



# NITROGEN REMOVAL PRR

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While the BOD and suspended solids removal capability of lagoon systems have been reasonably well-documented and reliable designs are possible, the nitrogen removal capability of wastewater lagoons has been given little consideration in system designs until the past ten years or so. Yet, E. Joe Middlebrooks<sup>(1)</sup>, along with a few other "pond scientists" devoted considerable attention to this subject for the past 20 years. With tightening regulations, their findings are now even more significant. This short paper consists mainly of excerpts from the work of Middlebrooks, and his colleagues: Sherwood

C. Reed, Abraham Pano, and V. Dean Adams.<sup>(2)</sup> It also addresses the functions of the

Progressive Reaeration and Recirculation Process (**PRR™ Process**) as provided by the Sunflo<sup>2</sup> technology and how, based on Sunflo<sup>2</sup>, scientists' translation of research by Middlebrooks, et al, these functions affect nitrogen removal in a stabilization pond environment.

Nitrogen is the major component of the air mixture of gasses. Just over 78% of dry air is oxygen /or nitrogen. Nitrogen is an indispensable element of the biosphere, the shell of living material around the earth. All living matter contains nitrogen in many complicated compounds, and all living matter requires nitrogen (nutrient) compounds in its food.

Despite the fact that nitrogen is not an abundant element, there is about 20 million tons of it in the air above each square mile of the earth's surface; far more than enough to supply the nitrogen needed.

Nitrogen is somewhat inert and is reluctant to combine with other elements to form ions that dissolve in water for plant food. We are fortunate that nitrogen is not more reactive; if it were, all the oxygen in the air would combine with nitrogen.

Because of the difficulty in gaining nitrogen compounds, nature passes its combined nitrogen from one living form to another in a complicated cycle. Nitrogen is absorbed as nitrates and ammonium salts into plant roots. In the plants, the nitrogen is incorporated into amino acids and proteins. Animals eating the plants gain the proteins for themselves. As one animal eats another, nitrogen-containing proteins are distributed throughout the animal kingdom. Dead and decaying plant and animal tissues and wastes are decomposed by bacteria to ammonia ( $NH_3$ ), amino acids and urea, which in turn are decomposed to nitrites, then to nitrates, which are absorbed by plants to begin the cycle again. This cycle is generally represented in Figure 1.

But the cycle is inefficient. Much of the ammonia from decaying proteins, as is the case in wastewater stabilization ponds, is lost to the atmosphere. Certain bacteria convert the nitrogen in amino acids to nitrogen gas, ( $N_2$ ), which is also lost to the atmosphere. This "inefficiency" by nature is good, when it comes to wastewater treatment as high concentrations of ammonia and/or nitrogen gas stimulate the growth of weeds, algae, and cause other treatment problems. Excess nitrogen and nitrogen compounds may reduce the desired treatment functions in a pond and adversely affect the medium that receives the effluent from that pond.

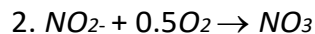
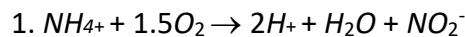
Nitrogen removal from wastewater as well as freshwater can be critical in many situations since ammonia-nitrogen (ammonia-N) in low concentrations can adversely affect young fish in receiving waters, and the addition of nitrogen to surface waters can cause eutrophication. Eutrophication is often the demise of small freshwater recreation lakes and protected bays that experience heavy nitrogen (or nitrogen compounds) inflows. Also, nitrogen is often the controlling parameter for design of land treatment systems. Any nitrogen removal in the preliminary lagoon units can result in very significant savings in land costs for the final land treatment site.

However difficult, there is evidence that nitrogen and nitrogen compounds can be controlled in a pond environment. Several US Environmental Protection Agency (EPA) sponsored studies of the late 1970's demonstrated that significant nitrogen removal does occur in lagoon systems. Early investigation indicated that removal rates were in some way related to pH, detention time, and temperature in the lagoon system. Subsequent studies show that under ideal conditions, up to 95% nitrogen removal can be achieved from facultative wastewater stabilization lagoons.

To appreciate how nitrogen may be controlled in a pond environment, one needs to review the rather complex interactions in which nitrogen participates while in the pond. In a stabilization pond, organic nitrogen (nitrogen associated with carbon compounds) and ammonium-nitrogen enter the pond with the influent wastewater. Organic nitrogen in fecal matter and other organic materials in the wastewater undergo conversion to ammonia ( $NH_3$ ) and ammonium ion ( $NH_4^+$ ) by microbial activity. Given enough time and under steady-state conditions, ammonia in water exists in two forms: ammonium ion and un-ionized ammonia.

At a high pH (low acid, high base), most of the ammonia solution is in the un-ionized form, whereas at a low pH (more acidic), the ammonia is mostly in the ionic form. The ionic form is more easily chemically combined into other compounds such as oxides. The ionic form is also preferred by nitrifying bacteria.

Although nitrogen does not readily react with other elements, oxygen on the other hand is quite aggressive. The reaction of oxygen with nitrogen compounds (oxidation) is referred to as "nitrification." It is a two-step process. The first is the aerobic process of ammonium ion oxidation to nitrite ( $NO_2^-$ ). The second involves the subsequent nitrite oxidation to nitrate ( $NO_3^-$ ). The stoichiometric (electron-balanced) equations for nitrification are:



These chemical reactions will not occur however, without the help of certain bacteria. Within the nitrification reactions, the specific bacteria of the genera Nitrosomonas and Nitrosococcus, play important roles in the first step, and Nitrobacter and related chemolithotrophic organisms (derive their energy from the oxidation of reduced inorganic compounds, such as ammonia, nitrite, and sulfide) carry out the second step. In addition, heterotrophic (organisms that require organic carbon as their carbon source for the formation of cell tissue) nitrification by bacteria and fungi contributes significantly to these processes in more acidic (low pH) environments where the chemolithotrophic nitrifiers are less able to function.

Neither the Nitrosomonas nor Nitrobacter bacteria (the predominate type of organisms that facilitate nitrification) originate in wastewater. These bacteria, along with many others, are found in the top two to three inches of soil throughout the world and are normally introduced (seeded) in the ponds via inflow and infiltration. These bacteria do not reproduce fast enough to maintain adequate populations until the water temperature is above 22<sup>o</sup> C. A continual seeding source is therefore essential for nitrification. As the source of nitrifying bacteria as well as their reproduction is inhibited in cold weather, the rate of nitrification predictably decreases with lower temperatures and nearly ceases at 4<sup>o</sup>C and below.

When they are introduced through inflow and infiltration, there is evidence that the relatively low mixing energy in an unassisted lagoon system allows most of the nitrifying bacteria to settle in the bottom. However, these organisms require fixed biofilm and aerobic conditions in which to become established.

Some investigators have concluded that nitrification in a typical facultative wastewater treatment pond is restricted by the very limited aerobic surface available for attachment and growth of a nitrifying biomass. Most of the bottom surface in an unassisted pond is anoxic or anaerobic, precluding nitrification over most of the submerged surface area. This means that, under ordinary circumstances in an unassisted stabilization pond, the necessary aerobic environment for the nitrifying biomass will be limited to the upper inside slopes of the berm, beneath the pond surface.

When there is adequate biofilm and the required bacteria are present, the availability of oxygen becomes a limiting factor. Since carbon is more reactive with oxygen than is nitrogen, the carbonaceous biochemical oxygen demand (CBOD) or "first stage" reactions must be satisfied before nitrification can take place. Assuming there are resources of oxygen sufficient to satisfy the first stage demands, the temperature of the wastewater, pH and alkalinity, influent flow, soluble BOD5 concentrations, and load variability also influence the rate of nitrification.

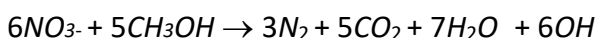
In any case, the first requirement is the development and sustenance of nitrifying organisms. Besides oxygen, the growth of nitrifiers depends primarily on the soluble organic concentration in the location of the biofilm. Typically, heterotrophic bacteria will dominate when the organic concentration is high. Heterotrophic bacteria thrive in biofilm surrounded by aerobic, low pH (more acidic) concentrations of wastewater. Field data obtained from rotating biological contractor nitrification designs indicate that nitrification is typically observed when the soluble BOD5 declines to 15 mg/l, and maximum nitrification occurs when soluble BOD5 concentrations drop to 10 mg/l or less.<sup>(3)</sup> Based on this finding, one might expect that when soluble BOD5 concentrations are higher than 15 mg/l in the vicinity of the biofilm (on the inside slopes of the berm), little or no nitrification will be observed.

Because there are a number of conditions required to promote nitrification, one can conclude that nitrification in any appreciable amounts will depend upon a controlled environment. Oswald found that oxidation to nitrate of ammonium rarely occurs in stabilization ponds because it is either lost to the air, assimilated by algae, or precipitated during periods of high pH before nitrification can become established.<sup>(4)</sup>

The nitrate produced in the nitrification process, as well as a portion of the ammonium produced from ammonification, can be assimilated by organisms as a nutrient to produce cell protein and other nitrogen-containing compounds. Nitrogen assimilation occurs when inorganic nitrogen (not associated with carbon, i.e.,  $NH_3$ ) is used as a nutrient and incorporated into new microbial biomass. The ammonium ion,  $NH_4^+$ , because it is already reduced, can be directly incorporated without major energy costs. However, when nitrate,  $NO_3^-$ , is assimilated, it must be reduced, wherein the oxygen is stripped off, with significant energy expenditure. In this process, nitrite may also accumulate as a transient intermediate.

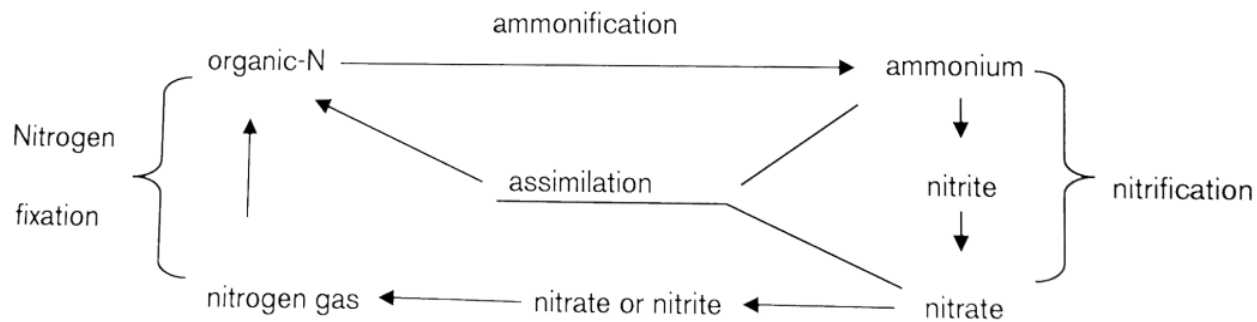
The removal of oxygen, or "denitrification," involves a different set of environmental conditions. In this case, nitrate is used as an oxidant in anaerobic respiration and usually involves heterotrophs such as *Pseudomonas denitrificans*. The major products of denitrification include nitrogen gas and nitrous oxide, although nitrite can also accumulate. Nitrite is of environmental concern because it can contribute to the formation of carcinogenic nitrosamines.

The nitrate is utilized in denitrification to form nitrite and then nitrogen gas. Several bacteria may be involved in the denitrification process including: *Pseudomonas*, *Micrococcus*, *Achromobacter*, and *Bacillus*. The overall denitrification reaction is:



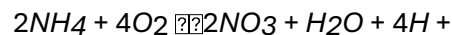
When elemental nitrogen, such as nitrogen gas ( $N_2$ ), is combined with carbon to form organic nitrogen, it is said to have been "fixed". This is an important step in the nitrogen cycle and is accomplished by certain bacteria (cyanobacteria) and algae. This makes the nitrogen available again as a nutrient completing the cycle shown in Figure 1.

**Figure 1. A simplified nitrogen cycle schematic.**



Nitrite and nitrate reduction is ordinarily not important in properly loaded ponds, because these anions are not present in significant concentration in domestic water supply, nor are they formed from ammonia in appreciable concentrations in heavily loaded stabilization ponds. The predominance of nitrate nitrogen in wastewater generally indicates that the waste has been stabilized.

However, at low organic loading rates, after the CBOD has been satisfied, nitrification can be expected, exerting a significant demand on the supply of molecular oxygen. The following reaction indicates that approximately 4.6 pounds of oxygen are required to convert one pound of ammonium-nitrogen to nitrate-nitrogen:



Besides this heavy additional demand on the oxygen resources, nitrates can be used by algae and other plants to form plant protein. Death and decomposition of the plant again yields ammonia. Since nitrogen in one form or another competes for precious oxygen and can perpetuate algal and other perhaps undesirable growth within the pond, and because it may have an adverse affect on the environment to which it is discharged, its removal or reduction from the pond is desirable.

As nitrogen concentrations in effluent becomes increasingly important, so has investigations into removal mechanisms. Various investigators have suggested that algae uptake, sludge deposition, adsorption by bottom soils, nitrification, denitrification, and loss of ammonia as a gas to the atmosphere (volatilization), account for nitrogen removal from stabilization ponds. Evaluations by Pano and Middlebrooks<sup>(5)</sup>, EPA<sup>(6)</sup>, Reed<sup>(7)</sup>, and Reed, et al<sup>(8)</sup>, suggest a combination of factors may be responsible, with the dominant mechanism (under favorable conditions) being volatilization, with the relative magnitude of losses as follows:

- 1 Gaseous ammonia ( $NH_3$ ) stripping to the atmosphere,
- 2 Ammonia assimilation in algal biomass, and
- 3 Biological nitrification-denitrification.

Middlebrooks suggests that, as evidenced by the low concentrations of nitrates and nitrites in pond effluents, nitrification generally does not account for any significant portion of ammonia-N removal. The assimilation of ammonia-N into the algal biomass depends on the biological activity in the system that in turn is affected by temperature, organic load, detention time, and wastewater characteristics.

The rate of gaseous ammonia losses depends mainly on the pH value, surface-to-volume ratio, temperature, and the mixing conditions in the pond. Ammonia-N exists in aqueous solutions as either the ammonium ion or ammonia, depending on the pH of the solution. At a pH value of 8.0, approximately 95% of the ammonia-N is in the form of ammonium ions ( $NH_4^+$ ). However, alkaline pH shifts the equilibrium equation;  $NH_3 + H_2O \leftrightarrow NH_4^+ + OH^-$ , toward the un-ionized, gaseous ammonia production. The shift appears to proceed rapidly, considering the fact that at a pH of 8.5, a majority of the ammonia-N is in the form of dissolved gas ( $NH_3$ ). The presence of algae in a pond can dramatically increase the pH value, especially in the stratum where algal growth is stimulated.

While experiments indicate that the pH must be greater than 8 for any appreciable formation of gaseous ammonia, mixing and temperature are also important factors. Mixing affects the magnitude of the mass transfer coefficient. Temperature affects both the equilibrium constant and the mass transfer coefficient.

Middlebrook's investigations indicate that at low temperatures, when biological activity decreases and the pond contents are generally well mixed from wind (or mechanical assistance), ammonia stripping will be the major process for ammonia-N removal in facultative wastewater stabilization ponds. He advances equations that demonstrate the importance that pH, temperature, and hydraulic loading rates have on ammonia-N removal via gassing off to the atmosphere.

The ammonia stripping in lagoons may be expressed by assuming a first-order reaction (Stratton).<sup>(9)(10)</sup>

The mass balance equation is:

$$VdC/dt = Q(C_0 - C_e) - kA(NH_3)$$

where,

$Q$  = flow rate ( $m^3/d$ ),

$C_0$  = influent concentration of  $NH_4^+ + NH_3$  (mg/l as N),  $C_e$  = effluent concentration of  $NH_4^+ + NH_3$  (mg/l as N),

$C$  = average pond contents concentration of  $NH_4^+ + NH_3$  (mg/l as N),  $V$  = volume of the pond ( $m^3$ ),

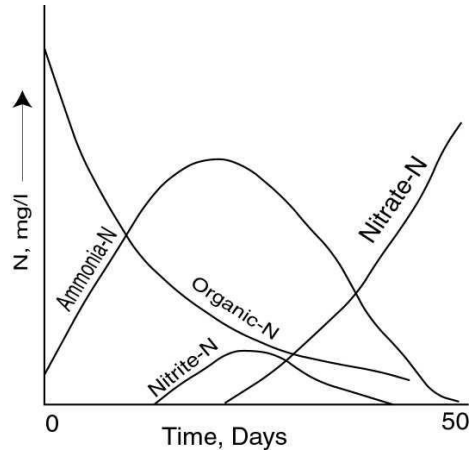
$k$  = mass transfer coefficient (m/d),

$A$  = surface area of the pond ( $m^2$ ), and

$T$  = time (days).

Figure 2 represents the various forms that nitrogen typically assumes over time in wastewater ponds. One can see that generally, ammonia-N, organic-N, and nitrite-N are minimal after 50 days, while nitrate-N concentrations begin to increase after about 25 days and continue to increase significantly thereafter. Depending on the temperature, loading and oxygen availability, total nitrogen in a quiescent pond reaches its lowest concentration at about 30 days.

**FIGURE 2. CHANGES OCCURRING IN FORMS OF NITROGEN PRESENT IN A POND ENVIRONMENT UNDER AEROBIC CONDITIONS.**



As algal growth is an important factor in pH concentrations, its control in a pond is desirable. The availability of nitrogen can be a limiting factor for the primary productivity of algae. However, at the same time, photosynthetic activity of algae in lagoon systems raises the concentration of the hydrogen ion,  $H^+$ , significantly, shifting a large fraction of the ammonia to an un-ionized state that is readily stripped (gassed off) to the atmosphere. In photosynthetic metabolism by algae, carbon dioxide is removed from the dissolved phase, decreasing the concentration of hydrogen ions and bicarbonate alkalinity. The effect of the resultant decrease in bicarbonate ( $HCO_3^-$ ) concentration is to decrease the total alkalinity of the pond.

The decreased alkalinity associated with photosynthesis will simultaneously reduce the carbonate hardness present in wastewater. Because of the close relationship between pH and photosynthetic activity, considerable diurnal fluctuation in pH is observed.

Ammonia assimilation into algal biomass depends on the biological activity in the system and is affected by several factors such as temperature, organic load, detention time, and wastewater characteristics. Oswald<sup>(4)</sup> suggests that if algae are present, they assimilate the ammonia before it can be oxidized.

It has been shown that the conversion of organic nitrogen to various other nitrogen forms results in a net loss in total nitrogen.<sup>(11)</sup> This loss may be due to either algal uptake for metabolic purposes or to bacterial action. It is likely that each mechanism contributes to the overall total nitrogen reduction. Regardless of the specific removal mechanism involved, ammonia removal in a facultative wastewater pond may exceed 95 percent, with the major removal occurring in the primary cell of a multi-cell system.<sup>(12)</sup>

Since gaseous stripping appears to be the most effective nitrogen removal mechanism, optimum conditions, especially pH and temperature, are of interest.

Experiments on ammonia stripping conducted by Stratton<sup>(9)(10)</sup> showed that the ammonia loss-rate constant was dependent upon the pH value and temperature (°C) as shown in the following relationships:

$$\text{Ammonia loss rate constant} \propto e^{1.57(\text{pH}-8.5)}$$

$$\text{Ammonia loss rate constant} \propto e^{0.13(T-20)}$$

Equation (6) shows the expected removal rate of ammonia gas and the ionic form. However, a modification of equation (6) becomes necessary when the total nitrogen, sometimes referred to as Total Kjeldahl Nitrogen (TKN), removal is to be calculated. TKN is composed of ammonia-N and organic nitrogen. Organic nitrogen is a potential source of ammonia-N resulting from reactions during the metabolism of organic matter in wastewater. When modified through observations of three full-scale facultative wastewater stabilization ponds, the calculation for TKN removal can be achieved with the following relationships:

$$\frac{C_e}{C_o} = \frac{1}{1 + A/Q K \cdot f(\text{pH})} \quad (7)$$

where,

$K$  = removal rate coefficient ( $1/t$ ), and

$f(\text{pH})$  = function of pH.

The  $K$  values are considered to be a function of temperature and mixing conditions. The function of pH, which is considered to be dependent on temperature, affects the removal rate coefficient ( $K$ ) and the ammonia dissociation constant ( $K_b$ ) values, as well as the biology of the pond. To incorporate the effect of the pH function on ammonia-N stripping (equation 6), the pH function was found to be an exponential relationship,

$$f(\text{pH}) = \frac{1}{1 + 10^{\text{pKw} - \text{pKb} - \text{pH}}} \quad (8)$$

The selection of an exponential function to describe the pH function was based on statistical analyses indicating that an exponential relationship best described the data. Also, most reaction rate and temperature relationships are described by exponential functions such as the Van't Hoff-Arrhenius equation, therefore, it is logical to assume that such a relationship would apply in the application of the theoretical equation to a practical problem.



Using data collected on a frequent schedule from every cell of several facultative wastewater treatment facilities, Middlebrooks developed a design model which fairly accurately predicts the total nitrogen removal in facultative lagoon systems. Validation of the model can be found in EPA(6), Reed(7), and Reed, et al. (8)

The Middlebrooks model shown in equations (9) and (10) is a first order equation, dependent upon pH, temperature, and detention time. It assumes a complete mix of pond contents. The development of the model suggests that volatilization of ammonia is the major pathway for nitrogen removal from wastewater stabilization ponds. This model should be useful for new or existing wastewater ponds when nitrogen removal and/or ammonia conversion is required. The design of new systems would typically base the desired detention time on BOD removal requirements. The total nitrogen removal that can be expected during the designed detention time can be calculated with this model. It is prudent to assume that the remaining nitrogen in the effluent will be ammonia. Middlebrooks suggests that additional detention time should be considered compared to the costs of other alternatives for nitrogen removal. The Middlebrooks approach, which heavily influenced by detention time, is demonstrated in equations (9) and (10) as follows:

$$N_e = \frac{N_o}{1 + t(0.0000576T - 0.00028)e^{(1.080 - 0.042T)(pH - 6.6)}} \quad (9)$$

where,

- $N_e$  = effluent total nitrogen (mg/l),
- $N_o$  = influent total nitrogen (mg/l),
- $t$  = detention time (days),
- $T$  = temperature of pond water (°C), and
- pH = pH of near surface bulk liquid.

The temperature,  $T$ , of the lagoon wastewater is calculated using Mancini and Barnhart's (1926) equation as follows:

$$T = \frac{0.5A T_a + T_i}{0.5A + Q} \quad (10)$$

where,

- $A$  = surface area of pond (m<sup>2</sup>),
- $T_a$  = ambient air temperature (°C),
- $T_i$  = influent temperature (°C), and
- $Q$  = influent flow rate (m<sup>3</sup>/day).

From the above discussion, one can conclude that the rate of nitrogen removal is influenced by: detention time; the depth and duration of the aerobic zone; conditions that facilitate gaseous exhaustion such as, surface area, reduced surface tension, surface rippling, and mixing; surface-to-volume ratio; pH level in the surface stratum; algal growth control; and ambient temperatures.

The Sunflo<sup>2</sup> technology, through the Progressive Reaeration and Recirculation Process (**PRR™ Process**), contributes to nitrogen removal by the following functions:

1. Reaeration through surface renewal (see Oxygen Transfer Via Enhanced Reaeration and Mixing), thereby establishing and maintaining an aerobic zone.
2. Establishing an aerobic zone that depending on BOD<sub>5</sub> loading conditions, may reach the full depth of the pond, thereby diminishing or replacing the facultative zone at the sludge surface, enhancing conditions for nitrification.
3. Mixing upper volume of pond to selected depths, thereby increasing the mass transfer coefficient.
4. Surface rippling, thereby increasing the surface area resulting in increased gas transfer capability for a given sized pond.
5. Surface renewal; a function that improves algal growth in the upper 8" stratum of the pond resulting in increased pH near the surface thereby increasing the conditions for gaseous stripping of  $NH_3$  to the atmosphere as well as increasing the photosynthetic supply of oxygen.
6. Controlled mixed volume from bottom to top, thereby destratifying temperature gradients and pH concentrations, resulting in homogenized contents and less fluctuation from the effects of temperature and pH within the mixed zone.

Experience has shown that the ammonia-N removal rate, by the Sunflo<sup>2</sup> technology, can be reasonably calculated using equation (6) during no-ice conditions. To the extent the surface area is covered by ice, and the temperature of the pond is reduced, stripping to the atmosphere will be limited. The area,  $A$ , in the formula must reflect the surface area of open water. Because the pH in the upper stratum is also a significant controlling factor, the average daily pH in this stratum is required in this formula. When considering TKN removal, equation (8) may be used with these same considerations of open surface area and surface stratum pH values to predict the total nitrogen removal from a facultative pond.

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