# PREDICTING EVAPORATION RATES FROM SPENT NUCLEAR FUEL STORAGE POOLS

**Bruce R. Hugo, William C. Kinsel**

*Washington State University, USA*

*brhugo@frontier.com , kinsel@tricity.wsu.edu*

### Abstract

Accurate prediction of evaporative losses from spent nuclear storage pools (SFPs) is important for activities ranging from sizing of water makeup systems during plant design to predicting the time available to supply emergency makeup water following severe accidents. Existing correlations for predicting evaporation from water surfaces are only optimized for conditions typical of swimming pools. This new approach that treats evaporation as a diffusion process has yielded an evaporation rate model that provided a better fit of published data and measurements from two SFPs. Insights from treating evaporation as a diffusion process include correcting for the effects of air flow and solutes on evaporation rate.

### Keywords

*spent fuel storage, evaporation, tritium*

## 1.0 Introduction

### 1.1 Applications of evaporation modeling

An accurate model of evaporation is desirable for a number of applications related to spent nuclear storage pool (SFP) design, operation, maintenance, and accident response. During plant design, sizing of the demineralized water source must consider the makeup requirements for the SFP based on the expected evaporation rate. In the absence of significant steam leaks, SFP evaporation will be the largest source of water loss from the nuclear power plant.

Once the plant has been operated, radioactive tritium builds up in all of the primary plant systems. Tritium is as one of the products of neutron capture by $^{10}B$. In pressurized water reactors (PWRs) the boron is dissolved directly in the coolant for most of the operating cycle; in boiling water reactors (BWRs) the boron is present in the control rods but either boron or tritium can leak from the control rods into the coolant. Tritium is also produced in the fuel as a product of ternary fission. Coolant is transferred between the reactor coolant system and other plant systems by a number of processes; during refueling outages the reactor coolant system and SFP are directly connected to support refueling. Evaporation will carry a portion of this tritium inventory to the SFP building's ventilation exhaust contributing to the site boundary dose. Predicting the impact of this contributor to offsite dose requires an accurate estimate of the evaporation rate from the SFP.

Maintenance on the SFP cooling system may require that the system be shut down to allow work on pumps, valves, and other components. The SFP water heats up as decay heat produced in the stored irradiated fuel exceeds the ambient losses. Usually an administrative limit is imposed on SFP temperature; an accurate estimate of the time available for the maintenance is needed to ensure the cooling equipment can be returned to service before this temperature limit is reached. Although some heat is lost from the pool due to conduction, convection, and radiation heat transfer, the dominant mode at higher water temperatures is evaporation. Overes-

timating the heat losses from evaporation could result in exceeding a temperature limit during maintenance; underestimation could result in deferring maintenance unnecessarily.

Following a severe accident such as a prolonged loss of electrical power as occurred during the 2011 Fukushima accident, neither cooling nor normal makeup to the SFP will be available. When emergency makeup capability is available, there will be competing needs for reactor pressure vessel (RPV) injection for core cooling, containment injection for containment cooling, and SFP makeup to maintain the spent fuel covered with water. An accurate method of determining cumulative evaporation losses from the SFP is essential for deciding when to shift emergency makeup from the RPV or containment to the SFP. Premature diversion of water to the SFP could result in unnecessary additional damage to the core or containment; late makeup could result in damage to spent fuel and significant additional releases of radioactivity.

## 1.2 Limitations of existing evaporation correlations

Measurements of evaporation from the SFP at the Columbia Generating Station nuclear power plant (CGS) indicated that the evaporation rate was larger than expected based on the Carrier correlation [1]. Three diverse methods were used to estimate the evaporation rate: tracking water makeup requirements, observing the reduction in SFP heatup rate when cooling was suspended for maintenance, and measurement of the amount of radioactive tritium released from the SFP building's ventilation system.

With the exception of a correlation proposed by Shah, existing proposed correlations were based on empirical considerations alone [1,2,4]. Shah's 1992 correlation, derived from the analogy between mass and heat transfer, had been shown to provide best overall fit for previously published data [2].

Although the 1992 Shah correlation applied to quiet air, he extended the model to include the effects of forced air flow over the surface [3]. Preliminary measurements of evaporation rate from the CGS SFP exceeded the predictions of both the Carrier and Shah correlations, especially at elevated temperatures (above 98°F).

## 2.0 Developing a new model of evaporation

The investigation into the divergence between the observed CGS SFP evaporation rates and those predicted by existing correlations led to a new model of evaporation that treats this form of mass transfer as a diffusion process. The goal was to predict evaporation rates using a correlation consistent with the mathematics of this physical process while using a minimum number of arbitrary parameters or different formulae for different ranges of water temperatures.

## 2.1 Nomenclature

$A$       arbitrary correlation constant;

$C_i$      concentration of component $i$, m$^{-3}$ or kg/m$^3$;

$CF$     SFP ventilation correction factor, dimensionless;

$D$       diffusion coefficient, m$^2$/sec;

$J_z$       mass flux, kg/m$^2$sec;

$k$        Boltzmann constant, J/K-mol;

$MW$   molecular weight, kg/mol;

$n$        arbitrary correlation constant, dimensionless;

$N$       number density, m$^{-3}$;

$\dot{N}''$      mass flux, molecules/m$^2$sec;

$Nu_F$    Nusselt number for forced convection, dimensionless;

$Nu_N$    Nusselt number for free convection, dimensionless;

$P$        total or atmospheric pressure, Pa;

$P_a$      partial pressure of water vapor in the ambient air, Pa;

$P_w$     partial pressure of water vapor at the water surface, Pa;

$P_{sat,w}$   saturation pressure of water at the pool water temperature, Pa;

$P_{sat,a}$   saturation pressure of water at the ambient air temperature, Pa;

$R$        ideal gas constant, J/K-mol;

$T$        temperature, K;

$v$        air velocity, m/sec;

$W$      mass flux, kg/m$^2$sec;

$X$       mole fraction, dimensionless

$z$        coordinate, m;

$\delta$       migration or boundary layer thickness, m;

$\phi$       relative humidity, dimensionless;

$\mu$       reduced mass, kg/mol;

$\sigma$       molecular collision cross section, m$^2$.

## 2.2 Diffusion model for evaporation with still air

When a concentration difference exists at two points in a mixture, there will be a migration of molecules from the high concentration region to the low concentration region in accordance with Fick's law:

$$J_z = -D\frac{dN}{dz} \qquad \textbf{Eq 1}$$

When *both* components in a mixture of gasses are migrating, the solution to Eq 1 for a partial pressure difference existing across a distance $\delta$ is:

$$W = \frac{\text{MW}}{P}\frac{D}{\delta}\left(P_w - P_a\right) \qquad \textbf{Eq 2}$$

This is the form of many empirical correlations (with most of the terms being absorbed into a single constant) for evaporation, including the Carrier correlation and the Shah correlation for forced convection. $P_w$ is the partial pressure of water vapor at the water surface, which is assumed to be saturation pressure for the water temperature. $P_a$ is usually the partial pressure of water vapor in the ambient air; the Carrier correlation instead uses the saturation pressure at the air dewpoint but the difference is small for typical swimming pool and SFP ambient conditions. Since this form does not provide a good fit of evaporation data over a large range of water temperatures, many correlations provide a better fit by using a correlation of the form:

$$W = A\left(P_w - P_a\right)^n \qquad \textbf{Eq 3}$$

The values of *n* used range from 1.0 to 1.3, but no physical reason was proposed for the evaporation rate to be a function of partial pressure difference raised to a power greater than unity. Values of *A* used ranged from $3\times10^{-10}$ kg/m$^2$·hr·Pa to $9\times10^{-12}$ kg/m$^2$·hr·Pa$^{1.3}$ [2,4].

The difficulty in fitting this solution of the diffusion equation to evaporation data is due to evaporation not involving the bidirectional migration of two components in a gas mixture. Only the water vapor generated at the water surface is diffusing, not the air above the water surface. In particular, air is not diffusing into the water surface. The solution to Eq 1 when only *one* component in a gas mixture is migrating is:

$$W = \text{MW}\,\frac{D}{T}\frac{1}{\delta}\frac{P}{R}\ln\frac{P - \phi P_{sat,a}}{P - P_{sat,w}} \qquad \textbf{Eq 4}$$

$P_{sat,a}$ is the saturation pressure at the air temperature and $\phi$ is relative humidity so that $\phi P_{sat,a}$ is just the value $P_a$ used earlier. $P_{sat,w}$ is the saturation pressure at the water temperature and is equal to $P_w$.

The logarithmic term $\ln\dfrac{P - \phi P_{sat,a}}{P - P_{sat,w}}$ is approximately equal to $\dfrac{P_w - P_a}{P}$ for small partial pressures of water vapor corresponding to low pool temperatures and typical ambient air conditions (i.e., when $P_a \ll P$). This explains the success of correlations of the form $W = A\left(P_w - P_a\right)$ in fitting evaporation data at low temperatures. But as water temperature and $P_{sat,w}$ rise, the nonlinearity of the logarithmic term results in most correlations *under-predicting* the evaporation rate even when the curve fit parameter *n* is introduced. Since SFP evaporation data were only available over a limited range of water temperatures, evaporation data from the 1946 experimental work of Boelter et al. [4] which ranged from 24°C to 94°C were used to test the validity of Eq 4.
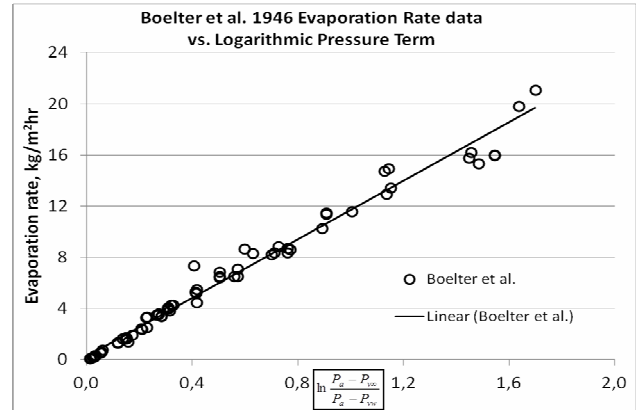


Figure 1. Boelter et al. 1946 data plotted against logarithmic temperature difference with least-squares fit.

The linearity of the data when plotted against the logarithmic term of Eq 4 is remarkable; the use of this term in place of $\left(P_w - P_a\right)$ alone provides a better fit of high temperature data than most existing correlations. The additional refinements to the diffusion model described below improve this fit further.

A second consequence of the diffusion model of evaporation is the temperature dependence of the diffusion coefficient $D$. From the kinetic theory of gasses, the relationship between $D$ and other gas mixture properties is:

$$D = \frac{2}{3}\frac{1}{\sigma P}\sqrt{\frac{k^3 T^3}{\mu\pi}} \qquad \textbf{Eq 5}$$

Due the simplifying assumptions made in developing Eq 5, it is not sufficiently accurate for use in predicting evaporation rates. For water vapor in air at 0°C the formula gives a diffusion coefficient of 0.14 cm$^2$/sec while the measured value is 0.22 cm$^2$/sec [6]. But it does show that $D$ should increase with temperature as $T^{1.5}$. Correlations for diffusion coefficients [6] show that the observed variation of $D$ with $T$ is actually in the range $T^{1.66}$ to $T^{2.0}$. Then $D(T)/T$ should be of the order $T^{0.66}$ to $T^{1.0}$.

Treating $P$ and $\delta$ as constant, a correlation for evaporation based on diffusion is now of the form:

$$W = A'T^n \ln\frac{P-\phi P_{sat,a}}{P-P_{sat,w}} = A\left(\frac{T}{T_0}\right)^n \ln\frac{P-\phi P_{sat,a}}{P-P_{sat,w}}$$
$$\textbf{Eq 6}$$

The form of Eq 6 on the right was used to make the temperature correction dimensionless. The coefficient $A$ and exponent $n$ are chosen to provide the best fit of experimental data, with the value of $n$ expected to be in the range $0.66 \le n \le 1.0$. The choice of $T_0$ is arbitrary but will be reflected in the value of $A$.

To determine the best values of $A$ and $n$ in Eq 6, the data of Boelter et al. [6] were used [4]. Omitting the temperature correction entirely (using a value of zero for $n$) and selecting $A$ to minimize the average absolute deviation between the prediction and the data resulted in a mean deviation of 11.2%. This mean deviation was reduced when larger values of $n$ were used (with a corresponding change in the parameter $A$); the best fit was obtained with $n = 1.3$ but $n = 1$ was selected since it is the highest value that can be supported by empirical correlations for $D(T)$. The resulting correlation is:

$$W = 8.64\frac{T}{273K}\ln\frac{P-\phi P_{sat,a}}{P-P_{sat,w}}\frac{kg}{m^2 hr} \quad \textbf{Eq 7}$$

$T$ must be in absolute temperature units; the units for $P$ are arbitrary due to cancellation.

## 2.3 Diffusion model for evaporation with forced air flow

Since the Boelter at al. data were for still air only, Eq 7 is accurate only in the absence of forced air flow over the water surface. This is not the situation for a SFP. At the CGS SFP, ventilation openings are spaced around three sides of the SFP; the bottom edges of these ducts are 15 cm above the water surface. Enough air is drawn off the surface of the pool to ensure that any radioactive particles emitted are drawn into the ventilation system and discharged at a monitored, elevated release point. Additionally, during severe accidents it is possible that the SFP water surface may be exposed directly to wind driven air flow. Hydrogen explosions such as occurred during the 2011 Fukushima accident are not the only mechanism for damaging the building enclosing the SFP – an earthquake, hurricane, or tornado beyond that for which a plant was designed could also result in a SFP being exposed directly to the atmosphere.

### 2.3.1 Evaporation from a SFP under normal conditions

Although determination of the air velocity distribution over the surface of the SFP is one objective of ongoing research by the authors, the average velocity does not exceed 0.5 m/sec based on the absence of ripples on the pool surface. The ventilation system should not cause disturbance of the water surface since extremely good water clarity is necessary to manipulate components stored nearly seven meters below the water surface. All SFP ventilation systems have similar design objectives; therefore, the evaporation predictions that follow should apply to most SFPs.

Evaporative and other losses from the CGS SFP result in a slow lowering of water level in two skimmer surge tanks (SSTs) that collect water flowing from a weir in the pool wall (see figure 2). Since the water level in the SFP is slightly above the height of this weir whenever the circulating pumps are running, inventory changes occur only in the SSTs. Each SST is instrumented with level detectors that provide local indication and remote alarms, control automatic makeup, and provide cavitation protection for the circulating pumps.
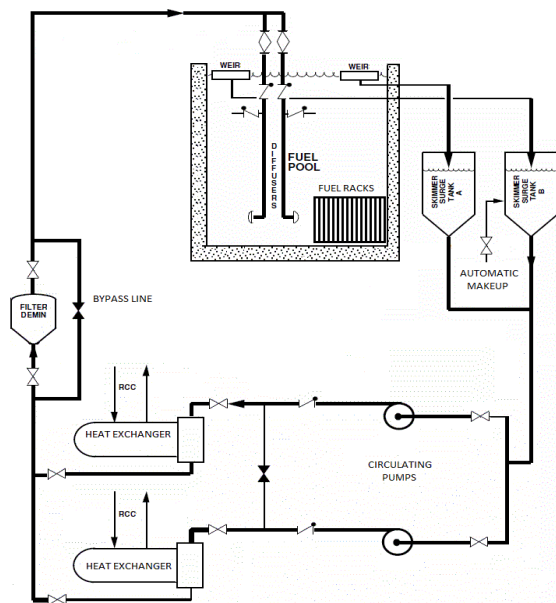
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Figure 2: Spent Fuel Pool Flow Diagram

Correction for automatic batch makeup can be made since the volume change is a function only of the SST dimensions and the level switch setpoints that control the opening and closing of the makeup valve. The effects of thermal expansion and contraction of the SFP water can also be determined since continuous SFP temperature monitoring is available using the process computer. Potential sources of error in this method include system leakage, which would result in an overestimate of evaporation, and makeup valve leakage which would result in an underestimate.

Approximately every two years when the SFP decay heat load is relatively low, the cooling system in shut down for preventive and corrective maintenance. SFP temperature rises since the decay heat input from the irradiated fuel exceeds the heat lost to the SFP building environment by conduction, radiation, and convection. The heatup rate is observed to decrease as water temperature and ambient heat losses rise. The effects of conduction through the concrete SFP walls, radiation loss to the space enclosing the SFP, and direct heating of the air above the water surface can be calculated. Any additional heat loss is due to increased evaporation. This method does not provide an absolute estimate of evaporation rate since the initial evaporation rate is unknown, but it does provide an estimate of the *change* in evaporation rate over the course of the cooling system outage.

The third method used was the use of tritium as a tracer. Since the tritium in the SFP water is combined with oxygen as $^3$H-OH and has nearly the same physical and chemical properties as water, it evaporates from the SFP and is exhausted from the SFP building at a monitored release point. The tritium activity released is measured monthly. Tritium concentration in the SFP is monitored and changes slowly; the amount of water evaporated from the SFP can be determined by dividing the total amount of tritium activity released by the tritium concentration in the SFP. There are normally no other comparable sources of water evaporation in the SFP building; this hypothesis was confirmed by comparing the average amount of tritium released during months when CGS was operating with the amount released during shutdown periods. To avoid attributing any minor contributors to tritium releases to SFP evaporation, only the change in estimated evaporation rate with SFP temperature (rather than the absolute amount estimated) was assumed to be valid.

The estimated evaporation rate from the CGS SFP was expressed as a multiple of the rate predicted by Eq 7, i.e., $W_{SFP} = CF \cdot W$ where CF is a correction factor that accounts for the increased evaporation rate due to air flow over the SFP surface.

The values of CF obtained by estimating evaporation using the inventory tracking method ranged from 1.04 to 1.18. These estimates are based on two tests of duration 2.2 and 0.8 days respectively. The amount of system leakage was found to be large compared with the expected evaporation rate except when the cooling system demineralizer is bypassed and used to purify water in the CGS suppression chamber. This system alignment is used only three days each month limiting the number of test runs that can be made.

A value of 1.45 was obtained for CF during a 52 hour cooling system shutdown. The initial SFP temperature was 36.7°C; the final temperature was 52.3°C. Heat losses to the 1.48 m thick concrete SFP wall were calculated using a one-dimensional transient heat conduction model and were found to account for 22% of the observed reduction in heatup rate over the course of the test. Radiation losses and convective heating of air at the surface of the SFP were each found to contribute about 7.5% of the total ambient heat losses. Average air flow was

estimated based on the known flow rates through the duct opening above the pool surface. Notably, free and forced convection heat transfer mechanisms were found to be important (Nusselt numbers were 487 and 374 respectively).

SFP building tritium release data from January 1998 through December 2011 were reviewed although electronically recorded values of SFP temperature were only available after June 2002. Tritium releases were the largest during those months when the reactor cavity was filled to support refueling operations which effectively triples the surface area available for evaporation. Rather than attempt to correct these data for the effect of increased surface area, only the data associated with normal plant operation at power were used in making the evaporation estimate. There was a substantial amount of variation in the evaporation rate estimates made using the tritium tracer method (see Figure 3), but the 98 data points indicated a correction factor of 1.22 to 1.31 depending on whether a curve fit based on least-squares or minimizing absolute deviations was used (the latter method assigns less weight to outlying data compared with least-squares).

Based on the three types of evaporation measurements made, the correction factor for SFP evaporation is about 1.25, i.e.,

$$W_{SFP} = 1.25 \left( 8.64 \frac{T}{273K} \ln \frac{P - \phi P_{sat,a}}{P - P_{sat,w}} \frac{kg}{m^2 hr} \right),$$

or

$$W_{SFP} = 10.8 \frac{T}{273K} \ln \frac{P - \phi P_{sat,a}}{P - P_{sat,w}} \frac{kg}{m^2 hr}$$
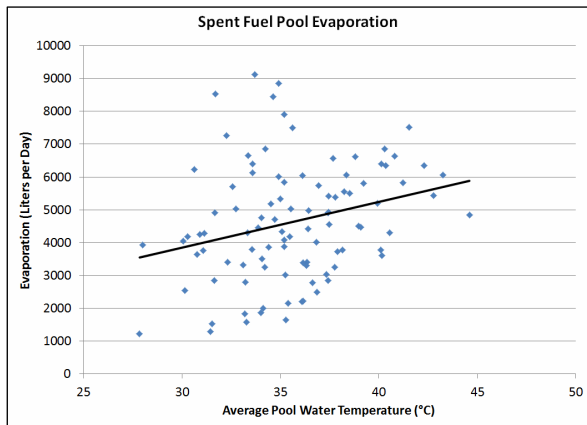
**Eq 8**



Figure 3: SFP Evaporation Rate vs. Temperature

## 2.3.2. Evaporation rate for other air velocities

The diffusion model of evaporation suggests (Eq 4) that forced air flow over the surface of the water promotes evaporation. The semi-stagnant air layer through which the diffusion is occurring, assumed to be a distance $\delta$ in the development of the model, should be affected by the air velocity over the water surface. This layer thickness was absorbed into the parameter $A$ earlier but will now be used to provide an insight into the expected form of a general correction factor CF($v$).

The Carrier correlation (1) accounts for air velocity with a multiplier of $A + Bv$; this essentially models $\delta \propto \dfrac{1}{A + Bv}$. But if $\delta$ is assumed to be comparable to the thickness of the turbulent boundary layer then $\delta$ should be proportional to either $v^{-0.2}$ (if $\delta$ is comparable to the velocity boundary layer) or $v^{-0.9}$ (if $\delta$ is comparable to the viscous sublayer) (5). A rough estimate of the diffusion distance using a diffusion coefficient of 0.22 cm$^2$/sec and the evaporation rate predicted by the Carrier correlation is $\delta$ = 0.38 cm. For the air velocity estimated at the CGS SFP surface the velocity layer thickness is about 22 cm based on a Reynolds number of 1.2 x $10^5$ and a distance of half the width of the SFP. In contrast, the viscous sublayer is about 3 cm thick under these conditions. Thus the diffusion distance should be of the order $v^{-0.9}$ and the correction factor should be of the order $v^{0.9}$.

Since the correction factor must have the value 1 when $v = 0$, the following empirical form was chosen:

$$CF(v) = \left( 1 + k \left[ v^{0.9} \right]^n \right)^{1/n} \quad \textbf{Eq 9}$$

The selection of this form was based on a method recommended for combining the effects of free and forced convection in heat transfer calculations [7]:

$$Nu^n = Nu_F^n \pm Nu_N^n \quad \textbf{Eq 10}$$

In Eq 10 recommended values of n range from 3 to 4 depending on the type of flow but this restriction was not imposed for fitting parameters to Eq 9.

Evaporation data as a function of air velocity were obtained from reference [8]; the best fit was obtained by using $k$ = 1.5 and $n$ = 1.5 when $v$ is in

m/sec. The general evaporation prediction using the diffusion model is therefore:

$$W = 8.64\left(1 + 1.5v^{1.35}\right)^{0.67} \frac{T}{273\text{K}} \ln \frac{P - \phi P_{sat,a}}{P - P_{sat,w}} \frac{\text{kg}}{\text{m}^2\text{hr}}$$

**Eq 11**

Eq 11 should produce accurate results at air velocities up to 12 km/hr for low pool temperatures (14°C-28°C) and for temperatures up to 94°C with still air. High temperature/high air velocity data were not available to test the diffusion model's predictions. Since the correlation was developed using established principles from diffusion, the kinetic theory of gasses, and boundary layer theory the evaporation rates predicted for other combinations of water temperature and air velocity should also be reasonably accurate.

Research is continuing to calculate the average air velocity over the CGS SFP and determine if this value along with Eq 11 can be used to obtain Eq 8.

## 2.4. The effect of solutes on evaporation

For water with a solute, such as the high concentrations of boric acid dissolved in a Pressurized Water Reactor SFP, the vapor pressure of the water is reduced as described by Raoult's law to $P_{sat}X_w$ where $X_w$ is the mole fraction of water in the solution.

This is normally a small correction since a boron concentration of 2000 ppm corresponds to $X_w = 0.9967$. If seawater has been used as an emergency source of SFP makeup water then the vapor pressure reduction would be more significant. Seawater has a water mole fraction of $X_w = 0.979$ due to the presence of nonvolatile sodium and chloride ions; subsequent evaporation of water during an accident would concentrate the seawater solution further.

## 3.0 Conclusion

The diffusion model of evaporation predicts evaporation rates from undisturbed pools with accuracy comparable to the best existing correlations. The functional form of the correlation (Eq 7) respects the underlying physical process of evaporation. Although the air flow velocity field over the SFP is not normally available, the common design requirements for SFP ventilation systems should result in similar average air velocities and thus permit use of the SFP-specific evaporation model that was developed using data from measurements of actual SFP evaporation rates. For unusual conditions such as wind driven air flow over a SFP, the diffusion model was extended using insights from boundary layer theory and experimental data from unoccupied swimming pools.

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