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Evaluation of Packed Towers for Removing Volatile Organics from Surface Waters

James W. Moore University of Arkansas, Fayetteville

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EVALUATION OF PACKED TOWERS FOR REMOVING VOLATILE ORGANICS FROM SURFACE WATERS

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Arkansas Water Resources Research Center University of Arkansas Fayetteville, Arkansas 72701

Arkansas Water Resources Research Center

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ABSTRACT

EVALUATION OF PACKED TOWERS FOR REMOVING VOLATILE ORGANICS FROM SURFACE WATERS

This study analyzes the potential of packed tower aeration as a remedial treatment process for the removal of trace organics on either an acute or chronic basis. Both pilot-scale installations $\,$ of the packed tower process were reviewed. Included are basic modeling considerations for the removal of volatile trace organics from raw water. Included also is an assessment of the state of the technology as applied to air stripping of water.

James W. Moore

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Keywords -- Packed Tower/Aeration/Surface Water/Water Treatment

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INTRODUCTION

The presence of various organic compounds in water supply sources has been a topic of increasing concern for the past decade. Although the principal focus of the attention has been on surface water supplies because of their perceived greater susceptibility to contamination by organic compounds, evidence has been accumulating that groundwaters are also subject to contamination. Recent sampling and analyses of groundwater supplies have indicated that many of these supplies contain a variety of organic chemical compounds. Since about eighty percent of all public water supplies in the United States utilize groundwater, the presence of various organic compounds in both ground and surface water supplies is of considerable concern.

The organic compounds include a variety of materials originating from agricultural, industrial, and waste disposal activities as well as from other sources. Although these compounds are ordinarily present in small concentrations, their presence may be significant because of toxicity or potential carcinogenicity considerations and because conventional water treatment plant processes are unable to remove them.

Because of the refractory nature of the trace organic compounds to most water treatment processes, one or more additional processes may be required to remove the compounds. Currently, the use of activated carbon in granular carbon columns is the most widely recognized treatment procedure. For retrofitting existing treatment plants, the use of the granular activated carbon columns will require repumping

the entire water flow through the treatment plant unless the columns are used as the initial treatment process. Usage as the initial process is not particularly advantageous because of the presence of numerous other materials, particularly in surface water sources, which will decrease the efficiency of the granular activated carbon columns. Consequently, the use of some other process more suitable as an, or the, initial process offers several advantages. Packed towers may be suitable for reducing the concentrations of the volatile trace organics and may be useful as the initial process.

Packed towers have been used for many years in the chemical process and air pollution control industries. In chemical process industry applications, generally the concentrations are much greater than encountered in water treatment applications. Additionally, the chemicals involved are usually more volatile than in the applications of interest for this project. Packed towers used in air pollution control applications are usually used as absorbers to transfer materials from the gas stream into the liquid. Consequently, the use of packed towers for reducing the concentrations of volatile trace organics in water treatment is a relatively new concept. There have been several recent installations for reducing the concentrations of several organic chemicals in groundwater usually as a remedial procedure.

The concept of the packed tower is relatively simple. The tower basically consists of a vessel filled with some type of media which will provide a relatively large surface area yet which has a large

void ratio. The large surface area is needed to allow the mass transfer of the chemical species from the gas to the liquid (absorbers) or from the liquid to the gas (strippers). The large void ratio is needed to provide a reasonably small headloss across the tower. Generally, it is also desirable to select a media which has a low weight to minimize structural costs.

A. Purpose and Objectives

The purpose of the research project was to evaluate the potential of packed towers for reducing the concentrations of selected trace organics from raw water. The potential usefulness included both sole and combined packed tower-granular activated carbon column applications.

B. Related Research or Activities

The use of absorbing and stripping devices for potable water treatment applications in the form of aerators has been practiced for many years. Examples include the absorption of oxygen from air for oxidizing iron and manganese, and the stripping of hydrogen sulfide, ammonia and methane from water. These applications include a variety of equipment such as waterfall aerators, spray nozzles, cascade aerators, multiple tray and plate aerators, diffusion or bubble aerators and mechanical aerators. For example, some water treatment plants have used spray systems in a fountain arrangement. The spray produces relatively small droplets of water which yields a large droplet surface area per unit volume. For the purposes for which the spray devices have been employed, the surface area of the droplets has been

adequate for transferring oxygen to water. Although not used as frequently, the spray systems would be useful for stripping highly volatile materials, such as ammonia and methane from water.

Similarly, the use of bubble aerators for diffusing air bubbles in water has been practiced in some water treatment applications, although less frequently. By utilizing small bubbles, a relatively large mass transfer surface area is obtained through which the contaminant can be reduced in concentration for stripping applications, or the oxygen concentration increased for absorption applications. Plate and tray towers operate on the principle of obtaining thin streams of water, and water droplets, to achieve the necessary surface area for transfer of the chemical species desired.

The spray, sparger, plate tower and tray tower systems have been used principally for either the transfer of highly volatile substances such as oxygen, ammonia, hydrogen sulfide and methane and/or in applications where high transfer efficiencies are not required. Their application for the stripping of moderate and low volatility substances is limited, particularly in instances where high removal efficiencies are required.

Packed Towers

The basic concept of packed towers is not new. Until recently, however, their principal applications have been for the transfer of species present in relatively large concentrations and for the transfer of highly volatile substances. The applications have changed somewhat by the need for devices to absorb air pollutants from vari-

ous gas streams. For example, packed towers have been utilized for the removal of sulfur dioxide from flue gas streams. Although the sulfur dioxide is highly volatile, the application of the packed tower concept for the smaller concentrations involved in trace organic chemicals is relatively new.

Figure 1 shows a cross-sectional view of three types of packed towers. In most applications, the tower is randomly packed with a light weight material which provides a large surface area, but which also has a large void ratio. The large surface area is necessary to obtain the mass transfer efficiencies needed. The large void ratio is needed to achieve the relatively low headlosses across the tower.

Figure 1. Schematic Cross-Sectional Views Of Packed Towers.

A variety of packings have been developed over the years. Tellerite, Raschig rings, Berl saddles and Intalox saddles are examples of packings which have been available for many years. Other packings have been developed in recent years.

Packed Tower Design

For stripping applications in which it is desired to remove one or more volatile contaminants from a solution, the operating line is below the equilibrium line as shown in Figure 2. The stripping of carbon dioxide, hydrogen sulfide, and volatile trace organics from water serve as examples of the process. For absorption applications, the operating line is selected so that it is above the equilibrium line as shown in Figure 3.

The overall approach used in designing packed towers is to determine the number of transfer units required which, when multiplied by the height of each transfer unit, yields the overall packing height. The calculation is as follows:

 $Tower Height = HTU \times NTU$ (1) where: HTU = the height of each transfer unit $NTU = the number of transfer units$

The height of each transfer unit can be defined for the gas and liquid phases as:

$$
HTG = \frac{G}{F_GA} = \frac{G}{k_{ya}(1-y)_{\text{IM}}}
$$
 = $\frac{G}{k_{G}ap_t(1-y)_{\text{IM}}}$ (2)
\nwhere: $y = \text{concentration of the contaminant in the gas, \text{ mole fraction}$
\n $G = \text{superficial molar mass velocity of the gas} \text{ k}_{y} = \text{gas mass transfer coefficient} \text{ a} = \text{specific interface surface area/packed} \text{ volume}$
\n $F_G = \text{gas-phase mass transfer coefficient} \text{ p}^{\text{obs}} = \text{total pressure}$

Figure 2. Operating and Equilibrium Lines For A Packed Tower Used In A Stripping Application.

Figure 3. Operating and Equilibrium Lines For A Packed Tower Used In An Absorption Application.

\n
$$
H_{\text{t}} = \frac{L}{F_{\text{t}}} = \frac{L}{k_{\text{r}}a(1-x)_{\text{t}}} = \frac{L}{k_{\text{t}}ac(1-x)_{\text{t}}}
$$
\n

\n\n where: $L = \text{total molar liquid rate}$ $F_{\text{t}} = \text{liquid-phase mass transfer coefficient}$ $k_{\text{r}} = \text{liquid mass transfer coefficient}$ $a = \text{specific interface surface area/packet}$ $x = \text{concentration in the liquid, mole fraction}$ $k_{\text{t}} = \text{liquid mass-transfer coefficient}$ $(1-x)_{\text{t}} = \text{logarithmic mean of 1-x and 1-x}_{\text{i}}$ \n

Corresponding equations for the gas phase are:

$$
N_{LG} = 2.303 \int_{\log y2}^{\log y1} \frac{y}{y-y_1} d \log y + 1.152 \log \frac{1-y_2}{1-y_1}
$$

and:

$$
N_{\rm{t}}L = \int_{x_2}^{x_1} \frac{dx}{x_1 - x} + \frac{1}{2} \ln \frac{1 - x_1}{1 - x_2} \tag{5}
$$

Two Resistance Theory.

The overall resistance to mass transfer between the two phases can be considered to be the sum of the gas phase and liquid phase resistances. In equation form, the expression would be as follows:

Total Resistance = Gas Phase Resistance + Liquid Phase Resistance (6)

If the resistances are defined as the reciprocals of the rate constants for the liquid and gas phases, the resulting equation would as follows (assuming phase equilibrium is governed by Henry's law at the interface):

> 1 = 1 + 1 (7) Ka K_La H_CK_Ga where: K = the overall coefficient K_I = liquid phase coefficent K_G^- = gas phase coefficient H $_{\rm C}$ = Henry's constant a = specific interface surface area/packed volum

The ratio of resistances can be given by:

$$
\frac{R_L}{R_G} = \frac{H_C K_G a}{K_L} + \frac{H_C K_G}{L_L}
$$
 (8)

and:

$$
\frac{R_{\underline{L}}}{R_{\underline{T}}} = (1 + \frac{K_{\underline{C}}}{H_{\underline{C}}K_{\underline{G}}})^{-1}
$$
 (9)

If the term R_L/R_G is very much greater than 1 the gas phase resistance can be ignored in some circumstances. If the resistances are approximately of the same magnitude, both must be considered. Models For Predicting Mass Transfer Rate Constants

Several models have been developed for predicting mass transfer rate constants. Among these are the Sherwood-Holloway, Shulman and Onda models. The Sherwood-Holloway model neglects the gas transfer resistance. Thus, it would be considered a one resistance model. The Shulman and Onda models evaluate both the gas phase and liquid phase resistances and, consequently, are considered two-resistance models.

Sherwood and Holloway evaluated the desorption of oxygen, hydrogen and carbon dioxide from water in countercurrent flow packed columns using a variety of packing materials over a relatively wide range of liquid and gas flow rates. Since the solutes were all gases with relatively large Henry's constants, the liquid resistance controlled mass transfer rather than gas resistance. Consequently, the transfer rate constant was correlated with the liquid rate and liquid-phase properties only as shown:

$$
\frac{k_{\rm L}a}{D_{\rm L}} = 10.746c \left(\frac{0.3048L_{\rm M}}{\mu_{\rm L}} \right)^{1-n} \left(\frac{\mu_{\rm L}}{\rho_{\rm L}D_{\rm L}} \right)^{0.8}
$$
(10)

where: $D =$ molecular diffusivity μ = fluid viscosity $p =$ fluid density α & n are empirically determined packing parameters

The Shulman model involves separate estimation of K_L , K_G and a. For the liquid phase coefficient, a relationship between the Sherwood number, the Reynolds number and the Schmidt number was developed:

$$
\frac{k_{\rm L}d_{\rm s}}{D_{\rm L}} = 25.1 \left(\frac{d_{\rm s}L_{\rm M}}{\mu_{\rm L}}\right)^{0.45} \left(\frac{\mu_{\rm L}}{\rho_{\rm L}D_{\rm L}}\right)^{0.6}
$$
\n(11)
\nwhere: $K_{\rm L} =$ liquid phase coefficient
\n $D_{\rm L} =$ molecular diffusivity
\n $d_{\rm S} =$ diameter of a sphere having the same surface
\narea as a unit of packing
\n $L_{\rm M} =$ liquid flow rate
\n $\mu_{\rm L} =$ fluid viscosity
\n $\rho_{\rm L} =$ fluid density

The gas phase coefficient is shown as follows:

$$
\frac{k_{\rm C}d_{\rm e}}{D_{\rm G}} = 1.195(1-\epsilon)^{0.36} \left(\frac{d_{\rm e}G_{\rm M}}{\mu_{\rm G}}\right)^{0.64} \left(\frac{\mu_{\rm G}}{D_{\rm G}\rho_{\rm G}}\right)^{0.33} \tag{12}
$$

where: $K_G = gas$ phase coefficient $d_{\mathcal{S}}$ = diameter of a sphere having the same surface area as a unit of packing G_M = gas flow rate μ_{G} = fluid viscosity P_{G} = fluid density D^-_G = molecular diffusivity

Both the gas and liquid phase coefficients involve relationships between the same dimensionless groups, the Sherwood, Reynolds and

Schmidt numbers. However, the liquid phase coefficient is proportional to the square root of the molecular diffusivity, whereas the gas phase coefficient is proportional to the 2/3 power of the molecular diffusivity.

The Onda model also entails separate estimation of K_G , K_L and a. For this model, the specific interfacial area is the specific wetted packing area, a_w . The specific wetted packing area is estimated as a function of the liquid flow rate, packing properties, and liquid properties.

$$
\frac{a_w}{a_t} = 1 - \exp[-1.45(\sigma_e/\sigma_L)^{0.76} \times [L_M/(\sigma_u\mu_L)]^{0.1}[L_M^2\sigma_t/(\rho_L^2g)]^{0.06}[L_M^2/(\rho_L\sigma_L\sigma_t)]^{0.7}] \qquad (13)
$$
\nwhere:
$$
a_t = \text{total specific surface area of packing}
$$
\n
$$
\begin{array}{rcl}\n\sigma_C &=& \text{critical surface area of packing} \\
\phi_L &=& \text{liquid density} \\
\sigma_L &=& \text{liquid surface tension}\n\end{array}
$$

The Reynolds, Froude and Weber numbers are included in the equation for estimating the specific wetted packing area. The correlation for the liquid phase coefficient is shown as follows:

$$
k_{\mathrm{L}}\left(\frac{p_{\mathrm{L}}}{\mu_{\mathrm{LS}}}\right)^{1/3} = 0.0051\left(\frac{L_{\mathrm{M}}}{a_{\mathrm{w}}\mu_{\mathrm{L}}}\right)^{1/3}\left(\frac{\mu_{\mathrm{L}}}{\rho_{\mathrm{L}}\mu_{\mathrm{L}}}\right)^{0.8}(a_{\mathrm{t}}d_{\mathrm{p}})^{0.4}
$$
(14)
where: $K_{\mathrm{L}} = 1$ iquid phase coefficient
 $d_{\mathrm{p}} = \text{nominal packing size}$
 $\rho_{\mathrm{L}} = 1$ iquid density
 $\mu_{\mathrm{L}} = 1$ iquid viscosity
 $L_{\mathrm{M}} = 1$ iquid flow rate
 $a_{\mathrm{w}} = \text{specific wetted packing area}$
 $D_{\mathrm{L}} = \text{molecular diffusivity}$

For the gas phase coefficient, the relationship is as follows:

$$
\frac{h_0}{a_t D_0} = 5.23 \left(\frac{G_M}{a_{t^1 G}} \right)^{0.7} \left(\frac{\mu_0}{\rho_0 D_0} \right)^{1/3} (a_t d_p)^{-1}
$$
\n(15)

where:
$$
K_G
$$
 = gas phase coefficient
\n a_t = total specific surface area of packing
\n D_G = molecular diffusivity
\n G_M = gas flow rate
\n μ_G = gas viscosity
\n ρ_G = gas density
\n d_D = packing diameter

In addition to these models, a variety of other single resistance models has been developed.

METHODS AND PROCEDURES

Both literature review and laboratory resources were used in evaluating the potential of packed tower aeration for stripping organic chemicals from water. The methods currently available to limit organic chemical concentrations in water supplies can be divided into two principal categories. These are: 1) control by management of the resource, and 2) control by reduction in the water treatment process.

Dyksen and Hess, 1982, identified three management techniques for reducing or eliminating the compounds from the water source. These are: 1) elimination of the source of the compound; 2) location of a new water supply source; and 3) blending of existing water supply sources. The first technique usually requires regulatory activities above the local level. The remaining two techniques are site specific in their application and are, thus, dependent on the availabil-

ity of a new water supply source, the availability of other water sources or both.

With respect to control of organic chemical concentrations by treatment, three general approaches are available. These are stripping of the organic chemicals by aeration, adsorption of the organic chemicals, and combined systems including both aeration and stripping.

Historically, aeration equipment used in water treatment has been either waterfall or spray aerators and diffused aerators. Included in the waterfall or spray aerator category are multiple tray, cascade, spray nozzles and packed column aerators. These operate on the principle of creating droplets, thin streams or thin films of water surrounded by air. The objective is to develop a large surface area of exposure between the air and water through which the contaminant can be stripped. The diffused air systems operate on the same principle of developing a large surface area for mass transfer except that air is bubbled through the water column.

The mass transfer efficiencies of aeration equipment used in water treatment for both stripping and absorption have not been of great concern for several reasons. Among these are the relatively low capital and operating costs of the equipment and the highly volatile nature of the substances which have ordinarily been transferred. For example, aeration is frequently used for transferring oxygen from the air into water for oxidizing iron and manganese. Since oxygen is very volatile and is present in large concentrations in the air, the

transfer efficiencies have been within acceptable ranges even for the less effective processes. Similarly, aeration can be used to strip ammonia, hydrogen sulfide, carbon dioxide, and methane from water. These are also very volatile substances. Consequently, the transfer efficiencies have also been acceptable. Many of the organic chemicals which may require removal have either intermediate or low volatilities. Consequently, greater mass transfer efficiencies will be required for acceptable performance. These greater mass transfer efficiencies will not only require optimal design of the aeration process, but will eliminate many of the aeration processes traditionally used.

With respect to operating characteristics, the two extremes in aeration process design are the diffused aerator and the packed tower aerator. Both have the capability of achieving the large surface to volume relationships and the detention times required for more efficient mass transfer. However, the diffused aerator accomplishes the task by injecting air bubbles in a column of water. Mass transfer of the substances occurs as the bubbles rise to the surface. The smaller the bubble size, the larger the area to volume relationship and, generally, the more efficient the process. The detention time is a function of the depth of the water column. That is, the length of time the air bubbles are in the water column. Within limits, the mass transfer efficiency is a function of the detention time.

The principal disadvantage of the diffused aeration process for removal of moderate and low volatility substances is the pressure

drop across the process. That is, the air must be compressed sufficiently to overcome the head of the column of water. Thus, as the detention time is increased to obtain greater efficiencies, the pressure losses also increase resulting in relatively large power costs. The process does have the inherent advantage of keeping suspended materials in suspension because of the turbulence created in the basin. Thus, application of the process on relatively turbid surface water sources does not create settling problems.

The packed tower process offers several advantages for stripping applications. These include the relatively low pressure loss across the tower, large surface area to volume ratios, variable flow rate ranges, and corrosion resistant packings. Usually the tower is operated in the countercurrent flow mode with the water flowing down through the tower by gravity and the air forced upward through the tower by pressure differential. Several full-scale packed tower systems have been constructed and operated for the reduction in concentration of organic substances in groundwater. Schilling, 1985, described a packed tower application for the reduction of $1, 1, 2, 2$ -tetrachloroethane, 1,2-trans-dichloroethylene, trichloroethylene and te trach loroe thylene in Washington. He reported removal efficiencies of about ninety-five percent for the $1, 1, 2, 2$ -tetrachloroethane and essentially complete removal of the other volatile organic substances. The $1,1,2,2$ -tetrach loroethane concentrations in the contaminated water were in the 17 to 300 parts per billion range. This application was designed to block a plume of contaminated water from

flowing into the well field used as a source of supply for the City of Tacoma.

A similar application for groundwater quality control was conducted by the U.S. Air Force at Wurtsmith Air Force base to control a plume of contaminated groundwater, Houel, et al., 1979.

Several laboratory studies have been conducted using packed towers for stripping various organic chemicals from water. Houel, et al., 1979, reported the results of stripping studies of chloroform. They reported removal rates of 97.5 percent and greater with residual concentrations of less than 0.2 micrograms per liter. Stallings, Rogers and Mullins, 1984, reported the results of a field pilot-scale investigation. They reported removal efficiencies of greater than ninety percent for groundwater containing sixteen different volatile organic chemicals, including hydrocarbons, chlorinated organics and aromatics. They concluded that the selection of the "best" packing material would not be solely be based on performance criteria, but rather would involve a comprehensive economic analysis to compare associated system capital and operating costs. Umphres, et al., 1983, presented the results of two pilot studies which evaluated trihalomethane removal by packed tower aeration.

Packed Tower Design

Although a variety of numerical models have been developed for packed tower design, their development was largely based on more volatile materials than the trace organic chemicals of interest in this study. For example, Sherwood and Holloway studied the stripping

of hydrogen, carbon dioxide and oxygen in countercurrent flow packed columns. These gases all have Henry's constants greater than one. For substances with smaller Henry's constants, however, the gas phase resistance rather than the liquid phase resistance may control. Generally, applications of the two-dimensional models have been more successful than the liquid phase resistance one-dimensional models. At the present time, pilot-scale studies are ordinarily conducted to develop design data for full-scale systems when either large removal efficiencies are required or when intermediate or low volatility substances are to be removed.

A variety of packing materials have been developed over the years for packed towers. These include Raschig rings, pall rings, Berl saddles, Intalox saddles, Tellerite, as well as newer packings. Saddle packing was used in the Tacoma, Washington system, Schilling, 1985. Pall rings were used in the Wurtsmith Air Force base groundwater cleanup system. Packing selection is still a function of the pilot-scale studies including the associated economic considerations.

PRINCIPAL FINDINGS AND SIGNIFICANCE

The concept of using packed towers for removal of selected volatile organic chemicals as part of the water treatment process is a valid one. Whether the process is used alone, or in conjunction with carbon (or other) absorption, is dependent on both the organic chemicals to be removed and site specific considerations. Although not the sole determinant of the removal characteristics of a specific organic chemical, the volatility of the substance is usually an

important consideration. Henry's constants have not been reported for several of the organic substances of interest. Procedures for determining Henry's constants have been identified.

The design of full-scale packed towers for removal of intermediate and low volatility organic chemicals should be based on pilotscale data at the present time. This includes packing selection as well as the other design parameters. Additional data is needed to allow full-scale design from existing one- and two-dimensional models for economically efficient design. Additional information is also needed concerning the difficulties which may be encountered in applying the packed tower process as the inital treatment process in relatively turbid waters.

CONCLUSIONS

The packed tower process has considerable potential for use in removing volatile organic chemicals in water as part of the treatment process. The application of the process is necessarily a site specific consideration. Depending on the specific circumstances, the process may be used either in conjunction with granular activated carbon columns or as a stand alone process for stripping certain substances from the water.

At the present time, pilot scale studies are needed to determine design criteria for full-scale systems when substances of intermediate and low volatility are to be stripped. When the process is applied correctly, high removal efficiencies have been obtained at relatively low cost. A major advantage of the process is the rela-

tively low capital and operating cost for circumstances where volat ile organic substances must be routinely stripped.

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 $\sim 10^{11}$

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