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Importance measures in global sensitivity analysis of nonlinear models

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The present paper deals with a new method of global sensitivity analysis of nonlinear models. This is based on a measure of importance to calculate the fractional contribution of the input parameters to the variance of the model prediction. Measures of importance in sensitivity analysis have been suggested by several authors, whose work is reviewed in this article. More emphasis is given to the developments of sensitivity indices by the Russian mathematician I.M. Sobol'. Given that Sobol' treatment of the measure of importance is the most general, his formalism is employed throughout this paper where conceptual and computational improvements of the method are presented. The computational novelty of this study is the introduction of the 'total effect' parameter index. This index provides a measure of the total effect of a given parameter, including all the possible synergetic terms between that parameter and all the others. Rank transformation of the data is also introduced in order to increase the reproducibility of the method. These methods are tested on a few analytical and computer models. The main conclusion of this work is the identification of a sensitivity analysis methodology which is both flexible, accurate and informative, and which can be achieved at reasonable computational cost. © 1996 Elsevier Science Limited.

1 INTRODUCTION

Sensitivity analysis (SA) of a model output aims to quantify the relative importance of each input model parameter in determining the value of an assigned output variable. Many different methods have been developed for SA, this discipline being very much application driven. The various techniques can be classified in two main branches, depending on the problem setting.

Global SA focuses on the output uncertainty over the entire range of values of the input parameters. Within this setting uncertainty ranges, different in principle for each parameter, are the input for the analysis. These ranges are valuable, they represent our knowledge or lack of it. SA can then help to identify key parameters whose uncertainty affects most the output. This in turn can be used to establish experimental (or field) research priorities, eventually leading to a better definition of the unknown parameter and hence to a reduction of its uncertainty range. The process can be iterated until an acceptable uncertainty range of the output is achieved.[1,3]

In the opposite problem setting the emphasis is on elucidating the key parameters in a complex system, not with respect to the output uncertainty, but with respect to the output itself. In this context, for instance, one may wish to investigate the inter-relationships between system description and different scales. In Rabitz,[4] the sensitivities of macroscopic quantities of a chemical system such as activation energies are investigated with respect to microscopic scale variables, such as the transition probabilities between quantum states for the same system. In this problem setting (*local* SA) one is interested in some kind of derivative (or Jacobian) of the model output with respect to the model input, possibly normalized by the means or standard deviations of the input/output variables themselves. In this context, aiming at the evaluation of the derivatives, model input parameters may be changed by a generally small fraction of their nominal value, the fraction being the same for all the parameters. The input parameter interval thus explored does not represent our uncertainty about that parameter.

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A review of global SA methods, including the Monte Carlo based regression-correlation measures, the Fourier amplitude sensitivity test (FAST) and various forms of differential analysis can be found in Helton,[5] which is also a good pointer to further references. A recent original work in the field of global sensitivity analysis is that of Welch *et al.*[6] where an efficient parameter screening, based on data adaptive modelling, is performed in order to build a computationally cheaper predictor to substitute for the original model. Recent progresses have been made in parameter screening by Andres & Hajas[7] (see also Saltelli *et al.*[8]). Those authors use an iterated fractional factorial design (IFFD), which appears capable of identifying a few active factors in systems with thousands of variable parameters. Another approach to SA not included in Helton[5] is the recent work of Cawlfield & Wu,[9] where global SA is performed within the frame of first order reliability analysis (FORM). SA for stochastic differential equations is discussed in Koda.[10] A comparison of different SA methods can be found in Iman & Helton,[11] Saltelli & Homma[12] and Saltelli *et al.*[13]

In the present work, a particular class of global sensitivity analysis techniques is explored. The 'measures of importance' addressed here are relatively recent in the SA literature, and are based on the partial or conditional variance of the model output, i.e., on a reduction in the variance of the model output corresponding to the 'fixing' of (set of) parameter(s). These conditional variances are usually obtained by averaging over the possible values of the fixed parameter(s). In Hora & Iman,[14] the 'uncertainty importance' of a variable X_j is defined as the expected reduction in the variance of output Y attributable to ascertaining the value of X_j.

$$l_j = \sqrt{\mathrm{Var}[Y] - E[\mathrm{Var}(Y|X_j)]}. \quad (1)$$

For numerical robustness reasons a new statistic is proposed in Iman & Hora,[15] which is based on estimating the quantity:

$$\mathrm{Var}_{X_j}[E(\log Y|X_j)]/\mathrm{Var}[\log Y] \quad (2)$$

where Var_{X_j} stands for variance over all the possible values of X_j and $E[\log Y|X_j]$ is estimated using linear regression. This solution has the advantage of robustness, but—as observed by the authors—the conclusions drawn on log Y are not easily converted back to Y. Similar considerations apply to the rank transformation suggested in this note. A rank transformed version of the importance measure is also discussed in McKay & Beckman.[16]

Other authors[13,17–19] have suggested computational improvements to the importance measure using the Monte Carlo approach. It will be shown that all those measures can be assimilated to Sobol' sensitivity indices of the first order.[20] In turn, Sobol' indices have a strong conceptual similarity with the FAST method.[21–23] The FAST procedure uses a search curve through the parameter space for evaluating the multi-dimensional integral instead of the Monte Carlo technique. Both using FAST and Sobol' series developments, the total variance D of the model output can be written as a sum of terms of increasing dimensionality, the first order terms describing the contribution to the total variance due to each parameter alone, the second order ones describing the contribution due to the two-ways parameter interactions and so on, i.e.:

$$D = \sum_{i=1}^{n} D_i + \sum_{i=1}^{n} \sum_{\substack{j=1 \\ j \neq i}}^{n} D_{ij} + \ldots \quad (3)$$

Somehow different, but still based on the same type of decomposition, is the technique suggested by Sacks *et al.*[24] and Welch *et al.*[6]

It may be worth mentioning that in the FAST applications mentioned above only the first order terms are usually explored, corresponding to that part of the total variance accounted for by each parameter when the output is averaged over the uncertainties in all other parameters (i.e., the D_i terms). The higher order terms are not often computed when using FAST. This is apparently justified when the sum of the first order terms D_i is close enough to the total variance D.[25] More generally, it can be said that higher order terms are very often neglected in SA (see Welch *et al.*[6] for an interesting exception).

In this article much emphasis is placed on the computation of the higher order terms; an amelioration is suggested to the existing version of the Sobol' sensitivity indices, that is based on computing for each parameter the total effect index. This index accounts for all the possible synergetic terms between the given parameters and all the others. Sobol' approach and formalism are described first. Then the global indices are introduced. Those are tested on a number of different test cases, using, in some instances, rank transformation of the input data. The computation of the indices is done by Monte Carlo, and accelerated convergence rates are obtained using quasirandom numbers.[26–28] Finally, the advantages and limitations of this technique are discussed.

2 METHODS

2.1 Mathematical description

A derivation of Sobol' global sensitivity estimates is given in Sobol'.[20] Its essential features are repeated here for the reader's convenience and also because they are needed to discuss the adaptations which have been made for the present work.

Assumption. The function $f(\mathbf{x}) \equiv f(x_1,...,x_n)$ under investigation is defined in the n-dimensional unit cube:

$$K^n = \{\mathbf{x} | 0 \le x_i \le 1; i = 1,...,n\}. \quad (4)$$

Definition. Let $\hat{\sum} T_{i_1...i_s}$ define the sum over all the combinations of indices in K^n

$$\hat{\sum} T_{i_1...i_s} \equiv \sum_{i=1}^{n} T_i + \sum\sum_{1 \le i \le j \le n} T_{ij} + ... + T_{12...n}. \quad (5)$$

Definition. The representation of $f(\mathbf{x})$ as a sum

$$f(x_1,...,x_n) = f_0 + \hat{\sum} f_{i_1...i_s}(x_{i_1},...x_{i_s}) \quad (6)$$

is called a decomposition into summands of different dimensions if

$$f_0 = \text{constant} \quad (7)$$

and the integral of every summand $f_{i_1...i_s}(x_{i_1},...x_{i_s})$ over any of its independent variables is zero, i.e.,

$$\int_0^1 f_{i_1...i_s}(x_{i_1},...x_{i_s}) \mathrm{d}x_{i_k} = 0, \text{ if } 1 \le k \le s. \quad (8)$$

Additional properties of the decomposition eqn (6) which descends from the definitions eqns (6)–(8) are:

Property. The sum in eqn (6) contains a number of summands equal to

$$\sum_{j=1}^{n} \binom{n}{j} = 2^n - 1. \quad (9)$$

Property.

$$f_0 = \int_{K^n} f(\mathbf{x}) \mathrm{d}\mathbf{x}. \quad (10)$$

Property (*orthogonality*). For any two different summands $f_{i_1...i_s}$ *and* $f_{j_1...j_t}$:

$$\int_{K^n} f_{i_1...i_s}(x_{i_1},...x_{i_s}) f_{j_1...j_t}(x_{j_1},...x_{j_t}) \mathrm{d}\mathbf{x} = 0 \quad (11)$$

because of the definition eqn (8), since at least one of the indices $i_1,...,i_s$, $j_1,...,j_t$ will not be repeated twice.

Theorem. The decomposition eqn (6) is unique whenever $f(\mathbf{x})$ is integrable over K^n. The terms in the decomposition can be also derived. f_0 is given by eqn (10). The one-indexed terms $f_i(x_i)$ can be obtained by integrating eqn (6) over all the indices but x_i, and using the definition eqn (8) to obtain:

$$\int_0^1 ... \int_0^1 f(\mathbf{x}) \{\mathrm{d}\mathbf{x}/\mathrm{d}x_i\} = f_0 + f_i(x_i) \quad (12)$$

where $\mathrm{d}\mathbf{x}/\mathrm{d}x_i$ indicates integration over all the variables except x_i. Analogously for the two-indexed summands $f_{ij}(x_i, x_j)$:

$$\int_0^1 ... \int_0^1 f(\mathbf{x}) \{\mathrm{d}\mathbf{x}/\mathrm{d}x_i \mathrm{d}x_j\} = f_0 + f_i(x_i) + f_j(x_j) + f_{ij}(x_i, x_j) \quad (13)$$

and so on for the higher dimension terms. The computation of any summand $f_{i_1...i_s}(x_{i_1},...,x_{i_s})$ is thus reduced to the integration of a multi-dimensional integral within K^n. It is important to stress here that in order to use Sobol' sensitivity indices one does not need to evaluate any of the $f_{i_1...i_s}(x_{i_1},...,x_{i_s})$, nor has one to know the form of $f(\mathbf{x})$, which may well be represented by a 'Computational model',[29] i.e., a function whose value is only obtained as the output of a computer programme.

The sensitivity estimates $S_{i_1...i_s}$ are:

$$S_{i_1...i_s} = \frac{D_{i_1...i_s}}{D} \quad (14)$$

where

$$D = \int_{K^n} f^2(\mathbf{x}) \mathrm{d}\mathbf{x} - f_0^2 \quad (15)$$

and

$$D_{i_1...i_s} = \int_0^1 ... \int_0^1 f^2_{i_1...i_s} \mathrm{d}x_{i_1}...\mathrm{d}x_{i_s}. \quad (16)$$

Squaring eqn (6) and using the orthogonality property eqn (11) it can be proved that

$$D = \hat{\sum} D_{i_1...i_s} \quad (17)$$

using again notation eqn (5) for the sum over the combinations of indices. From eqns (14)–(17):

$$\hat{\sum} S_{i_1...i_s} = 1. \quad (18)$$

It can be observed that D and $D_{i_1...i_s}$ are the variances of $f(\boldsymbol{x})$ and $f_{i_1...i_s}$, respectively. Hence the $S_{i_1...i_s}$ can be considered as true global sensitivity estimates, since they give the fraction of the total variance of $f(\boldsymbol{x})$ which is given by the individual summands in eqn (6). If one of the $S_{i_1...i_s}$ is nil, then the corresponding function $f_{i_1...i_s}$ is zero; if all the $S_{i_1...i_s}$ with $s \ge 2$ are nil then $f(\boldsymbol{x})$ can be expressed as

$$f(x_1,...,x_n) = f_0 + \sum_{i=1}^{n} f_i(x_i) \quad (19)$$

i.e., it is independent from all the cross-products of variables. If $f(\boldsymbol{x})$ is independent from variable x_i then all the $S_{i_1...i_s}$ terms that contain the index i will be nil and so on.

In the preceeding development, the variables x_i, i = 1,...,n, have been independent, meaning that no dependency or correlation exists among them for the system being modeled. The relation of Sobol' indices to FAST is evident. Even using FAST one may obtain eqn (17) above. The Fourier development is in fact also based on an orthogonal set of functions of increasing dimensionality as in eqn (6) (see also discussion in Sobol'[20]).

A similar series development for SA purposes was

also suggested by Sacks *et al.*[24] Those authors suggest the use of the functions $f_{i_1 \ldots i_s}$ themselves as sensitivity 'indicators'. For the first order terms (i.e., the $f_i(x_i)$'s) this implies visual inspection of the $f_i(x_i)$ vs x_i plots. For the second order terms (i.e., the $f_{ij}(x_i, x_j)$'s) a three-dimensional plot must be investigated. The method becomes impractical for higher order terms.

2.2 Monte Carlo computation

The applicability of the sensitivity estimates $S_{i_1 \ldots i_s}$ to a large class of functions $f(\mathbf{x})$ is linked to the possibility of evaluating the multidimensional integral associated with these estimates via Monte Carlo methods. For a given sample size N tending to ∞ the following estimates are straightforward:

$$\hat{f}_0 \doteq \frac{1}{N} \sum_{m=1}^{N} f(\mathbf{x}_m) \tag{20}$$

where $\mathbf{x}_m$ is a sampled point in the space K^n, and the hat is meant to distinguish between a quantity and its estimate. Another natural estimate is:

$$\hat{D} + \hat{f}_0^2 \doteq \frac{1}{N} \sum_{m=1}^{N} f^2(\mathbf{x}_m). \tag{21}$$

For the one-indexed terms S_i, an evaluation for D_i is needed from eqn (16):

$$\begin{aligned} D_i &= \int_0^1 f_i^2(x_i)\mathrm{d}x_i = \int_0^1 \left[\int_0^1 \ldots \int_0^1 f(\mathbf{x})\{\mathrm{d}\mathbf{x}/\mathrm{d}x_i\} - f_0 \right]^2 \mathrm{d}x_i \\ &= f_0^2 - 2f_0 \int f(\mathbf{x})\mathrm{d}\mathbf{x} + \int_0^1 \left[\int_0^1 \ldots \int_0^1 f(\mathbf{x})\{\mathrm{d}\mathbf{x}/\mathrm{d}x_i\} \right. \\ &\quad \left. \times \int_0^1 \ldots \int_0^1 f(\mathbf{x})\{\mathrm{d}\mathbf{x}/\mathrm{d}x_i\} \right] \mathrm{d}x_i \\ &= -f_0^2 + \int_0^1 \ldots \int_0^1 f(\mathbf{u},x_i) f(\mathbf{v},x_i)\mathrm{d}x_i \mathrm{d}\mathbf{u}\mathrm{d}\mathbf{v} \end{aligned} \tag{22}$$

where both $\mathbf{u}$ and $\mathbf{v}$ denote projections of $\mathbf{x}$ on $K^n - 1 = K^n$ minus the variable x_i. The integral has dimension $2(n-1) + 1 = 2n - 1$, and can be estimated via Monte Carlo so that

$$\hat{D}_i + \hat{f}_0^2 \doteq \frac{1}{N} \sum_{m=1}^{N} f(\mathbf{u}_m, x_{im}) f(\mathbf{v}_m, x_{im}). \tag{23}$$

In Monte Carlo terms D_i is thus generated by summing products of two function values: one with all the variables sampled and the other with all the variables re-sampled except the variable x_i.

In this form the Sobol' sensitivity estimate is very close to the importance measures discussed by other investigators.[13,17–19] In other words, the importance measures discussed by those authors are partial variances corresponding to a single parameter effect, i.e., they can be assimilated to sensitivity indices of the first order. The modified importance measure *HIM** discussed in Saltelli *et al.*[13] and Homma & Saltelli[19] is a rank based measure of importance which can be written as

$$HIM_i^* = \frac{\left[\frac{1}{N} \sum_{m=1}^{N} R(f(\mathbf{u}_m, x_{im}))R(f(\mathbf{v}_m, x_{im})) - \left(\frac{N+1}{2} \right)^2 \right]}{\left(\frac{N^2-1}{12} \right)} \tag{24}$$

where $R(f(\mathbf{x}_m))$ is the rank of $f(\mathbf{x}_m)$. In this formulation *HIM** is identical to Sobol's one indexed S_i for the function $f^* =$ 'Rank of $f(\mathbf{x})$', as for this function

$$f_0^* = \left(\frac{N+1}{2} \right) \text{ and } D = \left(\frac{N^2-1}{12} \right). \tag{25}$$

The coincidence in the formulation reached by different investigators, albeit the difference in the use of ranks, is remarkable.

An expression for the Monte Carlo evaluation of the second order terms D_{ij} was not given in Sobol',[20] but can be obtained using both definition eqn (8) and property eqn (11) to yield:

$$\begin{aligned} \hat{D}_{ij} + \hat{D}_i + \hat{D}_j + \hat{f}_0^2 \doteq \frac{1}{N} \sum_{m=1}^{N} & f(\mathbf{r}_m, x_{im}, x_{jm}) \\ & \times f(\mathbf{s}_m, x_{im}, x_{jm}) \end{aligned} \tag{26}$$

where now $\mathbf{r}_m$, $\mathbf{s}_m \in K^{n-2}$. This type of equation lends itself to an intuitive interpretation: the sum $D_{ij} + D_i + D_j$, i.e., the total variance due to variables x_i and x_j (including the cross product term), can be estimated by function values in which all the variables but x_i and x_j are re-sampled. If x_i, x_j are important variables, then high function values will be multiplied by high function values in eqn (26), resulting in large values of the sensitivity estimates.

Expressions analogous to eqn (23) and eqn (26) can be derived for the higher order terms. One important computational aspect linked to the Monte Carlo evaluation of the $\hat{D}i_1 \ldots i_s$ is that they must be derived from the summantions as eqn (23) and eqn (26). In order to describe the approach taken in the present work let the following notation be introduced:

$$\overline{Di_1 \ldots i_s} \doteq \frac{1}{N} \sum_{m=1}^{N} f(\mathbf{u}_m, \mathbf{x}_m) f(\mathbf{v}_m, \mathbf{x}_m) \tag{27}$$

where the vector $\mathbf{x}_m$ contains the variables $x_{i_1}, x_{i_2}, \ldots, x_{i_s}$; it is easy to see that

$$\hat{D}_i = \overline{D_i} - \hat{f}_0^2 \tag{28}$$

$$\begin{aligned} \hat{D}_{ij} &= \overline{D_{ij}} - \hat{D}_i - \hat{D}_j - \hat{f}_0^2 \\ &= \overline{D_{ij}} - \overline{D_i} - \overline{D_j} + \hat{f}_0^2 \end{aligned} \tag{29}$$

and

$$\hat{D}_{ijk} = \overline{D_{ijk}} - \overline{D_{ij}} - \overline{D_{ik}} - \overline{D_{jk}} + \overline{D_i} + \overline{D_j} + \overline{D_k} - \hat{f}_0^2. \tag{30}$$

Those relations can be generalized to

$$\hat{D}_{i_1\ldots i_s} = \overline{D_{i_1\ldots i_s}} - \sum^{>} \overline{D_{i_1\ldots i_{s-1}}} + \sum^{>} \overline{D_{i_1\ldots i_{s-2}}} \ldots (-1)^{s-r} \sum^{>} \overline{D_{i_1\ldots i_r}} \ldots (-1)^s \hat{f}_0^2 \quad (31)$$

where $\sum^{>} \overline{D_{i_1\ldots i_r}}$ indicates the sum over all the permutations (of size r) of the indices contained in $i_1, i_2 \ldots i_s$. Equation (31) allows the $\hat{D}$'s to be computed from the $\bar{D}$'s.

In the calculation of the various terms $\hat{D}_{i_1\ldots i_s}$ in the Monte Carlo scheme it is important that the re-sampled variables are always generated using the same random numbers. For instance, when comparing first order terms like $\hat{D}_i \hat{D}_j$ it is essential that the respective function values in eqn (23) only differ for the sample values of x_i, x_j, i.e., for each m, $m = 1,\ldots,n$, the terms x_{mk}, $k \neq i,j$ must be identical in the two sums for $\hat{D}_i, \hat{D}_j$, otherwise the difference between $\hat{D}_i$, $\hat{D}_j$ could be blurred by the noise associated with the different sampling of the x_{mk}, $k \neq i,j$. Similar considerations apply for the sums for the higher order terms (e.g., eqn (26)). In Sobol's terminology[20,28] the constructive dimension of the Monte Carlo algorithm needed to compute a complete set of $\hat{D}_{i_1\ldots i_s}$ is equal to $2 \times n$, where the constructive dimension equals the total number of random numbers which must be generated to evaluate all the random variables needed for a single trial. This is easily implemented, in practice, by generating a random numbers matrix of size (N, $2 \times n$). Then when computing, for instance, eqn (23) and eqn (26), the $\mathbf{u}_m$, $\mathbf{r}_m$, x_{im}, x_{jm} terms are generated using the first n columns of the random matrix; the $\mathbf{v}_m$, $\mathbf{s}_m$ terms (the variables to be re-sampled) are computed using the last n columns of the same matrix. If a reduced set of $D_{i_1\ldots i_s}$ are to be computed, then the constructive dimension is equal to $2 \times n$ minus the total number of variables which are never re-sampled in any of the requested $D_{i_1\ldots i_s}$.

2.3 Random points generation

The random data matrix can either be generated using crude Monte Carlo or some form of stratified sampling, such as for example the Latin hypercube sampling (LHS) in McKay *et al.*[30] Whenever possible, Sobol' LP_τ number sequences have been used in this work.[26,27] The performances of various sampling strategies for computing importance measures was investigated in a previous article,[19] where LP_τ sequences were found to perform better than both crude random sampling and LHS. It should be mentioned that LHS needed for computing the measures is perturbed, as two LHS matrices of row dimension n are used, and the partial variances are computed from columns of both matrices as outlined in Section 2.2. The estimates of the sensitivity indices are in fact multi-dimensional integrals, and the good performance of quasirandom sequences for this kind of numerical integration is known.[31]

As discussed in Sobol'[28] quasirandom numbers are characterized by an enhanced convergence, i.e., the $N^{-1/2}$ statistic convergence rate of the crude Monte Carlo can—in some cases and depending on the nature of the function under investigation—become as large as $N^{-1+\epsilon}$ with an arbitrary small $\epsilon > 0$. Subroutines to generate LP_τ sequences are available.[32,33] Unfortunately, convenient computational formulae are only available if the row dimension of the matrix to be generated is ≤ 51.[20] As discussed in the previous section the constructive dimension needed to compute the $D_{i_1\ldots i_s}$ is usually $2 \times n$, and this may limit the application of LP_τ algorithms when the number of independent variables is large. In the results section both crude Monte Carlo and Sobol' quasirandom numbers have been used, depending on the number of variables in the test case.

2.4 Error estimates

The quantities involved in the evaluation of the $S_{i_1\ldots i_s}$ can be regarded as 'means' of a given function; f_0 is the mean of $f(\mathbf{x})$ in eqn (20), $D + f_0^2$ is the mean of $f^2(\mathbf{x}_m)$ in eqn (21) and so on. Consequently

$$STD(f_0) = \frac{STD(f(\mathbf{x}))}{\sqrt{N}} = \frac{1}{\sqrt{N}} \sqrt{\frac{1}{N} \sum_{m=1}^{N} f^2(\mathbf{x}_m) - f_0^2} \quad (32)$$

where STD stands for standard error. Sobol' suggests the use of the probable error δ corresponding to the crude Monte Carlo method computed as

$$\delta f_0 = 0.6745 \times STD(f_0) \quad (33)$$

with the population f_0 having a 50% chance of falling in the interval $f_0 \pm \delta f_0$.[28] Analogously, the probable error on $\overline{D_{i_1\ldots i_s}}$ in eqn (27) is estimated as:

$$\overline{\delta D_{i_1\ldots i_s}} \doteq \frac{0.6745}{\sqrt{N}} \sqrt{F - I^2} \quad (34)$$

where

$$I \doteq \frac{1}{N} \sum_{m=1}^{N} f(\mathbf{u}_m, \mathbf{x}_m) f(\mathbf{v}_m, \mathbf{x}_m) \quad (35)$$

$$F \doteq \frac{1}{N} \sum_{m=1}^{N} [f(\mathbf{u}_m, \mathbf{x}_m) f(\mathbf{v}_m, \mathbf{x}_m)]^2. \quad (36)$$

As shown previously, $\hat{D}_{i_1\ldots i_s}$ can be expressed as a linear combination of terms $\overline{D_{i_1\ldots i_s}}$ and f_0^2. Thus the probable error on the $S_{i_1\ldots i_s}$ can be determined. In the

results section we have approximately computed the probable error as:

$$\delta\hat{S}_i \approx \frac{\delta\hat{D}_i}{\hat{D}} + \frac{\hat{D}_i \delta\hat{D}}{\hat{D}^2} \tag{37}$$

where the first order term has been taken as example and the errors on $\hat{D}_i$ and $\hat{D}$ are the probable errors. In this formula $\hat{f}_0^2$ and its error were neglected. This is justified since the problem is usually scaled before computing the variances, so that $\hat{f}_0^2$ is small. The probable error for the higher order terms can be also estimated from eqn (37) applying the maximum error propagation formula. The actual error, however, is likely to be much lower due to the opposite sign of the terms in eqn (31). Hence $\delta\overline{D_{i_1\ldots i_s}}$ in eqn (34) will be used as a yardstick for our computations instead of using $\delta\hat{D}_{i_1\ldots i_s}$ in the results section.

By testing $S_{i_1\ldots i_s}$ computations on a set of analytical functions given in Sobol'[20] it was seen that the results were affected by a systematic error which could be compensated for. Let x_i be a non influential variable and S_i be its first order sensitivity. According to the theory it should be $S_i = 0$. In fact

$$\hat{D}_i = \overline{D_i} - f_0^2 = \frac{1}{N}\sum_{m=1}^{N} f(\mathbf{u}_m, x_{im}) f(\mathbf{v}_m, x_{im}) - f_0^2. \tag{38}$$

If the two function values are completely uncorrelated, as should be the case for non influential parameters, then the summation in eqn (38) can be rewritten as

$$\frac{1}{N} f_0 \sum_{m=1}^{N} f(\mathbf{v}_m, x_{im}) = \frac{1}{N} f_0 N f_0 = f_0^2 \tag{39}$$

so that D_i vanishes. In practice this does not happen, due to the finite sample size employed in the estimation. The residual value of D_i can never be lower than

$$\frac{1}{N}\sum_{m=1}^{N} f(\mathbf{u}_m) f(\mathbf{v}_m) - f_0^2 \tag{40}$$

where a constructive dimension equal to $2 \times n$ has been assumed, and $f(\mathbf{u}_m)$, $f(\mathbf{v}_m)$ indicate evaluation of function values for the two sets of n columns of the input sample matrix. The summation in eqn (40) corresponds to the case in which all the variables are re-sampled. The correction term of eqn (40) applies to all the terms $D_{i_1\ldots i_s}$.

This is better illustrated by an example. The product of linear functions:[20]

$$f = \frac{(2x_1 + 1)\ldots(2x_n + 1)}{2^n} \tag{41}$$

has variances

$$D_{i_1\ldots i_s} = (12)^{-s} \text{ and } D = (13/12)^n - 1. \tag{42}$$

Results for the evaluation of the $S_{i_1\ldots i_s}$ for $n = 5$ and N (sample size) equal to 1024 are given in Table 1. Parameter number 6 is a dummy, with no influence on the output. The left hand side values were obtained using eqn (31), while the correction term eqn (40) was used for the right hand side values. Those results can be compared with the theoretical values

$$D_i = 0.083,\ D_{ij} = 0.0069,\ D_{ijk} = 0.00058,$$
$$D_{ijkl} = 4.8\,10^{-5},\ldots D = 0.492 \tag{43}$$
$$S_i = 0.17,\ S_{ij} = .014,\ S_{ijk} = 0.0012,\ S_{ijkl} = 9.8\,10^{-5},\ldots$$

As can be seen from the left hand side column in Table 1, a systematic error of about -0.004 is subtracted from the first order terms, added to the second order ones, subtracted from the third order ones and so on. This is due to the fact that each $D_{i_1\ldots i_s}$ is equal to the sum of an odd number of $\overline{D_{i_1\ldots i_s}}$ terms with alternate signs in eqn (31), and that the error is the same for each $\overline{D_{i_1\ldots i_s}}$. The right hand side in Table 1 was obtained correcting each term with eqn (40) multiplied by $(-1)^{s+1}$.

2.5 A new statistics: the global sensitivity indices

As mentioned in the introduction, the emphasis of the present note is on the computation of the higher order sensitivity indices, because of their relevance to nonlinear models. Unfortunately, one separate sample (of size N) is needed to compute each of the $S_{i_1\ldots i_s}$. Given that the number of terms in the development of eqn (6) and in eqn (17) as well is $2^n - 1$, and that one sample is needed for f_0, then Nx(2^n) model evaluations are to be computed. In applications with a large number of variables this number would be prohibitive.

For this reason all the variables have been partitioned into two subsets, one containing a given variable X_i alone, and the complementary set $\mathbf{X}_{ci}$ containing all the X_j with $j \neq i$. In this case the decomposition of $f(x)$ will turn into:

$$f(x) = f_0 + f_i(X_i) + f_{ci}(\mathbf{X}_{ci}) + f_{i,ci}(X_i, \mathbf{X}_{ci}). \tag{44}$$

Therefore, using the definitions eqn (15) and eqn (16) the total variance D can be given as:

$$D = D_i + D_{ci} + D_{i,ci}. \tag{45}$$

At this point the new statistics S_{Ti} can be introduced:

$$S_{Ti} \equiv S_i + S_{i,ci} = 1 - S_{ci} \tag{46}$$

where S_{ci} equals the sum of all the $S_{i_1\ldots i_s}$ terms where the index i is excluded. Consequently, S_{Ti} denotes the 'total' effect of variable X_i, which includes the fraction of variance accounted for by variable X_i alone and the fraction accounted for by any combination of X_i with

Table 1. Values of $S_{i_1,\ldots,i_s}$ for the function eqn(41) without (left) and with (right) the correction term eqn(40). $n = 5$ and sample size $N = 1024$. Parameter 6 is a dummy

$S_{i_1,\ldots,i_s}$ without correction term		$S_{i_1,\ldots,i_s}$ with correction term	
1	0.170579	1	0.174747
2	0.169207	2	0.173375
3	0.160731	3	0.164899
4	0.160707	4	0.164875
5	0.166913	5	0.171080
average of 1-5 = 0.1656		average of 1-5 = 0.1698	
6	-0.004167	6	0.000000
12	0.017214	12	0.013046
13	0.016702	13	0.012534
14	0.019198	14	0.015030
15	0.016804	15	0.012636
16	0.004167	16	-0.000000
23	0.020892	23	0.016724.
...		...	
average of second order terms not including 6 = 0.0187		average of second order terms not including 6 = 0.0142	
1 2 3	-0.003855	123	0.000312
1 2 4	-0.003037	124	0.001130
1 2 5	-0.003589	125	0.000578
1 2 6	-0.004167	126	0.000000.
...		...	
average of third order terms not including 6 = -0.00339		average of third order terms not including 6 = 0.00078	
1234	0.005261	1234	0.001093
1235	0.005228	1235	0.001060
1236	0.004167	1236	-0.000000
1245	0.004203	1245	0.000036.
...		...	
average of fourth order terms not including 6 = 0.00251		average of fourth order terms not including 6 = 0.00085	
12345	-0.003429	12345	0.000738
12346	-0.004167	12346	0.000000
12356	-0.004167	12356	0.000000
12456	-0.004167	12456	0.000000.
...		...	

the remaining variables. S_{ci} can be estimated with just one Monte Carlo integral as:

$$S_{ci} = \frac{1}{D}\frac{1}{N}\sum_{m=1}^{N} f(x_{im}, \mathbf{u_m}) f(x'_{im}, \mathbf{u_m}) - f_0^2 \qquad (47)$$

where $\mathbf{u}$ is the vector of all the variables but x_i and the prime indicates re-sampling. This approach reduces the number of model evaluations to $N \times (n + 1)$ i.e., one sample for f_0 plus one sample for each variable.

In the statistical literature this kind of parameter screening procedure, where the number of model simulations to be performed grows with the number of parameters, is commonly indicated as a OAT method, from one-factor-at-a-time. A textbook on experimental design[34] contrasts OAT with factorial design, the latter being indicated as a more appropriate technique to uncover two or three way effects (i.e., higher order terms). This kind of criticism does not hold for the approach presented in this note. An interesting example of an OAT method contrasted to LHS is given in Morris.[29] This author suggests an OAT factorial sampling plan to be used for preliminary parameter screening in local SA. The screening is aimed at distinguishing between (a) non influential parameter, (b) parameters influencing the output linearly and (c) parameters influencing the output non linearly or via higher order effect (association with other parameters). In a worked example with a nonlinear model and $n = 20$ input parameters Morris achieves the screening with just 84 runs.

Another example of OAT is given in Cotter,[35] using a two level factorial design. In Cotter's approach, as in the present note, the total effect of a parameter is considered to be due to a sum of terms of increasing

dimensionality, including a first order effect, a sum of second order terms, a sum of third order ones and so on. Using $(2n + 2)$ runs, Cotter can estimate for each parameter, the total algebraic sum of the 'even order' effects and the total algebraic sum of the 'odd order' effects. Parameters are ranked based on these sums. A shortcoming of this approach is a possible cancellation within the sums of terms of opposite sign.

Both Morris' and Cotter's approach can be considered as screening tests conducted at small sample size. The total sensitivity indices S_{Ti} suggested here are indeed 'large sample' methods. It may be worth stressing that this may render the approach non viable for applications with large numbers of parameters and computationally expensive models (with the 'too expensive' boundary moving at each new generation of workstations). On the other hand, S_{Ti} provides an information qualitatively superior to that of other global methods. A regression based SA, for instance, provides coefficients which may be used for assessing the importance of a given parameter, conditionally upon the efficiency of the regression model (i.e., on R^2_y). Those coefficients can be considered as clues of parameter influence. The sensitivity indices provide the actual fraction of variance accounted by each (combination of) parameter(s). S_{Ti} gives the total fraction of such a variance due to interactions of any order. This kind of information is more precise from the mathematical point of view and more informative of the model behaviour.

2.6 Rank based version of importance measures

In the results section data are presented for S_{Ti} and S_i, and for the rank based versions of the statistics, indicated by S^*_{Ti} and S^*_i. Rank transformation is a fairly common procedure in sensitivity analysis of a nonlinear model. For example, the standardized regression coefficients (SRC) are often replaced by their rank equivalent (the standardized rank regression coefficients SRRC)[36] when the regression based on the SRC's is poor. Nevertheless, the use of rank with the importance measure discussed here is conceptually different from the use of rank in a regression based sensitivity analysis technique.

As far as the regression technique (i.e., the SRRC) is concerned, the rank transformation is essential to the analysis, in that it allows the detection of parameters non linearly correlated with the output and which could otherwise be overlooked. In other words a sensitivity analysis based on the linear version of the technique (i.e., on the SRC) would be regarded with suspicion, unless the model under analysis were proven to be almost linear.

In the same spirit the use of the importance measure finds its justification in its ability to detect nonlinear *and* nonmonotonic relationships, which could escape detection even using the SRRC's.[12,13] Nevertheless, the importance measure could, in principle, yield the same result even without the rank transformation. The reason of the transformation lies, in this case, in the scarce robustness of the raw values version of the statistics.[13] Robustness means here the ability of the method to replicate its results with different input samples taken from the same population. This happens because, normally, the error associated with S_{Ti} is much larger than that associated with S^*_{Ti}. This implies that in order to achieve a given (target) probable error, a larger sample is needed for S_{Ti} than for S^*_{Ti} (the error in both cases decreases as $\frac{1}{\sqrt{N}}$). As mentioned in the introduction the scarce robustness of the raw value-based measure is discussed in Iman & Hora,[15] who note that the measure is highly influenced by outliers associated with long tailed input distributions.

3 RESULTS

3.1 First test case: analytical functions

To demonstrate the performance of the importance measures introduced, an artificial analytical model with 3 input variables is considered, after Ishigami & Homma[18]:

$$f(X_1,X_2,X_3) = \sin X_1 + a \sin^2 X_2 + bX_3^4 \sin X_1 \quad (48)$$

where its input probability density functions (pdf) are assumed as follows:

$$p_i(X_i) = \begin{cases} \dfrac{1}{2\pi}, \text{ when } -\pi \le X_i \le \pi \\ 0, \text{ when } X_i < -\pi, X_i > \pi \end{cases} \text{ for } i = 1,2,3. \quad (49)$$

From eqn (15) and eqn (16) the total variance D and partial variances $D_{i_1 \ldots i_s}$ can be obtained analytically as:

$$D = \frac{a^2}{8} + \frac{b\pi^4}{5} + \frac{b^2\pi^8}{18} + \frac{1}{2} \quad (50)$$

$$D_1 = \frac{b\pi^4}{5} + \frac{b^2\pi^8}{50} + \frac{1}{2} \quad (51)$$

$$D_2 = \frac{a^2}{8} \quad (52)$$

$$D_3 = 0 \quad (53)$$

$$D_{12} = 0 \quad (54)$$

$$D_{13} = \frac{b^2\pi^8}{18} - \frac{b^2\pi^8}{50} \quad (55)$$

$$D_{23} = 0 \quad (56)$$

$$D_{123} = 0. \quad (57)$$

The same functions were also used to investigate the effect of the sampling strategy.[19]

The primary purpose of this test case is to examine how the Monte Carlo computation of the sensitivity estimates works with the comparison to the exact values. A Sobol' quasirandom input sequence of constructive dimension equal to 2×3 has been generated for the 3 input parameters. A base sample of size 1024 was used. The D_{123} term was estimated as $D_{123} = D - \hat{\sum} D_{12}$. The results of $S_{i_1 \ldots i_s}$, $\hat{S}_{i_1 \ldots i_s}$ and $\hat{S}^* i_1 \ldots i_s$ are given in Table 2. The error terms in Table 2 were estimated with eqn (37). The constants in eqns (50)–(57) are given the values $a = 7$ and $b = 0.1$ in the present exercise.

The following remarks can be made:

- The values of $\hat{S}_{i_1 \ldots i_s}$ show good agreements with the exact values.
- The effect of the rank is to decrease the relative influence of the higher order term ($\hat{S}^*_{1,3} < \hat{S}_{1,3}$).

In particular, it is interesting that the sensitivity estimate related to combination of the variables X_1 and X_3 is non negligible, although the partial variance D_3 is negligible. This is an example of how the ranking of input parameters based on first order terms could give unreliable results.

For comparison the results of SRC and the SRRC are also presented in Table 3 with the model coefficients of determination R_y^2. As can be expected from the analytical form of eqn (48), strong nonlinear and nonmonotonic relationships between the input variables and the output result in a poor performance of the regression method, both using raw values and rank-transformed data. The resulting model coefficients of determination R_y^2 are very low. Hence the SRC and SRRC cannot provide a reliable ranking of input variables. The results from the SRRC contrast sharply with the predictions from S_{Ti} in Table 4. According to the SRRC's X_1 is ten times more important than either X_2 or X_3. This is not to be believed, due to the combined evidence of the S_{Ti} and the low R_y^2 associated with the SRRC. The computed and analytical values of S_{Ti} are in good agreement with the exact values.

Table 2. $S_{i_1 \ldots i_s}$, $\hat{S}_{i_1 \ldots i_s}$, and $\hat{S}^*_{i_1 \ldots i_s}$ values with error terms for the first test case

Variables	$S_{i_1 \ldots i_s}$, exact	$\hat{S}_{i_1 \ldots i_s}$ (error term) calculated	$\hat{S}^*_{i_1 \ldots i_s}$ (error term)
X_1	0.3138	0.3230(0.032)	0.3115(0.026)
X_2	0.4424	0.4390(0.034)	0.5106(0.029)
X_3	0.0	0.0078(0.029)	0.0077(0.023)
X_1X_2	0.0	0.(0.048)	0.0034(0.034)
X_1X_3	0.2436	0.2354(0.049)	0.1311(0.030)
X_2X_3	0.0	0.(0.043)	0.0190(0.031)
$X_1X_2X_3$	0.0		

Table 3. Standardized regression coefficients (SRC) and standardized rank regression coefficients (SRRC) with model coefficients of determination (R_y^2)

Variable	SRC	SRRC
$X1$	0.44	0.43
$X2$	−0.03	−0.03
$X3$	0.02	0.04
R_y^2	0.20	0.18

3.2 Second test case: KIM

KIM is a chemical kinetics model for the OH initiated oxidation of dimethylsulphide (DMS), a sulphur bearing compound which is naturally produced by oceanic biota over remote areas (see scheme in Fig. 1). The reactions considered are given in Table 5. KIM solves a system of 37 differential equations. Switches in KIM allow the non irreversible reactions, e.g.,

$$CH_3S + O_2 \underset{k_{-7}}{\overset{k_7}{\rightleftharpoons}} CH_3SOO$$

to be considered either kinetically (the results depend on both k_7 and k_{-7}) or at the equilibrium (the results only depend on the ratio k_7/k_{-7}). In the present analysis the equations involving k_{14} and k_{19} are considered at the equilibrium. The integration of the system equations is performed using Gear's method.[37] KIM has 37 input parameters (constructive dimension = 2×37) which are either kinetic constants, activation energies, or initial values for species concentration. Parameter distributions are given in Saltelli & Hjorth.[38]

A thorough sensitivity analysis of this model is reported elsewhere[38] and only a selected subset of results is discussed here. Models similar to KIM have often been the object of sensitivity analysis. The large

Table 4. Comparison of calculated $\hat{S}_{T_i}$ values to the exact values S_{T_i} for each variable

Variable	S_{Ti} exact	$\hat{S}_{Ti}$ calculated (error term)
X_1	0.5574	0.5532 (0.043)
X_2	0.4442	0.4338 (0.049)
X_3	0.2410	0.2380 (0.048)

Table 5. Reactions in KIM model. Only sulphur bearing reaction products are given

$CH_3SCH_3 \cdot + OH \xrightarrow{k_1} CH_3S \cdot$	$CH_3SOO \cdot + NO \xrightarrow{k_9} CH_3SO \cdot$	$CH_3SO_2 \cdot + NO_2 \xrightarrow{k_{17}} CH_3SO_3 \cdot$	$SO_2 + OH \xrightarrow{k_{25}} H_2SO_4$
$CH_3SCH_3 \cdot + OH \xrightarrow{k_2} CH_3S \cdot (OH)CH_3$	$CH_3SO \cdot + O_3 \xrightarrow{k_{10}} CH_3SO_2 \cdot$	$CH_3SO_2 \cdot + O_3 \xrightarrow{k_{18}} CH_3SO_3 \cdot$	$CH_3S(O)_2O_2 \cdot + NO \xrightarrow{k_{26}} CH_3SO_3 \cdot$
$CH_3S \cdot (OH)CH_3 \xrightarrow{k_3} CH_3SO \cdot$	$CH_3SO \cdot + O_3 \xrightarrow{k_{11}} SO_2 \cdot$	$CH_3SO_2 \cdot + O_2 \underset{k_{-19}}{\overset{k_{19}}{\rightleftharpoons}} CH_3S(O)_2O_2 \cdot$	$CH_3SOO \cdot \xrightarrow{k_{27}} SO_2$
$CH_3S \cdot (OH)CH_3 \xrightarrow{k_4} CH_3SO_2CH_3$	$CH_3SO \cdot + NO_2 \xrightarrow{k_{12}} CH_3SO_2 \cdot$	$CH_3S(O)_2O_2 \cdot + NO_2 \underset{k_{-20}}{\overset{k_{20}}{\rightleftharpoons}}$	$CH_3SOO \cdot + NO_2 \underset{k_{-28}}{\overset{k_{28}}{\rightleftharpoons}} CH_3SOONO_2$
$CH_3S \cdot + NO_2 \xrightarrow{k_5} CH_3SO \cdot$	$CH_3SO \cdot + NO_2 \xrightarrow{k_{13}} SO_2$	$CH_3S(O)_2O_2NO_2$	$CH_3SOO \cdot + O_3 \xrightarrow{k_{29}} CH_3SO \cdot$
$CH_3S \cdot + O_3 \xrightarrow{k_6} CH_3SO \cdot$	$CH_3SO \cdot + O_2 \underset{k_{-14}}{\overset{k_{14}}{\rightleftharpoons}} CH_3S(O)O_2$	$CH_3SO_2O_2 \cdot \xrightarrow{k_{21}} SO_2$	$CH_3S \cdot + O_2 \xrightarrow{k_{30}} SO_2$
$CH_3S \cdot + O_2 \underset{k_{-7}}{\overset{k_7}{\rightleftharpoons}} CH_3SOO \cdot$	$CH_3SO \cdot + NO_2 \underset{k_{-15}}{\overset{k_{15}}{\rightleftharpoons}} CH_3S(O)O_2NO_2$	$CH_3SO_3 \cdot \xrightarrow{k_{22}} SO_3$	$CH_3SOO \cdot + O_2 \xrightarrow{k_{31}} SO_2$
$CH_3SOO \cdot \xrightarrow{k_8} CH_3SO_2 \cdot$	$CH_3S(O)O_2 \cdot \xrightarrow{k_{16}} CH_3SO_2 \cdot$	$CH_3SO_3 \cdot \xrightarrow{k_{23}} CH_3SO_3H$	
		$SO_3 + H_2O \xrightarrow{k_{24}} H_2SO_4$	

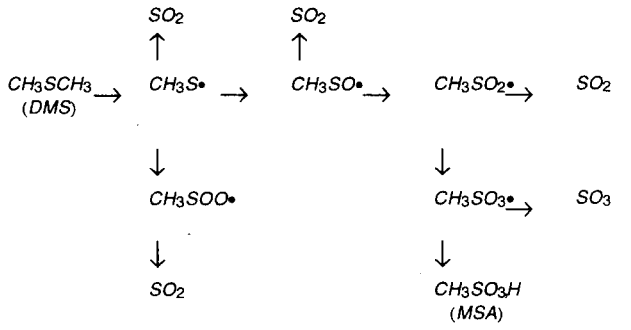

Fig. 1. Simplified flow diagram for the oxidation of DMS.

number of variables of this model has two important consequences:

- The constructive dimension is too high to use Sobol' quasirandom numbers; a crude Monte Carlo sampling is used instead.
- The cost of computing all the higher order terms is prohibitive. For this reason only the $\hat{S}_i$, $\hat{S}_i^*$ (first order) and $\hat{S}_{Ti}$, $\hat{S}_{Ti}^*$ (total effect) terms are computed.

A base sample of size $N = 1{,}000$ was used. Results are given for two output variables: the concentration of methane sulphonic acid (MSA) and the $[MSA]/([H_2SO_4] + [SO_2])$ ratio. MSA, SO_2 and H_2SO_4 are all possible end products of the oxidation of DMS. Results for $\hat{S}_i$, $\hat{S}_{Ti}$ are given in Fig. 2 for [MSA] and Fig. 3 for $[MSA]/([H_2SO_4] + [SO_2])$. Only those variables are given whose $\hat{S}_{Ti}$ value is in general greater than its $\delta\hat{S}_{Ti}$ in eqn (37).

The main purpose of this test case is to illustrate the difference between the first order terms and the total effect ones. We intend to show that using the former

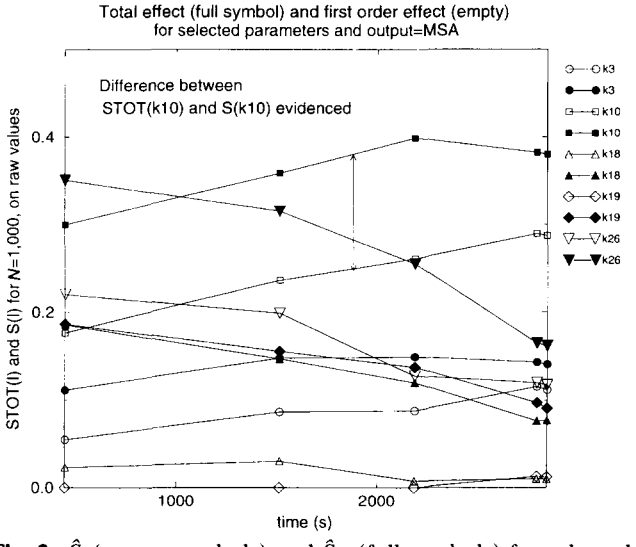

Fig. 2. $\hat{S}_i$ (empty symbols) and $\hat{S}_{Ti}$ (full symbols) for selected parameters for MSA.

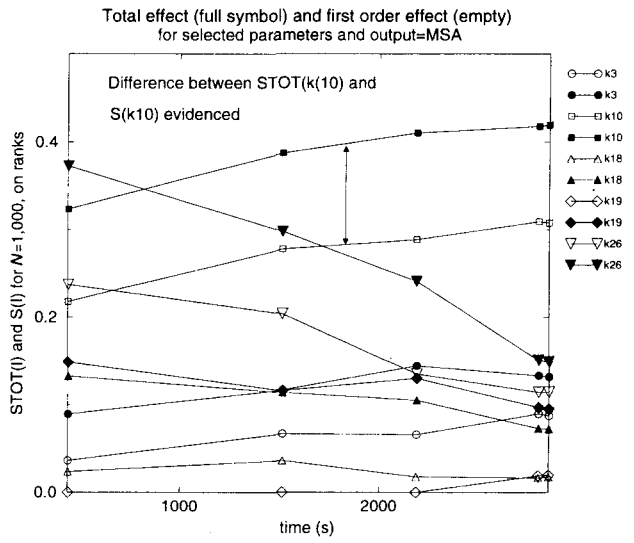

Fig. 3. $\hat{S}_i^*$ (empty symbols) and $\hat{S}_{Ti}^*$ (full symbols) for selected parameters for MSA.

can be misleading. The difference between S_i and S_{Ti} is a measure of the nonlinearity of the model. In fact the nonlinearity of a model also depends on which output variable is considered. It is intuitive that the model for [MSA] is more linear than the model for $[MSA]/([H_2SO_4] + [SO_2])$ ratio. This is reflected in the differences between the estimates $\hat{S}_i$ and $\hat{S}_{Ti}$ (Figs 2 and 3). It should be noted that, in Fig. 2, in spite of the differences between the first order indices and the total ones, the ranking of the parameters is substantially preserved. This is not the case for the more nonlinear model (Fig. 3), where the relative importance of k_{26} and k_{31} is reversed. This result is not due to the limited sample size, but to the relevance of higher order terms in this model (which—incidentally—is not dramatically nonlinear). This behaviour points to an intrinsic weakness of the importance measures, and of the use of first order indices alone.

The results for the rank based models are given in Fig. 4 for [MSA] and Fig. 5 for $[MSA]/([H_2SO_4] + [SO_2])$. As in all the other test cases the effect of the rank transformation is to 'flatten' the model, increasing the model linearity. This is reflected in an increase in the first order terms at the expenses of the higher order ones. The $\hat{S}_i^*$, $\hat{S}_{Ti}^*$ curves are, in general, much closer to each other than the $\hat{S}_i$, $\hat{S}_{Ti}$ ones. Inspection of Tables 6 and 7 reveals what a serious problem the estimation of the sensitivity indices is. In spite of the large base sample the error associated with $\hat{S}_i$, $\hat{S}_{Ti}$ is still large. According to our experience this is mainly due to the large scale of variation of the output (and less to the model nonlinearity). As expected, the error on the ranked measures is much lower, which makes ranks, in spite of their limitations, a popular alternative.

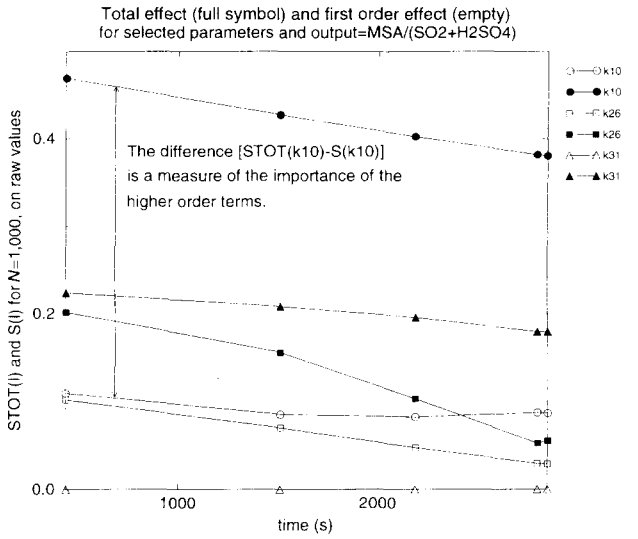

Fig. 4. $\hat{S}_i$ (empty symbols) and $\hat{S}_{Ti}$ (full symbols) for selected parameters for MSA/(SO_2 + H_2SO_4).

3.3 Third test case: simplified Level E

The test case employed here is a simplified version of the exercise already discussed in Saltelli *et al.*[13] The detailed description of that exercise, named Level E, is given in the OECD/NEA report.[39] It involves the computation of the dose to man resulting from migration of radionuclides. The release takes place from a nuclear waste repository in an idealized geological formation. The source term model consists of a delay for an initial containment time, T, followed by leaching at a constant fraction rate k. The governing equations for the inventory of radionuclides

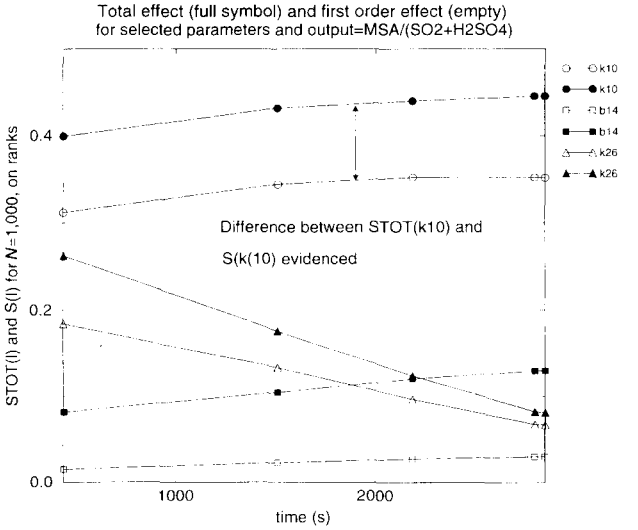

Fig. 5. $\hat{S}_i^*$ (empty symbols) and $\hat{S}_{Ti}^*$ (full symbols) for selected parameters for MSA/(SO_2 + H_2SO_4).

Table 6. Sensitivity indices (first order and total effect) for MSA for the same variables selected as in Figs 2 and 3, on raw values and ranks

Variable	Time (s)	$\hat{S}_i$	$\delta\hat{S}_i$	$\hat{S}_{Ti}$	$\delta\hat{S}_{Ti}$
k_3	676.9	0.0555	0.0273	0.1117	0.0777
	1427.0	0.0870	0.0268	0.1482	0.0676
	2275.0	0.0884	0.0283	0.1494	0.0611
	3475.0	0.1157	0.0285	0.1437	0.0531
	3600.0	0.1120	0.0285	0.1410	0.0539
k_{10}	676.9	0.1766	0.0310	0.2997	0.0668
	1427.0	0.2364	0.0306	0.3590	0.0577
	2275.0	0.2610	0.0320	0.3989	0.0512
	3475.0	0.2897	0.0313	0.3823	0.0468
	3600.0	0.2874	0.0316	0.3803	0.0481
k_{18}	676.9	0.0237	0.0242	0.1858	0.0688
	1427.0	0.0313	0.0225	0.1475	0.0661
	2275.0	0.0075	0.0227	0.1201	0.0617
	3475.0	0.0100	0.0227	0.0769	0.0576
	3600.0	0.0097	0.0230	0.0776	0.0588
k_{19}	676.9	-0.0197	0.0209	0.1866	0.0708
	1427.0	-0.0041	0.0213	0.1560	0.0654
	2275.0	-0.0114	0.0227	0.1377	0.0619
	3475.0	0.0128	0.0231	0.0975	0.0572
	3600.0	0.0124	0.0231	0.0912	0.0589
k_{26}	676.9	0.2202	0.0372	0.3512	0.0661
	1427.0	0.1988	0.0308	0.3153	0.0599
	2275.0	0.1277	0.0279	0.2550	0.0577
	3475.0	0.1199	0.0270	0.1651	0.0540
	3600.0	0.1175	0.0268	0.1622	0.0549
Variable	Time (s)	$\hat{S}_i^*$	$\delta\hat{S}_i^*$	$\hat{S}_{Ti}^*$	$\delta\hat{S}_{Ti}^*$
k_3	676.9	0.0365	0.0228	0.0898	0.0366
	1427.0	0.0674	0.0228	0.1172	0.0361
	2275.0	0.0667	0.0232	0.1448	0.0358
	3475.0	0.0905	0.0238	0.1336	0.0359
	3600.0	0.0885	0.0238	0.1323	0.0360
k_{10}	676.9	0.2174	0.0255	0.3236	0.0338
	1427.0	0.2777	0.0264	0.3873	0.0328
	2275.0	0.2888	0.0272	0.4105	0.0325
	3475.0	0.3092	0.0276	0.4178	0.0325
	3600.0	0.3076	0.0276	0.4190	0.0325
k_{18}	676.9	0.0238	0.0227	0.1332	0.0364
	1427.0	0.0364	0.0225	0.1144	0.0366
	2275.0	0.0178	0.0228	0.1059	0.0366
	3475.0	0.0167	0.0226	0.0737	0.0370
	3600.0	0.0174	0.0226	0.0730	0.0370
k_{19}	676.9	-0.0209	0.0216	0.1492	0.0357
	1427.0	-0.0058	0.0214	0.1167	0.0363
	2275.0	-0.0076	0.0221	0.1306	0.0361
	3475.0	0.0192	0.0224	0.0976	0.0367
	3600.0	0.0198	0.0224	0.0959	0.0367
k_{26}	676.9	0.2372	0.0266	0.3724	0.0332
	1427.0	0.2038	0.0255	0.2980	0.0342
	2275.0	0.1357	0.0250	0.2408	0.0349
	3475.0	0.1147	0.0245	0.1514	0.0363
	3600.0	0.1149	0.0245	0.1494	0.0362

at time t, $M(t)$ are

$$\frac{dM}{dt} = -\lambda M, \quad t < T \tag{58}$$

Table 7. Sensitivity indices (first order and total effect) for the ratio MSA/($SO_2 + H_2SO_4$) for the same variables selected as in Figs 4 and 5, on raw values and ranks. b_{14} is the exponential term in the Arrhenius equation for k_{-14}, i.e.:

$$k_{-14} = \exp(a_{14})\exp\left(\frac{b_{14}}{r_{cat} \cdot t_{kelv}}\right)$$

Variable	Time (s)	$\hat{S}_i$	$\delta\hat{S}_i$	$\hat{S}_{Ti}$	$\delta\hat{S}_{Ti}$
k_{10}	676.9	0.1096	0.0534	0.4695	0.1297
	1427.0	0.0857	0.0612	0.4278	0.1509
	2275.0	0.0824	0.0646	0.4027	0.1623
	3475.0	0.0873	0.0641	0.3815	0.1635
	3600.0	0.0865	0.0640	0.3799	0.1670
k_{26}	676.9	0.1020	0.0815	0.2017	0.2769
	1427.0	0.0704	0.1089	0.1560	0.2428
	2275.0	0.0478	0.1138	0.1032	0.2387
	3475.0	0.0292	0.1068	0.0526	0.2394
	3600.0	0.0286	0.1064	0.0553	0.2403
k_{31}	676.9	−0.0377	0.0299	0.2242	0.1869
	1427.0	−0.0569	0.0391	0.2084	0.1852
	2275.0	−0.0582	0.0433	0.1955	0.1915
	3475.0	−0.0453	0.0466	0.1792	0.1917
	3600.0	−0.0452	0.0464	0.1793	0.1949
Variable	Time (s)	$\hat{S}_i^*$	$\delta\hat{S}_i^*$	$\hat{S}_{Ti}^*$	$\delta\hat{S}_{Ti}^*$
k_{10}	676.9	0.3117	0.0270	0.3996	0.0329
	1427.0	0.3440	0.0276	0.4314	0.0322
	2275.0	0.3518	0.0280	0.4395	0.0321
	3475.0	0.3516	0.0282	0.4453	0.0321
	3600.0	0.3513	0.0282	0.4453	0.0321
b_{14}	676.9	0.0152	0.0221	0.0817	0.0370
	1427.0	0.0231	0.0222	0.1049	0.0369
	2275.0	0.0276	0.0224	0.1208	0.0368
	3475.0	0.0304	0.0226	0.1303	0.0367
	3600.0	0.0302	0.0226	0.1304	0.0367
k_{26}	676.9	0.1848	0.0254	0.2624	0.0346
	1427.0	0.1339	0.0243	0.1767	0.0358
	2275.0	0.0968	0.0238	0.1243	0.0365
	3475.0	0.0680	0.0233	0.0825	0.0370
	3600.0	0.0673	0.0233	0.0815	0.0370

$$\frac{dM}{dt} = -\lambda M + kM, \quad t \geq T \tag{59}$$

with the initial condition $M(0) = M_0$. The flux from the source term is then given by

$$S(t) = kM(t), \quad t \geq T \tag{60}$$

The geosphere model includes advection, longitudinal dispersion, equilibrium sorption and radioactive decay. The governing equation for the flux $F(x,t)$ is

$$R\frac{\partial F}{\partial t} + v\frac{\partial F}{\partial x} - dv\frac{\partial^2 F}{\partial x^2} = -\lambda RF \tag{61}$$

where R, v and d are retardation factor, flow velocity and dispersion length, respectively. The initial condition is

$$F(x,0) = 0 \tag{62}$$

and the boundary conditions are

$$F(0,t) = \delta(t) \tag{63}$$

$$F(\infty,t) = 0. \tag{64}$$

In the biosphere model the geosphere flux is assumed to enter a stream which is used for drinking water at the end of the geosphere layer whose length is l. The resultant dose $D(t)$ can be obtained analytically as[40]

$$D(T+t) = \frac{1}{2}\beta\frac{w}{W}kM_0 e^{-\lambda(T+t)} e^{l/2d} e^{-Rl^2/4dvt} e^{-vt/4dR} \times [\phi(\gamma) + \phi(\gamma')], \tag{65}$$

where β is an ingestion dose factor and

$$\gamma = \left(\frac{Rl^2}{4dvt}\right)^{1/2} + \left(\frac{vt}{4dR} - kt\right)^{1/2}, \tag{66}$$

$$\gamma' = \left(\frac{Rl^2}{4dvt}\right)^{1/2} - \left(\frac{vt}{4dR} - kt\right)^{1/2}, \tag{67}$$

$$\phi(z) = e^{z^2} erfc(z). \tag{68}$$

The isotope I-129 is considered in this test case. Six parameters are treated as uncertain variables with the form of probability distributions in Table 8.

For this exercise the full range of sensitivity indices is computed. A base sample size of 1024 was generated using Sobol' quasirandom sequences. Then sensitivity estimates were computed for all the $2^6 - 1 = 63$ combinations (see eqn (9)). $\hat{S}_{i_1,\ldots,i_s}$ values for the dose output at three time points are given in Table 9. At all time points the variance of the output is accounted for by two or three higher order terms plus one first order. The sum of the terms gives roughly 80% (or more) of the total variance. The predominance of the higher order terms is evident.

Although the simplified version of the test case used here considers only I-129 and one geosphere layer, rather than the nuclide chain plus multi-layered geosphere of the full test case, this model still has an interesting nonmonotonic feature. Figure 6 shows the model coefficients of determination R^2_y for the regression models based on the raw values and on the ranks. This coefficient provides a measure of how well the linear regression model based on either SRC's or SRRC's can reproduce the actual output vector. The large difference between the values of these two coefficients demonstrates the nonlineality of the model. This test model also has an interesting nonmonotonic feature which peaks at $t = 8 \times 10^4$ y.

Table 8. Input parameter for the simplified Level E test case

Notation	Definition	Distribution	Range	Units
v	water velocity in geosphere layer	log-uniform	$/10^{-3}, 10^{-1}/$	m/a
W	stream flow rate	log-uniform	$/10^{5}, 10^{7}/$	m^3/a
R	retardation factor for Iodine	uniform	/1, 5/	-
l	length of geosphere layer	uniform	/100, 500/	m
T	containment time	uniform	/100, 1,000/	a
k	leach rate for Iodine	log-uniform	$/10^{-3}, 10^{-2}/$	a^{-1}

The SRRC's as a function of time for all six variables are given in Fig. 7. The SRRCs in Fig. 7 show the changing pattern of importance over time. The absolute values of the SRRCs can be used to determine variable importance and the sign of a SRRC indicates the input and output correlation. Although Fig. 7 gives us information that the variables which govern the transit time in the geosphere have a positive correlation with the output at early time, which becomes negative at a later time, the SRRC's fail to yield proper ranking of uncertain input parameters at the presence of model nonmonotonicity.

$\hat{S}_{Ti}$ instead finds a proper ranking of input parameters even at the nonmonotonic point, $t = 8 \times 10^4$ year as shown in Fig. 8. Results for $\hat{S}^*_{Ti}$ as a function of time have also been plotted in Fig. 9. There are remarkable differences between S_{Ti} and S^*_{Ti} for the stream flow rate, W. This difference highlights an interesting 'pathology' linked to the use of rank transformation. As seen from Fig. 9, variable W is never important when using rank transformed indices. This stems from eqn (65), but this is not what concerns us here. The fact that W is only relevant in association with other variables makes W an ideal victim of the rank transformation which, as observed in the second test case, kills the higher order terms at the expense of the first order ones.

4 CONCLUSIONS

This work is mainly devoted to the study of Sobol' sensitivity indices, to their performances, and to the introduction of a new global index. This is also a sequel to earlier investigations of the performances of the measure of importance, originally developed by Hora & Iman.[14] In Saltelli *et al.*,[13] in particular, we have suggested a ranked version of the measure of importance, and in Homma & Saltelli,[19] we have tested different sampling strategies (crude MC, LHS, quasirandom) for its estimate. In this paper we show how this ranked measure exactly coincides with S_i^*, a Sobol' sensitivity index of the first order computed on the ranks.

Our finding highlights the value of the sensitivity indices. The $Si_1,...,i_s$ are informative as they yield the exact fraction of the output variance accounted for by any input parameter or combination of parameters. This variance analysis is indeed a rigorous form of sensitivity analysis. In this respect the sensitivity indices resembles the FAST approach. The computation of the $Si_1,...,i_s$ seems more straightforward than that of the FAST coefficients, especially as far as the higher order terms are concerned. There is no difference, from the computational point of view, between a first order term, S_i, and a higher order $S_{i_1,...i_s}$ or S_{Ti} term.

Our experience with the test case KIM is that the parameter ranking can be affected by the higher order terms, even when the sum of the first order terms is not far from unity. Even in the first test case, the sum of the first order terms was higher than 0.7, and yet this did not capture an interesting second order term. A sensitivity analysis based on the importance measure, or on Sobol' or FAST sensitivity indices of the first order, may thus be misleading. In this respect we would tend to disagree with a 'rule of the thumb'

Table 9. Sensitivity estimates $\hat{S}_{i_1,...i_s}$ for dose in decreasing order at three time points

$t = 10^4$ (y)		$t = 8 \times 10^4$ (y)		$t = 7 \times 10^5$ (y)	
vW	0.203(0.126)	vWR	0.286(0.166)	v	0.281(0.125)
$vWRI$	0.197(0.249)	vWI	0231(0.133)	vWR	0.154(0.187)
I	0.131(0.093)	v	0.119(0.048)	vWI	0.129(0.146)
vR	0.111(0.141)	W	0.109(0.044)	vRI	0.119(0.180)
vRI	0.109(0.164)	vW	0.102(0.101)	RI	0.109(0.074)
R	0.104(0.078)				
v	0.100(0.032)				

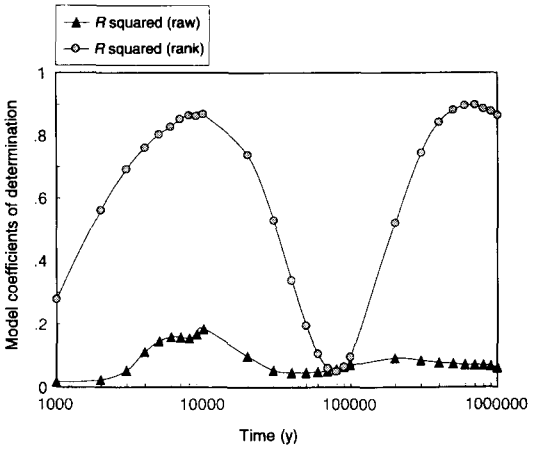

Fig. 6. Model coefficients of determination R^2_y for the simplified Level E test case.

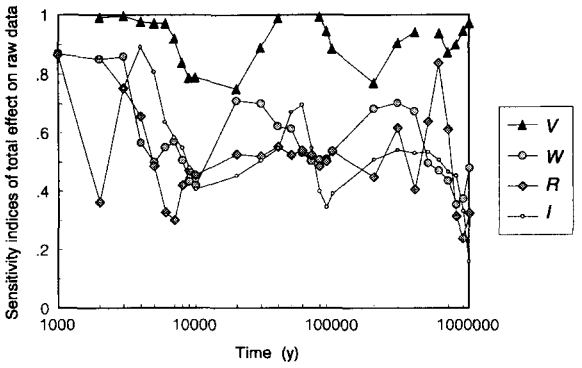

Fig. 8. $\hat{S}_{Ti}$ for selected parameters for the simplified Level E test case.

that says that a FAST based sensitivity analysis is good enough when the sum of the first order terms is greater than 0.6.[25]

The flexibility of the $S_{i_1,\ldots,i_s}$ derives from the possibility of adapting the computation's strategy to the model at hand, depending mostly on the number of input parameters involved. If the number of parameters is low and the model not too expensive to run, then all the $Si_{1,\ldots,i_s}$ can be computed (test cases 1 and 3) achieving a complete variance analysis. This corresponds to a complete 'killing' of the problem. For systems with a large number of parameters the S_{Ti} coefficients, derived in this article, can be computed (test case 2). In this latter case the amount of information collected is reduced, but the parameters are still accurately ranked. The $S_{i_1,\ldots,i_s}$ are also reliable and accurate in the sense discussed in Saltelli *et al.*[13] They can rank the input parameters when other tests (such as the PRCC, SRRC) fail due to model nonmonotonicity (test cases 1 and 3).

The main drawback in the use of the $S_{i_1,\ldots,i_s}$ is the large sample size needed for their evaluation. This is due on one hand to the difficulty of estimating a variance (scarce robustness of the estimate), and on the other hand to the fact that the base sample has to be replicated at least as many times as the number of variables. The scarce robustness of the importance measures, also discussed by Iman & Hora,[15] is particularly acute when, in the order, (a) there is a large range of variability in the output variables, (b) there are many input variables and (c) the model is strongly nonlinear. Our experience with the indices seems to indicate that (c) has a moderate impact on the robustness of $S_{i_1,\ldots,i_s}$. When the error on the $S_{i_1,\ldots,i_s}$ is excessive, the ranked version of the test can be used, which usually provides more stable results. This comes to the expenses of an alteration of the original model. The $S^*_{i_1,\ldots,i_s}$ forcefully linearizes the model, artificially increasing the fraction of the total variance accounted for by the first order terms (test case 3, in particular). Yet in the absence of computationally viable alternatives the S^*_{Ti} seems to offer a workable solution (maybe the only solution) to the problem.

Finally it can be worth stressing that the method presented in this article is not a screening test. It works better when the model allows a good thousand simulations per variable. Computational constraints

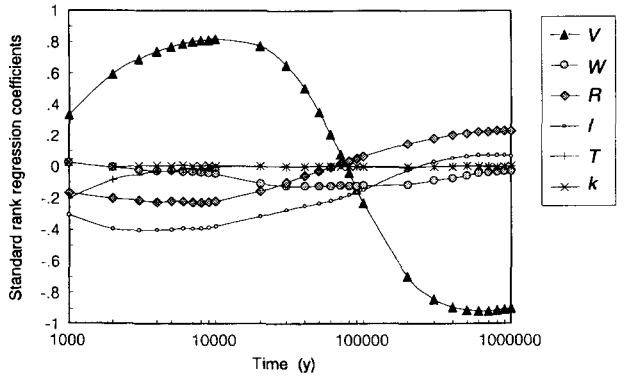

Fig. 7. Standardised rank regression coefficients for the simplified Level E test case.

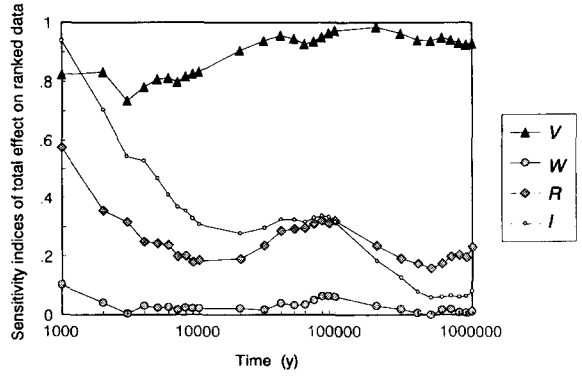

Fig. 9. $\hat{S}^*_{Ti}$ for selected parameters for the simplified Level E test case.

can make this impossible for many problems. Yet, once the model has been screened, and the variable parameters reduced to a manageable size, here the sensitivity indices can come into play, and yield an information as accurate as that which one could achieve using FAST, and as straightforward to compute as a standard deviation.

REFERENCES

1. Helton, J.C., Iman, R.L., Johnson, J.D. & Leigh, C.D., Uncertainty and sensitivity analysis of a dry containment test problem for the MAEROS aerosol model. *Nucl. Sci. & Engng*, **102** (1989) 22–42.
2. Helton, J.C., Rollstin, J.A., Sprung, J.L. & Johnson, J.D., An exploratory sensitivity study with the MACCS reactor accident consequence model. *Reliab. Engng System Safety*, **36** (1992) 137–164.
3. Helton, J.C., Garner, J.W., Marietta, M.G., Rechard, R.P., Rudeen, D.K. & Swift, P.N., Uncertainty and sensitivity analysis results obtained in a preliminary performance assessment for the waste isolation pilot plant. *Nucl. Sci. & Engng*, **114** (1993) 286–331.
4. Rabitz, H., System analysis at molecular scale. *Sci.*, **246** (1989) 221–226.
5. Helton, J.C., Uncertainty and sensitivity analysis techniques for use in performance assessment for radioactive waste disposal. *Reliab. Engng System Safety*, **42** (1993) 327–367.
6. Welch, W.J., Buck, R.J., Sacks, J., Wynn, H.P., Mitchell, T.J. & Morris, M.D., Screening, predicting, and computer experiments. *Technometrics*, **34** (1992) 15–47.
7. Andres, T.H. & Hajas, W.C., Using iterated fractional factorial design to screen parameters in sensitivity analysis of a probabilistic risk assessment model. In *Proceedings of Joint International Conference on Mathematical Methods and Supercomputing in Nuclear Applications*, Karlsruhe, Germany, 19–23 April 1993, Kernforschungszentrum, Karlsruhe, pp. 328–340.
8. Saltelli, A., Andres, T.H. & Homma, T., Sensitivity analysis of model output; performance of the iterated fractional factorial design (IFFD) method. *Computational Stat. & Data Anal.*, **20** (1995) 387–407.
9. Cawlfield, J.D. & Wu, M.-C., Probabilistic sensitivity analysis for one-dimensional reactive transport in porous media. *Water Resources Res.*, **29** (1993) 661–672.
10. Koda, M., Sensitivity analysis of stochastic dynamical systems. *Int. J. System Sci.*, **23** (1992) 2187–2195.
11. Iman, R.L. & Helton, J.C., An investigation of uncertainty and sensitivity analysis techniques for computer models. *Risk Anal.*, **8** (1988) 71–90.
12. Saltelli, A. & Homma, T., Sensitivity analysis for model output; performance of black box techniques on three international benchmark exercises. *Computational Stat. & Data Anal.*, **13** (1992) 73–94.
13. Saltelli, A., Andres, T.H. & Homma, T., Sensitivity analysis of model output; an investigation of new techniques. *Computational Stat. & Data Anal.*, **15** (1993) 211–238.
14. Hora, S.C. & Iman, R.L., A comparison of Maximum/Bounding and Bayesian/Monte Carlo for fault tree uncertainty analysis. *SAND*85-2839, Sandia National Laboratories, Albuquerque, NM, 1986.
15. Iman, R.L. & Hora, S.C., A robust measure of uncertainty importance for use in fault tree system analysis. *Risk Anal.*, **10** (1990) 401–406.
16. McKay, M.D. & Beckman, R.J., A procedure for assessing uncertainty in models. In *Proceedings of PSAM-II, An International Conference Devoted to Advancement of System-based Methods for the Design and Operation of Technological Systems and Processes*, San Diego, CA, USA, 20–25 March 1994, 009-13-18.
17. Ishigami, T. & Homma, T., An importance quantification technique in uncertainty analysis. *JAERI-M* 89-111, Japan Atomic Energy Research Institute, Tokai, 1989.
18. Ishigami, T. & Homma, T., An importance quantification technique in uncertainty analysis for computer models. In *Proceedings of the ISUMA '90, First International Symposium on Uncertainty Modelling and Analysis*, University of Maryland, USA, 3–5 December 1990, pp.398–403.
19. Homma, T. & Saltelli, A., Use of Sobol's quasirandom sequence generator for integration of modified uncertainty importance measure. *J. Nucl. Sci. Tech.*, **32** (1995) 1164–1173.
20. Sobol', I.M., Sensitivity estimates for nonlinear mathematical models. *Math. Modeling & Computational Experiment*, **1** (1993) 407–414.
21. Cukier, R.I., Fortuin, C.M., Schuler, K.E., Petschek, A.G. & Schaibly, J.K., Study of the sensitivity of coupled reaction systems to uncertainties in rate coefficients I: Theory. *J. Chemical Physics*, **59** (1973) 3873–3878.
22. Schaibly, J.K. & Schuler, K.E., Study of the sensitivity of coupled reaction systems to uncertainties in rate coefficients II: Applications. *J. Chemical Physics*, **59** (1973) 3879–3888.
23. Cukier, R.I., Levine, H.B. & Schuler, K.E., Nonlinear sensitivity analysis of multiparameter model systems. *J. Computational Physics*, **26** (1978) 1–42.
24. Sacks, J., Welch, W.J., Mithcell, T.J. & Wynn, H.P., Design and analysis of computer experiments. *Stat. Sci.*, **4** (1989) 409–435.
25. Liepman, D. & Stephanopoulos, G., Development and global sensitivity analysis of a closed ecosystem model. *Ecological Modelling*, **30** (1985) 13–47.
26. Sobol', I.M., On the distribution of points in a cube and the approximate evaluation of integrals. *USSR Computational Math. & Math. Physics*, **7** (1976) 86–112.
27. Sobol', I.M., Uniformly distributed sequences with an additional uniform property. *USSR Computational Math. & Math. Physics*, **16** (1976) 236–242.
28. Sobol', I.M., Quasi-Monte-Carlo methods. *Progress in Nucl. Energy*, **24** (1990) 55–61.
29. Morris, M.D., Factorial sampling plans for preliminary computational experiments. *Technometrics*, **33** (1991) 161–174.
30. McKay, M.D., Beckman, R.J. & Conover, W.J., A comparison of three methods for selecting values of input variables in the analysis of output from a computer code. *Technometrics*, **21** (1979) 239–245.
31. Davis, P.F. & Rabinowitz, P., *Methods of numerical integration*, 2nd ed., Academic Press, NY, 1984.
32. Bratley, P. & Fox, B.L., Algorithm 659, implementing Sobol's quasirandom sequence generator. *ACM Trans. Math. Software*, **14** (1988) 88–100.
33. Sobol', I.M., Turchaninov, V.I., Levitan, Yu. L. & Shukhman, B.V., *LPTAU. A FORTRAN subroutine for quasi random sequence generators. Keldysh Institute of Applied Mathematics*, Russian Academy of Sciences, (distributed by OECD/NEA Data Bank, 12 bd. des Iles 92130 Issy les Moulineaux, France) 1991.

34. Box, G.E.P., Hunter, W.G. & Hunter, J.S., *Statistics for experimenters*, John Wiley and Sons, New York, 1978.
35. Cotter, S.C., A screening design for factorial experiments with interactions. *Biometrika*, **66** (1979) 317–320.
36. Iman, R.L., Shortencarier, M.J. & Johnson, J.D., A FORTRAN 77 program and user's guide for the calculation of partial correlation and standardized regression coefficients. *SAND*85-0044, Sandia National Laboratories, Albuquerque, NM, 1985.
37. Gear, C.W., *Numerical initial value problems in ordinary differential equations*, Prentice-Hall, Englewood Cliffs, New Jersey, 1971.
38. Saltelli, A. & Hjorth, J., Uncertainty and sensitivity analyses of dimethylsulphide (DMS) oxidation kinetics. *J. Atmospheric Chem.*, **21** (1995) 187–221.
39. OECD/NEA PSAC User Group, *PSACOIN Level E Intercomparison, An international code intercomparison exercise on a hypothetical safety assessment case study for radioactive waste disposal systems*, OECD/NEA publication, Paris, 1989.
40. Robinson, P.C. & Hodgkinson, D.P., Exact solutions for radionuclide transport in the presence of parameter uncertainty. *Radioactive waste management and the nuclear fuel cycle*, **8** (1987) 283–311.