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Modeling and geometry optimization of photochemical reactors: single- and multi-lamp reactors for UV-H₂O₂ AOP systems

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Abstract:

This paper deals with the modeling and optimization of a photochemical reactor for the production of hydroxyl radicals. These radicals can be used in advanced oxidation processes for water purification. The UV-lamps used in the photochemical reactor lead to a relatively high energy and operating cost. Minimization of these costs is possible via a model-based optimization of the reactor geometry. First, a model is built taking into account the reaction kinetics, the reactor geometry and the radiation intensity distribution in the reactor. Afterwards, two different configurations of photochemical reactors are optimized. The first modeled reactor configuration is a single-lamp reactor and consists of a cylindrical UV-lamp that is present inside a quartz sleeve, outside of which the water flows between it and the external reactor wall (annular channel reactor). A rigorous optimization problem is formulated consisting of the calculation of the optimal reactor geometry (length and radius) in such a way that, for a given reactor volume and a given flow rate of the water to be treated, the mean outlet concentration of an organic pollutant is minimized. By solving this optimization problem, it has been shown that an optimal reactor length exists. If the absorption coefficient of the water or the reaction rate

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constant increases, the optimal reactor length increases as well. The second modeled
25 reactor configuration is similar to the first one but now contains multiple lamps
positioned symmetrically in a circular pattern (i.e. multi-lamp reactor). The optimization
problem consists of the calculation of the optimal lamp position inside the reactor in such
a way that the mean outlet concentration of an organic pollutant is minimized. The lamp
position is given by the radial distance between the centers of the reactor and a lamp.
30 Again a well-posed optimization problem is obtained and an optimal lamp position exists.
If the number of lamps or the reaction rate constant increases or the absorption coefficient
of the water decreases, the optimal distance between the centers of the reactor and the
lamp increases.

Keywords: optimization, photochemistry, mathematical modeling, chemical reactors,
35 advanced oxidation processes, multi-lamp photochemical reactor.

1. Introduction

The production of process and drinking water with strongly varying qualities has led to a
40 huge number of water purification techniques during the last decades (Arceivala and
Asolekar, 2008; Pizzi, 2010). One class of these techniques are the *Advanced Oxidation
Processes* (AOPs). These processes use hydroxyl radicals as (secondary) oxidant. These
radicals are formed from a less reactive, primary oxidant (e.g., hydrogen peroxide or
ozone) and have a very high standard reduction potential and so are able to oxidize a
45 large number of organic compounds in water, yielding very low rest concentrations of
these compounds (Mota et al., 2008; Munter, 2001; Stasinakis, 2008).

The performance of the AOP increases when the amount of hydroxyl radicals produced increases. A possible way to increase the hydroxyl radical production is by using high energy electromagnetic waves, usually UV-radiation. These are the so-called *photochemical AOPs* which are carried out in a photochemical reactor. This reactor consists of a vessel in which one or more UV-lamps are present in order to irradiate water. However, the use of these lamps gives rise to a relatively high energy and operating cost. A prediction of the operation and energy costs is possible based on mathematical models (Mota et al., 2008; Stasinakis, 2008).

In order to model a photochemical reactor, knowledge of the radiation intensity I_λ inside the reactor is needed. The radiation intensity I_λ is calculated using a radiation model. Radiation models are divided into two groups: (1) the incidence models and (2) the emission models. The incidence models assume the existence of a given radiant energy distribution in the vicinity of the reactor while the emission models propose a model for the source emission. Table 1 gives an overview of the main radiation models. Most of these models have been developed for tubular reactors (Alfano et al., 1986, Cassano et al., 1995; Elyasi and Taghipour, 2010; Imoberdorf et al., 2008; Romero et al., 1983).

The incidence models are further divided into two- and three-dimensional models. In the 2D-incidence models the radiation intensity I_λ generally depends on the radial and axial coordinate and the radiation travels in planes perpendicular to the reactor axis. In this case, the radiation propagation can be radial (RI), partially diffuse (PI) or diffuse (DI) (see Figure 1 and Table 1). In the 3D-incidence model the radiation intensity I_λ also

70 depends on the angular coordinate. The incidence models are developed for photochemical reactors with an external radiation source (Alfano et al., 1986; Cassano et al., 1995; Elyasi and Taghipour, 2010; Romero et al., 1983).

The classification of the emission models is based on the approximation of the radiation source as either a line source or an extense source. The line source approximation
75 assumes the lamp to have a zero volume while the extense source approximation assumes the lamp to be a cylinder with a certain volume. The line source emission models approximate the lamp as a sequence of point sources, emitting their radiation in either parallel planes perpendicular to the lamp axis (LSPP), in a diffuse way (LSDE) or in a spherical, uniform way (LSSE) (see Figure 2 and Table 1). In the extense source emission
80 models the emission can be volumetric (ESVE) (to be used with arc sources) or superficial (ESSE or ESDE) (to be used with fluorescent sources) (see Table 1) (Alfano et al., 1986; Cassano et al., 1995; Elyasi and Taghipour, 2010; Fullana-i-Palmer et al., 2009; Irazoqui et al., 2000; Romero et al., 1983).

Extense source emission models provide the most realistic description of the radiation
85 sources and consequently give the most accurate results for the radiation intensity I_L . However, these models have the highest complexity and lead to high computational efforts (Alfano et al., 1986; Cassano et al., 1995; Elyasi and Taghipour, 2010; Irazoqui et al., 2000; Romero et al., 1983).

90 Despite this large number of different radiation models available in literature, application of (one of) these models to optimize an annular, cylindrical, homogeneous, photochemical reactor has not been reported in literature. Therefore, this paper addresses

the formulation and solution of a well-posed optimization problem for the model-based optimization of a photochemical reactor geometry. First, a model is built taking into account the reaction kinetics, the reactor geometry and the radiation intensity in the reactor. Two photochemical reactor configurations are optimized, namely a single- and a multi-lamp reactor. The optimization of the former reactor consists of the calculation of the optimal reactor geometry (length and radius) in such a way that, for a given reactor volume and a given flow rate of the water to be treated, the mean outlet concentration of an organic pollutant is minimized. The multi-lamp reactor is similar to the single-lamp reactor but now contains multiple lamps positioned symmetrically in an circular pattern. The optimization problem consists of the calculation of the optimal lamp position inside the reactor in such a way that the mean outlet concentration of an organic pollutant is minimized. The lamp position is given by the radial distance between the centers of the reactor and a lamp. For both reactor configurations, the influence of several process parameters (i.e. the absorption coefficient of the water, the reaction rate constant and, in case of the multi-lamp reactor, the number of lamps) is evaluated.

The structure of the paper is as follows. Section 2 describes the configuration of the single- and multi-lamp photochemical reactors considered in this paper. Section 3 derives the mathematical models for these reactor types. Since knowledge of the reaction kinetics and radiation intensity is required, a general expression for the reaction rate of the organic pollutant and a simplified radiation model will be derived in this section. Section 4 formulates the optimization problem for these reactor types whereas Section 5 discusses the obtained optimization results. Section 6 briefly describes the numerical techniques

and software used to solve the optimization problems. Finally, Section 7 summarizes the main conclusions.

2. Reactor configurations

This section describes the reactor configurations studied.

The first reactor considered in this paper is a single-lamp, annular, cylindrical, homogeneous, photochemical reactor (see Figure 3). The reactor consists of a cylinder-shaped UV-lamp that is present inside a quartz sleeve with a thickness t and an inner radius s , outside of which water flows between it and the external reactor wall (annular channel reactor). Between the lamp and the quartz sleeve an air flow is present in order to cool the lamp. The reactor has an inner radius R_r . The second reactor is similar to the first one but now contains multiple lamps positioned symmetrically in an circular pattern (i.e. multi-lamp reactor) (see Figure 4).

3. Mathematical reactor model

A mathematical reactor model has to be based on the momentum, mass and energy balance equations. Since these equations require knowledge of the reaction kinetics and (in case of a photochemical reactor) of the radiation intensity, Sections 3.1 and 3.2 start with the description of these two aspects, respectively. Finally, Section 3.3 incorporates this knowledge into complete mathematical models for the reactors, which were described in Section 2.

3.1. Reaction kinetics

Advanced oxidation processes (AOPs) are technologies characterized by the generation of hydroxyl radicals ($\text{HO}\cdot$), which are highly reactive and non-selective substances used to degrade toxic organic compounds present in a medium (such as wastewater and soil). The hydroxyl radical has a high reduction potential ($E_0 = 2.80 \text{ V}$) and is able to react with practically all classes of organic compounds. Figure 5 depicts the general reaction mechanism of an AOP. The hydroxyl radicals are formed from a primary oxidant (e.g., hydrogen peroxide or ozone). Hereby, several energy sources (e.g., UV-radiation or ultrasonic waves) and/or several compounds (e.g., semi-conductors or metallic compounds) can *increase* (e.g., UV-radiation) and/or *accelerate* (e.g., TiO_2 -catalyst) the hydroxyl radical production. The organic compounds are broken down by the hydroxyl radicals, resulting in complete mineralization of these compounds, i.e., the formation of carbon dioxide, water and inorganic compounds, or their conversion into less aggressive products with often a higher biodegradability. When the primary oxidant is irradiated with high energy electromagnetic waves (usually UV-radiation) the decomposition of the primary oxidant will rise and so will the hydroxyl radical production. These are the so-called photochemical AOPs which are carried out in a photochemical reactor (Mota et al., 2008; Munter, 2001; Stasinakis, 2008). To simplify the optimization problems, a homogeneous, non-catalytic photochemical AOP (e.g., $\text{H}_2\text{O}_2/\text{UV}$ -process) will be considered. This means that all the reactant molecules are soluble in the water to be treated. In case of the $\text{H}_2\text{O}_2/\text{UV}$ -process the main reaction, responsible for the increased hydroxyl radical production, is the following (Crittenden et al., 1999):

The photon energy content is given by $h\nu$ where h is Planck's constant and ν is the photon wave frequency.

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In order to derive an expression for the reaction rate of the organic pollutant, the reaction rate of the primary oxidant has to be determined. The latter is based on the dimensionless quantum yield Φ_λ^{Ox} which is defined as the number of primary oxidant molecules n_{Ox} consumed per unit time (as a consequence of the photochemical reaction, e.g., Equation (1)) divided by the number of photons (with a certain wavelength λ) absorbed by the primary oxidant Ox during this period, i.e. ϕ_p^{abs} (Oppenländer, 2003):

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$$\Phi_\lambda^{Ox} = \frac{dn_{Ox}/dt}{\phi_p^{abs}} \quad (2)$$

If C_{Ox} , N_A and dV are the primary oxidant concentration, Avogadro's constant and an infinitesimal volume, respectively, the number of molecules dn_{Ox} can be written as (Oppenländer, 2003):

175

$$dn_{Ox} = C_{Ox} \cdot N_A \cdot dV \quad (3)$$

Substituting Equation (3) in Equation (2) yields:

$$\frac{dC_{Ox}}{dt} = \Phi_\lambda^{Ox} \cdot \frac{\phi_p^{abs}}{N_A \cdot V} = \Phi_\lambda^{Ox} \cdot e'_{\lambda, Ox} \quad (4)$$

Here, $e'_{\lambda, Ox}$ is the local, volumetric number rate of photons absorbed by the primary oxidant Ox (at a certain wavelength λ). Usually, Equation (4) is written in terms of energy rates. This can be done by using Planck's law, which yields the following expression for the primary oxidant reaction rate (Oppenländer, 2003):

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$$\frac{dC_{Ox}}{dt} = \Phi_\lambda^{Ox} \cdot \frac{\lambda}{h \cdot c \cdot N_A} \cdot e_{\lambda, Ox} \quad (5)$$

Here, $e_{\lambda, Ox}$ and c are the local, volumetric energy rate of photons absorbed by the primary
oxidant Ox (at a certain wavelength λ) and the speed of light (in water), respectively.

Section 3.2 discusses the calculation of $e_{\lambda, Ox}$.

The rate r_M of the reaction between the hydroxyl radical $HO^\cdot$ and the organic compound
M is given by (Oppenländer, 2003):

$$r_M = -\frac{dC_M}{dt} = k_{HO^\cdot, M} \cdot C_M \cdot C_{HO^\cdot} \quad (6)$$

Here C_M , $C_{HO^\cdot}$ and $k_{HO^\cdot, M}$ denote the concentrations of the organic compound M and the
hydroxyl radical $HO^\cdot$ and the reaction rate constant, respectively. The reaction
mechanism of the photochemical AOP, e.g., the H_2O_2/UV -process, denoted by Equations
(1) and (6), is however far from complete. First of all, other reactions between the
primary oxidant Ox, the hydroxyl radical $HO^\cdot$ and their subsequent products will occur
(see Table 2 for the H_2O_2/UV -process). Secondly, the products M_1^{ox} formed by the
reaction between the organic compound M and the hydroxyl radical $HO^\cdot$, can again react
with the hydroxyl radical $HO^\cdot$, yielding the products M_2^{ox} , and so on (see Figure 6). In
addition, species different from the organic compound M and the species M_i^{ox} can also
react with the hydroxyl radical $HO^\cdot$. These are the interfering compounds S_i which lower
the performance of the AOP (see Figure 6). Because of the complexity of this reaction
mechanism, a simplified approach, based on a pseudo steady state approximation (PSSA),
will be followed in order to derive an expression for the concentration $C_{HO^\cdot}$ and
subsequently the rate r_M . Applying this approach yields the following expression for the
reaction rate of the organic pollutant (Oppenländer, 2003):

$$-\frac{dC_M}{dt} = \frac{k_{HO\cdot,M} \cdot C_M \cdot v_{rx} \cdot \frac{dC_{Ox}}{dt}}{k_{HO\cdot,M} \cdot C_M + \sum_i k_{HO\cdot,S_i} \cdot C_{S_i}} \quad (7)$$

Here v_{rx} denotes the stoichiometric coefficient of the photochemical decomposition reaction and equals 2 in case of the H_2O_2 /UV-process, see Equation (1), whereas $\frac{dC_{Ox}}{dt}$ is denoted by Equation (5). $k_{HO\cdot,S_i}$ is the reaction rate constant of the reaction between the hydroxyl radical $HO\cdot$ and the interfering compounds S_i . Two limits of Equation (7) are of special interest:

- 1) If the concentration C_M is high, such that $k_{HO\cdot,M} \cdot C_M \gg \sum_i k_{HO\cdot,S_i} \cdot C_{S_i}$, then the reaction between the organic compound M and the hydroxyl radical $HO\cdot$ dominates the reaction between the hydroxyl radical $HO\cdot$ and the interfering compounds S_i and the overall rate expression (7) simplifies to (Oppenländer, 2003):

$$-\frac{dC_M}{dt} = v_{rx} \cdot \frac{dC_{Ox}}{dt} = k_0 \quad (8)$$

Here k_0 denotes the reaction rate constant of the zeroth order reaction.

- 2) If the concentration C_M is low, so that $k_{HO\cdot,M} \cdot C_M \ll \sum_i k_{HO\cdot,S_i} \cdot C_{S_i}$, then the reaction between the hydroxyl radical $HO\cdot$ and the interfering compounds S_i dominates the reaction between the organic compound M and the hydroxyl radical $HO\cdot$ and the overall rate expression (7) simplifies to (Oppenländer, 2003):

$$-\frac{dC_M}{dt} = \frac{k_{HO\cdot,M} \cdot C_M \cdot v_{rx} \cdot \frac{dC_{Ox}}{dt}}{\sum_i k_{HO\cdot,S_i} \cdot C_{S_i}} = k_1 \cdot C_M \quad (9)$$

Here k_1 denotes the reaction rate constant of the first order reaction. Equation (9) will be applied in the following sections of the current paper.

To summarize, the following assumptions were made when deriving Equation (7).

1. The following reactions are neglected, i.e., the reactions taking place between: (a) the primary oxidant Ox, the hydroxyl radical HO $\cdot$ and their subsequent products (see e.g. Equations (A.2) – (A.6) in Table 2), (b) the hydroxyl radical HO $\cdot$ and the species M $_i^{\text{ox}}$, and the hydroxyl radical HO $\cdot$ and the species S $_i^{\text{ox}}$, and (c) the organic compound M and other radicals, e.g., the hydroxyperoxyl radical HO $_2\cdot$.
2. The direct photolysis of the organic compound M (i.e. the photochemical decomposition of the organic compound M when the UV-radiation directly hits this compound) is also neglected. This is acceptable when the concentration C_M is low.
3. Mass transfer limitations are neglected. This is acceptable since the AOP is assumed to be homogeneous.

3.2. Radiation intensity

First of all, an expression for the rate $e_{\lambda,ox}$, used in Equation (5), has to be derived. By solving the *radiative transfer equation (RTE)* (i.e. the conservation equation for radiative energy), the following expression for the rate $e_{\lambda,ox}$ can be determined (Alfano and Cassano, 2008; Bandini et al., 1975; Cassano et al., 1995; Elyasi and Taghipour, 2010; Huang et al., 2011; Modest, 2003):

$$e_{\lambda,ox} = \alpha_{\lambda,ox} \cdot I_{\lambda} \quad (10)$$

Here $\alpha_{\lambda,ox}$ and I_{λ} denote the volumetric absorption coefficient of the oxidant and the radiation intensity (at a certain wavelength λ), respectively. When solving the RTE, yielding Equation (10), the following assumptions were made (Alfano and Cassano, 2008; Elyasi and Taghipour, 2010; Modest, 2003):

1. Since the AOP is assumed to be homogeneous, light scattering can be neglected.
- 250 2. Since the process temperature of the AOP is usually low, light emission can also be neglected.

If $\epsilon_{\lambda,ox}$ is the molar absorption coefficient of the oxidant, the absorption coefficient $\alpha_{\lambda,ox}$ is defined as follows (Oppenländer, 2003):

$$\alpha_{\lambda,ox} = \ln 10 \cdot \epsilon_{\lambda,ox} \cdot C_{ox} \quad (11)$$

255 The radiation intensity I_λ can be calculated using a radiation model. To simplify the calculations in the current paper and in order to obtain generic insight, a simple radiation model, namely the LSPP-model (Line Source with Parallel plane emission), is applied. The LSPP-model assumes the lamp as a line source where each point emits radiation in parallel planes perpendicular to the lamp axis. In the following, Sections 3.2.1 and 3.2.2 describe the derivation of the mathematical expression for the radiation intensity I_λ in the single-lamp and the multi-lamp reactor, based on the LSPP-model, respectively.

3.2.1. Single-lamp reactor

265 The LSPP-model is only valid for the reactor depicted in Figure 3. Performing a radiation balance for a homogeneous control volume, introducing Lambert-Beer's law and integrating along the radiation path, the final expression for the radiation intensity I_λ is as follows (Alfano et al., 1986, Placido and Nascimento, 2007):

$$I_\lambda = \frac{P_l}{2 \cdot \pi \cdot r \cdot L_l} \cdot \exp(-\alpha_{\lambda,q} \cdot t) \cdot \exp[-\alpha_{\lambda,w} \cdot (r - s - t)] \quad (12)$$

270 Here P_b , L_b , $\alpha_{\lambda,q}$, $\alpha_{\lambda,w}$, and r denote the effective radiation power of the lamp (assuming the lamp to be monochromatic), the length of the lamp, the volumetric absorption

coefficients of quartz and water and the radial distance between a point in the reactor and the center line of the lamp, respectively. The LSPP-model has the following limitations:

1. The lamp emitting radiation in a radial way, is a simplification of the real emitting character of a lamp.
2. The model neglects reflection and refraction of radiation at the interfaces in the reactor, namely the air/quartz-, quartz/water- and water/reactor wall-interface.
3. The use of the integrated form of Lambert-Beer's law implies the following assumptions (Ingle and Crouch, 1988):
 - a. The assumptions made when deriving Equation (10).
 - b. The irradiated water is homogeneous (i.e. the same composition and temperature) along the entire radiation path.
 - c. The irradiated water is not turbid.
 - d. The concentration of the species which absorb the radiation is low.
4. As a consequence of the radial radiation emission, the model neglects the loss of radiation at the reactor in- and outlet. This assumption is more permissible as the ratio of the reactor radius R_r over the reactor length L_r becomes smaller.
5. The length L_l needs to be at least as large as the length L_r (Placido and Nascimento, 2007).

In order to overcome the second limitation of the LSPP-model, the authors developed an adapted version of the LSPP-model, taking into account the (repeated) reflection and refraction of the radiation at the interfaces present in the reactor, yielding the following expression for the radiation intensity I_λ (Coenen, 2012):

$$I_\lambda = \frac{P_l}{2 \cdot \pi \cdot r \cdot L_l} \cdot \frac{(1-R_{aq}) \cdot (1-R_{qw}) \cdot T_q \cdot T_w [r-s-t] \cdot (R_{wr} \cdot T_w^2 [R_r-r] + 1)}{(1-T_q^2 \cdot R_{qw} \cdot R_{aq}) \cdot \left[1 - T_w^2 [R_r-s-t] \cdot R_{wr} \cdot \left[R_{qw} + (1-R_{qw})^2 \cdot \frac{T_q^2 \cdot R_{aq}}{1-T_q^2 \cdot R_{qw} \cdot R_{aq}} \cdot \frac{s}{s+t} \right] \right]} \quad (13)$$

Here R_{aq} , R_{qw} and R_{wr} denote the reflectivities of the air/quartz-, quartz/water- and the water/reactor wall-interface, respectively, and are calculated by means of the Fresnel equations. If an electromagnetic wave travels from a medium with refractive index n_1 to a medium with refractive index n_2 and perpendicular to the interface between this two media, then the reflectivity (i.e. the fraction of the radiative energy reflected back into medium 1 at this interface) R_{12} of this interface is given by (Modest, 2003):

$$R_{12} = \left(\frac{n_1 - n_2}{n_1 + n_2} \right)^2 \quad (14)$$

In Equation (13), T_q and T_w denote the transmittivities (i.e. the fraction of the radiative energy transmitted by an absorbing medium) of the quartz sleeve and the water, respectively. Based on Lambert-Beer's law, the transmittivities T_q and T_w are calculated as follows (Modest, 2003):

$$T_q = \exp(-\alpha_{\lambda,q} \cdot t) \quad (15)$$

$$T_w[r] = \exp(-\alpha_{\lambda,w} \cdot r) \quad (16)$$

The adapted LSPP-model of the authors (Coenen, 2012) will be applied to the single-lamp photochemical reactor.

3.2.2. Multi-lamp reactor

To simplify the calculations in the current paper, the calculation of the radiation intensity I_λ in the multi-lamp reactor will be also based on the LSPP-model. However, the model given by Equation (13) is only applicable to the reactor presented in Figure 3 in which the axes of the lamp and the reactor coincide. On the other hand, these axes do not coincide in the multi-lamp reactor. If, however, the reflectivity R_{wr} equals zero then the LSPP-

model is also applicable to the multi-lamp reactor. In addition, the following assumptions will be made when calculating the radiation intensity I_λ in the multi-lamp reactor:

1. The reactor wall absorbs all incoming radiation.
2. Concerning *the interaction of a lamp with the radiation emitted by the other lamps in the reactor*, each lamp is assumed to have a 100% transmittance. This means that shadow effects (i.e. the interception of the radiation by other lamps), absorption, re-emission, reflection and refraction of this radiation in a lamp are neglected.

From the conclusions made by Georges et al. (2008), Imoberdorf et al. (2008) and Jin et al. (2005), it follows that the above mentioned assumptions can be justified. In addition, neglecting the shadow effects will be more justified when (i) the (outer) radius of the lamps decreases, (ii) the distance between the lamps increases or (iii) the ratio of the volume occupied by the lamps in the reactor over the volume occupied by the water in the reactor decreases.

Taking into account the above mentioned considerations, the radiation intensity I_λ in the multi-lamp reactor will be calculated as follows: the radiation intensity $I_{\lambda,j}$ in point A as a result of the radiation emitted by the j^{th} lamp (see Figure 7) equals:

$$I_{\lambda,j} = \frac{P_{l,j}}{2\pi \cdot r_j \cdot L_l} \cdot \frac{(1-R_{aq}) \cdot (1-R_{qw}) \cdot T_q \cdot T_w [r_j - R_l]}{(1-T_q^2 \cdot R_{qw} \cdot R_{aq})} \quad (17)$$

$$P_{l,j} = \frac{P_{l,tot}}{N_l} \quad (18)$$

Here, $P_{l,j}$, N_l , $P_{l,tot}$ and r_j denote the effective radiation power of the j^{th} lamp (assuming the lamp to be monochromatic), the number of lamps in the reactor, the total effective radiation power of all the lamps in the reactor and the distance between point A and the center of the j^{th} lamp (see Figure 6), respectively. If (r, θ) and (r_j^*, θ_j^*) are the polar

340 coordinates of A and the center point of the j^{th} lamp, respectively, then the radial distance
 r_j can be written as:

$$r_j = \sqrt{(r \cdot \cos \theta - r_j^* \cdot \cos \theta_j^*)^2 + (r \cdot \sin \theta - r_j^* \cdot \sin \theta_j^*)^2} \quad (19)$$

$$\theta_j^* = \Delta\theta \cdot (j - 1) = \frac{2\pi}{N_l} \cdot (j - 1) \quad (20)$$

Finally, the total radiation intensity I_λ in A equals the sum of the radiation intensities in A,
345 resulting from each individual lamp:

$$I_\lambda = \sum_{j=1}^{N_l} I_{\lambda,j} \quad (21)$$

The radial distance r_j^* is equal for each lamp and will be denoted by the symbol r^* .

3.3.Balance equations

350 The reactor under study is a photochemical reactor, either a single- (see Figure 3) or a
multi-lamp reactor (see Figure 4) in which a homogeneous, non-catalytic AOP (e.g., the
 $\text{H}_2\text{O}_2/\text{UV}$ -process) takes place. The water contains an organic pollutant M (which has to
be degraded) and also some interfering components S_i . The process is stationary and
isothermal. The reactor and the lamp(s) have the same length. The flow in the reactor is a
355 plug flow and radial and axial dispersion of the species are neglected.

The mathematical reactor model is based on the momentum, mass and energy balance
equations. Since the flow in the reactor is assumed to be a plug flow, the momentum
balance equation is however unnecessary. Generally, the energy consists of mechanical,
thermal and radiative energy. Because of the isothermal conditions, thermal energy can
360 be eliminated from the energy balance equation. In addition, the mechanical and radiative
energy can be uncoupled, resulting in a balance equation for the radiative energy (i.e., the

RTE) (Alfano et al., 2008; Cassano et al., 1995). The RTE was already solved in Section 3.2, resulting in Equation (10). The only balance equation left is the mass balance equation of the organic compound M. In the following, Sections 3.3.1 and 3.3.2 describe the derivation of this mass balance equations for the single-lamp and the multi-lamp reactor, respectively.

3.3.1. Single-lamp reactor

Considering the above mentioned assumptions, the mass balance equation of the organic compound M can be written as:

$$0 = -u \cdot \frac{\partial C_M(r, z)}{\partial z} - r_M(r, z) \quad (22)$$

with:

$$R_i \leq r \leq R_r \quad \text{and} \quad 0 \leq z \leq L_r$$

and the following boundary condition:

$$C_M(r, z = 0) = C_{M,0} \quad , R_i \leq r \leq R_r \quad (23)$$

Here u , z , r_M and $C_{M,0}$ denote the plug flow speed, the axial coordinate (which varies from 0 to the length L_r), the reaction rate of the organic compound M (denoted by the right hand side of Equation (7)) and the inlet concentration of the organic compound M, respectively. Because of the radial dependence of the radiation intensity I_λ , the reaction rate r_M will also depend on the radial coordinate r . As a consequence the concentration C_M will depend on both the axial and the radial coordinate, z and r , respectively. To further simplify Equation (7), the following assumptions will be made:

1. The concentration C_M is low and much smaller than the concentration C_{S_i} .
2. The concentration C_{S_i} is very large and therefore constant throughout space.

385 3. The monochromatic radiation, emitted by the lamp(s), is not only absorbed by the
primary oxidant but also by other components which have a constant
concentration (in space).

From the above mentioned assumptions, it follows that the rate r_M is denoted by Equation
(9). As a consequence, the rate r_M can be written as:

390
$$r_M = k \cdot I_\lambda \cdot C_M \quad (24)$$

Here k is the constant reaction rate constant. In case of the single- and multi-lamp reactor,
the radiation intensity I_λ is denoted by Equations (13) and (21), respectively.

In order to obtain more physical insight in the (photo)chemical process, the non-
395 dimensional mathematical model, denoted by Equations (22) – (24), will be rewritten as a
function of dimensionless parameters. Assuming the reflectivities R_{aq} , R_{qw} , R_{wr} and the
absorption coefficient $\alpha_{\lambda,q}$ equal to zero and then performing a dimension analysis, based
on the Buckingham Pi-theorem (Bird et al., 2002; Graebel, 2001), the dimensionless
model for the single-lamp photochemical reactor can be written as:

400
$$0 = -\frac{\partial c'_M}{\partial z'} - Da \cdot I'_\lambda \cdot c'_M \quad (25)$$

$$I'_\lambda = \frac{\exp(-\Pi_2 \cdot r')}{1 + \left(\frac{1}{\Pi_1} - 1\right) \cdot r'} \quad (26)$$

with:

$$0 \leq r' \leq 1 \quad \text{and} \quad 0 \leq z' \leq 1$$

and the following boundary condition:

405
$$c'_M(r', z' = 0) = 1 \quad , 0 \leq r' \leq 1 \quad (27)$$

Here, the dimensionless parameters are defined as follows:

$$c'_M = \frac{c_M}{c_{M,0}}, r' = \frac{r-R_i}{R_r-R_i}, z' = \frac{z}{L_r}, \Pi_1 = \frac{R_i}{R_r}, \Pi_2 = \alpha_{\lambda,w} \cdot (R_r - R_i), \Pi_5 = \frac{R_r}{L_r}, I'_\lambda = \frac{I_\lambda}{I_{\lambda,b}} \quad (28)$$

$$Da = \frac{\tau}{t_{rx}}, \tau = \frac{L_r}{u}, t_{rx} = \frac{1}{k \cdot I_{\lambda,b}} \quad (29)$$

$$I_{\lambda,b} = \frac{P_l}{2 \cdot \pi \cdot R_i \cdot L_r} \quad (30)$$

410 and have the following meaning: c'_M , I'_λ , r' and z' denote the dimensionless concentration of the organic compound M, the dimensionless radiation intensity and the dimensionless radial and axial coordinate, respectively. $I_{\lambda,b}$ is the radiation intensity evaluated at the lamp boundary (i.e. at $r = R_i$). The parameters Π_1 and Π_5 are measures for the narrowness of the annulus and the reactor, respectively, whereas the parameter Π_2 is a measure for
415 the radiation absorption by the aqueous medium in the reactor. Finally, the parameter Da denotes the Damköhler number which is the ratio of the mean residence time τ over the characteristic reaction time t_{rx} (Levenspiel, 1999).

3.3.2. Multi-lamp reactor

420 For the multi-lamp reactor, the non-dimensionless form of the mass balance equation for the organic compound M and the boundary condition are similar to the single-lamp reactor (see Equations (22) and (23)):

$$0 = -u \cdot \frac{\partial c_M(r, \theta, z)}{\partial z} - r_M(r, \theta, z) \quad (31)$$

with:

$$425 \quad 0 \leq r \leq R_r, \quad 0 \leq \theta \leq 2\pi \quad \text{and} \quad 0 \leq z \leq L_r$$

and the following boundary condition:

$$c_M(r, \theta, z = 0) = c_{M,0}, \quad 0 \leq r \leq R_r, \quad 0 \leq \theta \leq 2\pi \quad (32)$$

Here, the only differences between the two reactor configurations are the boundary values for the radial coordinate r which, in case of the multi-lamp reactor, are 0 and R_r , and the reaction rate r_M and concentration C_M which now also depend on the angular coordinate θ due to the angular dependence of the radiation intensity I_λ .

The dimensionless mass balance equation for the multi-lamp reactor can be written as:

$$0 = -\frac{\partial c'_M}{\partial z'} - \frac{Da}{N_l} \cdot I'_\lambda \cdot c'_M \quad (33)$$

with:

$$0 \leq r' \leq 1, \quad 0 \leq \theta \leq 2\pi \quad \text{and} \quad 0 \leq z' \leq 1$$

and the following boundary condition:

$$c'_M(r', \theta, z' = 0) = 1, \quad 0 \leq r' \leq 1, 0 \leq \theta \leq 2\pi \quad (34)$$

Assuming the reflectivities R_{aq} , R_{qw} , R_{wr} and the absorption coefficient $\alpha_{\lambda,q}$ equal to zero, the expression for the radiation intensity I'_λ now becomes:

$$I'_\lambda = \Pi_1 \cdot \sum_{j=1}^{N_l} \frac{\exp\left[-\Pi_2 \cdot \left(\sqrt{r'^2 + r^{*2} - 2 \cdot r' \cdot r^* \cdot \cos(\theta - \theta_j^*)} - \Pi_1\right)\right]}{\sqrt{r'^2 + r^{*2} - 2 \cdot r' \cdot r^* \cdot \cos(\theta - \theta_j^*)}} \quad (35)$$

Here, the dimensionless parameters are defined as follows:

$$c'_M = \frac{C_M}{C_{M,0}}, r' = \frac{r}{R_r}, z' = \frac{z}{L_r}, \theta, \Pi_1 = \frac{R_i}{R_r}, \Pi_2 = \alpha_{\lambda,w} \cdot R_r, \Pi_5 = \frac{R_r}{L_r} \quad (36)$$

$$N_l, r^* = \frac{r^*}{R_r}, I'_\lambda = \frac{I_\lambda}{I_{\lambda,j,b}} \quad (37)$$

$$I_{\lambda,j,b} = \frac{P_{l,j}}{2 \cdot \pi \cdot R_i \cdot L_r} \quad (38)$$

$$I_{\lambda,b} = \frac{P_{l,tot}}{2 \cdot \pi \cdot R_i \cdot L_r} \quad (39)$$

$$Da = \frac{\tau}{t_{rx}}, \tau = \frac{L_r}{u}, t_{rx} = \frac{1}{k \cdot I_{\lambda,b}} \quad (40)$$

The physical meanings of the parameters c'_M , r' , z' , Π_1 , Π_2 , Π_5 , Da and I'_λ are the same as for the single-lamp photochemical reactor. r^{*} is the dimensionless distance between the center of a lamp and the reactor.

4. Optimization problem formulation

In this section a well-posed optimization problem is formulated for both a single- and multi-lamp reactor. To this end the available degrees of freedom have to be identified. In addition, an objective function, which mathematically quantifies the cost to be minimized, has to be specified. Finally, also constraints can be added.

4.1. Single-lamp reactor

The optimization problem for the single-lamp photochemical reactor is the following. Given is a photochemical reactor, depicted by Figure 3 and described mathematically in Section 3.3. This reactor has a constant volume V_r through which water flows with a given volumetric flow rate Q . The other parameters which were considered to remain constant are the radius R_i , the radiation power P_b , the absorption coefficient $\alpha_{\lambda,w}$ and the reaction rate constant k . The optimization problem consists of the calculation of the optimal reactor length $L_{r,opt}$ in such a way that the mean outlet concentration $c'_{M,av}$ of the organic pollutant is minimized.

The concentration $c'_{M,av}$ is calculated in the following way: Equation (25) has the following analytical solution:

$$c'_M(r', z') = \exp\left[-Da \cdot \frac{\exp(-\Pi_2 \cdot r')}{1 + \left(\frac{1}{\Pi_1} - 1\right) \cdot r'} \cdot z'\right] \quad (41)$$

At the reactor outlet (i.e. $z' = 1$), the following holds:

$$c'_M(r', z' = 1) = \exp[-Da \cdot \frac{\exp(-\Pi_2 \cdot r')}{1 + (\frac{1}{\Pi_1} - 1) \cdot r'}] \quad (42)$$

The calculation of the concentration $c'_{M,av}$ is based on the following equation:

$$C_{M,av} \cdot \int_{R_i}^{R_r} 2 \cdot \pi \cdot r \cdot u \cdot dr \triangleq \int_{R_i}^{R_r} 2 \cdot \pi \cdot r \cdot u \cdot C_M(r, z = L_r) \cdot dr \quad (43)$$

Here, $C_{M,av}$ is the average concentration of the organic compound M at the reactor outlet.

Substituting the dimensionless parameters (28) – (30) into Equation (43) finally leads to:

$$c'_{M,av} = \frac{2}{1 + 1/\Pi_1} \cdot \int_0^1 \left[1 + \left(\frac{1}{\Pi_1} - 1 \right) \cdot r' \right] \cdot c'_M(r', z' = 1) \cdot dr' \quad (44)$$

with $c'_M(r', z' = 1)$ given by Equation (42).

From the above mentioned constraints, it follows that the fluid velocity u and the radius R_r are no longer freely variable because they can be determined in the following way:

$$R_r = \sqrt{\frac{V_r}{\pi \cdot L_r} + R_i^2} \quad (45)$$

$$u = \frac{Q \cdot L_r}{V_r} \quad (46)$$

The mathematical procedure to perform the optimization is the following. For a certain value of the length L_r and the given values of the volume V_r , the volumetric rate Q , the radius R_i , the radiation power P_l , the absorption coefficient $\alpha_{\lambda,w}$ and the reaction rate constant k , first calculate the radius R_r and the velocity u by means of Equations (45) and (46). Secondly, compute the values of the dimensionless parameters, denoted by Equations (28) – (30), and finally the value of the concentration $c'_{M,av}$ by means of Equation (44). Repeat this procedure for different values of the length L_r until the minimum $c'_{M,av,min}$ of the concentration $c'_{M,av}$ as a function of the length L_r is found.

4.2. Multi-lamp reactor

The optimization problem for the multi-lamp photochemical reactor is defined as follows. Given is a photochemical reactor, depicted by Figure 4 and described mathematically in Section 3.3. This reactor contains N_l identical lamps with a total, effective radiation power $P_{l,tot}$ and each with an outer radius R_l . The reactor has a constant geometry, denoted by the length L_r and the radius R_r . Through this reactor, an aqueous medium flows with a plug flow velocity u and absorption coefficient $\alpha_{\lambda,w}$. The reaction rate constant k is also given. The optimization problem consists of the calculation of the optimal value r_{opt}^* of the radial distance r^* in such a way that the concentration $c'_{M,av}$ at the reactor outlet is minimized.

The calculation of the concentration $c'_{M,av}$ is analogous to Section 4.1: Equation (33) has the following analytical solution:

$$c'_M(r', \theta, z') = \exp\left[-\frac{Da}{N_l} \cdot I'_\lambda(r', \theta) \cdot z'\right] \quad (47)$$

At the reactor outlet (i.e. $z' = 1$), the following holds:

$$c'_M(r', \theta, z' = 1) = \exp\left[-\frac{Da}{N_l} \cdot I'_\lambda(r', \theta)\right] \quad (48)$$

The calculation of the concentration $c'_{M,av}$ is based on the following equation:

$$C_{M,av} \cdot \int_{A_{wet}} u \cdot dA \triangleq \int_{A_{wet}} u \cdot C_M(r, \theta, z = L_r) \cdot dA \quad (49)$$

with A_{wet} the 'wet surface' (i.e. the cross section area available for the flow of the water in the reactor) (see Figure 7). Substituting the dimensionless parameters (36) – (40) into Equation (49) and taking into account the symmetry of the cross section of the multi-lamp reactor (see Figure 7) finally leads to:

$$c'_{M,av} = \frac{\int_{A_{wet}} c'_M(r', \theta, z'=1) \cdot r' \cdot dr' \cdot d\theta}{\int_{A_{wet}} r' \cdot dr' \cdot d\theta} = \frac{\int_{A_{wet}} c'_M(r', \theta, z'=1) \cdot r' \cdot dr' \cdot d\theta}{\pi \cdot (1 - N_l \cdot \Pi_1^2)} =$$

$$\frac{2.N_l}{\pi.(1-N_l.\Pi_1^2)} \int_{A'} c'_M(r', \theta, z' = 1).r'.dr'.d\theta \quad (50)$$

with $c'_M(r', \theta, z' = 1)$ denoted by Equation (48).

During the optimization, two constraints on the radial distance r^* are imposed, namely a
 515 minimal and maximal value, r_{min}^* and r_{max}^* , respectively. If the radial distance r^* equals
 its minimal value r_{min}^* , the lamps touch each other in the reactor. If the radial distance r^*
 equals its maximal value r_{max}^* , the lamps touch the reactor wall. For this reason, the
 expressions for the radial distances r_{min}^* and r_{max}^* are the following:

$$r_{min}^* = \frac{R_i}{\sin \frac{\pi}{N_l}} \quad (51)$$

$$r_{max}^* = R_r - R_i \quad (52)$$

The mathematical procedure to perform the optimization is the following: for a certain
 value of the length L_r , the radius R_r , the radius R_i , the radiation power $P_{l,tot}$, the number of
 lamps N_l , the velocity u , the absorption coefficient $\alpha_{i,w}$ and the reaction rate constant k ,
 calculate the values of the dimensionless parameters, denoted by Equations (36) – (40),
 525 and finally the concentration $c'_{M,av}$ by means of Equation (50). Repeat this procedure for
 different values of the radial distance r^* until the minimum $c'_{M,av,min}$ of the concentration
 $c'_{M,av}$ as a function of the radial distance r^* is found.

5. Results and discussion

In this section, the obtained results are presented for both reactor configurations.

5.1. Single-lamp reactor

Figure 8 depicts the results of the optimization of a single-lamp photochemical reactor by performing the procedure, described in Section 4.1. Based on this figure, it follows that an optimal value of the length L_r exists (i.e., $L_r = 5,4$ m). The existence of an optimal reactor length $L_{r,opt}$ can be explained by the fact that due the constant value of the radiation power P_b , the ratio P_l/L_l increases when the length L_l decreases. As a consequence, the radiation intensity I_λ and the rate r_M will increase in the vicinity of the lamp (see Equations (13) and (24), respectively). However, from Equation (45) it follows that the radius R_r increases and also the parameter Π_2 (see Equation (28)). A rise of the parameter Π_2 leads to a decrease in the radiation intensity I_λ and also the rate r_M (see Equations (26) and (24), respectively). As a result, an optimal value of the length L_r exists.

Secondly, it is observed in Figure 8 that the concentration $c'_{M,av}$ converges to a value of one for very small and very large values of the reactor length L_r . The latter can be explained as follows: since the volume V_r and the volumetric rate Q are constant, the residence time τ is also constant (see Equations (29) and (46)). However, from Equations (29) and (30), it follows that the characteristic reaction time t_{rx} depends on the length L_r and goes to infinity when the length L_r goes to infinity. Consequently, the parameter Da also goes to zero (see Equation (29)) and the decomposition of the organic compound M as well (see Equation (25)). On the other hand, when the length L_r becomes extremely small, the annulus becomes very wide (i.e. the parameter Π_1 goes to zero, see Equations (28) and (45)) and the radiation absorption increases (i.e. the parameter Π_2 increases, see Equations (28) and (45)). These two effects imply a decrease in the radiation intensity I'_λ

and also of the rate r_M (see Equations (26) and (24), respectively). As a result, the concentration $c'_{M,av}$ will become close to one.

From the above explanation of the existence of the length $L_{r,opt}$ it follows that when the radiation absorption in the reactor increases, a smaller reactor (i.e. a reactor with a smaller value of the parameter Π_5) has a higher performance. This is in accordance with Figures 9 and 10 (left): the length $L_{r,opt}$ increases when the absorption coefficient $\alpha_{\lambda,w}$ increases. In addition, the length $L_{r,opt}$ goes to zero and infinity when the absorption coefficient $\alpha_{\lambda,w}$ goes to zero and infinity, respectively. When the radiation absorption increases, less radiation will be available to decompose the organic compound M and so the concentration $c'_{M,av,min}$ will increase and finally converge towards one. If no radiation absorption occurs, the organic compound M can be decomposed completely and the concentration $c'_{M,av,min}$ will converge towards zero (see Figure 10 (right)).

The influence of the reaction rate constant k on the optimization results is depicted in Figures 11 and 12. When the reaction rate constant k is smaller than $10^{-2} \text{ m}^2/\text{J}$, the length $L_{r,opt}$ increases when the reaction rate constant k increases. An explanation is the following: the only dimensionless parameter containing the reaction rate constant k is the Damköhler number Da (see Equations (28) – (30)). When the reaction rate constant k decreases, the Damköhler number Da will also decrease and so will the decomposition of the organic compound M. To compensate for this decrease in the decomposition, a decrease in the length L_r is necessary. After all, a decrease in the length L_r implies an increase in the radiation intensity I'_λ and also the rate r_M (see Equations (26) and (24), respectively). An explanation for the increase in the concentration $c'_{M,av,min}$ when the reaction rate constant k decreases (see Figure 12 (right)), is twofold. First of all, when the

reaction rate constant k decreases, the length $L_{r,opt}$ also decreases. As a consequence, the
 580 annulus of the reactor becomes wider and so more radiation absorption will occur in the
 aqueous medium. This implies an increase in the concentration $c'_{M,av,min}$. Secondly, when
 the reaction rate constant k decreases, the reaction rate r_M decreases and so will the
 decomposition of the organic compound M.

When the reaction rate constant k is very large, a very fast, total decomposition of the
 585 organic compound M will occur, leading to a very fast decline of the concentration of this
 compound throughout the reactor. As a consequence, the influence of the reactor
 geometry on the reactor performance is limited, resulting in a flat curve near the
 minimum (e.g., see the curve with $k = 10^{-1} \text{ m}^2/\text{J}$ in Figure 11) and an almost constant
 value for the optimal length as shown in Figure 12 (left). Analogously, this behavior also
 590 occurs for very small values of the reaction constant k , i.e. when no decomposition of the
 organic compound M occurs (see Figures 11 and 12 (left)).

5.2. Multi-lamp reactor

Figure 13 (left) depicts the results of the optimization of a multi-lamp photochemical
 595 reactor by performing the procedure, described in Section 4.2. From this figure, it can be
 concluded that an optimal value of the radial distance r^* exists. In this case the radial
 distance r_{opt}^* and the dimensionless concentration $c'_{M,av,min}$ equal 0.35 m and 0.54,
 respectively. The existence of the radial distance r^* can be explained by means of the
 second principle of *Process Intensification (PI)*, namely ‘Give each molecule the same
 600 *process experience*.’ According to Van Gerven and Stanckiewicz (2009) application of
 this principle will enhance the performance of the photochemical reactor. In case of a

homogeneous, non-catalytic, isothermal, photochemical reactor, a measure for the process experience of a molecule is its radiation dose D (Braslavsky, 2007):

$$D = \int_0^\tau I_\lambda \cdot dt \quad (53)$$

605 with τ the residence time of the molecule in the reactor. For the general case, in which the radiation intensity I_λ and the axial velocity u in the cylindrical reactor depend on the radial coordinate r , the angular coordinate θ and the axial coordinate z , the radiation dose D can be written as:

$$D(r, \theta) = \int_0^{L_r} \frac{I_\lambda(r, \theta, z)}{u(r, \theta, z)} \cdot dz \quad (54)$$

610 From the second principle of PI, it follows that the performance of the reactor will be enhanced if the spatial variation of the radiation dose $D(r, \theta)$ decreases. A measure for this spread is the variance of the radiation dose $D(r, \theta)$:

$$\sigma_D^2 = \frac{\int_{A_{wet}} (D(r, \theta) - D_{av})^2 \cdot dA}{A_{wet}} \quad (55)$$

with the surface A_{wet} as defined in Equation (49) and D_{av} the average radiation dose:

$$615 \quad D_{av} = \frac{\int_{A_{wet}} D(r, \theta) \cdot dA}{A_{wet}} \quad (56)$$

In case of a plug flow reactor (i.e., where the velocity u is independent of the coordinates r , θ and z) and the radiation intensity I_λ independent of the axial coordinate z , Equation (54) can be written as:

$$D(r, \theta) = I_\lambda(r, \theta) \cdot \frac{L_r}{u} \quad (57)$$

620 From Equation (57), it follows that the explanation of the optimization results can be based on the radiation intensity $I_\lambda(r, \theta)$ instead of the radiation dose $D(r, \theta)$. Analogous to

Equations (55) and (56), the variance $\sigma_{I_\lambda}^2$ and average $I'_{\lambda,av}$ of the radiation intensity $I'_\lambda(r, \theta)$ are defined as:

$$\sigma_{I_\lambda}^2 = \frac{\int_{A_{wet}} (I'_\lambda(r, \theta) - I'_{\lambda,av})^2 \cdot dA}{A_{wet}} \quad (58)$$

$$I'_{\lambda,av} = \frac{\int_{A_{wet}} I'_\lambda(r, \theta) \cdot dA}{A_{wet}} \quad (59)$$

Analogous to Equation (50), Equations (58) and (59) can be simplified to, respectively

$$\sigma_{I_\lambda}^2 = \frac{2 \cdot N_l}{\pi \cdot (1 - N_l \cdot \Pi_1^2)} \int_{A'} (I'_\lambda(r, \theta) - I'_{\lambda,av})^2 \cdot r' \cdot dr' \cdot d\theta \quad (60)$$

$$I'_{\lambda,av} = \frac{2 \cdot N_l}{\pi \cdot (1 - N_l \cdot \Pi_1^2)} \int_{A'} I'_\lambda(r, \theta) \cdot r' \cdot dr' \cdot d\theta \quad (61)$$

Because of the above mentioned second principle of PI, the variance $\sigma_{I_\lambda}^2$ and the average $I'_{\lambda,av}$ are calculated and plotted in Figure 13 (right). From this figure, it is observed that the variance $\sigma_{I_\lambda}^2$ has a minimum at the radial distance $r^* = 0.38$ m and the average $I'_{\lambda,av}$ a maximum at the radial distance $r^* = 0.28$ m. Despite the fact that the location of the local minimum of the variance $\sigma_{I_\lambda}^2$ differs from the radial distance r_{opt}^* , the value of the variance $\sigma_{I_\lambda}^2$ at the radial distance $r^* = r_{opt}^*$ does not differ strongly from its minimal value. In addition, the value of the average $I'_{\lambda,av}$ at the radial distance $r^* = r_{opt}^*$ does not differ strongly from its maximal value. For this reason, at the radial distance $r^* = r_{opt}^*$, the average $I'_{\lambda,av}$ is almost maximal (which leads to a higher decomposition of the organic compound M) and the variance $\sigma_{I_\lambda}^2$ is almost minimal. As a result, the explanation of the existence of the radial distance r_{opt}^* , based on the second principle of PI is fairly plausible.

Figure 14 and 15 show some profiles of the concentration c'_M and the radiation intensity I'_λ in the optimized multi-lamp reactor. At the lamp outer wall, the radiation intensity I'_λ is

very large and so is the rate r_M . As a result, the concentration c'_M will be very small at this location in the reactor.

From the above explanation of the existence of the radial distance r_{opt}^* , it follows that
645 when the number of lamps N_l in the reactor increases, a higher uniformity of the radiation intensity $I'_\lambda(r, \theta)$ can be reached inside the reactor (and so a better realization of the second principle of PI). If the number of lamps N_l increases, an increase in this uniformity is possible by increasing the radial distance r^* (and so the radial distance r_{opt}^*).

This reasoning is in accordance with the results in Figures 16 and 17. Notice from Figure
650 17 (right) that from a certain value of the number of lamps N_l (in this case about 16) the increase in the reactor performance when increasing the number of lamps N_l becomes very small. Therefore, using a huge number of lamps in the multi-lamp reactor is economically unfeasible.

If the radiation absorption in the aqueous medium increases (i.e. the absorption
655 coefficient $\alpha_{\lambda,w}$ increases), the radiation intensity I_λ will decline more strongly when moving further away from a lamp. Consequently, the uniformity of the radiation intensity I_λ in the reactor will decrease. This decrease can be compensated by decreasing the radial distance r^* (and so the radial distance r_{opt}^*) which will again lead to an increase in the reactor performance. This reasoning is in accordance with the results in Figures 18 and 19
660 (left). Analogous to the influence of the reaction rate constant k on the optimization results for the single-lamp reactor, it is observed that for large values of the absorption coefficient $\alpha_{\lambda,w}$ the calculation of the minimum of the $c'_{M,av}$ vs. r^* - curves is ill-conditioned (see Figure 18). The explanation of the path of the curve plotted in Figure 19 (right) is the same as in Figure 10 (right).

The influence of the reaction rate constant k on the optimization results is depicted in Figures 20 and 21. Analogous to the influence of the reaction rate constant k on the optimization results for the single-lamp reactor, it is observed that for very small and very large values of k , a flattening of the curves occurs, reflecting the decrease in the influence of the reactor geometry on the reactor performance. On the other hand, a general trend in Figure 21 (left) is observed, namely the radial distance r_{opt}^* increases when the reaction rate constant k increases. An explanation for this trend is the following. When in a given multi-lamp reactor the reaction rate constant k increases, the concentration of the organic compound M will decrease the strongest near the lamps and in the reactor center because the highest values of the radiation intensity I_λ (and so the reaction rate r_M) occur at these locations in the reactor. However, near the reactor wall the radiation intensity I_λ is smaller, resulting in a smaller decrease in the concentration of the organic compound M. Therefore, in order to increase the decomposition of the organic compound M near the reactor wall, an increase in the radial distance r^* (and as a result also the optimal radial distance r_{opt}^*) will enhance the reactor performance.

5.3. Significance of diffusive heat transfer

In the mathematical reactor models, stated in Section 3, diffusive heat transfer was not considered, i.e. isothermal conditions in the photochemical reactors were assumed. This section discusses if this simplification is realistic or not.

Two phenomena which can lead to non-isothermal conditions in the reactor are (i) the reaction heat of the AOP and (ii) the heat released by the UV-lamps.² The significance of

² In general, UV-lamps have an efficiency smaller than one and so do not convert all electrical input energy into radiation energy, but also release some heat.

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the heat effects, due to the reaction heat, can be expressed, using the following dimensionless number:

$$\delta = \frac{\Delta H_r \cdot C_{M,0}}{\rho \cdot c_p \cdot T_0} \quad (62)$$

690 with ρ , c_p and T_0 the mass density, the specific heat capacity of water and the initial reactor temperature, respectively. The parameter δ is a measure for the ratio of the reaction heat and the sensible heat of the reaction medium. However, in general, heat effects arising from the AOP reactions are overshadowed by the heat released from the UV-lamp(s). (Tang, 2004) A dimensionless number, expressing the significance of the
695 heat effects, due to the heat released by the UV-lamp(s) in a (flow-through) photochemical reactor is the following:

$$\gamma = \frac{P_l/Q}{\rho \cdot c_p \cdot T_0} \quad (63)$$

The parameter γ is a measure for the ratio of the heat released by the UV-lamps (in case of a UV-lamp which converts all electrical input energy into heat) and the sensible heat of
700 the reaction medium. Substituting the following numerical values (applied during the optimization of the single-lamp reactor in Section 5.1)

$$P_l = 1000 \text{ W}, \rho = 1000 \text{ kg/m}^3, c_p = 4184 \frac{\text{J}}{\text{kg}} \cdot \text{K}, T_0 = 298,15 \text{ K}, Q = 0,004 \text{ m}^3/\text{s} \quad (64)$$

into Equation (63), yields a value of $2 \cdot 10^{-4}$ for the parameter γ . Even for the more commonly used medium pressure mercury arc lamps with an electric power up to 60 kW
705 (Schalk et al., 2005), the value for the parameter γ remains very small, indicating minor heat effects in the photochemical reactor due to the UV-lamps. As a consequence, isothermal conditions in the photochemical reactor can be assumed.

6. Numerical techniques and computer software

This section briefly describes the numerical techniques and computer software used to optimize the photochemical reactors.

All the calculations have been made in MATLAB.

For the single-lamp reactor, the integral in Equation (44) has been calculated, using the `quad` command which uses recursive adaptive Simpson quadrature. The applied absolute tolerance was 10^{-6} . The concentration $c'_{M,av,min}$ has been calculated using the `fminsearch` command. Here, the underlying algorithm is based on the Nelder-Mead simplex search method of Lagarias et al (1998), a direct search method that does not use numerical or analytic gradients. The applied absolute termination tolerance on the concentration $c'_{M,av,min}$ and the decision variable L_r are both 10^{-4} .

For the multi-lamp reactor, the integral in Equation (50) has been calculated, using the `quad2d` command which, analogously to the `quad` command, applies a quadrature rule. The applied absolute tolerance was 10^{-8} . The decision variable in the optimization problem for the multi-lamp reactor is the radial distance r^* . The concentration $c'_{M,av,min}$ has been calculated using the `fminbnd` command. Here, the underlying algorithm is based on the golden section search and parabolic interpolation which are derivative free optimization methods. The applied absolute termination tolerance on the decision variable r^* equals 10^{-4} .

7. Conclusions

In this paper the modeling and optimization of two simple, homogeneous, non-catalytic, isothermal, photochemical plug flow reactor configurations were treated. The UV-lamps used in these reactors lead to a relatively high energy and operating cost. A minimization of these costs was performed via a model-based optimization of the reactor geometry. First, a simplified reactor model was built taking into account the reaction kinetics, the reactor geometry and the radiation intensity in the reactor.

The first modeled reactor consisted of a cylindrical UV-lamp that is present inside a quartz sleeve, outside of which the water flows between it and the external reactor wall (annular channel reactor). A rigorous, well-posed optimization problem was formulated consisting of the calculation of the optimal reactor geometry (length and radius) in such a way that, for a given reactor volume and a given flow rate of the water to be treated, the mean outlet concentration of an organic pollutant was minimized. By solving this optimization problem, it has been shown that an optimal reactor length existed. If the absorption coefficient of the water or the reaction rate constant increases, the optimal reactor length increases as well.

The second modeled reactor was similar to the first reactor but contained multiple lamps positioned symmetrically in a circular pattern (i.e. multi-lamp reactor). The optimization problem consisted of the calculation of the optimal lamp position inside the reactor in such a way that the mean outlet concentration of an organic pollutant was minimized. The lamp position was given by the distance between the centers of the reactor and a lamp. Again a well-posed optimization problem was obtained and an optimal lamp position existed. If the number of lamps or the reaction rate constant increases or the absorption

coefficient of the water decreases, the optimal distance between the centers of the reactor
755 and the lamp increases.

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Notations

AOP Advanced Oxidation Process

A_{wet} wet surface, i.e. the surface available for the flow of the water in the
770 reactor, [m²]

c speed of light (in water), [m/s]

c'_M dimensionless concentration of the organic compound M, [-]

$c'_{M,0}$ dimensionless reactor inlet concentration of the organic compound M, [-]

$c'_{M,av}$ dimensionless, mean outlet concentration of the organic compound M, [-]

775 $c'_{M,av,min}$ minimal, dimensionless, mean outlet concentration of the organic
compound M, [-]

	$C_{HO\cdot}$	concentration of the hydroxyl radical $HO\cdot$, [mol/m ³]
	C_M	concentration of the organic compound M, [mol/m ³]
	$C_{M,0}$	reactor inlet concentration of the organic compound M, [mol/m ³]
780	$C_{M,av}$	mean outlet concentration of the organic compound M, [mol/m ³]
	C_{Ox}	primary oxidant concentration, [mol/m ³]
	C_{Si}	concentration of the interfering compounds S_i , [mol/m ³]
	D	radiation dose, [J/m ²] D_{av} average value of the radiation dose D, [J/m ²]
	Da	Damköhler number, [-]
785	$e_{\lambda,Ox}$	local, volumetric energy rate of photons absorbed by the primary oxidant Ox (at a certain wavelength λ), [W/m ³]
	$e'_{\lambda,Ox}$	local, volumetric number rate of photons absorbed by the primary oxidant Ox (at a certain wavelength λ), [mol/m ³ .s]
	E_0	standard reduction potential, [V]
790	h	Planck's constant, [J.s]
	$HO\cdot$	hydroxyl radical
	I_λ	radiation intensity, [W/m ²]
	$I_{\lambda,j}$	radiation intensity in a certain point in the multi-lamp reactor as a result of the radiation emitted by the j^{th} lamp, [W/m ²]
795	I'_λ	dimensionless radiation intensity, [-]
	$I'_{\lambda,av}$	average value of the radiation intensity I'_λ , [-]
	$I_{\lambda,b}$	radiation intensity evaluated at the lamp boundary in the single-lamp reactor, [W/m ²]
	k	reaction rate constant, [m ² /J]

800	$k_{HO\cdot, M}$	reaction rate constant of the reaction between the organic compound M and the hydroxyl radical $HO\cdot$, [m^2/J]
	$k_{HO\cdot, Si}$	reaction rate constant of the reaction between the interfering compounds S_i and the hydroxyl radical $HO\cdot$, [m^2/J]
	L_l	UV-lamp length, [m]
805	L_r	reactor length, [m]
	$L_{r,opt}$	optimal length of the single-lamp reactor, [m]
	LSP	line source with parallel plane emission
	M	organic pollutant, broken down by the AOP
	N_A	Avogadro's constant
810	n_i	refractive index of medium i, [-]
	N_l	number of lamps in the multi-lamp reactor, [-]
	n_{Ox}	number of primary oxidant molecules, [-]
	Ox	primary oxidant of the AOPP ₁ effective, monochromatic radiation power of the UV-lamp in the single-lamp reactor, [W]
815	P_{el}	electrical power of a UV-lamp [W]
	$P_{l,j}$	effective, monochromatic radiation power of the j^{th} UV-lamp in the multi-lamp reactor, [W]
	$P_{l,tot}$	total, effective, monochromatic radiation power of all the UV-lamps in the multi-lamp reactor, [W]
820	PSSA	pseudo steady state approximation
	Q	volumetric flow rate of the water in the reactor, [m^3/s]

	r'	dimensionless, radial distance between a point in the reactor and the reactor center line, [m]
825	r	radial distance between a point in the reactor and the reactor center line, [m]
	r^*	radial distance between the center of a lamp in the multi-lamp reactor and the center of the multi-lamp reactor, [m]
	$r^{*'} $	dimensionless, radial distance between the center of a lamp and the center of the multi-lamp reactor, [-]
830	r_{max}^*	maximal value of the radial distance r^* , [m]
	r_{min}^*	minimal value of the radial distance r^* , [m]
	r_{opt}^*	optimal, radial distance between the center of a lamp in the multi-lamp reactor and the center of the multi-lamp reactor, [m]
	r_j^*	radial coordinate of the center of the j^{th} lamp in the multi-lamp reactor, [m]
835	R_{aq}	reflectivity of the air/quartz-interface, [-]
	R_i	outer radius of a UV-lamp, equals the sum of the thickness t and the radius s , [m]
	r_j	radial distance between a point in the multi-lamp reactor and the center of the j^{th} lamp, [m]
840	r_M	reaction rate of the organic compound M with the hydroxyl radical $\text{HO}^\cdot$, [mol/m ³ .s]
	R_{qw}	reflectivity of the quartz/water-interface, [-]
	R_r	inner radius of the reactor, [m]
	RTE	radiative transfer equation

845	R_{wr}	reflectivity of the water/reactor wall-interface, [-]
	s	quartz sleeve inner radius of a UV-lamp, [m]
	S_i	interfering compounds
	t	quartz sleeve thickness of a UV-lamp, [m]
	t	time, [s]
850	T_q	transmittivity of the quartz sleeve, [-]
	t_{rx}	characteristic reaction time, [s]
	T_w	transmittivity of the water, [-]
	u	plug flow speed of the water in the reactor, [m/s]
	UV	ultraviolet
855	V	volume, [m ³]
	V_r	volume of the reaction medium in the reactor, [m ³]
	z	axial distance between a point in the reactor and the reactor inlet, [m]
	z'	dimensionless, axial distance between a point in the reactor and the reactor inlet, [m]
860	$\alpha_{\lambda, Ox}$	volumetric absorption coefficient of the primary oxidant Ox (at a certain wavelength λ), [m ⁻¹]
	$\alpha_{\lambda, q}$	volumetric absorption coefficient of quartz (at a certain wavelength λ), [m ⁻¹]
	$\alpha_{\lambda, w}$	volumetric absorption coefficient of the aqueous medium (at a certain wavelength λ), [m ⁻¹]
865		

	γ	dimensionless number, expressing the ratio of the heat released by the UV-lamps (in case of zero-efficiency) and the sensible heat of the reaction medium
	δ	dimensionless number, expressing the ratio of the reaction heat and the sensible heat of the reaction medium, [-]
870	$\epsilon_{\lambda, \text{Ox}}$	molar absorption coefficient of the primary oxidant Ox, [m ² /mol]
	θ	angular coordinate, [rad]
	θ_j^*	angular coordinate of the center of the j^{th} lamp in the multi-lamp reactor, [rad]
875	λ	wavelength, [m]
	ν	photon wave frequency, [s ⁻¹]
	ν_{rx}	stoichiometric coefficient of a reaction, [-]
	Π_1	dimensionless parameter, which is a measure for the narrowness of the annulus, [-]
880	Π_2	dimensionless parameter, which is a measure for the radiation absorption by the aqueous medium in the reactor, [-]
	Π_5	dimensionless parameter, which is a measure for the narrowness of the reactor, [-]
	$\sigma_{I'_\lambda}^2$	variance of the radiation intensity I'_λ , [-]
885	σ_D^2	variance of the radiation dose D, [J ² /m ⁴]
	τ	residence time in the reactor, [s]
	ϕ_p^{abs}	number of photons (with a certain wavelength λ) absorbed by the primary oxidant Ox during an infinitesimal period, [s ⁻¹]

Φ_{λ}^{Ox} quantum yield of the primary oxidant Ox at the wavelength λ , [-]

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Table 1: Classification of the main radiation models (Alfano et al., 1986).

Type	Characteristics	Name	Abbreviation	References
Incidence model	2D	Radial	RI	Matsuura, 1970
		Partially diffuse	PDI	Matsuura, 1970
		Diffuse	DI	Matsuura, 1970; Harada et al., 1971; Märkl and Vortmeyer, 1971
	3D	Diffuse	TDI	Zolner and Williams, 1971; Williams, 1978
Emission model	Line source	Parallel Plane Emission	LSPP	Placid and Nascimento, 2007
		Spherical Emission	LSSE	Jacob and Dranoff, 1970; Blatchley, 1997; Bolton, 2000; Tsekov and Smirniotis, 1997
		Diffuse Emission	LSDE	Akehata and Shirai, 1972; Yokota and Suzuki, 1995; Yang et al., 2005a, 2005b
	Extense source	Volumetric Emission	ESVE	Irazoqui, 1973; Irazoqui et al., 1976, 2000
		Superficial Spherical Emission	ESSE	Foraboschi, 1959; Stramigioli et al., 1977
		Superficial Diffuse Emission	ESDE	Yokota, 1976

Table 2: Summary of the reactions between hydrogen peroxide (H_2O_2), hydroxyl radical ($HO\cdot$) and the subsequent species formed in the H_2O_2 /UV-process (Mota et al., 2008).

Chemical Reaction	
$H_2O_2 \xrightarrow{h\nu} 2 HO$	(A.1)
$H_2O_2 + HO\cdot \rightarrow HO_2\cdot + H_2O$	(A.2)
$H_2O_2 + HO_2\cdot \rightarrow HO\cdot + H_2O + O_2$	(A.3)
$2 HO\cdot \rightarrow H_2O_2$	(A.4)
$2 HO_2\cdot \rightarrow H_2O_2 + O_2$	(A.5)
$HO\cdot + HO_2\cdot \rightarrow H_2O + O_2$	(A.6)

Figure 1: Characteristics of the two-dimensional incidence models. (left) Radial, (middle) partially diffuse and (right) diffuse (Alfano et al., 1986).

Figure 2: Radiation emission characteristics of the lamps in the line source models. (left) parallel plane, (middle) diffuse and (right) spherical (Alfano et al., 1986).

Figure 3: 3D-view (left) and cross section (right) of the annular, cylindrical, photochemical, single-lamp reactor. In this case, s , t , R_i and R_r are the quartz sleeve inner radius, thickness and outer radius, and the reactor (inner) radius, respectively. The radius R_i equals the sum of the thickness t and the radius s (Bolton, 2000).

Figure 4: 3D-view (left) and cross section (right) of an annular, cylindrical, photochemical, multi-lamp reactor containing five lamps, positioned in a circular pattern. In this case points A and B are a random point in the reactor and the point at the center of a lamp, respectively. The location of A is determined by its polar coordinates (r, θ) . r_j and R_i are the distance between A and the center of the j^{th} lamp and the outer radius of the quartz sleeve of a lamp, respectively. The latter equals the sum of the thickness t and the radius s .

Figure 5: General reaction mechanism of an AOP.

Figure 6: Simplified scheme of the reaction mechanism between the hydroxyl radical $\text{HO}^\bullet$, the organic substrate molecule M and the interfering compounds (or scavenger molecules) S_i . The square brackets $[\]$ denote a molar concentration (Oppenländer, 2003).

Figure 7: Depiction of the wet surface A_{wet} and the surface A' used to calculate the concentration $C_{M,av}$ in the multi-lamp reactor.

Figure 8: The reactor outlet concentration $c'_{M,av}$ as a function the length L_r in the single-lamp photochemical reactor with $V_r = 0.4 \text{ m}^3$, $Q = 0.004 \text{ m}^3/\text{s}$, $k = 10^{-4} \text{ m}^2/\text{J}$, $R_i = 0.1 \text{ m}$, $P_l = 1000 \text{ W}$ and $\alpha_{z,w} = 20 \text{ m}^{-1}$.

Figure 9: The reactor outlet concentration $c'_{M,av}$ as a function the length L_r in the single-lamp photochemical reactor for different values of the absorption coefficient $\alpha_{\lambda,w}$ (in m^{-1}). The values of the remaining parameters are the same as in Figure 8.

Figure 10: The length $L_{r,opt}$ (left) and the reactor outlet concentration $c'_{M,av,min}$ (right) as a function of the absorption coefficient $\alpha_{\lambda,w}$ for the single-lamp reactor. The values of the remaining parameters are the same as in Figure 8.

Figure 11: The reactor outlet concentration $c'_{M,av}$ as a function the length L_r in the single-lamp photochemical reactor for different values of the reaction rate constant k (in m^2/J). The values of the remaining parameters are the same as in Figure 8.

Figure 12: The length $L_{r,opt}$ (left) and the reactor outlet concentration $c'_{M,av,min}$ (right) as a function of the reaction rate constant k for the single-lamp reactor. The values of the remaining parameters are the same as in Figure 8.

Figure 13: The reactor outlet concentration $c'_{M,av}$ (left) and the average radiation intensity $I'_{\lambda,av}$ and the variance $\sigma^2_{I'_{\lambda}}$ (right) as a function of the radial distance r^* in the multi-lamp photochemical reactor with $L_r = 6$ m, $R_r = 0.5$ m, $R_i = 0.03$ m, $N_l = 10$, $u = 0.06$ m/s, $k = 10^{-4}$ m²/J, $P_{l,tot} = 10\,000$ W and $\alpha_{\lambda,w} = 20$ m⁻¹.

Figure 14: The concentration c'_M in the multi-lamp photochemical reactor as a function of the radial distance r' and the axial distance z' for a certain value of the angle θ such that the cross section cuts a lamp in half. The values of the parameters are the same as in Figure 13 except the value of the radial distance r^* which now equals its optimal value r^*_{opt} .

Figure 15: The concentration $c'_M(r', \theta, z'=1)$ (left) and the radiation intensity $I'_\lambda(r', \theta, z'=1)$ (right) in the multi-lamp photochemical reactor. The values of the parameters are the same as in Figure 13 except the value of the radial distance r^* which now equals its optimal value r^*_{opt} .

Figure 16: The reactor outlet concentration $c'_{M,av}$ as a function the radial distance r^* in the multi-lamp photochemical reactor for different values of the number of lamps N_l . The values of the remaining parameters are the same as in Figure 13.

Figure 17: The radial distance r_{opt}^* (left) and the reactor outlet concentration $c'_{M,av,min}$ (right) in the multi-lamp photochemical reactor as a function of the number of lamps N_l . The values of the remaining parameters are the same as in Figure 13. The radial distances r_{min}^* and r_{max}^* are calculated by means of Equations (52) and (53), respectively.

Figure 18: The reactor outlet concentration $c'_{M,av}$ as a function the radial distance r^* in the multi-lamp photochemical reactor for different values of the absorption coefficient $\alpha_{\lambda,w}$ (in m^{-1}). The values of the remaining parameters are the same as in Figure 13.

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Figure 20: The reactor outlet concentration $c'_{M,av}$ as a function the radial distance r^* in the multi-lamp photochemical reactor for different values of the reaction rate constant k (in m^2/J). The values of the remaining parameters are the same as in Figure 13.

Figure 21: The radial distance r_{opt}^* (left) and the reactor outlet concentration $c'_{M,av,min}$ (right) in the multi-lamp photochemical reactor as a function of the reaction rate constant k . The values of the remaining parameters are the same as in Figure 13.

List of figure captions

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Figure 11: The reactor outlet concentration $c'_{M,av}$ as a function the length L_r in the single-lamp photochemical reactor for different values of the reaction rate constant k (in m^2/J). The values of the remaining parameters are the same as in Figure 8.

Figure 12: The length $L_{r,opt}$ (left) and the reactor outlet concentration $c'_{M,av,min}$ (right) as a function of the reaction rate constant k for the single-lamp reactor. The values of the remaining parameters are the same as in Figure 8.

Figure 13: The reactor outlet concentration $c'_{M,av}$ (left) and the average radiation intensity $I'_{\lambda,av}$ and the variance $\sigma_{I'_{\lambda}}^2$ (right) as a function of the radial distance r^* in the multi-lamp photochemical reactor with $L_r = 6 \text{ m}$, $R_r = 0.5 \text{ m}$, $R_i = 0.03 \text{ m}$, $N_l = 10$, $u = 0.06 \text{ m/s}$, $k = 10^{-4} \text{ m}^2/\text{J}$, $P_{l,tot} = 10\,000 \text{ W}$ and $\alpha_{\lambda,w} = 20 \text{ m}^{-1}$.

Figure 14: The concentration c'_M in the multi-lamp photochemical reactor as a function the radial distance r' and the axial distance z' for a certain value of the angle θ such that the cross section cuts a

lamp in half. The values of the parameters are the same as in Figure 13 except the value of the radial distance r^* which now equals its optimal value r_{opt}^* .

Figure 15: The concentration $c'_M(r', \theta, z'=1)$ (left) and the radiation intensity $I'_\lambda(r', \theta, z'=1)$ (right) in the multi-lamp photochemical reactor. The values of the parameters are the same as in Figure 13 except the value of the radial distance r^* which now equals its optimal value r_{opt}^* .

Figure 16: The reactor outlet concentration $c'_{M,av}$ as a function the radial distance r^* in the multi-lamp photochemical reactor for different values of the number of lamps N_L . The values of the remaining parameters are the same as in Figure 13.

Figure 17: The radial distance r_{opt}^* (left) and the reactor outlet concentration $c'_{M,av,min}$ (right) in the multi-lamp photochemical reactor as a function of the number of lamps N_L . The values of the remaining parameters are the same as in Figure 13. The radial distances r_{min}^* and r_{max}^* are calculated by means of Equations (52) and (53), respectively.

Figure 18: The reactor outlet concentration $c'_{M,av}$ as a function the radial distance r^* in the multi-lamp photochemical reactor for different values of the absorption coefficient $\alpha_{\lambda,w}$ (in m^{-1}). The values of the remaining parameters are the same as in Figure 13.

Figure 19: The radial distance r_{opt}^* (left) and the reactor outlet concentration $c'_{M,av,min}$ (right) in the multi-lamp photochemical reactor as a function of the absorption coefficient $\alpha_{\lambda,w}$. The values of the remaining parameters are the same as in Figure 13.

Figure 20: The reactor outlet concentration $c'_{M,av}$ as a function the radial distance r^* in the multi-lamp photochemical reactor for different values of the reaction rate constant k (in m^2/J). The values of the remaining parameters are the same as in Figure 13.

Figure 21: The radial distance r_{opt}^* (left) and the reactor outlet concentration $c'_{M,av,min}$ (right) in the multi-lamp photochemical reactor as a function of the reaction rate constant k . The values of the remaining parameters are the same as in Figure 13.