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Comprehensive analysis of the air quality impacts of switching a marine vessel from diesel fuel to natural gas☆

Weihan Peng [a, b], Jiacheng Yang [a, b], Joel Corbin [c], Una Trivanovic [d], Prem Lobo [c], Patrick Kirchen [d], Steven Rogak [d], Stéphanie Gagné [c], J. Wayne Miller [a, b], David Cocker [a, b, *]

[a] Department of Chemical and Environmental Engineering, Bourns College of Engineering, University of California, Riverside, CA, 92507, United States
[b] University of California, Bourns College of Engineering, Center for Environmental Research and Technology (CE-CERT), 1084 Columbia Avenue, Riverside, CA, 92507, United States
[c] Metrology Research Centre, National Research Council Canada, 1200, Montreal Road, Ottawa, ON, K1A 0R6, Canada
[d] Department of Mechanical Engineering, University of British Columbia, 2054-6250, Applied Science Lane, Vancouver, BC, V6T 1Z4, Canada

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ABSTRACT

New environmental regulations are mandating cleaner fuels and lower emissions from all maritime operations. Natural gas (NG) is a fuel that enables mariners to meet regulations; however, emissions data from maritime operations with natural gas is limited. We measured emissions of criteria, toxic and greenhouse pollutants from a dual-fuel marine engine running either on diesel fuel or NG as well as engine activity and analyzed the impacts on pollutants, health, and climate change. Results showed that particulate matter (PM), black carbon (BC), nitric oxides (NO_x), and carbon dioxide (CO_2) were reduced by about 93%, 97%, 92%, and 18%, respectively when switching from diesel to NG. Reductions of this magnitude provide a valuable tool for the many port communities struggling with meeting air quality standards. While these pollutants were reduced, formaldehyde (HCHO), carbon monoxide (CO) and methane (CH_4) increased several-fold. A health risk assessment of exhaust plume focused on when the vessel was stationary, and at-berth showed the diesel plume increased long-term health risk and the NG plume increased short-term health risk. An analysis of greenhouse gases (GHGs) and BC was performed and revealed that, on a hundred year basis, the whole fuel cycle global warming potential (GWP) per kWh including well-to-tank and exhaust was 50% to few times higher than that of diesel at lower engine loads, but that it was similar at 75% load and lower at higher loads. Mitigation strategies for further reducing pollutants from NG exhaust are discussed and showed potential for reducing short-term health risks and climate impacts.

1. Introduction

The global seaborne trade accounts for 80% volume of international trade and is continually growing (UNCTAD, 2019). This activity results in air pollution both at sea and in coastal regions. Emissions from shipping were estimated to be responsible for 14% nitric oxides (NO_x), 16% sulfur oxides (SO_x) and 5% particulate matter ($PM_{2.5}$) in coastal areas (European Environment Agency, 2013) and leading to environmental, health and climate impacts (Corbett et al., 2007; Huang et al., 2018; Liu et al., 2016; Matthias et al., 2010; Monteiro et al., 2018; Sharafian et al., 2019). It was estimated that around 60,000 cardiopulmonary and lung cancer deaths were caused by ship PM emissions every year and predicted that mortalities would increase by 40% from 2007 to 2012 (Corbett et al., 2007). Greenhouse gases (GHGs) such as CO_2 and CH_4, and aerosols from shipping emissions also play an important role in climate change (Lashof and Ahuja, 1990). Without increasingly stringent controls on emissions, marine-transport pollutants will lead to further degradation of air quality and human health, and exacerbation of global warming.

In order to control and limit emissions from marine vessels, the International Convention for the Prevention of Pollution from Ships

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* Corresponding author. Address: 1084 Columbia Avenue, Riverside, CA, 92507, United States.
E-mail address: dcocker@engr.ucr.edu (D. Cocker).

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(MARPOL) was adopted at the International Maritime Organization (IMO) to set emission standards and designate sulfur emission control areas (ECAs) where marine vessels must operate on fuels with sulfur content limited to 0.1%. Various control options are being used by marine vessel operators to meet the increasingly stringent standards, including exhaust aftertreatment technologies, or switching to cleaner fuels such as natural gas (Anderson et al., 2015; Khan et al., 2012).

Natural gas (NG) is widely used in trucks and buses due to its availability and often lower cost and lower emissions of $PM_{2.5}$ and NO_x (Ayala et al., 2002; Yang et al., 2013). However, NG only represented less than 3% of global shipping fuel through 2013 to 2015 (Olmer et al., 2017). The major challenge has been the lack of NG refueling infrastructure and the associated costs. Now with many vessels on order, larger scale facilities are being built (Thomson et al., 2015), thus creating availability of NG refueling at a lower cost. The number of NG-fueled vessels in operation has grown from 34 in 2013 to 121 in 2018, with 135 vessels under construction in addition to more than 400 LNG carriers that are largely fueled by natural gas. This growth has been driven by orders placed for vessels such as cruise ships, containers, and oil tankers, and was estimated to reach to about 600 in 2021. (Burel et al., 2013; Le Fevre, 2018; Pavlenko et al., 2020; Sharafian et al., 2019; Thomson et al., 2015).

There are limited measurement data on emissions from NG fueled marine engines. To the best of our knowledge, only one study previously reported on-board measurements of particle and gaseous emissions from an NG powered ship (Anderson et al., 2015). Anderson et al. shows that NG combustion in a marine engine results in a significant decrease in PM, particle number (PN), NO_x, and CO_2 emissions, and increase in total hydrocarbon, CH_4 and CO emissions. However, the overall environmental, climate, and health impacts requires further investigation. Further, the paper does not report the major incomplete combustion product formaldehyde (HCHO). HCHO is defined as a carcinogenic substance (International Agency for Research and Cancer (IARC)) and contributes to other severe health effects, including asthma and nasopharyngeal cancer (Krzyzanowski et al., 1990; McGwin et al., 2010; Nielsen and Wolkoff, 2010; Zhang et al., 2018). In addition, HCHO contributes to photochemical smog and ground-level ozone in atmosphere (Finlayson-Pitts and Pitts, 2000) which are also related to adverse health effects. Earlier results with on-road vehicles showed an increase in formaldehyde (HCHO) from NG (Ayala et al., 2003, 2002; Hesterberg et al., 2008), however, no carbonyl data from NG marine vessels has been reported thus far.

Given the limited number of reports comparing on-board marine-engine emissions from diesel and NG fuels, this study aims to provide a comprehensive analysis of the comparative emissions of criteria pollutants (NO_x, CO, SO_2, $PM_{2.5}$ mass, elemental carbon (EC) and organic carbon (OC)), greenhouse pollutants (CO_2, CH_4, black carbon (BC)) and toxic air pollutant (HCHO) from a large modern commercial vessel operating at sea during normal revenue service in the Vancouver, BC, Canada area in April 2018. In particular, combining the measured real-world dual-fuel engine activity or E2 cycle from ISO 8178 and modal emissions, this study provides an assessment on the impacts on local air quality, human health risks, and global climate change when switching from diesel to NG. We discuss the trade-offs in health risks for lower PM with increased HCHO, and the effect on the global climate when BC and CO_2 emissions are decreased but CH_4 emissions are increased.

2. Materials and methods

2.1. Test platform: vessel and propulsion system

The 6750-deadweight-tonnage (DWT) test vessel was the first LNG-battery hybrid roll-on/roll-off (RO/RO) cargo ferry operating in North America. The heart of the propulsion system was the twin NG-diesel dual fuel engines coupled to constant-speed generators. The engines employ direct injection of liquid fuel and indirect injection of NG fuel. The 4-stroke, 9-cylinder, turbocharged dual-fuel engines have a maximum power output and speed of 4320 kW and 720 rpm, respectively and can be operated in either NG or diesel mode. In NG mode, the diesel pilot fuel supplies less than 10% of the total fuel energy at 10% engine load and much less at higher loads (1% at 75% load).

2.2. Engine operating conditions

Emissions were measured while the vessel operated as closely as possible to the four certification loads specified in the ISO 8178 E2 cycle (International Organization for Standardization, 1996). A summary of test points is given in Table S1. Measurements at the certification loads allowed a check of the performance of this engine compared to certification values. Some deviation from the E2 cycle loads occurred as the vessel had to maintain published schedules. Operating at 100% was impractical hence the highest load was 90%. In addition to measurements at the four E2 cycle modes, tests were carried out at idle where the vessel spent considerable time during the loading and unloading of cargo. Usually idle emissions are not discussed in most studies because this type of operation is avoided; however, it is relevant to coastal vessel operations and is considered here. One to three repeat measurements at the same loads were carried out when possible.

The vessel used liquefied natural gas (LNG) with 92% mole fraction of methane. The boil-off gas was routed to engine and burned in the gas mode. The fuel used in diesel mode or as pilot fuel in gas mode was a regular Canadian on-road use ultra-low sulfur diesel fuel (ULSD) with <15 ppm sulfur. The typical fuel used by this specific engine was NG. The vessel was first tested on NG and then on diesel.

2.3. Emission measurements

The emission measurements were conducted following ISO 8178−2 protocol (International Organization for Standardization, 1996). The exhaust was sampled from a partial flow venturi dilution system as described by Agrawal et al. (2008). A key feature in the experimental design was for all diagnostic instruments to measure from the same dilution tunnel during the campaign to eliminate dilution ratio as a variable for instrument comparison. We measured exhaust concentrations of criteria pollutants, greenhouse pollutants, and toxic gas as described in the Supplementary material with the setup shown in Fig. 1. The volumetric exhaust flow rate was calculated following EPA Method 2 (United States Environmental Protection Agency, 2011) using a pitot tube, and the calculated value agreed with engine manufacturer data and calculated values using the carbon balance method (Figure S1).

2.4. Data analysis

Emission factors at each engine load were calculated using measured concentration values of each species, exhaust flow rate, and engine power using Equation S1. Real-world activity profiles of the vessel were collected for two weeks of routine operation from the vessel on-board control and data acquisition software and

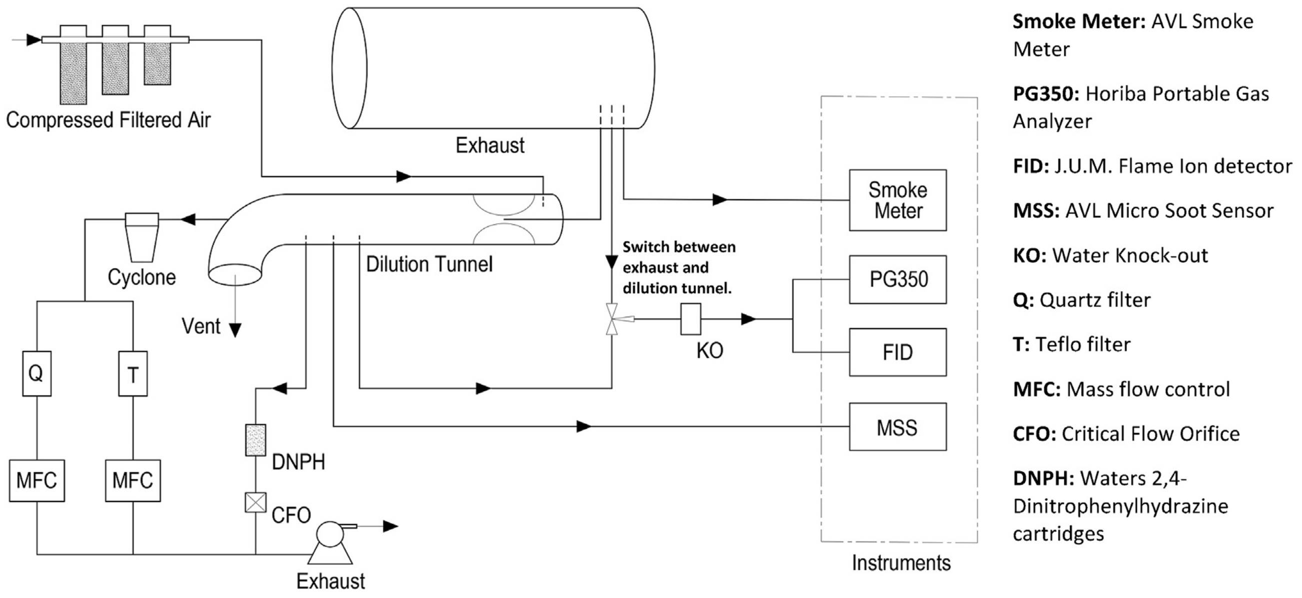

Fig. 1. Schematic of measurement setup.

categorized into 0–25%, 25–50%, 50–75%, and 75–100% engine load ranges. The engine modal activity was used as weighting factor to calculate average weighted emission factors using Equation S2. Average weighted emission factors were also calculated with certification weighting factors specified in ISO 8178- E2 cycle as comparison.

A health risk assessment of exhaust emissions for diesel and NG combustion was conducted following the guidelines from California Office of Environmental Health Hazard Assessment (OEHHA) and South Coast Air Quality Management District (SCAQMD) (California Office of Environmental Health Hazard Assessment, 2015; South Coast Air Quality Management District, 2017) for specifying operational and compliance requirements for a stationary air pollutant source by considering hazard identification, exposure assessment, dose-response assessment, and risk characterization. Emission factors when a vessel is at berth and assuming Gaussian dispersion from a stationary stack were used. The health risk assessment uses a complex model and the calculated health indices are highly dependent on the meteorological location and target population of the emissions. However, our study focuses on the impact of switching from diesel to NG; thus, we calculated the relative change of health index from diesel to NG exhaust, which allows the elimination of effects from meteorological and population factors (Supplemental Material). The carcinogenic and non-carcinogenic, acute, and chronic health risks, including maximum individual cancer risk (MICR), chronic hazard index (HIC), 8-hr chronic hazard index (HIC8) as well as acute hazard index (HIA) were calculated. HIC, HIC8 and HIA indicate the cumulative health impacts from multiple substances on the target organ in the long term, 8-hr exposure, and short term, respectively (Supplementary material).

The GWP for 20- and for 100-years were calculated: for CH_4, we used the GWP (CO_2 equivalent g/kWh) of 84 and 34 (Myhre et al., 2013) and for BC, we used 3200 and 900 (Bond et al., 2013), respectively. Global temperature change potential (GTP) (Shine et al., 2005), indicating the potential of global surface temperature change, is also presented in the Supplementary material.

3. Results and discussion

3.1. Real-world engine activity

To accurately calculate the emission contribution to an air mass, it is essential to know both the emissions at each engine load and the fraction of time that the vessel operates at the corresponding load. A concern in applying the weighting factors of ISO 8178 E2 Cycle (International Organization for Standardization, 1996) is that this vessel operated in harbor service; not the open sea. The actual ship weighting factors (Table 1) are significantly different from the standard E2 weighting factors due to the fraction of time when engine was at idle.

3.2. Modal and weighted average emission rates and factors

The modal emission rates (g/hr) and emission factors (g/kWh) for NO_x, CO_2, HCHO, CO, CH_4, total hydrocarbon (THC), and $PM_{2.5}$ as well as BC, EC and OC emissions at five engine modes are shown in Table 1. Measurement uncertainties were analyzed by considering duplicate measurement of exhaust flow, sample measurement, and instrument uncertainties (Farrance and Frenkel, 2012).

The average weighted emissions factors (EF) are listed in Table 1–b for both real-world operation and standard E2 cycle. Although Table 1–a showed that the percentage of time at each load was significantly different for these two cycles, the average weighted emission factors for most pollutants were similar. However, EF_{THC} and EF_{CH4} calculated from the E2 cycle were about 40% lower than those calculated from the actual vessel activity due to the high emissions at idle where the vessel spent 32% of operation time. Unless specified, the average weighted emission factors discussed here were calculated from the real-world cycle of this vessel. The switch from diesel to NG resulted in reductions in NO_x, $PM_{2.5}$, BC, OC and CO_2 by 92%, 93%, 97%, 92% and 18%, respectively, along with increases in CO and HCHO by 424% and 615%, respectively (Fig. 3). An average methane emission factor of 11.5 g/kWh was measured when the engine was in NG mode, while it was under limit of detection (<0.002 g/kWh) in diesel mode.

The EF_{NOx} for diesel was 9.6 ± 0.3 g/kWh, similar to the IMO Tier II certification value (9.7 g/kWh) and for NG was 0.76 ± 0.02 g/kWh,

Table 1
a: Engine activity: real-world and certification cycle. **b**: Modal and average weighted emission factors.

a	Engine Load				
	Idle	25%	50%	75%	100%
Actual Vessel Cycle	0.32	0.09	0.06	0.31	0.22
Standard E2 Cycle	0.00	0.15	0.15	0.50	0.20

b		Engine Load[a]						
Pollutant	Fuel Type	Idle	25%	50%	75%	100%	Actual Vessel Cycle	Standard E2 Cycle
NO_x (g/kWh)	**NG**	3.68 ± 0.30	1.12 ± 0.03	0.64 ± 0.02	0.52 ± 0.03	0.73 ± 0.03	0.76 ± 0.02	0.63 ± 0.02
	Diesel	15.7 ± 1.0	10.6 ± 0.5	10.9 ± 0.4	9.2 ± 0.4	9.2 ± 0.4	9.6 ± 0.2	9.4 ± 0.2
CO (g/kWh)	**NG**	36.3 ± 0.5	7 ± 0.1	4.7 ± 0.1	2.1 ± 0.1	1.5 ± 0.1	3.5 ± 0.1	2.5 ± 0.1
	Diesel	7.79 ± 0.17	0.83 ± 0.02	0.6 ± 0.01	0.36 ± 0.01	0.36 ± 0.01	0.67 ± 0.01	0.41 ± 0.01
CO_2 (g/kWh)	**NG**	1380 ± 90	572 ± 19	567 ± 18	490 ± 17	468 ± 12	521 ± 10	497 ± 10
	Diesel	1180 ± 60	588 ± 14	657 ± 11	613 ± 11	613 ± 11	635 ± 7	617 ± 7
THC[b] (g/kWh)	**NG**	188 ± 18	31 ± 1	15.6 ± 0.6	6 ± 0.3	4.4 ± 0.2	13.6 ± 0.7	8.0 ± 0.2
	Diesel	n/a	n/a	n/a	n/a	n/a	n/a	n/a
CH_4[c] (g/kWh)	**NG**	162 ± 16	25.5 ± 0.9	12.8 ± 0.5	5 ± 0.3	3.7 ± 0.2	11.5 ± 0.6	6.6 ± 0.2
	Diesel	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
HCHO (mg/kWh)	**NG**	2520 ± 520	466 ± 61	303 ± 41	139 ± 26	124 ± 17	244 ± 24	171 ± 16
	Diesel	337 ± 40	32 ± 4	16 ± 2	23 ± 2	23 ± 2	34 ± 2	22 ± 2
$PM_{2.5}$ (mg/kWh)	**NG**	126 ± 14[d]	9 ± 0.3	7.1 ± 0.2	13.9 ± 0.7	4.5 ± 0.3	13.5 ± 0.6	10.2 ± 0.4
	Diesel	2170 ± 120	212 ± 9	131 ± 4	119 ± 4	119 ± 4	199 ± 5	125 ± 3
OC (mg/kWh)	**NG**	110 ± 13[d]	13.9 ± 0.7	7.8 ± 0.4	12.6 ± 1.0	6.1 ± 0.4	13.2 ± 0.7	10.3 ± 0.6
	Diesel	2360 ± 160	151 ± 8	99 ± 4	85 ± 4	85 ± 4	172 ± 6	108 ± 3
EC (mg/kWh)	**NG**	6 ± 1[d]	1.0 ± 0.1	0.7 ± 0.1	0.7 ± 0.1	0.5 ± 0.1	0.8 ± 0.1	0.7 ± 0.1
	Diesel	277 ± 17	38 ± 2	28 ± 1	14.9 ± 0.8	14.9 ± 0.8	26.2 ± 0.8	17.6 ± 0.5
BC (mg/kWh)	**NG**	5.6 ± 2.4	1.0 ± 0.1	0.7 ± 0.1	0.8 ± 0.1	0.6 ± 0.1	0.9 ± 0.1	0.7 ± 0.1
	Diesel	296 ± 18	41 ± 2	27 ± 1	16 ± 1	16.2 ± 1	28 ± 1	19 ± 1

[a] Due to the practical limitations associated with measuring these emissions during commercial operation of the vessel, the exact engine loads for idle, 25%, 50%, 75% and 100% on this vessel were 6%, 29–31%, 47–50%, 75% and 90% for LNG mode and 5%, 26%, 50%, 75% and 75% for diesel mode.

[b] Total hydrocarbon emission factors from diesel exhaust were not reported here since a heated line was not used for the hydrocarbon analyzer.

[c] CH_4 concentration was under LOD (100 ppb) in diesel mode.

[d] Due to that only BC measurement was available at this test point, EC, OC and $PM_{2.5}$ were estimated from BC and average OC/EC ratio.

much lower than the Tier III standard (2.4 g/kWh) (Shallcross et al., 2012). The reductions of both NO_x and $PM_{2.5}$ provide immediate benefit in terms of air quality in non-attainment areas if a significant number of harbor craft and ocean-going vessels switched to NG. The EF_{CO2} for NG was 521 ± 10 g/kWh, with an 18% reduction compared to diesel. This reduction shows the potential of reducing global CO_2 emission inventory from shipping, while still far from the IMO CO_2 reduction goal of 70% by 2050 (Comer et al., 2018). The greatest reductions of NO_x (92–94%) and CO_2 (20–24%) were observed at >50% loads (Fig. 2), where more than half of engine operating time was spent. However, these emissions reduction benefits must be considered together with the HCHO and CH_4 emissions of NG, as discussed below.

Simultaneously observed with reduced NO_x and $PM_{2.5}$ were increased levels of CO, HCHO and CH_4. The higher emission factors of two major incomplete combustion products, CO and HCHO, are believed to be caused by imperfect flame propagation in uneven-temperature regions of combustion chamber from NG engines (CIMAC, 2014; Liu et al., 2013). EF_{CO} and EF_{HCHO} increased from 0.67 ± 0.01 g/kWh and 34 ± 2 mg/kWh to 3.5 ± 0.1 g/kWh and 244 ± 24 mg/kWh, respectively when switching from diesel to NG. This observation is consistent with earlier results for on-road applications for NG (Ayala et al., 2003, 2002). With respect to modal emission factors (Fig. 2), the CO and HCHO emissions for both NG and diesel modes becomes smaller as engine load increases but are still considerably higher in NG mode. With a similar trend to CO, HCHO modal emission factor data show more variability, which we attribute to the more complex sampling and analysis process for HCHO (chemical capture followed by transport to a laboratory for analysis) (Delgado et al., 2008; Sebaei et al., 2018).

Between 10^3 and 10^4 ppm of methane was measured from NG exhaust for engine loads from 90% to idle loads. This slip of unburned NG fuel is characteristic of pre-mixed natural gas combustion systems and results from similar sources as that for CO and HCHO. We calculated a coefficient of determination of 99% and 98% (Figure S2) between HCHO, CH_4, and CO concentrations at all test points, suggesting that these three incomplete combustion products share similar origins, unlike other emissions (Table S2). Near-zero methane emissions (<100 ppb) were detected from diesel exhaust since diesel contains no methane. Unlike the emission factor profiles of NO_x, CO_2 and $PM_{2.5}$ where EF is higher at idle and decreases and stabilizes at >20% engine load, EF_{CH4} decreases as engine load increases and combustion becomes less lean. This is due to an improved flame propagation with reduced excess air in combustion, leading to less unburnt methane emitted (Sommer et al., 2019; Woodyard, 2009). This observation indicates a potential for limiting CH_4 and other incomplete combustion products (CO and HCHO) from NG exhaust by advanced combustion techniques.

The average weighted emission factors of the $PM_{2.5}$ mass, EC, and OC are shown in Fig. 3. The gravimetric PM mass measurements show a good agreement with OC/EC results. The results show that OC is about and 80% of the mass emitted from diesel fuel and 95% with NG and the lubrication oil has been argued as the major source of the OC in NG exhaust (Anderson et al., 2015; Corbin et al., 2020). The average weighted $EF_{PM2.5}$ from NG is 13.5 ± 0.6 mg/kWh, 93% lower than that from diesel and well under the 100 mg/kWh Tier 4 emission standards for non-road engines. Comparing these results with a previous study (Anderson et al., 2015) shows that their $PM_{2.5}$ emission factors in NG mode from a different model of dual-fuel engine with maximum output of 7600 kW, were 1–2 orders of magnitude lower that those presented here. This difference can be explained by the difference between the engines and the approach used to measure $PM_{2.5}$ mass. This study used a standard $PM_{2.5}$ gravimetric analysis method (United States Environmental Protection Agency, 1998), and an Engine Exhaust Particle Sizer (EEPS) was used in the other study (Anderson et al.,

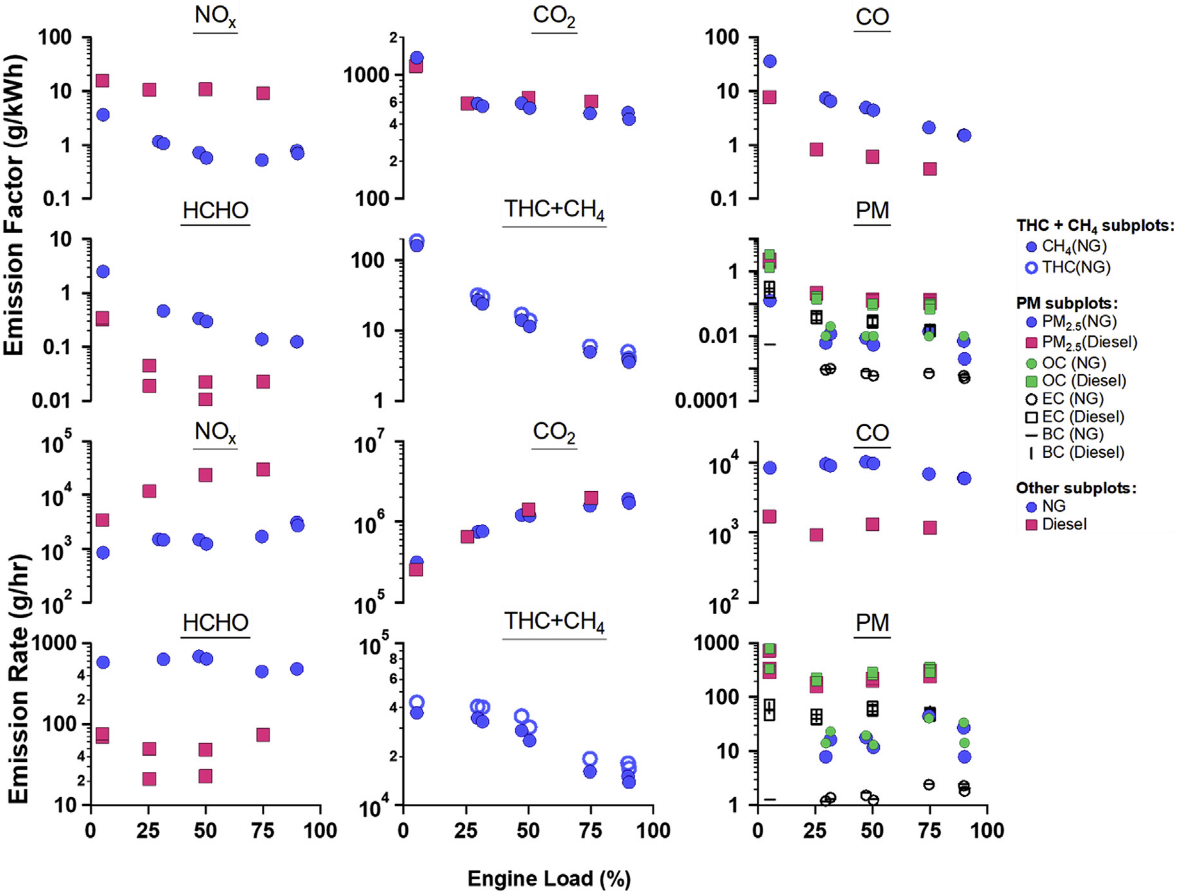

Fig. 2. Modal Emission Rates and Factors for NO$_x$, CO$_2$, CO, HCHO, CH$_4$, Total hydrocarbon (THC), and PM$_{2.5}$ as well as BC, EC and OC emissions. Square symbols represent diesel emissions and circle symbols represent NG emissions. In the "THC and CH$_4$" sub-figure, open and solid circles represent THC and CH$_4$ emissions from NG, respectively. CH$_4$ from diesel exhaust was below the detection limit (100 ppb) and THC from diesel was not measured. The "PM" sub-figure shows total PM mass, EC, OC, and BC.

2015) to measure particle number and PM$_{2.5}$ mass was calculated with particle size distribution and an assumed effective density. As stated by those authors and discussed previously (Corbin et al., 2020; Trivanovic et al., 2019), this approach is of limited accuracy. We note that the trends in PM$_{2.5}$ characteristics with load agree between the two studies. In addition (Lehtoranta et al., 2019), reported a PM emission factor of ~20 mg/kWh for a smaller marine engine (1.4 MW output) powered by NG, which is consistent with the PM$_{2.5}$ emission factor measured in this study.

3.3. *Health risks of exhaust*

Exhaust from internal combustion engines contains constituents that can harm human health and for that reason, pollutant levels in exhausts are regulated. For example, when burning NG, HCHO is the primary concern, therefore the US Environmental Protection Agency (EPA) has a HCHO exhaust limit for clean-fuel fleet for heavy duty engines of 0.067 g/kWh (United States Environmental Protection Agency, 1994) and the Occupational Safety and Health Administration (OSHA) established an 8-h maximum permissible exposure level (PEL) for HCHO (OSHA, 2011) for workers. Furthermore, IARC has classified diesel PM$_{2.5}$ as a carcinogen (Hesterberg et al., 2012; Nielsen and Wolkoff, 2010) resulting in limits on engine exhaust concentrations of PM$_{2.5}$ becoming more stringent. Both pollutants are carcinogenic and linked with a number of health effects related to eyes, skin, lung, and other human organ systems (Krzyzanowski et al., 1990; Lin et al., 2018; Nielsen and Wolkoff, 2010; Pope et al., 2002).

Assessing health issues for both fuels over the whole voyage of a vessel is complex given that for a considerable portion of time, the vessel is stationary at berth and near communities. Ship emissions at harbor have a significant impact on local pollutants level and subsequent health impacts (Alastuey et al., 2007; Xiao et al., 2018). We therefore decided to limit our comparative analysis of local health risks for both fuels to the time when the vessel was stationary and at-berth as that is when personal exposure would be the highest. When the vessel was at-berth, the engines operated at <10% load. At low loads, fuel combustion efficiency is the poorest and emissions of partial oxidation products like PM and HCHO are the highest as evidenced in the modal data from this study (Table 1—b). The health risk assessment results in Table 2 show the maximum individual cancer risk (MICR) and chronic non-carcinogenic health index are reduced by 92% and 35%, respectively when switching from diesel to NG, due to the PM reduction. However, when the non-carcinogenic shorter-term health risks were estimated, the acute hazard index (HIA) and 8-hr chronic hazard index (HIC8) increased more than 6-fold with NG due to increased HCHO levels. A difference not captured in the risk analysis is that though diluted, the aged diesel PM will remain in the atmosphere for a few days to a few weeks (Seinfeld, 2015). However, the lifetime of HCHO in atmosphere is only a few hours (Miller et al., 2008; Pamler et al., 2003) during daytime due to the reactions with OH radicals and photolysis in atmosphere, causing it to both dilute and get consumed.

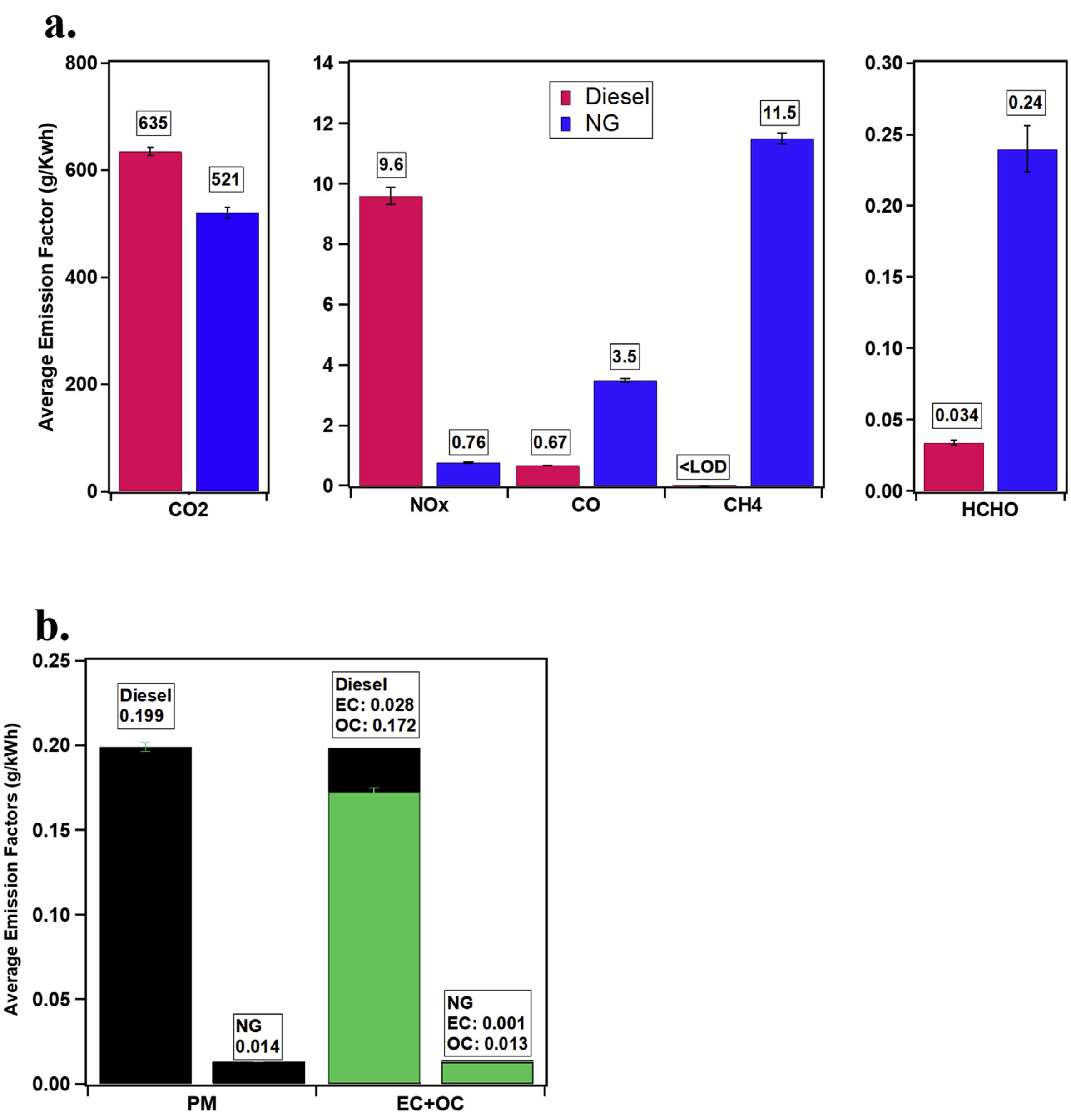

Fig. 3. Actual-ship-cycle-weighted emission factors for both fuel modes. **a**: gaseous emissions (g/kWh); **b**: particle emissions (g/kWh). EC and OC are represented by black and green color, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 2
Differences in hazards risk index and climate impacts of NG compared to diesel.

	Health Risk Index	Actual	Shore Power[a]	Cylinder Deactivation	Oxidation Catalyst
Carcinogenic	**MICR**	−92%	/	−93%	−94%
Non-Carcinogenic	**HIC**	−35%	/	−61%	−92%
	HIC8	649%	/	320%	−63%
	HIA	649%	/	320%	−63%

[a] With the use of shore power, emissions from exhaust were eliminated when vessel was at berth.

3.4. *Health risk mitigation/control strategies*

Given that harmful constituents are emitted in the engine exhaust from burning either NG or diesel, the original vessel design included mitigation/control measures to ensure that the exhaust plume was diluted before reaching public areas. For this vessel, the engineering control technology used a tall stack to move the hot, high-velocity plume away from the vessel, and reduce concentration and exposure in public areas. In this study, we discuss three mitigation strategies: 1) use of shore power at berth; 2) cylinder-deactivation; and 3) oxidation catalysts.

Following the measurements reported here, the vessel operator decided to use shore power while at-berth to eliminate the emissions from the internal combustion engines, thus eliminating the health risks associated with exhaust emissions.

For facilities where shore power is not an option, changing the engine operation, such as the use of cylinder deactivation, which improves fuel economy and reduces emissions by deactivating the fuel injection to a sub-set of cylinders (Kutlar et al., 2005; Vos et al., 2019), will reduce formaldehyde emissions at idle. During this study, the manufacturer reprogramming reduced CO by 30% and 44% respectively when two and three cylinders were deactivated at idle (Figure S3). While no HCHO measurements were performed during cylinder deactivation, we assumed the same reduction (44%) according to the correlation between CO and HCHO. With the HCHO reduction from cylinder deactivation technology, it was estimated that between NG and diesel, the reduction of the longer-term health risks such as MICR and HIC from NG would increase to 93% and 61%, respectively, and the shorter-term risks such as HIC8 and HIA would reduce from 6-fold to 3-fold (Table 2).

The use of an oxidation catalyst has been employed as a control measure to reduce HCHO emissions from NG engines. While there is no available data on the oxidation catalyst HCHO removal efficiency of emissions from a marine vessel, one study reported a 95% reduction of HCHO on NG buses with oxidation catalyst under different drive conditions, and another study on NG engines (2–5 MW) with similar power to this study, found 40%–95% HCHO removal efficiency with oxidation catalysts from various manufacturers. (Ayala et al., 2003; Kristensen, 2007). While the removal efficiency depends on the manufacturers and catalyst operation time, we assumed the maximum of reported value range (95%) HCHO removal to investigate the largest health benefit potential of using NG as a marine fuel. The health risk assessment estimated that, with catalyst installed on exhaust, all four health risk indexes from NG exhaust were 63%–94% lower compared to diesel exhaust (Table 2). This indicates the potential of short-term health benefits from NG when HCHO is properly controlled in the exhaust.

3.5. Climate impacts

In addition to the local and regional impacts of switching from diesel to NG, there are global climate impacts associated with an increase in CH_4 emissions and the reduction of CO_2 and BC emissions from shipping. These impacts result from the direct and indirect effects of radiative forcing (Chung and Seinfeld, 2002; Lashof and Ahuja, 1990) in the atmosphere for the long-term climate pollutant, CO_2, and the short-lived climate pollutants, CH_4 and BC. Although the atmospheric lifetime of black carbon is about 5–8 days (Cape et al., 2012), its climate effects may persist after deposition onto surfaces such as ice and snow (Bond et al., 2013) as the deposited BC continues to absorb solar radiation by changing the albedo of highly reflective and white surfaces. Bond et al. suggested a CO_2 equivalent global warming potential (GWP) –the amount of CO_2 equivalent climate forcing – for BC based on total climate forcing of 3200 for 20 years and 900 for 100 years with high uncertainties of −90%, +100% due to the large difficulties in BC lifetime and distribution estimation (Bond et al., 2013). Using these GWPs, we found that the BC GWP was reduced by 97% by switching from diesel fuel to NG. Such a reduction would make a significant difference for vessels sailing near snow or ice-covered surfaces, such as in the Arctic (Gong et al., 2018). We emphasize that the climate impacts of BC are more complex than GWP alone may capture (Bond et al., 2013).

Methane emissions are also of concern since their warming potential is many times that of CO_2. To describe the climate effects of methane emissions, we again use the GWP (IPCC, 2007), which is widely accepted for comparing the impact from greenhouse pollutant emissions relative to CO_2. The detailed exhaust-pollutant-based GWP modal analysis using values from Table S4, for both diesel fuel and NG at 20- and 100-years, are presented in Fig. 4. The GWP is greatest for both NG and diesel at idle with the GWP of NG being a factor of 7 that of diesel over 20 years (GWP20), and a factor of 4.8 over 100 years (GWP100), with the difference being largely attributable to the additional CH_4 emissions of the NG mode.

The additional GWP from CH_4 emissions largely outweighs the expected GWP benefit from the 18% reduction in CO_2 emissions from NG. While CH_4 accounts for the greatest fraction of GWP at lower engine loads, its contribution decreases significantly as engine load increases. The modal data shows that at loads above 75%, the GWP of NG exhaust is largely reduced. It is only 1.5 times that of diesel at 20-years, and the two fuels are equivalent at 100-years. The analysis shows the importance of NG emissions on GWP and the need to reduce the time spent at low engine loads, especially at idle.

The average GWP20 (Fig. 4) from NG exhaust is 1515 CO_2 equivalent g/kWh, about 109% higher than that from diesel exhaust, 725 CO_2 equivalent g/kWh. Due to the shorter lifetime of CH_4 in the atmosphere (Myhre et al., 2013) compared to CO_2, while still 38% higher than that of diesel, NG GWP100 (914 CO_2 equivalent g/kWh) decreases 40% relative to GWP20, reflecting the atmospheric oxidation of methane effects to CO_2. A similar GTP analysis is presented in the Supplementary material.

In order to understand the comprehensive climate change impacts of switching from diesel to NG for the whole fuel cycle, we estimated GWP from well-to-tank (WTT) emissions including emissions from production, purification, and distribution, using WTT emission values of NG (6.9 g CO_2 eq/MJ) and diesel (10.9 g CO_2 eq/MJ) from Global Transport Model of energy and emissions of shipping (MAN B&W, 2019). NG shows a GHG emission benefit from well to tank compared to diesel due to its lower WTT carbon emissions. We added the 100-year GWP from WTT emissions in Fig. 4 and found that when considering impacts of the whole fuel cycle, NG provides further GWP benefits when engine load was 75% or higher and the GWP gap between NG and diesel for smaller engine load was reduced.

3.6. Climate change mitigation/control strategies

While the CO_2 emissions are unavoidable when the engine is operating, reducing CH_4 emissions is possible via different strategies such as using shore power or cylinder deactivation. Table 3 shows that with these mitigation methods, the GWP increase from NG is reduced due to lower methane emissions.

As stated previously for health effects, use of shore power when at-berth is the simplest and most efficient control measure. With shore power, the average weighted emission factors of CO_2, CH_4 and BC were estimated to decrease by 6%, 50%, and 22% to 489 g/kWh, 5.82 g/kWh, and 7 mg/kWh, respectively. By reducing emissions of greenhouse pollutants, the use of shore power reduces the GWP20 and GWP100 from NG exhaust, leading to a comparable GWP100 between NG and diesel.

Cylinder deactivation technology is an option for vessels when no shore power is available. When the engine was at idle and three cylinders were deactivated, CH_4 emissions were reduced by ~56%–60% (Sommer et al., 2019), leading to the decrease of GWP20 and GWP100 from NG exhaust while still 67% and 20% larger compared to the GWP of diesel.

In addition, blending hydrogen in NG or using renewable NG (RNG) are also options to reduce carbon emissions and the corresponding climate impacts. While the emissions from RNG or NG and hydrogen blends were not measured and directly compared in this study, other studies have showed that blending 20% hydrogen in natural gas reduced brake-specific CO_2 emission factor by 5–15% (Akansu et al., 2004; Navarro et al., 2013), which reduces the

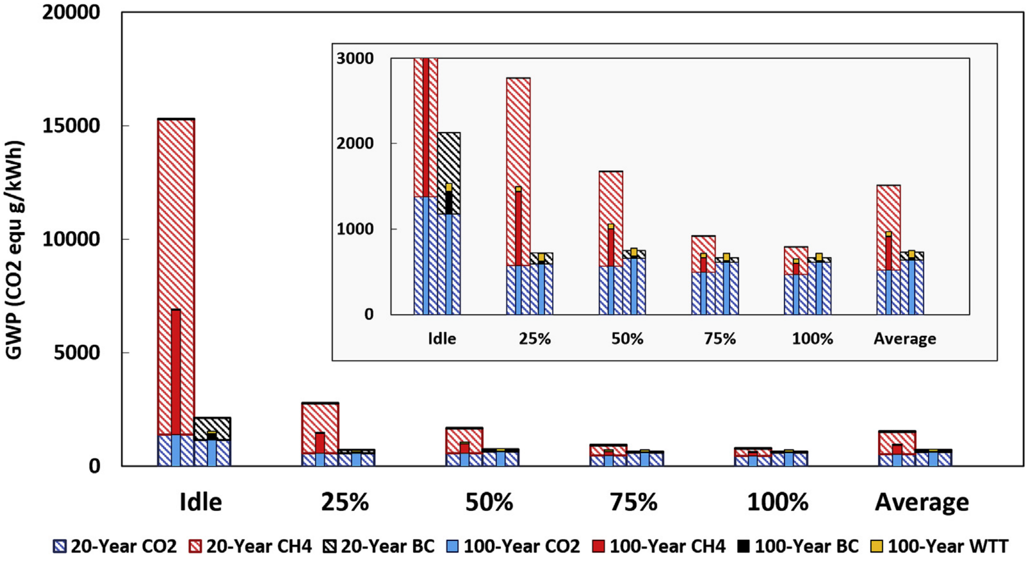

Fig. 4. Modal and average weighted CO_2 equivalent GWP with zoomed-in view (0–3000 CO_2 equivalent g/kWh zoomed-in subfigure) from NG and diesel fuel exhaust using 20-year and 100-year timeframe at different engine loads. Well-to-tank (WTT) GWP were estimated using emission values from GloTraM (MAN B&W, 2019) and shown in 100-year GWP only. Similar GTP calculations are presented in Figure S4.

Table 3
Differences in global warming potential of NG exhaust emissions compared to diesel (calculated as $[GWP_{NG} - GWP_{diesel}]/GWP_{diesel}$).

GWP	Actual	Shore Power	Cylinder Deactivation
20-year	109%	37%	67%
100-year	38%	4%	20%

corresponding GWP from CO_2, and liquefied biogas reduced ~70% life-cycle GWP100 (CO_2 equivalent g/km) compared to LNG (Bengtsson et al., 2012, 2014), indicating the potential climate benefits from NG.

4. Conclusion

Switching a dual-fuel marine vessel from diesel fuel to natural gas reduced emissions of NO_x, $PM_{2.5}$, CO_2, and BC by 92%, 93%, 18% and 97%, respectively, whereas CO and HCHO emissions increased by factors of 4 and 6, respectively, and CH_4 increased from <0.002 g/kWh to 11.5 g/kWh. The reductions in criteria pollutants such as PM and NO_x are significant and would have a notable effect on local air quality near coastal areas where a great fraction of these pollutants come from shipping. Over the long term, the reduction in PM from NG leads to a 92% lower cancer risk but the short-term effects of high levels of formaldehyde were of concern. However, mitigation measures are available to significantly reduce formaldehyde emission rates below that of diesel. The global warming analysis showed that NG increased 100-year fuel cycle GWP by 29%. However, when the engine operated at higher loads (>75%), the impact of the reduction of CO_2 outweighed that of CH_4 emissions, making GWP of NG comparable to diesel for these loads. In the long term, the global climate risks associated with unburned CH_4 and substantial CO_2 suggest the necessity of transitioning from fossil NG to renewable NG.

CRediT authorship contribution statement

Weihan Peng: Methodology, Investigation, Data curation, Formal analysis, Visualization, Writing - original draft. **Jiacheng Yang:** Methodology, Investigation. **Joel Corbin:** Investigation, Writing - review & editing. **Una Trivanovic:** Investigation, Writing - review & editing. **Prem Lobo:** Conceptualization, Investigation, Writing - review & editing. **Patrick Kirchen:** Methodology, Investigation, Resources. **Steven Rogak:** Visualization, Writing - review & editing. **Stéphanie Gagné:** Conceptualization, Project administration, Funding acquisition, Writing - review & editing. **J. Wayne Miller:** Conceptualization, Funding acquisition, Supervision, Investigation, Writing - review & editing. **David Cocker:** Supervision, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.115404.

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