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Vapour Emissions During Ex-situ Remediation Activities: Model Calibration, Evaluation of Biofilters, and Screening Tool Development

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1 INTRODUCTION

1.1 General

Under the authorization of the Petroleum Technology Alliance Canada (PTAC), Meridian Environmental Inc. (Meridian) has conducted a follow-up assessment of the environmental significance of vapour emissions during ex-situ remediation activities. The work was conducted under the Alberta Upstream Petroleum Research Fund (AUPRF), under the direction of the Canadian Association of Petroleum Producers (CAPP) and the Small Explorers and Producers Association of Canada (SEPAC), as indicated in a letter of approval from PTAC dated February 3, 2012. The following report documents the activities conducted in 2012.

1.2 Background

Onsite *ex-situ* soil remedial technologies involve the excavation of contaminated material and treatment onsite, potentially resulting in the release of volatile chemicals to ambient air. The resulting volatile emissions may include greenhouse gasses as well as compounds that may impact ambient air quality for humans or ecological receptors. Meridian previously completed a comparative assessment of emissions during *ex-situ* remedial activities and land fill disposal (Meridian 2011) for the PTAC. A modelling approach was developed for use in comparing lifecycle emissions from alternative remediation scenarios, and their impacts to human health and the environment.

Upon completion of the previous project key data gaps were identified. Based on these data gaps Meridian proposed a follow-up project consisting of three independent tasks. The tasks are: collection of emission data from *ex-situ* remediation for calibration of the previously developed model, evaluation of the effectiveness of biofilters in treating emissions from remediation activities using a lifecycle approach, and development of a spreadsheet tool for screening-level assessment of air quality and life cycle emissions for *ex-situ* and landfill disposal options. Each of these proposed tasks would add value to the previous work conducted by Meridian.

Collection of Emission Data from Ex-Situ Remediation

Emissions from industrial processes and remedial efforts in the oil and gas industry are subject to considerable uncertainty surrounding their quantity and composition. During development of the Meridian emission model, several assumptions were made that significantly influenced emission output values. In particular, there is a paucity of real-world data on emissions from *ex-situ* remediation and no previously existing modelling approaches were identified. Meridian developed a model to predict emissions from *ex-situ* remediation based on models derived for excavation; however, several of the model inputs could not be reliably quantified with existing data. Obtaining data on actual soil emissions during *ex-situ* remediation was therefore proposed to help quantify the relevant processes and calibrate the model.

Lifecycle Evaluation of Biofilters

Use of biofilters may significantly reduce emission outputs but will depend heavily on site, contaminant, and filter operation characteristics. However, the long-term fate of chemicals extracted using biofilters and emissions from the production and disposal of biofilters must also be quantified in order to evaluate the lifecycle effects of this approach. This aspect of the project evaluates the lifecycle emissions of VOCs, greenhouse gasses and priority pollutants associated with biofilter use based on a desktop review.

Spreadsheet Model

In order for the previously-developed model to be more useful to industry, a spreadsheet model has been developed to allow for a screening-level comparison of different remediation approaches with respect to ambient air quality and lifecycle pollutant emissions. The model was based on the previous Meridian modelling approach, adjusted based on the results of the current work. The spreadsheet is intended to be user-friendly and minimize the number of required inputs for simplicity, while still allowing for the use of additional site-specific data where available in order to refine the results.

1.4 Scope of Work

The scope of work was based on a proposal submitted by Meridian on August 25, 2011.

The collection of emissions data and model calibration involved the following tasks:

1. Identification of a candidate site and possible industry partnerships, along with appropriate data collection methodologies for each site

2. Quantification of emissions through soil and air sampling during *ex-situ* remediation

3. Calibration of the model based on the collected data.

The research on biofilters involved the following tasks:

- 1. Literature review in two distinct parts: (1) identify and assess current technology and applications of biofilters (2) identify the lifecycle emissions associated with biofilter use
- 2. Addition of biofilter considerations to the model in order to predict emission treatment effectiveness and lifecycle emissions during *ex-situ* remedial activities.
- 3. Life cycle analysis of a remediation scenario with and without biofilter application

The development of the spreadsheet tool involved the following tasks:

- 1. Alteration of the original model based on any updated information from the other proposed tasks
- 2. Development of a spreadsheet tool and internal validation of tool functionality
- 3. Completion of user guidance documents.



2 MODEL CALIBRATION

In order to calibrate the model, soil concentrations and volatile emissions from one *exsitu* remediation project were measured as a pilot test. Candidate sites were provided by the PTAC Project Manager. The available reports from these sites were evaluated to identify the site with the highest concentrations of volatile contaminants and appropriate remediation technology. The remediation project selected was a wellsite currently owned by ConocoPhilips Canada (Conoco), located within 06-23-034-08 W5M approximately 30 km northwest of Sundre, Alberta. Contamination at the wellsite consisted primarily of diesel fuel.

Remediation activities at the site included processing excavated soil piles through a trommel aerator. At the time of sampling, excavation and piling of soil had previously been completed and contaminated soil was being stored onsite. Soil was aerated by both allu bucket and trommel treatments. Emission sources at the site included: the trommel aeration exhaust output, the trommel diesel engine exhaust output, dump truck exhaust emissions, and excavator exhaust emissions.

2.1 Data Collection

Soil and air samples were collected simultaneously after a brief warm-up period for the operators and equipment. As the contaminated material had already been excavated and piled onsite, it was not possible to correlate the treated soil with the worst-case contamination identified in previous investigations; however, field readings with an RKI Eagle gas detector and odour observations by field personnel indicated that the sampled material was considered highly contaminated.

Pre-treatment soil samples were obtained directly from an excavator bucket containing approximately 1 m³ of contaminated soil before aeration and trommel treatments. It should be noted that as the soil had already been excavated and moved around site, it is likely that a significant amount of the volatile materials had been lost at this point. The material in the excavator bucket was then aerated and run through the trommel. Post-treatment soil samples were then obtained directly from the fine material ejected from the trommel after all the soil had passed through. This process was repeated five times over a 1-hour period, with before and after samples taken from each bucket load.

Over the course of the soil sampling process, three flow-regulated summa canisters were obtaining continuous 1-hour air samples at a height of 1.5 m above grade. Air samples were obtained from locations identified as "background," "source," and "downwind." The background sample was obtained offsite and upwind of the trommel, the source sample was collected adjacent to the trommel output exhaust, and the downwind sample was collected approximately 10 m downwind of the source sample. A distance of 10 m was utilized to provide room for moving equipment through the area.

Collected soil and air samples were submitted to ALS Laboratories in Calgary for analysis of benzene, toluene, ethylbenzene, xylenes, PHC F1, and PHC F2. Analysis of physical soil characteristics was also completed for pre-treatment soil samples.

2.2 Results

Concentrations of benzene, toluene, ethylbenzene, and xylenes were not detected in any of the pre-treatment or post-treatment soil samples, with the exception of ethylbenzene in two pre-treatment samples. PHC fractions F1 through F4 were detected in soil samples at concentrations below regulatory guidelines for vapour inhalation. Measureable differences in average pre- and post-treatment soil concentrations were only observed for PHC F2 (505 mg/kg to 443 mg/kg) and PHC F3 (492 mg/kg to 419 mg/kg).

No PHCs were detected at the background air sampling location, and only PHC F2 was detected in air downwind of the source area. Benzene, ethylbenzene, PHC F1 and PHC F1 detected in the air sample from the source area, but all detected parameters were well below occupational exposure limits and the tolerable or risk-specific concentrations specified by ESRD (2010).

The results of laboratory analysis for soil and air are presented in Tables 1 and 2, respectively.

2.3 Data Evaluation

The soil data were used to estimate the mass of contaminants lost to the atmosphere during the 1-hour remediation period. This data was incorporated into the previously developed Meridian emission model (2011) and emission outputs were compared to measured air concentrations obtained from the source and downwind air sampling location.

As the downwind sample did not contain detectable PHCs, a qualitative comparison to the laboratory detection limits was made instead. Concentrations of several parameters were below laboratory detection limits and could not be included in the model comparison. Based on the measured concentrations and observations on site activities, it appears that a significant portion of the volatile contaminants had been lost prior to obtaining the pre-treatment samples, either through excavation and handling disturbances or gradual volatilization to the atmosphere over time.

2.4 Model Comparison

Measured concentrations of PHC contaminants in soil were used as inputs for the Meridian (2011) model, and predicted air exposure concentrations were compared to the measured air concentrations. The model significantly overpredicted exposure concentrations for all parameters with data available, by a factor ranging from 3 to 200. While this overprediction ensures that the model is conservative when considering worker exposure to contaminants, it is likely that the previous loss of volatile mass was a major factor as well. Due to the limited amount of data available, no adjustments to the original model are recommended at this time. A comparison of measured air concentrations predicted by the model is included in Table 3.

2.5 Sources of Uncertainty

Sources of uncertainty were previously identified in the Meridian (2011) report; only sources of uncertainty specific to the data collection and model calibration are considered herein.

A limited number of soil and air samples were obtained from the site during remediation activities. While concentrations measured in soil were generally consistent and are likely representative of actual concentrations, there are insufficient data available to determine the accuracy of the summa canister air sampling. Due to the large size of the trommel emission output area, there was uncertainty regarding where the worst case sample should be collected from and how to address variability in wind direction. Other concerns related to the site include the presence of diesel exhaust emissions from additional onsite equipment not included in the model, including a bulldozer and additional excavator.

When considering uncertainty in the model itself, differences in operator technique, equipment used, and site conditions (such as a steep grade or access difficulties) can also invalidate the assumptions used in the model, or significantly affect emissions and exposure concentrations.

2.6 Recommendations

Additional data are required to complete model calibration. If more sampling is undertaken, the following recommendations are provided:

As mentioned in Section 2.2, it is suspected that a significant amount of the volatile materials had been lost before sampling began. A site where higher levels of volatile hydrocarbon contamination are present and pre-treatment handling is limited would maximize the potential for significant quantities of hydrocarbons to be removed from the soil during treatment.

As the majority of remediation equipment runs on diesel fuel, sampling from sites contaminated with diesel should be avoided, so that emissions from the contaminated material and from equipment can be distinguished. It is recommended that condensate spill sites be targeted in the future based on their expected composition of volatile PHCs.

The sites selected should ideally be relatively small, in order to ensure that the limited amount of samples taken are representative of overall site conditions. Alternatively, if larger sites are used, a site that has been well-characterized and where excavated soils can be more directly related to areas of identified high contamination would maximize the potential for sampling highly-impacted soils.

3. BIOFILTER EVALUATION

The biofilter literature review included academic research, government information and regulatory guidance, as well as publically available information on commercially available biofilters.

3.1 Background

A typical biofilter uses a three-phase system, consisting of a gas phase transporting the target chemicals through the reactor, a liquid phase which contains an aqueous biofilm, and a solid phase substrate on which the microorganisms are present (Ikemoto *et al.* 2006).

The gas phase often consists of ambient air, with the typical open biofilter being exposed to the atmosphere and limited by the rate of ascending gas flow (Delhomenie and Heitz 2005). The speed of the biofiltration process is often limited by the rate of diffusion of contaminants inside the biofilm layer. A prototype biofilter using a convective flow biofilm to speed up this process was tested by Fang and Govind (2007) and found to be more effective than a diffusive flow biofilter; however, the majority of biofilters use diffusive flow.

Biofilms are populations of microorganisms attached to the solid biofilter substrate surfaces through polymeric substances, and the biofilm matrix will contain: water, microbial cells, polymers, absorbed nutrients, and metabolic byproducts (Singh *et al.* 2006). Establishment of a biofilm within a biofilter requires an initial microbial population and sufficient nutrient flow to maintain that population (Singh *et al.* 2006).

The solid substrate, or filter bed, is generally in the form of inert packing material, and is required to increase the available area for surface transfer between phases (Delhomenie and Heitz 2005). Microorganisms are immobilized on this surface, which is often also infused with a nutrient solution (Delhomenie and Heitz 2005). A filter bed should have: high surface area, high porosity to allow for gas transport, good water retention, nutrients to support microbes, and diverse indigenous microflora (Delhomenie and Heitz 2005). Options for filter bed material are peats, soils, or compost, which can also have additional material added for structural support, such as wood chips. Typical biofilter substrates have a porosity of 40 to 50%, a surface area of 1 to 100 m²/g, and contain 1 to 5% organic matter (Nelson and Bohn 2011). Compost typically has a higher porosity (S0 to 80%) and organic matter content (50 to 80%) along with greater microbial density (Nelson and Bohn 2011).

Use of other organisms besides bacteria, such has fungi, has also been attempted. A study by Harms *et al.* (2001) identified potential fungal species capable of degrading PHCs; however, there were not considered to be ideal for biofilters as they were less resilient and grew slower than similar bacteria. Hasan and Sorial (2010) also looked at using fungi to enhance bioremediation in acidic environments, and found that adding fungi to a biofilter increased the available surface area, increasing the gas-phase uptake and elimination diffusion as a rate limiting step.

At this time most commercial biofilters operate using indigenous microbial populations.

3.2 Relevant Parameters

The parameters of greatest importance to biofilter operation are generally considered to be: air flow rate, temperature, pH, moisture, and substrate organic content (Delhomenie and Heitz 2005). The treatment rates are often limited by the mass transfer of target compounds to the biofilm phase, and mass transfer rates are most influenced by: target chemical properties, biofilter substrate properties, gas flow behavior, phase surface area, wettability of biofilter substrate, and environmental conditions (Kraakman *et al.* 2011).

During biofilter operation, moisture content often requires regular monitoring. The ideal moisture content is considered to be between 10 and 25% for soil substrate based biofilters, and between 20 and 20% for compost substrate based biofilters (Nelson and Bohn 2011). In order to maintain optimal humidity levels, active systems such as passing inlet air through a water column may be required depending on environmental conditions (Rani 2009).

3.3 Treatment Rates

Removal efficiencies over 99% have been obtained for BTEX in lab tests (Mathur *et al.* 2007, Pandey *et al.* 2010) and complete removal of hexane in a compost biofilter (Zamir *et al.* 2011); however, lower removal efficiencies closer to 80% are often encountered in practice (Namkoong *et al.* 2003). Treatment rates are typically limited by the rate of mass transfer between phases in a biofilter, and are influenced by the initial concentration of microbes and the air flow rate through the biofilter (Delhomenie and Heitz 2005). Mass transport in biofilms is described by Fick's Law (Delhomenie and Heitz 2005), and soil water partitioning in biofilters is generally the limiting factor in treatment rates, whereas air-water partitioning tends to reach equilibrium at typical flow rates (Massabo *et al.* 2007).

Remediation rates are typically proportional to environmental temperature and are inversely proportional to the molecular weight of the target compounds (Sanscartier *et al.* 2011). Microbial activity is linked to ambient temperature, with an optimum between 20 and 40 °C; however, degradation processes can cause a temperature gradient to occur within the filter which can hinder maintenance of the filter (Delhomenie and Heitz 2005). Achieved treatment efficiency will vary depending on input concentrations of target compounds and the biofilters internal conditions; however, addition of silicon rubber pellets in a bioreactor or other compounds that can absorb/desorb gaseous components can be used to smooth out input concentrations, preventing possibly damaging high transient loadings of contaminants (Littlejohns and Daugulis 2008) and maintaining a more consistent treatment rate. Despite its relatively low molecular weight, removal rates of benzene are commonly the lowest among BTEX and other petroleum hydrocarbons (Namkoong *et al.* 2001) as it is easier to degrade water soluble compounds (Nelson and Bohn 2011).

Other chemical parameters of target compounds also influence the treatment rates of biofiltres. Degradation rates tend to increase with the presence of compounds with double bonds, and higher oxygen, nitrogen, and sulphur content (Nelson and Bohn 2011). The presence of certain contaminants can also interfere with the biodegradation of target compounds. For example, Hasan and Sorial (2009) found the degradation of n-hexane to be negatively impacted by the presence of benzene. Shim *et al.* (2006) determined that the presence of methyl tert-butyl ether (MTBE) decreased the elimination capacity for benzene, toluene and xylenes, with benzene degradation most strongly inhibited by MTBE. It was also found that MTBE could not be degraded alone, as it was co-metabolically degraded with benzene, toluene, or xylene.

The materials used in biofilter construction can also significantly treatment rates. The use of granulated activated carbon can be used to enhance gas adsorption (Mathur *et al.* 2007), and has been show to benefit treatment efficiency, depending on humidity (Nabatilan and Moe 2011). Use of higher porosity substrates can increase removal rates while reducing plugging issues (Nikakhtari and Hill 2008). When trying to remediate hydrophobic substances that will not readily dissolve into the biofilm phase, the use of surfactants has been found to enhance solubility and increase biofilter effectiveness (Hasan and Sorial 2010), this result has not been found to be significant in all cases involving PHCs (Lee *et al.* 2011).. The use of organic solvents has also been attempted to achieve this effect (Kraakman *et al.* 2011)

Operational and maintenance considerations are also important when trying to determine treatment rates. High conversion rates of target compounds can result in microbial growth rates high enough to result in filter clogging (Kraakman *et al.* 2011). Shim *et al.* (2006) found that the presence of specific pollutants, such as MTBE, can significantly alter the established bacteria communities and the expected treatment rates for other target compounds. Additionally, high inlet concentrations of target parameters may also damage the degrading biomass, Barona *et al.* (2007) found this to be the case with even low concentrations of toluene.

3.4 Applicability to Emission Model

Design of biofilters based on models or small-scale testing often runs in to difficulties due to uncertainties in determining the rate-limiting step in larger systems (Kraakman *et al.* 2011). While treatment rates can often be established based on easily measurable parameters, such as biofilter design or flow rate in lab scale tests, determining the actual treatment rate of a biofilter under field conditions would require some form of validation.

However, for all biofilters, there is a range of operational parameters over which biofiltration is effective, dependent on the filter design, microbial community, and contaminants of concern (Fang and Govind 2007). Therefore, it is possible that a standard removal efficiency credit could be applied to the emission model if a generic biofilter design was confirmed to be operating within the defined operational range. This credit would also be contingent on some basic site conditions, such as the absence of

contaminants that may limit microbial growth, or adverse site conditions (such as extremes of temperature or humidity).

While overall removal efficiencies greater than 95% are possible and efficiencies of 80% or greater are expected to be achieved, actual removal efficiency will vary between contaminants and result in changes to the composition of volatile contaminants. Therefore application of an overall removal credit should be based on the lowest expected removal efficiency, in order to account for difficult to degrade chemicals and the potential for inhibitory interactions between compounds. Based on a review of published literature on biofilter treatment of petroleum hydrocarbons, a minimum expected efficiency of 60% is considered reasonable for difficult to treat compounds such as toluene (Barona *et al.* 2007; Fang and Govind 2007; Zmir et al. 2011), n-hexane (Hasan and Sorial 2010; Zamir et al. 2011; Zmir et al. 2011), methyl tert-butyl ether (Mudliar et al. 2010), or benzene (Namkoong et al. 2001; Pandey et al. 2010). A 60% reduction factor has been applied to long-term air concentration exposure during ex-situ remediation, the biofilter reduction factor was not applied to the short-term air concentration.

A case-study, similar to the case used for the original emission model in Meridian (2011) was used to demonstrate the effect of the biofilter reduction factor on long term exposure concentrations. As discussed above, presence of a biofilter is assumed to reduce long term exposure concentrations by a factor directly proportional to the biofilter treatment efficiency. Use of a biofilter also increases engine combustion emissions of criteria air contaminants by a small factor due to transport of the biofilter to and from the site. Model inputs and outputs for the case study are included in Appendix A.

3.5 Lifecycle Emissions

Lifecycle emissions for biofilters can be considered similarly to how landfill emissions are handled in the Meridian (2011) model, which involves determining the mass of carbon entering the biofilter and then applying a standard conversion to CO_2 factor. As biofilters will be operated aerobically, the calculation of emissions from ex-situ remediation does not change with the presence of a biofilter unit and it is assumed that 100% of contaminants will be converted to carbon dioxide.

Additional consideration for biofilters that require some active component, such as a fan to increase airflow or systems to maintain temperature or humidity has been incorporated into the 'hours of power generation' input ex-situ remediation.

The lifecycle emissions from construction and maintenance of biofilters are expected to be minimal, as soil or waste material is often used for the filter substrate, and indigenous microbes can be used under most conditions (Massabo *et al.* 2007), providing that an acclimatization period with the targeted contaminants is factored into the treatment rate (Delhomenie and Heitz 2005). For calculation purposes, it is assumed that the filter substrate is comprised of natural or waste materials and makes up the bulk of the biofilter mass. The remainder of the biofilter, consisting of the outer shell and and monitoring equipment, is assumed to be reused. Maintenance of the biofilter is

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necessary as bacteria have a tendency to accumulate at phase boundary interfaces (Hanzel *et al.* 2012); however, maintenance activities are not expected to represent a significant source of emissions. In order to account for additional transportation of the biofilter to the site and removal afterwards, if a biofilter is being used it is assumed by default that an additional truck is required to mobilize to the site twice based on the typical size of a biofilter.

4 SPREADSHEET TOOL

A spreadsheet tool was developed for Microsoft Excel that contains the full emission model developed by Meridian (2011). The inputs for the tool require information related to site conditions, contaminant concentrations, and remediation method. Additional secondary inputs for more complex parameters may also be used to refine estimates if site specific data is available. Default and/or recommended values are provided where applicable for input parameters based on expected site conditions in Alberta, current regulatory guidance, and common remedial strategies. The output of the tool will includes a screening-level air quality assessment and comparative life cycle emissions for *ex-situ* treatment and landfill disposal options. The assumptions and operation of the tool are described in the previous Meridian (2011) report, and a User Guidance document is provided in Appendix B.

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