

# Supplementary material

## International Maritime Regulation decreases sulfur dioxide but increases nitrogen oxide emissions in the North and Baltic Sea.

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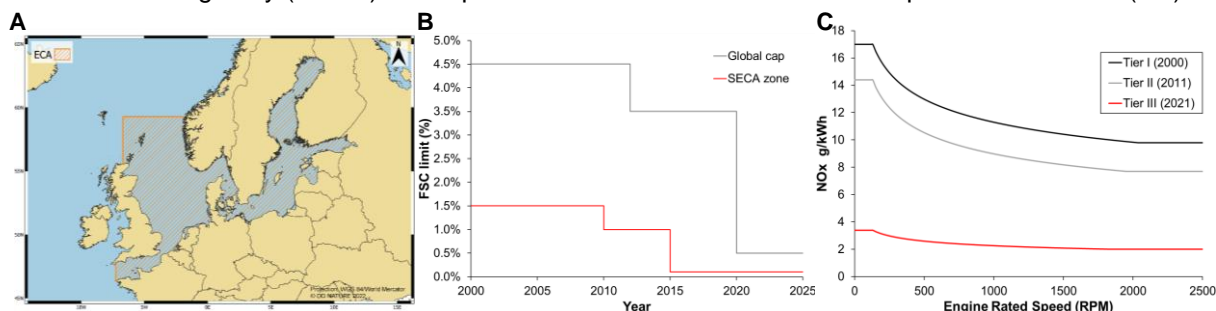
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## Supplementary Notes

### Supplementary Notes 1: Marpol Annex VI

#### SO<sub>x</sub> regulation

Regulation 14 of MARPOL Annex VI introduced limits on SO<sub>x</sub> emissions from Ocean Going Vessels (OGVs) and designated the Baltic Sea, the North Sea and English Channel as a Sulfur Emission Control Area (SECA) (Supplementary Figure 1A)<sup>1,2</sup>. From 2008 OGVs operating in the SECA are obliged to use compliant fuels or an Exhaust Gas Cleaning System to achieve the same amount of sulfur emission reduction as for using compliant fuel<sup>3,4</sup>. Since 2015 the maximum allowed Fuel Sulphur Content (FSC) inside SECAs is 0.10 %S m/m. In 2020, the FSC limits outside ECAs were also sharpened to 0.50 %S m/m by the so-called “global sulfur cap”<sup>5</sup> (Supplementary Figure 1B) and the “carriage ban”<sup>6</sup>. In addition, SO<sub>x</sub> emission regulations have been implemented at the level of the EU by the EU Sulphur Directive ((EU) 2016/802)<sup>7</sup>. Assessment of compliance by port inspection authorities is based on analysis of fuel samples or documentary checks in accordance with the Commission Implementing Decision ((EU) 2015/253)<sup>8</sup>. Inspection data is shared through Thetis-EU — the port inspection database managed by (EMSA) and reported on an annual basis to the European Commission (EC)<sup>9</sup>.



**Supplementary Figure 1. Emission Control Area (ECA) as defined by MARPOL Annex VI (A). Limits on the Fuel Sulfur Content of marine fuels according to the MARPOL Annex VI regulation 14 (B). NO<sub>x</sub> emission limits according to MARPOL Annex VI Regulation 13 (C).**

#### NO<sub>x</sub> regulation

Regulation 13 of MARPOL Annex VI introduced NO<sub>x</sub> emission limits<sup>2,10,11</sup> and dedicated Nitrogen Emission Control Areas (NECAs). In 2016, the first NECAs entered into force on the East and West coasts of North America and the Caribbean Sea. The Baltic and North Sea SECA was designated as NECA in 2021 and is referred to as an ECA from that moment<sup>12,13</sup> (Supplementary Figure 1A). The establishment of the ECAs required cooperation and agreement from many countries. The aim of the ECAs was not only to reduce the emissions of NO<sub>x</sub> and SO<sub>x</sub> in Europe’s most densely populated areas, but more importantly it was hoped that SO<sub>x</sub> and NO<sub>x</sub> emission restrictions would also lead to a reduction of the particulate matter (PM).

The NO<sub>x</sub> emission standards are determined by the amount of NO<sub>x</sub> emissions (in g) per brake horse power (BHP) on the crankshaft (in kWh) and are categorized into four tiers based on the Keel Laying Date (KLD) of the OGV. The limit for each tier decreases with the Engine Rated Speed (ERS or n) expressed in rotations per minute (rpm), although the limits per tier are uniform for OGVs with an ERS of less than 130 rpm, which in reality applies to the vast majority of the section of medium to large sized merchant vessels (> 5000 GT) (Supplementary Figure 1C). The engines are certified before and after installation on board, utilizing testing methods outlined in the NO<sub>x</sub> Technical Code<sup>14</sup>. For the two test cycles of the MEs, the verification procedures take into account a weighted average of four engine loads with corresponding weighting factors (Supplementary Table 1)<sup>2,14</sup>. As a result of legal restrictions and operational constraints, the port inspection authorities can rely solely on examining the mandatory onboard documentation to verify compliance to the NO<sub>x</sub> regulations. The use of emission measurements as legal evidence is currently not implemented nor is its use as an enforcement mechanism described in the regulations. Moreover, conducting an analysis in port would not necessarily yield meaningful results, as to verify compliance, the ME need to operate at an engine load of minimum 25%. In the case of Tier III OGVs, the port inspection authorities have the added option to verify the effective operation of abatement technology like selective catalytic reduction (SCR) and exhaust gas recirculation (EGR). However, there is a lack of explicit guidelines in the NO<sub>x</sub> technical code regarding the recording of onboard measured emission data and the tracking of abatement system operations. Moreover, it is important to highlight that malfunctioning SCR systems can generate N<sub>2</sub>O emissions<sup>15</sup>. This is currently not regulated by MARPOL Annex VI. This is a matter of great significance considering the impact of climate change, as N<sub>2</sub>O has a global warming potential (GWP) of approximately 300 times that of carbon dioxide (CO<sub>2</sub>)<sup>15,16</sup>.

**Supplementary Table.1. Test cycles and weighting factors according to the NO<sub>x</sub> Technical code<sup>14</sup>.**

	Speed	100%	100%	100%	100%
E2	Power	100%	75%	50%	25%
	WF	0.2	0.5	0.15	0.15
	Speed	100%	91%	80%	63%
E3	Power	100%	75%	50%	25%
	WF	0.2	0.5	0.15	0.15

Unlike SO<sub>2</sub>, NO<sub>x</sub> emissions from ships are not regulated by an additional EU directive, which means there are no common EU monitoring rules and enforcement procedures. As a result, the NO<sub>x</sub> inspection data of EU MSs are not shared through Thetis-EU, nor are inspection results reported to the EC. On board inspections executed by the port inspection authorities are currently limited to documentary checks.

## Supplementary Methods

### Supplementary Methods 1: Sniffer measurement technique

#### Remote FSC measurements

All of the measurements were conducted using a similar methodology based on comparable sniffer sensors<sup>17–20</sup>. The FSC can be calculated based on the ambient and exhaust plume SO<sub>2</sub> and CO<sub>2</sub> concentrations<sup>17,19–26</sup>. As 97.8% of the sulfur (S) in the fuel will be transformed to SO<sub>2</sub><sup>27–29</sup>, the SO<sub>2</sub> values are used to retrieve the S-content, the CO<sub>2</sub> is used to derive the carbon (C) content. To retrieve the amount of combusted fuel the C-content is multiplied by 87%, which is the average C-content for marine fuels<sup>20,23</sup>.

$$FSC = 0.232 \times \frac{\int [SO_2 - SO_{2,bkg}]_{ppb} dt}{\int [CO_2 - CO_{2,bkg}]_{ppm} dt} [\% \text{ Sulphur}] \quad (1)$$

Fixed stations and airborne sniffers have a NO cross sensitivity for the SO<sub>2</sub> sensor, therefore this formula is adapted by subtracting the measured SO<sub>2</sub> with the NO amount in the plume, multiplied by the NO cross sensitivity factor (CS<sub>NO</sub>).

$$FSC_{NO} = 0.232 \times \frac{\int [SO_2 - SO_{2,bkg}]_{ppb} - CS_{NO} \times [NO - NO_{bkg}]_{ppb} dt}{\int [CO_2 - CO_{2,bkg}]_{ppm} dt} [\% \text{ Sulphur}] \quad (2)$$

It is worth noting that the airborne measurements conducted with mini-sniffers use a similar sniffer measurement technique, but are based on more compact and less costly electrochemical sensors for SO<sub>2</sub>. As a result the measurement precisions is lower and the time response longer. Nevertheless, due to the longer duration RPAS and helicopters spent in the plume (hovering), their accuracy is higher, hence, the measurement uncertainty is comparable to the heavier sniffer sensors<sup>17,18</sup>.

#### Remote NO<sub>x</sub> measurements

A challenge in utilizing remote NO<sub>x</sub> measurements for assessing NO<sub>x</sub> non-compliance arises from the technical emission standards of the NO<sub>x</sub> Technical Code which express emission limits as weighted average NO<sub>x</sub> (in g) over brake horsepower (in kWh). The NO<sub>x</sub> emission factor (Fe<sub>no</sub>) in g NO<sub>x</sub>/kg fuel is calculated similarly to the FSC using the background and plume NO<sub>x</sub> and CO<sub>2</sub> concentrations<sup>23,25,30</sup>.

$$EF_{NO_x} = \frac{M_{NO_2} \frac{g}{mol}}{M_C \frac{g}{mol} \times 0.87} \times 1000 \times \frac{\int [NO_x - NO_{x,bkg}]_{ppm} dt}{\int [CO_2 - CO_{2,bkg}]_{ppm} dt} \left[ \frac{g}{kg \text{ fuel}} \right] \quad (3)$$

$$EF_{NO_x} = 3.33 \times \frac{\int [NO_x - NO_{x,bkg}]_{ppb} dt}{\int [CO_2 - CO_{2,bkg}]_{ppm} dt} \left[ \frac{g}{kg \text{ fuel}} \right] \quad (4)$$

For the calculation of the NO<sub>x</sub> emission in g NO<sub>x</sub>/kWh, the NO<sub>x</sub> emission factor (g NO<sub>x</sub>/kg fuel) is then multiplied by the Specific Fuel Consumption (SFC).

$$SFC = \frac{kg \text{ fuel}}{kWh \text{ BHP}} \quad (5)$$

$$EF_{P,NO_x} = EF_{NO_x} \times \left( \frac{kg \text{ fuel}}{kWh \text{ BHP}} \right) \quad (6)$$

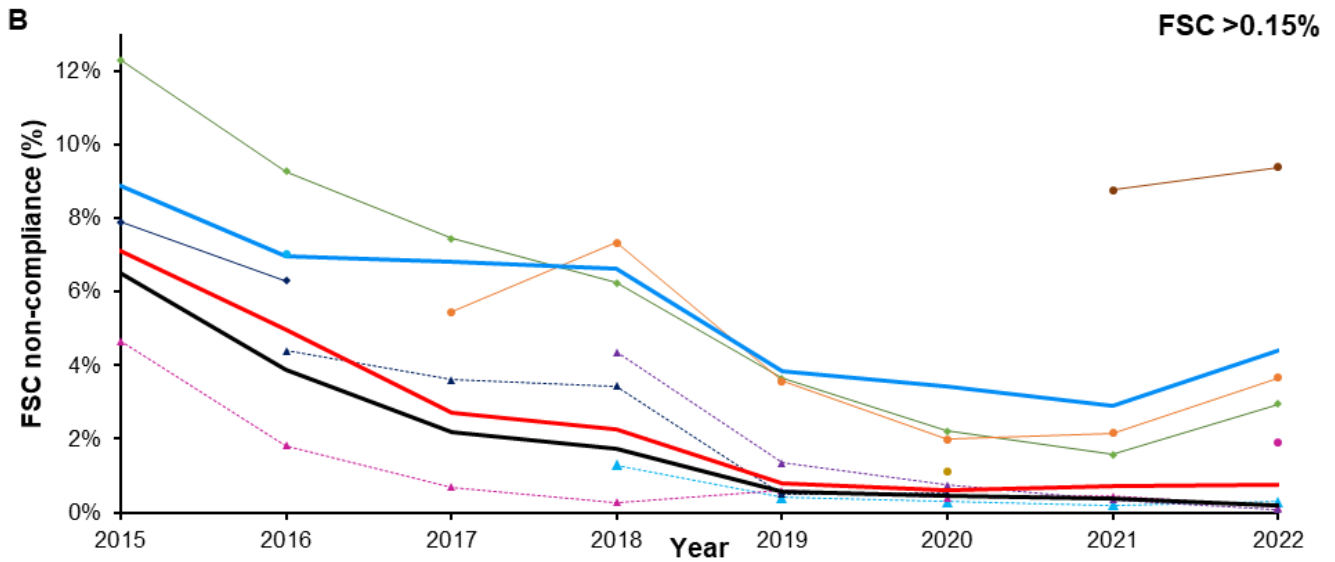
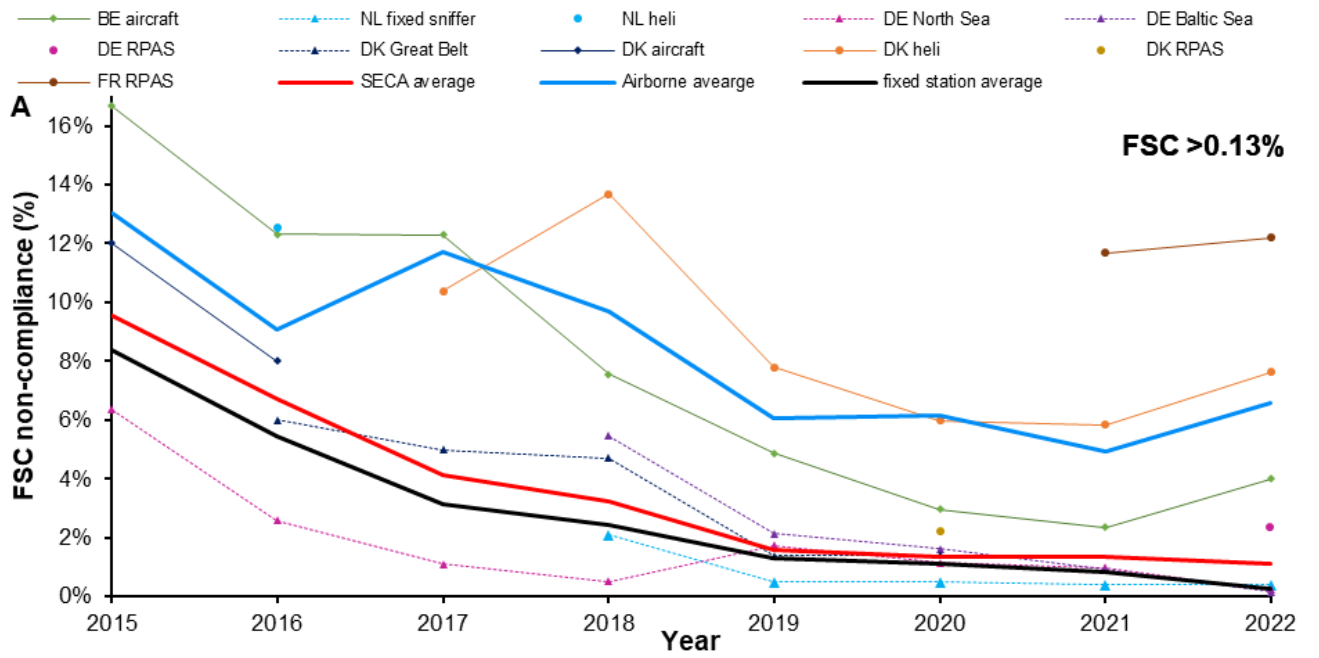
Typically, the SFC ranges from 0.16 kg/kWh to 0.24 kg/kWh<sup>21,29–35</sup>. The exact SFC can be obtained through radio communication (as done by the MUMM) or by applying a modeled approach based on voyage and OGV characteristics (as done by Explicit).

While Belgium has the ability to request information on engine parameters from the monitored OGVs, other remote monitoring agencies lack this option. As a result, these agencies face difficulties in directly assessing NO<sub>x</sub> emission levels expressed in g NO<sub>x</sub>/kWh based on remote measurements alone, highlighting the need for alternative approaches or additional data sources to accurately evaluate NO<sub>x</sub> emissions. In addition, as the emission limits for main engines are expressed as the weighted average over four engine loads, it is challenging to define non-compliance on remote measurements in particular for low exceedances of the emission limits and when OGVs operate at a low engine load (< 25-50%), due to the low weighting factor of these lower loads. Furthermore, it is worth noting that the RPAS of the European Maritime Safety Agency (EMSA) have the capacity to measure NO<sub>x</sub> emissions. However, since there is currently no specific EU Directive addressing NO<sub>x</sub> emissions from OGVs, EMSA has no legal mandate to monitor NO<sub>x</sub> emissions. As a result, the focus of EMSA's monitoring efforts remains primarily on SO<sub>2</sub> and other maritime surveillance tasks, which have established EU regulatory frameworks in place.

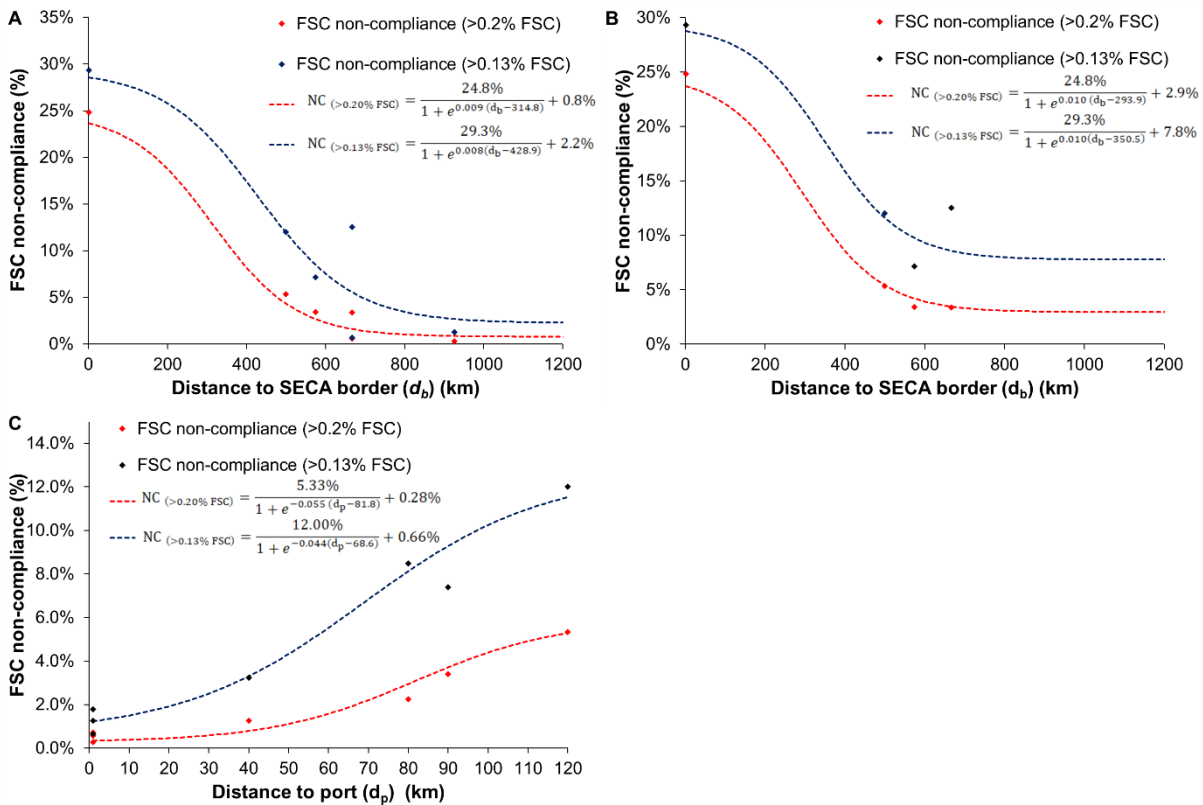
Supplementary figures



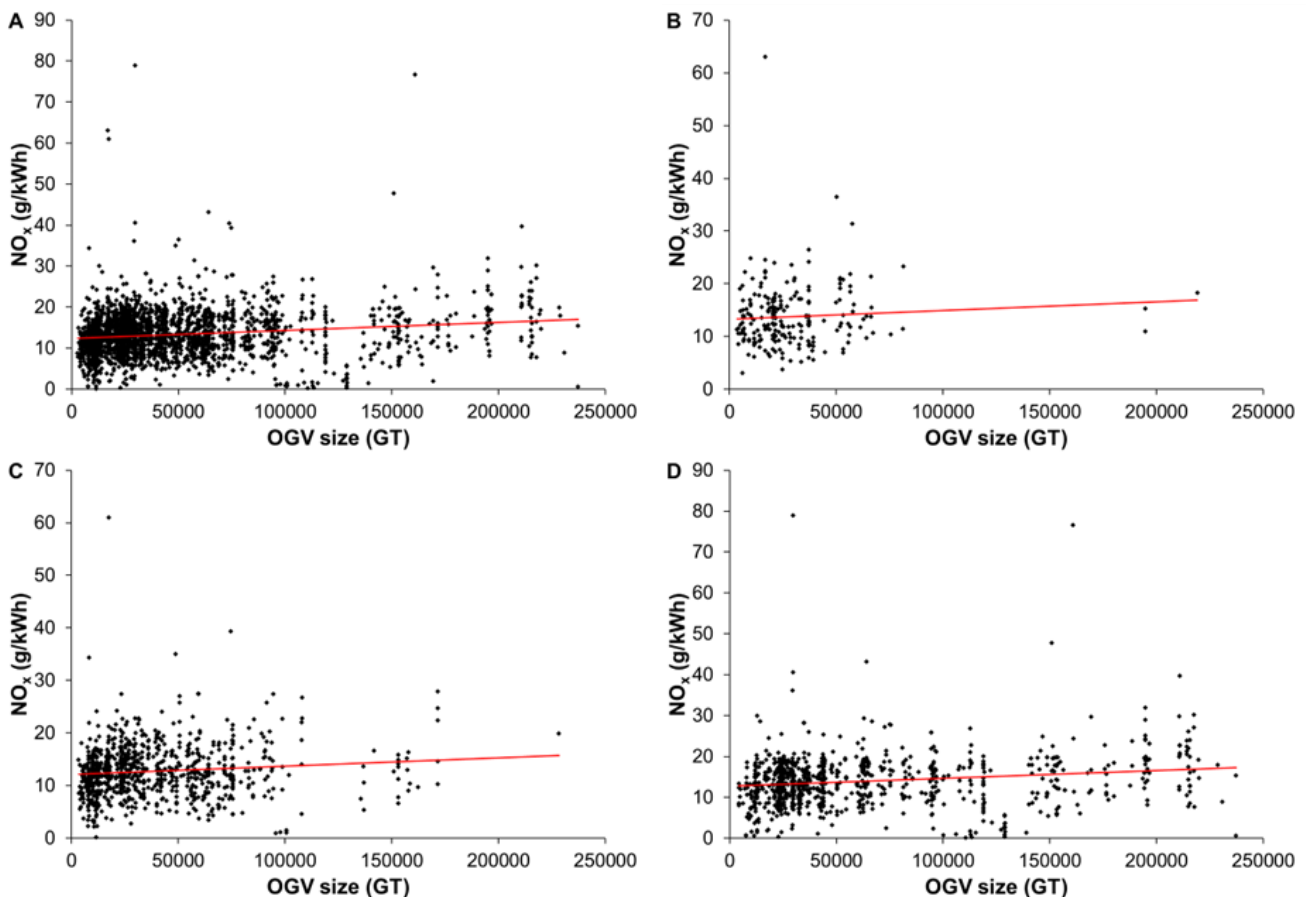
Supplementary Figure 2. The Bonn Agreement area © The Bonn Agreement <sup>36</sup>.



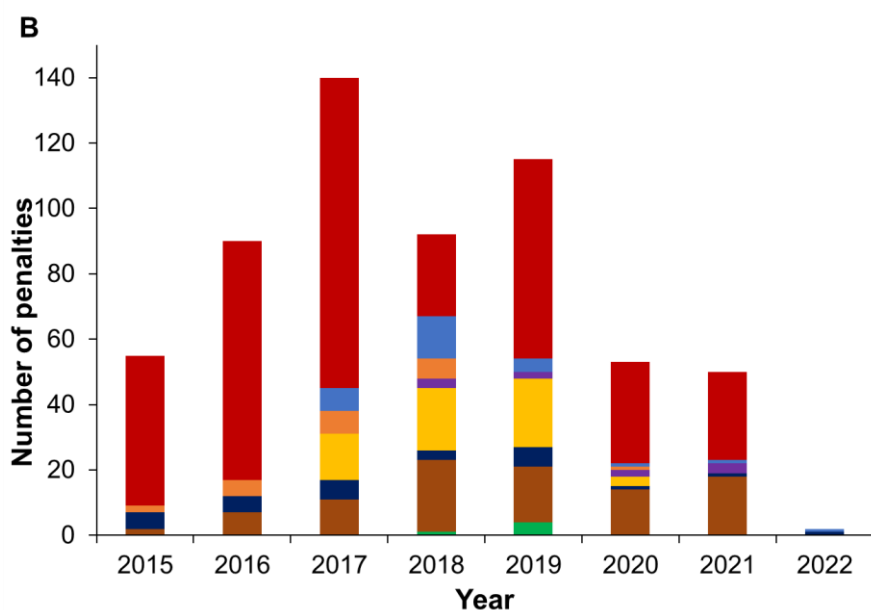
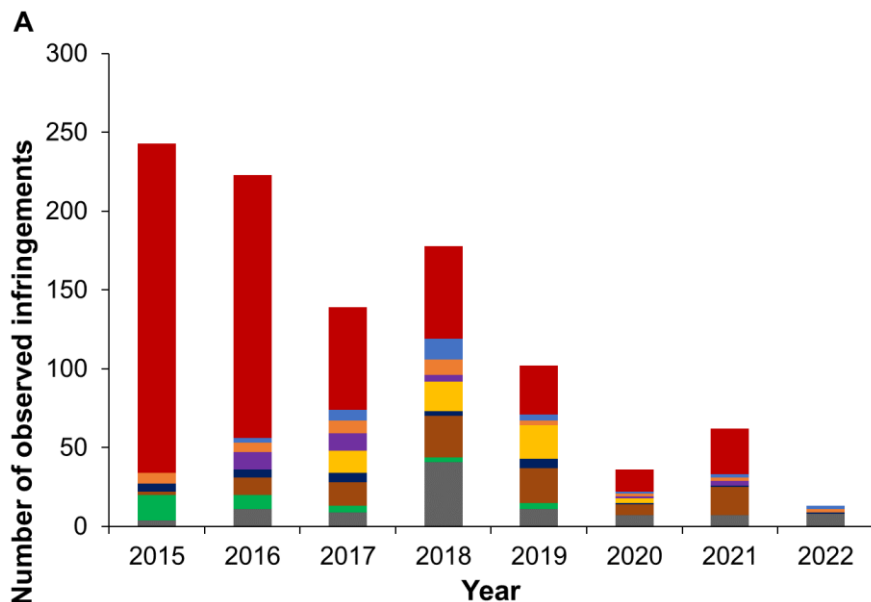
Supplementary Figure 3. FSC non-compliance for remote monitoring locations in the SECA for cut-off levels: 0.13% FSC (A) and 0.20% FSC (B). Measurements with fixed wing aircraft are displayed with full lines and diamond markers (◆), measurements using RPAS and helicopters are have full lines and circle markers (●), fixed sniffer measurements are displayed with dotted line and triangular markers (▲).



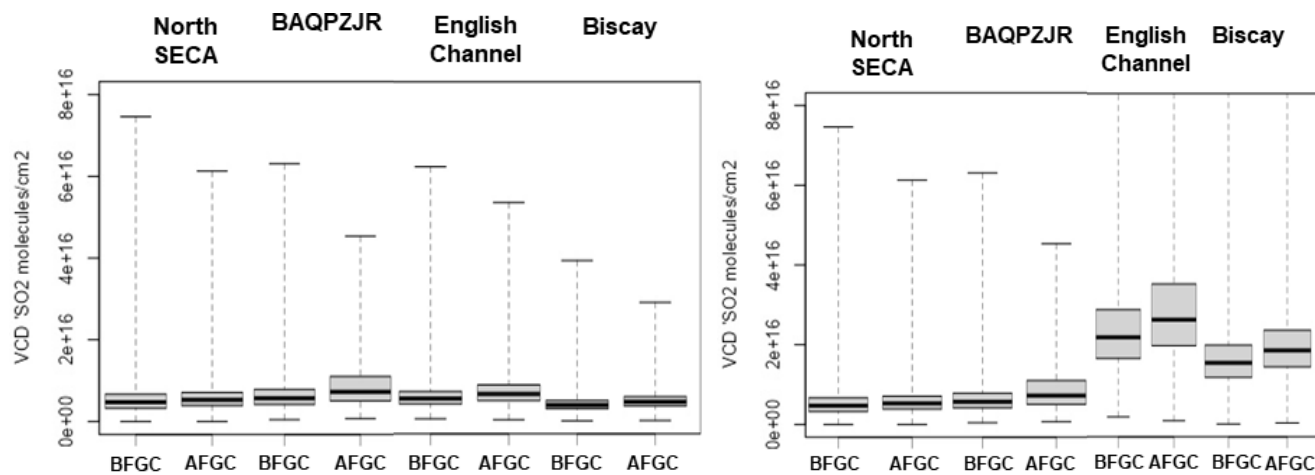
**Supplementary Figure 4. Non-compliance fitting based on the mean non-compliance of the different remote measurement locations in function of the distance to the SECA border ( $d_b$ ) for the 0.13 and 0.20% FSC cut-off levels, for the all remote measurements (A) and for when only the airborne measurements are considered (B). Non-Compliance fitting in function of the distance to port ( $d_p$ ) (C).**



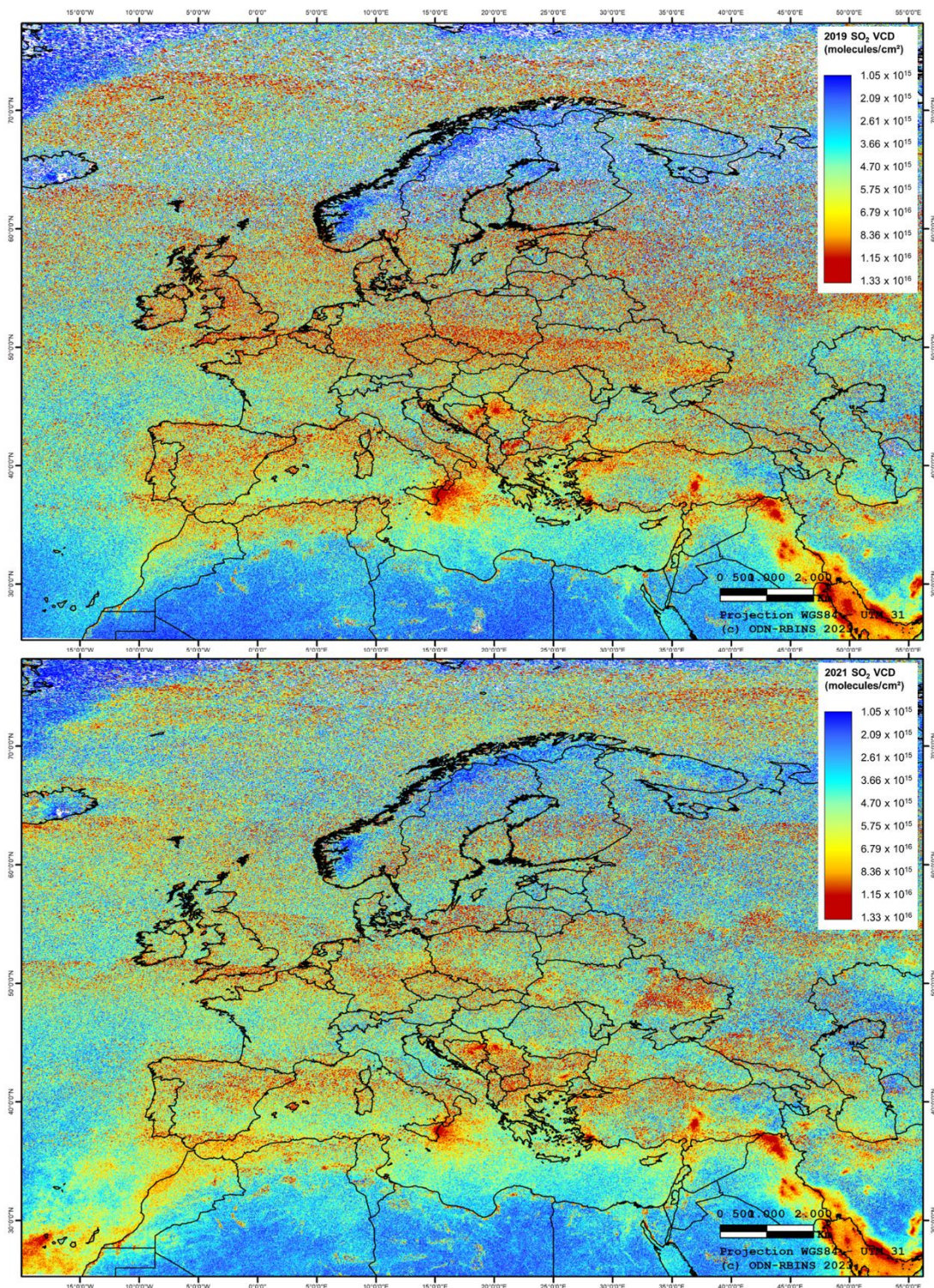
**Supplementary Figure 5. Results of the Belgium remote NO<sub>x</sub> emission expressed in function of the OGV size in gross tonnage (GT), for all measured OGVs (A), for only the Tier 0 OGVs (B), Tier I OGVs (C) and Tier II (OGVs).**



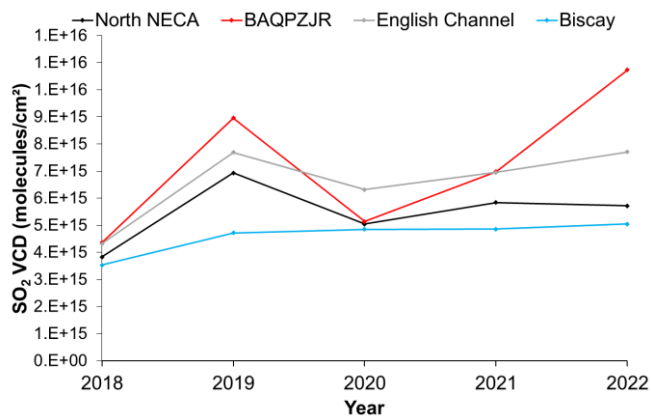
Supplementary Figure 6. Number of reported sulfur infringements (A) and penalties (B) for the BA CPs. The nationality of the BA CPs is not released on request of certain BA CPs.



Supplementary Figure 7. Box plot of SO<sub>2</sub> (VCD) between different areas before the global cap (2018-2019) and after the global cap came into effect (2021-2022), with minimum, 25% percentile, median, 75% percentile and maximum. The four upper graphs include the maximum value, while the four graphs at the bottom demonstrate a zoom on the 25% and 75% percentiles.



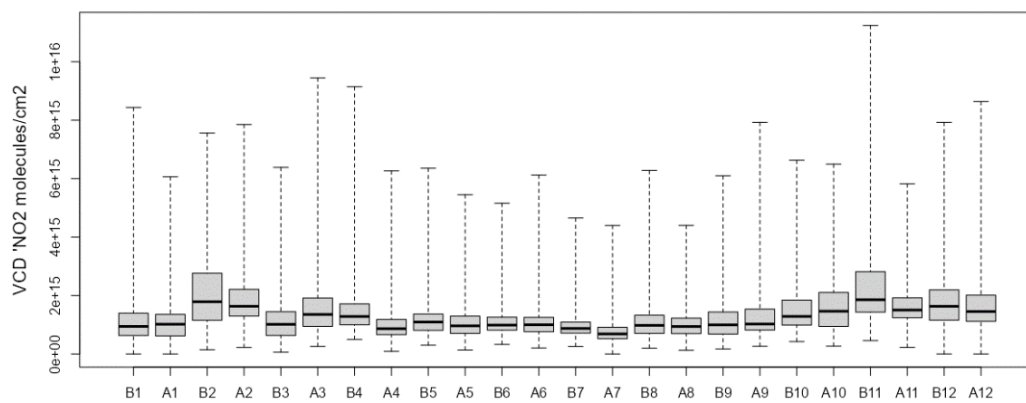
Supplementary Figure 8. TROPOMI Sentinel 5 combined SO<sub>2</sub> VCD maps for 2019 and 2021 (right).



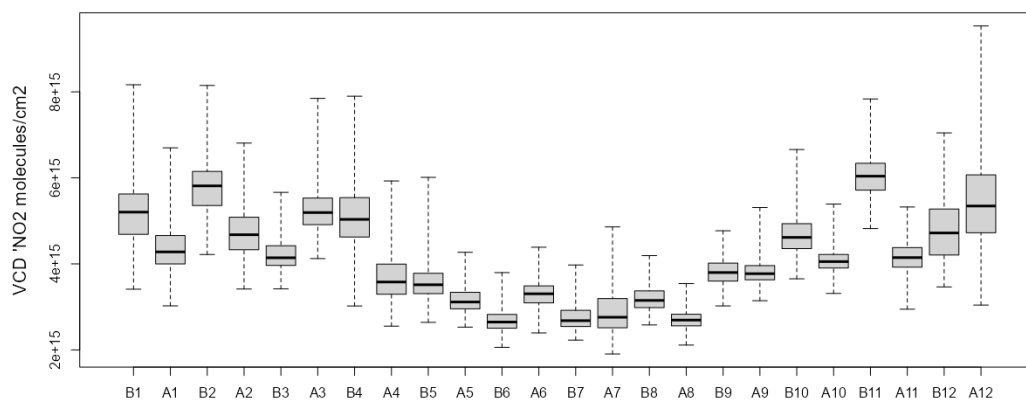
Supplementary Figure 9. SO<sub>2</sub> mean VCD levels from 2018-2022. For 2018 and 2022, not all months were available.



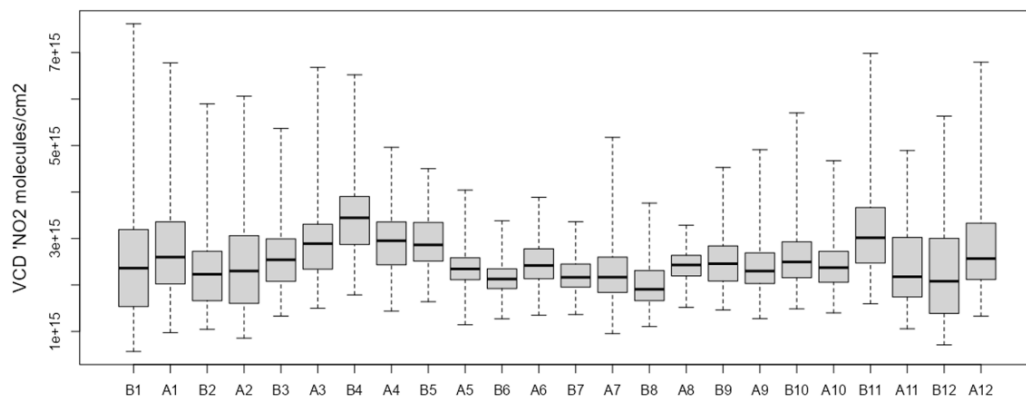
### North NECA



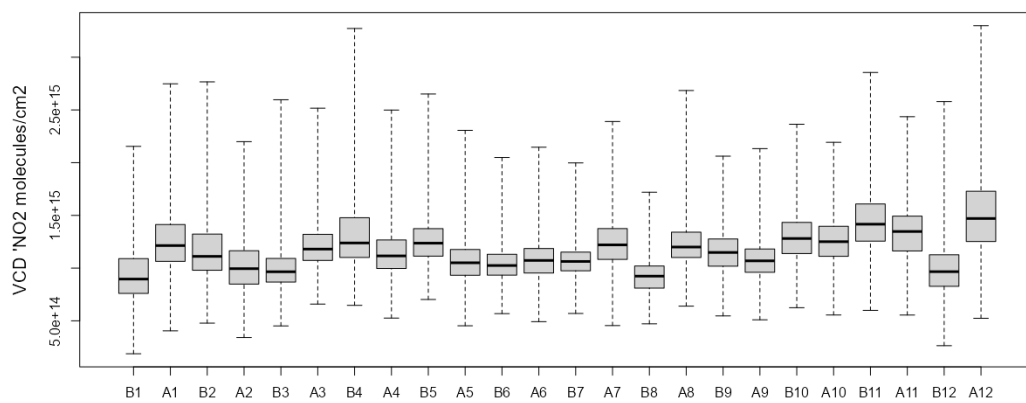
### BAQPZJR



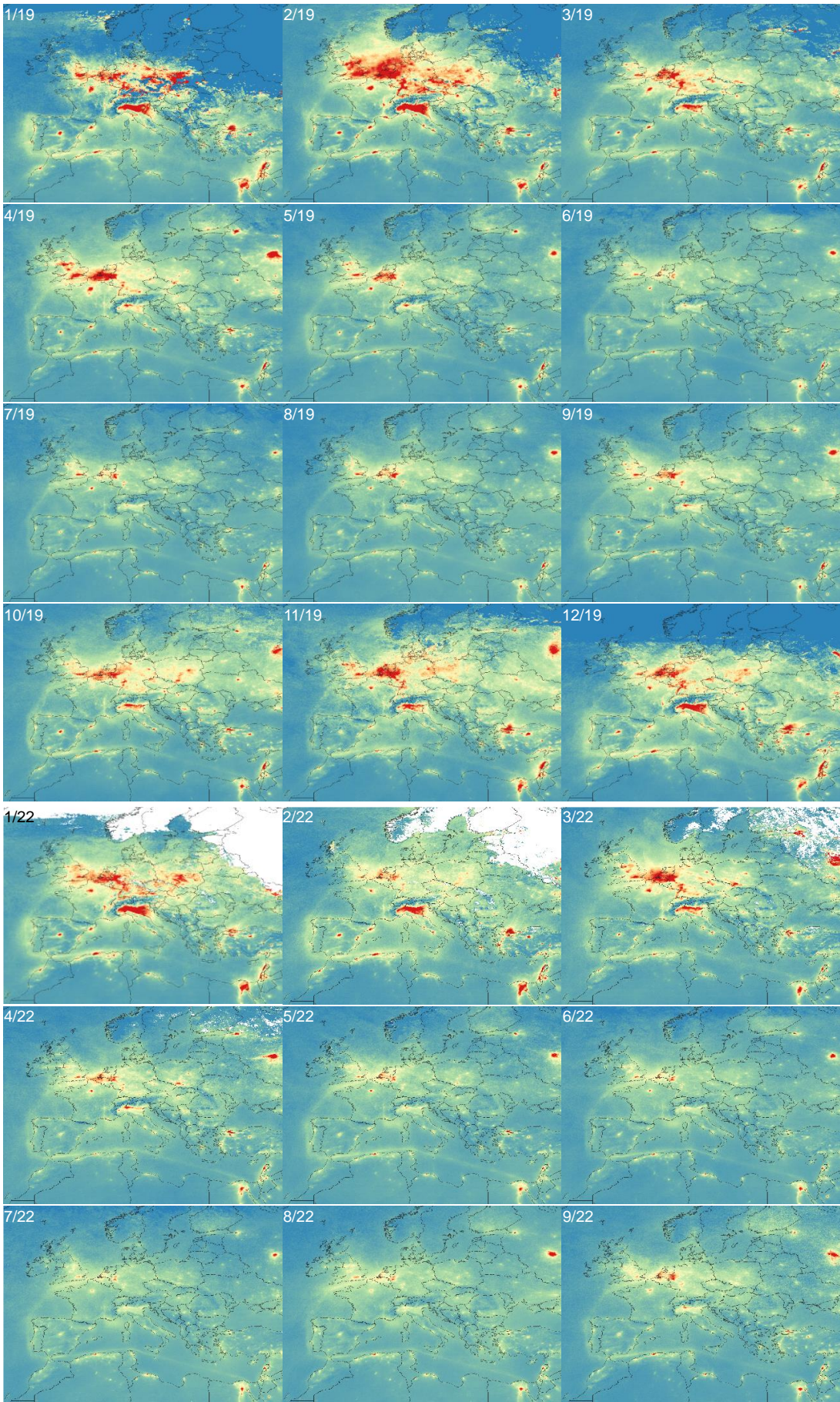
### English Channel

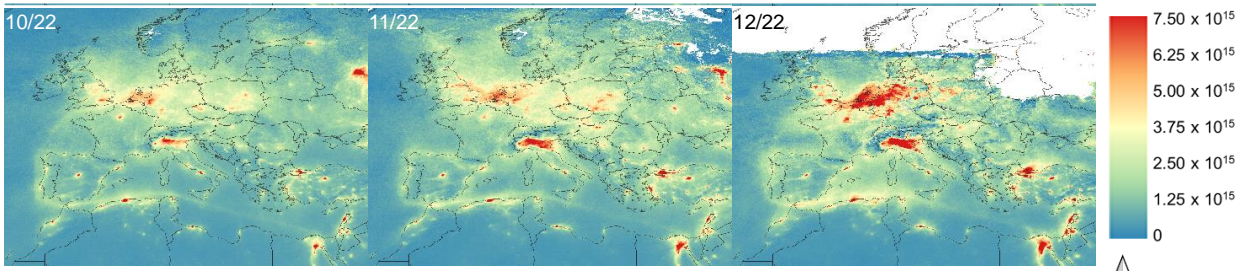


### Biscay

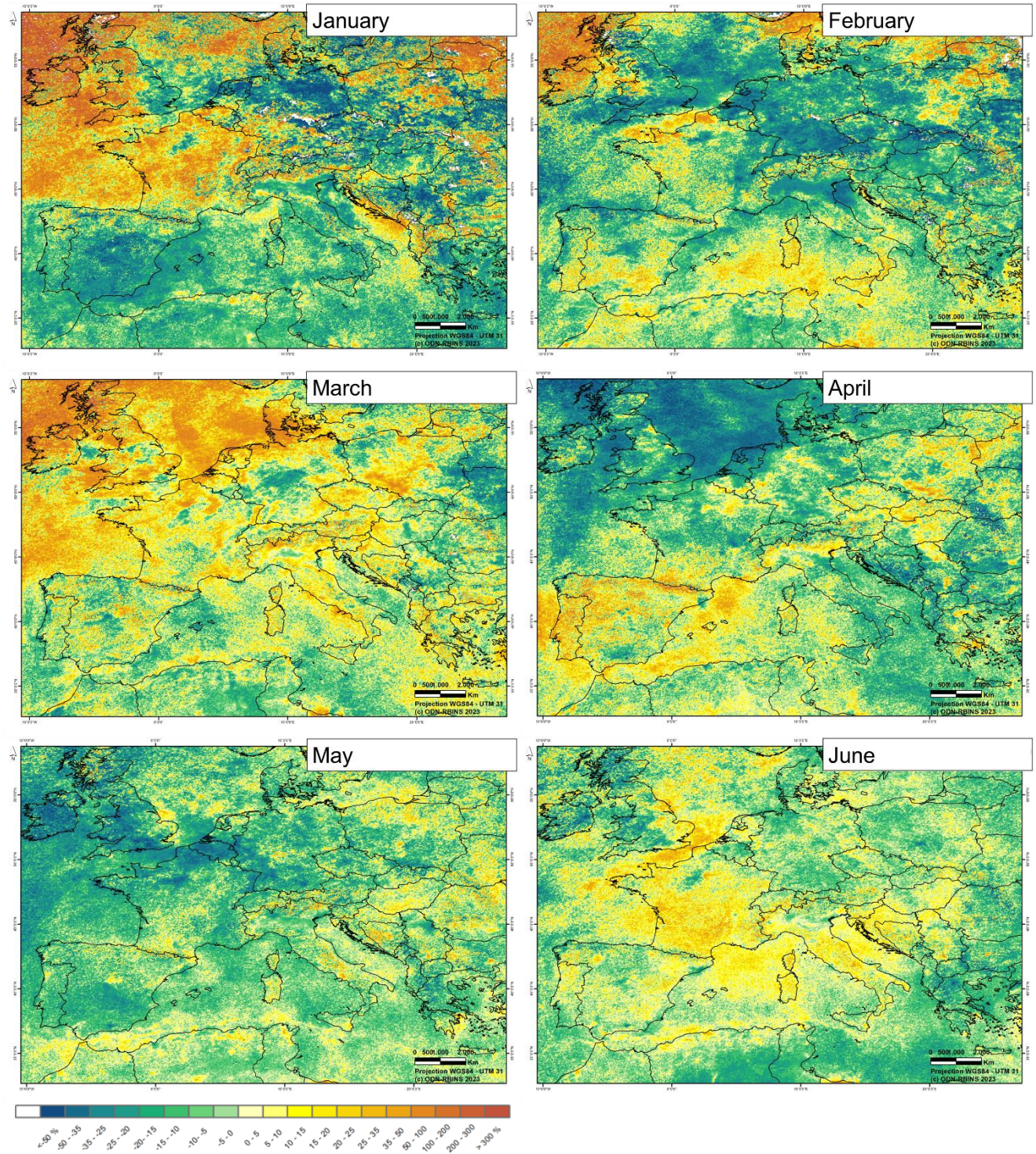


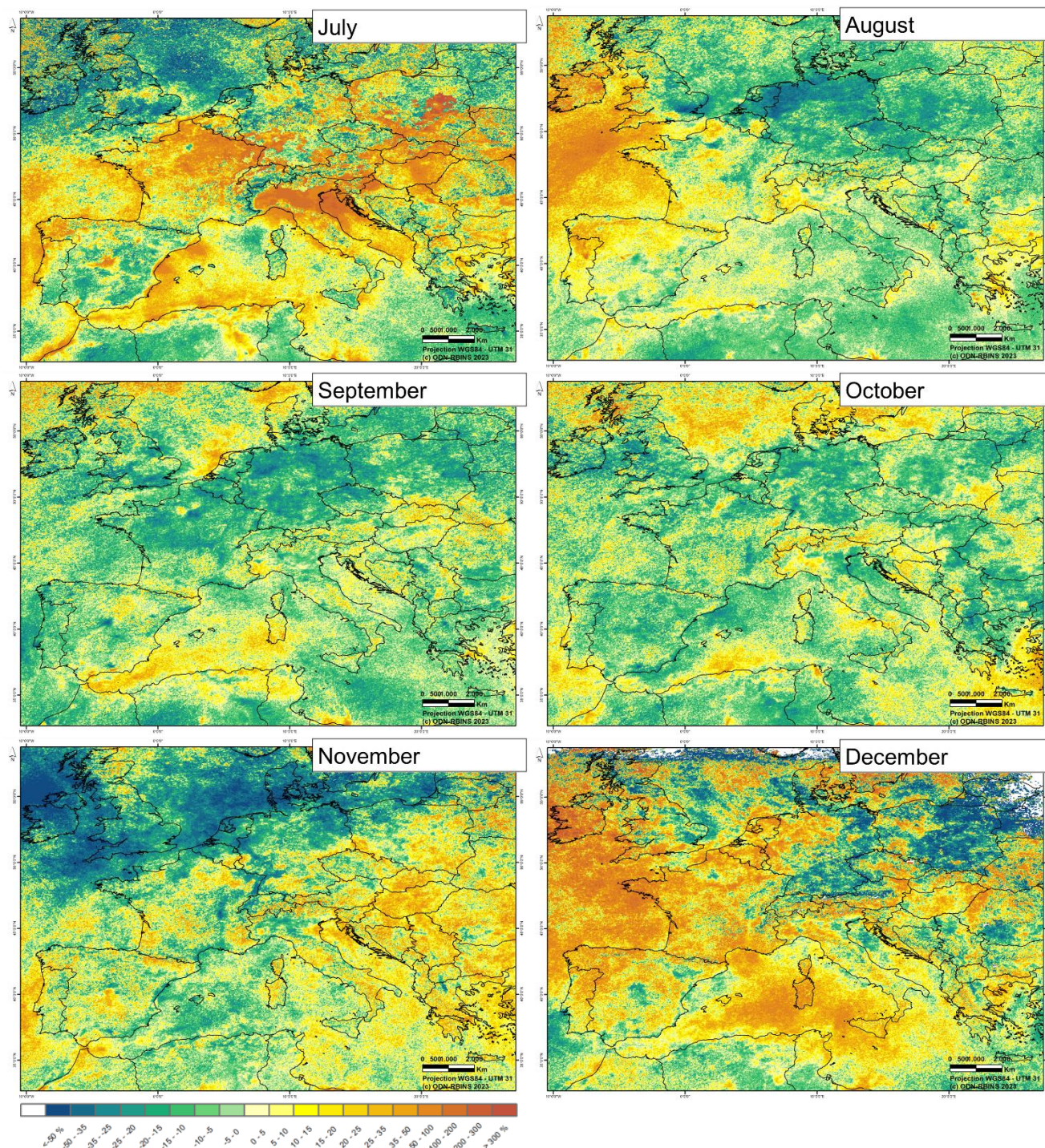
**Supplementary Figure 10. Boxplots of the NO<sub>2</sub> VCD for four zones in the ECA per month before (B#) and after (A#) the NECA came into force, with minimum, 25% percentile, median, 75% percentile and maximum.**



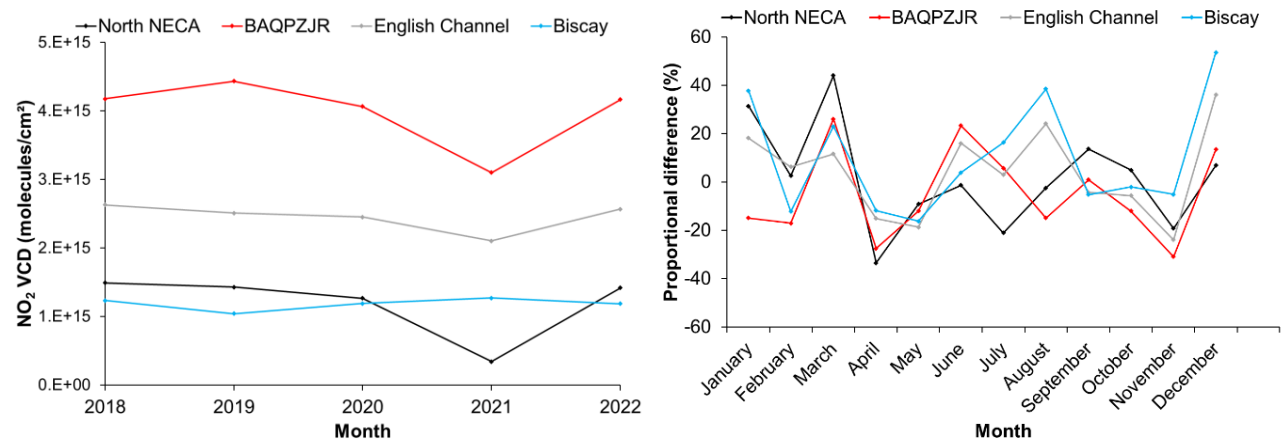


Supplementary Figure 11. Time series of NO<sub>2</sub> VCD per month 2022 based on TROPOMI Sentinel 5<sub>A</sub>

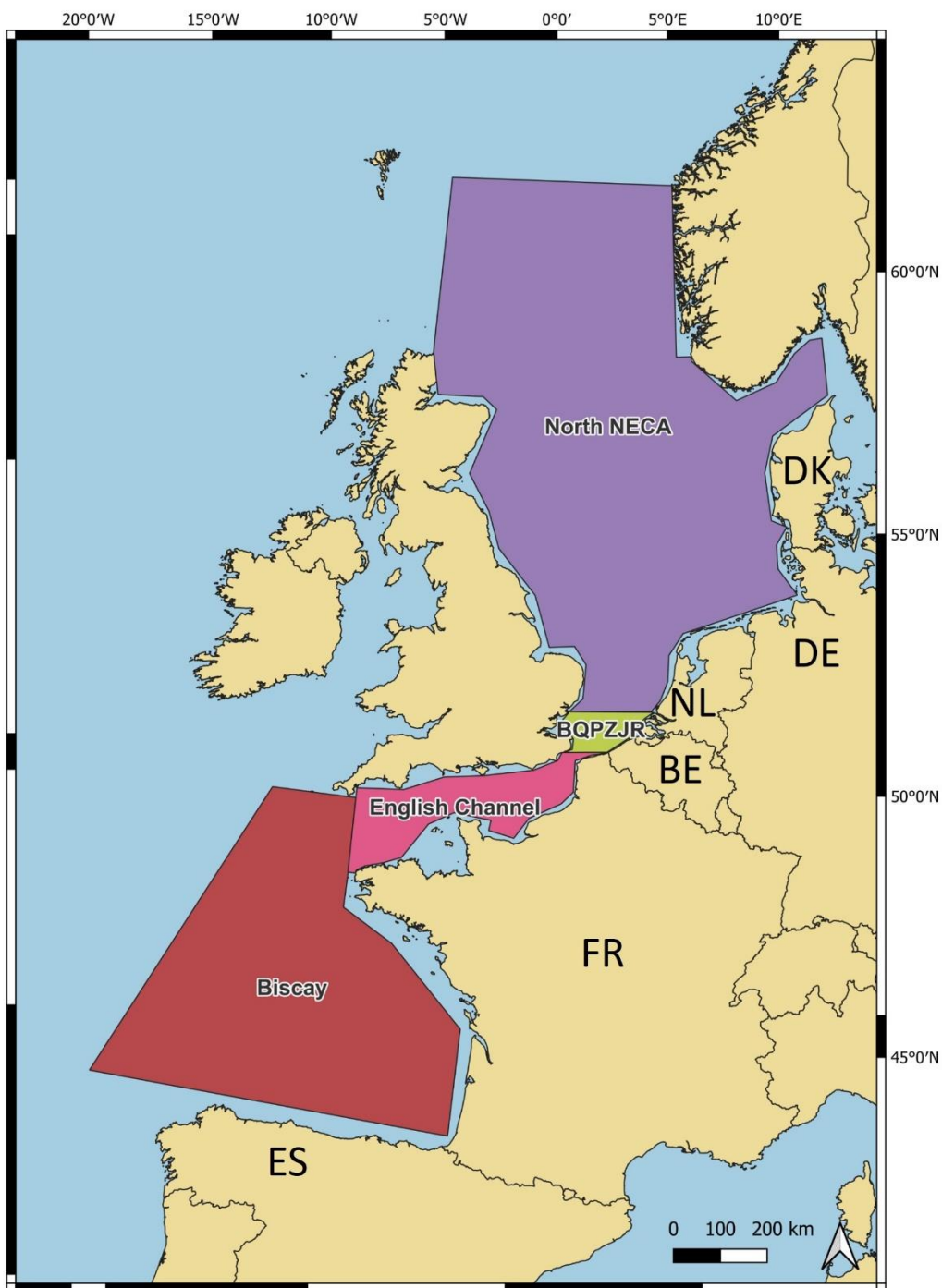




Supplementary Figure 12. Difference (expressed in %) in the average monthly NO<sub>2</sub> VCD from January until December between the period before (2018-2020) and after the implementation of the NECA (2021-2022).



Supplementary Figure 13. Average yearly NO<sub>2</sub> VCD per zone (A). Average proportional difference per month per zone after the NECA came into effect (B). For 2018 not all months were available.



Supplementary Figure 14. Definition of the areas used for the spatial analysis of emission levels for SO<sub>2</sub> and NO<sub>2</sub>. The purple area is the Northern ECA; the light gray area is the BAQPZJR; below in dark gray is the English Channel; the brown area is the Bay of Biscay used as reference outside the ECA.

## Supplementary tables

**Supplementary Table 2. Overview of remotely monitored OGVs for FSC in the SECA from 2015-2020 (note that the DE fixed data for the North Sea covers data from 2 stations).**

	BE AC	NL fixed	NL heli	DE fixed North Sea	DE fixed Baltic Sea	DE RPAS	DK heli	DK RPAS	DK fixed Great Belt	DK AC	FR RPAS	LT RPAS	Sum SECA
2015	114	70		1479						396			2059
2016	1219	199	327	2540					1691	853			6829
2017	887	147		5643			404		4155				11236
2018	1138	2958		6088	1557		614		3580				15935
2019	1232	8765		8815	2601		615	26	5458				27512
2020	405	4958		7713	2401		602	180	3910		67		20236
2021	1015	3343		6684	2506		600	3			308	142	14601
2022	951	4975		7103	2541	214	602				480		16866
Total	6961	25415	327	46065	11606	214	3437	209	18794	1249	855	142	115274

**Supplementary Table 3. Correlation ( $R^2$ ) between different remote measurement locations.**

	BE aircraft	DK mini sniffers	NL Rotterdam	DE combined	DK Great Belt	SECA fixed	SECA airborne
BE aircraft	-	0.76	0.89	0.64	0.90	0.91	-
DK mini sniffers	0.76	-	0.29	0.11	0.64	0.48	
NL Rotterdam	0.89	0.29	-	0.72	0.68	-	0.70
DE (combined)	0.64	0.11	0.72	-	0.35	-	0.51
DK fixed	0.90	0.64	0.68	0.35	-	-	0.84

**Supplementary Table 4. Correlation ( $R^2$ ) between Belgian airborne data and other remote measurement locations.**

Location	FSC>0.13	FSC>0.15	FSC>0.2	Mean
NL Rotterdam	0.91	0.91	0.85	0.89
DE (combined)	0.57	0.70	0.64	0.64
DK heli	0.51	0.78	0.62	0.64
DK Great Belt	0.85	0.93	0.93	0.90
DK Mini-sniffers	0.70	0.73	0.84	0.76
SECA average (excl. BE)	0.91	0.95	0.90	0.92

**Supplementary Table 5. Constants and correlation factor for the fitting of the non-compliance rate in function of the distance to the SECA border (nm) based on the three non-compliance cut-off levels.**

	All measurements			Airborne sniffers and mini-sniffers		
	FSC > 0.2%	FSC > 0.15%	FSC > 0.13%	FSC > 0.2%	FSC > 0.15%	FSC > 0.13
$R^2$	0.972	0.924	0.794	0.997	0.972	0.883
$k$	24.8%	29.2%	29.3%	24.8%	29.2%	29.3%
$o$	-0.009	-0.010	-0.008	-0.010	-0.009	-0.010
$m$	314.8	424.2	428.9	293.9	343.0	350.5
$p$	0.8%	1.4%	2.2%	2.9%	5.1%	7.8%

**Supplementary Table 6. Constants and correlation factors for the fitting of the non-compliance in function of the distance to port (nm) for the three non-compliance cut-off levels.**

	FSC > 0.2%	FSC > 0.15%	FSC > 0.13
$R^2$	0.963	0.969	0.962
$k$	5.33%	10.92%	12.00%
$o$	0.055	0.070	0.044
$m$	81.8	88.3	68.6
$p$	0.28%	0.64%	0.66%

**Supplementary Table 7. Proportional difference for the different zones after the implementation of regulatory measures.**

	SO <sub>2</sub> annual difference (%)	NO <sub>2</sub> annual difference (%)	NO <sub>2</sub> monthly difference (%)
Northern SECA	-15.9%	+1.0%	+7.15%
BAQPZJR	-22.5%	-5.8%	+6.2%
English channel	-9.5%	+4.1%	+5.86%
Bay of Biscay	+3.0%	+14.4%	+9.59%

## Supplementary references

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6. *Amendments to the Annex of the Protocol of 1997 to amend the International Convention for the Prevention of Pollution from Ships, 1973, as modified by the Protocol of 1978 relating thereto (Prohibition on the carriage of non-compliant fuel oil for combustion purposes for propulsion or operation on board a ship), Resolution MEPC.305(73), Adopted on 26 October 2018, Entered into Force on 1 March 2020.*
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