

The potential of using an acoustic source for enhanced remediation of NAPL

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Keywords: acoustic waves; DNAPL; LNAPL; NAPL; in situ soil remediation; optimization; field test

1 Introduction

Many cases of soil contamination involve the presence of light NAPL (LNAPL) or dense NAPL (DNAPL). It is the experience that the remediation of NAPL phases in soil can take decades when conventional pump-and-treat is used. If these phases can be dissolved faster or even mobilised, this could assist (in-situ) remediation efforts.

During the remediation using two-phase extraction, up to 10 times higher removal concentrations of oil components in groundwater were observed when concurrent sonic drilling (200 Hz) occurred on-site.

In the oil industry there has been interest in acoustic stimulation for decades and modest research has focused on the mobilization of oil phases in reservoirs by vibrations for enhanced oil extraction. This follows the observation of mobilizing effects on oil after earthquakes and explosions. The effectiveness of enhanced Oil recovery by applying vibration in the field is not considered to be convincingly proven (Kostrov and Wooden, 2005).

More recently, international interest from the environmental science perspective has focused on the potential of using acoustic waves to improve the extraction of pollutants in soil, such as mineral oil or perchlorethylene (PCE) NAPL. Lab studies from recent international scientific literature show no mobilization, but enhanced dissolution of NAPL (Kim and Wang, 2003; Chrysikopoulos and Vogler, 2004). None of these studies studied the effects of acoustic stimulation in the field. Therefore this study focused on understanding the behaviour of acoustic waves and their interaction with NAPL phases at field sites. In addition, we studied the potential mechanisms to enhance and optimize in-situ remediation approaches using acoustic stimulation.

2 Methods

2.1 Acoustic source

A standard vibrating needle, normally used for the compaction of concrete, was used as an acoustic source. The vibrating needle operates at a fixed acoustic frequency of 200 Hz (12,000 rpm). The vibrating needle is connected through an inverter

to a standard 240V - 50 Hz power supply. The vibrator consists of a rotating clapper in a cylindrical enclosure. The clapper rotates at high speed against the housing and generates a strong vibration in the acoustic range. Seismic receivers were installed to study the behaviour of the vibrations in the soil.

2.2 Acoustic Receivers

For the observation of the vibration intensity two types of vibration sensors were used, namely motion sensors (geophones) placed at ground level and pressure sensors (hydrophones) placed in monitoring wells. The geophones record a vector quantity (particle velocity and direction) and consist of three mutually perpendicularly placed motion sensors. The hydrophones record a scalar variable (pressure disturbance) and have no direction dependency. The sensors transform the pressure or particle velocity to a disruption (electrical) voltage variation over time. The hydrophones register a direct relationship between pressure and amplitude of 25V/bar. The seismic sensors detect a voltage variation which is recorded and stored as a function of time with a seismograph.

2.3 Data Acquisition and Processing

The SUMMIT seismic recording system was used for the data acquisition. Seismic activity was recorded during a number of measurement sessions, both during active vibration and at rest. At each session the seismic activity was repeatedly recorded (at least 4 times) during a few seconds at each sensor. Measurements of seismic activity were recorded when the vibrating needle was at rest to quantify the naturally occurring background noise signal.

2.4 Groundwater sampling

The first sampling round took place before starting any acoustic vibrations were introduced and were used as a reference for the following sampling rounds. Representative groundwater samples were taken from each monitoring well sampling after flushing the well (20-30 L) and then sent to the lab for analyses on oil compounds (LNAPL site) and chlorinated solvent compounds (DNAPL

site). For the DNAPL site also a well outside the zone of acoustic influence (>200 m, outside zone of acoustic influence) was monitored. No concentration changes were recorded for this well.

3 Description of Field Sites

Acoustic field tests of a day were conducted at two locations: a site in the Rotterdam harbour area (LNAPL location) and a site in the eastern part sandy part of the Netherlands (DNAPL location). Goal of the field tests was evaluate the effect of acoustic vibrations on the behaviour of NAPL at contaminated sites with differences in soil structure and contamination type.

3.1 Rotterdam Harbour LNAPL Site

The soil at the Rotterdam site consisted of silty sand with low permeability. Shallow mineral oil contamination existed in both soil and groundwater, with residual (smeared) pure phase NAPL. At the start of the acoustic field test no floating LNAPL layer was detected at the groundwater surface (1.5 m-bsl), but such a floating layer had existed at the site previously. At this site, an acoustic signal of 200 Hz was periodically introduced to the soil using a vibration needle. At this first field site the investigation focussed on the propagation and radius of influence of vibrations and monitoring changes in contaminant concentration. Measurements on the surface using geophones have been carried out in perpendicular directions at distances of 1, 2, 4, 8 and 16 m from the vibrating needle. Measurements in two monitoring wells to horizontal distances of 0.5 and 1.5 m from the vibrating needle were performed at a vertical distance of 1 m.

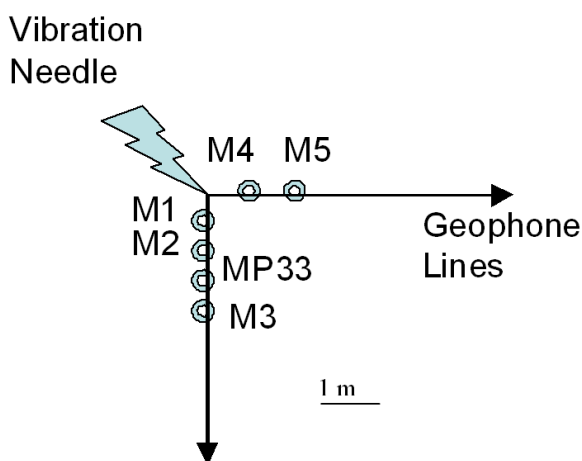


Figure 1: Layout of the acoustic measurements and groundwater monitoring wells for the LNAPL site.

3.2 Eastern Netherlands DNAPL Site

The soil at the site in the eastern part of the Netherlands consisted of coarse sand with a VOCI contamination at depth (0–5 mbsl for the target hotspot). At this site acoustic experiments were conducted at varying frequencies using a modified version of the vibration needle used at the LNAPL site. Groundwater monitoring occurred both near a DNAPL hotspot and downstream in the plume area. At this second field site the investigation focussed on the propagation and radius of influence of vibrations and monitoring changes in contaminant concentration. Unlike the LNAPL site, acoustic signal was continuously introduced into the soil (except during initial recording of the background noise signal) and the input frequencies were varied to establish a more detailed understanding of the method of application of acoustic waves and potential of further optimization of the effectiveness. Measurements on the surface using geophones have been carried in a south-westerly oriented line of 9 geophones.

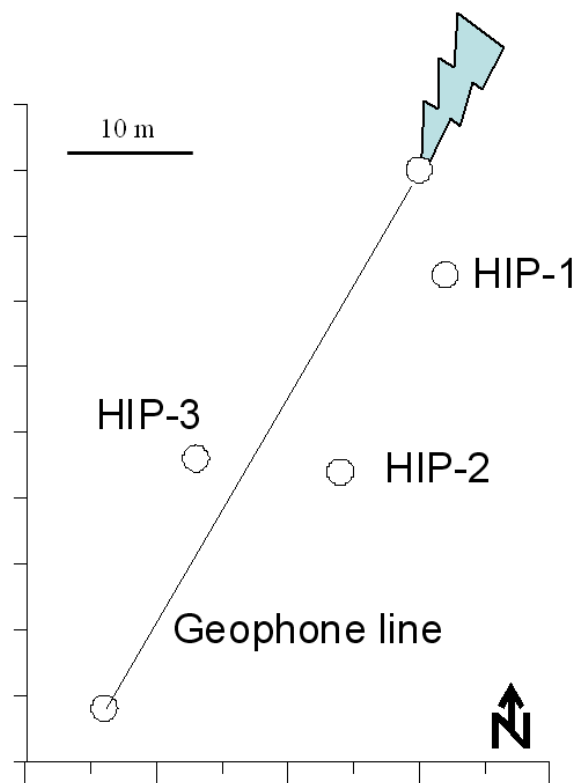


Figure 2: Layout of the acoustic measurements and groundwater monitoring wells for the DNAPL site.

4 Results

4.1 LNAPL Site

The monitoring wells M1-M5 and MP33 are within 4 meters away from the vibration source, well within the observed radius of influence of the acoustic vibrating. Therefore, if the vibration has any effect on the formation of a floating layer or dissolved concentrations, it is expected that these effects are noticeable in these wells.

After all acoustic periods a floating layer remained absent in the monitoring wells sampled. The results of the measured aqueous concentrations in the monitoring wells showed that:

- The highest concentrations of mineral oil components are found in MP3 and MP33, the lowest in M4 and M5
- BTEX at the start is 98% of the total mineral oil components, this does not change during the vibration experiments, ie the less soluble components (> C10) are not preferred mobilized.
- After each stage vibration / sampling stage, the concentrations increased gradually during the monitoring wells.
- The relative increase for the various monitoring wells compared to the initial concentrations of mineral oil components (C6-C40, Figure 10) in order of highest to lowest: M4 (622%), M1 (374%), M5 (319%) M2 (233%), M3 (155%). The concentrations in monitoring well MP33 increased by 113%.
- The relative increase from the initial concentrations of BTEX components in order of highest to lowest: M4 (661%), M1 (382%), M5 (321%), M2 (231%), M3 (153%). The concentrations in monitoring well MP-33 rose by only 115%.
- The monitoring wells showed relative concentration increases of both BTEX and total mineral oil components (C6-C40), consistent with the distance from the location of the acoustic needle, i.e. the relative concentration increases are strongest for the monitoring wells near with the acoustic needle.

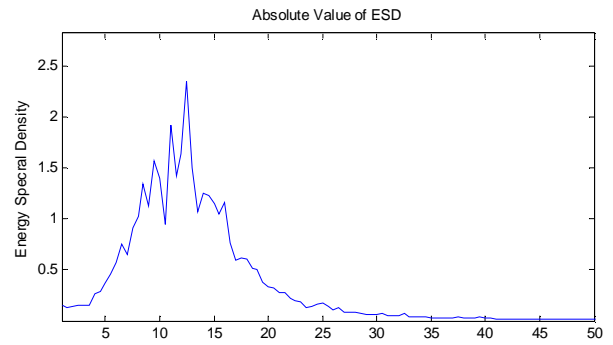


Figure 3: Recorded background frequencies (ambient noise) for the soil at the DNAPL site, indicating a resonance frequency between around 12 Hz.

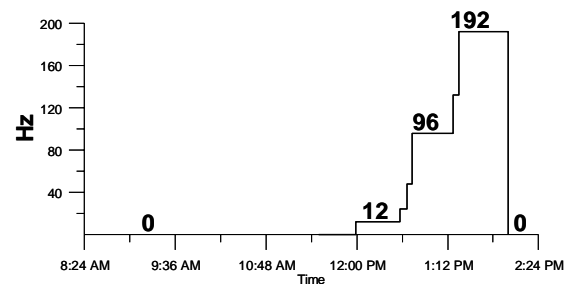


Figure 4: Evolution of applied acoustic frequencies during the field test at the DNAPL site. Frequencies are indicated for the periods when groundwater sampling took place.

4.2 DNAPL Site

4.2.1 Acoustics

The resonance frequency determined for the soil at this location was 12 Hz (Figure 3). The selected frequencies applied for testing at the site were multiples of this resonance frequency (Figure 4). The travel of the various frequencies through the soil is summarized in Figure 5. The results are summarized as follows:

- The observed amplitude of the vibration in the direct vicinity of the acoustic source is strongest above 48 Hz. This mainly reflects the characteristic of the modified vibrating needle used, generating lower amplitudes at lower frequencies (less energy).
- The soil vibrations showed the strongest amplitude at frequencies of 48 and 96 Hz. These frequencies were discernable from the background signal up to a distance of 40 m.
- At the resonance frequency of 12 Hz determined for this soil, the signal is hardly damped over the entire distance covered by the geophone line.
- The vibrations in the soil travel easiest at multiples of its resonant frequency.

- Higher frequencies (at least > 132 Hz) are hardly discernible from background at the furthest geophones and mute much faster than the lower frequencies.
- The vibration at 196 Hz (similar to the frequency used during "sonic drilling") had the smallest radius of influence, up to 25 meters.

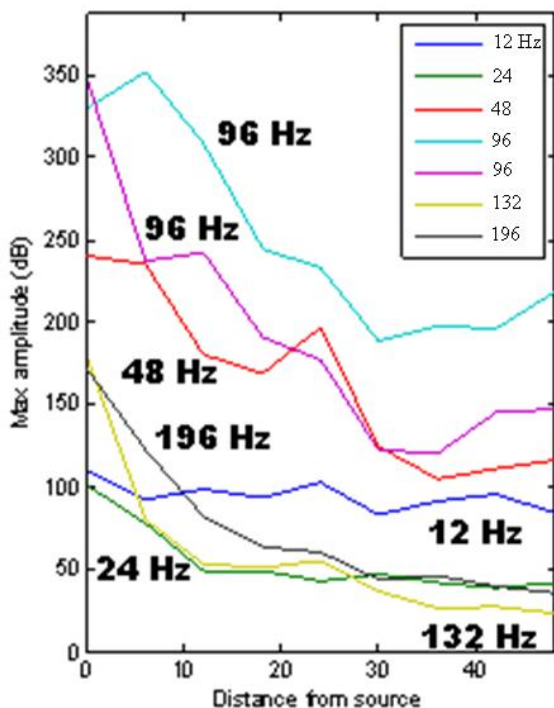


Figure 5: Maximum amplitude measured with increasing distances from the acoustic source operating at different input frequencies.

4.2.2 VOCl concentrations

Before, during and after the acoustic tests, TCE was the VOCl with the highest concentration in all analyses. In addition, cis-DCE and PCE were present in elevated concentrations. 1,1,1-TCA was only found in one monitoring well. 1,1-DCA and 1,2-DCA were not detected in any of the samples analyzed. Compared with the results at the LNAPL location there was little variation (<20%) in concentrations.

All monitoring wells, except reference monitoring well HIP-4, fell well within the range of zone of acoustic influence of 40m (compare Figure 2 and Figure 5). As the TCE concentrations in monitoring well HIP-1 were highest, at 15% of TCE solubility, this indicates the proximity of pure phase DNAPL. At the vibration frequency of 96 Hz, this monitoring well showed the highest concentration of cis-DCE and PCE (Figure 6). This is the vibration frequency with the highest amplitude in the geophone measurements (Figure 5). The concentrations at the other frequencies show no clear deviation from the background situation (0 Hz). Monitoring wells HIP-2 and HIP-3 are indeed in the range of the vibrating needle,

but the observed concentrations are too low to indicate to the proximity of DNAPL. In monitoring well 2 was an upward trend in concentrations observed. However, the concentrations in monitoring wells HIP-2 and HIP-3 do not clearly show up at a certain frequency (Figure 7 and Figure 8).

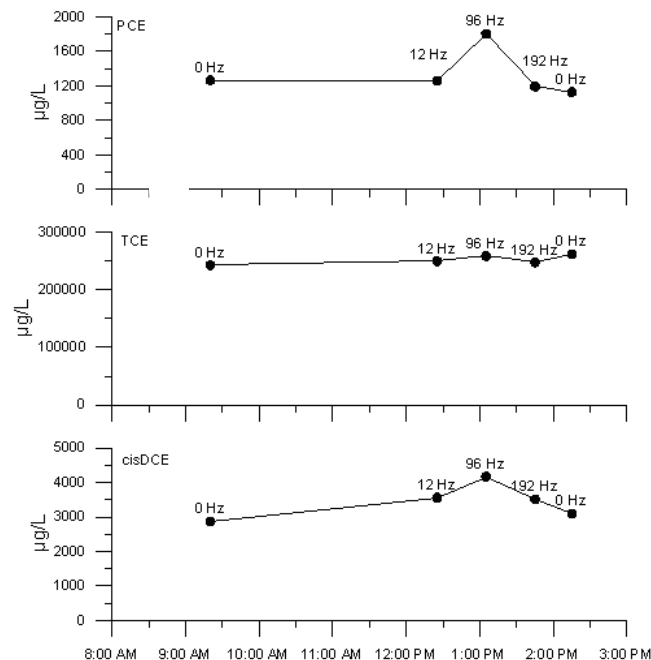


Figure 6: PCE, TCE, and cis-DC during the day for monitoring well monitoring well HIP-1. The labels indicate those frequency vibration was active at the time of sampling

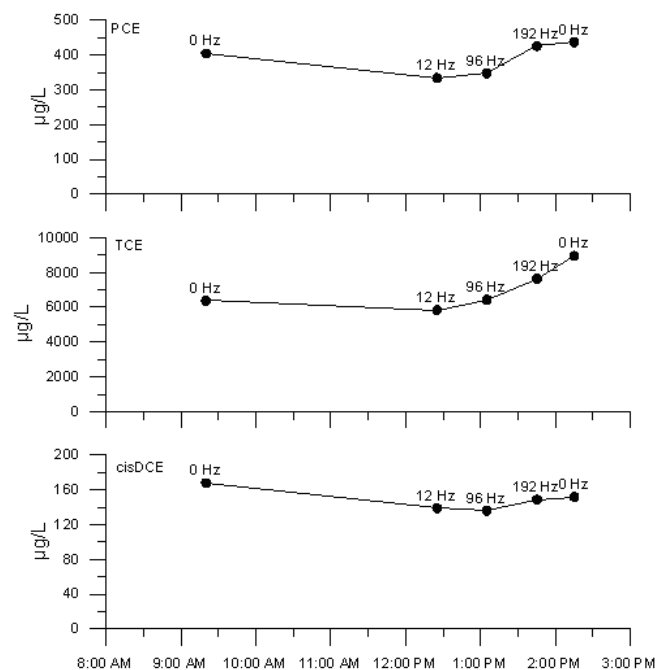


Figure 7: PCE, TCE, and cis-DCE concentrations during the day for monitoring well HIP-2. The labels indicate those frequency vibration was active at the time of sampling

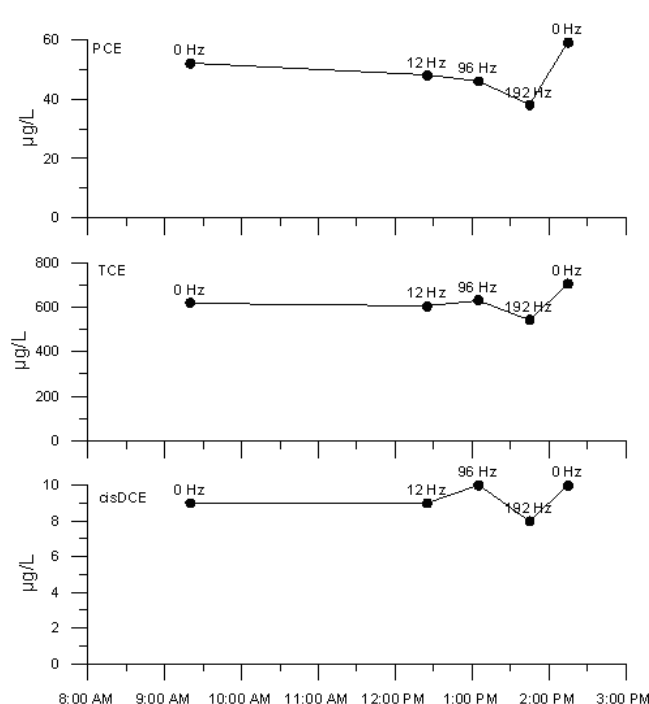


Figure 8: PCE, TCE, and cis-DCE concentrations during the day for monitoring well HIP-3. The labels indicate those frequency vibration was active at the time of sampling

5 Discussion

Although the relative variation in concentration measured over time at monitoring well HIP-1 is similar to those in HIP-2 and HIP-3, the absolute increase of concentrations in monitoring well HIP-1 is significantly greater and only occurs at 96 Hz. This suggests that this is probably due to acoustic disruption of the DNAPL. Although not conclusively demonstrated yet, if we assume that the increased concentration in monitoring well HIP-1 are due to vibrations at 96 Hz, three scenarios can be considered for the elevated concentrations in monitoring well HIP-1:

1. The vibrations resulted in disruption of DNAPL and the mobilized DNAPL droplets were sampled after transport in groundwater;
2. The vibrations resulted in disruption of DNAPL and the mobilization of DNAPL droplets, which dissolved during transport in groundwater before sampling;
3. The vibrations resulted in disruption of a DNAPL. The increased DNAPL-groundwater surface that resulted led to enhanced dissolution of VOCl components.

The observed concentrations of VOCl and their solubilities can be used to assess the most likely scenario. This is because the solubility of VOCl water depends on their solubility product and the

molar ratio present in the DNAPL (Raoult's law). As natural degradation of VOCl was absent due to oxic conditions at this location, the assumption is that cis-DCE was a component in the DNAPL itself. This is not unlikely since both cis-DCE and TCE and PCE component are available as solvents and can be present as minor component in DNAPL. The cis-DCE components VOCl, PCE and TCA are only present in very small amounts (> 2 orders of magnitude lower) in the groundwater than TCE. Based on these concentrations, it is expected that TCE accounts for more than 95% of the DNAPL composition. Therefore, in Scenario 1 an exceedence of the absolute solubility product of TCE (1100 mg / L) would be expected, which is not the case (Figure 6). The concentrations of TCE in monitoring well HIP-1 are 22-23% of the solubility product.

If the mobilized DNAPL droplets would completely dissolve in the groundwater before they reach the monitoring (scenario 2), we would expect that the concentrations in groundwater reflect the composition of the DNAPL. Since the water solubility of cis-DCE (5700 mg / L) is higher and the solubility PCE lower (200 mg / L) than that of TCE, the composition of the DNAPL phase is expected to be poorer in cis-DCE and richer in PCE. This would mean that the ratio between cis-DCE and TCE would go down and the ratio between PCE and TCE should go up. This is not the case, as these ratios both go up (Figure 6).

This suggests that scenario 3, the occurrence of stimulated dissolution of the DNAPL by vibrations, is the most likely scenario. However, it is then expected that all concentrations would rise in approximately equal proportion due to the enhanced dissolution of the DNAPL. This is not the case. The increase in TCE concentration is relatively smaller than the increase of cis-DCE and TCE. The total increase in VOCl concentration is 4%. TCE increases by only 3% while cis-DCE and PCE increase by 130% and 145%. This difference in relative increase may be associated with the breakup of DNAPL (scenario 3). Because cis-DCE and PCE are present in the DNAPL with small proportion relative to TCE (> 95%) t, there may be differences in concentrations within the DNAPL, for example due to diffusion gradients towards the DNAPL-groundwater interface. This could explain how the breaking up of DNAPL and enhanced dissolution can increase the relative concentration in groundwater for components that are present in the DNAPL as traces. So while VOCl concentrations in monitoring well HIP-1 could be caused by enhanced dissolution due to DNAPL disruption at, it is clear that the effects of mobilization of DNAPL was not observed.

Overall our results show that on both the DNAPL and LNAPL site no NAPL phase was mobilized.

However it is likely that both sites NAPL dissolution was enhanced. Based on new insights from the results of the vibration experiment at the DNAPL site, it is expected that the optimal vibration frequency for enhanced dissolution is likely less than 200 Hz. This is due to the lower resonance frequency of soil in most locations in the Netherlands.

With respect to the mechanism of mobilization, it seems unlikely that vibration of small DNAPL droplets themselves could cause NAPL as the required frequencies for droplet resonance would be very high. Simultaneously, the influence radius of the vibration in the ground goes down with higher frequencies. As expected the resonance frequency for a NAPL droplet on a grain scale is thousands of times smaller than the soil as a whole, the radius of influence of such high frequencies is expected to be minimal (mm). A disruptive influence on NAPL phases by acoustic vibration is therefore most likely due to resonance of the soil as a whole. As shown in this study, the frequency at which this occurs also has a much greater radius of influence (10s of meters).

At the DNAPL location likely a slightly enhanced dissolution (3%) of NAPL phase was observed, while at the NAPL site concentration increases of >600% were observed. The extent to which acoustic vibrations can disturb a NAPL, depends, in addition to the oscillation frequency, also on the energy with which they reach the NAPL. This is also dependant on the position of the vibration source relative to the spatial distribution of NAPL phases in the soil of interest. Although the frequency during sonic drilling (200 Hz) is not optimal, the high energy level at which this occurs will likely affect NAPL near the drill location. This probably explains the field observations that led to this investigation. The spatial distribution of NAPL phases, especially those of DNAPL, are often the biggest unknown in practice. Improved understanding of the distribution locations, can assist in the consideration of the (planned) sonic drilling location (s) possible disturbance of the NAPL phases can result. The impact of this disruption is also expected to depend on the degree of saturation in which the NAPL is present in the soil. For DNAPL phases with high pore saturation degrees, including in particular DNAPL pools, possible adverse effects should be taken into account, such as the further sinking of the DNAPL phase. For residual pore saturations it is most likely that the acoustic disturbance will solely lead to increased access of NAPL phase. This can speed up the dissolution. Based on the results of this study this is of unknown a priori impact, and it is potentially not cost effective to solely introduce vibrations in the soil for that purpose. Possibly, however, improved accessibility for remedial agents may improve remediation efficiency. The potential application,

operation and cost of this approach would have to be further investigated.

6 Conclusions and recommendations

Our detailed field studies on the effect of acoustic waves on NAPL in soil indicated that:

- The application of acoustic waves stimulated NAPL dissolution, not mobilisation, at the investigated field sites.
- The increase in relative dissolved contaminant concentrations decreased with distance from the acoustic source
- Optimal frequency for this process is related to the natural soil resonance frequency (e.g. 12 Hz or 96 Hz) and is site-specific
- Acoustic interaction with NAPL did not result in effects that can be ascribed to the mobilisation of NAPL droplets
- Use of acoustic waves at NAPL sites may increase remediation potential

To benefit from possible positive effects sonic drilling should be performed, where possible, to coincide with the active remediation measures (eg NAPL extraction or ISCO). For an improved understanding under what conditions vibration does cause mobilization of NAPL phases, remediation conditions (e.g. the amount of mass removal) before and during active sonic drilling can be compared.

Acknowledgements

This study was performed with the HIP (Holland In-situ Proeftuin) research program. We would like to acknowledge contractor HMVT, Netherlands Centre for Soil Quality Management and Knowledge Transfer (SKB) and the Dutch Ministry of Economic Affairs for financial support. The authors thank Klaas de Jong of HMVT and Arno Peekel of SKB for useful discussion and comments.

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