

Solvable model for chemical oscillations

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Abstract The Lotka–Volterra equation, proposed first with two variables by A. J. Lotka, underpins the well-known classic model for chemical oscillations. The general solutions of the Lotka–Volterra equation, with n variables, however, remain unknown. We describe a solvable nonlinear model and general solution, previously unstudied for chemical oscillations, that is analogous to the Lotka–Volterra equations with n variables. This model approximates the Lotka–Volterra equations in the neighbourhood of an equilibrium point and is solvable because it can be shown to be linearized to a set of first-order linear differential equations. The purpose of this report is a description of the general solution of the model.

Keywords Solvable model · Chemical oscillation · Lotka–Volterra equation · Nonlinear differential equation

Mathematics Subject Classification 80A30 · 92E20

1 Introduction

The Lotka–Volterra equation is used in a classic well-known model to describe chemical oscillations [1]. It was initially applied with two variables to a chemically oscillating system, and then generalized to a system with n variables. It has a number of important

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inherent nonlinear properties. Much literature has been published over the intervening years covering aspects such as dynamical invariants [1], heteroclinic cycle [2], strange attractors [3], analytic solutions with two variables [4,5], factorized canonical forms [6], diffusion and stability [7–10], front and travelling waves [11,12], turbulence [13]. Because the masses of interrelated biological species are often seen in nature to oscillate, the Lotka–Volterra equation has also become one of the most important models in biological ecology [7,11,14].

A class of two-variable models, with the Lotka–Volterra equations taking the form of ordinary differential equations, includes both the predator–prey and competitive models [1–6]. The predator–prey model oscillates whereas the competitive model does not. Models suitable for describing chemical oscillations are thus the predator–prey models, which have two equilibrium points: one is the trivial point (0, 0); the other is the so-called centre, which is defined as a point that has a neighbourhood in which any orbit returns exactly to its initial point in only one cycle around its centre. In other words, all orbits are periodic and the system in the neighbour of this centre circulates indefinitely. This behaviour underpins chemical oscillations. In a similar manner, the Lotka–Volterra equations described as both a pair of first-order differential equations and reaction–diffusion concentrations, have been investigated intensely [7–14]. In an oscillating model of reaction–diffusion, chemical substances react and oscillate, and then spatially diffuse along concentration gradients of the chemical substances. Because of the nonlinearity of the Lotka–Volterra equations, no explicit solutions are known for the Lotka–Volterra models of ordinary differential equations with n variables, or more precisely, a general solution expressed explicitly in terms of elementary functions of time are unknown.

We describe here a solvable nonlinear model, which has not been previously investigated for chemical oscillations, that is analogous to the Lotka–Volterra equations with n variables. The purpose of this study is to describe the general solution of the solvable model and moreover present a numerical example.

2 Solvable model

The solvable model investigated in this study is the following set of ordinary differential equations:

$$\begin{cases} \frac{dX_1}{dt} = X_1 \left(a_{11} \log \frac{X_1}{b_1} + a_{12} \log \frac{X_2}{b_2} + \cdots + a_{1N} \log \frac{X_N}{b_N} \right) \\ \vdots \\ \frac{dX_N}{dt} = X_N \left(a_{N1} \log \frac{X_1}{b_1} + a_{N2} \log \frac{X_2}{b_2} + \cdots + a_{NN} \log \frac{X_N}{b_N} \right) \end{cases}, \quad (1)$$

where X_1, \dots, X_N are concentrations of chemical substances (mol/L), a_{ij} rate constants (s^{-1}), and b_j equilibrium values in concentration of chemical substances, each of which makes the j -th logarithmic term be 0 (mol/L). With a specific set of a_{ij} , the concentrations of this system exhibit chemical oscillations in time.

We demonstrate that Eq. (1) is solvable. If the conditions

$$X_i \neq 0 \quad (i = 1, \dots, N) \quad (2)$$

hold, Eq. (1) can be transformed to

$$\begin{cases} \frac{d \log(X_1/b_1)}{dt} = \frac{1}{X_1/b_1} \frac{d(X_1/b_1)}{dt} = a_{11} \log \frac{X_1}{b_1} + a_{12} \log \frac{X_2}{b_2} + \dots + a_{1N} \log \frac{X_N}{b_N} \\ \vdots \\ \frac{d \log(X_N/b_N)}{dt} = \frac{1}{X_N/b_N} \frac{d(X_N/b_N)}{dt} = a_{N1} \log \frac{X_1}{b_1} + a_{N2} \log \frac{X_2}{b_2} + \dots + a_{NN} \log \frac{X_N}{b_N} \end{cases} \quad (3)$$

Using substitution

$$Y_i = \log \frac{X_i}{b_i} \quad (4)$$

Equation (3) can be expressed as

$$\begin{cases} \frac{dY_1}{dt} = a_{11}Y_1 + a_{12}Y_2 + \dots + a_{1N}Y_N \\ \vdots \\ \frac{dY_N}{dt} = a_{N1}Y_1 + a_{N2}Y_2 + \dots + a_{NN}Y_N \end{cases} \quad (5)$$

This is apparently a set of first-order ordinary linear differential equations. The solution of Eq. (5) is [15] therefore,

$$\vec{Y} = \exp(tA)\vec{Y}_0, \quad (6)$$

where $\vec{Y} = \begin{pmatrix} Y_1(t) \\ \vdots \\ Y_N(t) \end{pmatrix}$, $\vec{Y}_0 = \begin{pmatrix} Y_1(0) \\ \vdots \\ Y_N(0) \end{pmatrix}$, and $A = \begin{pmatrix} a_{11} & \dots & a_{1N} \\ \vdots & \ddots & \vdots \\ a_{N1} & \dots & a_{NN} \end{pmatrix}$. By substituting Eq. (4) for Eq. (6), we obtain

$$\begin{pmatrix} \log \frac{X_1(t)}{b_1} \\ \vdots \\ \log \frac{X_i(t)}{b_i} \\ \vdots \\ \log \frac{X_N(t)}{b_N} \end{pmatrix} = \exp(tA) \begin{pmatrix} \log \frac{X_1(0)}{b_1} \\ \vdots \\ \log \frac{X_N(0)}{b_N} \end{pmatrix} \quad (7)$$

Each i -th equation in Eq. (7) can be apparently transformed, by exponentiating both sides, to yield solution $X_i(t)$ on the left-hand side expressed explicitly in terms of elementary functions of time t on the right-hand side; therefore Eq. (1) is solvable with Eq. (7) as its general solution.

We will use four specific models derived from or related to Eq. (1), more specifically, the following solvable model (SV) with two variables,

$$\begin{cases} \frac{dX_1}{dt} = X_1 \left(a_{11} \log \frac{X_1}{b_1} + a_{12} \log \frac{X_2}{b_2} \right) \\ \frac{dX_2}{dt} = X_2 \left(a_{21} \log \frac{X_1}{b_1} + a_{22} \log \frac{X_2}{b_2} \right) \end{cases} \quad (8)$$

The first model is derived from Eq. (8) by substituting $a_{11} = a_{22} = 0$, $a_{12} = -a_{21} = 0.01$, $b_1 = b_2 = 1$ (SV, 0.01),

$$\begin{cases} \frac{dX_1}{dt} = 0.01X_1 \log X_2 \\ \frac{dX_2}{dt} = -0.01X_2 \log X_1 \end{cases}. \quad (9)$$

The second replaces the off-diagonal A values of Eq. (9) with $a_{12} = -a_{21} = 0.001$ (SV, 0.001),

$$\begin{cases} \frac{dX_1}{dt} = 0.001X_1 \log X_2 \\ \frac{dX_2}{dt} = -0.001X_2 \log X_1 \end{cases}. \quad (10)$$

The third is the Lotka–Volterra (LV) two-variable predator–prey model used for comparison with Eq. (9) (LV, 0.01),

$$\begin{cases} \frac{dX_1}{dt} = 0.01X_1(-1 + X_2) \\ \frac{dX_2}{dt} = 0.01X_2(1 - X_1) \end{cases}. \quad (11)$$

The fourth is another LV model with a smaller coefficient of 0.001 (LV, 0.001),

$$\begin{cases} \frac{dX_1}{dt} = 0.001X_1(-1 + X_2) \\ \frac{dX_2}{dt} = 0.001X_2(1 - X_1) \end{cases}. \quad (12)$$

All these models describe oscillatory behaviour of chemical rates for X_1 and X_2 .

Whereas the LV models Eqs. (11) and (12) are not solvable in a manner that solutions should be explicitly expressed as elementary functions of time [5], the SV models Eqs. (9) and (10) are solvable with solutions Eq. (7). This can be demonstrated as follows. Using Eq. (7), the general solution of Eq. (9) is

$$\begin{aligned} \begin{pmatrix} \log X_1(t) \\ \log X_2(t) \end{pmatrix} &= \exp \left[\begin{pmatrix} 0 & 0.01t \\ -0.01t & 0 \end{pmatrix} \right] \begin{pmatrix} \log X_1(0) \\ \log X_2(0) \end{pmatrix} \\ &= \begin{pmatrix} \cos 0.01t & \sin 0.01t \\ -\sin 0.01t & \cos 0.01t \end{pmatrix} \begin{pmatrix} \log X_1(0) \\ \log X_2(0) \end{pmatrix} \\ &= \begin{pmatrix} \log X_1(0) \cos 0.01t + \log X_2(0) \sin 0.01t \\ \log X_1(0) - \sin 0.01t + \log X_2(0) \cos 0.01t \end{pmatrix}. \end{aligned} \quad (13)$$

Therefore, by exponentiating both sides, we obtain

$$\begin{cases} X_1(t) = X_1(0) \cos 0.01t \cdot X_2(0) \sin 0.01t \\ X_2(t) = X_1(0) - \sin 0.01t \cdot X_2(0) \cos 0.01t \end{cases}, \quad (14)$$

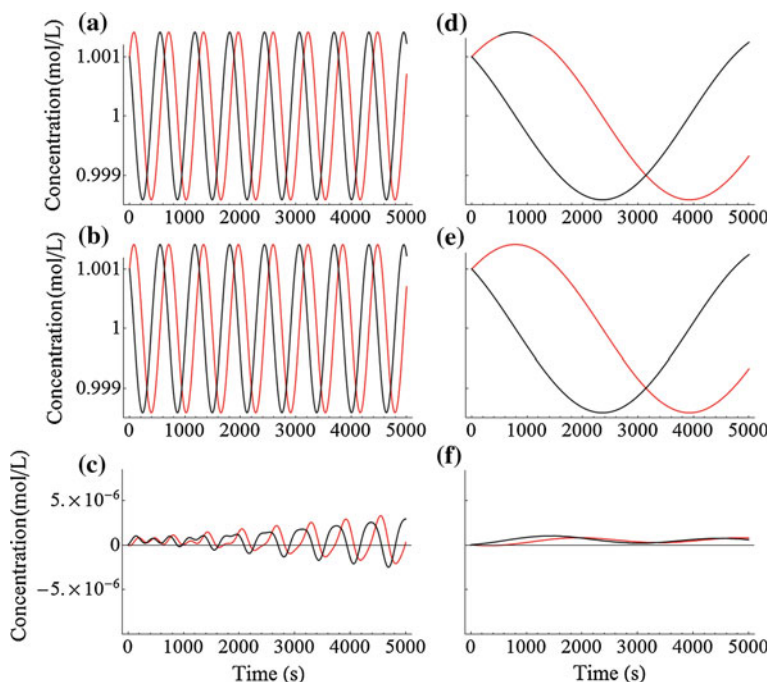


Fig. 1 Simulations for the SV and LV models in the neighbourhood of the equilibrium point (1, 1). The concentrations of X_1 (red) and X_2 (black) (mol/L) versus time (s). **a** The SV model with rate constant 0.01 s^{-1} . **b** The LV model with 0.01 s^{-1} . **c** Error between the SV and LV models. **d–f** Corresponding versions with rate constant 0.001 s^{-1} . NDSolve in Mathematica 8 (<http://www.wolfram.com/mathematica/>) and analytical solutions were used for the LV and SV models, respectively (Color figure online)

which is the general solution of Eq. (9) explicitly expressed in terms of elementary functions of t . Similarly, the general solution of Eq. (10) is

$$\begin{cases} X_1(t) = X_1(0)\cos 0.001t \cdot X_2(0)\sin 0.001t \\ X_2(t) = X_1(0)^{-\sin 0.001t} \cdot X_2(0)\cos 0.001t \end{cases} \quad (15)$$

3 Example: simulation with solvable model of ordinary differential equations

Following the classic Lotka analysis [1], we apply this solvable model to the simulation of chemical oscillations between two chemical substances. This example uses two fixed rate constants, 0.01 and 0.001 s^{-1} , which represent slow but natural reactions. Examples of chemical oscillations for the SV models [Eqs. (9), (10), (14), and (15)] are shown (Fig. 1a, d). Figure 1a presents the solution for the SV model with rate constant 0.01 s^{-1} [Eq. (9) or (14)]. The initial values of X_1 (red) and X_2 (black) are both set to 1.001 mol/L , sufficiently close to the equilibrium point 1 mol/L . The solution exhibits periodical chemical oscillations of X_1 and X_2 for $5,000 \text{ s}$, interacting and keeping a phase. In contrast, Fig. 1b corresponds to a LV model used for comparison [Eq. (11) or (15)] has the same rate constant 0.01 as the SV model; Fig. 1b is almost the same

as Fig. 1a. The LV model is well-known to have an equilibrium point at (1, 1) in the X_1 – X_2 plane with circulating orbits. With an initial point of (1.001, 1.001), which is close enough to (1, 1), an almost simple harmonic oscillation of X_1 and X_2 is expected and observed (Fig. 1b). Figure 1a, b are almost the same and thus the SV model Eq. (9) is expected to approximate the LV model Eq. (11). The errors between the SV (Fig. 1a) and LV (Fig. 1b) models, shown in Fig. 1c, are less than 1/100 of the amplitudes of the oscillations in Fig. 1a, b. This numerical example exhibits the proximity between the SV and LV models given the same rate constant 0.01. Although the errors oscillate and gradually increase, we found that the errors resulted from the difference in algorithms between the SV and LV models. The SV model Eq. (9) was calculated using the analytical solution Eq. (14) with no integration errors (Fig. 1a) whereas the LV model Eq. (11) is a numerical calculation that accumulated integration errors (Fig. 1b). We have made an additional confirmation by calculating the errors between the SV and LV models with the same numerical integration algorithm. Errors remained below 1/1000 of the amplitudes of the oscillations in Fig. 1a, b over 270,000 s (data not shown). No increases in time were observed in Fig. 1c with the errors linearly increasing to roughly 1/10 of the amplitudes of the oscillations in Fig. 1a, b at 270,000 s. An example with rate constant 0.001 (Fig. 1d–f) exhibits slower oscillations in time than that of Fig. 1a–c because the rate constant of this example is 1/10 of the other. In these systems, the frequencies of chemical oscillations thus depend on rate constant. Similar to the previous example, Fig. 1d, e are near equal, and the errors (Fig. 1f) are less than 1/100 of the amplitudes of the oscillations in Fig. 1d, e. Figure 1d–f (rate constant 0.001) are essentially the same as enlarged versions of the region 0–500 s of Fig. 1a–c (rate constant 0.01). In summary, the SV model approximates the LV model within errors of less than 1/100 of the amplitudes given initial conditions (1.001, 1.001), which was near to the equilibrium point, not depending on two examples of the rate constants.

Figure 2 shows global regions of the X_1 – X_2 planes for the SV and LV models. Figure 2a is the X_1 – X_2 planes, or phase plane, for the SV model with rate constant 0.01 [Eq. (9)], Fig. 2b for the corresponding LV model [Eq. (11)] for comparison, Fig. 2c for the SV model with rate constant 0.001 [Eq. (10)], and Fig. 2d for the corresponding LV model [Eq. (12)] for comparison. A phase plane defines an instant evolving direction for the relevant system at a given point, i.e., a pair of concentrations of X_1 and X_2 , in the plane. At each point (X_1 , X_2) in the plane, a vector calculated by the right-hand side of, e.g., Eq. (9) for Fig. 2a is drawn to globally obtain orbits of the system by connecting all the vectors. Note that the region around concentrations (X_1 , X_2) in Fig. 2a–d has been scaled up 5,000 times to that of Fig. 1a–f. As shown in the figure, the SV and LV models have equilibrium points at (1, 1) with orbits circulating around it. Global views and shapes of orbits in Fig. 2 are rather different between the SV and LV models whereas oscillations in Fig. 1 are almost the same. The reason is that in the neighbourhood of the equilibrium point (1, 1) the SV and LV models exhibit small-amplitude oscillations and precision from the approximation of the SV model is good (Fig. 1c, f). In a region distant from the equilibrium point, however, orbits of the SV and LV models are affected by different nonlinearities inherent in the SV and LV models (Fig. 2). In Fig. 2a, c, the SV model, angles of triangle-like orbits in the phase planes are entirely sharp and dented whereas in Fig. 2b, d, the LV model, those are entirely blunt and bulged. This tendency, as shown in the figures, becomes prominent

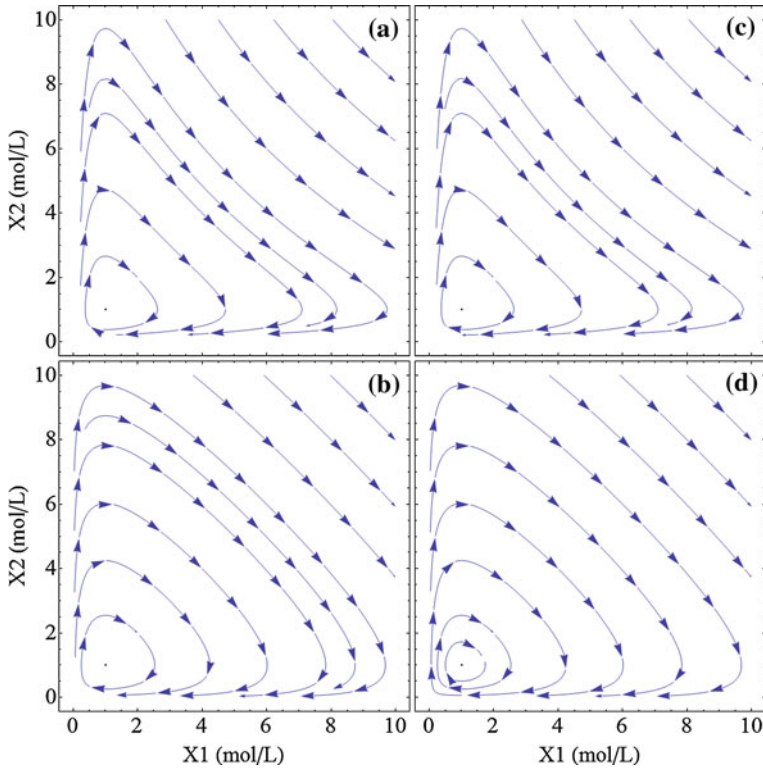


Fig. 2 Global phase planes for the SV and LV models. Concentrations of X_2 versus X_1 (mol/L). **a** The SV model with rate constant 0.01 s^{-1} . **b** The LV model with 0.01 s^{-1} . **c** The SV model with 0.001 s^{-1} . **d** The LV model with 0.001 s^{-1} . The centres are drawn as points at (1, 1). Vectors indicate the direction of evolution of the system; an orbit is formed by connecting vectors. StreamPlot in Mathematica 8 was used

in distant regions from the equilibrium point. Thus, we found that the orbits of the SV model were relatively distorted than those of the LV model.

We can mathematically demonstrate how the SV model can approximate the LV model in the neighbourhood of the equilibrium point (1, 1). A logarithmic function can be expanded as a series,

$$\log X = (X - 1) - \frac{1}{2}(X - 1)^2 + \frac{1}{3}(X - 1)^3 + \dots, \tag{16}$$

around $X = 1$. Replacing the second and higher-order terms in the right-hand side of Eq. (16) by ε , we can obtain

$$\log X = X - 1 + \varepsilon. \tag{17}$$

Supposing $\varepsilon = 0$ in Eq. (17) and then substituting Eq. (17) in the LV models, Eqs. (11) and (12), result in the SV models Eqs. (9) and (10). The solvable model Eq. (1) can be obtained by substituting Eq. (17) with $\varepsilon = 0$ for the Lotka–Volterra equations with n variables. Therefore, the solvable model Eq. (1) can generally approximate

Lotka–Volterra equations with constraint $\varepsilon = 0$, in other words, in the neighbourhood of the equilibrium point. In contrast, from a global view, the orbits of the SV model become distorted (Fig. 2a, c) compared with those of the LV models (Fig. 2b, d) due to the second and higher terms in Eq. (16).

4 Conclusion

A general solution of a solvable model for chemical oscillations that approximates the classic Lotka–Volterra equations with n variables was demonstrated. The general solution of the solvable model with two variables was described using only explicit expressions of elementary functions of time. We demonstrated an application of this model by approximating the Lotka–Volterra equations in the neighborhood of orbit centres.

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References

1. A.J. Lotka, J. Am. Chem. Soc. **42**, 1595 (1920)
2. R.M. May, W.J. Leonard, SIAM J. Appl. Math. **29**, 243 (1975)
3. A. Arneodo et al., J. Math. Biol. **14**, 153 (1982)
4. C.M. Evans, G.L. Findley, J. Math. Chem. **25**, 105 (1999)
5. C.M. Evans, G.L. Findley, J. Math. Chem. **25**, 181 (1999)
6. L. Brenig, Phys. Lett. A **133**, 378 (1988)
7. S.A. Levin, L.A. Segel, Nature **259**, 659 (1976)
8. L.J.S. Allen, Math. Biosci. **65**, 1 (1983)
9. F. Rothe, J. Math. Biol. **3**, 319 (1976)
10. A. Hastings, J. Math. Biol. **6**, 163 (1978)
11. E.E. Holmes et al., Ecology **75**, 17 (1994)
12. P.L. Chow, W.C. Tam, Bull. Math. Biol. **38**, 643 (1976)
13. K. Orihashi, Y. Aizawa, Phys. D **240**, 1853 (2011)
14. J.A. Sherratt et al., Proc. Nat. Acad. Sci. U.S.A. **92**, 2524 (1995)
15. M.W. Hirsch et al., *Differential Equations, Dynamical Systems, and an Introduction to Chaos*, 2nd edn. (Academic Press, San Diego, 2004)