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Impacts of the Large Increase in International Ship Traffic 2000–2007 on Tropospheric Ozone and Methane

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The increase in civil world fleet ship emissions during the period 2000–2007 and the effects on key tropospheric oxidants are quantified using a global Chemical Transport Model (CTM). We estimate a substantial increase of 33% in global ship emissions over this period. The impact of ship emissions on tropospheric oxidants is mainly caused by the relatively large fraction of NOx in ship exhaust. Typical increases in yearly average surface ozone concentrations in the most impacted areas are 0.5–2.5 ppbv. The global annual mean radiative forcing due to ozone increases in the troposphere is 10 mWm⁻² over the period 2000–2007. We find global average tropospheric OH increase of 1.03% over the same period. As a result of this the global average tropospheric methane concentration is reduced by approximately 2.2% over a period corresponding to the turnover time. The resulting methane radiative forcing is -14 mWm⁻² with an additional contribution of -6 mWm⁻² from methane induced reduction in ozone. The net forcing of the ozone and methane changes due to ship emissions changes between 2000 and 2007 is -10 mWm⁻². This is significant compared to the net forcing of these components in 2000. Our findings support earlier observational studies indicating that ship traffic may be a major contributor to recent enhancement of background ozone at some coastal stations. Furthermore, by reducing global mean tropospheric methane by 40 ppbv over its turnover time it is likely to contribute to the recent observed leveling off in global mean methane concentration.

Introduction

Previous emission inventories for the global ship fleet reported a significant contribution to global anthropogenic emissions of pollutants like CO₂, NO_x, and SO₂ (1–4). These studies presented emissions for the specific years 2000 and 2004. In this study we make an estimate of ship emissions over the period 2000–2007. This period was selected because it is characterized by large increases in traffic and thereby

emissions of pollutants. As in Dalsøren et al. (4), the fleet considered was the civil world fleet of ships above or equal 100 Gross Tons (GT). The gridded ship emission inventory constructed takes into account growth for the main ship types in the fleet.

Observations suggest that increasing emissions from international sea transportation may have a significant contribution to recent observed trends of pollutants at surface stations in various regions of the world (5–8), thereby offsetting the effect of land based regulations.

In this study the impact of increasing ship emissions on regional ozone and global hydroxyl (OH) and methane lifetime levels during the period 2000–2007 are quantified using the global Chemical Transport Model (Oslo CTM2). Ozone and OH levels are particular sensitive to NOx emissions from ships (9). We discuss implications for the oxidation potential further by calculating global hydroxyl changes and how this in turn influences recent trends of the greenhouse gas methane. Although there have been large year to year fluctuations in global methane (10, 11), the significant long-term trend has gradually leveled off since the 1980s. Several reasons for this have been discussed (12, 13), but despite findings that ship traffic has a quite large impact on hydroxyl and methane (1, 4, 9, 14, 15) the role of the recent increase in ship emissions was not quantified.

Model and Experiment

The OsloCTM2 model was used to calculate the effects on oxidants of increasing ship emissions. The setup is similar to Dalsøren et al. (4, 14), and the model was run in T42 resolution ($2.8^\circ \times 2.8^\circ$) with 60 vertical layers using meteorological data for 2000. Modeled distribution of ozone and ozone precursors have been evaluated and compared to observations for ship impacted air in previous studies (1, 14). The major loss of methane is tropospheric removal by reaction with hydroxyl. The impacts on hydroxyl and methane of emission changes of CO, NMVOCs, and NOx in different regions were studied by the model in Dalsøren et al. (16, 17). In order to study the response on methane a simple box model (18) was used to solve eq (1): $(dC)/(dt) = P - Q \cdot C = (E)/(\beta) - (1)/(\tau) \cdot C$. C is the mixing ratio of methane (ppbv), P is the production rate, E is the emission rate, β is a factor for conversion of emissions to mixing ratio (TgCH₄/ppbv), τ is the lifetime (in years), and Q is the loss rate. Assuming that the production (E/β) and loss (Q) terms are constant during the time step Δt , the differential equation above is solved to give eq (2): $C_t = (E)/(\beta \cdot Q) + (C_{t-1} - (E)/(\beta \cdot Q))e^{-Q \cdot \Delta t}$ where the loss term is $Q = 1/\tau$. τ is a function of the instantaneous chemical lifetime (τ_0 atm) and the lifetime due to soil uptake, the methane concentration at former time step and a factor accounting for the feedback of methane changes on its own lifetime (18–20). E was varied to study the impact of typical uncertainties in emission estimates reported by the IPCC AR4 report (21).

The radiative forcing concept is often used for quantitative comparisons of various mechanisms causing climate change (22). The ozone radiative forcing is calculated with solar and longwave radiative transfer schemes (23, 24) and shown to compare within 10–15% of other schemes used frequently for tropospheric ozone (24, 25). Monthly mean ozone fields from Oslo CTM2 were used in the calculations. The fields of temperature, water vapor, and cloud data are consistent with the data used in the Oslo CTM2. We also calculate the methane and the methane-induced ozone radiative forcing and follow the approach used in Hoor et al. (9). The calculated

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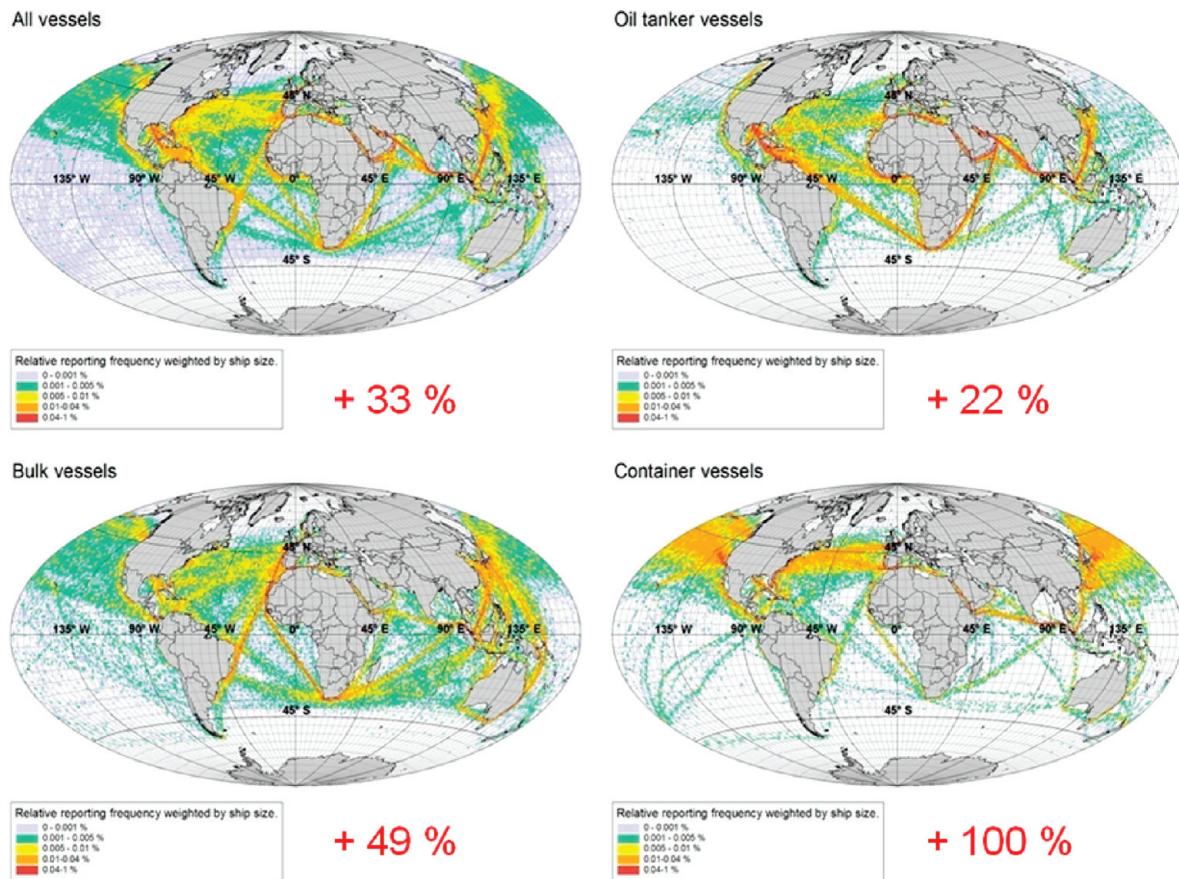


FIGURE 1. Vessel traffic densities (relative number of observations per grid cell) for year 2001/2002 based on the AMVER (2005) data. Upper left: all cargo and passenger ships in the AMVER merchant fleet, upper right: oil tankers, lower left: bulk carriers, lower right: container vessels. Red numbers denote the estimated increase in emissions in the period 2000–2007.

methane change from eq 2 due to 2000–2007 increased ship traffic is adopted. Based on detailed radiative transfer simulations a specific radiative forcing for methane changes of $0.37 \text{ mWm}^{-2} \text{ ppbv}^{-1}$ is used for a background level of methane of 1740 ppbv and nitrous oxide of 319 ppbv. Since a methane reduction also leads to a reduction in the ozone amount, we also estimate this radiative forcing. As described in ref 9 the methane-induced ozone radiative forcing is 0.42 times the methane forcing. This value is based on multimodel simulations calculating the increase in ozone and ozone radiative forcing due to a methane increase.

A new global ship emissions inventory reporting year 2004 emissions for major ship types and ports (4) serves as a basis for the ship emissions used in the CTM study. The emissions for the period 2000–2007 were obtained by scaling the 2004 numbers with reported changes in seaborne trade (tonne-miles) (26) for different ship types. This approach is based on former studies suggesting a close historical relationship between development in seaborne trade in tonne-miles and fuel consumption (15, 27). The approach was also used in the IMO GHG study (28) to backcast and forecast point estimates for emissions to compare between studies with different reference years. The increase in trade is assumed equal to the increase in fuel consumption (so X% increase in trade equals X% increase in fuel consumption). This is a simple approximation, but it is considered justifiable over the limited time period. Even if Marpol Annex VI (29) entered into force in 2005 and among other things regulates NO_x emissions from new or converted engines and sulfur emissions in special control areas (and globally) we assume limited changes in engine emission factors for the period in question. We also assume that the operational speed of the fleet had no overall change over the period 2000–2007. We therefore

inferred a one to one relation between changes in fuel consumption and emissions. The changes in emissions over the period are shown in Figure 1 and quantified for the total fleet and separately for the three major ship types (container, bulk, tanker) constituting approximately 50% of the 2004 fuel consumption. We calculate a total emission increase of 33% from international sea transportation over the period 2000–2007. These findings are supported by the IMO GHG study (28) which estimated an increase in fuel consumption of 34% in the period 2000–2007. The increase in emissions from container ships is estimated to be as high as 100% whereas bulk ship emissions increase by 49% and tanker with 22%. As shown in Figure 1 the major ship types have very different geographical operational patterns, and the increase in emissions differ. For the fleet not falling into one of the three major ship types we used one uniform scaling factor so that the change for this group plus the summed change for the three major types equaled the change for the total fleet, i.e. 33%. Based on the above assumptions, yearly gridded inventories were constructed for the period in question. Emissions from other sectors, taken from the RETRO database (30), were fixed at their 2000 levels to discern the impact of increasing ship emissions from other factors.

Results

The modeled surface ozone distribution for July 2000 is shown in Figure 2. High ozone concentrations are found in the vicinity of major precursor emissions and where the meteorological conditions are favorable for photochemical production. In the Northern Hemisphere this is typically downwind of industrial or populated regions of eastern U.S., central/southern Europe, the Middle East, and southeast Asia.

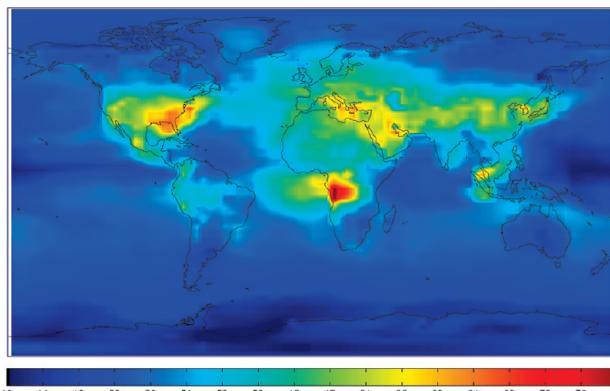


FIGURE 2. Surface ozone distribution in July 2000 (ppbv).

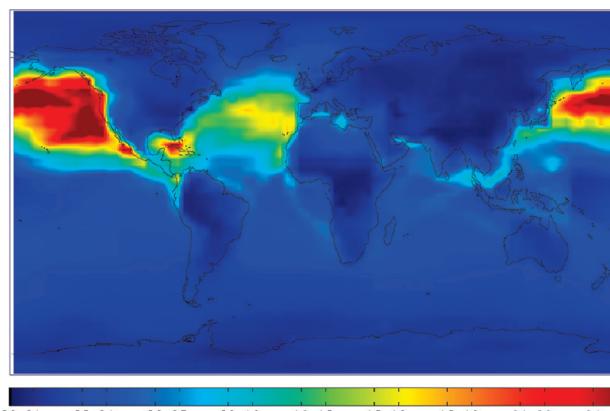


FIGURE 3. Absolute changes (ppbv) of surface ozone for July due to 2000–2007 increase in ship emissions.

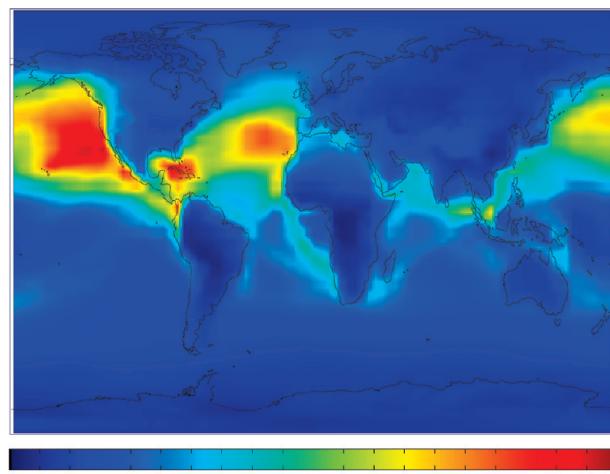


FIGURE 4. Yearly average absolute increase (ppbv) in surface ozone due to increase in ship emissions 2000–2007.

In the Southern Hemisphere high concentrations are found over a region with intense savanna burning and recirculation of air masses in Africa. The background concentrations over the oceans are low, but close to coastal regions near precursor source regions ozone is sufficiently high to be of concern with regards to guidelines for vegetation and health damage (31). As shown in Figures 3 and 4, the surface ozone changes due to increasing ship emissions in the period 2000–2007 are largest over the background oceans. This is due to the nonlinearity of the ozone chemistry (32–34) resulting in larger signals from perturbations in less polluted areas. There are also substantial increases of surface ozone in coastal regions of northwestern North America. The container traffic has the largest emission increase of all segments, and much of

it occurs in the North Pacific (Figure 1). Due to this and the nonlinearity we find the largest increase in surface ozone in the North Pacific. The influence on surface ozone of increased transport across the Pacific between Asia and North America is clearly evident both in Figures 3 and 4. Though the increases in coastal waters of Europe are smaller they could be of significance for the discussion of recent trends at coastal stations.

Since there has been a reduction of most land based regional emissions in Europe and North America (35, 36), a discussion evolves whether the increases found in background ozone are caused by long-range transport from Asia or ship emissions (8, 35, 37). In Figure 5 the modeled surface ozone in 2000 is compared to observed concentrations at the coastal stations Saturna in western Canada and Mace Head in Ireland. Mace Head experiences air masses directly off the North Atlantic Ocean to the immediate west and sometimes air masses from the polluted regions of continental Europe to the east (38). The Saturna island is situated in the midst of major international shipping channels and is frequently impacted by emissions from marine vessels but also emissions from surrounding urban areas (39). The model reproduces the timing of low and high values quite accurately though the diurnal amplitude is somewhat smaller than the observed. The calculated levels are also reasonable, the modeled yearly mean for Saturna is about 4.5 ppbv too high, and for Mace Head it is 2.7 ppbv too low. The changes in modeled surface ozone from 2000 to 2007 at the two stations solely due to ship emissions are shown in Figure 6. We find that ship emissions increase ozone with 0.13 ppbv/yr for Saturna (Figure 6) and 0.05–0.4 ppbv/yr along the northwestern North American coast in general (Figures 3 and 4). For Mace Head we find that increased ship emissions result in an averaged surface ozone increase of 0.23 ppbv/yr. Chan (40) estimates a meteorologically adjusted trend in daily average ozone from 1997–2006 for Pacific Canadian stations (including Saturna) of 0.32 ± 0.43 ppbv/yr. From the 1990s up to now, Parrish et al. (8) report an ozone increase of about 0.34 ± 0.09 ppbv/yr for marine boundary layer stations in northwestern North America (not including Saturna) when the wind flow is onshore from marine areas. In the same study a trend for marine air masses of 0.39 ± 0.11 ppbv/yr is found for Mace Head though this station show indications of stabilization for the last years. Comparing the modeled and measured figures it seems likely that increased ship emissions can explain some of the increase in background ozone. The numbers given by Parrish et al. (8) are exclusively for air masses of marine origin, the number from Chan (40) is meteorologically adjusted, while our estimates include all air masses using the meteorological year 2000. Zhang et al. (37) modeled an increase of approximately 0.16–0.25 ppbv/yr at the west coast of North America due to increasing Asian emissions over the 2000–2006 period. Combining this finding with our estimation of the impact from ship emissions one can get close to the reported increase for marine background air masses in Parrish et al. (8) and Chan (40).

Figure 7a shows the change in tropospheric ozone column in Dobson units (DU), and Figure 7b shows the radiative forcing. The changes in the ozone column roughly follow the same spatial pattern as the changes in the surface ozone (Figure 4), but the perturbation is more widespread due to the longer lifetime of ozone at higher altitudes. The radiative forcing of the ozone change from 2000 to 2007 is 10.3 mWm^{-2} . To put this in a perspective this radiative forcing is 1/3 of the radiative forcing due to ozone from shipping between year 2000 and preindustrial times (18). Due to the much stronger radiative forcing of ozone change in the upper troposphere and lower stratosphere (41, 42) and differences in the regional radiative forcing for a given ozone change (25), the pattern

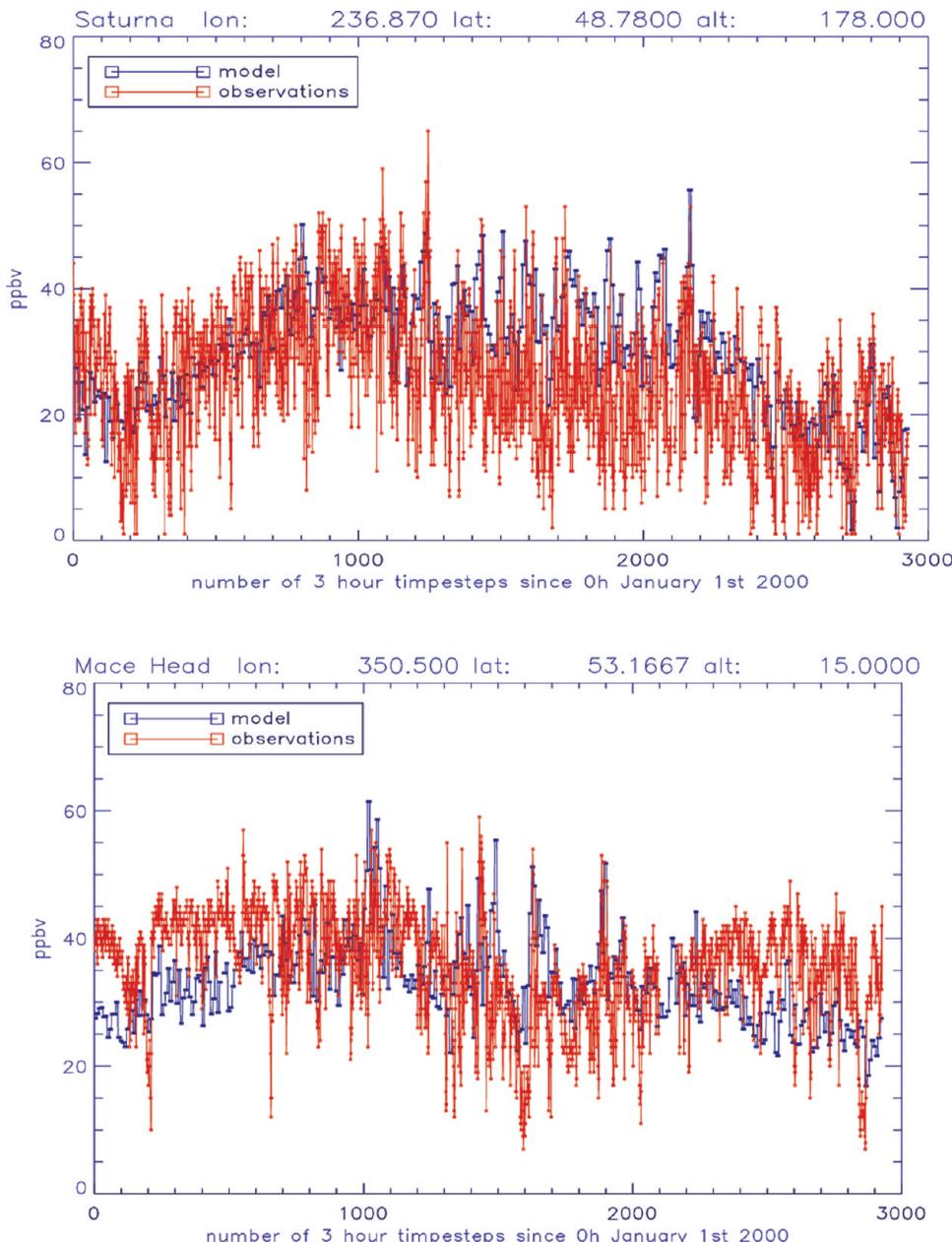


FIGURE 5. Comparison of observed (red) and modeled (blue) surface ozone in 2000 at two coastal stations. Saturna (upper) and Mace Head (lower). The temporal resolution is 3 h.

of radiative forcing follows the pattern of ozone change only to some extent.

With the CTM we calculated a global mean tropospheric OH increase of 1.03% due to the 2000–2007 increase in sea transportation. The resulting decrease in global mean tropospheric methane lifetime was 1.57%. In Figure 8 we have used eq 2 to calculate the development of global methane for different year 2007 starting points. Using the change in methane lifetime calculated by the CTM we have varied the emission term (E in eq 2) within its uncertainty to study three different and possible situations: One with production larger than loss, one where the production and loss are about equal, and one where the loss exceeds the production. We have then decreased the methane lifetime ($\tau_{\text{O}_3 \text{ atm}}$) in eq 2 by 1.57% and repeated the calculations to study the impact of the increasing ship emissions. For all situations we calculate that the methane concentration is reduced by almost 0.2%/yr over the typical tropospheric turnover time of methane. Compared to typical trends of methane of +0.5–1%/yr in the 1980s, +0–0.5%/yr in the

1990s, and only small changes after the millennium this is significant contribution from one single emission sector. The calculated methane change of 39 ppbv from eq 2 results in a RF of -14.4 mWm^{-2} . From 2000 to 2007 the additional methane-induced ozone RF is then -6.1 mWm^{-2} .

The overall net-global mean forcing due to the changes in shipping emissions over the period 2000–2007 is then $10.3 - 14.4 - 6.1 = -10.2 \text{ mWm}^{-2}$ (see Figure 9).

Discussion

The emission estimates used in this study are based on the most updated trade statistics available at the time, the UNCTAD 2007 (25) report. After finalizing the model runs described in this paper, the 2008 UNCTAD Review of maritime transport (43) was released. The updated UNCTAD (43) report presents updated figures, in particular for tank, bulk, and total trade. The updated figures show a growth between 2000 and 2007 of 22% for tank, 63% for dry bulk, and 39% for total trade. Hence, it is evident that the actual figures reported for

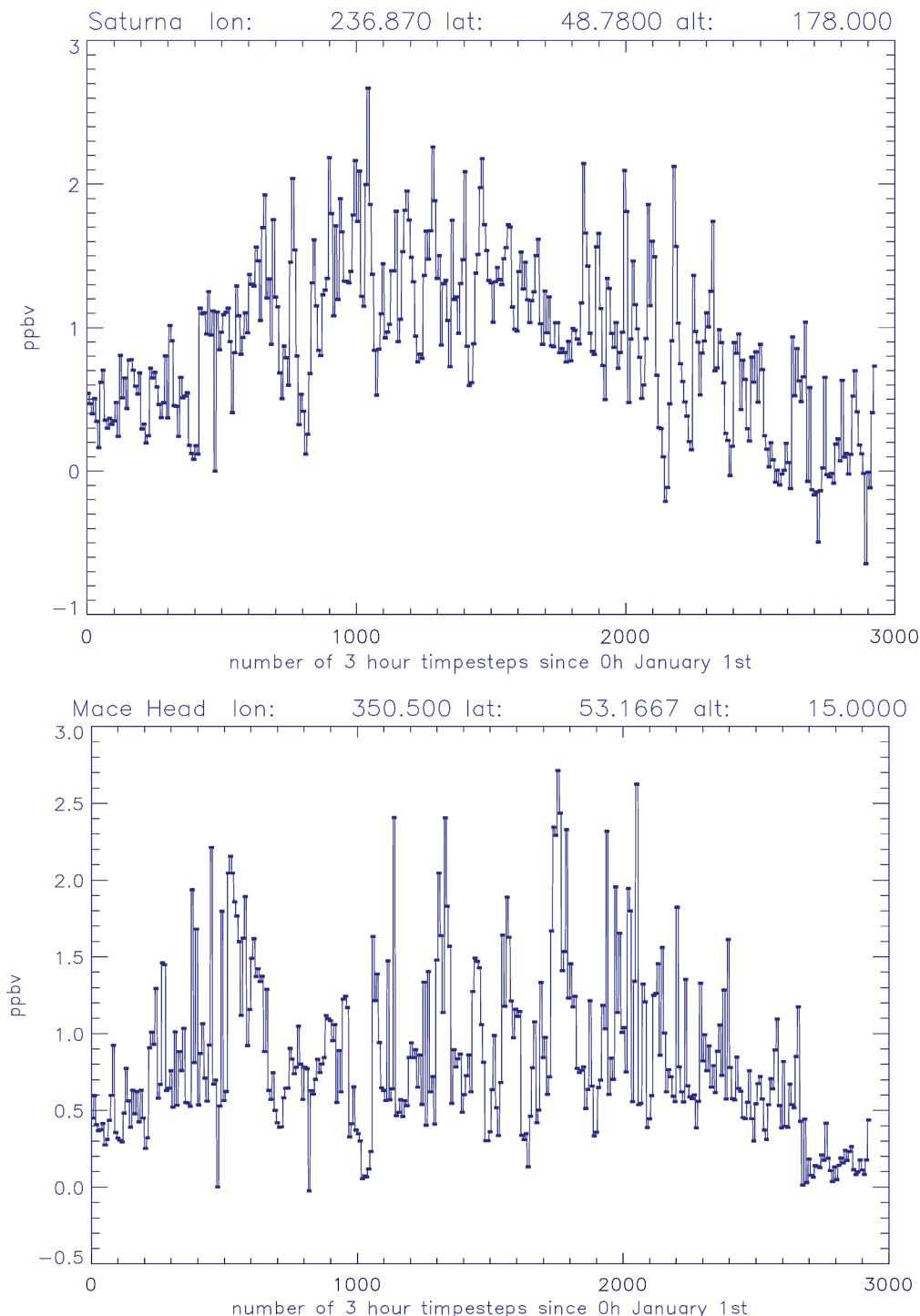


FIGURE 6. Modeled changes in surface ozone at the two coastal stations Saturna (upper) and Mace Head (lower) solely due to increasing ship emissions from 2000 to 2007. The meteorological year is 2000, and the temporal resolution is 3 h.

2007 are in good agreement with the estimates used herein for total trade and tank. For the bulk trade however, the used estimate is significantly lower than for the figures reported by UNCTAD 2008 (43).

The major cause of the 2000–2007 increase in seaborne trade is the blooming of economies in Asia, in particular China, and the large growth in container trade. However, with the financial crisis developing in 2008 the market for shipping has taken a dramatic turn. The collapse was unexpected and will result in substantial decreased growth rates the coming years. The true impact of this crisis on seaborne transport is not yet known. In an interim report on the impact of the financial crisis, OECD (44) reports an

expected 13.2% decline in worldwide trade for 2009 (not specific for seaborne trade) and a moderate growth of 1.5% in 2010. UNCTAD (45) currently estimates world merchandise trade to fall between 6 and 8% in 2009. However, most long-term future scenarios on international seaborne trade expect increases of 1–4%/year (15, 28, 46).

Although the traffic patterns are kept constant for the period in question, the split into major ship types and differentiated growth rates ensure that we to a large extent keep track of shifts in traffic patterns on the world oceans. There might however be different development of shorter intraregional transport, and the use of one global uniform scaling factor per ship type does not capture this. This is also

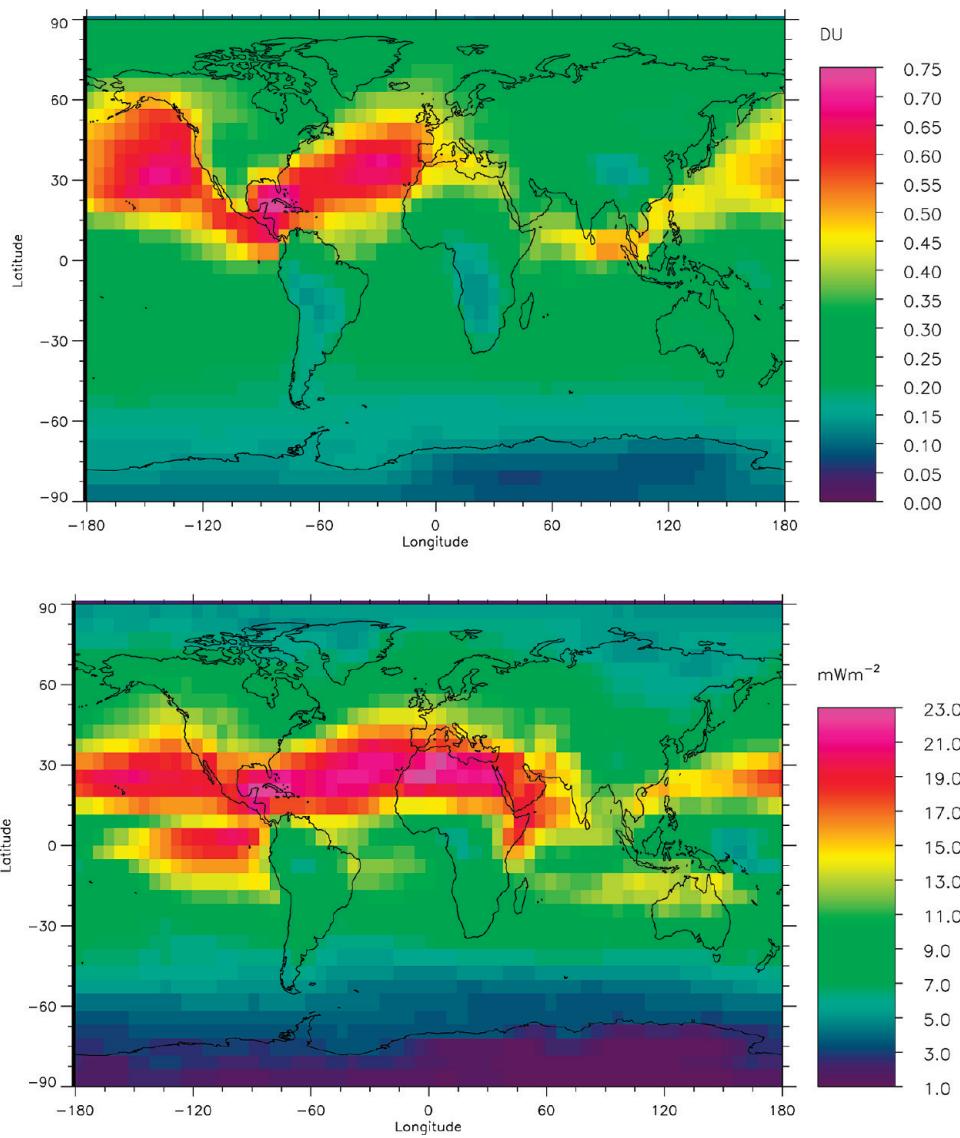


FIGURE 7. a: Change in tropospheric ozone column (Dobson Units) due to increasing ship emissions 2000–2007. b: Radiative forcing (mW m^{-2}) of ozone at the tropopause level due to increasing ship emissions 2000–2007.

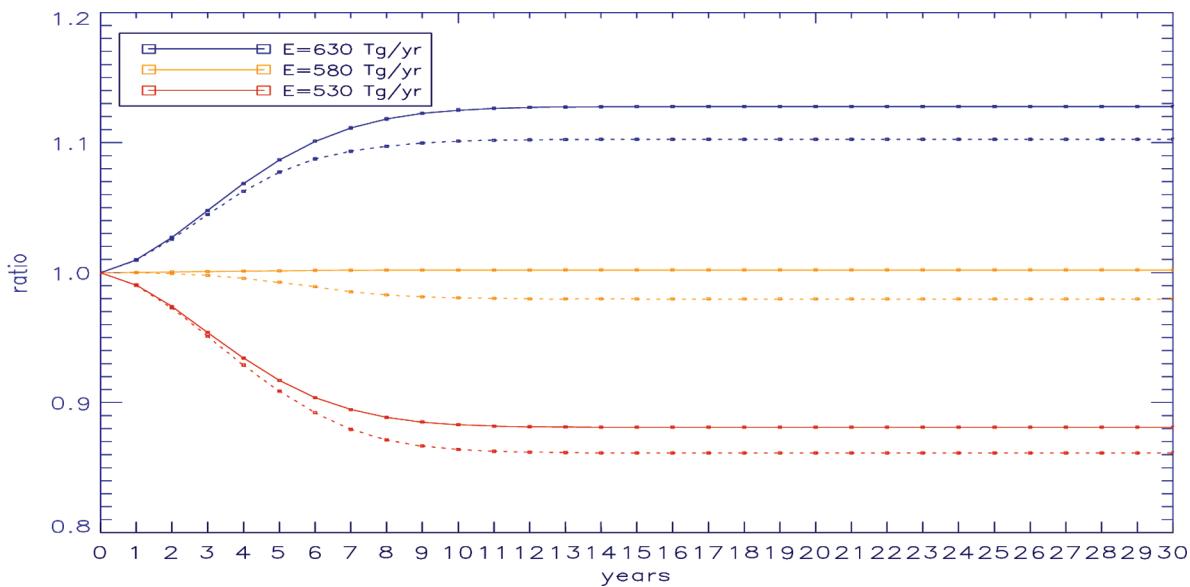


FIGURE 8. Relative changes in global averaged tropospheric methane over time solving eq 2 using the modeled change in lifetime and different emission (E) numbers. Lines show the development using global averaged methane lifetime from a simulation excluding an increase in ship emissions, and the dotted lines take into account the decrease in methane lifetime resulting from increased ship emissions in the period 2000–2007.

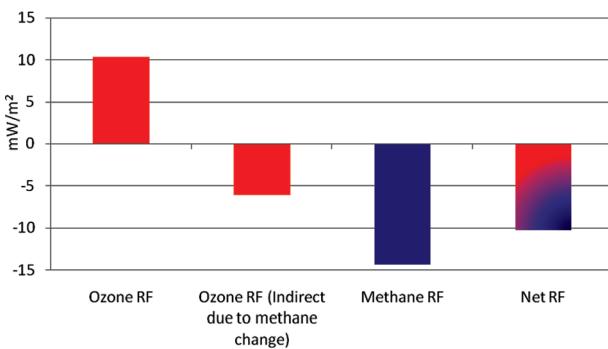


FIGURE 9. Radiative forcing (RF) (mWm^{-2}) of ozone and methane changes due to increasing ship emissions over the period 2000–2007. In the methane calculations the concentration change after a turnover time of 12 years was used.

the case for ports where we did a uniform scaling equal to the change (+33%) for the whole fleet not taking into account that certain ports are dominated by visits of specific ship types. The latter effect is likely less important as port emissions account for only 5% of total shipping emissions (4).

Box-model studies and measurements (47–53) indicate enhanced NO_x destruction within ship plumes and that global models might overestimate the effect of ship emissions on the NO_x , OH, and ozone budget. On the other hand, some studies (52, 53) find reasonable correspondence between global models and observations. Whereas Dalsøren et al. (14) states that limited observations in ship impacted air outside plumes inhibit clear conclusions regarding the performance of global models.

In this study we used meteorology for the year 2000 in all simulations. Meteorological variability may have an influence on calculated ozone trends in North America and Europe (35, 36, 40). However, this does not change the findings from several studies that ozone is increasing in background coastal air masses. It was for instance found by Chan (40) that the meteorologically adjusted ozone trend is stronger for the Pacific Canada than the nonadjusted trend. Land based emissions of ozone precursors have also changed the past decade. Unfortunately no global emission inventory exists with data after year 2000 suitable for a global model. It is possible that recent changes in nonship emissions change the contribution from shipping due to nonlinearities. Studies for the future (14, 52, 54) have shown some differences in the absolute and relative contribution from shipping between scenarios with very different changes of land based emissions. However, since our comparison is outside heavy polluted regions where nonlinearity is strongest and due to our short time frame (2000–2007), we believe the development in land emissions has limited influence on our overall conclusion on the shipping contribution.

Currently, the International Maritime Organisation (IMO) is imposing international regulations on emissions of key pollutants (55). Without a successful implementation of national or international regulations the experiences from this and other studies (56) indicate that ship emissions may have significant effects on the trends of greenhouse gases, key oxidants, and pollutants. In coastal zones of developed countries ship emissions, if measures are not taken, may strongly counteract the benefits from likely continuing reductions in emissions from some land based sectors.

Acknowledgments

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Literature Cited

- Endresen, Ø.; Sørgård, E.; Sundet, J. K.; Dalsøren, S. B.; Isaksen, I. S. A.; Berglen, T. F.; Gravir, G. Emission from international sea transportation and environmental impact. *J. Geophys. Res.* **2003**, *108* (D17), 4560. doi:10.1029/2002JD002898.
- Eyring, V.; Köhler, H. W.; van Aardenne, J.; Lauer, A. Emissions from international shipping: 1. The last 50 years. *J. Geophys. Res.* **2005**, *110*, D17305. doi:10.1029/2004JD005619.
- Corbett, J. J.; Koehler, H. W. Updated emissions from ocean shipping. *J. Geophys. Res. [Atmos.]* **2003**, *108*, 4650–4666.
- Dalsøren, S. B.; Eide, M. S.; Endresen, Ø.; Mjelde, A.; Gravir, G.; Isaksen, I. S. A. Update on emissions and environmental impacts from the international fleet of ships: the contribution from major ship types and ports. *Atmos. Chem. Phys.* **2009**, *9*, 2171–2194.
- Lelieveld, J.; van Aardenne, J.; Fischer, H.; de Reus, M.; Williams, J.; Winkler, P. Increasing Ozone over the Atlantic Ocean. *Science* **2004**, *304*, 1483–1487.
- Derwent, R. G.; Stevenson, D. S.; Doherty, R. M.; Collins, W. J.; Sanderson, M. G.; et al. The contribution from ship emissions to air quality and acid deposition in Europe. *Ambio* **2005**, *34* (1), 54–59.
- Devasthale, A.; Krüger, O.; Grassl, H. Impact of ship emissions on cloud properties over coastal areas. *Geophys. Res. Lett.* **2006**, *33*, L02811. doi:10.1029/2005GL024470.
- Parrish, D. D.; Millet, B. D.; Goldstein, A. H. Increasing ozone in marine boundary layer inflow at the west coasts of North America and Europe. *Atmos. Chem. Phys.* **2009**, *9*, 1303–1323.
- Hoor, P.; Borken-Kleefeld, J.; Caro, D.; Dessens, O.; Endresen, Ø.; et al. The impact of traffic emissions on atmospheric ozone and OH: results from QUANTIFY. *Atmos. Chem. Phys.* **2009**, *9*, 3113–3136.
- Dlugokencky, E. J.; Houweling, S.; Bruhwiler, L.; Masarie, K. A.; Lang, P. M.; Miller, J. B.; Tans, P. P. Atmospheric methane levels off: Temporary pause or a new steady state. *Geophys. Res. Lett.* **2003**, *30*, 148–149.
- Rigby, M.; et al. Renewed growth of atmospheric methane. *Geophys. Res. Lett.* **2008**, *35*, L22805. doi:10.1029/2008GL036037.
- Wang, J. S.; Logan, J. A.; McElroy, M. B.; Duncan, B. N.; Megretskaya, I. A.; Yantosca, R. M. A 3-D model analysis of the slowdown and interannual variability in the methane growth rate from 1988 to 1997. *Global Biogeochem. Cycles* **2004**, *18*.
- Bousquet, P.; Ciais, P.; Miller, J. B.; Dlugokencky, E. J.; Hauglustaine, D.; et al. Contribution of anthropogenic and natural sources methane emissions variability. *Nature* **2006**, *443*, 439–443.
- Dalsøren, S. B.; Endresen, Ø.; Isaksen, I. S. A.; Gravir, G.; Sørgård, E. Environmental impacts of the expected increase in sea transportation, with a particular focus on oil and gas scenarios for Norway and northwest Russia. *J. Geophys. Res.* **2007**, *112*, D02310. doi:10.1029/2005JD006927.
- Eyring, V.; Isaksen, I. S. A.; Berntsen, T.; Collins, W. J.; Corbett, J. J.; Endresen, Ø.; Grainger, R. G.; Moldanova, J.; Schlager, J.; Stevenson, D. S. Assessment of Transport Impacts on Climate and Ozone: Shipping. *Atmos. Environ.* **2010**, in press.
- Dalsøren, S. B.; Isaksen, I. S. A. CTM study of changes in tropospheric hydroxyl distribution 1990–2001 and its impact on methane. *Geophys. Res. Lett.* **2006**, *33*, L23811. doi:10.1029/2006GL027295.
- Dalsøren, S. B.; Isaksen, I. S. A.; Li, L.; Richter, A. Effect of emission changes in southeast Asia on global hydroxyl and methane levels. *Tellus, Ser. B* **2009**, *61* (4), 588–601.
- Fuglestvedt, J.; Berntsen, T.; Myhre, G.; Rypdal, K.; Skeie, R. B. Climate forcing from the transport sectors. *Proc. Natl. Acad. Sci.* **2008**, *105* (2), 454–458.
- Osborn, T. J.; Wigley, T. M. L. A simple model for estimating methane concentrations and lifetime variations. *Climate Dynamics* **1994**, *9*, 181–193.
- Wigley, T. M. L. The contribution of emissions of different gases to the enhanced greenhouse effect. *Climate Change and the Agenda for Research*; Westview Press: 1994.
- Denman, K. L.; Brasseur, G.; Chidthaisong, A.; Ciais, P.; Cox, P. M.; et al. Couplings Between Changes in the Climate System and Biogeochemistry. In *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*; Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K., Tignor, B. M., Miller, H. L., Eds.; Cambridge University Press: Cambridge, United Kingdom and New York, NY, U.S.A., 2007.

- (22) Forster, P.; Ramaswamy, V.; Artaxo, P.; Berntsen, T.; Betts, R.; et al. Changes in atmospheric constituents and in radiative forcing. In Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M. et al., Eds.; *Climate change 2007: The physical science basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge University Press: United Kingdom and New York, NY, U.S.A., 2007.
- (23) Myhre, G.; Karlsdottir, S.; Isaksen, I. S. A.; Stordal, F. Radiative forcing due to changes in tropospheric ozone in the period 1980 to 1996. *J. Geophys. Res.* **2000**, *105* (D23), 28935–28942.
- (24) Gauss, M.; Myhre, G.; Isaksen, I. S. A.; Collins, W. J.; Dentener, F. J.; et al. Radiative forcing since preindustrial times due to ozone change in the troposphere and the lower stratosphere. *Atmos. Chem. Phys.* **2006**, *6*, 575–599.
- (25) Berntsen, T. K.; Isaksen, I. S. A.; Myhre, G.; Fuglestvedt, J. S.; Stordal, F.; et al. Effects of anthropogenic emissions on tropospheric ozone and its radiative forcing. *J. Geophys. Res.* **1997**, *102* (D23), 28101–28126.
- (26) United Nations Conference on Trade and Development (UNCTAD). *Review of Maritime Transport 2007*; New York, 2007; ISBN: 978-92-1-112725-6 (online available at <http://www.unctad.org/Templates/WebFlyer.asp?intItemID=2598&lang=1> (accessed month day, year).
- (27) Endresen, Ø.; Sørgård, E.; Behrens, H. L.; Brett, P. O. A historical reconstruction of ships' fuel consumption and emissions. *J. Geophys. Res.* **2007**, *112*, D12301. doi:10.1029/2006JD007630.
- (28) Buhaug, Ø.; et al. Prevention of Air Pollution from Ships. Updated 2000 Study on Greenhouse Gas Emissions from Ships. Phase 1 Report - Executive Summary. 2008, International Maritime Organization IMO, Marine Environment Protection Committee 58th session Agenda item 4, MEPC 58/4/4. September 1, 2008.
- (29) International Maritime Organization (IMO). Regulations for the prevention of air pollution from ships and NOx technical code. 1998, ANNEX VI of MARPOL 73/78, London.
- (30) Schultz, M.; van het Bolscher, M.; Pulles, T.; Brand, R.; Pereira, J.; Spessa, A. RETRO report on emission data sets and methodologies for estimating emissions. Workpacage 1, Deliverable D1-6, 2007; EU-Contract No. EVK2-CT-2002-00170. Available at http://retro.enes.org/reports/D1-6_final.pdf (accessed month day, year).
- (31) Fowler, D. The Royal Society: Ground-level ozone in the 21st century: future trends, impacts and policy implications; REPORT 15/08; 2008;148 pp, ISBN: 978-0-85403-713-1 Issued: October 2008 RS1276.
- (32) Isaksen, I. S. A.; Hov, Ø; Hesstvedt, E. Ozone generation over rural areas. *Environ. Sci. Technol.* **1978**, *12*, 1279–1284.
- (33) Lui, S. C.; Trainer, M.; Fehsenfeld, F. C.; Parrish, D. D.; Williams, E. J.; Fahey, D. W.; Hubler, G.; Murphy, P. Ozone production in the rural troposphere and the implications for regional and global ozone distributions. *J. Geophys. Res.* **1987**, *92* (D4), 4191–4207.
- (34) Lin, X.; Trainer, M.; Liu, S. C. On the nonlinearity of the tropospheric ozone production. *J. Geophys. Res.* **1988**, *93* (D12), 15,879–15,888.
- (35) Jonson, J. E.; Simpson, D.; Fagerli, H.; Solberg, S. Can we explain the trends in European ozone levels? *Atmos. Chem. Phys.* **2006**, *6*, 51–66.
- (36) EPA (Environmental Protection Agency). *Latest Findings on National Air Quality Status and Trends through 2006*; Contract No. EP-D-05-004 Work Assignment No. 3-4 EPA-454/R-07-007; 2008.
- (37) Zhang, L.; et al. Transpacific transport of ozone pollution and the effect of recent Asian emission increases on air quality in North America: an integrated analysis using satellite, aircraft, ozonesonde, and surface observations. *Atmos. Chem. Phys.* **2009**, *8*, 6117–6136.
- (38) Simmonds, P. G.; Derwent, R. G.; Manning, A. L.; Spain, G. Significant growth in surface ozone at Mace Head, Ireland, 1987–2003. *Atmos. Environ.* **2004**, *38*, 4769–4778.
- (39) McLaren, R.; Wojtal, P.; Majonis, D.; McCourt, J.; Halla, J. D.; et al. NO₃ radical measurements in a polluted marine environment: links to ozone formation. *Atmos. Chem. Phys. Discuss.* **2009**, *9*, 24531–24585.
- (40) Chan, E. Regional ground-level ozone trends in the context of meteorological Influences across Canada and the eastern United States from 1997 to 2006. *J. Geophys. Res.* **2009**, *114*, D05301. doi:10.1029/2008JD010090.
- (41) Forster, P. M. D.; Shine, K. P. Radiative forcing and temperature trends from stratospheric ozone changes. *J. Geophys. Res.* **1997**, *102* (D9), 10841–10855.
- (42) Hansen, J.; Sato, M.; Ruedy, R. Radiative forcing and climate response. *J. Geophys. Res.* **1997**, *102* (D6), 6831–6864.
- (43) United Nations Conference on Trade and Development (UNCTAD). *Review of Maritime Transport 2008*; New York, 2008; ISBN 978-92-1-112758-4 (online available <http://www.unctad.org/Templates/WebFlyer.asp?intItemID=4659&lang=1> (accessed month day, year).
- (44) OECD. *OECD Economic Outlook, Interim Report*; 2009; ISBN 978-92-64-06136-1, March 2009.
- (45) United Nations Conference on Trade and Development (UNCTAD). *Global economic crisis: implications for trade and development*; Report by the UNCTAD secretariat; 2009;TD/B/C.I/CRP.1, May 7, 2009. Available at http://www.unctad.org/en/docs/cicrp1_en.pdf (accessed month day, year).
- (46) Endresen, Ø.; Dalsøren, S. B.; Eide, M.; Sørgård, E.; Isaksen, I. S. A. The environmental impacts of increased international maritime shipping - Past trends and future perspectives. *OECD/ITF Global Forum on Transport and Environment in a Globalising World*; November 10–12, 2008, Guadalajara, Mexico.
- (47) Kasibhatla, P.; Levy II, H.; Moxim, W. J.; Pandis, S. N.; Corbett, J. J.; et al. Do emissions from ships have a significant impact on concentrations of nitrogen oxides in the marine boundary layer. *Geophys. Res. Lett.* **2000**, *27*, 2229–2232.
- (48) Chen, G.; Huey, L. G.; Trainer, M.; Nicks, D.; Corbett, J.; et al. An investigation of the chemistry of ship emission plumes during ITCT 2002. *J. Geophys. Res.* **2005**, *110*, D10S90. doi:10.1029/2004JD005236.
- (49) von Glasow, R.; Lawrence, M. G.; Sander, R.; Crutzen, P. J. Modelling the chemical effects of ship exhaust in the cloud-free marine boundary layer. *Atmos. Chem. Phys.* **2003**, *3*, 233–250, 2003.
- (50) Charlton-Perez, C. L.; Evans, M. J.; Marsham, J. H.; Esler, J. G. The impact of resolution on ship plume simulations with NO_x chemistry. *Atmos. Chem. Phys.* **2009**, *9*, 7505–7518.
- (51) Kim, H. S.; Song, C. H.; Park, R. S.; Huey, G.; Ryu, J. Y. Investigation of ship-plume chemistry using a newly-developed photochemical/dynamic ship-plume model. *Atmos. Chem. Phys.* **2009**, *9*, 7531–7550.
- (52) Eyring, V.; Stevenson, D. S.; Lauer, A.; Dentener, F. J.; Butler, T.; et al. Multi-model simulations of the impact of international shipping on atmospheric chemistry and climate in 2000 and 2030. *Atmos. Chem. Phys.* **2007**, *757*, 780.
- (53) Franke, K.; Richter, A.; Bovensmann, H.; Eyring, V.; Jöckel, P.; et al. Ship emitted NO₂ in the Indian Ocean: comparison of model results with satellite data. *Atmos. Chem. Phys.* **2009**, *9*, 7289–7301.
- (54) Collins, W. J.; Sanderson, M. G.; Johnson, C. E. Impact of increasing ship emissions on air quality and deposition over Europe by 2030. *Meteorol. Z.* **2009**, *18*, 25–39.
- (55) International Maritime Organisation (IMO). *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*; 2008 (Revised MARPOL Annex VI), MEPC 58/23/Add.1 ANNEX 13 RESOLUTION MEPC.176(58), Adopted on October 10, 2008.
- (56) Fuglestvedt, J. S.; Berntsen, T.; Eyring, V.; Lee, D. S.; Sausen, R.; Isaksen, I. S. A. Shipping emissions: From cooling to warming of climate - and reducing impacts on health. *Environ. Sci. Technol.* **2009**, *43*, 9057–9062.

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