The Red Sea Deep Water is a potent source of atmospheric ethane and propane

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Non-methane hydrocarbons (NMHCs) such as ethane and propane are significant atmospheric pollutants and precursors of tropospheric ozone, while the Middle East is a global emission hotspot due to extensive oil and gas production. Here we compare in situ hydrocarbon measurements, performed around the Arabian Peninsula, with global model simulations that include current emission inventories (EDGAR) and state-of-the-art atmospheric circulation and chemistry mechanisms (EMAC model). While measurements of high mixing ratios over the Arabian Gulf are adequately simulated, strong underprediction by the model was found over the northern Red Sea. By examining the individual sources in the model and by utilizing air mass back-trajectory investigations and Positive Matrix Factorization (PMF) analysis, we deduce that Red Sea Deep Water (RSDW) is an unexpected, potent source of atmospheric NMHCs. This overlooked underwater source is comparable with total anthropogenic emissions from entire Middle Eastern countries, and significantly impacts the regional atmospheric chemistry.
The Middle East accommodates more than half of the world’s known oil and gas reserves. Fossil fuel exploitation in this region is responsible for the release of large amounts of gaseous pollutants into the atmosphere, including methane (CH$_4$) and non-methane hydrocarbons (NMHCs). Ethane and propane have the strongest sources, and being relatively long-lived (ethane ca. 2 months, propane ca. 2 weeks) are ubiquitous in the global atmosphere. Atmospheric oxidation of NMHCs in the presence of nitrogen oxides (NO$_x$) leads to production of tropospheric ozone and peroxyacetyl nitrates (PAN) that are phytotoxic and harmful to human health. The abundance of NMHCs and NO$_x$ in combination with the intense photochemistry in the Arabian Basin results in extremely high ozone mixing ratios that can reach up to 200 ppb.

Globaly, the atmospheric concentrations of ethane and propane exhibit temporal trends that are closely related to anthropogenic activities. The general decline in fossil fuel emissions toward the end of the twentieth century resulted in a decline of global atmospheric ethane and propane. Conversely, the subsequent expansion of US oil and natural gas production has led to a reversal of their global atmospheric trends, with emissions increasing since 2010.

While anthropogenic activities substantially influence the emission rate and composition of atmospheric hydrocarbons, Earth’s natural degassing is also a significant source. Natural geologic (i.e., mud volcanoes, onshore and marine seeps, and micro seepage, geothermal and volcanic) sources contribute to both ambient ethane and propane concentrations, and their inclusion in global emission inventories helps to better explain the reported values from the expanding global observation network. Indeed such sources will have dominated preindustrial emissions.

During the AQUABA ship campaign, which took place between July and August 2017, NMHCs were monitored around the Arabian Peninsula (Supplementary Fig. 1). By comparing the observations with model simulations, we aim to evaluate the emission inventories and atmospheric chemistry mechanisms while focusing on the most abundant anthropogenic hydrocarbons: ethane and propane. The largest measurement/model discrepancy was observed over the northern part of the Red Sea, which was investigated in terms of possible underestimation of existing sources and emission patterns (i.e., ratios between the measured hydrocarbons) that are derived by using positive matrix factorization analysis.

The observed ethane mixing ratios were reproduced by the model for most of the route (Fig. 1), indicating that emission sources and atmospheric processes in the Middle East region are generally well understood. Significant model underestimations (Suez Canal, northern Arabian Gulf) and overestimations (Gulf of Oman) occurred for short periods only during the first leg of the route, suggesting local, small-scale inconsistencies in the emission sources. The only region that was inadequately simulated during both legs of the route was the northern part of the Red Sea where measured mixing ratios of ethane and propane were up to about 20 (average ± standard deviation = 4.3 ± 3.8) and 40 (average ± standard deviation = 7.8 ± 5.9) times higher than model predictions. According to the model results, biomass burning, fuel production and transmission, and transformation industry emissions regulate the regional hydrocarbon abundance (Fig. 1d; Supplementary Fig. 5). However, neither the dominant nor any of the 15 inventory sources was able to explain the observations, even when the emission strength was varied (Supplementary Fig. 6).

Source apportionment. To derive the hydrocarbon signature of the potent unidentified source, a well-established receptor model (positive matrix factorization; US EPA PMF 5.0) was utilized. Receptor models use ambient observations to apportion the observed species concentrations to signature sources by assessing changes in species correlation with time and finding the optimum solution that explains the concentrations of all observed constituents.

The PMF analysis of the northern Red Sea data identified 4 distinct emission sources/factors (Fig. 2a). Factor 1 illustrates an emission source that is rich in C2−C6 hydrocarbons, alkene and acetonitrile (a biomass-burning tracer), with contributions mainly during the first leg when the air masses originate from the Suez Canal (Supplementary Fig. 7). In addition, it correlates with anthropogenic activity markers such as acetone, methanol, and acetaldehyde (Supplementary Fig. 8), confirming the urban character of this emission source. By contrast, factor 4 was significant only during the second leg (Supplementary Fig. 9) when the air originated from the Sinai Peninsula. The small concentration contribution and back-trajectory-specific direction suggest a background signature from a region with small emission sources. Factor 3 apportions marine traffic emissions that are clearly distinguishable from the sporadic occurrences (Supplementary Fig. 10), high ethene, large alkanes (>C4), and the absence of ethane in the emission pattern.

The remaining factor 2 is characterized by exceptionally high alkane concentrations that decrease with increasing carbon number. Alkene contribution is negligible, and in combination with the absence of acetonitrile in the emission signature, factor 2 points toward a non-anthropogenic emission source. The source contribution to the measured signal is expressed by the significance of the factor that is termed as factor strength. As shown in Fig. 2b, this source contributed most to the measurements over the northmost part of Red Sea and in particular between 23° and 27° latitude. Since the model underestimation (expressed by the ratio between measurements and models) increases with the strength of factor 2 (Fig. 2c), it becomes evident that it represents the missing source.

Summarizing, the high ethane and propane mixing ratios that were observed over the northern part of Red Sea could not be...
Distinct movements of the tectonic plates have led to a partly fractured sea floor and the formation of numerous brine-filled pools that are characterized by close-to-solubility limit halite (mineral form of NaCl) concentrations and strong temperature gradients. Generally, the water occupying depths from 300 to 2000 m in the Red Sea is recognized as the warmest and saltiest deep water in the world with pronounced seasonality, although the rates and mechanisms of its renewal remain uncertain.

Hydrocarbon release from the Red Sea floor can occur through direct fluid seepage from hydrocarbon reservoirs deposited above offshore rocks, located between 25 and 28°N (e.g., Rudeis and Kairen formation). In addition, the Gulf of Suez and Aqaba contribute to the RSDW through bottom-trapped density outflows. Considering that this region is known for the large oil and gas reserves, natural seepage and crude oil/gas seepage from leaky subsea wells could be a significant submarine source of NMHCs in this region. Finally, the numerous brine pools that are located on the sea floor need to be considered. Depending on their chemical composition, the brine pools are classified into two distinct types. The formation of Type I brine pools is controlled by evaporative dissolution and sediment alteration, characterized by exceptionally high methane and hydrocarbon concentrations. Short-chained hydrocarbons are formed by the degradation of long-chained hydrocarbons that originate from the organic-rich sedimentary rocks and the bioproduction in the brine’s water to sediment interface. Type II brine pools are controlled by volcanic/magmatic alterations and are poor in organic material. They are more common and are frequently found in the southern and middle part of the Red Sea floor. In contrast, only two brine pools are classified as type I: the Oceanographer deep (26°17.2’N, 35°01.0’E) and the Kebrat deep (24°43.1’N, 36°17.0’E). Oceanographer in particular is known to contain high methane...
The convective mixing can be especially deep and reaches the sea relatively weak with values up to 393 kg/yr for the Kebrit deep salinity gradient, and the reported methane emissions are brine pools and RSDW occurs via diffusion across the strong region with the highest discrepancy between NMHC measurements and models for ethane and propane, and in with the measured methane. Further explanation on the source assignment for each factor is shown in Supplementary Figs. S8-10.

concentrations (4921 μl/l39 (average methane inside type II brines is 0.04 ± 0.04 μl/l))31 and it is located directly underneath the region with the highest discrepancy between NMHC measurements and model simulations. The mixing between the dense brine pools and RSDW occurs via diffusion across the strong salinity gradient, and the reported methane emissions are relatively weak with values up to 393 kg/yr for the Kebrit deep. However, the impact of above-brine water currents that may enhance the fluxes has not been quantified, so this emission estimate remains uncertain.

Considering the aforementioned potential sources, it seems entirely plausible that the RSDW is highly enriched in ethane and propane from a deep sea source. The transfer from deep water to the surface can be relatively rapid due to the exceptionally effective vertical mixing of the Red Sea deep water and the outflows from the Gulfs of Suez and Aqaba that have been shown in Supplementary Figs. S8, S9. Model estimates of the renewal times range from 19 to 90 years, while tracer studies indicate somewhat faster renewal times to about 26 years33,42. Further, mesoscale eddies are particularly effective in the areas of the Oceanographer and Kebrit deeps46 and may contribute to the vertical transport of hydrocarbons. Eddies may also affect deep-water environment with downward effective transfer rate of 200–600 m day−1 as measured in the Pacific Ocean47.

While the relative significance of the various submarine hydrocarbon sources cannot be ascertained, we assume that their cumulative contribution represents the missing source derived in this study. This assumption is supported by the similarity in the PMF-derived chemical emission profiles (increased alkane concentrations and the absence of alkenes and other anthropogenic tracers in the emission signature). Therefore, we surmise that methane and non-methane hydrocarbons can potentially reach the surface and degas into the atmosphere following air–sea exchange mechanisms.

**Flux calculations.** To test this hypothesis, two source points (over two model resolution grids (1.1 × 1.1°) with intensity 2:1 from north to south) were added to the model simulation as additional point sources from the ocean surface at the location of the type I brine pools. While Type I brine pools were chosen as the reference location, the emission points cover a large area and thereby include emissions from all aforementioned potential sources. Initially, approximate emission rates that match the factor 2 signature were imported. The measurement to model ratio output for NMHCs was substantially improved with median values deviating from unity by only ca. ±30%. Therefore, the emission rates were fine-tuned so that the measurement/model median ratio was
equal to 1, and hence the emission strength of the RSDW could be ascertained. This resulted in predicted emission rates for ethane \((0.12 \pm 0.06 \text{ Tg yr}^{-1})\), propane \((0.12 \pm 0.06 \text{ Tg yr}^{-1})\), i-butane \(= 0.03 \pm 0.03 \text{ Tg yr}^{-1}\), and n-butane \(= 0.07 \pm 0.06 \text{ Tg yr}^{-1}\) (uncertainties are based on the standard deviation of the measurement/model ratio values within the 25th and 75th percentile). Increasing uncertainties will be introduced when considering the seasonality of the deep-water circulation, the wind speeds at the air–sea interface, and the potential temporal variability of the emissions (i.e., triggered events by the increased seismic activity in the region\(^{48}\)). As a final step, the derived emission rates were added to the model and the simulations were repeated. The inclusion of the RSDW emissions in the emission inventory significantly improves the model–measurement comparison, making it equivalent to the agreement seen elsewhere on the route (Fig. 3; Supplementary Fig. 12, Supplementary Fig. 6q). The uncertainties here are likely associated with water current circulation with depth and the exact location of the degassing points. Furthermore, the seabed emissions are likely higher than those reported due to oxidation and bacterial degraders in the water column\(^{49,50}\).

Considering the linearity between the measured ethane and methane mixing ratios (Supplementary Fig. 13) and by assuming common source origin, an emission rate of ca. 1.3 Tg CH\(_4\) yr\(^{-1}\) is derived. While this rate is only a small fraction of the global natural methane sources \((238–484 \text{ Tg CH}_4 \text{ yr}^{-1})\)\(^{51}\), it is responsible for the high ambient methane mixing ratios observed over the northern Red Sea (average \(= 1.94 \pm 0.03 \text{ ppm}\)).

Implications. The degassing rates for ethane and propane derived here are considerable and comparable in magnitude with the
emissions from several Middle Eastern countries, known to be exceptional sources related to the hydrocarbon industry (Fig. 4). The combined ethane and propane emission rates rival those from countries, with the most intense oil and gas activities, such as the United Arab Emirates (UAE), Kuwait, and Oman. In addition, the associated methane emissions from the RSDW are a hitherto unaccounted source of atmospheric methane. Considering the seasonality of the deep-water circulation, it is likely that the emissions to the atmosphere will be further enhanced during the wintertime.

While in much of the Middle East NOx abundance is a rate-limiting factor in oxidant photochemistry,12 it could be expected that over the Red Sea NMHCs are rate limiting due to small upwind anthropogenic sources. To evaluate the implications of the newly discovered NMHC emission source for atmospheric chemistry, the differences in key atmospheric constituents (hydroxyl radical (OH), ozone (O3), and PAN) were investigated for the entire area of Northern Red Sea using the model with and without the inclusion of the RSDW sources of ethane, propane, and butanes. Interestingly, summertime average OH depletion is significant over the degassing spots (≈−40%; max = −70%; Supplementary Fig. 14). Downwind of the source location, summertime average ozone production is somewhat enhanced (up to 11%; Supplementary Fig. 15) while there is a prodigious increase in PAN abundance (≈+102%; max = 750%; Supplementary Fig. 16). This represents a significant deterioration of regional air quality, as PAN, a lachrymator and urban smog component, is harmful to human health32,53, and directly related to ethane concentrations34.

In the coming decades, ship traffic through the Red Sea and Suez Canal is projected to increase strongly55, with a concomitant rise in NOx emissions. From the increase of NOx emissions in the model (comparing with and without the RSDW emissions) it is expected that the degassing hydrocarbons will amplify ozone formation in the future (Fig. 5). The photochemical pollution from anthropogenic NOx and degassing NMHCs from RSDW will directly affect air quality, for example in Neom city, a cross-border megaproject in the Tabuk Province of north-western Saudi Arabia56.

**Methods**

**The AQABA ship campaign.** To study the Air Quality and climate in the Arabian Basin (AQABA), a ship expedition was conducted in July and August 2017. The research vessel Kommandor Iona (IMO: 8401999, flag: UK, length overall x breadth extreme: 72.55 m x 14.9 m) was equipped with five air-conditioned laboratory containers that hosted a large suite of atmospheric gas and aerosol measurement equipment. The ship sailed from Toulon (France), crossed the Mediterranean Sea, and through the Suez Canal covered the periphery of the Arabian Peninsula to Kuwait and back. In total, 20,000 km of the marine route was covered with an average speed of 3.4 ± 1.8 m s−1 over the course of 60 days. Further information on the AQABA ship campaign can be found elsewhere in the literature46,57.

**Non-methane hydrocarbon measurements.** Non-methane hydrocarbons (C2–C8) were measured in situ with two coupled, commercial gas chromatography-flame ionization detectors (GC-FID; AMA Instruments GmbH, Germany). Detailed information on the instrumentation, experimental setup, sampling, and calibrations can be found elsewhere55. Briefly, atmospheric samples were collected through a common 5.5-m tall (3 m above the container), 0.2-m-diameter, high-flow (≈10 m3 min−1) stack with a subflow of 2.5 m3 min−1. The air passed through a PTFE filter (5-μm pore size, Sartorius Corporate Administration GmbH, Germany) and heated (40 °C) Teflon lines before it was drawn into the instruments with a flow of 90 scm (2 x 45 cm2 (stp) (scm)) (scm). An ozone scrubber (Na2S2O5-impregnated quartz filters) and a NaCl filter dryer (500-sccm counterflow) were used to eliminate the effects of ozone and humidity in sample collection. The sampling times and volumes were adjusted according to ambient NMHC concentrations and wave conditions. During polluted conditions (e.g., Arabian Gulf and the Suez Canal) short sampling times (10 min) and small volumes (450 mL) allowed higher time resolution (50 min per measurement), while under clean conditions, such as those found in the Arabian Sea, longer sampling times and volumes (30 min, 1350 mL, time resolution = 1 h) improved detection limits. For most of the route, the sampling time was 20 min, the sampling volume 900 mL, and the time resolution 50 min per measurement. The diverse conditions met during the campaign led to the geographical demarcation that was used during data analysis (Supplementary Fig. 1).

**Model simulations.** In this work the EMAC (ECHAM5/MESSy Atmospheric Chemistry) model has been used. The EMAC model is a numerical chemistry and climate simulation system that includes submodels describing tropospheric and middle atmosphere processes and their interaction with oceans, land, and human effects35. It uses the second version of the Modular Earth Submodel System (MESSy)34 to link multi-institutional computer code. The core atmospheric model is the fifth-generation European Centre Hamburg general circulation model (ECHAM56,58. For this study, we applied EMAC (ECHAM5 version 5.3.02, MESSy version 2.53.0) in the T106L31 resolution, i.e., with a spherical truncation of T106 and 31 vertical hybrid pressure levels up to 10 hPa. The simulations cover the period of the AQABA field campaign, i.e., from June to September 2017. The dynamics were weakly nudged by Newtonian relaxation toward ERA-Interim reanalysis data69. The model configuration of the chemical mechanism is similar to that of Lelieveld et al.60. The comprehensive Organic Mechanism (MaInz Organic Mechanism)61. NMHC chemistry representation has been used. Biomass-burning and anthropogenic emissions were prescribed based on the global fire assimilation system62 and EDGAR, v4.3.2,20,21 database, respectively. Furthermore, the emissions of ethane were subdivided into different source sectors as shown in Supplementary Figs. 2, 5, and 6. The ethane, propane, n-butanes, and i-butane emissions were scaled by factors of 1.9, 1.7, 1.0, and 0.43, respectively, to match recent global emission estimates in the literature6. Geothermal sources in the region14 were estimated by scaling sulfuric volcanic emissions to 0.2 Tg yr−1. All emissions were vertically distributed following the literature60. Gas flares were estimated based on the work of Caso et al.65.

**Positive matrix factorization.** PMF is a receptor model that uses an advanced multivariate factor analysis technique that is based on weighted least square regression using realistic error estimates to weight data values, and by imposing non-negativity constraints in the factor computational process. PMF is widely used to identify and quantify the main sources of atmospheric pollutants46,68. The mathematical background of PMF analysis is comprehensively described elsewhere65. Briefly, the statistical method uses a mass balance equation, which in the receptor model is expressed as

$$x_{ij} = \sum_{k} G_{ik} F_{kj} + E_{ij},$$

(1)

Here, $x_{ij}$ is the concentration of $j$ species measured in sample $i$ and $G_{ij}$ is the species contribution of the $k$ source to sample $i$. $F_{ij}$ (frequently reported as source profiles) is the fraction of $j$ species from the $k$ source, while $E_{ij}$ is a residual associated with the $j$ species concentration measured in the $i$ sample. Furthermore, $p$ is the total number of sources. The goal of the model is to reproduce $x_{ij}$ matrix by finding values for $G_{ij}$ and $F_{ij}$ matrices for a given $p$. The values of $G_{ij}$ and $F_{ij}$ matrices are adjusted until a minimum Q (the loss function)
for a given \( p \) is found\[^{29}\]. PMF solves the receptor modeling problem by minimizing the loss function \( Q \) based on the uncertainty of each observation by the following equation:

\[
Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left( \frac{e_{ij}}{p} \right)^2,
\]

where \( e_{ij} \) is an estimate of the uncertainty for the \( j \)th species in the \( i \) sample, \( n \) is the number of samples, and \( m \) is the number of species.

PMF application to volatile organic compound (VOC) source apportionment and profile contribution has been applied to a wide range of environments including urban and rural areas\[^{29-31}\]. A main advantage of PMF is that it can provide the source profile and contribution without any prior knowledge of VOC emission profiles. In this study, PMF was applied to the 50-min data samples of the AQABA campaign for identification and quantification of the major observed VOC sources, using the US EPA PMF 5.0 software\[^{32}\] (https://www.epa.gov/air-research).

Missing points were replaced with the median concentration of the corresponding species over the entire measurement matrix and they were accompanied by an uncertainty of 4 times the species-specific median, as suggested\[^{28}\]. It should be mentioned here that this is the first application of PMF to data from a moving platform (ship). This might introduce a small bias, despite the fact that the data were filtered for own ship exhaust. It should also be noted that the background was not removed from the measurements, since the background changes as the ship travels.

Since PMF is a weighted least-squares method, individual estimates of the uncertainty in each data value are necessary. The uncertainty input data matrix followed established approaches\[^{26,33}\] by including the measurement uncertainty of each sample and NMHC species\[^{16}\]. As a complementary criterion, a signal-to-noise condition was additionally applied in the data as suggested in the literature\[^{2}\]. Individual species that retained a significant signal were separated from those dominated by noise. When signal-to-noise (S/N) ratio was <0.2, the uncertainty of each sample and NMHC species\[^{16}\] was noted that factor 2 (Fig.1) NMHC signature remained relatively unchanged around the Arabian Peninsula.

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