

# Efficiency of Landfill Leachate Treatment by Freeze Crystallization and Natural Process of Snow Metamorphism

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in collaboration with

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## ABSTRACT

In the past, various attempts have been made to apply, in full-scale, the beneficial effect of the freeze crystallization phenomenon. However, in most of the studies, the process has not been widely accepted because of its high-energy consumption of the refrigeration system, or, in the case of freezing in natural conditions, because of the problem with distribution and freezing of large volumes of wastewater. These problems can be avoided by freezing the wastewater in the form of man-made snow.

The process responsible for segregation of chemical elements and nutrients in the snow pack, as well as its concentration, is snow metamorphism. Snow, as a thermodynamically unstable material, has the ability to change its physical characteristic even at very low temperatures. Fresh man-made snow is a composition of frozen droplets and in macro scale, is dissimilar to natural snowflakes. However, the process of metamorphism is governed by the same mechanisms. Based on this process, as well as the phenomenon of ions elution and its concentration in melt water, authors of the following paper hypothesized that the snow metamorphism process could be applied to treat/concentrate landfill leachate.

After wastewater atomization, the ice crystals first form from the pure water at the surface of each droplet, and trap dissolved compounds such as salts and gases, as well as solid particles in the ice globule. Growing further ice crystals, incorporate water molecules and concentrate compounds in the remaining liquid inside the droplet. As the ice crystal grows, the free space for liquid decreases and the concentration at the growing ice front is higher than in the bulk water. When the solubility of the gaseous compounds reaches the maximum and oversaturation takes place, gas bubbles nucleate and are trapped between the growing ice crystals. An analogous situation occurs with other compounds and some salts may precipitate. The pressure of liquid inside the ice droplet increases with the decreasing volume of remaining liquid. The ice globule may fracture and the concentrate could be released outside. This gives additional benefit for further concentration because the concentrate is removed from the inside of the droplets on their surface. At that moment, the separation and concentration is in macro scale of each droplet and the contaminants are equally distributed in the snow pack profile. However, soon after the man-made snow is deposited on the ground, the temperature gradient is established in the snow pack profile. The mechanism of snow metamorphism, based on vapor pressure gradient, takes place and is enhanced by fluctuations in ambient temperatures. Some ice grains or portions sublime and the water vapor condenses as pure ice on the surface of the ice grains located in the area with lower vapor pressure. As a result, solid particles and ionic compounds, as well as gas bubbles (volatile compounds of wastewater) are easily excluded and released from the ice. Most of the contaminants are deposited on the surface of the ice grains in the pores of the snow pack. When the melting begins, the rejected and accumulated soluble impurities on the ice crystals are similarly washed down as "cake" in a filtration process. Volatile compounds are stripped away during atomization and/or separated in frozen droplets as gas bubbles, and slowly released during snow metamorphism and melting.

The efficiency of concentration for non-volatile compounds is described as follows:

$$E_{(i)} = 1 - a(1 - V_c) - bd(e^{(-V_c/d)} - e^{(-1/d)});$$

Where: a, b, d = coefficients of concentration (based on experimental data).

Concentration of compound “i” in the effluent melt water and in the concentrate (Figure 1) can be calculated from the following equations:

$$C_{E(i)} = \frac{1 - E_{(i)}}{1 - V_c} C_{0(i)} \quad C_{C(i)} = \frac{E_{(i)}}{V_c} C_{0(i)}$$

Where:

$C_{E(i)}$  = Average concentration of compound (i) in effluent [mg/l];

$C_{0(i)}$  = Concentration of compound (i) in raw wastewater [mg/l].

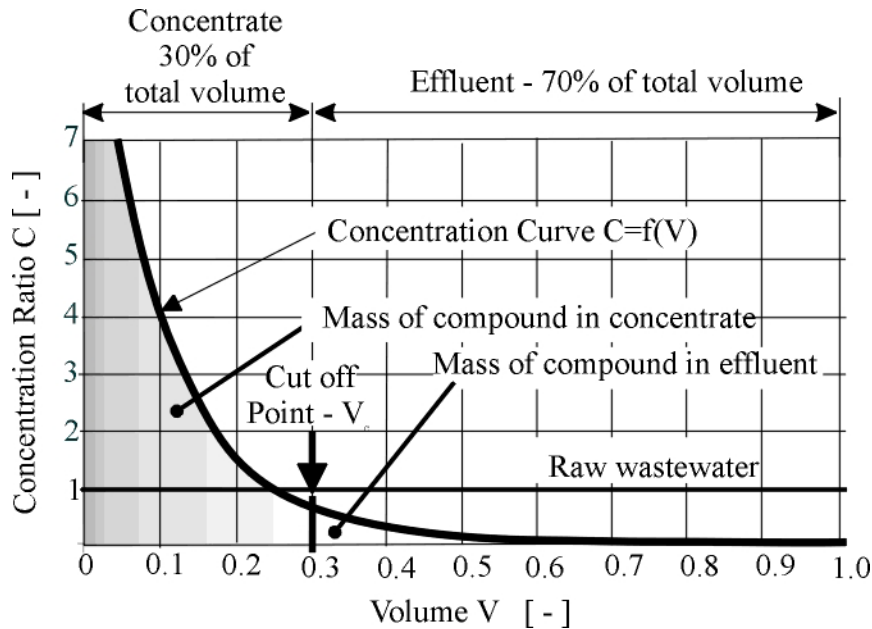


Figure 1 Concentration Curve

Selected results from concentration of landfill leachate in lab scale experiments are presented. An elution of anions and cations, as well as other parameters during the melting process, were monitored. A high concentration of contaminants was reported in the initial runoff of melt water. Most toxic elements and organics were concentrated in the first melt water runoff. The efficiency of the process was different for different elements. Sulphate was generally not detectable in the melt effluent. Very high efficiency of the treatment was also noted for chloride and such compounds as Boron, Potassium and Sodium. More than 95% of these elements were removed from the leachate. An increase in BOD, COD, DOC and TOC, in the first melt was very high and reached values of 92%, 94%, 95% and 97%, respectively. Satisfactory concentration efficiency of Barium was also reported (94%). Other elements such as Aluminum, Chromium, Nickel, Zinc and Iron were concentrated to 93%, 90%, 93%, 89% and 92%, respectively. Mercury, in raw leachate was 0.0007mg/L and was concentrated in the first 20% of melt to the value of 0.003 mg/L. In further samples of effluent, Mercury was below Method Detection Limit. The concentration efficiency of Copper was at the range of 77%. Lower concentration of 21% was reported for Fluoride. Fluoride is easily incorporated into the ice structure and thus its concentration was restricted.

The freeze crystallization process improves clarity of effluent. The colloidal particles undergo natural agglomeration and are easily separated from the melt water stream. The atomizing freeze crystallization process can be an excellent pretreatment unit system in winter operation or may work as a separate independent unit.