

A TRANSFER OPERATOR BASED COMPUTATIONAL STUDY OF REACTING FLUIDS

Motivation

In many industrial applications one aims for efficient chemical reactions. We use a toy model to mimic reactions of chemical fluids in a stirred tank reactor (STR) and investigate the dynamics under the influence of complex flow structures.

Transfer Operator Method

We analyze transport and reactions of ideal fluid particles. We assume that they move passively according to $\dot{x}(t) = u(x(t), t)$, with tracer trajectories $x(t) \in \mathbb{R}^2$. Let $S: M \rightarrow M$ be the flow map that maps a particle $x_0 = x(t_0)$ to its new position $S(x_0) = x(t_0 + \tau)$ over the time span $[t_0, t_0 + \tau]$.

In practice, we represent fluids by density vectors, which are evolved by means of a numerical transfer operator and are subsequently updated according to an underlying chemical reaction scheme. We model a STR as a compact domain $M \subset \mathbb{R}^2$ discretized into n disjoint, connected sets (boxes) B_i , $i = 1, \dots, n$ [1]. Each box B_i contains N uniformly distributed sample points of chemical substances $x_{0,k}^i$, $k = 1, \dots, N$. We use Ulam's method [2] for a finite-rank approximation of an infinite-dimensional Perron-Frobenius operator as a matrix $P \in \mathbb{R}^{n,n}$ as follows:

$$P_{ij} = \frac{\#\{k: S(x_{0,k}^i) \in B_j\}}{N},$$

i.e. the entry P_{ij} is estimated as the proportion of the fluid particles that gets mapped from box B_i to box B_j under the action of S . We can interpret the row-stochastic matrix P as the transition matrix of a Markov chain on a finite state space, where the boxes B_i represent the states. A fluid density vector $C_{A_t} \in \mathbb{R}^n$ representing a chemical substance A at time t evolves according to $C_{A_{t+\tau}} = C_{A_t} P$.

To simulate the diffusion process into neighboring boxes, we construct the diffusion matrix D by choosing K test points $x_{i,k} \in B_i$, $k = 1, \dots, K$, in each box B_i and representing the diffusion over an ϵ -ball centered at $x_{i,k}$. Therefore, we choose another grid of points $y_{i,k,k'}$, $k' = 1, \dots, K'$ uniformly spread over $B_\epsilon(x_{i,k})$ to estimate the diffusion matrix [3]

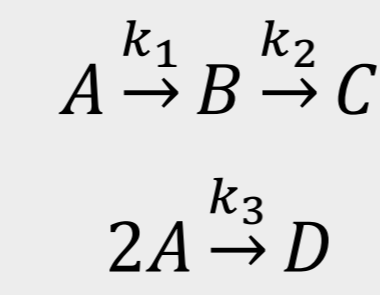
$$D_{ij} = \frac{\#\{y_{i,k,k'} \in B_j\}}{K \cdot K'}$$

and obtain the diffused transition matrix

$$P_D = P \cdot D.$$

Competitive Consecutive Reaction

We simulate Van de Vusse's competitive consecutive reaction [4] in an open DG reactor model:



The constant feed concentration C_{A0} of pure A is continuously fed into the STR (volume V per box, flow rate v_0) and the concentrations C_i are pumped out of the STR (red: input/output dynamics, see fig. 1). In each box B_i , the particles react under the following conditions:

$$\dot{C}_A = (C_{A0} - C_A) \cdot \frac{v_0}{V} - k_1 C_A - k_3 C_A^2$$

$$\dot{C}_B = -C_B \cdot \frac{v_0}{V} + k_1 C_A - k_2 C_B$$

$$\dot{C}_C = -C_C \cdot \frac{v_0}{V} + k_2 C_B$$

$$\dot{C}_D = -C_D \cdot \frac{v_0}{V} + \frac{1}{2} k_3 C_A^2,$$

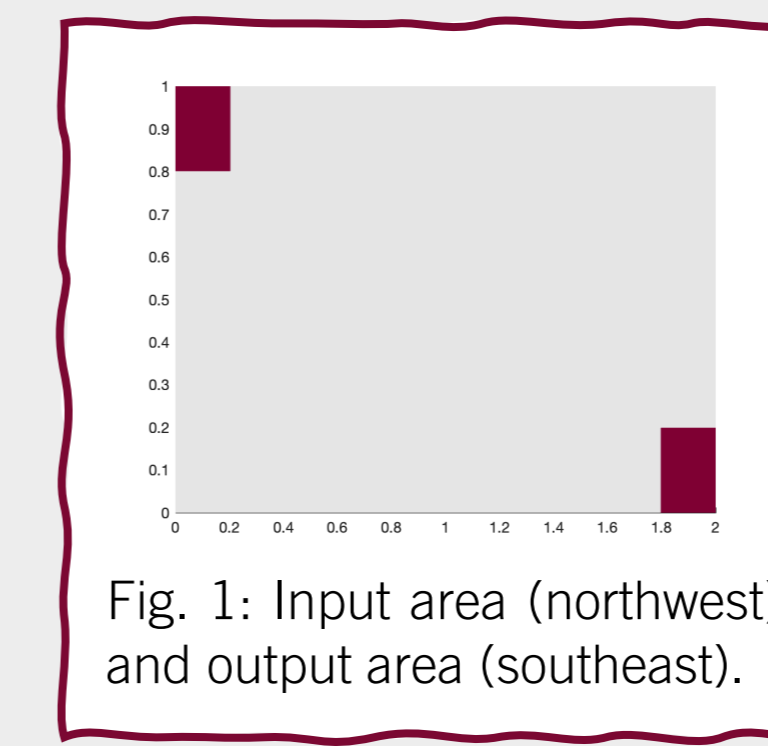


Fig. 1: Input area (northwest) and output area (southeast).

where $\pm C_i$ red is the input/output on the input/output area.

We consider a STR with $V = 1l$, $C_{A0} = 10 \text{ mol/l}$ and $v_0 = 10 \text{ l/min}$. For each time step $\tau = 0.01$, the particles are transported and then react (here $k_1 = \frac{5}{6}$, $k_2 = \frac{5}{3}$ and $k_3 = \frac{1}{6}$). The reaction is simulated using the explicit Euler method.

Double Gyre Mixer (DG)

The DG mixes fluids over the domain $[0,2] \times [0,1]$ with the velocity field u [5] defined by

$$\dot{x} = -0.5\pi \sin(\pi f(x, t)) \cos(\pi y)$$

$$\dot{y} = 0.5\pi \cos(\pi f(x, t)) \sin(\pi y) \frac{df}{dx}(x, t),$$

where $f(x, t) = \delta \sin(2\pi t)x^2 + (1 - 2 \cdot \delta \sin(2\pi t))x$.

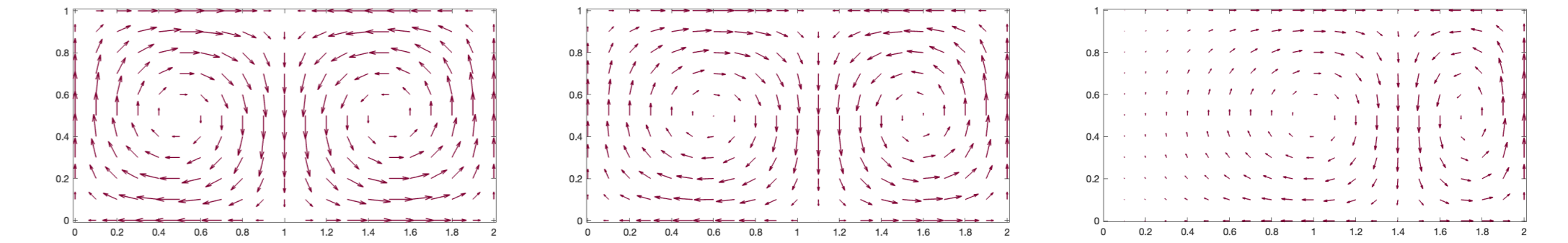


Fig. 2: $t = 0$, $t = 0.5$ or $t = 1$ (left) and $t = 0.25$ for $\delta = 0.1$ (middle) and $\delta = 0.5$ (right).

Results for $\delta = 0.1$

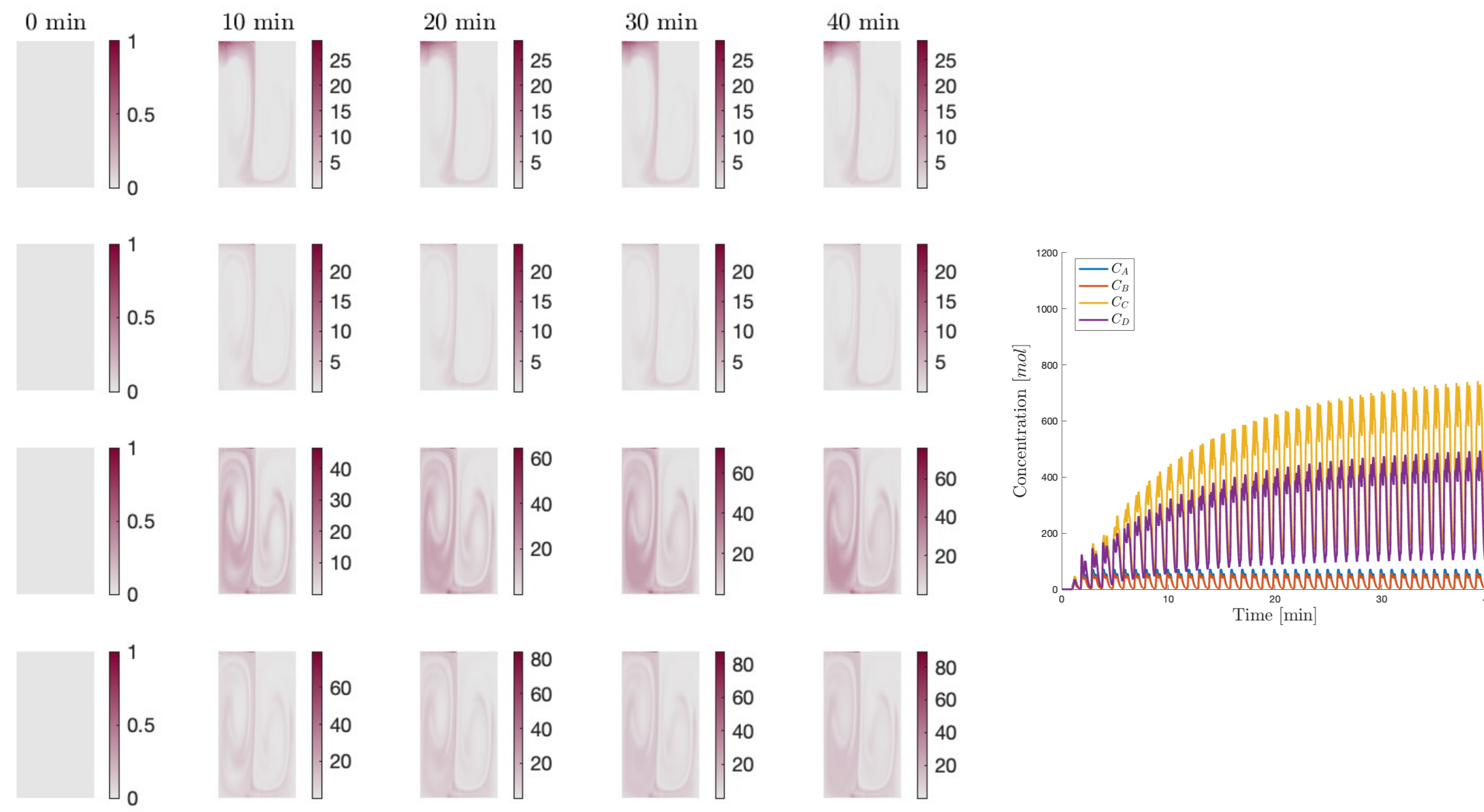


Fig. 3: $C_A, C_B, C_C, C_D [\text{mol}]$ (row 1 to 4, left) and output concentrations (right) without diffusion.

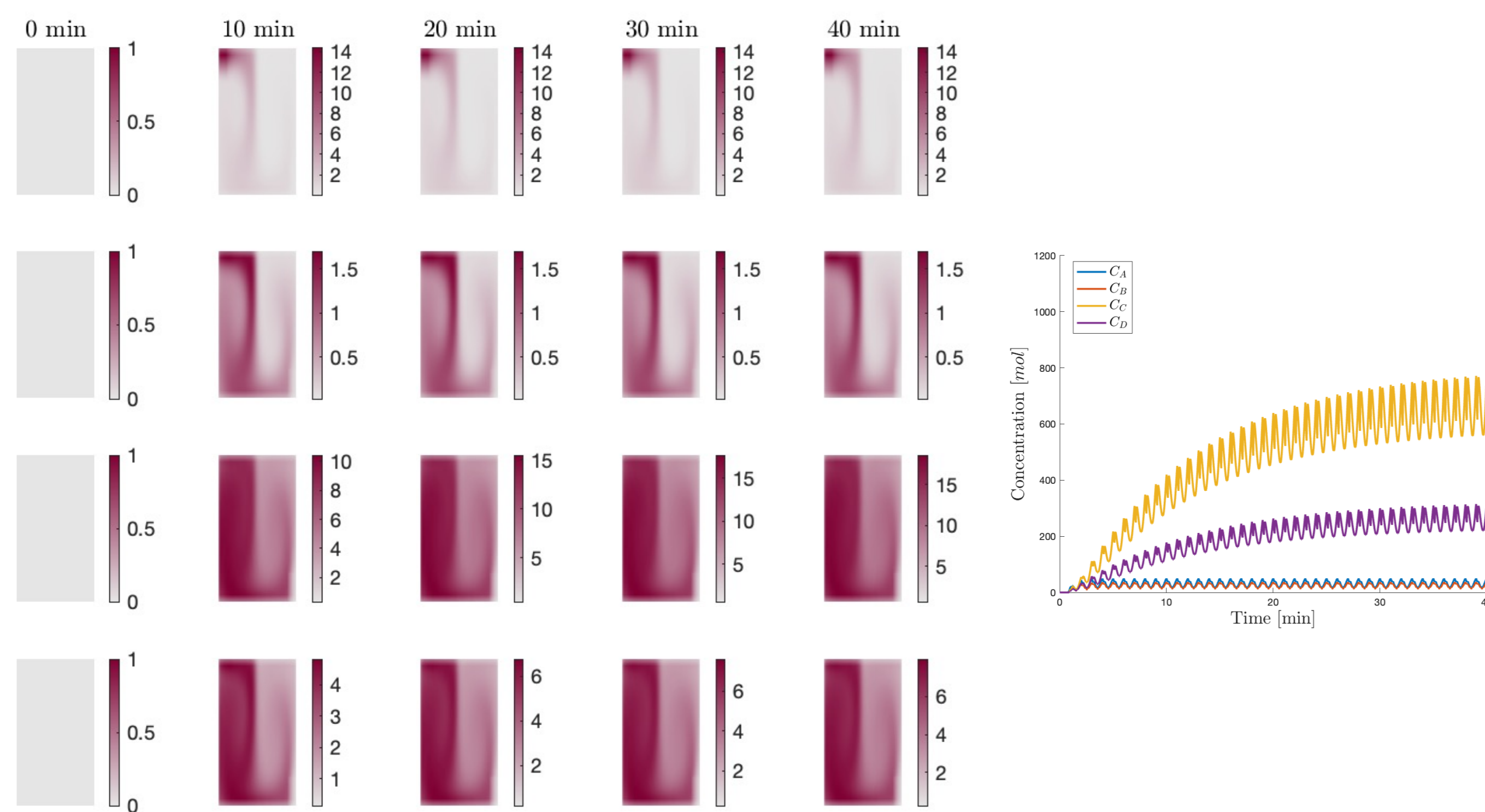


Fig. 4: $C_A, C_B, C_C, C_D [\text{mol}]$ (row 1 to 4, left) and output concentrations (right) with diffusion.

Results for $\delta = 0.5$

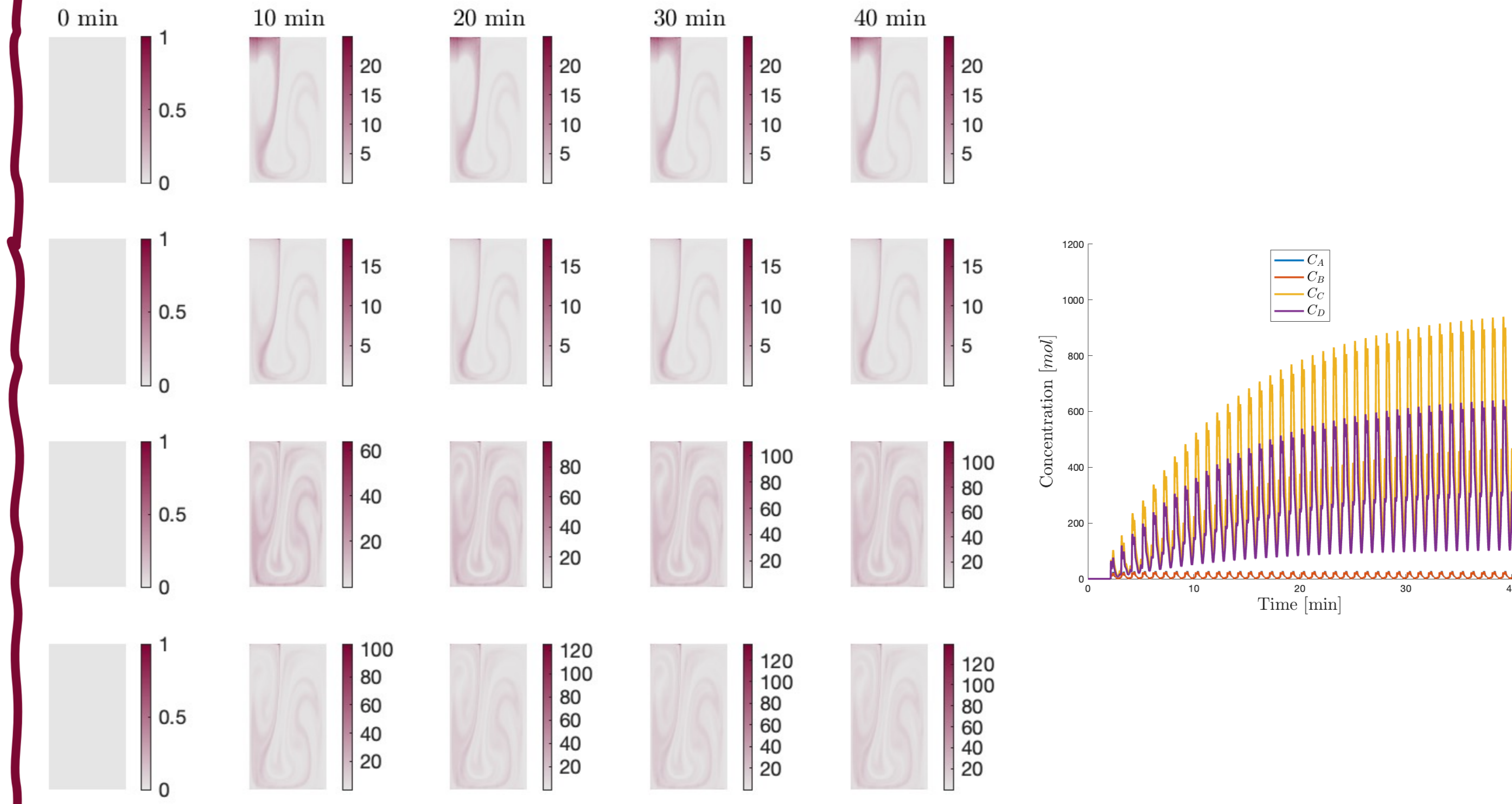


Fig. 5: $C_A, C_B, C_C, C_D [\text{mol}]$ (row 1 to 4, left) and output concentrations (right) without diffusion.

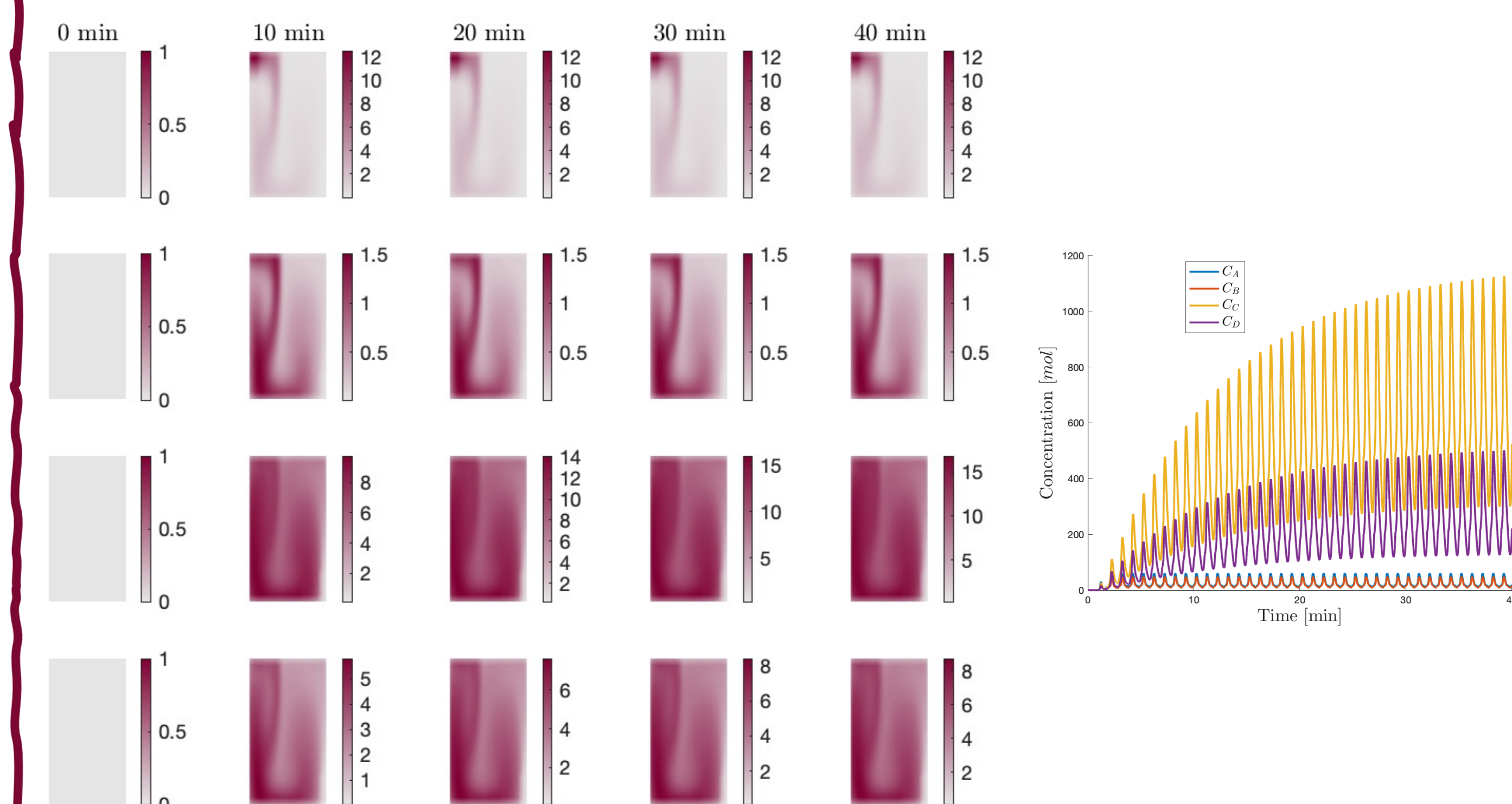


Fig. 6: $C_A, C_B, C_C, C_D [\text{mol}]$ (row 1 to 4, left) and output concentrations (right) with diffusion.

Outlook

In the future, we will compare experimental data with the transfer operator based results and visualize the distribution of particles and their concentrations as networks.

[1] Dellnitz, M., Junge, O. On the Approximation of Complicated Dynamical Behavior. *SIAM Journal on Numerical Analysis* 36.2 (1999), 491–515.

[2] Ulam, S. (1964). *Problems in Modern Mathematics*. Interscience.

[3] Froyland, G., Padberg-Gehle, K. (2014). Almost-Invariant and Finite-Time Coherent Sets: Directionality, Duration, and Diffusion. In: Bahsoun, W., Bose, C., Froyland, G. (eds) *Ergodic Theory, Open Dynamics, and Coherent Structures*. Springer Proceedings in Mathematics & Statistics, vol 70. Springer, New York, NY, 171–216.

[4] Van de Vusse, J. G. Plug-flow type reactor versus tank reactor. *Chemical Engineering Science* 19.12 (1964), 994–996.

[5] Shadden, S. C., Lekien, F., Marsden, J. E. Definition and properties of Lagrangian coherent structures from finite-time Lyapunov exponents in two-dimensional aperiodic flows. *Physica D: Nonlinear Phenomena* 212.3 (2005), 271–304.