POTENTIAL SOURCES OF MERCURY IN SOUTHERN PATTANI BASIN, THE GULF OF THAILAND

Ronnakrit Rattanasriampaipong*

Petroleum Geoscience Program, Department of Geology, Faculty of Science,
Chulalongkorn University, Bangkok, 10330, Thailand
*Corresponding author email: rattanasriampaipong.r@gmail.com

Abstract

Potential sources of mercury in producing oil and gas fields can be from either geogenic (hydrothermal fluid migration, coals and carbonaceous shales, etc.) or anthropogenic (water re-injection activities) sources. Due to lack of knowledge on mercury occurrence in the petroleum systems in SE Asia, no conclusive explanation has yet been made for the variable, sometimes elevated mercury concentrations measured production fluids in wells in the Pailin area, southern Pattani Basin, offshore Thailand. In prior work, petroleum engineers attempted to identify likely Hg contributing zones by conducting detailed investiga-tions using dedicated platforms with known perforated pay zones based on geological marker correlation. But none of engineering work considered the possibility of documentation of geological controls. This current research project is the first attempt to integrate conventional geological approaches (cuttings, petrography, isotopes, wireline and seismic) with engineering datasets (mercury surveillance data, gas composition, production data) in order to better interpret and evaluate potential sources of mercury in the Pailin operating area. Based on the series of geological evaluations applied in this integration, the following conclusions are drawn as to the origin of the anomalous Hg levels; (1) There is a poor correlation between levels of Hg and deeply-sourced CO_2 making a basement source less likely, (2) There is no obvious indication of hydrothermal fluid influence, based on the C-O isotopic signatures of various carbonate cements, specifically sampled from cuttings, (3) The low levels of reinjec-tion of produced water and a lack of correlation to this parameter across the area, means an anthropogenic cause for mercury-enriched hydrocarbons is unlikely, and (4) Intrabasinal geogenic sources, particularly from mature marginal marine coals and carbonaceous shales characterized by elevated levels of uranium, as indicated in spectral gamma ray log ("High Gamma Zones"), are considered the most likely source of elevated Hg Levels. Mature generating zones of this interval are most common in Upper Sequence 2 in the Gulf of Thailand (GOT) stratigraphy.This study concludes that mercury and its compounds were either sequestered in the remains of coal-forming land plants or deposited in associated muddy marine-influenced swamp and transitional marine deposits, where organic matter accumulated in reducing environments. Then, as these rocks were buried and matured the mercury co-migrated with the expelled hydrocarbon from its source to its reservoirs.

Keywords: mercury (Hg), mercury occurrence, carbonate cements, High Gamma Zones, the Gulf of Thailand

1. Introduction

Elevated mercury contents in hydrocarbons have had no distinct explanation in the Gulf of Thailand. There is no quantitative understanding on what form of mercury is present, and what the source is. Several conceptual models explaining the occurrence of Hg in hydrocarbon have been utilized in several oil and gas fields, and depending on the model the mercury can be derived from either a geogenic or an anthropogenic source.

2. Origin of Mercury and Its Cycle

Magmatic processes define the main geological pathway moving mercury from deeper sources into external settings via either; (a) atmospheric Hg deposition; as long-distant transported gaseous Hg(0) (Bagnato et al., 2011; Pyle and Mather, 2003; Witt et al., 2008), or (b) secondary migration of Hg-bearing hydrothermal fluids charging into subsurface rock formations; especially in organic-rich formations like coals or black shales (Diehl, 2004) (see Figure 1).

Over time the atmospheric form can be either, (a) oxidized into a water-soluble form, known as ionic mercury (Hg(II)), which can then be removed from atmosphere and accumulate in surface waters and landscapes via precipitation (Lindqvist, 1991), or (b) be

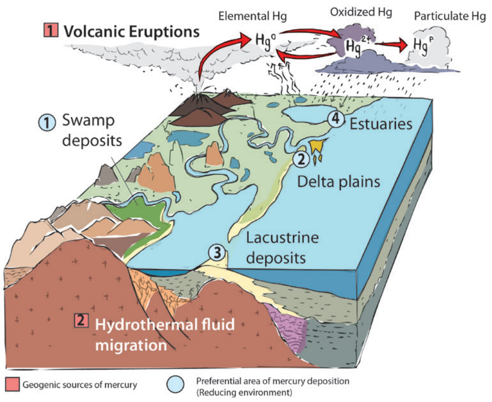

Figure 1. Geological origin of mercury and its deposits

adsorbed by clays as particulate Hg (HgS(P)) (Krupp, 1988). Additionally, the mobilized mercury can enter groundwater aquifers through multiple steps of biogeochemical transformation, whereby Hg-discharging coastal areas, especially muddy bays and estuaries, can be contaminated with elevated levels of dissolved Hg and MeHg (e.g., Bone et al, 2007; Black et al., 2009; Ganguli et al., 2012; Szymczycha et al., 2013).

Immobilization of mercury can occur in several ways, but requires specific conditions. Stable organic and inorganic Hg-rich complexes are chemically bonded with clay particles of either high organic or sulfur content, or with sediments containing elevated levels of humic and fulvic acids (Johansson et al., 1991; Yin et al. 1996, Font et al., 2016). Smith-Downey et al. (2010) notes that primary sequestration of gaseous Hg(0) in terrestrial plants occurs in the plant foliage stomata. Movable divalent mercury (Hg(II)) may also be mediated via biotic and abiotic factors to form methylmercury (MeHg) (Randall and Chattopadhyay, 2013). Microbial communities, especially those anoxic settings with elevated levels of sulfate-reducing bacteria (SRB), are thought to be primary agents for biotic methylation of mercury in estuarine settings (Barringer, et al., 2013, Ramond et al., 2011; Merritt and Amirbahman, 2009).

3. Overview of Global Mercury Deposits

Natural Hg-prone provinces typically occur along geological plate boundaries, especially in regions of increased hydrothermal activity or active magmatic extrusion, and in typically-anoxic sediment-hosted Hg-rich formations (Feng, 2003).

With ongoing petroleum exploration and production activities, progressively more hydrocarbon provinces with higher concentrations of mercury are encountered and developed (Boschee, 2013). Associated geological features frequently observed in Hg-rich producing hydrocarbon fields include; low hydrogen sulphides levels, high carbon dioxide levels, abnormally high geothermal gradients, coaly intervals, I-type tin granites, reservoir-in-contact volcanic rock layers, or carbonate source rocks (Ozerova, et al., 2004; Li et al., 2008). Southeast Asia, especially Thailand and Indonesia, are regions known to carry anomalously high mercury concentrations in a variety of hydrocarbon products.

4. General Settings and Stratigraphy

The Pailin operating area, which covers approximately 6,800 sq. km., is located in the southern part of the Pattani trough. Broad-scale tectonic elements are believed to develop in accordance with a pre-existing north-to-south oriented set of "Indosinian" fabrics and a series of northwest to southeast strike-slip faults developed during Indian-Eurasian plate collisions (Mountford, 1992). Accommodation space for post-Oligocene sedimentary sequences was created during the post-rifting episode (Crossley, 1990) (see Figure 3).

The Tertiary stratigraphy of the Pattani Basin can be divided into 5 sequences, termed "Sequence One" to "Sequence Five" in ascending order, based on bothlithostratigraphic and seismic stratigraphic facies (Jardine, 1997; Mountford 1992). Tectonics and Eustatic sea-level fluctuations influenced the facies development of each sequence, which ranges from continental alluvial to shallow/marginal marine. These "sequences" are litho-stratigraphic and not defined in the sense of conventional sequence stratigraphic analysis, which requires of all sequence-bounding surfaces. Jardine (1997) has proposed the widely-accepted Pattani depositional model, which generalized depositional settings and stratigraphic units (see Figure 2).

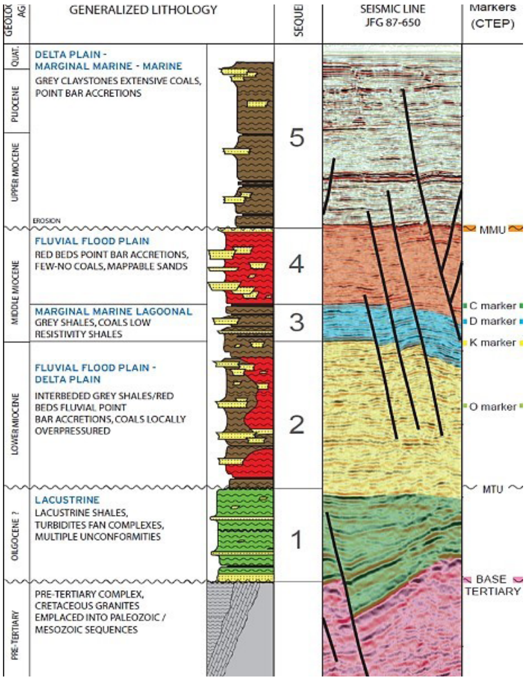

Figure 2. Generalized stratigraphy of the Pattani Basin (after Jardine, 1997)

5. Potential Sources of Mercury in Pailin Area

Origins of elevated mercury levels in the Gulf of Thailand are equivocal because of a lack of detailed scientific studies. Frankiewicz et al. (1997) proposed that mercury is derived from coal, carbonaceous shale, and adjacent tin (Type II) granites and associated with high temperature producing reservoirs. Similarly, Gallup and String (2006) also suggested mercury origins associated with coals and organic-rich sedimentary rocks. In addition, an anthropogenic re-injection activities can cause mercury accumulations in subsurface.

6. Analytical Approach

This study is the first attempt to incorporate several data sets from different disciplines in order to quantify existing hypotheses.

The study focuses on four dedicated platforms that have been collecting well-level mercury contents tied to known zones of perforation. The platforms are situated in two fields in the Pailin area including NPQ and NPR platforms located in the North Pailin area, plus the MOE and MOF platforms located in the Moragot area.

The approach used in this study is as follows. The initial step is to identify potential sources of mercury found in produced condensate. Several geological evaluations are considered in order to test individual hypotheses mentioned earlier (see Table 1). Next, a regional fault and horizon interpretation is compiled to model possible migration pathways of mercury.

According to data limitations, most of observations have been made on Moragot platforms.

7. Detailed Investigation on Potential Sources of Mercury in the Gulf of Thailand

By incorporating Hg-level information with geological data, a more conclusive explanation is expected that will either strengthen or weaken the former assumption that; "the elevated mercury concentration in produced condensate has been sourced from "massive sands" reservoir sections." Sequences of geological evaluation are described below.

7.1 Mercury Distribution

Mercury contents were determined from collected condensate samples by using industry standard mercury analysis techniques (via a Lumex Mercury Analyzer). The concentration of Hg was reported in total mercury content (THg, ppb). However, in this study, for reasons of confidentiality, all mercury measurements will not be reported as actual values. Relative comparison of Hg content will be illustrated as relative true-scale outputs, with no labels, bar charts and cross plots.

Condensate samples submitted for Hg determination are collected from outflows discharges at different locations on a platform: wellhead, platform, launcher platform and central processing platform; (1) some are single source fluids from a known production interval, (2) others are aggregations fluids from a number of production levels, and yet (3) others are aggregations of processed fluids from a number

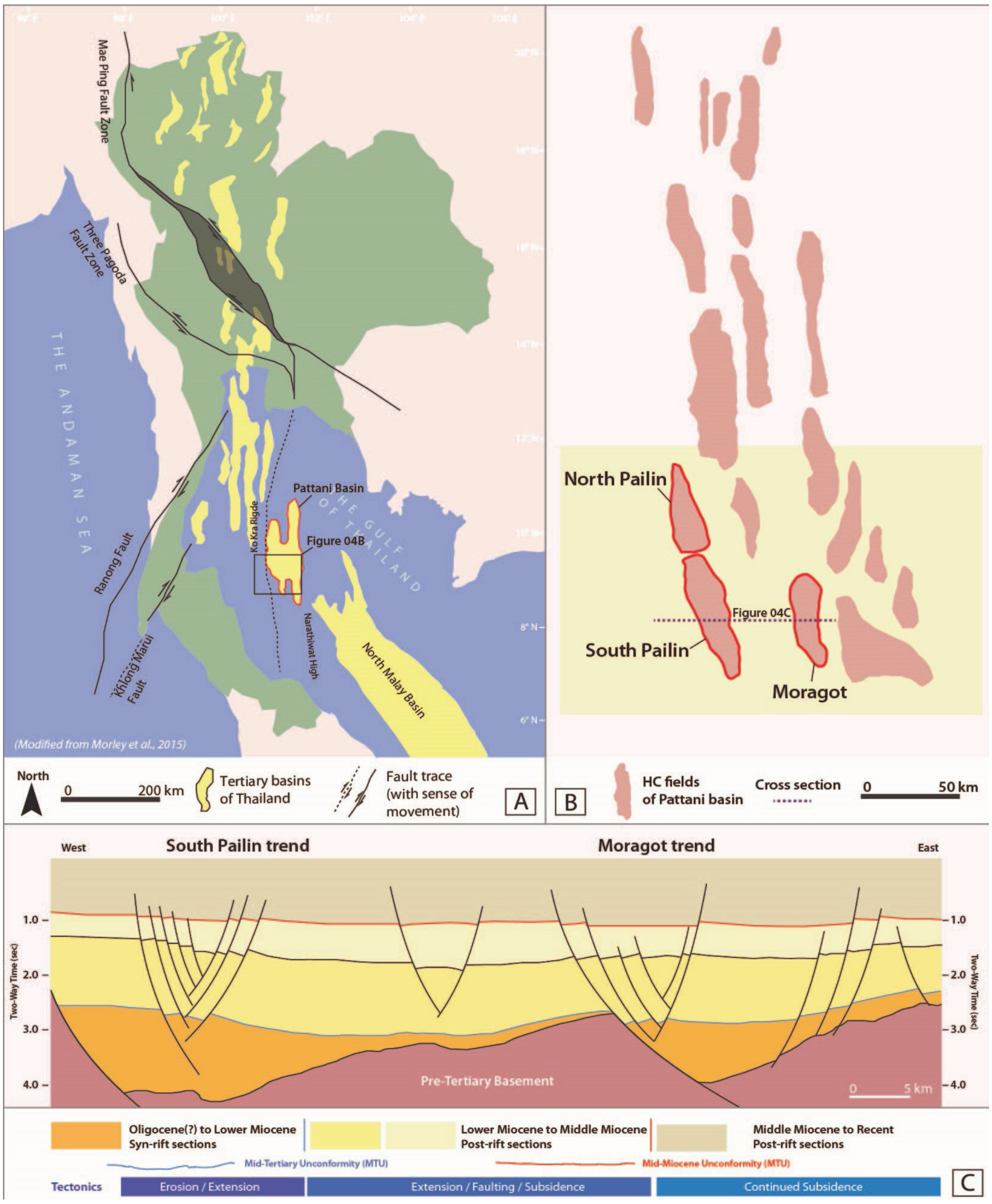

Figure 3. (A) Major tectonic elements map overlying with Tertiary basins of Thailand (Modified from Morley et al., 2015). The Pattani basin is located in the central part of the Gulf of Thailand. (B) Location of Pailin (PA) operating area (including North PA, South PA, and Moragot) relative to other hydrocarbon fields in the Pattani basin. (C) West-East structural cross section of Pailin area (from South Pailin to Moragot) with highlighted sequences.

of wells. Accordingly, there are inherent varia tions in determined Hg level tied to where in the hydrocarbon production process the sample was collected.

Regionally, Moragot and North Pailin platforms seems to have lower Hg contents than the South Pailin platforms (based on platform-scale Hg contents).

Based on Hg concentrations from dedi-cated platforms, Moragot trend shows relatively high Hg contents at the MOE platform location.

7.2 Hg-CO2 Correlation

Wilhelm et al. (2008) recommended that utilizing reservoir CO2 content may allow the prediction of mercury levels. To test this statement, Hg-CO2 cross plots are compiled by using the Hg sampling date as a time reference for CO2 content matching.No clear relationship can be established in the compiled log-linear cross plots of platform-level Hg and CO^2

Geological Assessment	Purpose of analytical approach
Hg distribution	To evaluate mercury distribution in a regional perspective
Hg-CO_2 correlation	To quantify the Hg-CO_2 relationship, if possible
Potential mercury-rich reservoir sections	To identify zones that contribute significant mercury
Hydrothermal influence evaluation	To test the mercury-rich hydrothermal fluid hypothesis by using cuttings-derived C-O isotope data
Water re-injection observations	To test the anthropogenic origin hypothesis by looking for Hg-anomalies that can be ties to water reinjection activities in the study area including the incorporation of formation testing data
Possible migration pathway recognition	To clarify possible Hg-source intervals, which could possibly be either *lacustrine shales in Sequence 1* (vertically-migrated fluid model) or *marginal/lagoonal coals and carbonaceous shales* (self-sourced fluid model)

Table 1. Summary of analytical approaches integrated to better understand the potential Hg source

contents (see Figure 4).

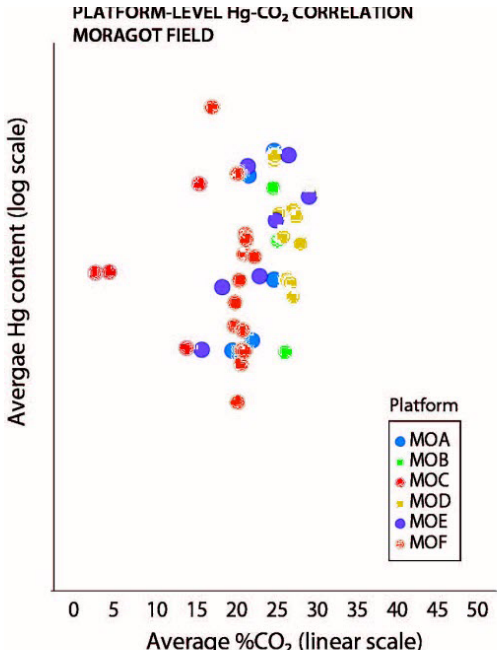

Figure 4. (A) Platform-level Hg-CO2 cross plot (coloured by platform) of Moragot field

7.3 Potential Mercury-Rich Reservoir Sections

The zone of perforation has been selected to represent narrower producing zone. Regional markers were used as a reference for categorizing "to-be-perforated" reservoirs into different stratigraphic sequences.

The zone of perforation has been selected to represent narrower producing zone. Regional markers were used as a reference for categorizing "to-be-perforated" reservoirs into different stratigraphic sequences.

Four stratigraphic intervals were nominated to represent various depositional settings in the Pailin area stratigraphy, namely: (1) C-K interval (Sequence 3), (2) K-M interval (late Upper Sequence 2), (3) M-O interval (High GR zone, early Upper Sequence 2), and (4) Below O interval (Lower Sequence 2). Due to an inherently complicated distribution of pay zones, some wells have been perforated across commingled intervals. These commingled-sequence producing wells are not considered as "zonal indicator" wells in this compilation.

MOE outboard wells were used to illustrate this concept. Figure 5 shows a well correlation panel highlighted with zones of perforation of the various MOE outboard wells. Hg contents of each well are displayed in both bar charts and as box plots below (see Figure 5B). Only four wells are considered to be reliable zonal indicators. Based on these observations, the K-M interval, represented by MOE-04, has contributed a minor amount of mercury. In contrast, reservoirs located below the M marker, either M-O (MOE-07) or the "Below O" (MOE-09 and -10) intervals, show relatively higher Hg concentrations.

7.4 Hydrothermal Influence Evaluation

This project is the first attempt to determine possible hydrothermal influences using cuttings-derived C-O stable isotope data of carbonate cements in a siliciclastic system from offshore Thailand. This determination may identify hydrothermal isotopic signatures in carbonate cements in the siliciclastic of carbonate cements in a siliciclastic system from offshore Thailand. This determination may identify hydrothermal isotopic signatures

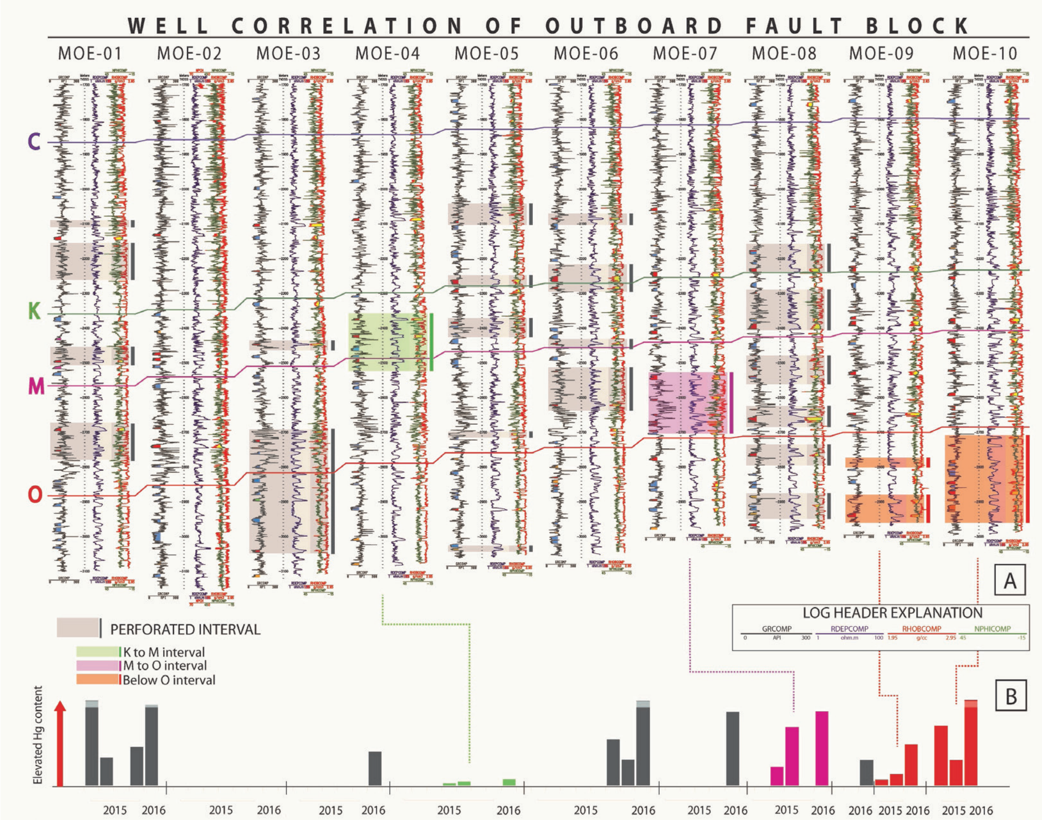

Figure 5 (A) Well correlation of drilled wells in an outboard fault block in MOE. The overlying shaded-interval shows perforated zones for each well (zonal perforations). Wells with grey shaded intervals have commingled-perforated zones of different intersected geological intervals (not interval specific). Wells with other colour-shaded zones (MGWE-04, -07, -09, and -10) have narrower zone of perforation (more interval specific). (B) Bar chart: Well-level mercury contents from each well in the fault block with shaded colour in accordance with perforated zones illustrated in Figure 10A. Variations in Hg content in each well can be recognized from the varying in height of bars.

in carbonate cements in the siliciclastic sedimentary sequence via the use of C-O covariant cross plots. This is a widely used technique in carbonate studies in Thailand, when defining the diagenetic fluid history (Warren et al., 2014).

In brief, covariant C-O isotopic signatures can be used as an indicator of carbonate rocks/cements origin. Depleted 13C value indicates either freshwater/ meteoric influences or elevated organic contents (possibly catagenic), whereas, the depletion of ^{18}O in Permian carbonates of onshore Thailand indicates precipitation from higher temperature fluids (Warren et al., 2014).

Overall, there is no consistent significant shift in ^{18}O values with respect to increasing depth in the studied well. A lack of any thermal anomaly signal in the isotope plots indicates that there was no consistently-hotter hydrothermal fluid coming from the deeper sections and moving upward through sedimentary sequences with widespread precipitated carbonate cements as it passed through the outboard area. More likely, the various carbonate cement values preserve isotope signature differentials that are related to differences in early burial pore fluid chemistries.

7.5 Water Re-Injection Observation based on Formation Pressure

Due to complex connectivity of fluvial-dominated thin bed stacked reservoirs in the Gulf of Thailand, the re-injected water could accidentally charge movable mercury into producing reservoirs, especially if the re-injected water had picked up mercury from Hg-rich sections. Currently, only 6 platforms

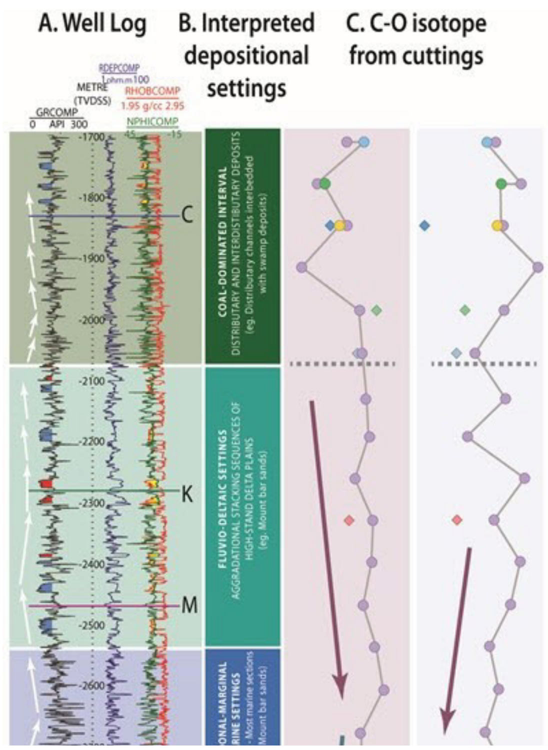

Figure 6 (A) Triple-combo log of MOE-05 with white arrows showing shaling- and sanding-upward sections. (B) Interpreted depositional settings according to lithofacies from Figure 10A. (C) Plots of cuttings-derived C-O isotope with depth overlain with arrows emphasizing the trend of each data output.

in Pailin area utilize re-injection for water disposal, all these platform are located more than 10 km away from any platform with high levels of Hg. No water flooding operation in use as the main produced hydrocarbon is natural gas.

8. Evaluation of Possible Mercury Sources

Due to data limitations, making strongly quantitative arguments for or against each hypotheses across all platforms is rather difficult. As it has so much more information available relative to the other platforms, the MOE and MOF platforms are used as the prime examples in ranking the relevance of the various Hg sources.

8.1 Lacustrine Shales (Sequence 1)

Because there are no well penetrations into this section, the most obvious illustrations of this sequence in the study area come from seismic data.

Typically, the lacustrine deposits occupy the depocentre of the basin (see Figure 7A). This suggests that, if mercury-rich condensate is sourced from the lacustrine shales, produced fluids from the MOF platform should have a relatively higher content of mercury than the MOE platform.

However, well-level monitored mercury concentrations show that MOE produced fluids are relatively richer in Hg content, especially from wells in the outboard area. Furthermore, as the producing reservoirs are located in shallower sequences, to bring the mercury-rich hydrocarbons up into these reservoirs requires a relatively long-distance migration. A corollary to the lacustrine source hypothesis would be that mercury and its compounds, which are heavy metals, should accumulate and remain in the vicinity of the source due to gravitational segregation. Thus, the likelihood of lacustrine shales being the source of mercury in the produced fluid is ranked as low.

8.2 Marginal Marine Coals and Carbonaceous Shales (Upper Sequence 2 and Lower Sequence 3)

In these sequences, high gamma-ray marginal marine coals and carbonaceous shales predominate due to fluvio-deltaic deposition during marine transgression. Integration of the total GR log with available spectral GR logs from nearby delineation wells shows thathigh GR signature positions on the GRCOMP curve commonly associate with high uranium spikes in the spectral log. These log characters suggest that coals and shales in M-O intervals are dominated by organic-rich materials (moremarine influence).

Coincidentally, based on "in-house" regional basin modelling studies (Liu and Czerniak, 2007), the level of vitrinite reflectance (Ro) value of 1.0 – known as main hydrocarbon generation window – is at the level of M marker in the outboard zone (see red dashed line, Figure 7B). This indicates that the Hg-rich condensates in M-O reservoirs were likely derived from the adjacent source rocks. Moreover, the relative

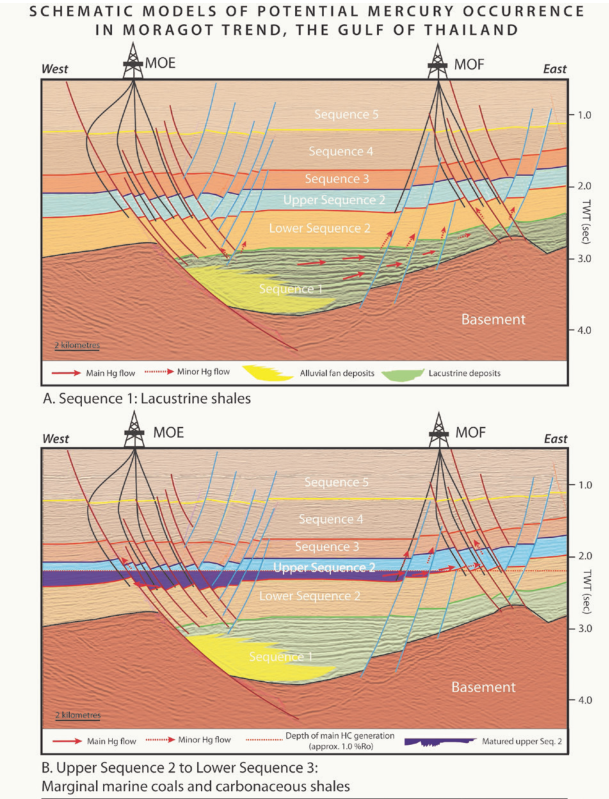

Figure 7 Schematic model of mercury occurrence in Moragot trend. (A) "Vertical-migrated hydrocarbon model" – the mercury-rich HC vertically migrated from lacustrine source rocks (Sequence 1) through faults. Regionally, the main migration moves toward MOF platform due to formation dips. (B)"Self-sourcing hydrocarbon model" – the mercury-rich HC migrated into reservoirs from adjacent marginal marine source rocks.

decrease in Hg contents of fluids produced from the MOF platform can be correlated with the presence of a lower hydrocarbon maturity level in these marginal marine (upper Sequence 2) sections located to the east of the Moragot trend. Below the O marker, uranium-rich shales are largely absent due to a transition from humic-rich to more fluvial-dominated depositional settings. To charge Hg-bearing hydrocarbon fluids into "O"-sands from the overlying (younger/ upper) stratigraphic sequences probably requires large-offset faulting. This is needed in order to move the marginal marine shales on down-thrown side to the equivalent depth of the sands, thus allowing Hg-enriched hydrocarbons generated in the shale section to charge adjacent "O"-sands juxtaposed across the fault on the up-thrown side (see interpretation in Figure 7B).

This model is a clarification of a previous study done by the petroleum engineering team that proposed massive sands as the main contributor to mercury-rich condensates. However, only suitable structures, like the bounding fault, would allow sufficient throw to facilitate the occurrence of Hg-enriched O"-pay sands.

This marginal marine shale source model is evaluated as the most-likely scenario for mercury occurrence in the Moragot trend.

8.3 Mercury-Rich Hydrothermal Fluids

This hypothesis postulated a trending of intense depletion in $\delta^{18}O$ stable isotope values with depth, utilizing cuttings-derived C-O isotopic plot fields from an appropriate well that was drilled along a deeply-cut bounding fault. But, the absence of obvious hydrothermal-influenced isotopic signature in most of the isotope values (see Figure 6C) suggests that the likelihood of hydrothermal-associated mercury origin is low. However, the result of the isotope study is evaluated as uncertain as the cuttings samples used in this research are from only one well, MOE-05.

Also, the newly-documented poor correlation between Hg contents and CO2 percentage in the produced fluids, suggests these components are likely derived from different sources, running counter to the notion of a deep vertically-migrated basement (hydrothermal) source for Hg. CO2 in the GOT is widely believed to have migrated vertically from deep sources, such as pre-Tertiary basement (Lin, 1997). If mercury was also derived directly from a deep basin source, then the two components (CO2 and Hg) should show better correlations in the produced fluids at the well-level measurements.

8.4 Unintended Mercury Accumulation due to Water Disposal Activities

As the level of water re-injection activity in the Pailin area is low, this hypothesis is considered to be the least likely explanation for the occurrence of elevated mercury levels across the region, especially in the vicinity of the Moragot trend.

However, this model could be relevant to producing platforms located in the vicinity of water disposal platforms. Further studies on those platforms are recommended.

9. Conclusions and Implications

This study is the first attempt to integrate various available data to investigate the possible source/occurrence of elevated mercury levels in southern part of the Gulf of Thailand and to attempt to determine whether the mercury is derived from geogenic or anthropogenic sources.

The results of detailed data integration suggest that the mercury contaminating produced hydrocarbons in the study area is likely to originate from geological sources. The most probable explanation for the occurrence of mercury-rich condensates is the "coals and carbonaceous shales origin" hypothesis, whereby mercury and its compounds are captured in the marginal marine organic-rich shales during deposition (Upper Sequence 2). These Hg prone sediment sources are indicated by elevated uranium levels best developed in the M-O intervals, known as the "high gamma ray" zone in the Pailin stratigraphy. At this level in the stratigraphy, Hg-enriched hydrocarbons are generated and charge adjacent reservoirs. Variations in Hg levels are tied to (1) the maturity of these M-O intervals, (2) the connectivity between reservoirs within thesame sequences (M-O interval reservoirs), and (3) the juxtaposition across faults of reservoirs deposited in different stratigraphic units (e.g. "Below O" reservoirs, aka "O"-sands). Other possible Hg sources are not supported by convincing results:

1) Lacustrine source rocks: less likely – this hypothesis is not supported as it conflicts with relative Hg content measured at the well level in fluids produced from MOE and MOF platforms. If the lacustrine shales were the dominant source, then there should be higher Hg accumulations at MOF location compared to MOE. The opposite is true.

2) Hg-rich hydrothermal fluids migrated through deep-cut faults: less likely – due to (1) absence of consistently hotter isotopic signatures with depth (more depleted in $\delta^{18}O$ value) and (2) a poor correlation between Hg and CO2, the latter is known to be sourced from pre-Tertiary basement migrating through deep-penetrated faults.

3) Anthropogenic sources by unintended water re-injection activities: less likely – due to (1) only a few water re-injection activities (only water disposal, no water flooding) in the Pailin area, and (2) examples where this is happening do not consistently tie back to platforms with fluid contents showing elevated Hg levels.

10. Recommendations

Based on observations made in this study, the following comments should be useful for future studies on mercury source investigation and relevant topics.

- Quality control on mercury concentrations is vital. Any outliers from measurements across all scales have to be rejected. Collaboration with petroleum engineers is highly recommended.
- A zonal perforation strategy in monitoring fluid content is useful. Narrow zones of perforation within the same stratigraphic units are recommended in regions known to be subject to elevated Hg levels.
- Depth of perforations should be checked. Some perforated sands that are listed in the corporate database seem to have miss-perforated. This will mislead the results attempting to identify zones of mercury contribution.
- Reservoir properties, such as porosity and shale volume (Vsh), of the perforated sands could reflect mercury concentrations collected at surface. Mercury held in the less sandy (higher Vsh) reservoir is expected to have less mobility. Due to commingled zones of perforation, this was too difficult to verify in this study.
- More isotope studies from several wells are required to confirm or reject any influnce on Hg levels coming from hydro thermal fluids

- Spectral gamma ray logs important in verifying degree of marine influence in the suspected shale intervals (separates U from K and Th). At a more regional scale, the effects of lithofacies change on Hg content – due to the extent of marine transgressive muds – should be an aim in further more regional assessments of likely Hg content
- Maturity level of suspected source rocks will likely express the degree of Hg enrichment
- Detailed basin modelling is needed to confirm timing of HC generation and migration, and whether or not it fits with the proposed model, which Hg levels are tied to the maturation of a specific type of source rock ("High Gamma zone").

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