Supplementary Material to 'Constraints on ship NO_x emissions in Europe using GEOS-Chem and OMI satellite NO_2 observations'

G. C. M. Vinken, K. F. Boersma, A. van Donkelaar, and L. Zhang

1. Updated DOMINO_GC retrieval

Supplementary Figure S1 shows the GEOS-Chem profile for June 2006 within the ship track in the Mediterranean Sea (simulated with a priori ship emissions), and the GEOS-Chem profile for the simulation using the top-down constrained ship emissions. For this location, the NO_2 tropospheric vertical column density decreases strongly (by almost a factor 5) in the lowest model layer. This is reflected in the differences between the AMFs (Fig. S2(a) and S2(b)) by higher AMFs in the Mediterranean Sea (+20%). We see comparable changes in Fig. S2(a) and S2(b) over the North Sea (+10%), and lower AMFs in the Bay of Biscay (-25%) and the Baltic Sea (-20%).

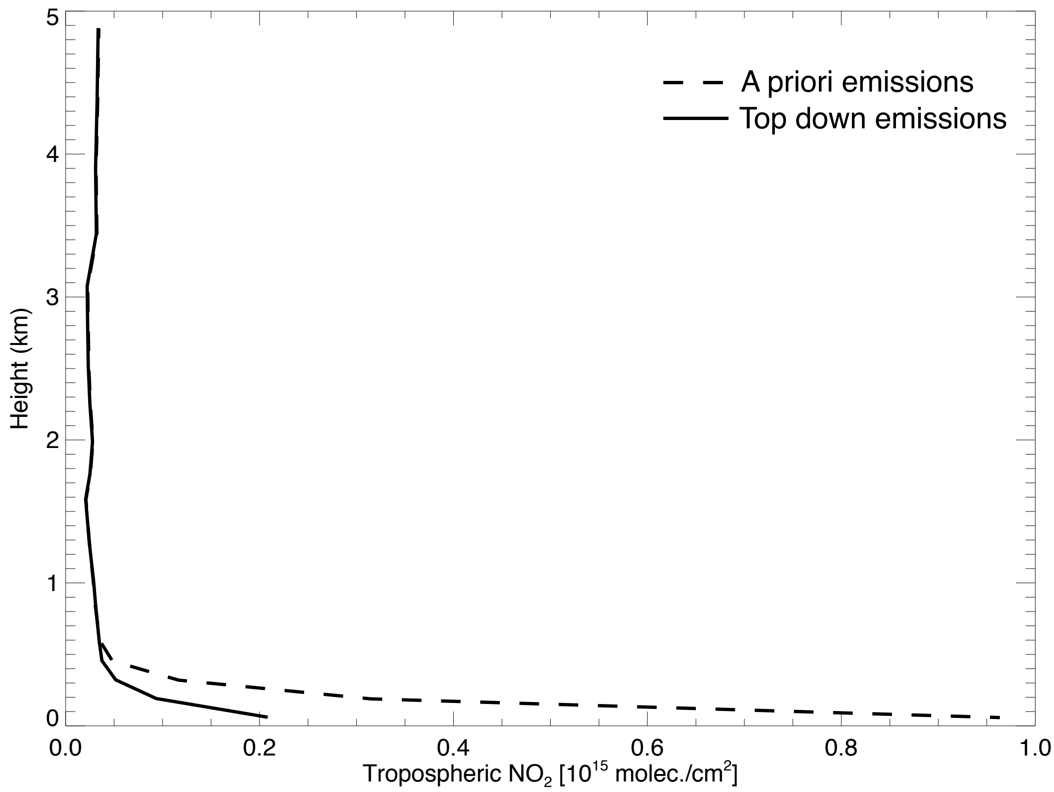
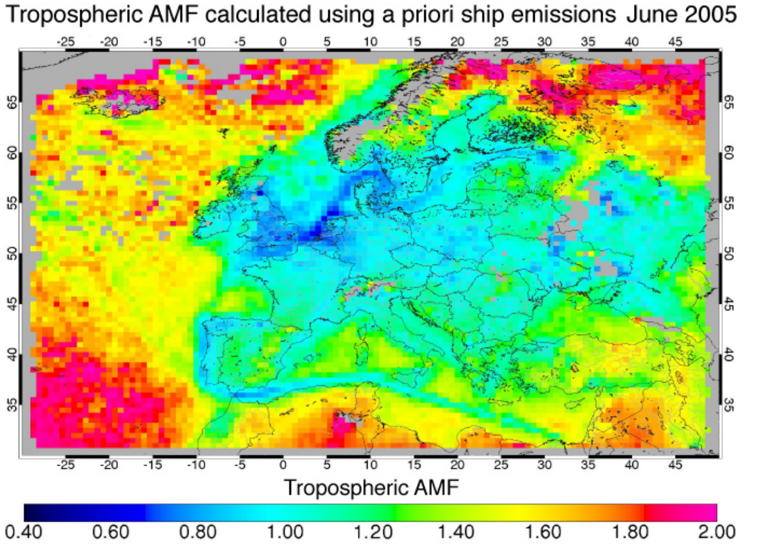
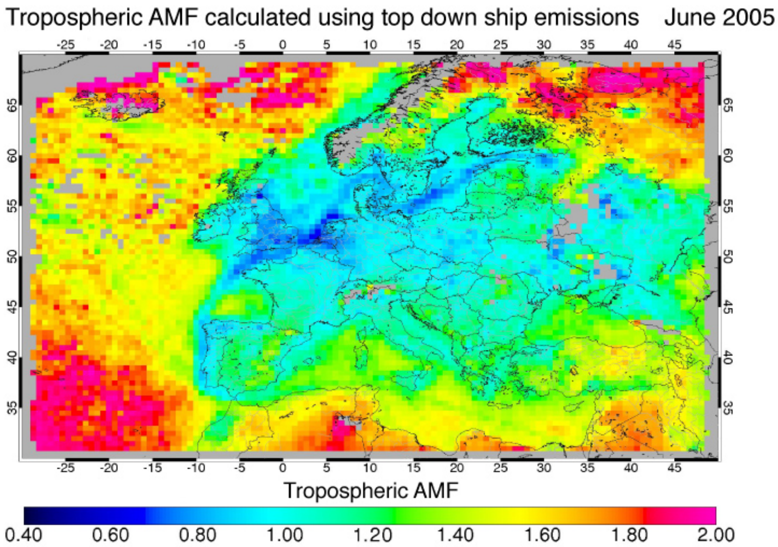


Figure S1: GEOS-Chem simulated NO_2 vertical profiles using the EMEP/AMVER-ICOADS a priori ship emission inventory (dashed line), and OMI constrained top-down emissions (solid line). This profile is averaged over June 2006 at OMI overpass time (13:00-15:00 LT) for a grid cell within the Mediterranean Sea ship track (25.3E,33.5N).

(a)



(b)



(c)

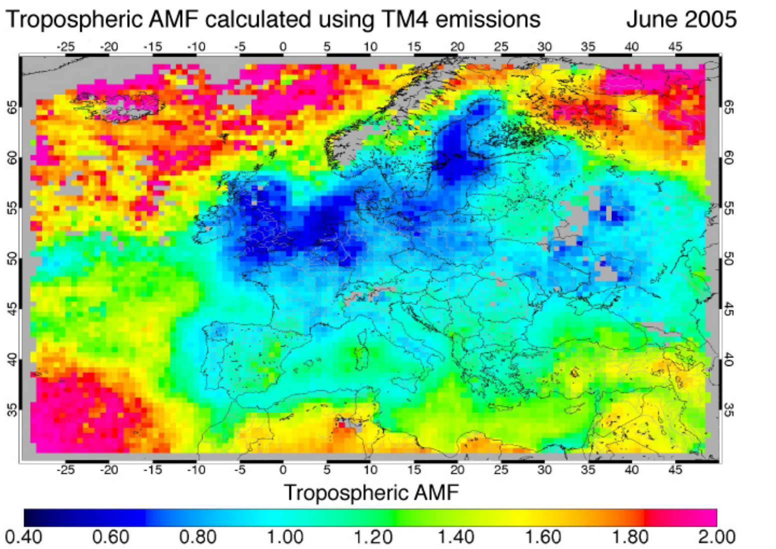


Figure S2: Tropospheric Air mass factors over Europe averaged for June 2005 calculated using the EMEP/AMVER-ICOADS a priori emissions **(a),** OMI top down emissions **(b)**, and the original DOMINO v2 AMFs calculated using the TM4 model **(c)**.

2. Background correction

Figure S3 shows the cross-section averages for the ship tracks indicated in Fig. 6 and 9. The dashed line represents the fitted (linear) background, that was subtracted from these NO_2 columns in Fig. 10. This background fit is a linear fit calculated between the same latitude or longitude points for all seasons / years.

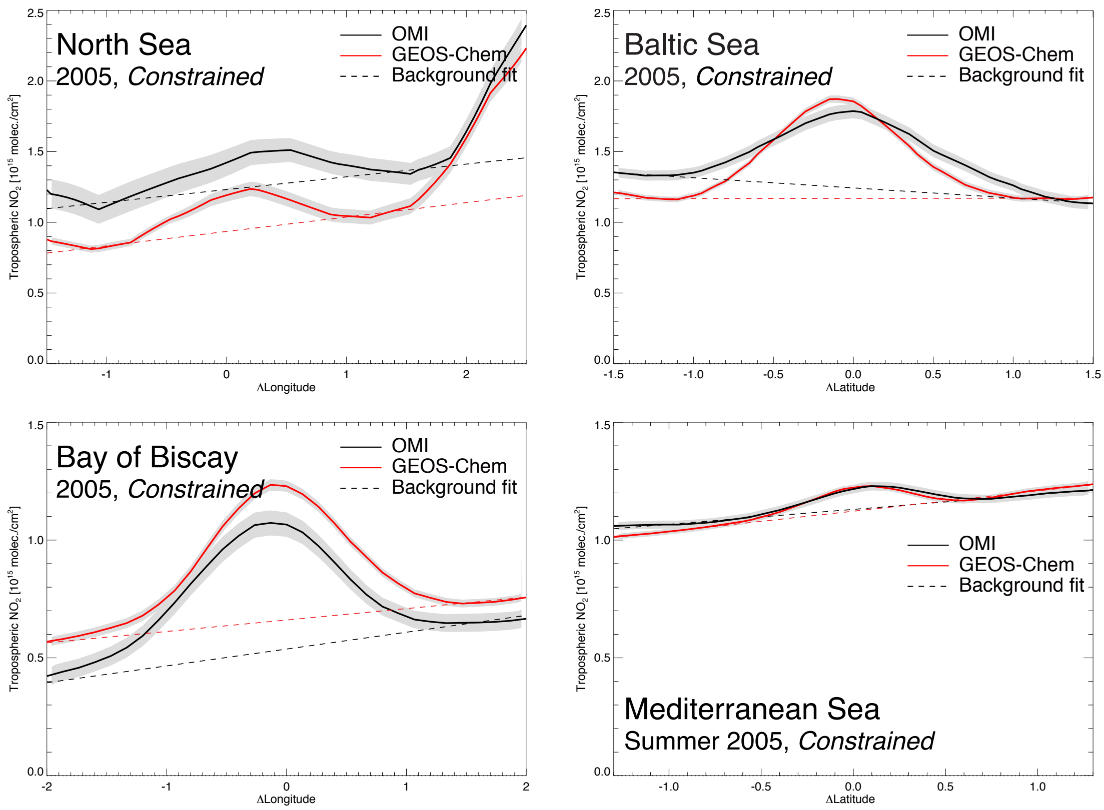


Figure S3: Along ship track averages of the tropospheric NO_2 columns over the areas in Fig. 6c for observed OMI (black line), and simulated GEOS-Chem columns (red line) using constrained emissions of Fig. 9b. Tropospheric NO_2 columns and averaging as in Fig. 10. A linear background fit is indicated by the dashed lines, and grey shadings represent the sample standard error.

3. Additional iteration of Equation 4

We found that for the Baltic Sea and some seasons in the Mediterranean Sea emission changes were large and an additional iteration was needed to ensure a match between GEOS-Chem and OMI within 10%. For this additional iteration we applied Eq. 4 again, but now using the top down emissions (calculated using Eq. 4) as a priori emissions for the next iteration. For consistency we based the OMI and GEOS-Chem columns on these top down emissions (indicated by subscript 3) by calculating β_2 and γ_2 (see Table S1) as follows:

$$\beta_2 = \frac{\Delta E/E}{\Delta N_{GC}/N_{GC,2}}$$

$$= \frac{\left. E_{a\,priori} + \left(\left(N_{OMI,1} - N_{GC,1}\right)/N_{GC,1}\right)\cdot\left(\beta_1 + \beta_1\cdot\gamma_1\right)\cdot E_{a\,priori} - \left(E_{a\,priori} + \left(\left(N_{OMI,1} - N_{GC,1}\right)/N_{GC,1}\right)\cdot E_{a\,priori}\right) \middle/ E_{a\,priori} + \left(\left(N_{OMI,1} - N_{GC,1}\right)/N_{GC,1}\right)\cdot E_{a\,priori}\right.}{\left(N_{GC,3} - N_{GC,2}\right)/N_{GC,2}}$$

$$= \frac{\left. \left(\left(N_{OMI,1} - N_{GC,1}\right)/N_{GC,1}\right)\cdot\left(\beta_1 + \beta_1\cdot\gamma_1\right) - \left(\left(N_{OMI,1} - N_{GC,1}\right)/N_{GC,1}\right) \middle/ \left(1 + \left(\left(N_{OMI,1} - N_{GC,1}\right)/N_{GC,1}\right)\right)\right.}{\left(N_{GC,3} - N_{GC,2}\right)/N_{GC,2}}$$

$$= \frac{\left. \left(\left(N_{OMI,1} - N_{GC,1}\right)/N_{GC,1}\right)\cdot\left(\beta_1 + \beta_1\cdot\gamma_1 - 1\right) \middle/ \left(1 + \left(\left(N_{OMI,1} - N_{GC,1}\right)/N_{GC,1}\right)\right)\right.}{\left(N_{GC,3} - N_{GC,2}\right)/N_{GC,2}}$$

and

$$\gamma_2 = \frac{\left(N_{OMI,3} - N_{OMI,2}\right)/N_{OMI,2}}{\left(N_{GC,3} - N_{GC,2}\right)/N_{GC,2}}$$

Then we proceed and calculate the final constraint on the emissions by:

$$E_{top\ down,2} = E_{top\ down,1} + E_{top\ down,1}\cdot\frac{\left(N_{OMI,3} - N_{GC,3}\right)}{N_{GC,3}}\cdot\beta_2 + E_{top\ down,1}\cdot\frac{\left(N_{OMI,3} - N_{GC,3}\right)}{N_{GC,3}}\cdot\beta_2\cdot\gamma_2$$

Simulations with this final run result in the cross-sections of Figure 10, and result in a close match (within 10%) between observed and simulated column enhancements relative to background for all 4 ship tracks.

Table S1: Table 2. Overview of constraints, beta and gamma values for the second iteration for the Mediterranean Sea, the North Sea, the Baltic Sea and the Bay of Biscay. Beta and gamma values are calculated as indicated above.

Ship track	Season/Year	$\frac{\left(N_{OMI,3} - N_{GC,3}\right)}{N_{GC,3}}$	β_2	γ_2
North Sea	2005	0.02	0.58	0.58
	2006	0.07	0.51	0.52
Baltic Sea	2005	0.46	0.30	0.23
	2006	0.62	0.37	0.27
Bay of Biscay	2005	0.21	0.75	0.29
	2006	0.27	0.65	0.27
Mediterranean Sea	Spring 2005	0.03	0.86	0.71
	Summer 2005	-0.04	0.71	0.87
	Autumn 2005	0.0	0.82	0.47
	Annual 2005	-0.09	0.80	0.68
	Spring 2006	-0.06	1.02	0.61
	Summer 2006	-0.41	0.95	0.97
	Autumn 2006	-0.14	0.88	0.67
	Annual 2006	-0.12	0.95	0.75