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COMMENT

Comment on "Numerical modeling of UV intensity: application to collimated-beam reactors and continuous-flow systems", by E. R. Blatchley III, *Wat. Res.* **31,** 2205–2218

This paper concerns computational methods and an apparatus design intended to improve the accuracy of measurements of photochemical reaction rate coefficients in UV disinfection of water and wastewater. This is an excellent paper that sets a high standard of care in measurement and theoretical development, and contains an excellent collection of informative references to related work.

Like other papers by Blatchley and various coauthors (e.g., Blatchley, *et al.*, 1994, 1995), this paper is an effort to bring greater scientific rigor to engineering application of UV irradiation. Several sources of error in past methods are addressed: variation in the output power of low-pressure mercury arc lamps, reflection in collimated beam devices, nonuniform flow in continuous-flow reactors, spatial variations in intensities in open-channel arrays, and imperfections in numeric models of the lamps. Not all of these sources of error are treated in equal depth in this paper, but all of them prevent accurate determination of the dose received by water samples that are then assayed for the chemical or biological effects of the irradiation.

As hydrodynamic behavior in flowing irradiation systems is poorly understood, and has been difficult to investigate, batch reactor setups with irradiation by collimated beam devices have been considered to provide the most accurate results. However, past designs of collimated beam devices (e.g., Qualls and Johnson, 1983; Sakamoto and Schwartzel, 1995) have performed collimation by tubes that unwantedly scatter some light into the sample. The new design in this paper performs collimation by using circular holes in several flat plates mounted in a flat-sided enclosure, with everything made of wood that reflects very little UV light as shown by measurements made not only on the wood but on several more reflective materials. Measurements on the new design are compared with the intensities provided by the line source integration (LSI) method, which for these long lamps is superior to the previously used point source summation (PSS) method. Other measurements were made of the outputs of four lamps with the new type of collimator so that each lamp output was sampled in a way that allowed reliable extrapolation of the sample to the whole lamp.

Since UV disinfection is increasingly important for water and wastewater treatment, the research in this paper is likely to have substantial practical importance. We invite the author's comments about the following items.

1. Figure 2 of the paper shows a collimator with three plates, which is apparently intended as a typical design, but Figure 8 shows only two plates in the collimator used for power measurement. Is there a typical number of plates that is recommended, or were different numbers used for collimators of different lengths, such as the ones used in making the measurements for Figure 7?
2. Has this apparatus been used to study any actual wastewater samples? Estimated doses of 80 mW-s/cm^2 or less have been sufficient to produce large inactivation factors for coliform bacteria and selected viruses in microfiltered (Jolis and Hirano, 1996) and multimedia filtered (Sakamoto and Schwartzel, 1995) reclaimed wastewater, which is much less than the standard of 140 mW-s/cm^2 in Title 22 of the California Code Regulation. Since Sakamoto and Schwartzel used a tube collimator of the type that the present design is intended to supercede, and Jolis and Hirano estimated their doses from a formula that invalidly assumes uniform plug flow, determining correct rate coefficients for disinfecting highly filtered tertiary effluent could have a rapid impact in reducing the operating costs of existing systems and the total costs of new ones by showing the true dose needed.
3. Since measurements made in open-channel systems underestimate the kinetic coefficients, it is clear that for a process that obeys first order kinetics, this underestimation will lead to design of systems that apply excessive irradiation. Is a comparable design bias present in systems for inactivating organisms obeying more complex kinetics?
4. Although this type of lamp is often reported to have an output power of 26.7 W, these four lamps under the conditions of the experiment had powers of 31.1–33.1 W. Underestimation of lamp output would also lead to designs that irradiate unnecessarily. On the other hand, since lamp output declines

with age (EPA, 1986), older lamps presumably come closer to the usually stated output. Would the author see any merit in a phased lamp replacement schedule that would guarantee a range of lamp ages in each unit so that the average output is always close to the expected level? This could be done easily in a system like those used in the author's 1993 and 1995 papers, where each lamp bank was composed of several modules of lamps.

5. Measurements agree with the calculations almost perfectly in the far field region (when the length of the collimator is greater than about 12 cm), but the calculations overestimate the intensity and shorter distances, in the near field. Inaccuracy of lamp output is proposed as the explanation for a large fraction of this discrepancy because of the sensitivity of low pressure, low intensity mercury vapour arc lamp output to surface temperature. Does the author have any suggestions for maintaining a more stable lamp temperature despite diurnal or seasonal changes in water temperature or flow rate?
6. Owing to the nonlinearity of plasma conduction, the most intense UV emission probably comes from near the lamp centerline, so that the success of the single line source model can be easily understood. However, the spatial extent of the emission region is likely to affect the intensity calculation for the near field. The passage from the point source summation method to the line source integration method implies that the light output of the lamp could be modeled more realistically by distributing several line sources through the cross-section of the lamp. For the collimator calculation one would then have an easy summation of the arctangent functions used in equations (5) and (8). It might even be possible to develop an area source integration method, which for collimator calculation involves an integration of the arctangent functions that might also have an analytic solution available in Gradshteyn and Ryzhik (1990). Numerical evaluation evidently is also possible. In short, one can build on the author's analytic solution to gain more accuracy in the near field.
7. This more realistic model of lamp emission would have an immediate application to the lamp output power measurements, since it does not require uniform intensity across the lamp cross-section, but the summation for integral could be weighted based on measurements made through a collimator with small apertures that viewed only small portions of the mercury vapor-field lamp lumen.
8. Could quartz optical fibers or any arrangement of lenses or mirrors be attached to the photometer to provide the photon collection through a wide solid angle mentioned on page 2215 of the paper as not having been available for the present study? The optical expertise demonstrated in this paper appears to be a good starting point for devising this innovation.
9. Fundamental hydrodynamic theory argues for the formation of boundary layers near wetted surfaces (Currie, 1974), and the paper by Blatchley, *et al.* (1995) include vertical profiles of horizontal velocity recorded in a large channel between the vertical lamp arrays, and downstream of them, showing clear evidence of a boundary layer near the bottom (Iranpour *et al.*, 1998). Does the presence of boundary layers provide a useful starting point for understanding the hydrodynamics and disinfection behaviour in flowing systems?
10. Is there any plan to attempt computational modeling of the hydrodynamics of flowing channels? This and previous publications from this author's research (Blatchley, *et al.*, 1995) indicate that little or no hydrodynamic modeling has been attempted in the past, and it undoubtedly would require much greater computational resources than have been applied in the previous studies. However, with the continuing decrease in computer hardware costs and advances in the capabilities of fluid dynamics software (to the point of recommending finite element decompositions of the volume, with user modifiability of the recommendation) much more could be done now than just a few years ago when the project with a large channel was carried out. Directing a numerical modeling software package to evaluate both equations of fluid flow and the lamp radiation model would provide predictions of dose distributions in flowing systems, thus overcoming one of the difficulties sited on page 2206 of the paper (Iranpour *et al.*, 1997).
11. The other objection, to the lack of a measurement technique for the dose distribution, might be overcome if the hydrodynamic tracer technology used by Anderson and Tchobanoglous (1995) could be modified by using a photochemically sensitive substance that changes color or opacity proportional to the ultraviolet dose that it receives. Is any such chemical known to be currently available, or has there ever been an effort to develop such a technique?

These observations could lead to complementary studies that would put UV disinfection and related technology on a firmer foundation of predictability. Let us repeat once again that this is an excellent report about an excellent project.

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AUTHOR'S REPLY

As compared with other, perhaps more-conventional reactors, current knowledge of photochemical reactor behavior is poorly developed. Yet, in water and wastewater treatment operations, photochemical reactors are being used with increasing frequency. To date, the most common application of photochemical reactors has been in the inactivation of pathogenic microorganisms (i.e. disinfection). However, many other photochemical endpoints are also possible, and processes employing these endpoints are also prevalent in the treatment of potable water, wastewater, hazardous wastes, and air.

While full-scale designs of photochemical reactors are common in engineering practice today, the methods used to design these systems rely heavily on empirical relationships; in general, these relationships provide qualitatively correct representations of the characteristics of photochemical reactor systems, but yield conservative designs. These circumstances have led to a situation in which the majority of currently operating photochemical reactors are vastly overdesigned. Furthermore, the reactor configurations which are available from manufacturers generally can be characterized as having low process efficiency when compared with theoretical estimates of optimum reactor performance. Even with these limitations, currently available reactor configurations are often able to compete effectively with more conventional reactors. For example, UV-based disinfection systems using "off-the-shelf designs" have been shown to be less expensive than chemical disinfection systems (i.e. chlorination or ozonation) for new system construction, and cost-competitive for situations involving retrofit (Blatchley *et al.*, 1996). Nonetheless, it is clear that improvements in our ability to predict reactor performance will yield more efficient system designs and configurations for photochemical reactor systems.

Fundamental process theory can be used to demonstrate that the basic elements needed for reactor system analysis are physically meaningful representations of intrinsic reaction kinetics and transport phenomena which characterize the system. In the case of photochemical reactors, this translates to a need for accurate representations of hydrodynamics and the radiation intensity (irradiance) field, as well as the aforementioned reaction kinetics. The goal of the paper in question was to provide a tool for prediction of the spatial distribution of radiation intensity in these systems, and to illustrate possible applications and limitations of the method. This method, and more sophisticated intensity field models to be developed in the future, will be employed in the development of fundamental photochemical reactor theory, thereby decreasing the need for empiricism in design of photochemical reactor systems.

Existing process models for UV disinfection have been useful as tools for the design of these systems. However, the implicit and explicit assumptions of these models prevent their application as the sole basis for design. Several research groups have recognized the limitations of these models and are working toward development of more refined models. In their comments, Iranpour *et al.* have identified several of the important issues which the research community faces in developing these new-generation models. The following responses are provided to the sequence of questions raised in their comment.

1. In a flat-plate collimator, the vast majority of radiation conditioning is accomplished by the top and bottom plates. Therefore, a minimum of two flat plates is required, but additional plates may be added. Regardless of the number of plates employed, it is critical that the openings in the plates be properly aligned and of a size which will guarantee complete, uniform irradiation of the target. The positions of the plates relative to each other and the lamp axis are also important considerations in the design of a collimator. The plate configurations and spacings illustrated in the paper were judged to provide acceptable collimation, as represented by the LSI model and radiometric measurements. Many other combinations of these parameters could be used to accomplish acceptable performance.
2. Flat-plate collimators are simple to build and have been used in all of the work conducted on UV disinfection within our group at Purdue. The advantage of a collimated radiation source is that radiation intensity can be easily measured (e.g. by radiometry) and the radiation "dose" can be accurately controlled over a broad range of values. Moreover, the physical interpretation of UV dose in a collimated-beam system is unambiguous. This last point has frequently been lost in the evaluation of disinfection kinetics and reactor behavior.

   The Title 22 Regulations used by the State of California stipulate a minimum UV dose to be delivered by a continuous-flow UV system for disinfection of wastewater effluents which are intended for reuse applications. The two most commonly used measures of "dose" in these systems are based on the "bioassay" and "point-source summation" (PSS) methods. The physical interpretations of these dose calculations have been described previously (Blatchley and Hunt, 1994). While both methods have been used to characterize the response of full-scale systems, neither provides an appropriate representation of the exposure histories experienced by microorganisms in traversing the irradiated zones of continuous-flow UV systems. However, the notion of a single-valued representation of dose in such a system has proven to be useful as a practical, empirically based tool for sizing of these systems, albeit with important implications in terms of design conservatism, as described previously.

The approach of using a single value of "dose" to represent these systems is analogous to the "CT concept" which has a long history of (mis-)use in chemical disinfection systems. Just as in the case of the CT concept, a careful examination of reactor behavior reveals important weaknesses of this approach. In recent years, the engineering community has recognized the importance of accurate representations of the physical phenomena which are known to govern reactor performance. In chemical disinfection systems, this translates to the inclusion of hydrodynamic behavior, disinfectant decay, and accurate representations of intrinsic reaction kinetics. In UV systems, an analogous approach, employing hydrodynamics and intensity field modelling, is being used to develop estimates of UV dose distributions (e.g. Chiu *et al.*, 1997, 1998). While more difficult to calculate than a single-valued representation of dose, the dose distribution approach has clear advantages in terms of a physical interpretation of process performance and accurate process predictions (Blatchley and Hunt, 1994).

Like most pilot investigations to date, the studies referred to by Iranpour *et al.* have employed these relatively crude, single-valued representations of UV dose for assessment of process performance. These analyses are useful in that empirical estimates of process performance can be made based on the vast experience of system manufacturers. However, these measurements cannot be used in a strict sense for prediction of process performance. Such a prediction will require an assessment of the dose distribution or an analogous measure of process behavior.

As a further complication in the analysis of UV systems, many new reactors are being built with radiation sources which provide polychromatic output. Strictly speaking, all wavelengths of radiation will display differences in terms of their spatial distribution in a reactor and the ability of corresponding photons to induce photobiochemical changes in microorganisms. To a first approximation, it appears that roughly equivalent dose-response relationships are achieved by monochromatic radiation at $\lambda = 253.7$ nm (the characteristic wavelength of conventional low-pressure Hg arc lamps) and the polychromatic spectrum of UV radiation which characterizes medium-pressure lamps. However, the methods used to assess "UV dose" in medium-pressure collimated-beam systems are somewhat ambiguously defined in the literature, such as the work by Sakamoto and Schwartzel (1995) which is referred to in the comment; moreover, the physical interpretation of a single-valued dose estimate for a continuous-flow system employing these lamps is more difficult to comprehend than those used for a system based on monochromatic radiation. It should be noted that these criticisms are based entirely on a theoretical perspective; in practical terms, UV systems employing lamps with polychromatic output have proven themselves to be effective in disinfection applications. Just as in the case of conventional low-pressure systems, it is likely that the development of more fundamentally sound tools for analysis of these systems will yield substantial benefits, particularly for the engineers who are called upon to design them.

Due to the highly non-linear behavior of photochemical reactor systems, it could be argued that if a single value of dose is to be used to represent their behavior, an estimate of ***minimum UV dose*** will yield a more significant measure of reactor performance than a dose estimate based on conventional PSS or bioassay methods. The vast majority of microorganisms which retain viability downstream of the irradiated zone in a UV system will have followed trajectories through the irradiated zone of the system which yield a low dose. Since areas of low intensity and high velocity are strongly correlated in the conventional designs which exist today (Blatchley *et al.*, 1998), a reasonable representation of minimum UV dose could be easily calculated as the product of minimum UV intensity within the irradiated zone and minimum detention time. Minimum intensity is easily calculated using the PSS or LSI methods (Blatchley, 1997). Minimum detention time can be measured easily using tracer tests (see discussion below).

3. For most kinetic expressions, experiments with continuous-flow systems will yield underestimates of kinetic coefficients unless an accurate representation of transport behavior (i.e. hydrodynamics) is included in the analysis. However, in the strictest sense, there may be some kinetic expressions for which this trend will not apply. Furthermore, in the limit of slow reaction rates, the effects of transport behavior become less significant in terms of overall process performance. For slow reactions, overall process behavior may be limited by reaction kinetics. Most reactions of interest in UV disinfection systems are extremely fast, and process performance in these systems is generally limited by transport

behavior rather than intrinsic reaction kinetics. Therefore, models which do not incorporate a detailed representation of hydrodynamic behavior will not yield accurate process predictions unless they are modified with empirical parameters which allow for model "calibration." Models which rely on these "fitting" parameters are unreliable for evaluation of conditions which extend beyond the limits of calibration. This fact represents an important limitation in the use of these models for scale-up of pilot-test results to full-scale systems.

4. In concept, phased lamp replacement would seem to represent a logical approach. The design of such an approach should be based on a combination of numerical analysis and measurement, and such an investigation would represent a valuable contribution to the industry. Such an investigation would also benefit from an analysis of the variability in lamp output. The method based on a flat-plate collimator and the LSI model described in the manuscript allows these measurements to be conducted rapidly and inexpensively. With a sufficiently large database of these measurements, it would be possible to develop an optimization scheme for lamp replacement, possibly based on a stochastic approach.
5. In conventional UV systems, the water being treated represents an overwhelming heat sink for the system. The effects of influent water temperature on lamp output will be difficult to overcome. However, given the strong influence of temperature on lamp output, there may be merit to the incorporation of temperature control for lamps in UV disinfection systems. Again, the lamp characterization protocol based on the flat-plate collimator and the LSI model would be a valuable tool in the assessment of such an approach.
6. Iranpour *et al.* are correct in pointing out that improvements in numerical modelling of the intensity field can be made. Moreover, these modelling improvements are likely to represent valuable components in future UV process models, especially if they can be shown to yield accurate representations of near-field intensity.
7. It is not entirely clear that model improvements will yield substantial benefits in the measurement or evaluation of lamp output. The relatively simple models based on point-source summation or line-source integration are quite accurate in the far-field, and if properly linked with (far-field) intensity measurements, should yield accurate measurements of lamp output power.
8. Wide-angle detection devices have been used for radiometric measurements and may represent an important improvement for characterization of the intensity field, especially in the near-field.
9. As suggested, boundary-layer theory is a reasonable starting point in the evaluation of hydrodynamics in UV systems. The boundary layers which exist in the vicinity of the wetted surfaces in these systems are responsible for the strong local gradients in velocity (momentum) which characterize these systems.
10. The inclusion of an accurate representation of hydrodynamic behavior is absolutely critical to the development of an accurate process model. Computational and experimental techniques are available for characterization of hydrodynamic behavior. Our group has employed both methods (computational and experimental) for detailed characterization of turbulent hydrodynamic behavior in these systems. The numerically and experimentally derived results have both been incorporated into process models for prediction of process performance. A series of manuscripts describing the results of this work have been submitted for publication in the refereed literature.

    The numerical and experimental characterizations have been based on computational fluid dynamics (CFD) and laser Doppler velocimetry (LDV), respectively. Valuable information has also been gained through the application of flow visualization techniques. While it is clear that these advanced methods hold great promise for improving our knowledge of process behavior, it must also be clearly understood that these approaches are not without problems themselves. The work we have completed to date using these methods, along with similar work being conducted by other research groups, will answer many important questions, but cannot be viewed as a panacea.
11. Tracer testing, as described in the work of Anderson and Tchobanoglous (1995), is useful as a tool for evaluation of one-dimensional behavior. These tests provide a gross representation of overall mixing behavior in one direction (usually the direction of flow). For UV systems, the primary use for these tests is in assessing exposure times within the irradiated zone; these tests also allow an assessment of the presence or absence of reactor short-circuiting. However, it is important to recognize that these results provide no information of transport behavior in the transverse direction. More importantly, the results of conventional tracer tests provide absolutely no information regarding the mixing processes which take place on the scale of true importance in UV systems. As an indication of this scale, one may consider the spatial variations in UV intensity which are characteristic of UV systems under normal operating conditions. It has been shown that intensity may vary by as much as two orders of magnitude over a distance of 5–7 cm (Blatchley, 1997; Chiu *et al.*, 1998). Since photochemical kinetics vary in direct proportion to radiation intensity, this simple analysis reveals that local reaction rates would also be expected to vary by a similar amount over this short distance. Clearly, any process analysis of a UV disinfection system must accurately represent transport behavior in similar detail.

It could be argued that the disinfection process itself provides an opportunity for the use of a photochemically active "tracer" for these systems (i.e. microorganisms). The responses of many microorganisms to UV irradiation are well known and can be used to assess an "equivalent dose," using the commonly applied bioassay method. The shortcomings of this approach have been addressed above. Qualls and Johnson (1983) developed a modification of this technique which allowed for a somewhat more detailed assessment of reactor behavior. Their bioassay technique, which involved a step-change in microbial concentration upstream of the irradiated zone of a system, with time-course sampling at a point downstream, allows a crude estimate of the dose distribution delivered by a system to be made.

It is likely that experimental methods will soon be developed which advance the technique developed by Qualls and Johnson (1983), and provide more detailed information regarding the dose distribution. Our group is currently developing experimental techniques which (we hope) will provide this sort of information. In general, we believe that a combination of numerically and experimentally based techniques can and should be used for analysis of photochemical reactor systems.

Iranpour *et al.* have raised a number of important questions about photochemical reactor performance, and in so doing have pointed out many important research topics. It is clear that the development of tools for the assessment of photochemical reactors will represent an important contribution for both manufacturers and users of these systems.

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