Environ. Sci. Technol. **2009,** *43,* 5592–5598

Assessment of Near-Future Policy Instruments for Oceangoing Shipping: Impact on Atmospheric Aerosol Burdens and the Earth's Radiation Budget

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Received November 14, 2008. Revised manuscript received June 9, 2009. Accepted June 18, 2009.

We apply the global climate model ECHAM5/MESSy1-MADE with detailed aerosol and cloud microphysics to study the impact of shipping on tropospheric aerosol burdens, clouds, and the radiation budget for four near-future ship emission policy scenarios for the year 2012. We compare a "*No Control*" scenario with global sulfur limits and regionally applied reductions. We show that, if no control measures are taken, near surface sulfate increases by about 10−20% over the main transoceanic shipping routes from 2002 to 2012. A reduction of the maximum fuel sulfur (S) content allowed within 200 nautical miles of coastal areas ("global emission control areas") to 0.5% or 0.1% (5000 or 1000 ppm S, respectively) results in a distinctive reduction in near surface sulfate from shipping in coastal regions compared with the year 2002. The model results also show that if emissions of nitrogen oxides (NO_x) remain unabated, a reduction of the fuel sulfur content favors a strong increase in aerosol nitrate (NO_3) which could counteract up to 20% of the decrease in sulfate mass achieved by sulfur emission reductions. The most important impact of shipping on the radiation budget is related to the modification of low maritime stratus clouds resulting in an increased reflectivity and enhanced shortwave cloud forcing. The direct aerosol effect from shipping is small. Our study shows that one can expect a less negative (less cooling) radiative forcing due to reductions in the current fuel sulfur content of ocean-going ships. The global annual average net cloud forcings due to shipping (year 2012) are in the range of −0.27 to −0.58 W/m^2 with regional cooling occurring most over the remote oceans.

Introduction

International shipping contributes significantly to emissions from the transportation sector (*1, 2*), thereby affecting the chemical composition of the atmosphere, climate and regional air quality, and health. Key compounds emitted are sulfur and nitrogen oxides (SO_x and NO_x, respectively) and particulate matter (PM) as well as greenhouse gases such as carbon dioxide (CO_2). Recent studies have shown that ship emissions could have a large impact on the Earth's radiation budget, particularly by altering the albedo of low maritime stratus clouds (*3, 4*). Corbett et al. (*5*) showed that PM from shipping has a negative impact on the health of the population in coastal areas, resulting in an increase in, for instance, heart attacks, lung cancers, hospital admissions, and premature mortality. World sea trade and hence the emissions from shipping are expected to grow rapidly throughout the next years (e.g. ref *6*). Since ship exhaust gases contribute to the worldwide pollution of air and sea, ships are now facing an increasing number of rules and regulations as well as voluntary appeals from international, national, and local legislators.

Emissions from ships in international trade are regulated by the International Maritime Organization (IMO) under Annex VI of the International Convention for the Prevention of Pollution from Ships (MARPOL) 73/78 (*7*). The Marine Environment Protection Committee (MEPC) of IMO has recently approved proposed amendments to the MARPOL Annex VI regulations to reduce harmful emissions from ships. The changes will focus on a progressive reduction in SO_x emissions from ships, with the maximum allowed fuel sulfur content reduced initially from the current 4.5% (45,000 ppm S) to 3.5% (35,000 ppm S) in 2012 and then progressively to 0.5% (5000 ppm S) by 2020. In addition, the sulfur limits in Emission Control Areas (ECA) will be reduced from the current level of 1.5% (15,000 ppm S) to 1% (10,000 ppm S) in 2010 and further reduced to 0.1% (1000 ppm S) in 2015.

Here we assess the impact of a 2012 "*No Control*" scenario with three emissions control scenarios on aerosol burdens and the Earth's radiation budget; a related paper evaluates potential health benefits from reducing ship emissions (*8*). The approach we use is similar to that described by Corbett et al. (*5*). Holding constant all parameters except those emissions mitigated by fuel switching, we isolate the impact on the Earth's radiation budget and the percent change in tropospheric aerosol burdens that can be expected from proposed action. We employ the same geospatial inventory (*9*), atmospheric model, and uncertainty factors as reported in ref *5*. This approach allows us to compare control scenarios with previous work assessing baseline impacts. We present our results for each mitigation case and show relative changes compared with the *No Control* case.

Near-Future Policy Instruments. This study compares a 2012 "*No Control*" scenario with three ship emissions control scenarios for the year 2012. The first step of the analysis was to create shipping emissions inventories for the year 2002 and the various scenarios in 2012. PM emissions estimates were obtained based on global representation of ship emissions inventories of sulfur dioxide (SO_2), black carbon (BC), and particulate organic matter (POM). We calculated two sets of 2002 emissions inventories, following the geographical distribution of reported ships positions from the

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10.1021/es900922h CCC: $40.75

Published on Web 07/13/2009

TABLE 1. Summary of Ship Emission Scenarios Showing Annual Emission Totals of Particulate Matter and Trace Gases from Shipping in Tg yr^{-1} in 2002 and in 2012[a]

ship emission scenario	fuel sulfur content	NO_x ($TgNO_2$)	SO_2 (Tg)	primary SO_4 (Tg)	CO (Tg)	BC (Tg)	POM (Tg)
			Global Emission (Adjustment Factors in Parentheses)				
2002	2.7%	16.40	9.20	0.350	1.08	0.070	0.710
			ECA Sulfur Reduction Scenarios				
2012 *No Control*	2.7%	24.50 (100%)	13.75 (100%)	0.523 (100%)	1.61 (100%)	0.105 (100%)	1.061 (100%)
2012 *Coastal 0.5*	0.5%	24.50 (100%)	7.15 (18.5%)	0.272 (18.5%)	1.61 (100%)	0.105 (100%)	0.637 (32.1%)
2012 *Coastal 0.1*	0.1%	24.50 (100%)	5.96 (3.7%)	0.227 (3.7%)	1.61 (100%)	0.105 (100%)	0.561 (20.0%)
			Global Sulfur Reduction Scenario				
2012 *Global 0.5*	0.5%	24.50 (100%)	2.55 (18.5%)	0.097 (18.5%)	1.61 (100%)	0.105 (100%)	0.341 (32.1%)

[a] Parentheses indicate the reductions assumed due to lower fuel sulfur contents.

Automated Mutual-assistance Vessel Rescue System (AMVER) and the International comprehensive Ocean-Atmosphere Data Set (ICOADS). Differences between the two data sets can be significant for individual regions such as the latitude belt 40°N-60°N and arise from different spatial and temporal sampling biases. Both data sets and their differences are discussed in detail by Wang et al. (*9*) and within their Supporting Information. Both ship emission inventories consider the seasonal cycle by specifying monthly mean emission totals as well as geographical distributions for each month. The annual emission totals of both ship emission inventories have been normalized to the same value, which allows us to assess the uncertainty stemming from the spatial distribution and seasonal cycle of the two ship emission inventory sets. These emissions are overlaid on an inventory without ship traffic to obtain the increased PM pollution attributed to ships operations.

Ship emissions for the 2012 *No Control* scenario were estimated from current inventories using a uniform global annual compound growth rate of 4.1% resembling the average annual growth rate for the last two decades (*10*). The *No Control* case used the present day average sulfur content of 2.7% (27,000 ppm S) (*11*) assuming residual fuel properties typical for particulate organic matter (POM). In the 2012 *No Control* scenario all compounds of ship emissions increase by approximately 50% relative to 2002.

Scenarios 2012 *Coastal 0.5* and 2012 *Coastal 0.1* represent impacts associated with reduced ship PM emissions corresponding to a 0.5% (5000 ppm S) and 0.1% (1000 ppm S) sulfur content for marine fuels within 200 nautical miles (nm) off coastlines in 2012, respectively. These fuel sulfur content limits conform with current international regulations to reduce emissions from ships in already established ECAs. Our scenarios include all coastal areas corresponding to a global ECA zone. Scenario 2012 *Global 0.5* is designed to assess global versus coastal action; this scenario explores impacts associated with reduced ship PM emissions corresponding to a 0.5% sulfur concentration for marine fuels globally. Such a scenario will come into force by 2020.

Our scenarios reduce inventory emissions of sulfur and particulate organic matter to correspond with properties for different marine fuel types. Control scenarios used sulfur contents of 0.1% S (~4% of *No Control* within coastlines) and 0.5% S (~19% of *No Control* globally), respectively. We reduced POM emissions by ~68% for marine distillate fuel oil (MDO) at 0.5% S and by ~80% for marine gas oil (MGO) at 0.1% S, respectively, in the control scenarios to correspond to typical differences among residual and distillate fuel POM measurements (*12*). This decreases SO_2 and primary sulfate (SO_4) by 81.5% in the *Coastal 0.5* and *Global 0.5* scenarios relative to the 2012 *No Control* scenario and by 96.3% in the *Coastal 0.1* scenario. Organic matter decreases by 67.9% relative to the 2012 *No Control* scenario in *Coastal 0.5* and *Global 0.5* and by 80% in *Coastal 0.1*. All other compounds (NO_x, carbon monoxide (CO), BC) remain unchanged at 100%. A summary of the scenario specifications is given in Table 1.

Model and Model Simulations. We applied the model system ECHAM5/MESSy1-MADE (hereafter referred to as E5/M1-MADE) as described in ref *4* to study the impacts on the Earth's radiation budget under the different ship emission scenarios. The core of E5/M1-MADE consists of the general circulation model (GCM) ECHAM5 (*13*) within the framework of the Modular Earth Submodel System MESSy (*14*). The aerosol submodel MADE (*15*) takes into account detailed aerosol microphyical processes. The aerosols are interactively coupled to the chemistry submodel MECCA (*16*) as well as the GCM's cloud microphysics (*17, 18*) and radiation scheme. Additional details on the model are given in the Supporting Information.

All model simulations presented in this study have been conducted in T42 horizontal resolution corresponding to about 2.8° × 2.8° longitude by latitude. The vertical dimension is resolved by 19 vertical nonequidistant layers from the surface up to 10 hPa. The impact of shipping on aerosols, clouds, and the radiation budget is estimated by calculating differences between model simulations with and without ship emissions. Nudging of model dynamics to operational analysis data from the European Centre for Medium-Range Weather Forecasts (ECMWF) from 1999 to 2004 allows us to obtain significant results within the 6 model years analyzed for each experiment. This meteorological forcing is also applied to the 2012 emission scenarios. Sea surface temperature and sea ice coverage are prescribed according to the ECMWF operational analysis data. We consider the signal from shipping to be significant if the t test applied to the annual mean values of the individual years provides significance at a confidence level of 99%.

Ship emissions are estimated from the fleet activity taking into account emissions from cargo and passenger ships, totaling 9.2 Tg SO_2 per year in 2002 and 2.6 to 13.8 Tg SO_2 per year in 2012 depending on the ship emission scenario. The annual emission totals for the trace gases and aerosol compounds considered are summarized in Table 1. Emissions of primary aerosols such as BC, POM, or mineral dust as well as SO_2 from all other sources are from the AeroCom data set for the year 2000 (*19*). Primary particles from shipping (BC, POM, SO_4) are assumed to be in the size-range of the Aitken mode, which is typically being observed for fossil fuel combustion processes. A flat fraction of 2.5% of the SO_2 emissions is assumed to be primary SO_4 (*19*). We apply these nonship emissions also to the 2012 model simulations. To estimate an upper limit for uncertainties introduced by using year 2000 background emissions for our 2012 scenarios, we conducted a sensitivity simulation for the ICOADS 2012 *No Control* scenario using anthropogenic nonship emissions scaled uniformly with growth rates projected by the Intergovernmental Panel of Climate Change (IPCC) from 2000 to 2012 for a scenario with fast growing nonship emissions (SRES

TABLE 2. Relative Contribution of Shipping to the Tropospheric Global Annual Average Burdens of Sulfate (SO_4), Ammonium (NH_4), Nitrate (NO_3), Black Carbon (BC), and Particulate Organic Matter (POM) in % for the 10 Ship Emission Scenarios Considered[a]

ship emission scenario	SO_4	NH_4	NO_3	BC	POM
		Geospatial Distribution AMVER			
2002	3.4% (40.9)	2.4% (6.8)	5.1% (5.4)	0.9% (0.9)	1.2% (10.7)
2012 *No Control*	5.2% (62.5)	3.4% (9.9)	6.7% (7.3)	1.4% (1.4)	1.8% (15.4)
2012 *Coastal 0.5*	2.2% (25.6)	1.4% (4.0)	7.9% (8.7)	1.5% (1.5)	1.1% (9.3)
2012 *Coastal 0.1*	1.6% (19.1)	1.0% (2.9)	8.1% (8.9)	1.6% (1.6)	1.0% (8.3)
2012 *Global 0.5*	1.1% (13.2)	0.7% (2.0)	8.7% (9.7)	1.6% (1.6)	0.6% (5.6)
		Geospatial Distribution ICOADS			
2002	2.9% (34.6)	2.1% (5.9)	6.2% (6.7)	0.9% (0.9)	1.1% (10.0)
2012 *No Control*	4.3% (51.0)	2.8% (8.0)	8.2% (9.0)	1.3% (1.3)	1.7% (14.6)
2012 *Coastal 0.5*	1.9% (22.7)	1.3% (3.7)	9.0% (10.1)	1.5% (1.5)	1.0% (9.1)
2012 *Coastal 0.1*	1.5% (17.1)	1.0% (2.8)	9.3% (10.3)	1.5% (1.5)	0.9% (8.0)
2012 *Global 0.5*	0.9% (10.1)	0.6% (1.8)	9.8% (11.0)	1.5% (1.5)	0.6% (5.0)

[a] Numbers in brackets are corresponding absolute changes in Gg (10^6 kg).

A1FI (*20*)). This sensitivity simulation showed that (absolute) changes in global average aerosol burdens as well as in the direct and indirect aerosol effects from shipping are usually less than 5% for both types of background emissions, but local changes can be up to 25% in some regions. This is in the range of current uncertainties in present-day emissions inventories for trace gases especially for particulate matter. However, the relative contribution of SO_4 from shipping, for instance, decreases for the ICOADS 2012 *No Control* scenario from 4.3% (year 2000 background emissions) to 3.9% (year 2012 background emissions) and that of NO_3 from 8.0% to 7.3%. A detailed discussion of this uncertainty is provided in the Supporting Information.

Results

$PM_{2.5}$ (particulate matter) refers to the total mass concentration of particles less than 2.5 μm in diameter, which is commonly used by regulatory agencies such as the United States Environmental Protection Agency (EPA) to define air quality standards. Particles of this size pose a health concern because they can be inhaled into and accumulate in the respiratory system. Comparing results of each model simulation with and without ship inventories, we quantify changes in ambient concentrations of $PM_{2.5}$ and the Earth radiation budget due to marine shipping in the various 2012 policy scenarios compared with the year 2002.

Near Surface Aerosol Mass Concentrations. For the *No Control, Coastal 0.5*, and *0.1* scenarios aerosol sulfate resulting from oxidation of SO_2 is the dominant aerosol compound from shipping (in terms of absolute mass), contributing between 1.5% (ICOADS, 2012 *Coastal 0.1*) and 5.2% (AMVER, 2012 *No Control*) to the total tropospheric sulfate burden. The second largest contribution to the total mass of aerosols from shipping stems from organic matter. The amount of aerosol nitrate from shipping depends strongly on the geospatial distribution used for the ship emissions, with relative contributions to the tropospheric aerosol NO_3 burden between about 5.1% (AMVER, 2002) and 9.8% (ICOADS, 2012 *Global 0.5*). For the ICOADS *Global 0.5* scenario, aerosol nitrate is the most important aerosol compound from shipping in terms of total mass. The model results indicate that on global annual average, aerosol nitrate increases with decreasing fuel sulfur content. A decrease in the fuel sulfur content corresponds to a decrease in the tropospheric SO_4 burden from shipping, which in turn decreases the amount of ammonia required to neutralize the sulfate (formation of ammonium sulfate). If less ammonia is bound by sulfate, the occurrence of free excess ammonia, which is required to form ammonium nitrate in the model, becomes more frequent. Thus, a decreasing fuel sulfur content with NO_x emissions unchanged allows for higher aerosol nitrate concentrations. In contrast to the other aerosol compounds (SO_4, BC, POM), aerosol nitrate shows a strong dependence on the geospatial distribution in the model implying a high level of uncertainty. Compared with the AMVER geospatial distribution, the ICOADS geospatial distribution favors greater formation of aerosol nitrate.

Table 2 summarizes the contribution of shipping to the total tropospheric aerosol burdens for the two geospatial distributions and the emission scenarios considered. Figure 1 shows the relative changes of the annual mean near surface sulfate mass concentrations for all 2012 emission scenarios compared with the corresponding 2002 cases. If no control measures are taken and the average fuel sulfur content in marine heavy fuels remains at its present level of about 2.7% (*11*), near surface sulfate (all sources) is expected to increase by about 10−20% over the main shipping routes in the Pacific, Atlantic, and Indian Ocean compared with the year 2002. In general, the relative increases in near surface sulfate are more pronounced in the ICOADS emission scenarios than in the AMVER scenarios, which can be attributed to the differences between the geospatial distribution of the ship emissions. Results from scenarios *Coastal 0.5* and *0.1* show a distinctive reduction in near surface sulfate in coastal waters as well as in the coastal regions of western North America, Europe, and North Africa by about 10−20%. The reduction of the fuel sulfur content within 200 nm off-coast also decreases sulfate from shipping in the coastal regions of the nations along the rim of the Western Pacific and Japan. A global reduction of the fuel sulfur content from 2.7% to 0.5% shown in scenario *Global 0.5* will lower the near surface sulfate mass concentrations from shipping by up to 50% above the oceans compared with 2002 levels. The reduction of sulfate in coastal areas in *Global 0.5* is more pronounced than in *Coastal 0.5*. These differences within coastal areas can be attributed to the decreased long-range transport of sulfate, i.e. advection from high seas to coastal waters, in case of a global reduction in fuel sulfur content (*Global 0.5*) compared with a reduction within 200 nm off-coast only (*Coastal 0.5)*.

If NO_x emissions remain unabated, the reduction in fuel sulfur content results in an increase in aerosol nitrate from shipping as more free ammonium is available for formation of ammonium nitrate (Figure 2). Compared with 2002, near surface aerosol nitrate concentrations increase by about 5−10%, in particular in the coastal regions of Europe and North Africa. While changes in near surface aerosol nitrate remain small over the open ocean along the main transoceanic shipping routes due to high sulfate concentrations in *Coastal 0.1* and *Coastal 0.5*, respectively, near surface

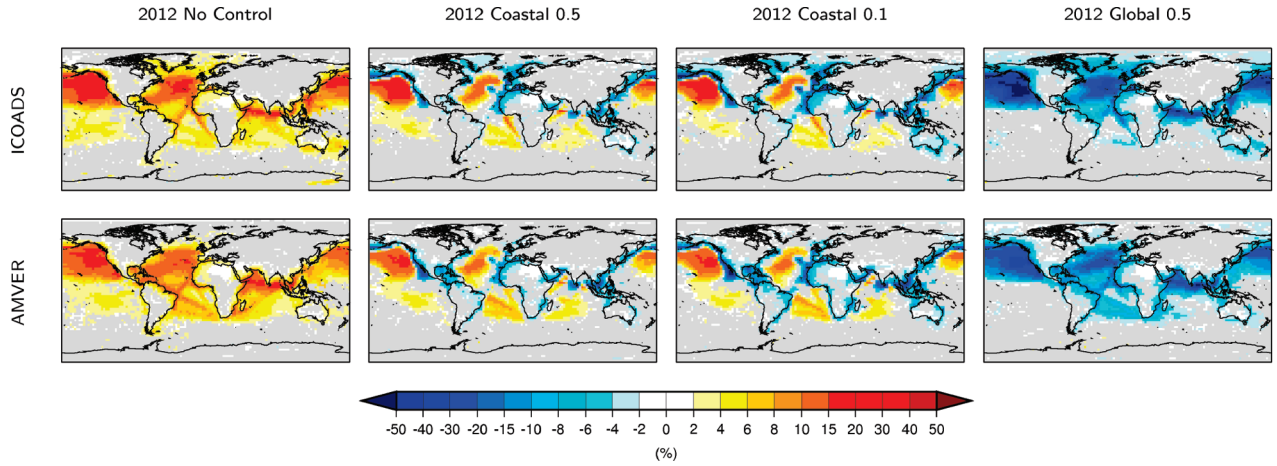

FIGURE 1. Simulated relative changes (annual mean) in % of near surface sulfate (SO_4) mass concentration in fine particles due to shipping compared with 2002. From left to right: ship emission scenario 2012 *No Control*, 2012 *Coastal 0.5*, 2012 *Coastal 0.1*, and 2012 *Global 0.5*. Upper row: ICOADS spatial distribution, lower row: AMVER spatial distribution. Differences that are not significant at 99% confidence level compared with their interannual variability are masked out in gray.

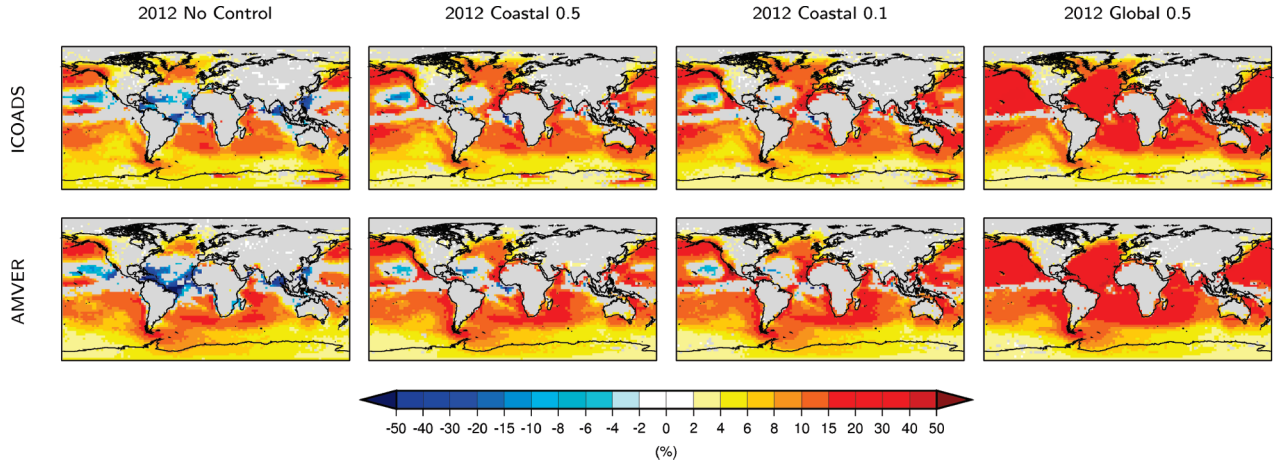

FIGURE 2. As in Figure 1 but for total aerosol nitrate (NO_3) mass concentration in fine particles.

aerosol nitrate increases by about 20−30% in *Global 0.5* with local changes up to about 40% on annual average.

The 2012 *Coastal 0.5* scenario shows $PM_{2.5}$ in coastal areas which are about the same level or slightly below the 2002 scenario, whereas the *Coastal 0.1* scenario shows decreases up to 8% in Western Europe compared with the year 2002. Because of a reduction in long-range transport of aerosol particles from shipping emitted over the open oceans, $PM_{2.5}$ concentrations in the coastal areas are up to 0.03 μg m^{-3} lower along the west coast of North America in *Global 0.5* than in *Coastal 0.5*. Despite increasing ship traffic, $PM_{2.5}$ concentrations from shipping remain at or below 2002 levels in all three future emissions control scenarios particularly in the coastal regions of North America and Europe. Additional details on changes in $PM_{2.5}$ can be found in the Supporting Information.

Direct Aerosol Forcing. We calculate the direct aerosol forcing as the differences between top of the atmosphere (ToA) radiative fluxes in the solar spectral range. These changes are mainly related to enhanced scattering of solar radiation by sulfate, ammonium, nitrate, and associated aerosol liquid water. A discussion on the uncertainties of separating direct and indirect aerosol effect as well as a table summarizing the global annual mean direct aerosol forcing due to shipping for each of the ten ship emission scenarios can be found in the Supporting Information. The global annual average clear sky direct aerosol forcing amounts to −28 mW/m^2 in the year 2002 for both, the AMVER and the ICOADS geospatial distributions. The corresponding all sky direct aerosol forcings amount to about −11 mW/m^2. These are much lower than the clear sky values, because most particles from shipping are emitted in cloudy areas below the cloud tops and thus contributing little to the global annual average. The 2012 *No Control* scenario shows an increase in the all sky direct aerosol forcing of about 10−20% compared with the year 2002. A reduction of the fuel sulfur content in the ECA areas (*2012 Coastal 0.5* and *0.1*) has only a small impact on the global annual average all sky direct aerosol forcings, which remain in the range −12 and −14 mW/m^2. The reduction in sulfur and POM emissions by about 80% and 70%, respectively, in *Global 0.5* reduces the annual average all sky direct aerosol forcing to −4 to −6 mW/m^2. This is slightly more than what could be expected from solely the reduction in sulfur emissions assuming that sulfate accounts for about 75% of the direct aerosol effect from shipping (*4*). Again, the decrease in sulfate due to reduction in SO_2 emissions is partly compensated by increasing aerosol nitrate and associated aerosol water in *Coastal 0.5* and *0.1* and most pronounced in the *Global 0.5*. In all 2012 scenarios emissions of NO_x (precursor gas for aerosol nitrate) remain at high levels counteracting the reduced scattering of solar radiation by sulfate and POM particles compared with the year 2002.

Indirect Aerosol Forcing. Emissions from shipping have the potential to increase the number of condensation nuclei available for cloud formation. Geographical regions with a frequent high amount of low maritime stratus clouds are most susceptible for modifications of the cloud microphysical

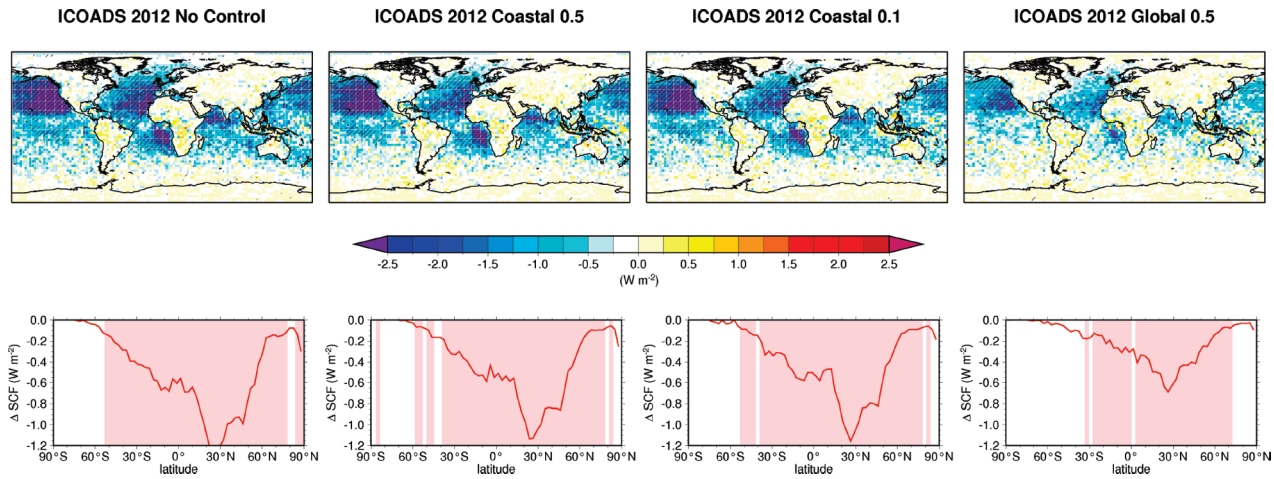

FIGURE 3. Multiyear average of simulated changes in shortwave cloud forcing due to shipping for ICOADS emission scenarios at the top of the atmosphere (ToA) in W m^{-2}. From left to right: 2012 *No Control*, 2012 *Coastal 0.5*, 2012 *Coastal 0.1*, and 2012 *Global 0.5*. Upper row shows the geographical distribution, lower row zonal averages. Hatched areas (upper row) and light-red shaded areas (lower row) show differences which are significant at the 99% confidence level compared with the interannual variability. The corresponding figure for the AMVER emissions scenarios can be found in the Supporting Information (Figure S2).

properties. Such regions coinciding with high ship traffic density are located over the Pacific west of North America, the Atlantic west of southern Africa, and over the northeastern Atlantic (*4*). Additional condensation nuclei from shipping increase the number of cloud droplets, while the liquid water content is only slightly affected. This results in smaller cloud droplets increasing the reflectivity of the marine stratus clouds (first indirect aerosol effect). The increased reflectivity of the marine stratus clouds results in an increased shortwave cloud forcing, calculated as the difference in the all sky shortwave minus clear sky shortwave radiative fluxes at ToA. The impact of shipping on the thermal emissivity of the marine stratus clouds is low as the temperature differences between the cloud tops and the sea surface temperatures are only small. Changes in cloud microphysical and optical properties as well as uncertainties associated with an assessment of the indirect aerosol effect (from shipping) are discussed in Lauer et al. (*4*).

The model results show no significant changes in the longwave cloud forcings (thermal spectral range) due to shipping. Changes in the precipitation formation efficiency (second indirect aerosol effect) due to shipping affecting geographical precipitation patterns and total precipitation are too small compared with their interannual variability to provide a statistically significant signal within the 6-year period of our analysis. Longer model integrations are needed to study the second indirect aerosol effect from shipping. Figure 3 shows the geographical distributions of the 6-year annual average changes in ToA shortwave cloud forcing and the corresponding zonal means for all 2012 ICOADS scenarios (see Figure S2 in the Supporting Information for the AMVER geospatial distribution).

On average, maximum changes in shortwave cloud forcing are found at about 30°N. In this latitude region, high ship traffic coincides with persistent maritime stratocumulus decks, which are highly susceptible for additional cloud condensation nuclei from shipping. Cold sea surface temperatures in combination with warm, dry air aloft lead to the formation of these subtropical stratocumulus decks. In contrast to the ICOADS geospatial distributions, the changes in the shortwave cloud forcing of the AMVER scenarios show a second maximum at about 15°S. This results from enhanced scattering of solar radiation by maritime stratus clouds off the west coast of southern Africa due to higher emissions in this region compared with the ICOADS scenarios. Despite these local differences between the two geospatial distributions (AMVER and ICOADS), the global annual average changes in the net cloud forcing (sum of shortwave and longwave cloud forcing) are almost identical, amounting to −0.43 and −0.44 W/m^2 for the year 2002 and −0.57 and −0.58 W/m^2 (2012 *No Control*) to −0.27 W/m^2 (*Global 0.5*). The impact of ship emissions on the net cloud forcing over the remote oceans is only slightly affected in the *Coastal 0.5* and *0.1* scenarios but reduced on average by about 50% in the *Global 0.5* scenario compared with the 2012 *No Control* scenario. This reduction in global annual average changes of the net cloud forcing due to shipping in the *Global 0.5* scenarios is less than expected from Lauer et al. (*4*), who found that SO_2 emissions are responsible for about 75% of the total indirect aerosol forcing from ship emissions. This could be related to the strong increase in aerosol nitrate, which compensates for some of the decrease in aerosol sulfate. Table 3 summarizes the global annual mean indirect aerosol forcing due to shipping.

TABLE 3. Changes in Global Annual Average Net Cloud Forcing at the Top of the Atmosphere Due to Indirect Aerosol Effect from Shipping in W m^{-2}

ship emission scenario	Atlantic (75°W-15°E, 40°S-60°N)	Pacific (120°E-80°W, 40°S-60°N)	global
	Geospatial Distribution AMVER		
2002	−0.965	−0.829	−0.430
2012 *No Control*	−1.268	−1.084	−0.569
2012 *Coastal 0.5*	−1.086	−0.909	−0.471
2012 *Coastal 0.1*	−1.035	−0.855	−0.449
2012 *Global 0.5*	−0.613	−0.518	−0.269
	Geospatial Distribution ICOADS		
2002	−0.844	−0.735	−0.439
2012 *No Control*	−1.136	−0.974	−0.578
2012 *Coastal 0.5*	−0.969	−0.807	−0.482
2012 *Coastal 0.1*	−0.921	−0.776	−0.459
2012 *Global 0.5*	−0.515	−0.436	−0.265

Discussion

Our model results show, that if no control measures are taken, near surface sulfate increases by about 10−20% over the main shipping routes in the Pacific, Atlantic, and Indian Ocean in 2012 compared with the year 2002 contributing up

to 5.2% (63 Gg) to the total tropospheric sulfate burden. Reduction of the fuel sulfur content in the ECA area results in a distinctive reduction in near surface sulfate from shipping in the coastal regions of North America, Europe, and North Africa by about 10−20%. The *Global 0.5* scenarios reduce near surface sulfate over the oceans by up to 50% compared with 2002. Because of a decreased long-range transport of sulfate in *Global 0.5*, SO_4 concentrations in coastal areas are lower in the *Global 0.5* scenario than in the *Coastal 0.5* scenarios. Aerosol nitrate has a strong dependence on the geospatial distribution in the model implying a high level of uncertainty. As sulfate concentrations drop, more excess ammonia becomes available to form ammonium nitrate which has otherwise been bound by sulfate forming ammonium sulfate. Formation of aerosol nitrate is greater in the ICOADS model runs than for the AMVER geospatial distribution.

The spatial changes in $PM_{2.5}$ due to shipping are dominated by SO_4 and POM. $PM_{2.5}$ is commonly used by regulatory agencies to define air quality standards. In the *No Control* scenario, the increase in $PM_{2.5}$ from the year 2002 to 2012 is in the range of 1−4% annually for most coastal areas of North America and Western Europe, whereas the 2012 *Coastal 0.5* scenario shows similar $PM_{2.5}$ concentrations as the 2002 scenario. *Global 0.5* and *Coastal 0.1* show lower $PM_{2.5}$ concentrations in coastal areas compared with 2002 levels.

Changes in aerosol composition, particle numbers, and particle sizes due to shipping result in modified ToA radiative fluxes (direct aerosol effect) due to increased scattering and absorption of solar radiation. Model results for the 2012 *No Control* scenarios show an increase in the all sky direct aerosol forcing of about 10−20% compared with the year 2002. Our model results indicate that a reduction of the fuel sulfur content within 200 nm off-coast (*Coastal 0.5* and *Coastal 0.1*) has only little impact on the global annual average all sky direct aerosol forcings. The decrease in SO_4 is partly compensated by increasing NO_3 and associated aerosol liquid water counteracting the reduction in scattering by sulfate and POM aerosols compared with the year 2002.

The additional number of cloud condensation nuclei from shipping results in an increased number of smaller cloud droplets, which can reflect solar radiation more efficiently than unmodified clouds. The model results suggest that the persistent maritime stratocumulus decks in the subtropics are highly susceptible for the additional cloud condensation nuclei from ship emissions. Changes in the clouds precipitation formation efficiency and total precipitation are too small to provide a statistically significant signal from shipping within the period of our analysis. The two geospatial distributions (AMVER, ICOADS) show local differences, for instance, off the west coast of southern Africa, but the global annual average changes in the net cloud forcing due to shipping are small. Compared with 2002, the net cloud forcing of −0.57 to −0.58 W/m^2 in the 2012 *No Control* scenarios is increased by about 30%. Model results for the *Coastal 0.5* and *Coastal 0.1* scenarios indicate a larger (absolute) net cloud forcing of −0.45 to −0.48 W/m^2 compared with 2002 (−0.43 to −0.44 W/m^2). On global annual average, net cloud forcing due to shipping amounts to −0.27 W/m^2 in the *Global 0.5* scenarios which is less than calculated for the 2002 cases. This reduction in global annual average changes of the net cloud forcing due to shipping in the *Global 0.5* scenarios is less than expected from previous studies which suggested that sulfate is responsible for about 75% of the total indirect aerosol forcing from ship emissions. This can be explained by the strong increase in aerosol nitrate, which compensates for some of the decrease in aerosol sulfate.

The largest individual contribution to the total radiative forcing of shipping is related to the modification of low maritime stratus clouds resulting in an increased reflectivity and enhanced shortwave cloud forcing. Despite significant local differences, the global annual average cloud forcing due to shipping does not depend strongly on the geospatial distribution for the two cases studied (AMVER, ICOADS). This is also the case for the direct aerosol forcing. Our model results suggest that a reduction in the ECA areas 200 nm off coast is less efficient than a global reduction of ship emissions, as a major contribution to the global averages comes from modified cloud properties over the remote oceans. The ECA areas could be efficient to reduce the near surface mass concentrations of aerosols in coastal regions but can be counteracted by increasing aerosol nitrate concentrations if NO_x emissions remain unabated. An efficient strategy for emission reduction from shipping should consider both SO_2 and NO_x.

In our complementary paper (*8*), we show that without any control strategies in place, one could expect about 83,000 to 90,000 premature deaths annually by 2012 due to cardiopulmonary illness and lung cancer. These numbers could be reduced by 31,000−36,000 for the *Coastal 0.5* scenario and 42,000−45,000 for the *Coastal 0.1* case.

Based on our study, policies to reduce the current fuel sulfur content of ocean going ships will produce less negative radiative forcing (less cooling). CO_2 remains in the atmosphere for a long time and will continue to have a warming effect long after its emission. In contrast, sulfate has a residence time in the atmosphere of approximately 1 week, and the climate response from sulfate is of the order of decades while that of CO_2 is of the order of centuries (*21, 22*). Possible cancellation of positive and negative radiative forcing remains a topic of scientific research, but our model shows that an asymmetry exists among relatively cooler marine regions and land-based populated regions where warming impacts are unabated by ship aerosol effects. This strongly suggests that simple cancellation of global means is inappropriate as a measure of offsetting effects, and a more comprehensive assessment metric is required. While the control of NO_x, SO_2, and particle emissions from ships will have beneficial impacts on human health, acidification, and eutrophication, CO_2 reductions from all sources, including ships and other freight modes, are urgently required to reduce global warming.

Acknowledgments

This work was supported by the German Helmholtz-Gemeinschaft Deutscher Forschungszentren (HGF) and the Deutsches Zentrum für Luft- und Raumfahrt (DLR) within the Young Investigators Group SeaKLIM as well as by JAMSTEC through its sponsorship of the International Pacific Research Center (IPRC) in the School of Ocean Earth Science and Technology (SOEST) at the University of Hawaii. We thank Johannes Hendricks (DLR, Germany) for helpful comments on the manuscript. We also thank the whole MESSy team of the Max Planck Institute for Chemistry in Mainz, Germany for their support and help and the Max Planck Institute for Meteorology in Hamburg, Germany for the development and provision of ECHAM5. We also kindly acknowledge the provision of MADE by the University of Cologne, Germany (RIU/EURAD-project). All model simulations were performed on the High Performance Computing Facility (HPCF) at the European Centre for Medium-Range Weather Forecasts (ECMWF) (IPRC Publication #618).

Supporting Information Available

Additional details on the model, changes in $PM_{2.5}$, and discussion of separation of direct and indirect aerosol effect as well as the impact of changes in anthropogenic nonship emissions between 2002 and 2012. This material is available free of charge via the Internet at http://pubs.acs.org.

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ES900922H