



Review

Current understanding of the ecological risk of mercury from subsea oil and gas infrastructure to marine ecosystems

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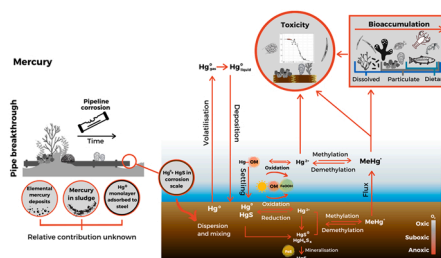
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HIGHLIGHTS

- Mercury (Hg) species associated with offshore infrastructure are determined.
- Hg accumulation and ecological risks are addressed in the decommissioning context.
- Potential bioaccumulation risks and impacts on humans are discussed.
- Understanding of Hg risk to the marine ecosystem is needed for decision making.

GRAPHICAL ABSTRACT



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ABSTRACT

Many oil and gas fields are nearing production cessation and will require decommissioning, with the preferred method being complete infrastructure removal in most jurisdictions. However, decommissioning *in situ*, leaving some disused components in place, is an option that may be agreed to by the regulators and reservoir titleholders in some circumstances. To understand this option's viability, the environmental impacts and risks of any residual contaminants assessed. Mercury, a contaminant of concern, is naturally present in hydrocarbon reservoirs, may contaminate offshore processing and transmission infrastructure, and can biomagnify in marine ecosystems. Mercury's impact is dependent on its speciation, concentration, and the exposure duration. However, research characterising and quantifying the amount of mercury in offshore infrastructure and the efficacy of decontamination is limited. This review describes the formation of mercury-contaminated products within oil and gas

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infrastructure, expected exposure pathways after environmental release, possible impacts, and key research gaps regarding the ecological risk of *in situ* decommissioned contaminated infrastructure. Suggestions are made to overcome these gaps, improving the *in situ* mercury quantification in infrastructure, understanding environmental controls on, and forecasting of, mercury methylation and bioaccumulation, and the cumulative impacts of multiple stressors within decommissioned infrastructures.

1. Introduction

As production in offshore oil and gas fields near the end of their life, oil and gas infrastructure will require decommissioning (Wood Mackenzie, 2017). The current methods for decommissioning entails either (1) the removal of all infrastructure, (2) the removal of topside with the submerged sections of the platform structure left *in situ*, or (3) the partial removal or modification options of the infrastructure (Bull and Love, 2019). Approximately 12,000 offshore assets that facilitate the extraction and transportation of produced oil and gas resources have been installed globally as of 2017 (Ars and Rios, 2017). Pipelines are a major component of the asset inventory and therefore represent a large proportion of the decommissioning cost. Within the producing basins of Australia and the North Sea, pipelines such as infield flowlines and export and inter-field pipelines contribute to 33 % and 10–15 % of the total decommissioning liability, respectively (Advisian Pty Ltd, 2020; Shen et al., 2017).

There are many factors considered when decommissioning, including environmental impacts, financial costs, human safety, local regulations and social acceptability (Shaw et al., 2018). It has been proposed that a multi-criteria decision matrix should be adopted when deciding a decommissioning approach. Many such frameworks have been proposed and reviewed (see Capobianco et al., 2021, Fowler et al., 2014, Li and Hu, 2021 and Tan et al., 2021 for more details). However, the lack of quantitative data on the potential inventory of residual contaminants at the point of decommissioning (i.e., post cleaning) and likely environmental impacts preclude effective multi-criteria assessments (Capobianco et al., 2021; Fowler et al., 2014).

There has been a growing interest in the *in situ* decommissioning of offshore infrastructure. Recent studies have identified a range of benefits from this approach, including the formation of artificial reef habitats leading to increased marine biomass and biodiversity (Fowler et al., 2018; Macreadie et al., 2011). However, studies of the detrimental impacts of oil and gas production systems to the marine environment have mainly focused on hydrocarbon contaminants from the operational phase of oil and gas production, rather than the impacts from any residual contaminants left *in situ*. For example, recent reviews have highlighted the limited study and understanding of the potential impacts of residual naturally occurring radioactive materials that may be left within decommissioned offshore infrastructure (Koppel et al., 2022; MacIntosh et al., 2021). Similar knowledge gaps exist for other contaminants, including mercury.

Mercury has been identified as a contaminant of concern for the *in situ* decommissioning of oil and gas infrastructure because of its prevalence within most oil and gas reservoirs around the world (Wilhelm, 2001b). Mercury presents as several different species in oil and gas systems, namely elemental mercury (Hg^0), inorganic mercury (e.g., $HgCl_2$, HgS) and organic mercury (e.g., dimethylmercury, methylmercury) (Enrico et al., 2020). As these different species co-exist within the production fluids (gas, hydrocarbon, and water), they interact differently with the infrastructure used to transport and process these fluids. This leads to the formation of various products that can accumulate within the infrastructure that are subjected to decommissioning decisions. This is of concern for subsea pipelines because they have direct and long-term exposure to both raw well stream (i.e., reservoir fluid) and processed hydrocarbons. Coupled with their large spatial scale, they may be a significant source of mercury pollution to marine environments depending on whether they are removed, repurposed, relocated,

or left *in situ*.

Current research into the impacts of mercury related to oil and gas production mainly focus on consequences arising from produced water discharges, drilling muds, or cutting piles (Bakke et al., 2013; Neff, 2008). Findings have been mixed, with some studies reporting elevated mercury having a limited impact on the environment (Gillett et al., 2020; Regoli et al., 2019). Other studies have shown increasing contaminant concentrations negatively alter natural eco-systems by affecting the abundance and diversity of certain organisms near the oil and gas production area (Montagna and Harper, 1996; Olsgard and Gray, 1995). However, most of these studies only consider mercury contaminants from the operational phase of oil and gas production, there is a lack of research about the ecological risk of residual mercury in decommissioned infrastructure left *in situ*. The understanding of mercury transport, exposure pathways and toxicity in marine environments and biota is also relatively limited compared to freshwater or terrestrial ecosystems.

This information is vital in assessing future risks of decommissioning, as mercury is highly toxic to marine organisms. Its effects on marine ecosystems are highly dependent on mercury speciation, concentration, and exposure duration (Wiener, 2013). For example, methylmercury is bioavailable and readily absorbed by soft tissues in organisms, where it is not easily eliminated (Harding et al., 2018a). This leads to its biomagnification across the food web. This has led to devastating effects not only on wildlife but also on humans (Yorifuji, 2020), as seen by incidents such as the Minamata Bay disaster in the 1950s and 60s (Kitamura et al., 2020).

This review describes the formation of mercury-contaminated products in oil and gas production systems focusing on subsea pipelines. The expected mercury exposure pathways in the marine environment, the possible impacts of mercury to the marine environment, and key research gaps in knowledge relating to the assessment of the ecological risk of decommissioning *in situ* mercury-contaminated oil and gas infrastructure are then discussed. Recommendations of research needed to improve ecological risk assessments and *in situ* mercury measurement techniques are then given to ensure decommissioning decision making is informed by a holistic understanding of benefits and risks.

2. Roadmap

This review will address the accumulation and speciation of mercury in oil and gas infrastructure (Section 3)Section 3.1Sections 3.2 and 3.3. Following this, the ecological risks of mercury in the marine ecosystem will be discussed (Section 4), focussing on the exposure pathways (Section 4.1) and potential hazards to the marine environment as well as humans (Section 4.2). Other considerations that are discussed include mitigation options (Section 5.1) and the current measurement techniques of mercury in subsea pipelines (Section 5.2). Next, research needs are assessed (Section 6), which deals with aspects of the development of environmental risk assessment frameworks (Section 6.1), subsea pipeline inventory and *in situ* quantification of mercury (Section 6.2), the variability of mercury bioavailability and methylation in marine environments (Section 6.3), the biomagnification in food webs (Section 6.4), and the need to consider cumulative impacts and multiple stressors (Section 6.5).

3. Mercury in oil and gas infrastructure

Mercury is found ubiquitously in oil and gas systems, existing as elemental, inorganic, and organic species, each with different chemical and physical properties. An understanding of mercury in terms of its sources and behaviour within oil and gas systems is important to understanding its partitioning tendencies within the production fluids and the formation of discrete products that accumulate within the infrastructure.

3.1. Mercury in oil and gas reservoirs

Mercury is a well-known trace component of production fluids in oil and gas reservoirs worldwide. There are competing theories attempting to explain how mercury concentrates in oil and gas basins. One possibility is that it originates from accumulated mercury in the organisms that lead to the formation of natural gas and hydrocarbons (Lang et al., 2012; Wilhelm, 2001a). Others suggest that mercury migrates into the oil and gas reservoirs from source rocks and metal-rich fluids. This

migration process may be encouraged by secondary processes such as geological and geothermal activities (Abai et al., 2015; Yan et al., 2017). Several authors believe that regions with higher tectonic and geological activity are responsible for higher mercury content within natural gas reservoirs (Gallup, 2014; Lang et al., 2012; Yan et al., 2017) because of increased interactions with metal-rich fluids, mineral material, and formation waters (Filby, 1994).

Independent of the origin, the mercury content of oil and gas basins and its global distribution may be affected by geographical location, geology, age of the basins, as well as tectonic and seismic activity of the region (Ryzhov et al., 2003; Wilhelm and Bloom, 2000). The geographical variability of mercury in oil and gas basins is shown in Fig. 1. Mercury in gas ranges from as low as 0.01 $\mu\text{g}/\text{m}^3$ in South America to as high as 1930 $\mu\text{g}/\text{m}^3$ in China. Levels of mercury may also vary temporally, as shown by Ryzhov et al. (2003) who measured considerable variability in natural gas mercury concentrations ranging from 1.6 to 17 times over 20 years at the same location.

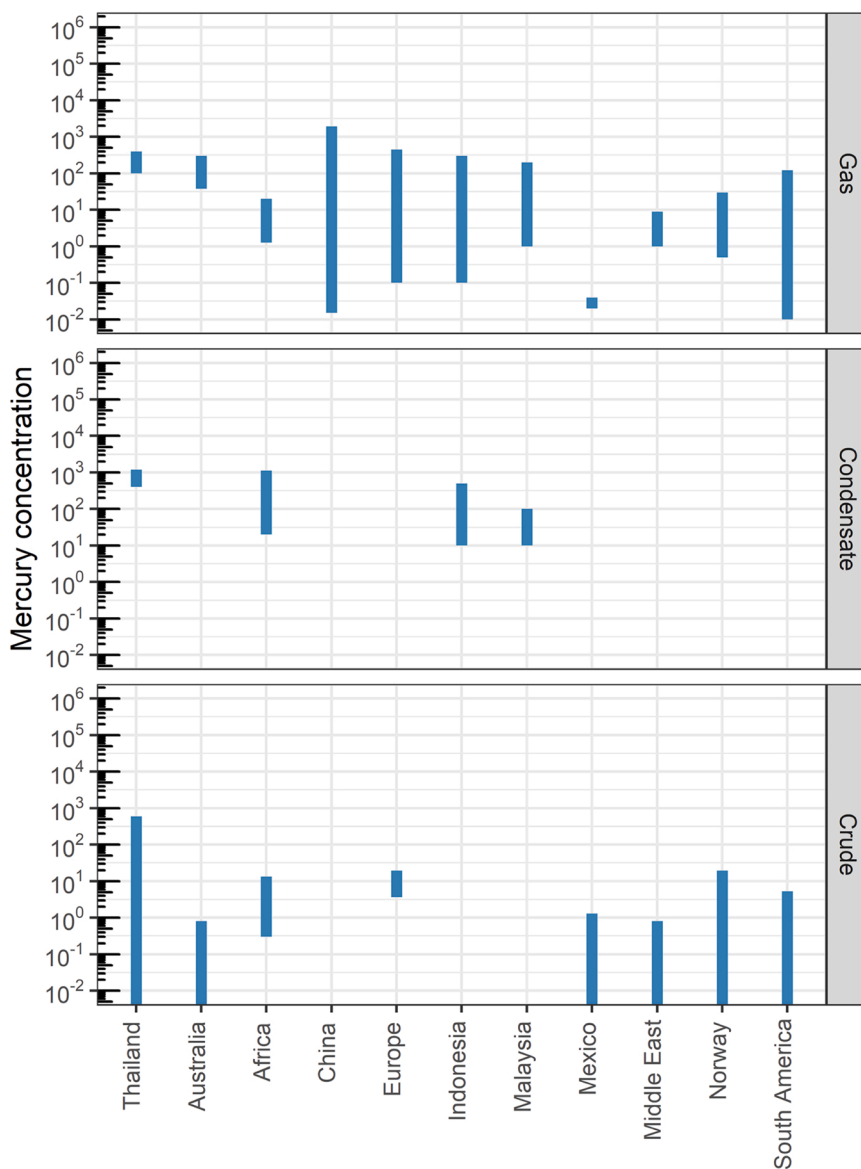


Fig. 1. Mercury concentration detected in oil and gas products at different geological regions. Units are $\mu\text{g}/\text{m}^3$ for gas and $\mu\text{g}/\text{kg}$ for condensate and crude products. Line segments represent concentration ranges summarised from various sources including Ezzeldin et al. (2016), Mussig and Rothmann (1997), Shafawi (1999), Wilhelm et al. (2007), Crafts and Williams (2020), and Lang et al. (2012).

3.2. Mercury speciation in oil and gas production systems

The behaviour of different mercury species in oil and gas processes are specific to their physical and chemical properties. These behavioural differences lead to variations in their partitioning in oil and gas phases or products in the production system (Wilhelm, 1999a). Different solubilities lead to preferential partitioning in the production fluids while different volatilities may lead to separation due to temperature and pressure changes within the extraction and processing system.

The speciation of mercury (and therefore its propensity to partition into different fluid phases or to different infrastructure) may also change within the oil and gas production systems. The oxidation of Hg^0 in crude oil leads to the formation of water-soluble ionic mercury compounds and complexes. Hg^0 and ionic mercury species are known to be very reactive with sulphur, forming the generally insoluble HgS species (Wilhelm, 2001b).

Production fluids contain several co-existing mercury species due to the multiphase nature of the fluids, which are transported within the production tubing, subsea equipment, and flowlines/risers prior to the upstream processing facility. Mercury speciation that has been reported within the different phases have been reviewed and summarised in Table 1, these phases will now be discussed in more detail.

3.2.1. Mercury speciation in the gas phase

Mercury occurs in gas phase in its elemental and organic form. Hg^0 is believed to be the dominant species in natural gas, making up over 50 %

Table 1

Mercury species detected in different phases of oil and gas production fluids. R represents alkyl groups such as CH_3 and C_2H_5 . X represents inorganic anions such as chloride and other halides.

Elemental Hg	Inorganic Hg	Organic Hg	Source
Gas phases			
Hg^0	–	$Hg(CH_3)_2$	(Wilhelm, 2001b)
Hg^0	Ionic Hg	–	(Nengkoda et al., 2009)
Hg^0	–	–	(Bingham, 1990)
Hg^0	–	–	(Schickling and Broekaert, 1995)
Hg^0	–	R-Hg-X and R-Hg-R	(Abbas et al., 2016)
Hg^0	–	–	(Mussig and Rothmann, 1997)
Hg^0	–	$Hg(CH_3)_2$	(Wilhelm and Bloom, 2000)
Water phases			
–	$HgCl_2$	–	(Lothongkum et al., 2011)
Hg^0	$HgCl_2$, Hg_2Cl_2 , HgS , HgO , $HgSR_2$	$Hg(CH_3)_2$, CH_3HgCl	(Gallup, 2014)
–	$HgCl_2$	$HgCH_3$	(Zettlitzer et al., 1997)
Hg^0	$HgCl_2$	–	(Tao et al., 1998)
Gas condensates and crude oil phases			
Hg^0	Ionic Hg (Hg^+ , Hg^{2+})	$HgCH_3$, $Hg(CH_3)_2$, HgC_2H_5 , $Hg(C_2H_5)_2$	(Frech et al., 1996)
Hg^0	$HgCl_2$	$Hg(CH_3)_2$, HgC_3H_8 , $Hg(C_2H_5)_2$	(Tao et al., 1998)
Hg^0	$HgCl_2$, Hg_2Cl_2 , HgS	$Hg(C_6H_5)_2$, CH_3HgCl , $HgCH_3$, $Hg(CH_3)_2$	(Schickling and Broekaert, 1995)
Hg^0	Hg^{2+}	–	(Bouyssiere et al., 2002)
Hg^0	Ionic Hg (Not specified)	$HgCH_3$	(Ezzeldin et al., 2016)
Hg^0	$HgCl_2$, Hg_2Cl_2	–	(Snell et al., 1996)
Volatile Hg	Ionic Hg (Hg^+ , Hg^{2+})	Non-ionic organic Hg (Not specified)	(Gaulier et al., 2015)
Hg^0	Hg^{2+} , HgS	$HgCH_3$, HgC_2H_5 , HgC_6H_5	(Zettlitzer et al., 1997)
Hg^0	Ionic Hg	R-Hg ionic, R-Hg-R	(Nengkoda et al., 2009)

of total mercury detected (Wilhelm, 2001b) due to its volatile nature. Gaseous Hg^0 can also partition from the gas phase into the liquid phase of multiphase well fluids (i.e., containing gas and liquids) based on its solubility limit. Once dissolved, Hg^0 is unstable and easily converted to other species through oxidation and reduction processes (Corns et al., 2020; Yan et al., 2017). In the presence of ions such as chlorides, sulphates, and other sulphur-bound compounds, oxidised Hg^0 may react to form more stable compounds such as $HgCl_2$ and other chloride species (Kozin and Hansen, 2013; Ma et al., 2014), and other types of organic mercury with R-Hg-X structures (Zettlitzer et al., 1997).

Ionic mercury is rarely detected in gas phases because of its low volatility and its high solubility in production fluids (Wilhelm, 2001b). Volatile organic mercury species such as dimethylmercury ($Hg(CH_3)_2$) and organometallic mercury (i.e., R-Hg-X where R represents alkyl group and X represents inorganic monovalent cation) have been reported to coexist in natural gas (Table 1) but only in low proportions (<1 %).

The speciation of mercury in gas phases may also be affected by basin geochemistry. Natural gas containing elevated amounts of hydrogen sulphide (H_2S) and carbon dioxide (CO_2) is commonly termed as 'sour' gas. When these two sour gases are present at low concentrations, the gas is termed as 'sweet' gas. Various authors have observed that in sour gas reservoirs, mercury is usually detected as its sulphur-bound species (HgS , HgS_x^+). On the other hand, mainly Hg^0 and several other species are usually detected in sweet gas reservoirs (Gallup, 2014).

3.2.2. Mercury speciation in the water phase

Mercury is present in both the produced water stream from the primary separation unit and the waste water discharge stream (Gallup, 2014; Lothongkum et al., 2011; Tao et al., 1998; Zettlitzer et al., 1997). Typical mercury concentrations detected in produced waters from several well-known gas and oil fields are between 0.001 and 0.3 mg/L (Ahmadun et al., 2009; Gallup and Strong, 2007; Zettlitzer et al., 1997). Mercury occurs in the water phase in all forms (Table 1), namely elemental, ionic, organic, and adsorbed to particulate matter (O'rear et al., 2016; Gustafsson et al., 2007). Inorganic species, such as $HgCl_2$, are very water soluble and have the highest abundance at 10–50 % of total mercury detected in the water phase (Wilhelm, 2001b). For example, of all published records of mercury speciation in the water phases of oil and gas production systems reviewed here, $HgCl_2$ was present in all except for the samples analysed by Zettlitzer et al. (1997) (Table 1). The results of Zettlitzer et al. (1997), however, are only based on two samples, one in which all mercury species were below the instruments' detection limit of 0.1 mg/L.

Not all inorganic species are soluble; HgS is insoluble in hydrocarbon and water phases under typical process conditions. The presence of sulphur and its compounds in gas streams has resulted in the formation of mercury complexes such as thiol complexes, $HgSR_2$ where R represents alkyl groups (Gallup, 2014).

3.2.3. Mercury speciation in gas condensate/crude oil

All mercury forms have been detected in gas condensates and crude oil samples (Table 1). Detected species include Hg^0 , methyl mercury $HgCH_3$, ethyl mercury HgC_2H_5 , dimethyl mercury $Hg(CH_3)_2$, diethyl mercury $Hg(C_2H_5)_2$, phenyl mercury HgC_6H_5 , diphenyl mercury $Hg(C_6H_5)_2$, $HgCl_2$, CH_3HgCl , Hg_2Cl_2 , and particulate mercury (as solid HgS).

Suspended insoluble HgS is found in both condensate and crude oil, making up ~60 % of the total mercury detected by Zettlitzer et al. (1997). Avellan et al. (2018) and Ruhland et al. (2019) reported that HgS was present as nanoparticles of various sizes (mostly < 100 nm but some larger) in condensate. Inorganic mercury, such as $HgCl_2$, is highly soluble in gas condensates and crude oil. Tao et al. (1998) detected the presence of $HgCl_2$, which represented 53–97 % of the total dissolved mercury in several condensate samples. Similarly, 58–85 % of the total mercury in gas condensates detected by Zettlitzer et al. (1997) was

found to be HgCl_2 .

The occurrence of other forms of organic mercury is still debated because of the challenges associated with sampling and preserving mercury species that are unstable outside oil and gas system conditions. For example, organic mercury tends to undergo species interconversion and photolysis to form Hg^0 , which might explain the presence of Hg^0 in condensate (Bouyssiere et al., 2000; Dessy and Lee, 1960; McAuliffe, 1977). Other organic species are possible. For example, Schickling and Broekaert (1995) have observed reactions between $\text{Hg}(\text{C}_6\text{H}_5)_2$ with HgCl_2 and CH_3HgCl to form other types of organic mercury.

The likelihood of organic mercury in condensate is higher due to the fact that it is more soluble in organic solutions compared to water (Haynes, 2017). Several authors have suggested that the partitioning of mercury into gas condensate is generally related to the boiling point of the gas condensate itself (Edmonds et al., 1996; Shafawi, 1999). As gas condensates are made up of several straight chain alkanes (C2-C6 +) with different boiling points, it is suspected that organic mercury will partition into those hydrocarbon phases as they condense from the gas phase.

3.3. Mercury accumulation in subsea pipelines

Mercury partitions from production fluids to oil and gas infrastructure during production due to its chemical behaviour at different points of separation and extraction process. This partitioning leads to the potential for mercury-species specific accumulation in some infrastructure, such as in pipelines carrying raw-well stream fluid.

The accumulation of mercury is also known to depend on the pipeline material and any internal coatings, the pipeline temperature (i.e., ambient external temperature), pressure conditions, and mercury speciation. Pipelines coated with an internal epoxy layer may prevent the build-up of corrosion products that otherwise are known to accumulate mercury (Collet and Chizat, 2015).

Several mechanisms may lead to mercury accumulation in production infrastructure:

- Deposition of Hg^0 to pipeline surfaces by condensation (coated, corrosion-resistant, and uncoated).
- Adsorption to steel surfaces (in uncoated steel pipelines).
- Adsorption into corrosion products (in uncoated steel pipelines).
- Accumulation in sludges such as asphaltenes.

Evidence of mercury accumulation within the oil and gas infrastructure is usually indicated by a delay in the detection of mercury in downstream oil and gas processing facilities. Wilhelm and Nelson (2010) reported that for an offshore subsea development connecting wells to an onshore processing plant via a subsea pipeline, operating for approximately 4 years, the mercury gas concentrations at the onshore facility measured less than $1 \mu\text{g Hg}/\text{m}^3$ compared to the $200\text{--}300 \mu\text{g Hg}/\text{m}^3$ at the wellhead. Based on the measured mercury concentration,

internal pipeline geometry and the average gas throughput at the plant, an estimated capacity of $1.5\text{--}2 \text{ g}/\text{m}^2$ of mercury was retained in the pipeline in the 4 years of operation. Mercury accumulation in pipeline systems is further evidenced by Zettlitzer and Kleinitz (1997), who reported total mercury concentrations of $1\text{--}80 \text{ mg}/\text{kg}$ of tubing and piping used for a natural gas production system. Fig. 2 summarises the mercury species associated with different aspects of the oil and gas installation compartments offshore. They will be discussed in turn.

3.3.1. Deposition and adsorption of mercury

Hg^0 cannot penetrate (absorb) into steel because of its large atomic size relative to the steel crystal lattice (Wilhelm and Nelson, 2010). Instead, Hg^0 is reported to adsorb to steel surfaces forming a monolayer (Linderoth and Morup, 1992; Roseborough et al., 2006b), or as adsorbed Hg^0 droplets, approximately $1\text{--}10 \mu\text{m}$ in size (Chaiyasit et al., 2009). The process of Hg^0 gas adsorption to pipe walls has been modelled by the firm Genesis (Crafts and Williams, 2020). They report that the process is reversible with a temperature dependency based on Fick's first law of diffusion. It has also been reported that mercury adsorption capacity on iron surfaces increases when the system's temperature is reduced (Roseborough et al., 2006a). Hg^0 may also condense on surfaces when experiencing changing pressures and temperatures. This may lead to the accumulation of Hg^0 depositions, particularly in low points of pipeline systems and areas of stagnation and changes in flow regime at pipe bends and changes in pipe diameter (Wilhelm, 1999a).

Where H_2S is present in the gas stream, the formation of particulate HgS may occur. This reaction is not favoured in the gas phase at process temperatures, so its reaction on surfaces or in solution following mercury oxidation is more likely (Wilhelm and Nelson, 2010). HgS may settle in pipelines if not carried by production fluids or adsorbed to steel, where it has been measured as a dominant mercury phase at concentrations of $0.05\text{--}0.1 \text{ g Hg}/\text{m}^2$ at a gas production unit (Wilhelm and Nelson, 2010).

3.3.2. Adsorption of mercury on corrosion products

Corrosion products are typically oxidised steel components and may include iron oxides (Fe_3O_4 , FeOOH) in carbon-steel pipes, oxides of chromium and nickel in corrosion resistance pipes, or iron sulphide (FeS , FeS_2) where H_2S is present. Additionally, the presence of CO_2 can also lead to the formation of iron carbonate (FeCO_3) within the corrosion layer (Popoola et al., 2013). These form a corrosion layer sometimes referred to as a scale on the internal surface of pipes.

Various parameters such as changes in temperature, pressure and flow regimes at pipe bends during production activities, presence of internal coating, pH, the chemistry of the production fluids, etc. have been reported to influence mercury species that are associated with corrosion and scale formation on the internal surface of pipelines (Yang et al., 2021). In uncoated pipelines, Hg^0 reacts with iron oxide and iron sulphide scales, which form a mercury-rich layer on the pipe walls (Wilhelm, 1999b). FeS is known to interact with Hg^{2+} via adsorption

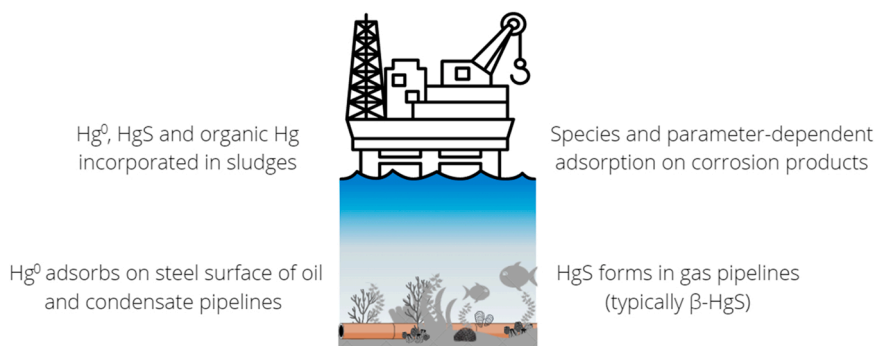


Fig. 2. Summary of mercury deposition and adsorption in different components of the offshore oil and gas installation.

and co-precipitation of HgS on its surface (Coulibaly et al., 2016; Rivera et al., 2019; Wolfenden et al., 2005) while FeS₂ mainly interacts with Hg²⁺ through surface adsorption forming a structured monolayer (Bower et al., 2008). Adsorption rates vary with changes in pH whereby low pH promotes the formation of monolayer Hg-Cl complexes on FeS₂ while mercury Hg-OH complexes mostly form at higher pH. Findings also suggest that mercury adsorption rate and capacity are enhanced at higher pH (Bower et al., 2008).

In gas production systems, HgS in the form of metacinnabar (β -HgS) is often reported. HgS was found to contribute up to ~25 wt% of the corrosion scale comprising FeOOH, FeCO₃ and Fe₃O₄ (Wilhelm and Nelson, 2010). Adsorbed Hg⁰ on the corrosion product likely reacts with gaseous H₂S to form HgS (Hall et al., 1991). Other mercury species such as HgCl, HgCl₂, Hg₂Cl₂, HgO₂, and HgO have also been reported within scale associated with gas transmission pipelines, but at much lower concentrations than Hg⁰ or HgS (Chanvanichskul et al., 2017).

The adsorption of mercury to corrosion products is superficial to the pipeline steel, as mercury cannot penetrate non-corroded steel. This has been confirmed by measuring mercury at different depths in the steel, which was found to be < 0.1 % Hg at 10 nm under the corrosion product layer (Chaayasit et al., 2009). Similarly, Chanvanichskul et al. (2017) reported no mercury inside the pipe base metal after removal of the corrosion product.

Wilhelm and Nelson (2010) also performed a characterisation study of a carbon steel pipe collected downstream of the primary separator in a gas production unit. They found that mercury concentration of < 0.2 g Hg/m² of pipe was both in the scale and on the surface of the metal as HgS and as Hg²⁺ incorporated into the iron oxide. The deposited HgS layer accounted for 0.05–0.1 g Hg/m² pipe. There was no accumulation of mercury at the interface between the scale and the metal.

The adsorption of mercury to corrosion products may be increased depending on the production fluid constituents and environment contributing to corrosion of carbon steel. For example, chloride ions and CO₂ are reported to accelerate steel corrosion creating a greater surface area for mercury adsorption (Pojtanabuntoeng et al., 2011). This type of mercury accumulation should not occur in pipelines that have internal coating to prevent corrosion (Bittrich et al., 2011). Having said that, welded field joints in subsea pipelines often do not have internal coating and so corrosion products may still occur and adsorb mercury over time.

3.3.3. Incorporation of mercury in sludges

Sludges are categorised as a heterogeneous mixture of waste oils and paraffin/wax, resin, asphaltene, inorganic minerals, and other depositions that are not in scales. Heavy hydrocarbons such as asphaltene and paraffin/wax are deposited on the pipeline due to surface adsorption or through precipitation and settling due to gravity.

Sludges are reported to have higher mercury content compared to the process fluid itself, with total mercury concentrations of 42.5–376.6 mg/kg reported (Camera et al., 2015). A range of mercury species may exist in sludges, including Hg⁰, inorganic (typically HgS) and organic species. Condensed droplets of Hg⁰ or insoluble HgS particulate matter can be physically incorporated into sludges in pipelines (Salvá and Gallup, 2010). Organic mercury species may be more soluble in the organic matrix and inorganic mercury may adsorb or complex to sludge components (Wilhelm, 1999b).

The type of sludge may also affect mercury accumulation. For example, a significant fraction (10–50 %) of the total mercury concentration within crude oils has also been reported to be associated with the asphaltene component of sludge, which was present in crude oil as both dissolved and suspended colloids (Wilhelm et al., 2006).

3.3.4. Expected forms and types of mercury contamination products

As outlined in Section 3.1, several mercury species co-exists and partition into different types of fluids within oil and gas systems. Therefore, depending on the operating conditions, location and fluid phases flowing through oil and gas pipelines, mercury species may be

detected in different contamination products such as sludges and scales.

Gas export pipelines tend to accumulate Hg⁰ from its deposition as liquid Hg⁰ and adsorption on steel surfaces as a monolayer. If these pipelines carry wet gas and/or produced water and corrode, scales of iron oxides and carbonates may be present. This may lead to a higher portion of inorganic mercury, such as mercury chloride and mercury oxide. Where H₂S is produced, adsorbed Hg⁰ or inorganic mercury may react to form HgS, typically as β -HgS.

Oil and condensate carrying pipelines may contain sludges or adsorbed Hg⁰, but typically at lower concentrations than in gas pipelines. Mercury accumulation in these pipelines is easier to clean through routine flushing and pigging operations, and so may be less of a concern than mercury products in gas-carrying pipelines.

It is currently unclear whether organic mercury can accumulate within offshore infrastructures due to sampling and measurement challenges (see Section 5.2). Having said that, organic mercury has been reported to exist within hydrocarbon phases such as gas condensates and crude oil, hence pipelines associated with those fluids may have the potential to accumulate organic mercury either incorporated into sludges and/or dissolved in residual hydrocarbon on steel surfaces.

Interactions between mercury and other types of pipeline materials are not well understood, such as corrosion-resistant alloy (CRA) and unbonded flexible pipes containing multiple layers of CRA and thermoplastics exposed to production fluids (Khalid et al., 2020). These may form different mercury contamination products that behave differently in the marine environment. Plastics such as polyethylene, polypropylene and high-density polyethylene have been known to readily adsorb mercury from solutions (Zhang et al., 2020). However, the mechanisms of mercury adsorption onto plastics in terms of capacity and rates of adsorption are not well defined. Turner and Holmes (2015) reported that the adsorption of mercury to microplastics in the aquatic environment involved interactions between metal cations, oxyanions, and organic complexes as a function of time, pH, and metal concentrations. How these interactions affect the release of mercury to the marine environment as the pipeline degrades needs to be better understood.

4. The ecological risk of mercury in the marine ecosystem

Residual mercury in subsea pipelines may cause negative impacts to surrounding marine ecosystems if left *in situ*. Ecological risk assessments are used to understand the likelihood and consequence of these impacts to receptors in the ecosystem. An ecological risk assessment typically considers the following steps: problem formulation, exposure characterisation, hazard characterisation, and risk characterisation (Suter and Norton, 2019). Problem formulation requires an understanding of mercury species, their behaviour in the marine environment, their concentration within the contaminated products, the types of ecosystem receptors that may be impacted, and the legislative and societal context defining the requirements for environmental protection. Exposure characterisation investigates the likely concentration or dose of mercury that will be experienced by ecosystem receptors. A hazard characterisation quantifies the potential magnitude of impact from such an exposure. The risk is then characterised by combining hazard and exposure information for a particular scenario and comparing it against the goals of environmental protection.

4.1. Exposure pathways

The exposure pathway of mercury from contaminated products described in Section 3.3 to ecosystem receptors will be controlled by its chemical speciation, which in turn is controlled by environmental conditions. Understanding contaminant speciation, partitioning, and transformation processes in the environment is a crucial aspect of an exposure characterisation. The behaviour of mercury in marine environments, with respect to its biogeochemical cycling, is generally well understood, with the presence of iron and sulphate reducing bacteria,

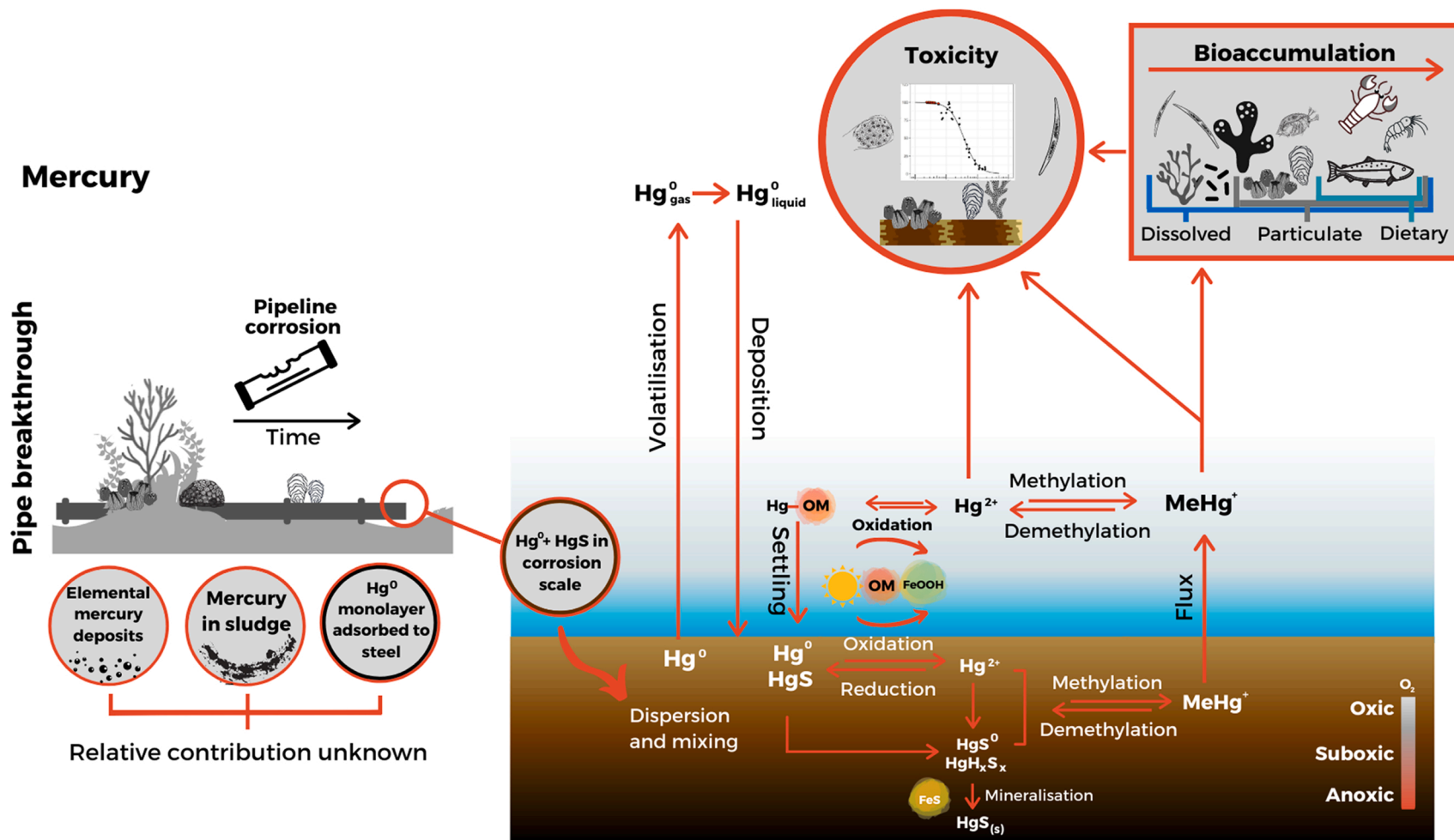


Fig. 3. Conceptual model of mercury (Hg) behaviour, environmental transformations, and exposure pathways in the marine environment Key environmental transformations such as oxidation, methylation, and mineralisation reactions are included but not intended to be exhaustive. OM refers to organic matter.

sediment organic matter, pH, temperature, and sediment redox potential known to affect mercury speciation (Gworek et al., 2016). However, the fate and likely behaviour of mercury-contaminated products from offshore oil and gas pipelines has not yet been considered.

Mercury-contaminated products occur in a variety of forms and species such as elemental or inorganic mercury species adsorbed to steel surfaces, adsorbed to corrosion products, or entrained in hydrocarbon sludges and asphaltenes (Section 3.3). A theoretical conceptual model of possible mercury environmental transformations and exposure pathways in the marine environment from contaminated offshore pipelines is shown in Fig. 3.

The following subsections discuss the key environmental conditions and/or processes that may contribute to the transformations of the identified mercury-contamination products when introduced to the marine ecosystem before and after pipeline breakthrough (i.e., seawater ingress to pipe due to corrosion). The general behaviour of mercury species and the important environmental processes including reduction, oxidation and methylation reactions are reviewed here to infer likely mercury exposure pathways.

4.1.1. Mercury contained in pipeline before breach

At the time a pipeline is decommissioned; mercury will likely be contained inside the pipe and would be inaccessible to ecosystem receptors (i.e., assuming the pipeline is capped). The rate at which pipeline degrades is dependent on the pipeline material, the presence of any external coatings, and the physical environment it is decommissioned in. As a result, estimates of when corrosive breakthrough may occur vary widely. One semiquantitative degradation assessment of an oil and gas asset in Australian waters suggested that steel pipelines will fully degrade between 1500 and 2000 years (Raitt et al., 2019). However, corrosive breakthrough leading to seawater intrusion at sites of localised corrosion may occur after 200 years (Raitt et al., 2019).

It is currently unclear whether there will be changes to the speciation of mercury from the point it is decommissioned to the corrosive breakthrough of the pipeline steel. Speciation changes will likely depend on the redox potential of the internal pipeline environment, which in turn will be determined by decisions such as pipeline flushing, the addition of biocides and corrosion inhibitors prior to decommissioning (Byron and Nichols, 2017; Manouchehri, 2017).

Decommissioned pipelines are normally filled with treated seawater, as empty pipelines may float or move on the seabed and cause a

navigation hazard. Seawater contains high concentrations of chloride (~480 mM) which promotes the formation of inorganic ionic mercury from Hg^0 and $HgS_{(s)}$ (Amyot et al., 2005). Over time Hg^0 and $HgS_{(s)}$ may leach to the dissolved phase. If anoxic conditions are formed, sulphate present within seawater may be reduced to sulphide which can either increase the solubility of Hg^0 through the formation of HgS_x species at lower concentrations or lead to the mineralisation of mercury as insoluble HgS (Morel et al., 1998). As the reduction of sulphate to sulphide is microbially mediated, this may be prohibited by the addition of biocides in seawater used to fill the pipeline. Insoluble FeS may also reduce ionic mercury to Hg^0 or promote the formation of $HgS_{(s)}$ (Coulibaly et al., 2016).

4.1.2. Environmental parameters controlling mercury bioavailability and methylation in the marine environment

When the pipeline is breached and seawater ingress occurs, any dissolved mercury will be flushed and less soluble mercury species, such as Hg^0 and $HgS_{(s)}$, will disperse or mix into the surrounding sediments, mostly under the pipeline's footprint. The ecological risk of this mercury will depend on its initial inventory, transformation to bioavailable inorganic mercury species, and subsequent methylation. Methylmercury poses the greatest risk to marine ecosystems because of its greater solubility and bioavailability (Harding et al., 2018a). As methylation is a microbially-mediated process, factors influencing the bioavailability of mercury to methylating microorganisms (such as the oxidation of insoluble mercury species) (Ma et al., 2019), or environmental conditions that promote the activity of the methylating microorganisms (Ndungu et al., 2016) will be important considerations for risk (Fig. 3). The variability of these factors is responsible for high variability of mercury methylation rates reported in Fig. 4.

Methylation typically occurs in the upper layers of sediments under weakly reducing conditions (i.e., reduced oxygen). It may also occur in the atmosphere or water column when mercury is associated with particulate organic matter. Anaerobic bacteria are largely responsible for mercury methylation and may include sulphate-reducing, iron-reducing, and methanogenic bacteria (Ma et al., 2019; Slowey and Brown, 2007). Anaerobic bacteria are usually found in anoxic sediments where they use oxidised sulphate, iron, manganese, or nitrate as electron acceptors for biochemical respiration reactions. However, due to the higher concentration of sulphate in seawater relative to iron and manganese oxides or nitrate, sulphate respiration is a dominant process in

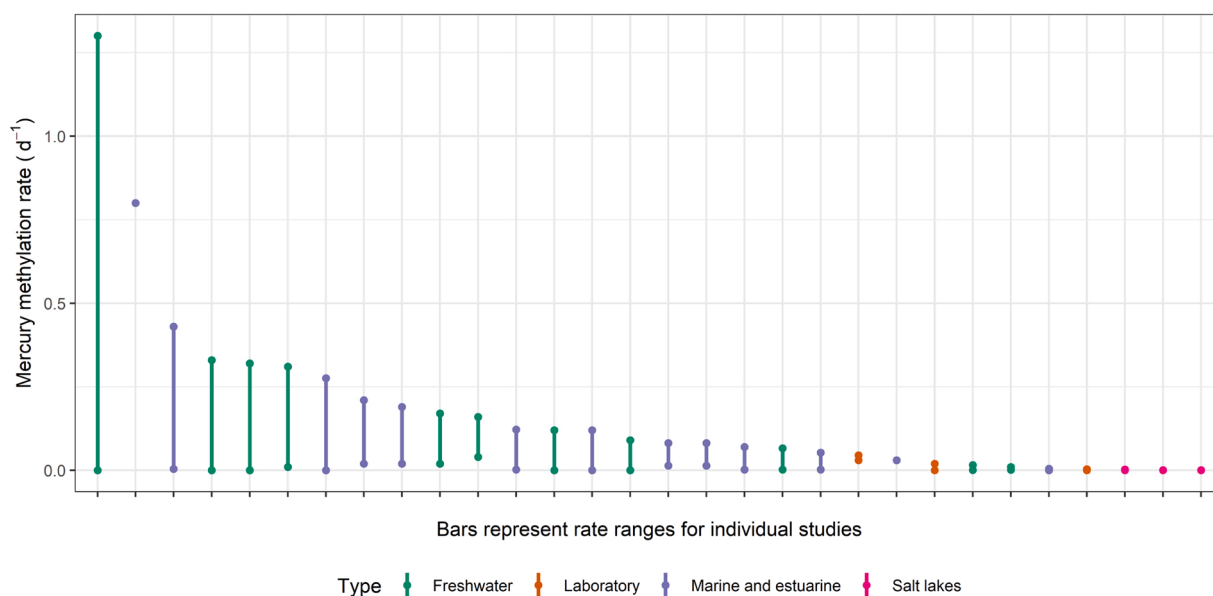


Fig. 4. Mercury methylation rates found in different environments reported by Paranajpe et al. (2017) and references therein. Individual bars and points represent the values for individual studies (maximum and minimum reported values).

suboxic and anoxic conditions.

Mercury-methylating microorganisms can only accumulate dissolved forms of mercury (Choi et al., 1994). Therefore, mercury present within the pipeline such as adsorbed Hg^0 and $\text{HgS}_{(s)}$ need to undergo reactions transforming them into bioavailable forms, such as dissolved neutral complexes or ionic species. Several environmental parameters have been identified as having a strong effect on methylation rates including pore water sulphide concentrations, pH, temperature, redox potential, and the mineralisation of mercury (i.e., its aging).

4.1.2.1. Pore water sulphide concentration and pH. Sulphide (typically as HS^-) has the strongest binding affinity to ionic mercury of any abundant ligand in the marine ecosystem (Fitzgerald et al., 2007). However, its role in determining mercury speciation is concentration dependent and may appear contradictory due to various soluble, insoluble, charged, and neutral species. At very low sulphide concentrations (such as those found in oxic seawater), soluble mercuric sulphide complexes form which increase the dissolved fraction of mercury. At moderate to high concentrations of sulphide, mercury sulphide forms insoluble $\text{HgS}_{(s)}$ or neutral dissolved HgS^0 complexes (Morse and Luther, 1999). At higher sulphide concentrations, the concentration of dissolved HgS^0 may decrease as polysulfide species, such as HgHS_2 and HgHS_2 , are formed (Benoit et al., 1999; Dyrssen and Wedborg, 1989), hence the neutral dissolved HgS^0 species is suggested to be the predominant bioavailable mercury species to methylating microorganisms in the marine environment.

The exact mechanisms behind these precipitation and dissolution reactions, or mechanisms behind mercury bioaccumulation by microorganisms, are not fully understood. These reactions are subject to multiple competing reactions and important knowledge gaps remain around the role of high-surface area nanoparticulate iron sulphide species, such as framboidal pyrite, mackinawite, and the role of organic matter (Tian et al., 2021). However, the presence of binding phases including the iron sulphide mineral mackinawite may promote mineralisation of mercury to insoluble $\text{HgS}_{(s)}$ forms (Johnson et al., 2015; Skyllberg et al., 2021).

Pore water pH has an influence on mercury speciation, with lower pH generally increasing the uptake of ionic mercury to methylating microorganisms that likely increases methylmercury production (Kelly et al., 2003). Pore water pH has also been shown to affect the ratio of methylated mercury species produced, with lower pH's increasing the proportion of mono-methylmercury compared to dimethylmercury (Bystrom, 2008), and reducing the amount of methylmercury that binds to sediment (Rudd, 1995).

4.1.2.2. Organic matter. Organic matter functions in multiple ways to promote and enhance mercury methylation through its capacity to stimulate microbial activity (i.e., as a food source) and as a complexing agent influencing mercury speciation and bioavailability (Chiasson-Gould et al., 2014). The influence of dissolved organic matter (DOM) on methylation is often confounded by sulphur geochemistry (Benoit et al., 1999). In environments where sulphate is not limited, methylation is primarily determined by the availability of organic matter. However, in environments with a sulphide concentration of over 0.01 μM , mercury sulphide complexes form preferentially, making binding with DOM less significant in determining methylation (Benoit et al., 1999).

While concentrations of DOM in marine systems are lower than in freshwater systems, the availability of organic carbon has been correlated to methylmercury distribution in marine surface and intermediate waters in the Arctic (Cossa et al., 2009; Kirk et al., 2012; Sunderland et al., 2009), north Pacific (Sunderland et al., 2009), and Mediterranean (Cossa et al., 2009), as well as in estuarine (Lambertsson and Nilsson, 2006) and marine (Mazrui et al., 2016) sediment.

4.1.2.3. Temperature. Temperature has a complicated relationship with

mercury methylation, but it has been shown that mercury methylation overall peaks during the summer months, which may correlate with the optimal temperature range for bacterial activity (Bystrom, 2008; Castro et al., 2000). Furthermore, it has been reported that ionic mercury has a tendency to partition to pore water with increasing temperature, which in turn affects its availability for methylation (Boszke et al., 2003). In contrast, the greatest mercury biomagnification occurs in high latitude environments where temperatures are often much colder (Lavoie et al., 2013). The net effect of temperature on methylmercury production is, however, less than other environmental parameters such as the influence of organic matter (Buckman et al., 2019).

4.1.2.4. Redox potential. Anoxic sediments have been found to correlate to increased mercury methylation rates. This is likely because anoxic sediments promote the formation of bioavailable mercury species and because they are characteristic of methylating microorganisms, such as sulphate reducing bacteria. For example, in oxygen minima zones, mercury methylation rates increased with dissolved oxygen concentrations up to 80 μM in the Arabian Sea (Chakraborty et al., 2016), whilst others measured the highest methylmercury concentration in Arctic waters around 290 μM dissolved oxygen (Heimbürger et al., 2015).

Redox potential may also be affected by other environmental parameters. Finer sediment texture (i.e., higher silt/clay fractions) can reduce sediment porosity and the penetration of overlying water leading to anoxic/suboxic conditions at shallower depths in sediments. Climate change is leading to reduced oxygen in coastal and ocean waters which is expanding anoxic zones above sediments (Breitburg et al., 2018). This is because higher temperatures reduce the amount of oxygen that can be dissolved in seawater and increases metabolic rates of microorganisms consuming oxygen for respiration. The influence of these global shifts may be important as mercury release will only occur following pipeline corrosion, which may take hundreds of years.

4.1.2.5. Morphology and age of pipe scale. Background mercury species are methylated in the environment at much lower rates compared to more recent inputs, indicating an aging process of mercury species which reduces its ability to be methylated (Chen et al., 2017; Gustafsson et al., 2007). HgS is an important mercury species in this process as the initial sulfidation of mercury may form bioavailable HgS^0 species but its ongoing mineralisation, such as to $\beta\text{-HgS}_{(s)}$, reduces its bioavailability and therefore methylation. For example, Jonsson et al. (2012) reported that $\beta\text{-HgS}$ in anoxic estuarine sediments had a methylation rate constant of $\sim 0.001 \text{ d}^{-1}$, up to two orders of magnitude lower than other geochemically relevant mercury compounds.

$\beta\text{-HgS}$ is the major species found within contaminated pipelines and has a lower tendency to be methylated in comparison to the other species such as cinnabar ($\alpha\text{-HgS}$) and FeS-adsorbed mercury (Jonsson et al., 2012; Sumner et al., 2020). The size of $\beta\text{-HgS}$ particles has been shown to affect its ability to be methylated, with increasing sizes having lower methylation rates (Zhang et al., 2019, 2012). This may relate to the changing surface structure of the mineral (Tian et al., 2021).

The aging process of HgS may occur quickly, compared to timescales of pipeline corrosion. Zhang et al. (2012) found that particulate HgS methylation decreased by 1.6–4.9x at an 'age' of 21 days. The ecological risks from mercury methylation should also consider the wall thickness and material, as it will influence the corrosion timeline of > 200 years.

4.1.3. Mercury released from decommissioned oil and gas pipelines

Mercury contained in pipelines will not pose a risk to the marine ecosystem until it is released. Dissolved-phase mercury will readily disperse while solid-phase mercury will deposit to sediments in oxic or anoxic conditions. Changes to mercury speciation and the propensity for its methylation will be highly dependent on site-specific factors (as described above in Section 4.1.2 and Fig. 3). Environmental parameters (pore water sulphide concentration and pH, organic matter,

temperature, redox potential) will vary site to site. The morphology and age of pipe scale will be different depending on the local reservoir, production life and method of pipeline cleaning and preservation for decommissioning. This makes it difficult to draw general conclusions about individual environmental parameters that may be used to predict risk in the future.

The redox potential of sediments is a consistent indicator for the potential for methylation to occur. Indications of sediment conditions from suboxic to anoxic can include a reduction in potential < 0 mV, dissolved oxygen concentrations in overlying water < 2 mg/L, and the presence of HS^- . In sub-/anoxic conditions, the microorganisms are most active, increasing the likelihood of methylation (Benoit et al., 1999; Harding et al., 2018a; Ma et al., 2019). Oxidic sediments aid the formation of chloride or organic ligand mercury complexes (Lamborg et al., 2004), and reduced the activity/prevalence of microbes (Ma et al., 2019). In highly sulfidic anoxic sediments, the bioavailability of mercury and the activity of the bacteria decrease (Benoit et al., 1999). The speciation of mercury will affect its bioavailability and thus its methylation potential (Zhang et al., 2012). Analytical methods to detect mercury speciation are further discussed in Section 5.2.

Some environmental parameters have multiple effects to mercury methylation (Ma et al., 2019). Sediment organic matter affects the activity of local microbial communities, in turn affecting the reducing conditions and sulphide production. Sediment organic matter also provides strong adsorbent sites for mercury. This may facilitate more bioavailable mercury as it is generally correlated to increased methylmercury production (Taylor et al., 2019).

The current understanding of mercury speciation in oil and gas pipelines comes from samples collected from coupons or pig dust and may not reflect the likely speciation of mercury when released to the environment following corrosive breakthrough of a pipeline in 10–100 s of years after it is decommissioned. This is particularly important given the recalcitrance of $\text{HgS}_{(s)}$ compared to Hg^0 or recently precipitated HgS^0 and other sulphide species.

Other questions still remain around the role of adsorbents such as nanoparticulate iron sulphides (Rivera et al., 2019), the uptake mechanisms of mercury from sediments, and the diffusion of methylmercury from sediments to the overlying water. Additionally, factors that lead to

demethylation in an environment may mitigate some risk of methylmercury (Paranjape et al., 2017), but are generally less understood. Finally, the current understanding of mercury methylation and risk in marine environments is largely based on studies of estuarine conditions which are very different to coastal and open ocean environment conditions.

4.2. Potential Hazards of Mercury to Marine Ecosystems and Humans

Mercury-contaminated products within oil and gas pipelines could give rise to several hazards to marine organisms when released to marine ecosystems. Mercury is known to be toxic and is persistent in the environment. The environmental impact of mercury is considered in the subsequent subsections including its bioaccumulation and biomagnification in the food web and toxicity to marine organisms. Fig. 5 summarises the hazards of mercury to marine ecosystems and human health, which are discussed in detail below.

4.2.1. Bioaccumulation and biomagnification

Methylmercury is the mercury species of greatest concern as it readily bioaccumulates to tissue (Harding et al., 2018a). Marine organisms are exposed to mercury throughout their life from both natural and anthropogenic mercury sources. Direct absorption from seawater and accumulation from dietary sources are the main exposure routes (Atwell et al., 1998; Campbell et al., 2005). Methylmercury is efficiently transferred from prey to predator via a dietary exposure and so biomagnifies up the food web (Bowles et al., 2001; Gray, 2002; Scheuhammer, 2012). A recent meta-analysis found that the bioconcentration of methylmercury from water to plankton resulted in the greatest single step increase in mercury concentration in the food web, a 10^5 increase in mercury concentration from seawater to phytoplankton (Booth et al., 2020; Streit, 1998). Subsequent increases of approximately 10x per trophic level reflect the dietary-based biomagnification in the food web, Fig. 6 (Wu et al., 2019). As a result, fish methylmercury concentrations are 10,000 to 100,000 + times higher than in the surrounding water.

Trophic position and dietary preference are important determinants of mercury accumulation. An example of the latter is provided by Gilmour et al. (2019) who investigated mercury concentrations in blood

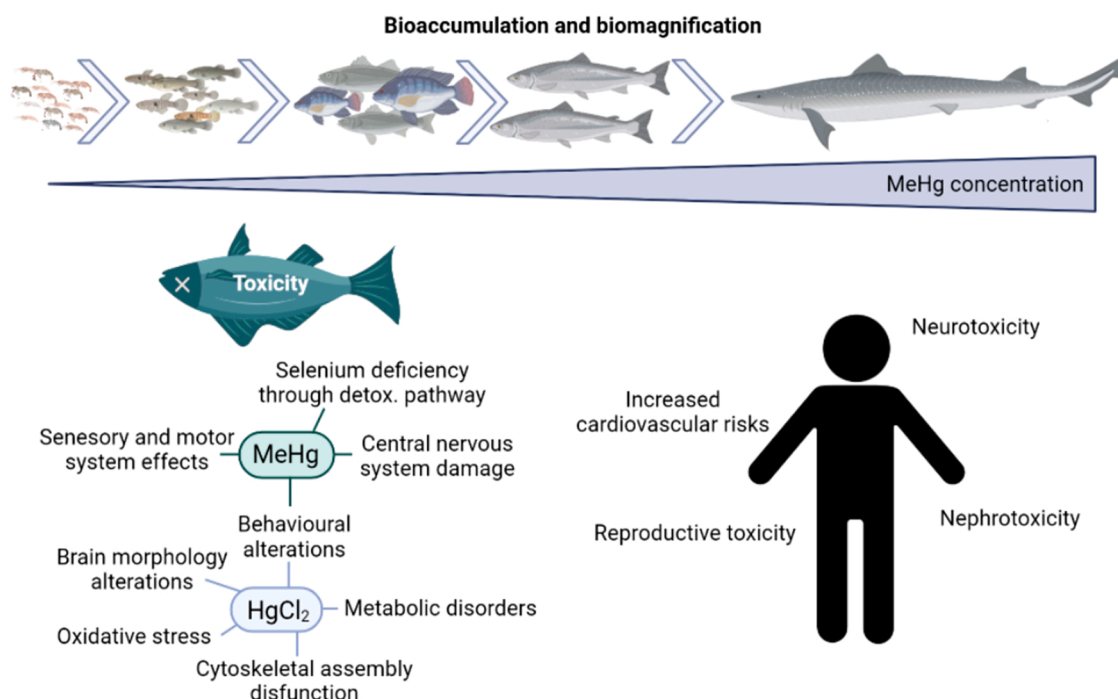


Fig. 5. Summary of the potential hazards of mercury released into the marine environment for ecosystems and humans.

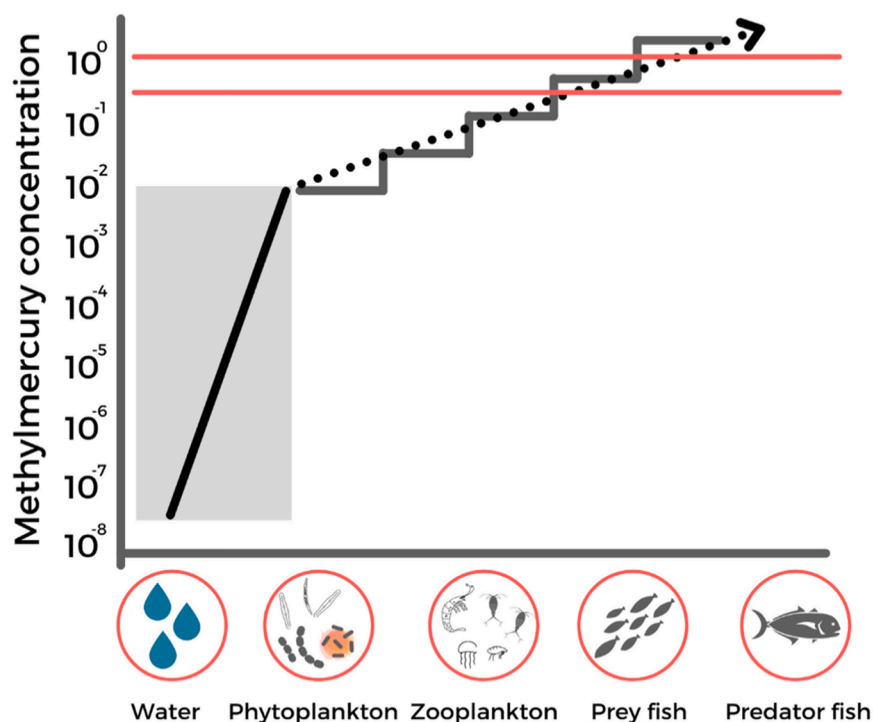


Fig. 6. Conceptual model of methylmercury bioaccumulation showing that bioconcentration to phytoplankton (inclusive of particulate organic matter) accounts for the greatest single biomagnification step (Wu et al., 2019). Transfer up trophic levels occurs in a stepwise manner is summarised by a predictive equation represented as the dotted black line. The two red lines represent the Food Standards Australia and New Zealand seafood safety mercury guidelines (FSANZ (2017)). Australian and New Zealand food standard for fish known to accumulate mercury (1 mg/kg) and all other seafood (0.5 mg/kg).

samples from two seabird species that exhibited different foraging strategies in Western Australia: the great-winged petrel, *Pterodroma macroptera*, which feed on pelagic squid, and the flesh-footed shearwater, *Ardenna carneipes*, that forage fish close to the coast. Mercury was found to be six times higher in great-winged petrels ($3.36 \pm 0.18 \mu\text{g/g ww}$) than flesh-footed shearwaters ($0.55 \pm 0.11 \mu\text{g/g ww}$).

Once methylmercury is absorbed by organisms, it is not easily eliminated (Harding et al., 2018a). This coupled to its ability to transfer from prey to predator enables mercury's biomagnification. Large and long-lived organisms occupying predatory trophic positions tend to have the highest mercury concentrations (Driscoll et al., 2013; Gray, 2002). This poses a unique challenge for mercury released from offshore oil and gas pipelines as the spatial range of impact may extend well beyond the footprint of the pipeline. For example, Australian fur seals travelled 65–175 km to forage along subsea oil and gas infrastructure (Arnould et al., 2015).

One approach to estimating mercury biomagnification in food webs is using an ecosystem model, such as 'EcoTracer' in 'Ecopath with Ecosim' (EwE). EwE combines static mass balance analysis ('Ecopath'), with temporal dynamics ('Ecosim'), and spatiotemporal scales ('Ecospace') (Christensen and Walters, 2004). These types of models require parameters describing the direct uptake rate of mercury from the environment to different species representing functional groups in the food web, their diet transfer between organisms, and their excretion (Booth et al., 2020; Walters and Christensen, 2018). These parameters are species specific and location specific (Streit, 1998), possibly reflecting different dietary preferences, food web structures, and the influence of different environments on contaminant bioavailability (Rajeshkumar and Li, 2018).

The application of EwE has been reported by Booth and Zeller (2005) who modelled the effect of climate change on the bioaccumulation of mercury in the food web of the Faroe Islands. They modelled 21 functional groups of the Faroe Island food web for 100 years to match baseline concentrations of methylmercury that were comparable to field observations. Simulations were then run to evaluate how seawater temperature rises would affect the biomagnification of mercury. They found that increased seawater temperatures would lead to an increase in

methylmercury concentration in the food web by 1.7 % and 4.4 % and therefore increase the dietary mercury exposure of the local population. To our knowledge, food web modelling has not been applied to the context of residual mercury in subsea oil and gas pipelines.

4.2.2. Toxicity

All forms of mercury may cause toxicity to marine organisms and mercury is one of the most toxic metals in the environment. The magnitude of a toxic effect to an organism depends on the mercury speciation, concentration, and exposure duration (Wiener, 2013). The mechanism of mercury toxicity is through neurotoxic effects. Observations of marine medaka (*Oriyza melastigma*) exposed to HgCl_2 indicated that inorganic mercury may cause neurotoxicity by inducing oxidative stress, cytoskeletal assembly dysfunction, and metabolic disorders (Wang et al., 2015). Mercury may also induce morphological changes in the brain including the number and volume of neurons and glial cells, which in turn may induce changes in swimming behaviour and long-term neurological effects (Wang et al., 2015). Methylmercury is able to cross the blood-brain barrier (Evans et al., 2016) and accumulate in the brain, leading to central nervous system damage (Roos et al., 2010). This can have adverse effects on organism sensory and motor system, as well as their behaviour (Das et al., 2003).

Toxic effects of mercury to marine organisms are life-stage dependent with embryonic and larval stages more sensitive to mercury than adult stages. For example, toxicity tests on red sea bream, *Pagrus major* suggested mercury (as inorganic ionic mercury) concentrations exceeding $20 \mu\text{g/L}$ can reduce hatching success, increase mortality, and induce teratogenicity (a disruption of development) in both embryo and larvae (Huang et al., 2011). Other impacts resulting from several different species of mercury (i.e. methylmercury, HgCl_2 and HgS) exposure caused malformations to the spine, bladder, head, and fins with these deformities leading to reduced chances of survival of medaka (*O. latipes*) embryos (Dong et al., 2016).

Mercury may be intracellularly detoxified by the formation of complexes with selenium (Ralston et al., 2012). This is commonly observed in marine mammals such as dolphins and whales (Caurant et al., 1996; Wagemann et al., 1998). However, although binding to selenium

Table 2

No observed effect concentrations (NOEC, the maximum concentration where no toxicity is observed) from chronic exposure of mercury for different taxonomic groups aggregated at the phylum level (ANZECC/ARMCANZ, 2000). *The source has only specified NOEC values for 7 species of algae with no indication of micro or macro algae species.

Taxonomic Group	NOEC ($\mu\text{g/L}$)
Fish	7.4–160
Crustaceans	0.8–10
Echinoderms	4
Molluscs	0.12–10.14
Annelids	3.4–18
Algae*	0.9–88

detoxifies methylmercury, Hg-Se induces oxidative damage and can lead to neurotoxic effects (Ralston et al., 2012; Ralston and Raymond, 2010).

The toxicity of mercury can be measured by ecotoxicological tests. These are usually carried out in a laboratory setting to control or investigate environmental parameters that may modify toxicity. These tests expose organisms to increasing concentrations of a known mercury species and measure the physiological response (Depew et al., 2012). Examples of the mercury concentration where no toxic effects are observed (NOEC) to an organism are summarised for individual species at the phylum level and are given in Table 2.

In oil and gas pipelines, mercury may not be the only residual contaminant. Mercury is often found alongside radioactive lead, ^{210}Pb , which is a decay product of radon, ^{222}Rn . Radon is extracted with natural gas in oil and gas systems but due to its short half-life of 3.8 days will often decay into a number of unstable radioisotopes before depositing as ^{210}Pb (Koppel et al., 2022). Other contaminants may include residual hydrocarbons, metals such as nickel and chrome from corroding steel, or the components of plastic coatings. In such circumstances the toxicity of all contaminants should be considered jointly (Cedergreen et al., 2008).

In humans, mercury can have long-term health implications including neurological damage, reproductive effects, and increased cardiovascular risks. Exposure to all forms of mercury may lead to nephrotoxicity as it is primarily accumulated in the kidneys (Zalups, 2000). HgCl_2 is the most nephrotoxic form of mercury and exposure to a moderate or high dose can lead to acute cellular necrosis (Bridges and Zalups, 2017). Humans are at risk from dietary exposure to mercury in seafood, especially in the form of methylmercury. Typical symptoms of methylmercury poisoning in humans (known as Minamata disease) include sensory disturbances, ataxia, dysarthria, constriction of the visual field, auditory disturbances, and tremors were also documented. Foetuses may also be poisoned by methylmercury when mothers ingest contaminated marine organisms (Sakamoto et al., 2018).

5. Other considerations for ecological risk assessment

5.1. Mitigation options

A range of mitigation strategies currently exists and are well adopted by oil and gas operators to reduce the risk of mercury in pipelines. Current mitigation strategies can be categorised into three main options: flushing, pigging and chemical cleaning. Understanding the types of mercury-contaminated products that can be removed and the efficacy of these mitigation strategies are important considerations to environmental risk assessments and hence drive the need to determine and understand the contamination inventory.

Before decommissioning, pipelines are depressurised and hydrocarbons within the structure are removed in line with relevant regional regulations by flushing and pigging. Pipelines are thoroughly cleaned whether being removed from the seabed or left *in situ*. In some cases,

pigging cannot be carried out for pipelines with a small diameter or of unfavourable infrastructure. Typically, the pipeline is flushed with treated seawater to reduce its hydrocarbon content to the same level or lower than seawater concentration. Flushing activities will help remove mercury species that are associated with fairly mobile contaminants within the pipeline such as liquid hydrocarbons, loose corrosion products or sludges. It is not clear the extent to which flushing and pigging can remove Hg^0 adsorbed to the steel pipeline wall, rigid corrosion products containing mercury, or mineral scales containing HgS .

Larger diameter pipelines are typically cleaned during routine pigging campaigns. A recent pigging development by Chanvanichskul et al. (2017) utilised specialised pigs for the *in situ* decontamination and sampling of mercury from pipelines. The pig design constitutes a train of one chemical and two mechanical pigs. The first pig is designed to be used with chemicals, while the second removes residual mercury mechanically by grinding and scraping the corrosion scale. These pigs also allow for simultaneous sampling of the deposited pipe scale containing mercury during the decontamination process collected for subsequent analysis in the lab. Multiple samples could be obtained during multiple runs of the pigs to verify the amount of mercury within the pipeline prior to decommissioning.

In cases where it is known that high levels of residual mercury exist, for example in the Gulf of Thailand, it is common to employ an additional chemical cleaning step using lixiviants to reduce any potential long-term risk of methylation that could arise from residual insoluble mercury. Lixiviants refer to liquid chemicals that can be used to scavenge any residual mercury from the pipe after flushing and pigging. The majority of these lixiviants are either acidic, strong oxidants and/or a complexing agent which removes mercury from the pipelines *via* oxidation to its more soluble forms. Acidic solutions can dissolve mercury alongside the iron oxide and iron sulphide but not HgS that are present within the pipe scale. In mercury-contaminated pipelines that contain primarily HgS , inorganic sulphide/polysulfide solutions are commonly employed due to their ability to dissolve both HgS and Hg^0 to yield soluble HgS_2H^+ and HgS_2^{2-} complexes. A 1 wt% sodium sulphide solution has been reported to dissolve HgS to yield a 1663 $\mu\text{g/L}$ mercury in solution (O'rear et al., 2016). The use of 0.2 M Iodine (I_2)/2 M potassium iodide (KI) has been reported to successfully remove adsorbed various mercury species such as Hg^0 , HgS and HgO from contaminated carbon steel (Chaiyasit et al., 2009) following oxidation and complex formation mechanism, forming soluble HgI_4^{2-} (Ebadian, 2001). Execution of chemical cleaning of mercury-contaminated pipelines can be achieved using a pig train containing the appropriate lixiviant at a given contact time to achieve a desired removal percentage (Baker et al., 2021).

The complexity and local environmental controls on mercury's bioavailability and toxicity across marine conditions means that mitigation options such as the use of mercury removal technology will be important components of any decommissioning plan. What is still missing, however, is an understanding of how low the mercury inventory needs to be to pose an acceptable risk to the marine environment.

5.2. Current measurement techniques for mercury in pipelines

The identification and quantification of mercury in oil and gas pipelines is an important step to understanding its risk from decommissioning activities through the potential exposure to the marine environment. It is also important from a verification perspective that contamination mitigation options are as effective as specified, providing the regulator assurance of the assets 'as is' condition for decommissioning. A summary of the techniques can be found in Fig. 7, and all methods will be discussed in turn.

The measurement and speciation of mercury contamination within a pipeline is a function of the sampling method, which can range from collecting pipe scale from pigging campaigns, extracting a removable

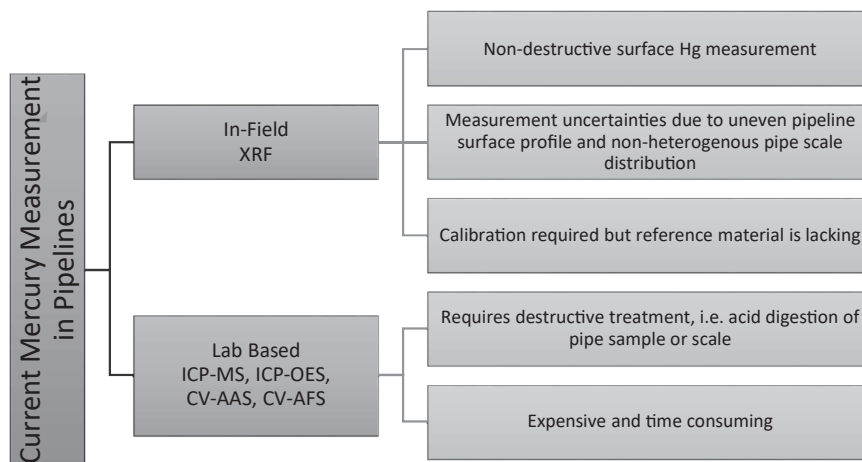


Fig. 7. Summary of current measurement techniques for mercury in pipelines.

coupon or pipeline spool, or cutting a sample of pipework. The testing methods can be classed as either destructive or non-destructive. Destructive testing entails an acid digestion step to extract mercury from either the collected pipe scale or the pipe section itself to its soluble form. The extracted mercury then follow conventional laboratory analysis methods of using either cold vapour atomic absorption spectrometry (CV-AAS); (US EPA, 2007a), cold vapour atomic fluorescence spectrometry (CV-AFS); US EPA (2007b), inductively coupled plasma-mass spectrometry (ICP-MS); (US EPA, 2014), or inductively coupled plasma-optical emission spectrometry (ICP-OES); (US EPA, 2018). Cold vapour techniques utilise reductants such as SnCl_2 (Clevenger et al., 1997) or NaBH_4 (Schickling and Broekaert, 1995) to reduce the divalent mercury species in the solution into the more volatile Hg^0 . The gaseous Hg^0 is then introduced into either the AAS or AFS cell for detection. Despite the existence of well-established laboratory techniques, current mercury measurement techniques for pipelines are expensive, time-consuming, and typically require retrieval of a physical sample. This limits the amount of sampling that can be done over the entire pipeline and may result in permanent mechanical damage to the pipeline.

Non-destructive testing involves the use of x-ray fluorescence (XRF); (Chanvanichskul et al., 2017; IPIECA, 2014) to measure the total amount of mercury on the surface of the sample. However, measurements using XRF are susceptible to measurement uncertainties and require proper calibration. This is difficult given the lack of reference standards for mercury scale on pipe steel. Most mercury will commonly exist in the surface scale layer therefore the dilution factor contribution from the bulk of the pipe needs to be taken into consideration to get a representative mercury concentration within a given pipe section. This is possible if the steel pipe thickness and the thickness of the scale layer are known. However, XRF's x-ray penetration depth depends on the density of the sample and varies with the depth and type of scale present on the pipe. The mercury scale thickness is usually reported as approximately 0.01–20 μm in steel (Chaiyasit et al., 2009; Chanvanichskul et al., 2017). However, uncertainties remain, with scale thickness to vary from several microns to several millimetres (Chanvanichskul et al., 2017). Furthermore, its distribution is unlikely to be homogenous due to the turbulent flow of the fluids during service. This means that there may be uncertainty converting the mercury content of the surface scale to a whole-steel mercury concentration.

The current research focus involves developing technologies that can be deployed *in situ* or in field. For example, SWASV (Square Wave Anodic Stripping Voltammetry) can quantify total mercury and is comparable to analysis using ICP-OES (Silakorn et al., 2019). The device is portable in comparison to those used in traditional labs; however, it still requires an acid digestion step as the analysis is on liquid samples. *In*

situ developments involve the application of sampling techniques to pig devices (Silakorn et al., 2018) or neutron activated gamma analyses (Vickery et al., 2019) which can measure internal mercury concentrations subsea from the outside of the pipe.

6. Knowledge gaps

Mercury is prevalent in oil and gas systems; however, little research has been undertaken to quantify the *in situ* inventories for subsea infrastructure nor understand how accumulation of mercury within decommissioned infrastructure will affect marine ecosystems. The following sub-sections highlight the knowledge gaps and recommend a series of research needs to better understand the risk of mercury from decommissioned oil and gas infrastructures to develop and conduct a well-informed ecological risk assessment.

6.1. Development of an ecological risk assessment framework

Ecological risk assessments of contaminants should consider all potential impacts to ecosystem receptors, direct and indirect. For mercury, this is complicated by its biogeochemistry, the extent to which local environmental conditions can affect rates of mercury methylation, and its ability to biomagnify in food webs (i.e., its hazards and exposure pathways in the marine environment). Based on its potential hazards, these impacts may include persistent sediment and water contamination, toxicity to local organisms leading to loss of ecosystem services and biodiversity, and impacts to indigenous, recreational, and commercial fishing communities because of mercury-contaminated marine resources. Additionally, decommissioning infrastructure containing contaminants means that the contaminant release and environmental exposures will occur in the future on timelines reflecting infrastructure corrosion rates (Raitt et al., 2019). This limits the potential for monitoring options and retrospective studies of impacts following release. These unique factors for ecological risk assessments for decommissioning need to be explicitly considered.

Environmental quality standards are important components of existing ecological risk assessment frameworks and are well adopted because of their simplicity and ease of application. These typically take the form of a concentration value for a contaminant in water or sediments, below which the risk(s) of negative impacts occurring is considered low. However, these are typically developed to be protective of direct toxicity to marine organisms and so may not be protective of other potential impacts. Site-specific investigations looking at multiple lines of evidence to understand contaminant risks, including the quantification of mercury partitioning and methylation rates in the receiving environment, the potential for bioaccumulation in local organisms, and

potential trophic transfer in food webs, would be the best practice approach to understand all risk and impacts. However, such studies are rare, possibly because of costs and uncertainty around how regulators will interpret such data. That is, it is easier to demonstrate regulatory compliance if there is a single numerical value to compare against.

The development of a single numerical guideline value that is protective of all potential impacts would require a significant research effort to ensure that it protects all environments (across the range of environmental conditions that can affect mercury speciation) and contaminant release scenarios. It would also require a level of conservatism that reflects a precautionary approach to ensure future generations do not inherit environmental harm from decommissioning decisions. The value of such a guideline is in its simplicity, ease of application, and the regulatory certainty it would provide. Exceedance of such a guideline would then prompt further mitigation (cleaning) activities or more detailed site-specific investigations. The development of an ecological risk assessment framework to support decommissioning decisions requires syndication of stakeholder views and alignment on the impacts and risks that need to be considered. Given the current understanding of mercury behaviour and impact in the marine environment this will require additional research, particularly in the areas discussed in the following subsections.

6.2. The inventory and *in situ* quantification of mercury in subsea infrastructure

The inventory of residual mercury left in pipelines, following *in situ* decommissioning, may be required to be reported under the Minamata Convention on Mercury (Article 9, 'Releases').

No published data exists that defines this inventory in subsea pipelines. This limits the ability to understand the underlying drivers of mercury formation and accumulation in oil and gas systems, which likely relate to basin reservoir fluid properties, operating parameters, pipeline materials, and internal coatings. This also limits the ability to determine *a priori* the cleaning and mitigation needs.

Inventories could be better established when *in situ* measurement technologies are fully developed and qualified for subsea deployment (i.e., at a technology readiness level for operational deployment). Current 'off-the-shelf' measurement technologies require direct access to the source of contaminant and a series of preparation steps. When subsea *in situ* mercury measurement technologies are verified "ready for deployment", they have the potential to efficiently, accurately and cost effectively detect and measure mercury species along the entire length of a pipeline, both spatially and temporality (i.e., pre and post contamination mitigation) to characterise the mercury inventory.

Research is needed to develop and validate new techniques to quantify the inventory of mercury in pipelines. This may be coupled to models to better understand mercury accumulation in oil and gas systems that can then be used to predict cleaning and mitigation needs in other infrastructure.

6.3. Environmental controls on mercury bioavailability and methylation

Key environmental transformations control whether sediments become sinks or sources of bioavailable mercury in the marine environment, including: (1) speciation changes of Hg^0 and insoluble HgS to bioavailable forms, the (2) methylation of mercury by endemic microorganisms, and the (3) availability of binding phases including sulphides, iron and manganese oxides, and organic matter that may control organic and inorganic mercury mobility. These reactions will depend on the initial mercury species, and while the likely species are Hg^0 and β - HgS , an understanding of how speciation changes between decommissioning and corrosive breakthrough is still missing.

Environmental transformations of mercury are controlled by highly localised environmental conditions and so the actual mercury risk must be considered on a site-specific basis. Predictive models of mercury

methylation potentials are generally not applicable across different environments for this reason. Research is required to understand under what conditions Hg^0 and $HgS_{(s)}$ react to form bioavailable mercury species. The constraints of these reactions, such as the sediment reduction potentials and pore water sulphide concentrations which promote methylation or mineralisation, should be quantified, and better understood. This will allow more robust predictions of mercury behaviour in a particular environment and will therefore reduce the uncertainty in the quantification of risks. This is required because mercury release from pipelines will only occur following corrosive breakthrough of the pipelines which may be 100–1000s of years in the future.

6.4. Mercury biomagnification

The methylation, bioaccumulation, and biomagnification of mercury means that impacts from decommissioned oil and gas pipelines may be observed on much wider spatial scales than the footprint of the infrastructure itself. Ideally, measures that link mercury biogeochemistry to biomagnification for a given environment and food web is required to predict future impacts from mercury release.

Biomagnification models, such as EwE may be used to explore potential impacts to food webs. However the influence of local species interactions and food webs, environmental conditions on the uptake of mercury from the environment, and excretion rates need to be considered to accurately portray accumulation scenarios (Christensen et al., 2005; Walters and Christensen, 2018).

6.5. Consideration of cumulative impacts and multiple stressors

Ecological risk assessments should consider the broader environmental context to get a holistic understanding of risk. This includes assessing the potential impact arising from multiple stressors as well as the effect of having cumulative impacts to an ecosystem. Multiple stressors can interact with one another and affect risk in unanticipated ways. For example, climate change impacts to environmental parameters, such as increase seawater temperatures, expansion of areas with low dissolved oxygen concentrations, and ocean acidification may affect mercury exposure and hazards in ways that will not be apparent when the risks of mercury are assessed using the present marine conditions.

Mercury may not be the only contaminant present in decommissioned oil and gas pipelines and the impact of multiple stressors to organisms may be unexpected. Multiple stressors may lead to ameliorative effects, such as observed between selenium and mercury to toxicity. Alternatively, they may lead to synergistic effects where the impact is greater than what would be expected from the sum of the individual contaminants.

Cumulative impacts may also arise where multiple or repeated local impacts affect a broader ecosystem. Current frameworks assess the ecological risk of contaminants on a case-by-case basis. Yet the cumulative impact of mercury release from multiple assets should also be considered, particularly because of mercury's propensity to biomagnify.

7. Conclusions

Regardless of the decommissioning methodology, the impact of contaminants, especially mercury, to the marine ecosystem from oil and gas infrastructure needs further research. This review identifies the species of mercury that can be found in subsea pipelines, likely exposure pathways, and methylation of mercury. Several key research gaps have been discussed, as well as the research needed to overcome these gaps. An improved understanding of the risk of mercury to the marine ecosystem will enable better decision making, support comparative assessments of disposal options, and promote societal acceptance of decommissioning decisions.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Co-authors TC and FG are undertaking consultancy work on mercury contamination for offshore pipelines.

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Environmental implication

Past research has shown that mercury associates with offshore oil and gas pipelines as well as other products associated with the infrastructure, deeming such materials "hazardous". However, the current environmental risk assessments for decommissioning activities of such contaminated materials does not take into account the complexity of the compound's nature and the potential harmful effects on e.g., marine food webs. This review paper has outlined these gaps in our current understanding, as well as providing advice on addressing these gaps to ensure that the marine environmental risk assessments reflect the hazardous nature of mercury-contaminated offshore infrastructure.

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