Effect of Temporal Changes in Air Injection Rate on Air Sparging Performance Groundwater Remediation

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Abstract

Air sparging (AS) is a commonly applied method for treating groundwater contaminated with volatile organic compounds (VOCs). When using a constant injection of air (continuous mode), a decline in remediation efficiency is often observed, resulting from insufficient mixing of contaminants at the pore scale. It is well known that turning the injection on and off (pulsed mode) may lead to a better remediation performance. In this article, we investigate groundwater mixing and contaminant removal efficiency in different injection modes (i.e., continuous and pulsed), and compare them to those achieved in a third mode, which we denote as "rate changing." In this mode, injection is always on, and its rate is varying with time by abrupt changes. For the purpose of this investigation, we conducted two separate sets of experiments in a laboratory tank. In the first set of experiments, we used dye plume tracing to characterize the mixing induced by AS. In the second set of experiments, we contaminated the tank with a VOC and compared the remediation efficiency between the different injection modes. As expected, we observed that time-variable injection modes led to enhanced mixing and contaminant removal. The decrease in contaminant concentrations during the experiment was found to be double for the "rate changing" and "pulsed" modes compared to the continuous mode, with a slightly preferable performance for the "rate changing" mode. These results highlight the critical role that mixing plays in AS, and support the need for further investigation of the proposed "rate changing" injection mode.

Introduction

The presence of volatile organic compounds (VOCs) in groundwater is a significant human health risk (Avramov et al. 2013; Martí et al. 2014). Air sparging (AS) is considered a reliable, proved, and effective method for the removal of VOCs in contaminated sites (Suthersan 1999; Benner et al. 2002; Khan et al. 2004; Da et al. 2015). In AS, air is injected into a well whose screen is located at the saturated zone, below the contaminated zone. The injected air enhances volatilization as it travels through the subsurface pore network (Johnson 1993; Reddy et al. 1995). Air moves laterally from the well and upwards toward the unsaturated zone. Contaminants, once trapped in the liquid phase, are transformed to the gas phase and are transported by the injected air. Typically, a soil vapor extraction (SVE) system is installed in the unsaturated zone, and the extracted air is treated ex situ to remove the contaminants (e.g., using activated carbon filters). In addition to the physical removal of contaminants by AS, the supplement of oxygen to the groundwater promotes biodegradation of organic contaminants (Johnson 1998; Semer and Reddy 1998).

Depending mainly on the medium's particle size distribution and pore geometry, the injected air will move in channel type flow or in the form of discrete bubbles (Ji et al. 1993; Braida and Ong 2001; Selker et al. 2006; Hu et al. 2010), forming a cone or parabolic shape above the injection screen (Reddy et al. 1995; Chao et al. 2008). For example, for fine sands with grain size smaller than 2 mm, the main air flow mechanism was found to be pore scale channeling (Ji et al. 1993; Clayton 1998; Hu et al. 2010).

Typically, above the injection point, a cone shaped zone with multiple air channels is developed. This zone is roughly defined as the zone of influence (ZOI) (Hu et al. 2010). The borders of the ZOI are defined by the channels reaching most laterally far from the injection point. However, due to channelized flow, not all of the volume within the ZOI is actually impacted by the

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Air Injection. The *mass transfer zone* (MTZ) is a thin layer next to a single air channel where VOCs rapidly deplete, while outside this layer, concentration of VOCs remains fairly constant (Braida and Ong 2000, 2001). If the distance between two adjacent channels is larger than twice the width of the MTZ, portions of the aquifer within ZOI will not be affected by AS. This may provide an explanation for the observation of a significant decline in contaminant removal rate under prolonged operation (Braida and Ong 2001).

### Air Sparging Stages

As air is introduced into the saturated subsurface, it displaces groundwater, leading to a local mounding of the water table around the injection well and within the ZOI. When the injected air reaches the vadoze zone, preferred pathways of higher air permeability are formed and the pressure within the flow channels is reduced. Following this decrease in pressure, some smaller channels collapse and eventually steady state conditions are established. It is common to address three distinct stages in AS (Lundegard and Andersen 1996; Gao et al. 2013): (1) an initial transient growing stage where airflow channels expand to the lateral and vertical directions; (2) a second transient stage where the airflow region shrinks and some channels collapse and merge into larger channels—leading to a size reduction of the ZOI in the lateral direction; and (3) a steady state stage during which formed air channels remain practically static as long as system parameters remain unchanged (See Figure 1 for illustration). We denote the characteristic time for the system to reach steady state by \( T \). For example, for the case described in Figure 1, this characteristic time can be estimated as \( T \approx 70 \) [min].

In the third stage, upon reaching steady state conditions, contaminants within the MTZ, in the immediate vicinity of the air channels, are rapidly removed, while beyond this zone, contaminant diffusion from the bulk toward the MTZ becomes the limiting factor for the remediation process (Elder and Benson 1999; Braida and Ong 2000, 2001; Kirtland and Aelion 2000; Peterson et al. 2000; Yang et al. 2005; Baker and Benson 2007). Mei et al. (2002) showed that in the third stage, water motion within the ZOI is negligible.

Once the sparging system is turned off, a “shutdown” stage begins and a temporary depression in the groundwater level (GWL) is observed, as water flows back into formerly air filled pores.

### Effect of Changes in the Injection Rate on AS Performance

Previous studies pointed out pronounced advantages applying AS in a pulsed operation rather than using it in a continuous operation mode (Shah et al. 1995; Johnson et al. 1999; Suthersan 1999; Kirtland and Aelion 2000; Heron and Gierke 2002; Yang et al. 2005; Baker and Benson 2007; Balleke et al. 2009). A field scale study, reported by Heron and Gierke (2002), showed that applying AS in a pulsed mode accelerated the time weighted average mass removal by 40% to 600% (depending on the “aggressiveness” of the pulsing). Yang et al. (2005) reported an increase of the average hydrocarbon removal rate by a factor of up to 3 when compared with pulsed operation and continuous operation. Kirtland and Aelion (2000) found significantly higher mass removal rates of Benzene, Toluene, Ethyl-benzene, and Xylene (BTEX) in pulsed aeration in comparison with continuous operation. Abdel-Moghny et al. (2012) showed similar results when remediating soils contaminated with waste-lubricant oil in a laboratory model.

Pulsed AS utilizes the transient stages of mounding and depression (stages 1 and 2 and the shut-down phase, respectively) to induce groundwater mixing and the consequential redistribution of contaminants relative to air channels (Elder and Benson 1999; Kirtland and Aelion 2000; Heron and Gierke 2002; Baker and Benson 2007). As a result, the mass transfer limitations associated with contaminant removal are reduced. Mixing occurs both at the micro scale as air channels form and collapse, and at the macro scale due to induced back-and-forth groundwater flow within the ZOI and its vicinity (Yang et al. 2005). It seems reasonable to hypothesize that in order to obtain such induced mixing and enhanced remediation, the time interval associated with the pulsation must be of the same order of \( T \), the characteristic time of reaching steady state in the flow.

In this research we suggest, in addition to the continuous and pulsed modes, a different mode for AS which we coin “rate changing” injection mode. In this mode, air injection rate is varied within a given range of injection rates by periodically changing the applied injection pressure. The difference between the “rate changing” mode and the pulsed mode is that in the first, air is constantly injected while changing the rate within the given range, whereas in pulsed operation mode, injection rate shifts between zero and a constant value.
changing” mode relies on earlier research showing that by increasing the injection rate, additional channels are formed, and the ZOI increases (Ji et al. 1993; Semer et al. 1998; Chao et al. 2008). We hypothesize that the new investigated mode may (1) induce mixing of groundwater without the need to periodically shut-down the injection system, and (2) lead to improved contaminant removal rates, when compared to the pulsed mode operation.

Although past research showed that pulsed operation is superior to continuous, limited experimental confirmation of the mechanisms behind this phenomenon exists in the literature. Thus, the objective of this work is to explore and compare qualitatively and quantitatively the effect of the injection mode (i.e., “continuous,” “pulsed,” and “rate changing”) on groundwater mixing and contaminant removal. In order to examine the effect of changes in air injection mode on the remediation effectiveness, we compare both the magnitude of contaminant mixing and the removal efficiency. We investigate this using a quasi-2D transparent laboratory tank, where two sets of experiments are performed. In the first set, mixing is quantified by injecting a small volume of water with dye into the tank, injecting air in one of the three modes, and tracking the spreading of the dye with time. In the second set of experiments, the tank is initially contaminated with a constant known concentration of VOC. Then, the temporal variations of the VOC concentrations are measured in the pore water in various locations in the tank. This allows to directly quantify the removal efficiency.

Experimental Procedures

To study AS under different injection modes, a transparent tank with dimensions $50 \times 50 \times 2$ [cm$^3$] was constructed to allow visualization of the multiphase flow during sparging (Figure 2). The tank was packed with glass beads in the diameter range of $0.8 - 1$ mm. The average porosity was measured as $\emptyset = 0.35 \pm 0.01$. The tank was initially saturated by water. Compressed air was injected from the bottom center of the tank using a cylindrical diffuser (Aignep S.P.A, BS Italy) with a shaft length of 28 mm and a diameter of 15 mm, made from polyethylene porous material with pore diameter smaller than 75 $\mu$m. Injection pressure was regulated using a digital pressure controller, Alicat model PC-30PSIG-D (Tuscon, Arizona) and injection rates were measured using a Dwyer model VBA flowmeter (Chicago, Illinois). The transparent tank was illuminated from behind.

Two types of experiments were conducted in the experimental tank: dye spreading, denoted herein as hydrodynamic experiments, and VOC removal experiments. Details are given in the following.

Hydrodynamic Experiments

To qualitatively assess the mixing phenomena resulting from AS under different injection modes, we performed dye tracer experiments. We used a non-reactive blue food dye (Maimons food industry Inc.). Prior to initiation of AS, a small volume of the dye solution was injected at a point outside the anticipated air plume (Figure 2a). Dye movement during the sparging experiment was recorded by acquiring images using a digital camera. Image analysis was performed using Matlab (The MathWorks Inc., Natick, Massachusetts).

Air injection in the 2D hydrodynamic experiment was comprised of two stages. In the first stage, air was injected at a constant rate for 60 min (continuous mode). In the second stage, air injection varied in one of the two modes—pulsed or “rate changing”—for an additional 60 min (see Table 1). The various injection pressures and corresponding rates comprising the “rate changing” mode are illustrated in Figure 3. The different injection modes were tested for two general cases: “low” injection rates, $Q_{\text{max}} \approx 1.75$ LPM (Liter per minute) and “high” injection rates, $Q_{\text{max}} \approx 10$ LPM, where $Q_{\text{max}}$ is the maximal injection rate. The temporal average of the
VOC Removal Experiments

The injection rate in all time-variable modes (i.e., pulsed and “rate changing”) was approximately 0.5 $Q_{\text{max}}$. The injection pressures were 4.76 to 5.72 kPa and 4 to 7 kPa, respectively, for the low and high injection rates. Note that the pressure corresponding to 0 LPM differs between low and high injection experiments (4.76 vs. 4 kPa). This can be explained by the air channel “memory”: during subsequent sparging cycle, trapped air provides less resistance to flow than the original water filled pores. Thus, an initial higher injection pressure lowers the entry pressure in the following cycles (Baker and Benson 2007).

We conducted preliminary experiments and observed that the characteristic time of reaching steady state in flow in the experimental tank after initiating air sparging was $T \approx 20$ [s]. Therefore, in both time-variable modes, we adopted an interval length of the same order, namely 1 min. In part, the reason for this choice is technical: The water sampling procedure takes almost 1 min, and thus we were able to complete the sampling during a single interval of injection. Hence, in the pulsed mode, 1 min of injection is followed by 1 min of shut-down (no injection; $Q = 0$). In the rate-changing mode, the injection rate was changed abruptly every minute as depicted in Figure 3. The injection scheme was repeated periodically, with a cycle time of 6 min. The rationale behind the specific sets of injection rates was to have a significant variation in pixel intensity due to changing air saturation with each lasting 4 h (see Table 2). Water samples were taken at times $t = 0, 1, 2,$ and $4$ h and analyzed by gas chromatography (for details on the sampling and the analysis see Appendix A). The VOC experiments were carried out only for the “low” injection case, due to technical restrictions. Three repetitions were conducted for each mode. After each experiment, the unit was washed by 5 volumes of DI water and was then air dried.

### Results

**Hydrodynamic Experiments**

The results of the hydrodynamic experiments are given for two general cases: high and low injection rates. In each three injection modes were explored. In Figure 3c, we plot the injection rate vs. injection pressure, which are approximately linearly correlated for the considered injection rates.

Acquired images of the dye plume were analyzed by decomposing into the RGB (red, green, and blue) channels, as shown in Figure 4. For the analysis, it was assumed that the concentration ($C$) of the dye is linear with the pixel intensity of the green component ($I_G$). This rather crude assumption does not take into account the effect of changes in air saturation on the intensity, as the random formation of air channels within the domain did not enable us to calibrate for this effect. However, the variation in pixel intensity due to changing air saturation was limited to $\pm 5\%$. We used the red component ($I_R$) to delineate the edge of the plume, due to the high sensitivity of the red component to low concentrations, and its low sensitivity at high concentrations. Thus, the dye concentration (with arbitrary units) is approximated by the following expression:

$$C = \begin{cases} I_{G,0} - I_G & : I_R \leq I_{R,0} \\ 0 & : \text{otherwise} \end{cases}$$

where $I_{R,0} = 125$ and $I_{G,0} = 160$ are the red and green background intensity values, respectively. The zero subscript is used here to mark that these values approximately correspond to $C = 0$.

In Figure 6, we plot the resulting concentration map images in the region enclosed by the white rectangle in

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**Table 1**

Parameters of the Hydrodynamic (Dye Spreading) Experiments

<table>
<thead>
<tr>
<th>#</th>
<th>Experiment name</th>
<th>$Q_{\text{max}}$ [LPM]</th>
<th>$0 &lt; t &lt; 60$ [min]</th>
<th>$60 &lt; t &lt; 120$ [min]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Pulsed/low</td>
<td>1.75</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Rate changing/low</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>Pulsed/high</td>
<td>10</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>Rate changing/high</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Continuous mode, $Q = Q_{\text{max}}$
Table 2

Parameters of the VOC Removal Experiments

<table>
<thead>
<tr>
<th>#</th>
<th>Experiment Name</th>
<th>$Q_{\text{max}}$ [LPM]</th>
<th>$Q(0 &lt; t &lt; 240 \text{[min]})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>Continuous mode (VOC)</td>
<td>1.75</td>
<td>$Q = Q_{\text{max}}$ (continuous)</td>
</tr>
<tr>
<td>6</td>
<td>Rate changing mode (VOC)</td>
<td>1.75</td>
<td>Rate changing</td>
</tr>
<tr>
<td>7</td>
<td>Pulsed mode (VOC)</td>
<td>Pulsed</td>
<td></td>
</tr>
</tbody>
</table>

Figure 5. The images are shown for both the high and low injection rates. The first 60 min of all cases were conducted in continuous mode, and so, for simplicity, only one set of results is shown. At $t = 60$ [min], the mode changes from continuous to either pulsed or “rate changing.”

As seen in Figure 6, under continuous injection mode the plume grows slowly, and after 60 min of injection changes in the dye plume are relatively small. For “low” injection rates (Figure 6a), the size of the plume in the end of the continuous stage shows no significant changes, and for “high” injection rate a moderate increase is seen (Figure 6b). Switching to pulsed injection mode at $t = 60$ [min], the plume grows rapidly and after 20 min of pulsed sparging (i.e., at $t = 80$ [min]) a significant increase in its size is observed. A similar behavior is seen when switching to “rate changing”—the plume grows rapidly compared to the continuous mode.

In order to compare the dye spreading dynamics under each of the three modes (i.e., continuous, pulsed, and “rate changing”) we define a dimensionless area index ($AI$). $AI$ is a metric of the size of the plume, and is defined as the number of pixels ($\Sigma np_R$) within the plume (where $I_R > I_{R,0}$) divided by the total number of pixels in the frame ($\Sigma np_{tot}$):

$$AI = \frac{\Sigma np_R}{\Sigma np_{tot}}$$  \hspace{1cm} (2)
Figure 4. RGB intensity (I_{R,G,B}) along a single horizontal line of pixels across the dye plume. The red component was used to define the plume limits, due to its high sensitivity at low concentrations. Then, the green component was used for quantifying the concentration within the plume limits. As seen in this figure, it is difficult to correlate the blue component with dye concentration; therefore it was neglected in the analysis.

$$E = \exp \left(- \int_{\Omega} p(x,t) \ln p(x,t) \, dv \right)$$  

(3)

where $\nu$ is the total volume of the domain and $p(x,t)$ is the concentration distribution function normalized by the total mass of the solute:

$$p(x,t) = \frac{c(x,t)}{\int_{\Omega} c(x,t) \, dv}$$  

(4)

Figure 7 shows the dilution index and area index during the different injection modes in the hydrodynamic experiments.

It is clear from looking at these figures that DI and AI are roughly linearly related, with some possible deviation at late times. This observation is consistent with the mathematical definition of these metrics (see details in Appendix B). With this similarity between the two metrics in mind, we shall refer in the following only to DI for brevity, and assume it is a good metric of the mixing in the system as a whole.

During the continuous injection mode (first 60 min of injection), we observe an increase in the DI. This increase is relatively rapid in the first minutes after the air injection starts, and later on slows down considerably. However the DI keeps on slowly increasing. At $t = 60$ [min], the injection mode is changed to a transient injection mode (pulsed or “rate changing”). After this change, DI increases rapidly for both modes, but not at the same rate: the DI increases faster in the pulsed injection mode, relative to the “rate changing” mode. This effect is more prominent for the low injection rates, compared to the high injection rates (Figure 7). The results clearly indicate that both transient injection modes produce increased mixing rate in the system relative to the continuous mode. This increased mixing rate is more evident in the pulsed mode.

VOC Removal Experiments

Three repetitions were conducted in each of the VOC removal experiments. Concentrations of toluene in the samples are given in Table 3 for one of the repetitions as a representative case (for a quantification of the relative variability in measured concentrations at the three types of wells among repetitions see Appendix A). Note that the “center” well could not be sampled during sparging, as it is was located right above the air diffuser and was dry due to high air content. The normalized concentration $C/C_0$ (normalized by the initial concentration at a given sampling point) is shown in Figure 8.

Toluene concentrations for the “near” wells are shown in Figure 8a. Toluene concentrations in these wells decline in an almost similar way in the pulsed and “rate changing” modes. After 240 min (4 h) of injection, for the pulsed and “rate changing” mode, toluene concentration is dropped to 16% to 17% (respectively) of the initial value. For continuous injection mode, toluene is only moderately reduced to 59% of the initial concentration. The removal
efficiency is, accordingly, 84 and 83% for pulsed and “rate changing” modes, respectively, and just 31% for continuous mode.

The concentrations in the “far” wells (shown in Figure 8b) show no substantial reductions over time under all injection modes. For all modes, after 120 min of sparging, concentration is reduced by 3% to 8%, only to increase back to nearly the initial concentration when the injection is terminated (240 min). This phenomenon is well documented in the literature as the tailing effect, where when the sparging is turned off, untreated water flows from beyond the ZOI toward the injection point, leading to an increase in concentrations (Braida and Ong 2001). Indeed, we observe that the “far” wells are located about 5 cm outside the ZOI in these experiments. The overall removal rate after 4 h of sparging is 0 to 5% with superior performance for the “rate changing.” However, due to the significant relative error in these measurements, a clear trend cannot be concluded.

Finally, in Figure 8c we compare the average concentration of all wells (near, far and center wells) at the termination of the experiments. The “rate changing”

Figure 6. Computed dye plume concentration, \( C \) (Equation 1), within the white rectangle shown in Figure 5, for (a) “low” Injection rates \( (Q_{\text{max}} = 1.75 \text{ LPM}) \) and (b) “high” Injection rates \( (Q_{\text{max}} = 10 \text{ LPM}) \).
and pulsed sparging modes show the best results with reduction of VOC by 53 and 48%, respectively. Thus, the “rate changing” mode seems to have a slightly better remediation performance. Continuous injection results in only 33% removal of VOC. Keeping in mind that the average injection rate was nearly double in the continuous case (see Figure 3), it is clear that the efficiency of continuous sparging is substantially low, compared to the other two modes.

Conclusions

As seen from the results, continuous sparging is characterized by a relatively static condition and a limited movement of water, except for the very beginning of the injection. This was illustrated in the dye plume spreading experiments, by the slow dilution and limited movement of the dye tracer. In the VOC removal experiments, continuous sparging resulted in relative low remediation efficiency.

Switching to a time-dependent injection scheme (pulsed or “rate changing” mode) resulted in a significant increase in the plume dilution rate in the dye spreading experiments. Comparing the pulsed mode and the “rate changing” mode, we see that the pulsed mode resulted in a faster dilution of the dye tracer. By contrast, VOC removal experiments showed that the remediation efficiency is slightly higher for the “rate changing” mode, compared to the pulsed mode. At this stage, we cannot provide a complete explanation to this seemingly contradiction in the results. We can merely hypothesize that in the “rate changing” mode, the added variety of injection rates (six discrete values vs. two in the pulsed mode) leads to more variability in the size of the ZOI, and more variability in the location of air channels, which enhances the mixing of the water within the ZOI, homogenizes it, and leads to better remediation efficiency. This claim is somewhat supported by the smaller deviation of the measured VOC concentrations at $t = 240$ [min], as depicted in the error bars in Figure 8.

We emphasize here that the relative dilution was determined on the basis of the spreading of a dye tracer injection at just one point adjacent to the edge of the ZOI.

Our basic hypothesis in this research is that a rate changing mode can be superior in terms of remediation, relative to pulsed mode. The work described here is a
first attempt to study this assertion. Indeed, our results show that a slight increase in remediation efficiency is obtained in the rate changing mode; however, we anticipate that additional research (e.g., using a 3-D, large scale apparatus, and/or numerical simulations) is in place to further test this conclusion.

In this work, we focused on a case of a single well. In many field applications, multiple wells are used to inject the air. In such cases, a transient injection mode may be applied in such a manner that, for example, the total air injection rate in all wells will be constant, but the rate in each specific well will be time variable. Such coordinated time-variable rate can result in intermittent groundwater flow between the wells and this may enhance the mixing in the system and ultimately lead to a substantial increase of the remediation efficiency. In this context, the single well experiment we described here is a first step toward a better understanding of a multi-well system.

Finally, we would like to shortly address the cost-benefit of the “rate changing” approach. In a multi-well setup, the system can be designed such that the total discharge of air in all wells is constant, and the fraction injected in each well is time-dependent (i.e., rate changing or pulsed). It is reasonable to assume that the head losses in the distribution junction are similar for pulsed and rate changing approaches. Then, the energy cost to operate a rate-changing system is expected to be similar to that required to operate a pulsed system. On the other hand, the initial cost of the “rate changing” system may be higher because a special regulator is needed to generate multiple injection rates in all the wells.

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**Supporting Information**
Additional Supporting Information may be found in the online version of this article:

**Appendix A.** Sampling and analysis of toluene in water samples.
**Appendix B.** Comparison of the dilution index and area index, for the case of an instantaneous point source.
**Appendix C.** Analysis of dye plume transport during air sparging.

**References**


