the economic feasibility of any such proposed system and should be kept in mind in the evaluation of all such processes. However, the mechanics of this one-step process are quite simple. These losses are not likely to be as great as they will be for multistep processes. It may be that clever design and arrangements of components will permit us to reduce reradiation losses, making this portion of our analysis unduly pessimistic. The idea is worthy of further consideration and may even acquire economic and social value in the future.

Summary

The limitations of thermochemical energy storage devices are the limitations of Carnot devices. Entropy production entailed in product separation further limits the efficiency of thermochemical processes. Thus, high upper temperatures and few reaction steps are desirable. In this article, the one-step effusional separation of water into hydrogen and oxygen is considered. Membrane materials, design, and fabrication techniques are suggested. A parametric analysis of the process suggests that the idea is a tantalizing possibility.

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21. If the open area of the membrane is too small, for example, to permit the flow of enough effusate to yield the amount of hydrogen required by the energy balance, the system will not be stable. The membrane will overheat and be destroyed.
22. An estimate of the irreversibility associated with the separation process may be made by calculating the entropy change (ΔS) associated with it. There are many ways the reactor might be run, and many different avenues of heat transfer within the reactor and separator which might be selected. Since all of the governing equations are linked, each of these requires a separate, iterated, time-consuming solution. However, we can use the example of Fig. 5b to illustrate how this problem may be handled. The overall system efficiency, with irreversibilities neglected, is 0.58. If the effusion barrier permits the flow of gas, carrying its energy, but not the flow of heat, the effused gas, finding itself at a lower pressure, will dissociate some more, reducing its temperature from 2300° to about 2265°K, and will acquire a composition corresponding to chemical equilibrium at its new temperature and pressure. The entropy produced by the separation process can be calculated from the difference between the entropies of the two product streams, the entropy of the reactant stream, and the concomitant heat flow into the reactor. For the example given, this comes to 68.62 calories per degree Kelvin per mole of hydrogen produced. The lost work, $T_L\Delta S$, is 68.62 × 300 calories. Thus, at this operating condition, the production of 1 mole of hydrogen, equivalent to 56.69 kilocalories of work, entails the irrevocable loss of 20.59 kilocalories, even for the otherwise reversible process. The maximum overall efficiency is thus reduced from 0.58 to 0.37. If the device were made to operate with a smaller pressure drop (it was 90 percent in this example), the loss would be smaller.
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Secondary Sewage Treatment Versus Ocean Outfalls: An Assessment

Charles B. Officer and John H. Ryther

About one-third of the population of the United States lives or works close to estuarine and coastal waters (*1*). Understandably, there is concern about disposal of municipal and industrial wastes in the waterways adjacent to the larger metropolitan coastal communities—concern about how much and what types of waste can be assimilated by these waters and about what corrective actions should be taken to improve their quality.

These are not easy problems to solve. They require appropriate contributions from the marine sciences of physical oceanography, biology, chemistry, and geology as well as the related branches of civil engineering. The Environmental Protection Agency, under the terms of its enabling legislation, has taken the position that the solution shall be in the construction of secondary sewage treatment facilities (*2*). However, environmental scientists and engineers alike have argued that secondary treatment of wastewater is unneeded and ineffective if discharges are made into open coastal waters, where there is adequate mixing, dilution, and advection to prevent oxygen depletion, eutrophication, and other adverse effects resulting from high concentrations of organic wastes (*3–6*).

Generally, there is agreement among environmentalists and the practitioners of waste disposal that untreated sewage should not be discharged into harbors, estuaries, or other confined coastal marine waters with restricted circulation and water exchange. A basic question is whether secondary treatment is the appropriate solution in such situations, or whether the high capital and operating costs of advanced treatment would be better spent on a comparable investment in ocean outfalls.

The construction costs related to the solution of these pollution problems are immense. For the coastal metropolitan areas of the United States they are measured in billions of dollars. For example, for one metropolitan area, Boston, the costs for secondary sewage treatment plants are estimated to be several hundred million dollars, and those for ocean outfalls are comparable (*7*).

In brief, secondary sewage treatment biologically, through the action of microorganisms, decomposes and oxidizes the dissolved and particulate organic matter in wastewater. This action, carried out under vigorous aeration followed by sedimentation of inorganic solids and the microorganisms themselves, results in the removal of 80 to 90 percent of the biochemical oxygen demand (BOD) and suspended solids, leaving a relatively clear, well-oxygenated effluent containing the inorganic compounds of nitrogen, phosphorus, and other constituents. Before discharge, the effluent is normally disinfected by chlorination.

Secondary treatment has no direct effect on eutrophication of the receiving waters, which may result from uptake of the mineralized nitrogen and phosphorus compounds by phytoplankton and the subsequent oxygen demand associated with respiration or decomposition of that secondary crop of organic matter. Further, it is applicable only to point sources of wastewater and has no potential impact on distributed sources. The questions are, then, how much effect does BOD removal by secondary treatment have on the possible pollution, measured in terms of dissolved oxygen deficits, of a particular area, and relatively how important is the eutrophication potential of the secondary effluent?

In this article we make a scientific assessment of this problem, using order of magnitude estimates. Although we appreciate that numerical modeling is often a necessity for final engineering planning, we feel that such approximate considerations are appropriate for the general problem addressed here and that they facilitate the separation and examination of the important variables.

Marine Biological and Chemical Oxygen Demands

In most of the discussion that follows we will be concerned with the direct oxygen demand of the waste materials and the indirect oxygen demand associated with any eutrophication effects. The life history and ultimate fate of possible trace contaminants such as hydrocarbons, chlorinated hydrocarbons, and heavy metals or of pathogens such as viruses and bacteria will be considered only briefly. We do appreciate that these are also important problems for waste treatment and disposal.

The potential oxygen demands for the two processes—waste oxidation and eutrophication—are comparable. From Thomann (*8*), the total oxygen required for the ultimate biochemical oxidation of the organic wastes in an untreated municipal effluent—the waste biochemical oxygen demand (WBOD)—is about 400 milligrams per liter. Ryther and Dunstan (*9*) argue that inorganic nitrogen, rather than inorganic phosphorus, is the limiting nutrient in marine phytoplankton growth. From Redfield *et al.* (*10*) we have that the combining atomic ratio of oxygen to nitrogen in the reversible equation for marine phytoplankton photosynthesis, or respiration, is 276:15.5, or 20:1 by weight. Again from Thomann (*8*), the inorganic nitrogen ($NH_4^+ + NO_2^- + NO_3^-$) in a municipal effluent is characteristically in excess of 20 mg/liter. Thus, the ultimate phytoplankton photosynthetic oxygen production or eutrophication oxygen demand [phytoplankton biochemical oxygen demand (PBOD)] of untreated wastewater will be in excess of 400 mg/liter.

In addition to the inorganic nitrogen referred to above, untreated effluents have an organic nitrogen content of 20 mg/liter (*8*). In the secondary treatment process some nitrogen is usually lost, presumably to the atmosphere as NH_3, and a typical secondary effluent has an inorganic nitrogen content of about 20 mg/liter (*11*). The PBOD of a secondary effluent is thus at least as high as the WBOD of raw sewage and may, if nitrogen is not lost in the treatment, be double that amount.

Summary

Simplified models have been developed to obtain order of magnitude estimates of the oxygen demand of municipal and industrial wastes and of their potential eutrophication effects in the marine environment. The models have been applied to assess two major corrective actions that might be considered for such pollution problems—secondary sewage treatment and ocean outfalls. It is concluded that the arguments for secondary sewage treatment as the proper corrective action are not compelling and that the problem should be reexamined with appropriate scientific and engineering evaluations. One goal of such scientific evaluations should be a more thorough understanding of the life histories of possible trace contaminants and pathogens which may have long biological, chemical, or geological retention times in the marine environment.

Assimilation Characteristics of Rivers, Estuaries, and Coastal Waters

We are interested in the average conditions within a definable volume of water, V, of a river, estuary, or coastal region. The dilution and reaction rate phenomena are treated in terms of a steady state box model. The important physical oceanographic variable is the flushing or retention time, τ, defined as the time necessary to replace any particular conservative quantity in the volume at the rate at which that quantity is being injected into the volume (*12*). For rivers and lakes τ expresses the effect of the advective river flow, R, through the volume and is given simply by V/R. For estuaries it also includes the important effects of tidal mixing and density gradient- and wind-induced circulation flows. For coastal waters it includes all three of these effects plus that due to the coastal current structure.

The flushing time enters the calculations in two ways—in terms of the effective dilution and in connection with the reaction rates for oxidation, reaeration, or respiration. The volume-averaged concentration of a conservative quantity, c, is given in terms of τ as $C/(V/\tau)$, where C is the rate at which the quantity is being added. The flushing time here enters in terms of the quantity V/τ, which we refer to as the assimilation, and it is a direct measure of the dilution characteristics of the volume. The flushing time also enters in the first-order exponential reaction terms in the form $k\tau$, where k is the reaction constant. For small $k\tau$ values little reaction takes place

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Table 1. Flushing time (τ), volume (V), and dilution potential ($1/f$) for various rivers, estuaries, and coastal regions.

Region	τ, (days)	V, (10^6 m^3)	$1/f$	Reference
Raritan River	8	36	2	(*13*)
Houston Ship Channel	56	67	1	(*15*)
Delaware River	28	1,330	2	(*24*)
Boston Harbor	2	67	24	(*25*)
Mersey Estuary	4	147	7	(*26*)
Raritan Bay	14	860	12	(*13*)
Passamaquoddy Bay	16	4,700	41	(*27*)
New York Bight	8	23,000	63	(*22*)
Bay of Fundy	76	880,000	52	(*27*)

within the defined volume; for large $k\tau$ the reaction is essentially complete.

Ketchum (*13*) showed that the freshwater flow or river discharge, R, into a tidal river, estuary, or coastal region can be used as a natural tracer to determine the flushing time. The flushing time is given by fV/R, where f is the volume-averaged freshwater fraction, and the assimilation is given by R/f. The quantity $1/f$ is then a measure of the dilution potential over that due to river advection only.

Values that have been determined for τ, V, and $1/f$ for various selected rivers, estuaries, and coastal regions are given in Table 1. The substantial increase in the dilution potential from rivers to estuaries and from estuaries to coastal regions should be noted.

Waste Oxidation Model

The waste biochemical oxidation can be considered, in a first-order approximation, to decrease in proportion to the amount of pollutant present, so that the WBOD will be proportional to $\exp(-k_1t)$, where k_1 is the WBOD decay coefficient and t is the time from discharge. We take as an estimate of the total demand within the volume the average value of the exponential over the flushing time τ, or $[1 - \exp(k_1\tau)]/k_1\tau$, multiplied by the initial demand averaged over the volume, or $W/(V/\tau)$, where W is the combined waste discharge oxygen demand. This method of estimation is subject to question but is in keeping with the flow-through box model and order of magnitude considerations that are of interest here. The dissolved oxygen utilization within V, or the waste dissolved oxygen deficit (WDOD) with respect to a saturated condition, will then be

$$\mathrm{WDOD} = \frac{W}{\frac{V}{\tau}}\left(1 - \frac{1 - e^{-k_1\tau}}{k_1\tau}\right) \quad (1)$$

where W is in units of grams per day, V in cubic meters, τ in days, k_1 in 1/days, and WDOD in milligrams of oxygen per liter. The oxygen will be replenished within V through reaeration; the rate of oxygen replenishment will, again in a first-order approximation, be proportional to the dissolved oxygen deficit. Then following the same procedures as before, we have for WDOD including reaeration,

$$\mathrm{WDOD} = \frac{W}{\frac{V}{\tau}}\left(\frac{1 - e^{-k_2\tau}}{k_2\tau} - \frac{1 - e^{-k_1\tau}}{k_1\tau}\right)\frac{k_1}{k_1 - k_2} \quad (2)$$

where k_2 is the reaeration coefficient in 1/days. For large $k_1\tau$ and $k_2\tau$, Eq. 2 approaches $(W/V)(1/k_2)$, independent of τ.

To apply this simplified model, we take as our reference condition a saturated dissolved oxygen content, at a temperate latitude in summer, of 9 to 11 mg/liter. Thus, for values of WDOD > 10 mg/liter calculated from Eq. 2, we would anticipate that all or a substantial portion of V was anoxic. For values < 1 mg/liter, we would anticipate that there was no appreciable oxygen demand related to waste oxidation. And for values between 1 and 10 mg/liter, we would anticipate that there was a measurable oxygen demand, which for engineering planning should be further examined through numerical modeling techniques. Figure 1 is a graph of Eq. 2 with W/V plotted against τ for WDOD = 1 and 10 mg/liter. In this graph we used the characteristic values $k_1 = 0.4$ day^{-1} and $k_2 = 0.2$ day^{-1} obtained by numerical model simulation of field data (*14*).

It is of interest to compare these simplified results with observations. O'Connor *et al.* (*15*) give waste-loading values of 207×10^6 grams of oxygen per day for the Houston Ship Channel and 624×10^6 g/day for the Delaware River. As shown in Fig. 1, the Houston Ship Channel plots above the 10-mg/liter curve and the Delaware River between the 1- and 10-mg/liter curves. The Houston Ship Channel is anoxic and the Delaware River has dissolved oxygen deficit values ranging from 2 to 8 mg/liter along its length from Trenton to Delaware Bay (*15*). For Boston inner harbor the waste loading is only from storm-sewer overflows along the Charles River (*15*). We have taken an excess loading of 100×10^6 g/day for this source. The determined value, as shown in Fig. 1, plots below the 1-mg/liter oxygen curve. The observed dissolved oxygen in Boston Harbor approaches saturation.

Figure 1 provides us with a convenient index for estimating the effects of alterations in the waste loading, outfall location, or other environmental factors within the system. We see that, in general, small changes will have little effect. If conditions are becoming anoxic, major changes will be necessary to attempt to return the receiving waters to a measurably uncontaminated condition.

One such major change would be secondary sewage treatment. For example, suppose we initially have an anoxic condition in an urban estuary with a substantial waste loading of $W = 400 \times 10^6$ g/day, a volume $V = 40 \times 10^6$ m^3, and a flushing time $\tau = 10$ days. This original condition would then be as shown in Fig. 1. If secondary sewage treatment facilities were installed at all point sources, it might optimistically be hoped to reduce the loading by 90 percent (*4*, p. 9; *16*). In the resulting condition, as shown in Fig. 1, there would still be appreciable waste biochemical oxygen utilization.

Another major change would be the use of ocean outfalls. Here we take a coastal volume $V = 4000 \times 10^6$ m^3 and a flushing time $\tau = 5$ days. In the resulting condition, as shown in Fig. 1, the wastes now have a negligible effect on the natural dissolved oxygen content of the receiving environment. In essence, the ocean outfall, in this example, permits an improvement of two orders of magnitude whereas the secondary treatment, at best, permits an improvement of only one order of magnitude.

Eutrophication Oxidation Potential

We are interested here in developing a nutrient-limited model for the eutrophication potential. As discussed above, we can consider this limitation in terms of the inorganic nitrogen waste loading (N, in grams of nitrogen per day) or, more conveniently for our purposes, in terms of the potential utilization of the oxygen in these waste nutrients by phytoplankton in their respiration or decay (P, in grams of oxygen per day). For this condition P is approximately $20N$. This approach to eutrophication is similar to that used by Garside *et al.* (*17*) and others.

The eutrophication phenomenon is not one of a miscible material, involving simple flow-through considerations and decay, as is the case for waste oxidation of a clear effluent. Here we have biological retention effects as the phytoplankton material sinks to deep water and to the bottom and, subsequently, decays. The addition of a sink to the problem greatly increases the potential pollution hazard.

Considering the time constants of the system and system components, we treat the problem as one of a quasi steady state. We do appreciate that for longer time periods, measured in terms of seasons of the year, phytoplankton blooms are not steady state phenomena.

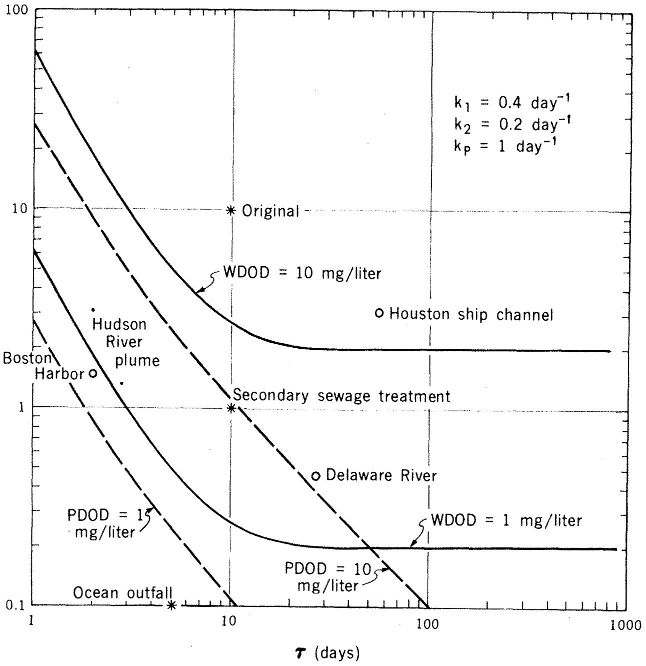

Fig. 1. Waste oxidation potential (WDOD) and eutrophication oxidation potential (PDOD) plotted in terms of waste loading (W/V and P/V) against flushing time (τ). The results for waste oxidation are compared with observations (○) for the Houston Ship Channel, the Delaware River, and Boston Harbor. Eutrophication oxidation results are compared with observation (●) for the Hudson River plume off New Jersey. (*) Possible effects of secondary sewage treatment (WDOD only) and ocean outfall (WDOD and PDOD) on an original pollution condition (see text).

For nutrients, particularly in our case inorganic nitrogen, to be the limiting constituents in phytoplankton blooms, a number of other conditions have to be met. For marine photosynthesis to occur at an optimum rate, it is necessary to have sufficient daytime solar insolation and sufficiently warm receiving waters; both of these conditions are generally met in temperate latitudes in the summer (*18*). In addition, the natural turbidity of the water must be low (*18*). This is a more serious limitation for urban estuaries, where the suspended sediment and detrital loads are often high, and precludes nutrients being the limiting factor in photosynthesis in these situations (*17*). When light, temperature, and turbidity conditions are favorable, phytoplankton growth rates can be rapid; optimum values are characteristically around a doubling per day, or a corresponding value for the phytoplankton production coefficient of $k_p = 1$ day^{-1} (*18*).

For eutrophication, or excess oxygen utilization in the phytoplankton respiration or decay, to then occur, it is necessary that the phytoplankton be physically separated from the waters in which they are produced. For example, if the phytoplankton characteristic of a particular bloom are small and essentially miscible with the water, they will utilize for respiration the photosynthetically produced oxygen and that reaerated in the surface waters as conditions become undersaturated. On the other hand, if there is nearly complete zooplankton grazing of the bloom, which is usually not the case, the ultimate oxygen utilization will be transferred to higher members in the aquatic food chain and their wastes. We are interested in the case where the plants or animals of the plankton or their solid wastes sink out of the photosynthetic upper waters and accumulate in the deeper subeuphotic waters and on the bottom. This corresponds to the situation that occurred in two examples of marine eutrophication that have been well studied. In one case the phytoplankton accumulated on the bottom (*19*), and in the other the phytodetritus from zooplankton grazing accumulated on the bottom (*20*).

The decay rates for phytoplankton respiration are generally low in comparison with the optimum production rates. A characteristic value for the respiration coefficient is $k_d = 0.05$ day^{-1} (*21*). Thus, if all of the available inorganic nitrogen, N, is continuously converted to phytoplankton and all of this material sinks out of the euphotic zone and is retained within the reference volume, the steady state accumulation will be N/k_d in terms of nitrogen content. The accumulation can be substantial; for $k_d = 0.05$ the accumulated material will be 20 times the daily production. This material decays at the rate k_d so that the oxygen utilization will be at the rate P.

To take the most adverse condition, we assume that there is no oxygen replenishment to the bottom waters from the supersaturated, or reaerated, water near the surface. This implies that there is a substantial thermocline or halocline with a consequent lowering of the vertical mixing coefficient in this zone, such that the hydrodynamic exchange is small in comparison with the normal flushing of the bottom waters. Then, including the rate of phytoplankton production in terms of the flushing time of the defined volume, the phytoplankton dissolved oxygen deficit (PDOD) for the bottom waters will be

$$\text{PDOD} = \frac{\frac{P}{V}}{\tau}\left(1 - \frac{1 - e^{-k_p\tau}}{k_p\tau}\right)\frac{\beta}{\alpha} \qquad (3)$$

where β is the fraction of the phytoplankton bloom that is retained and decays within the system out of the euphotic zone and α is the ratio of the volume of the bottom waters to the total volume, V. For large values of $k_p\tau$, Eq. 3 approaches $(P/V)\tau(\beta/\alpha)$ and is proportional to τ.

Equation 3 is graphed in Fig. 1 for PDOD = 1 and 10 mg/liter. Values of $\alpha = \beta = 1/2$ have been assumed for these calculations. The choice of the value of β is certainly subject to question. Not all of the plankton or plankton wastes will sink out of the euphotic zone and be retained within the reference system for decay. Considering that for a characteristic sewage effluent W and P have approximately the same numerical value, we see that the BOD potential from eutrophication is greater than that from direct waste oxidation, particularly at large values of τ. This conclusion, however, should be tempered: eutrophication is only a potential, and for many marine areas the determining criteria of solar insolation, water temperature, turbidity, and reduced vertical mixing will not be met.

As with the waste oxidation model, it

is of interest to compare these simplified calculations with observations. There is one example of cultural eutrophication in the marine environment for which there are sufficient data to make such a comparison. This is the anoxic condition that occurred off the coast of New Jersey during the summer of 1976 (*19*). The mixed Hudson River water exits from the lower bay into the apex of the New York Bight and moves as a definable plume southward along the shore of New Jersey for some considerable distance. Ketchum *et al.* (*22*) define a volume $V = 750 \times 10^6$ m^3 with a flushing time $\tau = 2.0$ days, or alternatively a volume $V = 1760 \times 10^6$ m^3 with a flushing time $\tau = 2.8$ days, for the mixed Hudson River plume along the New Jersey shore. From Garside *et al.* (*17*), we have that 160×10^6 g of inorganic nitrogen is released each day into the New York waterways from the sewage effluents, and during the summer 117×10^6 g/day exits through lower bay.

The points for these two defined volumes are shown in Fig. 1. We see that the mixed Hudson River water falls in the critical region between the 1- and 10-mg/liter eutrophication curves. For this case the flushing time for the bottom waters beneath the Hudson River plume will not, in general, be the same as that for the plume itself but will depend more on the prevailing coastal current conditions. In the summer of 1976 the wind and current conditions were abnormal and may well have led to an extended residence time over the 2 to 3 days assumed in these calculations, as has been speculated (*19*). It is pertinent to note that the anoxic conditions which occurred in 1976 were anticipated by Segar and Berberian (*23*) from measurements taken in 1974.

Let us now consider the possible effects of secondary sewage treatment or ocean outfalls on the eutrophication potential. Secondary sewage treatment will have no direct beneficial effect on the eutrophication potential; if anything, the eutrophication potential may be enhanced and accelerated because inorganic nutrients will be more rapidly and more completely available to the phytoplankton than they are when provided by the natural decomposition of wastewater in the sea. As shown in Fig. 1, the original condition will be unchanged by secondary sewage treatment. The installation of ocean outfalls, however, can virtually eliminate the eutrophication potential; in addition, the sewage nutrients can have the beneficial effect of serving as fertilizers for the aquatic food chain.

Trace Contaminants and Pathogens

The life history and ultimate fate of trace contaminants and pathogens introduced into the marine environment from municipal wastewaters comprise an important, albeit complex, subject. Initial source control, particularly for contaminants of industrial origin, may well be the most effective corrective action when it is possible (*3–5*). In this section we consider the effects of secondary sewage treatment and ocean outfalls when such control is not possible.

Part of the concern about such trace toxic materials is due to their stability and their biological, chemical, or geological transformations which lead to longer retention times within the marine environment. If a toxic material is miscible with the receiving waters but has a decay time long compared with the time constants of the marine system, it may be considered to be conservative and its average concentration will be given by the simple relation $c = C/(V/\tau)$. According to Bascom (*4*, p. 9), secondary treatment may reduce C by 45 percent. Taking the example we considered in the section on waste oxidation for comparison, the quantity V/τ will be increased by a factor of 200 for an ocean outfall. The reduction in c for an ocean outfall is clearly superior to that for secondary treatment. If the toxic material is not miscible but has a longer retention time, T, in the system, T will replace τ in the defining equation. Although the values of T for an urban estuary and an ocean outfall may be considerably different, the advantage in the reduction of c would still appear to lie with the ocean outfall, considering the hundredfold increase in the dilution volume.

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