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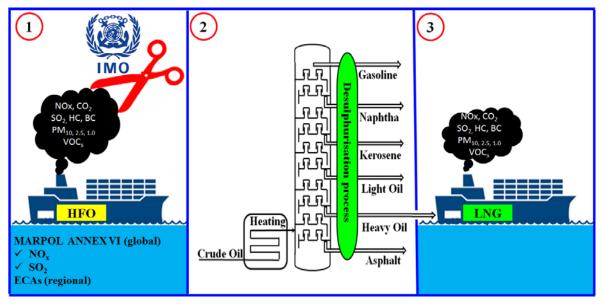
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1	Global Impacts of Recent IMO Regulations on Marine Fuel Oil Refining
2	Processes and Ship Emissions
3	
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19	Highlights
20	• Newly implemented IMO regulations require a significant reduction of ship emissions;
21	• Ship emission studies related to switching to low-sulphur fuels are thoroughly reviewed;
22	Refinery processes to produce low-sulphur fuels require extra energy consumption;
23	• Scope for new regulations to be added to reduce particle number and methane slip;
24	• Future marine fuel mix needs to be involved to meet fuel regulations and emission reduction;
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37 Abstract

38 This study presents an overview of the context and global impacts of recent International Maritime 39 Organization (IMO) regulations on the marine fuel oil refining industry, future marine fuel mix and ship 40 emissions. IMO limited marine fuel sulphur content in both Sulphur Emission Control Areas (SECAs) and 41 Nitrogen Oxide Emission Control Areas (NECAs) to 0.1% (wt. %) by 2015, and to 0.5% globally by 2020. It 42 is anticipated that the newly implemented IMO regulations will help to mitigate negative impact of ship 43 emissions on public health and environment. IMO regulations require significant changes to refineries to 44 increase the production of low sulphur fuels through a shift to distillates, use of novel deep desulphurization 45 techniques, or fuel blending. Changes to the refinery processes can bring forth increases in greenhouse gas 46 emissions and high capital investments. Alternative fuels will need to meet the required reduction of air 47pollutants and greenhouse gas emissions in coastal areas. Alternative marine fuels consisting of liquefied 48 nature gas (LNG) and biofuel may be suitable fuels to meet both targets. These two fuels are predicted to 49 account for 50% of shipping energy demand by 2050, while the remainder will still be supplied by 50 conventional heavy fuel oil (HFO)/marine gas oil (MGO). Switching to low sulphur fuels as a results of the 51 new IMO regulations has led to measureable reductions in ship emissions generally. This fuel switching also 52 resulted in changes in engine emission characteristics, especially on particulate matter chemical composition. 53

Key words: Shipping Emissions, IMO Regulations, Air Pollution, Oil Refinery, Heavy Fuel Oil, Alternative
 Marine Fuels.

56	Acronym Tab	le
57	bbl/d	Barrels of oil per day
58	BC	Black Carbon
59	CFR	Code of Federal Regulations
60	CH_4	Methane
61	CO	Carbon Monoxide
62	CO_2	Carbon Dioxide
63	CONCAWE	Oil Companies International Study Group for Conservation of Clean Air and Water in
64		Europe
65	EC	Elemental Carbon
66	ECAs	Emission Control Areas
67	EGR	Exhaust Gas Recirculation
68	ELTs	End-of-life tyres
69	EPA	United Sates Environmental Protection Agency
70	EU	European Union
71	FAME	Fatty Acid Methyl Esters
72	FCC	Fluid Catalytic Cracker
73	GHGs	Greenhouse Gases
74	HC	Hydrocarbons
75	HCO	Heavy Cycle Oil
76	HDS	Hydrodesulphurisation
77	HFO	Heavy Fuel Oil
78	HTL	Hydrothermal Liquefaction
79	IARC	International Agency for Research on Cancer
80	IMO	International Maritime Organization
81	LCO	Light Cycle Oil
82	LNG	Liquefied Natural Gas
83	LSHFO	Low-sulphur Heavy Fuel Oil
84	MARPOL	Marine Pollution Convention
85	MDO	Marine Diesel Oil
86	MGO	Marine Gas Oil
87	N_2O	Nitrous Oxide
88	NECAs	Nitrogen Oxide Emission Control Areas
89	NO _x	Nitrogen Oxides
90	OC	Organic Carbon
91	ODS	Oxidative Desulphurisation
92	PM	Particulate Matter
93	PN	Particle Number

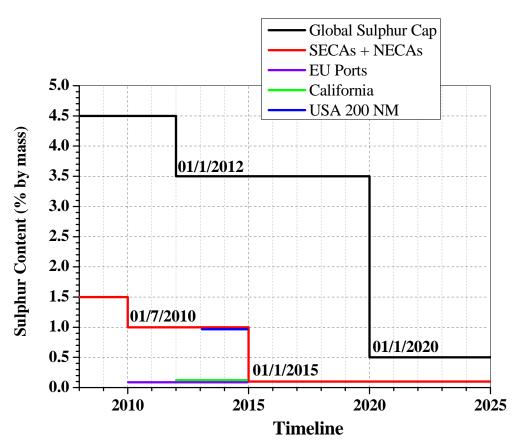
94	RMB30	Hybrids Fuel
95	SCR	Selective Catalytic Reduction
96	SECAs	Sulphur Emission Control Areas
97	SO _x	Sulphur Oxides
98	UN	United Nations
99	VOCs	Volatile Organic Compounds
100	WHO	World Health Organization

101 **1. The context of regulating global ship emissions**

102 **1.1. IMO regulations and current ship emission abatement systems**

103 IMO – the International Maritime Organization is a specialised branch of the United Nations (UN) which 104 issues global regulations on the safety, security and environmental performance of global shipping. In particular, Annex VI of the International Convention for the Prevention of Pollution from Ships - the Marine 105 106 Pollution Convention (MARPOL) was adopted by the 1973 Convention, and then modified by the 1978 107 Protocol with regard to limit the harmful impacts of ship emissions on air quality (IMO, 1997). These regulations were effective as of May 19th 2005, and aimed to reduce nitrous oxides (NO_x), sulphur oxides 108 109 (SO_x) and particulate matter (PM) from marine engines. Responding to the desire of some coastal nations for 110 further reduction of SO_x emissions from ships in their regions, Sulphur Emission Control Areas (SECAs) 111 have been instituted by applying provisions contained in the Regulation 14 of MARPOL Annex VI. SECAs 112 consist of the Baltic Sea area, the North Sea area, the North American region (containing the coastal sector of 113 the United States and Canada), and the United States Caribbean Sea areas (around Puerto Rico and the 114 United States Virgin Islands). These regulations limit the marine fuel sulphur content to 0.1% by mass in 115 SECAs by 2015, and to 0.5% globally by 2020 as shown in Fig. 1 (IMO, 2016a). These IMO regulations will 116 have a strong impact on marine oil refining, the future marine fuel market and ship emissions, which will be 117 discussed in the present paper.

118



119

120Fig. 1. IMO regulations on marine fuel sulphur content (this Figure is adopted from Cullinane and121Bergqvist (2014) and then updated from IMO (2016a))

122 In order to control emissions of SO_x and PM, fuel sulphur content must be reduced as it significantly 123 increases the emissions. SO_x emitted from marine diesel engines will form sulphate aerosols or sulphur-124 containing particles, which are the major component of PM. In addition, SO_x emissions are also able to 125 condense to form small-size particles in nucleation mode which may stick to coarse particles resulting in the 126 growth of particle composites. These secondary sulphate particles are an important contribution to land based 127 air pollution and also have a role in marine aerosol budget. The IMO therefore does not particularly limit PM 128 emissions directly, but regulates the sulphur-related portion of PM formation. It does this through the fuel 129 sulphur content requirements of Regulation 14 to Annex VI. The maximum value for sulphur content of 130 marine fuels used for vessels operating in SECAs was reduced from 1% to 0.1% on 1 January 2015 as seen 131 in Fig. 1. As an alternative to using low- sulphur fuel oil, approved systems for the abatement of emissions, 132 such as SO_x (wet and dry) scrubbers are likely to be dominantly used (Chu-Van et al., 2018b). In case these 133 systems for the abatement of emissions are used, the approval of the ship's administration (the flag state) is 134 needed. Scrubber systems play a role as filters of the engine exhaust gasses to remove SO_x by using washing-135 water that will be discharged directly into ocean (open loop), treated with chemicals and reused for a time 136 before discharging (closed loop), or treated through a hybrid mode of the above (Lindstad et al., 2017). A 137 cost efficiency analysis of three abatement options - retrofitting of scrubbers in ships using HFO, using 138 LSHFO, and using diesel – found that using HFO combined with scrubber installation gives the lowest cost. 139 However, this option encourages ships operating at high speeds which will increase fuel consumption and 140 greenhouse gas emissions (CO_2). In addition, CO_2 will be released through the neutralization process of the 141 acidic washing water after using at scrubbers (<u>Williams, 2010</u>). Therefore, reducing SO_x by using scrubber 142 systems may result in increase of CO₂.

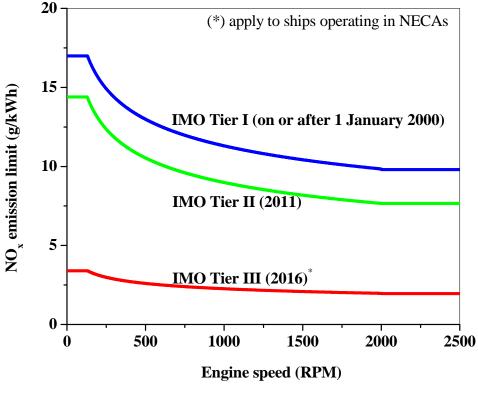
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144 Regulation 13 of MARPOL Annex VI controls NO_x emissions from both used and new marine diesel engines 145 that are over 130 kW output power. Different levers or tiers depend upon the ship construction date and 146 engine speed as presented in Fig. 2. It should be noted that Tier III requirements apply only to new vessels 147 operating in Nitrogen Oxide Emission Control Areas (NECAs) established to limit NO_x emissions, namely 148 the North American and the United States Caribbean Sea regions. It is well-known that NO_x emissions are a 149 function of various engine parameters such as combustion temperature, residence time of combustion 150 process, oxygen availability and ignition delay (Patel et al., 2014). NO_x emissions are mainly formed by 151 reaction of atmospheric nitrogen with oxygen through the Zeldovich mechanism which is strongly influenced 152 by the combustion temperature (Mwangi et al., 2015). Exhaust gas recirculation (EGR) arrangements and 153 selective catalytic reduction (SCR) systems seem to be a feasible abatement technology to achieve NO_x 154 reduction. By recirculating a part of exhaust gas to the scavenging air intake, EGR decreases oxygen 155 availability in the engine combustion chamber resulting in a reduction of peak combustion temperature. 156 However, this system can cause an increase in engine emissions such as black carbon (BC) due to the lack of 157 oxygen during engine combustion process (Nielsen et al., 2018). Using metal catalyst combined with 158 ammonia as a reduction element, SCR system reduces NO_x in engine exhaust by converting NO_x into N_2 and 159 water. To maintain required NO_x reduction, exhaust temperature needs to be kept as high as possible.

160 Therefore, engine low load or low exhaust temperature conditions normally resulted in less efficiency of

161 SCR system (Brynolf et al., 2014). It reveals that the practical challenges for the use of exhaust cleaning

162 systems are still existed.



163 164

Fig. 2. IMO regulations on NO_x emissions (IMO, 1999)

165

166 **1.2. Regional and national regulations**

167In addition to the regulations issued by the IMO, fuel sulphur content limits and ship emission regulations 168 have been introduced by other countries and jurisdictions such as the State of California, United States 169 Environmental Protection Agency (US EPA) and the European Union (EU) in order to further reduce the 170 negative impacts of emissions from ships on air environment. As can be seen in Fig. 1, from 1 January 2010, 171European directive 2005/33/EC demandes all ships at berth or anchoring in European ports to use fuel oil 172 with a sulphur content of less than 0.1% by mass (European Parliament, 2005). However, passenger ships in the EU's non-ECA waters can use marine fuels with S content of 1.5% from January 1st 2015 to January 1st 173 1742020 (Cullinane and Bergqvist, 2014). The use of cleaner marine distillate fuels in ocean-going ships 175 operating in the Californian coastal areas was mandated by the State of California. In particular, "Fuel 176 sulphur and other operational requirements for ocean-going vessels within California waters and 24 nautical 177 miles of the California baseline" has been adopted on 24 July 2008, and enforced since July 2009 (ARB, 1782008). In addition, some types of vessels such as container vessels, passenger ships and refrigerated-cargo 179 ship fleets that visit Californian ports need to comply with Californian at-berth regulations with the target of 180 80% emission reduction by 2020.

181

182 US Environmental Protection Agency (EPA) regulations for marine applications are available at Title 40 of 183 the Code of Federal Regulations (CFR), Parts 94 and 1042. Three categories of engines are identified based 184 on per-cylinder displacement (US-EPA, 2012). Marine diesel engines which belong to categories 1 and 2 185 normally have engine output power in the range of 500 to 8000 kW. These engines can be either used as 186 marine main engines on many kinds of small vessels such as tugboats, supply and fishing vessels, or used as 187 marine auxiliary diesel engines on larger vessels. Category 3 marine diesel engines, which range in output 188 power size from 2500 to 70000 kW, are generally used to provide propulsion power for ocean-going ships 189 consisting of container ships, oil tankers, bulk carriers and cruise ships. Application of emission control systems on category 3 marine diesel engines is limited (US-EPA, 2012). The EPA Tier 2 limits are similar to 190 191 Tier II limits of Annex VI for NO_x, but also consist of limits on hydrocarbons (HC), particulate matter (PM) 192 and carbon monoxide (CO).

193

194 **1.3. Negative impact of ship emissions**

195 The primary drivers for regulating ship exhaust emissions are their harmful effects on both public health and 196 environment (Anderson et al., 2015b; Blasco et al., 2014; Chu-Van et al., 2018a; Corbett et al., 2007; Mueller 197 et al., 2015; Reda et al., 2015; Ristovski et al., 2012). Ship emissions consist of both gaseous and particulate 198 emissions (Mueller et al., 2011). Viana et al. (2014) noted that emissions from ships are one of the most 199 significant contributors to poor air quality, especially in coastal environment around the globe. A study in 200 2010 reported that over 70% of shipping emissions have been detected up to 400 km inshore, significantly 201 contributing to air quality degradation in areas closer to harbours (Eyring et al., 2010). Ship emissions caused 202 an increase in concentrations of particulate matter as well as of gaseous pollutants, which consequently form 203 new secondary particles in more densely-populated areas (González et al., 2011; Viana et al., 2014). 204 Particulate matter generated by engine combustion is a complicated mix of particularly tiny liquid and solid 205 particles consisting of numerous ingredients, including acids (e.g. nitric and sulphuric acids), metals and 206 organic compounds. A previous study indicated that BC emitted from international ships accounted for 1 to 207 2% of total BC around the world (Lack et al., 2008). Gaseous emissions such as SO_x form sulphate aerosols 208 - small particles that reflect sunlight and contribute to radiative cooling of the planet (Sofiev et al., 2018). In 209 contrast, CO₂ emissions increased warming effect (Lack et al., 2008; Lack et al., 2011). Additionally, SO_x 210 and NO_x emissions from global ships are also significant contributors to the acidification of the oceans 211 (Hassellöv et al., 2013).

212

213 In 2012, diesel engine exhaust emissions were classified as carcinogenic substance by the World Health

Organization (WHO) International Agency for Research on Cancer (IARC) (<u>IARC, 2012</u>) in the same group

as asbestos. The links between fine particle $(PM_{2,5})$ emissions and increased mortality due to cardiovascular

- and lung problems has been noted in several studies (Abramesko and Tartakovsky, 2017; Corbett et al., 2007;
- 217 Pope Iii and Dockery, 2006; Ristovski et al., 2012). In details, almost 64,000 cardiovascular and lung cancer

- 218 deaths annually can be attributed to $PM_{2.5}$ ship emissions (<u>Corbett et al., 2007</u>). In Europe, 5–10% of total 219 PM emissions are derived from shipping activities, which are responsible for approximately 400,000 220 premature deaths annually (Andersson et al., 2009). Through the use of low-sulphur fuel, the reduction in 221 PM_{2.5} emissions (by mass) by the year 2020, cleaner marine fuels will significantly contribute to the 222 reduction of global ship-related morbidity and premature mortality by 54 and 34%, respectively (Sofiev et 223 al., 2018). However, marine fuels with lower sulphur content will still significantly contribute to the number 224 of related fatalities (~250,000 deaths) as well as ~6.4 million cases of childhood asthma (Sofiev et al., 2018). 225 Therefore, a need for new regulations will be necessary, because reducing in-fuel sulphur will be insufficient 226 to diminish emissions of ultrafine particle emissions (Winnes and Fridell, 2012). As noted in a further study 227 by Winnes et al. (2016), the sulphur content of the fuel influenced the observed particle mass, but not the 228 particle number emissions. Authors suggested that fuel chemical- and physical-related studies (e.g. metal and 229 ash content, viscosity) are needed so as to understand this phenomenon.
- 230

231 **2. Impact on marine fuel oil refining processes**

232 Following the more stringent regulatory requirements, refineries bear the brunt of the increasing demand for 233 low sulphur marine fuels. Around 2 million barrels of oil per day (bbl/d) of fuel oil will need to be switched 234 to distillate fuels to meet the global sulphur cap in 2020, with an annual growth trend predicted at 500,000 235 bbl/d of distillate and a decline of 150,000 bbl/d in residual demand (CONCAWE, 2016). This requires 236 significant changes to the refinery to increase the capacity of residue conversion processes to produce 237 cracked gasoil, the development of larger sulphur-removal processes for residues (IEA, 2016), or the use of 238 sweeter crude feedstock. An analysis of potential refinery modifications to meet the targets requires capital 239 investments of \$2-5 billion for 350,000 bbl/d refinery, with payout periods of 4.2-5 years (Abdel-Halim and 240 Yu, 2018).

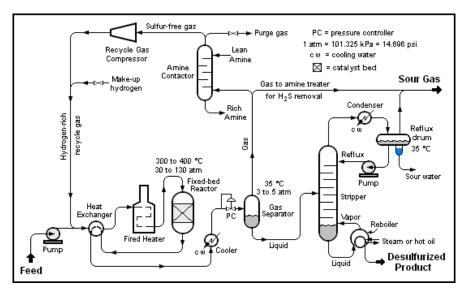
241 Around 50% of crude oil usually proceeds to the heavy fuel fractions; however, with increasing light fuel 242 (e.g. gasoline) demand and declining heavy fuel demands, complex refinery processes are often attuned to 243 produce lighter components. Marine fuels are composed from diesel, kerosene, light and heavy gasoil, and 244 atmospheric residue fractions. Kerosene can be used as feed for producing straight run marine gasoil and 245 distillate marine diesel, and these products are used to decrease the viscosity of residues through blending. 246 From the bottom of the atmospheric crude distillation part, the atmospheric residue proceeds to vacuum 247 distillation, where it is further separated to a light fraction fed to a fluid catalytic cracker (FCC) and a heavy 248 fraction that proceeds to a visbreaking process. The FCC process involves high temperatures and an 249 aluminium silicate catalyst to produce more gasoline, light cycle oil (LCO), which is a distillate product, and 250 heavy cycle oil (HCO), which is added to the visbreaker residue. The visbreaker uses a mild thermal 251 cracking process to yield blendstock gasoil. Blending of these different product streams with streams of less 252 desirable properties (i.e. high viscosity and density) to achieve marine fuel standards maximises total 253 refinery yield. The blends consider the maximum density limit, which affects ignition quality; maximum 254 aluminium and silicon limit, to avoid abrasive damage in the fuel system; and maximum total sediment aged limit, to minimise impurities stemming from residual and unstable components generated in visbreaking
 (Vermeire, 2012).

Sulphur is present in crude oil from 0.03 to 7.89% weight in various forms (Agarwal and Sharma, 2010). Smaller sulphurous molecules such as thiols, sulphides and thiophenes have lower boiling points and are distilled with naphtha, kerosene and diesel, while the heavier sulphur-containing molecules such as benzothiophenes, dibenzothiophenes and heavy sulphides are usually found in vacuum gas oil and vacuum residue (Leprince, 2001). The chemical structures of sulphur-containing compounds dictate the ease or difficulty of their removal from crude oil. Aliphatic sulphurs are easier to remove, while aromatic sulphurs are more challenging (Javadli and de Klerk, 2012).

Within the refinery, the cracking, coking and hydrotreating processes remove sulphur from product streams as hydrogen sulphide (Elvers, 2008). These processes result in low sulphur content products to meet statutory

requirements. Hydrodesulphurisation (HDS), a specific hydrotreatment process, uses hydrogen to remove

- sulphur in the middle distillates of kerosene and gasoil. A typical HDS process is shown in Fig. 3.
- Hydrodesulphurisation of residues is also feasible, although conventionally not considered, due to cost. This
- 269 is due to frequent catalyst replacements borne from a higher level of vanadium in residues that irrevocably
- deposit on the hydrotreating catalyst (<u>Speight, 2011</u>).



271



Fig. 3. Typical hydrodesulphurisation process flow diagram (Beychok, 2012).

273

Hydrodesulphurisation uses catalysts with molybdenum, cobalt and nickel supported on alumina (i.e. CoMo-Al₂O₃ and NiMo-Al₂O₃) to assist in the elimination of the heteroatom and hydrogenation of the hydrocarbons (Topsøe et al., 1996). Unlike catalysts used for the lighter fractions, heavier fractions require catalysts that are meso- and macroporous to allow adequate contact of larger molecules with the active catalyst sites. Heavy oil hydroprocessing also requires catalysts that remove metals in the feed to protect the HDS catalysts (Furimsky, 1998). The reactors are designed to contain beds of different catalysts to minimise catalyst deactivation and maintain the reaction rates (<u>Takahashi et al., 2005</u>). The process also requires high temperatures and high hydrogen pressure, which contributes to high production costs (<u>Kressmann et al.,</u> 1998).

283 Various technologies are proposed for reducing sulphur to the required lower levels. Ebullated bed reactors 284 with catalysts smaller than 1 mm can accommodate heavy feeds with high asphaltene, sulphur and metal 285 content, and achieve up to 90% HDS conversion (Rana et al., 2007). Moving bed reactors, which can 286 accommodate online catalyst replacement, have also been designed for partial removal of contaminants prior 287 to processing in conventional fixed bed reactors (Yuandong et al., 2009). Another technique suggested is to 288 ensure a low H₂S partial pressure towards the HDS reactor outlet to prevent inhibition of the desulphurisation 289 reaction. Hydrogen with some amount of H₂S is recycled from the reactor outlet back to the inlet to 290 maximise the hydrogen supply. This presents issues in the reaction at low sulphur levels, since the 291 equilibrium can easily be tipped towards formation of sulphurous compounds if H₂S partial pressure is 292 sufficient. Reactor configurations, such as interstage removal of H₂S, countercurrent feed-hydrogen flows, 293 and a separate catalyst bed fed with fresh hydrogen are suggested to minimise the effect of H₂S (Ancheyta 294 and Speight, 2007). Aside from HDS reactors, liquid-liquid extraction of the sulphur containing compounds 295 has also been explored using organic solvents (Kumar et al., 2014), ionic liquids (Chen et al., 2014), and 296 deep eutectic solvents (Warrag et al., 2018). The latter was developed as a less toxic, greener alternative to 297 conventional solvents and ionic liquids, and can remove up to 99.5% sulphur in multistage reactors to 10 298 ppm. However, this has been demonstrated only for gasoline and diesel fuels (Chandran et al., 2019). A 299 combination of conventional refinery processes of distillation, extraction and hydrotreatment, has also been 300 proposed to facilitate deep desulphurization down to 73 ppm (Kumar et al., 2018).

301 A new technique proposed for removal of refractory sulphur compounds such as thiophenes, is oxidative 302 desulphurisation (ODS). This process involves addition of an oxidising agent such as hydrogen peroxide, and 303 a catalyst to convert aromatic sulphur to sulphones or sulfoxides, which can then be easily extracted by a 304 polar solvent (Ismagilov et al., 2011). The use of oxygen in air as an oxidant with the use of cobalt and 305 manganese oxide catalysts has also been explored to lower the cost of the process (Sain et al., 2003). The key 306 advantage of ODS against HDS is the high reactivity of dibenzothiophenes in ODS, whereas in HDS they are 307 the least reactive (Ismagilov et al., 2011). This process, combined with HDS has the potential to produce 308 low-sulphur products because they target different sulphur species in heavy oil.

Another method that has been recently explored is biodesulphurisation, using bacteria that consumes dibenzothiophenes as substrates in mild conditions with low hydrogen requirements (<u>Chen et al., 2018</u>). High sulphur removal was reported up to 76% from crude oil and 98% from hydrodesulphurised diesel with a residence time of four days (<u>Adlakha et al., 2016</u>). There are challenges associated with the complexity and cost of cultivating bacteria in specialized culture media and isolating specific bacteria for the process, however, a more commercially-feasible bacterial consortium can be implemented (Porto et al., 2018). 315 The new technology introduced in refineries can suffer from unsustained reliability and lack of experience, 316 which are both critical in production of large volumes of marine fuel. In order to avoid the complexities and 317 costs of installing additional specialised refinery sections for the HDS of heavy fractions, residues are also 318 blended with distillates. This is an established practice to extend the value of crude oil, however in this case, 319 blending can also be done to meet new fuel standards. This affects refinery operations because there is added 320 pressure to produce more distillates that can serve as blendstock. The processes to remove sulphur described 321 above all require additional energy and hydrogen, and intensification of those processes to produce lighter, 322 lower-sulphur marine fuels also increases energy and hydrogen consumption. This translates to potentially 323 higher CO_2 emissions (Silva, 2017). However, new installations to cope with the higher demand for distillate 324 fuels will have higher refinery efficiencies, which dampen the increase in energy and CO₂ emissions. A 325 model by the Oil Companies International Study Group for Conservation of Clean Air and Water in Europe 326 (CONCAWE) for the European scenario illustrates the effect of the new sulphur standard by showing the 327 resultant marginal increase or decrease in the demands of marine fuels. Marginal increases (10-20%) in 328 demand of low sulphur residual marine fuels cause small increases in energy requirements and CO₂ 329 emissions. In contrast, a proportionate decrease in high sulphur marine fuel demand, also increases CO_2 330 emissions, due to the liberation of CO_2 in refining heavy fractions, which used to be a carbon sink. 331 Furthermore, scenarios of switching to a 50% or 100% distillate marine fuel scenario show CO₂ emissions 332 7.5-15 times higher than the low sulphur residual marine fuel case. The model considers the balance of the 333 entire refinery and economic optimisation, so the higher CO₂ emissions also reflect the difficulty of 334 producing distillate marine fuels, which is already a high-demand product (Dastillung et al., 2009). On the 335 global scale, a study estimates that a total switch from residual fuels to distillate fuels will increase total 336 global anthropogenic CO_2 emissions by 0.01%, while decreasing total global sulphur emissions by 6% 337 (Corbett and Winebrake, 2008), presenting a minor trade-off to mitigate the effect of sulphur in the 338 atmosphere.

The increased sulphur removal is expected to increase nondiscretionary sulphur production only incrementally, similar to changes that occurred when road and jet fuel standards required lower sulphur content (Ober, 2000). Other factors such as the increased use of sour crude and gas, and improvement of environmental standards in developing countries can also contribute to an increase in sulphur production. This is expected to meet increasing demand in fertilizer processing and other industrial areas (Apodaca, 2017).

345 **3. Impact on the future marine fuel mix for the shipping sector**

346 To meet growing future demand and the regulated emissions reduction, it can be expected that the mix of 347 marine fuels used will change. A forecast for shipping industry fuels has been recently reported by DNV-GL 348 (2017). Several major types of cargoes, such as crude oil, oil products, natural gas, bulk, container and other 349 cargoes have been included in the study as can be seen in Fig. 4. It has been predicted that demand for 350 seaborne transport will increase 60% by 2050. The increase in demand of seaborne has also been reported in 351 a two years study (GMT, 2013), which predicted that international seaborne trade will be nearly doubled by 352 2030 compared to 2010. It is hard to predict exactly the number, but overall an increase in the future 353 seaborne demand has been predicted. It shows the important role of cleaner fuels for the marine sector in 354 order to satisfy future seaborne demand and to mitigate emissions. The shipping fuel mix has been predicted 355 up to 2050 as shown in Fig. 5. As can be seen in Fig. 5, alternative marine fuels, namely liquefied natural gas 356 (LNG) and biofuel, may account for 50% of shipping energy demand, while the remainder will still be 357 supplied by conventional HFO/MGO (DNV-GL, 2017). Similarly, LNG is predicted to reach the maximum 358 of 11% in 2030, while 47-66% is of HFO (GMFT, 2014). It shows the vital role of emission regulations on

359 shipping with the increasing utilization of alternative fuels.

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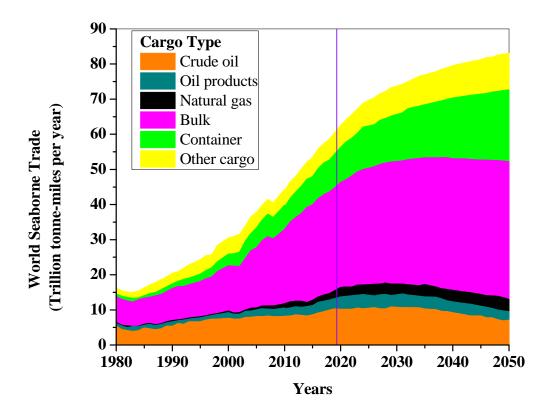
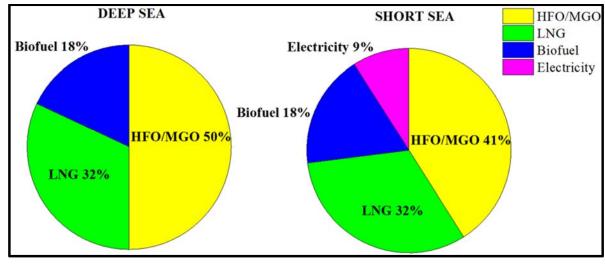




Fig. 4. Demand forecast for seaborne transport (adopted from DNV-GL (2017))

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Fig. 5. Shipping fuel mix 2050 (DNV-GL, 2017)

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367 Alternative fuel potential for use in the marine sector will need to satisfy the required reduction in coastal 368 area air pollutants and greenhouse gas emissions, and to comply with IMO regulations (Gilbert et al., 2018). 369 A recent study assessed the full life-cycle air emissions of a set of alternative shipping fuels, including LNG, 370 methanol, liquid hydrogen, biodiesel, straight vegetable oil and bio-LNG. Results showed that no available 371 fuel exists to meet both short-term and long-term ship emission reductions (Gilbert et al., 2018). Residual 372 fish oil and MGO blends have been used in a recent study to investigate combustion and emission profiles 373 (Ushakov et al., 2013). Burning fish oil showed a reduction up to 56% of CO, 70% of HC, and 70-90% of 374 total number and PM mass compared to MGO, which may be strongly associated with high oxygen content 375 and lack of aromatic compounds in fish oil. However, the lower heating value of fish oil resulted in an 376 increase in specific fuel consumption observed at all load conditions (Ushakov et al., 2013). Recently, a 377 study suggested that waste cooking oil biodiesel (fatty acid methyl esters-FAME) blended with MGO can be 378 considered as an alternative fuel for auxiliary marine engines because it helps to reduce NO_x emissions (Wei 379 et al., 2018).

380

381 Methanol has recently been given attention by researchers as a low carbon alternative fuel to conventional 382 fuels. The total life-cycle emissions of methanol use were compared to MGO and HFO in order to identify 383 the environmental benefits of using methanol (IMO, 2016b). Generally, emissions of SO_x, NO_x, PM and 384 GHGs (CO₂, CH₄ and N₂O) were used in the total life-cycle model. It is reported that although methanol's 385 cost is slightly lower than that of MGO, it is not a preferable fuel for shipping in the long-term. This is 386 because its use neither gives a favorable payback period for the capital investment of its propulsion, nor 387 meets the price of its production. The Stena Germanica ferry is reported as the first large marine vessel 388 running methanol after a conversion in 2015 in Poland. Methanol use is expected to reduce emissions for 389 SO_x, NO_x, CO₂ and PM by up to 99%, 60%, 25% and 95% respectively (MKC, 2018). It is obvious that 390 methanol has a lower investment and space requirement, making it attractive compared to LNG, however a 391 major barrier to its use is its high fuel cost (Motorship, 2018).

392	Natural gas in the form of LNG is an alternative fuel for shipping is considered the most likely option in
393	order to comply with IMO regulations and to reduce air pollution and climate change (IMO, 2016c). Table 1
394	reveals that the use of LNG as a marine fuel for shipping would be preferable from an environmental
395	perspective; however other factors such as finance need to be considered to choose the compliance strategy
396	for a vessel. A recent study investigated both particulate and gaseous emissions from an LNG-powered
397	vessel (Anderson et al., 2015a). Results showed that a significant reduction of particles, NO _x , SO ₂ , and CO ₂
398	was observed for LNG use compared to MGO, while emissions of HC and CO showed the opposite trend.
399	HC analysis also revealed 90% of HC was unburnt methane, which shows the need to control and limit
400	methane slip when using LNG (Anderson et al., 2015a), since the global warming potential of methane is 25
401	times higher than that of CO ₂ over a 100-year scale. The energy policy requirements are necessary in order to
402	reduce methane leakage through better processing infrastructure designs and during engine combustion
403	(Thomson et al., 2015). A total life-cycle emission inventory for air emissions from two vessels operating
404	between China and Taiwan using both HFO and LNG for each ship was conducted (Hua et al., 2017). Both
405	ships present a considerable decrease in the total fuel life-cycle emissions of NO _x (38-39%), CO (42-43%),
406	SO_2 (99.8%) and PM_{10} (97.5%), however methane emissions were significant in the case when LNG was
407	used compared to HFO. This study also indicated that LNG as a marine fuel enables improved life-cycle
408	GHG emissions and reduces SO_x and PM across the Taiwan Strait (<u>Hua et al., 2017</u>). Recently, the
409	assessment on using LNG as an alternative marine fuel has suggested there is a need for regulating methane
410	slip from LNG engines (Brynolf et al., 2014). In addition, uncertainties related to future price and global
411	availability of LNG, methanol and MGO are actually questionable, while issues with HFO/Scrubber are
412	located at the reliability/corrosion during operation. Infrastructure development for bunkering and safety in
413	use would also be considered as uncertainties since LNG and methanol are used.

414

Table 1. Comparison between the alternatives: LNG, Methanol, MGO and HFO/Scrubber with HFO
 (IMO, 2016b, c)

Fuel types	Environmental factors			ctors	Other factors			
	SO _x	NO _x	PM	CO ₂	Cargo capacity	Capital investments	Operating costs	
LNG	++	++	++	+	Restricted	Very high	Very low	
Methanol	++	+	++	++	Restricted	Very high	High	
MGO	+	-	-	-	Unrestricted	Low	Very high	
HFO/Scrubber	+		+	-	Slightly restricted	High	Medium	
HFO					Unrestricted	Low	Low	

417 ++ very positively, + positive, - negative, -- very negative.

418 Biofuels are of increasing interest and a promising alternative for both road and seawater transport sector.

419 Potential biofuels should be required to provide no competition with food crops or land use and to offer a

420 reduction in greenhouse gas emissions (Obeid et al., 2019). Microalgae-based biofuels have recently

421 received great attention from researchers because of their potential for large-scale fuel production on non-

422 arable land (Islam et al., 2017). Oil extraction from microalgae can be obtained through a variety of methods

423 such as mechanical disruption, ultrasonic-assisted extraction, solvent extraction, hydrothermal liquefaction 424 (HTL) and supercritical fluid extraction. Among these, HTL has been identified as a very promising method 425 to convert biomass into biofuels owing to its high energy efficiency (Islam et al., 2017) and ability to process 426 a wide variety of locally available waste biomass (Kosinkova et al., 2017). Physicochemical properties of 427 microalgae biofuels resulted in engine combustion reaction changes, which consequently will have a large 428 effect on engine performance and emission profile (Hossain et al., 2018b). These properties are dependent on 429 microalgae species (Islam et al., 2013), process conditions (e.g. temperature, pressure, retention time and the 430 choice of catalyst) (Ross et al., 2010; Xu et al., 2015). The variation of microalgae biofuel properties also 431 affects their potential application. In particular, microalgae HTL bio-crude may be more suitable for low-432 speed marine diesel engines because its properties are similar to HFO (Hossain et al., 2018b). In contrast, 433 microalgae biodiesel consisting of FAME mostly can be used as an alternative fuel for high speed diesel 434 engines (cars, buses and vehicles) owing to its properties being similar to that of conventional diesel (Islam 435 et al., 2015). HTL bio-crude is a dark and energy-dense liquid with high viscosity (Hossain et al., 2018b; 436 Kosinkova et al., 2015), and is comparable with HFO as regards fuel properties. The fuel property 437 comparison between microalgae FAME, HTL bio-crude, tyre oil, MGO and HFO is presented in Table 2. It 438 can be seen that HTL bio-crude properties are closer to that of HFO, which reveals the promising low-439 sulphur alternative fuel for shipping in the future. This fuel can also be upgraded to reduce oxygen content 440 and improve calorific value (Ramirez et al., 2015) and can be used as alternative fuel for auxiliary marine 441 diesel engines. The storage stability of HTL bio-crudes must also be considered to anticipate the effects of 442 long-term storage of fuels (Kosinkova et al., 2016).

443

444Table 2. Microalgae FAME, HTL bio-crude, Tyre oil and Licella properties in comparison with the445conventional marine fuels: MGO and HFO.

Fuel	Unit	Microalgae		Tyre Oil	Licella	MGO	HFO
Parameter		Biodiesel	HTL bio-crude	(<u>Hossain et</u>	(<u>Nabi et</u>	(<u>Nabi et</u>	(<u>Chu-Van et</u>
		(FAME)	(<u>Hossain et al.,</u>	<u>al., 2018a</u>)	<u>al., 2015</u>)	<u>al., 2012</u>)	<u>al., 2018a</u>)
		(Islam et al.,	<u>2017a</u>)				
		<u>2015</u>)					
Density at 15 °C	kg/L	0.912	0.97-0.104	0.847	0.89	0.855	0.986
Viscosity at 40 °C	mm ² /s	5.06	73.8	3.43	5.11	2.60	377*
Calorific value	MJ/kg	39.86	30-35	42.28	42.63	42.8	42.74
Cetane number		53	N/A	51	N/A	47	N/A
Sulphur (S)	% mass	7.5x10 ⁻⁴	0	0.35	0.010	0.005	3.13
Carbon (C)	% mass	78.41	75.56	84.1	84.46	86.2	88.14
Hydrogen (H)	% mass	11.12	10.14	15.9	12.66	13.6	9.36
Oxygen (O)	% mass	10.47	10.33	N/A	2.88	N/A	N/A
Nitrogen (N)	% mass	0	3.97	N/A	0.05	N/A	0.68

446 *measured at 50 °C; N/A: not available.

447 Tyre oil may also be a promising fuel for marine diesel engines. The physicochemical properties of the tyre 448 oil are similar to MGO and this fuel can be miscible with conventional diesel in any blended ratio. A very 449 recent study investigated engine performance and emissions from a non-road diesel engine using tyre oil 450 (Hossain et al., 2018a). The results showed a significant reduction in NO_x, PM mass and number, while CO 451 emissions slightly increased. Tyre oil is produced from end-of-life tyres (ELTs). It has been reported that 1-452 1.5 billion tons of ELTs are generated annually around the world (Hossain et al., 2018a). The ELTs to fuel 453 technology not only can help to deal with problem-related waste disposal from ELTs in the world, but also 454 offers a potential fuel solution for the marine sector.

455

456 Companies such as Licella, which convert numerous low-cost products, waste and non-edible biomass 457 residues through thermochemical conversion processes, are helping to produce renewable fuels that can be 458 used for the transportation sector. A previous study investigated fuel characterisation, engine performance 459 and exhaust emissions with a new renewable Licella biofuel (Nabi et al., 2015). The renewable component of 460 the Licella biofuel in this study was made from Australian pinus radiata wood flour by using the HTL 461 conversion method. Results showed a reduction in both PM mass and PN emissions for all Licella blends, 462 with no significant changes in engine performance compared to conventional diesel. However, HC and NO_x 463 emissions were observed to be higher for the Licella biofuel. This study also highlights a need for further 464 studies on the future use of Licella biofuel in marine and stationary generation applications (Nabi et al., 465 2015).

466

467 **4. Potential impact to ship emissions**

468 To mitigate emissions from ships, the IMO has issued and implemented more stringent regulations related to 469 marine sulphur content and NO_x emissions. IMO compliant fuels or marine fuels with low sulphur content 470 normally refer to marine gas oil (MGO), marine diesel oil (MDO), and low-sulphur residual fuel (LSHFO), 471 which have been used to meet fuel sulphur content IMO regulations. For the reduction in NO_x , the use of fuel 472 additives can be considered as a feasible measure though it did not be officially recognised by IMO. In this 473 section, the effect by using these fuels compared to heavy fuel oil (HFO) on engine emissions will be 474 thoroughly investigated to assess the efficiency of IMO regulations. A number of recent on-board and test-475 bench measurements will be used in the present study.

476

Switching to low-sulphur fuels such as MGO, MDO or LSHFO from HFO results in a variety of fuel properties changing, and consequently may affect ship emissions because these are significantly affected by fuel type, fuel quality and fuel properties (Winnes et al., 2016). It is evident that fuel sulphur content has a strong impact on PM mass, particle number (PN), and particulate matter chemical composition. On-board measurements were carried out on a ship operating in the Baltic Sea (an SECA) (Zetterdahl et al., 2016). This study investigated the impact of 0.1% sulphur content fuels (LSHFO or hybrid fuel called RMB30) on emissions compared to HFO. This fuel change from 0.48% to 0.1% sulphur content resulted in a reduction of

nearly 67% PM mass, and 80% SO₂ as well as a reduction in total volatile organic compounds (VOCs)

485 (Zetterdahl et al., 2016). This is in agreement with the results of other on-board measurements (Moldanová et 486 al., 2013), which showed that HFO emitted the higher PM mass compared to MGO. Particulate organic 487 carbon (OC) emissions for HFO combustion were three times higher than that of MGO, while elemental 488 carbon (EC) revealed no significant differences between the two fuels. A recent test-bed study investigated 489 the effect of sulphur- and vanadium-spiked fuels on particle characteristics and engine performance (Chu-490 Van et al., 2018a). This study found that higher sulphur content leads to significant increase in PM mass, PN 491 and sulphate fraction. However, PN results in this study are in contrast with previous on-board measurement 492 (Zetterdahl et al., 2016), which showed unchanged values of PN for both LSHFO and HFO. It indicates the 493 uncertainty in PN emission measurement results which highly affected by other factors such as dilution 494 systems used, engine types and engine load conditions. In the case when HFO was used, the major 495 components of PM_{2.5} are hydrated sulfates, OC and trace metals, which accounted for 80-97% of total PM_{2.5}. 496 However, changing fuel from HFO to MDO revealed that OC is the main component of PM_{2.5}, accounting 497 for 65% of PM_{2.5} (Celo et al., 2015), and over 90% of PM found was OC for both MGO and LSHFO (Gysel 498 et al., 2017). It indicated that using IMO compliant fuels will result in a significant reduction of sulphate and 499 trace metal proportions of $PM_{2.5}$.

500

501 In addition to the impact on PM mass and PN, switching to IMO compliant fuels strongly affects particulate 502 matter chemical composition. A previous on-board measurement found that metals detected in exhaust when 503 using HFO were dominated by compositions from both fuel used (V, Ni, Fe) and lubricating oil (Ca, Zn), 504 while for MGO use, most metals from exhaust emissions were associated with lubricant compositions 505 (Moldanová et al., 2013). These findings are in good agreement with a very recent test-bed study (Chu-Van 506 et al., 2018b). In this study, a detailed investigation into the metallic elemental composition at different 507 particle diameter sizes was undertaken. A multivariate statistical analysis was performed to elucidate 508 relationships between the measured engine performance (engine power, thermal efficiency, specific fuel 509 consumption,...), engine emission variables (gaseous and particle emissions) and particular test conditions 510 such as different engine load conditions, the fuel sulphur and vanadium contents, and experimental 511 replication. The major sources of metallic elements in the emitted particles are considered in this study, 512 including the fuel and lubricating oil compositions, engine wear emissions and metal-containing dust in the 513 ambient air. Solid ultrafine-particles (Dp < 100 nm) are strongly associated with metallic compounds derived 514 from lubricating oil (Ca, Zn, Mg and K), while the fuel-related metallic compounds and engine wear 515 emissions are represented in the coarse particle fraction ($Dp > 1 \mu m$) (Chu-Van et al., 2018b).

516

In the case both ECA-compliant fuels used, it showed the difference in particle emissions. Particularly, MGO and LSHFO have been used in a recent on-board measurement to provide insight into particle emissions from a very large crude vessel (Gysel et al., 2017). These two fuels have small sulphur and no ash or vanadium content. The significant difference between the two fuels is in their viscosity, with a 4-times higher viscosity for LSHFO compared to MGO. Results showed that MGO emitted 5% higher NO_x but three times lower for PM_{2.5} compared to LSHFO combustion. It can reveal the important role of fuel viscosity on engine

- 523 emissions, however further study is needed to make this clear.
- 524

525 BC, a component of elemental carbon (EC), is an important climate forcing agent (Lack et al., 2008). BC 526 emissions from marine traffic in the Arctic also greatly influences climate warming through direct light 527 absorption and increase of surface albedo through darkening of snow (Bond et al., 2013). A reduction in 528 aromatic and long chain hydrocarbon constituents of fuels owing to switching to low sulphur marine fuels 529 (e.g. RMB30) may be associated with a decrease in BC (Lack et al., 2011). However, a previous study found 530 that using low sulphur distillate fuels emitted higher EC than HFO, although a reduction of hazardous 531 species such as PAHs, Oxy-PAHs, N-PAHs and transition metals was observed (Sippula et al., 2014). To 532 make the BC emission mechanism clear, Jiang et al. (2018) investigated the effects of BC measurement 533 method, fuel type and engine loads on BC emissions from a 187 kW, 2-stroke high-speed marine engine. The 534 variation in BC concentration due to different measurement methods ranged by a factor of 2, and did not be 535 responsible for a ten-time range of BC suggested in the literature review. Jiang and co-workers' view is that 536 fuel properties, engine load and engine characteristics are most likely to be the driving factors towards a 537 large variation in BC emissions (Jiang et al., 2018). Surprisingly, RMB30 or LSHFO, a new low-sulphur 538 residual fuel widely used in SECAs and NECAs, presents the highest level of BC emissions among three 539 tested fuels. There is not a clear link between BC and fuel properties like fuel sulphur content, viscosity and 540 carbon residue. Consequently, this calls for a need of further study on fuel parameters. In addition, this study 541 shows a positive trend, that using distillate fuels may help to reduce BC emissions from ships (Jiang et al., 542 2018).

543

544 Using fuel additives in order to obtain both fuel savings and environmental benefits, so adding additives to 545 HFO, has been attractive to both shipping companies and researchers recently. One study used oil-soluble 546 Ca- and Fe-based organometallic compounds as fuel additives in HFO (Ryu et al., 2016). Both reductions in 547 fuel consumption and exhaust emissions were observed. In particular, 23-32.6% and 16.5-19.7% reductions 548 in NO_x emissions obtained for Ca- and Fe-related additives, respectively. Similarly, for PM, added Ca-based 549 compound resulted in a 57.4-59.7% reduction, with 38.9-40.9% reduction for Fe-related additive (Ryu et al., 550 2016). Similarly, dispersant cetane and combustion improvers showed an improvement in fuel stability 551 during storage and a significant reduction of PM emissions (Jang and Choi, 2016). However, this method is 552 not considered as an alternative method to satisfy IMO regulations. Official alternatives to meet the SO_x and 553 NO_x limits set by the IMO are to use the approved exhaust cleaning systems.

554

In summary, the potential impact of IMO regulations on ship emissions can be seen through switching the fuel from HFO to low sulphur fuels. This changing in the fuel resulted in a significant reduction of overall engine emissions including PM mass and number emissions, sulphate fraction and BC. In addition, the fuel change has strongly influenced to engine emission characteristics such as particulate matter chemical composition. In the future, switching the fuel combined with the modification of marine engines such as using electronic fuel injection system (common rail system) can help to save fuel consumption and further 561 reduce engine emissions.

562

563 5. Conclusion

564 In this study, an overview of the context and global impacts of recent IMO regulations implemented on the 565 marine oil refining industry, future marine fuel mix and ship emissions has been investigated. The marine 566 fuel limit of 0.1% (wt. %) in both Sulphur Emission Control Areas (SECAs) and Nitrogen Oxide Emission 567 Control Areas (NECAs), and of 0.5% globally are in force by 2015 and 2020, respectively. Moreover, new 568 vessels which built on or after 2016 and operate in SECAs must also comply with the NO_x emission limit. It 569 has been reported that the new implemented IMO regulations will be beneficial for both public health and 570 environment. Owing to IMO impact, the demand for low sulphur marine fuel is increasing, which requires 571 significant changes to the refinery to increase the capacity of residue conversion processes to produce 572 cracked gasoil, the development of larger sulphur-removal processes for residues, or the use of sweeter crude 573 feedstock. These changes to the refinery processes can lead to an increase in greenhouse gas emissions and 574 high capital investments. Alternative marine fuels consisting of LNG and biofuel may be suitable fuels to 575 meet the required reduction in coastal area air pollutants and greenhouse gas emissions. It has been predicted 576 that these two fuels can account for 50% of shipping energy demand by 2050 while the rest will still be 577 fulfilled by conventional HFO/MGO. Switching from HFO to low sulphur fuels under IMO impact resulted 578 in a significant reduction of engine emissions and emission chemical characteristics.

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