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A DIRECT SEARCH ALGORITHM FOR THE NONLINEAR REGRESSION AND ITS APPLICATION TO SOME CHEMICAL PROBLEMS

(Presented by J. Kann)

Suppose we have a random observable dependent variable Y_i, $i=1, 2, \ldots, n$, and several nonrandom independent (controllable) variables x_k, $k=1, 2, \ldots, q$. Both Y_i and x_k are presumed to be continuous variables, i. e. real numbers in some finite range. Let β_j, $j=1, 2, \ldots, m$ be the parameters in the nonlinear model

$$Y=\eta(x_1, x_2, \ldots, x_q; \beta_1, \beta_2, \ldots, \beta_m)+\varepsilon, \tag{1}$$

where ε is the vector of random errors assuming that the form of model (1) has been chosen properly [1]. The term "nonlinear" means that model (1) suggested to describe the measured quantity Y_i is nonlinear in the parameters to be estimated (β_j). To estimate m unknown parameters (β_j) it is necessary that the number of experiments (values of dependent variable) $n>m$. In each experiment i each of the independent variables x_k, $k=1, 2, \ldots, q$, has the corresponding values $x_{1,i}, x_{2,i}, \ldots, x_{q,i}$. The estimates of β_j are obtained minimizing

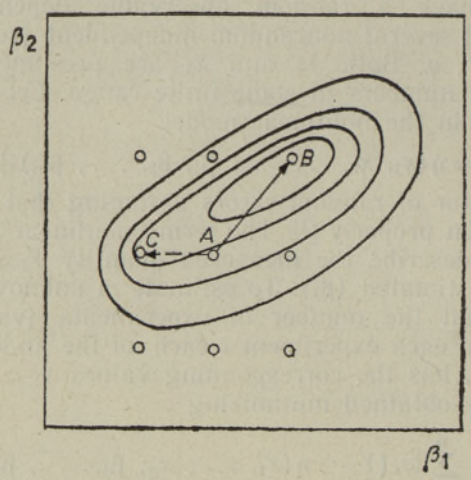
$$\Phi=\sum_{i=1}^{n} w_i(Y_i-\eta(x_1, \ldots, x_q; \beta_1, \ldots, \beta_m))^2, \tag{2}$$

where w_i represents appropriate weights [1].

Direct search methods [2, 3] have some advantages in nonlinear estimation from the viewpoint of the user. No derivatives need to be calculated and the respective computer programs are rather short. For instance, the direct search algorithm [3] may be realized in a FORTRAN IV program of 15 kB ($n=50$, $m=2$, $q=5$). As a result, such a program may be used in a small personal computer which is sufficiently fast and has a limited RAM (64 kB in many cases).

The disadvantage of simple direct search methods [2, 3] is that they deal with each β_j separately, improving the value of one β_j estimation at a time only, keeping all other β_j estimations constant at that period. In this case the iteration may not be optimal and the number of iteration steps is rather large. Now we suggest another iterative algorithm for the estimation of the β_j values using a direct search method: if m is rather large ($m>3$) β_j-s may be divided into groups, 2, 3 or 4 in each, and all the β_j estimates in a group will be improved simultaneously; the β_j estimates not involved in the group considered are kept constant at that time. All the groups of β_j estimates are consequently treated in the same way. In the suggested algorithm the changes in β_j estimates are made in accordance with each other in order to reach the lowest point in the Φ surface available in the step. The procedure consists of modifying the β_j estimates involved in the group to three levels: β_j, $(1+\alpha_j)\beta_j$ and $(1-\alpha_j)\beta_j$ which is followed by calculating the Φ values with all combinations of these modified estimates (see Fig. 1). The individual variation coefficients $\alpha_j<1$, for β_j estimations ensure that all the β_j estimates obtained have the

same sign ("+" or "−") as the initial β_j estimate given by the user. On the other hand, the individual α_j values will reflect the range where β_j is expected to vary. The number of combinations of three-level β_j values is 3^g where g is the number of β_j-s in the group considered. For $g=4$, the number of combinations is 81. This means that the suggested algorithm needs more computer time than simple methods [2, 3] where only one parameter, β_j, is varied at the time. To treat a group of four β_j-s ($g=4$) the computer time is approximately $81/3\times4=6.75$ times larger than in the case of simple methods [2, 3]. But this seems not to be a serious problem now because personal computers are becoming rather fast. It should also be taken into account that in the suggested algorithm the number of iteration steps is less than in simple direct search methods [2, 3] due to more optimal changes in β_j estimates.



Scheme of the Φ (2) surface, giving Φ as a function of two parameters (β_1 and β_2) only. The starting point with the initial β_1 and β_2 estimates is A. After 9 attempts ($g=2$), the lowest available point B on the Φ surface is chosen for the starting point in the next cycle. If the parameters are changed, one at a time, as in simple methods [2, 3], the starting point in the next cycle will not be B but C.

All the Φ values obtained with the 3^g combinations in the group considered are compared to each other and the lowest among them is chosen. The combination of β_j, $(1+\alpha_j)\beta_j$, $(1-\alpha_j)\beta_j$ values corresponding to this lowest Φ is assigned to the respective β_j-s as their new, improved estimates. If the best combination of β_j estimates contains one or more unmodified β_j-s, the respective α_j-s will decrease: $\alpha_j=\alpha_j\times C_1$, $0<C_1<1$, usually $C_1=0.5$). This is done because the α_j used was obviously too large to approach the Φ minimum. After that the next group of β_j estimates is treated in the same way. It should be noted that each group may consist of a different number of β_j-s. When all β_j groups are treated as described above the question about the proper α_j values arises again. In order to settle this question the Φ obtained is compared to the corresponding value before this iteration cycle. If the relative decrease in Φ is less than E, given by the user, a new set of α_j values is calculated:

$$\alpha_j^{(r+1)}=\alpha_j^{(r)}\cdot C_2, \tag{3}$$

where $C_2<1$ ($C_2=0.7$ usually). The new set of $\alpha_j^{(r+1)}$-s is compared to the respective lower limits of A_j-s given by the user. If for some $\alpha_j<A_j$, the respective β_j is further treated as a constant (not changed any more).

The g value for the group containing already some constant β_j is respectively reduced that makes the number of combinations smaller and the program faster. Only after these checks and changes have been made, then, if necessary, the next iteration cycle starts with the improved β_j estimates. When all β_j-s reach the status of constants the iterations will be completed.

The algorithm suggested here has been used in our laboratory in several computer programs (FORTRAN IV) for solving different chemical problems. Among them were:

— deconvulation of Raman spectra into the Voigt profiles;
— description of ions retention times in ion chromatography by nonlinear equations;
— estimation of the basicity constant (pK_{BH^+}), solvation parameter (m^*) and chemical shift values for ionized and unionized forms in the basicity studies employing carbon-13 NMR spectroscopy;
— a more complicated case in the basicity studies of weak bases, involving two protonated forms of the base studied. The parameters estimated were the same as in the above problem but their number was twice as large.

Experience in the application of these computer programs allows us to make two conclusions. First, the convergence to lower Φ-values has always been observed, even if the β_j zero estimates were chosen far from the β_j final values. Secondly, in order to obtain the best fit (i. e. the lowest Φ-value), in some cases several consecutive runs were carried out using the β_j values obtained in k-th run as zero estimates of β_j-s in the next, $(k+1)$-th run.

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MITTELINEAARSE REGRESSIOONI OTSESE ITERATSIOONI ALGORITM JA SELLE KASUTAMINE MÕNEDE KEEMIAÜLESANNETE LAHENDAMISEL

Mittelineaarse regressiooni ülesannete lahendamiseks on esitatud otsene parameetrite täpsustamise algoritm. Korraga toimub mitme parameetri varieerimine. See tagab optimaalsema lahendustee.

Юло ХАЛДНА, Райво ЮГА

АЛГОРИТМ ПРЯМОГО ПОИСКА НЕЛИНЕЙНОЙ РЕГРЕССИИ И ЕГО ПРИМЕНЕНИЕ ДЛЯ РЕШЕНИЯ НЕКОТОРЫХ ХИМИЧЕСКИХ ЗАДАЧ

Предложен алгоритм прямого поиска для решения задач нелинейной регрессии. Алгоритм не требует вычисления частных производных. Улучшение оценок происходит одновременно для нескольких параметров, чем достигается более оптимальный путь решения.