Satellite measurements of NO_2 from international shipping emissions

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[1] Measurements from the SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY) instrument on board the ENVISAT satellite have been analyzed for tropospheric NO_2 signatures of shipping emissions. Clear indication for NO_2 produced from ship emissions has been found over the Red Sea and along the main shipping lane to the southern tip of India, to Indonesia and north towards China and Japan where the signal is lost. Using simple assumptions for the NO_x loss, emission strengths were estimated and compared to an updated ship emission inventory. Good agreement was found in the spatial distribution and the absolute values for the Red Sea agree within a factor of 2, but larger discrepancies exist in other areas. Although the fluxes calculated still have large uncertainties, the results highlight the importance of ship emissions for the marine boundary layer and at the same time demonstrate the potential of satellite observations. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0394 Atmospheric Composition and Structure: Instruments and techniques; 3360 Meteorology and Atmospheric Dynamics: Remote sensing. **Citation:** Richter, A., V. Eyring, J. P. Burrows, H. Bovensmann, A. Lauer, B. Sierk, and P. J. Crutzen (2004), Satellite measurements of NO_2 from international shipping emissions, *Geophys. Res. Lett.*, *31*, L23110, doi:10.1029/2004GL020822.

1. Introduction

[2] Emissions of nitrogen oxides (NO_x = NO + NO_2) from the combustion of fossil fuels and biomass burning are well-established global sources of pollution. Recently, the influence of NO_x released by ships on tropospheric chemistry has been discussed [*Lawrence and Crutzen*, 1999; *Kasibhatla et al.*, 2000; *Davis et al.*, 2001; *Endresen et al.*, 2003]. The principal gaseous and particle emissions from ships include CO_2, H_2O, NO_x, SO_x, CO, unburned hydrocarbons, and particulate matter. The plumes from ship stacks effectively release these species at relatively high local concentrations into the marine boundary layer. While all these components do have an impact on the atmosphere and on climate [*Kasibhatla et al.*, 2000; *Davis et al.*, 2001; *Endresen et al.*, 2003; *Capaldo et al.*, 1999; *Durkee et al.*, 2000], in this paper we concentrate on NO_2 as this can now be observed by satellite measurements.

[3] NO_x emissions are dominated by anthropogenic sources, such as heating, electricity generation, and road transportation systems. To a lesser degree, biomass burning, shipping, soil emissions, lightning, and aviation also contribute to the observed concentrations. To assess accurately the impact of the emissions from shipping on the troposphere, and predict its future impact, detailed knowledge of the emission patterns and fluxes is required. Several emission inventories for shipping have been established in the last few years [*Corbett et al.*, 1999; *Olivier and Berdowski*, 2001; *Endresen et al.*, 2003]. *Corbett and Köhler* [2003] published an updated inventory for global fuel burn by internationally registered ships. Their bottom-up analysis suggests a total annual fuel burn of 289 Mt for ships greater 100 gross registered tons, nearly twice as much as the fuel consumption published by the international marine bunker industry. This highlights the degree of uncertainty in current emission estimates. Even though the first model studies used the lower total fuel consumption [*Corbett et al.*, 1999], an overestimate of the impact of shipping on the NO_x and ozone distribution appears to result [*Lawrence and Crutzen*, 1999; *Kasibhatla et al.*, 2000; *Davis et al.*, 2001; *Endresen et al.*, 2003]. This discrepancy can partly be attributed to an inadequate or oversimplified parameterization of chemical dilution within ship plumes, but inaccuracies in emission inventories could also play a role.

[4] Remote sensing measurements of NO_2 from space offer the potential for unique insight into this issue. NO_2 from anthropogenic emissions has been detected in Global Ozone Monitoring Experiment (GOME) measurements [*Leue et al.*, 2001; *Richter and Burrows*, 2002; *Martin et al.*, 2002], but the NO_2 signature of shipping is difficult to identify unambiguously because of the low spatial resolution of the GOME instrument (40 $\times$ 320 km^2). With the smaller ground scenes of the SCIAMACHY instrument (30 $\times$ 60 km^2), much more localized emissions can be detected in otherwise clean regions.

2. Instrument and Data Analysis

[5] The SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY) [*Burrows et al.*, 1995; *Bovensmann et al.*, 1999] was launched on ESAs ENVISAT satellite on February 28th, 2002. The instrument observes the upwelling radiation from the atmosphere and surface, and the extra terrestrial solar irradiance. During an orbit, SCIAMACHY measures alternately in nadir and limb viewing geometries in the spectral region between 220 and 2380 nm in eight channels at spectral resolutions between

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0.2 and 1.4 nm. The swath width of SCIAMACHY is 960 km providing global coverage at the equator within 6 days. ENVISAT flies in a sun synchronous orbit having an equator crossing time of 10:00 am.

[6] The retrieval approach taken for NO_2 nadir measurements is based on the Differential Optical Absorption Spectroscopy (DOAS) method. Briefly this technique determines the slant column density, SCD, along the light path through the atmosphere in a given spectral window by separating high frequency molecular signatures from broadband absorption and scattering [*Noxon*, 1975]. For NO_2 the spectral window between 425 and 450 nm is used in analogy to the GOME data analysis [*Richter and Burrows*, 2002]. In order to retrieve the tropospheric amounts of trace gases, the technique known as tropospheric excess method or reference sector method has been used. The method relies on the longitudinal homogeneity of the stratospheric column. Comparison of the measurements with those from regions having small concentrations of tropospheric NO_2 yields the tropospheric SCD, which can be converted to a vertical column density VCD by an Air Mass Factor, AMF, The AMF corrects for the different sensitivity of the measurements to absorption in different altitudes, which is of particular importance for absorbers located close to the surface. For the calculation of AMF undertaken here the radiative transfer program SCIATRAN [*Rozanov et al.*, 1997] has been used assuming a surface albedo of 4%, a maritime aerosol and a well mixed boundary layer of 700 m height. The analysis is similar to that applied to GOME data; more details of the retrieval of the tropospheric columns of NO_2 from GOME are provided by *Richter and Burrows* [2002] and *Burrows et al.* [1999].

3. Results and Discussion

[7] The Red Sea has been selected as a suitable region to investigate initially, the NO_2 from shipping. This is because a relatively large volume of traffic (approx. 14,000 ships per year with an average of 32,000 gross tonnage [*Institut für Seeverkehrswirtschaft und Logistik*, 2003]) passes through the Suez Canal between Europe and Asia and ship emissions are localized along a narrow track. Also, with the exception of a limited number of cities, the NO_2 emissions on the shores of the Red Sea are comparatively low reducing the risk of mixing between the two sources. Finally, the low cloud probabilities found in this region are favorable for tropospheric observations from space.

[8] In Figure 1a, the average of the NO_2 tropospheric column amount, derived from all available SCIAMACHY measurements from August 2002 to April 2004 for the region from 5°N to 35°N and 30°E to 60°E is shown. An intensity threshold was used to reject measurements with more than about 30% clouds. The most clearly distinguishable features can be attributed to the urban centers and the oil producing regions. Human release of NO_x is determining the amount of NO_2 in the Nile valley up to the Aswan dam. The impact of biomass burning and biofuel burning and soil emissions of NO_x in the African savannah, where the population density is low, is also clear. The desert regions of Egypt, Sudan, Ethiopia, Eritrea and Somalia have in comparison tropospheric amounts of NO_2 below the detection limit of about 1×10^{14} molecule cm^{-2}. Along the shores of the Red Sea and the Indian Ocean, cities such as Jedah, Aden, Djidda etc. are readily identifiable.

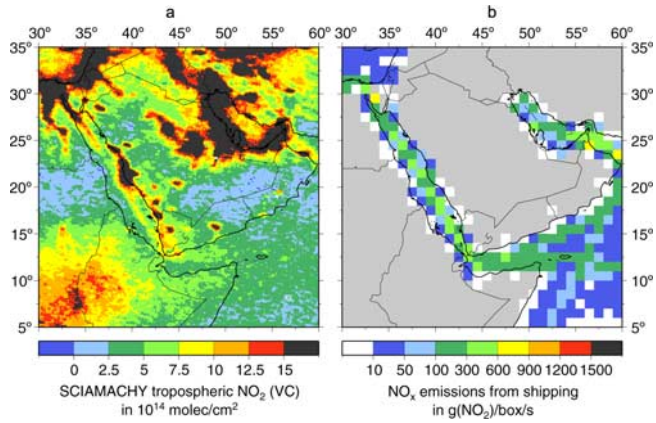

Figure 1. NO_x signature of shipping in the Red Sea (a) Tropospheric NO_2 columns derived from SCIAMACHY data from August 2002 to April 2004 using the Differential Optical Absorption Spectroscopy (DOAS) technique and the reference sector method for the region of the Red Sea (5°N to 35°N and 30°E to 60°E). (b) Estimated distribution of NO_x emissions by shipping in the same region. The global inventory has been updated using the *Corbett and Köhler* [2003] estimate for the year 2000 (6.87 Tg(N)/yr globally) and the global distribution for vessel traffic densities for the same year based on the reported distributions from AMVER data [*Endresen et al.*, 2003].

[9] In addition to the features described above, what can clearly be recognized in the measurements is a plume of NO_2 going down the middle of the Red Sea, and a similar signature in the Gulf of Persia. We attribute this to the NO_x release from ships.

[10] We have calculated an update of the global NO_x emission inventory from shipping using the *Corbett and Köhler* [2003] estimate of 6.87 Tg(N) per year and the global distribution for vessel traffic densities for the year 2000 based on the reported distributions from AMVER data with a $1° \times 1°$ spatial resolution [*Endresen et al.*, 2003]. Figure 1b shows the NO_x emissions from international shipping in the area of the Red Sea. The overall pattern in the emission inventory is in good agreement with the measurements. To compare the values in the emission inventory and the column amounts retrieved from SCIAMACHY, the emission flux has to be determined. This requires an estimate of the removal processes for NO_x. Here we provide only a rough estimate which will have to be followed by a quantitative model simulation based on in-situ measurements in the ship plumes as they become available. Assuming that $OH + NO_2 + M$ is the only loss process for NO_x, its effective second order reaction rate coefficient for the boundary layer is 2.5×10^{-11} cm^3 $molecule^{-1}$ s^{-1} [*Sander et al.*, 2000], and 24 h averaged background OH concentration is about 1.5×10^6 molecule cm^{-3} in this region [*Song et al.*, 2003], then a first order loss of NO_x is around 3.75×10^{-5} s^{-1}. Furthermore assuming a stationary state in NO_2, then the emission flux resulting from SCIAMACHY NO_2 columns of 5.5×10^{14} molecule cm^{-2} on a $1° \times 1°$ grid is $P_{NOx,SCIAMACHY} = 2.0 \times 10^{10}$ molecules cm^{-2} s^{-1} in the Red Sea shipping lanes.

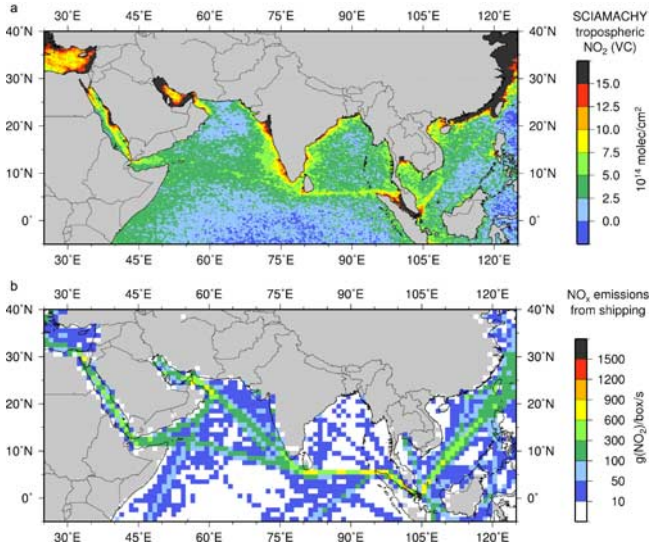

Figure 2. NO_x signature of shipping in the Indian Ocean. Same as Figure 1, but for an extended region that includes the Indian Ocean (5°S to 40°N and 25°E to 125°E).

[11] Clearly, the assumption of a background OH concentration for the NO_2 lifetime is a strong simplification of the complex loss processes in a diluting plume. As discussed by *Song et al.* [2003], the instantaneous NO_x lifetime depends on NO_x concentration and will be short right after emission but increase to background values as the plume evolves. The satellite observations are dominated by the aged plume as a result of their poor spatial resolution and thus the background value is more appropriate. Still, an uncertainty of up to a factor of 2 is introduced and the emissions are probably underestimated. The emission inventory for the same region is around 260 g (NO_2) s^{-1} into a box of 1° × 1°, which is about 10^{14} cm^2. Therefore the emission flux for the inventory is $P_{NOx,inventory} = 3.4 \times 10^{10}$ molecule cm^{-2} s^{-1} which is in the same order of magnitude as the SCIAMACHY measurements. Thus, the inventory does not only compare well with the geographical distribution of the measurements, but also the NO_x fluxes are comparable in the Red Sea.

[12] This result is further supported by the data from a larger geographical region (5°S to 40°N and 25°E to 125°E) shown in Figure 2a. Elevated NO_2 columns can be observed from the Red Sea to the southern tip of India, further to Indonesia and then up to the northeast where it separates into two branches. Other, weaker signals can be discerned between the Persian Gulf and India, and also in the Mediterranean and off the cost of Hong Kong. The very large values west of India and east of China are outflow of pollution from the continent and probably not related to ship emissions. Again the pattern of the satellite data is in good agreement with the inventory (Figure 2b), although it is worthwhile to point out that the inventory does not capture the slope of the lane between Sri Lanka and Sumatra (R3) and is clearly too far east of the coast of Singapore (R4). This is also reflected in the correlation coefficients between measurement and inventory that are 0.77 for R3 and 0.21 (0.83) for R4 without (and with) a one-degree shift to the west. The average estimates of emissions for the different shipping lines identifiable are compared in Table 1 using background OH concentrations of 2×10^6 molecule cm^{-3} for the tropical regions [*Song et al.*, 2003]. For all three regions, the agreement between measurement and emission inventory is reasonable, in particular when considering the large uncertainties in the assumptions made for the NO_x loss mechanism. The least satisfactory agreement is found in the part from India to Indonesia, where SCIAMACHY data suggest emissions comparable to those in the Red Sea whereas the inventory predicts a larger flux. This could be indication for a shorter than assumed NO_2 lifetime in this area but could also be related to inaccuracies in the emission inventory.

[13] Two factors contribute to the uncertainty of the satellite-retrieved emissions: the NO_2 columns used and the assumptions made for the lifetime of NO_2. Many parameters contribute to the accuracy of the satellite data [see *Richter and Burrows*, 2002; *Boersma et al.*, 2004]. In the case of ship emissions, the uncertainty in the spectral fit for the average used here is of the order of 10%. The uncertainty in the airmass factor is of the order of 25% which is mainly due to cloud contamination, possible aerosol effects and the uncertainty in the boundary layer height. An additional uncertainty arises from the identification of the shipping contribution (subtraction of stratosphere and background values) which we estimate to be about 20%. Assuming independent errors, the uncertainty of the SCIAMACHY derived NO_2 columns is thus 34%.

[14] The lifetime adopted could be too long if additional NO_x loss processes are active in the ship plumes or if the OH concentrations are larger than assumed, making the estimate a conservative lower limit. A further complication arises from the diurnal variation of the instantaneous NO_x lifetime which is long at night but short at daylight. At the SCIAMACHY measurement time of 10 am, the night time build-up of NO_2 might not yet be fully removed which would lead to an overestimation of NO_x emissions. Combination of the NO_2 column errors with the conservative estimate of a NO_2 lifetime overestimation of up to a factor of 2 leads to the error limits indicated in Table 1.

[15] Support for the lifetime adopted here comes from a recently published paper [*Beirle et al.*, 2004] that reports on

Table 1. Comparison of Ship Emission Inventory and SCIAMACHY Estimate

Shipping lane	Latitude Longitude (Start) to Latitude/Longitude (End)	Mean Inventory Estimate, molecule cm^{-2} s^{-1}	Assumed Lifetime, h	SCIAMACHY Estimate and Lower and Upper Limits, molecule cm^{-2} s^{-1}
R1 Red Sea	17.5°N 32°E to 30°N 44°E	3.4e + 10	7.4	2.0e + 10 (1.3..5.3e + 10)
R2 Indian Ocean West	5°N 44°E to 14°N 78°E	2.3e + 10	5.6	<1e + 10 (0.7..2.3e + 10)
R3 India to Indonesia	4°N 78°E to 7°N 98°E	7.1e + 10	5.6	2.8e + 10 (1.8..7.5e + 10)
R4 East of Indonesia	2°N 105°E to 8°N 110°E	6.2e + 10	5.6	2.7e + 10 (1.8..7.2e + 10)

the detection of NOx emissions from the shipping lane between India and Indonesia in 6 years of GOME data. In that study, the shape of the plume and the average wind speed was used to derive an effective lifetime of NO_2 of 3.7 (1.9–6.0) hours in reasonable agreement with the 5.6 hours used here. Using that lifetime, Beirle et al. found agreement between GOME derived emissions and inventories within the error limits, the satellite measurements being at the lower end of the estimates as in this work (see Table 1).

4. Summary and Conclusions

[16] The satellite data presented here show that emissions from ships do have a discernible impact on NO_2 and thus NO_x levels in clean regions on a more than local scale. The observed NO_2 pattern provides independent support for the emission distributions from current inventories at least for the region studied. Based on the assessment given here, it appears that the observed NO_2 column amounts are consistent with the updated inventory of ship emission fluxes based on the work of *Corbett and Köhler* [2003] within the uncertainties of the method (see Table 1). However, a final judgment for one or another emission estimate will only be possible after substantial reduction of the error bars. The accuracy of the space based emission estimates could be much improved with less uncertain assumptions for the NO_2 lifetime. This necessitates better knowledge of the horizontal and vertical distribution of the plumes and detailed modeling of NO_x removal. Use of more data from SCIAMACHY and in the future OMI will reduce the statistical errors and possibly facilitates the identification of shipping tracks in other regions of the world which has not been possible with the available data set.

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