Environmental Science: Atmospheres

ROYAL SOCIETY OF CHEMISTRY

PAPER

View Article Online
View Journal | View Issue



Cite this: *Environ. Sci.: Atmos.*, 2025, **5**, 1282

# $SO_2$ and $NO_x$ emissions from ships in North-East Atlantic waters: *in situ* measurements and comparison with an emission model

James D. Lee, *[ab] Dominika Pasternak, [ac] Shona E. Wilde, [ad] Will S. Drysdale, [ab] Stuart E. Lacy,[a] Sarah J. Moller, [ab] Marvin Shaw, [ab] Freya A. Squires, [ae] Pete Edwards, [ab] Loren G. Temple, [ak] Hugh Coe, [fg] Huihui Wu, [fh] Stephanie E. Batten, [ij] Stephane Bauguitte, [i] Chris Reed, [i] Thomas G. Bell, [k] Mingxi Yang, [k] Jukka-Pekka Jalkanen [l] and Javier Buhigas[m]

Measurements of apparent fuel sulfur content in ship exhausts (aFSC) and $NO_x/CO_2$ ratios were made from an airborne and ground-based platforms in the open Atlantic Ocean and the European sulfur emission control area (SECA) during multiple field campaigns from 2019 to 2023. In the open ocean a nearly 10-fold decrease in the mean aFSC demonstrates the strong impact the International Maritime Organization regulation change in 2020 had on sulfur emissions from ships. In 2019, 8 ships out of 19 showed a measured aFSC higher than the 3.5% limit at the time and in 2021 and 2022, 5 ships out of 78 were observed to be higher than the new 0.5% limit. In the SECA in the English Channel, the average aFSC across both 2019 and 2021 measurements was $0.04 \pm 0.01\%$, well below the more stringent 0.1% limit. In the port of Valencia, Spain, which is not in a SECA, observed aFSC was on average much lower than in the open ocean and close to the EU Sulfur directive of 0.1% fuel sulfur content in port areas if the ship stays more than 2 hours in port. In the Port of Tyne (within the European SECA), the aFSC is virtually identical to those measured in the English Channel, with no ships breaching the 0.1% limit. On average, measured aFSCs agree well with the estimates of the Ship Traffic Emission Assessment Model (STEAM3), although the model does not pick up outliers that breach limits. In the open ocean in 2019 the $NO_x/CO_2$ ratio was $0.021 \pm 0.002$, with ratios observed in port significantly lower (Port of Tyne $0.009 \pm 0.001$, Port of Valencia $0.011 \pm 0.001$), with a switch to auxiliary engines in ports a potential reason for this lower emission ratio. This work presents the first aircraft-based measurements of aFSC from ships outside of sulfur control zones since the change in sulfur emission regulations in 2020 and largely justifies the assumption that is often made that ships now emit around 7 times less sulfur than before 2020.

Received 1st August 2025
Accepted 2nd October 2025

DOI: 10.1039/d5ea00089k

rsc.li/esatmospheres



## Environmental significance

As 70% of shipping emissions are produced within 400 km of the coast, emissions of air pollutants (including $SO_2$, $NO_x$, VOCs and PM) can cause severe health and environmental problems to these regions. It is for this reason that, from 2005 the International Maritime Organisation (IMO) started creating Sulphur Emission Control Areas (SECA) *e.g.* in the North Sea, Baltic Sea or Caribbean Sea. Sulphur content in fuel by mass in these areas had a limit of 1.5% until 2010, 1.0% until 2015 and 0.1% since then. When sulphur emissions from ships occur in remote environments, the sulfate aerosol produced acts to cool the planet both directly by scattering sunlight through the formation of so-called ship tracks and more widely by increasing the albedo of clouds. This cooling due to sulfate aerosol offsets some of the warming effect of greenhouse gasses and is the largest uncertainty in determining the change in the Earth's radiative balance by human activity. The IMO also regulates sulphur emissions in international waters, setting the sulphur content limits at 4.5% until 2012, 3.5% up until 2020 and 0.5% since then. In order to assess the effect of shipping emissions on both local and regional air quality and the global climate, it is important to have robust experimental evidence, especially after regulations change. Emissions from ships are difficult to study, especially in regions a long distance from land. Here we present the first experimental evidence of the drop in sulphur emissions from ships, concurrent with the change in IMO regulations in international waters. This largely justifies most model studies that have tried to predict the change in global radiative forcing post regulation, which often just assume a 7× reduction in sulfur.

[a]*Department of Chemistry, University of York, York, UK. E-mail: james.lee@york.ac.uk*
[b]*National Centre for Atmospheric Science, University of York, York, UK*
[c]*Now at Deutsches Zentrum für Luft- und Raumfahrt (DLR) Oberpfaffenhofen, Germany*
[d]*Now at Energy & Emissions Research Laboratory, Department of Mechanical and Aerospace Engineering, Carleton University, Ottawa, Canada*
[e]*Now at British Antarctic Survey, Madingley Road, Cambridge, UK*
[f]*Department of Earth and Environmental Sciences, University of Manchester, UK*
[g]*National Centre for Atmospheric Science, University of Manchester, UK*
[h]*Now at Université Paris Est Créteil and Université Paris Cité, CNRS, LISA, F-94010 Créteil, France*
[i]*Facility of Airborne Atmospheric Measurements, National Centre for Atmospheric Science, University of Cranfield, Cranfield, UK*
[j]*Now at Department of Earth Sciences, Royal Holloway, University of London, UK*
[k]*Plymouth Marine Laboratory, Prospect Place, Plymouth, UK*
[l]*Atmospheric Composition Research, Finnish Meteorological Institute, Helsinki, Finland*
[m]*Opus RS Europe, Spain*

# 1 Introduction

Large amounts of sulfur in the atmosphere have detrimental effects on human health and the environment. Gas phase sulfur dioxide ($SO_2$) has a direct association with human respiratory and cardiovascular diseases.[1–3] $SO_2$ is also a key precursor of sulfate aerosol, which has a cooling effect on climate by increasing cloud droplet number and hence cloud brightness.[4] The sulfate particulate from marine emissions not only contributes to air pollution but also increases the risk of acid rain formation.[5] It is therefore vital to understand emissions of $SO_2$ in order to provide accurate predictions of current and future air quality and climate.

Human activity is responsible for the emission of the majority of atmospheric sulfur, which in nature is not particularly abundant.[6] These emissions come mostly from the Northern Hemisphere, however in the Southern Hemisphere they are also the dominant source of sulfur.[7] In the last two hundred years atmospheric sulfur trends were driven by anthropogenic emissions.[8] After peaking in 1970 a substantial reduction in $SO_2$ emissions was observed globally from 1980s to 2000s.[6] It was driven by legislation limiting sulfur emissions in Europe and North America, who were the major contributors at the time. Between 2000–2006 emissions increased primarily due to growth in China. However, these were soon addressed and reduced, so the global trend returned to a decline. Shipping is the most important anthropogenic source of sulfur in marine environments (largely as $SO_2$) and is estimated to be responsible for around 13% of global sulfur emissions.[9] When sulfur emissions from ships occur in remote environments, the sulfate aerosol acts to cool the planet both directly by scattering sunlight through the formation of so-called ship tracks,[10,11] and more widely by increasing the albedo of clouds.[12] This cooling due to sulfate aerosol offsets some of the warming effect of greenhouse gasses and is the largest uncertainty in determining the change in the Earth's radiative balance by human activity.[13,47,49] In addition, as 70% of shipping emissions are produced within 400 km of the coastlines[14] they can cause severe health and environmental problems to these coastal regions.[15–17]

The shift to clean fuels came to shipping much later than to land based engines. However, recent changes in international regulations have resulted in rapid changes in emissions from the sector. The International Maritime Organisation (IMO) is responsible for regulating shipping emissions. From the late 1980s the IMO started creating Special Areas, where for oceanographical or ecological reasons additional measures are taken to prevent pollution from oil, sewage or garbage. The first such area created was the Gulf of Aden. From 2005 Sulfur Emission Control Areas (SECA) started being introduced *e.g.* in the North Sea, Baltic Sea or Caribbean Sea. The legal basis for all of these areas is the International Convention for the Prevention of Pollution from Ships (MARPOL) which came into force in 1983. SECAs predominantly limit sulfur emissions but some also have regulations targeting $NO_x$ emissions. From 2015 sulfur content in fuel (Fuel Sulfur Content – FSC) in these areas should not exceed a strict limit of 0.1% by mass. In the past this limit was more relaxed: 1.5% until 2010 and 1.0% until 2015. However, the IMO also regulates sulfur emissions in international waters, setting the sulfur content limits at 4.5% until 2012, 3.5% up until 2020 and 0.5% since then.[18]

Model studies using the new sulfur limit to investigate the indirect effects of aerosol–cloud interactions,[12,47] have shown a potential increase in radiative forcing of $0.2 \pm 0.11$ $Wm^{-2}$ and there have also been observable effects in the amount of observed ship tracks.[25,45,46,48] This has led to some debate about the possibility of using artificially injected sulfur as a geo-engineering solution to global warming. Measurements of the true amount of sulfur now being emitted from shipping (rather than assuming the limit is being adhered to) are important to properly assess the effect on the sulfur reduction on radiative forcing.

Because of their location, often a long distance from land, emissions from ships are difficult to study. However, a number of studies quantify the FSC of ship emissions. In general, the amount of emitted sulfur is compared to the amount of emitted carbon, with the assumption that carbon constitutes 87% $\pm$ 1.5%[19,20] of typical ship fuel. An aircraft was deployed to measure particulate emissions from ships in the English Channel and Bay of Biscay as early as 2004. The study was preceded by a ground test with an engine rig. The authors did not attempt to quantify FSC, perhaps since it was already known to be 2.4% in the targeted ship.[21] Lack *et al.*[22,23] used NOAA's WP-3D aircraft to target a Maersk Line ship entering Californian waters as it changed fuel from 3.15% to 0.07% sulfur content. The intercepted plume was approximately 2–5 minutes old. They used $CO_2$ to derive emission factors. Kattner *et al.*[20] performed ground measurements in Hamburg harbour and analysed over 1400 ship plumes of 2–10 min age. They found that almost 100% of ships met the sulfur fuel content limit of 1% in 2014 and the much stricter 0.1% limit in 2015 and showed that up to 40% of ships entering the harbour could have their FSC quantified using this method. Beecken *et al.*, developed a technique to measure ship emissions of $SO_2$, $NO_x$, $CO_2$ and particles using small aircraft.[24] They measured 158 ships in the in the Baltic and North Sea (inside the SECA) and found that approximately 85% of measured ships complied with the sulfur fuel content limit at the time (1%). Yang *et al.*,[25] developed an Unmanned Aerial Vehicle (UAV)-based Microsensor Sniffing System (MSS) for real-time FSC monitoring. They showed that the system was effective in measuring FSC's for ships in the waters off Hong Kong, with 125 ships measured and mean FSC of 0.39%, well within the 0.5% regulatory limit. Mahajan *et al.*,[26] measured shipping emissions in Dunkirk, France using a multi-axis DOAS instrument (MAX-DOAS). Elevated aerosol extinction coefficients (AEC), nitrogen dioxide ($NO_2$) and $SO_2$ were observed up to 500 m from the surface in ship plumes. Whilst they could not calculate FSCs due to the lack of a $CO_2$ measurement, they did observe the $SO_2/NO_2$ ratio to be low throughout the campaign, confirming that the new fuel content regulations were being followed by most ships in the region. All these measurements are either in ports, inland waterways or special sulfur control zones and therefore are not representative

of emissions in the open ocean. These areas often have more stringent emission regulations than the open ocean, hence requiring ships to take additional methods to reduce emissions (*e.g.* exhaust scrubbers). Ships are also operating at a lower speed and engine load in these areas which is likely to have an effect on their emission ratios. In addition, ships spend most of their time, and hence produce most of their emissions, when in the open ocean. Having such open ocean emission measurements will be important for assessing the global effect of changes in sulfur emissions from shipping, especially with regard to the effect on radiative forcing.

Apart from being the main anthropogenic source of sulfur in the atmosphere, ships emit large volumes of Nitrogen Oxides ($NO_x = NO + NO_2$), with around 15% of global $NO_x$ emissions attributable to ships.[13] These emissions affect both local air quality and production of secondary pollutants such as ozone which can have an effect on the climate through changing oxidising capacity. It is important to understand $NO_x$ emissions from shipping, both close to land and in the open ocean, as shipping is likely to have a slower transition to non-combustion fuels than other transportation, making them an increasingly important contributor. Burgard and Bria[27] used a remote sensing system originally designed to measure land vehicles emissions to show that ships typically emit $72 \pm 24$ g $NO_x$ per kg of fuel burned, depending on engine specifications and in particular engine speed. Alternatively, $NO_x$ to $CO_2$ ratios can be used as a measure of emissions per unit of fuel enabling comparison with other vessels or vehicles.[28,29] Ratios for road vehicles are around 0.002–0.008 g $kg^{-1}$ for cars or 0.004–0.016 g $kg^{-1}$ for HGVs.[28] $NO_x$ emissions from shipping can be actively controlled either by optimising fuel combustion or by after-treatment of the exhausts *i.e.* catalytic removal.[30] $NO_x$ emissions are sensitive to ship speed and engine load and in some places, there are regulations for ship to steam slowly, which reduces emissions.

This paper presents what we believe are the first post-2020 IMO sulfur regulation measurements outside of a SECA zone since the change in IMO sulfur emission regulations in 2020, along with measurements within the European SECA and port-based measurements in Valencia, Spain and Tyne, UK. It describes the methodology for the aircraft and land-based measurements, including the calculation of apparent fuel sulfur content (aFSC) and $NO_x/CO_2$ emission ratios and compares these measurements to the output of a model that estimates the annual emissions of individual ships. There follows a discussion on the wider implications of the measurements, the global representativeness of the sampled ships and considerations for future work.

## 2 Methods

The "Atmospheric Composition and Radiative forcing changes due to UN International Ship Emissions regulations (ACRUISE)" project was funded by the UK Natural Environment Research Council to examine the composition of ship plumes pre and post the 2020 changes in the IMO regulations of sulfur emissions (referred further as IMO2020) and the effect the change has on cloud formation, radiative forcing and air quality. It involved measurements of ship plumes in and out of the North Atlantic SECA in 2019, 2021 and 2022 using a large research aircraft. The "Enabling the remote measurement of air pollution emissions in UK ports" project aimed to show that point measurements of ship plumes in ports could provide a useful measure of emissions using fixed, land-based measurements. It involved measurements at the port of Valencia in 2022 (outside a SECA) and Tyne, UK (inside the European SECA) in 2023. All flight and fixed locations, along with the extent of the SECA, are shown in Fig. 1.

### 2.1 Aircraft measurements of ship emissions

The U.K. Facility for Airborne Atmospheric Measurements (FAAM) Airborne Laboratory BAe-146 research aircraft[31,32] was

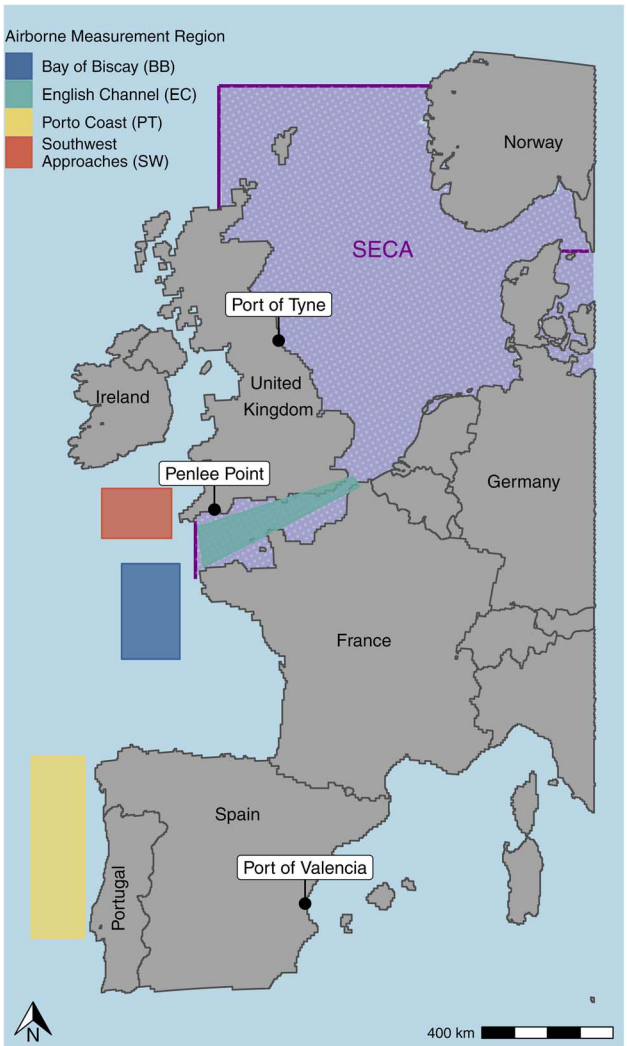


Fig. 1 Map of the study area showing regions where airborne measurements were conducted (Bay of Biscay, English Channel, Porto Coast, and Southwest Approaches) and point-sampling measurements at the Port of Tyne and the Port of Valencia. The location of the Penlee Point Atmospheric Observatory (PPAO) is also shown. The Sulfur Emissions Control Area (SECA) is represented by the purple shaded region.

used to sample ship plumes over the English Channel and North Atlantic Ocean as part of the ACRUISE project. The extensive range (1500 km) of the research aircraft makes it unique in its ability to sample ships in the open ocean outside of any SECA zone. There were 30 dedicated ACRUISE flights: 13 in the 2019 campaign (9 off the coast of Portugal, 2 to the SW of the English Channel and 2 inside the English Channel), 13 in the 2021 campaign (7 in the Bay of Biscay, 3 in the English Channel, 3 to the SW of the English Channel) and 4 in the 2022 campaign (2 to the SW of the English Channel, 2 in the Bay of Biscay). Flight numbers, dates and locations are given in SI1.

$SO_2$ was measured using a Thermo Scientific model 43i Trace Level Enhanced Pulsed Fluorescence $SO_2$ Analyser (TEi43i) during all three campaigns at 1 Hz (uncertainty greater of 5% or 4 ppb for ACRUISE-1, greater of 9% or 3 ppb for ACRUISE-2 and -3). $CO_2$ was measured using Fast Greenhouse Gas Analyser (FGGA, Los Gatos Research Inc.) model 907-0010 (ACRUISE-1, 1 Hz, precision ($1\sigma$) $\pm$0.391 ppm, accuracy $\pm$0.432 ppm) and model 907-0011 (ACRUISE-2 and ACRUISE-3, 10 Hz, precision ($1\sigma$) $\pm$0.599 ppm, accuracy $\pm$0.574 ppm). $NO_x$ was measured during ACRUISE-1 only using a custom built NO chemiluminescence two channel analyser measuring $NO_x$ by photolytic conversion at 385 nm and NO on a second channel.[33]

Ships were located pre and during the flight using the Automatic Identification System (AIS) data from **https://www.marinetraffic.com**. Once the ship was visually located from the flight deck, the plume direction was estimated and multiple passes perpendicular to the plume were carried out. Measurements of $NO_x$ or $CO_2$ were used to confirm that the plumes had been intercepted. On some occasions, after transecting the plume perpendicularly, the plume was followed from the ship as long as possible downwind to give an extended amount of time in the plume. The perpendicular transects are the main source of information used in this study. All the flight tracks from the three campaigns are shown in SI2. A total of 20 ships were identified and measured during ACRUISE-1 (2019), 95 during ACRUISE-2 (2021) and 15 during ACRUISE-3 (2022).

### 2.2 Ground based measurements of ship emission in ports

Ground-based point sampling measurements were conducted at two European ports. Both $NO_x$ and $CO_2$ were measured using an Iterative Cavity enhanced Differential optical absorption spectrometer (ICAD), developed by Airyx.[37] The instrument provides a direct measurement of $NO_2$, which is measured in the spectral range between ~430 and 465 nm. An internal converter based on gas phase titration with a $NO_x$-free $O_3$ source converts NO to $NO_2$ and allows for the measurement of total $NO_x$. Parallel $CO_2$ measurements are made using a smartGAS Non-Dispersive Infra-Red (NDIR) gas sensor (F3-212205-05000). The instrument has a response time of 2 s in the standard configuration. $SO_2$ was measured using an identical TEi43i to the airborne measurements. The instruments were housed inside a mobile laboratory (trailer for Valencia, van for Newcastle).[29,34]

The port of Valencia, Spain is situated to the SE of the city and is the fifth busiest seaport in Europe and the busiest in the Mediterranean. Measurements were made on a quay inside the port (39°26.5′N 0°19.1′W), where prevailing easterly winds would bring the plume from ships passing in and out of the port over the sample inlet while remaining unaffected by emissions from the wider port. A map of the sampling location is shown in SI3. Measurements were over 6 days (11th–16th October 2022) and after filtering the data to isolate plumes during easterly winds only, 58 plumes from 36 unique vessels were sampled. The port of Tyne, UK comprises the commercial docks on and around the River Tyne in Newcastle, UK. The majority of the docks for the larger ships, cruise liners and ferries are around 5 km up the river, therefore it was possible to position the mobile laboratory at various points along the river depending on the wind direction. A map of the sampling location is also shown in SI3. Measurements took place from 14th–18th May 2023, during which 26 plumes were sampled from 18 unique vessels, 4 of which were tugboats. All sampling was within the SECA therefore data is included in the SECA averages discussed below.

The Penlee Point Atmospheric Observatory (PPAO, 50° 19.08′ N, 4°11.35′ W)[35] receives UK input from the NE, Continental Europe emission/Channel shipping from SE, and Atlantic air from SW. Predominant wind directions are SW and NE. This allows quantification of the impact of anthropogenic activity, such as emissions from ships, on coastal environments as well as the influence of the sea on nearby land. $SO_2$ was measured using a TEi43i analyser, with a data series stretching back to 2015.

### 2.3 Calculation of apparent fuel sulfur content and emission ratios

With the assumption that fuel contains 87 $\pm$ 1.5% carbon[19] and 100% of the sulfur and the carbon content of the fuel are emitted as $SO_2$ and $CO_2$ respectively, the apparent fuel sulfur content (aFSC) mass percent can be calculated using eqn (1).[20] We use the term apparent fuel sulfur content as it does not take into account where the ship is running on high sulfur fuel but using an exhaust scrubber to meet regulations (an accepted means to meet the sulfur fuel limit).

$$\mathrm{aFSC}[\%] = \frac{\mathrm{SO_2[ppb]}}{\mathrm{CO_2[ppm]}} \times 0.232[\%] \quad (1)$$

Yu *et al.*[36] show that directly emitted sulfate aerosol ($SO_4$) increases with increasing FSC and the conversion to $SO_4$ also increases with plumes ageing. Since a precise position of the ship at the time of interception of its plume is not known, we cannot calculate the age of the plume. For the flight data, Yu *et al.* estimate the age of most of the plumes are a maximum of 15 minutes old and the sulfate percentage for this age of plume is 6% of the ship emitted sulfur. Grigoriadis *et al.* show that the percentage of fuel sulfur that gets converted to particulate $SO_4$ is a function of engine load, with the overall contribution of $SO_4$ to total $SO_x$ ($=SO_4 + SO_2$) is estimated to be between 2 and 5%.[37] In addition, according to Corbett *et al.* 95% of the overall sulfur

emission is in the form of $SO_2$.[14] There is a potential low bias to our aFSC estimate due to the presence of sulfate aerosol, but we do not correct our data as we do not have engine load information at the time of measurement. $NO_x/CO_2$ ratios are taken as the numerical ratio of the enhancement above background of the mixing ratios of $NO_x$ and $CO_2$.

For the airborne measurements, to account for the differences between instrument response time, peak areas rather than peak heights were used. Emission ratios were derived in a five-step process: background identification, plume identification, background refitting, peak integration and peak matching with QA/QC. To define plumes the concentration time series is assumed to consist of background, characterised by normal distribution and constant variance, and irregular peaks distinguished by different variance and higher mean. A Generalised Additive Model (GAM) with an adjustable smoothing parameter $k$ was fitted to the entire time series. Next, two flight specific thresholds based on standard deviation were chosen to identify the suitable size peaks and separate the background. Once the peaks were identified and extended to the background threshold, the baseline was refitted using only the data classified as background. The refitted baseline was then subtracted from the data (with the remainder signified using $\Delta$). The area under each peak was then integrated using trapezoidal approximation (see Barker *et al.*[38]). Measurement uncertainties are also propagated. Additionally, a 6% positive uncertainty is added to each aFSC to account for maximal sulfate conversion rate.[36] During the ACRUISE-3 flights, a custom-built laser-induced fluorescence (LIF) instrument was also deployed to measure $SO_2$. It has been demonstrated that the LIF is a more sensitive and faster technique compared to the commercial pulsed fluorescence analyser, with a $3\sigma$ detection limit of 0.07 ppb (at 10 seconds) and a 3 e-folding response time of 2 seconds, compared to 0.40 ppb and 17 seconds. Despite these differences in instrument performance, the aFSCs calculated for these two instruments using the integration analysis method have been shown to agree within the combined measurement uncertainties.[50] Uncertainties for $NO_x/CO_2$ ratios were also calculated and propagated, with a typical mean relative error of 11%, largely driven by uncertainty in the $CO_2$ enhancement.

For the ground-based point sampling plume measurements, enhancement ratios of $NO_x/CO_2$ and $SO_2/CO_2$ were calculated using ordinary least squares (OLS) regression. Plumes for each ship were isolated within the time series and background mixing ratios were calculated as the 1st percentile measurement in a rolling 5 min, centred window.[29] This approach produces a background estimate for every measurement point in the time series, which can then be subtracted to leave the remaining enhancement due to the ship plume. Prior to fitting the models, the time series of each variable ($\Delta NO_x$, $\Delta SO_2$ and $\Delta CO_2$) was aligned using cross correlation to account for differing response times of the instruments. For each plume, the time series was shifted to determine the offset which gave the strongest correlation with $CO_2$. The optimum offsets for $NO_x$ and $SO_2$ were then applied to the data for each plume. aFSCs were then calculated using eqn (1). Examples of typical plumes for the aerial and point sampling datasets are shown in Fig. 2. Standard errors of the model fit were calculated as the average vertical distance (error) between the data points and the regression line.

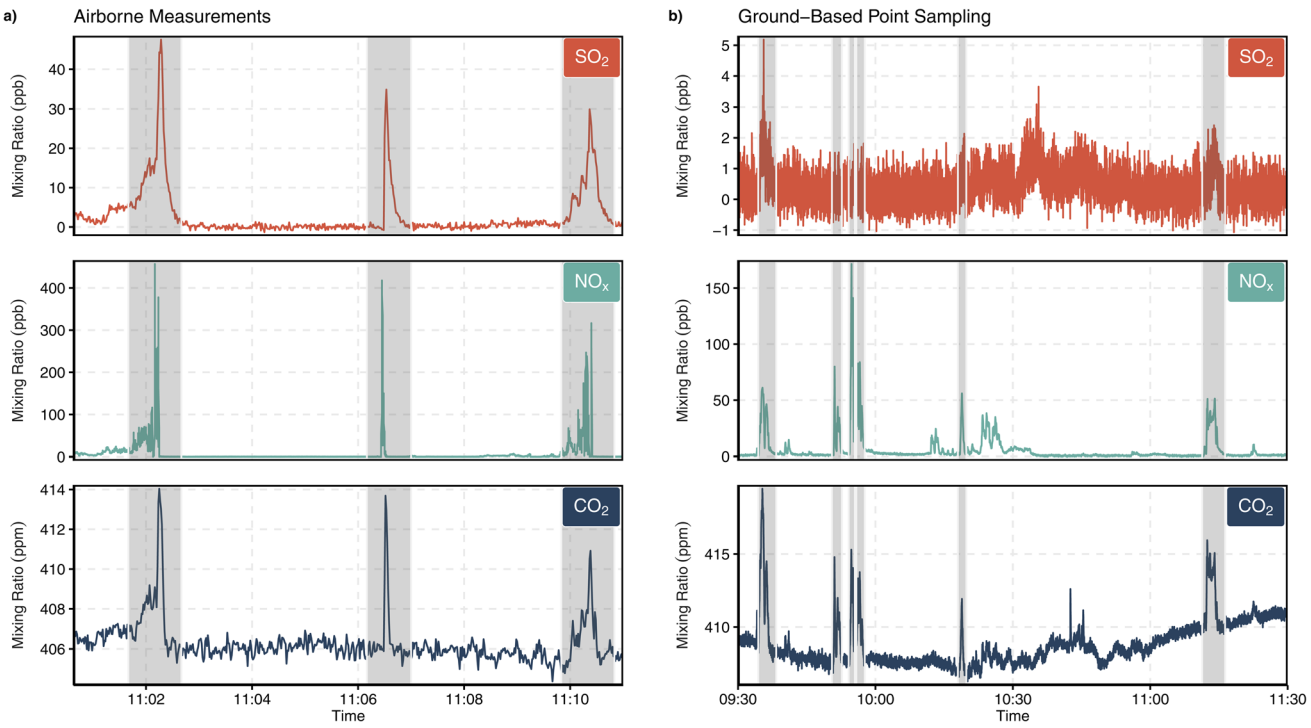


Fig. 2 (a and b) Time series data showing co-emitted plumes (grey shaded regions) of $SO_2$, $NO_x$, and $CO_2$ observed downwind of passing ships during an airborne survey on July 12, 2019 (left), and stationary point sampling measurements taken near the Port of Tyne on May 15, 2023 (right).

We investigated the potential interference on the TEi43i UV pulsed fluorescence $SO_2$ analysers and found it to be 145 $SO_2$ ppbv per NO ppbv. Based on the $NO_x$ measurements made in the 2019 flying campaign and during the ground-based measurements, this would lead to a potential interference of between 0.7 and 9.1% on the $SO_2$ measurement. We do not have measurements for the 2021 or 2022 flying campaigns but we estimate that there would be up to a 15% interference during these campaigns. Therefore, we consider that there is a potential positive bias on the measured aFSC of 15%.

### 2.4 STEAM model

The Ship Traffic Emission Assessment Model (currently third version - STEAM3) developed by the Finnish Meteorological Institute uses Automatic Identification System (AIS) data to evaluate exhaust emissions of individual ships. The model estimates emissions of gaseous species such as $SO_2$, $NO_x$ or $CO_2$, but also aerosols and particulate matter. The input for the model, apart from AIS data (*e.g.* speed, heading, load), includes ship parameters (especially engine specifications, such as model, fuel type, rpm) and meteorological data (to estimate impact of waves). Engine-specific fuel consumption is especially important in determining $SO_2$ and $CO_2$ emissions.[39] The model setup used in this study provides an estimate of the annual emission of $SO_2$, $CO_2$ and $NO_x$. The values obtained from the model are expressed in kilograms of each compound. Eqn (2) is a modification of eqn (1) and was used to convert mass ratios to mixing ratios to calculate average yearly FSCs.[20]

$$\mathrm{FSC}[\%] = \frac{\mathrm{SO_2[kg]}}{\mathrm{CO_2[kg]}} \times 1.60[\%] \quad (2)$$

For $NO_x/CO_2$ (ppm/ppm) ratios an assumption was made that total $NO_x$ can be treated as $NO_2$ to convert mass ratio to mixing ratio. Modelled mixing ratios were calculated as follows eqn (3).

$$\frac{\mathrm{NO_x[ppm]}}{\mathrm{CO_2[ppm]}} = \frac{\mathrm{NO_x[kg]}}{\mathrm{CO_2[kg]}} \times 1.05 \quad (3)$$

where the factor of 1.05 is the ratio of the molecular mass of $NO_2$ to $CO_2$.

## 3 Results

### 3.1 Apparent fuel sulfur content

In all years and measurement sites the aFSC from most ships remain within the IMO limits (Fig. 3). A clear reduction in mean open ocean aFSC from 3.03 ± 0.53% in 2019 to 0.31 ± 0.05% in 2021 (nearly 10-fold decrease) and 0.25 ± 0.08% in 2022 demonstrates the strong impact the IMO2020 regulation change had on sulphur emissions. In 2019, 8 ships out of 19 had an aFSC higher than the 3.5% limit, including a 61 900 tonne container ship built in 2011 (aFSC = 9.8 ± 0.35%) and an 86 100 crude oil tanker built in 2000 (aFSC = 8.95 ± 0.31%). In 2021 and 2022, 4 ships out of 78 were observed to be breaching the 0.5% limit by more than the error in the observed aFSC and the potential 15% positive bias due to NO interference. There is no obvious reason why these particular ships breach the limit, either in their size, age or type of vessel. In the Port of Valencia

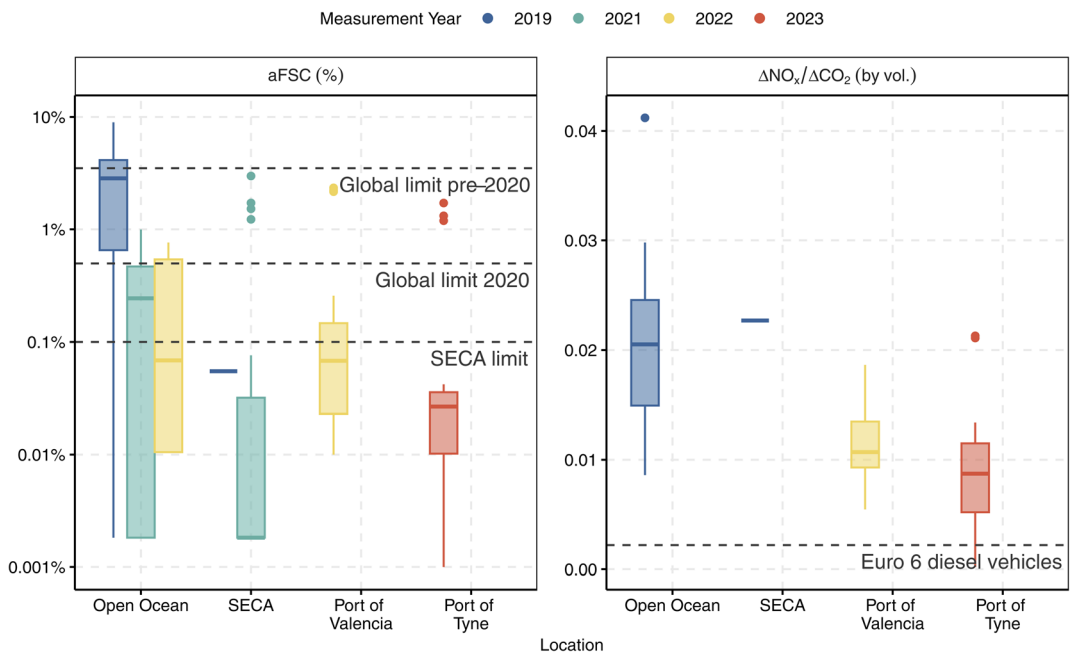


Fig. 3 Boxplots displaying the aFSC (left) and $\Delta NO_x/\Delta CO_2$ ratio (right) for individual vessels measured across different years and sampling locations. In cases where multiple flight passes of a plume from the same vessel were conducted on a single day, mean values were computed beforehand. Zero values, caused by minor enhancements near the detection limit of the instruments, were substituted with half of the minimum values for both aFSC and the $\Delta NO_x/\Delta CO_2$ ratio, respectively.

in 2022, observed aFSC was on average much lower than in the open ocean (0.10 ± 0.02%), which is close to the EU Sulfur directive (0.1% S fuel in port areas if the ship stays more than 2 hours in port). In SECAs, the airborne measurements suggest that ships generally adhere to the strict 0.1% limits (both before and after the 2020 regulatory change for the open ocean) with only 2 ships exceeding the limit (out of 33). The average aFSC inside the SECA across both 2019 and 2021 measurements was 0.04 ± 0.01%, slightly lower than values measured by Van Roy *et al.*, 2023 (0.068%).[51] An interesting example in 2019 was a large container ship (~200 000 tonne) that was sampled in both the Porto shipping lanes and in the English Channel SECA. A large change was measured in aFSC with 4.29 ± 0.36% in the open ocean (exceeding the regulatory limit) and 0.06 ± 0.02% in the SECA. We believe the aFSC reduction is due to a change in fuel because the ship is not listed as having a scrubber fitted. In the Port of Tyne (within the SECA), the observed aFSC is again considerably lower than those measured in the SECA in the English Channel (0.04 ± 0.01%), with no ships breaching the 0.1% limit. It should be noted that our plume measurements are a weighted average of emissions from main and auxiliary engines. Auxiliary engines are smaller than main engines and usually use lighter fuels than the main engines. Plume measurements that include auxiliary engine emissions will most likely bring the aFSC estimate down.

As shown in Fig. 4, there is no particular trend between aFSC and age of vessel. The open ocean 2021/2022 data show a higher average for ships more than 30 years old (mean of 0.33 ± 0.08% ($n = 2$) compared to 0.2 ± 0.05% for younger ships). However, this average is from a sample size of two. In the SECA, ships over 30 years old showed a mean aFSC of 0.045 ± 0.011% ($n = 3$), compared to 0.035 ± 0.015% for ships younger than 30 years. Fig. 5 shows that, in the open ocean and SECA, there is no strong correlation between aFSC and gross tonnage of the vessel. For 2021/2022 measurements, the highest mean aFSC in the open ocean was for 50 000–100,000 tonne ships (0.38 ± 0.09%), which is slightly higher than the 100 000–200,000 tonne ships (0.21 ± 0.05%) and the 10 000–50 000 tonne ships (0.23 ± 0.04%). Interestingly, in 2019, larger ships showed considerably greater mean aFSC values (3.48 ± 0.47% for 100 001–200 000 tonne ships and 4.49 ± 1.31% for 50 000–100,000 tonne ships). Inside the SECA, the highest mean aFSC was for 100 000–200,000 tonne ships (0.07 ± 0.03%). There is no intrinsic ship feature that prevents the retrofitting of a scrubber or a switch to

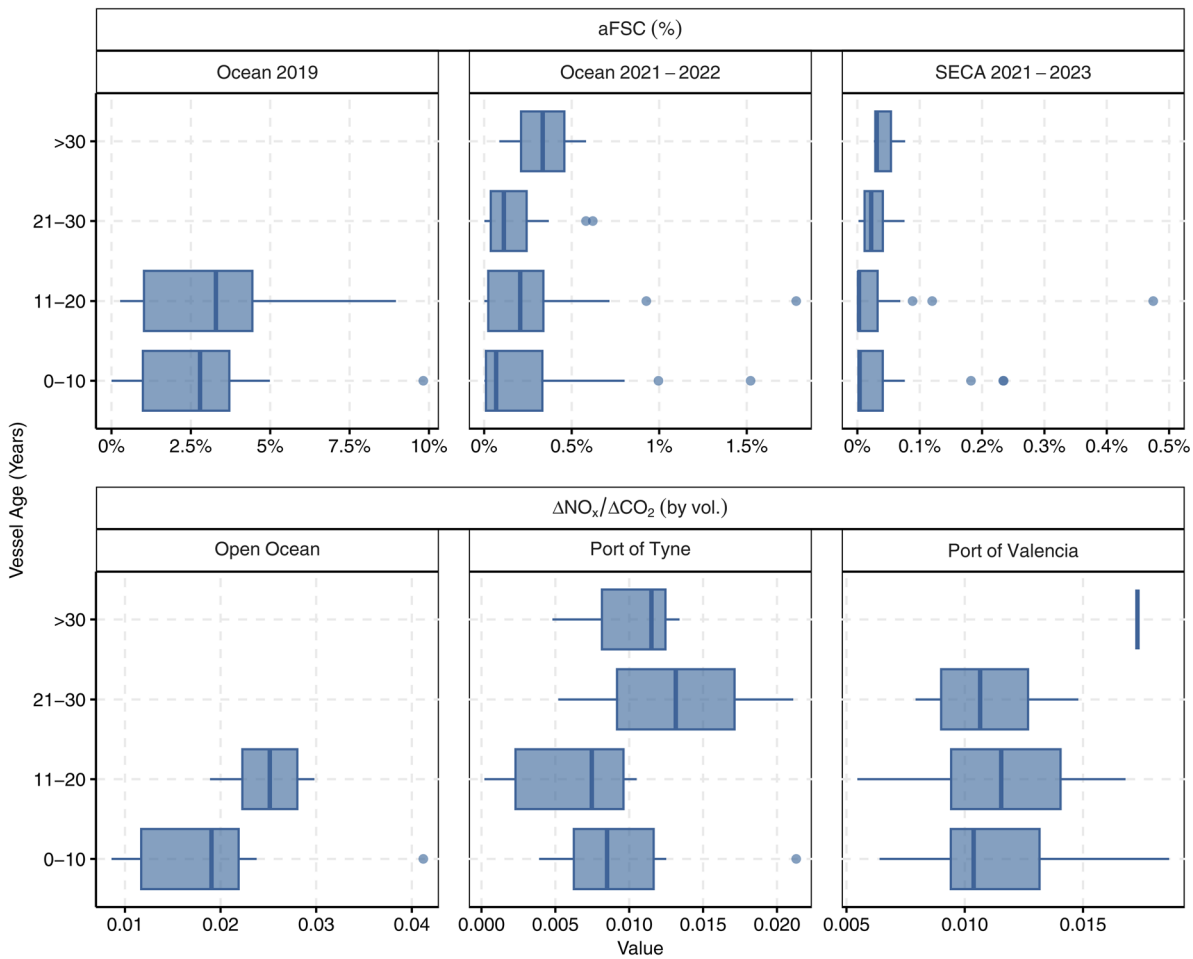


Fig. 4 Boxplots displaying the aFSC (top) and $\Delta NO_x/\Delta CO_2$ ratio (bottom) for individual vessels as a function of vessel age measured in each sampling location. In cases where multiple flight passes of a plume from the same vessel were conducted on a single day, mean values were computed beforehand.

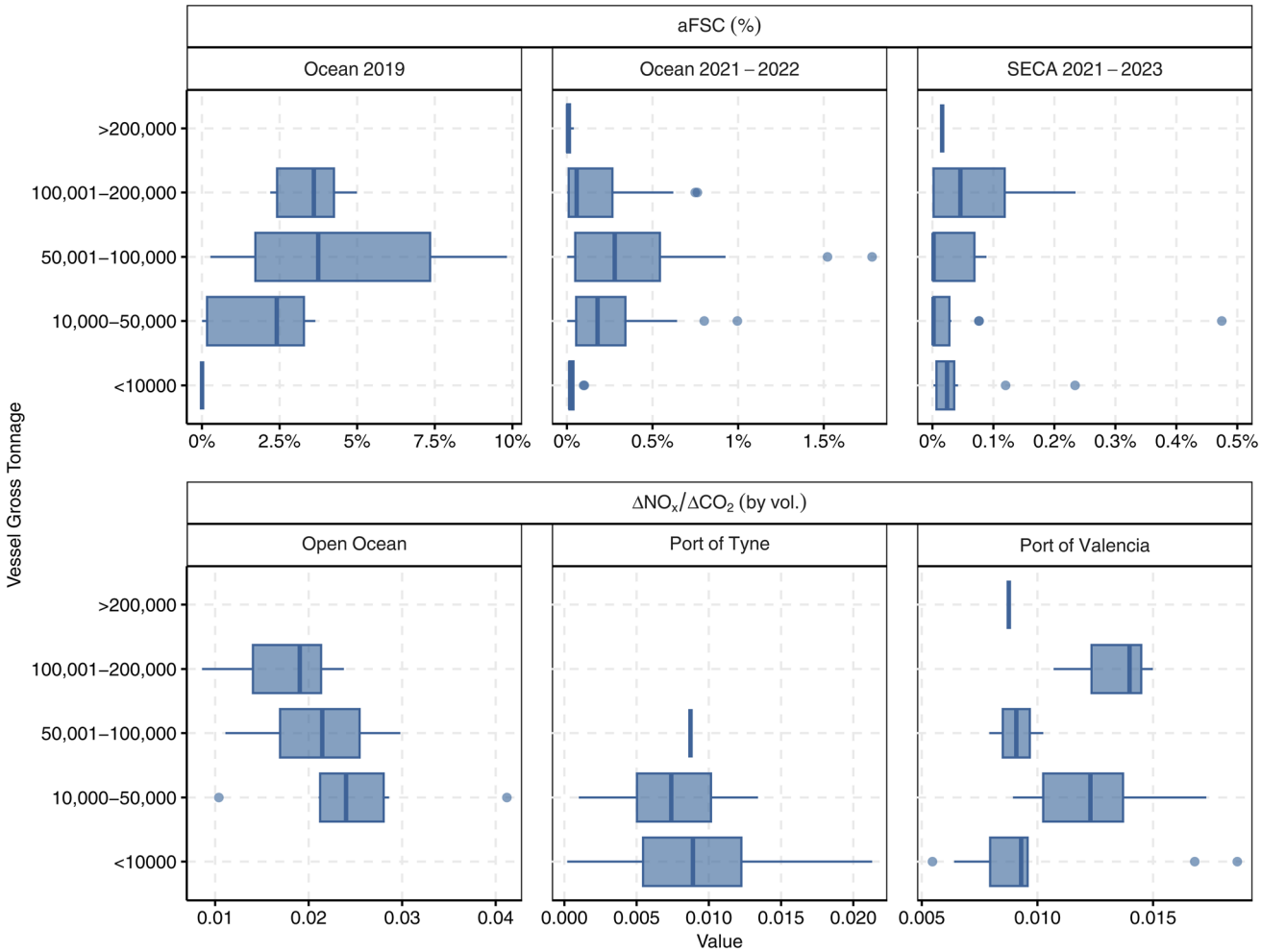


Fig. 5 Boxplots displaying the aFSC (top) and $\Delta NO_x/\Delta CO_2$ ratio (bottom) for individual vessels as a function of each vessels total internal volume (gross tonnage) measured in each sampling location. In cases where multiple flight passes of a plume from the same vessel were conducted on a single day, mean values were computed beforehand.

cleaner fuel, which is the likely reason for the lack of correlation between age or size of ship with aFSC.

The aFSC in this sample of ships seems to depend predominantly on the owner's or manager's decision to implement a suitable sulfur reduction solution. Ships opting for scrubbers and for lower sulfur fuel were sampled during the ACRUISE campaigns (Fig. 6). Although there were fewer ships with scrubbers than without, the percentage of ships with scrubbers increased after the IMO2020 regulation change (9% with scrubbers in 2019, 28% with scrubbers in 2021 and 2022). Fig. 6 shows clear differences in the aFSC between ships with and without a scrubber. In the open ocean in 2021–2022, ships equipped with a scrubber had a mean aFSC of 0.20 ± 0.07%, compared to 0.35 ± 0.05% for ships without scrubbers. However, a two-sample *t*-test showed that this difference was not statistically significant ($t(43) = -1.6$, $p = 0.112$), for ships measured in SECA, the mean aFSC for ships with a scrubber was 0.01 ± 0.01% compared to 0.06 ± 0.02% for those without ($t(24) = -2.1$, $p = 0.047$). As expected, in the open ocean in 2019, before the IMO 2020 regulation came into effect, there was a large, statistically significant difference in the aFSC for ships with and without a scrubber. The mean aFSC with a scrubber was 0.7 ± 0.65% compared to 3.61 ± 0.57% without ($t(11.3) = -3.36$, $p = 0.006$). Although the mean value for ships with a scrubber was influenced by a single outlier, the median aFSC was actually lower than that for ships with a scrubber in the open ocean in 2021–2022 (0.002%, compared to 0.07%) (Fig. 6). From this small sample size, it suggests that a scrubber cleans exhaust emissions to a level well below the regulatory limits, even though the sulfur content of the fuel itself may be significantly greater than the regulation allows. The mean aFSC for ships with scrubbers could still be overestimated, as it's only possible to determine whether a ship has a scrubber, not whether the scrubber is actually turned on. There is some evidence that if the ship is fitted with a scrubber the aerosol size distribution bifurcates to a small mode and a larger mode.[40,41] This is attributed to not all of the spray injected into the stack to remove the $SO_2$ being drained into the sea but a proportion being vented to the atmosphere having taken up significant $SO_2$ and converting it to $SO_4$. This may result in our measured aFSC

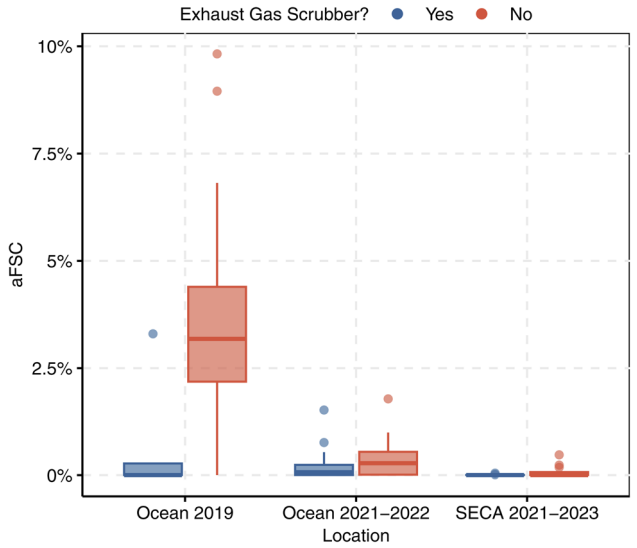


Fig. 6 Boxplots displaying the aFSC for individual vessels with and without exhaust gas cleaning systems (scrubbers) measured during the ACRUISE flight campaigns across different sampling locations and years. In cases where multiple flight passes of a plume from the same vessel were conducted on a single day, mean values were computed beforehand.

decreasing when the total S in the plume remains the same. However, concurrent measurements using an Aerosol Mass Spectrometer (see Yu *et al.*, 2020 for details[36]) indicate that the fraction of sulfur emitted as $SO_4$ in near-field plumes of ships without scrubbers ranged from 0 to ~11%. While this fraction was up to 70% for ships with scrubbers, the inclusion of $SO_4$ still resulted in FSC largely within the 0.5% regulatory limit. We only consider the aFSC calculated from the gas-phase $SO_2$ data in this work. Targeted analyses, including the emission factor of $SO_4$ in different cases, will be addressed in future studies.

### 3.2 *Trends in the* $\Delta NO_x/\Delta CO_2$ ratio in ship plumes

$\Delta NO_x/\Delta CO_2$ ratios (ppm ppm$^{-1}$) were quantified only during the first ACRUISE campaign in 2019 and during both the port measurement campaigns (Fig. 3). The open ocean $\Delta NO_x/\Delta CO_2$ ratio was 0.021 ± 0.002. The average values for identified ships ranged from 0.009 to 0.041. A 200 000 tonne container ship observed both in the open ocean and in SECA demonstrated a large change in measured aFSC (Section 3.1). However, the average $\Delta NO_x/\Delta CO_2$ ratio for this ship remained the same (0.020 ± 0.002 and 0.020 ± 0.006, respectively), suggesting that the steps taken to reduce sulfur content had little or no effect on the $NO_x$ emissions. It is likely that the aFSC changes was achieved using a change in fuel type. Ratios observed in the port measurements were lower than for ships at sea. The average $\Delta NO_x/\Delta CO_2$ ratio for ships in the Port of Tyne was 0.009 ± 0.001 and 0.011 ± 0.001 in the Port of Valencia. Ships travel slower in ports or switch to auxiliary engines, a potential reason for the $NO_x$ emissions lower than the open ocean. Similar to aFSC, the $\Delta NO_x/\Delta CO_2$ ratio seems to be independent of ship age and gross tonnage (Fig. 4 and 5). Ships built after 2021 are required to have lower $NO_x$ emissions within certain areas (including the European SECA).[42] Unfortunately, no measurements of $NO_x$ were made on ships underway in the English channel in 2021 or 2022. Future work should assess $NO_x$ emissions in an emission control area.

## 4 Discussion

### 4.1 Representativeness of the measured shipping fleet composition

Across the 3 aircraft (ACRUISE) and 2 ground based campaigns emissions of 171 identified ships were successfully quantified. During ACRUISE-1 the identification rate was relatively low because retrospective attribution of plumes to specific ships based on Marine Traffic data proved to be a non-trivial task in a busy shipping lane. The strategy was improved during subsequent campaigns when ships were identified visually in-flight and targeted multiple times. During the ground-based measurements all sampled ships were identified.

According to the Review of Maritime Transport 2021,[43] the majority of ships sailing all over the world are bulk carriers (43%), oil tankers (29%) and container ships (13%). Currently oil tankers are the fastest growing fleet. During ACRUISE the dominant type were container ships (38%), followed by crude oil and chemical tankers (26% together), specifically targeted LNG tankers (9%) and bulk carriers (7%). For the ground measurements passenger ships were the most represented type (40%), followed by tugs (22%) and container ships (20%). The differences from the global statistics reflect both regional characteristics as well as measurement platform limitations in case of ACRUISE. The dominance of container ships comes from their large size and tall stacks that make them excellent targets to sample from a research aircraft. Meanwhile, fishing vessels were avoided due to the high numbers of sea birds following them. For the SAQN measurements in Valencia, 46% of the 36 vessels were passenger vessels (cruise ships or ferries), 23% were container ships, 23% were tugs and there were 1 each of oil tanker, chemical tanker and a fishing vessel. For the Port of Tyne measurements, 30% were passenger vessels, 30% were tugs, with single samples of a container ship, offshore supply vessel, military frigate and a vehicle carrier.

The current global average ship age is 22 years and it varies by vessel type.[43] For bulk carriers the average age is 11 years, for oil tankers 20 years and for container ships 13 years. Ships have been and continue to become bigger, with more mega-vessels being built. The average age is 12 years for all ships globally, 10 years for bulk carriers and container ships and 11 years for oil tankers. For our dataset, the average age is 11 (±8) years, considerably younger than the global average, which possibly relates to the relatively large size of ships sampled (2500 tonnes minimum, 57 300 tonnes median) and the region we sampled. In our sampled ships 30% of ships had sulfur scrubbers fitted in 2021 and 20% in 2022 whereas globally it is around 10%.

We have also looked at how the fleet of ships we measured compares to the global fleet in terms of their contribution to total sulfur emissions. Fig. 7 shows the percentage of sulfur emissions (as given in the STEAM model inventory) by gross

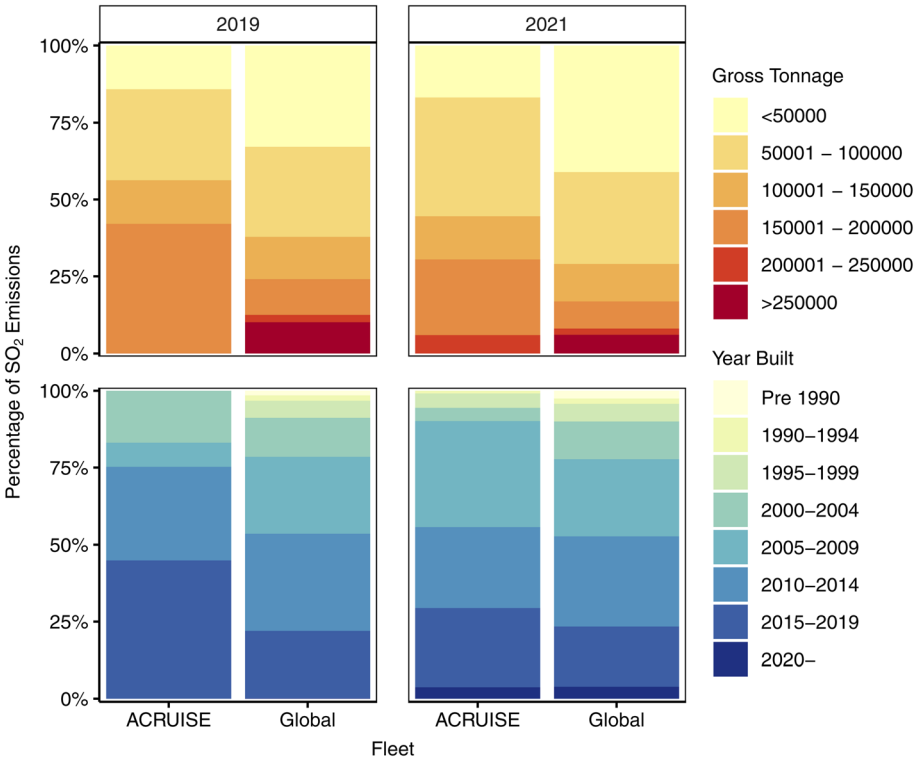


Fig. 7 Comparison of the percentage contribution to total $SO_s$ emissions between the ACRUISE dataset and the global fleet, categorised by gross tonnage (top) and vessel construction year (bottom) in 2019 and 2021.

tonnage and vessel age for both the ACRUISE sampled data and the global fleet (for both 2019 and 2021). The main difference in terms of tonnage, is that the ACRUISE data seems to undersample both the smaller and very large ships. In 2019 the measured ships showed that 13% of the sulfur emissions come from ships <50 000 tonnes, whereas in the global fleet this is 32%. In 2021 it is 15% from ships <50 000 for the measured ships and 40% for the global fleet. This is likely due to it being easier for the aircraft to spot and measure the larger ships. Also, 45% in 2019 and 30% in 2021 of sulfur emissions in the ACRUISE sample were from ships between 150 000 and 250 000 tonnes. For the global fleet this number is 11% in 2019 and 9% in 2021, with a further 10% (2019) and 8% (2021) coming from ships >250 000 tonnes, which were not sampled at all in this study. This is likely because the very heavy oil tankers and bulk carriers (which make up the majority of ships > 200 000 tonnes) do not typically sail up the coast of Europe and in the English Channel. In terms of vessel age, the main difference between the measured and global fleet is that the measurements typically do not include as many older vessels. For example, in 2019, 25% of the sulfur emissions in the measured fleet come from vessels built before 2004 but for the global fleet this number is 48%. In 2021, the age profile of the sulfur emissions is very similar between the measured and STEAM inventory. Our results described in Section 3.1 indicate that there is not a string correlation between ship age or tonnage and hence we do not believe that the differences in the measured and global fleet are important when extrapolating our results globally.

### 4.2 Comparison of measured and modelled aFSC

We compared measured aFSCs with the output of the STEAM model for all ships measured during the 2019 and 2021 ACRUISE campaigns, with the comparison shown in Fig. 8 (individual ship names removed). The STEAM model assumes that every ship complies with the SECA rules, therefore we only compared ships in the Porto or Bay of Biscay (open ocean) areas.

In the 2019 data set the model places most of the ships at about 2.0–2.5% FSC with two notable exceptions: a 2014 built LNG tanker (0.03%) and a 2006 built ro–ro cargo ship (0.92%). Neither of these ships were fitted with a scrubber. In the case of the LNG tanker, modelled $SO_2$ emissions are lower than other ships but $CO_2$ emissions were typical for the fleet. The model assumes that the tanker uses LNG as fuel, however this seems either not the case or the ship was running additional generators. The cargo ship does not stand out in terms of absolute emissions of either $SO_2$ or $CO_2$. For the LNG tanker the measured value is more in line with the typical model output range for non-LNG ships (2.20%), while for the cargo ship the measured value ($0.65 \pm 0.25\%$) is within uncertainty of the modelled one. In 2021, the model value has all ships achieving the 0.5% sulfur limit, therefore the 13 ships that were observed breaching the limit are underestimated by the model. There were 2 cases where observed aFSC was greater than the modelled FSC by > 1%: a 61 700 tonne bulk carrier built in 2014 ($1.52 \pm 0.13\%$) and a 66 400 tonne container ship built in 2008 ($1.78 \pm 0.34\%$).

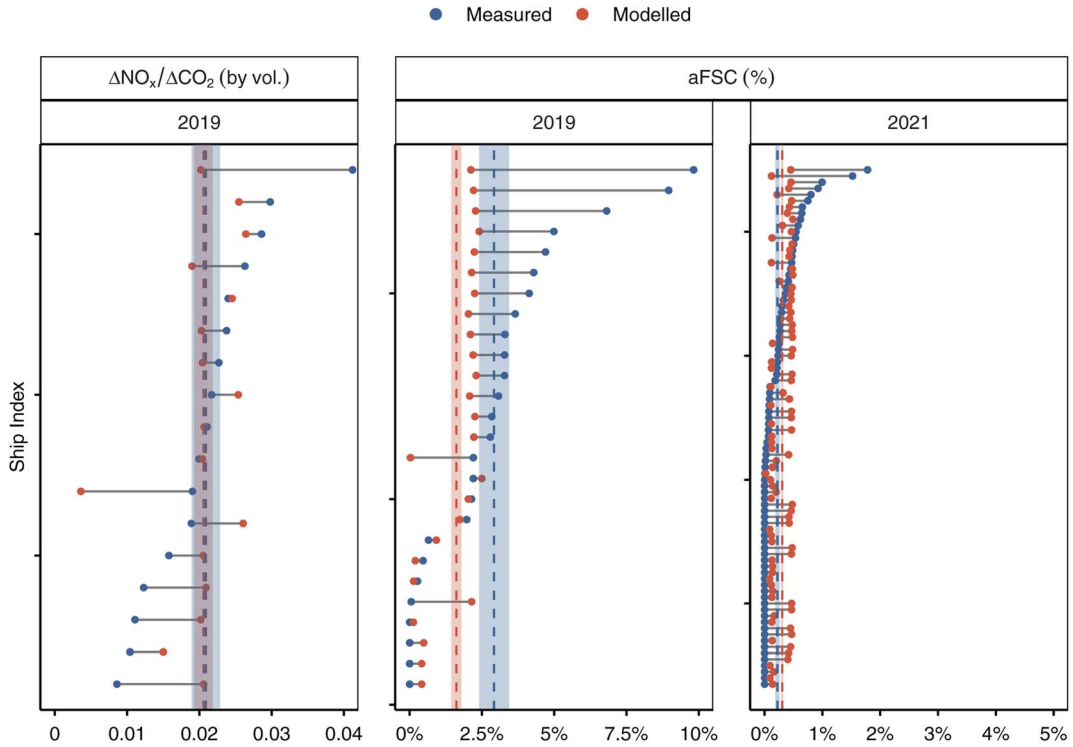


Fig. 8 Comparison of measured values of aFSC and the $\Delta NO_x/\Delta CO_2$ ratio from ships observed during the ACRUISE-1 and ACRUISE-2 campaigns and modelled values calculated using STEAM3. The *y*-axis represents a unique index assigned to each measured ship with different values for 2019 and 2021 ships. The dashed vertical lines indicate the mean measured and modelled values, where the shaded area shows the 95% confidence intervals.

Taking the fleet averages, the 2019 average modelled FSC for the sampled ships (2.00 ± 0.58%) is significantly lower than observed average aFSC (3.95 ± 2.33%). This is because 8 out of 20 ships were observed exceeding the 3.5% limit, which is not shown in the model values. For the 2021 ships, the average modelled FSC was 0.37 ± 0.15% which compared very well to the average observed aFSC for these ships 0.34 ± 0.21%. STEAM also reflects the observation that FSC is lower when a ship has a scrubber fitted. As described in Section 3.1, ships with a scrubber showed an average observed aFSC of 0.20 ± 0.0.07% whereas those without averaged 0.35 ± 0.05%. STEAM output suggests an average FSC of 0.18 ± 0.02% and 0.36 ± 0.13 for ships with and without a scrubber respectively. This analysis demonstrates that, at least for the small sample size of measured ships examined in this study, the assumptions in the model, on average, align well with the measured sulfur emissions, especially for measurements collected from 2021 onwards.

### 4.3 Comparison of ship emissions in ports *versus* open ocean

An interesting result to come from this study is the difference in aFSC between ships measured while manoeuvring in and out of port and those at sea. Fig. 3 shows that average observed aFSC for open ocean ships was 0.31 ± 0.05% and 0.25 ± 0.07% in 2021 and 2022 respectively, whereas for the port of Valencia in 2022 (not in a SECA, but under an EU directive stipulating a maximum FSC of 0.1% for ships staying >2 hours in port) it is 0.11%. For English Channel (within the European SECA) measurements in 2021, the average aFSC was 0.09 ± 0.04%, whereas in the Port of Tyne in 2023 it was 0.04 ± 0.01%. Reasons for this difference could be ships operating scrubbers in port where there is a greater likelihood of regulatory monitoring and/or a greater percentage of passenger vessels measured in ports. We also see a difference in the $\Delta NO_x/\Delta CO_2$ ratio for ships in the open ocean in 2019 (0.023 ± 0.004) compared to those moving in and out of the Port of Valencia in 2022 (0.011 ± 0.001). The reason for this difference is not immediately clear. $NO_x$ emission ratios have been shown to be higher when engines are at lower load,[37] which is likely when in port. It is possible that ships in port are also using auxiliary engines, potentially producing less $NO_x$ per unit fuel burnt.

The difference between aFSC and $\Delta NO_x/\Delta CO_2$ emission ratios in port and at sea is likely not important on a global scale as the vast majority of the emitted $SO_2$ and $NO_x$ will take place while the ship is at sea. However, it is important to consider the difference when looking at the effect of ship emissions on local air quality. Ship emissions make up the majority of $SO_2$ observed in and around ports and will likely also have a significant effect on $NO_x$. It is worth noting that the mean $\Delta NO_x/\Delta CO_2$ ratio measured in both the Ports of Valencia and Tyne (0.011 ± 0.001 and 0.008 ± 0.001 respectively) are significantly higher than the ratio seen from road vehicles, with studies showing that even the latest Euro 6 diesel vehicles have a $\Delta NO_x/\Delta CO_2$

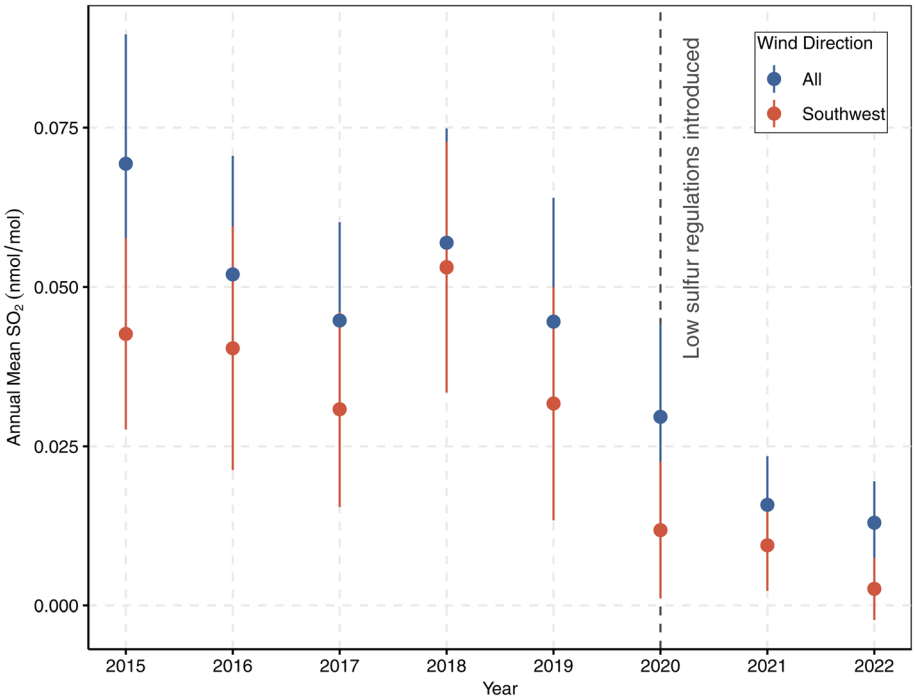


Fig. 9 Annual mean $SO_2$ mixing ratios at The Penlee Point Atmospheric Observatory (PPAO) between 2015 and 2022 shown for the entire data set (blue) and a filtered subset containing observations during south westerly winds. The error bars represent the 95% confidence intervals in the mean. The dashed line indicates the year when the International Maritime Organization (IMO) implemented new sulfur regulations, restricting sulfur content in ships' fuel oil to a maximum of 0.50%.

ratio between 0.0017–0.0026.[44] With the likely continued reduction of $NO_x$ emissions from road vehicles due to fleet electrification, shipping could become a very important $NO_x$ source in port and coastal urban areas in the future. Of course, if ships are able to switch to shore power while in port, their emissions would also be reduced effectively to 'zero'.

### 4.4 Wider effect of shipping emissions on ambient $SO_2$ concentrations

$SO_2$ measurements from the Penlee Point Atmospheric Observatory over the past 7 years can be used to assess the effect that changes in ship emissions have had on background $SO_2$ concentrations. Fig. 9 shows annual mean $SO_2$ concentrations from 2015 to 2022 for all wind directions and for when winds are from the south-west, bringing air from the shipping lanes in the English Channel and Bay of Biscay. Measurements at the site have previously been used to show the reduction in background $SO_2$ from the end of 2014 when the FSC regulation changed from 1% to 0.1% in the European SECA.[35] Whilst $SO_2$ levels remained reasonably constant from 2015 to 2019, there was another significant drop from $0.035 \pm 0.01$ nmol mol$^{-1}$ to $0.015 \pm 0.007$ nmol mol$^{-1}$ between 2019 and 2020, with further decreases in 2021 (to $0.012 \pm 0.04$ nmol mol$^{-1}$) and 2022 (to $0.007 \pm 0.001$ nmol mol$^{-1}$). The drop from 2019 to 2021 of 65% is lower than the 86% reduction observed in the aFSC measurements between the two years, which is likely due to the air at the observatory being influenced by ships both inside and outside of the SECA (where the regulations did not change in 2020) and by local emissions from small vessels unaffected by the regulations. $SO_2$ levels are also affected by natural $SO_2$ formed as a result of naturally-produced dimethylsulfide oxidation, so the observed change is reasonable.

## 5 Conclusions

This paper presents a series of measurements of sulfur emissions (in the form of aFSCs) and $NO_x$ ($\Delta NO_x/\Delta CO_2$ ratios) from ships in a variety of locations in Europe, both in and out of sulfur emission control areas. Measurements were taken before and after the change in IMO sulfur fuel content regulations from 3.5% to 0.5% in 2019 and observed a drop in mean measured aFSC in the open ocean from $3.03 \pm 0.52$% in 2019 to $0.31 \pm 0.05$% in 2021 and $0.25 \pm 0.07$% in 2022. Other studies have shown the knock-on effects of this reduction (*e.g.* in ship tracks), these are the first aircraft-based observations demonstrating the reduction outside of ports or coastal emission control areas. This study largely justifies most model studies that have tried to predict the change in global radiative forcing post regulation, which often just assume a 7 fold reduction in sulfur. It is worth noting that, in the open ocean, 5 ships out of 78 breached the 0.5% FSC limit in 2021 and 2022. These are quite a substantial proportion of the observed fleet, suggesting that whilst it is reasonable to assume the fleet average FSC is below the 0.5% limit, there could be a significant number of outlier ships breaching that limit, potentially having a large effect on local air quality out of SECAs. Our data also show that within the European SECA, FSC were even lower ($0.09 \pm 0.04$%

in the English Channel and 0.04 ± 0.01% for ships coming in and out of the Port of Tyne), with no ships breaching the 0.1% limit. The main variables that affect aFSC appear to be the choice of fuel/scrubber, rather than ship type, age or tonnage. The majority of ships have complied with regulation, which has led to a noticeable reduction in the background $SO_2$ burden in the northeast Atlantic. The limited sample size makes it difficult to draw any conclusions about what might cause a ship to breach the limit (*e.g.* age, tonnage) and more measurements are needed to investigate the cause of these outliers, which would contribute disproportionately to emissions. In addition, this study is limited to the Eastern North Atlantic Ocean, English Channel and two European ports. Measurements in other parts of the world, especially in the open ocean are required to see if this result is representative of the global industry.

## Author contributions

JL and DP designed the flights and the ground based measurements and wrote the manuscript. SW, WD and SL carried out the data analysis and produced the figures. HC helped design the flights and provided insight into the outcomes. MS, FS, HW, SB, SBaug and CR, LT and PE made the measurements. TB and MY designed the overall experiment, J-PJ did the model calculations and JB provided support for the ground-based measurements.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

FAAM data described in this analysis can be accessed at the Centre for Environmental Data Analysis **http://catalogue.ceda.ac.uk/uuid/affe775e8d8890a4556aec5bc4e0b45c**, CEDA. The code for downloading FAAM data, performing the analysis and producing the figures, along with data from the ground based measurements is archived on Zenodo at **https://doi.org/10.5281/zenodo.16682384**. The version of the code employed for this study is version v1.0.0.

Supplementary information: SI1 for FAAM flight numbers, dates and locations. SI2 for visualisations of flight tracks. SI3 for ground based sampling locations. SI4 for tabulated aFSC data. See DOI: **https://doi.org/10.1039/d5ea00089k**.

## Acknowledgements

Airborne data was obtained using the FAAM Airborne Laboratory BAe-146-301 Atmospheric Research Aircraft [ARA], operated by Airtask Ltd, managed by the National Centre for Atmospheric Science, leased through University of Leeds, and owned by UK Research and Innovation and the Natural Environment Research Council (NE/T014849/1). Thanks to the FAAM team for supporting the fights, including Maureen Smith, Doug Anderson and Alan Woolley. Thanks to the staff from Airtask Ltd for campaign organisation, flight planning and resolving any safety issues: Barney Black, Finbarre Brennan, Peter Chappell, Steve James, Mark Robinson and David Simpson. The authors gratefully acknowledge the financial support provided by the UK Natural Environment Research Council (grant numbers NE/S005390/1, NE/S004564/1 and NE/S004467/1 (ACRUISE), NE/N018044/1 (ACSIS), ST/S005366/1 (SAQN)) and the European Union H2020 NEMO project. DP and LT thank the NERC Spheres Doctoral Training Program for studentships. The Penlee Point Atmospheric Observatory (PPAO) and the work of TGB and MY were supported by NERC ACSIS and ACRUISE projects. JL, PE and MS thank the National Centre for Atmospheric Science (NCAS) for continued funding. JPJ thanks the support of the Strategic Research Council at the Academy of Finland: Marine waterways as a sustainable source of wellbeing, security, and safety (Decision number: #365647).

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