Measurement Report: The effects of SECA regulations on the atmospheric SO_2 concentrations in the Baltic Sea, based on long-term observations at the Finnish Utö Island.

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Abstract. The designation of the Baltic Sea as a Sulfur Emission Control Area (SECA) in May 2006, with subsequent tightening of regulations in 2010 and 2015 has reduced the sulfuric emission from shipping traffic. This study assesses the ~~, focusing on~~ impacts of SECA on observed SO_2 concentrations by~~,~~ providinge~~s~~ a long-term analysis of 1–minute time resolution air quality data from 2006 to 2020 at Utö island (Baltic Sea)~~,~~ supported by the predictions from the Ship Traffic Emission Assessment Model (STEAM). Additionally, 1~~-~~–hour resolution~~hourly~~ data from 2003 to 2005 is utilized to investigate changes due to the SECA limits set in 2006. The observed SO_2 concentrations at Utö have continuously decreased since 2003 due to an overall decrease in SO_2 emissions in Northern Europe, combined with reduced emissions from shipping traffic due to SECA regulations. Three–year average SO_2 concentrations dropped from pre–SECA (2003–2005) to post–SECA periods (2007–2009, 2011–2013, 2016–2018) by 38 %, 39 %, and 67 %, respectively. No clear trends were observed in the concentrations of other measured pollutants ~~measured~~. Furthermore,~~In addition to time series analysis,~~ we investigated wind direction resolved SO_2 concentrations for two selected years (2014 and 2019) ~~and studied the changes in ship plumes of one vessel regularly passing by Utö. ,~~ and the results showed~~ing~~ a significant decrease in high SO_2 ~~-~~concentration shipping plumes due to the implementation of SECA in 2015~~ implementation~~. Our~~This~~ study brings out the importance of long–term, high time~~-~~–resolution air quality observations at remote marine research station~~s, in the vicinity of a heavily trafficked ship lane,~~ in providing means~~possibility~~ for both quantitative and qualitative analyses of the impacts of regulatory environmental legislation.

1 Introduction

In recent years, international maritime trade has ~~seen~~ experienced sustained growth, primarily due to its recognized economic efficiency, accounting for over 90 % of global trade (IMO, 2016). This growth has been projected to continue, with ~~. Projections from~~ the United Nations Conference on Trade and Development (UNCTAD, 2019) indicatinge~~d~~ a prospective annual growth

rate of 2.6 % to 3.4 % until 2024. In 2019, 11.08 billion tonnes of goods were shipped, and the global commercial fleet, consisting of 98 140 vessels exceeding 100 gross tonnes, had a total capacity of 2.06 billion deadweight tonnes (UNCTAD, 2020). In the Baltic Sea, the shipping activity has increased fairly steadily during 2006–2020 (HELCOM, 2021). For example, in 2020 the total number of vessel fleet was increased by 271 %, 140 % and 46 % in comparison to 2006, 2010 and 2015, respectively.

However, maritime activities have led to significant environmental challenges. In particular,~~Johansson et al. (2017) reported that on a global scale,~~ shipping emissions are a major source of air pollution, contributinge annually to approximately 20.8~~8~~ × 10^6 tonnes NO_x, 9.7 × 10^6 tonnes SO_2 and 1.5 × 10^6 tonnes $PM_{2.5}$ globally (Johansson et al., 2017). ~~, whereas~~ Smith et al. (2015) evaluated that global shipping is responsible for approximately 13 % and 12 % of total global anthropogenic emissions of NO_x and SO_x, respectively. On European scale, the European Environment Agency (EEA, 2017) evaluated that 90 % of SO_2 emissions from transport were attributed to maritime transport, while $PM_{2.5}$, PM_{10} and NO_x transport-associated emissions accounted for 45 %, 28 %, and 35 %, respectively.

In ~~some locations within~~ the Baltic Sea region, ~~the~~ emissions of SO_2, NO_x, and NO_2 from international shipping were estimated to account for as much as 80 % of the overall concentrations of these pollutants in the near-surface environment (Haglund et al., 2016)~~,. Jonson et al. (2015) reported that shipping emissions in the Baltic Sea could~~ deteriorat~~ing~~e the air quality in coastal areas (Jonson et al., 2015). Apart from their air quality impacts, t~~T~~hese emissions had also the potential to cause acidification and eutrophication of marine waters and surrounding terrestrial ecosystems, ~~significantly impacting the~~ with serious implications in the Baltic Sea environment (HELCOM, 2009; Hunter et al., 2011; Raudsepp et al., 2013).

Notably, t~~T~~he impacts of shipping emissions are not limited to sea areas. ~~In fact, a~~Around 70 % of ship emissions occur within 400 km of land (Corbett et al., 1999), while more than one-third of the world's population lives within 100 km from the coast (UNEP, 2024). Pollution from shipping can reach the inhabited land areas and have a severe impact on human health. Shipping emissions have been shown to negatively impact the air quality, especially on coastal regions and portside (Chen et al., 2019; Donateo et al., 2014; Liu et al., 2017), the climate (Contini et al., 2015; Merico et al., 2016; Sofiev et al., 2018) and the economy (Jalkanen et al., 2014). Andersson et al. (2009) and Brandt et al. (2013) evaluated that international shipping was responsible for approximately 50 000 premature deaths annually in Europe. Furthermore, Barregard et al. (2019) ~~showed~~ reported that shipping in the Baltic Sea and the North Sea alone may have resulted in approximately 14 000 premature deaths in Europe in 2011.

While sulfur reductions have ~~undoubtedly~~ improved air quality and subsequently human health (Jonson et al. 2015; Sofiev et al., 2018; Barregard et al., 2019)~~-~~, it should be kept in mind that reducing sulfuric emissions from shipping may lead to~~also~~

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~~have~~ unintended adverse impacts on climate and marine ecosystems. The impact on climate, due to decrease in the cooling effect of ship-emitted aerosols, is likely leading to an increase in global sea surface and atmospheric temperatures (Lauer et al., 2009; Partanen et al., 2013; Hausfather and Forster, 2023). The impacts of sulfur reduction on marine ecosystems includes concentrated aquatic phase pollutant emissions from the exhaust gas cleaning systems as discussed in Picone et al. (2023) and Hermansson et al. (2024). Thus, the potential success of sulfur reduction regulations discussed in this study may require further investigations in a wider context not focusing only on direct human health benefits.

As international maritime trade and shipping emissions are increasing, measures to limit and monitor their adverse impacts are necessary. To mitigate shipping emissions, the International Maritime Organization (IMO) has implemented a series of regulations aimed at reducing emissions from ships. These regulations include the establishment of Sulfur Emission Control Areas (SECAs), which require ships to use low -sulfur fuel in designated areas, or, alternatively, reduce the sulfuric emission to air with exhaust gas cleaning system. In May 2006, the Baltic Sea was designated as a SECA region along with the North Sea and the English Channel. SECA regions have also been set up along the North American East and West Coast and the U.S Caribbean. The maximum allowed sulfur content of fuel in the SECA areas was decreased in 2006 from 2.7 to 1.5 %. Since 1 of July 2010, the International Maritime Organization (IMO) implemented a rule that required ships sailing in SECA regions to use fuel with a sulfur content no higher than 1.0 % (Van Aardenne et al., 2013). This rule was further tightened in 2015, reducing the sulfur limit to 0.1 %.

Given these regulations, it is essential to assess their effectiveness through compliance studies across various regions. For instance, Beecken et al. (2015) conducted two campaigns in the Gulf of Finland and Neva Bay during the summers of 2011 and 2012, sampling 466 plumes from 311 individual vessels using both ground–based and helicopter–borne measurements. Their analysis revealed that 90 % of the observed plumes complied with the 1 % sulfur content limit set for SECAs in 2011, with this compliance increasing to 97 % in 2012. Similarly, Yang et al. (2016) observed a significant decrease in ship–emitted SO_2 following the implementation of SECA regulations in January 2015 in the English Channel, underscoring the substantial impact of these policies on air quality. Mellqvist et al. (2017) reported compliance levels of 92–94% around Denmark during 2015–2016, and Jonson et al. (2019) revealed strong compliance with SECA regulations in the Baltic Sea based on emission modelling. Additionally, Repka et al. (2019) conducted a comprehensive study and found compliance rates exceeding 98 % in Göteborg and Gdynia, 95–97 % in St. Petersburg, and 94% in the central Baltic Sea. However, compliance rates were slightly lower at SECA borders, around 87 %.

~~However, while the SECA regulations aim at improving human health, it should be kept in mind that reducing sulfuric emissions from shipping may also have significant adverse impacts on climate and marine ecosystems. The impact on climate, due to decrease in the cooling effect of ship-emitted aerosols (Partanen et al., 2013) is likely leading to an increase in global~~

~~sea surface temperatures (Hausfather and Forster, 2023). For instance, Lauer et al. (2009) estimated that limiting fuel sulfur content (FSC) of ships globally to 0.5 % would decrease the negative radiative forcing (less cooling) of shipping emissions from -0.6 W m^{-2} to -0.3 W m^{-2}, potentially accelerating global warming. In addition, Hausfather and Forster (2023) argued that transition to low-sulfur marine fuels has intensified global warming, as sulfur particles in ship emissions have been mitigating greenhouse gas-induced warming. Furthermore, the authors claimed that estimates by Carbon Brief indicated that the 2020 regulations could contribute to a temperature increase of approximately 0.05 °C by 2050, equivalent to two additional years of emissions. The impacts of SECA on marine ecosystems include concentrated aquatic phase pollutant emissions from the exhaust gas cleaning systems as discussed in Picone et al. (2023) and Hermansson et al. (2024).~~

~~In this study,~~In this study, the impacts of SECA regulations on SO_2 concentrations in a small island in the Baltic Sea are studied. More specifically, this research will present unique fine temporal resolution data of SO_2, NO_x, NO, $PM_{2.5}$ and O_3 concentrations measured ~~on~~ at Utö island at the outer edge of the Archipelago Sea in the Baltic Sea, providing further insights into the impacts and the effectiveness of SECA regulations. The observations were carried out during the period 2006~~3~~-2020, covering the period with three separate changes in SECA limits. Additionally, sparser hourly data from 2003 to 2005 has also been utilized. ~~The focus of the study is on the impacts of SECA regulations on SO_2 concentrations.~~

Moreover, t~~T~~he observations of air quality parameters ~~Utö~~ are ~~supported~~ supported with local meteorological data and emission estimates utilizing AIS data. To further contextualize these measurements, we also computed the emissions attributed to shipping in the Baltic Sea during the target period using the STEAM model.

It is also noteworthy that the dataset used in this study, which includes wind direction analysis of SO_2 concentrations relative to a major shipping lane, has not been previously published or analyzed. ~~A qualitative case study is also presented on the changes due to the SECA regulations based on an analysis of pollution plumes originating from an individual Ro-Ro cargo ship.~~

2 Measurement location and site characteristics

The observations utilized in this study have been carried out ~~in~~ at Utö (59º 46’ 50 N, 21º 22’ 23 E), a small island in the Baltic Sea (Fig. 1a) with an area less than 1 km^2. It is located 70 km from the coast of the mainland of Finland and surrounded only by the open sea and a few smaller islands. The vegetation between the bare rocky areas consists mainly of different shrubs and a few trees. The island is flat, the highest point is less than 15 m above the sea surface. Less than 40 inhabitants live ~~in~~ at Utö during the winter. Population, including tourists during the summer months, varies between 100 and 200. Ships that pass Utö are pilot, passenger, military, fishing, cargo, commercial and tanker vessels. An important shipping lane to or from Finland is located at a distance of approximately 1 km west of the island (Fig. 1b).

Meteorological observations at Utö started in 1881. Since the beginning of observations, there has been a gradual increase in measured atmospheric, air quality, marine and electromagnetic parameters (Ahlnäs, ~~K.,~~ 1961; Laurila and Hakola, 1996; Komppula et al., 2007; Kyllönen et al., 2009; Kilkki et al., 2015; Laapas and Venäläinen, 2017; Laakso et al. 2018; Honkanen et al., 2018; 2021; Grönholm et al., 2021; Kraft et al., 2021; Seppälä et al., 2021; Rautiainen et al., 2023; Honkanen et al., 2024; ~~,~~ Hellén et al., 2024). As the island is sparsely populated and there is very little land traffic, there are no local sources of NO_x and SO_2. Consequently, Utö is an ideal location for plume tracking.

The long–term marine and meteorological observations and other characteristics of Utö Atmospheric and Marine Research Station are discussed in Laakso et al. (2018) and the references listed above, and thus only few results relevant for ~~shipping plume dispersion~~this study are summarized here. The prevailing meteorological conditions in the area are characterized by high wind speeds, with monthly averages ranging from 5.6 m s^{-1} in July to 8.9 m s^{-1} in December, resulting in an annual average of 7.1 m s^{-1}. The prevailing wind is from the southwest, while the sector from south (clockwise) to northwest dominates, accounting for approximately 60 % of the observed wind patterns (Honkanen et al., 2018). An examination of the wind time series from 1959 to 2016 (Laapas and Venäläinen, 2017) indicated no substantial alterations in wind direction or wind speed, aligning with a recent investigation covering the period of 1979 to 2008.

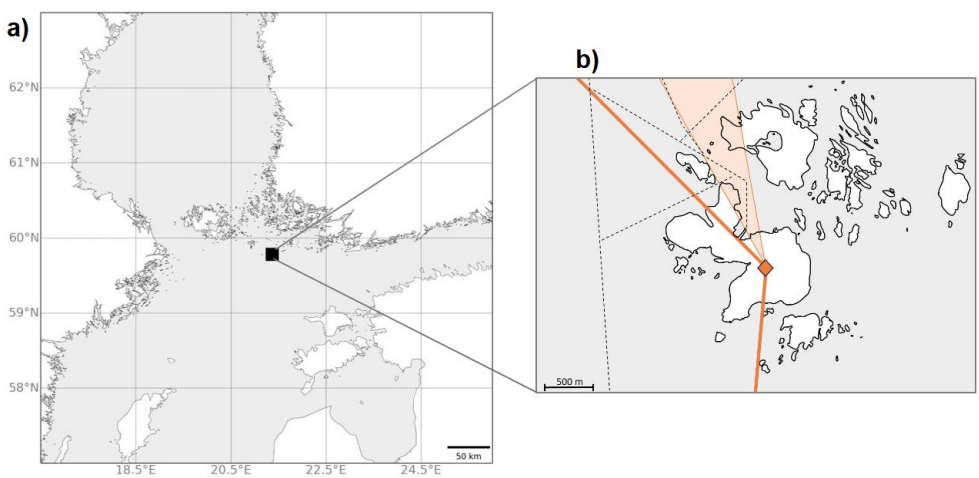


Figure 1: a) The location of the Utö island in the Baltic Sea and b) Utö air quality station (orange diamond) and the shipping lanes adjacent to the island (black dashed lines). Major ships operate on the north-south shipping lane at a distance of approximately 500 m west of Utö (or 1 km from air the quality station). The wind sector representing the directions from the main shipping lane (185°–

315°), is indicated by the orange lines.~~-~~ The ship traffic to and from the harbour of the island is presented by the shaded orange sector.

3 Measurements and methods

3.1 Air quality and wind observations

To study the effects of SECA regulations on air quality, ~~observations~~ concentrations of SO_2, $PM_{2.5}$, NO, NO_x and O_3 were ~~collected~~measured ~~were~~at 1–minute time resolution and analysed. The period of the study spanned 15 years, from 2006 to 2020 (supporting hourly data from 2003 to 2005). During this time, some of the instruments were changed (Table 1). The data has gone through normal quality control procedures as defined by EMEP and World Meteorological Organization (WMO) standards (see e.g. Anttila and Tuovinen, 2010 and references therein). In addition to the air quality observations, wind data on site was used. The temporal resolution of the wind data was 10 minutes and therefore it was interpolated linearly for intervals of 1 minute to match the air quality data.

Table 1. Air quality observations.

Observation	Device	Start	End	Notes
SO_2	Thermo 43i-TLE	09/09/2011		
	Thermo 43s	01/01/1996	08/09/2011	
PM_{25}	Fidas 200E	11/06/2021		
	5030 Sharp	22/11/2017		
	Thermo FH62 I-R	01/06/2003	22/11/2017	
NO_X	Thermo 42i-TL	07/02/2014		
	Horiba APNA-370	18/08/2007	07/02/2014	
	TEI 42CTL		18/08/2007	Data gap 2006-2007
O_3	Thermo 49i	12/02/2015		

Horiba Ltd APOA-360	03/10/2003	11/02/2015

Calibration of the instruments is carried out with precision to ensure accuracy and reliability. Initially, instruments are calibrated at the National Reference Laboratory for Air Quality at the Finnish Meteorological Institute (FMI). Following this, instruments are calibrated in the field~~filed~~ using reference values from the laboratory, involving zero level and three span points to confirm linearity.

SO_2 instrument is calibrated with the Gasmet Ansyco system, which utilizes a permeation chamber to produce SO_2 test gas and can also generate zero air. For PM2.5/10, the Fidas instrument is calibrated according to the manufacturer's procedure using a Monodust (reference particles) provided with the instrument to define the number of particles entering the device. Zero calibration is achieved with a HEPA filter, eliminating the need for reference values from the National Reference Laboratory.

NO_x is calibrated using the Sonimix 3012/3022 multigas calibrator, which includes a GPT, ozone, and a self-regenerating zero air generator. Calibration for NO specifically is conducted with externally supplied diluted NO in N_2. O_3 is calibrated with the 49i-PS, a UV photometric primary standard designed for the calibration of ozone analyzers. These procedures ensure that measurements are both accurate and reliable.

3.2 Automatic Identification System (AIS) data

The Automatic Identification System (AIS) is a mechanism for the automated generation and transmission of vessel-related information to both other vessels and coastal authorities (IMO, 2020). The AIS data encompassed main ship attributes, including Maritime Mobile Service Identity (MMSI), latitude, longitude, true heading, course over ground, and speed over ground. Based on the HELCOM AIS data, vessels were categorized into six distinct types that best characterized their specific roles: cargo ships, large passenger ships, medium-sized passenger vessels, large work vessels, small vessels, and others. In the vicinity of Utö, cargo and large passenger ships constituted the majority of vessels.

In our study, AIS data was utilized to calculate emissions using the STEAM model and to investigate potential changes in the shipping traffic density close to the Utö measurement site. ~~and to monitor ship movements of one Ro-Ro cargo in the vicinity of Utö. Data collection for the studied ship comprised AIS data sourced from the Helsinki Commission (HELCOM) for the period from 2013 to 2015 and from the AIS receiver at the Utö marine station for the period from 2016 to 2019.~~

3.3. The STEAM model

The STEAM model integrates AIS-derived data alongside technical insights into the global fleet and fundamental naval architecture principles (Jalkanen et al., 2009, 2012, 2014; Johansson et al., 2017). The model is subsequently used to predict vessel water resistance and instantaneous engine power of the main and auxiliary engines. The STEAM model facilitates the prediction of instantaneous fuel consumption and the emissions of specific pollutants (Jalkanen et al., 2016).

The input data for the STEAM model concerning ship properties, encompasses, among other factors, measured emission factors (when available), shaft generators, specific fuel oil consumption, and fuel type and sulfur content for main and auxiliary engines, along with installed abatement techniques (Jalkanen et al., 2009). The computed emissions for NO_x used in this study encompass the International Maritime Organization (IMO) registered traffic. The assessment includes all vessel traffic equipped with AIS transceivers while specifically excluding those navigating in inland waterways from the dataset. In this work, STEAM was used to run the Baltic Sea ship emission timeseries between years 2006 and 2020, including the regulatory changes to sulfur content of marine fuels. Further, STEAM was used to study the vessel traffic statistics near the Utö island, and determine hourly vessel counts. Further, STEAM was also used as a supporting tool in interpretation of air quality measurement results, to determine the emission intensity of local plumes

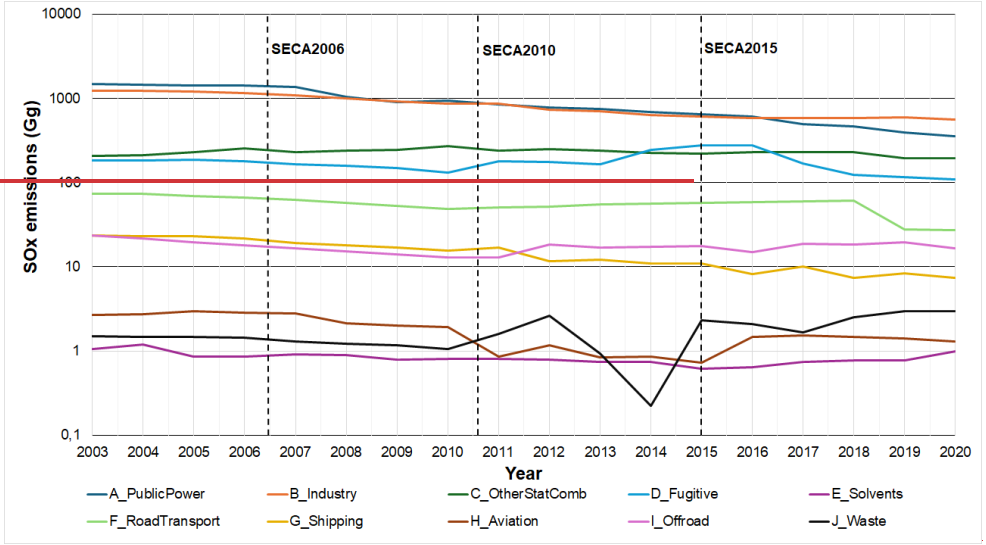
4 Results and discussion

The results are divided in subsections addressing the changes in long-range transport and shipping, investigating (i) the air quality time series and, (ii) the wind-direction dependence of observations. For convenience, we will use the following abbreviations throughout the text: SECA2006, SECA2010, and SECA2015 to refer to the SECA regulations established on 19 May 2006, 1 July 2010, and 1 January 2015, respectively. and (iii) a qualitative case study based on one regular Ro-Ro ship that has passed by the Utö Island.

4.1 Changes of long-range transport and shipping emissions

The SO_2 emissions in the Northen Europe have shown a decreasing trend in most of the emission sectors. Figure 2 illustrates the combined emissions in each emission sector from countries that have a major impact on the considered areaon the northern Baltic Proper / southern border of Archipelago Sea during the period 2003-2020. The dominant emissions originate from the energy (indicated as "public power" in Fig. 2) and industry sectors, exceeding the emissions from shipping by more than an order of magnitude. The typical average lifetime of SO_2 in the lower atmosphere is of the order of a fewapproximately 1–3 days (Leet al., 2011; Beirle et al., 2014). Part of the SO_2 observed at Utö is therefore originatesd from long-range transport of regional background pollution, while another portion is attributed to shipping traffic in the vicinity of Utö (e.g., Lee et al.,

2011). Long–range transported background pollution is well–diluted, whereas local emissions tend to manifest as short–term, high–concentration peaks.


SECA2006
SECA2010
SECA2015
SOx emissions (Gg)
10000
1000
100
10
1
0,1
2003
2004
2005
2006
2007
2008
2009
2010
2011
2012
2013
2014
2015
2016
2017
2018
2019
2020
Year
A_PublicPower
B_Industry
C_OtherStatComb
D_Fugitive
E_Solvents
F_RoadTransport
G_Shipping
H_Aviation
I_Offroad
J_Waste

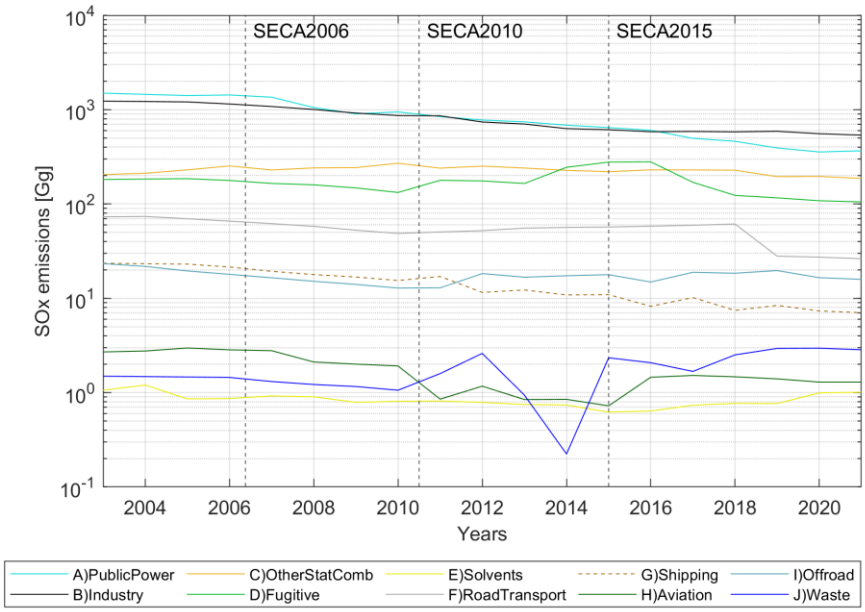


Figure 2: The combined annual SO_x emissions of Estonia, Finland, Lithuania, Latvia, Sweden, Russia and Poland divided in source sectors during the period 2003-2020. The implementation of subsequent SECA regulations is indicated with vertical dashed lines. Note that the figure is presented on a logarithmic scale (y--axis). The source of SO_x emissions data for the different sectors is EMEP (European Monitoring and Evaluation Programme).

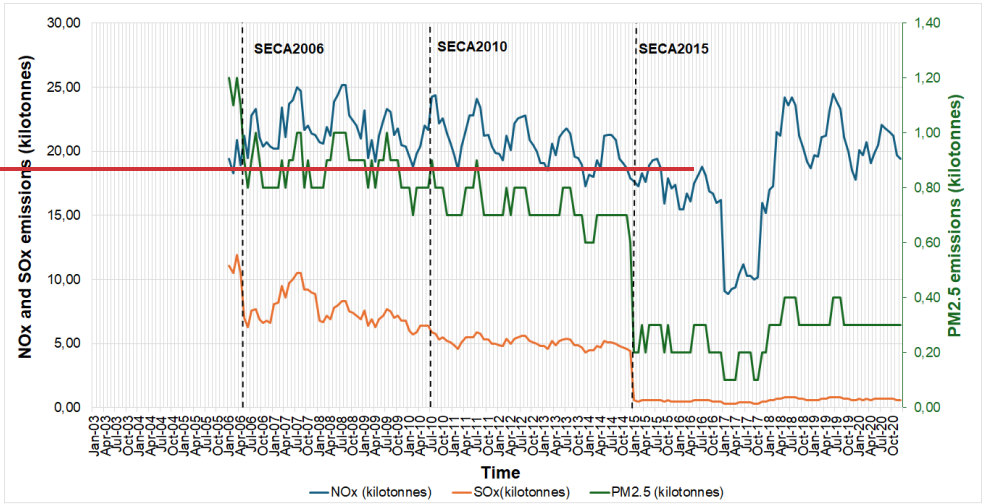
Figure 3 displays the monthly shipping emissions in the Baltic Sea calculated using the STEAM model. The changes in SO_2 emissions are a combination of changes in emissions due to the implementation of SECA regulations, and changes in shipping traffic density.

~~. The maximum allowed sulfur content of fuel was decreased from 2.7 to 1.5 % in May 2006, and it was further reduced to 1.0 % in July 2010, and finally, to 0.1 % in January 2015. The shipping activity in the Baltic Sea, both in terms of the total number of ships and the transport work, has increased steadily during 2006-2020, except for a few minor exceptions such as the economic recession, which started in 2008 and continued until 2009 in most areas of the neighbouring countries of the Baltic Sea (Jalkanen et al., 2014) and the COVID-19 pandemic in 2019 (HELCOM, 2021). According to emission model~~

~~computations from STEAM (Fig. 3), there has been a gradual decrease of SO_x and $PM_{2.5}$ emissions originating from shipping in the Baltic Sea since 2006.~~

From 2006 onward, shipping activity in the Baltic Sea has generally increased, with some exceptions. In particular, notable downturns in the total number of ships and the transport work were observed during the economic recession, which started in 2008 and continued until 2009 affecting most areas of the neighbouring countries of the Baltic Sea (Jalkanen et al., 2014), and again during the COVID–19 pandemic in 2019 (HELCOM, 2021). An exception to this upward trend occurred in 2013, with approximately 350 000 ship crossings in the used passage lines compared to 2012, likely due to the reductions in cargo ships attributed to reduced shipping activity resulting from the economic recession during these years (HELCOM, 2014). Finland also experienced a unique gross domestic product (GDP) decline from 2011 to 2012, largely due to weak export performance (Bank of Finland, 2018).

Emission computations from STEAM (Fig. 3) indicate a ~~This~~ gradual decline in ~~the modelled~~ SO_x and $PM_{2.5}$ emissions attributed to maritime traffic in the Baltic Sea starting in 2006. This decline in SO_x and $PM_{2.5}$ emissions continued gradually until 2014, at which point Baltic economies began to recover, reaching pre–crisis production levels (Bank of Finland, 2018). Following the introduction of SECA2015, ~~followed by~~ a sharp decrease in SO_x and $PM_{2.5}$ emissions is observed, highlighting the impact of stricter emissions controls. ~~from 2014 till 2015; the latter decrease aligns with the implementation of limiting the maximum allowed sulfur content of fuel in 2015.~~ The magnitude of the relative reduction of $PM_{2.5}$ emissions attributed to shipping is smaller than that of the SO_x emissions. The predicted annual NO_x emissions during the period between 2006 and 2020 have remained relatively stable, except for between 6 November 2017 and 31 December 2017, when a 95 % decrease in AIS data was observed due to an issue with AIS data reception.


SECA2006
SECA2010
SECA2015
NOx and SOx emissions (kilotonnes)
PM2.5 emissions (kilotonnes)
Time
NOx (kilotonnes)
SOx(kilotonnes)
PM2.5 (kilotonnes)

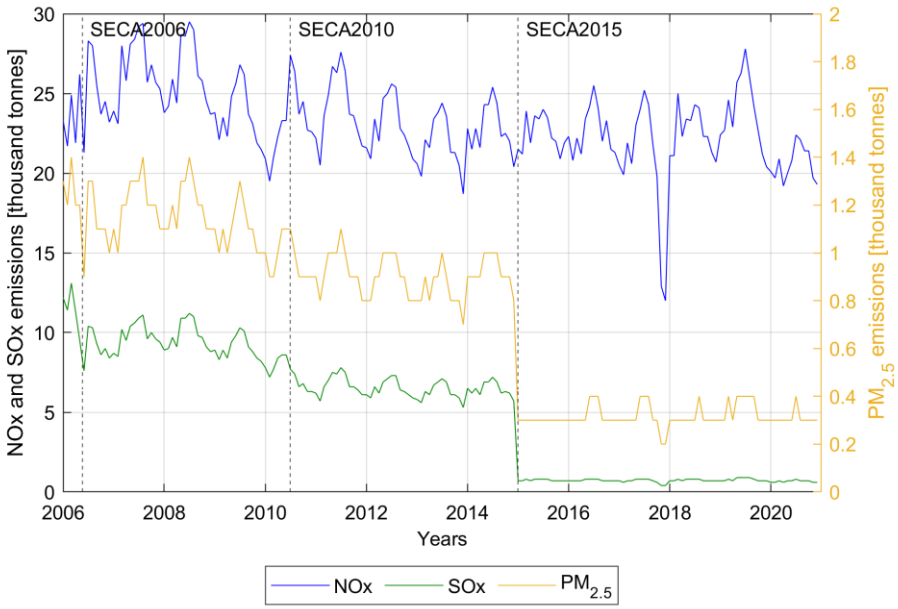


Figure 3: Predicted emissions of NO_x, SO_x, and $PM_{2.5}$ attributed to maritime traffic in the Baltic Sea from January 2006~~3~~ to December 2020, computed using the STEAM model. The SECA regulations ~~were~~ introduced in 2006, 2010 and 2015 are~~have~~ represented ~~(indicated with~~ ~~by~~ vertical dashed lines~~)~~.

4.2 Observed changes in ship traffic and concentrations of SO_2 and other air quality parameters at Utö

At Utö, local marine traffic typically consists of a few larger vessels per day. We ~~have~~ analysed the AIS data to quantify ~~of~~ the number of ships passing within 5 km of the island of Utö ~~per day at distances closer than 5 km~~on a daily and yearly basis (Fig. 4). As expected, results showed a declining trend in the yearly number of ships up to 2010, which coincided with the 2008–2009 economic recession affecting the majority of the Baltic Sea region. The decline continued through 2013, with a slight increase observed in 2014, as productions levels across the region began to recover (Bank of Finland, 2018).

From 2015 to 2020, however, no clear increasing or decreasing trend in the number of ships was detected. ~~On average, around 5–10 ships passed the island daily. The number of ships per day did not experience either an increasing or decreasing trend during 2013–2020. The amount of ship traffic has been fairly constant during this period. There have been a few local maxima, in particular during the summers of 2013 and 2014. Currently, we do not have access to the AIS data before 2013.~~

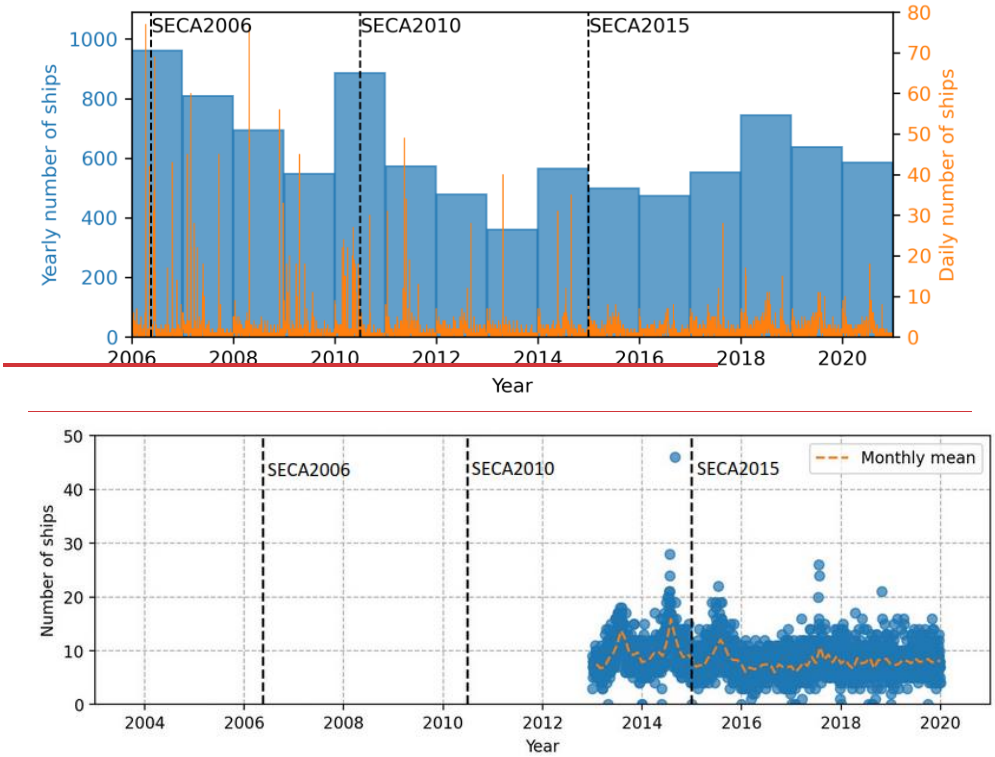


Figure 4: The total number of ships per ~~day~~ year (left y–axis) that have passed the island of Utö at distances closer than 5 km (blue ~~circles~~vertical bars~~lines~~). The total number of ships per day (orange ~~vertical~~ lines) is shown on the right y–axis~~orange dashed line presents the monthly mean of daily values~~.

The observed 30–day moving averages and moving percentiles of the concentration data for SO_2, $PM_{2.5}$, NO, NO_x and O_3 from 2006 to the end of 2020 are presented in Fig. 5. The dashed vertical lines represent the introduction of SECA2006, SECA2010

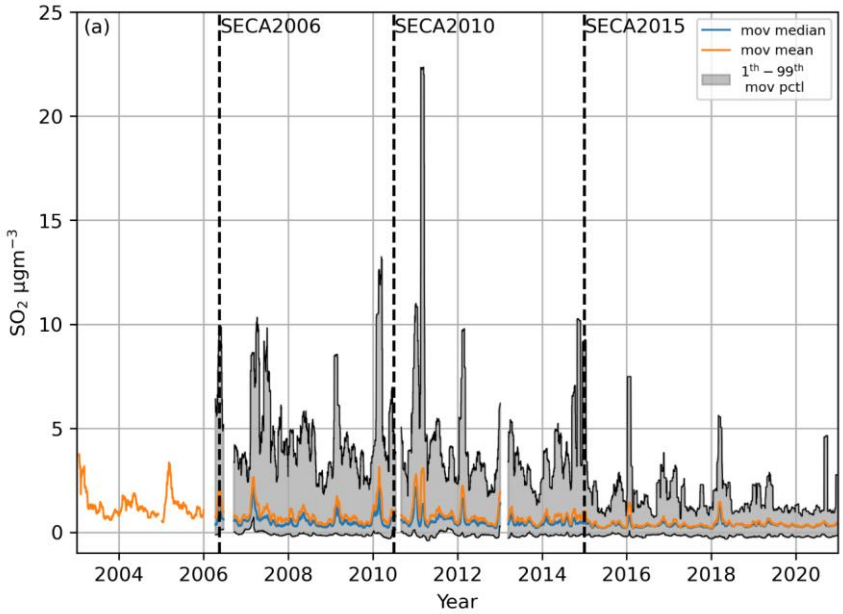
and SECA2015 regulations in 2006, 2010 and 2015, corresponding to 1.5 %, 1.0 % and 0.1 % sulfur fuel content limits, respectively. The effects of the sulfur cap in 2006 and 2015 are clearly visible, while the impact of 2010 is negligible. The change in 2015 is apparent in the shifts of the 1st and 99th percentiles corresponding to short–duration local ship plumes. Additionally, the mean and median values after 2015 are closer to each other, indicating that the higher peaks in the data occur less frequently (Fig. 5a).

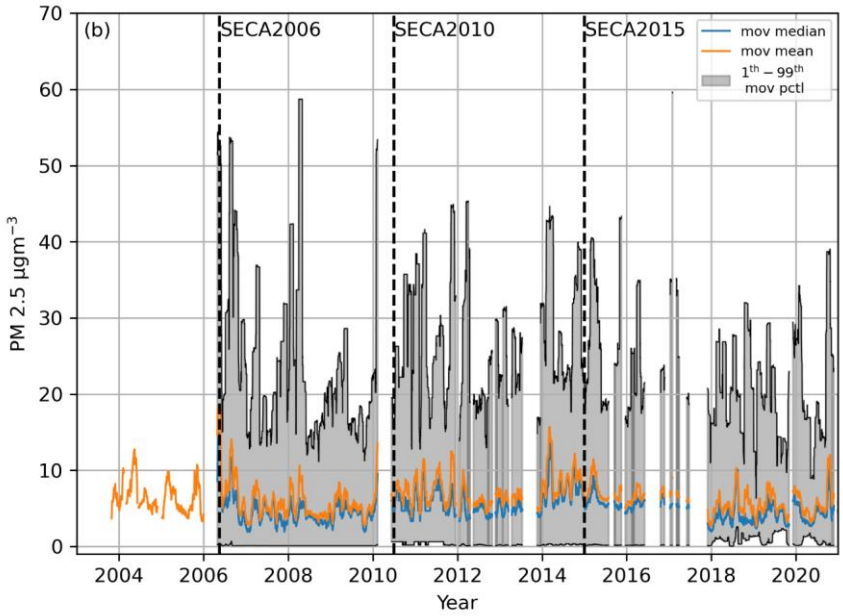
Table 2: Annual average and median concentrations of SO_2 at Utö. Values for the period 2003–2005 are based on hourly averages, while for the period 2006–2020, 1–minute time resolution data has been used. STD is the standard deviation. N (%) represents the fraction of the year for which high–quality data is available, expressed as a percentage. High–quality data, used in our analysis, are defined as valid data recorded, excluding those compromised by factors such as instrument malfunctions, environmental interference, or calibration issues.

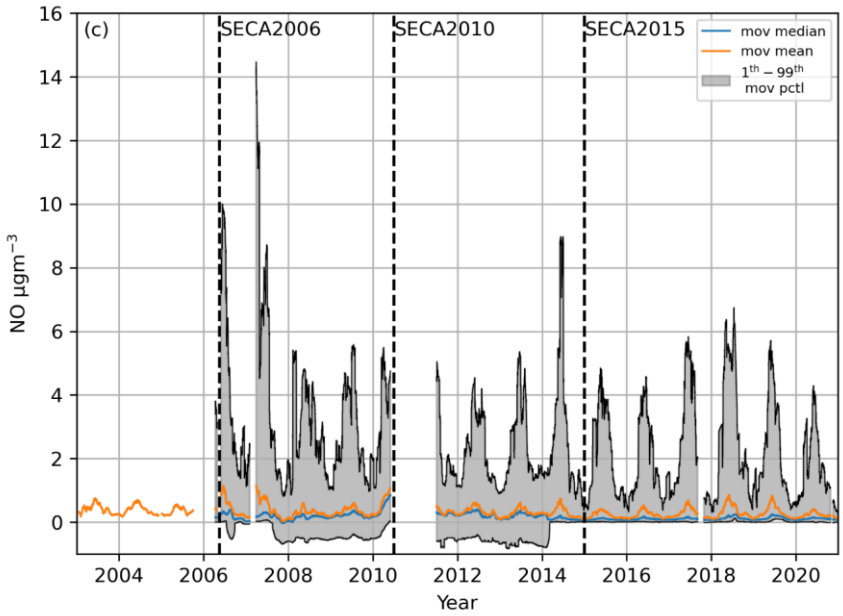
SO_2	Mean [µg m^{-3}]	Median [µg m^{-3}]	STD [µg m^{-3}]	N (%)
2003	1.38	0.70	2.15	98.3TBA
2004	1.17	0.80	1.13	91.5TBA
2005	1.32	0.80	1.45	97.1TBA
2006	0.93	0.55	1.51	55.7
2007	0.97	0.56	1.58	99.8
2008	0.70	0.40	1.19	98.9
2009	0.73	0.42	1.07	98.4
2010	1.12	0.58	1.80	89.0
2011	1.00	0.57	1.81	93.0
2012	0.78	0.45	1.18	95.86.1
2013	0.58	0.389	0.81	84.54
2014	0.75	0.46	1.08	99.5
2015	0.37	0.29	0.47	99.4
2016	0.45	0.32	0.68	99.45
2017	0.34	0.27	0.54	98.3
2018	0.50	0.367	0.60	99.3
2019	0.41	0.36	0.50	99.1
2020	0.34	0.32	0.35	99.1

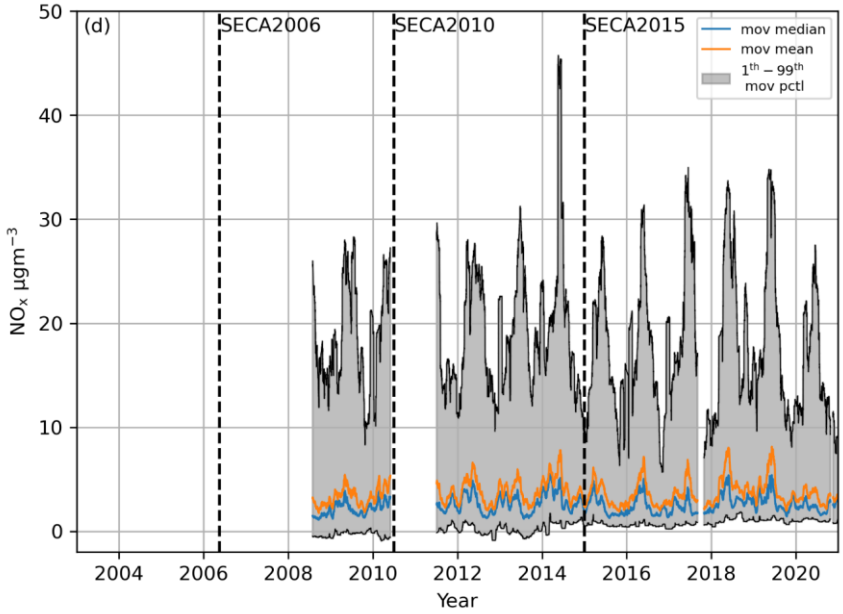
In general, the reduction in the release of SO_2 emissions attributed to shipping could also result in a decline in the production of particulate matter originating from precursor compounds from shipping. However, as the results (Fig. 5 and Appendix A)

demonstrate, there were no step changes in the $PM_{2.5}$ concentration during the considered period. This does not mean that the emissions of particulate matter from ships were at the same level as before the introduction of SECA regulations, but rather that the measurement of $PM_{2.5}$ is not sensitive enough to show the changes in the ultrafine particle size range. $PM_{2.5}$ concentrations also had some negative values, which were removed, leading to gaps in the time series (Fig. 5b). Seppälä et al. (2021) studied the impact of SECA on particle number size distribution (size range 7–537 nm) at Utö and found that the main changes due to SECA in particle number ~~were~~ occurred i~~o~~n sizes between 33 and 144 nm. ~~, and~~ T~~t~~hus, the impact on particle mass, resulting mainly from larger particles, is minor. Similarly to $PM_{2.5}$, no decreases were observed in the NO_x, NO and O_3 concentrations. However, for NO and NO_x, a period of data from 22 May 2010 to 15 June 2011 was removed (Figs. 5c and 5d) due to abnormally low values, likely caused by overly strict data processing. The year–to–year temporal variation of $PM_{2.5}$, NO, NO_x and O_3 (yearly mean and median values and standard deviation) are presented in Appendix A.




(b)
SECA2006
SECA2010
SECA2015
mov median
mov mean
1th – 99th
mov pctl
PM 2.5 μgm−3
Year
0
10
20
30
40
50
60
70
2004
2006
2008
2010
2012
2014
2016
2018
2020


(c)
SECA2006
SECA2010
SECA2015
mov median
mov mean
1th – 99th
mov pctl
NO µgm−3
Year
16
14
12
10
8
6
4
2
0
2004
2006
2008
2010
2012
2014
2016
2018
2020


(d)
SECA2006
SECA2010
SECA2015
mov median
mov mean
1th – 99th
mov pctl
NOx µgm−3
Year
0
10
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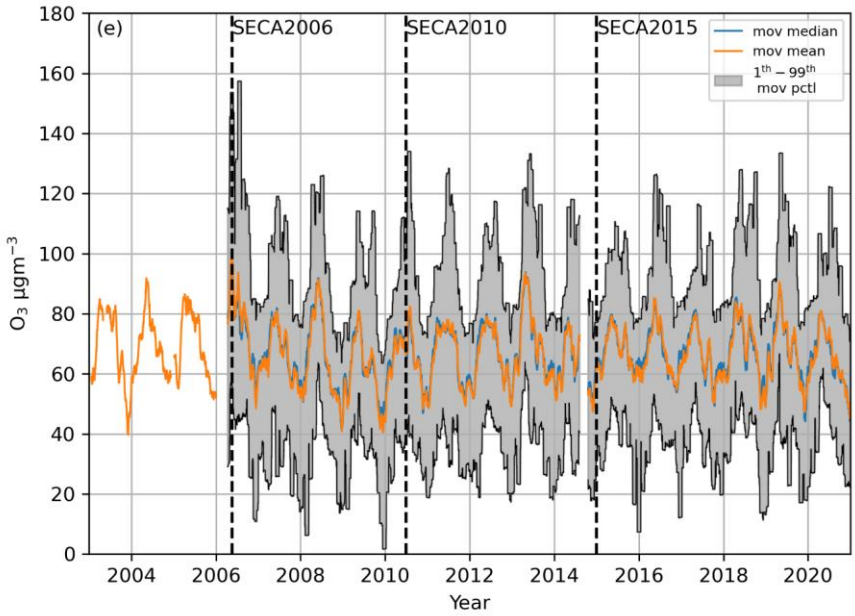
30
40
50
2004
2006
2008
2010
2012
2014
2016
2018
2020


(e)
SECA2006
SECA2010
SECA2015
mov median
mov mean
1th – 99th
mov pctl
O3 µgm−3
Year
0
20
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60
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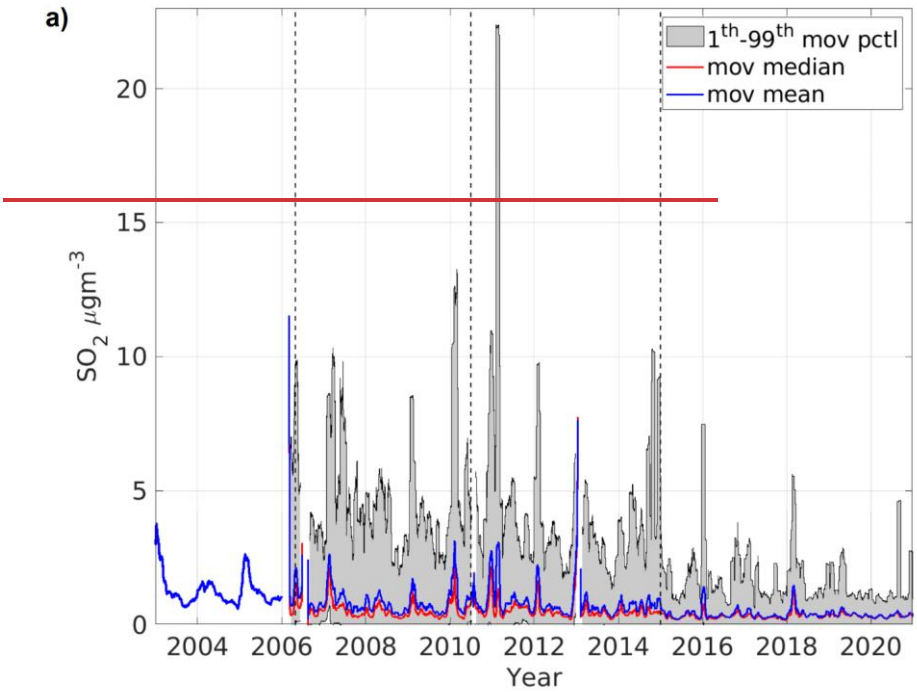
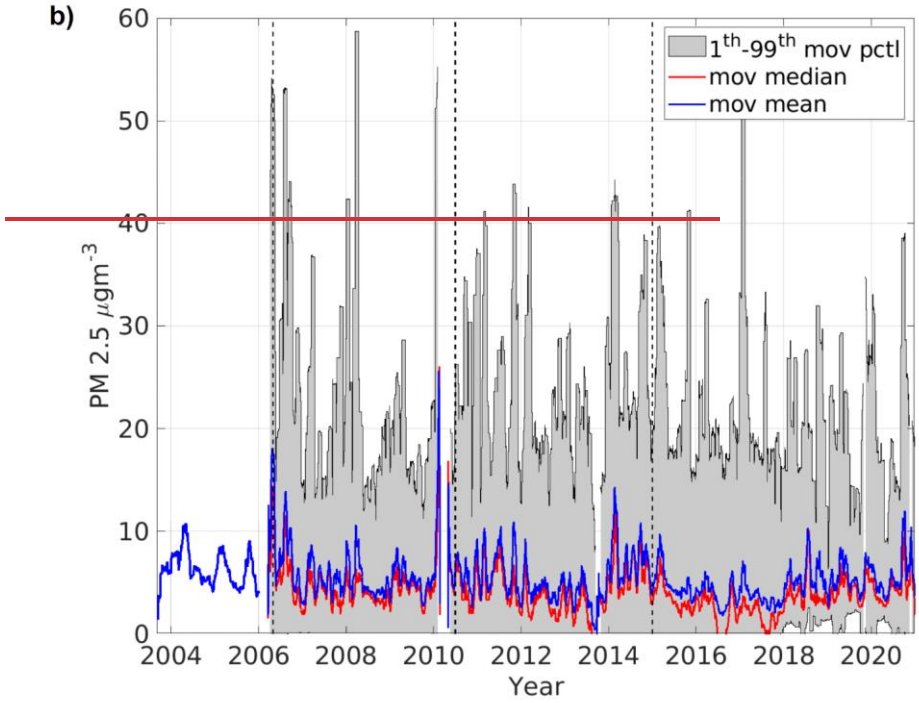
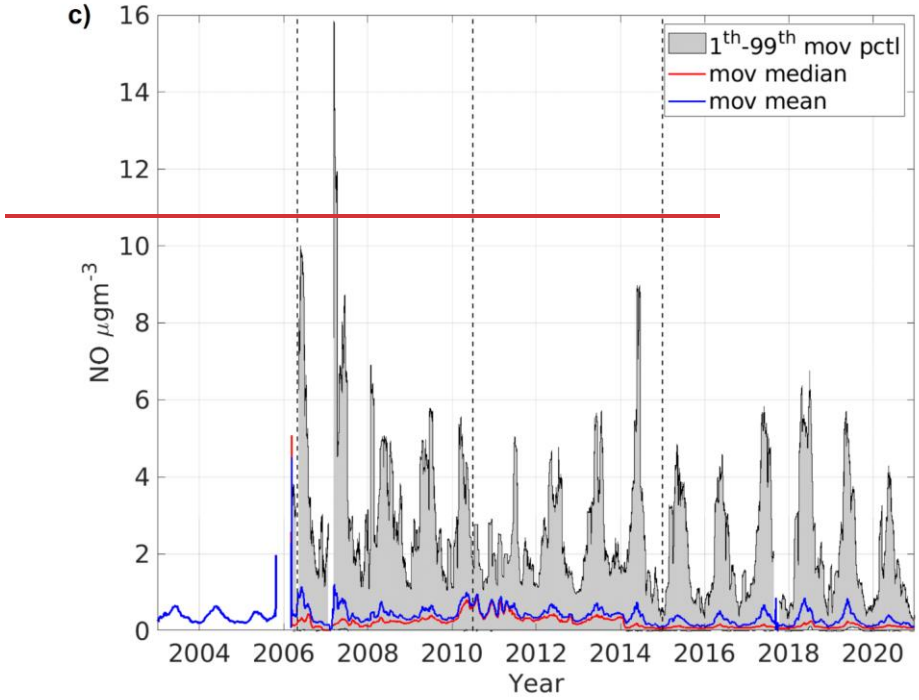
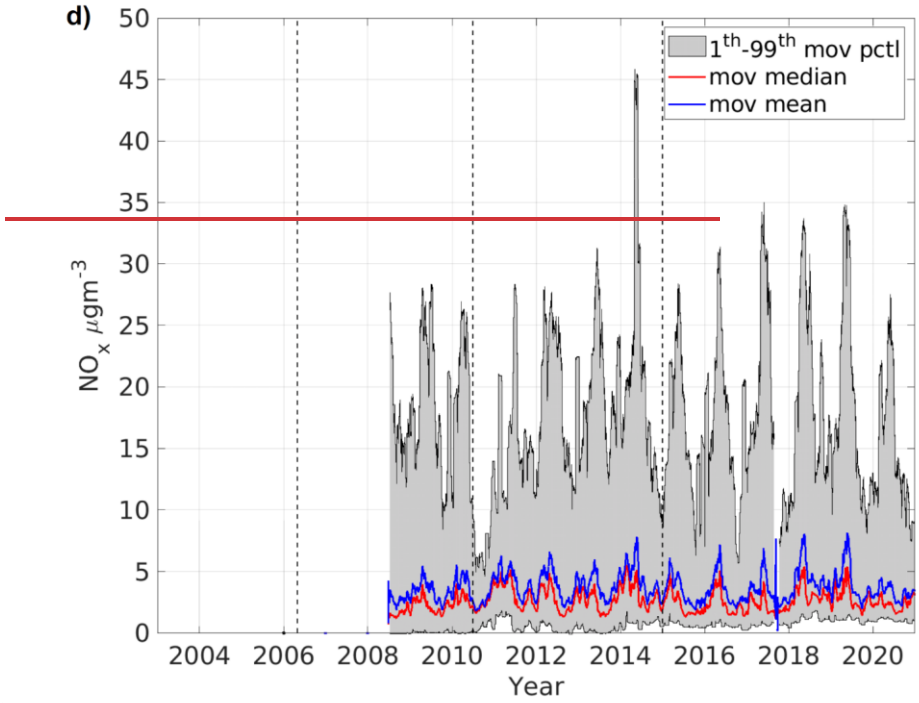
160
180
2004
2006
2008
2010
2012
2014
2016
2018
2020


a)
1th-99th mov pctl
mov median
mov mean
SO2 μgm-3
0
5
10
15
20
2004
2006
2008
2010
2012
2014
2016
2018
2020
Year


b)
1th-99th mov pctl
mov median
mov mean
PM 2.5 μgm-3
Year
0
10
20
30
40
50
60
2004
2006
2008
2010
2012
2014
2016
2018
2020


c)
1th-99th mov pctl
mov median
mov mean
NO μgm-3
Year
0
2
4
6
8
10
12
14
16
2004
2006
2008
2010
2012
2014
2016
2018
2020


d)
1th-99th mov pctl
mov median
mov mean
NOx μgm-3
Year
0
5
10
15
20
25
30
35
40
45
50
2004
2006
2008
2010
2012
2014
2016
2018
2020

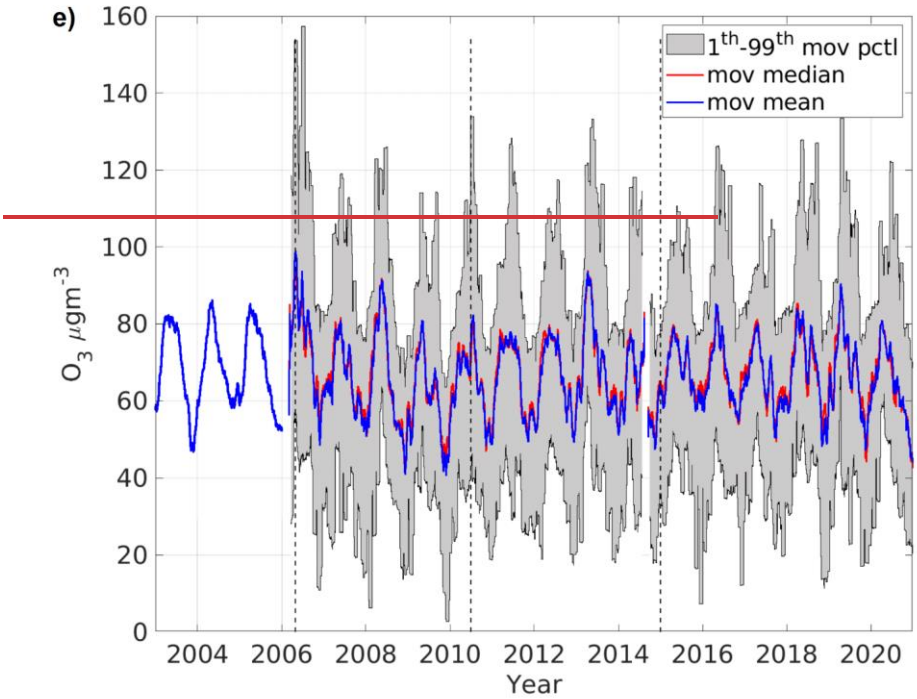


Figure 5: ~~Time series of measured concentrations presented as moving percentiles based on 1-minute temporal resolution in 30 days window for a) SO_2, b) $PM_{2.5}$, c) NO, d) NO_x and e) O_3 from 2003 to the end of 2020 at the air quality station in Utö. The implementation of the SECA regulations in 2006, 2010 and 2015 have been presented by three vertical dashed lines.~~ Time series of measured concentrations for (a) SO_2, (b) $PM_{2.5}$, (c) NO, (d) NO_x, and (e) O_3 from 2003 to the end of 2020 at the air quality station in Utö. The concentrations are presented as moving percentiles based on 1-minute resolution data within a 30-day window, with 30–day moving mean, median, and 1st–99th percentiles. For the years 2003–2005, only the moving mean is shown, based on 1-~~-~~hour~~0-minute~~ resolution data. The implementation of SECA regulations in 2006, 2010, and 2015 is indicated by three vertical dashed lines.

Since the years of 2006, 2010 and 2015 marked significant milestones, as they represented the introduction of the new SECA regulations, we decided to focus on the periods that occurred before and after the implementation of SECAs: 2003–2005, 2007–2009, 2011–2013 and 2016–2018. Specifically, we compared the three-~~-~~year average of SO_2 concentrations before the SECA introduction (2003–2005) to those following each SECA enactment (2007–2009, 2011–2013, and 2016–2018) (Table 2). ~~Our findings revealed notable reductions in three-year average SO_2 concentrations during these periods: 38% (after SECA~~

~~2006), 39% (after SECA 2010), and 67% (after SECA 2015), respectively~~ Our findings revealed that three–year average SO_2 concentrations from the pre-SECA period (2003–2005) decreased by 38 %, 39 %, and 67 % in comparison to the post-SECA periods (2007–2009, 2011–2013, and 2016–2018), respectively.

As indicated in the previous section, SO_2 emissions have been widely reduced across various industrial sectors. To estimate the local effect of shipping, we utilized NO, which is highly reactive and has a short lifetime in the air. Since it is one of the main gases in ship exhaust, it can serve as a marker for ship plumes originating near Utö. Therefore, we selected periods when SO_2 and NO both showed clear peaks simultaneously in the data, and when the wind was blowing from the western sector (180–360 degrees). The prominence of the peak indicating the peak height relative to other data was chosen to be 2- μg m^{-3} for both NO and SO_2. Figure 6 depicts the SO_2 concentration normalized by NO concentration during these plumes. Normalization using CO_2 concentrations would have allowed further analysis of fuel sulfur content, unfortunately such data has not been measured at Utö in a location suitable for ship SO_2 plume research prior to implementation of SECA in 2015. There is a clear difference before and after January 2015. Prior to the implementation of the strictest SECA regulation (i.e. SECA-2015), the SO_2 concentrations during local pollution plumes were of the same magnitude as NO. However, after the SECA–2015 regulation was enacted, the SO_2 concentrations decreased significantly ~~during~~ within these near–source plumes.

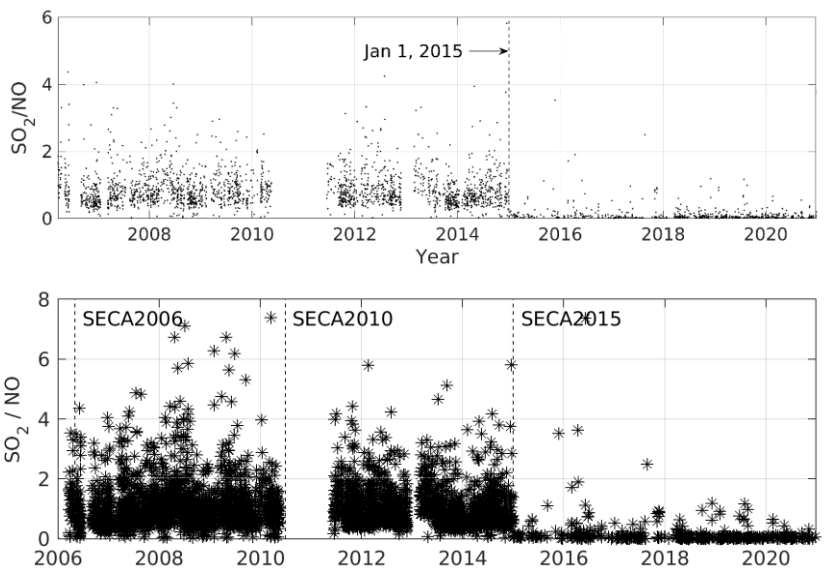
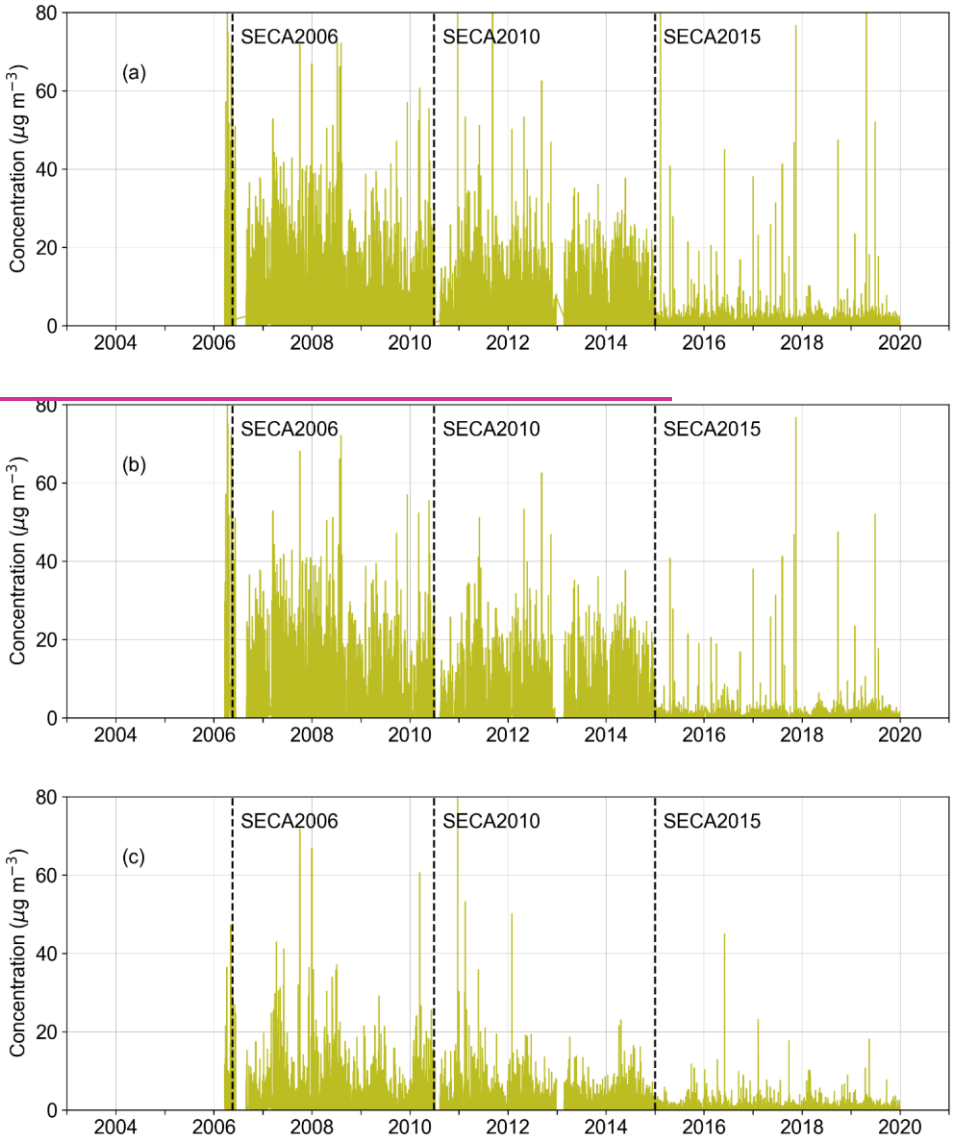


Figure 6: Time series of the observed SO_2/NO concentration ratio, when the wind was blowing from the westerly directions (wind direction 180-360º) and the plume from the passing ships was transported over the Utö island. Only the periods with simultaneous peaks in both SO_2 and NO signals were included into the analysis.

4.3. Dependence of concentrations on local wind direction

To study the impacts of ~~the~~ ships passing by Utö, we selected ~~the~~ SO_2 concentration data based on ~~the~~ wind direction. First, we separated the datapoints ~~that were~~ measured ~~during~~ when the wind was blowing from the shipping lane (~~includes~~ covering ~~the~~ wind directions from 185° to 315°) towards the measurement site from the rest of the data.

All data, ~~as well as~~along with data from when the wind was blowing from the direction of the shipping lane~~,~~ and data from the background sector (wind directions excluding the shipping lane sector) are shown in Figs. 7a–c. Similar~~ly~~ to the previously presented results, there is an evident decrease ~~of~~ in ~~the~~ SO_2 concentrations after the SECA ~~in~~ 2015, and a slight decrease after the SECA ~~in~~ 2010. Thi~~s~~e decreasing trend ~~can be seen~~is visible in all three plots, but the ~~strongest~~ most pronounced decrease ~~is~~ occurs after 2015 when only wind directions ~~of~~ from shipping lane were ~~selected~~considered.


(a)
(b)
(c)
SECA2006
SECA2010
SECA2015
Concentration (μg m⁻³)
0
20
40
60
80
2004
2006
2008
2010
2012
2014
2016
2018
2020

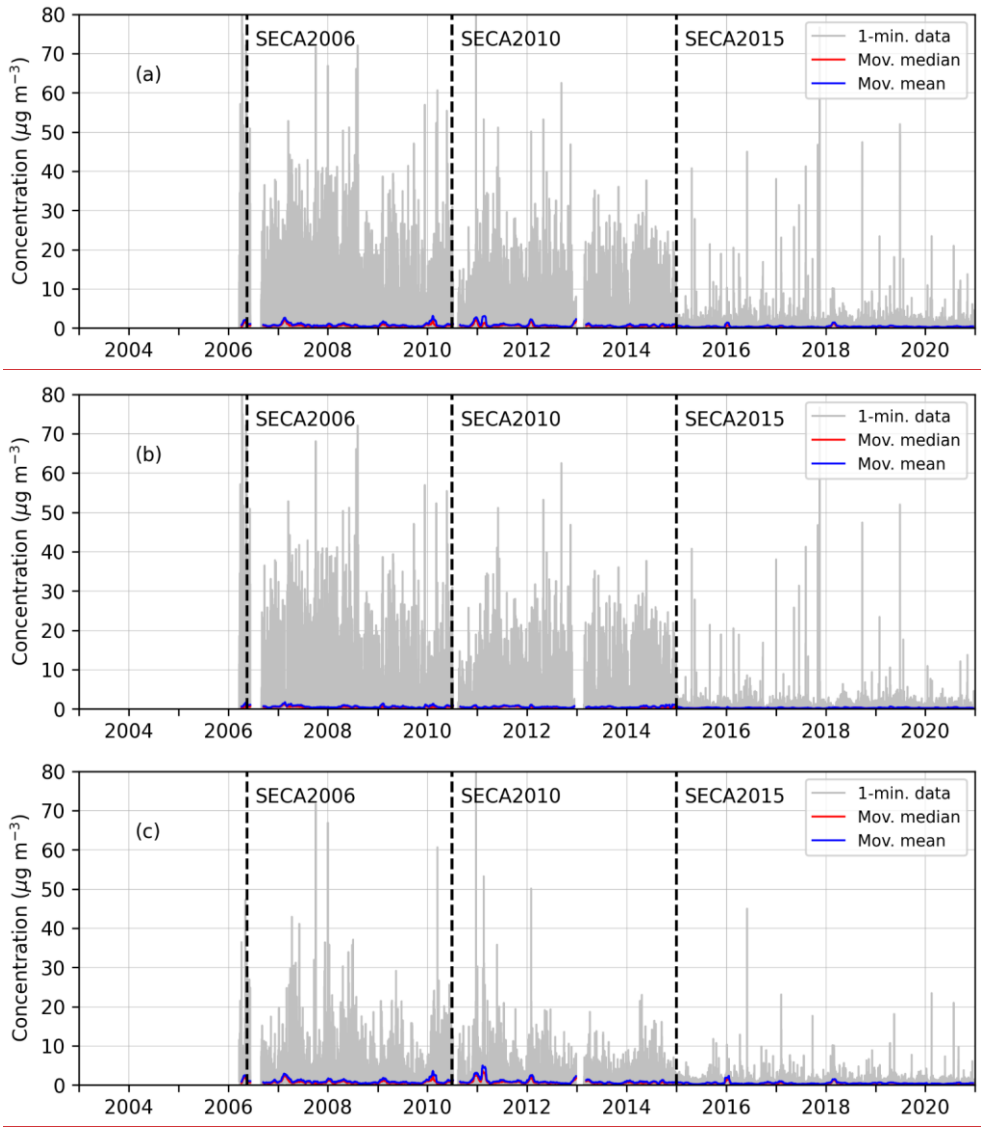
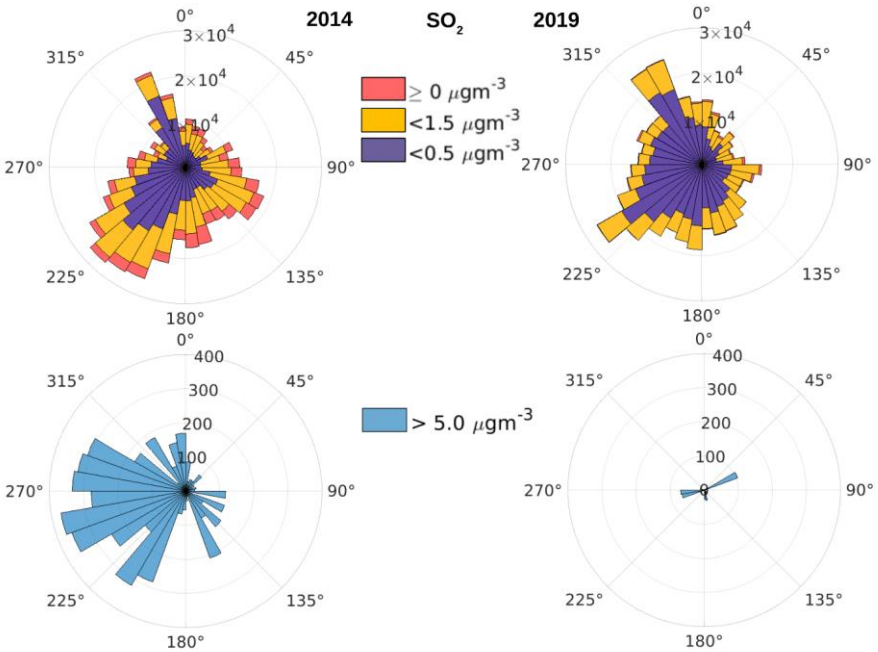

(a)
(b)
(c)
SECA2006
SECA2010
SECA2015
1-min. data
Mov. median
Mov. mean
Concentration (μg m⁻³)
0
10
20
30
40
50
60
70
80
2004
2006
2008
2010
2012
2014
2016
2018
2020

Figure 7: The time series of measured SO_2 concentrations (1–min. temporal resolution; grey solid lines) and their moving means (blue solid lines) and medians (red solid lines) in 30 days window with a) all the data points included (unstratified), b) data only from 130º wide wind sector from direction of shipping lane, extending from 185º to 315º and c) data only from background wind sectors excluding shipping lane, less than 185º and over 315º. The temporal resolution of the SO_2 concentration data was 1 minute on average.

To analyse the effect of wind direction in more detail, we generated concentration rose plots in finer 10–degree intervals (Fig. 8). To assess the effects of the implementation of SECA regulation in 2015, we chose two years: one immediately preceding the new regulation, 2014, and the other representing the 5–year period after, 2019. The years 2016, 2017, 2018 and 2019 were not substantially different in terms of the SO_x emissions from shipping (cf. Fig. 2) or the number of ships (Fig. 4). In this respectregard, any of these years could have been selected as an example year for the post–SECA 2015 analysis. The selected year 2019 was prior to the also not substantially different due to the COVID–19 pandemic and selected for comparison. The pandemic did not yet affect the emissions in Europe in 2019; these its effects were felt only during the subsequent years.

The measured annual distribution of wind directions is determined by the local climate at the site and potentially by local factors affecting wind measurements. The most common wind directions in this region are southwesterly, with relatively high contributions also from northwesterly directions, at angles from 320º to 350º (Fig. 8a and Fig. 8c). This northwesterly sector coincidentally corresponds to the routes of shipping to and from the harbor and wharf area of the island. The shipping traffic to the island typically consists of smaller vessels on average compared to the average marine transport in the neighboring ship lane. The upper panels in Figs. 8a–b include all measured concentrations. The observed SO_2 concentrations were systematically and substantially higher in 2014 compared to the values in 2019. Furthermore, if only peak concentrations during the plumes are considered (lower panels, Figs. 8c–d), it is evident that the numbers of the highest concentrations were drastically higher in 2014 compared to the corresponding values in 2019 and originated from the direction of the adjacent shipping lane (i.e., from the westerly directions).


2014
SO2
2019
≥ 0 μgm-3
<1.5 μgm-3
<0.5 μgm-3
> 5.0 μgm-3
0°
45°
90°
135°
180°
225°
270°
315°
3×10^4
2×10^4
1×10^4
400
300
200
100
0

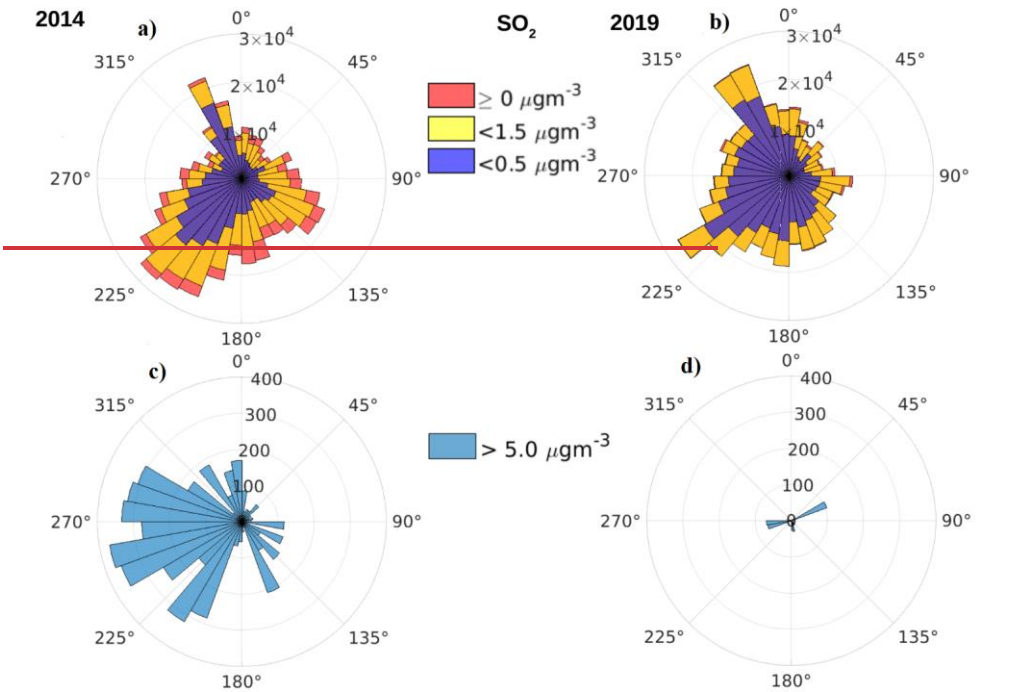


Figure 8: The measured concentrations of SO_2 at the island of Utö as a function of the wind direction, as polar histograms with 1-minute resolution. The panels a) and c) correspond to the data in 2014 and the panels b) and d) to those in in 2019. These two years were selected to represent the situation before and after the SECA regulation in 2015. The upper panels (a and b) include all datapoints, whereas the lower panels (c and d) include only the data, for which the concentrations were higher than a selected threshold value (5 µg m^{-3}). In panels a) and b), the red colour in the legend represents all data points with values larger than 0 µg m^{-3}. Overlaid on the red, the yellow colour shows data points with values below 1.5 µg m^{-3}, and the violet colour, which overlays the yellow, indicates data points with values below 0.5 µg m^{-3}. The radial axis represents the number of measured cases for each wind direction sector. ~~The red colour in the legend refers to all data, except for the regimes shown by the yellow and blue colours.~~

4.4. Uncertainties

Given the nearly two–decade long observation period, several factors could introduce uncertainties in the results. Instrumentation for all variables changed over time, and quality assurance methods improved. Furthermore, there were changes in the personnel responsible for maintaining and ensuring data quality. Although standard protocols were followed and measurement diaries were kept, these changes may have affected the results.

While our observations are crucial for understanding long–term, high temporal resolution air quality trends at remote marine research stations near shipping lanes, they represent only a specific norther region of the Baltic Sea; therefore, observations shown in our study cannot be directly described to represent other areas of the Baltic Sea.

~~4.5. Case study: Analysis of individual ship plumes of a selected ship prior and post the SECA regulation in 2015~~

~~We also analysed individual ship plumes of one Ro-Ro cargo ship that has regularly transited the sea route in the vicinity of the Utö island. The chosen vessel was classified as a Hazardous Category A cargo ship, with dimensions of 188 m in length, 26 m in width, and a draught of 6.6 m. Table 3 presents detailed information of the wind speed and direction, plume direction and the distance between the Ro-Ro cargo ship and the measurement tower, along with other parameters. The ship's speed during all passes was approximately 19 knots. The study provides only qualitative results, as the data shown has not been processed to take into account dilution of the ship plume.~~

~~Table 3: Wind and plume parameters, and ship location data recorded during its passing in the vicinity of the Utö island for the selected case study. The ship was equipped with a scrubber in June 2015.~~

Time (date and time)*	Window (TOA)**	Wind speed (m s^{-1})	Wind direction (º)	Plume direction (º)	Ship latitude (º)	Ship longitude (º)	Distance (m)***
2/11/2013 2:11	2/11/2013 2:17	10.2	251	71	59.75	21.35	3490
24/4/2014 21:48	24/4/2014 21:58	2.8	275	95	59.77	21.35	1690
23/10/2015 7:23	23/10/2015 7:31	7.8	283	103	59.74	21.34	4090
15/2/2017 22:16	15/2/2017 22:20	6.5	249	69	59.77	21.35	1580

~~* Time is the AIS file timestamp.~~

~~** Window (TOA= Time Of Arrival) is when the plume is estimated to reach the measurement tower located in Utö.~~

~~*** Distance is the distance between the ship and the measurement tower.~~

~~First, we identified the time periods, during which the plume originating from the Ro-Ro cargo ship was detectable at the measurement station at the island, i.e., the so-called plume windows. This analysis was done for selected recorded passage of the vessel during the years 2013–2017. The AIS data for this ship was not available during 2006–2012 and in 2016.~~

~~Second, we compiled and examined the temporal variations of the SO_2 and NO_x concentrations, which were measured at 1-minute intervals. The considered passages occurred approximately three and four years after the implementation of SECA regulations in 2010 (2.11.2013 and 24.4.2014), and at ten months and two years after the enforcement of the latest SECA~~

regulations in 2015 (23.10.2015 and 15.2.2017). The temporal evolution of the concentrations of SO_2 and NO_x during these plume windows have been presented in Figs. 9a–d.

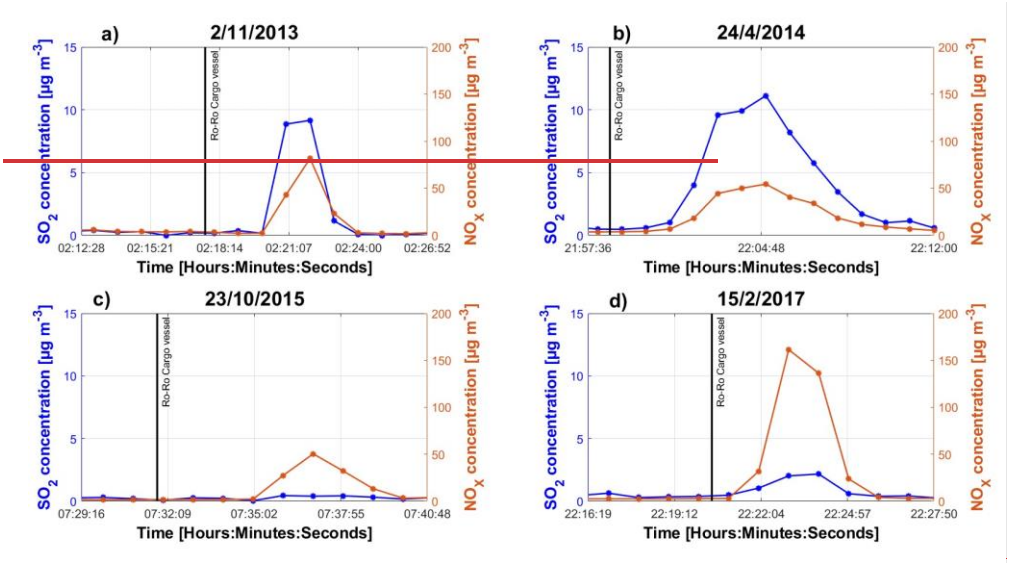


Figure 9: Temporal variation of the measured concentrations of SO_2 and NO_x at the Utö island before and after the passage of a Ro-Ro cargo ship along a shipping lane located west of the island. The concentrations are presented at two times before the implementation of SECA regulation on 1 January 2015, presented in panels (a) and (b) and at two times post this regulation presented in panels (c) and (d). Solid black vertical line indicates the ship's passing time, i.e., the time, during which the ship is closest to the measurement station. The time series of the concentrations of SO_2 (blue curves, left vertical axis) and NO_x (dark orange curves, right vertical axis) were measured at 1–minute intervals. The durations of the horizontal scales range from 9 to 15 minutes.

Prior to the passage of the Ro-Ro cargo vessel past Utö island, the concentrations of both NO_x and SO_2 consistently remained below the regional background values of approximately 6 µg m^{-3} and 1 µg m^{-3}, respectively. However, after some time from the moment, at which the ship had travelled past the closest point of the measurement station of Utö, there was a significant short-term rise in NO_x concentrations for all four considered cases, from a few tens to up to approximately 160 µg m^{-3} (Figs. 9a–d).

In contrast to the increases of the NO_x concentrations, there was a systematic difference in the rise of the SO_2 concentrations after the passing of the ship, when one considers the cases before and after the latest SECA regulations. After the studied

~~vessel's transit past Utö before the SECA regulation in 2015, the SO_2 concentrations increased to approximately 9 µg m^{-3} and 11 µg m^{-3}, respectively (Figs. 9a–b). The corresponding increases of the SO_2 concentrations after the 2015 SECA (and installation of a scrubber to the ship studied) were approximately 0.45 µg m^{-3} and 2.20 µg m^{-3}, respectively (Figs. 9c–d). These observations provide evidence on the efficacy of the SECA regulation implemented in 2015.~~

5 Conclusions

In this study, we have presented and analysed a high temporal resolution ~~of dataset that covers the data on~~ air quality dataset of SO_2, $PM_{2.5}$, NO, NO_x, and O_3 concentrations and of wind data for one and a half decades at the island of Utö in the Baltic Sea. This dataset is particularly unique, as there are no other similar long–term air quality observations~~datasets~~ from remote locations in the Baltic Sea, and the data set ~~it~~ has not been previously published or analyzed. Utö's central location next to northern Baltic~~t~~ Proper ~~in the middle of the open sea~~, with minimal influence from other pollution sources, makes it an ideal site to study long–term pollution trends from shipping. The period of the study includes all three~~,~~ consecutive revisions of the SECA regulations.

The findings derived from the analysis of the air quality and meteorological datasets and STEAM modelling indicated that the SECA regulations, introduced in 2006, tightened in 2010 and further strengthened in 2015, have been successful in reducing the observed SO_2 concentrations at Utö during the studied period ~~-~~2003~~–~~–2020. ~~In fact, c~~Comparing three–year average SO_2 concentrations pre–SECA (2003–2005) to post–SECA periods (2007–2009, 2011–2013, 2016–2018) revealed reductions of 38 % (SECA 2006), 39 % (SECA 2010), and 67 % (SECA 2015), respectively. During the target period, there were no clear decreasing trends for the concentrations of $PM_{2.5}$, NO_x, NO and O_3~~, or $PM_{2.5}$~~. ~~The year–to–year variations of the concentrations were substantial for all pollutants and may be attributed to differences in regional meteorology.~~ The year–to–year variations of the concentrations were substantial for all pollutants; these ~~are~~ were attributed partly to the variations in regional meteorology, partly to the variations of emissions.

We also analysed polar histograms of SO_2–concentration during two representative years, 2014 and 2019, i.e., before and after the implementation of the SECA ~~regulations in~~ 2015. This analysis was done both for the whole dataset and for selecting only the highest measured concentrations, above a specified threshold. The numbers of the cases with the highest SO_2 concentrations were drastically higher in 2014 compared to the corresponding values in 2019. The highest concentrations were mostly originating from the direction of the adjacent shipping lane. This analysis therefore indicates that the highest measured concentrations attributed to shipping have substantially decreased after the SECA ~~regulations in~~ 2015.

By filling an important gap in the current literature, this study provides a thorough view of the air quality trends in the Baltic Sea and highlights the importance of good quality, high temporal resolution, long–term air quality data at remote marine

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research stations~~, especially those near heavily trafficked shipping lanes with minimal influence from other pollution sources~~. Such observations are crucial for both quantitative and qualitative analyses of the impacts of regulatory environmental ~~regulations~~legislation. ~~While the scope of this study does not include employing an atmospheric transport model~~The findings of this work will provide valuable insights into the effectiveness of SECA regulations and serve as a benchmark for local and regional dispersion modelling efforts for future research utilizing the data of this study. ~~The present study highlights the importance of good quality, high temporal resolution, long-term air quality data to support the evaluation of the impacts of changes in environmental regulations and legislation, in this study impacts of SECA~~.

Appendix A

Table A1: Annual average and median concentrations of $PM_{2.5}$ at Utö. Values for the period 2003–2005 ~~Values for year 2004 and 2005~~ are based on hourly averages, while ~~and~~ for the period 2006–~~-~~2020 on 1–~~-~~minute time resolution data has been used. STD is the standard deviation. N (%) represents the fraction of the year for which high–quality data is available, expressed as a percentage. High–quality data, used in our analysis, are defined as valid data recorded, excluding those compromised by factors such as instrument malfunctions, environmental interference, or calibration issues.

$PM_{2.5}$	Mean [µg m^{-3}]	Median [µg m^{-3}]	STD [µg m^{-3}]	N (%)
2003	5.97	~~4.30~~	5.57	TBA
2004	6.82	~~5.20~~	6.07	TBA
2005	6.07	~~4.50~~	6.13	TBA
2006	8.35	5.55	9.43	70.3
2007	5.28	3.93	5.49	99.9
2008	5.26	3.88	6.00	98.8
2009	5.30	4.37	4.84	98.0
2010	6.39	4.67	7.18	69.2
2011	6.35	4.67	7.28	93.8
2012	4.51	3.05	5.58	96.0
2013	3.85	2.24	5.29	86.7
2014	7.35	5.73	7.56	99.5
2015	4.97	3.48	6.21	98.9
2016	3.85	2.19	5.25	98.7
2017	3.66	1.96	5.91	95.9
2018	5.58	3.83	4.94	97.9
2019	5.49	4.09	4.54	90.6

2020	5.42	3.93	5.02	87.5
$PM_{2.5}$	**Mean [µg m^{-3}]**	**Median [µg m^{-3}]**	**STD [µg m^{-3}]**	**N (%)**
2003	5.97	-	5.57	20.0
2004	6.82	-	6.07	84.8
2005	6.07	-	6.13	93.8
2006	8.53	5.67	9.44	68.9
2007	5.28	3.93	5.49	99.9
2008	5.25	3.87	5.99	98.8
2009	5.30	4.37	4.84	98.0
2010	7.73	6.02	7.07	58.2
2011	7.60	6.02	7.23	79.3
2012	6.14	4.70	5.54	72.0
2013	5.92	4.54	5.32	58.5
2014	8.97	7.24	7.34	82.4
2015	7.06	5.66	6.15	71.3
2016	6.38	5.28	5.15	62.1
2017	5.78	4.27	6.31	63.1
2018	5.61	3.85	4.93	97.3
2019	5.56	4.14	4.53	89.5
2020	5.51	3.99	5.00	86.2

Table A2: Annual average and median concentrations of NO at Utö. Values for the period 2003–2005 ~~Values for year 2004 and 2005~~ are based on hourly averages, while ~~and~~ for the period 2006–2020 on 1–minute time resolution data has been used. STD is the standard deviation. N (%) represents the fraction of the year for which high–quality data is available, expressed as a percentage. High–quality data, used in our analysis, are defined as valid data recorded, excluding those compromised by factors such as instrument malfunctions, environmental interference, or calibration issues.

NO	**Mean [µg m^{-3}]**	**Median [µg m^{-3}]**	**STD [µg m^{-3}]**	**N(%)**
2003	0.40	~~0.10~~	0.76	

2004	0.39	~~0.20~~	0.60	TBA
2005	0.35	~~0.10~~	0.57	TBA
2006	0.47	0.14	1.87	71.2
2007	0.50	0.19	1.70	86.9
2008	0.43	0.23	1.42	99.1
2009	0.43	0.28	1.09	97.9
2010	0.66	0.56	0.87	97.0
2011	0.53	0.41	0.81	95.0
2012	0.44	0.31	1.10	98.1
2013	0.46	0.31	1.23	98.0
2014	0.30	0.09	1.10	99.5
2015	0.22	0.08	1.09	98.7
2016	0.23	0.09	1.20	99.5
2017	0.29	0.09	1.86	86.0
2018	0.34	0.11	1.76	99.3
2019	0.31	0.12	1.73	99.1
2020	0.22	0.10	1.41	95.9
NO	**Mean [µg m^{-3}]**	**Median [µg m^{-3}]**	**STD [µg m^{-3}]**	**N (%)**
2003	0.40	-	0.76	98.3
2004	0.39	-	0.60	91.1
2005	0.35	-	0.57	70.4
2006	0.46	0.14	1.85	71.3
2007	0.40	0.16	1.62	86.9
2008	0.29	0.15	1.30	99.1
2009	0.33	0.22	1.04	97.9
2010	0.64	0.48	1.27	35.9 *
2011	0.33	0.25	0.98	50.2 *
2012	0.33	0.24	1.11	98.1
2013	0.30	0.21	1.14	98.0

2014	0.29	0.09	1.09	99.4
2015	0.22	0.08	1.08	98.6
2016	0.23	0.09	1.25	99.5
2017	0.29	0.09	1.86	86.0
2018	0.34	0.11	1.76	99.3
2019	0.31	0.12	1.73	99.1
2020	0.22	0.10	1.41	95.9

**Low N (%) is attributed to the removal of data from 22 May 2010 to 15 June 2011, due to abnormally low values, likely caused by overly strict data processing.*

Table A3: Annual average and median concentrations of NO_x at Utö. Values for the period 2003–2005 ~~Values for year 2004 and 2005~~ are based on hourly averages, while ~~and~~ for the period 2006–2020 on 1-minute time resolution data has been used. STD is the standard deviation. N (%) represents the fraction of the year for which high–quality data is available, expressed as a percentage. High–quality data, used in our analysis, are defined as valid data recorded, excluding those compromised by factors such as instrument malfunctions, environmental interference, or calibration issues.

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NO_x	Mean [µg m^{-3}]	Median [µg m^{-3}]	STD [µg m^{-3}]	N (%)
2003	-	-	-	-
2004	-	-	-	-
2005	-	-	-	-
2006	-	-	-	-
2007	-	-	-	-
2008	2.73	1.62	4.50	46.3
2009	3.37	2.15	4.47	97.8
2010	3.34	2.42	3.82	96.4
2011	4.13	3.33	3.64	94.9
2012	3.93	2.64	4.86	97.7
2013	3.76	2.50	4.76	97.5
2014	4.32	2.99	4.97	99.4

2015	3.31	2.13	4.35	98.7
2016	3.47	2.25	4.92	99.5
2017	3.50	2.13	5.97	86.0
2018	4.49	3.01	5.92	99.3
2019	4.06	2.59	5.91	99.1
2020	3.27	2.30	4.78	95.9
NO_x	**Mean [µg m^{-3}]**	**Median [µg m^{-3}]**	**STD [µg m^{-3}]**	**N (%)**
2003	-	-	-	-
2004	-	-	-	-
2005	-	-	-	-
2006	-	-	-	-
2007	-	-	-	-
2008	2.57	1.51	4.43	47.1
2009	3.32	2.12	4.46	98.1
2010	4.20	2.72	5.66	35.9 *
2011	3.41	2.39	4.09	50.2 *
2012	3.87	2.59	4.93	98.1
2013	3.69	2.46	4.75	98.0
2014	4.32	2.98	4.97	99.4
2015	3.31	2.13	4.35	98.6
2016	3.47	2.25	4.91	99.5
2017	3.50	2.13	5.97	86.0
2018	4.49	3.01	5.92	99.3
2019	4.06	2.59	5.91	99.1
2020	3.27	2.31	4.78	95.9

**Low N (%) is attributed to the removal of data from 22 May 2010 to 15 June 2011, due to abnormally low values, likely caused by overly strict data processing.*

Table A4: Annual average and median concentrations of O_3 at Utö. Values for the period 2003–2005 ~~Values for year 2004 and 2005~~ are based on hourly averages, while ~~and~~ for the period 2006–2020 on 1–minute time resolution data has been used. STD is the standard deviation. N (%) represents the fraction of the year for which high–quality data is available, expressed as a percentage. High–quality data, used in our analysis, are defined as valid data recorded, excluding those compromised by factors such as instrument malfunctions, environmental interference, or calibration issues.

O_3	Mean [µg m^{-3}]	Median [µg m^{-3}]	STD [µg m^{-3}]	N (%)
2003	67.04	~~68.00~~	19.97	
2004	69.73	~~69.00~~	16.47	TBA
2005	68.71	~~67.00~~	18.20	TBA
2006	74.95	74.08	22.23	77.6
2007	65.66	65.87	16.46	99.8
2008	65.01	63.53	20.41	98.8
2009	60.12	59.91	16.87	98.3
2010	64.42	64.48	16.36	99.1
2011	66.17	66.11	16.43	95.1
2012	65.62	66.24	16.02	97.5
2013	70.28	68.93	18.05	98.4
2014	62.36	62.42	15.98	82.0
2015	67.57	68.12	13.66	99.3
2016	66.70	66.58	15.88	99.5
2017	65.61	66.83	14.53	98.7
2018	67.63	67.59	17.67	99.3
2019	67.24	67.71	17.91	99.1
2020	64.59	65.41	15.55	99.1
O_3	**Mean [µg m^{-3}]**	**Median [µg m^{-3}]**	**STD [µg m^{-3}]**	**N (%)**
2003	67.04	-	19.97	92.3
2004	69.73	-	16.47	90.0
2005	68.71	-	18.20	97.1

2006	74.94	74.08	22.23	77.6
2007	65.66	65.87	16.46	99.8
2008	64.99	63.46	20.38	98.8
2009	60.09	59.91	16.90	98.3
2010	64.43	64.49	16.35	99.1
2011	66.17	66.10	16.42	95.1
2012	65.64	66.28	16.00	97.5
2013	70.28	68.93	18.05	98.4
2014	62.36	62.43	15.98	82.0
2015	67.58	68.13	13.65	99.3
2016	66.68	66.51	15.86	99.5
2017	65.61	66.83	14.53	98.7
2018	67.64	67.59	17.67	99.3
2019	67.25	67.72	17.92	99.1
2020	64.55	65.35	15.55	99.1

Code availability

Codes for the analysis of long-term pollutant concentrations ~~and of individual ship plumes in case of one selected ship prior and post SECA~~ are available upon request from Androniki Maragkidou (androniki.maragkidou@fmi.fi). Codes for the concentration analyses are available from Tiia Grönholm (Tiia.Gronholm@fmi.fi).

Data availability

~~The measured data are~~The 1–minute air quality data from 2006-2020 is available in Zenodo [DOI to be added]. The data set also includes 10–minute resolution meteorological data (wind speed, wind direction, air temperature, air pressure, relative humidity and precipitation) and 1-hour air quality data from 2003-2005. Meteorological data is also available from FMI Open Data (https://en.ilmatieteenlaitos.fi/open-data).

Author contributions

AM conducted part of the analyses, wrote the first draft of the manuscript, and lead the processing of the manuscript to its final format. TG performed most of the data analysis and part of the writing. LR processed the AQ data and computed some of the figures. LR, LL, and JK contributed to the planning of the study and the writing of the article. TM and TA were responsible for the measurements at the Utö island and contributed to the analysis of the measurements. J-PJ has done the computations on the emissions of shipping using the STEAM model. JN is responsible of the computer analyses underlying Fig. 7. All authors commented on the manuscript and provided feedback.

Competing interests

The authors declare that they have no conflict of interest.

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