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NUMERICAL SOLUTION OF CONTAMINANT TRANSPORT PROBLEMS WITH NON-EQUILIBRIUM ADSORPTION IN 2D

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Abstract. In this paper, an efficient operator splitting scheme for solving 2D convection-diffusion problems with adsorption is introduced. Particularly, we consider a practical problem of soil parameters identification using dual-well tests. We use a general mathematical model including contaminant transport, mechanical dispersion and molecular diffusion and adsorption in both equilibrium and non-equilibrium modes. First, the original half-plane domain is transformed to a rectangle using a bipolar orthogonal transformation. Then in each time step we solve separately the transport, dispersion and adsorption parts. Due to the transformation, the transport problem (linear or non-linear) is reduced to 1D and can be solved in an analytical form. The dispersion part is solved using standard finite volume method. For the system of ODE's representing adsorption we derive an implicit scheme.

Key words. convection-diffusion problem, non-equilibrium adsorption, operator splitting

AMS subject classifications. 76S05, 65M99, 65-06

1. Introduction. Groundwater contamination is one of the most typical hydrogeological and environmental problems. The general model of a groundwater layer includes various hydrogeological processes as contaminant transport, mechanical dispersion, molecular diffusion, sorption, chemical reactions etc. In many practical situations we need to predict the time behaviour of a contaminated groundwater layer. In order to obtain realistic results, it's necessary to have realistic data in the mathematical model. Various soil parameters (porosity, dispersivities, sorption coefficients etc.) can be precisely determined using systems of monitor wells, where by monitoring the contaminant concentration in the wells it's possible to reconstruct the groundwater layer properties. Solving of such inverse problems requires repeated solving of direct problems and therefore it's crucial to have an efficient numerical method for the direct problem.

In this paper we consider a system of two monitor wells and a mathematical model including contaminant transport, dispersion and adsorption. If we use Dupuit-Forchheimer approximation, we have a 2D convection-diffusion-adsorption problem. In e.g. [3], the authors describe an efficient method for solving problems with adsorption in equilibrium mode. The main goal of this paper is to introduce a method also for non-equilibrium sorption problems, based on a similar idea.

2. Mathematical model. Let us consider two wells (injection and extraction well) situated at points $(-d, 0)$ and $(d, 0)$ in Cartesian coordinates, with given radii r_1, r_2. Moreover, let us assume that the pumping rate of the injection well is equal to the discharge in the extraction well. The contaminant transport problem with dispersion and adsorption (reaction) is represented by the following system of differential equations

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$$
\begin{aligned}
h_{eff}\partial_t(C+\Psi_e(C)) - \mathrm{div}(Dh_{eff}\nabla C) - \mathrm{div}(h_{eff}\vec{v}C) + h_{eff}\partial_t S = 0\\
\partial_t S = K(\Psi_n(C) - S)
\end{aligned}
$$

where $((x,y),t) \in \Omega\times < 0,T >$, $C(x,y,t)$ represents the contaminant concentration and $S(x,y,t)$ is the concentration of adsorbed pollution. We consider the adsorption in both equilibrium mode (represented by sorption isotherm $\Psi_e(C)$) and non-equilibrium mode (represented by $\Psi_n(C)$). Both isotherms are considered to be of Freundlich type, $\Psi(C) = AC^p$, $A > 0$, $0 < p < 1$. Here D is the dispersivity tensor

$$D_{ij} = \left\{(D_0 + \alpha_T|v|)\delta_{ij} + \frac{v_i v_j}{|v|}(\alpha_L - \alpha_T)\right\}$$

where D_0 is the molecular diffusion coefficient and δ_{ij} the Kronecker symbol. α_L and α_T are longitudinal and transversal dispersivities. $\vec{v}$ is defined by

$$\vec{v} = -\frac{1}{h_{eff}\theta_0}\nabla\Phi$$

where θ_0 is porosity, Φ is the flow potential and h_{eff} is the groundwater acquifer height (if we consider saturated layer) or piezometric head (for unconfined zone).

As the original two-dimensional domain Ω is symmetric along x-axis, we can restrict ourselves only to one of its half-planes. Using a bipolar transformation, this domain can be transformed to a rectangle $\Omega_R = (0,\pi)\times(v^{(1)},v^{(2)})$ (sides $u=0$, $u=\pi$ corresponding to well borders), where the equipotential curves and streamlines of the flow are parallel to coordinate axes and orthogonal to each other (see [1]). The bipolar coordinates (u,v) are defined by

$$(2.1)\quad x = \frac{\delta}{2}\frac{\sinh v}{\cosh v - \cos u}, \quad y = \frac{\delta}{2}\frac{\sin u}{\cosh v - \cos u}, \quad \sqrt{r_1^2 + \frac{1}{4}\delta^2} + \sqrt{r_2^2 + \frac{1}{4}\delta^2} = 2d$$

The values $v^{(1)}$, $v^{(2)}$ are obtained from

$$(2.2)\qquad \sinh v^{(1)} = -\frac{\delta}{2r_1}, \qquad \sinh v^{(2)} = \frac{\delta}{2r_2}$$

Applying the transformation described above to the problem in (x,y) coordinates, we obtain the following convection-diffusion-adsorption problem in (u,v) coordinates

$$
\begin{aligned}
&(2.3)\quad \partial_t(C+\Psi_e(C)) - F\partial_v C - g(\partial_u(a\partial_u C) + \partial_v(b\partial_v C)) + \partial_t S = 0\\
&(2.4)\quad \partial_t S = K(\Psi_n(C) - S)
\end{aligned}
$$

where $(t,(u,v)) \in < 0,T > \times\Omega_R$. Terms g, a, b and F are known functions depending on u and v and the soil parameters (α_L, α_T, D_0, θ_0 (see [1])). We consider the boundary conditions

$$(2.5)\qquad C = C_0(t) \text{ on } \Gamma_1, \quad \partial_u C = 0 \text{ on } \Gamma_2\cup\Gamma_4, \quad \partial_v C = 0 \text{ on } \Gamma_3$$

where $\Gamma_1 = (0,\pi)\times\{v = v^{(2)}\}$, $\Gamma_2 = \{0\}\times(v^{(1)},v^{(2)})$, $\Gamma_3 = (0,\pi)\times\{v = v^{(1)}\}$, $\Gamma_4 = \{\pi\}\times(v^{(1)},v^{(2)})$.

The initial conditions are

$$(2.6)\qquad C((u,v),0) = 0, \quad S((u,v),0) = 0$$

3. The operator splitting scheme.

3.1. Equilibrium mode problem. In [3], the authors describe a method for 1D problem of the form

$$\partial_t(C+\Psi(C))+v(x)\partial_x C-\partial_x(D(x,t)\partial_x C)=0$$

for $(x,t)\in(0,L)\times(0,T)$ with boundary and initial conditions

$$C(0,t)=C^0(t),\qquad \partial_x C(L,t)=0,\qquad C(x,0)=C_0(x)$$

Here, $\Psi(C)$ represents adsorption in equilibrium mode.

According to [3], the problem can be solved using operator splitting approach. We use time discretization $t_0=0$, t_1, $t_2\ldots t_{n-1}$, $t_n=T$, and then in each time step we separately solve two parts of the problem corresponding to transport and diffusion, i.e. first

$$\partial_t(\phi+\Psi(\phi))+v(x)\partial_x\phi=0,\qquad t\in(t_{j-1},t_j)$$

with the above inflow boundary condition and with initial condition $\phi(x,t_{j-1})=C_{j-1}$. We denote the solution of this problem by $C_j^{1/2}$. Then we continue with the next step and solve

$$\partial_t(\phi+\Psi(\phi))-\partial_x(D(x,t)\partial_x\phi)=0,\qquad t\in(t_{j-1},t_j)$$

with initial condition $\phi(x,t_{j-1})=C_j^{1/2}$. Finally, the solution of this problem we denote by C_j $(\approx C(x,t_j))$.

3.2. Non-equilibrium mode problem. The idea mentioned in previous section can be extended to problem described in sec. 2, which was done in [4] (for 1D case). Again, we use time discretization $t_0=0$, t_1, $t_2\ldots t_{n-1}$, $t_n=T$, and then in each time step we solve three parts of the problem: transport, dispersion and adsorption part.

The transport part presents a hyperbolic problem of the form

$$\partial_t(\phi+\Psi_e(\phi))-F\partial_v\phi=0,\qquad t\in(t_{j-1},t_j),\ j=1\ldots m \tag{3.1}$$

with boundary conditions of the form (2.5) and initial condition $\phi(u,v,t_{j-1})=C_{j-1}$. By $C_j^{1/3}$ we denote the obtained solution, i.e. $C_j^{1/3}:=\phi(u,v,t_j)$. Now we solve the problem (the diffusion part)

$$\partial_t(\phi+\Psi_e(\phi))-g(\partial_u(a\partial_u\phi)+\partial_v(b\partial_v\phi))=0,\qquad t\in(t_{j-1},t_j) \tag{3.2}$$

with the same boundary conditions and initial condition $\phi(u,v,t_{j-1})=C_j^{1/3}$. The obtained solution is denoted by $C_j^{2/3}$.

The last part of the procedure is solving the reaction part represented by the system

$$\partial_t(\phi+\Psi_e(\phi))+\partial_t S=0 \tag{3.3}$$

$$\partial_t S=K(\Psi_n(\phi)-S) \tag{3.4}$$

with the initial conditions $\phi(u,v,t_{j-1})=C_j^{2/3}$, $S(u,v,t_{j-1})=S_{j-1}$.

Finally, we put

$$C_j:=\phi(u,v,t_j),\qquad S_j:=S(u,v,t_j)$$

where $\phi(u,v,t_j)$, $S(u,v,t_j)$ represent the solution of (3.3)–(3.4).

For some theoretical results concerning this technique see e.g. [2].

3.3. Linear and non-linear transport problem. When we consider the equilibrium adsorption to be of Freundlich type as above and the initial profile is piecewise constant, the solution of the transport problem can be found in an analytical form. Let us consider a space discretization of rectangle Ω_R with nodes $u_0 = 0, u_1, \ldots, u_m = \pi$, $v_0 = v^{(2)}, v_1, \ldots, v_n = v^{(1)}$. We shall solve (3.1) in the strip $(u_{i_0-1/2}, u_{i_0+1/2}) \times (v^{(1)}, v^{(2)})$, with shocks in $v_0, v_{3/2}, v_{5/2}, \ldots, v_{n-1/2}, v_n$.

In general, F in (3.1) is not a constant but depends on v. Therefore we first use the transformation

$$z = G(v) = \int_{v^{(1)}}^{v} \frac{ds}{F(s)} \tag{3.5}$$

and we obtain

$$\partial_t(\bar{\phi} + \Psi_e(\bar{\phi})) - \partial_z \bar{\phi} = 0, \qquad \bar{\phi}(z, 0) = \phi_0(v) \tag{3.6}$$

First, let us consider the simplest case $\Psi_e(\phi) \equiv 0$, e.g. no equilibrium adsorption is present. Then the problem is reduced to linear transport problem

$$\partial_t \bar{\phi} - \partial_z \bar{\phi} = 0$$

that can be solved easily by shifting the initial profile, i.e.

$$\bar{\phi}(z, t) = \bar{\phi}(z + t, 0) = \phi_0(z + t)$$

In case $\Psi_e(\phi) \neq 0$, (3.6) represents a multiple Riemann problem. It's possible to find an analytical solution that consists of acceptable shocks and rarefaction waves (see e.g. [3]).

Finally we project the solution $\phi(v, t)$ to a piecewise constant function on intervals (v_j, v_{j-1}), $j = 1, \ldots, n$, which is done by taking averages over (v_j, v_{j-1}). This projection can now be used as input for the diffusion part.

3.4. Dispersion problem. Now we solve the problem

$$\partial_t(\phi + \Psi_e(\phi)) - g(\partial_u(a\partial_u \phi) + \partial_v(b\partial_v \phi)) = 0, \qquad t \in (t_{j-1}, t_j) \tag{3.7}$$

with boundary conditions of the form (2.5) and initial condition $\phi(u, v, t_{j-1}) = C_j^{1/3}$.

Here we use the standard finite volume method (see [1]) that leads to a system of equations:

1. In case with linear transport ($\Psi_e \equiv 0$) and 1D diffusion the system is linear and tridiagonal
2. In case with linear transport and 2D diffusion the system is linear, sparse, symmetric and positive definite, and we use a conjugated gradients method
3. When the transport is nonlinear, we obtain a nonlinear system and we use Newton method

3.5. Adsorption problem. Here we sketch a method for a system of ODE's of the form

$$\partial_t(\phi + \Psi_e(\phi)) + \partial_t S = 0 \tag{3.8}$$

$$\partial_t S = K(\Psi_n(\phi) - S) \tag{3.9}$$

on $< 0, t >$, with the initial conditions $\phi(0) = \phi_0$ (in our case we use $C_j^{2/3}$), $S(0) = S_0$ (we use S_{j-1}).

Integrating (3.8), using initial conditions, solving (3.9) in S and substituting for S we obtain an integral equation

$$f(\phi(t)) + S_0 e^{-Kt} + K \int_0^t e^{-K(t-z)} \Psi_n(\phi(z))\, dz = f(\phi_0) + S_0 \tag{3.10}$$

where $f(\phi) = \Psi_e(\phi) + \phi$.

Now we use time discretization $0 = \tau_0, \tau_1, \dots, \tau_{k-1}, \tau_k = t$ and moreover we apply a transformation $w = \Psi_n(\phi)$, denoting $f(\Psi_n^{-1}(w))$ by $\varphi(w)$. Thus we obtain for any $i \le k$

$$\varphi(w) = f(\phi_0) + S_0 - S_0 e^{-K\tau_i} - K \sum_{j=1}^{i} \int_{\tau_{j-1}}^{\tau_j} e^{-K(\tau_i - z)} w(z)\, dz \tag{3.11}$$

Finally, when we approximate the integrals in (3.11) (see [4]), it is possible to derive an implicit scheme (with m being the iterative index)

$$\varphi_{i+1}^{(m+1)} = f(\phi_0) + S_0 - S_0 e^{-K\tau_{i+1}} - K I_{i+1} - K \alpha_{i+1,i+1} w_{i+1}^{(m)},\ m = 0, 1... \tag{3.12}$$

where $w_{i+1} \approx w(\tau_{i+1})$, $\varphi_{i+1} = \varphi(w_{i+1})$ and terms $I_{i+1}(w_0, \dots, w_i)$, $\alpha_{i+1,i+1}$ come from the approximation of the integrals in (3.11). As the starting value for the implicit process we take $w_{i+1}^{(0)} = w_i$.

4. Computational aspects of the problem.

4.1. Time discretization. The considered time interval $< 0, T >$ doesn't have to be discretized equidistantly. In fact, while solving the transport problem, we determine the time step at run-time. Though it is theoretically unlimited, we go on with computation only till the moment when a collision between a rarefaction wave and a shock in front appears (see [3]). Some limitation is also required by the dispersion part when Newton method is used.

The adsorption part can represent the source of the most significant time step limitation. Especially when the adsorption coefficient K is large, it is necessary to use smaller time steps. Experience shows that this is also the case when there is a big difference between the powers in equilibrium and non-equilibrium sorption isotherms $\Psi_e(C)$ and $\Psi_n(C)$.

4.2. Solution of the discretized 2D problem. Once the rectangle Ω_R is discretized as described in 3.3, we have two possibilities for solving the 2D problem on this area. If the dispersion is considered to be a 1D problem (i.e. $\alpha_T = 0$, $D_0 = 0$), it is more convenient to solve the problem separately in each strip, so that each strip can use its own time steps. If we have to treat a 2D dispersion problem, we have to solve the whole problem simultaneously in all strips.

4.3. Time step for the adsorption part. In 3.5 we introduced a scheme for the adsorption part. This scheme allows us to solve the problem in several time substeps. Although in many cases it is sufficient to solve the adsorption in a single (global) time step, in many of the more complicated situations mentioned in 4.1 it helps to use more substeps for the adsorption (which doesn't affect the global time step used for transport and dispersion).

5. Numerical experiments.

Experiment 1: Comparison with analytical solution and finite difference scheme in 1D. For a simple 1D problem

$$\partial_t C + v(x)\partial_x C - \partial_x(D(x,t)\partial_x C) + \partial_t S = 0$$
$$\partial_t S = K(C - S)$$

it is possible to construct an analytical solution according to [5] and compare it with the solution obtained by our numerical scheme. In order to verify the efficiency of the method, we add also a comparison with the solution obtained by a simple finite difference scheme, where we put $\partial_t C \approx (C_i^k - C_i^{k-1})/\tau$, $\partial_x C \approx (C_i^k - C_{i-1}^k)/h$, $\partial_x^2 C \approx (C_{i+1}^k - 2C_i^k + C_{i-1}^k)/h^2$, $\partial_t S \approx (S_i^k - S_i^{k-1})/\tau$, $C_i^k = C(x_i, t_k)$, $S_i^k = S(x_i, t_k)$. In this experiment we set v and D constant, $v(x) \equiv 1$, $D(x,t) \equiv 10^{-4}$ and we use boundary condition $C_0(t) \equiv 1$. Sorption parameter $K = 6.95$. We use time step $\tau = 0.1$ and space grid step $h = 0.1$ for both finite difference and operator splitting schemes. As we can see in fig. 5.1, even with these relatively large time and space steps, the operator splitting method was able to obtain very precise results, while the finite difference scheme already suffered from a significant numerical dispersion.

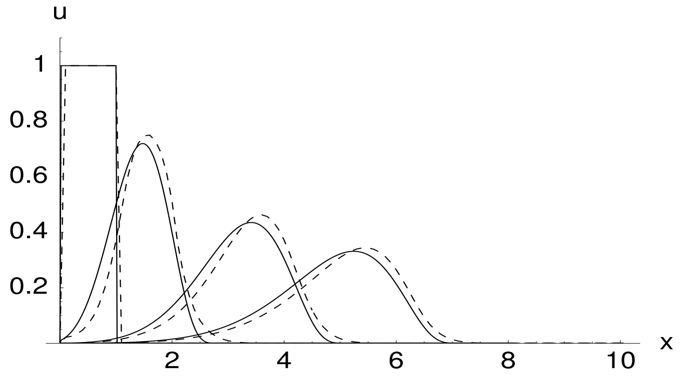

FIG. 5.1. *Comparison of analytical solution (solid line) with solution obtained by operator splitting scheme (large dots) and finite difference scheme (dotted line) at time levels 10, 30, 50*

Experiment 2: Comparison with the solution of an equilibrium mode problem in 1D. Here we put $\Psi_e(C) = C^p$, $\Psi_n(C) = C^q$ with $p = q = 0.75$ and the initial condition is $C(x,0) = 1$ for $x < 1$ and $C(x,0) = 0$ otherwise. Boundary condition is $C(0,t) = 0$. The method was tested for a wide range of values of the coefficient K, starting with $K = 10^{-5}$, when the non-equilibrium adsorption is almost negligible, and ending with $K = 100$ which makes the non-equilibrium adsorption behave as an almost equilibrium mode process. In both cases we have a possibility to verify the results. For small K the results shouldn't be very different from the ones of the equilibrium mode problem. On the other hand, when K is large and we consider $p = q$, the solution should be similar to the solution of the problem with $F(u) = u + 2u^p$ and no reaction part (again the equilibrium mode problem). The tests that have been done show that this is true.

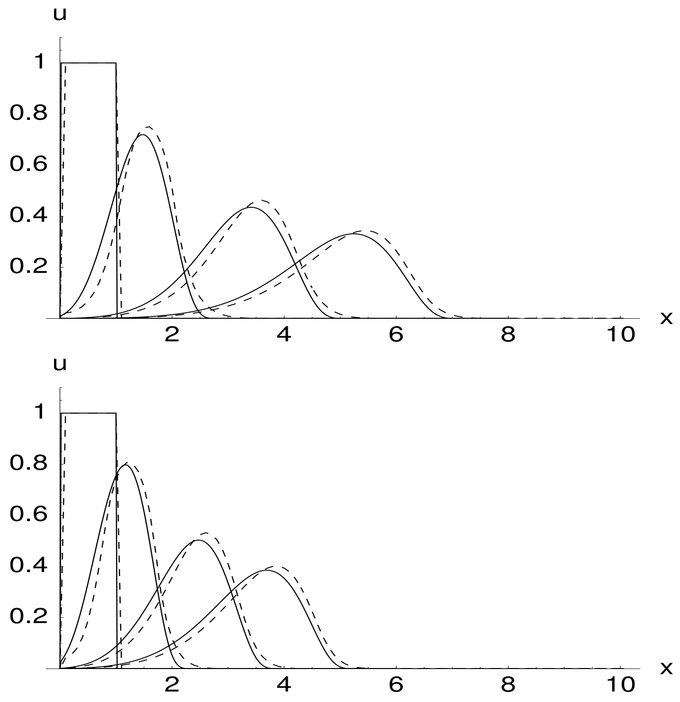

FIG. 5.2. *Solutions of the non-equilibrium mode problems for* $D = 10^{-1}$, $K = 10^{-5}$ *and* $K = 100$, *at time levels 0, 2, 6, 10 (solid line) compared with the solutions of the equilibrium mode problems (dashed line)*

Experiment 3: Solution of a 2D problem. Now we take a 2D problem with two wells situated at points $(5, 0)$ (injection well) and $(-5, 0)$ (extraction well). We consider both adsorption isotherms of the form $\Psi(C) = C^{0.75}$, dispersivities $\alpha_L = 0.1$, $\alpha_T = 0$, molecular diffusion with $D_0 = 0$, sorption coefficient $K = 10^{-5}$ and $K = 1.0$. Boundary condition is of pulse shape form, $C_0(t) = 1$ for $t < 2.0$ and $C_0(t) = 0$ otherwise. We use a space grid with 80×400 grid points and time step $\tau = 0.04$. The lines in the pictures represent concentration levels ($C = 0.05, 0.1 \ldots, 1.0$) at time $t = 8.0$ (fig. 5.3).

Experiment 4: Breakthrough curves

Breakthrough curves represent the discharge of contaminant in the extraction well. In fig. 5.4 are displayed breakthrough curves (plot of average contaminant concentration in the extraction well versus time) in time interval $< 0, 15 >$ for various values of sorption coefficient K. For the other parameters and for space discretization we use the same data as in Experiment 3.

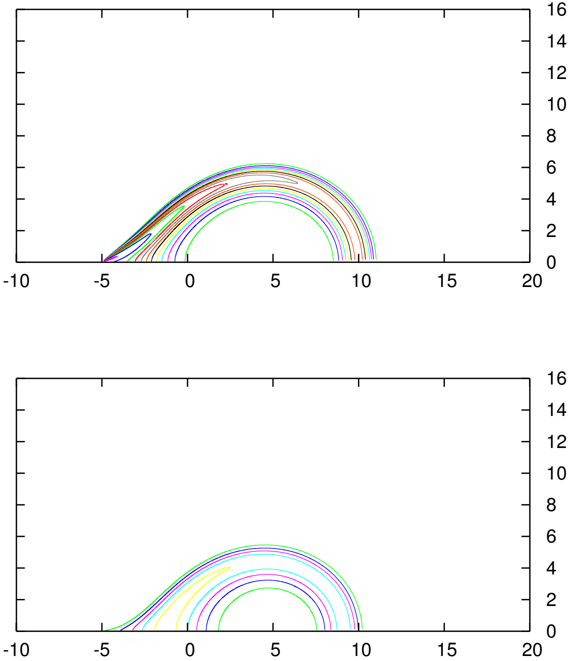

FIG. 5.3. *Solutions of 2D problems with very slow adsorption ($K = 10^{-5}$) and faster adsorption ($K = 1.0$) after 8 days.*

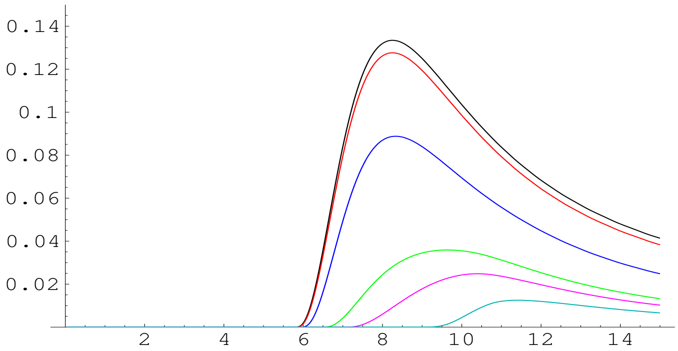

FIG. 5.4. *Breakthrough curves for sorption coefficients* $K = 10^{-5}$, 0.01, 0.1, 0.5, 1.0 *and* 10.0

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