



Assessing fuel spill risks in polar waters: Temporal dynamics and behaviour of hydrocarbons from Antarctic diesel, marine gas oil and residual fuel oil



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ABSTRACT

As part of risk assessment of fuel oil spills in Antarctic and subantarctic waters, this study describes partitioning of hydrocarbons from three fuels (Special Antarctic Blend diesel, SAB; marine gas oil, MGO; and intermediate grade fuel oil, IFO 180) into seawater at 0 and 5 °C and subsequent depletion over 7 days. Initial total hydrocarbon content (THC) of water accommodated fraction (WAF) in seawater was highest for SAB. Rates of THC loss and proportions in equivalent carbon number fractions differed between fuels and over time. THC was most persistent in IFO 180 WAFs and most rapidly depleted in MGO WAF, with depletion for SAB WAF strongly affected by temperature. Concentration and composition remained proportionate in dilution series over time. This study significantly enhances our understanding of fuel behaviour in Antarctic and subantarctic waters, enabling improved predictions for estimates of sensitivities of marine organisms to toxic contaminants from fuels in the region.

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1. Introduction

The seas surrounding the Antarctic continent are some of the most remote and unpolluted marine environments on earth (Halpern et al., 2008). A key concern for the Southern Ocean and for Antarctic marine waters is the accidental release of large volumes of petroleum fuel (Poland et al., 2003; Ruoppolo et al., 2013). The risk of such contamination is rising with increasing vessel traffic from an expanding tourism industry, fishing, and research and supply vessels operating in the area. Ship-to-shore fuel transfer and storage of fuel at Antarctic research stations poses a controlled but additional risk (Snape et al., 2006; Aronson et al., 2011). Polar environmental factors such as extreme weather, ice and isolation increase the risk of accidents and contamination and are also likely to impede or restrict responses to fuel pollution incidents.

The Antarctic near-shore environment is ice-covered for much of the year which reduces wave action resulting in low energy sea conditions. This creates the potential for spills to be enclosed or encapsulated in ice and thus persist in the environment for extended periods. Low temperatures and presence of ice can significantly reduce oil spreading, increase viscosity and reduce the evaporation rate of the volatile components of oils in polar waters (Payne et al., 1991; Fingas and Hollebone, 2003; Faksness and Brandvik, 2008b; Brandvik and Faksness, 2009). In addition, biodegradation rates are much slower than might be expected in temperate areas

(Siron et al., 1995; Delille et al., 1998; Garrett et al., 2003; McFarlin et al., 2014). These factors may result in prolonged exposure of marine organisms to hydrocarbons in Antarctic waters (Stark et al., 2003).

In the event of a spill, only a certain proportion of the petroleum compounds within a fuel are water-soluble, and will dissolve into seawater producing a water accommodated fraction (WAF) (Singer et al., 2000). The hydrocarbon concentration of a WAF is dependent on fuel composition and the solubility of the various compounds within it, and is influenced by environmental conditions, particularly temperature. Hydrocarbon solubility decreases at low temperatures, however, aromatics become enhanced in seawater due to reduced evaporation (Payne et al., 1991). The components in a WAF are considered to be a major contributor to ecological impacts of an oil spill as soluble compounds are bioavailable to marine organisms in the water column and therefore have the potential to cause toxic effects (Neff, 2002; Faksness and Brandvik, 2008a).

An important consideration in the assessment of the impact of fuel spills in Antarctic and subantarctic waters is the toxicity of these water-soluble hydrocarbons to marine organisms. However, there is a paucity of such toxicity data in general for polar species and particularly for Southern Ocean organisms (Chapman and Riddle, 2005; Olsen et al., 2013; Bejarano et al., 2014). Studies of the effects of petroleum spills in cold and ice-covered waters have largely tested effects of crude oils on Arctic and boreal species (e.g. Barron et al., 2003; Perkins et al., 2003; Hansen et al., 2011; Gardiner et al., 2013). However, under the terms of the current Protocol on Environmental Protection to the Antarctic Treaty (Madrid Protocol), oil exploration is not undertaken and crude oil is not

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carried in bulk in Antarctic waters. The carriage of heavy residual fuel oil is also regulated (International Maritime Organisation, 2010) and the heaviest fuel oil now allowed in Antarctic Treaty waters is an intermediate grade (IFO 180). Light diesel and gas oils, and intermediate residual fuel oils are commonly carried as bunker fuels in Antarctic and subantarctic marine areas (ATCM, 2005) and are transferred from ship to shore to supply the energy needs of research stations. Although there have been some studies with the types of fuel oils used and carried in the Southern Ocean (e.g. McDonald et al., 1995; Smith and Simpson, 1995; Stark et al., 2003; Lane and Riddle, 2004; Payne et al., 2014) current understanding of fuel toxicity to subantarctic and Antarctic species is very limited.

Toxicity tests with polar ectotherms generally need to be conducted over extended exposure durations to account for the slower response of organisms at low temperatures (King and Riddle, 2001; Chapman and Riddle, 2005). One challenge in conducting such tests with petroleum fuel contaminants is that the concentration of volatile components in WAFs of petroleum fuels change through time, which result in changing concentrations and compositions of treatments during static tests (Redman and Parkerton, 2015). The initial measured concentration at the start of a toxicity test is commonly used in determining point estimates. For fuels however, this initial concentration does not adequately represent the actual exposure concentration throughout the test duration as there is likely to be considerable loss of hydrocarbons from test solutions over time. Its use is therefore likely to underestimate toxicity. A more accurate estimation of the overall exposure concentrations can be gained by measuring hydrocarbons during the course of exposure in toxicity testing and modelling total hydrocarbon content (THC) of WAFs through time to determine exposure concentrations, which are then used to derive point estimates (Tsvetnenko and Evans, 2002; Landrum et al., 2012).

Measuring the THC in a petroleum contaminated water sample quantifies the contributions of all hydrocarbons in the dissolved and accommodated fraction. This metric is often used to estimate the sensitivities of organisms to petroleum products. It is also utilised for risk assessments in a spill situation, including the development of species sensitivity distributions (Barron et al., 2013; Bejarano et al., 2014) and setting remediation targets and water quality guidelines for hydrocarbon contamination (Tsvetnenko, 1998; Tong et al., 1999). However, individual hydrocarbon compounds within the THC are usually not identified and the detailed chemical composition is often unknown, which limits our understanding of these complex mixtures. An increased understanding can be gained from data obtained using methods such as high performance gas chromatography-flame ionisation detection (GC-FID), which allows separation of THC into equivalent carbon number (ECN) ranges defined by approximate carbon numbers and boiling point in the GC column (Tong and Karasek, 1984; TPHCWG, 1997). ECNs can be expressed as groups (e.g. F1–3), which allows the complex hydrocarbon content within a fuel to be grouped into fractions based on individual compound mass. The relative contribution of each carbon number group to the total WAF content can be measured and temporal patterns in proportions in each fraction described. Fractions defined by ECN provide a useful metric for use in toxicity testing (Erlacher et al., 2013).

The aim of this study was to investigate hydrocarbon concentration and composition in WAFs generated at Antarctic and subantarctic marine temperatures (0 and 5 °C) from Special Antarctic Blend diesel (SAB), marine gas oil (MGO), and an intermediate grade of residual fuel oil (IFO 180). The patterns of subsequent hydrocarbon losses from these WAFs are compared in time series exposures over seven days at 0 and 5 °C, including the relative proportions in ECN groups.

2. Methods

2.1. Test fuels description and source

Two diesel fuels (SAB and MGO) and a residual fuel oil (IFO 180) were selected for this study based on their common use within the Antarctic and subantarctic region. Table 1 shows physical properties of the three

fuels. SAB is a middle distillate produced by a straight run refinery process and contains *n*-alkanes in the range *n*-C_{9–14}, with the highest peak around *n*-C₁₂, along with other branched and cyclic alkanes and aromatic hydrocarbons (Snape et al., 2005). MGO is a marine distillate composed mainly of *n*-alkanes, cycloalkanes and aromatic hydrocarbons, predominantly in the *n*-C_{9–25} range (Shell, 2010). IFO 180 is produced by straight run and catalytic cracking processes and is a combination of mainly residual oil blended with around 6 to 7% of middle distillate components of *n*-alkanes, branched and cyclic alkanes and aromatic hydrocarbons. Composition of an IFO 180 can vary according to source oil, refinery process and percentage of distillate added (Uhler et al., 2007). This complex mixture can have a carbon range from C₆ to >*n*-C₄₀ and may contain polycyclic aromatic hydrocarbon compounds from cracked components (BP, 2006).

SAB was obtained from a storage tank at Davis Station, East Antarctica in January 2010. The MGO sample was drawn from the bunker of the Australian research and supply vessel *Aurora Australis*, after refuelling in Hobart in December 2009. IFO 180 was supplied by BP Australia from the Whinstanes Terminal, Brisbane in October 2009.

2.2. Production of the water accommodated fraction

Replicate experiments were conducted to determine the THC and ECN ranges in WAFs from SAB, MGO and IFO 180 fuels in seawater at Antarctic (0 °C; *n* = 7) and subantarctic (5 °C; *n* = 1) temperatures. The methods for preparing WAFs were based on standard protocol (Singer et al., 2001) with some recommended modifications (Barron and Ka'aihue, 2003). These experiments were run in conjunction with toxicity tests with Antarctic and subantarctic marine invertebrates (Harrison et al., 2013; Brown et al., 2015a).

Natural seawater was filtered (0.5 µm) to produce filtered seawater (FSW) and chilled to test temperature. Average salinity of FSW was 34.9‰, pH 8.10, and dissolved oxygen 10.98 mg/L.

Water accommodated fractions of each fuel made at 0 °C had a fuel to seawater loading of 1:25 (*n* = 4) or 1:40 (*n* = 3). The WAFs made at 5 °C (*n* = 1) had a fuel to seawater loading of 1:40. Chilled 10 L glass aspirator bottles were filled with FSW at the required temperature leaving 20% headspace. Fuel was carefully added to the FSW at the volume specific to the loading ratio for each experiment. The bottles were sealed and set on magnetic stirrers in unlit temperature-controlled cabinets at either 0 ± 1 or 5 ± 1 °C. The magnetic bar rotation speed was ~200 rpm, which created a minimal vortex of <1 cm depth. The 1:40 WAFs (at 0 and 5 °C) were stirred for ~26 h and drawn off after a 10 min settling period (after Singer et al., 2000). The 1:25 WAFs (at 0 °C only) were stirred for 18 h and WAFs were drawn off after a 6 h settling period. These WAF generating methods replicated methods used in companion toxicity tests. The difference in stirring times reflected a change in methodology between two series of toxicity tests when a WAF settlement period was incorporated.

WAFs were drawn off into chilled glass bottles and samples were either, extracted immediately for measurement of initial THC and ECN fractions (WAF initial concentration), or placed into climate controlled cabinets in time series tests. See Fig. 1 and Table 2 for test design.

2.3. Measurement of initial concentrations in water accommodated fractions

Samples were extracted immediately after WAFs were decanted for measurement of the initial THC and ECN fractions in WAFs (Time 0).

Table 1
Physical properties of the three test fuels.

Test fuel		Vapour pressure (kPa)	Viscosity (cSt)	Density (g/cm ³ at 15 °C)
Special Antarctic Blend diesel	SAB	<0.1	<7 at 40 °C	0.81
Marine gas oil	MGO	<0.1	3.0 at 40 °C	0.85
Intermediate residual fuel oil	IFO 180	<0.1	180 at 50 °C	0.99

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