on the filter and the optical attenuation is measured with high time resolution from 1 second to 1 or more minutes. Attenuation is measured on two spots with different sample flows and on the reference spot without the flow (Figure 2.12). The black carbon mass

concentration is calculated from the change in optical attenuation at 880 nm in the selected time interval using the mass absorption cross-section of  $7.77 \text{ m}^2 \text{ g}^{-1}$ . At this wavelength, other aerosol particles (carbonaceous or mineral) absorb significantly less and absorption can be attributed to black carbon alone (Sandradewi *et al.*, 2008a). Measurements at distinct wavelengths (370, 470, 520, 590, 660, 880 and 950 nm) allow for spectral analysis of the data – the analysis of the dependence of absorption on the wavelength can be of importance for research on direct effects of aerosol black carbon, climate forcing (Bond *et al.*, 2013) and mineral dust detection (Collaud Coen *et al.*, 2004), through the determination of the single scattering albedo dependence on the wavelength, or source apportionment (Sandradewi *et al.*, 2008b). When the attenuation reaches a certain threshold, a tape advance is induced so that measurements start on a clean spot. The attenuation threshold, first reached at the lowest wavelength of 370 nm, is called ATNTA (with a default value of 120); this can be changed by the user, for example a higher value can be set to conserve the filter tape. However, setting the ATNTA value too high will impact on the measurements and make them excessively noisy at high attenuation levels. The airflow is measured by two mass flowmeters after the aerosol particles are deposited on the filter. The flow can be reported at ambient conditions, conditions set by the user or different standard conditions, with the default being  $20.11^\circ\text{C}$  and  $1013.25 \text{ hPa}$ . Several valves are incorporated in the manifold to allow for bypass (air is not flowing through the filter during the tape advance procedure), clean air (air is filtered before reaching the optical chamber; used during the warm-up procedure and during automated quality control tests) and measurement modes. The flow diagram describing the different flow modes is provided in Figure 2.12.

## 2.3 Condensation Particle Counter

The CPC acts very much like an optical particle counter. However, the particles are first enlarged by a condensing vapour to form easily detectable droplets. The science behind the counter is focused on how to condense the vapour onto the particles. When the vapour surrounding the particles reaches a certain degree of supersaturation, it begins to condense onto the particles. This is called heterogeneous condensation. If supersaturation is too high,

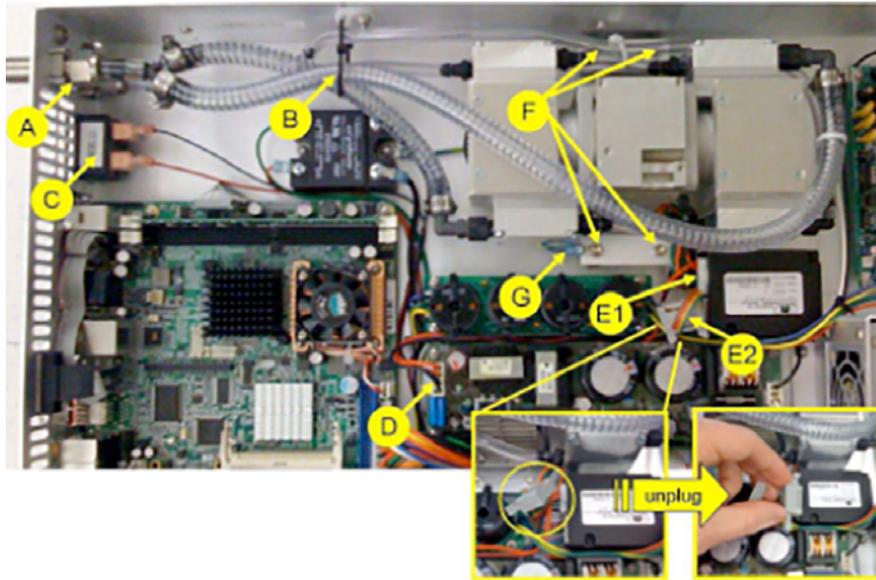
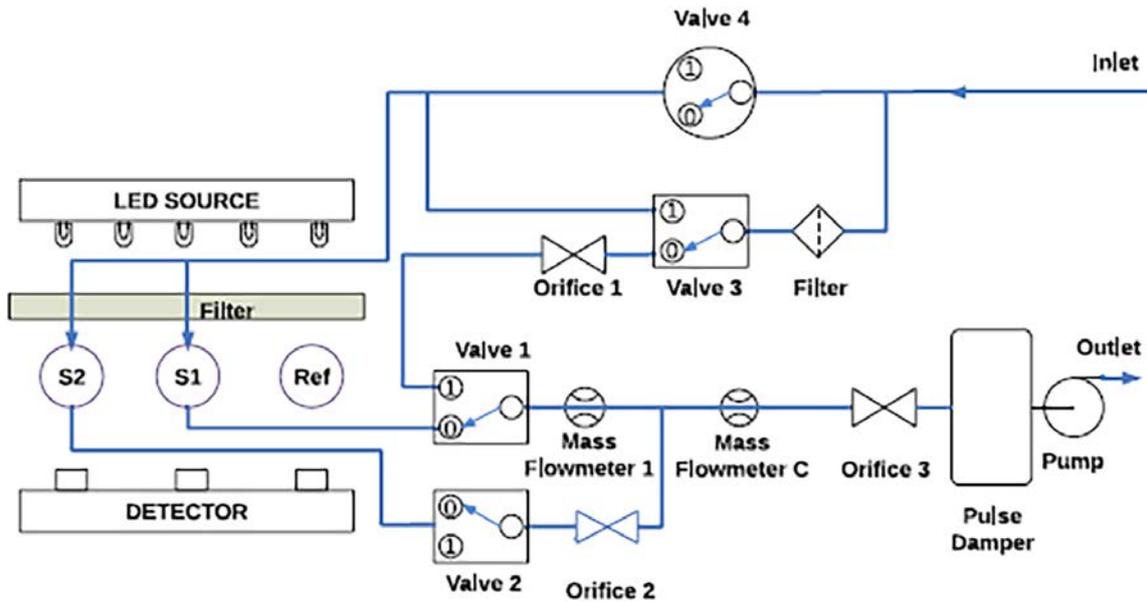


Figure 2.11. Internal pump (F) of the G1301 instrument.



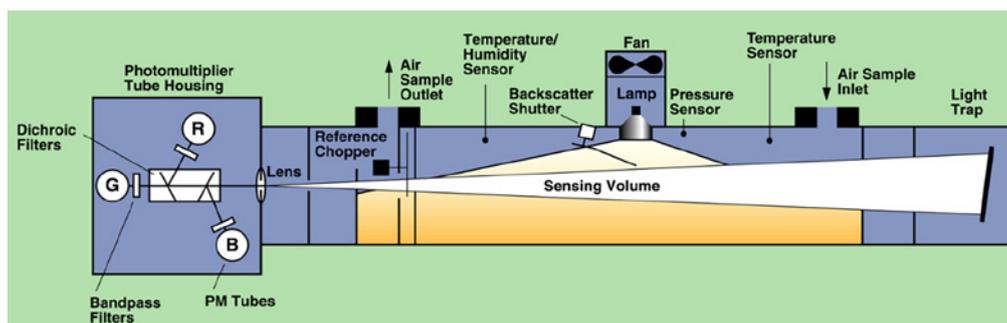
Mode	Valve 1	Valve 2	Valve 3	Valve 4
Bypass	1	1	0	1
Warm-up / clean air	0	0	1	1
Measurement	0	0	0	0
Flowmeter calibration	0	1	0	0

Figure 2.12. Schematic of the AE33 aethalometer.

condensation can take place even if no particles are present. This is referred to as homogeneous nucleation or self-nucleation, whereby molecules of the vapour form clusters that act as nucleation sites as a result of the natural motion of the gas and attractive van der Waals forces. This condition is avoided by accurately controlling the operating temperature.

## 2.4 Three-wavelength Integrating Nephelometer

The nephelometer is an instrument that measures aerosol light scattering (Figure 2.13). It detects scattering properties by measuring light scattered by the aerosol and subtracting light scattered by the gas



**Figure 2.13. Schematic of the nephelometer.** Image provided by NOAA Global Monitoring Laboratory, Boulder, CO (<https://esrl.noaa.gov/gmd/>). Image courtesy of TSI Incorporated, Shoreview, MN, and protected by copyright.

and the walls of the instrument and the background noise in the detector. The three-wavelength model (Integrating Nephelometer 3563, TSI) splits the scattered light into red (700 nm), green (550 nm) and blue (450 nm) wavelengths. The TSI nephelometer also measures backscattered light at these wavelengths. The one-wavelength Radiance Research nephelometer (M903) measures forward scattering at 550 nm only.

The main body of the TSI 3563 nephelometer is a 10-cm-diameter aluminium tube, 90 cm long (Figure 2.14). Along the axis is an 8-cm-diameter tube set with aperture plates, which give the lamp an angular horizontal range from 7° to 170°. The backscatter shutter allows blocking of the angles from 7° to 90° so that only backscattering is measured. The light trap provides a dark reference against which to measure the scattered light.

The receiving optics are located on the other side of the tube from the trap. The light that passes through the lens is separated by dichroic filters into three wavelengths. The first splitter is a colour splitter that passes 500- to 800-nm light and reflects 400- to 500-nm light. The reflected light passes through a filter centred at 450 nm (blue) into a photomultiplier tube (PMT). The light that passes through the first splitter goes to a second splitter that passes 500- to 600-nm light and reflects 600- to 800-nm light. The reflected light passes through a filter centred at 700 nm (red) into a second PMT. The light that passes through both splitters passes through a filter centred at 550 nm (green) into a third PMT.

The reference chopper is used for calibration of the nephelometer. It consists of three zones. The signal zone allows light to pass through unaltered.

The dark zone allows no light to pass and is used to measure the background noise of the instrument. The calibrate zone allows approximately 0.1% of the light to pass and is used to measure lamp stability over time. The chopper rotates 23 times per second (see Figure 2.13).

## 2.5 High-volume Filter Samples

In order to obtain information about the chemical composition of the ambient aerosol particles, high-volume filter samples were collected at Carnsore Point, Oak Park and Malin Head. A DIGITEL model DHA 80 HTD automated high-volume filter sampler deployed at each of these sites collects daily filter samples on 150-mm-diameter Whatman No 41 filters with an exposed area of about 140 mm diameter at a flow rate of approximately 500 l minute<sup>-1</sup>. The instrument is equipped with a PM<sub>10</sub> sample head, which restricts the size of particles sampled to those that have an aerodynamic equivalent diameter of 10 µm or less. Particles present in a daily sample volume of air of about 82.320 m<sup>3</sup> are thus collected on a single filter. These filter samples are collected weekly and returned to Met Éireann for analysis by ion chromatography of a number of well-known aerosol chemical species (SO<sub>4</sub>-S, NO<sub>3</sub>-N, NH<sub>4</sub>-N, Na, Mg, K, Ca and Cl), which yields daily values of mass concentration (µg m<sup>-3</sup>) of these ions.

## 2.6 Precipitation Samples

Precipitation samples are collected by an automated precipitation wet-only sampler (NSA 181, Eigenbrodt). This instrument is equipped with a precipitation sensor that controls the opening and closing of a lid



**Figure 2.14. Components of the nephelometer. Photo credit: Mick Geever.**

on the collecting funnel so that the funnel is open only during precipitation events. This ensures that only precipitation is sampled and excludes dry aerosol deposition from the samples. A rotating base on the sampling funnel directs the flow to one of eight plastic bottles mounted underneath it in a refrigerated compartment; this rotates by one position to a new bottle at midnight each day. The instrument holds nine sample bottles: one for each day of the week, with one

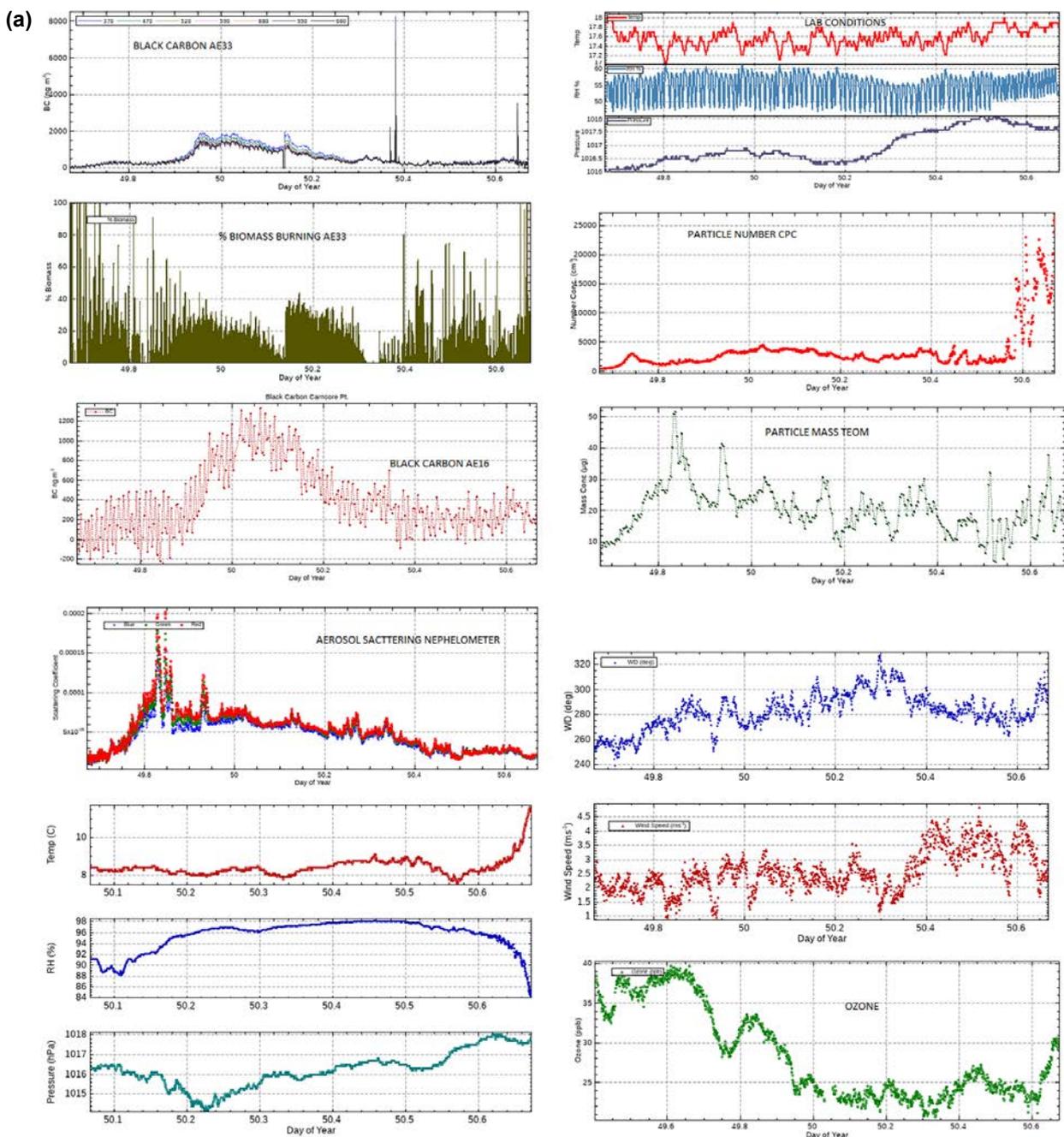
overlap bottle for the day on which the samples are collected and one blank. The samples are collected on a weekly basis and returned to Met Éireann for analysis by ion chromatography of a number of chemical species ( $\text{SO}_4\text{-S}$ ,  $\text{NO}_3\text{-N}$ ,  $\text{NH}_4\text{-N}$ , Na, Mg, K, Ca and Cl) and for measurement of conductivity and pH. This analysis produces daily values of mass concentration ( $\mu\text{g m}^{-3}$ ) of these ions, which form the basic data set obtained from these measurements.

# 3 Visualisation, Infrastructure Development, Instrument Validation and Installation of New Equipment

## 3.1 Visualisation

In order to facilitate continuous operation of the network, a real-time data transfer and visualisation system was put in place. The data streamed from the stations are updated every 10 minutes and transferred to a dedicated server at the National University of

Ireland Galway (NUIG). Automatic scripts upload plotted measurement data as well as instrumental diagnostics to a website to facilitate data inspection and instrument performance (Figure 3.1). Examples of the online data platforms of the instruments at the various sampling sites can be found at the links in the following sections.



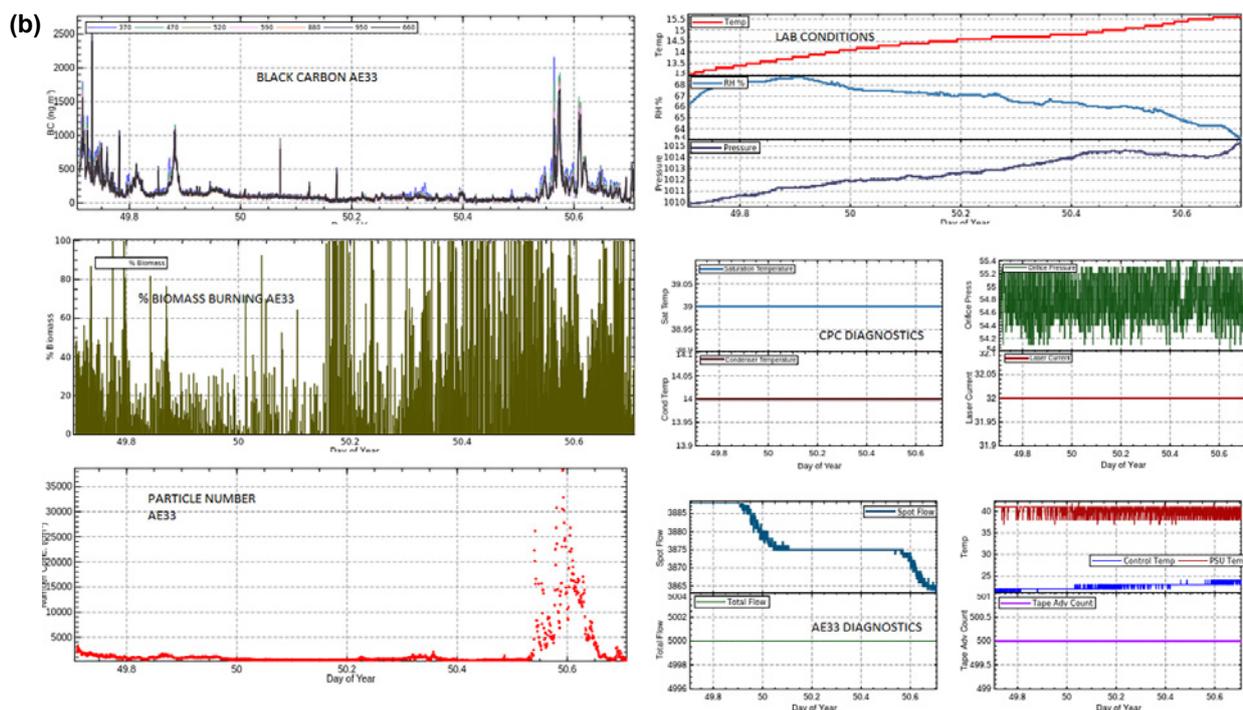


Figure 3.1. Real-time data network visualisation from (a) Carnsore Point and (b) Malin Head.

### 3.1.1 Carnsore Point

- AE33 aethalometer: operational, data transfer and data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=194%3Acrp-aerosol-optical-properties&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=194%3Acrp-aerosol-optical-properties&catid=106%3Atxb&Itemid=30)).
- AE16 aethalometer: operational, data transfer and data visualisation in place (now obsolete; URL not available).
- CPC: installed, operational, data transfer and data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=193%3Acrp-aerosol-physical-properties&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=193%3Acrp-aerosol-physical-properties&catid=106%3Atxb&Itemid=30)).
- TSI nephelometer: operational, data transfer and data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=194%3Acrp-aerosol-optical-properties&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=194%3Acrp-aerosol-optical-properties&catid=106%3Atxb&Itemid=30)).
- TEOM: operational, near real-time (NRT) data transfer and NRT data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=193%3Acrp-aerosol-physical-properties&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=193%3Acrp-aerosol-physical-properties&catid=106%3Atxb&Itemid=30)).
- ICOS-compliant WindSonic wind sensor: operational, data transfer and data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=185%3Acarnsore-pt-met&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=185%3Acarnsore-pt-met&catid=106%3Atxb&Itemid=30)).
- Temperature, pressure, humidity sensors: NRT data transfer and data visualisation in place from previously installed anemometer ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=185%3Acarnsore-pt-met&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=185%3Acarnsore-pt-met&catid=106%3Atxb&Itemid=30)).
- Ozone measurements: operational, data transfer and data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=207%3Acrp-ozone&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=207%3Acrp-ozone&catid=106%3Atxb&Itemid=30)).
- Picarro G1301: operational, NRT data transfer and NRT data visualisation in place (<https://icos-atc.lscce.ipsl.fr/?q=CRP>).
- Laboratory temperature, humidity and pressure measurements: operational, NRT data transfer and NRT data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=210%3Acrp-lab-conditions&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=210%3Acrp-lab-conditions&catid=106%3Atxb&Itemid=30)).

### 3.1.2 Malin Head

- AE33 aethalometer: operational, NRT data transfer and data visualisation in place ([http://macehead.org/index.php?option=com\\_content&view=article&id=197%3Amalin-head-ae33&catid=106%3Atxb&Itemid=30](http://macehead.org/index.php?option=com_content&view=article&id=197%3Amalin-head-ae33&catid=106%3Atxb&Itemid=30)).
- Picarro G1301: operational, data transfer and data visualisation in place (<https://icos-atc.lsce.ipsl.fr/?q=MLH>).
- CPC: installed, operational, data transfer and data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=203%3Amlh-aerosol-physical-properties&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=203%3Amlh-aerosol-physical-properties&catid=106%3Atxb&Itemid=30)).

### 3.1.3 Mace Head

- Picarro G1301: the stability of the cavity temperature started to fluctuate significantly and after testing it was deemed appropriate to return the instrument for repair; this was done in January 2017 and the instrument was back in operation in June 2017 (<https://icos-atc.lsce.ipsl.fr/?q=MHD>).
- AE33 aethalometer (installed in 21 December 2017): operational, NRT data transfer and data visualisation in place ([http://www.macehead.org/index.php?option=com\\_content&view=article&id=29%3Amhd-ae33&catid=106%3Atxb&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=article&id=29%3Amhd-ae33&catid=106%3Atxb&Itemid=30)).

## 3.2 Infrastructure Development

### 3.2.1 Carnsore Point

Since the inception of this study, there has been an impetus to make use of the tower adjacent to the Carnsore Point site in order to ensure the quality of the measurements. There has been a suggestion that the proximity of livestock (cattle) to the sampling location may have an effect on CH<sub>4</sub> levels at the site. Sampling at multiple heights has been recommended as a possible method of dealing with this issue. This would be facilitated by movement of the sampling site from its current location to a different location, as shown in Figure 3.2. The gradient of measurements at multiple heights will allow for easier identification of air masses of a regional or local nature.

It has recently been shown that local spikes in CO<sub>2</sub> and CH<sub>4</sub> high-resolution data can be successfully

removed using a statistical technique. The standard deviation (SD) method (Drewnick *et al.*, 2012) considers that a time series is a combination of a smooth signal superimposed with a fast variable signal.

One of the conclusions from this study, however, was that action should be taken where appropriate to position sampling sites so as to minimise local interference. To this end numerous meetings were held with the Electricity Supply Board (ESB) to agree on the use of the tower facility at Carnsore for sampling. Once this was agreed planning permission was obtained to facilitate the movement of the EPA facility from its existing location (see Figure 3.2) This was agreed in February 2017. There were a number of health and safety issues surrounding the construction work carried out on site and these were resolved at a number of on-site meetings.

The new facility will be a significant upgrade to the current configuration.

Figure 3.3 shows the current status of the Carnsore Point site, with the existing containers and tower infrastructure (the green box at the base of the tower is to provide power).

### 3.2.2 Malin Head

The laboratory facilities at Malin Head were extensively refurbished during summer 2017 to make it a more suitable site for atmospheric measurements. An aerosol inlet was installed that allows simultaneous measurement of aerosol species. New bench space was also fitted to allow for the installation of new equipment (Figure 3.4).

## 3.3 Instrument Validation

### 3.3.1 Validation of meteorology equipment at Malin Head

Meteorological equipment was installed at the lower building at Malin Head because of the delay in relocating ICOS measurements to Met Éireann's main tower. Figure 3.5 provides comparisons between the two observing systems. The difference in height and horizontal distance between the installed system and the Met Éireann system is approximately 22 m and 45 m, respectively.

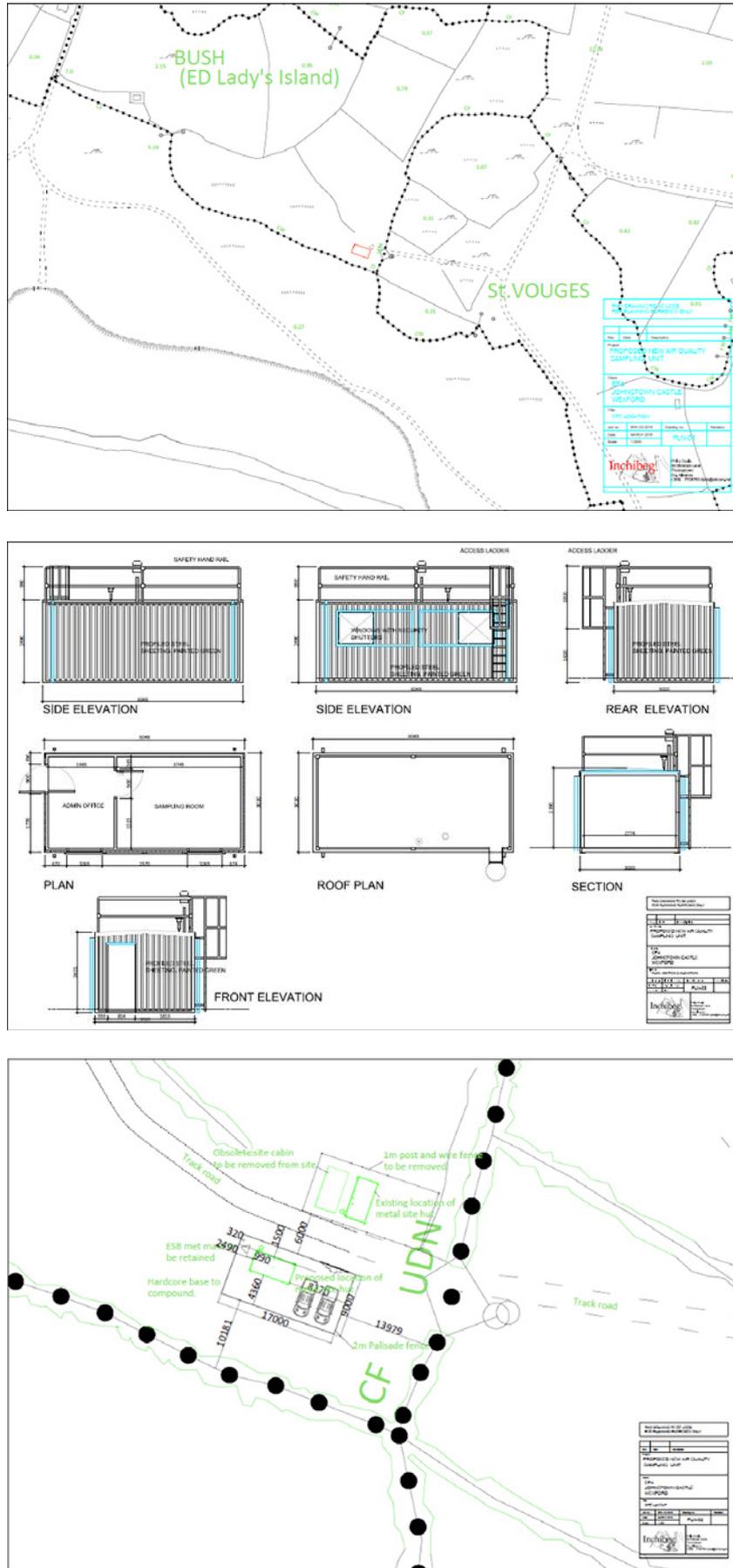
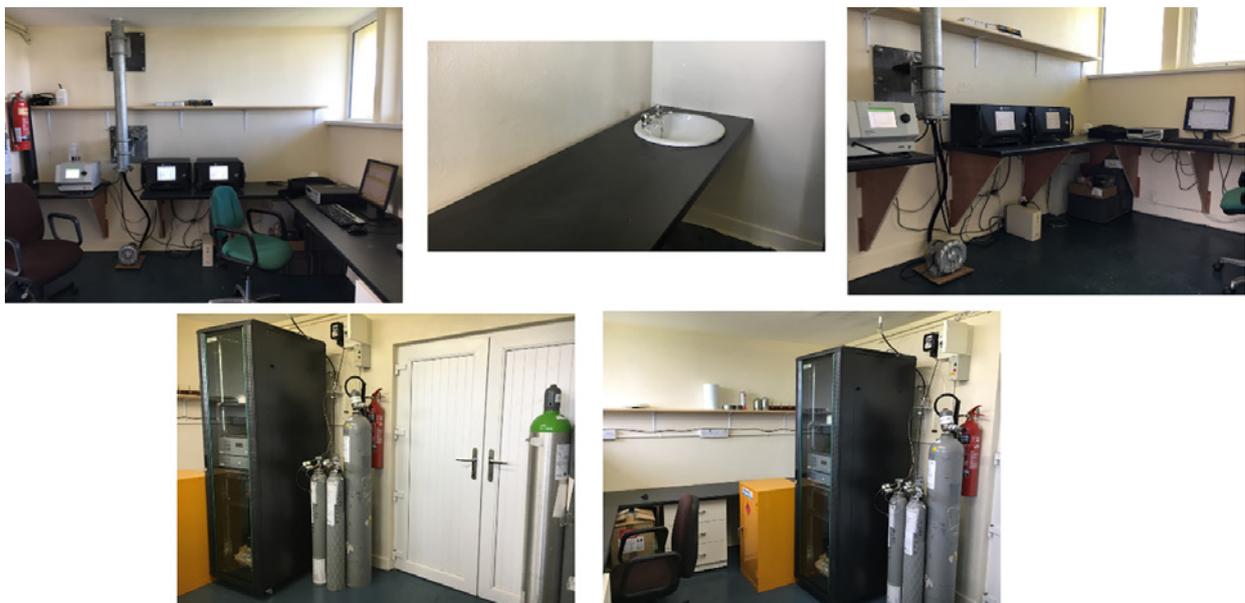


Figure 3.2. Planned infrastructural developments at Carnsore Point.



**Figure 3.3. Current Carnsore Point infrastructure.**



**Figure 3.4. Refurbishment of the laboratory at Malin Head.**

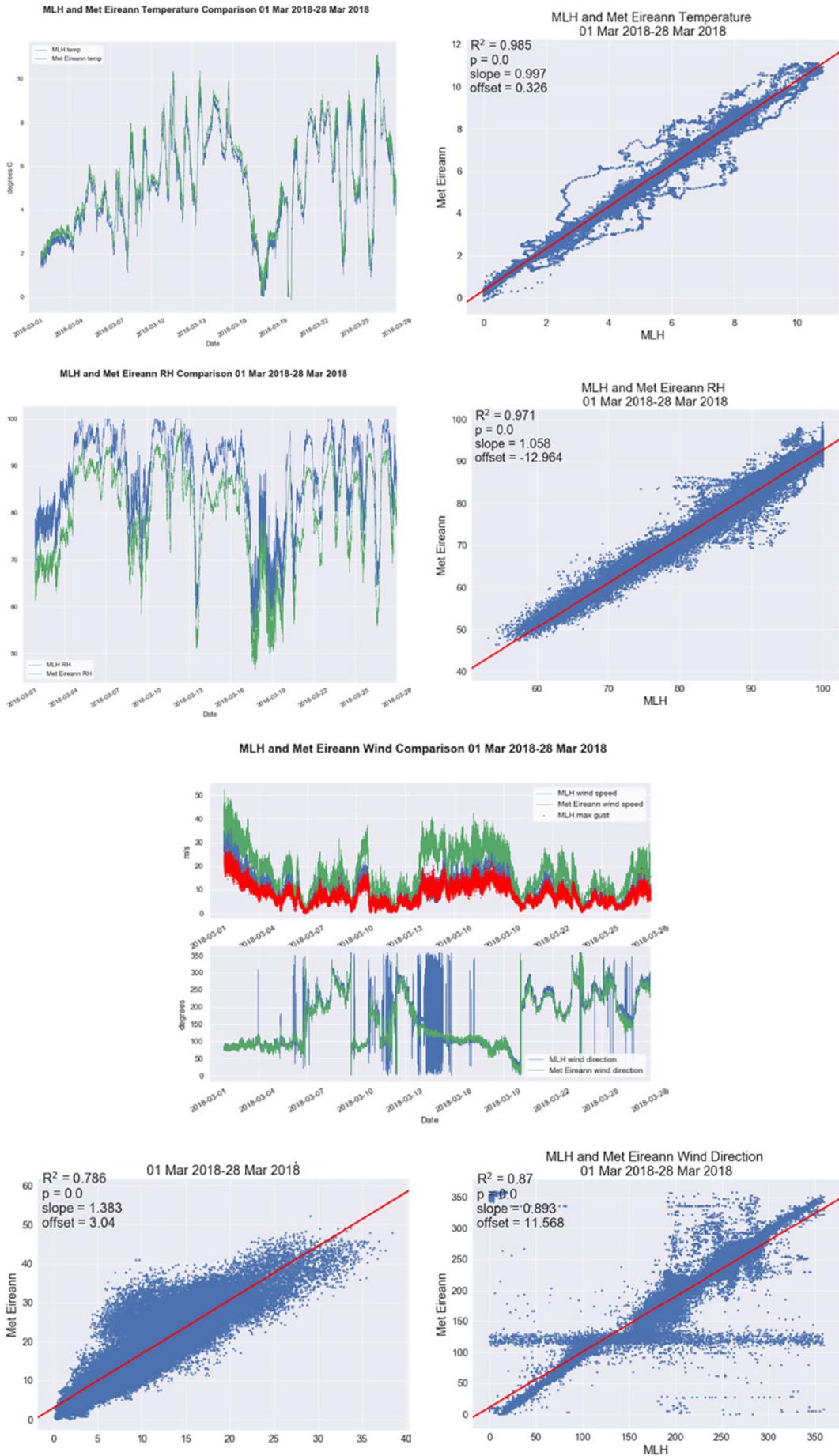


Figure 3.5. Validation of ICOS-compliant anemometry equipment at Malin Head.

Given the difference in height and horizontal distance between the sampling points, there is good agreement between temperature and wind direction for the two sites. Differences in wind speed and humidity can be attributable to height differences in the two sampling positions.

### 3.3.2 Comparison of the AE16 legacy aethalometer and new AE33 aethalometer at Carnsore Point

Figure 3.6 provides a comparison between AE33 aethalometer and AE16 legacy instrument data.

The plots in Figure 3.6 below show excellent agreement between the two instruments; however, the slope indicates that the AE33 is reading about 30% higher BC than the AE16. A comparison of the AE16 BC to the uncorrected (spot 1) 880nm data of the AE33 shows slightly better agreement between the two instruments.

### 3.3.3 Comparison of the Mace Head AE33 aethalometer and the reference instrument

On arrival at Mace Head, the AE33 was compared with the multi-angle absorption photometer (MAAP). The two instruments agree extremely well, but the AE33 measures consistently higher than the MAAP, particularly during times of low black carbon concentrations, as shown in Figure 3.7. This

instrument also measures about 15% higher than the Malin Head AE33 aethalometer, so it is most likely calibrated differently. A scatterplot of hourly averages of the two instruments (not shown) had an  $R^2$  value of 0.956 and a slope of 0.851. According to an intercomparison study by the Leibniz Institute for Tropospheric Research and the European Center for Aerosol Calibration (ECAC), AE33 aethalometers typically measure up to 35% more black carbon than MAAPs and “such differences can be caused by different sensitivities depending on aerosol type” (ECAC, n.d.).

### 3.3.4 Comparison of two AE33 aethalometers

For validation purposes, two AE33 instruments were run in tandem to compare their performance over the typical range of measurements encountered at monitoring sites. Differences of ~20% between instruments for BC, Spot 1 and Spot 2 were noted (Figure 3.8); these are within the range seen in previous assessments of the AE33 at the ECAC.

## 3.4 Installation of New Instrumentation

A number of new instruments have been added to the network since 2017. These include a G2401 GHG analyser at Malin Head, an ACSM at Carnsore Point and an SMPS/FIDAS fine dust measurement system at Malin Head. A description of each of these installations is provided in the following sections.

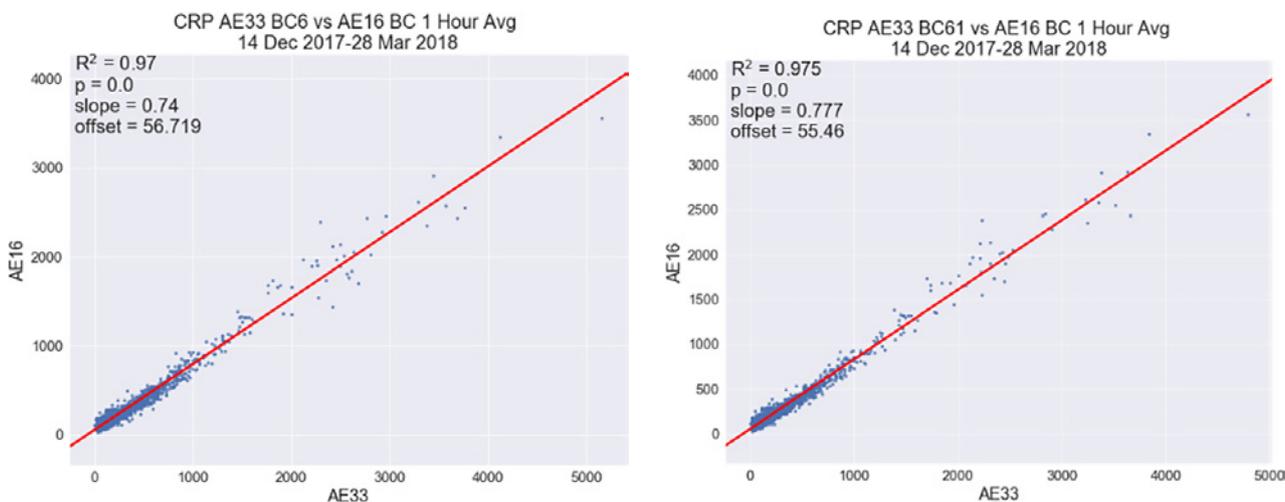


Figure 3.6. Correlation plots of CRP AE16 vs AE33 880nm BC (left) and CRP AE16 vs AE33 spot 1-uncorrected (right).

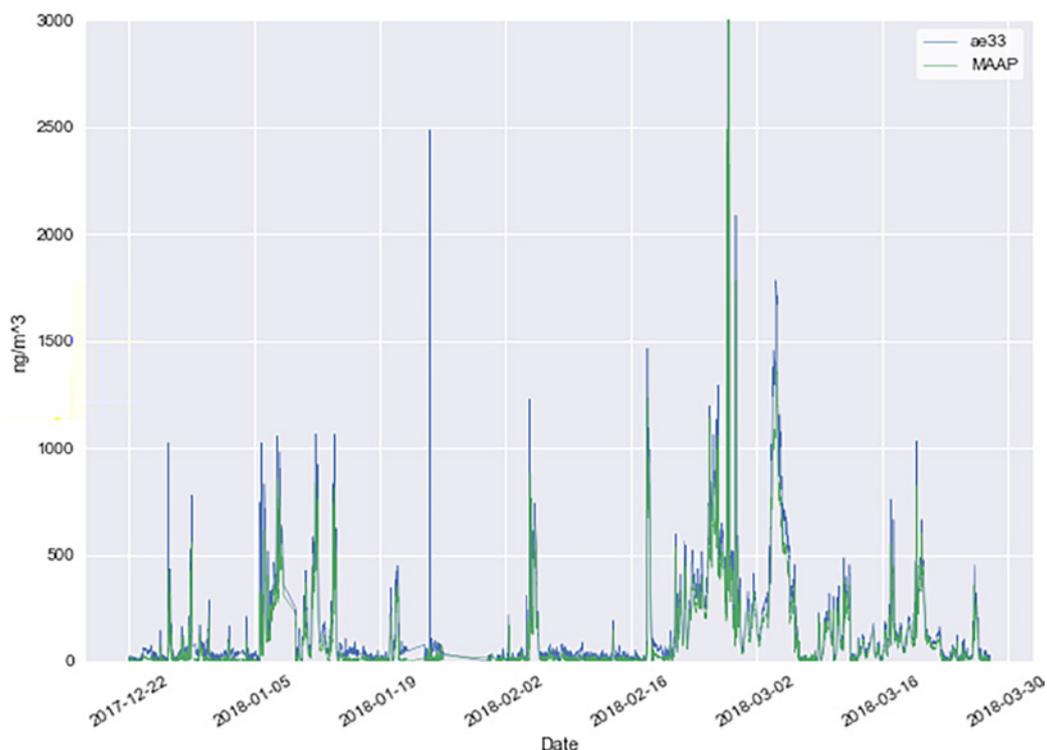


Figure 3.7. Comparison of AE33 and MAAP black carbon measurements at Mace Head (30-minute average), 22 December 2017–28 March 2018.

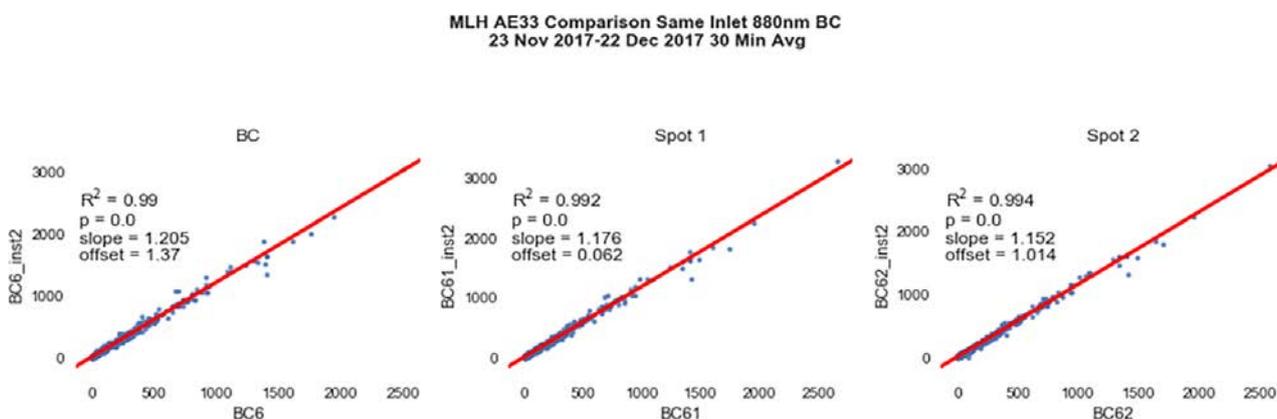


Figure 3.8. Comparison of two AE33 aethalometers for BC (black carbon) Spot 1 and Spot 2.

### 3.4.1 Installation of the G2401 analyser at Malin Head

The G2401 is a next-generation Picarro instrument, which, in addition to measuring CH<sub>4</sub> and CO<sub>2</sub>, has the capability to measure CO (which is a very useful anthropogenic tracer). The main purpose of installing new GHG monitoring instrumentation at the sites is to reduce down time, as the lead time for repair of these instruments is quite long. The original instruments are over 8 years old and it is likely that servicing

and repair for the G1301 will not be available after 2019. The G2401 instrument was deployed at Malin Head and run for a period of 8 weeks to determine stability (Figure 3.9). GCWerks (a bespoke software package used for processing GHG data) will be used in conjunction with this instrument prior to embedding into the full ICOS network. The G1301 has been working quite satisfactorily at Malin Head and will continue to do so until its replacement by the G2401 is necessitated.

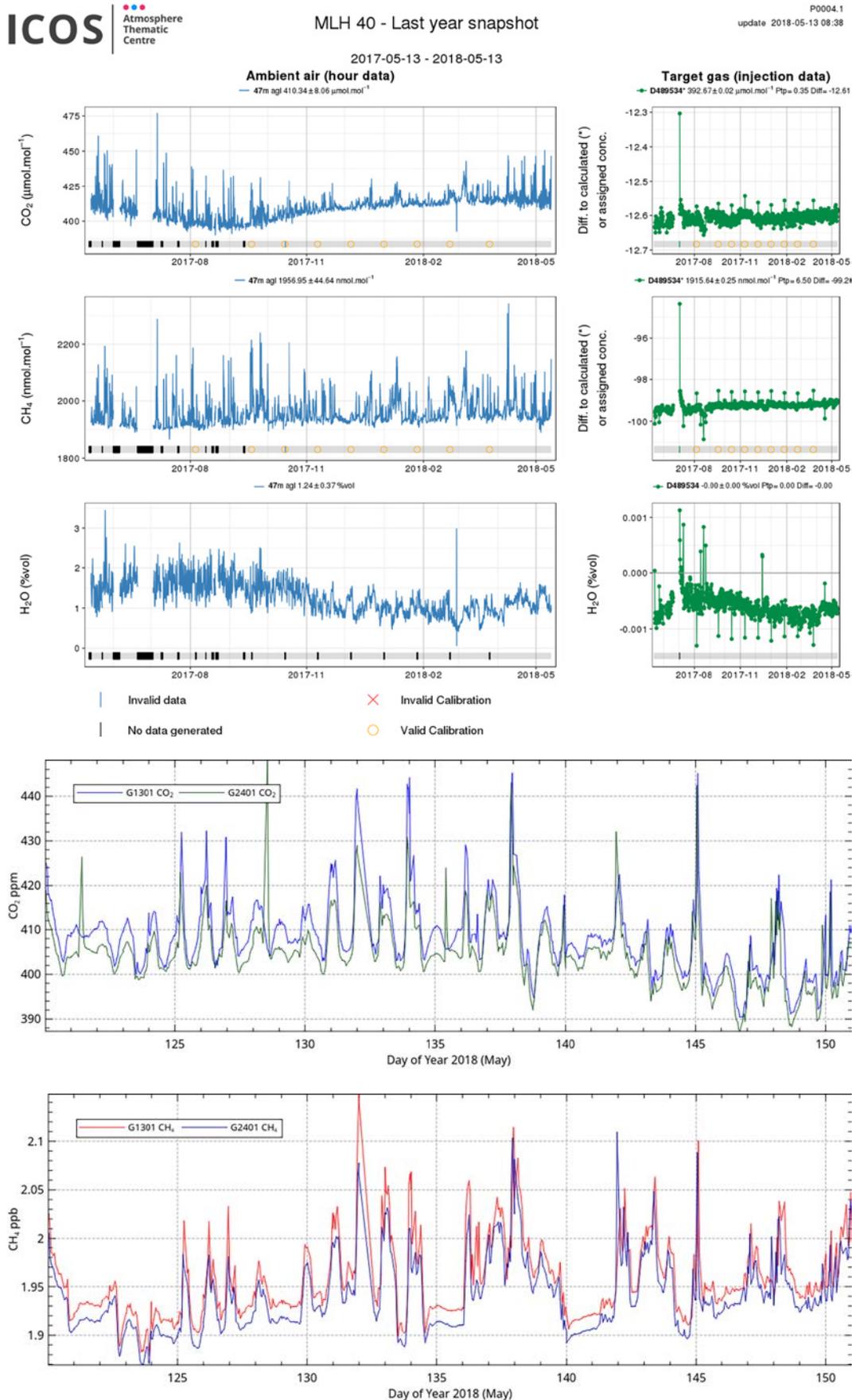


Figure 3.9. GHG measurements from Malin Head (<https://icos-atc.lsce.ipsl.fr/?q=MLH>). ppb, parts per billion; ppm, parts per million. ICOS data are licensed under the Creative Commons Attribution 4.0 International licence (<https://creativecommons.org/licenses/by/4.0/>). ICOS RI, licensed under CC4BY.

### 3.4.2 Installation of the ACSM at Carnsore Point

An ACSM was purchased for installation at Carnsore Point. Because of infrastructural developments being undertaken at Carnsore Point in early 2018, the instrument was deployed at University College Dublin (UCD) to enable a provisional assessment of its performance to be carried out. Figure 3.10 shows a comparison between the new PM<sub>2.5</sub> (particulate matter  $\leq 2.5 \mu\text{m}$ )-capable ACSM and the existing PM<sub>1</sub> (particulate matter  $\leq 1 \mu\text{m}$ ) ACSM. The two instruments show good agreement, providing confidence in its field deployment.

### 3.4.3 Installation of the SMPS/FIDAS at Malin Head

The installation of the SMPS/FIDAS was undertaken after resolving a number of issues surrounding the installation of radioactive sources. The FIDAS (Figure 3.11) was installed at Malin Head in May 2018 in our existing aerosol inlet whilst the SMPS was installed at the end of 2018. The common inlet has been designed to facilitate assessment of smaller particle sizes and has not been optimised for larger particles. We are currently investigating the provisional data to assess the suitability of the inlet. It may be necessary to install a roof inlet to optimise the measurements.

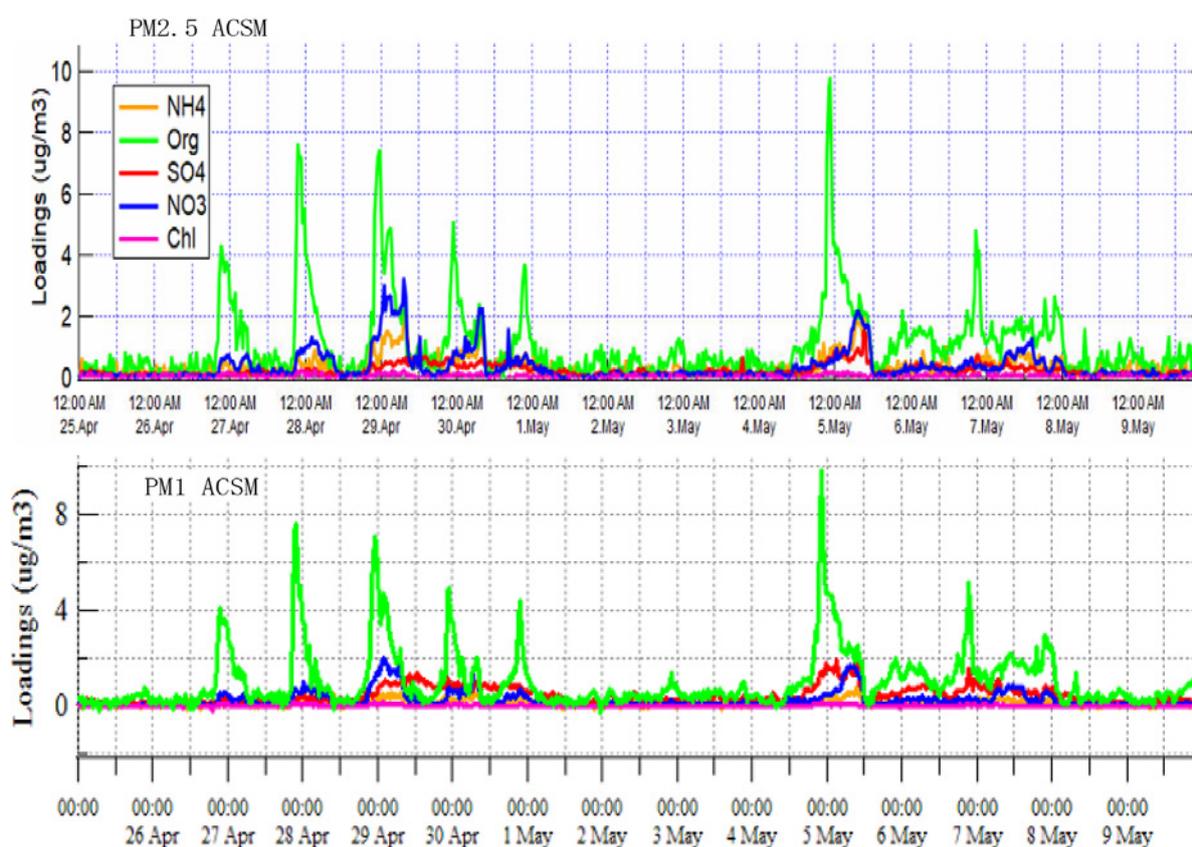


Figure 3.10. Preliminary comparison between the new PM<sub>2.5</sub> ACSM (top) and the existing PM<sub>1</sub> ACSM (bottom). These measurements were conducted at UCD from 25 April 2018 to 9 May 2018. Both instruments show similar evolution of the measured organic compounds (Org), sulfate (SO<sub>4</sub>), nitrate (NO<sub>3</sub>), ammonium (NH<sub>4</sub>) and chloride (Chl). Regular calibration of the PM<sub>2.5</sub> ACSM is needed to obtain more accurate measurements.

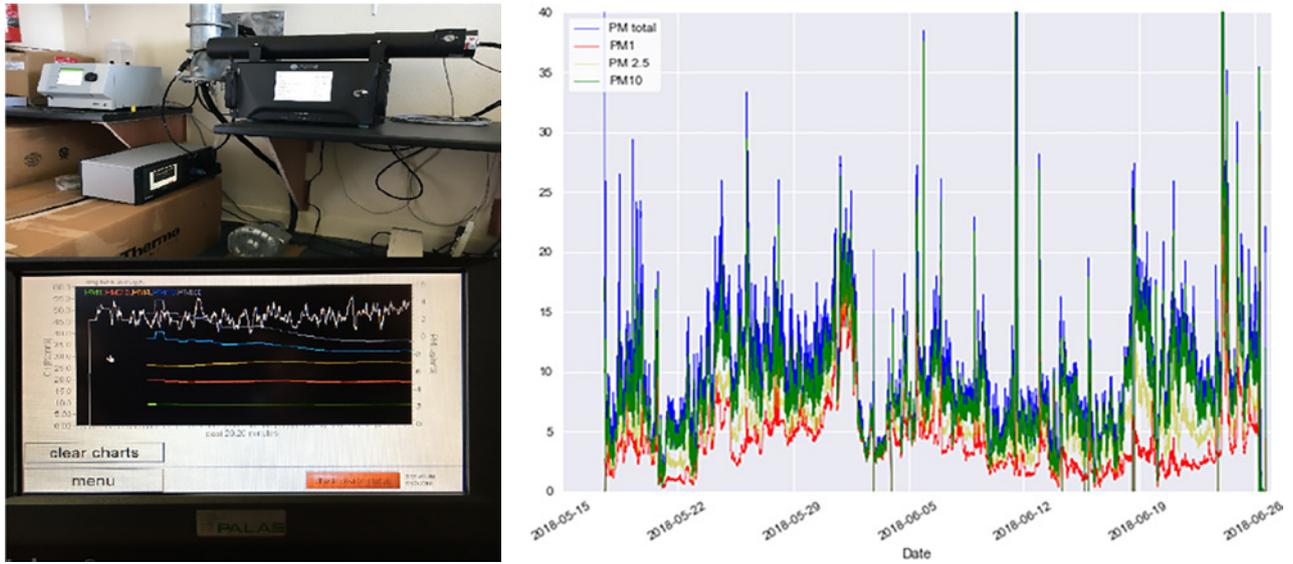


Figure 3.11. Installation of the FIDAS instrument (top left, laboratory installation; bottom left, instrument interface; right, example of time series from the instrument).

## 4 Results

### 4.1 Aerosol Chemical Composition from High-volume Filter Samples at Malin Head and Carnsore Point

Table 4.1 provides yearly statistics for the species thus analysed from the filters sampled at Carnsore Point (seven species) and Malin Head (eight species). Mean values and variability are broadly similar over the 11 years sampled for all species except SO<sub>4</sub>, which shows a general downwards trend.

### 4.2 Wind Sectoring of Aerosol Chemical Composition at Carnsore Point and Malin Head

Figure 4.1 shows wind sectoring of the 2015 ion concentrations at Carnsore Point, Malin Head and Valentia Observatory (Valentia has been included here as EMEP measurements are made at this site, although they do not fall under the responsibility of this study).

**Table 4.1. Aerosol chemical composition (Ca, Cl, K, Mg, Na, NH<sub>4</sub>, NO<sub>3</sub> and SO<sub>4</sub>) from high-volume filter samples at Carnsore Point and Malin Head**

Year	Mean	SD	Min	50%	Max
<b>CRP Ca</b>					
2005	0.13	0.10	0.00	0.11	0.61
2006	0.18	0.12	-0.01	0.14	0.72
2007	0.17	0.12	0.02	0.14	0.66
2008	0.19	0.17	0.01	0.16	2.73
2009	0.16	0.09	0.02	0.13	0.51
2010	0.12	0.10	0.01	0.10	0.70
2011	0.17	0.12	0.01	0.14	0.67
2012	0.13	0.09	0.01	0.11	0.62
2013	0.15	0.12	0.01	0.12	0.66
2014	0.17	0.12	0.02	0.13	0.76
2015	0.16	0.12	0.01	0.12	0.76
<b>CRP Cl</b>					
2005	nan	nan	nan	nan	nan
2006	nan	nan	nan	nan	nan
2007	nan	nan	nan	nan	nan
2008	nan	nan	nan	nan	nan
2009	nan	nan	nan	nan	nan
2010	nan	nan	nan	nan	nan
2011	nan	nan	nan	nan	nan
2012	5.08	4.99	0.27	3.71	61.46
2013	5.57	5.02	0.05	3.8	25.19
2014	8.66	52.32	0.33	3.88	999.99
2015	6.31	5.20	0.06	4.68	32.97
<b>CRP K</b>					
2005	0.15	0.12	0.00	0.11	0.74
2006	0.16	0.11	0.00	0.13	0.60

Table 4.1. Continued

Year	Mean	SD	Min	50%	Max
2007	0.15	0.11	0.02	0.11	0.56
2008	0.16	0.10	0.01	0.13	0.45
2009	0.16	0.09	0.02	0.13	0.48
2010	0.13	0.10	0.01	0.09	0.63
2011	0.21	0.24	0.02	0.15	1.85
2012	0.14	0.17	0.02	0.11	2.97
2013	0.15	0.11	0.02	0.12	0.56
2014	0.16	0.12	0.02	0.11	0.68
2015	0.16	0.11	0.02	0.13	0.73
CRP Mg					
2005	0.34	0.29	0.00	0.26	1.55
2006	0.46	0.38	-0.01	0.32	2.07
2007	0.41	0.39	0.00	0.28	1.93
2008	0.42	0.33	0.02	0.30	1.51
2009	0.41	0.31	0.00	0.33	1.58
2010	0.32	0.33	0.00	0.20	2.11
2011	0.47	0.44	0.01	0.37	3.47
2012	0.35	0.36	0.02	0.26	4.82
2013	0.36	0.34	0.01	0.25	1.71
2014	0.43	0.40	0.02	0.29	2.28
2015	0.43	0.35	0.01	0.31	2.18
CRP Na					
2005	2.87	2.44	0.08	2.18	14.98
2006	3.66	2.92	0.02	2.66	15.92
2007	3.51	3.17	0.11	2.42	15.35
2008	3.46	2.65	0.06	2.55	12.41
2009	3.35	2.38	0.10	2.73	12.32
2010	2.52	2.58	0.02	1.58	16.84
2011	3.32	2.68	0.05	2.86	16.07
2012	2.92	3.04	0.10	2.13	42.30
2013	2.96	2.61	0.05	2.08	13.81
2014	3.45	3.11	0.12	2.32	18.29
2015	3.51	2.77	0.05	2.66	16.82
CRP NH <sub>4</sub>					
2005	0.65	0.63	0.12	0.42	5.12
2006	0.70	0.89	0.00	0.38	9.49
2007	0.73	1.12	0.00	0.39	9.83
2008	0.75	1.03	0.06	0.36	9.98
2009	0.61	0.87	0.09	0.33	7.91
2010	0.63	0.60	0.10	0.42	4.00
2011	1.02	1.67	0.05	0.35	11.07
2012	0.57	0.77	0.04	0.33	8.49
2013	0.69	0.96	0.04	0.34	8.48
2014	0.62	0.74	0.04	0.37	5.33
2015	0.58	0.72	0.07	0.29	4.80

Table 4.1. Continued

Year	Mean	SD	Min	50%	Max
<b>CRP NO<sub>3</sub></b>					
2005	0.36	0.45	0.02	0.20	4.07
2006	0.38	0.39	0.00	0.21	2.02
2007	0.45	0.7	0.02	0.18	6.72
2008	0.39	0.56	0.02	0.14	2.99
2009	0.35	0.64	0.01	0.15	6.32
2010	0.39	0.46	0.01	0.21	3.93
2011	0.39	0.54	0.01	0.15	3.44
2012	0.37	0.66	0.01	0.15	7.03
2013	0.46	0.73	0.01	0.16	6.93
2014	0.60	5.23	0.00	0.14	99.99
2015	0.31	0.43	0.01	0.12	2.40
<b>CRP SO<sub>4</sub></b>					
2005	0.67	0.44	0.14	0.56	3.01
2006	0.81	0.56	0.00	0.66	4.38
2007	0.72	0.46	0.13	0.61	2.83
2008	0.68	0.39	0.00	0.58	2.40
2009	0.59	0.34	0.11	0.52	2.83
2010	0.56	0.31	0.02	0.48	1.98
2011	0.67	0.44	0.02	0.56	2.97
2012	0.53	0.47	0.02	0.45	7.71
2013	0.57	0.34	0.02	0.49	2.21
2014	0.93	5.22	0.02	0.56	99.99
2015	0.16	0.12	0.01	0.12	0.76
<b>MLH Ca</b>					
2005	0.1	0.07	0.00	0.10	0.42
2006	0.12	0.08	0.00	0.10	0.41
2007	0.13	0.07	0.00	0.11	0.48
2008	0.13	0.08	0.01	0.11	0.58
2009	0.11	0.14	0.02	0.09	2.02
2010	0.09	0.06	0.01	0.08	0.47
2011	0.09	0.06	0.01	0.08	0.37
2012	0.11	0.10	0.01	0.08	1.37
2013	0.10	0.08	0.01	0.08	1.03
2014	0.09	0.07	0.01	0.08	0.74
2015	0.09	0.06	0.01	0.07	0.38
<b>MLH Cl</b>					
2005	nan	nan	nan	nan	nan
2006	nan	nan	nan	nan	nan
2007	4.59	3.25	0.37	3.6	19.59
2008	4.37	3.24	0.45	3.36	17.56
2009	3.63	2.44	0.21	3.11	13.23
2010	3.08	2.12	0.34	2.57	12.46
2011	nan	nan	nan	nan	nan
2012	3.45	2.72	0.29	2.68	19.99

Table 4.1. Continued

Year	Mean	SD	Min	50%	Max
2013	3.64	2.7	0.25	2.99	18.77
2014	4.12	4.46	0.10	2.69	16.93
2015	3.74	2.57	0.46	3.13	13.01
MLH K					
2005	0.10	0.07	0.00	0.08	0.48
2006	0.11	0.07	0.00	0.09	0.47
2007	0.11	0.07	0.02	0.09	0.53
2008	0.11	0.07	0.03	0.10	0.40
2009	0.10	0.05	0.02	0.09	0.34
2010	0.08	0.05	0.01	0.08	0.28
2011	0.11	0.08	0.02	0.09	0.89
2012	0.09	0.06	0.01	0.08	0.40
2013	0.09	0.05	0.02	0.08	0.38
2014	0.36	0.87	0.01	0.08	3.29
2015	0.09	0.06	0.02	0.08	0.33
MLH Mg					
2005	0.24	0.21	0.00	0.2	1.17
2006	0.29	0.22	-0.01	0.23	1.28
2007	0.29	0.21	0.00	0.22	1.30
2008	0.29	0.22	0.03	0.21	1.19
2009	0.24	0.17	0.02	0.21	0.95
2010	0.21	0.15	0.01	0.18	0.92
2011	0.32	0.48	0.02	0.21	4.46
2012	0.22	0.18	0.02	0.17	1.27
2013	0.22	0.18	0.01	0.18	1.22
2014	0.22	0.17	0.01	0.18	1.21
2015	0.23	0.18	0.02	0.18	0.93
MLH Na					
2005	2.16	1.66	0.03	1.78	9.20
2006	2.39	1.69	0.14	1.96	9.67
2007	2.65	1.86	0.22	2.07	11.07
2008	2.46	1.80	0.17	1.92	9.57
2009	2.05	1.38	0.10	1.79	7.42
2010	1.72	1.18	0.11	1.47	7.05
2011	2.11	1.80	0.07	1.69	18.77
2012	1.94	1.54	0.16	1.52	11.37
2013	1.90	1.40	0.04	1.56	9.07
2014	1.66	1.29	0.03	1.32	7.16
2015	2.01	1.43	0.17	1.63	7.32
MLH NH <sub>4</sub>					
2005	0.45	0.69	0.00	0.26	7.13
2006	0.65	1.12	-0.02	0.32	11.25
2007	0.55	0.92	0.01	0.25	9.10
2008	0.59	0.91	0.08	0.26	8.13
2009	0.47	0.63	0.09	0.26	6.23

Table 4.1. Continued

Year	Mean	SD	Min	50%	Max
2010	0.47	0.54	0.04	0.26	3.19
2011	0.55	0.76	0.09	0.27	6.13
2012	0.48	0.68	0.04	0.26	6.39
2013	0.56	0.81	0.08	0.26	6.52
2014	0.73	1.02	0.03	0.34	6.83
2015	0.47	0.66	0.10	0.23	5.19
MLH NO <sub>3</sub>					
2005	0.23	0.56	0.01	0.07	6.63
2006	0.27	0.38	0.00	0.1	2.16
2007	0.25	0.45	0.00	0.06	3.47
2008	0.28	0.55	0.01	0.08	5.03
2009	0.23	0.49	0.00	0.07	5.53
2010	0.23	0.35	0.00	0.07	1.91
2011	0.22	0.39	0.01	0.07	3.45
2012	0.22	0.38	0.01	0.08	2.84
2013	0.28	0.53	0.01	0.07	3.68
2014	0.25	0.44	0.00	0.09	3.54
2015	0.22	0.41	0.00	0.04	2.39
MLH SO <sub>4</sub>					
2005	0.46	0.31	0.04	0.38	2.40
2006	0.58	0.45	0.00	0.44	3.35
2007	0.55	0.39	0.00	0.43	2.92
2008	0.53	0.34	0.07	0.42	2.05
2009	0.4	0.25	0.11	0.34	1.85
2010	0.41	0.26	0.11	0.34	2.03
2011	0.4	0.24	0.08	0.36	2.15
2012	0.4	0.26	0.05	0.34	2.13
2013	0.44	0.31	0.08	0.35	2.23
2014	0.56	0.56	0.02	0.39	4.18
2015	0.38	0.23	0.11	0.32	1.83

CRP, Carnsore Point; MLH, Malin Head; nan, not a number.

Differences in the trends at each of the stations is reflective of the large clean sectors at both Malin Head and Valentia. Carnsore Point is more subject to regimes that have passed over the island of Ireland and this is reflected in the data. The trend in SO<sub>4</sub> is looked at in more depth in the following section.

### 4.3 Trends in Aerosol SO<sub>4</sub>

Measurements of various atmospheric parameters have been conducted at the Met Éireann Valentia Observatory at Caherciveen, County Kerry, since the 1980s. One such parameter is aerosol SO<sub>4</sub>, for which

there is a long-term record from 1981 to the present. Bashir *et al.* (2006) have reported these data up to 2003. The data presented here extend the series to 2016. The entire data series is plotted in Figure 4.2. The downwards trend observed by Bashir *et al.* (2006) is continued in the more recent data from 2007 to 2016. Although there are no physical processes parameterised by such a linear fit, it highlights the consistent downwards trend. The very large SO<sub>4</sub> value in 1982 was most likely due to the eruption of the El Chicón volcano in Mexico in March and April 1982, which produced a volcanic plume with a particularly high sulfur content.

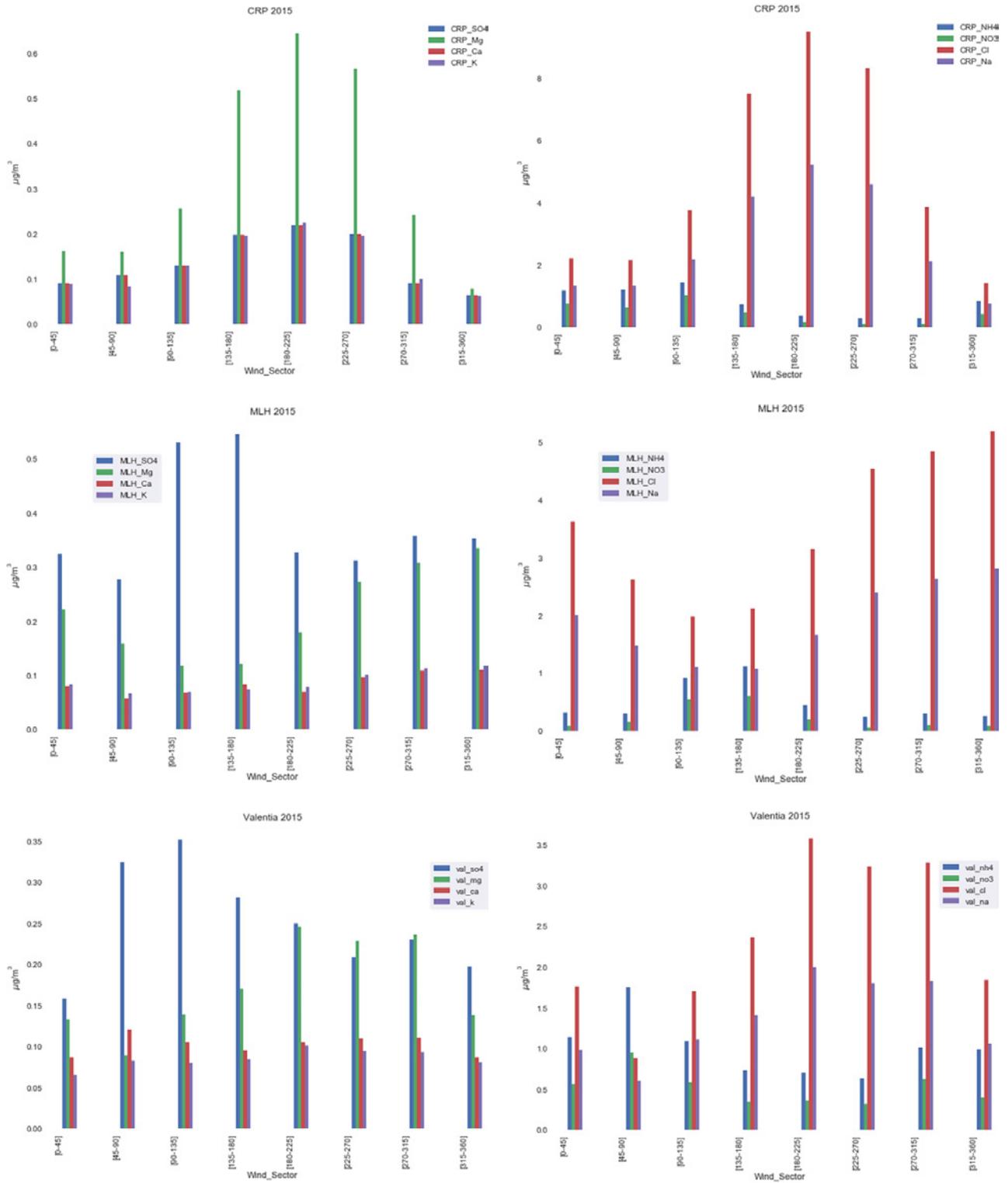
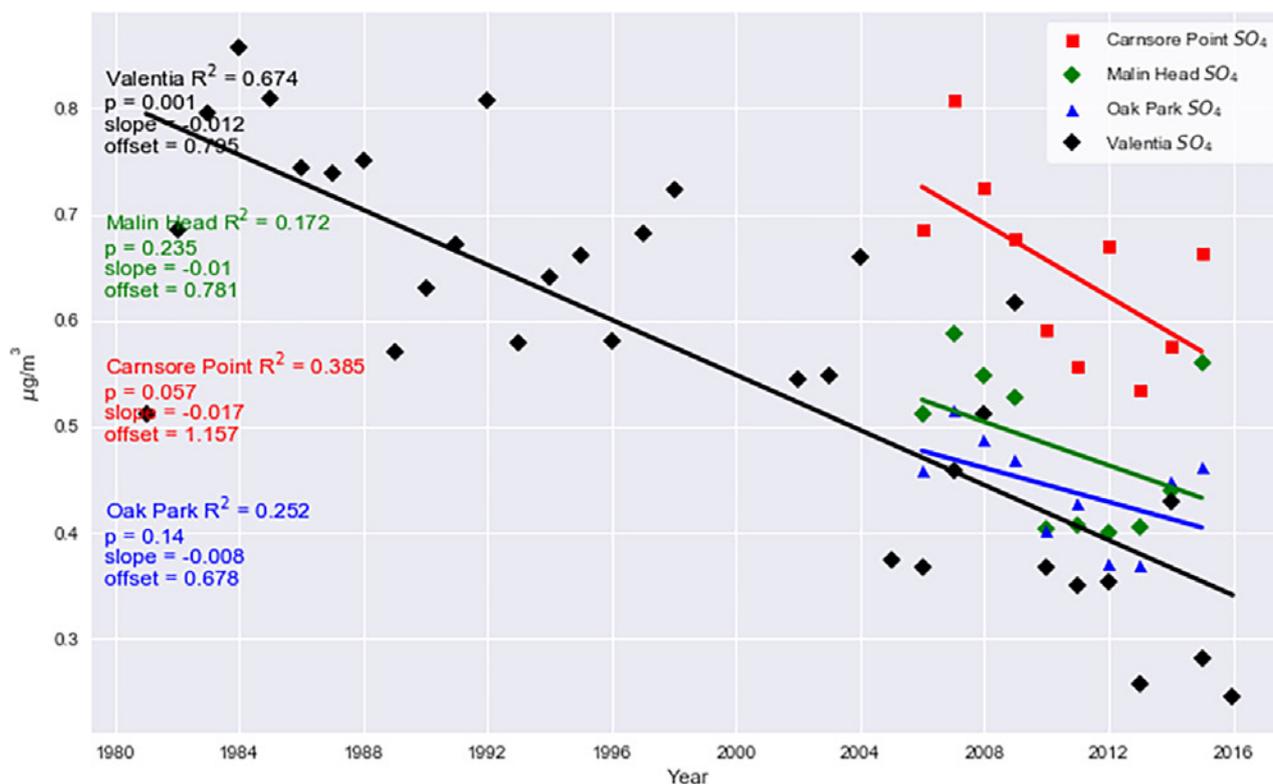


Figure 4.1. Wind sectoring of the ion concentrations at Carnsore Point (CRP), Malin Head (MLH) and Valentia for 2015.



**Figure 4.2. Aerosol SO<sub>4</sub> concentrations from high-volume filter samples from Valentia, Carnsore Point, Malin Head and Oak Park.**

Overlaid on the Valentia data set are the results from Carnsore Point, Malin Head and Oak Park for aerosol SO<sub>4</sub> high-volume filter sample measurements for the years 2005–2016. Although it would be difficult to detect a long-term trend from these data sets on their own, when seen against the Valentia data set it is clear that these more recent measurements do follow the generally downwards trend and, with the exception of Carnsore Point, show generally similar SO<sub>4</sub> levels to those recorded at Valentia.

## 4.4 Black Carbon

### 4.4.1 Malin Head

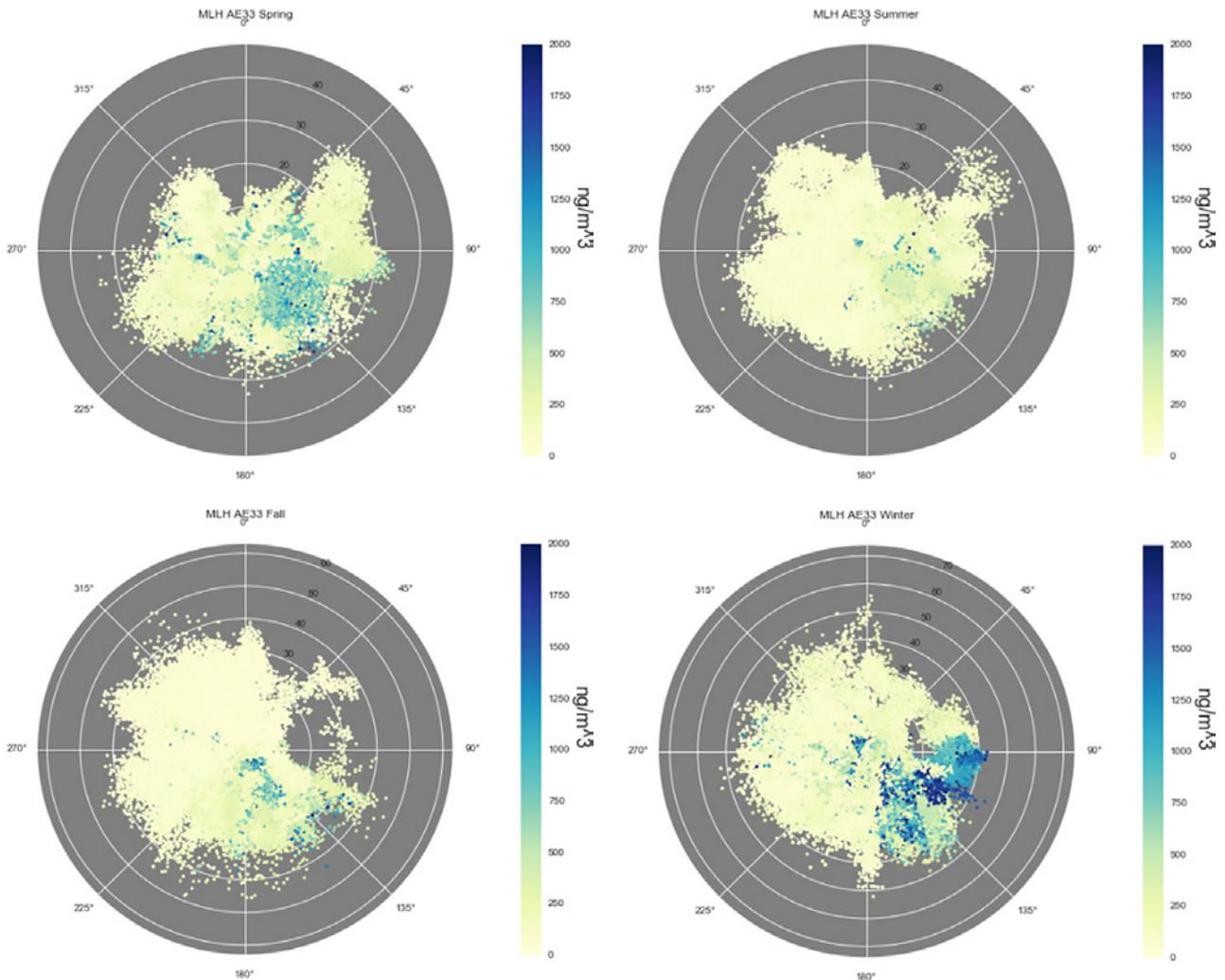
Figure 4.3 shows the seasonal wind roses for black carbon measurements made at Malin Head observatory in 2017. These clearly show the increase in black carbon concentrations during the winter. This is most likely associated with increased use of fossil fuels during the winter. This is also evidenced by the equivalent wind rose plot for biomass burning (not shown here). The increase in black carbon emissions from this sector during the winter is a good illustration

of the increased particulate matter loading associated with domestic fuel burning.

### 4.4.2 Carnsore Point

Figure 4.4 shows the seasonal wind roses for black carbon measurements made at Carnsore Point over the same time period (2017). The seasonal nature of black carbon emissions is well illustrated again, with increased levels during the winter. The wind direction of these increased emissions suggests that these relate predominantly to wintertime UK and European pollution events. The increase in biomass burning makes a significant contribution to the annual black carbon mixing ratios.

The cumulative frequency distribution for the Malin Head and Carnsore Point black carbon measurements is shown in Figures 4.5 and 4.6, respectively, and illustrates the difference in the siting of the two instruments. The Malin Head data are more reflective of a large amount of clean air punctuated by high black carbon concentrations associated with local sources (there is traffic in close proximity to the sampling point, at ~25 m), whereas the Carnsore Point data have a



**Figure 4.3. Seasonal wind roses for black carbon measurements made at Malin Head in 2017.**

wider distribution that is more reflective of regional sources, with few local sources because of the location of the site.

#### 4.5 Particulate Matter at Carnsore Point

Figure 4.7 shows the seasonal wind roses for the TEOM particulate matter measurements made at Carnsore Point during 2017. It is clear from these measurements that in both summer and spring particulate matter is correlated with wind sectors that are more associated with air mass trajectories that have passed over the mainland. This suggests that the transboundary contribution as an overall contribution may be less than that associated with black carbon. A mass reconstruction using TEOM, aethalometer and high-volume filter sample data is under way.

Figures 4.8 and 4.9 show the wind roses for particle counts for 2017 at Carnsore Point and Malin Head, respectively. The data clearly show a seasonal cycle, with enhanced particle numbers during the spring and summer. This is probably related to open ocean nucleation events.

Figures 4.10 and 4.11 show the number of seasonal exceedances of particle counts for two different thresholds (30,000 and 40,000, respectively) for a clean 90° air sector at both Carnsore Point and Malin Head in 2017/2018. This excludes any data with a black carbon concentration of over 70 ng m<sup>-3</sup>, which may be reflective of anthropogenic pollution. This illustrates the possible influence of seasonal open ocean or littoral zone nucleation on particle number levels at these remote sites. This is currently the focus of further research.

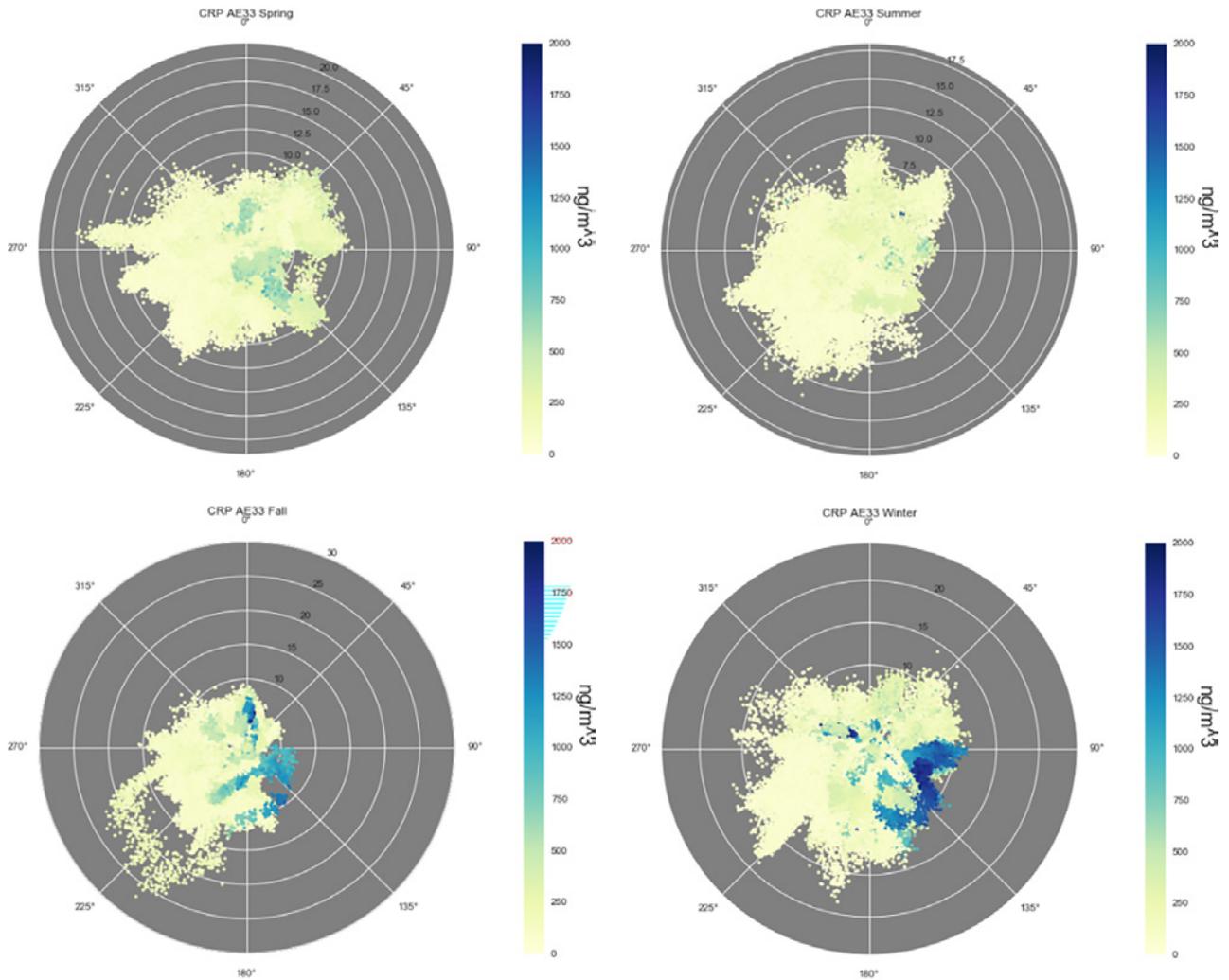


Figure 4.4. Seasonal wind roses for black carbon measurements made at Carnsore Point in 2017.

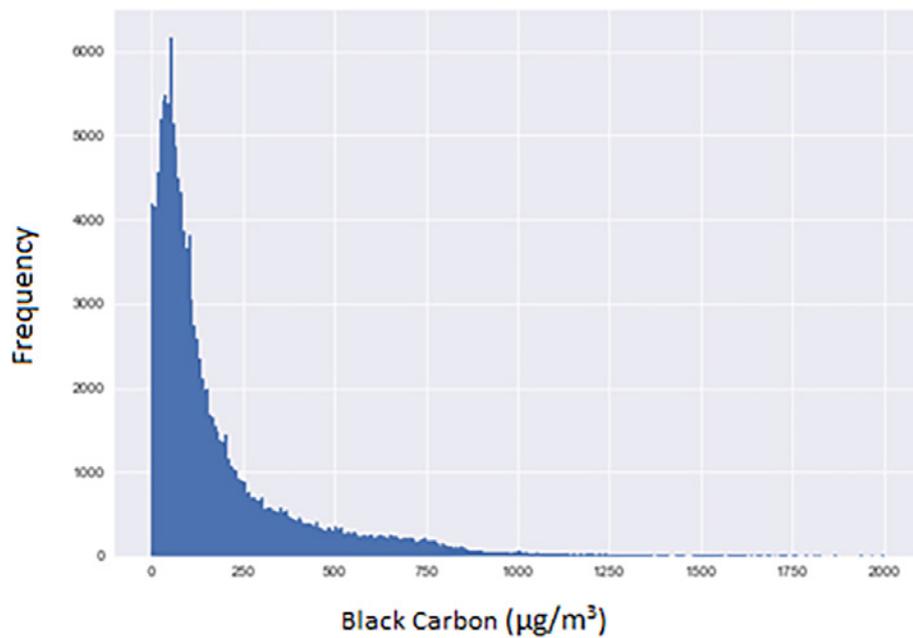


Figure 4.5. Cumulative frequency distribution for Malin Head black carbon measurements using the AE33 aethalometer, 27 April 2017–31 August 2017.

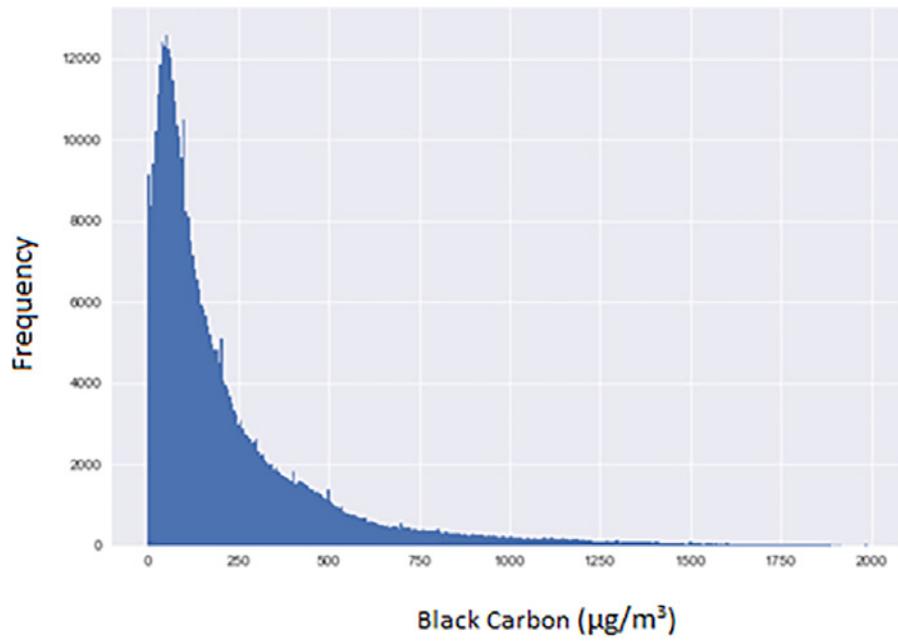


Figure 4.6. Cumulative frequency distribution for Carnsore Point black carbon measurements using the AE33 aethalometer, 1 January 2017–31 December 2017.

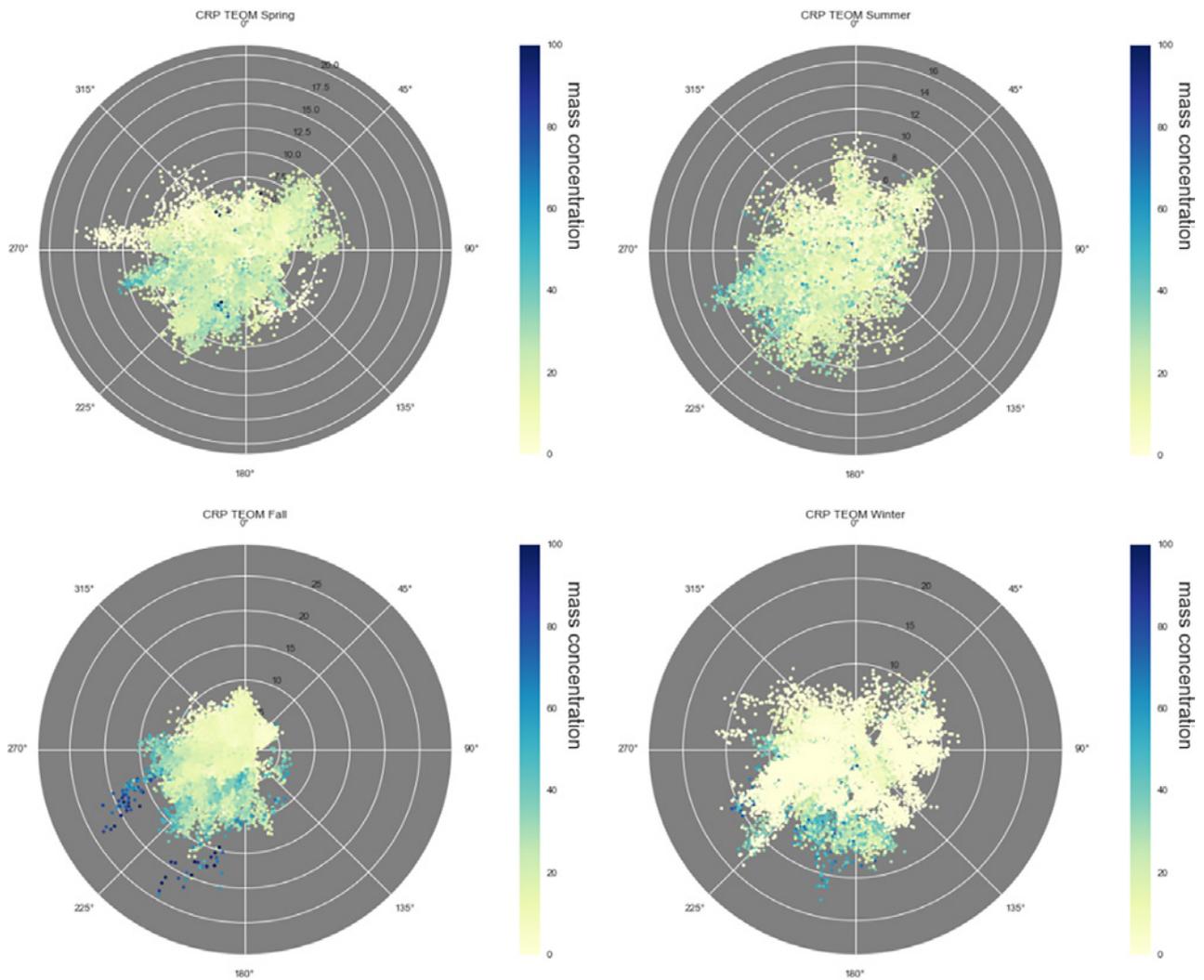


Figure 4.7. Seasonal wind roses for particulate matter measurements at Carnsore Point.

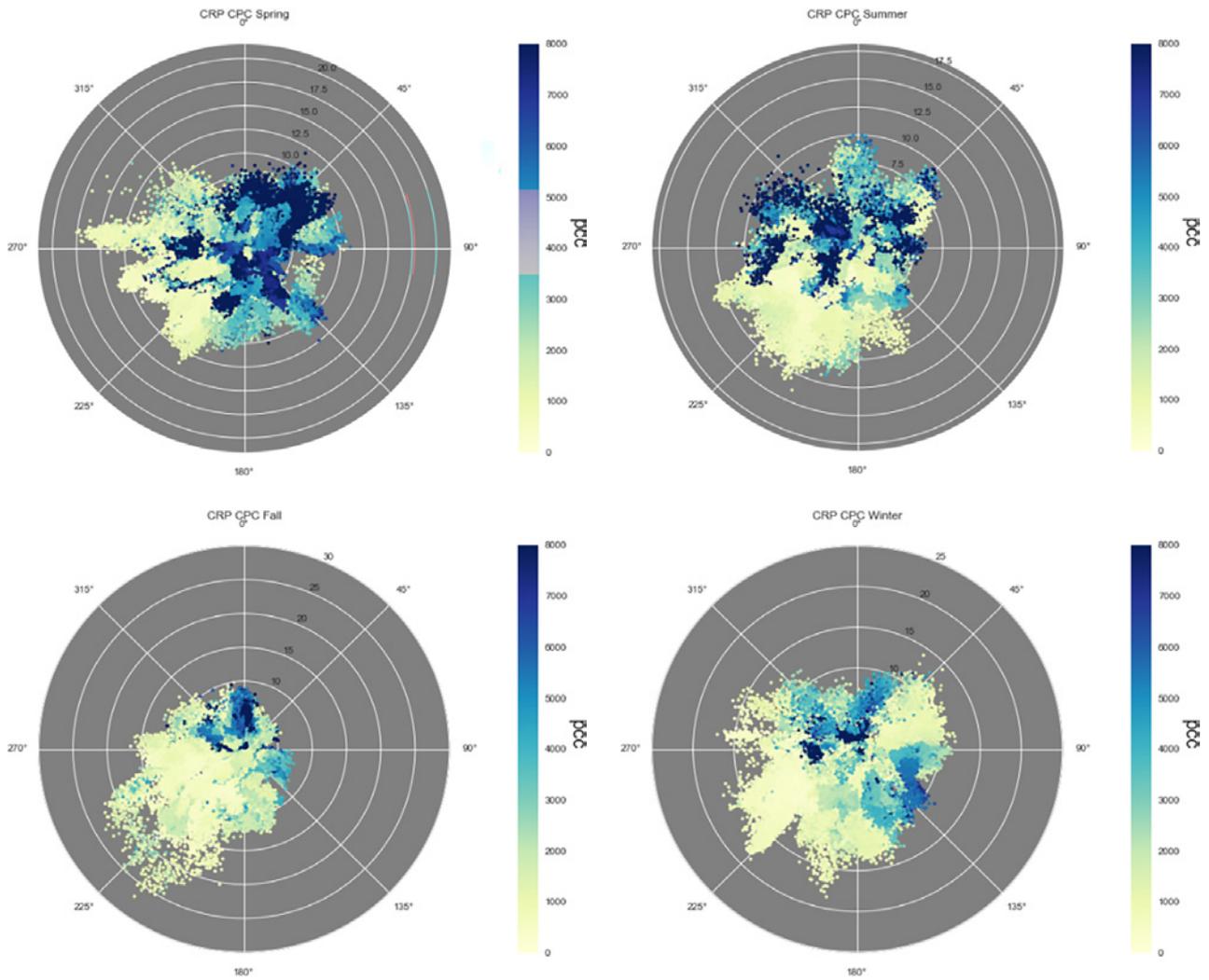


Figure 4.8. Seasonal wind roses for CPC measurements at Carnsore Point.

#### 4.6 Ozone Measurements at Carnsore Point

Figure 4.12 provides a time series of ozone data since the reinstallation of the ozone monitoring instrument at Carnsore Point, classified according to wind direction, and Figure 4.13 shows wind rose data classified according to wind sectors. In general, higher concentrations are associated with wind sectors that have seen the air mass trajectory pass over the country.

Initial analysis of the ozone wind rose measurements at Carnsore Point suggests that the predominant wind sector for ozone measurements is to the west of the site. Given that this wind direction is

associated with air masses that have passed over the island of Ireland this suggests that much of the ozone seen at Carnsore Point is generated *in situ*, with a possible contribution from intercontinental transport.

#### 4.7 Nephelometer Measurements at Carnsore Point

Figure 4.14 shows the seasonal wind roses for nephelometer measurements at Carnsore Point for 2017. It is clear from these plots that there is a large winter component associated with these measurements, which may be reflective of highly absorbing material such as black carbon.

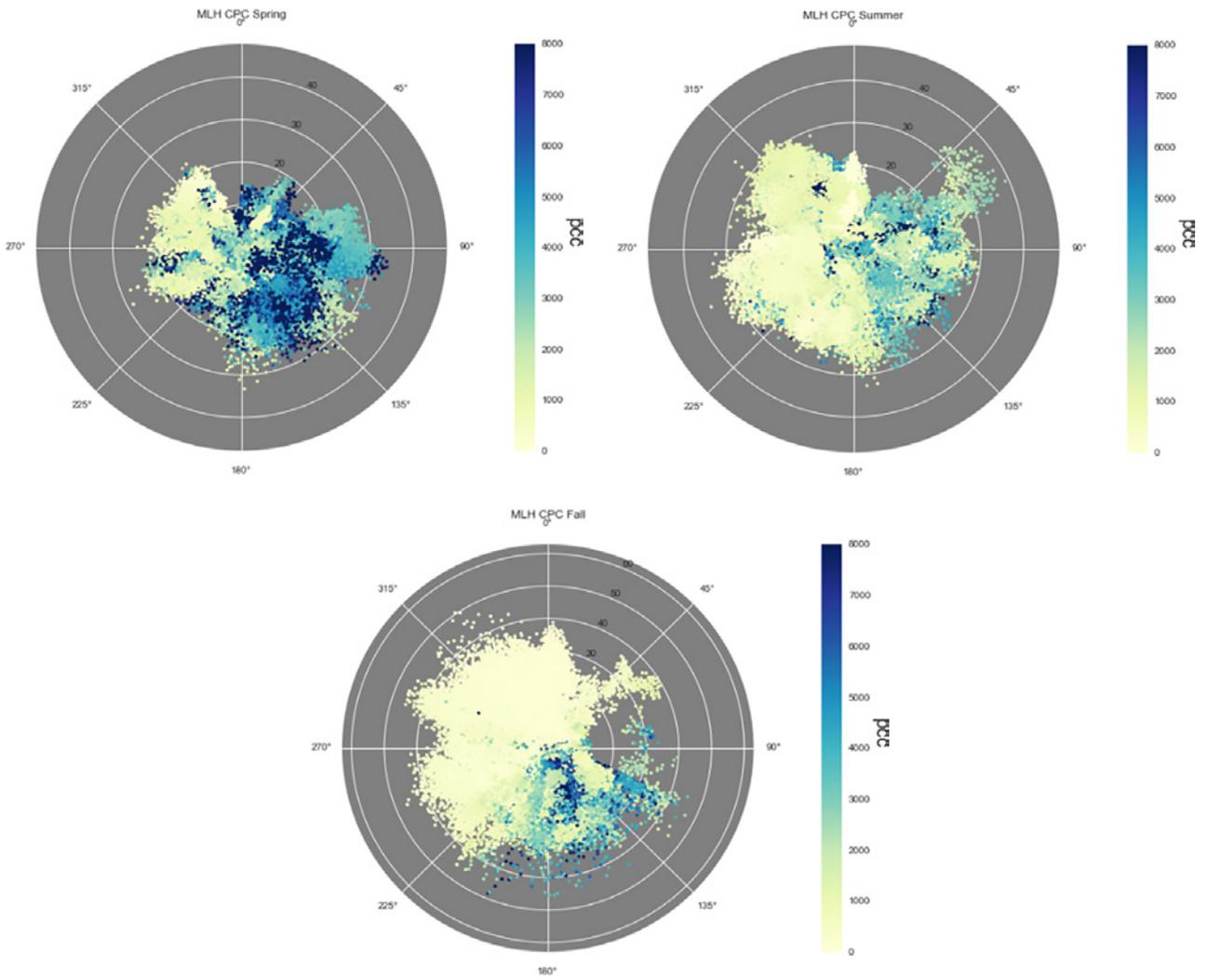


Figure 4.9. Seasonal wind roses for CPC measurements at Malin Head.

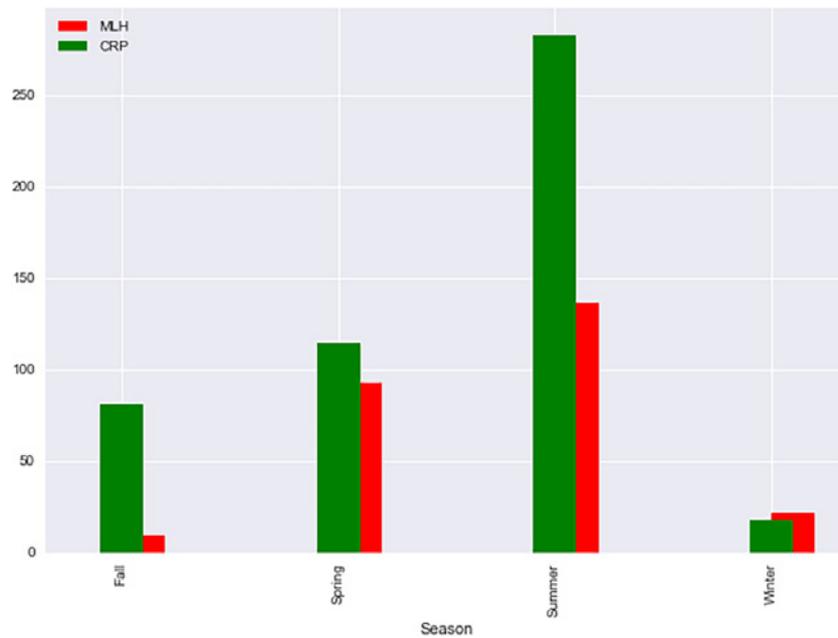


Figure 4.10. Seasonal number of CPC events above 30,000 counts for clean air sectors at Carnsore Point (CRP) and Malin Head (MLH) from 1 May 2017 to 1 May 2018 (black carbon <math>< 70 \text{ ng m}^{-3}</math>).

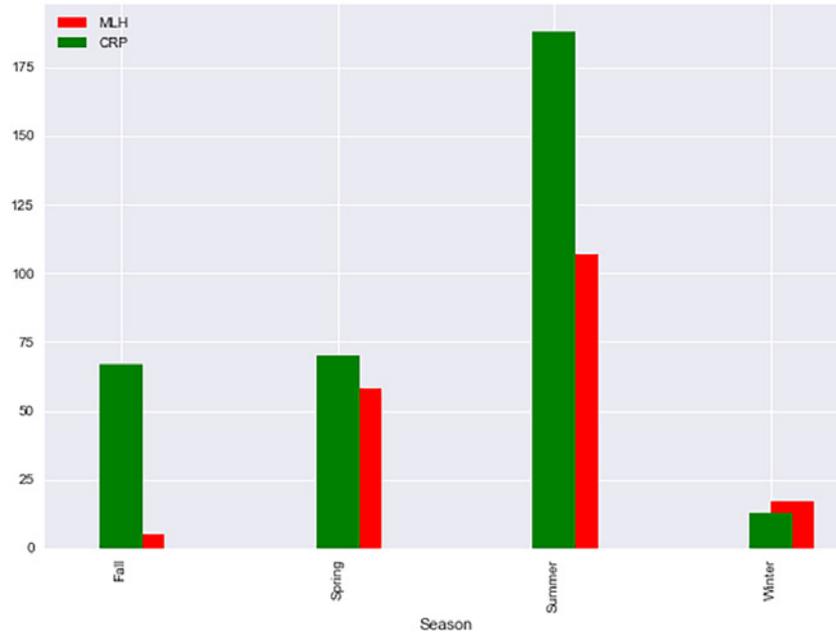


Figure 4.11. Seasonal number of CPC events above 40,000 counts for clean air sectors at Carnsore Point (CRP) and Malin Head (MLH) from 1 May 2017 to 1 May 2018 (black carbon <math>70 \text{ ng m}^{-3}</math>).

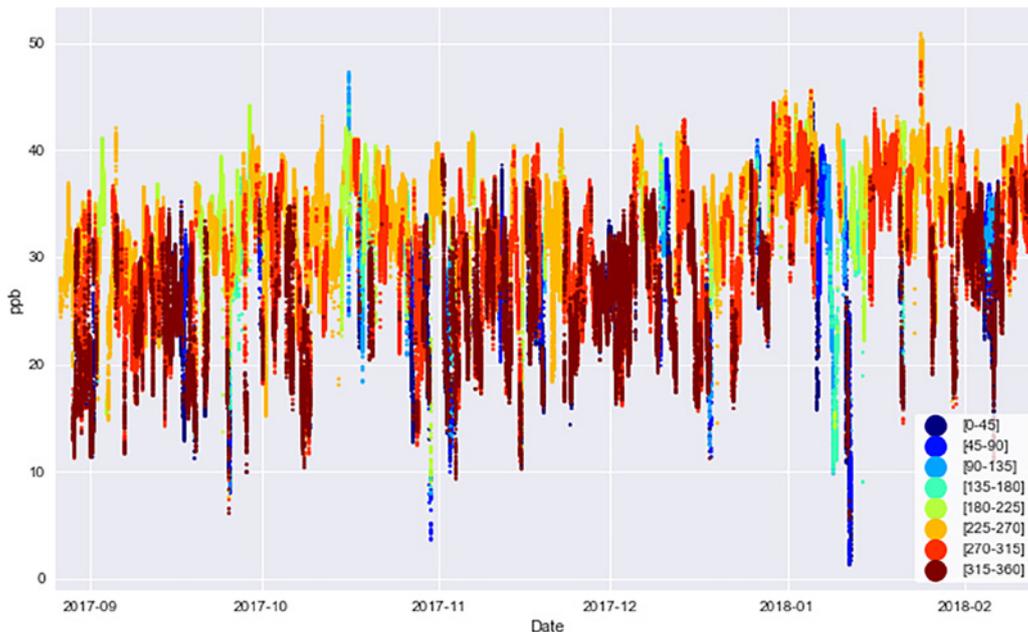


Figure 4.12. Wind sectoring of an ozone time series at Carnsore Point, 26 August 2017–14 February 2018.

#### 4.8 Greenhouse Gas Measurements

Figure 4.15 shows data from the GHG observation network for June 2017–June 2018. Data are captured at the three sites more than 97% of the time. The instruments are serviced every 2 months, which involves cleaning the valves and replacing the rotors and filters. The instruments are also leak checked regularly. The first-generation instruments (G1301)

are reliable instruments. The main issue with these instruments is the significant lead time for repairs at the US repair facility. In order to minimise the need for repairs it is important to reduce heat stress on the instruments and this is one of the main reasons behind the installation of air conditioning at the sites. Calibration cylinders are currently being produced at the central ICOS laboratory in Germany.

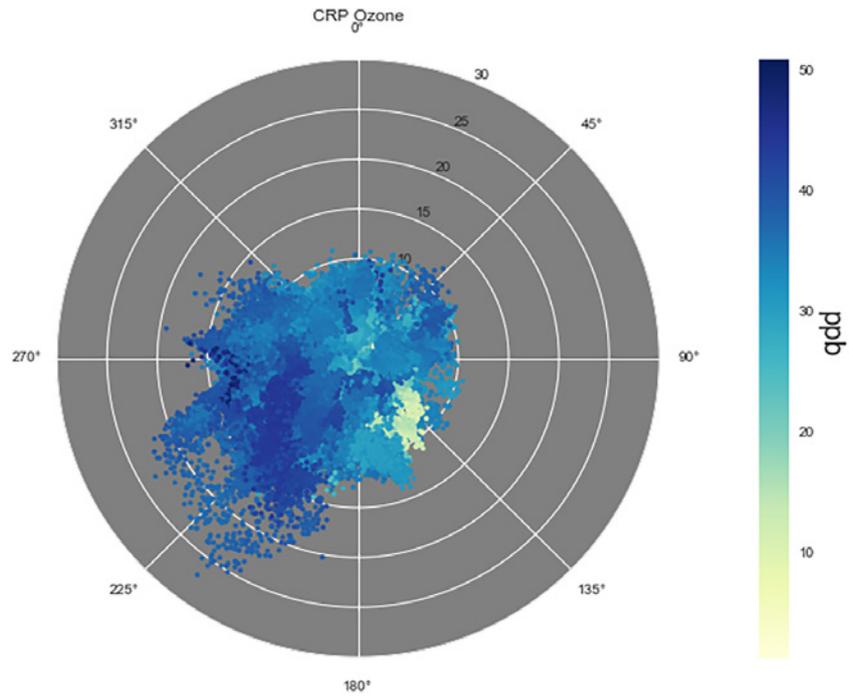


Figure 4.13. Wind rose for Carnsore Point ozone measurements for 2017.

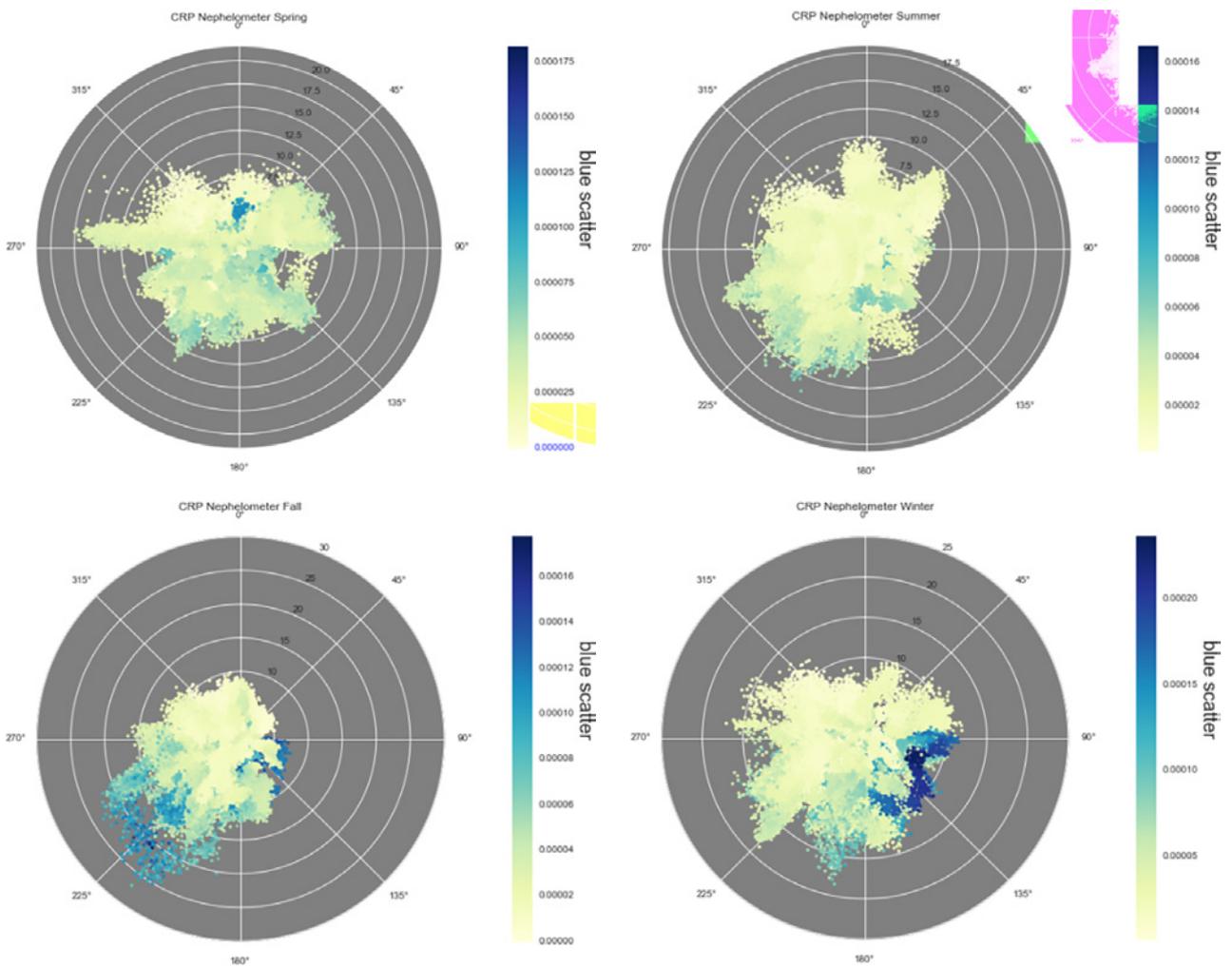
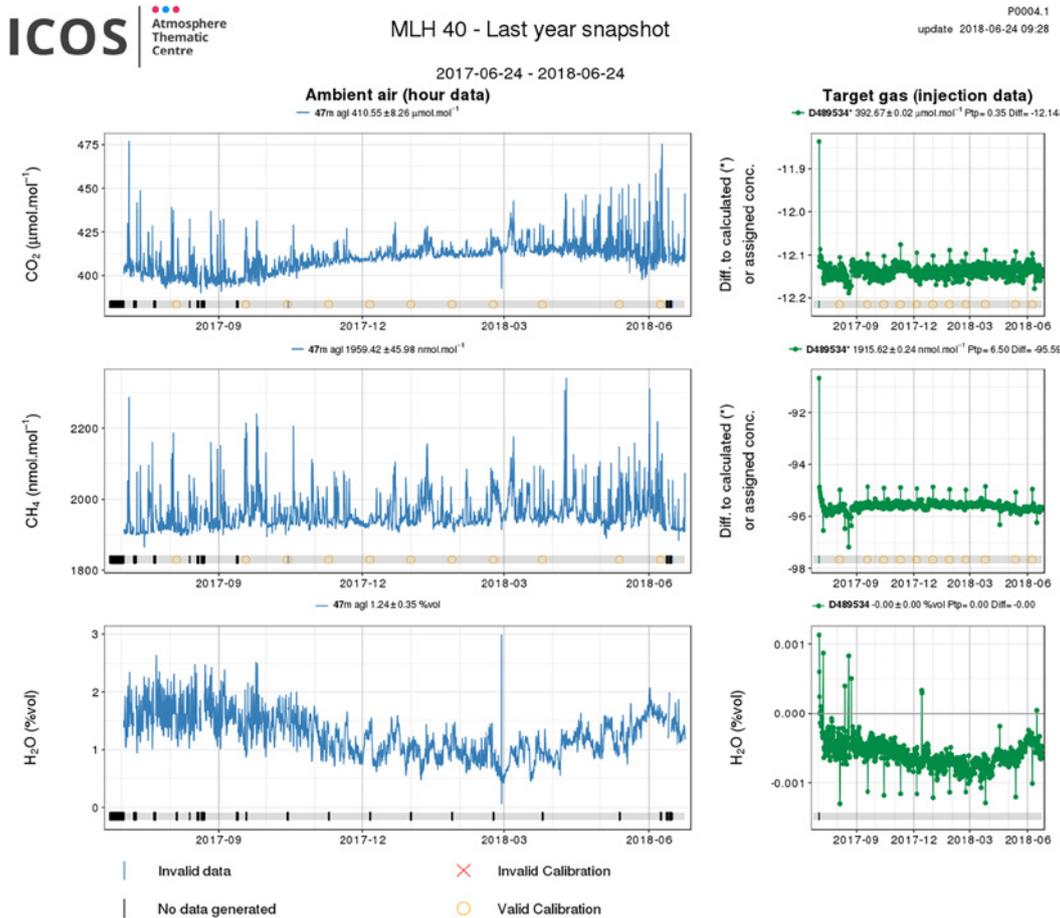
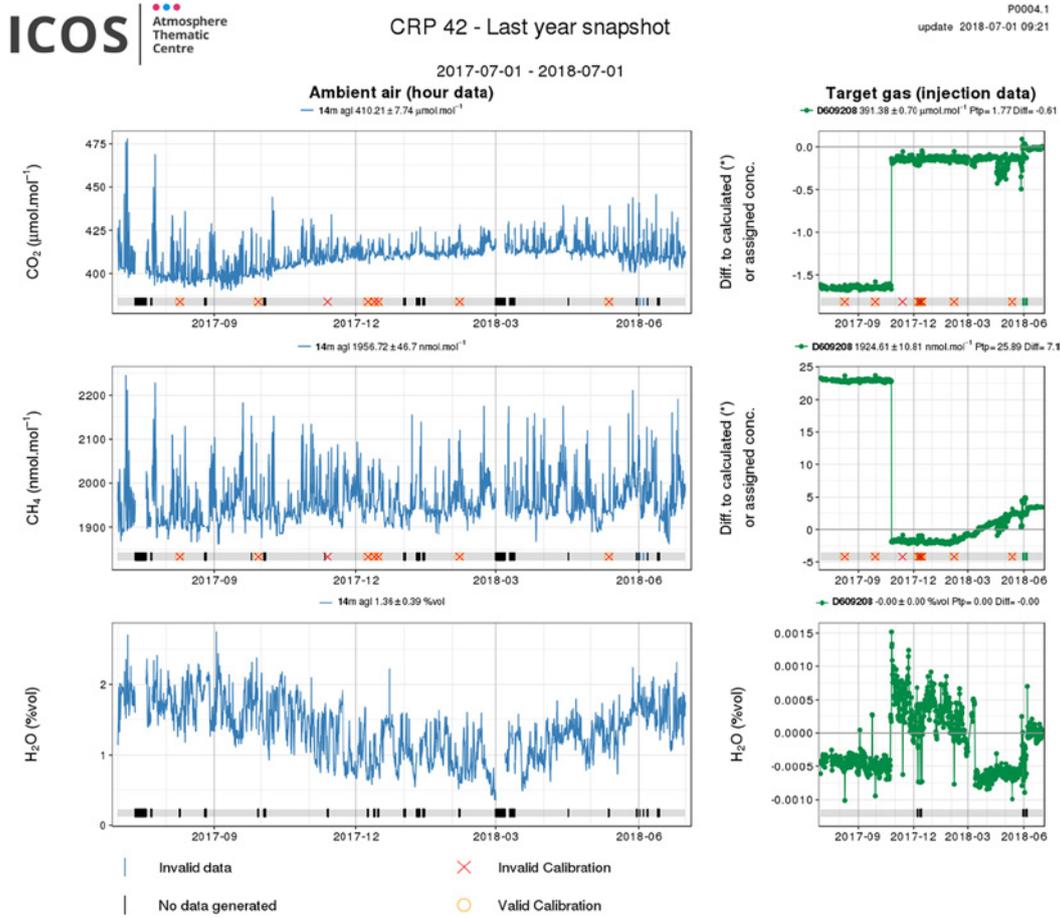


Figure 4.14. Seasonal wind roses for Carnsore Point nephelometer measurements.



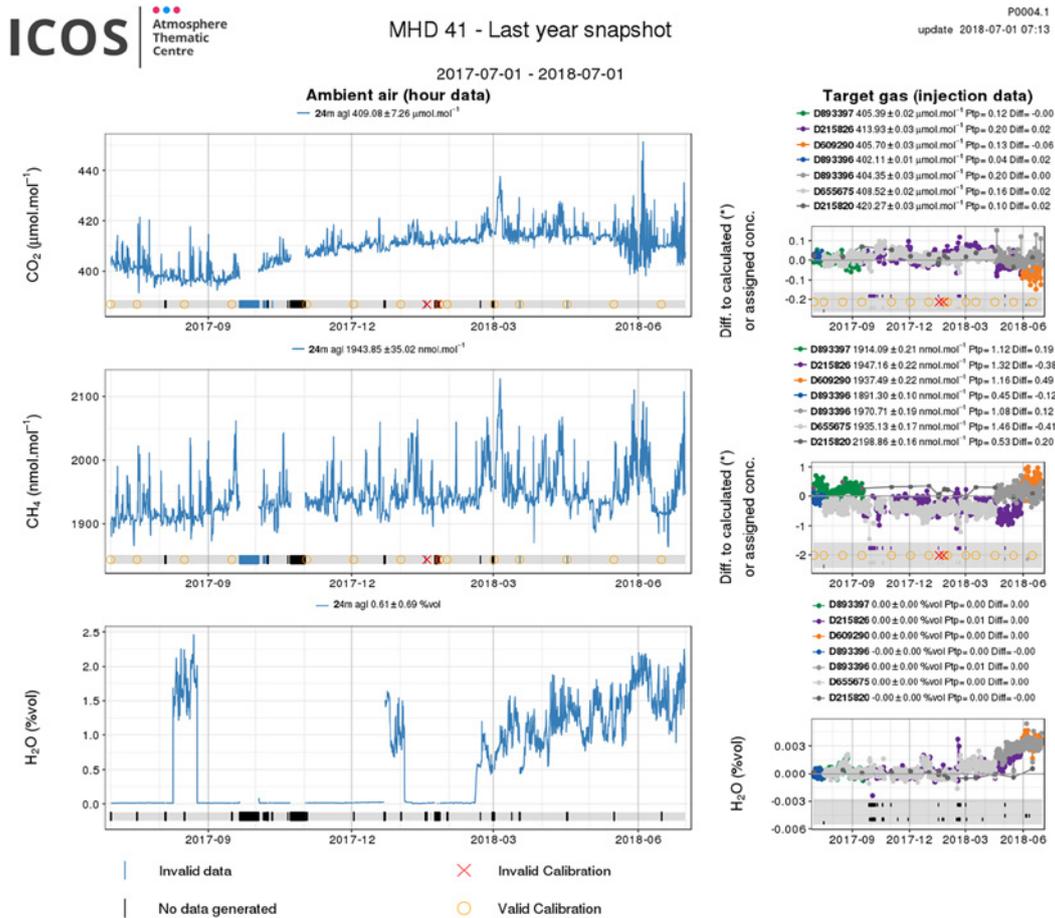


Figure 4.15. GHG measurements from Ireland's Atmospheric Composition and Climate Change Network (<https://icos-atc.lscce.ipsl.fr>). CRP, Carnsore Point; MHD, Mace Head; MLH, Malin Head. ICOS data are licensed under the Creative Commons Attribution 4.0 International licence (<https://creativecommons.org/licenses/by/4.0/>). ICOS RI, licensed under CC4BY.

To minimise down time on the network, it would be prudent to have a spare instrument at each site that has been tested and validated and that can be installed at a site if the primary instrument needs to be repaired.

These data were used to constrain emissions of CH<sub>4</sub> for Ireland in 2012 (Figure 4.16). This shows the capability of network data to help in the determination of Irish GHG emissions.

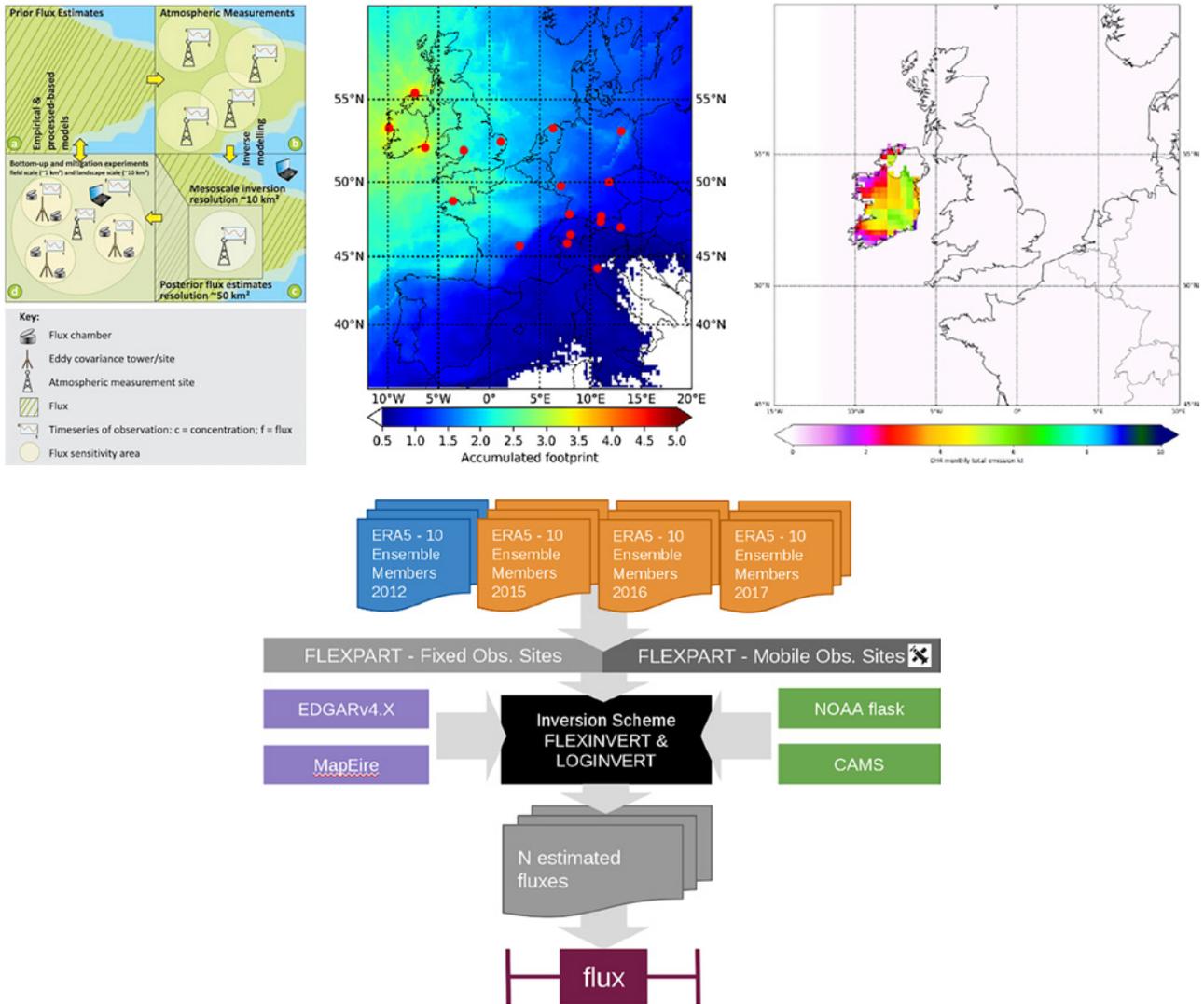


Figure 4.16. Ireland's GHG inversion system. CAMS, Copernicus Atmospheric Monitoring Service; EDGAR, Emissions Database for Global Atmospheric Research; NOAA, National Oceanic and Atmospheric Administration.

# 5 Recommendations for EPA Climate Change and Atmospheric Composition Network and Recent Developments

## 5.1 Recommendations

The main recommendations for the ensured continuation of this network are:

- continued development of the network infrastructure;
- movement of the network to full ICOS compliance (see section 5.1.1);
- continued investment in the updating of equipment and replacement of obsolete equipment;
- development of a set of ICOS complementary measurements.

### 5.1.1 ICOS technical requirements

These are outstanding compliance issues that need to be addressed with respect to the ICOS network.

#### *Water removal for instruments*

The ICOS specification document contains the following recommendation regarding water removal from the air sampling stream at the three Irish stations. In light of this recommendation it is proposed that dryers are installed at the three planned ICOS sites.

The following recommendations were presented and discussed during the Atmosphere MSA (Monitoring Station Assembly) of June 2015 in EMPA. The presence of water vapour in the sample can induce important errors in measurements if it is not properly dealt with (for example by drying the sample or applying water vapour corrections). If there is a risk of water condensation at the site (especially in southern Europe for example), it is recommended to use a dryer installed close to the sampling tubing entrance point in the room. Two drying options are proposed:

1. Cryogenic water trap. This option offers the best performance but is demanding in terms of maintenance and the risk of leakage is an issue.

2. Nafion. The performance is lower compared to cryogenic water trap but brings nevertheless satisfactory results.

If there is no risk of water condensation (most of the European sites): Remote station: drying with Nafion followed by manufactory water corrections or instrument specific H<sub>2</sub>O correction determined by ATC (Atmospheric Thematic Centre) can be implemented.

For other stations: instrument specific H<sub>2</sub>O correction determined by ATC, Nafion, or Cryogenic water trap can be implemented.

Laurent, 2016, p. 32

#### *Digital flowmeters*

One of the more recent requirements from ICOS is the installation of digital flowmeters for monitoring both the sampling line flow (to confirm flow rate, velocity and, by implication, residence time) and the instrument sampling flow.

#### *Swagelok fittings, Valco fittings, tools and consumables, spare instrument pumps and line flushing pumps*

A limited supply of Swagelok fittings for lines, sampling line connections, pump connections, etc., is available for the installation of proposed modifications, as well as routine maintenance. For the sampling valve systems, a very limited supply of Valco fittings, valve replacement parts and spare sampling valves is available. In addition, there are other consumables associated with sampling systems and instrument maintenance that are required to minimise any down time.

### *ICOS cylinder storage*

The ICOS specification document states that:

At the station, it is highly recommended that cylinders and associated pressure regulators are installed in an air temperature controlled room in order to avoid potential bias induced by temperature variation. Moreover, it is recommended that tanks and pressure regulators are installed within a dedicated enclosed rack to buffer temperature changes potentially induced by the air conditioning air flow. The ideal cylinder position within the rack is supine (horizontal) in order to limit stratification and fractionation effects.

Laurent, 2016, p. 35

According to this recommendation it is proposed that compliant racking is installed at each of the planned ICOS sites.

### *ICOS-compliant regulators*

Two sets of ICOS-compliant regulators were procured in 2018 and are currently being tested. It is recommended that two more sets of regulators are bought for the other two instruments currently on the network.

### *GCWerks for Malin Head and Mace Head.*

In 2017 GCWerks was installed at Carnsore Point for ICOS-independent analysis of network GHG data. It is proposed that this installation is extended to Mace Head and Malin Head for 2018.

### *Full set of ICOS-compliant meteorology required at Malin Head*

The exact time frame for installation of tower sampling at Malin Head remains unclear because a public tender is being negotiated to carry out electrical work at the site. The tower utilises a full set of ICOS-compliant anemometry while a temporary set of anemometry equipment was installed at the current sampling location (in the separate lower building) at the end of 2017. It is recommended that this should be replaced with a complete set of ICOS-compliant anemometry, which can be transferred to the tower when tower sampling is installed.

### *Mirrored ICOS-dedicated server*

Currently, all ICOS data are streamed to a dedicated server at NUIG and a weekly back-up copy is made on a different server. It is recommended that a second mirrored server is installed to preclude the possibility of data loss. This would be installed off site and be managed by NUIG personnel.

### *Temperature sensors at Mace Head*

These are required to document the performance of the air conditioning unit at the site. It is important to understand this performance in real time as it can have a negative effect on the performance of the GHG instrumentation.

## **5.1.2 Recommended instrumentation for measurements and characterisation of short-lived climate forcers**

Currently, we are in the process of staged replacement of the GHG analysers at the proposed ICOS sites (Mace Head, Malin Head and Carnsore Point). A new instrument was installed at Malin Head in early 2018 and this is currently being evaluated and compared with the existing instrument. The replacement of the GHG instruments at Mace Head and Carnsore Point should be undertaken as these are currently nearing the end of service. Given the importance to Ireland of agriculture-related N<sub>2</sub>O emissions, it is also recommended that N<sub>2</sub>O instrumentation is installed to constrain Irish emissions. In addition, we propose mirroring of the measurements carried out at Carnsore Point at Malin Head and recommend the installation of an SMPS/FIDAS for particle mass and size distribution characterisation, as well as a nephelometer and an ACSM.

## **5.1.3 Recommendations for ICOS infrastructural requirements**

### *Dedicated ICOS website for atmospheric monitoring stations*

Currently, the output from the Irish ICOS network (trace gas concentrations, meteorological measurements and supporting measurements) is available on the Mace Head website ([http://www.macehead.org/index.php?option=com\\_content&view=category&layout=blog&id=96&Itemid=30](http://www.macehead.org/index.php?option=com_content&view=category&layout=blog&id=96&Itemid=30)). In

the interest of better transparency, a more visible web portal that provides a more complete description of the network would be useful and facilitate further engagement with stakeholders.

#### *Air conditioning at Malin Head*

The current ICOS instrumentation resides in a non-air conditioned building. The current plan is to relocate the instrument to the upper building to facilitate access to the main meteorological tower on site. This has been a relatively slow process because of the need for complete rewiring of the site and the associated tender process. The precision of the instrumentation is dictated in part by the stability of the room temperature. It is apparent from temperature measurements taken that there is quite a large seasonal temperature fluctuation in the room, as well as some daily variability. It is proposed that air conditioning is installed in both the lower and the upper building.

#### *Air conditioning at Mace Head*

The efficiency of the current air conditioning system at Mace Head has started to decline and during the summer period the temperature variation is beginning to deviate considerably from ICOS recommendations. The air conditioning system should therefore be

replaced. Traditional hydrofluorocarbon refrigerants are not suitable because of ongoing measurements being carried out under the Advanced Global Atmospheric Gases Experiment (AGAGE) programme; therefore, an alternative type of air conditioner is required (water cooled).

## 5.2 Recent Developments

### 5.2.1 *Ozone data incorporated into EPA measurements*

The ozone data from the three sites in the network (Malin Head, Mace Head and Carnsore Point) are now provided in the form of hourly averages that are uploaded to the EPA's data-handling system. This has been working well although the satellite broadband connection at Carnsore Point is not as reliable as connections at the other sites (this should be improved as part of the redevelopment of Carnsore Point). Figure 5.1 shows EPA visualisations of ozone data from Malin Head from October 2018.

### 5.2.2 *Building work at Carnsore Point*

After meeting with various stakeholders over a number of years (ESB and EPA) and going through the planning process, work started on the new EPA facility in October 2018 (Figure 5.2). A number of delays were

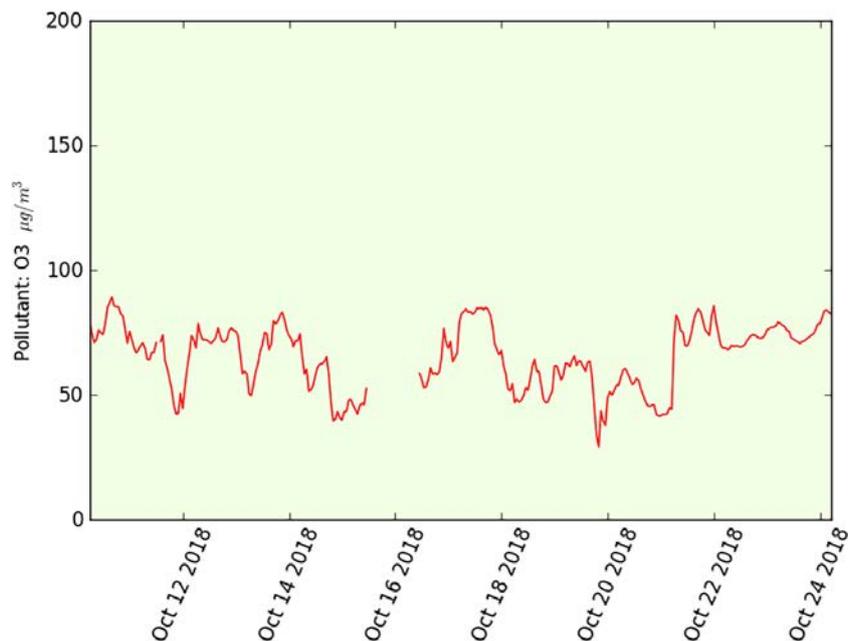


Figure 5.1. Ozone measurements at Malin Head from 10 October to 24 October 2018 visualised through the EPA portal.



**Figure 5.2. Start of site redevelopment at Carnsore Point.**

encountered in the instigation of this work. These delays were as follows:

- A delay in the planning process. There had been no plans for the original site and this made the application more time-consuming than it might have been otherwise.
- A delay in tower certification, which is provided by the ESB. There was an issue with a faulty guide wire that needed to be replaced and the ESB would not provide access to the site until this replacement was undertaken. This ended up taking over 9 months.
- A delay in receiving permits to work on the site. Permits to work on the site are issued by the wind farm site manager. A new site manager was employed and the necessary qualifications to issue these permits had to be obtained.

### 5.2.3 *EMEP campaign*

#### *Background*

Carbonaceous aerosol sources are abundant and both anthropogenic and natural; thus, identifying

and quantifying sources are important to develop efficient abatement strategies. In particular, there is an interest in distinguishing the relative contribution of combustion sources using either fossil fuels (such as road transportation) or biomass (such as residential wood burning). Source apportionment of the carbonaceous aerosol is usually based on filter samples with a 24-hour time resolution or longer, and typically includes several species that are not part of regular monitoring. Detailed separation of sources is challenging and a certain overlap between apportioned sources is likely. Further, the various source apportionment approaches are costly and the results are available only after sample collection. This necessitates high time resolution and online information on aerosol particles from specific sources. This demand is not only from scientists wanting to understand atmospheric dynamics and composition, but also from air quality managers, to enable them to inform, educate and increase public awareness of air quality-related issues and to design air quality management plans. Online and high time resolution measurements from, for example, aerosol mass spectrometers have become available in recent years. However, the mass spectra must be processed post

sampling to obtain source-apportioned data on the carbonaceous aerosol and these data are thus not available online.

Separation of equivalent black carbon (EBC) into fossil fuel (EBCff) and wood-burning (EBCwb) sources is possible using multi-wavelength measurement of the absorption coefficient (Sandradewi *et al.*, 2008a), and for the AE33 aethalometer this is an online feature. A few European studies have reported using the multi-wavelength aethalometer for source apportionment of EBC, even in the rural background environment and for an extended period of time (Herich *et al.*, 2011). With an increasing number of multi-wavelength aethalometers employed at European rural background sites, as part of regular monitoring, and the substantial focus on black carbon and its sources (wood burning in particular), it appeared timely to focus on such measurements in a wintertime EMEP/ACTRIS intensive measurement period. The intensive measurement period aimed to test the multi-wavelength aethalometer source apportionment approach in the European rural background environment, including low loading areas in Scandinavia and more polluted regions in Central Europe, and in areas differing in source composition,

preferably also with an influence of coal combustion. Further, it compared EBCwb and EBCff apportioned by the multi-wavelength aethalometer approach with filter-based measurements of the biomass-burning tracer levoglucosan and elemental carbon for validation purposes.

#### *Aims*

- To quantify EBCff and EBCwb using multi-wavelength aethalometer measurement and to validate this approach using concurrent offline measurements of the biomass-burning tracer levoglucosan (and elemental carbon, organic carbon and total carbon) for a wider part of Europe.
- To provide a harmonised data set for model validation.
- To initiate regular monitoring of EBCff and EBCwb and the reporting of such data to EBAS.

Three EMEP sites (Mace Head, Carnsore Point and Malin Head) and the AEROSOURCE site at UCD took part in this campaign. Black carbon data have been reported and we are currently awaiting the results of the organic carbon analyses.

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# Abbreviations

<b>ACSM</b>	Aerosol chemical speciation monitor
<b>ATNTA</b>	Attenuation threshold
<b>CLRTAP</b>	Convention on Long-range Transboundary Air Pollution
<b>CPC</b>	Condensation particle counter
<b>CPVU</b>	Computer pump vacuum unit
<b>CRDS</b>	Cavity ring-down spectroscopy
<b>DAS</b>	Data acquisition system
<b>EBC</b>	Equivalent black carbon
<b>EBCff</b>	Equivalent black carbon from fossil fuel
<b>EBCwb</b>	Equivalent black carbon from wood burning
<b>EMEP</b>	Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe
<b>EPA</b>	Environmental Protection Agency
<b>ERIC</b>	European Research Infrastructure Consortium
<b>ESB</b>	Electricity Supply Board
<b>EU</b>	European Union
<b>GHG</b>	Greenhouse gas
<b>ICOS</b>	Integrated Carbon Observation System
<b>ICOS RI</b>	Integrated Carbon Observation System Research Infrastructure
<b>LSCE</b>	Laboratory for Sciences of Climate and Environment
<b>MAAP</b>	Multi-angle absorption photometer
<b>NUIG</b>	National University of Ireland Galway
<b>PM<sub>1</sub></b>	Particulate matter $\leq 1 \mu\text{m}$
<b>PM<sub>2.5</sub></b>	Particulate matter $\leq 2.5 \mu\text{m}$
<b>PM<sub>10</sub></b>	Particulate matter $\leq 10 \mu\text{m}$
<b>PMT</b>	Photomultiplier tube
<b>SLCF</b>	Short-lived climate forcer
<b>SMPS</b>	Scanning mobility particle sizer
<b>TEOM</b>	Tapered element oscillating microbalance
<b>TGT</b>	Target (cylinder)
<b>UCD</b>	University College Dublin
<b>UN</b>	United Nations

## AN GHNÍOMHAIREACHT UM CHAOMHNÚ COMHSHAOIL

Tá an Gníomhaireacht um Chaomhnú Comhshaoil (GCC) freagrach as an gcomhshaoil a chaomhnú agus a fheabhsú mar shócmhainn luachmhar do mhuintir na hÉireann. Táimid tiomanta do dhaoine agus don chomhshaoil a chosaint ó éifeachtaí díobhálacha na radaíochta agus an truaillithe.

## Is féidir obair na Gníomhaireachta a roinnt ina trí phríomhréimse:

**Rialú:** Déanaimid córais éifeachtacha rialaithe agus comhlionta comhshaoil a chur i bhfeidhm chun torthaí maithe comhshaoil a sholáthar agus chun díriú orthu siúd nach gcloíonn leis na córais sin.

**Eolas:** Soláthraimid sonraí, faisnéis agus measúnú comhshaoil atá ar ardchaighdeán, spriocdhírthe agus tráthúil chun bonn eolais a chur faoin gcinnteoireacht ar gach leibhéal.

**Tacaíocht:** Bimid ag saothrú i gcomhar le grúpaí eile chun tacú le comhshaoil atá glan, táirgiúil agus cosanta go maith, agus le hiompar a chuirfidh le comhshaoil inbhuanaithe.

## Ár bhFreagrachtaí

### Ceadúnú

Déanaimid na gníomhaíochtaí seo a leanas a rialú ionas nach ndéanann siad dochar do shláinte an phobail ná don chomhshaoil:

- saoráidí dramhaíola (*m.sh. láithreáin líonta talún, loisceoirí, stáisiúin aistriúcháin dramhaíola*);
- gníomhaíochtaí tionsclaíocha ar scála mór (*m.sh. déantúsaíocht cógaisíochta, déantúsaíocht stroighne, stáisiúin chumhachta*);
- an diantalmhaíocht (*m.sh. muca, éanlaith*);
- úsáid shrianta agus scaoileadh rialaithe Orgánach Géinmhodhnaithe (*OGM*);
- foinsí radaíochta ianúcháin (*m.sh. trealamh x-gha agus radaiteiripe, foinsí tionsclaíocha*);
- áiseanna móra stórála peitрил;
- scardadh dramhuisece;
- gníomhaíochtaí dumpála ar farraige.

### Forfheidhmiú Náisiúnta i leith Cúrsaí Comhshaoil

- Clár náisiúnta iniúchtaí agus cigireachtaí a dhéanamh gach bliain ar shaoráidí a bhfuil ceadúnas ón nGníomhaireacht acu.
- Maoirseacht a dhéanamh ar fhreagrachtaí cosanta comhshaoil na n-údarás áitiúil.
- Caighdeán an uisce óil, arna sholáthar ag soláthraithe uisce phoiblí, a mhaoirsiú.
- Obair le húdarás áitiúla agus le gníomhaireachtaí eile chun dul i ngleic le coireanna comhshaoil trí chomhordú a dhéanamh ar líonra forfheidhmiúcháin náisiúnta, trí dhírú ar chiontóirí, agus trí mhaoirsiú a dhéanamh ar leasúchán.
- Cur i bhfeidhm rialachán ar nós na Rialachán um Dhramhthrealamh Leictreach agus Leictreonach (DTLL), um Shrian ar Shubstaintí Guaiseacha agus na Rialachán um rialú ar shubstaintí a ídionn an ciseal ózóin.
- An dlí a chur orthu siúd a bhriseann dlí an chomhshaoil agus a dhéanann dochar don chomhshaoil.

### Bainistíocht Uisce

- Monatóireacht agus tuairisciú a dhéanamh ar cháilíocht aibhneacha, lochanna, uisce idirchriosacha agus cósta na hÉireann, agus screamhuisec; leibhéal uisce agus sruthanna aibhneacha a thomhas.
- Comhordú náisiúnta agus maoirsiú a dhéanamh ar an gCreat-Treoir Uisce.
- Monatóireacht agus tuairisciú a dhéanamh ar Cháilíocht an Uisce Snámha.

## Monatóireacht, Anailís agus Tuairisciú ar an gComhshaoil

- Monatóireacht a dhéanamh ar cháilíocht an aeir agus Treoir an AE maidir le hAer Glan don Eoraip (CAFÉ) a chur chun feidhme.
- Tuairisciú neamhspleách le cabhrú le cinnteoireacht an rialtais náisiúnta agus na n-údarás áitiúil (*m.sh. tuairisciú tréimhsiúil ar staid Chomhshaoil na hÉireann agus Tuarascálacha ar Tháscairí*).

## Rialú Astaíochtaí na nGás Ceaptha Teasa in Éirinn

- Fardail agus réamh-mheastacháin na hÉireann maidir le gáis ceaptha teasa a ullmhú.
- An Treoir maidir le Trádáil Astaíochtaí a chur chun feidhme i gcomhar breis agus 100 de na táirgeoirí dé-ocsaíde carbóin is mó in Éirinn.

## Taighde agus Forbairt Comhshaoil

- Taighde comhshaoil a chistiú chun brúnna a shainiú, bonn eolais a chur faoi bheartais, agus réitigh a sholáthar i réimsí na haeráide, an uisce agus na hinbhuanaitheachta.

## Measúnacht Straitéiseach Timpeallachta

- Measúnacht a dhéanamh ar thionchar pleananna agus clár beartaithe ar an gcomhshaoil in Éirinn (*m.sh. mórfheananna forbartha*).

## Cosaint Raideolaíoch

- Monatóireacht a dhéanamh ar leibhéal radaíochta, measúnacht a dhéanamh ar nochtadh mhuintir na hÉireann don radaíocht ianúcháin.
- Cabhrú le pleananna náisiúnta a fhorbairt le haghaidh éigeandálaí ag eascairt as tairmí núicléacha.
- Monatóireacht a dhéanamh ar fhorbairtí thar lear a bhaineann le saoráidí núicléacha agus leis an tsábháilteacht raideolaíochta.
- Sainseirbhísí cosanta ar an radaíocht a sholáthar, nó maoirsiú a dhéanamh ar sholáthar na seirbhísí sin.

## Treoir, Faisnéis Inrochtana agus Oideachas

- Comhairle agus treoir a chur ar fáil d'earnáil na tionsclaíochta agus don phobal maidir le hábhair a bhaineann le caomhnú an chomhshaoil agus leis an gcosaint raideolaíoch.
- Faisnéis thráthúil ar an gcomhshaoil ar a bhfuil fáil éasca a chur ar fáil chun rannpháirtíocht an phobail a spreagadh sa chinn-teoireacht i ndáil leis an gcomhshaoil (*m.sh. Timpeall an Tí, léarscáileanna radóin*).
- Comhairle a chur ar fáil don Rialtas maidir le hábhair a bhaineann leis an tsábháilteacht raideolaíoch agus le cúrsaí práinnfhreagartha.
- Plean Náisiúnta Bainistíochta Dramhaíola Guaisí a fhorbairt chun dramhaíl ghuaiseach a chosaint agus a bhainistiú.

## Múscaill Feasachta agus Athrú Iompraíochta

- Feasacht comhshaoil níos fearr a ghiniúint agus dul i bhfeidhm ar athrú iompraíochta dearfach trí thacú le gnóthais, le pobail agus le teaghlaigh a bheith níos éifeachtúla ar acmhainní.
- Tástáil le haghaidh radóin a chur chun cinn i dtithe agus in ionaid oibre, agus gníomhartha leasúcháin a spreagadh nuair is gá.

## Bainistíocht agus struchtúr na Gníomhaireachta um Chaomhnú Comhshaoil

Tá an ghníomhaíocht á bainistiú ag Bord Iáinimseartha, ar a bhfuil Ard-Stiúrthóir agus cúigear Stiúrthóirí. Déantar an obair ar fud cúig cinn d'Oifigí:

- An Oifig um Inmharthanacht Comhshaoil
- An Oifig Forfheidhmithe i leith cúrsaí Comhshaoil
- An Oifig um Fianaise is Measúnú
- Oifig um Chosaint Radaíochta agus Monatóireachta Comhshaoil
- An Oifig Cumarsáide agus Seirbhísí Corparáideacha

Tá Coiste Comhairleach ag an nGníomhaireacht le cabhrú léi. Tá dáréag comhaltáí air agus tagann siad le chéile go rialta le plé a dhéanamh ar ábhair inní agus le comhairle a chur ar an mBord.

Authors: Damien Martin and Colin O'Dowd

The Atmospheric Composition and Climate Change network is a valuable established national research and monitoring infrastructure. It has been developed incrementally based on measurements of greenhouse gases (GHG's) and short-lived climate forcers (SLCF's) including aerosol/particulate matter (PM) chemical and physical characteristics in line with best practice from both pan-European and global monitoring programmes. GHG measurements are undertaken under the umbrella of the ICOS (Integrated Carbon Observing System) pan-European research infrastructure, while additional observations are conducted under EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe). The report describes the operation, development and expansion of the network activities and infrastructure.

### Identifying Pressures

The 2015 Paris Agreement established the global policy response to climate change. A key objective of that agreement is that global GHG emissions are balanced by removals from the atmosphere during the second half of this century. Achieving both national and global pathways will require an increased understanding of emissions and removals by sinks and the processes by which the latter can be enhanced. Measurements of GHGs can be used with modelling techniques to verify emission inventories and in particular to assess the balance of emissions and removals from the land sector.

Air pollution levels in Ireland are determined by local emissions and emissions in Europe, North America and further afield through the hemispheric transport of pollutants. Actions to address air pollution are taken at these levels and include the Convention on Long-range Transboundary Air Pollution (CLRTAP) and its protocols, which link to the EU Clean Air for Europe (CAFE) programme, including the National Emissions Ceiling Directive. EMEP is a programme under CLRTAP that addresses the requirement for Parties to the Convention to undertake air quality monitoring. A fundamental understanding of the nature, scope and magnitude of transboundary air pollution, which is monitored using this network, is essential to understanding its relative source contribution and to supporting national and international efforts to improve air quality.

### Informing Policy

A national GHG monitoring and analysis network, especially one linked to the ICOS European Research Infrastructure Consortium, can help to resolve fundamental questions relating to Ireland's GHG emissions which can inform future climate change policy, for example:

1. Reduce uncertainties in determining the nature and extent of the sources and sinks for GHG emissions and removals in Ireland.
2. Assess how meteorological and other factors influence these sinks over timescales ranging from seasons to decades and the interplay between these and management systems.
3. Highlight geographical areas with high levels of uncertainty in bottom-up analysis of gases such as nitrous oxide and methane.
4. Produce an independent integrated and comprehensive analysis of emissions and removals in Ireland in the context of meeting the goal of climate neutrality.

Monitoring of aerosol chemical and physical characteristics and other SLCFs can be used to elucidate transboundary air pollution and underpin national and international monitoring and response strategies.

### Developing Solutions

This fellowship has enabled and sustained scientific work on a national monitoring network. Infrastructure has been continually developed over the course of the fellowship that will facilitate long-term sustainable measurements. Given the national and international importance of climate change, it is critical to maintain a level of investment in infrastructure, analytical systems and associated complementary measurements to ensure that Ireland is at the forefront of this critical area in order to inform policy and facilitate meaningful solutions. Ireland is already at the forefront of GHG and transboundary air pollution monitoring. Further developing the inversion modelling techniques to include SLCFs would be an important extension of network capability, particularly in the areas of source apportionment and emission verification. Robust source apportionment of air pollution is essential to understand the complex nature of its sources and identify where to target policies to improve air quality and maximise societal benefit.