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## Ships going slow in reducing their NO<sub>x</sub> emissions: changes in 2005–2012 ship exhaust inferred from satellite measurements over Europe

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### Abstract

We address the lack of temporal information on ship emissions, and report on rapid short-term variations of satellite-derived ship NO<sub>x</sub> emissions between 2005 and 2012 over European seas. Our inversion is based on OMI observed tropospheric NO<sub>2</sub> columns and GEOS-Chem simulations.

Average European ship NO<sub>x</sub> emissions increased by ~15% from 2005 to 2008. This increase was followed by a reduction of ~12% in 2009, a direct result of the global economic downturn in 2008–2009, and steady emissions from 2009 to 2012. Observations of ship passages through the Suez Canal and satellite altimeter derived ship densities suggests that ships in the Mediterranean Sea have reduced their speed by more than 30% since 2008. This reduction in ship speed is accompanied by a persistent 45% reduction of average, per ship NO<sub>x</sub> emission factors. Our results indicate that the practice of ‘slow steaming’, i.e. the lowering of vessel speed to reduce fuel consumption, has indeed been implemented since 2008, and can be detected from space. In spite of the implementation of slow steaming, one in seven of all NO<sub>x</sub> molecules emitted in Europe in 2012 originated from the shipping sector, up from one in nine in 2005. The growing share of the shipping contributions to the overall European NO<sub>x</sub> emissions suggests a need for the shipping sector to implement additional measures to reduce pollutant emissions at rates that are achieved by the road transport and energy producing sectors in Europe.

### 1. Introduction

Current emission inventories suggest ship NO<sub>x</sub> (NO<sub>x</sub>=NO+NO<sub>2</sub>) emissions account for approximately 15% (3.0–10.4 Tg N yr<sup>-1</sup>) of total anthropogenic NO<sub>x</sub> emissions (e.g. Corbett *et al* 2007, Paxian *et al* 2010, IMO 2014). NO<sub>x</sub> emissions lead to tropospheric ozone (O<sub>3</sub>) production and aerosol formation, both of which deteriorate air quality and influence climate change. Over the past decade, numerous studies have used satellite NO<sub>2</sub> observations to constrain various NO<sub>x</sub> emission sources, like those from anthropogenic (e.g. Martin *et al* 2006, Stavrakou *et al* 2013) and soil activity (e.g. Jaegle *et al* 2005, Vinken *et al* 2014a), or from biomass burning (e.g.

Mebust *et al* 2011, Castellanos *et al* 2014) and lightning (e.g. Boersma *et al* 2005, Bucsela *et al* 2010). However, few researchers have yet attempted to reduce the substantial uncertainties in the shipping sector NO<sub>x</sub> emission inventories by space-based observations of NO<sub>2</sub>. Bottom-up uncertainties arise from the extrapolation of only a few measurements and assumptions about important emission drivers, such as fuel consumption, and the number and velocity of ships. More recent AIS data-based ship emission estimates use highly accurate information on ship position and speed, but contend with uncertainties on ship engine speed and power, and depend on the availability of AIS data and up-to-date information on the vessels (e.g. Jalkanen *et al* 2009). Furthermore, existing spatially-

resolved emission inventories often cover only one year per decade, and miss the considerable year-to-year increases in ship emissions due to increased world trade over the past decade (estimated at 5% per year (Eyring *et al* 2005)). Such temporal changes, however, are covered in the IMO reports (IMO 2009, 2014). According to Faber *et al* (2012) and IMO (2014), and references therein, fuel consumption and emissions decreased sharply in response to the economic downturn in 2008–2009, but as of yet, this decrease has not been confirmed by independent observations on ship emissions or on ship speed. Here we document rapid changes in ship NO<sub>x</sub> emissions inferred from space in a period of economic turmoil. This will improve our understanding of what drives these changes over areas where air quality is currently not monitored by other methods.

Busy ship lanes have been identified using satellite observations of tropospheric NO<sub>2</sub> columns. Over the Indian Ocean, Beirle *et al* (2004) demonstrated that enhanced pollution levels could be detected along well-known shipping lanes in maps of tropospheric NO<sub>2</sub> columns from the Global Ozone Monitoring Experiment (GOME) instrument. Several more ship tracks have been identified over the Red Sea and China Sea using the SCanning Imaging Absorption spectrometer for Atmospheric CHartographY (SCIAMACHY) (Richter *et al* 2004). Recently, Vinken *et al* (2014b) used NO<sub>2</sub> observations from the Ozone Monitoring Instrument (OMI) to constrain European ship emissions for 2005 and 2006 in ship lanes in the Baltic Sea, the North Sea, the Bay of Biscay, and in the Mediterranean Sea. The temporal evolution of OMI tropospheric NO<sub>2</sub> columns over the Baltic Sea was also shown by Ialongo *et al* (2014). These studies demonstrate that satellite instruments are able to observe localised NO<sub>2</sub> pollution over ship lanes, and can provide worldwide, robust observations that are useful to provide constraints on these emissions over long time periods.

Several papers reported on trends in tropospheric NO<sub>2</sub> columns from satellite data sets (e.g. Richter *et al* 2005, van der A *et al* 2006). In Europe, Castellanos and Boersma (2012) found reductions in OMI NO<sub>2</sub> columns of at least 20% between 2004 and 2010. These reductions are the result of both the 2008–2009 global economic recession and tighter European NO<sub>x</sub> emission controls. Also, de Ruyter de Wildt *et al* (2012) found signatures of the economic recession in satellite NO<sub>2</sub> measurements over major ship tracks in Europe and Asia. Here we infer NO<sub>x</sub> emissions from the shipping sector over European seas between 2005 and 2012. These variations are calculated by a mass-balance approach using OMI satellite observed NO<sub>2</sub> columns and GEOS-Chem simulations over the Baltic Sea, the North Sea, the Bay of Biscay and the Mediterranean Sea. The derived temporal variations are compared to shipping statistics and trade volumes to better understand the relationship between ship

emissions and economic activity. Furthermore, we study how ship NO<sub>x</sub> emissions change as the shipping sector implemented a new operational practice in response to the economic downturn in 2009 (Corbett *et al* 2009).

## 2. Method

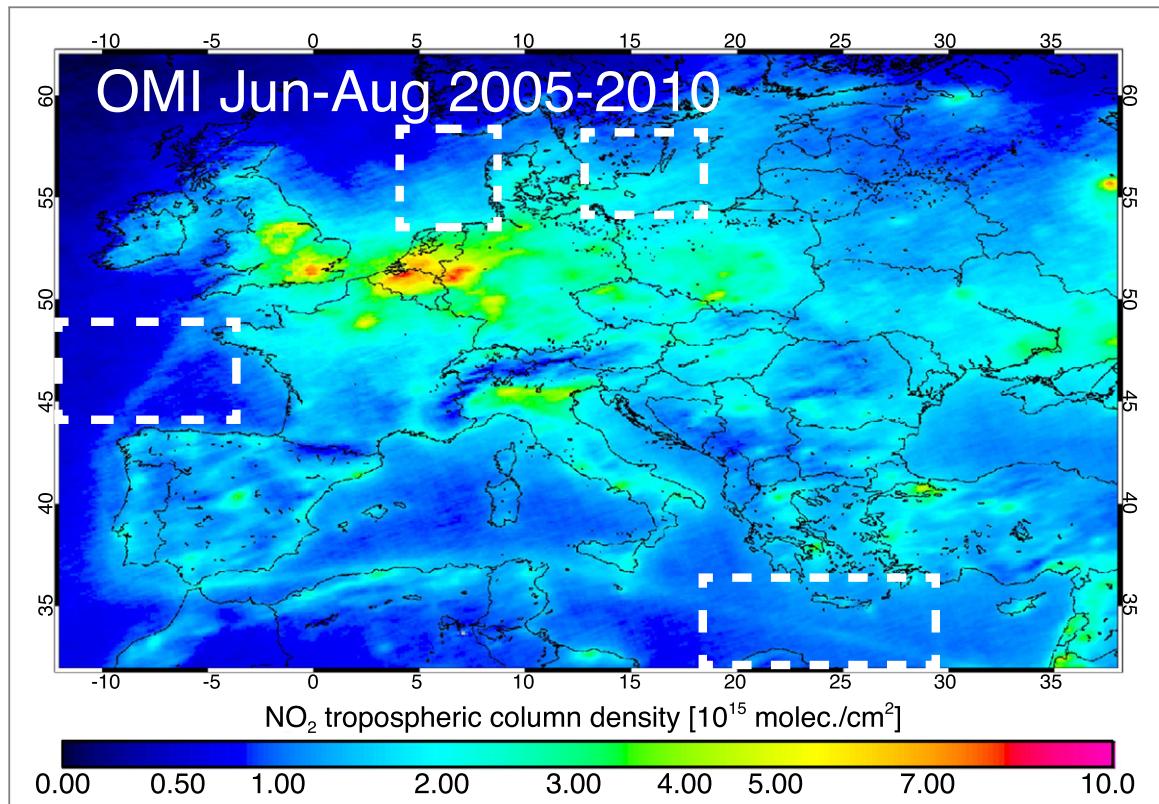
### 2.1. Satellite observations

We use observed tropospheric NO<sub>2</sub> columns from the Dutch-Finnish Ozone Monitoring Instrument (OMI). OMI provides daily worldwide measurements, and has a spatial resolution as small as  $13 \times 24 \text{ km}^2$  for nadir pixels. We use retrieved NO<sub>2</sub> column densities from the Dutch OMI tropospheric NO<sub>2</sub> (DOMINO) v2.0 product (Boersma *et al* 2011). The error in individual OMI observations is estimated to be  $1.0 \times 10^{15}$  molecules  $\text{cm}^{-2} + 25\%$  (Boersma *et al* 2011). The DOMINO v2.0 NO<sub>2</sub> retrieval has been validated with in-situ Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements (Irie *et al* 2012, Ma *et al* 2013). Several recent studies have used the DOMINO v2.0 retrieval to constrain NO<sub>x</sub> emissions (e.g. Stavrakou *et al* 2013, McLinden *et al* 2014, Vinken *et al* 2014b).

Figure 1 shows an average map of OMI tropospheric NO<sub>2</sub> columns for June–August, 2005–2010. Pollution from a mixture of several sources (e.g. traffic and industry) can be observed over land, but over remote seas ship emissions can directly be identified. The map shows clear enhancements over ship tracks in the Bay of Biscay and the Mediterranean Sea. NO<sub>2</sub> pollution from ships in the North Sea and the Baltic Sea is not immediately distinguishable in this all-measurement average map of tropospheric NO<sub>2</sub> columns. To optimize the detection of ship tracks in the North Sea and the Baltic Sea, we use a filter technique to exclude observations which: (1) are influenced by strong outflow of pollution from land; (2) cover the area of the ship track only to limited extend (less than 75%, caused by cloud cover); or (3) have strong negative values (lower than  $-0.2 \times 10^{15}$  molecules  $\text{cm}^{-2}$ ).

In this study retrieval errors are minimized by excluding clouded, snow, or ice covered pixels, and we only use observations with a cloud radiance fraction below 0.5, and a surface albedo below 0.2. Furthermore, we remove the outer 2 (large) pixels on each side of the swath to reduce spatial smearing. We re-gridded the OMI NO<sub>2</sub> observations onto the GEOS-Chem nested horizontal grid ( $1/2^\circ \times 2/3^\circ$ ), and require that over 75% of a grid cell is covered by OMI observations, with more than 3 valid observations per seasonal or yearly average in each grid cell.

Reports in the peer-reviewed literature (e.g. Corbett *et al* (2009)) and from the shipping sector indicate that shipping companies started adopting ‘slow steaming’ from the year 2008 onwards, when companies lowered the speed of ships to reduce fuel usage and



**Figure 1.** OMI-observed tropospheric NO<sub>2</sub> columns (clear-sky) for June–July–August averaged from 2005 to 2010 on a  $0.1^\circ \times 0.1^\circ$  resolution. The dashed rectangles indicate the areas for the Baltic, North Sea, eastern Atlantic, and the Mediterranean Sea where ship NO<sub>x</sub> emissions are inferred from OMI and GEOS-Chem NO<sub>2</sub> columns.

overcapacity (Bonney 2010, Rodrigue *et al* 2013). This is the first self-imposed emission reduction regime for an international industry sector, and is highlighted in the last IPCC report as a ‘model for future international cooperation for other sectors’ (Sims *et al* 2014). Combining the OMI-inferred ship NO<sub>x</sub> emissions with information on the number of ships in a sea provides information on the average emission factor per ship, which should have decreased as a result of adopting slow steaming. In this study, we derive the ship density (number of ships in a pre-defined maritime area) from observations of satellite-born altimeters as recently reported by Tournadre (2014). The detection method is based on the detectable signature in the portion of the echo waveform above the sea of high-resolution satellite altimeter waveforms (Tournadre 2007). These waveforms are constructed by measuring the backscattered power of a nadir looking radar by the sea surface (or ship) as a function of time. Tournadre *et al* (2012) demonstrated that this method could be used to detect the distribution of small icebergs in the Southern Ocean, and Tournadre (2014) recently combined observations from seven altimeter satellite instruments to present a 2-decade database of ship densities for the global oceans. The altimeter data are representative for the density of all ships (commercial vessels, ferries, cruise and military ships) in a particular stretch of the world’s seas, but over narrow shipping lanes the altimeter data are

probably more representative for those ships that travel along these lanes (e.g. tankers, containerships). Information on the speed of ships in the Mediterranean Sea ( $v_{\text{ship}}$ ) can be derived (see appendix A) by:

$$v_{\text{ship}} \propto \frac{F_{\text{in}}}{C_{\text{ship}}} \quad (1)$$

where  $F_{\text{in}}$  represents the inflow of ships in the Mediterranean Sea (i.e. ships passing through the Suez Canal (Suez Canal Authority 2014)), and  $C_{\text{ship}}$  is the ship density (number of ships per unit area) detected by the altimeter instruments over the Mediterranean Sea (Tournadre 2014).

## 2.2. Estimating ship NO<sub>x</sub> emissions from OMI satellite observations

We use a mass-balance approach to estimate ship NO<sub>x</sub> emissions using the local ratio of OMI NO<sub>2</sub> observations to NO<sub>2</sub> columns simulated with the GEOS-Chem chemistry transport model (appendix B). We do this for 4 areas characterized by a distinct enhancement in tropospheric NO<sub>2</sub> as indicated by the dashed rectangles in figure 1. These areas are similar to those used in Vinken *et al* (2014b). We first simulate tropospheric NO<sub>2</sub> columns with the GEOS-Chem model for the period 2005–2012 with fixed, year 2005, ship NO<sub>x</sub> emissions from Vinken *et al* (2014b) as prior. We then evaluate the GEOS-Chem NO<sub>2</sub> columns with the OMI-observed NO<sub>2</sub> columns for the years 2005–2012 and obtain a scaling factor for the

prior NO<sub>x</sub> emission inventory from the ratio of observed-to-simulated NO<sub>2</sub> columns. In the second step, we simulate NO<sub>2</sub> columns with GEOS-Chem with the scaled-up prior inventory. From the difference between the initial and scaled runs we calculate the sensitivity of the simulated NO<sub>2</sub> column to local NO<sub>x</sub> emissions changes. This sensitivity, the  $\beta$ -factor (Lamsal *et al* 2011), accounts for the non-linear response of NO<sub>2</sub> columns to changing NO<sub>x</sub> emissions, and details of its calculation can be found in Vinken *et al* (2014b). In the final step, we scale the prior inventory ( $E_p$ ) by the ratio calculated in step one, multiplied with the  $\beta$ -factor, to obtain top down OMI ship NO<sub>x</sub> emissions ( $E_t$ ) taking into account non-linear chemistry:

$$E_t = E_p + \left( \frac{N_O - N_G}{N_G} \right) \cdot \beta \cdot E_p \quad (2)$$

where  $N_O$  and  $N_G$  represent the local OMI and GEOS-Chem tropospheric NO<sub>2</sub> columns that have been integrated along the shipping lanes, and corrected for background, non-shipping contributions (Vinken *et al* 2014b).

We eliminate the effect of *a priori* NO<sub>2</sub> profile shape on the comparison of model and observations, by applying the averaging kernel (Eskes and Boersma 2003) on the GEOS-Chem simulated NO<sub>2</sub> columns. The averaging kernel, provided along with the OMI retrieval, improves consistency between model and observations by accounting for the vertical sensitivity of the satellite instrument. Our top down NO<sub>x</sub> emission estimates for 2005 and 2006 agree to within 10% with earlier results presented in Vinken *et al* (2014b), where we did not apply the kernel but replaced the TM4 with GEOS-Chem *a priori* profiles. The top-down ship NO<sub>x</sub> emissions are lower because of the use of the averaging kernel, but patterns are consistent. We note, however, that ship lanes are less resolved in the maps of NO<sub>2</sub> columns due to the use of the averaging kernel (as a result of the rather coarse TM4 *a priori* NO<sub>2</sub> profiles at 3° × 2° resolution).

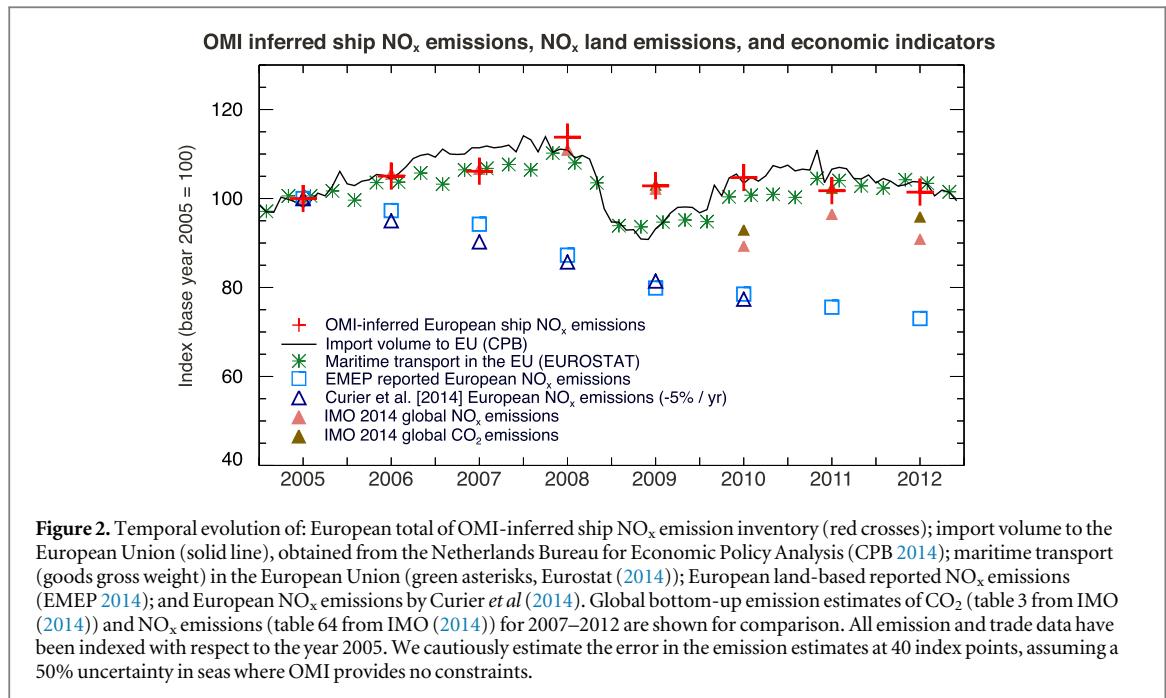
### 3. Results

We proceed and calculate constraints on NO<sub>x</sub> emissions over the four European seas, which represent 26% of all European ship NO<sub>x</sub> emissions, based on equation (2). The resulting European totals are presented in figure 2 (indexed with 2005 as base year), and per sea emissions are given in table 1. The figure shows that OMI-inferred European ship NO<sub>x</sub> emissions (red plusses) have increased by approximately 5% yr<sup>-1</sup> from 2005 to 2008, followed by a sharp reduction (−12%) between 2008 and 2009, and stayed relatively constant from 2009 onwards.

In 2008–2009, a global downturn in economic activity caused the import volume of the European

Union to fall steeply according to the statistical information from the Netherlands Bureau for Economic Policy Analysis (CPB; black solid line), as reflected in a decrease of 12% in OMI-inferred emissions for 2009 relative to 2008. The short-term variation in OMI NO<sub>x</sub> emissions corresponds with maritime transport (weight of goods) in the European Union (reported by Eurostat; green asterisks), another independent indicator of European economic activity. From 2009 onwards our OMI-inferred ship NO<sub>x</sub> emissions stay more or less constant, in line with the small variability in import volume and maritime transport in that period. Both OMI (European) and IMO GHG Study 2014 (global) top-down NO<sub>x</sub> emissions decrease between 2009 and 2007 (by −3% and −5%, respectively), and, more generally, the OMI and IMO estimates agree reasonably well for the period from 2007 to 2012 for which IMO estimates are available. Although the IMO emission estimates represent global totals, not directly comparable to our OMI European estimates, they have been extensively quality assured (through uncertainty analyses on input parameters, and by comparisons to IMO GHG Study 2009). Our results for the first time show the short-term variation in ship NO<sub>x</sub> emissions over European seas. Our results are in line with an earlier report on changes in satellite-observed NO<sub>2</sub> columns by de Ruyter de Wildt *et al* (2012), who found substantial increases in observed NO<sub>2</sub> columns between 2005 and 2008 for the Mediterranean Sea, followed by a decrease in 2009.

The increase and subsequent strong decrease of ship NO<sub>x</sub> emissions stands in clear contrast with trends in continental European NO<sub>2</sub> concentrations (Castellanos and Boersma 2012) and land-based NO<sub>x</sub> emissions (Zhou *et al* 2012, Curier *et al* 2014). Figure 2 shows that emissions over the European mainland, as reported to the EMEP programme (as part of the Convention on Long Range Transboundary Air Pollution (EMEP 2014)), decreased by about 4% per year over the 2005 to 2012 period (light blue squares), as a result of national and European emission control policies. A similar decrease in European land-based NO<sub>x</sub> emissions has been reported by Curier *et al* (2014), based on an inversion with OMI NO<sub>2</sub> columns for 2005–2010 (dark blue triangles in figure 2). Figure 3 illustrates this contrast between sea- and land-based annual changes in emissions between 2005 and 2008. This figure is a composite map of the variation in OMI-inferred ship NO<sub>x</sub> emissions reported in this study, and the OMI-inferred emissions decreases of 5% per year reported by Curier *et al* (2014) for land-based sources (converted to absolute changes using the TNO/MACC-II inventory (Kuenen *et al* 2014)). While land-based NO<sub>x</sub> emissions have decreased sharply over Spain, Great Britain, and Italy, we find substantial increases in ship NO<sub>x</sub> emissions over European seas between 2005 and 2008. That the OMI ship NO<sub>x</sub> emissions do not follow the land-based



**Table 1.** Overview of ship NO<sub>x</sub> emissions for different ship lanes and the total of all European ship NO<sub>x</sub> emissions (in Tg N yr<sup>-1</sup>) from 2005 to 2012. The overall error in the OMI top-down ship NO<sub>x</sub> emission inventory is estimated to be ~40% (Vinken *et al* 2014b).

Ship lane <sup>a</sup>	2005	2006	2007	2008	2009	2010	2011	2012
Mediterranean Sea	0.087	0.118	0.116	0.149	0.088	0.103 <sup>b</sup>	0.092 <sup>b</sup>	0.089 <sup>b</sup>
Bay of Biscay	0.068	0.074	0.079	0.114	0.073 <sup>c</sup>	0.073 <sup>c</sup>	0.065 <sup>c</sup>	0.065 <sup>c</sup>
North Sea <sup>d</sup>	0.042			0.051				
Baltic Sea	0.038	0.048	0.052	0.043	0.054 <sup>c</sup>	0.054 <sup>c</sup>	0.053 <sup>c</sup>	0.053 <sup>c</sup>
European total	0.91	0.96	0.97	1.04	0.94	0.95	0.93	0.93

<sup>a</sup> Emission strengths are aggregated over the ship lane as in Vinken *et al* (2014b).

<sup>b</sup> Because of the reduced number of valid observations in 2010–2012 compared to earlier years, annual constraints have been used for 2010–2012.

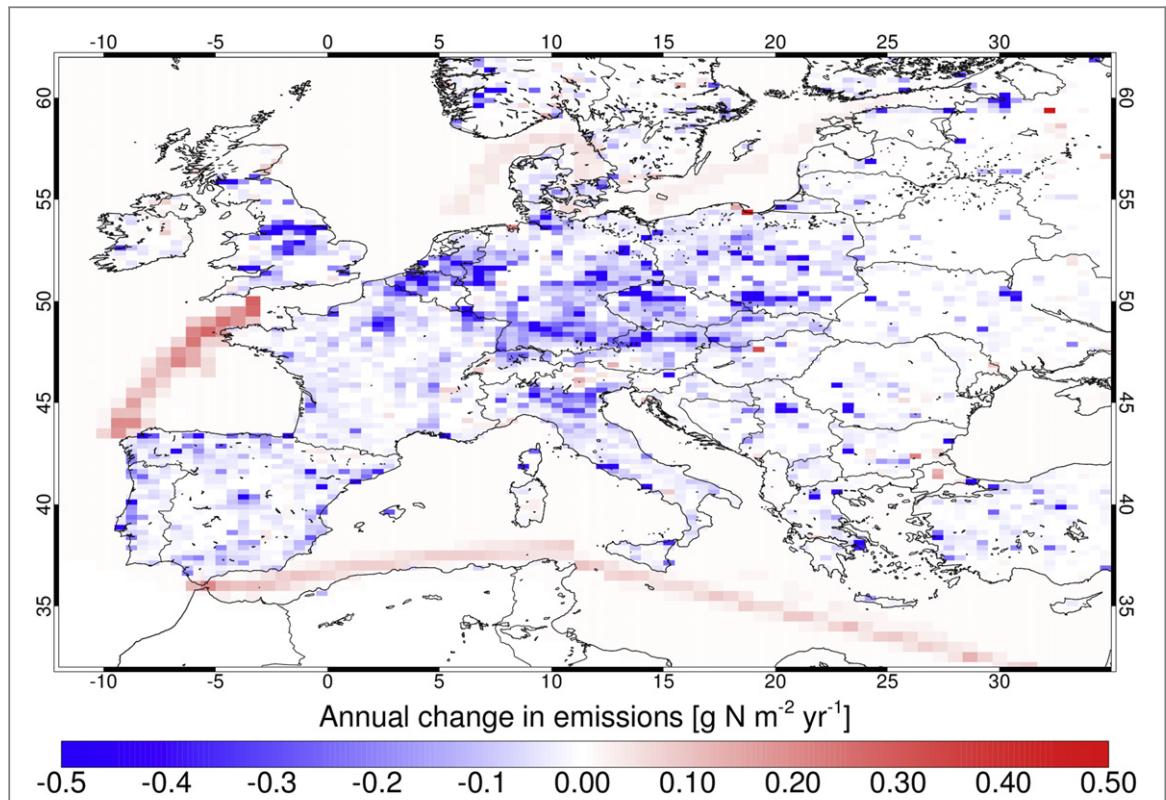
<sup>c</sup> Because of the limited number of valid observations in those years, data from 2009–2010 and 2011–2012 have been combined in one, single estimate.

<sup>d</sup> Because of the small number of valid observations over the North Sea, ship NO<sub>x</sub> emissions could only be determined for the years 2005 and 2008.

emission trends, provides compelling evidence for the increase of ship NO<sub>x</sub> emissions over the Bay of Biscay. It indicates that our method succeeds in screening out the influence of land-based emission sources in the inversion. Due to the steadily decreasing land emissions, and increases in the ship emissions, the share of the shipping sector to total European NO<sub>x</sub> emissions increased from 11% to 14% between 2005 and 2008. In 2009 it decreased to 13%, to slowly increase again to 14% in 2012.

As a direct result of the economic downturn in 2008–2009, demand for container shipping plummeted, and profits for ocean shippers fell sharply (Faber *et al* 2012, UNCTAD 2011). Shippers started experimenting with slow steaming of ships to: (1) cut costs by saving on fuel, as fuel prices doubled between

2005 and 2008 (Notteboom and Vernimmen 2009); and (2) reduce overcapacity, as demand fell and many new vessels, ordered before the global economic recession, became operational. Slow steaming refers to reducing the speed of a vessel from about 20–25 knots to 16–19 knots (Bonney 2010, Rodrigue *et al* 2013). As fuel consumption is approximately a quadratic function of ship speed (Faber *et al* 2012), fuel use (and thus costs) for ship voyages are reduced significantly by slow steaming (e.g. Eide *et al* 2009, Fagerholt *et al* 2009, Notteboom and Rodrigue, 2009). Also, CO<sub>2</sub> and NO<sub>x</sub> emissions have been reported to decrease sharply with reduced ship speed (EPA 2000, Agrawal *et al* 2008, Khan *et al* 2012). If slow steaming has indeed been implemented by the shipping sector, per ship NO<sub>x</sub> emissions are expected to have dropped



**Figure 3.** Composite map of annual changes (between 2005 and 2008) in  $\text{NO}_x$  emissions derived from OMI  $\text{NO}_2$  observations. Annual changes in OMI-inferred ship  $\text{NO}_x$  emissions (this work,  $1/2^\circ \times 2/3^\circ$  resolution) are combined with annual changes in OMI-inferred land-based  $\text{NO}_x$  emissions based on the annual trends reported by Curier *et al* (2014) (at  $1/4^\circ \times 1/2^\circ$  resolution) and absolute land-based emissions from the TNO/MACC-II inventory for the year 2009 (Kuenen *et al* 2014).

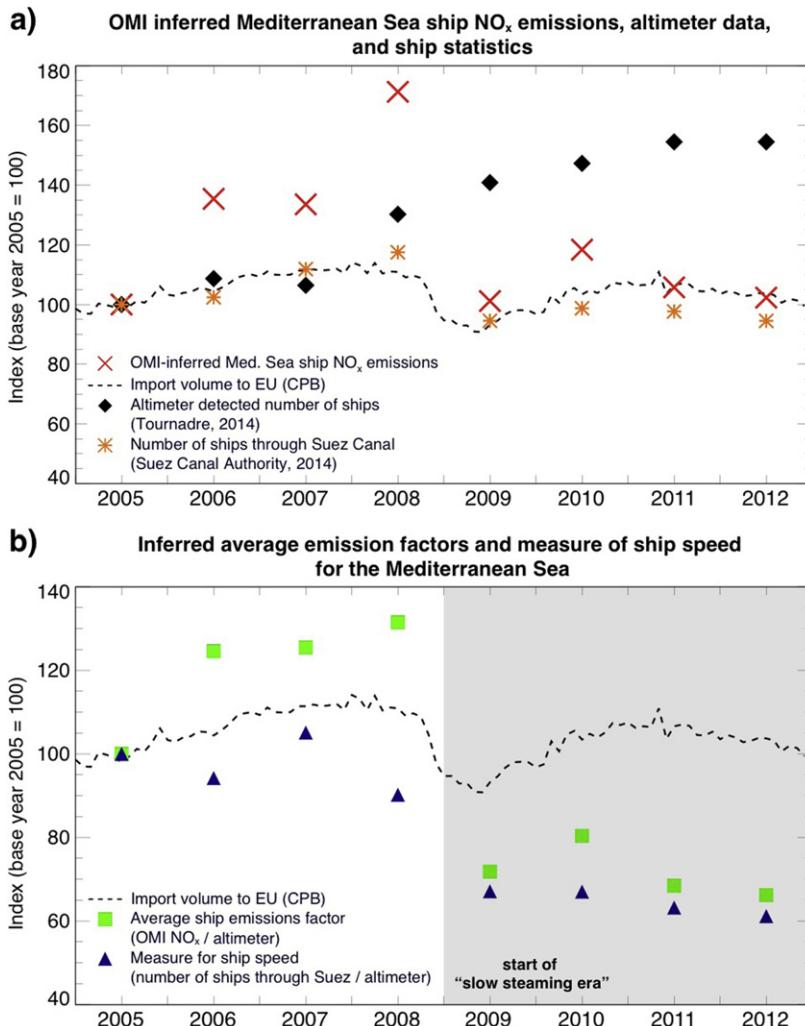
since 2008–2009. For the Mediterranean Sea, the availability of independent information on the ship density detected by satellite-borne altimeter instruments, and the counted number of ships passing through the Suez Canal, provides a unique opportunity to determine the changes in ship speed and per ship  $\text{NO}_x$  emissions in this sea.

Figure 4(a) shows the temporal evolution of ship  $\text{NO}_x$  emission over the Mediterranean Sea between 2005 and 2012 (red crosses). This figure shows a strong overall increase in the emissions of 71% between 2005 and 2008, followed by reduction back to 2005 levels in 2009. Ship  $\text{NO}_x$  emissions over the Mediterranean Sea remain around the 2009 level for subsequent years. The temporal evolution of the ship density detected by the satellite-borne altimeter (Tournadre 2014) over the Mediterranean Sea is also shown in figure 4(a) (black diamonds). The altimeter-detected ship density increases over the Mediterranean Sea, most notably after 2007. This increase in shipping implies that:

- (1) ever more or ever larger ships are sailing through the Mediterranean Sea; or
- (2) ships are sailing at lower speeds, which would increase their residence time in the Mediterranean Sea.

In both cases, the probability of ships being detected by the altimeter instruments has increased throughout the 2005–2012 period. Figure 4(a) also shows the temporal variation of the numbers of ships passing through the Suez Canal (Suez Canal Authority 2014) (orange asterisks). In the period 2005–2008, the counted number of ships passing through the Suez Canal increases in line with the altimeter-detected ship density until 2008, indicating that the number of ships sailing through the Mediterranean Sea indeed increased between 2005 and 2008. In 2009, the number of ships through the Suez Canal falls by 18%, whereas the altimeter-detected ship density continues to increase. This indicates that from this point in time on ships have likely decreased their speed in the Mediterranean Sea (via equation (1)), as the traffic through the Suez Canal represents a large fraction (but not all) of the Mediterranean traffic.

The reported number of ships through the Suez Canal allows us to analyze the change in ship speed, by calculating the derived measure for the average speed of ships in the Mediterranean Sea using equation (1). The average speed of ships is proportional to the ratio of the number of ships passing through the Suez Canal to the altimeter-detected ship density in the Mediterranean Sea. The resulting space-based indicator for ship speed is shown in figure 4(b) by blue triangles, and indicates that the average speed of ships varied by



**Figure 4.** (a) Temporal evolution of: OMI-inferred ship NO<sub>x</sub> emissions for the Mediterranean Sea (red crosses); import volume to the European Union (dashed line, (CPB 2014)); number of ships detected by the satellite-borne altimeter over the Mediterranean Sea (black diamonds) (Tournadre 2014); and the number of ships passing through the Suez Canal per year (orange asterisks (Suez Canal Authority 2014)). (b) Average ship NO<sub>x</sub> emission factor indicated by the green squares. The average ship speed of ships in the Mediterranean Sea (equation (2)) indicated by dark blue triangles. All data in both plots were indexed with respect to the year 2005.

less than 10% until 2008, but decreased by about 30% in 2009, and remained low until 2012. This is consistent with the reported start of slow steaming in 2008–2009, and vessel speed reductions of about 12–30% reported in literature (Faber *et al* 2012, Maltoni *et al* 2013, Rodrigue *et al* 2013, IMO 2014).

We make full use of the altimeter ship densities by also inferring information on the change of (average) NO<sub>x</sub> emissions per ship. The ratio of OMI-inferred ship NO<sub>x</sub> emissions to the altimeter-detected ship density over the Mediterranean Sea can be interpreted as a measure of the average ship NO<sub>x</sub> emission factor (green squares in figure 4(b)). Ship average NO<sub>x</sub> emissions increased from 2005–2008, then decreased by 46% from 2008 to 2009, to stay relatively constant, at 30% below the 2005 levels, in the years from 2009 onwards. The persistently lower ship NO<sub>x</sub> emission factors from 2009 onwards coincide with the implementation of the practice of slow steaming in 2008–2009 (Rodrigue *et al* 2013). In conclusion,

average emission factors per ship remain low since 2009, indicating that slow steaming has become a permanent operational practice in the shipping sector in the Mediterranean Sea.

#### 4. Discussion

Uncertainties in the absolute top-down ship NO<sub>x</sub> emission estimates originate from possible systematic errors in the OMI NO<sub>2</sub> retrievals, in GEOS-Chem NO<sub>2</sub> simulations, and in the inversion method. In line with earlier work by Vinken *et al* (2014b) we estimate the overall error in the OMI ship NO<sub>x</sub> emission estimates at 40–60%, assuming that the uncertainty in emissions for seas without OMI constraints is about 50%. However, there is much less uncertainty in the temporal evolution of the NO<sub>x</sub> emissions than in their absolute levels. Many of the model and retrieval errors are constant in time (e.g. identical assumptions on

vertical transport in the model, assumptions on albedo in the calculation of the AMF), and these will express themselves in NO<sub>x</sub> emission errors in a similar manner from one year to the other. The temporal variability in ship NO<sub>x</sub> emissions is thus mostly driven by variability in the retrieved NO<sub>2</sub> slant columns (spectral fit), as has been discussed in many earlier papers (Richter *et al* 2005, van der A *et al* 2006). The uncertainty in these changes is therefore mostly related to year-to-year differences in sampling, and instrumental effects such as the row anomaly. This detection ‘limit’ is very difficult to calculate, but we estimate it to be ±10%, in line with previous studies, which indicated that OMI is capable of capturing trends of this magnitude, and from the good consistency between time series of NO<sub>2</sub> columns over shipping lanes derived from GOME and SCIAMACHY with those of OMI (de Ruyter de Wildt *et al* 2012). The uncertainty is most likely somewhat higher for emission estimates after the row-anomaly. The year-to-year variation in NO<sub>x</sub> emissions is near the detection ‘limit’ (of ±10%) but substantial. Our results contribute to better estimates of the magnitude and temporal variation of regional ship NO<sub>x</sub> emissions for which currently no other estimates are available.

## 5. Conclusions

We report on short-term variability in European ship NO<sub>x</sub> emission estimates for the period from 2005 to 2012. We estimated ship NO<sub>x</sub> emissions over four distinct European shipping lanes in the Mediterranean, Bay of Biscay, North Sea, and Baltic Sea based on a mass-balance inversion, tropospheric NO<sub>2</sub> columns retrieved from OMI, and GEOS-Chem model simulations. Ship NO<sub>x</sub> emissions in European seas have increased from 2005 to 2008, in clear contrast to decreases in land-based NO<sub>x</sub> emissions in that period. In 2009, average ship NO<sub>x</sub> emissions over European seas dropped sharply in response to the global economic downturn, and have remained at the 2009 level until 2012, so there is little net change in overall shipping emissions between 2005 and 2012.

We found that the temporal variation in OMI-inferred ship NO<sub>x</sub> emissions corresponds with independent indicators of economic activity. However, variations in ship NO<sub>x</sub> emissions are not just the result of changes in the European trade volume, but also originate from changes in operational practice by the shipping sector, driven by increasing fuel prices, falling profits, and overcapacity. We used information from independent sources to determine how ship NO<sub>x</sub> emissions have changed in response to the new industry practices. Using the number of ships per unit area detected by satellite-borne altimeter instruments, we found that average per ship NO<sub>x</sub> emission factors fell by ~46% in 2009 (overall emissions fell by 69%) in the Mediterranean Sea and stayed relatively constant afterwards. By combining altimeter ship density data

with statistics on ships passing through the Suez Canal, we inferred the average speed of ships in the Mediterranean Sea. The temporal evolution of average ship speed shows a distinct, 30% reduction from 2008 to 2009, and persistently lower ship speeds in successive years. We interpret this as direct evidence that the practice of slow steaming, i.e. reducing ship speed to save fuel, has indeed been implemented widely, resulting in detectable reductions in ship NO<sub>x</sub> emissions.

Our results indicate that the implementation of slow steaming in 2009 has contributed to offsetting the 2005–2007 increase in NO<sub>x</sub> emissions over European shipping lanes, but the relative contribution of the shipping sector to total European NO<sub>x</sub> emissions increased from 11% in 2005 to 14% in 2012. This suggests that, in spite of the implementation of slow steaming, the shipping sector is responsible for an ever-larger share of the European NO<sub>x</sub> emissions, and additional measures might be required to reduce pollutant emissions at rates that are achieved by the road transport and energy producing sectors. Demand for waterborne freight transport is anticipated to grow deep into the 21st century, and because ships will keep using oil as fuel (Sims *et al* 2014), achieving (NO<sub>x</sub>) emission reductions to benefit climate and human health will be unlikely in the next decades. The improved capacity to monitor ship NO<sub>x</sub> emissions of space-borne sensors such as the S5P TROPOMI and geostationary sensors (GEMS over Asia, Sentinel-4 over Europe, and TEMPO over North America), all due for launch in the years to come, will provide timely and detailed information on the air pollution caused by ships, which can be used for policy decisions and to better inform the public.

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## Appendix A. Ship speed in the Mediterranean Sea

We use an analogy with a one-box model to derive the average speed of ships in the Mediterranean Sea. We interpret the number of ships detected by the altimeter satellite instrument ( $C_{\text{ship}}$ ) as a density of ships (unit: number of ships per m<sup>2</sup>) in the predefined area of the Mediterranean (rectangle in figure 1). The change rate of the ship density can then be written as:

$$\frac{dC_{\text{ship}}}{dt} = F_{\text{in}} - F_{\text{out}} = F_{\text{in}} - k \cdot C_{\text{ship}} \quad (\text{A1})$$

with  $F_{\text{in}}$  as the inflow of ships (in ships  $\text{m}^{-2} \text{s}^{-1}$ ), and  $F_{\text{out}}$ , the outflow from the Mediterranean Sea.  $k$  represents the loss (departure) rate ( $\text{s}^{-1}$ ) of ships from the Mediterranean Sea. Assuming steady state within a particular year, and no major changes loss processes in the Mediterranean Sea (such as the sinking of ships or the shifting of routes), the steady-state solution can be written as:

$$C_{\text{ship}} = \frac{F_{\text{in}}}{k} \quad (\text{A2})$$

With  $C_{\text{ship}}$  representing the density of ships in the Mediterranean Sea, and  $F_{\text{in}}$  representing the number of ships counted as passing the Suez Canal in that year. The loss rate  $k$  can be expressed as the ratio of the ships flowing out of the box per unit time to the ships within the box:

$$k = \frac{C_{\text{ship}} \cdot W \cdot v_{\text{ship}}}{C_{\text{ship}} \cdot L \cdot W} = \frac{v_{\text{ship}}}{L} \quad (\text{A3})$$

with  $W$  the width and  $L$  the length of the Mediterranean Sea box, and  $v_{\text{ship}}$  the speed of the ships. Combining equations (A2) and (A3) the speed of ships for in the Mediterranean Sea for a particular year can be derived as:

$$v_{\text{ship}} = L \cdot \frac{F_{\text{in}}}{C_{\text{ship}}} \quad (\text{A4})$$

The only assumptions we make here is that the fraction of ships sailing through the Mediterranean Sea (on its way to or coming from the Suez Canal) to those from or towards the Black Sea does not change between 2005 and 2012, and that the route ships take through the Mediterranean Sea has not changed (i.e. ships take the shortest route).

## Appendix B. GEOS-Chem chemistry transport model

We simulate tropospheric  $\text{NO}_2$  columns over Europe for 2005 to 2012 using the nested grid version of the GEOS-Chem chemistry transport model (v8-03-02)<sup>5</sup> (Wang et al 2004, Chen et al 2009). The model is operated at  $1/2^\circ \times 2/3^\circ$  resolution with 47 vertical layers, and extends from  $30^\circ$  to  $70^\circ \text{N}$  and  $30^\circ \text{W}$  to  $50^\circ \text{E}$ . We use the same model settings, and plume-in-grid approach (Vinken et al 2011) to account for non-linear chemistry in expanding ship plumes as previously described by Vinken et al (2014b). Here, we use the 2005 OMI derived top-down ship  $\text{NO}_x$  European emissions reported in that study, which amounts to  $1.0 \text{ TgN yr}^{-1}$ , as prior inventory. Ozone- $\text{NO}_x$ -hydrocarbon-aerosol chemistry in GEOS-Chem has recently been discussed by Lin et al (2012). In this study, we spin the model up for one year (2004) and

run simulations for 2005 to 2012. Daily tropospheric  $\text{NO}_2$  columns are averaged corresponding to the satellite overpass time (between 13:00–15:00 h local time). We only include days for which valid satellite observations are available to ensure a consistent comparison.

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