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Corrigendum

# Gas phase carbonyl compounds in ship emissions: Differences between diesel fuel and heavy fuel oil operation



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## ABSTRACT

Gas phase emission samples of carbonyl compounds (CCs) were collected from a research ship diesel engine at Rostock University, Germany. The ship engine was operated using two different types of fuels, heavy fuel oil (HFO) and diesel fuel (DF). Sampling of CCs was performed from diluted exhaust using cartridges and impingers. Both sampling methods involved the derivatization of CCs with 2,4-Dinitrophenylhydrazine (DNPH). The CCs-hydrazone derivatives were analyzed by two analytical techniques: High Performance Liquid Chromatography-Diode Array Detector (HPLC-DAD) and Gas Chromatography-Selective Ion Monitoring-Mass Spectrometry (GC-SIM-MS). Analysis of DNPH cartridges by GC-SIM-MS method has resulted in the identification of 19 CCs in both fuel operations. These CCs include ten aliphatic aldehydes (formaldehyde, acetaldehyde, propanal, isobutanal, butanal, isopentanal, pentanal, hexanal, octanal, nonanal), three unsaturated aldehydes (acrolein, methacrolein, crotonaldehyde), three aromatic aldehyde (benzaldehyde, *p*-tolualdehyde, *m.o*-molualdehyde), two ketones (acetone, butanone) and one heterocyclic aldehyde (furfural). In general, all CCs under investigation were detected with higher emission factors in HFO than DF. The total carbonyl emission factor was determined and found to be 6700 and 2300  $\mu$ g kWh<sup>-1</sup> for the operation with HFO and DF respectively. Formaldehyde and acetaldehyde were found to be the dominant carbonyls in the gas phase of ship engine emission. Formaldehyde emissions factor varied from 3870  $\mu$ g kWh<sup>-1</sup> in HFO operation to 1540  $\mu$ g kWh<sup>-1</sup> in DF operation, which is 4-30 times higher than those of other carbonyls. Emission profile contribution of CCs showed also a different pattern between HFO and DF operation. The contribution of formaldehyde was found to be 58% of the emission profile of HFO and about 67% of the emission profile of DF. Acetaldehyde showed opposite behavior with higher contribution of 16% in HFO compared to 11% for DF. Heavier carbonyls (more than two carbon atoms) showed also more contribution in the emission profile of the HFO fuel (26%) than in DF (22%).

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## 1. Introduction

Ships and marines emissions make a considerable contribution to both gaseous and particulate air pollutants in the atmosphere, particularly on the ports and coastal regions (Miola and Ciuffo, 2011). These toxic pollutants have been associated with serious adverse health effects, including premature death and respiratory symptoms (Corbett et al., 2007; Song, 2014). Carbonyl compounds (CCs) are well known to be critically important in atmospheric chemistry (Sawant et al., 2007) and they attract immense attention due to their adverse health effects on humans (Weng et al., 2010). Certain CCs such as formaldehyde, acetaldehyde and acrolein are known to be toxic, mutagenic or carcinogenic and thus have been

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identified as hazardous air pollutants. (Bhattacharya and Tulsawani, 2008), (IARC, 2006). A summary of health hazards associated with carbonyl compounds are listed Table 1 (Karavalakis et al., 2010). The primary emission sources of CCs are diverse and include: motor vehicles (Pang and Mu, 2007), incomplete combustion of hydrocarbon fuels in industrial processes (Liu et al., 2006), cigarette smoke (Pang and Lewis, 2011), biomass burning (Seco et al., 2007), the frying process as a by-product of vegetable oil degradation (Katragadda et al., 2010), wine production (Culleré et al., 2009) and urban incineration (Dai et al., 2012). CCs can also be produced as secondary airborne pollutants via the photochemical oxidation of atmospheric hydrocarbons (Moussa et al., 2006).

Sampling and analysis of CCs started in the middle of the fifties during the last century. Earliest measurements were performed for CCs in cigarette smoke by (Touey, 1955), who used a precipitation method to assess the total aldehyde content by their reaction with (5,5-dimethyl-1,3-cyclohexanedione) in gas-absorption flasks to form dimedon. Later techniques resolved CCs in tobacco smoke using liquid chromatography based on their 24dinitrophenylhydrazine derivatives (Houlgate et al., 1989). Recently, different methods were used to analyze CCs in different sources. High performance liquid chromatography (HPLC) represent the most convenient method for the analysis with conventional UV–VIS detectors (Feng et al., 2011: Karavalakis et al., 2011). Moreover, on-line capillary liquid chromatography (Prieto-Blanco et al., 2011) and gas chromatography mass spectrometry (GC-MS) (Pang and Lewis, 2011) were employed in the investigation of the CCs. Most of these analytical methods depend on the same principle of sampling, which involves the derivatization of CCs with 2.4-dinitrophenylhydrazine (DNPH) to produce stable CCs-hydrazone derivatives. Carbonyl compounds emission from heavy-duty diesel engines have been intensively investigated recently using different fuel blends such as ethanol-diesel blends (Song et al., 2010) or biodiesel blends (Karavalakis et al., 2011). These studies revealed that the use of biodiesel or ethanol-diesel fuels led to significant increases in the emission of most carbonyl compounds due to the high oxygen containing additives. To our knowledge there is no any study discussing the carbonyl compounds in the emissions of ship engines operated with heavy fuel oil or distillate oils. In this study we investigated carbonyl compounds emission in the gas phase of a ship diesel engine fueled with heavy fuel oil (HFO) and standard diesel fuel (DF, contained 3.2% biodiesel) respectively. The study was carried out as part of a project of the Helmholtz Virtual Institute for Complex Molecular System in Environmental Health (HICE). CCs were collected using 2,4-dinitrophenylhydrazine (DNPH) cartridges and impingers. The

Table 1

Health hazards associated with carbonyl compounds emissions.

Compound	NOAEL <sup>a</sup>	LOAEL <sup>b</sup>	Health hazard	l	Reference(s)
Formaldehyde	15 mg/kg day	82 mg/kg day	Recognized:	Carcinogen	(Til et al., 1989; EPA-HEN, 1994)
			suspected:	Gastrointestinal or liver toxicant	(ATSDR, 2004)
				Immunotoxicant	(EEC, HAZMAP)
				Neurotoxicant	(RTECS)
				Reproductive toxicant	(Frazier and Hage, 1998)
				Respiratory toxicant	(EPA-HEN, 1994; ATSDR, 2004)
				Skin or sense organ toxicant	(EEC, EPA-HEN, 1994)
Acetaldehyde	$8.7 \text{ mg/m}^3$	$16.9 \text{ mg/m}^3$	Recognized:	Carcinogen	(EPA-HEN, 1994)
	01	0,	suspected:	Developmental toxicant	(Jankovic and Drake, 1996)
				Immunotoxicant	(HAZMAP)
				Kidney toxicant	(RTECS)
				Neurotoxicant	(RTECS)
				Respiratory toxicant	(HAZMAP, EPA-HEN, 1994)
				Skin or sense organ toxicant	(HAZMAP, EPA-HEN, 1994)
Acrolein	0.05 mg/kg day	_	Recognized:	-	
Actor	0.05 mg/kg duy		suspected:	Carcinogen	(EPA-HEN, 1994)
			suspected.	Cardiovascular or blood toxicant	(ATSDR, 2004)
				Developmental toxicant	(Jankovic and Drake, 1996)
				Gastrointestinal or liver toxicant	(M. J. Malachowski, 2013)
				Neurotoxicant	(RTECS)
				Respiratory toxicant	(ATSDR, 2004)
				Skin or sense organ toxicant	
Methacrolein	4.00 nnm	15.3 ppm	Recognized:	Highly irritating to the eyes, nose, throat and lungs.	(HAZMAP, ATSDR, 2004) (Richard and Lewis, 2007)
	4.99 ppm	13.5 hhii	Recognized.		(Richard and Lewis, 2007)
				Epithelial inflammatory, atrophic and metaplastic changes in the dorsal meatus	
A	000 mm/lim day	1700 mm m/line days	Deservised	8	(66016)
Acetone	900 mg/kg-day	1700 mg/kg day	Recognized:	Inhalation: can irritate the nose and throat.	(CCOHS)
				Skin contact: may cause mild irritation.	
				EYE IRRITANT. Causes moderate to severe irritation.	
				Effects of long-term (chronic) exposure: can cause	
	2			dry, red, cracked skin (dermatitis) following skin contact	
Propanal	8 mg/m <sup>3</sup>	-	Recognized:	-	—
			suspected:	Neurotoxicant incidence of atrophy of the olfactory	(RTECS)
				epithelium in male rats	
Crotonaldehyde	0.23 μg/m <sup>3</sup>	-	Recognized:	-	-
			suspected:	Carcinogen	(EPA-TRI, 1994)
				Respiratory toxicant	(HAZMAP)
				Skin or sense organ toxicant	(HAZMAP)
Benzaldehyde	200 mg/kg	400 mg/kg day	Recognized:	Forestomach lesions, kidney toxicity	(Kluwe et al., 1983)
	day or			Necrotic and degenerative lesions of the brain,	
	26 mg/m <sup>3</sup>			renal tubular necrosis	
				and epithelial hyperplasia, hyperkeratosis of the	
				forestomach (in rats)	

<sup>a</sup> No-observed-adverse-effect level.

<sup>b</sup> Lowest-observed-adverse-effect level.

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