An adaptive least-squares global sensitivity method and application to a plasma-coupled combustion prediction with parametric correlation

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\textbf{ABSTRACT}

We introduce an efficient non-intrusive surrogate-based methodology for global sensitivity analysis and uncertainty quantification. Modified covariance-based sensitivity indices (mCov-SI) are defined for outputs that reflect correlated effects. The overall approach is applied to simulations of a complex plasma-coupled combustion system with disparate uncertain parameters in sub-models for chemical kinetics and a laser-induced breakdown ignition seed. The surrogate is based on an Analysis of Variance (ANOVA) expansion, such as widely used in statistics, with orthogonal polynomials representing the ANOVA subspaces and a polynomial dimensional decomposition (PDD) representing its multi-dimensional components. The coefficients of the PDD expansion are obtained using a least-squares regression, which both avoids the direct computation of high-dimensional integrals and affords an attractive flexibility in choosing sampling points. This facilitates importance sampling using a Bayesian calibrated posterior distribution, which is fast and thus particularly advantageous in common practical cases, such as our large-scale demonstration, for which the asymptotic convergence properties of polynomial expansions cannot be realized due to computation expense. Effort, instead, is focused on efficient finite-resolution sampling. Standard covariance-based sensitivity indices (Cov-SI) are employed to account for correlation of the uncertain parameters. Magnitude of Cov-SI is unfortunately unbounded, which can produce extremely large indices that limit their utility. Alternatively, mCov-SI are then proposed in order to bound this magnitude \( \in [0, 1] \). The polynomial expansion is coupled with an adaptive ANOVA strategy to provide an accurate surrogate as the union of several low-dimensional spaces, avoiding the typical computational cost of a high-dimensional expansion. It is also adaptively simplified according to the relative contribution of the different polynomials to the total variance. The approach is demonstrated for a laser-induced turbulent combustion simulation model, which includes parameters with correlated effects.

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1. Introduction

This paper presents an uncertainty quantification (UQ) and global sensitivity analysis (GSA) method designed for relatively high-dimensional problems. It is based on variance-based GSA with the global sensitivity indices (SI) introduced by Sobol' [1] using the Analysis of Variance (ANOVA) expansion for independent inputs. The use of Sobol' indices is now one of the most common GSA techniques. It uses a functional decomposition to incorporate component functions involving single or groups of uncertain parameters, and the computation of the sensitivity measures of each component function is usually done by Monte Carlo (MC) or quasi Monte Carlo (quasi-MC) methods. Theoretical properties of the global SI are established [2–6], and it has proven useful in applications [7]. Similar sensitivity measures were developed independently by Wagner [8] in operations research. However, here at the outset, we should recognize that sensitivity analysis is limited to global methods and to variance-based definitions. Borgonovo and Plischke [9] and Ghanem et al. [10] review local sensitivity measures, derivative-based global measures, moment-independent (density-based) measures, and value-of-information-based measures. Several global measures (including variance-, density-, and value-of-information-based sensitivities) satisfy a common rationale [11], and quantify the discrepancy between unconditional and conditional probability. Borgonovo et al. [11] address the estimation methods for this class of global measures.

It is well-understood that GSA seeks to quantify the overall influence of input parameters on output quantities of interest (see for example Borgonovo et al. [12]). However, a broad evaluation of the parameter space can lead to high cost in, for example, MC or quasi-MC methods. Sudret [13] used generalized Polynomial Chaos expansions (gPC) to build surrogate models for computing Sobol' sensitivity indices based on the surrogate form. Blatman and Sudret [14] further introduced the sparse gPC expansions in order to efficiently estimate the global SI. To avoid the cost of brute-force evaluation in high dimensions, they employ a stepwise approach to select only most significant polynomials by recursively resolving a least-squares regression (LSR) system. The LSR approach is an efficient tool to compute polynomial expansion coefficients, by minimizing the error of the surrogate model representation in the mean-square sense [13–15]. Compared to a projection approach [16–18], where each polynomial coefficient is obtained by computing a multi-dimensional integral, the LSR approach is more flexible (in choosing sampling points), which seems to be generally advantageous for problems involving a potentially large number of uncertain parameters.

In contrast to the Blatman and Sudret [14] approach, Tang et al. [19] used ANOVA expansion and Polynomial Dimensional Decomposition (PDD) [20–24] to represent the expansion’s component subspace functions. The main challenge, and motivation for improving PDD methods, is reducing the required number of samples, which corresponds to the cost of the method. Standard ANOVA components increase exponentially with the number of uncertain parameters, with a corresponding polynomial increase of the number of PDD terms for each component function. This limits the LSR approach even for a truncated low-order ANOVA expansion; indeed, for the LSR problem to be well-posed, the number of deterministic model evaluations is necessarily larger than the total polynomial expansion size [14]. Thus, Tang et al. [19] introduced an advantageous combination of the adaptive ANOVA method [25,26] and the stepwise regression technique [14], with which a sparse surrogate model representation can be efficiently constructed. A key to doing this is the use of variance contribution to select the most important polynomials. Tang et al. [19] demonstrated that this can be more efficient. Thus, the present work is based on the use of this approach to take advantage of its efficiency for building sparse surrogates when the superconvergence of PDD/gPC expansions cannot be achieved, as is often the case in practice.

Correlative effects are particularly important in the current combustion application introduced in Sections 9 and 10. Definitions and numerical techniques can be developed for global sensitivity analysis with correlated inputs [27–29]. Kucherenko et al. [29] employed the same definition as Sobol' SI [1] for cases with correlated inputs, by using the conditional expectations for first-order SI, and the conditional variance for total sensitivity indices (TSI). These variance-based SI (Var-SI) [29] are consistent with Sobol' SI in case of independency, and in case of correlation their first-order SI are still strictly positive and bounded by [0, 1]. The computation relies on the use of MC methods with the knowledge of conditional probability density function (pdf). Alternatively, Li et al. [27] originally introduced the covariance-based sensitivity indices (Cov-SI) to account for correlation effects among ANOVA component functions due to input correlation. They introduced three Cov-SI (structural, correlation, and total contribution) for each component function, which reduce to a single index for independent inputs. Rahman [30] later defined the same sensitivity indices [31]. Under a boundedness type assumption on the joint pdf of inputs, Chastaing et al. [28] showed the availability of a generalized ANOVA decomposition, based on which they defined generalized sensitivity indices that are very similar to Cov-SI [27]. Li and Rabitz [31] recently established for correlated inputs a relationship between Var-SI and Cov-SI, and showed that Var-SI can be estimated from Cov-SI by using a modest number of model evaluations. An advantage of the Cov-SI [27] compared to Var-SI [29] is that a conditional pdf is not required when computing component functions, which is an attractive property since conditional distribution is rarely known completely, such as in our combustion application. Thus, we use the approach of Li et al. [27]. However, a disadvantage of this approach is that neither the Cov-SI are strictly non-negative nor their magnitude bounded. Consequently, one can obtain extremely large Cov-SI in presence of strong negative correlation between ANOVA components. We thus introduce generalized covariance-based sensitivity indices (mCov-SI), with magnitude $\in [0, 1]$.

The specific formulation we use is developed in Section 2, with emphasis on the new contributions. Overall it includes the ANOVA decomposition, polynomial representation of ANOVA components, and LSR approach for computation of coefficients. The adaptive sparse meta-modeling approach with stepwise LSR is summarized in Section 3. The variance-based SI (Var-SI) are presented in Section 4 and the original Cov-SI for correlated inputs in Section 5 as a foundation. Only key
aspects for the proposed generalization are summarized. The new mCov-SI are then introduced in Section 6, with a demonstration on a model mathematical function in Section 7. The combustion model and sources of uncertainties are presented in Section 8. We demonstrate the new method on a realistically challenging simulation-based prediction in Sections 9 and 10. This is done in two stages. First, in Section 9, a two-dimensional axisymmetric model of our full problem is introduced and used to demonstrate the methods in this context. We then show results for the costlier three-dimensional target application case, having reduced the dimension of the uncertain-parameter space in two dimensions. Conclusions are summarized following this two-stage demonstration.

2. ANOVA, orthogonal polynomials, and least-squares regression (LSR)

Let \( \mathbf{x} = (x_1, \ldots, x_N) \) denote a list of correlated random variables with uniform marginal distribution \((0 \leq x_i \leq 1 \text{ for } i = 1, \ldots, N)\) and the system response be a random variable \( y = f(\mathbf{x}) \). A physical uncertain parameter \( \zeta_i \) with an arbitrary marginal cumulative distribution function (cdf) \( F_i \) can be transformed into a uniform variable by [29]:

\[
\quad x_i = F_i(\zeta_i),
\]

which is used throughout this paper.

2.1. ANOVA expansion

The basic representation we use is its functional expansion

\[
f(\mathbf{x})_{\text{ANOVA}} = f_0 + \sum_{1 \leq i \leq N} f_i(x_i) + \sum_{1 \leq i < j \leq N} f_{ij}(x_i, x_j) + \cdots + f_{1,2,\ldots,N}(x_1, x_2, \ldots, x_N)
\]

Thus an ANOVA expansion decomposes the original \( N \)-dimensional space into a union of subspaces, with each subspace representing a particular order of interaction among parameters. Many problems only have low-order interactions and are amenable to such a representation, making an ANOVA expansion useful despite the nominally poor asymptotic scaling of the number of terms required in the complete ANOVA expansion:

\[
P_{\text{ANOVA}} = 2^N.
\]

2.2. ANOVA subspace representation by orthogonal polynomials

A set of orthogonal univariate polynomials is defined as an infinite sequence of polynomials, \( \{\psi^j(x); j = 0, 1, \ldots\} \), where \( \psi^j(x) \) has degree \( j \) and any two polynomials in the set are orthogonal so [32]

\[
\int_{\mathbb{R}} \psi^j(x) \psi^k(x) p_X(x) \, dx = \gamma^j_X \delta_{jk},
\]

where

\[
\delta_{jk} = \begin{cases} 1 & j = k, \\ 0 & j \neq k \end{cases} \quad \text{and} \quad \gamma^j_X = \int_{\mathbb{R}} \left(\psi^j(x)\right)^2 p_X(x) \, dx.
\]

For UQ, the particular polynomials used are often selected to have kernels that correspond to particular distributions. In this paper, transformed uniform variables and Legendre polynomials are used.

The methods we introduce will require bases that span multiple subspaces. Let

\[
\psi^j_{\mathbf{x}_T} = \prod_{k=1}^T \psi^{j_k}(x_{ik})
\]

represent a multivariate polynomial basis in the \( T \)-dimensional subspace, for which \( j_k \geq 1 \) is the degree of polynomial for the variable \( x_{ik} \), and \( \mathbf{j}_T = \{j_1, j_2, \ldots, j_T\} \). Following Rahman [20], the component functions in (2) can be expanded using an infinite number of basis functions in a tensor product for multi-indices \( \mathbf{j}_T \):

\[ f_{T_i}(x_T) = \sum_{j_1 \geq 1} C_{T_i}^{j_1} q_{T_i}^{j_1}, \] (7)

where the \( C_{T_i}^{j_1} \) are the expansion coefficients. The regression for computing these is introduced in Section 2.3.

In practice, (7) must be truncated. Following the Polynomial Dimensional Decomposition (PDD) of Rahman [20], we truncate (7) to \( m \) terms for each dimension:

\[ f_{T_i}(x_T) = \sum_{1 \leq j_1 \leq m} C_{T_i}^{j_1} q_{T_i}^{j_1}. \] (8)

Hence, the total size of the order-\( m \) full PDD expansion of an \( N \)-dimensional function is

\[ P_{\text{PDD}, m} = 1 + Nm + \binom{N}{2} m^2 + \cdots + \binom{N}{N} m^N = (1 + m)^N. \] (9)

PDD and generalized Polynomial Chaos (gPC) are closely related, so either could be used for the design of the least-squares adaptive strategy used in this paper. Building on previous work [19], we employ PDD to facilitate flexibility in adaptation. PDD can also readily support anisotropy in polynomial order, since the degree bound can be specified independently for each dimension, which is not straightforward for gPC [33].

2.3. Computation of polynomial coefficients

For independent inputs \( x \), PDD is an orthogonal expansion [20], and the corresponding functional decomposition (2) is also orthogonal. The coefficients involved in (8) can then be obtained by projection [16,34,35]

\[ C_{T_i}^{j_1} = \frac{E[f(x)q_{T_i}^{j_1}]}{E[q_{T_i}^{j_1}]}, \] (10)

though this is expensive for a large number of input variables, since it requires high-dimensional integration. For correlated inputs, the orthogonality of both PDD and (2) is lost, so the projection by (10) is not viable.

These challenges motivate a least-squares regression (LSR) approach [14], which also affords flexibility in choosing sampling points. Our approach can be regarded as forming a response surface that provides a least-squares optimal PDD expansion surrogate. As an LSR approach, it is essentially a fitting procedure, so it offers flexibility for complex and correlated uncertain inputs. It is thus introduced as potentially advantageous in common cases where cost precludes realization of asymptotic convergence of polynomial expansions.

The training set size \( Q \) for a regression problem must exceed the PDD expansion size \( P \). Experience shows that

\[ 1.5P \less B Q \less 3P \] (11)

leads to a sufficiently well-conditioned regression [19]. The numerical techniques for linear regression problems are standard, and we refer to Blatman and Sudret [14].

The standard approach of using independent, identically distributed (i.i.d.) univariate random variables and a transformation, e.g. Gentle [36, Section 7.4] and Kucherenko et al. [29], is an efficient way to generate training sets for our mathematical examples with multivariate normal distributions. In fact, to sample a normal distribution \( x \sim N(\mu, \Sigma) \), one only needs to do a Cholesky (or other adequate) decomposition of the covariance matrix: \( AA^T = \Sigma \), and the transformation is simply \( x = \mu + AX \), where \( x \) is an independent standard normal vector with zero mean and unit variance. To obtain a faster rate of convergence than standard MC, a quasi-random Sobol’ sequence [37] (or other low-discrepancy sequences) can be used to generate a uniform random vector, which can then be transformed into \( x \) using the inverse normal cdf (1). It is argued that at least a quadratic over-sampling rate is required for an \( \text{asymptotically} \) stable approximation using quasi-MC sampling techniques [38,39], though LSR with a linear over-sampling (11) can still provide good approximations [40]. Recently, LSR has been analyzed mathematically, seeking to ensure the stability and accuracy of polynomial approximations, leading to new sampling strategies with a low over-sampling rate and optimal convergence [38,40–45]. For some of these methods, the stability and convergence can be significantly improved even with a linear over-sampling rate (11). Use of these advanced methods remains our ongoing effort, and a standard quasi-MC (Sobol’ sequence) is used in this paper. For our combustion application with complex inputs, a Bayesian calibration procedure will be used.

We emphasize that \( Q \) sets most of the computational cost. Given that \( Q \) follows from \( P \) per (11), a necessary objective of this work is to reduce the expansion size \( P \) by including only the influential polynomial terms of the expansion.
3. Adaptive sparse ANOVA meta-modeling

In many cases, particularly those with a large number of uncertain parameters, reducing the size of a PDD or gPC representation of Section 2 can make uncertain analysis more efficient. To do this, Blatman and Sudret [14] proposed a gPC approach coupled with a stepwise regression method to build a sparse polynomial model of the output. Tang et al. [19] enhanced this stepwise regression technique with an adaptive ANOVA decomposition, so that the model can be constructed from low-dimensional spaces instead of a single high-dimensional one. They introduced additional levels of dimension reduction, which rely on the relative importance of expansion functions in terms of their variance relative to the total. This generalizes the approach of Blatman and Sudret [14], who introduced an adaptive strategy based on the representation error of the polynomial representation. Numerical examples confirm that the variance-based criterion can be more efficient for high-dimensional problems than its representation-error-based counterpart [19]. With this adaptive method, we are able to utilize high-degree polynomials (e.g. \( m = 10 \)) in our full-scale three-dimensional plasma-coupled turbulent combustion prediction (Fig. 31), while nearly all existing complex LSR applications use low polynomial degrees (e.g. \( m \leq 3 \)) [40].

This adaptive sparse meta-modeling method is used in this work and summarized in the following subsections. The standard truncation dimension \( \nu \) (i.e. the largest input interaction order) is introduced in Section 3.1, variance-based adaptive ANOVA in Section 3.2, and the stepwise regression in Section 3.3.

3.1. Truncation dimension \( \nu \)

The low-order interactions of input variables are known to impact output [1,46,47], and it might be argued that this is a characteristic of a well-designed physical model. This motivates truncation of the full ANOVA expansion (2) as

\[
\tilde{f}(\mathbf{x}) \approx f(\mathbf{x})_{\text{ANOVA, } \nu} = f_0 + \sum_{\nu=1}^{N} \sum_{\nu_1 < \cdots < \nu_T} f_{\nu_1, \nu_2, \cdots , \nu_T}(x_{\nu_1}, x_{\nu_2}, \cdots , x_{\nu_T}), \quad \text{with } \nu \ll N,
\]

(12)

where \( \nu \) is the truncation (or effective) dimension [25]. This obvious first step reduces the size of the ANOVA expansion from (3) to

\[
P_{\text{ANOVA, } \nu} = 1 + N + \binom{N}{2} + \cdots + \binom{N}{\nu},
\]

(13)

which will typically be much smaller. For example, with \( N = 50 \) and \( \nu = 2 \), \( P_{\text{ANOVA, } \nu} \approx 10^{-12} \times P_{\text{ANOVA}} \).

3.2. Variance-based adaptive ANOVA

Even with a sparse ANOVA expansion, the standard PDD expansion of the component functions of (8) can be made still more efficient. Blatman and Sudret [14] show that, for a large number of engineering problems, many polynomial terms in a gPC expansion are negligible in the constructed meta-model. Numerical results confirm that a sparse PDD representation can be effective [19]. The variance-based adaptive method originally proposed by Tang et al. [19] is employed in this work. Steps essential to the proposed generalization are summarized in this subsection.

We use LSR to construct a first-order ANOVA-based PDD expansion

\[
f_{\text{PDD, } m}^{(1)}(\mathbf{x}) = f_0 + \sum_{i=1}^{N} \left[ \sum_{j=1}^{m} c_{ij} \psi_j(x_i) \right],
\]

(14)

which has \( P^{(1)} = 1 + Nm \) polynomial terms. After choosing a truncation dimension \( \nu \), the full set of second- and higher-order PDD polynomial bases is constructed from the tensor product rule (8). The element polynomial index varies within the range of

\[
\left\{ 2 + Nm, \cdots , 1 + Nm + \binom{N}{2} m^2 + \cdots + \binom{N}{\nu} m^\nu \right\},
\]

(15)

where the first and last index corresponds to the first bi-variate polynomial and last \( \nu \)-variate polynomial, respectively.

The set (15) can be large even for relatively modest \( N, \nu, m \). To seek a high-accuracy model representation \( (\nu \geq 3, \ m \geq 5) \) with acceptable computational cost for problems with \( N \geq 20 \), we recommend to further reduce the size of (15) by using the concept of active dimension [19]. This requires retention of fewer parameters for the construction of set (15) by looking at each parameter’s variance contribution (second-order moment) in (14) represented by a summation of squares of coefficients. More in detail, choosing \( D < N \) to satisfy

\[
\sum_{i=1}^{D} \mathcal{S}_i \geq p, \quad \mathcal{S}_i = \frac{\sum_{j=1}^{m} \sum_{k=1}^{N} (C_{ij}^k)^2 \gamma_i^j}{\sum_{k=1}^{N} \sum_{j=1}^{m} (C_{kj}^i)^2 \gamma_k^i},
\]

(16)
and replacing \( N \) by \( D \) for any second- and higher-order ANOVA components, yields a reduced set
\[
\left\{ 2 + Nm, \ldots, 1 + Nm + \left( \frac{D}{2} \right)m^2 + \cdots + \left( \frac{D}{v} \right)m^v \right\}.
\]

The threshold \( p \) in (16) is a constant close to the unity (e.g. \( p = 0.999 \)). We also have assumed that \( \{S_i\} \) are monotonically decreasing with respect to \( i \). The rationale of using (16) is in that we exclude—for second- and higher-order ANOVA components—all parameters whose total variance contribution is small (less than 0.1% for \( p = 0.999 \)) in a first-order ANOVA expansion. As will be presented in Section 4, \( S_i \) is indeed the first-order variance-based sensitivity index. Experience [19] shows a smaller active dimension \( D < N \) is efficient for very high-dimensional applications. The present paper considers \( N = 7 \), and we here set \( D = N \) to explore the full dimensional space.

### 3.3. Adaptive least-squares regression

To keep the model size small (i.e. the system sparse), we augment the existing polynomial set (14) by one at a time recursively adding new polynomial candidates from the set of higher-order ANOVA functions, using (15) or (17) if using active dimension.

During this recursive procedure, we eliminate polynomials that are quantified as unimportant. To measure this relative importance we use a variance-based criterion. For this procedure, a LSR problem needs to be solved, which eliminates polynomial \( \Phi_{ij} \) satisfying
\[
\frac{(C_{ij})^2 \gamma_{ij}}{\sum_k (C_{ik})^2 \gamma_{ik}} < \theta,
\]
where \( \theta \) is a pre-selected threshold. Note \( \{\alpha_k, k = 0, \ldots, P - 1\} \) represents multi-indices corresponding to the PDD formulation. The multivariate normalization constant is \( \gamma_{ij} = E[\Phi_{ij}(\mathbf{x})] \). Thus, these nominally unimportant polynomials are eliminated following each LSR problem during the recursive procedure. Since we keep the ANOVA polynomial surrogate sparse enough during this adaptive step, the size of a regression problem is restricted in a controlled way, so the needed number of model evaluations according to (11) is kept low.

The resulting surrogate model can be evaluated quickly enough to facilitate Monte Carlo importance sampling and provide estimates of output statistics. The cost of the recursive resolution of the regression linear system is negligible compared to the deterministic model evaluations.

For evaluation of surrogate model accuracy, we use two estimates (see Appendix A and presentation by Blatman and Sudret [14]): one is empirical error \( R^2 \), and the other is leave-one-out cross validation \( Q^2 \). We recognized that \( R^2 \) can be misleading, especially for a surrogate model that overfits the training data. A \( Q^2 = 1 \) value indicates a perfect surrogate prediction performance, while \( Q^2 \approx 0 \) indicates poor model accuracy.

### 4. Variance-based sensitivity indices (Var-SI)

We include this section to link between the well-known Var-SI, which are predicated on independence, and correlation-related sensitivity measures that will be introduced and discussed in Sections 5 and 6.

For independent inputs, the total variance \( V(y) \) of output \( y = f(\mathbf{x}) \) is the summation of all partial variances
\[
V(y) = \sum_{T=1}^{N} \sum_{i_1 < \cdots < i_T} \text{Var}(f_{i_1, \cdots, i_T}).
\]

The global Var-SI [1–3] of an order-\( T \) component function \( f_{i_1, \cdots, i_T} \) is then defined by the ratio
\[
S_{i_1, \cdots, i_T} = \frac{\text{Var}(f_{i_1, \cdots, i_T})}{V(y)}.
\]

From (19), all Var-SI are non-negative and sum to unity. Furthermore, the variance-based total sensitivity index (Var-TSI) \( S_{i,T} \) of a single parameter \( x_i \) is estimated by adding all Var-SI containing \( x_i \). Both \( 0 \leq S_{i,T} \leq 1 \) and \( \sum_{i=1}^{N} S_{i,T} \geq 1 \) are guaranteed.

The Var-SI can be evaluated with MC or other sampling methods [2,3,5,48,49]. In our calculations, we can take advantage of the previously presented polynomial spectral expansion and carry out a post-processing of the expansion coefficients. Once the PDD coefficients are determined, the second-order moment and global Var-SI can also be obtained directly. Thus, it is straightforward to write the Var-SI by using the PDD expansion as [19]
\[
S_{i_1, \cdots, i_T}^{\text{Var}} = \frac{\text{Var}(f_{i_1, \cdots, i_T})}{\text{Var}(f_m(\mathbf{x}))} = \frac{1}{\sum_{k=1}^{P-1} (C_{\alpha_k})^2 \gamma_{\alpha_k}} \sum_{\alpha_j \subseteq \{i_1, \cdots, i_T\}} C_{\alpha_j}^2 \gamma_{\alpha_j}.
\]

The Var-TSI \( S_{i,T}^{\text{Var}} \) can be obtained simply by adding all the measured \( S_{i_1, \cdots, i_T}^{\text{Var}} \) whose index indicates inclusions of \( x_i \).
5. Standard covariance-based sensitivity indices (Cov-SI)

It is important to recognize that the uniqueness of the ANOVA expansions depends upon the independence of the samples [50]. Otherwise, the correlated samples in general lead to spurious terms in the decomposition [51,52] and total sensitivity estimates can be smaller than the contributing indices [29]. This is particular concern for our combustion problem, since the chemical reaction rate parameters are anticipated to be (and are) highly correlated so the Var-SI of (20) cannot be directly employed, since (21) cannot provide accurate variance or Var-SI. Component orthogonality is lost.

Kucherenko et al. [29] used (20) to define first-order Var-SI and Var-TSI for correlated inputs. These sensitivities are $\in [0, 1]$, and consistent with Sobol’ indices in limit of independence, but rely on the knowledge of conditional probability density function (pdf). With our combustion application with complex joint pdf, it is impractical to compute conditional pdf based integration.

Our approach uses and further develops the approach proposed by Li et al. [27], which is based on a covariance decomposition of the unconditional variance that defines three Cov-SI: the total, structural, and correlative contributions. This approach is compatible with our least-squares surrogate method since only a joint distribution is required, and one of our goals is to evaluate it for our challenging prediction problem. Knowledge of conditional pdf’s is not required when computing the ANOVA component functions. Computing component functions using surrogates is also more efficient than standard approaches that calculate the conditional variance using a large number of samples [27].

The covariance approach is attractive in that, with correlated inputs, the unconditional variance $V(y)$ can be decomposed as the sum of the covariances of each component function $f_{sj}$ ($s_j$ being a general multi-index for ANOVA expansion [26]) with the model output $y$:

$$ V(y) = \sum_{j=1}^{N} \left[ \frac{\text{Var}(f_{sj})}{V(y)} + \sum_{k=1, k \neq j}^{N} \frac{\text{Cov}(f_{sj}, f_{sk})}{V(y)} \right], \text{ with } N = 2^N = P_{\text{ANOVA}}. \quad (22) $$

Based on (22), the structural, correlative, and total Cov-SI can be defined in terms of the component functions $f_{sj}$:

$$ S_{sj}^{\text{cov}} = \frac{\text{Var}(f_{sj})}{V(y)}, $$

$$ S_{sj}^{b, \text{cov}} = \left[ \sum_{k=1, k \neq j}^{N} \frac{\text{Cov}(f_{sj}, f_{sk})}{V(y)} \right], $$

$$ S_{sj}^{\text{cov}} = S_{sj}^{\text{cov}, a} + S_{sj}^{b, \text{cov}} = \frac{\text{Cov}(f_{sj}, y)}{V(y)}, \quad (23) $$

where only the structural indices $S_{sj}^{\text{cov}, a}$ are strictly non-negative. Overall, they are only subject to the constraint

$$ \sum_{j}^{N} S_{sj}^{\text{cov}} = \sum_{j}^{N} \left( S_{sj}^{\text{cov}, a} + S_{sj}^{b, \text{cov}} \right) = 1. \quad (24) $$

Chastaing et al. [28] and Rahman [30] define similar covariance-based indices using a generalized ANOVA decomposition; Li and Rabitz [31] recently showed that Var-SI can be estimated from Cov-SI by using a modest number of input–output samples.

For independent inputs, it should be clear that (23) reduces to one single index that converges to Var-SI (20). While the traditional Var-SI have become accepted as quantifying relative importance of inputs, Cov-SI for correlated inputs warrant additional interpretation. Nevertheless, given their derivation, it is clear that large values indicate an important component function, either independently or correlative with other components. Thus, they provide a useful tool for developing uncertainty estimates.

6. Generalized covariance-based sensitivity indices (mCov-SI)

We illustrate a limitation of Cov-SI with a simple example, and provide our generalization to avoid it.

6.1. A bivariate example

The limitation we seek to address can be shown with a bivariate normal distribution $\mathbf{x} \sim \mathcal{N}(\mu, \Sigma)$ into a simple two-dimensional linear additive model,

$$ y = a_1 x_1 + a_2 x_2, \quad a_1 \geq a_2 > 0 \quad (25) $$

with

$$ \mu = \begin{bmatrix} 0 \\ 0 \end{bmatrix}, \quad \Sigma = \begin{bmatrix} 1 & \rho \\ \rho & 1 \end{bmatrix}, \quad -1 \leq \rho \leq 1. $$

Larger $a_1$ augments the importance of $x_1$ and vice versa.
Fig. 1. Analytical Cov-SI (23) for \(\alpha_1/\alpha_2 = 2\), which emphasizes the relative importance of \(x_1\).

Fig. 2. Cov-SI (23) for \(\rho = 1\).

6.1.1. Var-SI

Computing Var-SI by surrogate coefficients using (21) gives

\[
S_{1,\text{var}} = \frac{a_1^2}{a_1^2 + a_2^2}, \quad S_{2,\text{var}} = \frac{a_2^2}{a_1^2 + a_2^2},
\]

which are independent of the correlation coefficient \(\rho\). These indices show the expected behavior: for larger \(a_i\) the variable \(x_i\) is indicated as more important. Note these Var-SI estimates are both bounded by \([0, 1]\), and use only coefficient information, so they directly reflect the structure and are useful to analyze the surrogate structure.

6.1.2. Original Cov-SI

Information needed for the total variance should be computed using (22):

\[
V = a_1^2 + a_2^2 + 2\rho a_1 a_2 \in \left[ (a_1 - a_2)^2, (a_1 + a_2)^2 \right].
\]

For variable \(x_1\), the three Cov-SI are then

\[
S_{1,\text{cov}}^{a} = \frac{a_1^2}{V}, \quad S_{1,\text{cov}}^{b} = \frac{\rho a_1 a_2}{V}, \quad S_{1,\text{cov}}^c = \frac{a_1^2 + \rho a_1 a_2}{V},
\]

and for \(x_2\)

\[
S_{2,\text{cov}}^{a} = \frac{a_2^2}{V}, \quad S_{2,\text{cov}}^{b} = \frac{\rho a_1 a_2}{V}, \quad S_{2,\text{cov}}^c = \frac{a_2^2 + \rho a_1 a_2}{V}.
\]

These Cov-SI rightly depend on the correlation \(\rho\) and ratio \(\alpha_1/\alpha_2\). Fig. 1 illustrates this. Of course, \(\rho = 0\) recovers the Var-SI of (26).

If \(0 < \rho \leq 1\), all three Cov-SI are positive and \(\in [0, 1]\). The correlative contributions are equal with \(S_{1,\text{cov}}^{b} = S_{2,\text{cov}}^{b}\), and for \(\alpha_1 > \alpha_2\) then \(S_{1,\text{cov}}^{a} > S_{2,\text{cov}}^{a}\) and \(S_{1,\text{cov}}^c > S_{2,\text{cov}}^c\). The correct relative importance is thus quantified by the Cov-SI. For high positive correlation, the original Cov-SI are plotted in Fig. 2. When \(\alpha_1 \gg \alpha_2\), we have \(S_{1,\text{cov}}^{a} \rightarrow 1\), and all other
Cov-SI vanish. This success in reproducing these expected features of the sensitivity makes them attractive for quantifying sensitivity. Some limitations of Cov-SI are seen for \( \rho \in [-1, 0) \). For one, there are negative correlative contributions \( S_1^{a, cov} = S_2^{b, cov} < 0 \). Although we still have relative importance correctly ordered (\( S_1^{a, cov} > S_2^{b, cov} \), \( S_1^{cov} > S_2^{cov} \) for \( a_1 > a_2 \)), the case of \( \rho \to -1 \) (and \( a_1 > a_2 \)) yields

\[
S_1^{a, cov} > S_2^{a, cov} > 1, \quad S_1^{cov} > 1, \quad S_2^{cov} = 1 - S_1^{cov} < 0.
\]

Thus the importance ranking is consistent, though Cov-SI are no longer \( \in [0, 1] \). The original Cov-SI are plotted in Fig. 3. The limit case of \( a_1/a_2 \to \infty \) still yields \( S_1^{a, cov}, S_1^{cov} \to 1 \), and all other Cov-SI vanish, as expected for \( \rho \to 1 \). However, taking the limit of equal importance \( a_1 \to a_2 \), we have zero total variance \( V \to 0 \) and all Cov-SI diverge

\[
\left\{ S_1^{a, cov}, S_1^{b, cov}, S_1^{cov} \right\}_{a_1 \to a_2, \rho \to -1} \to \pm \infty.
\]

These characteristics limit the utility of these metrics. Although we illustrate this for a simple function, only uncertain parameters of comparable importance and negative correlations are required to realize the behavior. We have seen extremely large Cov-SI for uncertain kinetic parameters, which obfuscate interpreting sensitivity results. In order to overcome this, we propose a new variant of the Cov-SI.

### 6.2. mCov-SI

To avoid consequences of negative correlation, we modify the total variance formulation (22) to

\[
\tilde{V}(y) = \sum_{j=1}^{N} \text{Var}(f_{s_j}) + \left[ \sum_{k=1, k \neq j}^{N} \text{Cov}(f_{s_j}, f_{s_k}) \right],
\]

and re-define the corresponding structural, correlative, and total mCov-SI as

\[
\begin{align*}
\tilde{S}_j^{a, cov} &= \text{Var}(f_{s_j})/\tilde{V}(y), \\
\tilde{S}_j^{b, cov} &= \left[ \sum_{k=1, k \neq j}^{N} \text{Cov}(f_{s_j}, f_{s_k}) \right]/\tilde{V}(y), \\
\tilde{S}_j^{cov} &= \tilde{S}_j^{a, cov} + \tilde{S}_j^{b, cov} = \text{Cov}(f_{s_j}, y)/\tilde{V}(y).
\end{align*}
\]

The modified covariance-based total sensitivity index (mCov-TSI)

\[
\left\{ \tilde{S}_j^{a, cov}, \tilde{S}_j^{b, cov}, \tilde{S}_j^{cov} \right\}
\]

is the sum of all parts involving \( x_i \).

It should be clear that (32) is consistent with output variance (22) if and only if \( \sum_{k=1, k \neq j}^{N} \text{Cov}(f_{s_j}, f_{s_k}) \geq 0 \) for all \( f_{s_j} \), in which case the mCov-SI (33) are identical to the original (23). In general, we have \( \tilde{V}(y) \geq V(y) \). In case of independent model parameter inputs, (33) and (23) both converge to (21). A clear advantage of (33) for correlated inputs is that the magnitude of three mCov-SI is \( \in [0, 1] \), even when \( \tilde{V}(y) \to 0 \). In more detail, we have
\[ 0 \leq \tilde{S}_{a,j}^{\text{cov}} \leq 1, \quad 0 \leq \sum_j \tilde{S}_{a,j}^{\text{cov}} \leq 1, \]
\[ -1 < \tilde{S}_{b,j}^{\text{cov}} < 1, \quad -1 < \sum_j \tilde{S}_{b,j}^{\text{cov}} < 1, \]
\[ -1 < \tilde{S}_{j}^{\text{cov}} \leq 1, \quad 0 \leq \sum_j \tilde{S}_{j}^{\text{cov}} = V(y)/\tilde{V}(y) \leq 1. \]

The total contribution \( \tilde{S}_{j}^{\text{cov}} \) measures the overall \( f_{j} \) contributions to the variance of the model output, which can be positive or negative. It is thus a robust estimate of ANOVA subspace importance. We do recognize, however, that the total contributions do not sum to unity unless \( \tilde{V}(y) = V(y) \), though we do note that
\[
\sum_j \left( \tilde{S}_{j}^{\text{cov}} + \left| \tilde{S}_{j}^{\text{cov}} \right| \right) = 1,
\]
though \( \left( \tilde{S}_{j}^{\text{cov}} + \left| \tilde{S}_{j}^{\text{cov}} \right| \right) \) itself does not provide a useful sensitivity measure. To understand this limitation, consider \( y = x_1 + x_2 + x_3 \) with \( x \sim \mathcal{N}(0, \Sigma) \) and
\[
\Sigma = \begin{bmatrix}
1 & -1 & 0 \\
-1 & 1 & 0 \\
0 & 0 & 1
\end{bmatrix}.
\]

It is easy to see that the total contribution of \( x_1 \) and \( x_2 \) to \( V(y) \) is zero; \( x_3 \) solely contributes to \( V(y) \). However, the metric \( \left( \tilde{S}_{j}^{\text{cov}} + \left| \tilde{S}_{j}^{\text{cov}} \right| \right) \) would wrongly suggest that \( x_3 \) is unimportant.

6.2.1. The bivariate example revisited

To illustrate the properties and advantage of mCov-SI, we revisit the bivariate example (25) of Section 6.1. Using (33), Fig. 4a shows the mCov-SI that are no longer smooth in \( \rho \), compared to standard Cov-SI shown in Fig. 1. When \( \rho \) is positive and increases, mCov-SI remain identical to Cov-SI, and importance is correctly quantified. However, when \( \rho \) is negative,
mCov-SI are all bounded in \([0, 1]\), in contrast to Cov-SI in Fig. 1. Because of negative correlative contribution, total \(S_{1}^{\text{cov}}\) is smaller than structural \(S_{1}^{\text{cov},1}\) for both variables, and \(S_{1}^{\text{cov}}\) still indicates that \(x_{1}\) is more important than \(x_{2}\). For \(a_{1} = a_{2}\), Fig. 4b shows that mCov-SI can successfully quantify the equal importance in the entire \(\rho\) space with all indices bounded in \([-1, 1]\).

The mCov-SI for the limit case of \(\rho = -1\) for a varying \(a_{1}/a_{2}\) are further plotted in Fig. 5, compared to Fig. 3 showing Cov-SI. We see that mCov-SI do not diverge in the limit of equal importance. We notice the structural Cov-SI \(S_{1}^{\text{cov},1}\) (Fig. 3) increases while structural mCov-SI \(S_{1}^{\text{cov}}\) decreases. Total mCov-SI \(S^{\text{cov}}\) still indicate correct importance in the entire space of \(a_{1}/a_{2}\).

6.3. mCov-SI computation

A surrogate-based MC sampling method can be used to estimate the \(\tilde{S}\) values. Knowledge of joint pdf \(p(\xi)\), which generally is the same pdf used to generate samples for the surrogate construction, for correlated physical inputs is needed to generate samples. The transformation (1) provides a training set for the surrogate: \(x^{(i)}, i = 1, \ldots, M\). We use \(M = 10^5\). For \(i\)-th function output \(y^{(i)}\) and \(f_{s_{j}}(x_{s_{j}})\) as a \(T\)-dimensional subspace function \(f_{T_{s}}(x_{T})\), the \(i\)-th component function evaluation \(f_{s_{j}}(x_{s_{j}}^{(i)})\) can be computed by adding all corresponding multivariate polynomial terms:

\[
f_{s_{j}}(x_{s_{j}}^{(i)}) = \sum_{j_{r}} c_{j_{r}}^{i} y_{j_{r}}^{i} f_{T_{s}}^{j_{r}}(x_{s_{j}}^{(i)}).
\]

The variance and covariance can thus be computed for \(f_{s_{j}}\) as

\[
\begin{align*}
\text{Var}(f_{s_{j}}) &= \frac{1}{M - 1} \sum_{i=1}^{M} \left( f_{s_{j}}(x_{s_{j}}^{(i)}) - \bar{f}_{s_{j}} \right)^{2}, \\
\text{Cov}(f_{s_{j}}, y) &= \frac{1}{M - 1} \sum_{i=1}^{M} \left( f_{s_{j}}(x_{s_{j}}^{(i)}) - \bar{f}_{s_{j}} \right) (y^{(i)} - \bar{y}), \\
\sum_{k=1, k\neq j}^{N} \text{Cov}(f_{s_{j}}, f_{s_{k}}) &= \text{Cov}(f_{s_{j}}, y) - \text{Var}(f_{s_{j}}),
\end{align*}
\]

with

\[
\bar{y} = \frac{1}{M} \sum_{i=1}^{M} y^{(i)} \quad \text{and} \quad \bar{f}_{s_{j}} = \frac{1}{M} \sum_{i=1}^{M} f_{s_{j}}(x_{s_{j}}^{(i)}).
\]

Note in particular that unlike in a standard ANOVA expansion the mean of a component function \(\bar{f}_{s_{j}}\) is not zero here because of correlated input sampling.

For the variance of model output \(V(y)\), (22) should be used. Concerning mCov-SI, \(\tilde{S}^{\text{cov},1}, \tilde{S}^{\text{cov},2}, \tilde{S}^{\text{cov},3}\) can be readily estimated by their definitions (33). The mCov-TSI \(\tilde{S}_{1,1}^{\text{cov},1}, \tilde{S}_{1,1}^{\text{cov},2}, \tilde{S}_{1,1}^{\text{cov},3}\) can be computed by adding all indices that include \(x_{i}\).
7. A mathematical example

Before proceeding to application in Sections 9 and 10, we illustrate key properties with a linear model with correlated sampling from a multivariate normal distribution.

Consider

$$y = 5x_1 + 4x_2 + 3x_3 + 2x_4 + x_5,$$

with input vector $\mathbf{x} \sim \mathcal{N}(0, \Sigma)$ and covariance matrix

$$\Sigma = \begin{pmatrix}
1 & \rho & 0.2 & 0 & 0 \\
\rho & 1 & 0.2 & 0 & 0 \\
0.2 & 0.2 & 1 & 0 & 0 \\
0 & 0 & 0 & 0.2 & 1 \\
0 & 0 & 0 & 0 & 1
\end{pmatrix}.\quad(37)$$

Li et al. [27] studied this problem with $\rho = 0.6$ using the original Cov-SI (23).

The Var-SI (21) ascribe importance based on the square of coefficients, and thus indicate immediately the ranking of importance: $x_1 > x_2 > x_3 > x_4 > x_5$. As will be shown, mCov-SI inform different importance measure for different $\rho$. The analytical values of mCov-SI can be readily obtained as for the bivariate case in Section 6.1. Fig. 6 shows the three analytical mCov-SI versus $\rho$. A critical correlation $\rho_0 = -0.12$ can be obtained by setting $\text{Cov}(f_1, f_2) + \text{Cov}(f_2, f_3) = 0$ which corresponds to $S_2^{\text{b,cov}} = 0$. If $\rho \geq \rho_0$, mCov-SI function as the original Cov-SI. Otherwise, mCov-SI become smaller than their original counterparts.

The structural mCov-SI reflect directly the model coefficients, similarly as Var-SI. The greater a coefficient, the greater the corresponding $S_1^{\text{b,cov}}$. Correlative $S_1^{\text{b,cov}}, S_2^{\text{b,cov}}$ are greater than all other inputs with a high positive $\rho$, and decrease with a decreasing $\rho$. For $\rho \leq \rho_0$, they both become negative. These negative effects influence the total contributions of $x_1$ and $x_2$. For high positive $\rho$, $x_1$ is the most significant contributor, followed by $x_2$. Their importance decreases below $x_3$ for large negative $\rho$. For $\rho \to -1$, $x_2$ is indicated as the least important input, despite the fact that neither its structural nor correlative effect is the smallest. This importance assessment using the total measure $S_i^{\text{cov}}$ of mCov-SI is qualitatively confirmed by examining output $y$ against inputs $x_2, x_3$ in Fig. 7. Inputs are normalized using transformation (1). It is clear that $x_2$ produces more output variations than $x_3$ for $\rho = 0.8$, whereas $x_3$ is more important for $\rho = -0.8$.

If course, 5 first-order Hermite polynomials could serve as an effective PDD surrogate for this simple linear case. However, the goal is to treat arbitrary input distribution, we thus systematically use the transformation (1) to recover a uniform input distribution. In this case, the use of (1) makes the linear problem artificially nonlinear, and thus high-order Legendre polynomials are required. In this sense, this transformation also helps to test the performance. Table 1 shows the numerical results are in very good agreement with the analytical results. We have set $m = 4$ (8), and $\theta = 1 \times 10^{-5}$ (18). With only
Fig. 7. Sensitivity of model output (36) to $x_2$ and $x_3$.

(b) Output of (36) showing stronger sensitivity to $x_2$ with positive correlation of $\rho = 0.8$.

Fig. 7. Sensitivity of model output (36) to $x_2$ and $x_3$.

Table 1
Estimated and exact mCov-SI for (36).

<table>
<thead>
<tr>
<th>Estimate (33)</th>
<th>Exact</th>
</tr>
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<tbody>
<tr>
<td>$S_{\text{cov}}^1$</td>
<td>$S_{\text{cov}}^1$</td>
</tr>
<tr>
<td>$S_{\text{cov}}^2$</td>
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</tbody>
</table>

<table>
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<tr>
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<th>$\rho = -0.8$</th>
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<tbody>
<tr>
<td>$x_1$</td>
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</tr>
<tr>
<td>$x_2$</td>
<td>0.027139</td>
</tr>
<tr>
<td>$x_3$</td>
<td>0.170910</td>
</tr>
<tr>
<td>$x_4$</td>
<td>0.051893</td>
</tr>
<tr>
<td>$x_5$</td>
<td>0.016949</td>
</tr>
</tbody>
</table>

<table>
<thead>
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<th>$\rho = -0.6$</th>
<th>$\rho = -0.6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x_1$</td>
<td>0.202177</td>
</tr>
<tr>
<td>$x_2$</td>
<td>0.079404</td>
</tr>
<tr>
<td>$x_3$</td>
<td>0.184881</td>
</tr>
<tr>
<td>$x_4$</td>
<td>0.056819</td>
</tr>
<tr>
<td>$x_5$</td>
<td>0.018437</td>
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</table>

<table>
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<tbody>
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<td>$x_1$</td>
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</tr>
<tr>
<td>$x_2$</td>
<td>0.274771</td>
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<tr>
<td>$x_3$</td>
<td>0.180451</td>
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<tr>
<td>$x_4$</td>
<td>0.055138</td>
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<tr>
<td>$x_5$</td>
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<table>
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<th>$\rho = 0.6$</th>
<th>$\rho = 0.6$</th>
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<tr>
<td>$x_1$</td>
<td>0.433618</td>
</tr>
<tr>
<td>$x_2$</td>
<td>0.336064</td>
</tr>
<tr>
<td>$x_3$</td>
<td>0.15108</td>
</tr>
<tr>
<td>$x_4$</td>
<td>0.050798</td>
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<tr>
<td>$x_5$</td>
<td>0.013512</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$\rho = 0.8$</th>
<th>$\rho = 0.8$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x_1$</td>
<td>0.437015</td>
</tr>
<tr>
<td>$x_2$</td>
<td>0.359002</td>
</tr>
<tr>
<td>$x_3$</td>
<td>0.145919</td>
</tr>
<tr>
<td>$x_4$</td>
<td>0.044784</td>
</tr>
<tr>
<td>$x_5$</td>
<td>0.013280</td>
</tr>
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</table>
Thus, Q = 50 data points, the adaptive surrogate is able to produce an approximation with $R^2 > 0.998$ and $Q^2 > 0.993$ in all cases. The PDD model representation size is $P \in [18, 20]$. The computational cost based on number of function evaluations is significantly lower (by a factor of about 6) than with the methods of Li et al. [27]. This is despite the fact that we have made the problem, in a sense, more challenging by working with uniformly distributed random variables.

7.1. Summary of sensitivity indices

Based on this 5-dimensional model and the characteristics of the formulation, we summarize Var-SI (21) versus mCov-SI (33).

- Structural mCov-SI $\tilde{S}^{\text{cov}}$ functions similarly as Var-SI $S^{\text{var}}$. Both rely on the variance contribution of each ANOVA component function to the output’s total variance, and they are identical for independent inputs. With correlated inputs, variation of $\tilde{S}^{\text{cov}}$ exists in $\rho$ (Fig. 6). This variation is neglected by Var-SI $S^{\text{var}}$ using (21) because correlated effects are neglected. For this particular model, because the variation of $\tilde{S}^{\text{cov}}$ does not yield a different parameter ranking, Var-SI and the structural mCov-SI provide the same characterization. However, in general structural mCov-SI $\tilde{S}^{\text{cov}}$ provides more reliable variance-based sensitivity estimates than Var-SI (21).
- Three indices of mCov-SI (structural $\tilde{S}^{\text{cov}}$, correlative $\tilde{S}^{\text{cov}}$, and total $\tilde{S}^{\text{cov}}$) provide a complete description of an ANOVA subspace’s contribution to output’s total variance. The total mCov-SI is the most reliable sensitivity measure among three, since it accounts for the total contribution (see (33)). We note that it can happen that structural mCov-SI and total mCov-SI give the same parameter ranking, as for this 5-dimensional case with a positive $\rho$ (Fig. 6). However, when correlative effects are strong enough among ANOVA components, total and structural mCov-SI can indicate different parameter importance, as shown in Fig. 6 for $\rho < -0.5$.

Thus, for the subsequent analysis for our combustion application, mCov-SI and mCov-TSI are used for UQ, sensitivity analysis, and dimension reduction.

8. Combustion model & sources of uncertainty

8.1. Governing equations

The combustion systems we consider are represented by the single-fluid multi-species Navier–Stokes equations with corresponding sources,

$$
\begin{align*}
\frac{\partial \rho}{\partial t} + \frac{\partial \rho u_i}{\partial x_i} &= 0, \\
\frac{\partial \rho u_i}{\partial t} + \frac{\partial}{\partial x_j} \left( \rho u_i u_j + p \delta_{ij} - \tau_{ij} \right) &= S_{M,i}, \\
\frac{\partial \rho E_T}{\partial t} + \frac{\partial}{\partial x_j} \left( \rho E_T + p \right) u_j + q_j - \sum_i \rho u_i \tau_{ij} &= S_T, \\
\frac{\partial \rho Y_s}{\partial t} + \frac{\partial}{\partial x_j} \left( \rho Y_s u_j + \rho Y_s \nabla Y_s \right) &= \omega_s, \quad s = 1, \ldots, N_{sp} - 1,
\end{align*}
$$

(38)

where $i$ and $j$ index coordinate directions and summation is assumed over repeated $j$. In (38), $u_i$ is the flow velocity, $\rho$ the mass density, $p$ the pressure, $q_j$ the heat flux, $\tau_{ij}$ the viscous stress, and $V_s$ is the diffusion velocity of species $s$ with $N_{sp}$ the total number of species.

The mass-specific total energy

$$
E_T = \frac{1}{2} u_j u_j + \sum_{s=1}^{N_{sp}} h_s Y_s - \frac{p}{\rho}
$$

is based on mass fractions $Y_s$, and mass-specific enthalpy,

$$
h_s = \Delta h_{f,s}^0 + \int_{T_0}^{T} c_{p,s}(\tau) \, d\tau,
$$

(39)

includes reference enthalpy of formation, $\Delta h_{f,s}^0$ at $T_0 = 298.15$ K. The gas is assumed ideal, with

$$
p = \rho \left( \sum_{s=1}^{N_{sp}} \frac{Y_s}{W_s} \right) T.
$$
where $R_i$ is the gas constant and $W_s$ is the molar mass of species $s$. These models for the gas properties are expected to be accurate at the not-too-extreme conditions we consider and are thus considered to include only trusted parameters.

Important uncertainties are anticipated for the reaction mass source $\dot{\omega}_s$. $S_T$ models the thermal contribution of the Laser Induced Breakdown (LIB) that seeds ignition, and $S_M,i$ is the vortical contribution of the LIB. The model parameter uncertainties considered in this work mainly come from these source terms. The modeling input uncertainties will be presented in the following subsections.

The system (38) is discretized by high-order finite-difference methods on structured grids using the software PlasComCM. The numerical techniques and transport formulae $(\tau_{ij}, q_i, V_{i,j})$ are accurate and appropriate though standard and therefore we rely on previous publications regarding their details [53–57].

8.2. Chemical kinetics

The system of reactions is

$$
\sum_{s=1}^{N_{ip}} v'_{s,r} \mu_s = \sum_{s=1}^{N_{ip}} v''_{s,r} \mu_s, \quad r = 1, \ldots, N_{re},
$$

(40)

where $\mu_s$ represents the $s$-th species, $N_{re}$ is number of reactions, and $v'_{s,r}$ and $v''_{s,r}$ are stoichiometric coefficients in reaction $r$.

The reaction source $\dot{\omega}_s$ for species $s$ in (38) is

$$
\dot{\omega}_s = W_s \sum_{r=1}^{N_{re}} (v''_{s,r} - v'_{s,r}) \left[ k_r(T) \prod_{j \in \text{reactants}} \left( \frac{\rho_j V_j}{W_j} \right)^{\eta_{j,r}} \right],
$$

where $k_r(T)$ is the rate

$$
k_r(T) = A_r T^{\beta_r} \exp \left( -\frac{E_r}{R_u T} \right).
$$

The model includes three reactions with relatively slow time scales to facilitate computations coupled with large-scale flow simulations:

$$
\begin{align*}
H_2 + M & \rightarrow 2H + M, \\
2H + M & \rightarrow H_2 + M, \\
2H_2 + O_2 + H & \rightarrow 2H_2O + H,
\end{align*}
$$

(41)

among which we consider the uncertainties arising from only the third reaction. The other two are detailed reactions and therefore have rates that are based on specific experimental measurements, as discussed by Massa and Freund [56]. As a global reaction, its reaction orders are set to

$$
\eta_{H_2} = 0.544; \quad \eta_{O_2} = 0.945; \quad \eta_{H} = 0.110,
$$

based on a separate, auxiliary calibration of {ln $A_3$, $b_3$, ln $E_3$, $\eta_{H_2}$, $\eta_{O_2}$, $\eta_{H}$} using the present configuration. However, for the present calibration involving only {ln $A_3$, $b_3$, ln $E_3$} as uncertain parameters, the values of {ln $\eta_{H_2}$, $\eta_{O_2}$, $\eta_{H}$} are fixed to the maximum a posteriori (MAP) point of the auxiliary calibration. This reduces the dimensionality of the propagation problem and was deemed acceptable because, for an autoignition simulation governed by the kinetics, the uncertainty of the reaction orders is redundant with that of the Arrhenius parameters, which have been identified to introduce significant parametric uncertainty. With this assumption only the Arrhenius parameters $A_3$, $b_3$, $E_3$ are treated as uncertain parameters. These are calibrated against constant-pressure autoignition simulations using the GRI-Mech 3.0 detailed chemical mechanism [58]. Because of their mathematical form and the consequences on the predicted rates, following previous work [59], we calibrate ln $A_3$ and ln $E_3$ rather than $A_3$ and $E_3$. Assuming zero observation error and an additive Gaussian error model for the model-data discrepancy implies the following relation between the model output $y_k$ and calibration data $d_k$:

$$
d_k = \gamma_k(\kappa) + \delta(\sigma): \quad \delta \sim N(0, \sigma^2),
$$

(42)

where $\kappa \equiv \{\ln A_3, b_3, \ln E_3\}$ are the calibrated kinetic parameters. The data are assumed to be statistically independent, so only one inadequacy parameter $\sigma$ is included.

Each scenario $x_k$ specifies the initial conditions—temperature, radical mass fraction, and equivalence ratio—of a zero-dimensional simulation:

$$
x_k = \{T_0^{(k)}, y_{H,0}^{(k)}, \phi_0^{(k)}\}; \quad k = 1, \ldots, N_d.
The above were chosen as scenario variables in order to represent local conditions of the LIB ignition seed, which acts as a thermal and radical source. To quantify the uncertainty of predictions near an ignition threshold, we include scenarios that correspond to both igniting and non-igniting cases. An igniting case is defined as cases for which the temperature \( T(t) \) reaches its inflection point before \( t = t_f = 10^{-3} \) s. To map out the ignition threshold in scenario space, we consider 5 uniformly spaced values of \( T_0, Y_{H,0}, \) and \( \phi_0 \) in the ranges

\[
T_0 \in [900K, \ 1200K], \quad \log_{10} Y_{H,0} \in [-12, -4], \quad \phi_0 \in [0.8, 1.2],
\]

for a total of \( N_d = 125 \) calibration scenarios. This range of conditions were also chosen to overlap with the local conditions near the flame front in the standburner simulation, as ignition of the gas occurs in that region of the flow. The calibration target is

\[
y_k(\kappa) = \int_0^{t_f} T(t; \kappa, x_k) \, dt,
\]

which serves as an indicator of ignition and is plotted for a range of relevant scenarios in Fig. 8.

The inadequacy parameter \( \sigma \) is calibrated along with the kinetic parameters \( \kappa \). The relation given by (42) yields the following likelihood function:

\[
p(\mathbf{d} | \kappa, \sigma) = \frac{1}{(\sigma \sqrt{2\pi})^{N_d}} \exp \left[ -\frac{1}{2\sigma^2} \sum_{k=1}^{N_d} (y_k(\kappa) - d_k)^2 \right].
\]

The prior distribution is taken to be uniform but with a short-time-scale restriction. This is based on the details of the three-dimensional turbulent flow simulation [56]. Rate parameters that require time step of \( \Delta t < 10^{-5} \) s are precluded by restricting the calibration space (specifying restrictions on the prior for this model) as

\[
p(\kappa, \sigma) = 1, \text{ if } \begin{cases} 
\ln A_3 < 41.5, \text{ and} \\
\ln E_3 < 10.5, \text{ and} \\
b_3 < -6.6667 \times 10^{-10} (\ln E_3)^2 + 6.5 \times 10^{-5} \ln E_3 - 3.3583, \text{ and} \\
\sigma > 0,
\end{cases}
\]

\[
p(\kappa, \sigma) = 0 \text{ otherwise},
\]

where \( A_3 \) is in MKS units and \( E_3 \) is in units of J/kmol. These criteria are based on empirical analysis of the parameters.

Bayesian calibration is performed by sampling the posterior distribution

\[
p(\kappa, \sigma | \mathbf{d}) \propto p(\mathbf{d} | \kappa, \sigma) p(\kappa, \sigma)
\]

using a Delayed Rejection Adaptive Monte Carlo (DRAM) method [60] with 5 delayed stages and \( 10^6 \) samples. Fig. 9 shows that the sample chain for \( b_3 \) is well-mixed; the other calibrated parameters displayed similar behavior.

Fig. 10 shows the marginal pdf of these parameters, which are well-approximated using Beta distributions in order to facilitate the input transformation (1).
8.3. Laser Induced Breakdown (LIB) ignition seed

Ignition is seeded with a laser-induced plasma breakdown, which is modeled via the source term $S_T$ in the governing equations (38), which introduces additional model parameters. Though this will have an essential thermal source, visualizations of isolated breakdowns suggest significant rotation and mixing, which potentially affects the ignition seed. The modeled vorticity source [56], the term $S_{M,i}$ in (38), introduces additional uncertain parameters. These and their corresponding uncertainty are discussed in the following sections.

8.3.1. LIB: thermal source

Based on space- and time-resolved experimental measurements of axisymmetric breakdowns in pure air, we select the teardrop-shaped fit of the luminous region visualized in Fig. 11. We assume that the deposition of thermal energy $S_T$ follows this same pattern, with functional form

$$
S_T(x, t) = E_{abs} W_T(t) f_T(x),
$$
$$
f_T(x) = L_r(r) L_y(y) F,
$$
$$
W_T(t) = \frac{1}{2\delta t} \left[ \tanh \frac{t}{t_r} - \tanh \frac{t - \delta t}{t_r} \right],
$$

where we have used a cylindrical coordinate system $(r = \sqrt{x^2 + z^2}, \varphi, y)$, with $y$ the laser axial direction. This is shown in Fig. 12.

The total power transferred from the laser to the gas, $E_{abs}$, was deduced from the laser power pre- versus post-breakdown as averaged over 3160 measurements. The variation among trials was relatively small, with a nearly Gaussian distribution such that 95% of the measurements were within $17.6 \pm 6.2$ mJ; for propagating uncertainty we assume

$$
E_{abs} \sim \mathcal{N}(17.6 \text{ mJ}, 3.1^2 \text{ mJ}^2),
$$

which is shown with the data in Fig. 13.
Fig. 11. Broad-banded light emission from a laser-induced breakdown in air for a 520 nm wavelength laser with the focal length is 250 mm and the incident power 50 mJ/pulse [56].

Fig. 12. Fit of the luminosity region of the LIB [56]. $L_{TW} = 1.986$ mm and $D_{TW} = 0.672$ mm are the length and width of the luminosity region.

Fig. 13. Distribution of measured LIB energy $E_{abs}$.

The time envelop $W_T(t)$ in (43) is modeled as symmetric, with $t_r = 5 \times 10^{-6}$ s and $\delta_t = 2.5 \times 10^{-4}$ s. The fitted profiles $L_r$ and $L_y$ in the space factor $f_T(x)$ are

$$L_r = \sum_j^4 a_{r,j} \exp \left[ -\frac{(r - b_{r,j})^2}{c_{r,j}^2} \right],$$  \hspace{1cm} (45a)$$

$$L_y = \sum_j^4 a_{y,j} \exp \left[ -\frac{(y - b_{y,j})^2}{c_{y,j}^2} \right],$$  \hspace{1cm} (45b)$$

with parameters given in Table 2. The corresponding fit is visualized in Fig. 12. As a normalization factor to satisfy $\int f_T(x) \, dx = 1$, the factor $F$ is proportional to $L_{TW} D_{TW}^2$ [56] with $L_{TW}$ and $D_{TW}$ the length and diameter of the observed luminosity profile.
Table 2
Tabulated values for the luminosity curve-fit \([45]\).

<table>
<thead>
<tr>
<th>(i)</th>
<th>(a_{i,1})</th>
<th>(b_{i,1})</th>
<th>(c_{i,1})</th>
<th>(a_{i,2})</th>
<th>(b_{i,2})</th>
<th>(c_{i,2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.3474</td>
<td>0.5</td>
<td>0.0537</td>
<td>0.7115</td>
<td>0.8592</td>
<td>0.08489</td>
</tr>
<tr>
<td>2</td>
<td>1.427</td>
<td>19.72</td>
<td>1.434 \times 10^4</td>
<td>0.2481</td>
<td>0.943</td>
<td>0.03945</td>
</tr>
<tr>
<td>3</td>
<td>-1.407</td>
<td>0.02523</td>
<td>0.4204</td>
<td>0.2837</td>
<td>0.7523</td>
<td>0.1307</td>
</tr>
<tr>
<td>4</td>
<td>-1.407</td>
<td>0.9747</td>
<td>0.4203</td>
<td>0.4878</td>
<td>0.6014</td>
<td>0.2105</td>
</tr>
</tbody>
</table>

That the laser power distribution matches the luminosity is a model, and \(R_{TW} = D_{TW}/2\) and \(L_{TW}\) are considered to be uncertain parameters for purposes of propagating uncertainty to model predictions. A uniform distribution is assumed for each of these inputs. The mean measurements \(\mu_{L_{TW}} = 1.986 \text{ mm}\) and \(\mu_{R_{TW}} = 0.336 \text{ mm}\) are assumed to be in their means, while a range of variation \(\pm 50\%\) is selected to reflect a relatively large uncertainty in how the energy is actually deposited \([61]\):

\[
L_{TW} \sim \mathcal{U}(0.5\mu_{L_{TW}}, 1.5\mu_{L_{TW}}),
\]

\[
R_{TW} \sim \mathcal{U}(0.5\mu_{R_{TW}}, 1.5\mu_{R_{TW}}).
\]

In a worst-case scenario analysis, the power density deposited per unit length in the irradiation direction can be assumed constant, allowing us to associate the shot-to-shot variability of the energy deposited (approximately 50\% \([62]\)) to the uncertainty measure for \(L_{TW}\). The initial \((t < 100 \mu\text{s})\) radial extension of the region of deposited energy is linked to the width of the beam waist \([61]\), but the relationship between the radius of the fluence volume (the volume where the fluence is above the threshold to generate plasma in air) and the power deposited is strongly nonlinear \([63]\). In the spirit of a worst-case scenario analysis, we use the energy-shot variability as uncertainty measure for \(R_{TW}\) as well.

8.3.2. LIB: vorticity source

The principal uncertain parameter in the vortical contribution is a Mach number \(M_v\), which parameterizes effects associated with the formation and propagation of non-spherical shocks \([56,64]\). LIB images, such as from Glumac et al. \([61]\), show a strong temperature wave propagating with speed \(\approx 2500\) m/s from the nominal focal point towards the laser source. Thus, shock formation is thought to be phenomenologically similar to that of a detonation \([65]\), with synchronization of the thermal and shock waves. This is assumed to strengthen the shock propagating towards the laser source though not the one propagating away from it.

Shock-generation of vorticity is due to two primary contributions: shock-curvature and baroclinic torque \([66]\). Baroclinic torque is expected to derive primarily from the gradients in the density ahead of the shock. However, the present shock is expected to encounter primarily unheated fluid, so we focus only on the effects of shock curvature.

Following Kevlahan \([66]\), we assume a parabolic shock \(y/R_{TW} = -(r/R_{TW})^2/2\) with \(R_{TW} = D_{TW}/2\). We also assume a steady translation of the shock in the \(y\) direction. The vorticity is \([56]\)

\[
\omega = \frac{4M_g r (M_g^2 + r^2 - 1)^2}{(r^2 + 1)^2 ((\gamma - 1)M_g^2 + 2(r^2 + 1))((\gamma + 1)M_g^2 + 4r^2)},
\]

where \(M_g\) is the normal velocity Mach number based on the free-stream speed of sound. \(M_v\) is a sub-grid-scale parameter, which will be used to inform \(M_v\).

Assuming steady flow \(\nabla \cdot (\rho \vec{u}) = 0\) in the frame of the shock, with \(\rho = \rho (r)\), axisymmetry and \(y\)-independence yields zero radial velocity \(u_r = 0\), and thus an axial velocity source \(\frac{\partial v}{\partial r} = \omega\). Integrating in \(r\), the parabolic shock produces an axial jet with velocity \(v\):

\[
(v - v_0) [(\gamma - 1)^2(\gamma + 1)M_0] = \frac{2(\gamma - 1)M_0^2}{r^2 + 1} - (\gamma + 1)(\gamma + 1)^2 \ln (\gamma - 1) + 2(\gamma + 2) + 4\gamma \ln (r^2 + 1).
\]

The constant \(v_0\) is determined so that no net momentum is transferred to the fluid \([56]\).

The physics underlying the parameter \(M_0\) is associated with coupling between fluid mechanics and energy deposition during the \(\sim 20\) ns laser pulse. The detonation-like wave theory of laser induced breakdown relates the shock speed to the interaction between shock heating and absorption of laser energy by the ionized gas behind the shock, which acts as a piston to support a fast moving detonation \([67]\). This theory predicts \(M_0 \approx 18\), so the shock residence time is much smaller than the ignition time scales. Thus, we consider a larger representative volume to model vorticity deposition and an averaging procedure to represent the contribution of the shock-passing from the laser scale to the resolved scale in the flow simulation. This leads to a nominal Mach number \(M_e = 2\) \([56]\), with uncertainty that will be accounted for. Vorticity deposition is sensitive to \(M_v\) and it was observed in simulation tests that \(M_e = 3\) segments the stoichiometric surface in a manner that was not observed \([61,68]\). Thus the interval is taken to be uniform

\[M_e \sim \mathcal{U}(1, 3).\]
Table 3
Uncertain model parameters.

<table>
<thead>
<tr>
<th>Kinetics</th>
<th>Controls</th>
<th>LIB</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\zeta_1$</td>
<td>$\ln A_3$</td>
<td>Calibrated pdf (see text)</td>
</tr>
<tr>
<td>$\zeta_2$</td>
<td>$b_3$</td>
<td>$\zeta_3$ $\ln E_3$</td>
</tr>
<tr>
<td>$\zeta_4$</td>
<td>$L_{TW}$</td>
<td>$\sim U(0.993, 2.979)$ [mm]</td>
</tr>
<tr>
<td>$\zeta_5$</td>
<td>$R_{TW}$</td>
<td>$\sim U(0.168, 0.504)$ [mm]</td>
</tr>
<tr>
<td>$\zeta_6$</td>
<td>$M_e$</td>
<td>$\sim U(1.3)$ [-]</td>
</tr>
<tr>
<td>$\zeta_7$</td>
<td>$E_{abs}$</td>
<td>$\sim N(17.642, 3.115^2)$ [mJ]</td>
</tr>
</tbody>
</table>

Fig. 14. 100 input kinetic samples used for UQ analysis.

The uniform distribution reflects the fact that vorticity generation in the detonation-like regime of laser breakdown is still not well understood, thus the anticipated model error is a combination of model deficiency and data uncertainty, for which a most probable value does not exist.

The vorticity is mollified in time as for the energy source term:

$$S_{M,k} = \rho v(r) W_T(t).$$

The density $\rho$ is taken as the instantaneous value from the flow solver, which only changes significantly subsequently, when there is a successful ignition.

8.4. Synopsis of multiphysics uncertain parameters

We summarize the 7 uncertain parameters in Table 3, and denote them collectively as

$$\zeta = \left( \ln A_3, b_3, \ln E_3, L_{TW}, R_{TW}, M_e, E_{abs} \right),$$

with their corresponding transformed uniform variables $\xi \in [0, 1]^7$ using (1):

$$\xi = (F_1(\zeta_1), \ldots, F_7(\zeta_7)).$$

When choosing the training set size $Q$ for subsequent sensitivity analysis and uncertainty quantification, we have two principal considerations: (1) $Q$ should be as small as possible, since it sets the computational cost, and (2) it should be larger than the surrogate model size and make sure the least-squares matrix is well-conditioned. Experience (e.g. Tang et al. [19]) suggests that $Q = 100$ is a reasonable number for a 7-dimensional problem, though we remain mindful that this might need to be increased to avoid ill-conditioning. Fig. 14 shows 100 samples for the kinetics parameters, which have been selected from the DRAM chain and are representative of the distribution curve in Fig. 10. We see that these are moderately correlated following the DRAM calibration.
For the other parameters, aside from the kinetics parameters, we use a standard nested Sobol’ sequence [37]. Input samples of \( \ln E_3 \), \( M_e \), and \( E_{\text{abs}} \) are shown in Fig. 15 as scatter points. As expected, there is no strong correlation in these transformed coordinates.

9. Standburner: an axisymmetric model system

9.1. Axisymmetric model configuration

We introduce a model combustion system to demonstrate our methods and further reduce the dimensionality of the uncertain space. It is attractive because it requires significantly less simulation time than the full three-dimensional flow to be considered in the following section, though it is realistic and does correspond to a particular experimental setup, which should be well modeled by our axisymmetric constraint.

The geometry and dimensions are shown in Fig. 16. Full details of this configuration and experiments are reported elsewhere [62,69], including analysis of the effects of a co-annular dielectric-barrier discharge. The fuel (H\(_2\)) is injected through a diameter 4.826 mm orifice at 1 L/min at ambient conditions.

The full computational domain to represent this setup extends from the origin, centered on the orifice, out to \( r = 40 \) mm and \( y = 162.5 \) mm. It also extends into the fuel delivery tube. For the particular tests we examine a steady state is reached before ignition is attempted. To establish this in our simulations, fuel is delivered in a cold and otherwise quiescent air until oxidizer-to-fuel equivalence ratio

\[
\phi = \frac{Y_{O_2}}{8Y_{H_2}}
\]
is stationary. Fig. 17 shows this stabilized flow. To seed ignition, the LIB model is imposed in the stabilized fuel/air mixture, and at \( y = 103.40 \) mm on the \( r = 0 \) axis based on corresponding experiments [62,69].

9.2. Quantity of Interest (QoI)

Our specific QoI used to quantify ignition is the average temperature weighted to the location of the stoichiometric surface at the time of the LIB:

\[
\mathcal{J}_T = \frac{1}{T_0} \int T(\mathbf{x}, t) w(\mathbf{x}, t_0) \, d\mathbf{x},
\]

where \( T_0 \) is a normalization factor related to cold flow average temperature.
Fig. 16. Two-dimensional burner experiment for axisymmetric simulations with dimensions in millimeters [62,69].

Fig. 17. Axisymmetric computational domain and stationary pre-LIB flow: (a) the mixture density $\rho$, (b) hydrogen mass fraction $Y_{H_2}$, and (c) vertical flow velocity $V$. The white contour represents the stationary stoichiometric surface computed by letting $\phi = 1$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

\[ T_0 = \int T(x, t_0) w(x, t_0) \, dx. \]  

(51)

$\phi$ is the oxidizer-to-fuel equivalence ratio, and

\[ w(x, t_0) = \frac{1}{\sigma_0 \sqrt{2\pi}} \exp \left( -\frac{(\phi - 1)^2}{2\sigma_0^2} \right), \]  

(52)

where $\sigma_0 = 0.001$. It should be clear that $J_T \approx 1$ indicates a temperature field close to that of cold flow and a failed ignition. For ignited samples $J_T$ is much larger.

9.3. Uncertainty propagation and sensitivity analysis

9.3.1. Demonstration of ignition and non-ignition

The 100 input sample parameters of Figs. 14 and 15, are used in axisymmetric flow model with laser ignition seed and combustion. The temperature field for a typical ignition case is shown in Fig. 18. For any non-ignited case, the high-temperature region diffuses before reactions become significant.

The (50) results are shown in Fig. 19. Some of the general trends seen in these samples can be anticipated. For example, we observe that stronger or more narrowly focused LIB (greater $E_{\text{abs}}$ or smaller $R_{TW}$), makes ignition more likely. It turns
out that greater value of Mach number parameter $M_e$ makes is more difficult for ignition to happen. This is because of extinction due to the corresponding increased local strain. However, if ignition does occur in such cases, say due to larger $E_{abs}$, vorticity-driven mixing accelerates the initial flame propagation.

9.3.2. Surrogate construction

To build a sparse polynomial surrogate via the methods of Sections 2 and 3, several methodology parameters need to be specified, which have been extensively studied for mathematical model problems [19]. Here we assess sensitivity to the ANOVA order $\nu$ of (12) and polynomial degree $m$ of (8). This is important for confirming that our surrogate is insensitive
to the specific choices made in its construction. We take \( \nu = 2 \) and \( m = 2, \ldots, 8 \), and the threshold for selection of important polynomials is set to \( \theta = 2 \times 10^{-4} \) from (18). Two measures of surrogate approximation error \( 1 - R^2 \) (A.2), \( 1 - Q^2 \) (A.7) and the size of the sparse representation \( P_{\text{sparse}} \) are reported in Fig. 20. The error estimates show that accounting for third-order interactions makes the surrogate quality significantly better. We then use \( \nu = 3 \) for the axisymmetric flow, and neglect \( \nu = 4 \) terms because the error for \( \nu = 3 \) is deemed sufficiently small.

9.3.3. Surrogate output: ignition probability

The prediction output with uncertainty distribution can be easily approximated by performing MC (or quasi-MC) importance sampling of inputs and propagating them through the surrogate. Fig. 21 shows the pdf/cdf, with the kinetics parameter samples selected from a well-mixed DRAM sample chain. The (50) QoI has \( f_T \gtrsim 1 \), though our surrogate procedure is not so constrained, and in Fig. 21 we see that it can yield negative values. However, this has no consequence since these cases can be interpreted as unambiguously non-igniting cases. The distribution of the surrogate output indicates an ignition probability of approximately 60%, which agrees with the training data.

9.3.4. Sensitivity analysis & dimension reduction

Sensitivity analysis will be used to reduce the number of uncertain parameters that will be propagated through our three-dimensional predictions in Section 10. This is important since the computational cost of a single simulation is significantly greater. Each axisymmetric sample using 250k mesh-points requires 256 CPU core-hours on a computer cluster with compute nodes having two eight-core 2.6GHz Intel Sandybridge processors, 64 Gbytes of memory, and a high-speed Infiniband interconnect network.

For dimension reduction, we will use mCov-SI (33) and mCov-TSI (34). Fig. 22 compares the structural and correlative mCov-SI for the 63 components \( (m = 5) \) of the ANOVA expansion of order \( \nu = 3 \), and Fig. 23 shows the corresponding total mCov-SI. We see that the high-order interactions are comparable to some of the first-order interactions. In order to select the most important parameters for the three-dimensional case, we compare the total sensitivity indices that are presented in Fig. 24 for structural and correlative mCov-TSI and in Fig. 25 for total mCov-TSI. As expected, the structural mCov-TSI \( \hat{S}^{(0)}_{I,T} \) are consistent with Var-TSI (21) (not shown), since structural mCov-TSI provide ranking based on variance of each ANOVA component, just as Var-TSI. Concerning the correlative mCov-TSI \( \hat{S}^{(c)}_{I,T} \), only the contributions of parameters \( b_3 \) and \( \ln E_3 \) are significant and negative. It is interesting to notice that the input correlation between \( b_3 \) and \( \ln E_3 \) is also the most important (Fig. 14). Combining \( \hat{S}^{(0)}_{I,T} \) and \( \hat{S}^{(c)}_{I,T} \) the total mCov-TSI \( \hat{S}^{(T)}_{I,T} \) shown in Fig. 25 provide the sensitivity tool we use for dimension reduction. Overall, \( \ln E_3 \) and \( b_3 \) contribute the most parametric uncertainty. Also, \( \ln A_3 \) has more impact than the LIB parameters \( L_{TW}, R_{TW}, M_e, E_{abs} \), however its magnitude is comparable to LIB parameters. Since \( \ln A_3 \) is also correlated with \( \ln E_3 \) and \( b_3 \), it is thus deemed important to propagate \( \ln E_3, b_3, \ln A_3 \) through three-dimensional simulations.
Fig. 22. First-, second-, and third-order structural and correlative mCov-SI (33).

Fig. 23. First-, second-, and third-order total mCov-SI (33).

Fig. 24. Structural and correlative mCov-TSI (34).

Fig. 25. Total mCov-TSI (34).
The main outcome of this axisymmetric case is that we have determined useful \( m \) and \( \nu \) (Fig. 20) and that a third-order ANOVA (\( \nu = 3 \)) provides in general an accurate surrogate. Also, global sensitivities support reduction from 7 uncertain parameters to 3, allowing us to focus only on the kinetic parameters (\( \ln A_3, b_3, \ln E_3 \)).

10. Turbulent combustion application

The specific configuration was developed in laboratory experiments [62]. Our primary aim is to add uncertainty to the predictive simulation methodology developed for this [56,57]. Fig. 26 shows the simulation domain. It is a sub-section of an overall \( 35 \times 38 \) cm\(^2\) test section downstream of a 25:1 contraction ratio wind tunnel. For the particular cases we target, the wind tunnel flows air at 15 m/s. A 40-grit sandpaper strip at the beginning of the test section initiates a rapid transition to a turbulent boundary layer. At \( x = 0.28 \) m downstream from the trailing edge of the sandpaper, the round hydrogen jet, introduced in Section 9 exhausts normal to the wall with a mean velocity of 1 m/s. With the mixing fuel and oxidizer cold-flow statistically stationary, a laser beam is focused from above on the axis of the fuel orifice to potentially seed ignition. If ignition is sustained, the flame propagates towards the jet orifice and anchors at its upstream edge.

The governing flow equations and LIB model are the same as introduced in Section 9. The computational domain is discretized with the locally structured overset grid system shown in Fig. 27. There are 3 grids to discretize the boundary layer in Cartesian coordinates (Grid 1: \( 1025 \times 257 \times 97 \)), the jet region in cylindrical coordinates (Grid 2: \( 133 \times 385 \times 65 \)), and a Cartesian grid located at the center of the jet to avoid the axis coordinate singularity of Grid 2 (Grid 3: \( 25 \times 385 \times 25 \)). The stationary cold-flow state is the same for all UQ sample simulations. Fig. 28 visualizes the turbulent boundary layer. Fig. 29 shows the corresponding hydrogen mass-fraction isosurface.

Dimension reduction is particularly important for so large of a simulation, and we only propagate the \( \ln A_3, b_3, \ln E_3 \) parameters determined important based on the sensitivity analysis of Section 9.3.4. In total, 48 runs are made. A typical ignition event is shown in Fig. 30. We see that the high temperature region, originating from the laser breakdown, develops into combustion and propagates as a flame toward the orifice. The QoI is the same as (50) used for the stand-burner simulations.

Based on axisymmetric model, we again set \( \nu = 3 \) in (12), and vary polynomial degree \( m \) in (8) to assess accuracy. The selection criterion is set to \( \theta = 10^{-3} \) in (18). The surrogate approximation error and representation size are shown in Fig. 31. Fig. 31a shows that the error \( 1 - R^2 \) (defined by (A.2)) consistently decreases with a higher degree \( m \). For \( m = 3 \) and 4, the leave-one-out cross validation based error \( 1 - Q^2 \) (defined by (A.7)) suggests poor model prediction performance. Starting from \( m = 5 \), the surrogate gives a better performance, with \( m = 8 \) providing the best accuracy with \( R^2 = 0.998957 \) and \( Q^2 = 0.984142 \). Fig. 31b illustrates the number of polynomial terms \( P_{\text{sparse}} \) retained in the surrogate. We note \( P_{\text{sparse}} \) has an upper bound of 34, which does not preclude acceptable numerical conditioning for \( Q = 48 \).

Taking \( m = 8 \), Fig. 32 shows the detailed adaptive procedure. The surrogate building iteration index varies from \( 2 + 16m = 26 \) to \( (1 + m)^{3} = 729 \), corresponding to the first bi-variate polynomial and last tri-variate polynomial, respectively (see (15)). Criterion (18) adaptively eliminates polynomial terms in each iteration, and then keeps the model sparse (less than or equal to 35), with sizes plotted in Fig. 32a. Fig. 32b shows the error \( 1 - R^2 \) before and after applying (18). This decreases from above 0.3 to near zero in the end. The application of Criterion (18) does not affect \( 1 - R^2 \) significantly. However, \( 1 - Q^2 \), shown in Fig. 32c, is visibly decreased by applying (18), which confirms that Criterion (18), which is based on variance contribution of polynomial terms, is capable of improving the model prediction performance. At the end of this adaptive procedure, \( 1 - Q^2 \) attains the value of 0.016, indicating a good fit of surrogate to the data.
Finally, we propagate $10^5$ samples through the surrogate. The mCov-SI can be easily computed using the MC approach described in Section 6.3. The results are presented in Fig. 33, showing first-, second-, and third-order indices. For each parameter, or group of parameters, three indices are presented: structural, correlative, and total indices (33). The second-order group [ln $E_3$, ln $A_3$] is the most significant, based on both the structural and total index values, while the first-order indices of ln $A_3$ are nearly zero. When first-order indices of $b_3$ and ln $E_3$ are non-zero, their second-order interaction is negligible, but the third-order term involving also ln $A_3$ is significant regarding all its three indices. Correlative effects are less significant than their structural counterparts in general, and mostly present in second- and third-order terms. Based on
this analysis, we conclude that all kinetic parameters are non-negligible, either independently or through interaction with others, motivating further effort on kinetics modeling.

11. Summary & conclusions

We have evaluated a new technique for managing uncertainty quantification for cases in which the simulation model is too expensive for extensive sampling, demonstrating it on a 29M mesh-point simulation of the ignition of hydrogen-air jet in cross-flow configuration by a laser-induced breakdown. We introduced a novel augmentation of ANOVA with PDD to facilitate this analysis. It now includes two levels of adaptation: truncation of ANOVA dimension (its maximum interaction order) and enrichment of the surrogate with the identified significant polynomials of higher-order ANOVA. Recursive LSR is used for this final task. The resulting surrogate is a sparse, which facilitates evaluation, but faithful representation of the full simulation model. Since the surrogate size is updated recursively, the number of the required sample runs is well-controlled, which is necessary for use for expensive computations, and its final value is significantly smaller than when employing Monte Carlo methods.

The particular combustion problem we consider has kinetic models that lead to input correlations, which introduce particular challenges for more standard methods. These correlations are included in ANOVA analysis by employing a covariance decomposition of the unconditional variance, which introduces three sensitivity indices to take account of input correlations. In order to avoid extremely large index values in case of high negative correlations between ANOVA components, we have
defined a simple mCov variant, which provides sensitivity indices (mCov-SI and mCov-TSI). Their properties are such that they provide useful sensitivity measures for dimension reduction. We use them for dimensionality reduction. In our case, we have found it is important to include third-order interaction where three kinetic parameters provide correlated samples.

The uncertainty propagation has been done in both axisymmetric and three-dimensional turbulent flow configurations, with the sensitivity metrics for the axisymmetric model used to both assess the effectiveness of the surrogate in a configuration for which more samples could be obtained and reduce the dimension of the uncertainty space. This then guided its application in the three-dimensional configurations, and similarly increased confidence in its effectiveness.

While the present sensitivity analysis is based on a covariance decomposition of the output variance, the moment-independent (density-based) methods first proposed by Borgonovo [70] consider the entire probability distribution of the model output without referring to any particular statistical moment. We refer to Borgonovo and Plischke [9] and Ghanem et al. [10] for a review of these methods. The definitions of this class of methods do not require input independence, and their computation can be realized conveniently via Monte Carlo sampling of our adaptive model representation. Among our future efforts is to compare mCov-SI and mCov-TSI with moment-independent measures using the present adaptive ANOVA surrogate.
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Appendix A. Estimator of accuracy $R^2$ & $Q^2$

Our use of the empirical error based accuracy estimator $R^2$ and leave-one-out cross validation based estimator $Q^2$ presented by Blatman and Sudret [14] are summarized here.

Let $f_X(x)$ denote the surrogate representation obtained by adaptive sparse polynomial dimensional decomposition (PDD) with coefficients determined by regression approach with the training points $X = (x_1, x_2, \cdots, x_Q)$ and corresponding data outputs $Y = (y_1, y_2, \cdots, y_Q)$.

The following empirical error is usually computed to estimate approximation error [14]:

$$I_X[f_X] = \frac{1}{Q} \sum_{i=1}^{Q} \left( y_i - f_X(x_i) \right)^2.$$  \hfill (A.1)

Of common use is the related accuracy estimator (i.e. determination coefficient [14]):

$$R^2[f_X] = 1 - \frac{I_X[f_X]}{\hat{\gamma}[Y]},$$  \hfill (A.2)

where

$$\hat{\gamma}[Y] = \frac{1}{Q-1} \sum_{i=1}^{Q} \left( y_i - \bar{y} \right)^2, \quad \text{with} \quad \bar{y} = \frac{1}{Q} \sum_{i=1}^{Q} y_i.$$  \hfill (A.3)

$R^2 = 1$ indicates a perfect fit, while $R^2 \approx 0$ or $R^2 < 0$ reflects a poor model accuracy. We also use $1 - R^2$ to represent the modeling error. It is known that $R^2$ systematically tends to unity as the size of surrogate model tends to the size $Q$ of the training set, and overfitting phenomenon occurs in such cases. Let us present an alternative estimator based on leave-one-out cross validation [14].

Let $f_{X \setminus i}(x)$ denote the sparse PDD representation constructed from the training set $X \setminus \{x_i\}$. We modify the empirical error (A.1) to be

$$I_X^*[f_X] = \frac{1}{Q} \sum_{i=1}^{Q} \left( y_i - f_{X \setminus i}(x_i) \right)^2.$$  \hfill (A.4)

In the case of linearly parameterized regression, we have the following result [14] useful for the computation of (A.4):

$$y_i - f_{X \setminus i}(x_i) = \frac{y_i - f_X(x_i)}{1 - h_i},$$  \hfill (A.5)

where $h_i$ is the $i$-th diagonal term of the projection matrix [14]. Hence, (A.4) can be expressed as

$$I_X^*[f_X] = \frac{1}{Q} \sum_{i=1}^{Q} \left( \frac{y_i - f_X(x_i)}{1 - h_i} \right)^2.$$  \hfill (A.6)

Note the use of (A.6) avoids any additional resolution of regression problems concerning $f_{X \setminus i}(x)$.

The determination coefficient based on leave-one-out cross validation is analogously:

$$Q^2[f_X] = 1 - \frac{I_X^*[f_X]}{\hat{\gamma}[Y]},$$  \hfill (A.7)

$Q^2 = 1$ indicates a perfect fit, while $Q^2 \lesssim 0$ indicates a poor accuracy. We also use $1 - Q^2$ to represent the approximation error.

References


