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Illustrating Important Effects of Second-Order Sensitivities on Response Uncertainties in Reactor Physics

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Abstract: This paper illustrates the relative importance of the largest first- and second-order sensitivities of the leakage response of an OECD/NEA reactor physics benchmark (a polyethylene-reflected plutonium sphere) to the benchmark's underlying total cross sections. It will be shown that numerous 2nd-order sensitivities of the leakage response with respect to the total cross sections are significantly larger than the largest corresponding 1st-order sensitivities. In particular, the contributions of the 2nd-order sensitivities cause the mean (expected) value of the response to differ appreciably from its computed value and also cause the response distribution to be skewed towards positive values relative to the mean. Neglecting these large 2nd-order sensitivities would cause very large non-conservative errors by under-reporting the response's variance and expected value. The results presented in this paper also underscore the need for obtaining reliable cross section covariance data, which are currently unavailable. Finally, comparing the CPU-times needed for computations, this paper demonstrates that the Second-Order Adjoint Sensitivity Analysis Methodology is the only practical method for computing 2nd-order sensitivities exactly, without introducing methodological errors, for large-scale systems characterized by many uncertain parameters.

Keywords: polyethylene-reflected plutonium sphere (PERP) benchmark; 2nd-order adjoint sensitivity analysis; 1st- and 2nd-order sensitivities; total cross sections; expected value; variance and skewness of PERP response

1. Introduction

The work reported in this work is based on the PHYSOR-2020 plenary invited conference paper entitled "On the importance of second-order response sensitivities to nuclear data in reactor physics uncertainty analysis." This paper was selected by the PHYSOR-2020 Technical Committee to be published in the *Special Issue* of the *Journal of Nuclear Engineering*. To avoid duplicate publication, the contents and the title of the conference paper have been re-written to make the present work suitable for publication in this journal's *Special Issue*.

The OECD Nuclear Energy Agency (OECD/NEA) International Criticality Safety Benchmark Evaluation Project (ICSBEP) Handbook [1] describes several fundamental subcritical reactor physics benchmarks that use a 4.5 kg alpha-phase plutonium sphere constructed at Los Alamos National Laboratory in 1980 [2]. This sphere was initially used as a neutron source for conducting subcritical experiments aimed at estimating the reactivity worth of beryllium reflectors (and was therefore colloquially called the "BeRP ball") but was subsequently also used for other subcritical experiments using tungsten, nickel and/or polyethylene reflectors. Miller et al. [3] evaluated computationally neutron multiplicity measurements for the "polyethylene-reflected BeRP ball", noticing significant disagreement between their computational results and the corresponding measurements.

In addition to the nuclear data investigated in [3], the "polyethylene-reflected plutonium sphere" benchmark contains other uncertain data, the most prominent being the total neutron cross sections. The 1st- and 2nd-order sensitivities of this benchmark's leakage response to the benchmark's cross sections have been computed by applying Cacuci's

2nd-ASAM [4]; the most significant results thus obtained are summarized in this work. Many of the 2nd-order sensitivities were found to be much larger than the corresponding 1st-order sensitivities, shifting the leakage response's mean value significantly away from its computed value and causing asymmetries in the response distribution. All in all, the effects on the benchmark's leakage response's expected value, variance, and skewness of the 2nd-order sensitivities with respect to the total cross sections are much larger than the corresponding effects stemming from the corresponding 1st-order sensitivities.

2. Largest 1st- and 2nd-Order Sensitivities of the Perp Benchmark's Leakage with Respect to Perp's Nuclear Data

The "polyethylene-reflected plutonium sphere benchmark" which will henceforth be called using the acronym "PERP" (benchmark) contains two materials, as follows: "Material 1" (core) contains four isotopes: ^{239}Pu, ^{240}Pu, ^{69}Ga, ^{71}Ga; "Material 2" (reflector) contains two isotopes: C and ^{1}H. The neutron flux distribution within the PERP benchmark was computed by using the PARTISN [5] multigroup discrete ordinates particle transport code, which solves the following multi-group approximation of the neutron transport equation with a spontaneous fission source:

$$B^g(\boldsymbol{\alpha})\varphi^g(r,\boldsymbol{\Omega}) = Q^g(r), \; g = 1,\ldots,G, \tag{1}$$

$$\varphi^g(r_d,\boldsymbol{\Omega}) = 0, r_d \in S_b, \boldsymbol{\Omega}\cdot\mathbf{n} < 0, \; g = 1,\ldots,G, \tag{2}$$

where r_d denotes the radius of the spherical benchmark, and

$$B^g(\boldsymbol{\alpha})\varphi^g(r,\boldsymbol{\Omega}) \triangleq \boldsymbol{\Omega}\cdot\nabla\varphi^g(r,\boldsymbol{\Omega}) + \Sigma_t^g(r)\,\varphi^g(r,\boldsymbol{\Omega})$$
$$- \sum_{g'=1}^{G}\int_{4\pi}\Sigma_s^{g'\to g}\left(r,\boldsymbol{\Omega}'\to\boldsymbol{\Omega}\right)\varphi^{g'}\left(r,\boldsymbol{\Omega}'\right)d\boldsymbol{\Omega}' - \chi^g(r)\sum_{g'=1}^{G}\int_{4\pi}(\nu\Sigma)_f^{g'}(r)\,\varphi^{g'}\left(r,\boldsymbol{\Omega}'\right)d\boldsymbol{\Omega}', \tag{3}$$

$$Q^g(r) \triangleq \sum_{k=1}^{N_f}\lambda_k N_{k,1}F_k^{SF}\nu_k^{SF}e^{-E^g/a_k}\sinh\sqrt{b_k E^g}, \; g = 1,\ldots,G. \tag{4}$$

The PARTISN [5] computations of the neutron flux used the MENDF71X [6] 618-group cross section data collapsed to $G = 30$ energy groups, as well as a P$_3$ Legendre expansion of the scattering cross section, an angular quadrature of S$_{256}$, and a fine-mesh spacing of 0.005 cm (comprising 759 meshes for the plutonium sphere of radius of 3.794 cm, and 762 meshes for the polyethylene shell of thickness of 3.81 cm). The quantities appearing in Equations (1)–(4) are defined as follows:

1. The quantity $\varphi^g(r,\boldsymbol{\Omega})$ is the customary "group-flux" for group g, and is the unknown state-function obtained by solving Equations (1) and (2), where r_d is the radius of the PERP sphere while the vector $\mathbf{n}$ denotes the outward unit normal vector at each point on the sphere's outer boundary, denoted as S_b.

2. The total cross section $\Sigma_t^g(r)$ for energy group g, $g = 1,\ldots,G$, and material m, is computed using the following expression:

$$\Sigma_t^g(r) = \sum_{m=1}^{M=2}\Sigma_{t,m}^g(r); \; \Sigma_{t,m}^g(r) = \sum_i N_{i,m}\sigma_{t,i}^g(r) = \sum_i N_{i,m}\left[\sigma_{f,i}^g(r) + \sigma_{c,i}^g(r) + \sum_{g'=1}^{G}\sigma_{s,l=0,i}^{g\to g'}(r)\right], \; m = 1,2, \tag{5}$$

where $\sigma_{f,i}^g(r)$ and $\sigma_{c,i}^g(r)$ denote, respectively, the tabulated group microscopic fission and neutron capture cross sections for group g, $g = 1,\ldots,G$. Other nuclear reactions are negligible in the PERP benchmark.

3. The quantity $N_{i,m}$ denotes the atom density of isotope i in material m; $i = 1,\ldots,I$, $m = 1,\ldots,M$, where I denotes the total number of isotopes, and M denotes the total number of materials. The computation of $N_{i,m}$ uses the well-known expression:

$$N_{i,m} \triangleq \frac{\rho_m w_{i,m} N_A}{A_i}, \tag{6}$$

where ρ_m denotes the mass density of material m, $m = 1, \ldots, M$; $w_{i,m}$ denotes the weight fraction of isotope i in material m; A_i denotes the atomic weight of isotope i, $i = 1, \ldots, I$; N_A denotes Avogadro's number. For the PERP benchmark, $I = 6$ and $M = 2$.

4. The quantity $\Sigma_s^{g' \to g}\left(r, \Omega' \to \Omega\right)$ represents the scattering transfer cross section from energy group g', $g' = 1, \ldots, G$ into energy group g, $g = 1, \ldots, G$, and is computed in terms of the l-th order Legendre coefficient $\sigma_{s,l,i}^{g' \to g}$ (of the Legendre-expanded microscopic scattering cross section from energy group g' into energy group g for isotope i), which are tabulated parameters, in the following finite-order expansion:

$$
\begin{aligned}
\Sigma_s^{g' \to g}\left(r, \Omega' \to \Omega\right) &= \sum_{m=1}^{M=2} \Sigma_{s,m}^{g' \to g}\left(r, \Omega' \to \Omega\right), \\
\Sigma_{s,m}^{g' \to g}\left(r, \Omega' \to \Omega\right) &\cong \sum_{i=1}^{I=6} N_{i,m} \sum_{l=0}^{ISCT=3} (2l+1)\, \sigma_{s,l,i}^{g' \to g}(r) P_l\left(\Omega' \cdot \Omega\right), m = 1,2,
\end{aligned}
\tag{7}
$$

where $ISCT = 3$ denotes the order of the respective finite expansion in Legendre polynomial. The expressions in Equations (5) and (7) indicate that the zeroth order (i.e., $l = 0$) scattering cross sections must be considered separately from the higher order (i.e., $l \geq 1$) scattering cross sections, since the former contribute to the total cross sections, while the latter do not.

5. The quantity N_f denotes the total number of spontaneous-fission isotopes. The spontaneous-fission isotopes in the PERP benchmark are "isotope 1" (^{239}Pu) and "isotope 2" (^{240}Pu), so $N_f = 2$, and the spontaneous fission neutron spectrum of ^{239}Pu and ^{240}Pu, respectively, is approximated by a Watts fission spectrum using the evaluated parameters a_k and b_k. The decay constant for actinide nuclide k is denoted as λ_k, and F_k^{SF} denotes the fraction of decays that are spontaneous fission (the "spontaneous-fission branching fraction").

6. PARTISN [5] computes the quantity $\left(\nu\Sigma_f\right)^g$ by directly using the quantities $(\nu\sigma)_{f,i}^g$, which are provided in nuclear data files for each isotope i, and energy group g, as follows

$$
\left(\nu\Sigma_f\right)^g = \sum_{m=1}^{M=2} \left(\nu\Sigma_f\right)_m^g; \quad \left(\nu\Sigma_f\right)_m^g = \sum_{i=1}^{I=6} N_{i,m} \left(\nu\sigma_f\right)_i^g, \quad m = 1,2.
\tag{8}
$$

7. The quantity $\chi^g(r)$ quantifies the fission spectrum in energy group g.

8. The numerical model of the PERP benchmark contains 7477 nonzero parameters which are subject to uncertainties, as follows: (i) 180 group-averaged microscopic total cross sections; (ii) 7101 group-averaged microscopic scattering cross sections; (iii) 60 group-averaged microscopic fission cross sections; (iv) 60 "averaged" number of neutron per fission; (v) 60 group-averaged fission spectrum constants; (vi) 10 external neutron source parameters; (vii) 6 isotopic number densities. The vector $\boldsymbol{\alpha}$, which appears in the expression of the Boltzmann-operator $B^g(\boldsymbol{\alpha})$, represents the "vector of uncertain model parameters".

The fundamental quantity underlying the neutron measurements is the total leakage of neutrons leaving the PERP sphere, represented mathematically as follows:

$$
L(\boldsymbol{\alpha}) \triangleq \int_{S_b} dS \sum_{g=1}^{G} \int_{\Omega \cdot \mathbf{n} > 0} d\Omega\, \Omega \cdot \mathbf{n}\, \varphi^g(r, \Omega).
\tag{9}
$$

The expressions of the 1st-order sensitivities of the leakage response to the model parameters underlying the total cross section are as follows:

$$\frac{\partial L(\boldsymbol{\alpha})}{\partial \sigma_{t,i}^g} = -\sum_{g=1}^{G} \int dV \int_{4\pi} d\boldsymbol{\Omega}\, \psi^{(1),g}(\mathbf{r},\boldsymbol{\Omega})\varphi^g(\mathbf{r},\boldsymbol{\Omega})\frac{\partial \Sigma_t^g(\mathbf{t};\mathbf{r})}{\partial \sigma_{t,i}^g}\,, \; i = 1,\ldots,I;\; g = 1,\ldots,G, \quad (10)$$

where the multigroup adjoint fluxes $\psi^{(1),g}(r,\boldsymbol{\Omega})$ are the solutions of the following 1st-Level Adjoint Sensitivity System (1st-LASS):

$$A^{(1),g}(\boldsymbol{\alpha})\psi^{(1),g}(r,\boldsymbol{\Omega}) = \boldsymbol{\Omega}\cdot\mathbf{n}\delta(r-r_d),\; g = 1,\ldots,G, \quad (11)$$

$$\psi^{(1),g}(r_d,\boldsymbol{\Omega}) = 0,\, \boldsymbol{\Omega}\cdot\mathbf{n} > 0,\; g = 1,\ldots,G, \quad (12)$$

where
$$A^{(1),g}(\boldsymbol{\alpha})\psi^{(1),g}(r,\boldsymbol{\Omega})$$
$$\triangleq -\boldsymbol{\Omega}\cdot\nabla\psi^{(1),g}(r,\boldsymbol{\Omega}) + \Sigma_t^g(\mathbf{t};r)\,\psi^{(1),g}(r,\boldsymbol{\Omega}) - \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\Sigma_s^{g\to g'}\left(\mathbf{s};r,\boldsymbol{\Omega}\to\boldsymbol{\Omega}'\right)\psi^{(1),g'}\left(r,\boldsymbol{\Omega}'\right)$$
$$-\nu\Sigma_f^g(\mathbf{f};r)\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\chi^{g'}(\mathbf{p};r)\,\psi^{(1),g'}\left(r,\boldsymbol{\Omega}'\right),\; g = 1,\ldots,G. \quad (13)$$

The 2nd-order sensitivities of the leakage response to the parameters involved in the definitions of the total cross sections are given by the following expression:

$$\frac{\partial^2 L}{\partial t_j\partial t_{m_2}} = -\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\left[\psi_{1,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega})\psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega}) + \psi_{2,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega})\varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\right]N_{i_{m_2},m_{m_2}},\quad (14)$$
$$for\; j = 1,\ldots,J_{\sigma t};\; m_2 = 1,\ldots,J_{\sigma t}\,,$$

where the 2nd-level adjoint functions $\psi_{1,j}^{(2),g}$, $j = 1,\ldots,J_{\sigma t};\; g = 1,\ldots,G$, and $\psi_{2,j}^{(2),g}$, $j = 1,\ldots,J_{\sigma t};\; g = 1,\ldots,G$, are the solutions of the following Second-Level adjoint Sensitivity system (2nd-LASS):

$$B^g\left(\boldsymbol{\alpha}^0\right)\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = -\delta_{g_j g}N_{i_j,m_j}\varphi^g(r,\boldsymbol{\Omega}),\; j = 1,\ldots,J_{\sigma t};\; g = 1,\ldots,G, \quad (15)$$

$$\psi_{1,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0,\; \boldsymbol{\Omega}\cdot\mathbf{n} < 0;\; j = 1,\ldots,J_{\sigma t};\; g = 1,\ldots,G, \quad (16)$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) = -\delta_{g_j g}N_{i_j,m_j}\psi^{(1),g}(r,\boldsymbol{\Omega}),\; j = 1,\ldots,J_{\sigma t};\; g = 1,\ldots,G, \quad (17)$$

$$\psi_{2,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0,\; \boldsymbol{\Omega}\cdot\mathbf{n} > 0;\; j = 1,\ldots,J_{\sigma t};\; g = 1,\ldots,G. \quad (18)$$

All of the quantities appearing in Equations (14)–(17) are evaluated at the nominal values of all model parameters. The details of these computations are presented in [7]. All of the computations were performed using a DELL computer AMD FX-8350 having an 8-core processor.

The leakage response displayed the largest 1st- and 2nd-order sensitivities with respect to the (group-averaged) *total* microscopic cross sections for ^{1}H; these sensitivities are presented in Tables 1 and 2; complete results are presented in [7–12].

Table 1. First-order relative sensitivities, $\left(\partial L/\partial \sigma_{t,i=6}^g\right)\left(\sigma_{t,i=6}^g/L\right)$, $g = 1,\ldots,30$ of the leakage to the total cross sections of ^{1}H (isotope i = 6).

g	1st-Order	G	1st-Order	g	1st-Order	g	1st-Order	G	1st-Order
1–6	<−0.01	11	−0.19	16	−1.16	21	−9.69	26	−0.65
7	−0.07	12	−0.44	17	−1.17	22	−0.92	27	−0.58
8	−0.09	13	−0.52	18	−1.14	23	−0.89	28	−0.55
9	−0.14	14	−0.57	19	−1.09	24	−0.75	29	−0.55
10	−0.17	15	−0.58	20	−1.03	25	−0.71	30	**−9.366**

Table 2. Selected 2nd-order relative sensitivities $\left(\partial^2 L / \partial \sigma^g_{t,i=6} \partial \sigma^g_{t,k=6} \right) \left(\sigma^g_{t,i=6} \sigma^g_{t,k=6} / L \right)$, $g = 1, \ldots, 30$, having values greater than 1.0, for ^{1}H.

	g' = 12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30
g = 12	0.653	0.315	0.340	0.356	0.740	0.763	0.751	0.725	0.688	0.648	0.597	0.554	0.502	0.477	0.440	0.393	0.369	0.372	**6.432**
13	0.315	0.974	0.471	0.471	0.976	**1.005**	0.988	0.953	0.904	0.851	0.784	0.728	0.659	0.627	0.577	0.516	0.484	0.487	**8.424**
14	0.340	0.471	**1.261**	0.579	**1.158**	**1.192**	**1.172**	**1.130**	**1.072**	**1.009**	0.930	0.863	0.782	0.742	0.684	0.611	0.574	0.576	**9.968**
15	0.356	0.471	0.579	**1.391**	**1.255**	**1.277**	**1.255**	**1.210**	**1.148**	**1.081**	0.996	0.924	0.837	0.795	0.733	0.655	0.615	0.617	**10.67**
16	0.740	0.976	**1.158**	**1.255**	**4.461**	**2.700**	**2.647**	**2.553**	**2.421**	**2.280**	**2.100**	**1.949**	**1.767**	**1.677**	**1.546**	**1.381**	**1.296**	**1.300**	**22.48**
17	0.763	**1.005**	**1.192**	**1.277**	**2.700**	**4.853**	**2.789**	**2.684**	**2.546**	**2.398**	**2.209**	**2.050**	**1.858**	**1.764**	**1.625**	**1.452**	**1.363**	**1.367**	**23.62**
18	0.751	0.988	**1.172**	**1.255**	**2.647**	**2.789**	**4.828**	**2.689**	**2.546**	**2.399**	**2.210**	**2.051**	**1.859**	**1.764**	**1.626**	**1.453**	**1.363**	**1.367**	**23.62**
19	0.725	0.953	**1.130**	**1.210**	**2.553**	**2.684**	**2.689**	**4.619**	**2.498**	**2.349**	**2.165**	**2.010**	**1.822**	**1.729**	**1.594**	**1.424**	**1.336**	**1.340**	**23.15**
20	0.688	0.904	**1.072**	**1.148**	**2.421**	**2.546**	**2.546**	**2.498**	**4.284**	**2.266**	**2.085**	**1.936**	**1.755**	**1.666**	**1.535**	**1.372**	**1.287**	**1.290**	**22.29**
21	0.648	0.851	**1.009**	**1.081**	**2.280**	**2.398**	**2.399**	**2.349**	**2.266**	**3.937**	**2.004**	**1.857**	**1.684**	**1.599**	**1.474**	**1.317**	**1.236**	**1.238**	**21.40**
22	0.597	0.784	0.930	0.996	**2.100**	**2.209**	**2.210**	**2.165**	**2.085**	**2.004**	**3.515**	**1.760**	**1.593**	**1.512**	**1.394**	**1.246**	**1.169**	**1.171**	**20.24**
23	0.554	0.728	0.863	0.924	**1.949**	**2.050**	**2.051**	**2.010**	**1.936**	**1.857**	**1.760**	**3.177**	**1.521**	**1.440**	**1.328**	**1.187**	**1.114**	**1.116**	**19.28**
24	0.502	0.659	0.782	0.837	**1.767**	**1.858**	**1.859**	**1.822**	**1.755**	**1.684**	**1.593**	**1.521**	**2.792**	**1.358**	**1.249**	**1.117**	**1.048**	**1.049**	**18.13**
25	0.477	0.627	0.742	0.795	**1.677**	**1.764**	**1.764**	**1.729**	**1.666**	**1.599**	**1.512**	**1.440**	**1.358**	**2.604**	**1.214**	**1.082**	**1.016**	**1.017**	**17.58**
26	0.440	0.577	0.684	0.733	**1.546**	**1.625**	**1.626**	**1.594**	**1.535**	**1.474**	**1.394**	**1.328**	**1.249**	**1.214**	**2.349**	**1.037**	0.971	0.972	**16.79**
27	0.393	0.516	0.611	0.655	**1.381**	**1.452**	**1.453**	**1.424**	**1.372**	**1.317**	**1.246**	**1.187**	**1.117**	**1.082**	**1.037**	**2.039**	0.913	0.912	**15.76**
28	0.369	0.484	0.574	0.615	**1.296**	**1.363**	**1.363**	**1.336**	**1.287**	**1.236**	**1.169**	**1.114**	**1.048**	**1.016**	0.971	0.913	**1.885**	0.888	**15.30**
29	0.372	0.487	0.576	0.617	**1.300**	**1.367**	**1.367**	**1.340**	**1.290**	**1.238**	**1.171**	**1.116**	**1.049**	**1.017**	0.972	0.912	0.888	**1.891**	**15.39**
30	**6.432**	**8.424**	**9.97**	**10.67**	**22.48**	**23.62**	**23.62**	**23.15**	**22.29**	**21.40**	**20.24**	**19.28**	**18.13**	**17.58**	**16.79**	**15.76**	**15.30**	**15.39**	**429.6**

For the *first-order sensitivities, the 2nd-ASAM required* ca. *5 min CPU time* was needed for computing: (a) the forward group-fluxes $\varphi^g(r, \Omega)$ by solving Equations (1) and (2); (b) the adjoint group-fluxes $\psi^{(1),g}(r, \Omega)$ by solving Equations (11) and (12); and (c) the 7477 integrals expressed by Equation (10) to obtain all of the first-order sensitivities.

By comparison, it would require 694 h-CPU to compute approximately the first-order responses sensitivities using the two-point finite difference scheme shown below:

$$\frac{\partial R}{\partial \alpha_i} \simeq \frac{R_{i+1} - R_{i-1}}{2(\delta \alpha_i)} = O(\delta \alpha_i)^2, \, i = 1, \ldots, N_\alpha, \tag{19}$$

where $R_{i+1} \triangleq R(\alpha_i^0 + \delta \alpha_i)$ and $R_{i-1} \triangleq R(\alpha_i^0 - \delta \alpha_i)$. Note that Equation (19) *introduces its own intrinsic methodological error when approximating the respective derivative*; this error would be present even if the numerical computation of the response $R(\alpha_i)$ at the respective "sampling point" α_i were perfect. Additionally, one needs to "play around" to find the "correct" value to use for $\delta \alpha_i$, because if $\delta \alpha_i$ is either too large or too small, Equation (19) would produce erroneous results. *The 694 h-CPU required to compute approximately the first-order responses sensitivities using Equation* (19) *does not include the time needed to find the "appropriate" $\delta \alpha_i$ which gives a "satisfactory" approximation to the respective derivative.*

As indicated by Equation (14), the computation of each of the *27,956,503 distinct non-zero* second-order sensitivities of the response (to two non-zero parameters) requires one forward PARTISN computation to obtain the function $\psi_{1,j}^{(2),g}$ by solving Equations (15) and (16), as well as one adjoint PARTISN computation to obtain the function $\psi_{2,j}^{(2),g}$ by solving Equations (17) and (18). The total CPU time needed to compute the 27,956,503 distinct non-zero second-order sensitivities is ca. 929 h-CPU, as follows (i) 735 h-CPU needed to perform the 7427 forward PARTISN runs + 7415 PARTISN adjoint runs to compute the 2nd-level adjoint functions $\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}$; (ii) 194 CPU-hours needed to compute the integrals in Equation (14) to obtain the unmixed and mixed second-order sensitivities.

Consider, for comparison, the simplest finite-difference scheme for computing the second-order responses sensitivities, namely:

$$\frac{\partial^2 R}{\partial \alpha_i \partial \alpha_i} \simeq \frac{R_{i+1} - 2R_i + R_{i-1}}{(\delta \alpha_i)^2} = O(\delta \alpha_i)^2, \, i = 1, \ldots, N_\alpha, \tag{20}$$

$$\frac{\partial^2 R}{\partial \alpha_i \partial \alpha_j} \simeq \frac{R_{i+1,j+1} - R_{i-1,j+1} - R_{i+1,j-1} + R_{i-1,j-1}}{4 \delta \alpha_i \delta \alpha_j} = O\left[(\delta \alpha_i)^2, (\delta \alpha_j)^2\right], \, i = 1, \ldots, N_\alpha, \, j = 1, \ldots, i, \tag{21}$$

where $R_{i,j} \triangleq R\left(\alpha_i^0 + \delta \alpha_i, \alpha_j^0 + \delta \alpha_j\right)$, etc., are computed response values at the indicated "sampling points." Note again that the *finite-difference schemes introduce their own intrinsic methodological error when approximating the respective derivative*; this error would be present even if the numerical computation of the response $R(\alpha_i)$ at the respective "sampling point" α_i were perfect. Furthermore, one needs to find by "trial and error" the "correct" value to use for $\delta \alpha_i$, because if $\delta \alpha_i$ is either too large or too small, the *finite-difference schemes* would produce wrong numbers. *Not counting the "trial and error" the "correct" value to use for $\delta \alpha_i$, the CPU time (using a DELL computer with an 8-core processor, AMD FX-8350) needed to compute the 27,956,503 distinct non-zero 2nd-order sensitivities by using Equations (20) and (21) at the respective 111,811,058 "sampling points" would be 592 YEARS-CPU time! Furthermore, these sensitivities would not be exact (as produced by the 2nd-ASAM), but would contain second-order errors. Evidently, it is not feasible to compute the 2nd-order sensitivities using "sampling approaches".*

3. Uncertainty Quantification

The numerical results reported in this paper comprise just the effects of the group-averaged total microscopic cross sections, which will be indicated by using the subscript "*t*". Since correlations among the group total cross sections are not available for the

PERP benchmark, two extreme situations can be considered: (i) all cross sections are uncorrelated, which will be considered in Section 3.1 below; (ii) all cross sections are fully correlated, which will be considered in Section 3.2 below. The formulas used in this section are as originally used in [12]; for convenient referencing, they are reproduced in Sections 3.1 and 3.2.

3.1. Uncorrelated Total Microscopic Cross Sections

The expected value of the leakage response has the expression $[E(L)]_t^{(U)} = L(\boldsymbol{\alpha}^0) + [E(L)]_t^{(2,U)}$, where the superscript "(2,U)" indicates "2nd-order uncorrelated" cross sections and where: (i) $L(\boldsymbol{\alpha}^0)$ denotes the nominal value of the computed leakage response; and (ii) $[E(L)]_t^{(2,U)} = \frac{1}{2} \sum_{g=1}^{30} \sum_{i=1}^{6} \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \sigma_{t,i}^g \partial \sigma_{t,i}^g} \left(s_{t,i}^g \right)^2$, where $s_{t,i}^g$ denotes the standard deviation associated with the group-averaged total microscopic cross sections $\sigma_{t,i}^g$, for each isotope i. If the total microscopic cross sections are both *uncorrelated and normally distributed*, which will be indicated using the superscript "(U,N)", then the following expressions hold: (i) the variance, $[\text{var}\,(L)]_t^{(U,N)}$, of the leakage response of the PERP benchmark becomes $[\text{var}\,(L)]_t^{(U,N)} = [\text{var}\,(L)]_t^{(1,U,N)} + [\text{var}\,(L)]_t^{(2,U,N)}$, where the 1st-order term is defined as $[\text{var}\,(L)]_t^{(1,U,N)} \triangleq \sum_{g=1}^{30} \sum_{i=1}^{6} \left[\frac{\partial L(\boldsymbol{\alpha})}{\partial \sigma_{t,i}^g} \right]^2 \left(s_{t,i}^g \right)^2$ and the 2nd-order term is defined as: $[\text{var}\,(L)]_t^{(2,U,N)} \triangleq \frac{1}{2} \sum_{g=1}^{30} \sum_{i=1}^{6} \left[\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \sigma_{t,i}^g \partial \sigma_{t,i}^g} \left(s_{t,i}^g \right)^2 \right]^2$. Furthermore, the 3rd-order moment, $[\mu_3(L)]_t^{(U,N)}$, of the leakage response has the form $[\mu_3(L)]_t^{(U,N)} = 3 \sum_{g=1}^{30} \sum_{i=1}^{6} \left[\frac{\partial L(\boldsymbol{\alpha})}{\partial \sigma_{t,i}^g} \right]^2 \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \sigma_{t,i}^g \partial \sigma_{t,i}^g} \left(s_{t,i}^g \right)^4$.

The skewness, $[\gamma_1(L)]_t^{(U,N)}$, of the leakage response, L, due to the variances of uncorrelated and normally distributed microscopic total cross sections is defined as $[\gamma_1(L)]_t^{(U,N)} = [\mu_3(L)]_t^{(U,N)} / \left\{ [\text{var}(L)]_t^{(U,N)} \right\}^{3/2}$. If the 2nd-order sensitivities were unavailable, the third moment $[\mu_3(L)]_t^{(U,N)}$ and hence the skewness γ_1 of the leakage response would vanish and the response distribution would by default be assumed to be Gaussian.

3.2. Fully Correlated Total Microscopic Cross Sections

The effects of *correlations* among the group-averaged microscopic total cross sections are transmitted to the response moments (mean value, variance, skewness) through the 2nd-order *mixed* sensitivities of the leakage response to these cross sections. The exact effect of such correlations cannot be assessed exactly since they are unavailable. When the cross sections are *fully correlated* (an extreme case denoted by using the superscript "*FC*"), the effects of the 2nd-order sensitivities can be quantified as follows:

(i)   The mean value of the leakage response becomes $[E(L)]_t^{(FC)} = L(\boldsymbol{\alpha}^0) + [E(L)]_t^{(2,FC)}$, where $[E(L)]_t^{(2,FC)} \triangleq \frac{1}{2} \sum_{g=1}^{30} \sum_{g'=1}^{30} \sum_{i=1}^{6} \sum_{k=1}^{6} \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \sigma_{t,i}^g \partial \sigma_{t,k}^{g'}} \left(s_{t,i}^g s_{t,k}^{g'} \right)$. The quantity $[E(L)]_t^{(2,MSC)} = [E(L)]_t^{(2,FC)} - [E(L)]_t^{(2,U)}$, where the superscript "MSC" stands for "mixed second-order correlated," quantifies the 2nd-order contributions to the mean value of the stemming from the mixed 2nd-order sensitivities when the total cross sections are fully correlated.

(ii) When the cross sections are *normally distributed and fully correlated*, the variance of the response is $[\text{var}\,(L)]_t^{(FC,N)} = [\text{var}\,(L)]_t^{(1,FC,N)} + [\text{var}\,(L)]_t^{(2,FC,N)}$, where the contributions from the 1st-order sensitivities are contained in the term $[\text{var}\,(L)]_t^{(1,FC,N)} \triangleq \sum_{g=1}^{30} \sum_{g'=1}^{30} \sum_{i=1}^{6} \sum_{k=1}^{6} \left[\frac{\partial L(\boldsymbol{\alpha})}{\partial \sigma_{t,i}^g} \frac{\partial L(\boldsymbol{\alpha})}{\partial \sigma_{t,k}^{g'}} \right] \left(s_{t,i}^g s_{t,k}^{g'} \right)$ while the contributions from the 2nd-order sen-

sitivities are contained in the term $[\text{var}\,(L)]_t^{(2,FC,N)} \triangleq \dfrac{1}{2}\left[\displaystyle\sum_{g=1}^{30}\sum_{g'=1}^{30}\sum_{i=1}^{6}\sum_{k=1}^{6}\dfrac{\partial^2 L(\boldsymbol{\alpha})}{\partial\sigma_{t,i}^g\partial\sigma_{t,k}^{g'}}s_{t,i}^g s_{t,k}^{g'}\right]$

$\left[\displaystyle\sum_{g=1}^{G}\sum_{g'=1}^{G}\sum_{\mu=1}^{I}\sum_{\nu=1}^{I}\dfrac{\partial^2 L(\boldsymbol{\alpha})}{\partial\sigma_{t,\mu}^g\partial\sigma_{t,\nu}^{g'}}s_{t,\mu}^g s_{t,\nu}^{g'}\right]$. The superscript "*(FC,N)*" indicates "fully correlated, normally distributed." The contributions to $[\text{var}\,(L)]_t^{(FC,N)}$ involving the first-order sensitivities will be denoted as $[\text{var}\,(L)]_t^{(1,MSC)}$ and are obtained by subtracting the uncorrelated terms from the fully correlated ones, i.e., $[\text{var}\,(L)]_t^{(1,MSC)} = [\text{var}\,(L)]_t^{(1,FC,N)} - [\text{var}\,(L)]_t^{(1,U,N)}$. The superscript "(1, MSC)" denotes "<u>f</u>irst-order, <u>m</u>ixed <u>s</u>ensitivities, <u>c</u>orrelated." Similarly, the quantity $[\text{var}\,(L)]_t^{(2,MSC)} = [\text{var}\,(L)]_t^{(2,FC)} - [\text{var}\,(L)]_t^{(2,U,N)}$ represents the contributions to $[\text{var}\,(L)]_t^{(FC,N)}$ involving the 2nd-order mixed and correlated sensitivities.

When the total cross sections are fully correlated, the skewness, $[\gamma_1(L)]_t^{(FC,N)} = [\mu_3(L)]_t^{(FC,N)}/\left\{[\text{var}(L)]_t^{(FC,N)}\right\}^{3/2}$ of the response distribution response involve a third order cumulant that has the following expression:

$$[\mu_3(L)]_t^{(FC,N)} = \left[\sum_{g=1}^{30}\sum_{g'=1}^{30}\sum_{i=1}^{6}\sum_{k=1}^{6}\frac{\partial L(\boldsymbol{\alpha})}{\partial\sigma_{t,i}^g}s_{t,i}^g\frac{\partial L(\boldsymbol{\alpha})}{\partial\sigma_{t,k}^{g'}}s_{t,k}^{g'}\right]\left[\sum_{g=1}^{30}\sum_{g'=1}^{30}\sum_{\mu=1}^{6}\sum_{\nu=1}^{6}\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial\sigma_{t,\mu}^g\partial\sigma_{t,\nu}^{g'}}s_{t,\mu}^g s_{t,\nu}^{g'}\right]$$

$$+\left[\sum_{g=1}^{30}\sum_{g'=1}^{30}\sum_{i=1}^{6}\sum_{\nu=1}^{6}\frac{\partial L(\boldsymbol{\alpha})}{\partial\sigma_{t,i}^g}s_{t,i}^g\frac{\partial L(\boldsymbol{\alpha})}{\partial\sigma_{t,\nu}^{g'}}s_{t,\nu}^{g'}\right]\left[\sum_{g=1}^{30}\sum_{g'=1}^{30}\sum_{k=1}^{6}\sum_{\mu=1}^{6}\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial\sigma_{t,k}^g\partial\sigma_{t,\mu}^{g'}}s_{t,k}^{g'}s_{t,\mu}^g\right]$$

$$+\left[\sum_{g=1}^{30}\sum_{g'=1}^{30}\sum_{i=1}^{6}\sum_{k=1}^{6}\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial\sigma_{t,i}^g\partial\sigma_{t,k}^{g'}}s_{t,i}^g s_{t,k}^{g'}\right]\left[\sum_{g=1}^{30}\sum_{g'=1}^{30}\sum_{\mu=1}^{6}\sum_{\nu=1}^{6}\frac{\partial L(\boldsymbol{\alpha})}{\partial\sigma_{t,\mu}^g}s_{t,\mu}^g\frac{\partial L(\boldsymbol{\alpha})}{\partial\sigma_{t,\nu}^{g'}}s_{t,\nu}^{g'}\right].$$

When the total cross sections are fully correlated and normally distributed, the quantity $[\mu_3(L)]_t^{(MSC,N)} = [\mu_3(L)]_t^{(FC,N)} - [\mu_3(L)]_t^{(U,N)}$ provides the contributions from the "<u>m</u>ixed <u>s</u>econd-order sensitivities and fully <u>c</u>orrelated <u>n</u>ormally distributed parameters," which is indicated using the superscript "*(MSC,N)*".

Table 3 presents illustrative results when considering uniform relative standard deviations $s_{t,i}^g$ of 5% and 10%, respectively. These numerical results highlight the importance of both the unmixed and mixed 2nd-order sensitivities for: (a) causing the mean value of the leakage response to differ from its computed value; (b) contributing significantly to increase the total response variance $[\text{var}\,(L)]_t^{(U)}$; (c) causing the response distribution to become non-Gaussian.

Table 3. Response moments for various cases.

Fully Correlated Cross Sections			Uncorrelated Cross Sections		
Rel. St. Dev.	**10%**	**5%**	**Rel. St. Dev.**	**10%**	**5%**
$L(\alpha^0)$	1.7648×10^6	1.7648×10^6	$L(\alpha^0)$	1.7648×10^6	1.7648×10^6
$[E(L)]_t^{(2,FC)}$	2.9451×10^7	7.3627×10^6	$[E(L)]_t^{(2,U)}$	4.5980×10^6	1.1495×10^6
$[E(L)]_t^{(2,MSC)}$	2.4853×10^7	6.2132×10^6	$[E(L)]_t^{(U)}$	6.3628×10^6	2.9143×10^6
$[E(L)]_t^{(FC)}$	3.1216×10^7	9.1275×10^6	$[var\,(L)]_t^{(1,U,N)}$	3.4196×10^{12}	8.5490×10^{11}
$[var\,(L)]_t^{(1,FC,N)}$	4.7601×10^{13}	1.1900×10^{13}	$[var\,(L)]_t^{(2,U,N)}$	2.8789×10^{13}	1.7993×10^{12}
$[var\,(L)]_t^{(1,MSC)}$	4.4181×10^{13}	1.1045×10^{13}	$[var\,(L)]_t^{(U,N)}$	3.2208×10^{13}	2.6542×10^{12}
$[var\,(L)]_t^{(2,FC,N)}$	1.7347×10^{15}	1.0842×10^{14}	$[\mu_3(L)]_t^{(U,N)}$	6.2267×10^{19}	3.8917×10^{18}
$[var\,(L)]_t^{(2,MSC)}$	1.7059×10^{15}	1.0662×10^{14}	$[\gamma_1(L)]_t^{(U,N)}$	0.3407	0.8999
$[var\,(L)]_t^{(FC,N)}$	1.7823×10^{15}	1.2302×10^{14}			
$[\mu_3(L)]_t^{(FC,N)}$	8.4113×10^{21}	5.2571×10^{20}			
$[\mu_3(L)]_t^{(MSC,N)}$	8.3490×10^{21}	5.2181×10^{20}			
$[\gamma_1(L)]_t^{(FC,N)}$	0.1118	0.3983			

4. Concluding Remarks

The following conclusions can be drawn based on the results presented in Table 3:

1. The 2nd-order sensitivities become more important than the 1st-order sensitivities when the parameters' standard deviations and correlation become larger.
2. The 2nd-order sensitivities cause the mean value of the leakage response to differ from its computed value.
3. The 2nd-order sensitivities cause the leakage distribution in parameter space to be skewed towards positive values relative to the response's mean value.
4. In reality, the total cross sections are partially correlated, but these correlations are currently unavailable. The results (presented in Table 3) involving the mixed 2nd-order sensitivities highlight the need for future experimental research for quantifying these currently unavailable correlations.
5. It has also been shown that the Second Order Adjoint Sensitivity Analysis Methodology (2nd-ASAM) conceived by Cacuci [13–15] is the only practical method for computing the second-order sensitivities exactly (i.e., without introducing methodological errors) for realistic and practical models of large-scale systems, which invariably involve many uncertain parameters. In particular, it has been shown that the simplest "2-point sampling method" requires for computing the 1st-order sensitivities (while introducing second-order errors!) almost as much time as the 2nd-ASAM requires for computing (without methodological errors!) all of the 2nd-order sensitivities. Additionally, it has also been shown that even the simplest "sampling approach" requires unthinkably large amount of computational time, while still producing approximate results, subject to "sampling approach errors".

The complete 2nd-order sensitivity analysis of the PERP benchmark can be found in References [7–12]. The complete neutron transport computational model of this benchmark comprises 21,886 imprecisely known nuclear data parameters, which give rise to 21,886 first-order and 478,996,996 second-order sensitivities. The analysis presented in [7–12] indicates that many of the 2nd-order sensitivities of the leakage response to the nuclear data underlying this benchmark are more important than the 1st-order ones. While the effects of 2nd-order response sensitivities may be less markedly important for other reactor physics systems than they are for the leakage response of the PERP benchmark, the results presented in this work and in [7–12] clearly underscore the need for computing

them, rather than considering them a priori to be negligible, as has been the practice thus far in the published literature.

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