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REPORT*RAPPORT*

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ABSTRACT

Operator or time splitting is often used in the numerical solution of initial boundary value problems for differential equations. It is, for example, standard practice in computational air pollution modelling where we encounter systems of three-dimensional, time-dependent partial differential equations of the advection-diffusion-reaction type. For such systems little attention has been devoted to the analysis of splitting and to the question why splitting can work so well. From the theoretical point of view, the success of splitting is primarily determined by the splitting error. This paper presents an analysis of operator splitting aimed at providing insight into the splitting error. Using the Lie operator formalism, a general expression is derived for a three-term Strang splitting in the pure initial value case. For a class of advection-diffusion-reaction problems the splitting error is analyzed in greater detail. A special case is discussed in which the splitting error can be reduced. Also some attention is paid to the use of operator splitting in initial boundary value problems.

1. INTRODUCTION

Virtually all processes modelled by time-dependent partial differential equations (PDEs) split additively in subprocesses for which simpler PDEs exist. This greater simplicity also carries over to their numerical counterparts, which already a long time ago has led to the use of operator splitting or time splitting. Within operator splitting subprocesses are treated on their own in numerical time-stepping while adopting a certain order of reappearance. An early influential paper is Strang [10], where a symmetrical order of reappearance was proposed, which formally yields 2nd-order consistency.

In this paper we focus on this form of symmetrical Strang splitting for systems of advection-diffusion-reaction equations

$$\frac{\partial c}{\partial t} + \nabla \cdot (\underline{u}c) = \nabla \cdot (K \nabla c) + R(c), \quad c = c(\underline{x}, t), \quad \underline{x} \in \mathbb{R}^3. \tag{1.1}$$

Although our findings do have a wider scope, our motivating application is atmospheric air quality modelling where PDE systems like (1.1) lie at the heart of complicated models employed in studies on the chemical composition of the atmosphere. The societal motivation for these studies concerns air pollution. Throughout we suppose that the velocity vector $\underline{u}$ and the diffusion coefficient matrix K are given. Hence the problem is linear with respect to advection and diffusion, but nonlinear in the chemical reaction term R. The dependent variable c represents a vector of chemical species

concentrations, which evolve in time due to advection, diffusion, chemical interactions, emissions, and depositions, the latter three all contained in R.

To the best of our knowledge, one of the first influential papers on computational air quality modelling discussing splitting is McRae, Goodin and Seinfeld [9]. More references specifically concerning air quality modelling can be found in Zlatev [14]. Nowadays operator splitting is standard practice in this field. However, for PDE systems like (1.1), in the literature very little attention has been devoted to the analysis of splitting and to the question why splitting can work so well. From the theoretical point of view, the success of splitting is primarily determined by the splitting error, which is introduced by solving subproblems one after another in a completely decoupled manner. In general this splitting error always exists, also when all subproblems are solved exactly. The aim of this paper is to present an analysis of operator splitting and to provide insight into the splitting error.

In Section 2 we derive an expression for the Strang splitting error for arbitrary autonomous systems of differential equations using the Lie operator formalism, including the notion of commutators for nonlinear problems, the notion of the modified problem and the celebrated Baker-Campbell-Hausdorff formula. Here we have made fruitful use of material from Sanz-Serna and Calvo [7] and Sanz-Serna [8]. Section 3 focuses on the advection-diffusion-reaction problem (1.1). The body of this section consists of a theorem, which shows under which circumstances advection, diffusion and reaction commute with one another, assuming exact integration. This commutativity is of great importance, because when all processes commute, we have a zero splitting error. In Section 4 the splitting error is discussed in greater detail for a number of simplified test models. Simplifications cannot be avoided since for the general problem class (1.1) the error expressions are much too long to handle. Further we discuss ways to reduce the splitting error and address the subject of inconsistencies, which can occur if Strang splitting is used in case of initial boundary value problems. The final Section 5 summarizes our findings and contains a number of general remarks.

2. Strang splitting and the Lie operator formalism

In this section we will derive an expression for the Strang splitting error for the general, nonlinear, autonomous system of differential equations,

$$c_t = f\left(\underline{x}, c\right) \equiv f_1\left(\underline{x}, c\right) + f_2\left(\underline{x}, c\right) + f_3\left(\underline{x}, c\right), \quad t \in [t_0, T], \quad \underline{x} \in \mathbb{R}^d, \quad c\left(\underline{x}, t_0\right) = c_0\left(\underline{x}\right). \tag{2.1}$$

The solution $c\left(\underline{x}, t\right)$ is supposed to be vector-valued in $\mathbb{R}^m$ and f and its parts f_1, f_2 and f_3 can represent a nonlinear vector function in $\mathbb{R}^m$ or some spatial derivative operator. In our notation we will mostly, just for convenience, suppress the dependence on the spatial variable $\underline{x} = (x, y, z)$. The spatial dimension d is not yet fixed. To derive the splitting error expression, at this stage we merely consider an abstract initial value problem (2.1) in the function space $\mathbf{S}$ of real, sufficiently often differentiable vector-valued functions c on $\mathbb{R}^d \times [t_0, T]$. In addition we assume that all operators encountered in our derivations, are sufficiently differentiable in all their variables. Our starting problem (1.1) provides a particular example for (2.1).

2.1 Strang splitting

Let $S\left(\tau\right)$ denote the solution (semigroup) operator for (2.1), that is

$$c\left(t + \tau\right) = S\left(\tau\right) c\left(t\right),$$

and $S_k\left(\tau\right)$ the solution operator for the subproblem $c_t = f_k\left(c\right)$. Let $\tilde{S}_k\left(\tau\right)$ denote a consistent, numerical approximation to $S_k\left(\tau\right)$, for example defined by a Runge-Kutta type method. For the abstract initial value problem (2.1), we then compactly represent the celebrated Strang splitting scheme [10] by

$$\tilde{c}\left(t + \tau\right) = \tilde{S}\left(\tau\right) \tilde{c}\left(t\right), \quad \tilde{S}\left(\tau\right) \equiv \tilde{S}_1\left(\tfrac{1}{2}\tau\right) \tilde{S}_2\left(\tfrac{1}{2}\tau\right) \tilde{S}_3\left(\tau\right) \tilde{S}_2\left(\tfrac{1}{2}\tau\right) \tilde{S}_1\left(\tfrac{1}{2}\tau\right). \tag{2.2}$$

The solution $\tilde{c}\,(t+\tau)$ denotes the approximation to $c\,(t+\tau)$ resulting from approximately solving the subproblems $c_t = f_k\,(c)$ in the given sequential order. The solution operator $\tilde{S}$ is the resulting splitting approximation to S. Note that $\tilde{S}_k$ is still thought to be space continuous, that is without spatial discretization. In our derivation we will not specify $\tilde{S}_k$, but instead we assume that with $\tilde{S}_k$ we may associate the modified problem [7, 8][1],

$$c_t = F_k\,(c) \equiv f_k\,(c) + \tau^{p_k} E_k\,(c)\,, \tag{2.3}$$

where $\tau^{p_k} E_k\,(c)$ represents the local truncation error of the integration method defining $\tilde{S}_k$. The integer p_k is the order of consistency. By definition, as the local error of integration schemes is normally an infinite series expansion in τ, E_k itself may still depend on the step size τ. The modified problem concept is very convenient when it is combined with the Lie operator formalism introduced below. Adopting the modified problem concept means that we act as if we apply Strang splitting to the modified problem,

$$c_t = F\,(c) \equiv F_1\,(c) + F_2\,(c) + F_3\,(c)\,, \tag{2.4}$$

while solving the subproblems $c_t = F_k\,(c)$ exactly. Trivially, with $\tilde{S}_k$ one may associate the exact solution operator S_k, in which case the original subproblems $c_t = f_k\,(c)$ are supposed to be solved exactly, that is without time integration error.

2.2 The Lie operator formalism
Strang splitting always leads to a 2nd-order approximation, at least in a formal sense. We are interested in the structure of the splitting error. Albeit tedious, local splitting errors can always be obtained by straightforward Taylor expansions (see for example [5, 10]). This, however, leads to an expression which does not reveal in a clear way the structure of the error. For its derivation we therefore adopt the Lie operator formalism. This formalism will enable use of the celebrated Baker-Campbell-Hausdorff formula. The BCH formula yields a lot of insight in the particular structure of splitting errors. The authors learned the Lie operator formalism from [7, 8]. For selfcontainedness we here repeat the material from [7, 8] needed for our purpose. We also made fruitful use of a brief unpublished note of our colleague W. Hundsdorfer, who also refers to [7]. A nice introduction to Lie operators can also be found in [3].

Consider the general differential equation (2.4). With each given operator F, a Lie operator is associated, which we denote by $\mathcal{F}$. This Lie operator is a linear operator acting on the space of operators defined on $\mathbf{S}$. $\mathcal{F}$ maps each operator G into the new operator $\mathcal{F}G$, such that for any element $c \in \mathbf{S}$,

$$(\mathcal{F}G)\,(c) = G'\,(c)\,F\,(c)\,. \tag{2.5}$$

($'$ denotes differentiation with respect to c). For the solution $c\,(t)$ of (2.4) it easily follows that

$$(\mathcal{F}G)\,(c\,(t)) = \frac{\partial}{\partial t}G\,(c\,(t))\,, \tag{2.6}$$

and from induction to k that

$$\frac{\partial^k}{\partial t^k}G\,(c\,(t)) = \left(\mathcal{F}^k G\right)(c\,(t))\,. \tag{2.7}$$

The above relations (2.6) and (2.7) hold for any G defined on $\mathbf{S}$, in particular for the identity I. Inserting I for G and using the Taylor expansion of the true solution, we can write $c\,(t+\tau)$ in terms of the exponentiated Lie operator form or Lie-Taylor series,

$$c\,(t+\tau) = \left(e^{\tau \mathcal{F}} I\right)(c\,(t))\,.$$

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[1] Throughout we use $c \in \mathbf{S}$ to denote the solution of any differential equation. From the context it will be clear to which equation we are referring, for example our original problem (2.1) or a different problem such as (2.3). Likewise, c can denote an arbitrary element $\in \mathbf{S}$.

The same argument concerning this exponentiated Lie operator applies to each of the subproblems $c_t = F_k(c)$. When we compose the resulting exponentiated Lie operators in the same order as the solution operators in the splitting procedure, with which they are associated, we can reveal that the Strang splitting solution (2.2) can be expressed as

$$\tilde{c}(t+\tau) = \left(e^{\frac{1}{2}\tau\mathcal{F}_1} e^{\frac{1}{2}\tau\mathcal{F}_2} e^{\tau\mathcal{F}_3} e^{\frac{1}{2}\tau\mathcal{F}_2} e^{\frac{1}{2}\tau\mathcal{F}_1} I \right) (\tilde{c}(t)). \tag{2.8}$$

At this stage the BCH formula proves to be useful. Let X, Y be linear operators. According to this formula, the product $e^X e^Y$ can then be written as the exponential e^Z of

$$Z = X + Y + \frac{1}{2}[X,Y] + \frac{1}{12}([X,X,Y] + [Y,Y,X]) + \frac{1}{24}[X,Y,Y,X] + \cdots, \tag{2.9}$$

where $[X,Y]$ is the commutator $[X,Y] = XY - YX$ and $[X,X,Y]$ is recursively defined by $[X,X,Y] = [X,[X,Y]]$, etc. Note that, if X and Y are Lie operators, Z is also a Lie operator.

We put $X = \frac{1}{2}\tau\mathcal{F}_1$ etc. and apply (2.9) four times, or Yoshida's formula [7] twice, resulting in an expression for the symmetrical Strang splitting solution (2.8),

$$\tilde{c}(t+\tau) = \left(e^{\tau\tilde{\mathcal{F}}} I \right) (\tilde{c}(t)), \quad e^{\tau\tilde{\mathcal{F}}} \equiv e^{\frac{1}{2}\tau\mathcal{F}_1} e^{\frac{1}{2}\tau\mathcal{F}_2} e^{\tau\mathcal{F}_3} e^{\frac{1}{2}\tau\mathcal{F}_2} e^{\frac{1}{2}\tau\mathcal{F}_1},$$

where the new Lie operator $\tilde{\mathcal{F}}$ is formally defined by an infinite series expansion which is even in τ. Its leading part reads

$$\begin{aligned}
\tilde{\mathcal{F}} = {}& \mathcal{F}_1 + \mathcal{F}_2 + \mathcal{F}_3 - \frac{1}{24}\tau^2[\mathcal{F}_1,\mathcal{F}_1,\mathcal{F}_2] - \frac{1}{24}\tau^2[\mathcal{F}_1,\mathcal{F}_1,\mathcal{F}_3] + \\
&+ \frac{1}{12}\tau^2[\mathcal{F}_2,\mathcal{F}_2,\mathcal{F}_1] - \frac{1}{24}\tau^2[\mathcal{F}_2,\mathcal{F}_2,\mathcal{F}_3] + \frac{1}{12}\tau^2[\mathcal{F}_3,\mathcal{F}_3,\mathcal{F}_1] + \\
&+ \frac{1}{12}\tau^2[\mathcal{F}_3,\mathcal{F}_3,\mathcal{F}_2] + \frac{1}{12}\tau^2[\mathcal{F}_2,\mathcal{F}_3,\mathcal{F}_1] + \frac{1}{12}\tau^2[\mathcal{F}_3,\mathcal{F}_2,\mathcal{F}_1] + O\left(\tau^4\right).
\end{aligned} \tag{2.10}$$

If we are able to recover the operator $\tilde{F}$ corresponding with $\tilde{\mathcal{F}}$, we are led to the modified problem,

$$c_t = \tilde{F}(c),$$

associated with the symmetrical Strang splitting scheme.

We first derive the operators associated with the commutators (the so-called Lie or Poisson brackets). Direct application of (2.5) to the commutator $[\mathcal{F}_l, \mathcal{F}_m]$ yields for any G and any $c \in \mathbf{S}$,

$$[\mathcal{F}_l, \mathcal{F}_m] G(c) = (G'(c) F_m(c))' F_l(c) - (G'(c) F_l(c))' F_m(c).$$

Repeating this for $[\mathcal{F}_k, \mathcal{F}_l, \mathcal{F}_m]$ and inserting the identity I for G, gives

$$[\mathcal{F}_k, \mathcal{F}_l, \mathcal{F}_m] I(c) = (F'_m F_l)' F_k - (F'_l F_m)' F_k - (F'_k F_m)' F_l + (F'_k F_l)' F_m,$$

where all operators at the right-hand side are evaluated at c. We rewrite this expression as

$$[\mathcal{F}_k, \mathcal{F}_l, \mathcal{F}_m] I(c) = F'_{lm} F_k - F'_k F_{lm}, \quad F_{lm} \equiv F'_m F_l - F'_l F_m, \tag{2.11}$$

where, naturally, the new operator F_{lm} is called the commutator for F_l and F_m. To find $\tilde{F}$ we insert expression (2.11) for all commutators occurring in (2.10), which results in the modified problem for the Strang splitting (2.2),

$$c_t = \tilde{F}(c) \equiv F(c) + \tau^2 E_F(c) + O\left(\tau^4\right), \tag{2.12}$$

where $\tau^2 E_F(c)$ is the counterpart of the τ^2-term of (2.10). Remember here equation (2.6). After rearranging the terms, to make the contribution of splitting F_1 from F_2, F_1 from F_3 and F_2 from

F_3 to the splitting error more precise, E_F is written as

$$E_F \quad \equiv \quad -\frac{1}{24}F_{12}' \left(F_1 + 2F_2 + 2F_3\right) + \frac{1}{24}\left(F_1' + 2F_2' + 2F_3'\right) F_{12} \tag{2.13}$$
$$-\frac{1}{24}F_{13}' \left(F_1 + 2F_2 + 2F_3\right) + \frac{1}{24}\left(F_1' + 2F_2' + 2F_3'\right) F_{13}$$
$$-\frac{1}{24}F_{23}' \left(F_2 + 2F_3\right) + \frac{1}{24}\left(F_2' + 2F_3'\right) F_{23}.$$

The solution of the modified problem (2.12), assuming it exists, may be interpreted as the Strang splitting solution (backward analysis interpretation [8]).

The term $\tau^2 E_F \left(c\left(t\right)\right)$ represents the leading term of the local error of the Strang splitting scheme evaluated at $c\left(t\right)$. Note that the global error, $\tilde{c}\left(t+\tau\right) - c\left(t+\tau\right)$, can be directly seen to satisfy

$$\tilde{c}\left(t+\tau\right) - c\left(t+\tau\right) = \left(e^{\tau\bar{\mathcal{F}}}I\right)\left(\tilde{c}\left(t\right) - c\left(t\right)\right) + \left(e^{\tau\bar{\mathcal{F}}}I - e^{\tau\mathcal{F}}I\right)\left(c\left(t\right)\right),$$

where $(e^{\tau\bar{\mathcal{F}}}I - e^{\tau\mathcal{F}}I)c(t))$ is the complete local splitting error. The local splitting error is even in τ provided that the Lie operators are independent of τ or also even in τ. The leading τ^2-term is of course equal to the τ^2-term in (2.10).

A few important aspects concerning the splitting error should already be mentioned. When the three split operators F_1, F_2, F_3 commute with one another, $\tilde{F} = F$, no splitting error occurs. When, for example, only F_1 and F_2 commute, the first and second term connected with the commutator F_{12} cancel and no error occurs due to splitting F_1 from F_2. It is the Lie operator approach that attends to this clarity. The beauty of this approach is that it can be formulated for any autonomous operator F with its split parts F_1, F_2, F_3.

What remains to be done is to identify the local splitting error for the original problem (2.1) that would arise if the substeps would be integrated exactly. For that purpose we work the modified problem expression (2.3) into (2.12) and (2.13). A straightforward computation then leads to

$$c_t = \tilde{f}\left(c\right) \equiv f\left(c\right) + \tau^2 E_f\left(c\right) + O\left(\tau^{2+p_1}\right) + O\left(\tau^{2+p_2}\right) + O\left(\tau^{2+p_3}\right) + O\left(\tau^4\right), \tag{2.14}$$

where

$$\tau^2 E_f\left(c\right) = \tau^2 E_s\left(c\right) + \tau^{p_1} E_1\left(c\right) + \tau^{p_2} E_2\left(c\right) + \tau^{p_3} E_3\left(c\right),$$

with E_s defined by

$$E_s \quad \equiv \quad -\frac{1}{24}f_{12}'\left(f_1 + 2f_2 + 2f_3\right) + \frac{1}{24}\left(f_1' + 2f_2' + 2f_3'\right) f_{12} \tag{2.15}$$
$$-\frac{1}{24}f_{13}'\left(f_1 + 2f_2 + 2f_3\right) + \frac{1}{24}\left(f_1' + 2f_2' + 2f_3'\right) f_{13}$$
$$-\frac{1}{24}f_{23}'\left(f_2 + 2f_3\right) + \frac{1}{24}\left(f_2' + 2f_3'\right) f_{23}.$$

We see that in (2.14) the leading term consists of the sum of the three local integration errors introduced in (2.3) and the error term $\tau^2 E_s\left(c\right)$. The operator E_s obviously defines the leading term of the local splitting error for exact integration. That is, if all split steps would be integrated exactly, or just very accurately, then this term will dominate the local splitting error. On the other hand, if f_1, f_2, f_3 commute with one another, E_s will completely vanish. This means that the success of Strang splitting in terms of local accuracy is determined by E_s in the first place.

3. ADVECTION-DIFFUSION-REACTION PROBLEMS

In this section we will consider the advection-diffusion-reaction problem (1.1). In relation to (2.1) we associate f_1 with advection, f_2 with diffusion and f_3 with chemistry, that is

$$f_1\left(c\right) = -\nabla \cdot \left(\underline{u}c\right), \quad f_2\left(c\right) = \nabla \cdot \left(K \nabla c\right), \quad f_3\left(c\right) = R\left(c\right).$$

Observe that the velocity $\underline{u} = \left(u, v, w\right)$, the diffusion matrix coefficient K and the reaction term $R\left(c\right)$ do depend on the spatial variable $\underline{x} = \left(x, y, z\right)$. Also note that no component coupling exists in the advection and diffusion parts as opposed to the chemistry part $R(c)$ $(R\left(c\right) \in \mathbb{R}^m)$.

3.1 Commutativity

First we will answer the question when true commutativity occurs between the advection, diffusion and chemistry operators. In that case no splitting error exists between the commutating processes. To find the answer we have to elaborate the commutators

$$f_{lm}\left(c\right) = f'_m\left(c\right)f_l\left(c\right) - f'_l\left(c\right)f_m\left(c\right), \quad \left(l,m\right) = \left(1,2\right),\left(1,3\right),\left(2,3\right),$$

and equate them to zero. In this elaboration the derivatives $f'_1\left(c\right)$ and $f'_2\left(c\right)$ are to be interpreted componentwise. They in fact act as diagonal matrix differential operators having equal entries. More precisely, owing to their linearity we have, for any element $s \in \mathbf{S}$,

$$f'_1\left(c\right)s \equiv f_1\left(s\right) = -\nabla \cdot \left(\underline{u}s\right), \quad f'_2\left(c\right)s \equiv f_2\left(s\right) = \nabla \cdot \left(K\,\nabla\,s\right).$$

Trivially, the derivative $f'_3\left(c\right)$ is the $m \times m$ Jacobian matrix $R'\left(c\right)$. Our elaboration leads to the following theorem.

Theorem 1

a) *Advection commutes with diffusion if $\underline{u}$ and K are independent of $\underline{x}$.*

b) *Advection commutes with chemistry if $\nabla \cdot \underline{u} = 0$ and R is independent of $\underline{x}$.*

c) *Diffusion commutes with chemistry if R is linear in c and independent of $\underline{x}$.*

d) *With exact integration no splitting error exists if R is linear in c and $\underline{u}$, K and R are independent of $\underline{x}$.*

Result d) is based on a), b), c) for which the proof is given below. Results a) and d) can also be concluded from Fourier analysis (the standard constant coefficient case). Note that the requirement R independent of $\underline{x}$ does not mean that R is independent of $c = c(\underline{x}, t)$.

Proof.

a) For commutativity of advection and diffusion we need equality of

$$f'_2\left(c\right)f_1\left(c\right) = -\nabla \cdot \left(K\,\nabla\,\left(\nabla \cdot \left(\underline{u}c\right)\right)\right),$$

and

$$f'_1\left(c\right)f_2\left(c\right) = -\nabla \cdot \left(\underline{u}\,\left(\nabla \cdot \left(K\,\nabla\,c\right)\right)\right).$$

Recall that c is a vector but that $\underline{u}$ and K act componentwise. Further elaborating these two expressions trivially shows equality, if both $\underline{u}$ and K are independent of $\underline{x}$. In general the two expressions are not equal.
b) We need to compare

$$f'_3\left(c\right)f_1\left(c\right) = -R'\left(c\right)\nabla \cdot \left(\underline{u}c\right),$$

and

$$f'_1\left(c\right)f_3\left(c\right) = -\nabla \cdot \left(\underline{u}R\left(c\right)\right).$$

Let $R_x\left(c\right)$ denote the partial derivative vector of $R\left(\underline{x},c\right)$ with respect to x. Introduce a similar meaning for $R_y\left(c\right)$ and $R_z\left(c\right)$. An elementary calculation yields

$$f'_3\left(c\right)f_1\left(c\right) = -R'\left(c\right)\left(\underline{u} \cdot \nabla c\right) - R'\left(c\right)\left(\nabla \cdot \underline{u}\right)c,$$

and

$$\begin{aligned} f'_1\left(c\right)f_3\left(c\right) &= -\left(uR\left(c\right)\right)_x - \left(vR\left(c\right)\right)_y - \left(wR\left(c\right)\right)_z \\ &= -R'\left(c\right)\left(\underline{u} \cdot \nabla c\right) - \left(\nabla \cdot \underline{u}\right)R\left(c\right) - \left(uR_x\left(c\right) + vR_y\left(c\right) + wR_z\left(c\right)\right). \end{aligned}$$

The two expressions are equal if the velocity field is divergence-free and R is independent of x, y and z. This proves part b) of the theorem. Note that in this case R is allowed to depend on c.

c) For commutativity of diffusion and chemistry we need equality of

$$f_3'(c) f_2(c) = R'(c) \left(\nabla \cdot (K \nabla c) \right),$$

and

$$f_2'(c) f_3(c) = \left(\nabla \cdot (K \nabla) \right) R(c).$$

Introduce the vectors,

$$X = R_x(c) + R'(c) c_x, \quad Y = R_y(c) + R'(c) c_y, \quad Z = R_z(c) + R'(c) c_z.$$

Then we can write

$$
\begin{aligned}
f_2'(c) f_3(c) &= \frac{\partial}{\partial x} \left(K_{11} X + K_{12} Y + K_{13} Z \right) + \frac{\partial}{\partial y} \left(K_{21} X + K_{22} Y + K_{23} Z \right) + \\
&+ \frac{\partial}{\partial z} \left(K_{13} X + K_{23} Y + K_{33} Z \right),
\end{aligned}
$$

and

$$
\begin{aligned}
f_3'(c) f_2(c) &= R'(c) \left[\frac{\partial}{\partial x} \left(K_{11} c_x + K_{12} c_y + K_{13} c_z \right) + \right. \\
&+ \left. \frac{\partial}{\partial y} \left(K_{21} c_x + K_{22} c_y + K_{23} c_z \right) + \frac{\partial}{\partial z} \left(K_{31} c_x + K_{32} c_y + K_{33} c_z \right) \right].
\end{aligned}
$$

It immediately follows that in general the two expressions will differ in value. However, in the special case that R is linear in c and explicitly independent of $\underline{x}$, we do have equality and hence commutativity. Note that in this case dependence of K on $\underline{x}$ is permitted. □

We have to conclude that in almost every practical situation splitting errors arise, since the case of a space independent velocity field $\underline{u}$ and diffusion matrix K, combined with a space independent and linear chemistry process R, hardly occurs. On the other hand, the extended use of Strang splitting in computational air pollution modelling leads to the conjecture that in this field splitting errors are kept within reasonable bounds, something which is confirmed for the examples presented in [13]. The following interpretation of the results of Theorem 1, based on relevant practical properties of $\underline{u}, K$ and R, is in further support of this conjecture.

An important feature for air pollution models of the state of the atmosphere [1] is the diurnal cycle of sunsets and sunrises. This cycle obviously introduces a space-time dependency which manifests itself in two ways relevant to operator splitting errors, viz. through the photochemical reactions and the vertical transport. Let us first consider the photochemistry. After sunset, photochemical reactions are switched off. This not only simplifies the chemistry, but also strongly diminishes the spatial dependency of R. If also temperature and humidity hardly vary in $\underline{x}$, then at nightly periods R is often totally independent of x. Hence, if $\nabla \cdot \underline{u} = 0$, advection will commute with chemistry according to result b) of Theorem 1, diminishing the splitting error. The vertical transport is modelled by parameterized turbulent diffusion through the coefficient K. Since at night the stability of the atmosphere often increases, in many models K decreases to very small values after sunset. This means that the commutators f_{12} and f_{13} between diffusion and advection and diffusion and chemistry strongly decrease, which will lead to a strong decrease of the splitting error. It also often occurs that the velocity field $\underline{u}$ and the diffusion coefficient K vary slowly in $\underline{x}$, so that even during day time f_{12} can get small in large parts of the space domain.

Summarizing, the diurnal cycle strongly influences the commutators leading to a relatively small local splitting error over nightly periods. During these periods the global splitting error will also decrease owing to stability. In other words, the splitting error will oscillate with the diurnal cycle and not amplify beyond bound for evolving time. Specific circumstances will of course determine actual values.

4. ILLUSTRATIONS

We now proceed with simplified test models from class (1.1) so as to further study the local splitting error, in particular the leading error term $\tau^2 E_s$ defined in equation (2.15). Furthermore, we look at ways to reduce the splitting error in these cases and we pay attention to initial boundary value problems. Simplified models are used to avoid error terms too long to handle.

4.1 Examples of commutators

First consider the 3D problem,

$$c_t + uc_x + vc_y = (\kappa c_z)_z + R(c), \quad u_x + v_y = 0, \tag{4.1}$$

in which the transport is based on a divergence-free, horizontal velocity field, $\underline{u} = (u, v, 0)$, and on vertical diffusion with diffusion coefficient κ. This problem is relevant to many practical studies in the field of atmospheric air quality modelling where horizontal wind patterns dominate advection by wind and one-dimensional parameterized turbulent diffusion is used to simulate transport in the vertical direction. Putting

$$f_1(c) = -uc_x - vc_y, \quad f_2(c) = (\kappa c_z)_z, \quad f_3(c) = R(c),$$

we derive the commutators,

$$
\begin{aligned}
f_{12}(c) &= -\left(\kappa(uc_x + v_y)_z\right)_z + u(\kappa c_z)_{xz} + v(\kappa c_z)_{yz}, \\
f_{13}(c) &= uR_x(c) + vR_y(c), \\
f_{23}(c) &= -\kappa_z R_z(c) - \kappa R_z z(c) - 2\kappa R'_z(c)c_z - \kappa R''(c)c_z c_z.
\end{aligned}
$$

Despite the simplifications introduced in (4.1), these commutators still turn out to be rather complicated. The associated splitting error term E_s becomes too long to provide even little insight. Therefore a further simplification is introduced below. In passing we note that f_{12}, rewritten as

$$
\begin{aligned}
f_{12}(c) &= -\kappa_z u_z c_x - \kappa_z v_z c_y - 2\kappa u_z c_{xz} - 2\kappa v_z c_{yz} - \kappa u_{zz} c_x - \kappa v_{zz} c_y \\
&\quad + \kappa_x u c_{zz} + \kappa_{xz} u c_z + \kappa_y v c_{zz} + \kappa_{yz} v c_z,
\end{aligned}
$$

reveals that when u and v are constant in z and κ is constant in x and y, the commutator f_{12} vanishes yielding a zero advection-diffusion splitting error.

We now proceed with the 2D problem,

$$c_t + uc_x = \kappa c_{zz} + R(c), \quad u \text{ constant}, \quad \kappa = \kappa(x), \quad R(c) = R(x, c), \tag{4.2}$$

with x and z as the independent space variables. Only a constant velocity in the x-direction exists, the diffusion coefficient κ is restricted to a x-dependent function, and the reaction term R may only depend on x, but not on z. For this model the split functions read

$$f_1(c) = -uc_x, \quad f_2(c) = \kappa c_{zz}, \quad f_3(c) = R(c).$$

Of importance is that all three commutators,

$$f_{12}(c) = u\kappa_x c_{zz}, \quad f_{13}(c) = u R_x(c), \quad f_{23}(c) = -\kappa R''(c)c_z c_z,$$

are unequal to zero, with the exception of special cases of course. In this sense sufficient generality is maintained compared to (4.1). According to (2.15), after a long calculation, $\tau^2 E_s(c)$ is found equal to

$$\tau^2 E_s(c) = \tau^2 (E_{12}(c) + E_{13}(c) + E_{23}(c)), \tag{4.3}$$

where

$$E_{12}(c) = -\frac{1}{24}u^2\kappa_{xx}c_{zz} - \frac{1}{12}u\kappa_x R''(c)c_z c_z, \tag{4.4}$$

$$E_{13}(c) = -\frac{1}{24}u^2 R_{xx}(c) + \frac{1}{12}u\left(R'(c)R_x(c) - R'_x(c)R(c)\right) + \frac{1}{12}u\kappa R''_x(c)c_z c_z, \tag{4.5}$$

$$E_{23}(c) = \frac{\kappa}{24}\left((R''(c)c_z c_z)'(\kappa c_{zz} + 2R(c)) - \left(\kappa\frac{\partial^2}{\partial z^2} + 2R'(c)\right)(R''(c)c_z c_z)\right). \tag{4.6}$$

Even for the simplified model problem (4.2) E_s is still a rather complicated expression, providing again little insight into the splitting error. We have to reckon with stiff chemistry, in which case R and its derivatives can possess extremely large entries. Whether these large entries will actually diminish the accuracy, depends in part on the size of $R''(c)\,c_z c_z$, being present in E_{12}, E_{13} and E_{23}. Observe here that $R''(c)$ is a tensor, $R''(c)\,c_z$ a matrix and c_z a vector, so that componentwise

$$\left(R''(c)\,c_z c_z\right)^{(i)} = \sum_{j,k=1}^{m} \frac{\partial^2 R^{(i)}(c)}{\partial c^{(j)}\,\partial c^{(k)}}\,c_z^{(j)}\,c_z^{(k)}.$$

If the chemistry is based on at most second order reactions, which is normal in atmospheric chemistry, the second derivative operator R'' is constant, that is independent of c. Further, many of the entries will be zero since chemistry normally gives rise to very sparse Jacobian matrices (species react with only a few others). However, at least a few large entries will always remain and the coupling between fast (stiff) and slowly (non-stiff) reacting species will determine how these large entries enter the local error.

Observe also that, in accordance with Theorem 1, E_{12} vanishes if κ is constant and E_{13} vanishes if R is independent of x. In general, E_{23} vanishes if and only if all entries of R'' are zero. This is the case for linear chemistry, that is for

$$R(c) = G\,c + B(x,z)\,,$$

with G a constant matrix. The source and sink vector B can still be space dependent. However, in contrast to the diffusion-chemistry error, in this case the advection-chemistry error E_{13} does not vanish as it is given by

$$E_{13}(c) = -\frac{1}{24}u^2 B_{xx} + \frac{1}{12}u G B_x. \tag{4.7}$$

The advection-diffusion error reads

$$E_{12}(c) = -\frac{1}{24}u^2 \kappa_{xx} c_{zz}.$$

As the error (4.7) illustrates, strong spatial variations in the sources and sinks contribute to the splitting error.

4.2 Splitting advection and diffusion

We next examine the effect of only Strang splitting advection and diffusion for the 2D model problem (4.2). In this case we are able to say more about the splitting error in relation to spatial and time integration errors. So we consider the model problem,

$$c_t + u c_x = \kappa c_{zz}, \quad u \text{ constant}, \quad \kappa = \kappa(x)\,. \tag{4.8}$$

According to (4.4), the modified equation for (4.8) reads

$$c_t + u c_x = \kappa c_{zz} - \frac{1}{24}\tau^2 u^2 \kappa_{xx} c_{zz} + O\left(\tau^4\right)\,.$$

The error $-\frac{1}{24}\tau^2 u^2 \kappa_{xx} c_{zz}$ can be seen as artificial diffusion due to splitting. To keep the local splitting error sufficiently small, it turns out to be necessary that in first approximation

$$\frac{1}{24}\tau^2 u^2 |\kappa(x)_{xx}| << \kappa(x)\,. \tag{4.9}$$

The explicit quadratic dependence on τu is clarifying as it reveals that in an actual application the Strang splitting should work well, as long as for the numerical advection integration a normal CFL condition holds and the split step size τ is taken equal to the advection step size Δt.

Let Δx denote a mesh width in the x-direction. A normal CFL condition then is

$$\frac{\Delta t \, |u|}{\Delta x} \leq C_{CFL} \approx 1.$$

Inserting this condition and the equality $\tau = \Delta t$ in (4.9) gives

$$\frac{1}{24} \, C_{CFL}^2 \, (\Delta x)^2 \, |\kappa\,(x)_{xx}\,| << \kappa\,(x)\,.$$

If $C_{CFL} \approx 1$ and $|\kappa\,(x)_{xx}\,|$ is of moderate size compared to $\kappa\,(x)$, the leading local splitting error contribution will behave like $O\,(\Delta x)^2$. This order of accuracy is satisfactory in the sense that many numerical advection schemes also generate $O\,(\Delta x)^2$ errors by the spatial discretization of the advection operator and $O\,(\Delta t)^2 = O\,(\tau^2) = O\,(\Delta x)^2$ errors by the temporal integration. On the other hand, if very large values for τu are allowed, as for example made possible by the use of an implicit unconditionally stable advection integrator, or by many successive steps within split intervals with a conditionally stable explicit one, then large splitting errors can arise.

Would we allow κ in (4.8) to also depend on z, the modified equation is given by

$$\begin{aligned}
c_t + uc_x &= (\kappa c_z)_z - 1/24\,\tau^2 u^2\,(\kappa_{xx} c_z)_z + 1/12\,\tau^2 u\{(-\,(\kappa_x \kappa_{zz})_z + (\kappa \kappa_{xzz})_z)\,c_z \\
&\quad + (-3\kappa_x \kappa_{zz} + 3\kappa \kappa_{xzz})\,c_{zz} + (-2\kappa_x \kappa_z + 2\kappa \kappa_{xz})\,c_{zzz}\} + O\,(\tau^4)\,.
\end{aligned}$$

Obviously, with appropriate modifications the above statements also hold for the case $\kappa = \kappa\,(x, z)$.

4.3 Reducing splitting errors

The error expressions (4.4) to (4.6) once again show that in general splitting errors will exist, because they depend on very different solution and problem properties. However, in actual applications it is sometimes possible to eliminate at least part of the splitting error. In this paragraph we will consider some of these possibilities.

For problem (4.1) one sometimes decides to solve chemistry and vertical diffusion coupled [2, 11, 12] so as to avoid error terms like E_{23} resulting from splitting diffusion and chemistry. This coupled solving involves the solution of a 1D diffusion-reaction system for every vertical column in a 3D grid. Unfortunately, when the number of chemical species is large [12], in spite of the 1D nature, a direct solution method using a standard band-solver in the linear algebra is costly. An iterative tridiagonal Gauss-Seidel type process is a very competitive alternative though, but this type of solution process only works for gas-phase chemistry [11]. Coupling between diffusion and chemistry yields in some, but not in every case, an acceptable possibility to reduce the splitting error.

Part of the splitting error can be truly eliminated for problems of the form,

$$c_t + uc_x = f\,(x, c)\,, \quad u \text{ constant.} \tag{4.10}$$

We restrict ourselves to the 1D case, but the theory can easily be extended to 2D and 3D problems with a non-constant velocity field. Although f can represent any arbitrary nonlinear vector function in $\mathbb{R}^m$, we shall associate with f a chemical process. Note that our following derivation can also be applied to problems like (4.1), where $f(x, y, z, c)$ stands for vertical diffusion and chemistry. Observe at last, as proved in Theorem 1, that the dependence of f on x in (4.10) is essential, because otherwise no splitting error exists and our derivation is redundant.

We consider a special splitting technique for equation (4.10) similar to a semi-Lagrangian method. The underlying idea has been discussed previously in [5, 6] and in [4]. A Lagrangian methods solves

$$\frac{dc}{dt} = f\,(x(t), c)\,, \quad \dot{x} = u, \tag{4.11}$$

along the characteristics, using a moving grid to keep track of them. In case of a semi-Lagrangian method one still solves (4.11) along the characteristics, but with this difference that no moving

grid is used and the solutions $c(x^* - u\tau, 0)$, needed as initial values for integration along the characteristics to calculate the solutions $c(x^*, \tau)$ in the gridpoints x^*, are found by interpolation between known solutions in neighbouring gridpoints. Hence, within each time step a semi-Lagrangian method maps the Lagrangian solution to an Eulerian grid.

Our splitting variant of this semi-Lagrangian method over an interval $[0, \tau]$ is described as

$$
\begin{array}{llll}
\frac{\partial c_1}{\partial t} + u \frac{\partial c_1}{\partial x} & = & 0, & c_1\,(x,0) & = & \tilde{c}\,(x,0) & (a) \\[2mm]
\frac{dc_2}{dt} & = & f\,(x(t), c_2)\,, \quad \dot{x} = u, & c_2\,(x - u\tau, 0) & = & c_1\,(x, \tau) & (b) \\[2mm]
\tilde{c}\,(x, \tau) & = & c_2\,(x, \tau)\,. &&&
\end{array}
$$

$$(4.12)$$

First the advection step (4.12a) is carried out on an Eulerian grid. Then the second equation (4.12b) is integrated on the same grid, but using $x = x(t)$, with as initial value the solution obtained from the proceeding advection step. Note here the resemblance with the semi-Lagrangian method. The initial values needed for integration along the characteristics are determined in a proceeding step apart from the actual integration. If the advection step is solved exactly on the grid, no splitting error occurs between advection and chemistry. When no exact advection step is achieved, the errors, which arise in an actual Eulerian advection step, resemble the interpolation errors of the semi-Lagrangian method.

The way in which we obtain the solution to (4.12b) is not prescribed. One can think for instance of applying a splitting scheme to split diffusion from chemistry or in case of gas-phase chemistry one can decide to use the earlier mentioned iterative tridiagonal Gauss-Seidel solution method.

4.4 Strang splitting in initial boundary value problems.

Till now, we restricted ourselves to pure initial value problems. In practical applications though, we mostly encounter initial boundary value problems. When we use operator splitting in these situations, we have to reckon with boundary errors. We will now focus on the subject of prescribing boundary conditions in the intermediate steps of the Strang splitting and on the resulting possibility of inconsistencies between these boundary conditions and the solutions calculated in the proceeding intermediate steps. These inconsistencies can lead to numerical errors.

We consider once more the 2D autonomous problem (4.1) ($v = 0$) now described over a bounded domain $\{(x, z) \mid 0 \leq x \leq 2\pi, \; 0 \leq z \leq z_H\}$,

$$c_t + uc_x = (\kappa c_z)_z + R\,(c)\,, \tag{4.13}$$

where u is constant in x and κ and R can depend on x and z. As boundary conditions we prescribe 2π-periodicity in x-direction, and on $z = 0$ (the earth surface) and $z = z_H$ we prescribe

$$
\kappa c_z = d(x)c + E(x), \quad d(x) < 0 \qquad \text{at} \;\; z = 0, \tag{4.14}
$$

$$
\kappa c_z = 0, \qquad\qquad\qquad\qquad\;\; \text{at} \;\; z = z_H. \tag{4.15}
$$

The first condition describes the flux κc_z at the earth surface in terms of deposition dc and emission E. The second condition describes a no flux condition at the upper boundary of our domain. Our boundary conditions are chosen in close relation with boundary conditions found in practical applications. κ, d, E and R are assumed 2π-periodic in x, which occurs in true global models if x is associated with the longitudinal direction [13].

We apply Strang splitting to system (4.13) over the interval $[0, \tau]$, which yields

$$
\begin{array}{llll}
\frac{\partial c_1}{\partial t} + u \frac{\partial c_1}{\partial x} & = & 0, & c_1\,(x, z, 0) & = & c(x, z, 0) & (a) \\[2mm]
\frac{\partial c_2}{\partial t} & = & \frac{\partial}{\partial z}\left(\kappa \frac{\partial c_2}{\partial z}\right) \;\; + \text{b.c.} & c_2\,(x, z, 0) & = & c_1(x, z, \frac{\tau}{2}) & (b) \\[2mm]
\frac{\partial c_3}{\partial t} & = & R\,(c_3) & c_3\,(x, z, 0) & = & c_2(x, z, \frac{\tau}{2}) & (c) \\[2mm]
\frac{\partial c_4}{\partial t} & = & \frac{\partial}{\partial z}\left(\kappa \frac{\partial c_4}{\partial z}\right) \;\; + \text{b.c.} & c_4\,(x, z, \frac{\tau}{2}) & = & c_3(x, z, \tau) & (d) \\[2mm]
\frac{\partial c_5}{\partial t} + u \frac{\partial c_5}{\partial x} & = & 0, & c_5\,(x, z, \frac{\tau}{2}) & = & c_4(x, z, \tau) & (e),
\end{array}
$$

$$(4.16)$$

where the initial value $c(x, z, 0)$ in (4.16) satisfies the boundary conditions. Note that the boundary conditions are prescribed in step (4.16b) and (4.16d), so the solutions $c_2(x, z, \frac{\tau}{2})$ and $c_4(x, z, \tau)$ always satisfy the given conditions.

Consider the initial value for step (4.16b) delivered after exact time and space integration of step (4.16a),

$$c_2(x, z, 0) = c_1(x, z, \frac{\tau}{2}) = c(x - u\frac{\tau}{2}, z, 0).$$

If $u_z \neq 0$ for $z = 0$ and $z = z_H$, then at time $t = 0$ in step (4.16b) the boundary conditions (4.14) and (4.15) are not met, as can be seen from

$$\frac{\partial}{\partial z}(c_2(x, z, 0)) = c_z(x - u\frac{\tau}{2}, z, 0) - u_z\frac{\tau}{2}c_x(x - u\frac{\tau}{2}, z, 0). \tag{4.17}$$

The initial value for step (4.16b) is inconsistent with the boundary conditions prescribed in this step. Numerical errors will exist if we don't choose the time step τ large enough to damp out the initial error due to this inconsistency. Note however that at the end of step (4.16b) the boundary conditions are always met.

Now take $u_z = 0$, then $c_{2_z}(x, z, 0) = 0$ holds when $c_z(x - u\frac{\tau}{2}) = 0$ as can be concluded from (4.17). At a large distance from the earth surface $u_z = 0$ is likely to happen, thus no boundary condition inconsistency will exist at $z = z_H$, when z_H is chosen large enough. However, at the earth surface we must satisfy

$$\kappa(x, 0)c_{2_z}(x, 0, 0) = d(x)c_{2_z}(x, 0, 0) + E(x), \tag{4.18}$$

or, inserting (4.17) into (4.18), where still $u_z = 0$, we must satisfy

$$\kappa(x, 0)c_z\left(x - u\frac{\tau}{2}, 0, 0\right) = d(x)c\left(x - u\frac{\tau}{2}, 0, 0\right) + E(x).$$

In general this relation will only hold if κ, d and E are independent of x.

Similarly we can show that in general the solution of the chemistry step (4.16c) used as initial value in step (4.16d) introduces an inconsistency with the prescribed boundary conditions in this step. If at $z = z_H$

$$c_z = 0 \quad \text{and} \quad R_z(x, z, c) = 0, \tag{4.19}$$

no inconsistency is obtained, because the solution of step (4.16c) satisfies

$$\frac{\partial c_3}{\partial t \partial z} = R'(x, z, c_3)\frac{\partial c_3}{\partial z} + R_z(x, z, c_3).$$

For $z = z_H$ large enough, the assumptions (4.19) represent the realistic case. On the earth surface, however, we expect an inconsistency, for $R_z(x, z, c) = 0$ and also $c_z = 0$ may be violated there. Further, it is possible that due to the prescribed emission and deposition condition (4.14) in step (4.16b) strong transient exists, which can lead to a disturbance from the chemical equilibrium solution.

In [13] a comparison was made between solving the 3D problem (4.1) with a Rosenbrock method in combination with approximate factorization, and with the Strang splitting method. Approximate factorization can be seen as a form of splitting performed at the numerical algebra level rather than at the operator level as is done in Strang splitting. As boundary conditions were used

$$\kappa c_z = 0, \quad \text{at } z = 0 \text{ and } z = z_H,$$

while for the imposed windfield, $u_z = v_z = 0$. In [13] was argued that due to this form of splitting at the numerical algebra level, operator splitting errors as well as errors, arising from inconsistencies between the boundary conditions and the initial values prescribed in the intermediate steps in

Strang splitting, could be avoided. This should lead to more accurate solutions in favor of the Rosenbrock method with approximate factorization. Results proved them right, but the gain in accuracy was not as great as was expected. However, the results in [13] might have been too positive where the Strang splitting method was concerned. The specific choice of the boundary conditions led to no inconsistencies, while also the property $u_z = v_z = 0$ contributed to reduction of the splitting error between advection and diffusion. In other words, in a more realistic situation, where boundary conditions such as (4.14) and (4.15) can occur, the Rosenbrock method with approximate factorization might be a good alternative to Strang splitting. Future research has to throw light on this aspect.

5. Conclusions

In this paper we focussed on operator splitting, where we mainly restricted ourselves to three-term symmetrical Strang splitting primarily applied to time-dependent advection-diffusion-reaction (ADR) problems. For pure initial value problems the Lie operator formalism proves to be very useful to derive the structure of the splitting error. Through the notion of commutativity we are able to state in which cases the usage of Strang splitting leads to no splitting error. Application of a three-term symmetrical Strang splitting to pure initial value problems of the ADR-type leads to no splitting error between advection, diffusion and chemistry, when, with exact integration of the intermediate steps in the Strang splitting, the chemistry $R(c)$ is linear in c, and the windfield $\underline{u}$, the diffusion coefficient matrix K and R are independent of the spatial variable $\underline{x}$.

However, in most applications splitting errors will occur. By relating the physics of the problem with the commutators, we have conjectured that in air pollution models the splitting error will oscillate with the diurnal cycle and will not grow beyond bound for evolving time. Unfortunately, the splitting error expression is too complicated for real insight into its actual magnitude.

To avoid or reduce the splitting error several techniques can be applied. One concerns problems of the form (4.1), where diffusion and chemistry can be solved coupled, so only a 1D diffusion-reaction system has to be solved for every vertical column in 3D, avoiding an error due to splitting diffusion and chemistry. Secondly, for problems of the form (4.10) an alternative splitting technique exists, similar to a semi-Lagrangian method. A chemistry step is integrated along the characteristics proceeded by an advection step on an Eulerian grid, leaving no splitting error if the advection step is solved exactly and else resulting in an error similar to the interpolation errors of the semi-Lagrangian method.

Several questions concerning operator splitting remain. A good start for further research is the analysis of the splitting error in practical situations by using global Richardson extrapolation to estimate the splitting error for evolving time.

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