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Emissions of Volatile Organic Compounds from Crude Oil Processing – Global Emission Inventory and Environmental Release

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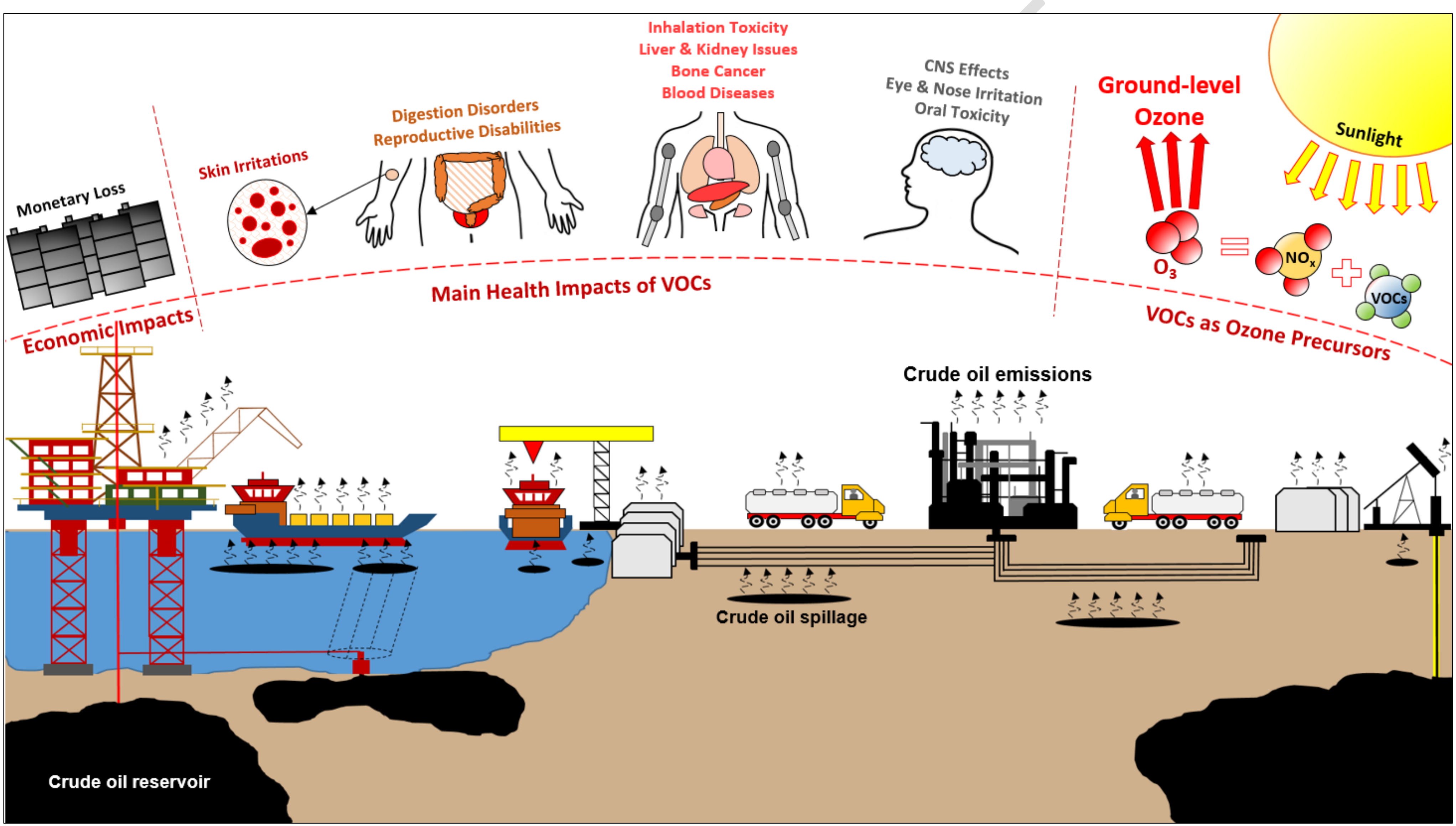
Graphical Abstract



Abstract

Airborne Volatile organic compounds (VOCs) are known to have strong and adverse impacts on human health and the environment by contributing to the formation of tropospheric ozone. VOCs can escape during various stages of crude oil processing, from extraction to refinery, hence the crude oil industry is recognised as one of the major sources of VOC release into the environment. In the last few decades, volatile emissions from crude oil have been investigated either directly by means of laboratory and field-based analyses, or indirectly via emission inventories (EIs) which have been used to develop regulatory and controlling measures in the petroleum industry. There is a vast amount of scattered data in the literature for both regional emissions from crude oil processing and scientific measurements of VOC releases. This paper aims to provide a critical analysis of the overall scale of global emissions of VOCs from all stages of oil processing based on data reported in the literature. The volatile compounds, identified via EIs of the crude oil industry or through direct emissions from oil mass, are collected and analysed to present a global-scale evaluation of type, average concentration and detection frequency of the most prevalent VOCs. We provide a critical analysis on the total averages of VOCs and key pieces of evidence which highlights the necessity of implementing control measures to regulate crude oil volatile emissions (CVEs) in primary steps of extraction-to-refinery pathways of crude oil processing. We have identified knowledge gaps in this field which are of importance to control the release of VOCs from crude oil, independent of oil type, location, operating conditions and metrological parameters.

Keywords: Crude oil; VOCs; Emission inventory; Health effects; Control measures.

1. Introduction

Crude oil, apart from being an ever-increasing demand of global industry (Herrera et al., 2018; Kilian and Murphy, 2014), is also recognised as one of the major origin of pollutants detected in terrestrial (Rajabi and Sharifipour, 2018), atmospheric (Afshar-Mohajer et al., 2018) and ocean ecosystem (Afshar-Mohajer et al., 2019). The world proven crude oil reserves have been estimated to be more than 1000 billion barrels (bb) in 1997 and 1498 bb at the end of 2018 (≈44% during the last 20 years) (OPEC, 2019) which demonstrates the global desire and need to recover more crude oil for energy, transport, material and manufacturing purposes. Crude oil exploitations also known as upstream operations comprise various steps of exploration, extraction, storage, transportation and refinement (Masnadi et al., 2018b) which can seriously affect human health and the environment by polluting the soil, water and air directly via spillage and indirectly through hazardous emissions and deforestation (Ramirez et al., 2017). The severity of hydrocarbon contamination released by crude oil is proportional to the scale of oil processing as a larger size of well-to-refinery operations would result in a higher possibility of crude oil contamination (Zhang et al., 2019b). Oil spillage usually occurs due to equipment malfunctions, human errors, and unavoidable hazardous emissions when oil mass is open-to-air without any control measures (Pelta et al., 2019). Human error is reported to be the main reason (at least 80%) of all accidental oil-tanker spills worldwide in 2018 of which about 70% happened in shipboard operations, hence the more crude oil is being shipped the more number of causalities and/or incidents is probable to take place (ITOPF, 2018). The adverse effects of crude oil pollution are not limited to the affected areas as some of the hydrocarbons (HCs) within the crude oil mass are classified as volatile substances. These substances have low boiling points which potentially cause free an escape from the oil into the surrounding atmosphere and create public health problems. These volatile substances are a subset of broader gaseous materials called Volatile Organic Compounds (VOCs) with well-known detrimental influences on our health and ecosystem (Dai et al., 2017).

VOCs are defined as organic chemicals (excluding CO, CO_2, H_2CO_3, $(NH_4)_2CO_3$, and carbonates) with high vapour pressures which can escape from solids or liquids in the form of gases at standard temperatures and pressures (Pellizzari et al., 1987). The VOCs originate from a wide variety of sources classified as natural (1150 Teragrams of Carbon per Year ($TgC.year^{-1}$)) and anthropogenic sources (142 $TgC.year^{-1}$) (Zhang et al., 2017b). Naturally-sourced VOCs are usually originated from the terrestrial and ocean biogenic reactions, while anthropogenic VOCs emitted from numerous man-made causes primarily due to the evaporation of organic solvents and fossil fuels combustion. Crude oil processing is considered to be capable of around 16% of the total VOC emissions into the atmosphere in the late 20th century (Masnadi et al., 2018a). Based on an airborne emission inventory reported in 2010, approximately 258 tonnes of HCs could have been evaporated per day from the Deepwater Horizon (DWH) Oil Spill in the Gulf of Mexico (Ryerson et al., 2011) which itself signifies the scale and importance of the problem.

Volatile compounds can escape from the oil mass during all steps of the crude oil industry from extraction sites and transportation facilities to storage tanks and refineries. Therefore, the production (exploration/extraction) sites and oil refineries have been recognised to be the second major origins of the VOCs after vehicle exhausts in the transportation sector (Khoramfar et al., 2018). As an example, the emission inventories in China (with sever and extensive air quality problem across the country), indicates that the petroleum industry (including crude oil processing (Bo et al., 2008; Wei et al., 2008)), road gasoline vehicles, biomass burning and paint coating are among top four sources of ambient air VOCs in the country (Wei et al., 2014b) in addtion to VOCs emitteid from major landfills in China (Wang et al., 2019; Xie et al., 2018).

From composition viewpoints, approximately 85% (weight) of the crude oil compounds consists of HCs (mostly with five or more carbon atoms (i.e. $\geq C_5$), with an average composition of alkanes (30%), cycloalkanes (49 %), aromatics (15 %) and asphaltics (6 %) (Simpson et al., 2010)). Light HCs in crude oil (e.g. alkanes and aromatic with ≤C15) can readily escape into the air due to their high vapour pressures and low boiling points to form the main subset of atmospheric VOCs. It is also noted that VOCs can cause major global-scale contributions to produce photochemical ozone (O_3) and other harmful oxidants which adversely affect the air quality and human health (Wei et al., 2014b). In addition to methane, some VOCs emitted from crude oil such as ethane, propane, butane, pentane, and hexane, can also interact with NOx in the air forming ground-level ozone named as the tropospheric ozone (Mustafa Salih et al., 2018). Hence, crude oil emissions are also considered as "ozone precursors" (Ras et al., 2009; Wei et al., 2014a) and "global warming agents" (Bolaji and Huan, 2013; Cetin et al., 2003).

In the last three decades, the Crude oil VOC Emissions (hereafter CVEs), have been investigated in literature either directly by means of laboratory and field-based analyses or indirectly via emission inventories (EIs) of the petroleum industry to regulate their contributions to the atmospheric pollutions (Table 1). However, only a few studies have focused on analysing the major mechanisms of CVEs and their influential parameters under different conditions such as water soluble fraction of HCs in crude oil contaminated soil (Saeed and Al-Mutairi, 2000; Saeed et al., 2013), ocean oil spillage (Afshar-Mohajer et al., 2018; Hanna and Drivas, 1993; Tonacci et al., 2015) and contaminated lands (Yang et al., 2007). The majority of these studies are associated with the fate of VOC emissions from petroleum industry in which VOCs emitted from various stages of crude oil processing including exploration, extraction, and production (Aklilu et al., 2018; DeLuchi, 1993; Helmig et al., 2014; Huang et al., 2018; Koss et al., 2017; Papailias and Mavroidis, 2018; Salih et al., 2018; Simpson et al., 2010; Villasenor et al., 2003; Wang et al., 2014; Warneke et al., 2014), storage

(DeLuchi, 1993; Jackson, 2006; Khoramfar et al., 2018; Paulauskiene et al., 2009; Theophanides et al., 2007), transportation (de Vos et al., 2007; DeLuchi, 1993; Howard and Nikolas, 2001; Karbasian et al., 2017; Lee and Chang, 2014; Lee et al., 2013; Martens et al., 2001; Milazzo et al., 2017; Paulauskiene et al., 2008; Sani and Mohanty, 2009; Sani and Mohanty, 2008; Tamaddoni et al., 2014), refineries (Baltrėnas et al., 2011; Cetin et al., 2003; Lin and Lee, 2006; Lin et al., 2004; Liu et al., 2008; Pandya et al., 2006; Ragothaman and Anderson, 2017; Sonibare et al., 2007; Wei et al., 2014b; Wei et al., 2016; Zargar et al., 2013; Zhang et al., 2017c; Zhang et al., 2018), offshore (Afshar-Mohajer et al., 2019; Afshar-Mohajer et al., 2018; Bahreini et al., 2012; Han et al., 2007; Hanna and Drivas, 1993; Müller and Sedláčková, 2003; Saeed and Al-Mutairi, 2000; Saeed et al., 2013; Schädle et al., 2014; Tonacci et al., 2015; Uhler et al., 2010; Yagi et al., 2004; Yuan et al., 2014) and onshore activities (Almudhhi, 2016; Lawlor et al., 1997; Moyer et al., 1994; Müller and Sedláčková, 2003; Mustafa Salih et al., 2018; Soukup et al., 2007; Wang et al., 2015a; Wang et al., 2015b; Yang et al., 2007) as well as accidental spillages. In addition to the source-specific properties of CVEs (i.e. crude oil characteristics (Saeed and Al-Mutairi, 2000)), metrological parameters such as wind speed/direction and temperature (Jackson, 2006), ground surface patterns at production sites and refineries (Huang et al., 2018; Salih et al., 2018) and even ocean currents (for ocean oil spillage (Afshar-Mohajer et al., 2018)) are critical factors which govern the CVE distributions and their fate in atmosphere.

Despite the significant importance of the VOC emission from crude oils, we have identified the lack of a comprehensive review of the state-of-the-art scientific findings related to key aspects of this topic. There is a large body of scattered data and research studies in the literature which included i) the scale of emissions from crude oil processing and ii) evaluations of the VOC emanations from crude oil. An essence and drive for preparation of the state-of-the-art review presented here are related to extensive need for developing feasible and sustainable solutions for remediation and/or containment strategies. The scale of the adverse impact of VOC emission on public health is significant in many areas and countries in the world. Niger Delta (Nigeria) is an example and one of the most affected places with regards to the scale and severity of land contamination by crude oil spillage. It has been reported that children born within 10 km of an oil spillage site in Niger Delta were twice as likely to die in their first month, which has been linked mainly to the exposure of pregnant women to the VOCs such as benzene and toluene (Elum et al., 2016).

This paper aims to provide a comprehensive and critical review of the published studies related to the CVEs. Critical information with regard to nature and origin of CVEs and summary of their background knowledge and scientific achievements are presented. The literature related to the VOCs emitted directly from crude oil are discussed, and the detected CVEs are quantitatively/qualitatively analysed. The published EIs, covering CVEs in petroleum sites as well as relevant controlling methods, are briefly reviewed.

2. Effects of VOCs on human health

The health effects of VOCs emitted from crude oil into the atmosphere have been well documented (Afshar-Mohajer et al., 2019; Brand et al., 2016; D'Andrea and Reddy, 2014; Jernelöv, 2010; Kwok et al., 2017; Merhi, 2010; Pappas et al., 2000; Pérez-Cadahía et al., 2006; Sebastián et al., 2001; Tomatis, 2000). The major detrimental effects of CVEs on human health are described in Fig. 1 by which we have demonstrated the health problems reported by certain VOCs such as BTEX (benzene, toluene, ethylbenzene and xylenes) and the widely detected VOCs near crude oil processing zones (Dehghani et al., 2018).

Benzene has been confirmed by the U.S. EPA Integrated Risk Information System (EPA IRIS) as a carcinogen gas (Group A), and an increment in lymphocyte count was also reported by the EPA IRIS via oral and inhalation of benzene due to its toxicity. In addition, benzene is recognised to be a major cause of leukaemia and other haematological cancers as well as blood-forming disorders (Talibov et al., 2018). All levels of exposure to benzene are considered to be a potential health risk (Smith, 2010). The human central nervous system can be affected by exposure to high concentrations of benzene, causing dizziness, nausea and headaches. Long exposure to benzene may also cause haematotoxicity, genotoxicity, chromosome aberrations, reproductive weakness and mortality (Edokpolo et al., 2015). Edokpolo et al. (2015) assessed the health risk of different scenarios of exposure to benzene emitted from petroleum refineries. The results indicated a cancer risk (CR) of 48,000 per 1 million of refinery workers (Edokpolo et al., 2015). The effects of exposure to benzene, released from petroleum refinery in Texas, on children’s haematological health were discovered by the signs of reductions in their white blood cell, haemoglobin and growth in platelet counts (D'Andrea and Reddy, 2014).

EPA IRIS has classified ethylbenzene (EB) with a potential of being a carcinogen (Group D) along with non-cancerous effects on liver and kidney, while the International Agency for Research on Cancer (a part of the World Health Organization) has confirmed the cancerous effects of EB. Short-term exposure to EB may cause eye and/or throat irritation, vertigo and dizziness. Long-term exposure to EB may result in more severe problems such as irreversible damages to the ear and kidney (Huff et al., 2010). On the other hand, toluene and xylenes have not been confirmed by the EPA IRIS as cancerous VOCs. However, some damages to human health have been identified by being exposed to toluene and all xylene isomers (Dhada et al., 2016). Immediate contact with toluene at high concentrations can cause temporary headache and dizziness, while permanent hearing/vision loss, retardation of mental capabilities, reduction in reproductive abilities and disorders in kidney/liver/immune system may be symptoms of repetitive exposures to toluene (Meek and Chan, 1994). Xylenes can also cause disruptions in the human nervous system from nausea/headache (in 100-200 ppm) to unconsciousness or even death (≥10000 ppm). Throat/nose irritation, accidental splash in eye, chest pain and shortness of breath, dryness and cracking in the skin have been reported as the results of 3-5 min exposure to 200 ppm of xylenes. Continuous exposure to a high level of xylenes may damage the liver and kidney (Kandyala et al., 2010). Health assessment of communities residing in the neighbourhood of petroleum refineries also confirms cough, wheeze, nausea, sinus/nose congestion, throat irritation, earache, skin rashes, nosebleed, headache, sleep problem, dizziness and stomach pain as common symptoms of inhalation of odours and emissions from crude oil-related industrial zones (Luginaah et al., 2002; Taylor et al., 1997).

Occupational exposure to CVEs can adversely affect those workers engaged in various steps of crude oil processing (Johnston, Lim et al. 2019). For example, a recent review on occupational exposure to BTEX in upstream petrochemical facilities in Italy showed that both the maximum values for long-term exposure and the ambient sampling were higher than the occupational exposure limits (OELs) for BTEX (Pedone, Lucaroni et al. 2019). Heibati et al. (2017) studied occupational exposure to BTEX in four main oil distribution companies in Iran. This study showed that those individuals responsible for tanker loading, tank-gauging, driving, firefighting and office works were the most exposed to CVEs among whom the highest levels of cancer and non-cancer health issues were detected for the tanker-loading workers (Heibati, Pollitt et al. 2017). On the other hand, the workers in the crude oil processing industry are mostly aware and informed of potential risks of being exposed to hazardous chemicals, and therefore various levels of systematic and personal protective strategies are usually used to protect this category of the work force. However, CVEs can silently damage the health of those communities living nearby major oilfields, petrochemical sites and oil spillage to an extent at which the CVEs have been named as “hidden Killer” (Hegarty 2017) to highlight the high rate of infant mortality and severe health disorders frequently reported near major oil-contaminated sites (e.g. Nigeria).

Although existing studies provide a good understanding of adverse health effects from VOCs exposure, more comprehensive and in-depth studies are required with greater statistical analysis and refined exposure assessments to accurately evaluate the CVE impacts on human health (mortality, illness, mental disorders, etc.) and to update occupational exposure (Johnston et al., 2019). Moreover, health surveillance of those communities living near major crude oil exploitation development should be carefully studied to better understand the relationship between oil extraction developments, regional air pollutions, and their adverse effect on human health (Finkel and Hays, 2016). Future studies are also required to cover gaps in the fundamental science of VOC monitoring technologies to achieve effective, low-cost and portable analytical instruments in order to elevate real-time monitoring process and help policy-makers to update/establish more efficient restrictive regulations in both regional and global scale (Collier-Oxandale et al., 2018).

3. Policies and regulations

Volatile organic compounds have various definitions and limitations provided by regional and/or international regulations to control indoor air quality (IQA) and outdoor air pollution (OAP). These standards aim to impose restrictive policies on specific industries (i) to reduce their VOC emissions, and (ii) to protect their VOC-exposed workers by effective personnel protective equipment (PPEs) (Wi, Kim et al. 2020). Crude oil exploitation industries have been classified by a number of regional regulations as one of the main sources of VOCs so far (Feldstein 1974, Zhang, Hao et al. 2011, Zhang, Wei et al. 2019). However, a comprehensive standard is still missing to control CVEs at a global scale and to pose restrictive policies on OPEC nations as well as industrial countries with high crude oil imports. As one of the recent regional standards for CVEs, the 40 CFR Part 60 entitled “Oil and Natural Gas Sector: Emission Standards for New, Reconstructed, and Modified Sources Reconsideration” proposed by the U.S. EPA provides federal-level regulatory measures for various sectors of U.S. crude oil and natural gas industry. The “Crude Petroleum Extraction” (NAICS code of 211120) and “Pipeline Distribution of Crude Oil” (NAICS code of 486110) were highly affected by 40 CFR Part 60 to regulate the amount of VOCs releasing form crude oil in the U.S. along with some other state-level standards of VOCs (e.g., section 01350 of California CDPH standard). The Directive 94/63/EC of European Parliament and Council was published on 20 Dec 1994 is another relevant example of CVEs regulation aiming to control VOCs emitted from petrol storage and distribution. Due to a significant amount of VOCs released in China, several restrictive policies have been proposed by Chinese authorities in which petroleum refinery industries (GB 31570-2015) have been restricted.

Although there are regional policies restricting CVEs in Northern America, China and European nations, a comprehensive international treaty does not exist that can limit global emissions of VOCs from crude oil processing worldwide since the current regulations lack extensive controls over specific regions such as the Middle East, Africa and Latin America with high proven reservoirs of crude oil. There are measures in crude oil loading and transit sectors to control VOC emissions including vapour recovery systems (VRS) (Bennett, 1993), vapour emission control system (VECS) (Martens, Oldervik et al. 2001), a sequential biotrickling–biofiltration (Khoramfar, Jones et al. 2018), gelling material foam (Corino and Canevari 1972), polyurethane type foam (Sani and Mohanty 2008), a thin film of surface-active materials (Canevari and Cooper 1974), aqueous foam (Gautam and Mohanty 2004) and clay nanoparticle embedded aqueous foam (Sani and Mohanty 2009, Sani, Gu et al. 2012). However, the overall pathway of crude oil processing has remained uncontrolled. A significant amount of VOCs is annually released into the atmosphere from crude oil sites in these regions which are detrimental agents to human health. Hence, there is considerable potential for proposing a global regulation to crude oil industry limiting volatile emissions and for exploring high-tech low-cost controlling methods since existing strategies are mostly time-consuming and expensive to be implemented by private and public sectors in petroleum industries. Although their initial costs of installation and maintenance are expensive, these systems in their optimum design would be considered economically profitable in the long term (Lee, Choi et al. 2013)."

4. Emission inventories of the crude oil industry

Emission inventory (EI) as a monitoring catalogue of atmospheric pollutants is usually produced as an annual or regular report prepared by the international bodies and individual industrial organisations. The first type can be very beneficial for policymakers and enable them to evaluate/improve the effectiveness of current regulations (UNFCCC (Ford et al., 2016) and CLRTAP (Byrne, 2015)). The second category, which is more focused on hazardous emissions from industrial zones (PRTRs (DeVito et al., 2015)) is useful for studies on specific sources and for proposing/improving dispersion models. From 1992-2019, several EIs have been carried out to evaluate CVEs all around the world, as depicted in Fig. 2. Table S1 in Supplementary Information presents details of these studies.

A range of point sources has been found to be responsible for a wide variety of VOCs, including CVEs; however, the correlation between specific VOCs and their probable sources were not established. The considered temporal domains in these studies were week(s) to several months and up to a year. Based on geographical and meteorological pre-evaluation of source site showing topographic maps and dominant wind/temperature currents, several points within each site were considered for CVE sampling. In order to understand and describe VOC dispersion phenomena and also source appointments which are highly affected by the meteorological factors (specifically wind and temperature) or terrain surface complexity, dispersion and source-receptor models have been employed in studies reported which include CALPUFF

(Jackson, 2006) and EPA PMF (Aklilu et al., 2018; Leuchner and Rappenglück, 2010; Wei et al., 2014a), respectively. It is noted that the emission data used in EIs are to a large extent based on estimates and statistical calculations. Therefore, it is postulated that there is a considerable level of uncertainty in EIs reported (Rypdal, 2002). Hence some statistical approaches and methods have been proposed to limit these uncertainties such as bootstrap techniques (Aklilu et al., 2018; Huang et al., 2018; Leuchner and Rappenglück, 2010) and bottom-up approach (Huang et al., 2011) and Monte Carlo simulation (Zhang et al., 2017a; Zhang et al., 2018)

With regards to the temporal variations in the VOCs originated from petroleum industries, VOC concentrations in the atmosphere around the relevant sites were found to be highly sensitive to the seasonal changes. This observation is based on the fact that the detected contents of VOCs were found to be higher in summer and then autumn (Cetin et al., 2003; Liu et al., 2008) or spring (Aklilu et al., 2018) which can be considered as a result of an increase in evaporation of volatile compounds in higher temperature. For highly volatile compounds such as benzene and toluene, lower temperature and wind speed in cold months led to their high concentrations and atmospheric lifetime (Baltrėnas et al., 2011). Moreover, the detected amounts of VOCs were higher in day time compared to night time (Lin et al., 2004). In long-term evaluations of VOCs, for example, due to seasonal variations, the amount of VOCs in the atmosphere is dominated by meteorological parameters, photochemical reactions and source characteristics. The changes in atmospheric Boundary Layer Height (BLH) were found to be more influential than other factors so that a lower BLH and fewer photochemical oxidations in cold seasons resulted in higher VOC concentrations in the atmosphere (Huang et al., 2018). These temperature-sensitive variations in atmospheric EI of HCs near petroleum refineries have been linked with the that evaporation which is the main mechanism of emissions for saturated HCs, while there was no diurnal variation in atmospheric aromatic HCs (Kalabokas et al., 2001). On the other hand, in some research, the day/night ratios of CVEs is less than 1.0, indicating higher emissions at nights (Leuchner and Rappenglück, 2010). Wind speed and direction over VOC-emitting sites have been other meteorological factors affecting the EIs of petroleum industries.

Due to the logical expectation of higher desperation rate, it is anticipated that high-speed wind would reduce VOC concentrations in the sampling points. However, we found reports that indicate the concentrations of VOCs in high-speed winds have been higher than that in slow-speed wind cases (Cetin et al., 2003). In this regard, the detected concentrations were also higher than those sampling points, which were on the direction of dominant winds when compared by other cases (Ragothaman and Anderson, 2017).

All in all, apart from several EIs carried out in northern American nations and Chinese petroleum industrial zones (Fig. 2), there are no comprehensive studies investigating emissions from crude oil processing all around the world, particularly in the Middle East, Central Africa, Russia and Latin America which are today responsible for producing about 60% of global production of crude oil (OPEC, 2019). Therefore, EIs of the crude oil industry in these areas would present regional data and could assist in forming a global picture of CVEs. According to the OPEC, the Middle East, Central Africa, Russia and Latin America are the regions with highest proven oil reserves of which Saudi Arabia, Russia, Iraq, UAE, Kuwait, Iran, Nigeria, Mexico and Venezuela are among top 15 nations that exported the highest dollar value worth of crude oil during 2019. A quick glance at Fig.2 reveals that there is an urgent need to assess CVEs in those regions which produce and distribute significant amounts of crude oil every year. Direct analyses, i.e. lab-based and in-situ tests, should be carried out to identify and quantify the VOCs emitting from various types of crude oil in those regions and also to evaluate health problems for those workers exposed to CVEs in oilfields. In addition, indirect assessments via EIs should be carried out to understand the effects of crude oil processing on regional atmospheric pollution and to detect the potential of health problems for nearby communities.

Based on studies reported in the literature, it would be difficult to provide a clear and factual explanation of why there is a lack of data for the EIs in those regions. However, we believe that such gaps in knowledge can be related to (i) lack of implementation of restrictive environmental regulations and policies due to their considerable financial demands, (ii) the significant scale of the crude oil industry in those regions (e.g. the number of oilfields, long pipelines, refineries, loading/unloading stations) (iii) strong competitions among major oil producers to reduce production cost and to maximise their profit, and (iv) access to cheap labour market.

5. Study methods

An extensive review was carried out on research reported which directly or indirectly evaluating the CVEs from all steps of crude oil processing to provide an overview of the most emitted VOCs from crude oil. Differences in analytical methods and apparatus types were taken into consideration when reporting the range of VOCs detected. This was important as some apparatus may have limitations to detect a certain range of VOC components. Hence, in some studies, more than one analytical approach was employed to detect a wider range of CVEs (Aklilu et al., 2018). Vacuum pumped canister with VOC adsorbent and simple gas-sampling nonreactive canisters with flow-limiting valves were found to be the most common types of sampling equipment installed, for example in ground levels (Pandya et al., 2006; Wei et al., 2014a) and at higher levels (Gentner et al., 2014; Huang et al., 2018). Apart from the specific EIs discussing CVEs from oceanic and terrestrial sources, continuous sampling/analysing apparatus installed on-board on ships (Gilman et al., 2013; Yagi et al., 2004) and aircrafts (Gentner et al., 2014; Koss et al., 2017; Yuan et al., 2014) were found to be most applicable methods. In this review, the relevant data supplied by other studies were utilised in some cases to assess the emissions of petroleum refineries and crude oil production areas (Gentner et al., 2014; Johnson et al., 2017; Koss et al., 2017; Warneke et al., 2014; Yuan et al., 2014). Gas Chromatograph-Mass Spectrometry (GC-MS) and Gas Chromatography with Flame-Ionization Detection (GC-FID) were found to be normally used in the labs for analysing the specimens or as online/offline analyser attached to the sampling apparatus.

In addition, each study may have used some specific units to detect VOC since they aimed at particular goals and approached the study according to their regional regulations. Therefore, it was not possible to directly compare the results to create a general picture for each volatile compound. This complexity was addressed by extensive data collection and proposing new indices dealing with concentration and detection possibility of VOCs to go beyond the limitations. The temporal and regional metrological parameters were carefully considered, and types of volatile chemicals escaping from crude oil were separately studied to ensure a comprehensive outcome for each chemical. The most effective factor on CVEs is the type of crude oil as its characteristics which highly govern the type and quantity of the VOCs released from crude oil(Saeed and Al-Mutairi, 2000).

We found that only a few studies have discussed the CVEs by considering the type of crude oil. For instance, numerous VOC-based EIs confirmed crude oil processing as one of their major sources of VOCs, e.g. Alaska crude oil (Hanna and Drivas, 1993; Sani and Mohanty, 2009), Arabian crude oil (Kalabokas et al., 2001; Salih et al., 2018; Tonacci et al., 2015), Mexican crude oil (Bahreini et al., 2012; Villasenor et al., 2003; Yuan et al., 2014), Canadian crude oil (Simpson et al., 2010; Yang et al., 2007), Nigerian crude oil (Sonibare et al., 2007) and the U.S. crude oil (Gilman et al., 2013; Helmig et al., 2014; Koss et al., 2017; Warneke et al., 2014); however, it was not possible to provide any relationships between the type of crude oil and detected CVEs from these data owing to the different analytical methods and existing uncertainties. Therefore, it appears that there is a lack of comprehensive experimental research on CVEs with respect to various types of crude oil and operating conditions (temperature, humidity, etc.). Another factor was lack of enough information related to the initial stages of crude oil production (i.e., prior to refineries), in which highly volatile compounds may escape into the atmosphere due to lack of appropriate measures.

Based on the literature review carried in this paper, it was realised that the VOCs emitted from crude oil at a global scale can be summarised based on two particular indices: i) “the possibility of detecting each compound” and ii) “its plausible concentration”. Table S2 in Supplementary Information provides details of the mentioned studies. In this regard, two parameters were defined to calculate both possibilities of detecting and plausible concentration for each VOC emitted from crude oil processing and reported in the literature. These definitions enable one to step beyond the aforementioned complexity in data analysis and to present a total outcome for each volatile chemical at a global scale. The first parameter is “Frequency of detection” (FoD) describing the number of detection of each volatile compound in all reviewed literature in the field. This parameter demonstrates to what extent it is plausible to identify a specific VOC emanated from crude oil processing. The second parameter is total average (TA) showing the ratio of the detected amount of each VOC to the total detected in each study (detection percentage) which is then averaged for each compound through all the values captured in the literature. This parameter describes the probable concentration of each compound emitting from crude oil into the atmosphere and intensifies the effects of each compound on our health and air quality. The detection percentage and total average measured for four common VOCs emitted from crude oil have been depicted in Fig. 3 as an example of data analysis and comparison procedure used in this study.

6. Crude oil VOC emissions

The FoD and TA for the detected VOCs of crude oil processing in the literature were calculated and analysed for better illustration of the problem. Fig. 4 presents the FoD and TA for all VOCs with significant total average of concentration and frequency of detection (circles are in-scale to compare chemical's detection percentage). As shown in Fig. 4, a number of compounds (in the highlighted area) have been found to have the highest FoDs and TAs, i.e. high-detected high-concentrated VOCs (Fig. 5(a-c)). These compounds are toluene, benzene, hexane, heptane, cyclohexane and pentane. Some VOCs have been found to have either higher FoDs or TAs as placed in top-left or bottom-right section of the graph, i.e. high-concentrated or high-detected VOCs, respectively (Fig. 5(b)). Ethane and Dodecane possess higher TAs than the average, however, they have not been frequently detected in the literature. On the other hand, chemicals such as ethylbenzene, m,p,o-xylene, trimethylbenzene isomers, 3-methylpentane, octane, methylcyclopentane and methylcyclohexane were more detected with lower TAs.

Fig. 5 (C) shows those compounds with the lowest FoDs and TAs (low-detected low-concentrated VOCs). The fluctuation of the data demonstrated in Fig. 5(a-c) is inevitable due to previously mentioned reasons for dissimilarities in analytical methods and regional operating conditions. The information also confirms the significant of BTEX in emission analysis of crude oil processing. As discussed, they are well-known to be extremely hazardous to human wellbeing, and EPA IRIS provided strong limitations about exposure to BTEX (Baltrėnas et al., 2011; Durmusoglu et al., 2010). Boiling point is an influential parameter on evaporative emissions of organic chemicals from crude oil(Hanna and Drivas, 1993). Detected CVEs illustrated in Fig. 6 are based on their boiling points along with their TAs and FoDs. As shown in Fig. 6, the majority of high-FoD compounds with a wide variety of TAs are placed within a specific range of boiling points, i.e. 48-180℃ (highlighted in green). It seems that a major part of those VOCs which are not within this area have low boiling points and release from oil mass during earlier steps of crude oil processing (extraction). This emphasises on the necessity of implementing control measures to regulate CVEs in the primary steps of extraction-to-refinery pathways of crude oil processing.

7. Control measures of crude oil VOC emissions

CVEs released during various steps of crude oil processing have been reported to be a significant volumetric loss of HCs (Michaelowa and Krause, 2000). According to (Choi et al., 2019), up to 2.4 million tons of CVEs are annually escaped to the environment from crude oil during loading/unloading and transportation resulting in a financial loss up to about 700 million US dollars. However, according to recent OPEC report discussing the countries with the highest amounts of crude oil import or export, it is possible to identify those nations on which the financial loss due to the release of CVEs would annually have significant impact; however, based on the findings of this review, it appears that the CVEs are being less controlled in exporting countries than those developed importing nations which usually utilise engineering control measures to take the highest advantages of imported crude oil.

Based on a recent statistical report by the International Tanker Owners Pollution Federation (ITOPF), approximately 50% of major crude oil incidences (incidents with oil spillage larger than 700 tonnes) between 1970 and 2017 have happened in open water. This significance of total spillage proportion can hardly be minimised and restricted using controlling/recovering CVEs systems. The rest of the oil spillage is related to inland transportation incidences (17%), loading/discharging (15%) and unknown sources (18%). Moreover, the major amounts of CVEs which should be controlled are emitted through charging/discharging and transportation of crude oil, especially in ocean tankers due to their high capacities (Karbasian et al., 2017). The CVEs during ballast water loading were found to be negligible; however, crude oil washing (COW) operations of tankers are also considered as another source of CVEs which is highly dependent on operating conditions (Gunner, 2002). It is noted that unloading crude oil has not been reported as a major cause for CVEs since discharging of bulk liquid is inevitably coincident with the inflow of air into reservoirs as a natural control of CVEs. However, sometimes operations are poorly managed, and excessive pumping rates have been used in the unloading process (Howard and Nikolas, 2001).

The control measures of CVEs are case-specific and highly dependent on crude oil properties and operating conditions; hence, for each case, the volatility of CVEs, temperature of both crude oil and gaseous phase, an estimation of the plausible amount of CVEs and the vent-stream characteristics are influential factors which have to be fully comprehended(Karbasian et al., 2017). Apart from the modification of equipment involved in the CVE emissions, most control methods are derived from VOC abatement procedures which can be broadly classified as recovery methods and destruction techniques. Recovery procedures aim to collect, store and reuse VOCs and include membrane separations (Zhang et al., 2019a), condensation (including refrigeration condensation (Xiong et al., 2014) and cryogenic condensation (Masetto, 2011)), adsorption (Zhang et al., 2017b) and absorption (Yen and Jeng, 1996). Destruction methods aiming at converting VOCs into simpler and safe compounds (CO_2 and H_2O) are thermal incineration (including regenerative and recuperative (Salvador et al., 2006)), catalytic incineration (Tseng et al., 2011), photo-catalytic oxidation (Lee, 2000), plasma technology (Du et al., 2018), electron beam technology (Son et al., 2010), biofiltration (Nikiema et al., 2007) and flares (McGowan, 2016). The most common strategies in CVE controlling systems are vapour recovery (pressure and temperature swing adsorption) and vapour suppression. The recovery methods require considerable energy consumption and incineration by-product (CO_2) (Sani and Mohanty, 2009). Suppression techniques aim at providing a barrier against VOCs by the use of gelling material foam (Corino and Canevari, 1972), polyurethane type foam (Sani and Mohanty, 2008), thin film of surface-active materials (Canevari and Cooper, 1974), aqueous foam (Gautam and Mohanty, 2004) and clay nanoparticle embedded aqueous foam (Sani and Mohanty, 2009; Sani et al., 2012). Thus, the intended foam must have a high persistency over time, a low permeability towards VOCs, a desirable fluidity and acceptable flexibility to move (Gautam and Mohanty, 2004). Compared with destruction methods which result in loss of significant amounts of VOCs, recovery and suppression methods seem to be more profitable options for collecting VOCs (Tamaddoni et al., 2014). These methods would be utilised for designing specific CVE control systems if their performance and stabilities were to be satisfactory (Gautam and Mohanty, 2004). There are several control methods (adapted from recovery procedures) for CVEs in inland and offshore transportations including absorption in crude oil, refrigeration/pressurisation condensation (to reuse as fuel), HC blanketing/recovering and vapour balancing (Table 1).

The control necessities of CVEs have been the subject of various investigations. In 1978, the Air Quality Planning and Standard Office of EPA published a guideline to control VOCs leaked from US petroleum refinery equipment which consisted of two main steps (i.e. monitoring and replacing/repairing) (HUSTVEDT et al., 1978). The manual was updated in 1982 to provide background information for proposing standards (GOODWIN, 1982). The report was only about refinery VOC emissions, and the fixed roof storage tankers were found to be the main source of CVEs. In this regard, the pump and compressor seals, pipeline valves, open-ended valves, drains, pressure relief equipment, flanges and other connections were a major local sources of CVEs (HUSTVEDT et al., 1978). Martens et al. (2001) carried out a modelling of the optimum performance (energy consumption and power demands) of some CVE controlling actions used by Norwegian authorities on their crude oil shuttle tankers during offshore loading. The CVE recovery system, reabsorption unit, re-condensation plant and collecting pipeline system (for Sequential Transfer Tank Atmospheres, STTA) were considered methods of their study for controlling CVEs in storage and shuttle tankers (Martens et al., 2001). The efficiency of an aqueous foam to control CVEs was evaluated by Gautam et al. (Gautam and Mohanty, 2004) which was followed by further developments were also tested using incorporated nano-clay (Sani and Mohanty, 2009; Sani et al., 2012). Tamddoni et al. (2014) provided an efficient absorption strategy of VOC recovery with referencing to their experimental and numerical studies (Tamaddoni et al., 2014). In 2018, a sequential biotrickling–biofiltration method to remove VOCs from crude oil tankers was tested by Khoramfar et al. for about three months as an applicable biological solution to CVE issues (Khoramfar et al., 2018). Karbasian et al. also performed an extensive numerical and physical modelling to propose a new swirl unit in crude oil loading stage, reducing the formation of VOCs (Karbasian et al., 2017).

These controlling measures, as a whole, are time-consuming and costly to be carried out for private and public sectors engaged with petroleum industries. Although their initial costs of installation and maintenance are perceived to be high, in the long term, these systems in their optimum design would be economically profitable(Lee and Chang, 2014; Lee et al., 2013). In total, there are direct relationships between the petroleum end-use demands and the inevitable amount of released CVEs (from displacement, spillage, refinery and storage facilities). It seems that the majority of existing scenarios is allocated to crude oil shipment and transportation sector, and other steps of well-to-refinery pathways of crude oil processing are still uncontrolled and need to be more studied to propose competent methods of VOC abatement methods.

8. Conclusion

This paper provides, for the first time, a global-scale insight into VOC emissions from the crude oil industry. The data in the literature were carefully collected and analysed to provide an illustrative outcome for global VOC emissions from crude oil processing. Inherent limitations in the used analytical approaches, discrepancies in types of crude oil and meteorological influences on crude oil VOC emissions (CVEs) were the main challenges to accurately quantify the CVEs. Two representative parameters were introduced to manage these complexities in the data, including frequency of detection (FoD) and total average (TA). The CVEs were classified into four different groups based on their FoDs and TAs: (i) high-detected high-concentrated, (ii) high-detected, (iii) high-concentrated and (iv) low-detected low-concentrated. Among all VOCs emitted from crude oil, toluene, benzene, hexane, heptane, cyclohexane and pentane were found to be high-detected high-concentrated compounds. The majority of the detected VOCs have relatively high boiling points which emphasise the importance of implementing control measures at early stages of crude oil extraction. Restrictive policies and regulations are vital to control the VOC emissions worldwide, particularly those related to the oil and gas industry. The reported oral/inhalation toxicity and carcinogenic effects from VOCs in crude oil production sites are still a matter of debate and should be further explored. Based on the reviewed literature, several gaps in the current knowledge have been identified and suggested to be investigated in future studies:

- Understanding of the mechanisms involved in the VOC emissions from crude oil by considering the effects of both crude oil types and operating conditions.
- In-situ investigations to monitor VOCs during various stages of crude oil processing, particularly during the early stages of extraction.
- In-depth assessment of influential parameters on VOC emissions from oil contaminated soils.

Supplementary information

Following supplementary pieces of information are provided in a separate document:

- Summary of research studies on emission inventories (EIs) of petroleum industries.

- Summary of the CVEs detected and reported in the literature.

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Table 1. Summary on experimental, field-based and EI studies of crude oil VOC emissions.

Focus of study	Source(s)	Research outline	Crude oil type(s)	Reference
Experimental and field-based studies	Massive oil-spills from damaged well heads during 1991 Gulf War	Due to huge consumption of seawater to control fire in exploded oil wells in the 1991 Gulf War, VOC emission from the water-soluble fraction of ten types of Kuwaiti crude oil was assessed by lab-oriented GC apparatus. About 40 VOCs were identified in WSFs of crude oil.	10 types of Kuwaiti crude oil	(Saeed and Al-Mutairi, 2000)
	Crude oil tankers	Novel aqueous foams were proposed as a way of controlling COEs from crude oil tankers and storage facilities.	Not mentioned	(Gautam and Mohanty, 2004)
	Fresh and weathered crude oil	A new automatic sampler-headspace-GC/MS (ALS-HS GC/MS) was proposed for VOC detection over crude oil samples. VOC emissions were reduced as crude oil aged for two weeks.	Canadian crude oil	(Yang et al., 2007)
	Crude oil tankers	Nano-clay incorporated aqueous foam was fabricated as a novel foam to control COEs in crude oil storage tanks.	Alaska North Slope crude oil	(Sani and Mohanty, 2009)
	Alberta's oil sands	Airborne whole air sampling collected during 2008 Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) field mission was used to detect VOCs, CO_2, CH_4, CO, NO, NO_2, NOx, O_3 and SO_2	Canada oil sands	(Simpson et al., 2010)
	Crude oil refinery in Lithuania	An experimental survey was carried out to determine gaseous BTEX in the vicinity of crude oil refinery and the effects of metrological parameters.	Not mentioned	(Baltrėnas et al., 2011)
	Provided crude oil from North Sea Friedrichskoog, Germany	This research focused on new a static headspace gas chromatography flame ionization detection method for detecting super VOCs in the field.	Light crude oil Heavy crude oil Mixed crude oil	(Wang et al., 2014)
	n-Pentane and n-hexane purchased from Baker and Merck	An experiential study was carried out to propose a new method of detecting binary VOCs of crude oil using SPME which is suitable for in-situ assessment.	NA	(Wang et al., 2015)
	Crude oil spillage over sea water	A new smart senor was proposed to detect hydrocarbon VOCs emitted from contaminated seawater.	Arab Light from Saudi Arabia Sarir crude oil from Libya	(Tonacci et al., 2015)
	Crude oil tankers	A new sequential biotrickling–biofiltration unit to remove VOCs from crude oil storage areas was proposed.	Not mentioned	(Khoramfar et al., 2018)
	Ocean crude oil spillage	An experimental study was carried out to investigate the effects of breaking waves on VOCs emitted from crude oil and crude oil-dispersant contaminated seawater.	Louisiana Light Sweet crude oil	(Afshar-Mohajer et al., 2018)
	Crude oil spillage	VOC emissions of crude oil were monitored by GC-FID to assess crude oil emissions in surface and deep soil in short- and long-term (3 months).	Provided from pipeline in the location Friedrigskoog, Germany	(Mustafa Salih et al., 2018)
	Crude oil land spillage	VOCs emitted from crude oil with different sulphur contents were measured indoor and outdoor circumstances.	Taq Taq crude oil Khurmala crude oil	(Salih et al., 2018)
	Ocean crude oil spillage	Human health risk of VOCs emitted from crude oil and crude oil-dispersant contaminated seawater under breaking waves was assessed via an experimental study.	Louisiana Light Sweet crude oil	(Afshar-Mohajer et al., 2019)

Table 1. Summary on experimental, field-based and EI studies of crude oil VOC emissions (continued).

Focus of study	Source(s)	Research outline	Crude oil type(s)	Reference
Studies based on emission inventories	Crude oil production Crude oil storage Crude oil transport	The 1993 VOC emissions in all steps of gasoline production and marketing cycle were analysed to provide an estimation for upstream VOC and petroleum refinery emissions from the use of gasoline in the United States in 2000.	All types of crude oil used in the US were considered, but their names and characteristics were not mentioned,	(DeLuchi, 1993)
	Crude oil refinery	An EI was prepared to investigate seasonal, diurnal and spatial variations of saturated and aromatic HCs around an oil refinery in Greece.	Mostly Arabian light and Arabian medium crude oil	(Kalabokas et al., 2001)
	Offshore production crude oil	An EI was proposed for the first time regarding the offshore petroleum industry in Mexico covering 174 platforms of crude oil extraction and production.	Mexican Maya and Istmo crude oil	(Villasenor et al., 2003)
	Crude oil refinery	VOC concentrations of ambient air were detected for a year around a petrochemical complex and an oil refinery (processing $\approx 10\times10^6$ tonnes crude oil annually) in Izmir (Turkey).	Not mentioned	(Cetin et al., 2003)
	Global-scale-background concentrations of VOCs originating from Petroleum over oceans	A global EI of petroleum VOCs (aromatic hydrocarbons, halogenated aromatic hydrocarbons, halogenated aliphalic hydrocarbons, chlorofluorocarbons etc.) was provided via on board sampling devices on ships travelling through two routes between Japan and Australia.	Not mentioned	(Yagi et al., 2004)
	Crude oil refinery	The air quality of an oil refinery in Kaohsiung, located in southern Taiwan, was assessed via emission inventory prepared by in-situ sampling and GC/UV-DOAS analyses. The most abundant COEs were Benzene and Toluene.	Not mentioned	(Lin et al., 2004)
	Crude oil refinery	Individual VOCs, total hydrocarbon and total VOCs at an Indian oil refinery were analysed in its crude distillation unit, vacuum distillation unit, catalytic reforming unit, new delayed cocker unit, LPG recovery unit, LPG dispatch unit, wax hydro finishing unit and solvent dewaxing unit.	Not mentioned	(Pandya et al., 2006)
	Crude oil tankers	US EPA standard regulatory storage tanks emission model (TANKS 4.9b) was used to estimate emission of 9 VOCs from 8 organic liquids storage tanks located in Dar-es-Salaam City Tanzania	Not mentioned	(Jackson, 2006)
	Crude oil refinery	Refinery emission prediction was investigated by the emission factor approach to evaluate VOC contribution of petroleum refineries to Nigeria's airshed.	Nigerian crude oil	(Sonibare et al., 2007)
	Crude oil terminals	PHOENICS 3.5 software package was utilised based on the data of oil terminals in Lithuania	Not mentioned	(Paulauskiene et al., 2009)
	Crude oil production and refineries	The data provided by a part of TexAQS-II Radical and Aerosol Measurement Project (TRAMP) was analysed to provide EI of VOCs along with receptor model to find their major contributions.	Not mentioned	(Leuchner and Rappenglück, 2010)
	Crude oil storages and refineries	An EI of anthropogenic sources of VOCs in the Yangtze River Delta region, China was presented.	Not mentioned	(Huang et al., 2011)
	The Deepwater Horizon Oil Spill	The data of air quality collected by The NOAA WP-3D research aircraft over The Deepwater Horizon Oil Spill was utilised to assess VOCs and SOA in the Gulf of Mexico.	Mexican crude oil	(Bahreini et al., 2012)

Table 1. Scientific literature of crude oil volatile organic compounds emissions (continued).

Focus of study	Source(s)	Research outline	Crude oil type(s)	Reference
Studies based on emission inventories	Crude oil production	VOCs emitted from crude oil, and natural gas production areas of Colorado were studied by the use of data collected by NOAA's Boulder Atmospheric Observatory as part of the NACHTT (Nitrogen, Aerosol Composition, and Halogens on a Tall Tower) campaign.	Colorado crude oil	(Gilman et al., 2013)
	Crude oil storages and refineries	VOCs emitted from China, processing 10 million tonnes of crude oil annually were analysed. PMF receptor model was employed to investigate sources of VOCs.	Not mentioned	(Wei et al., 2014)
	- The Deepwater Horizon Oil Spill Crude oil lab-based analyses	The provided data by on-board PTR-MS installed on the NOAA WP-3D aircraft over the Deepwater Horizon (DWH) oil spill in the Gulf of Mexico were compared with VOC emission of same crude oil analysed by lab-based PTR-MS.	Mexican crude oil	(Yuan et al., 2014)
	Crude oil production	The data provided by UBWOS2012 campaign was used to evaluate VOC emission from crude oil and natural gas production sites in the Uintah Basin, Utah	Utah crude oil	(Warneke et al., 2014)
		The highly elevated VOCs originated from heavy crude oil and natural gas were evaluated during 2012 Uintah Basin Winter Ozone Studies.		(Helmig et al., 2014)
	Crude oil storages and refineries	Based on the data collected by the NOAA WP-3D Orion research aircraft during the Shale Oil and Natural Gas Nexus (SONGNEX) campaign in March and April 2015 over nine large central US crude oil and gas production region, major VOCs emitted from petroleum industries have been reviewed.	American crude oil	(Koss et al., 2017)
	Crude oil storages and refineries	An EI was prepared to detect VOC emission from petroleum refinery located in the Pearl River Delta (PRD) region in China which processes about 13.2 million tons of crude oil.	Not mentioned	(Zhang et al., 2017)
	Crude oil storages and refineries	An EI was prepared to monitor VOC emissions from a petroleum refinery in Pearl River Delta of China with the aim of health risk assessment	Not mentioned	(Zhang et al., 2018)
	Crude oil storages and refineries	A one-year campaign was arranged for the assessment of VOC emissions from crude oil and gas production in the north-western margin of the Junggar Basin, China.	Not mentioned	(Huang et al., 2018)
	Peace River Oil Sands Area (PROSA)	Positive matrix factorization (PMF) was used to study sources of VOCs monitored in the cold heavy oil production area in Alberta, Canada.	Canadian crude oil	(Aklilu et al., 2018)
	Crude oil storages and refineries	VOC EI of offshore and onshore crude oil and gas production areas in the Gulf of Kavala was presented	Not mentioned	(Papailias and Mavroidis, 2018)

Table 2. Control procedures during oceanic tanker loading and transport (Ministry of Infrastructure and Water Management, 2009)

Circumstances	Procedures		Brief explanations
Crude oil loading	Vapour Emission Control System (VECS)		VECS, introduced in 1990, uses the inert gas piping to collect/deliver COEs to VECS manifolds for processing at front and rear of vessels in-shore.
	Vapour Pressure Release Control System (VOCON Valve)		VOCON Valve, a single valve facility and located at the bottom of the mast riser, limits COEs releases and involves two pieces of equipment: constant pressure valves and an automated release valve operating according to the VOCON procedure.
	Cargo Pipeline Partial Pressure Control System (KVOC)		KVOC system diminishes COEs by interrupting their productions during loading and also transit using a new high-diameter drop pipeline column reducing crude oil flow.
	Vapour Recovery Systems (VRS)	Condensation System	Pressurized/liquefied COEs are discharged to shore, or be used as fuel on-board for boilers or engines.
		Absorption System	Utilization of crude oil as an absorbent of COEs in a counter-current flow of crude oil in an absorber column.
		Absorption Carbon Vacuum-Regenerated Adsorption (CVA)	CVA system firstly uses activated carbon as adsorbent of COEs, and adsorbed COEs ultimately desorbed from AC by vacuum and deliver to an absorber column of crude oil.
		Direct Absorption of VOC in Crude Oil (CVOC System)	CVOC system employs swirl absorber (a combined ejector and mixing unit) to create a low-pressure domain so as to mix again COEs with crude oil.
	Increased Pressure Relief Settings (Applicable also for transit conditions)		To curb further COEs, tank pressure is maintained to obtain an equilibrium between the liquid and vapour phase of the cargo.
Crude oil transit	Vapour Pressure Release Control System (VOCON Valve)		VOCON Valve, a single valve facility and located at the bottom of the mast riser, limits COEs releases and involves two pieces of equipment: constant pressure valves and an automated release valve operating according to the VOCON procedure.
	Recovery of excess VOC and tank absorption (Venturi system)		Venturi system (consisting of a pressure controlled pump, feeding oil to a unit with Venturi(s)) is a process in which COEs is reabsorbed back into the crude oil.
	Increased Pressure Relief Settings		To curb further COEs, tank pressure is maintained to obtain an equilibrium between the liquid and vapour phase of the cargo.

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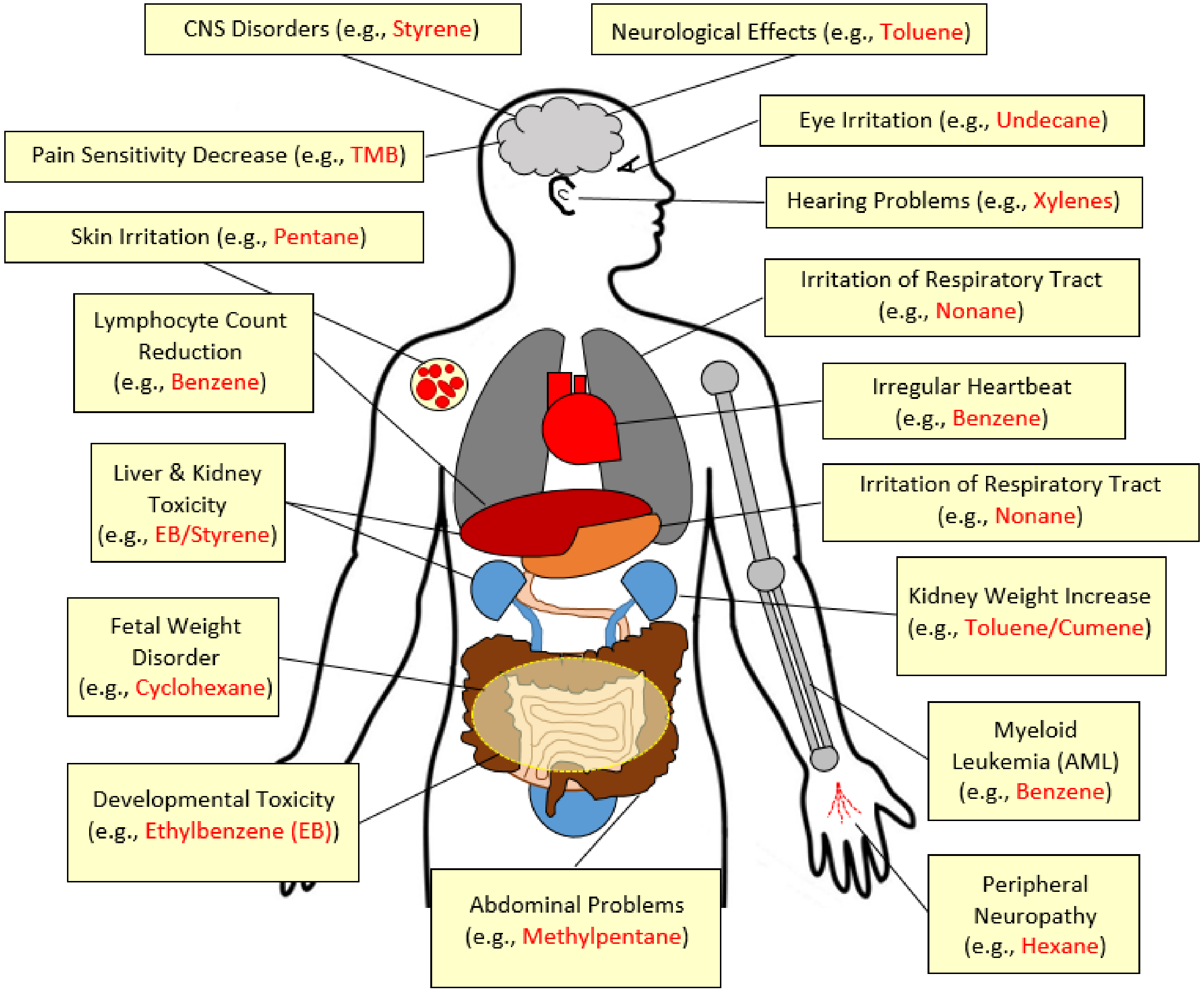


Fig. 1. Major impacts of VOCs on human health

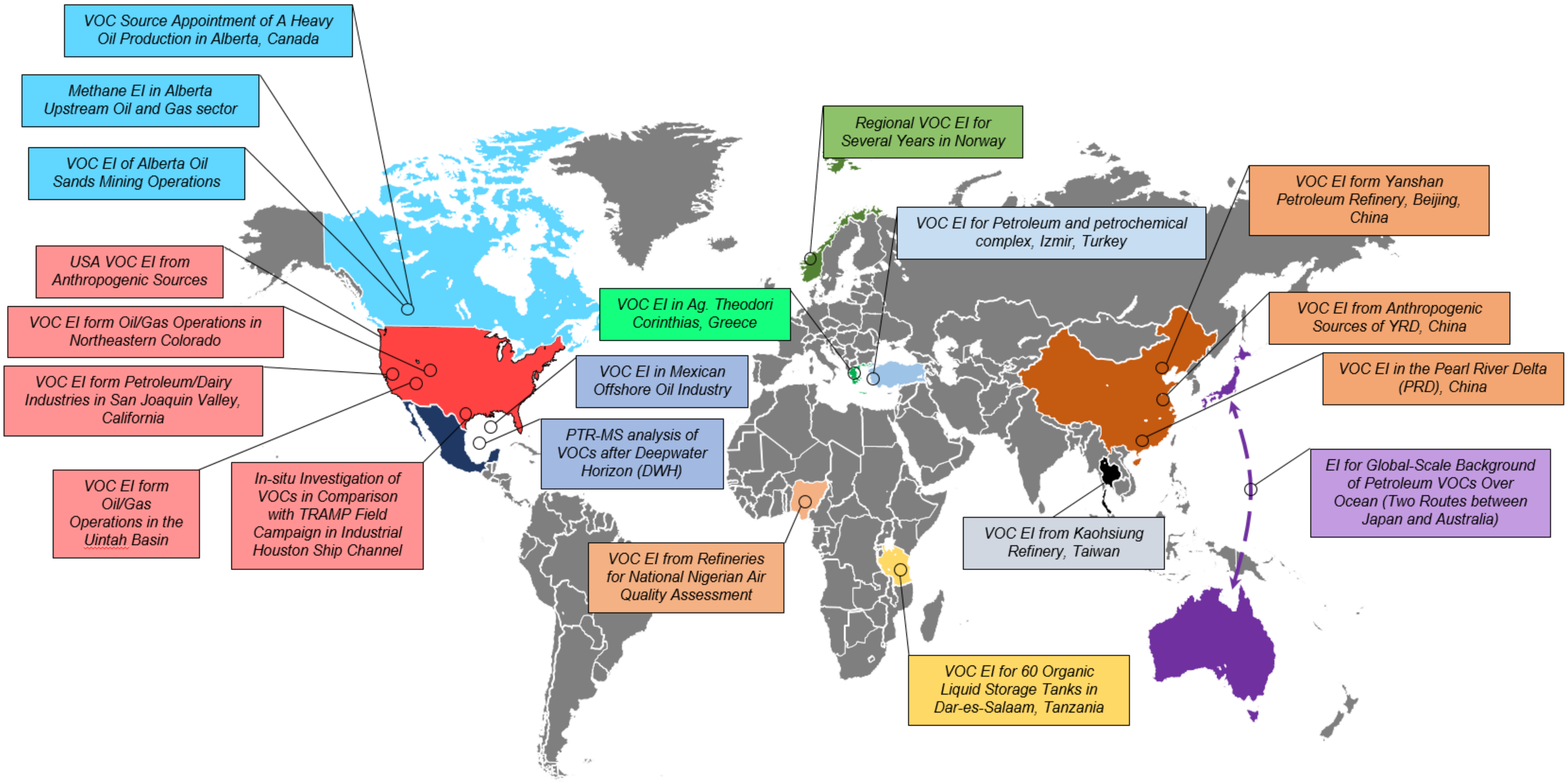


Fig. 2. Emission inventories of crude oil industry worldwide.

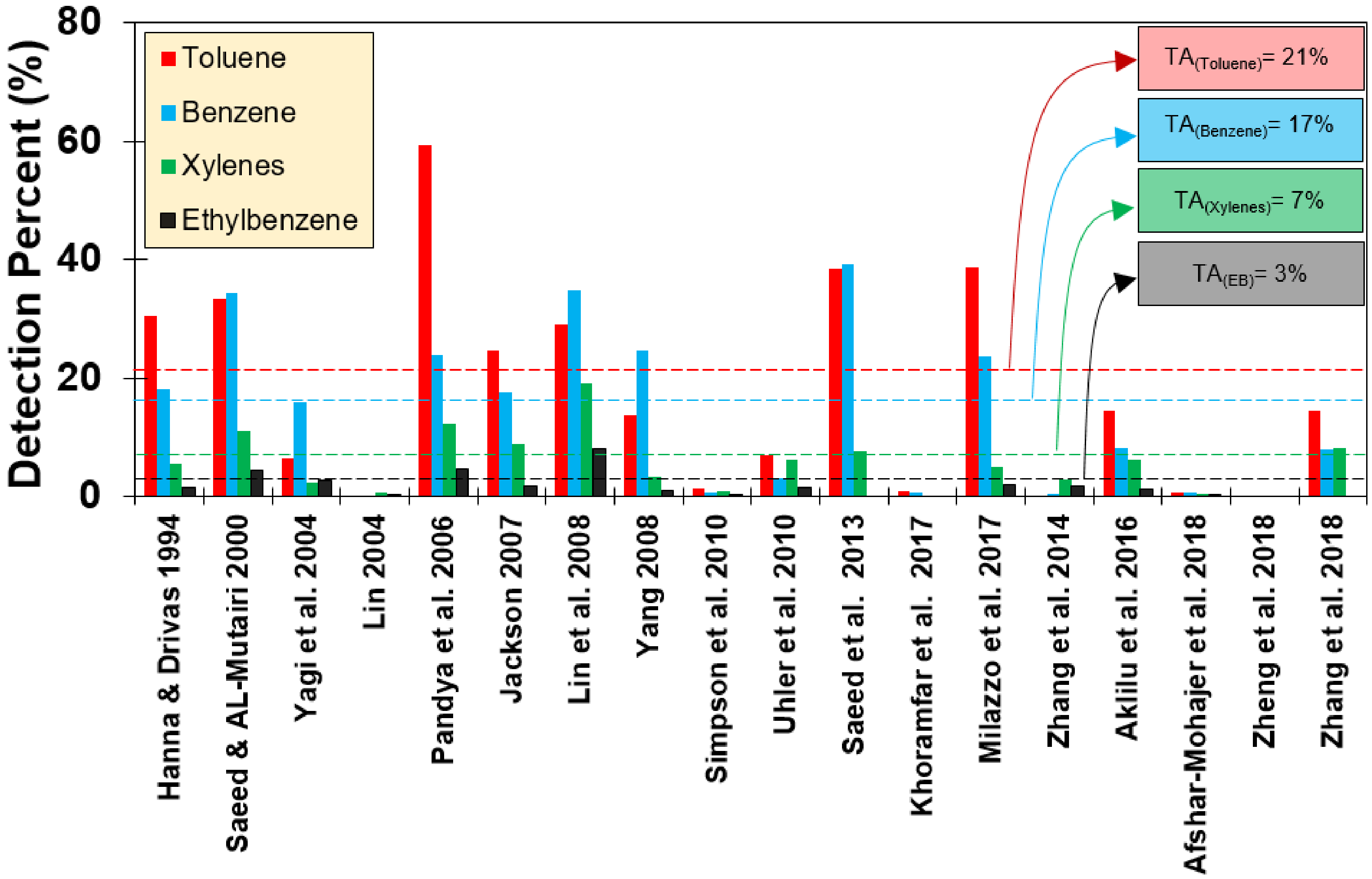


Fig. 3. Total average and detection percentage for toluene, benzene, xylenes and ethylbenzene

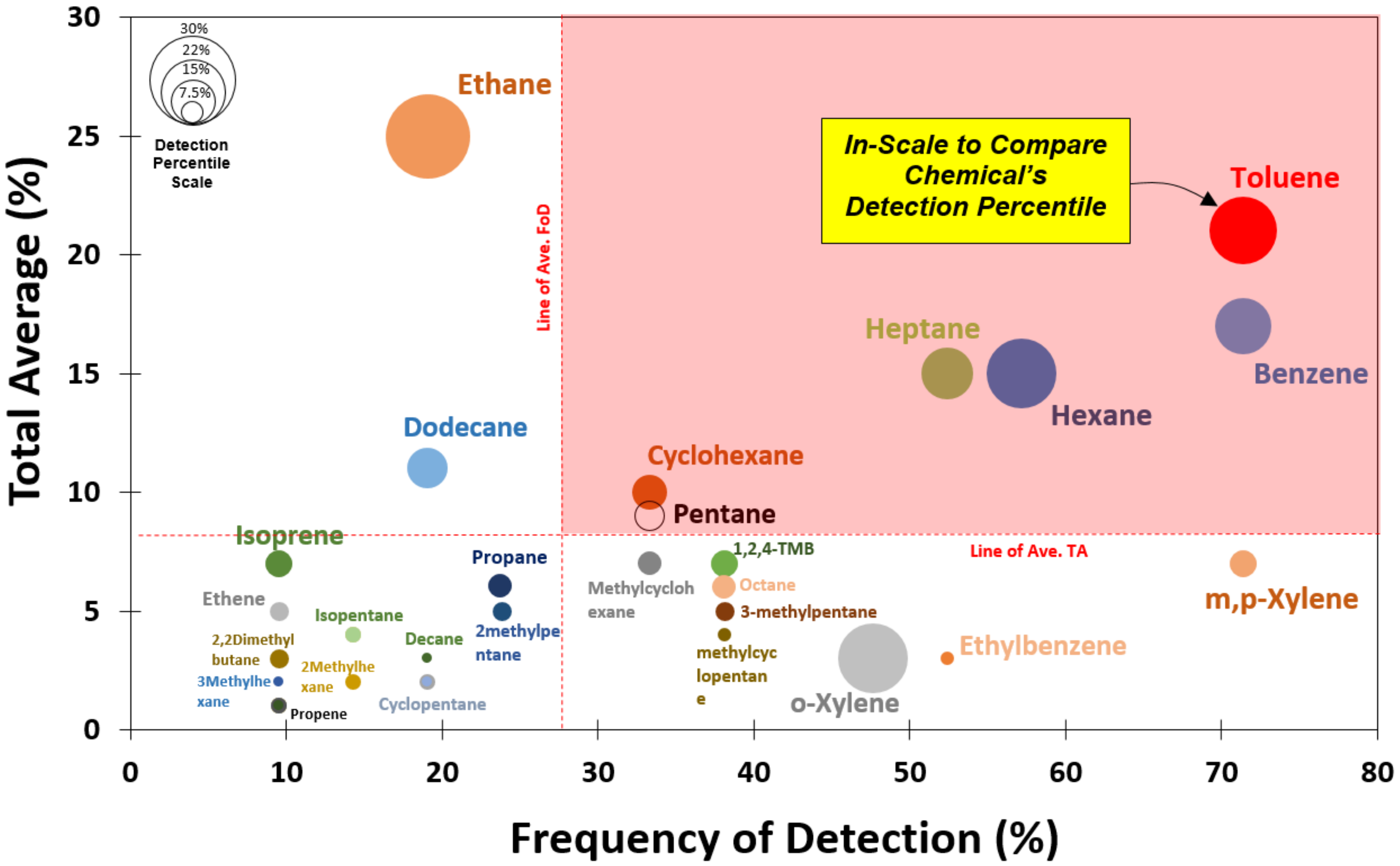
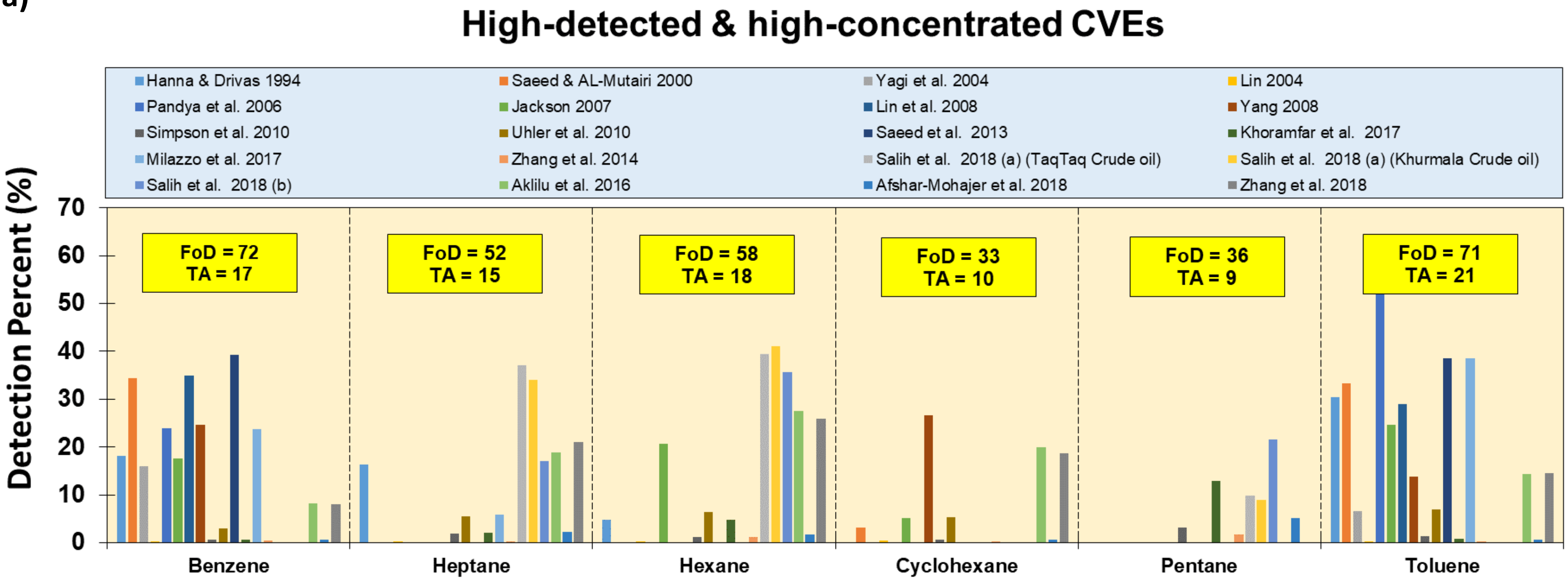
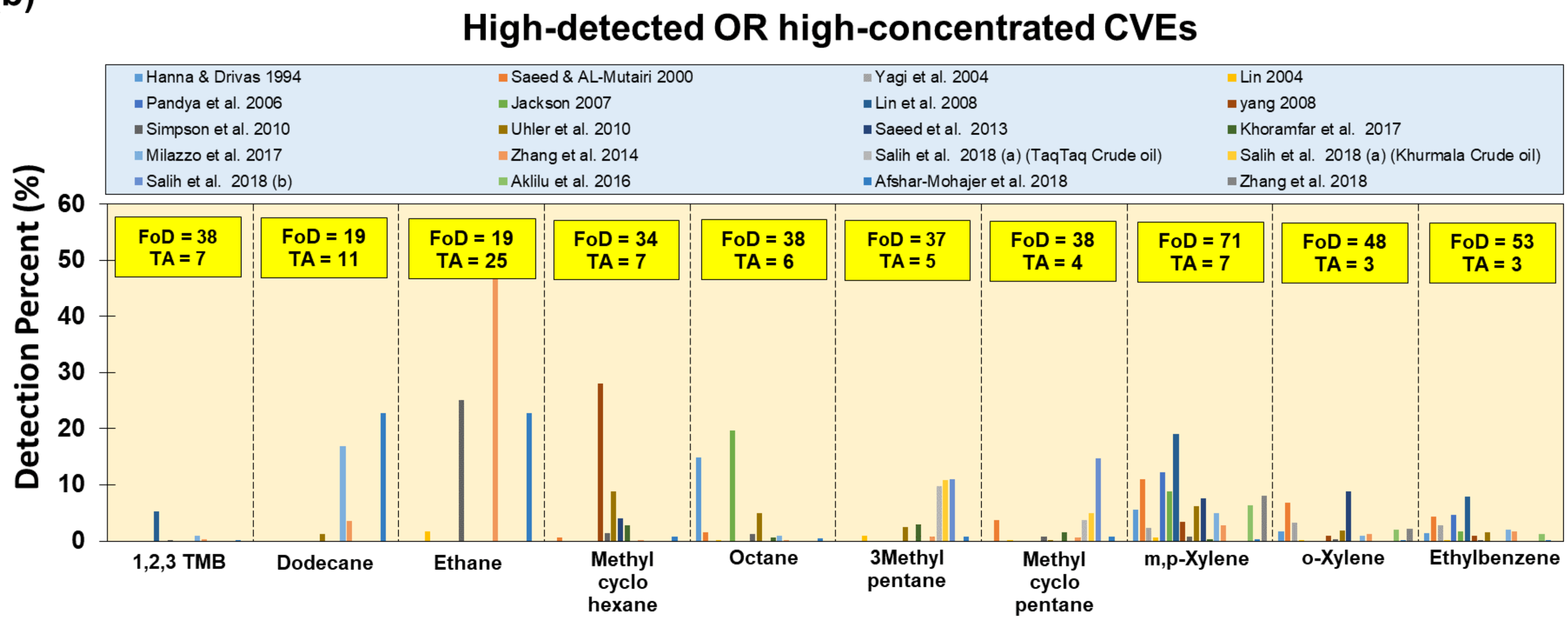


Fig. 4. Frequency of detection and total average of VOCs emitted from crude oil.

(a)


High-detected & high-concentrated CVEs
Hanna & Drivas 1994
Saeed & AL-Mutairi 2000
Yagi et al. 2004
Lin 2004
Pandya et al. 2006
Jackson 2007
Lin et al. 2008
Yang 2008
Simpson et al. 2010
Uhler et al. 2010
Saeed et al. 2013
Khoramfar et al. 2017
Milazzo et al. 2017
Zhang et al. 2014
Salih et al. 2018 (a) (TaqTaq Crude oil)
Salih et al. 2018 (a) (Khurmala Crude oil)
Salih et al. 2018 (b)
Aklilu et al. 2016
Afshar-Mohajer et al. 2018
Zhang et al. 2018
Detection Percent (%)
0
10
20
30
40
50
60
70
FoD = 72
TA = 17
FoD = 52
TA = 15
FoD = 58
TA = 18
FoD = 33
TA = 10
FoD = 36
TA = 9
FoD = 71
TA = 21
Benzene
Heptane
Hexane
Cyclohexane
Pentane
Toluene

(b)


High-detected OR high-concentrated CVEs
Hanna & Drivas 1994
Saeed & AL-Mutairi 2000
Yagi et al. 2004
Lin 2004
Pandya et al. 2006
Jackson 2007
Lin et al. 2008
yang 2008
Simpson et al. 2010
Uhler et al. 2010
Saeed et al. 2013
Khoramfar et al. 2017
Milazzo et al. 2017
Zhang et al. 2014
Salih et al. 2018 (a) (TaqTaq Crude oil)
Salih et al. 2018 (a) (Khurmala Crude oil)
Salih et al. 2018 (b)
Aklilu et al. 2016
Afshar-Mohajer et al. 2018
Zhang et al. 2018
Detection Percent (%)
60
50
40
30
20
10
0
FoD = 38
TA = 7
FoD = 19
TA = 11
FoD = 19
TA = 25
FoD = 34
TA = 7
FoD = 38
TA = 6
FoD = 37
TA = 5
FoD = 38
TA = 4
FoD = 71
TA = 7
FoD = 48
TA = 3
FoD = 53
TA = 3
1,2,3 TMB
Dodecane
Ethane
Methyl cyclo hexane
Octane
3Methyl pentane
Methyl cyclo pentane
m,p-Xylene
o-Xylene
Ethylbenzene

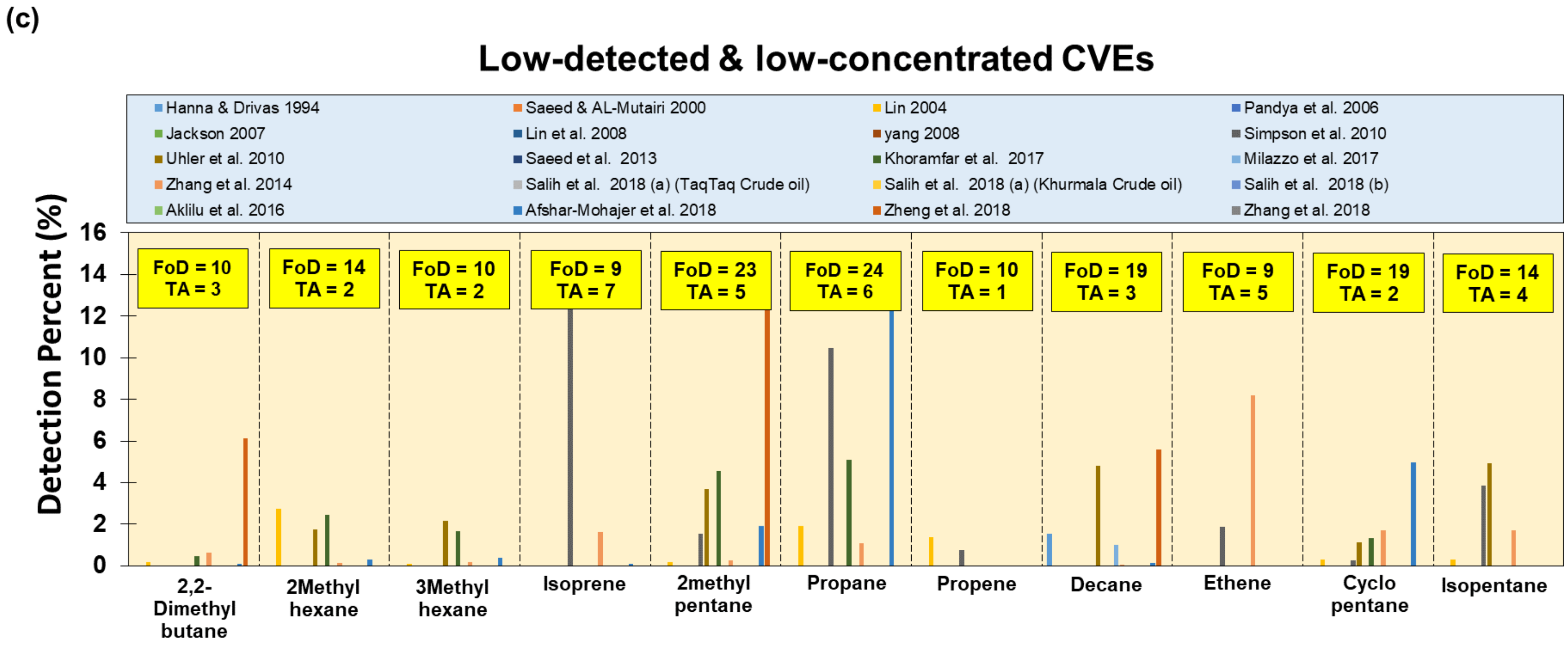


Fig. 5. (a) High-detected high-concentrated, (b) high-detected or high-concentrated, and (c) low-detected low-concentrated VOCs emitted from crude oil

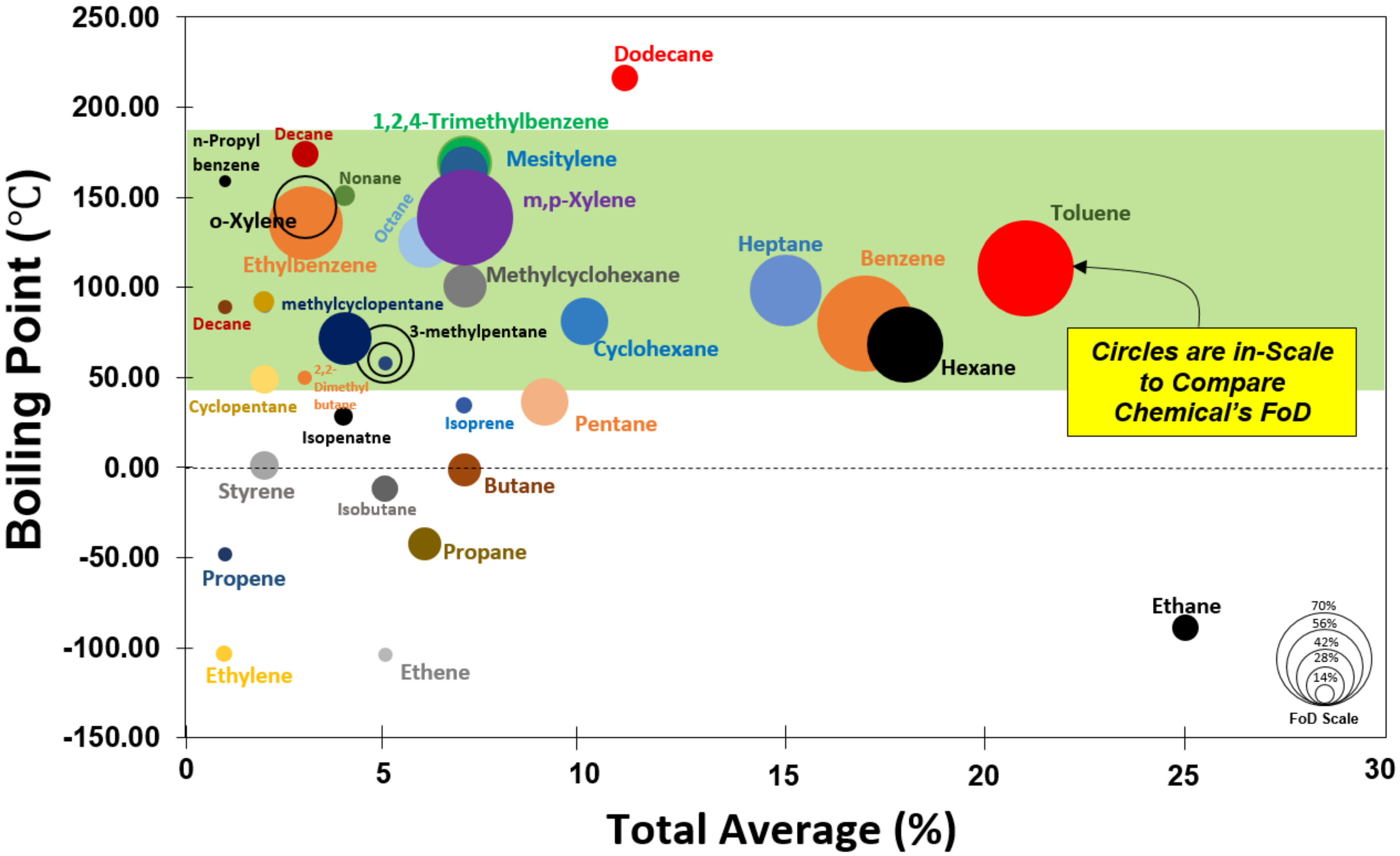


Fig. 6. High FoD-TA CVEs with specific range of boiling points

SUPPORTING INFORMATION

Emissions of Volatile Organic Compounds from Crude Oil Processing – Global Emission Inventory and Environmental Release

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Number of Pages: 12

Number of Figures: 0

Number of Tables: 2

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Table S2. The CVEs detected and reported in the literature.

Table S1. Research studies on emission inventories (EIs) of petroleum industries.

Aim(s)	Location	Time	Methods of Data Collection, EI and modelling	Uncertainty analysis or reasons	Crude oil related sources	Reference
- Global VOC EI from Anthropogenic sources	- The United States as an illustrative example of the globe	- NA	- The U.S. EPA NAPAP EI Database (Version 2 - 1989) - The United Nations (UN) Statistical Office Database	- Due to use of the U.S. EI as a base for global EI due to different ranges of industrialisation, population, etc. - Weighted average quality ratings provided for describing uncertainties	- The considered sources are: **Crude oil**/Gasoline/Fuel oil/Others marketing, storage, transportation and consumption; Oil & gas production; Coke production; Petroleum refining; Paper/Surface coating operations; Printing & publishing; Rubber & plastics; Solvent use; Paint production; Automobile industries; Paper industry; Asphalt paving; Diesel vehicles; Bakeries; Aircrafts LTOs; Fuelwood utilisation; Refuse disposal; Deforestation; Forest fires; Managed/Savanna burning; etc.	(Piccot et al., 1992)
- Air pollution levels of saturated and aromatic HCs	- Oil refinery in Ag. Theodori Corinthias, Greece	- 22 Feb-2 Mar - 4 May-12 May - 14 Sep-21 Sep - 25 Oct-2 Nov	- Sample collection (6 points of sampling; total 130 VOC samples) - Sampling (glass adsorption tubes (Chrompack) filled with Tenax-TA) - Analyser (GC-FID) - Metrological data (wind speed, wind direction, temperature)	NA	- The considered oil refinery had a processing capacity of **Crude oil** about 4.75 million tons per year.	(Kalabokas et al., 2001)
- Uncertainty analysis Norwegian EI - Projected EI for 2010	- Norway (national EI)	- 1990 - 1998 - 2000	- Data from national EI of Norway published by Selected Nomenclature for Air Pollution (SNAP)	- Uncertainty propagation or inverse modelling	- **Crude oil** loading selected as source for non-methane VOCs for	(Rypdal, 2002)
- EI of Mexican offshore oil industry	- The Gulf of Mexico	- 1999-2000	- Data from Air Quality Division personnel of the Mexican oil company at Atasta and Dos Bocas - Model (CALMET/CALPUFF/Regional Atmospheric Modelling System) - Metrological data (National Oceanic Atmospheric Administration (NOAA)) and Upper air soundings from IMP	- Recommended for future work	- Studied offshore operations were **crude oil** wells, re-pumping/compression stations, pipelines, and shipment	(Villasenor et al., 2003)
- VOC EI of Petroleum and petrochemical complex - Metrological effects	- Petroleum refinery and a connected petrochemical complex, Izmir, Turkey	- 12 months	- Sample collection (3 points of sampling; total 26 VOC samples) - Sampling (Vacuum pumped using carbon adsorbent (activated carbon)) - Analyser (GC-FID) - Metrological data (wind speed, wind direction, temperature and relative humidity) - Multiple linear regression analysis proposed for relations between individual VOC and meteorological parameters	NA	- No specific sources listed. - The studied refinery processed 9.14×10^6 tonnes of **Crude Oil** per annum	(Cetin et al., 2003)
- Global-scale-background petroleum VOCs over ocean	- Two routes between Japan and Australia - One round-the-world route	- Mar 2003 - Jan-Mar 2004 - Jun-Aug 2004	- Sample collection (on-ship) - Sampling (Canister and Solid-absorption-thermal desorption) - Analyser (GC-MS)	- Due to storage period of the sample on-board before analyse	- **Petroleum** mentioned as source of 9 detected aromatic HCs	(Yagi et al., 2004)

Table S1. Research studies on emission inventories (EIs) of petroleum industries (continued).

Aim(s)	Location	Time	Methods of Data Collection, EI and modelling	Uncertainty analysis or reasons	Crude oil related sources	Reference
- Air VOC concentrations at petroleum refinery	- Kaohsiung Refinery, Taiwan	- 28 April to 4 - May 2001	- Sample collection (26 points of sampling; total 52 VOC samples totally per day & night) - Sampling (Silicon lining canister) - Analyser #1 (GC-FID) - Analyser #2 (UV-DOAS system)	- NA	- Production processes (incl. **Crude oil**) - Storage tanks (incl. **Crude oil**) - Transport pipelines (incl. **Crude oil)** Waste areas	(Lin et al., 2004)
- Air VOCs at an oil refinery	- NA	Eight hours	- Sample collection (12 points of sampling) - Sampling (Vacuum pumped using carbon adsorbent (activated charcoal)) - Analyser (GC-MS) - Metrological data (wind speed)	- NA	- **Crude distillation unit** - Vacuum distillation unit - Catalytic reforming unit - New delayed cocker unit - LPG recovery unit - LPG dispatch unit - Wax hydrofinishing unit - Solvent dewaxing unit (SDU).	(Pandya et al., 2006)
- EI for 60 organic liquid storage tanks located in City of Dar-es-Salaam - Health risk assessment	- Dar-es-Salaam city, Tanzania	- 12 months	- Metrological data (wind speed, wind direction, temperature) - Model (CALPUFF model using the U.S. EPA standard regulatory storage tanks emission model (TANKS 4.09b)	- NA	- Petrol - Diesel, - **Crude oil** - Residue oils.	(Jackson, 2006)
- Contribution of VOCs from refineries to Nigeria's air quality	- Nigeria	- 12 months	- Data of emission from Department of Petroleum Resources (DPR) of Nigeria - Data of human population and land mass from National Population Commission (NPC) of Nigeria - Emission factor approach	- NA	- **Crude oil** processes in refinery incl. storage, pipelines, transport, etc.	(Sonibare et al., 2007)
- Experimental determination of main sources of VOCs in China	- The Pearl River Delta (PRD), China	- 2004–2005	- Sample collection (3 points of sampling; total 7 VOC samples) ONLY at refineries - Sampling (Canister) - Analyser (GC-MS & GC-FID) - Metrological data (sample collections in upwind/downwind areas)	- ONLY mentioned: "the uncertainty may be high due to the limited number of vehicles tested for exhaust VOCs"	- Two of the 10 refineries in China with **Crude oil** processing capacity at the 10 million ton level are located in the PRD.	(Liu et al., 2008)
- In-situ investigation of VOCs in comparison with TRAMP field campaign in industrial Houston Ship Channel	- Houston, USA	- Aug-Sep 2006	- Data of emission from TexAQS-II Radical and Aerosol Measurement Project (TRAMP) - Receptor sample collection (continuous online measurement on Moody Tower (MT) at the University of Houston) - Source sample collection on GC-auto sites (A6 points of sampling; total 209 VOC samples) - Sampling (glass-lined stainless steel tube with Peltier-cooled cold trap) - Receptor model (positive matrix factorization (PMF)) - Analyser (GC-FID) - Model (EPA PMF 1.1 receptor model based on multilinear engine ME-2)	- Bootstrap technique with 200 runs for PMF model's uncertainties	- Natural gas/**Crude oil** contributed about 27.4% to the VOC mass	(Leuchner and Rappenglück, 2010)

Table S1. Research studies on emission inventories (EIs) of petroleum industries (continued).

Aim(s)	Location	Time	Methods of Data Collection, EI and modelling	Uncertainty analysis or reasons	Crude oil related sources	Reference
- VOC EI of Alberta oil sands mining operations	- Alberta's oil sands deposits in the Athabasca, Peace River and Cold Lake regions	- July 2008	- Data from the 2008 Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) - Sampling as whole air sampling (WAS) (2-L electropolished, conditioned stainless canisters) - Analyser (GC-FID)	- NA	- **Crude oil** reserves in Canada's oil sands deposits are 2nd worldwide after Saudi Arabia.	(Simpson et al., 2010)
- BTEX EI of crude oil refinery in Baltic States, Lithuania	- Mažeikiai region, Lithuania	- 2 weeks in Nov 2009 - 2 weeks in Feb 2010	- Sample collection (13 points of sampling) - Sampling (Passam AG diffusion tubes using activated charcoal) - Analyser (GC) - Metrological data (wind direction, temperature, humidity, atmospheric pressure and wind speed)	- NA	- A very large **Crude oil** refinery (200,000 bbl/day) was major source of BTEX	(Baltrėnas et al., 2011)
- EI from anthropogenic sources of YRD China	- Yangtze River Delta (YRD) region in China	- 12 months (2007)	- Data from previous works and international report - Emission factor approach	- Bottom up approach	- **Crude oil** process at refineries considered as one of non-combustion industrial sources	(Huang et al., 2011)
- VOC emission form oil and gas operations	- Northeastern Colorado, USA	- 7-9 Sep 2010 - 18 Feb-7 Mar 2011 - Aug-Sep 2006 (shipborne measurement)	- Data (for comparison) from TexAQS/GoMACCS 2006 and CalNex 2010 - Sampling/Analyser (in-situ custom-built two-channel GC-MS)	- NA	- More than 15,000 active **Crude oil** and natural gas wells were in Northeastern Colorado	(Gilman et al., 2013)
- VOC emission form petroleum refinery in China (incl. inner device sources area as well)	- Yanshan petroleum refinery, Beijing, China	- Jun-Sep 2011	- Sample collection (8 points of sampling; total 258 sample) - Sampling (vacuum summa canister) - Analyser (GC-MS/GC-FID) - Metrological data (wind direction/speed, temperature) - Model (Positive Matrix Factorization (PMF))	- The determination of uncertainty was done based on Norris and Vedantham (2008)	- The refinery refines 10 million tons of **Crude oil** per annum.	(Wei et al., 2014) (see also (Wei et al., 2016))
- PTR-MS analysis of VOCs after Deepwater Horizon (DWH)	- Mexican Gulf	- April 2010	- Data (for comparison) from info. collected on board by NOAA WP-3D aircraft equipped with PTR-MS in 2010 - Analyser (GC–PTR-MS)	-	- 19 **Crude oil** samples (extra-light, light, light-medium, medium, medium-heavy, heavy and extra-heavy) were used.	(Yuan et al., 2014)
- VOCs from oil and gas well	- Uintah Basin in Utah, USA	- 15 Jan.- 29 Feb. 2012	- Data (from a part of a broader study called UBWOS2012) incl. NOAA PTR-MS & ULW-PTR-MS (ground/on-board) installed on Horse Pool, Utah - Analyser (online GC-MS)	-	- The Uintah Basin in Utah had about 2000 **Crude oil** wells are in operation.	(Warneke et al., 2014) (see also (Helmig et al., 2014))
- Organic carbon emissions in the San Joaquin Valley from petroleum/dairy industries	- San Joaquin Valley, California, USA	- 18 May- 30 Jun. 2010	- Data from CalNex project from the National Oceanic and Atmospheric Administration (NOAA) WD-P3 aircraft - Sample collection (at 18 m local tower and ground level collection) - Sampling (gas-sampling canisters) - Models (source receptor model/FLEXPART-WRF/HYSPLIT/ FLEXPART)	- Reduced by average data and standard errors (US EPA CMB 8.2 model)	- **Crude oil** production in San Joaquin Valley, was 450 000 barrels per day−1 about 8% of US production	(Gentner et al., 2014)

Table S1. Research studies on emission inventories (EIs) of petroleum industries (continued).

Aim(s)	Location	Time	Methods of Data Collection, EI and modelling	Uncertainty analysis or reasons	Crude oil related sources	Reference
- Methane EI in Alberta upstream oil and gas sector	- Lloydminster and Red Deer. Alberta, Canada	- 27 Oct.-5 Nov 2016	- Data from AER Products and Services Catalogue (containing 312 654 useable samples associated with 117 206 well segments) -	- Reduced by average data and standard errors	- The Province of Alberta is Canada's largest producer of fossil fuel resources congaing 80% of all **Crude oil** and equivalent production	(Johnson et al., 2017)
- PTR-ToF-MS VOC EI over oil and gas areas in the USA	- USA (Bakken, Upper Green River, Uintah, Denver–Julesburg, San Juan , Permian, Barnett, Eagle Ford, and aynesville)	- Mar. & Apr. 2015	- Data from Shale Oil and Natural Gas Nexus (SONGNEX) campaign detected by OAA WP-3D Orion research aircraft and Ionicon PTR-ToF-MS instrument in the Uintah Basin, Utah, oil and gas field (for extensive description of instrumentation please refer to ref.)	-	- The Uintah Basin in Utah had about 2000 **Crude oil** wells are in operation.	(Koss et al., 2017)
- VOC EI of a refinery in Pearl River Delta of China	- Huangpu district of Guangzhou city, China	- 5-15 Aug 2014	- Sample collection (35 points of sampling; total 54 sample) - Sampling (3.2 L silica vacuum SUMMA - canisters with flow-limiting valves) - Analyser (GC-MS/GC-FID) - SOA formation estimation (FAC method/SOAP method/ SOA yield method)	- Monte Carlo simulation using the Crystal Ball software (v11.1.2.4)	- The refinery has a capacity of refining 13.2 million tons of **Crude oil** per annum	(Zhang et al., 2017) (see also (Zhang et al., 2018))
- VOC long-term monitoring over oil and gas areas in China	- Northwestern margin of the Junggar Basin. China	- Sep – Aug 2015	- Sample collection (continuous sampling from top of 15 m building) - Analyser (GC-FID) - Metrological data (atmospheric pressure, temperature, relative humidity, wind speed and direction) - VOC Geographic origins (Conditional probability function, Backward trajectory analysis, Local and regional transport contribution)	- Bootstrap run (BS)	- The proven reservoir of crude oil in this area is about 2.41×10^9 t	(Huang et al., 2018)
- VOC source appointment of a heavy oil production in Canada	- Alberta, Canada	- 12 months	- Sample collection (2 monitoring stations and total 124 samples) - Sampling (fully inert lined SilcoCanTM or SiloniteTM canisters) - Analyser (GC-MS/GC-FID) - Model (Positive Matrix Factorization (PMF)) -	- Bootstrap run (BS)	- The Peace River Oil Sands area of Canada includes 21% of Alberta's heavy oil sands.	(Aklilu et al., 2018)

Table S2. The CVEs detected and reported in the literature.

VOCs emitted from crude oil	**ppmv**	**ppb**	**µg/m^3**	**ppbv**	**ppb**	**wt.%**	**ppbv**	**ppb**	**pptv**	**mg/kg**	**ppb**	**ppmv**	**mg/m^3**	**(TaqTaq crude oil) mg/kg**	**(Khurmala crude oil) mg/kg**	**mg/kg**	**ppm**	**ppbv**	**wt.%**	**mg/m^3**
Acetone									644											
Benzene	4.9	3188	0.034	0.03	29.27	17.5	15790	1970	24	2290	1595	197.33	0.24				69	1.13		249
Chlorobenzene			0.008																	
m-Dichlorobenzene			0.009																	
o-Dichlorobenzene			0.017																	
p-Dichlorobenzene			0.056																	
1,2,4-Trichlorobenzene			0.041																	
m-Diethylbenzene													0.06					0.07	6.8	
p-Diethylbenzene													0.05					0.1	9.3	
Ethylbenzene	0.4	401	0.006	0.04	5.72	1.7	3620	71.9	8	1140			0.82				11	0.3		
Ethenylbenzene (Styrene)			0.007	0.06			1735						0.26					0.4		
Isopropylbenzene (Cumene)		29				0.3		9.87		242	17							0.06		
Propylbenzene		32		0.04				15.4					0.04						4	
n-Propylbenzene									2	471	21		0.12					0.05		
1,2,3-Trimethylbenzene									5				0.19					0.09		
1,2,4-Trimethylbenzene			0.004	10.62		1.6	2362	38.3	6	1430			0.19				7	0.14	5.6	32
1,3,5-Trimethylbenzene (Mesitylene)			0.003	10.56				19	1	537	28		0.13				5	0.09	3.2	18
Butane				0.19					202			6709.15	0.59					15.8		
1-Butene				0.12					15				0.89					0.63		
Isobutane				0.28					89			5118.77	0.52							
Trans-2-Butene									3				1.28					0.6		
Cis-2-Butene													0.9					0.7		
2,2-Dimethylbutane				0.05								152.5	0.31					0.2	3.3	
2,3-Dimethylbutane				0.05					5	124			0.14					1.81	4.7	
Reference	**(Hanna and Drivas, 1993)**	**(Saeed and Al-Mutairi, 2000)**	**(Yagi et al., 2004)**	**(Lin et al., 2004)**	**(Pandya et al., 2006)**	**(Jackson, 2006)**	**(Liu et al., 2008)**	**(Yang et al., 2007)**	**(Simpson et al., 2010)**	**(Uhler et al., 2010)**	**(Saeed et al., 2013)**	**(Khoramfar et al., 2018)**	**(Zhang et al., 2017)**	**(Salih et al., 2018)**		**(Mustafa Salih et al., 2018)**	**(Aklilu et al., 2018)**	**(Afshar-Mohajer et al., 2018)**	**(Huang et al., 2018)**	**(Zhang et al., 2018)**

Table S2. The CVEs detected and reported in the literature.

VOCs emitted from crude oil	**ppmv**	**ppb**	**µg/m^3**	**ppbv**	**ppb**	**wt.%**	**ppbv**	**ppb**	**pptv**	**mg/kg**	**ppb**	**ppmv**	**mg/m^3**	**(TaqTaq crude oil) mg/kg**	**(Khurmala crude oil) mg/kg**	**mg/kg**	**ppm**	**ppbv**	**wt.%**	**mg/m^3**
2-Methylbutane												4189.04								
Decane	0.42									3590			0.02					0.23	3	
Dodecane										957			1.72					39.7		
Ethane				0.45					917				24.58					39.7		
Ethene									69				3.94							
Ethylene				0.43														1.42		
Ethyne				0.29					74				0.8							
Heptane	4.4			0.04					70	4050		649.84	0.05	11	8.8	4.2	158	4.07		656
2-methylheptane				0.04						1580		162.38	0.03					0.87		
3-methylheptane				0.03								116.67	0.08					0.2		
Hexane	1.3			0.06		20.6			44	4790		1553.86	0.54	11.7	10.6	8.79	231	3.08		811
1-Hexane																		1.36		
Cyclohexane		287		0.1		5.2		2117	23	3970			0.08				167	1.23		585
2,4-Dimethylhexane										209		80.75								
1,1-Dimethylcyclohexane												54.74								
cis-1,3-Dimethylcyclohexane												215.04								
Trans-1,2-Dimethycyclohexane												111.36								
Ethylcyclohexane																				
Methylcyclohexane		56						2241	52	6580	165	896.03	0.1					1.43		
Ethyl, methylcyclohexane		12																		
2-Methylhexane				0.72						1300		805.47	0.07					0.57		
3-Methylhexane				0.03						1620		550.81	0.08					0.7		
1,1,3-Trimethylcyclohexane												63.9								
Isoprene									468				0.78					0.2		
Reference	**(Hanna and Drivas, 1993)**	**(Saeed and Al-Mutairi, 2000)**	**(Yagi et al., 2004)**	**(Lin et al., 2004)**	**(Pandya et al., 2006)**	**(Jackson, 2006)**	**(Liu et al., 2008)**	**(Yang et al., 2007)**	**(Simpson et al., 2010)**	**(Uhler et al., 2010)**	**(Saeed et al., 2013)**	**(Khoramfar et al., 2018)**	**(Zhang et al., 2017)**	**(Salih et al., 2018)**		**(Mustafa Salih et al., 2018)**	**(Aklilu et al., 2018)**	**(Afshar-Mohajer et al., 2018)**	**(Huang et al., 2018)**	**(Zhang et al., 2018)**

Table S2. The CVEs detected and reported in the literature.

VOCs emitted from crude oil	**ppmv**	**ppb**	**µg/m^3**	**ppbv**	**ppb**	**wt.%**	**ppbv**	**ppb**	**pptv**	**mg/kg**	**ppb**	**ppmv**	**mg/m^3**	**(TaqTaq crude oil) mg/kg**	**(Khurmala crude oil) mg/kg**	**mg/kg**	**ppm**	**ppbv**	**wt.%**	**mg/m^3**
Nonane	1.4			0.06					13	4040		110.92	0.07					0.29		
Octane	4	145		0.05		19.6			45	3680		218.27	0.04					0.97		
Pentane									116			4233.2	0.86	2.9	2.3	5.35		8.81		
1-pentene				0.08						5480			0.51					4.47		
Cyclopentane				0.08					9	854		435.17	0.82					8.64		
Cis-2-pentene				0.06														0.09		
isopentane				0.08					141	3690			0.83							
Trans-2-pentene				0.06														0.19		
2-methylpentane				0.05					57	2750		1497.54	0.13					3.36	8.1	
3-methylpentane				0.25						1820		984.85	0.37	2.9	2.8	2.71		1.4		
2,2-dimethylpentane										3630										
2,3-dimethylpentane				0.07						548		223.08	0.07					0.85		
2,4-dimethylpentane													0.02					0.13		
methylcyclopentane		349		0.04					31	101		534.43	0.28	1.1	1.3	3.65		1.28		
1,2-Dimethylcyclopentane												250.48								
1,3-Dimethylcyclopentane		27										213.93								
trans-1,3-Dimethylcyclopentane												176.04								
2,2,4-Trimethylpentane				0.04									0.03					0.05		
2,3,4-Trimethylpentane				0.04									1.6					0.06		
1,2,3-Trimethylcyclopentane												74.68								
1,2,4-Trimethylcyclopentane												98.57								
Propane				0.5					382			1667.51	0.52					22.6		
2-methylpropane				0.07																
Propene				0.36					28											
Reference	**(Hanna and Drivas, 1993)**	**(Saeed and Al-Mutairi, 2000)**	**(Yagi et al., 2004)**	**(Lin et al., 2004)**	**(Pandya et al., 2006)**	**(Jackson, 2006)**	**(Liu et al., 2008)**	**(Yang et al., 2007)**	**(Simpson et al., 2010)**	**(Uhler et al., 2010)**	**(Saeed et al., 2013)**	**(Khoramfar et al., 2018)**	**(Zhang et al., 2017)**	**(Salih et al., 2018)**		**(Mustafa Salih et al., 2018)**	**(Aklilu et al., 2018)**	**(Afshar-Mohajer et al., 2018)**	**(Huang et al., 2018)**	**(Zhang et al., 2018)**

Table S2. The CVEs detected and reported in the literature.

VOCs emitted from crude oil	**ppmv**	**ppb**	**µg/m^3**	**ppbv**	**ppb**	**wt.%**	**ppbv**	**ppb**	**pptv**	**mg/kg**	**ppb**	**ppmv**	**mg/m^3**	**(TaqTaq crude oil) mg/kg**	**(Khurmala crude oil) mg/kg**	**mg/kg**	**ppm**	**ppbv**	**wt.%**	**mg/m^3**
Propylene																		1.88		
Toluene	8.2	3086	0.014	0.02	72.54	24.6	13120	1097	50	5160	1564	282.37	0.08				121	1.06		452
2-Ethyltoluene								12.2												
4-Ethyltoluene			0.002					36.7												
p-Ethyltoluene									2				0.05					0.08		
m-Ethyltoluene									4				0.08					0.09		
o-ethyltoluene									2				0.08					0.07	5.7	
Undecane										2160			0.13					0.19		
m,p-Xylene	1.5	1014	0.005	0.17	15	8.8	8630	271	29	4620	311	87.43	1.34				53	0.72		252
o-Xylene	0.45	635	0.007	0.04				74.1	14	1460	359		0.6				17	0.2		69
Reference	**(Hanna and Drivas, 1993)**	**(Saeed and Al-Mutairi, 2000)**	**(Yagi et al., 2004)**	**(Lin et al., 2004)**	**(Pandya et al., 2006)**	**(Jackson, 2006)**	**(Liu et al., 2008)**	**(Yang et al., 2007)**	**(Simpson et al., 2010)**	**(Uhler et al., 2010)**	**(Saeed et al., 2013)**	**(Khoramfar et al., 2018)**	**(Zhang et al., 2017)**	**(Salih et al., 2018)**		**(Mustafa Salih et al., 2018)**	**(Aklilu et al., 2018)**	**(Afshar-Mohajer et al., 2018)**	**(Huang et al., 2018)**	**(Zhang et al., 2018)**

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