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Comprehensive Second-Order Adjoint Sensitivity Analysis Methodology (2nd-ASAM) Applied to a Subcritical Experimental Reactor Physics Benchmark: V. Computation of Mixed 2nd-Order Sensitivities Involving Isotopic Number Densities

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Abstract: This work applies the Second-Order Adjoint Sensitivity Analysis Methodology (2nd-ASAM) to compute the mixed 2nd-order sensitivities of a polyethylene-reflected plutonium (PERP) benchmark's leakage response with respect to the benchmark's imprecisely known isotopic number densities and the other benchmark imprecisely known parameters, including: (i) the 6×180 mixed 2nd-order sensitivities involving the total microscopic cross sections; (ii) the $6 \times 21{,}600$ mixed 2nd-order sensitivities involving the scattering microscopic cross sections; (iii) the 6×60 mixed 2nd-order sensitivities involving the fission microscopic cross sections; and (iv) the 6×60 mixed 2nd-order sensitivities involving the average number of neutrons produced per fission. It is shown that many of these mixed 2nd-order sensitivities involving the isotopic number densities have very large values. Most of the large sensitivities involve the isotopic number density of ^{239}Pu, and the microscopic total, scattering or fission cross sections for the 12th or 30th energy groups of ^{239}Pu or ^{1}H, respectively. The 2nd-order mixed sensitivity of the PERP leakage response with respect to the isotopic number density of ^{239}Pu and the microscopic total cross section for the 30th energy group of ^{1}H is the largest of the above mentioned sensitivities, attaining the value −94.91.

Keywords: polyethylene-reflected plutonium sphere; 1st- and 2nd-order sensitivities; isotopic number density; fission spectrum; expected value; variance and skewness of leakage response

1. Introduction

In Parts I–IV [1–4], which are precursors of this work, the Second-Order Adjoint Sensitivity Analysis Methodology (2nd-ASAM) conceived by Cacuci [5–7] has been successfully applied to the subcritical polyethylene-reflected plutonium (acronym: PERP) metal fundamental physics benchmark [8], to compute the exact values of the 1st-order and 2nd-order sensitivities of the PERP's benchmark leakage response with respect to the 180 group-averaged total microscopic cross sections [1], 21,600 group-averaged scattering microscopic cross sections [2], 120 fission process parameters [3], and 10 source parameters [4]. This work presents the results obtained for the mixed 2nd-order sensitivities of the PERP benchmark's leakage response with respect to the benchmark's six isotopic number densities. Table 1 summarizes the dimensions and material composition of the PERP benchmark; additional details are presented in Part I [1]. As shown in Table 1, the six isotopic number densities correspond to each of the isotopes contained in the PERP benchmark, respectively. The isotopic number density is one of important parameters that contribute to the accuracy of the neutron transport calculation, as it appears in the definitions/constructions of the total, scattering and fission macroscopic cross sections, as well as the source term of the neutron transport equation.

Table 1. Dimensions and material composition of the PERP benchmark.

Materials	Isotopes	Weight Fraction	Density (g/cm^3)	Zones
Material 1 (plutonium metal)	Isotope 1 (^{239}Pu) Isotope 2 (^{240}Pu) Isotope 3 (^{69}Ga) Isotope 4 (^{71}Ga)	9.3804×10^{-1} 5.9411×10^{-2} 1.5152×10^{-3} 1.0346×10^{-3}	19.6	Material 1 is assigned to zone 1, which has a radius of 3.794 cm.
Material 2 (polyethylene)	Isotope 5 (C) Isotope 6 (^{1}H)	8.5630×10^{-1} 1.4370×10^{-1}	0.95	Material 2 is assigned to zone 2, which has an inner radius of 3.794 cm and an outer radius of 7.604 cm.

The mixed 2nd-order sensitivities of the leakage response with respect to the isotopic number densities are computed by specializing the general expressions derived by Cacuci [5–7] to the PERP benchmark. This work is organized as follows: Section 2 presents the numerical results for the 6×180 mixed 2nd-order sensitivities with respect to the isotopic number densities and total microscopic cross sections. Section 3 summarizes the numerical results for the 6×21600 matrix of mixed 2nd-order sensitivities to the isotopic number densities and scattering microscopic cross sections. Section 4 presents the numerical results for the 6×60 mixed 2nd-order sensitivities to the isotopic number densities and fission microscopic cross sections. Section 5 reports the numerical results for the 6×60 mixed 2nd-order sensitivities to the isotopic number densities and the average number of neutrons per fission. The conclusions drawn from the numerical results presented in this work are summarized in Section 6.

2. Mixed Second-Order Sensitivities of the PERP Total Leakage Response with Respect to the Parameters Underlying the Benchmark's Isotopic Number Densities and Total Cross Sections

As described in Part I [1], the neutron flux is computed by solving numerically the neutron transport equation using the PARTISN [9] multigroup discrete ordinates transport code. For the PERP benchmark under consideration, PARTISN [9] solves the following multi-group approximation of the neutron transport equation with a spontaneous fission source provided by the code SOURCES4C [10]:

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

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

where r_d denotes the external radius of the PERP benchmark, and where

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

$$Q^g(r) \triangleq \sum_{k=1}^{N_f}\lambda_k N_{k,1}F_k^{SF}\nu_k^{SF}\left(\frac{2}{\sqrt{\pi a_k{}^3 b_k}}e^{-\frac{a_k b_k}{4}}\right)\int_{E^{g+1}}^{E^g} dE e^{-E/a_k}\sinh\sqrt{b_k E}. \tag{4}$$

In Equation (1), the vector $\boldsymbol{\alpha}$ denotes the "vector of imprecisely known model parameters", which has been defined in Part I [1] as $\boldsymbol{\alpha} \triangleq \left[\boldsymbol{\sigma}_t;\boldsymbol{\sigma}_s;\boldsymbol{\sigma}_f;\boldsymbol{\nu};\mathbf{p};\mathbf{q};\mathbf{N}\right]^{\dagger}$. The vector-valued components $\boldsymbol{\sigma}_t$, $\boldsymbol{\sigma}_s$, $\boldsymbol{\sigma}_f$, $\boldsymbol{\nu}$, $\mathbf{p}$, $\mathbf{q}$ and $\mathbf{N}$ comprise the various model parameters for the microscopic total cross sections, scattering cross sections, fission cross sections, average number of neutrons per fission, fission spectra, sources, and isotopic number densities, respectively. For convenient reference, the components of the vector of model parameters $\boldsymbol{\alpha}$ are reproduced in Appendix A.

The PARTISN [9] calculations used the MENDF71X [11] 618-group cross sections collapsed to $G = 30$ energy groups, with group boundaries, E^g, as presented in [1]. The MENDF71X library uses ENDF/B-VII.1 Nuclear Data [12].

The total neutron leakage from the PERP sphere, denoted as $L(\boldsymbol{\alpha})$, will depend (indirectly, through the neutron flux) on the imprecisely known model parameters and is defined as follows:

$$L(\boldsymbol{\alpha}) \triangleq \int\limits_{S_b} dS \sum_{g=1}^{G} \int\limits_{\boldsymbol{\Omega}\cdot\mathbf{n}>0} d\boldsymbol{\Omega}\boldsymbol{\Omega}\cdot\mathbf{n}\,\varphi^g(r,\boldsymbol{\Omega}). \tag{5}$$

As has been shown by Cacuci [5], the 2nd-order mixed sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$ can be computed using two distinct expressions, involving distinct 2nd-level adjoint systems and corresponding 2nd-level adjoint functions, by considering either the computation of $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$ or the computation of $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_t\partial\mathbf{N}$. These two distinct paths will be presented in Sections 2.1 and 2.2, respectively. The end results produced by these two distinct paths must be identical to one another, thus providing a mutual "solution verification" which ensures that the respective computations were performed correctly. Moreover, the computation of $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$ can be significantly more efficient than the computation of $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_t\partial\mathbf{N}$, as will be further illustrated by the numerical results presented in Section 2.3.

2.1. Computing the Second-Order Sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$

The PERP benchmark comprises six distinct isotopes and two distinct materials; the respective isotopes are not repeated in the two materials. Therefore, only the following isotopic number densities exist for this benchmark: $N_{1,1}, N_{2,1}, N_{3,1}, N_{4,1}, N_{5,2}, N_{6,2}$, so that the vector $\mathbf{N}$ is defined as follows:

$$\mathbf{N} \triangleq \left[n_1, \ldots, n_{J_n}\right]^{\dagger} \triangleq \left[N_{1,1}, N_{2,1}, N_{3,1}, N_{4,1}, N_{5,2}, N_{6,2}\right]^{\dagger}, \quad J_n = 6. \tag{6}$$

The vector $\boldsymbol{\sigma}_t$ is defined in Part I [1] and in Equation (A6) in Appendix A.

The equations needed for deriving the expression of the 2nd-order sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$ are obtained by particularizing Equations (158), (167), (177) and (204) in [5] to the PERP benchmark and adding their respective contributions. The expression obtained by particularizing Equation (158) in [5] takes on the following form:

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial t_{m_2}}\right)^{(1)} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi^{(1),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega}) \frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial n_j \partial t_{m_2}} \\
&-\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega}\left[\psi_{1,i}^{(2),g}(r,\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega}) + \psi_{2,i}^{(2),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega})\right]\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}}, \\
&for j = 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma t}.
\end{aligned} \tag{7}$$

The multigroup adjoint fluxes $\psi^{(1),g}(r,\boldsymbol{\Omega}), g = 1, \ldots, G$, appearing in Equation (7) are the solutions of the following 1st-Level Adjoint Sensitivity System (1st-LASS) presented in Equations (156) and (157) of [5]:

$$A^{(1),g}(\boldsymbol{\alpha})\psi^{(1),g}(r,\boldsymbol{\Omega}) = \boldsymbol{\Omega}\cdot\mathbf{n}\delta(r - r_d), \quad g = 1, \ldots, G, \tag{8}$$

$$\psi^{(1),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} > 0, g = 1, \ldots, G, \tag{9}$$

where the adjoint operator $A^{(1),g}(\boldsymbol{\alpha})$ takes on the following particular form of Equation (149) in [5]:

$$
\begin{aligned}
&A^{(1),g}(\boldsymbol{\alpha})\psi^{(1),g}(r,\boldsymbol{\Omega}) \\
&\triangleq -\boldsymbol{\Omega}\cdot\nabla\psi^{(1),g}(r,\boldsymbol{\Omega}) + \Sigma_t^g(\mathbf{t})\,\psi^{(1),g}(r,\boldsymbol{\Omega}) - \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\Sigma_s^{g\to g'}\left(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}'\right)\psi^{(1),g'}\left(r,\boldsymbol{\Omega}'\right) \\
&-\nu\Sigma_f^g(\mathbf{f})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\chi^{g'}\,\psi^{(1),g'}\left(r,\boldsymbol{\Omega}'\right), \quad g=1,\ldots,G.
\end{aligned}
\tag{10}
$$

The 2nd-level adjoint functions $\psi_{1,i}^{(2),g}$ and $\psi_{2,j}^{(2),g}, j = 1,\ldots,J_n; g = 1,\ldots,G$, appearing in Equation (7) are the solutions of the following 2nd-Level Adjoint Sensitivity System (2nd-LASS) obtained from Equations (164)–(166) of [5]:

$$
B^g\left(\boldsymbol{\alpha}^0\right)\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = -\sigma_{t,i_j}^g\varphi^g(r,\boldsymbol{\Omega}),\ j=1,\ldots,J_n;\ g=1,\ldots,G,
\tag{11}
$$

$$
\psi_{1,i}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} < 0; i=1,\ldots,J_n; g=1,\ldots,G,
\tag{12}
$$

$$
A^{(1),g}\left(\boldsymbol{\alpha}^0\right)\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) = -\sigma_{t,i_j}^g\psi^{(1),g}(r,\boldsymbol{\Omega}),\ j=1,\ldots,J_n;\ g=1,\ldots,G,
\tag{13}
$$

$$
\psi_{2,i}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} > 0; i=1,\ldots,J_n; g=1,\ldots,G.
\tag{14}
$$

The parameters n_j and t_{m_2} in Equation (7) correspond to the isotopic number densities and microscopic total cross sections, respectively, and will therefore be denoted as $n_j \equiv N_{i_j,m_j}$ and $t_{m_2} \equiv \sigma_{t,i_{m_2}}^{g_{m_2}}$, where the subscripts i_j and m_j denote the isotope and material associated with the parameter n_j, while the subscripts i_{m_2}, g_{m_2} and m_{m_2} denote the isotope, energy group and material associated with the parameter t_{m_2}, respectively. The following derivatives will be used in Equation (7) and subsequently in this work:

$$
\frac{\partial\Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}} = \frac{\partial\Sigma_t{}^g(\mathbf{t})}{\partial\sigma_{t,i_{m_2}}^{g_{m_2}}} = \frac{\partial\left(\sum_{m=1}^{M}\sum_{i=1}^{I}N_{i,m}\sigma_{t,i}^g\right)}{\partial\sigma_{t,i_{m_2}}^{g_{m_2}}} = \delta_{g_{m_2}g}N_{i_{m_2},m_{m_2}},
\tag{15}
$$

$$
\begin{aligned}
&\frac{\partial^2\Sigma_t{}^g(\mathbf{t})}{\partial n_j\partial t_{m_2}} = \frac{\partial^2\Sigma_t{}^g(\mathbf{t})}{\partial N_{i_j,m_j}\partial\sigma_{t,i_{m_2}}^{g_{m_2}}} = \frac{\partial\left[\partial\Sigma_t{}^g(\mathbf{t})/\partial N_{i_j,m_j}\right]}{\partial\sigma_{t,i_{m_2}}^{g_{m_2}}} = \frac{\partial\left[\partial\left(\sum_{m=1}^{M}\sum_{i=1}^{I}N_{i,m}\sigma_{t,i}^g\right)/\partial N_{i_j,m_j}\right]}{\partial\sigma_{t,i_{m_2}}^{g_{m_2}}} \\
&= \frac{\partial\left[\sigma_{t,i_j}^g\right]}{\partial\sigma_{t,i_{m_2}}^{g_{m_2}}} = \delta_{i_j i_{m_2}}\delta_{g_{m_2}g},
\end{aligned}
\tag{16}
$$

where $\delta_{g_{m_2}g}$ denotes the Kronecker-delta functionals (e.g., $\delta_{g_{m_2}g} = 1$ if $g_{m_2} = g$; $\delta_{g_{m_2}g} = 0$ if $g_{m_2} \neq g$). Inserting the results obtained in Equations (15) and (16) into Equation (7), yields:

$$
\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j\partial t_{m_2}}\right)^{(1)} = -\delta_{i_j i_{m_2}}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega})\varphi^{g_{m_2}}(r,\boldsymbol{\Omega}) \\
&-N_{i_{m_2},m_{m_2}}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\left[\psi_{1,i}^{(2),g_{m_2}}(r,\boldsymbol{\Omega})\psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega}) + \psi_{2,i}^{(2),g_{m_2}}(r,\boldsymbol{\Omega})\varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\right], \\
&for j = 1,\ldots,J_n; m_2 = 1,\ldots,J_{\sigma t}.
\end{aligned}
\tag{17}
$$

The contributions stemming from Equation (167) in [5] have the following expression:

$$
\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j\partial t_{m_2}}\right)^{(2)} = -\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\left[\theta_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega}) + \theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega})\right]\frac{\partial\Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}}, \\
&for j = 1,\ldots,J_n;\ m_2 = 1,\ldots,J_{\sigma t},
\end{aligned}
\tag{18}
$$

where the 2nd-level adjoint functions $\theta_{1,j}^{(2),g}$, and $\theta_{2,j}^{(2),g}$, $j = 1,\ldots,J_n; g = 1,\ldots,G$, are the solutions of the following 2nd-Level Adjoint Sensitivity System resulted from Equations (164)–(166) of [5]:

$$B^g\left(\boldsymbol{\alpha}^0\right)\theta_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = \sum_{g'=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\sigma_{s,l,i_j}^{g'\to g}P_l(\boldsymbol{\Omega})\phi_l^{g'}(r), j = 1,\ldots,J_n;\ g = 1,\ldots,G, \tag{19}$$

$$\theta_{1,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} < 0; j = 1,\ldots,J_n;\ g = 1,\ldots,G, \tag{20}$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)\theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) = \sum_{g'=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\sigma_{s,l,i_j}^{g\to g'}P_l(\boldsymbol{\Omega})\xi_l^{(1),g'}(r), j = 1,\ldots,J_n; g = 1,\ldots,G, \tag{21}$$

$$\theta_{2,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} > 0; j = 1,\ldots,J_n; g = 1,\ldots,G. \tag{22}$$

In Equations (19) and (21), $\sigma_{s,l,i_j}^{g'\to g}$ denotes the l^{th}-order Legendre-expanded microscopic scattering cross section from energy group g' into energy group g for isotope i_j, as defined in Equation (A7) in Appendix A. The l^{th}-moments $\varphi_l^g(r)$ and $\xi_l^{(1),g}(r)$ are defined as follows:

$$\varphi_l^g(r) \triangleq \int_{4\pi} d\boldsymbol{\Omega}P_l(\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega}), \tag{23}$$

$$\xi_l^{(1),g}(r) \triangleq \int_{4\pi} d\boldsymbol{\Omega}P_l(\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega}). \tag{24}$$

Inserting Equation (15) into Equation (18) yields:

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j \partial t_{m_2}}\right)^{(2)} = -N_{i_{m_2},m_{m_2}}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\left[\theta_{1,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega})\psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega}) + \theta_{2,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega})\varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\right], \\ for j = 1,\ldots,J_n;\ m_2 = 1,\ldots,J_{\sigma t}.\end{gathered} \tag{25}$$

The contributions stemming from Equation (177) in [5] have the following expression:

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j \partial t_{m_2}}\right)^{(3)} = -\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\left[u_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega}) + u_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega})\right]\frac{\partial\Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}}, \\ for j = 1,\ldots,J_n;\ m_2 = 1,\ldots,J_{\sigma t},\end{gathered} \tag{26}$$

where the 2nd-level adjoint functions $u_{1,j}^{(2),g}$, and $u_{2,j}^{(2),g}$, $j = 1,\ldots,J_n; g = 1,\ldots,G$, are the solutions of the following 2nd-Level Adjoint Sensitivity System obtained from Equations (183)–(185) of [5]:

$$B^g\left(\boldsymbol{\alpha}^0\right)u_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = \chi^g\sum_{g'=1}^{G}\nu_{i_j}^{g'}\sigma_{f,i_j}^{g'}\varphi_0^{g'}(r), j = 1,\ldots,J_n;\ g = 1,\ldots,G, \tag{27}$$

$$u_{1,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} < 0; j = 1,\ldots,J_n;\ g = 1,\ldots,G, \tag{28}$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)u_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) = \nu_{i_j}^{g}\sigma_{f,i_j}^{g}\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r), j = 1,\ldots,J_n; g = 1,\ldots,G, \tag{29}$$

$$u_{2,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} > 0; j = 1,\ldots,J_n; g = 1,\ldots,G. \tag{30}$$

In Equations (27) and (29), the 0th-moments are defined as follows:

$$\varphi_0^g(r) \triangleq \int_{4\pi} d\boldsymbol{\Omega}\varphi^g(r,\boldsymbol{\Omega}), \tag{31}$$

$$\xi_0^{(1),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega}\psi^{(1),g}(r,\mathbf{\Omega}). \tag{32}$$

Inserting Equation (15) into Equation (26) yields:

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j \partial t_{m_2}}\right)^{(3)} = -N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[u_{1,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g_{m_2}}(r,\mathbf{\Omega}) + u_{2,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\varphi^{g_{m_2}}(r,\mathbf{\Omega})\right], \\ for j = 1,\ldots,J_n;\ \ m_2 = 1,\ldots,J_{\sigma t}.\end{gathered} \tag{33}$$

Finally, the contribution stemming from Equation (204) in [5] has the following expression:

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j \partial t_{m_2}}\right)^{(4)} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} g_{1,j}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega})\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}}, \\ for j = 1,\ldots,J_n;\ \ m_2 = 1,\ldots,J_{\sigma t},\end{gathered} \tag{34}$$

where the 2nd-level adjoint functions $g_{1,j}^{(2),g}, j = 1,\ldots,J_n;\ g = 1,\ldots,G$, are the solutions of the following 2nd-Level Adjoint Sensitivity System resulted from Equations (200) and (202) of [5]:

$$B^g\left(\boldsymbol{\alpha}^0\right)g_{1,j}^{(2),g}(r,\mathbf{\Omega}) = \frac{Q_{SF,i_j}^g}{n_j}, j = 1,\ldots,J_n;\ g = 1,\ldots,G, \tag{35}$$

$$g_{1,j}^{(2),g}(r_d,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} < 0; j = 1,\ldots,J_n;\ \ g = 1,\ldots,G. \tag{36}$$

The (α, n) source is zero for the PERP benchmark. Only the spontaneous fission source is present in the PERP benchmark, which implies that

$$Q^g = Q_{SF}^g = \sum_{m=1}^{M}\sum_{k=1}^{N_f} Q_{SF,k}^g, \tag{37}$$

where the spontaneous source rate density in group g for isotope k is defined as follows [10]:

$$Q_{SF,k}^g = \lambda_k N_{k,m}\chi_{SF,k}^g = \lambda_k N_{k,m} F_k^{SF}\nu_k^{SF}\left(\frac{2}{\sqrt{\pi a_k{}^3 b_k}}e^{-\frac{a_k b_k}{4}}\right)\int_{E^{g+1}}^{E^g} dE e^{-E/a_k}\sinh\sqrt{b_k E}. \tag{38}$$

In Equation (38), the quantity λ_k denotes the decay constant for isotope k, while $\chi_{SF,k}^g$ includes the spontaneous fission branch ratio and the spontaneous fission neutron spectra, which are approximated by a Watt's fission spectra using two evaluated parameters (a_k and b_k).

Inserting Equation (15) into Equation (34) yields:

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j \partial t_{m_2}}\right)^{(4)} = -N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[g_{1,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g_{m_2}}(r,\mathbf{\Omega})\right], \\ for j = 1,\ldots,J_n,\ \ m_2 = 1,\ldots,J_{\sigma t}.\end{gathered} \tag{39}$$

Combining the partial contributions obtained in Equations (17), (25), (33) and (39) yields the following expression:

$$\begin{aligned}
&\frac{\partial^2 L}{\partial n_j \partial t_{m_2}} = \sum_{i=1}^{4}\left(\frac{\partial^2 L}{\partial n_j \partial t_{m_2}}\right)^{(i)} = -\delta_{i_j i_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\psi^{(1),g_{m_2}}(r,\mathbf{\Omega})\varphi^{g_{m_2}}(r,\mathbf{\Omega}) \\
&-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[\psi_{1,i}^{(2),g_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g_{m_2}}(r,\mathbf{\Omega}) + \psi_{2,i}^{(2),g_{m_2}}(r,\mathbf{\Omega})\varphi^{g_{m_2}}(r,\mathbf{\Omega})\right] \\
&-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[\theta_{1,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g_{m_2}}(r,\mathbf{\Omega}) + \theta_{2,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\varphi^{g_{m_2}}(r,\mathbf{\Omega})\right] \\
&-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[u_{1,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g_{m_2}}(r,\mathbf{\Omega}) + u_{2,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\varphi^{g_{m_2}}(r,\mathbf{\Omega})\right] \\
&-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[g_{1,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g_{m_2}}(r,\mathbf{\Omega})\right],\ for\ \ j = 1,\ldots,J_n; m_2 = 1,\ldots,J_{\sigma t}.
\end{aligned} \tag{40}$$

2.2. Alternative Path: Computing the Second-Order Sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_t\partial\mathbf{N}$

As mentioned earlier, the mixed 2nd-order sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$ can be alternatively computed by using the symmetric expression $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_t\partial\mathbf{N}$. The equations needed for deriving the expression for $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_t\partial\mathbf{N}$ are obtained by particularizing Equations (158), (159), (160) and (162) in [5] to the PERP benchmark. The combined expression obtained by particularizing these equations takes on the following form:

$$\begin{aligned}
&\frac{\partial^2 L}{\partial t_j \partial n_{m_2}} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\psi^{(1),g}(r,\mathbf{\Omega})\varphi^{g}(r,\mathbf{\Omega}) \frac{\partial^2 \Sigma_t{}^{g}(\mathbf{t})}{\partial t_j \partial n_{m_2}} \\
&-\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[\psi_{1,i}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega}) + \psi_{2,i}^{(2),g}(r,\mathbf{\Omega})\varphi^{g}(r,\mathbf{\Omega})\right]\frac{\partial \Sigma_t{}^{g}(\mathbf{t})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\psi_{1,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\psi^{(1),g'}(r,\mathbf{\Omega}') \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\psi_{2,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\varphi^{g'}(r,\mathbf{\Omega}') \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\psi_{2,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\varphi^{g'}(r,\mathbf{\Omega}')\chi^{g} \frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f})\right]}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\psi_{1,j}^{(2),g}(r,\mathbf{\Omega}) \frac{\partial\left[(\nu\Sigma_f)^{g}(\mathbf{f})\right]}{\partial n_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}'\chi^{g'}\psi^{(1),g'}(r,\mathbf{\Omega}') \\
&+\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\psi_{2,j}^{(2),g}(r,\mathbf{\Omega}) \frac{\partial Q^{g}(\mathbf{q};r,\mathbf{\Omega})}{\partial n_{m_2}},\ for\ j = 1,\ldots,J_{\sigma t}; m_2 = 1,\ldots,J_n,
\end{aligned} \tag{41}$$

where the adjoint functions $\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}$, $j = 1,\ldots,J_{\sigma t}; g = 1,\ldots,G$ are the solutions of the 2nd-LASS presented in Equations (32), (34), (39) and (40) of Part I [1], which are reproduced below for convenient reference:

$$B^{g}\left(\boldsymbol{\alpha}^0\right)\psi_{1,j}^{(2),g}(r,\mathbf{\Omega}) = -\delta_{g_j g} N_{i_j,m_j}\varphi^{g}(r,\mathbf{\Omega}),\ j = 1,\ldots,J_{\sigma t};\ \ g = 1,\ldots,G, \tag{42}$$

$$\psi_{1,j}^{(2),g}(r_d,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} < 0; j = 1,\ldots,J_{\sigma t}; g = 1,\ldots,G, \tag{43}$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)\psi_{2,j}^{(2),g}(r,\mathbf{\Omega}) = -\delta_{g_j g} N_{i_j,m_j}\psi^{(1),g}(r,\mathbf{\Omega}),\ j = 1,\ldots,J_{\sigma t};\ \ g = 1,\ldots,G, \tag{44}$$

$$\psi_{2,j}^{(2),g}(r_d,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} > 0; j = 1,\ldots,J_{\sigma t}; g = 1,\ldots,G. \tag{45}$$

The parameters t_j and n_{m_2} in Equation (41) correspond to the total cross sections and isotopic number densities, and are therefore denoted as $t_j \equiv \sigma_{t,i_j}^{g_j}$ and $n_{m_2} \equiv N_{i_{m_2},m_{m_2}}$, respectively. The following results will be used in subsequent derivations:

$$\frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial t_j \partial n_{m_2}} = \frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial \sigma_{t,i_j}^{g_j} \partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left[\partial\left(\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\sigma_{t,i}^{g}\right)/\partial\sigma_{t,i_j}^{g_j}\right]}{\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left[\delta_{g_j g} N_{i_j,m_j}\right]}{\partial N_{i_{m_2},m_{m_2}}} = \delta_{i_j i_{m_2}}\delta_{g_j g}, \tag{46}$$

$$\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial n_{m_2}} = \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left(\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\sigma_{t,i}^{g}\right)}{\partial N_{i_{m_2},m_{m_2}}} = \sigma_{t,i_{m_2}}^{g}, \tag{47}$$

$$\frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial n_{m_2}} = \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial N_{i_{m_2},m_{m_2}}} = \sum_{l=0}^{ISCT}(2l+1)\sigma_{s,l,i_{m_2}}^{g\to g'}P_l\left(\mathbf{\Omega}'\cdot\mathbf{\Omega}\right), \tag{48}$$

$$\frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial n_{m_2}} = \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial N_{i_{m_2},m_{m_2}}} = \sum_{l=0}^{ISCT}(2l+1)\sigma_{s,l,i_{m_2}}^{g'\to g}P_l\left(\mathbf{\Omega}'\cdot\mathbf{\Omega}\right), \tag{49}$$

$$\frac{\partial\left(\nu\Sigma_f\right)^g(\mathbf{f})}{\partial n_{m_2}} = \frac{\partial\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\left(\nu\sigma_f\right)_i^g}{\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\nu_i^g\sigma_{f,i}^g}{\partial N_{i_{m_2},m_{m_2}}} = \nu_{i_{m_2}}^g\sigma_{f,i_{m_2}}^g, \tag{50}$$

$$\frac{\partial\left(\nu\Sigma_f\right)^{g'}(\mathbf{f})}{\partial n_{m_2}} = \frac{\partial\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\left(\nu\sigma_f\right)_i^{g'}}{\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\nu_i^{g'}\sigma_{f,i}^{g'}}{\partial N_{i_{m_2},m_{m_2}}} = \nu_{i_{m_2}}^{g'}\sigma_{f,i_{m_2}}^{g'}, \tag{51}$$

$$\frac{\partial Q^g(\mathbf{q};r,\mathbf{\Omega})}{\partial n_{m_2}} = \frac{\partial Q_{SF}^g}{\partial n_{m_2}} = \frac{\partial\sum\limits_{m=1}^{M}\sum\limits_{k=1}^{N_f}\lambda_k N_{k,m}\chi_{SF,k}^g}{\partial N_{i_{m_2},m_{m_2}}} = \lambda_{m_2}\chi_{SF,i_{m_2}}^g = \frac{Q_{SF,i_{m_2}}^g}{N_{i_{m_2},m_{m_2}}} = \frac{Q_{SF,i_{m_2}}^g}{n_{m_2}}. \tag{52}$$

Inserting the results obtained in Equations (46)–(52) into Equation (41), and performing the respective angular integrations yields the following expression for Equation (41):

$$\begin{aligned}
&\tfrac{\partial^2 L}{\partial t_j \partial n_{m_2}} = -\delta_{i_j i_{m_2}}\int_V dV\int_{4\pi} d\mathbf{\Omega}\,\psi^{(1),g_j}(r,\mathbf{\Omega})\varphi^{g_j}(r,\mathbf{\Omega})\\
&-\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\mathbf{\Omega}\left[\psi_{1,j}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega})+\psi_{2,j}^{(2),g}(r,\mathbf{\Omega})\varphi^g(r,\mathbf{\Omega})\right]\sigma_{t,i_{m_2}}^g\\
&+\sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV\xi_{1,j;l}^{(2),g}(r)\sum_{g'=1}^{G}\sigma_{s,l,i_{m_2}}^{g\to g'}\xi_l^{(1),g'}(r)+\sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV\xi_{2,j;l}^{(2),g}(r)\sum_{g'=1}^{G}\sigma_{s,l,i_{m_2}}^{g'\to g}\varphi_l^{g'}(r)\\
&+\sum_{g=1}^{G}\int_V dV\chi^g\xi_{2,j;0}^{(2),g}(r)\sum_{g'=1}^{G}\nu_{i_{m_2}}^{g'}\sigma_{f,i_{m_2}}^{g'}\varphi_0^{g'}(r)+\sum_{g=1}^{G}\int_V dV\nu_{i_{m_2}}^g\sigma_{f,i_{m_2}}^g\xi_{1,j;0}^{(2),g}(r)\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r)\\
&+\tfrac{1}{n_{m_2}}\sum_{g=1}^{G}\int_V dV\xi_{2,j;0}^{(2),g}(r)Q_{SF,i_{m_2}}^g,\ for\ j=1,\ldots,J_{\sigma t};\quad m_2=1,\ldots,J_n,
\end{aligned} \tag{53}$$

where

$$\xi_{1,j;l}^{(2),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega}P_l(\mathbf{\Omega})\psi_{1,j}^{(2),g}(r,\mathbf{\Omega}), \tag{54}$$

$$\xi_{2,j;l}^{(2),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega}P_l(\mathbf{\Omega})\psi_{2,j}^{(2),g}(r,\mathbf{\Omega}), \tag{55}$$

$$\xi_{1,j;0}^{(2),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega}\psi_{1,j}^{(2),g}(r,\mathbf{\Omega}), \tag{56}$$

$$\xi_{2,j;0}^{(2),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega}\psi_{2,j}^{(2),g}(r,\mathbf{\Omega}). \tag{57}$$

2.3. Numerical Results for $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$

The second-order absolute sensitivities, $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$, of the leakage response with respect to the isotopic number densities and the total cross sections for all isotopes of the PERP benchmark have been computed using Equation (40), and have been independently verified by computing $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_t\partial\mathbf{N}$ using Equation (53). Regarding computational requirements: computing $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$ requires 16 forward and adjoint PARTISN transport computations for obtaining the various 2nd-level adjoint functions $\psi_{1,j}^{(2),g}, \psi_{2,j}^{(2),g}, \theta_{1,j}^{(2),g}, \theta_{2,j}^{(2),g}, u_{1,j}^{(2),g}, u_{2,j}^{(2),g}$ and $g_{1,j}^{(2),g}$, $j = 1, \ldots, J_n; g = 1, \ldots, G$ needed in Equation (40). In contradistinction, computing $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_t\partial\mathbf{N}$ using Equation (53) would require $J_{\sigma t} = G \times I = 30 \times 6 = 180$ forward and adjoint PARTISN computations for obtaining the adjoint functions $\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}$, $j = 1, \ldots, J_{\sigma t}; g = 1, \ldots, G$ which are needed in Equation (53). It is thus evident that computing $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$ using Equation (40) is about 10 times more efficient than computing $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_t\partial\mathbf{N}$ using Equation (53).

The matrix $\partial^2 L/\partial n_j \partial t_{m_2}$, $j = 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma t}$ has dimensions $J_n \times J_{\sigma t} (= 6 \times 180)$; corresponding to this matrix is the matrix denoted as $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{t,k}^g\right)$ of 2nd-order relative sensitivities, which is defined as follows:

$$\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{t,k}^g\right) \triangleq \frac{\partial^2 L}{\partial N_{i,m}\partial\sigma_{t,k}^g}\left(\frac{N_{i,m}\sigma_{t,k}^g}{L}\right), \; i, k = 1, \ldots, 6; m = 1, 2; \; g = 1, \ldots, 30. \tag{58}$$

To facilitate the presentation and interpretation of the numerical results, the $J_n \times J_{\sigma t} (= 6 \times 180)$ matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{t,k}^g\right)$ has been partitioned into $J_n \times I = 6 \times 6$ submatrices, each of dimensions $1 \times G = 1 \times 30$. The summary of the main features of each of these submatrices is presented in Table 2 in the following form: when a submatrix comprises elements with relative sensitivities with absolute values that are greater than 1.0, the total number of such elements are counted and shown in the shaded cells of the table. Otherwise, if the relative sensitivities of all the elements of a submatrix have values that lie in the interval $(-1.0, 1.0)$, only the element having the largest absolute value in the submatrix is listed in Table 2, together with the phase-space coordinates of that element. The sub-matrices in Table 2, which comprise components with absolute values greater than 1.0, will be discussed in detail in subsequent sub-sections of this section.

Almost all [i.e., 1072 out of $J_n \times J_{\sigma t} (= 1080)$] of the elements of the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{t,k}^g\right)$, $i, k = 1, \ldots, 6$; $m = 1, 2; g = 1, \ldots, 30$ have negative values. The remaining 8 elements have very small (of the order of 10^{-4} or less) positive values; they are all related to the isotopic number densities of isotopes ^{240}Pu or ^{71}Ga. The results shown in Table 2 indicate that, 125 elements (of the total of 1080 elements) have very large relative sensitivities, greater than 1.0. The majority (123 out of 125) of those large sensitivities involve the isotopic number densities of ^{239}Pu or ^{1}H (namely, $N_{1,1}$ and $N_{6,2}$) and/or the microscopic total cross sections of isotopes ^{239}Pu or ^{1}H (namely, $\sigma_{t,1}^g$ and $\sigma_{t,6}^g$). Of the sensitivities summarized in Table 2, the single largest relative value is $S^{(2)}\left(N_{1,1}, \sigma_{t,6}^{30}\right) = -94.91$. The results in Table 2 also indicate that when the 2nd-order mixed relative sensitivities $S^{(2)}\left(N_{i,m}, \sigma_{t,k}^g\right)$ involve the isotopic number densities of isotopes ^{69}Ga and ^{71}Ga or the microscopic total cross sections of isotopes ^{69}Ga and ^{71}Ga, their absolute values are all smaller than 1.0. Moreover, as shown in Table 2, the element with the most negative value in each of the submatrices involves the microscopic total cross sections for the 12th or the 30th energy group of the isotopes.

Table 2. Summary presentation of the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{t,k}^{g}\right),\ i,k = 1,\ldots,6;\ m = 1,2; g = 1,\ldots,30$

	k **= 1 (^{239}Pu)**	***k*** **= 2 (^{240}Pu)**	***k*** **= 3 (^{69}Ga)**	***k*** **= 4 (^{71}Ga)**	***k*** **= 5 (C)**	***k*** **= 6 (^{1}H)**
$i = 1$ (^{239}Pu)	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,1}^{g}\right)$ 18 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,2}^{g}\right)$ 1 element with absolute value >1.0	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,3}^{g}\right)$ Min. value = -4.51×10^{-2} at g = 12	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,4}^{g}\right)$ Min. value = -3.06×10^{-2} at g = 12	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,5}^{g}\right)$ 12 elements with absolute value >1.0	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,6}^{g}\right)$ 22 elements with absolute values >1.0
$i = 2$ (^{240}Pu)	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,1}^{g}\right)$ 10 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,2}^{g}\right)$ Min. value = -2.05×10^{-1} at g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,3}^{g}\right)$ Min. value = -5.45×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,4}^{g}\right)$ Min. value = -3.70×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,5}^{g}\right)$ 1 element with absolute value >1.0	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,6}^{g}\right)$ 9 elements with absolute values >1.0
$i = 3$ (^{69}Ga)	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{t,1}^{g}\right)$ Min. value = -7.14×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{t,2}^{g}\right)$ Min. value = -4.52×10^{-4} at g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{t,3}^{g}\right)$ Min. value = -3.78×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{t,4}^{g}\right)$ Min. value = -1.40×10^{-5} at g = 13	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{t,5}^{g}\right)$ Min. value = -3.06×10^{-3} at g = 30	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{t,6}^{g}\right)$ Min. value = -3.66×10^{-2} at g = 30
$i = 4$ (^{71}Ga)	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{t,1}^{g}\right)$ Min. value = -4.51×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{t,2}^{g}\right)$ Min. value = -2.85×10^{-4} at g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{t,3}^{g}\right)$ Min. value = -1.32×10^{-5} at g = 13	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{t,4}^{g}\right)$ Min. value = -2.56×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{t,5}^{g}\right)$ Min. value = -1.95×10^{-3} at g = 30	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{t,6}^{g}\right)$ Min. value = -2.33×10^{-2} at g = 30
$i = 5$ (C)	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,1}^{g}\right)$ 9 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,2}^{g}\right)$ Min. value = -1.14×10^{-1} at g = 12	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,3}^{g}\right)$ Min. value = -5.13×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,4}^{g}\right)$ Min. value = -3.51×10^{-3} at g = 22	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,5}^{g}\right)$ 1 element with absolute value >1.0	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,6}^{g}\right)$ 11 elements with absolute values >1.0
$i = 6$ (^{1}H)	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,1}^{g}\right)$ 11 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,2}^{g}\right)$ Min. value = -1.83×10^{-1} at g = 12	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,3}^{g}\right)$ Min. value = -8.21×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,4}^{g}\right)$ Min. value = -6.23×10^{-3} at g = 22	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,5}^{g}\right)$ 1 element with absolute value >1.0	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,6}^{g}\right)$ 19 elements with absolute values >1.0

2.3.1. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,1}^{g}\right), g = 1,\ldots,30$

Table 3 shows the results obtained for the 2nd-order mixed relative sensitivity of the leakage response with respect to the isotopic number density and the microscopic total cross sections of isotope 1 (^{239}Pu), denoted as $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,1}^{g}\right) \triangleq \left(\partial^2 L/\partial N_{i=1,m=1}\partial\sigma_{t,k=1}^{g}\right)\left(N_{1,1}\sigma_{t,1}^{g}/L\right), g = 1,\ldots,30$. The 18 elements that have values greater than 1.0, shown bold in this table, involve the total cross sections of isotope ^{239}Pu for the energy groups $g = 6,\ldots,22$ and $g = 30$. The largest negative value in this submatrix is attained by the relative 2nd-order mixed sensitivity $S^{(2)}\left(N_{1,1}, \sigma_{t,1}^{g=12}\right) = -17.172$, which involves the isotopic number density and the 12th energy group of the total cross sections of isotope ^{239}Pu.

Table 3. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,1}^{g}\right)$, $g = 1, \ldots, 30$.

g	Relative Sensitivities	g	Relative Sensitivities
1	−0.005	16	**−10.430**
2	−0.009	17	**−4.783**
3	−0.026	18	**−2.885**
4	−0.122	19	**−2.242**
5	−0.621	20	**−1.883**
6	**−1.795**	21	**−1.631**
7	**−10.307**	22	**−1.168**
8	**−9.440**	23	−0.934
9	**−10.951**	24	−0.597
10	**−10.978**	25	−0.687
11	**−10.064**	26	−0.732
12	**−17.172**	27	−0.219
13	**−15.138**	28	−0.044
14	**−12.627**	29	−0.392
15	**−9.217**	30	**−5.241**

2.3.2. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,2}^{g}\right), g = 1, \ldots, 30$

The matrix $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,2}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=1,m=1} \partial \sigma_{t,k=2}^{g}\right)\left(N_{1,1} \sigma_{t,2}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of isotope 1 (^{239}Pu) and the microscopic total cross sections of isotope 2 (^{240}Pu), contains a single large element that has an absolute value greater than 1.0, which is $S^{(2)}\left(N_{1,1}, \sigma_{t,2}^{g=12}\right) = -1.005$.

2.3.3. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,5}^{g}\right), g = 1, \ldots, 30$

The submatrix $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,5}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=1,m=1} \partial \sigma_{t,k=5}^{g}\right)\left(N_{1,1} \sigma_{t,5}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2$^{\text{nd}}$-order relative sensitivities of the leakage response with respect to the isotopic number density of isotope 1 (^{239}Pu) and the microscopic total cross sections of isotope 5 (C), is presented in Table 4. This submatrix includes 12 elements that have absolute values greater than 1.0, noted in bold, involving the total cross sections of isotope C for the energy groups $g = 12, \ldots, 22$ and $g = 30$. The largest negative value is displayed by the 2nd-order relative sensitivity of the leakage response with respect to the isotopic number density for ^{239}Pu and the 30th energy group of the total cross section for C, namely, $S^{(2)}\left(N_{1,1}, \sigma_{t,5}^{g=30}\right) = -7.952$.

Table 4. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,5}^{g}\right)$, $g = 1, \ldots, 30$.

g	Relative Sensitivities	g	Relative Sensitivities
1	-4.720×10^{-6}	16	**−2.034**
2	-2.276×10^{-5}	17	**−1.657**
3	-1.017×10^{-4}	18	**−1.441**
4	-6.664×10^{-4}	19	**−1.315**
5	-7.945×10^{-3}	20	**−1.219**
6	−0.019	21	**−1.136**
7	−0.218	22	**−1.040**
8	−0.328	23	−0.962
9	−0.332	24	−0.870
10	−0.387	25	−0.824
11	−0.439	26	−0.758
12	**−1.118**	27	−0.674
13	**−1.363**	28	−0.623
14	**−1.402**	29	−0.605
15	**−1.245**	30	**−7.952**

2.3.4. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,6}^{g}\right), g = 1, \ldots, 30$

Table 5 lists the values of the components of the submatrix $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,6}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=1,m=1} \partial \sigma_{t,k=6}^{g}\right)\left(N_{1,1} \sigma_{t,6}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2nd-order mixed relative sensitivities of the leakage response with respect to the isotopic number density of isotope 1 (^{239}Pu) and the microscopic total cross sections of isotope 6 (^{1}H). This submatrix includes 22 elements that have absolute values greater than 1.0, shown in bold; the large sensitivities involve the total cross sections of isotope ^{1}H for energy groups $g = 9, \ldots, 30$. The largest negative value is for the 2nd-order relative sensitivity of the leakage response with respect to isotopic number density of isotope ^{239}Pu and the 30th energy group of the total cross section of isotope ^{1}H, i.e., $S^{(2)}\left(N_{1,1}, \sigma_{t,6}^{g=30}\right) = -94.909$.

Table 5. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{t,6}^{g}\right),\ g = 1, \ldots, 30$.

g	**Relative Sensitivities**	g	**Relative Sensitivities**
1	-4.001×10^{-6}	16	**−11.421**
2	-2.325×10^{-5}	17	**−11.674**
3	-1.087×10^{-4}	18	**−11.427**
4	-8.747×10^{-4}	19	**−10.979**
5	-7.585×10^{-3}	20	**−10.380**
6	−0.040	21	**−9.744**
7	−0.447	22	**−8.947**
8	−0.646	23	**−8.287**
9	**−1.001**	24	**−7.499**
10	**−1.250**	25	**−7.115**
11	**−1.478**	26	**−6.559**
12	**−3.696**	27	**−5.860**
13	**−4.693**	28	**−5.479**
14	**−5.379**	29	**−5.490**
15	**−5.577**	30	**−94.909**

2.3.5. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,1}^{g}\right), g = 1, \ldots, 30$

Table 6 shows the results obtained for the matrix $\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,1}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=2,m=1} \partial \sigma_{t,k=1}^{g}\right)\left(N_{2,1} \sigma_{t,k=1}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2nd-order mixed relative sensitivity of the leakage response with respect to the isotopic number density of ^{240}Pu and the total cross sections of ^{239}Pu. As highlighted in bold in this table, 10 elements of this submatrix have relative sensitivities with absolute values greater than 1.0. These large mixed relative sensitivities involve the total cross sections of isotope ^{239}Pu for energy groups $g = 7, \ldots, 16$, respectively. The largest negative value in this submatrix is $S^{(2)}\left(N_{2,1}, \sigma_{t,1}^{g=12}\right) = -1.914$, also occurring in the 12th energy group of the total cross sections.

2.3.6. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,5}^{g}\right), g = 1, \ldots, 30$

The matrix $\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,5}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=2,m=1} \partial \sigma_{t,k=5}^{g}\right)\left(N_{2,1} \sigma_{t,5}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of isotope 2 (^{240}Pu) and the microscopic total cross sections of isotope 5 (C), has only one large element with absolute value greater than 1.0, namely, $S^{(2)}\left(N_{2,1}, \sigma_{t,5}^{g=30}\right) = -1.067$, which occurs for the 30th energy group of the total cross sections of isotope C.

Table 6. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,1}^{g}\right), \ g = 1, \ldots, 30.$

g	Relative Sensitivities	g	Relative Sensitivities
1	-4.881×10^{-4}	16	**−1.136**
2	-9.701×10^{-4}	17	−0.525
3	-2.796×10^{-3}	18	−0.320
4	−0.013	19	−0.254
5	−0.068	20	−0.216
6	−0.198	21	−0.190
7	**−1.148**	22	−0.137
8	**−1.052**	23	−0.106
9	**−1.221**	24	−0.067
10	**−1.223**	25	−0.084
11	**−1.121**	26	−0.090
12	**−1.914**	27	−0.005
13	**−1.676**	28	0.0002
14	**−1.386**	29	−0.049
15	**−1.007**	30	−0.639

2.3.7. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,6}^{g}\right), g = 1, \ldots, 30$

Table 7 summaries the 2nd-order relative sensitivities in submatrix $\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,6}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=2,m=1} \partial \sigma_{t,k=6}^{g}\right)\left(N_{2,1} \sigma_{t,6}^{g} / L\right), g = 1, \ldots, 30$, of the leakage response with respect to the isotopic number density of isotope 2 (^{240}Pu) and the microscopic total cross sections of isotope 6 (^{1}H). As shown in bold characters in this table, 9 elements in this submatrix have absolute values greater than 1.0. These 9 elements are related to the energy groups $g = 16, \ldots, 23$ and $g = 30$ of the total cross sections of isotope ^{1}H, respectively. The largest 2nd-order mixed relative sensitivity attained in this submatrix involves the isotopic number density for ^{240}Pu and the 30th energy group of the total cross section for ^{1}H, i.e., $S^{(2)}\left(N_{2,1}, \sigma_{t,6}^{g=30}\right) = -12.741$.

Table 7. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{t,6}^{g}\right), \ g = 1, \ldots, 30.$

g	Relative Sensitivities	g	Relative Sensitivities
1	-4.446×10^{-6}	16	**−1.553**
2	-1.222×10^{-5}	17	**−1.568**
3	-4.406×10^{-5}	18	**−1.526**
4	-2.817×10^{-4}	19	**−1.463**
5	-1.977×10^{-3}	20	**−1.381**
6	-8.680×10^{-3}	21	**−1.295**
7	−0.083	22	**−1.189**
8	−0.107	23	**−1.099**
9	−0.162	24	−0.996
10	−0.201	25	−0.944
11	−0.234	26	−0.868
12	−0.563	27	−0.763
13	−0.685	28	−0.730
14	−0.762	29	−0.740
15	−0.773	30	**−12.741**

2.3.8. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,1}^{g}\right), g = 1, \ldots, 30$

Table 8 presents the results for the submatrix $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,1}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=5,m=2} \partial \sigma_{t,k=1}^{g}\right)\left(N_{5,2} \sigma_{t,1}^{g} / L\right), g = 1, \ldots, 30$, which comprises the 2nd-order relative sensitivities of the leakage response with respect to the isotopic number density of isotope 5 (C) and the microscopic total cross sections of isotope 1 (^{239}Pu). This submatrix contains 9 elements that have absolute values greater than 1.0, as shown in bold in this table. These 9 elements are related to the total cross sections of isotope C for the energy groups $g = 7$ and $g = 9, \ldots, 16$, respectively. The most negative value is $S^{(2)}\left(N_{5,2}, \sigma_{t,1}^{g=12}\right) = -1.803$ for the 2nd-order relative sensitivity with respect to isotopic number density of isotope C and the 12th energy group of the total cross section of isotope ^{239}Pu.

Table 8. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,1}^{g}\right), \ g = 1, \ldots, 30.$

g	Relative Sensitivities	g	Relative Sensitivities
1	-4.471×10^{-4}	16	**−1.177**
2	-8.863×10^{-4}	17	−0.593
3	-2.548×10^{-3}	18	−0.385
4	−0.012	19	−0.306
5	−0.061	20	−0.257
6	−0.178	21	−0.223
7	**−1.044**	22	−0.161
8	−0.982	23	−0.129
9	**−1.146**	24	−0.082
10	**−1.143**	25	−0.096
11	**−1.047**	26	−0.100
12	**−1.803**	27	−0.027
13	**−1.613**	28	−0.005
14	**−1.360**	29	−0.055
15	**−1.004**	30	−0.761

2.3.9. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,5}^{g}\right), g = 1, \ldots, 30$

The submatrix $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,5}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=5,m=2} \partial \sigma_{t,k=5}^{g}\right)\left(N_{5,2} \sigma_{t,5}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2nd-order mixed relative sensitivities of the leakage response with respect to the isotopic number density of isotope 5 (C) and the total cross sections of isotope 5 (C), has a single large element that has an absolute value greater than 1.0, namely $S^{(2)}\left(N_{5,2}, \sigma_{t,5}^{g=30}\right) = -2.016$.

2.3.10. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,6}^{g}\right), g = 1, \ldots, 30$

Table 9 lists the values for the elements of the submatrix $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,6}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=5,m=2} \partial \sigma_{t,k=6}^{g}\right)\left(N_{5,2} \sigma_{t,6}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2nd-order mixed relative sensitivities of the leakage response with respect to the isotopic number density of isotope 5 (C) and the microscopic total cross sections of isotope 6 (^{1}H). This submatrix includes 11 elements, highlighted in bold, which have absolute values greater than 1.0. These 11 elements involve the total cross sections of isotope ^{1}H for energy groups $g = 16, \ldots, 25$, and $g = 30$, respectively. The largest negative value in this submatrix is $S^{(2)}\left(N_{5,2}, \sigma_{t,6}^{g=30}\right) = -14.695$.

Table 9. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{t,6}^{g}\right), \ g = 1, \ldots, 30.$

g	Relative Sensitivities	g	Relative Sensitivities
1	-3.183×10^{-6}	16	**−1.665**
2	-8.920×10^{-6}	17	**−1.676**
3	-3.198×10^{-5}	18	**−1.626**
4	-2.007×10^{-4}	19	**−1.557**
5	-1.438×10^{-3}	20	**−1.471**
6	-6.575×10^{-3}	21	**−1.383**
7	−0.070	22	**−1.273**
8	−0.102	23	**−1.183**
9	−0.161	24	**−1.076**
10	−0.200	25	**−1.024**
11	−0.232	26	−0.947
12	−0.567	27	−0.850
13	−0.715	28	−0.801
14	−0.812	29	−0.804
15	−0.827	30	**−14.695**

2.3.11. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,1}^{g}\right), g = 1, \ldots, 30$

Table 10 presents the results obtained for the submatrix $\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,1}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=6,m=2} \partial \sigma_{t,k=1}^{g}\right)\left(N_{6,2} \sigma_{t,k=1}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2nd-order mixed relative sensitivities of the leakage response with respect to the isotopic number density of isotope 6 (^{1}H) and to the microscopic total cross sections of isotope 1 (^{239}Pu). As highlighted in bold in this table, 11 elements have relative sensitivities with absolute values greater than 1.0. These large 2nd-order mixed relative sensitivities pertain to the total cross sections of isotope ^{239}Pu for energy groups $g = 7, \ldots, 16$ and $g = 30$, respectively. The largest negative value in this submatrix is attained by the relative 2nd-order mixed sensitivity $S^{(2)}\left(N_{6,2}, \sigma_{t,1}^{g=12}\right) = -2.884$.

Table 10. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,1}^{g}\right),\ g = 1, \ldots, 30$.

g	**Relative Sensitivities**	g	**Relative Sensitivities**
1	-7.340×10^{-4}	16	**−1.787**
2	-1.456×10^{-3}	17	−0.892
3	-4.186×10^{-3}	18	−0.597
4	−0.020	19	−0.496
5	−0.101	20	−0.431
6	−0.293	21	−0.386
7	**−1.697**	22	−0.286
8	**−1.554**	23	−0.235
9	**−1.808**	24	−0.153
10	**−1.821**	25	**−0.183**
11	**−1.680**	26	−0.196
12	**−2.884**	27	−0.055
13	**−2.542**	28	−0.011
14	**−2.114**	29	−0.114
15	**−1.546**	30	**−1.933**

2.3.12. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,5}^{g}\right), g = 1, \ldots, 30$

The matrix $\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,5}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=6,m=2} \partial \sigma_{t,k=5}^{g}\right)\left(N_{6,2} \sigma_{t,5}^{g} / L\right), g = 1, \ldots, 30$, comprising the 2$^{\text{nd}}$-order relative sensitivities of the leakage response with respect to the isotopic number density of isotope 6 (^{1}H) and the microscopic total cross sections of isotope 5 (C), contains a single large element that has an absolute value greater than 1.0, which is $S^{(2)}\left(N_{6,2}, \sigma_{t,5}^{g=30}\right) = -3.186$.

2.3.13. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,6}^{g}\right), g = 1, \ldots, 30$

Table 11 lists the values obtained for the 2nd-order relative sensitivities of the leakage response with respect to the isotopic number density of isotope 6 (^{1}H) and the microscopic total cross sections of ^{1}H, which are components of $\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,6}^{g}\right) \triangleq \left(\partial^2 L / \partial N_{i=6,m=2} \partial \sigma_{t,k=6}^{g}\right)\left(N_{6,2} \sigma_{t,6}^{g} / L\right), g = 1, \ldots, 30$. In this submatrix, 19 elements have relative sensitivities with absolute values greater than 1.0, all involving the total cross sections of isotope ^{1}H for energy groups $g = 12, \ldots, 30$. The largest negative value is attained by the 2nd-order mixed relative sensitivity $S^{(2)}\left(N_{6,2}, \sigma_{t,6}^{g=30}\right) = -47.398$, occurring in the 30th energy group of the total cross section of isotope ^{1}H.

Table 11. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{t,6}^{g}\right),\ g = 1, \ldots, 30$.

g	**Relative Sensitivities**	g	**Relative Sensitivities**
1	-9.518×10^{-6}	16	**−3.558**
2	-2.606×10^{-5}	17	**−3.679**
3	-9.353×10^{-5}	18	**−3.656**
4	-5.963×10^{-4}	19	**−3.574**
5	-4.196×10^{-4}	20	**−3.440**
6	−0.018	21	**−3.296**
7	−0.169	22	**−3.102**
8	−0.220	23	**−2.937**
9	−0.332	24	**−2.730**
10	−0.415	25	**−2.630**
11	−0.489	26	**−2.476**
12	**−1.190**	27	**−2.272**
13	**−1.465**	28	**−2.170**
14	**−1.659**	29	**−2.176**
15	**−1.724**	30	**−47.398**

3. Mixed Second-Order Sensitivities of the PERP Total Leakage Response with Respect to the Parameters Underlying the Benchmark's Isotopic Number Densities and Scattering Cross Sections

This Section presents the computation and analysis of the numerical results for the 2nd-order mixed sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N} \partial \boldsymbol{\sigma}_s$ of the leakage response with respect to the isotopic number densities and group-averaged scattering microscopic cross sections for all isotopes of the PERP benchmark. These 2nd-order mixed sensitivities can also be computed by using the symmetric expression $\partial^2 L(\boldsymbol{\alpha})/\partial \boldsymbol{\sigma}_s \partial \mathbf{N}$. These two distinct paths for computing the 2nd-order sensitivities with respect to the isotopic number densities and group-averaged scattering microscopic cross sections will be presented in Section 3.1 and, respectively, Section 3.2. As shown in Section 3.3, below, the pathway for computing $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N} \partial \boldsymbol{\sigma}_s$ turns out to be about 450 times more efficient than the pathway for computing $\partial^2 L(\boldsymbol{\alpha})/\partial \boldsymbol{\sigma}_s \partial \mathbf{N}$.

3.1. Computing the Second-Order Sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N} \partial \boldsymbol{\sigma}_s$

The equations needed for deriving the expressions of the 2nd-order sensitivities $\partial^2 L/\partial n_j \partial s_{m_2}$, $j = 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma s}$ will differ from each other depending on whether the parameter s_{m_2} corresponds to the 0th-order $(l = 0)$ scattering cross sections or to the higher-order $(l \geq 1)$ scattering cross sections. This is because, as shown in Equation (A3) of Appendix A, the zeroth-order scattering cross sections contribute to the total cross sections while the higher-order scattering cross sections do not. Therefore, the zeroth-order scattering cross sections must be considered separately from the higher order scattering cross sections. As described in [1–3] and Appendix A, the total number of zeroth-order $(l = 0)$ scattering cross section comprised in $\boldsymbol{\sigma}_s$ is denoted as $J_{\sigma s,l=0}$, where $J_{\sigma s,l=0} = G \times G \times I$. The total number of higher order (i.e., $l \geq 1$) scattering cross sections comprised in $\boldsymbol{\sigma}_s$ is denoted as $J_{\sigma s,l\geq 1}$, where $J_{\sigma s,l\geq 1} = G \times G \times I \times ISCT$, with $J_{\sigma s,l=0} + J_{\sigma s,l\geq 1} = J_{\sigma s}$. There are two distinct cases, as follows:

Case 1: $\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)_{(n=N, s=\sigma_{s,l=0})}$, $j = 1, \ldots, J_n;\ \ m_2 = 1, \ldots, J_{\sigma s,l=0}$, where the quantities n_j refer to the isotopic number densities, while the quantities s_{m_2} refer to the parameters underlying the 0th-order scattering microscopic cross sections; and

Case 2: $\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)_{(n=N, s=\sigma_{s,l\geq 1})}$, $j = 1, \ldots, J_n;\ \ m_2 = 1, \ldots, \sigma_{s,l\geq 1},$, where the quantities n_j refer to the isotopic number densities, and the quantities s_{m_2} refer to the parameters underlying the l^{th}-order $(l \geq 1)$ scattering microscopic cross sections.

3.1.1. Second-Order Sensitivities $\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)_{(n=N, s=\sigma_{s,l=0})}$, $j = 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma s,l=0}$

The equations needed for deriving the expression of the 2nd-order mixed sensitivities $\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)_{(n=N, s=\sigma_{s,l=0})}$ are obtained by particularizing Equations (158), (159), (167), (168), (177), (178), (204) and (205) in [5] to the PERP benchmark. Specifically, using Equation (158) in [5] to the PERP benchmark yields the following expression:

$$
\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(1)}_{(n=N, s=\sigma_{s,l=0})} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega}\, \psi^{(1),g}(r, \boldsymbol{\Omega}) \varphi^g(r, \boldsymbol{\Omega}) \frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial n_j \partial s_{m_2}} \\
&- \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} \left[\psi_{1,j}^{(2),g}(r, \boldsymbol{\Omega}) \psi^{(1),g}(r, \boldsymbol{\Omega}) + \psi_{2,j}^{(2),g}(r, \boldsymbol{\Omega}) \varphi^g(r, \boldsymbol{\Omega}) \right] \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial s_{m_2}}, \\
&for j = 1, \ldots, J_n\,,\ \ m_2 = 1, \ldots, J_{\sigma s,l=0}\,,
\end{aligned}
\tag{59}
$$

where the adjoint functions $\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}$, $j = 1, \ldots, J_{\sigma f}; g = 1, \ldots, G$ are the solutions of the 2nd-LASS presented previously in Equations (11)–(14). In Equation (59), the parameters n_j and s_{m_2} correspond to the isotopic number densities and microscopic total cross sections, respectively, and are therefore denoted as $n_j \equiv N_{i_j,m_j}$ and $s_{m_2} \equiv \sigma_{s,l_{m_2}=0,i_{m_2}}^{g'_{m_2} \to g_{m_2}}$, where the subscripts $i_{m_2}, l_{m_2}, g'_{m_2}$ and g_{m_2} refer to the isotope, order of Legendre expansion, and energy groups associated with the parameter s_{m_2}, respectively. Noting that

$$\begin{aligned}
&\frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial n_j \partial s_{m_2}} = \frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial N_{i_j,m_j} \partial \sigma_{s,l_{m_2}=0,i_{m_2}}^{g'_{m_2} \to g_{m_2}}} = \frac{\partial \left\{ \partial \left[\sum_{m=1}^{M} \sum_{i=1}^{I} N_{i,m} \left(\sigma_{f,i}^g + \sigma_{c,i}^g + \sum_{g'=1}^{G} \sigma_{s,l=0,i}^{g \to g'} \right) \right] / \partial N_{i_j,m_j} \right\}}{\partial \sigma_{s,l_{m_2}=0,i_{m_2}}^{g'_{m_2} \to g_{m_2}}} \\
&= \frac{\partial \left\{ \partial \left[\sum_{m=1}^{M} \sum_{i=1}^{I} \sum_{g'=1}^{G} N_{i,m} \sigma_{s,l=0,i}^{g \to g'} \right] / \partial N_{i_j,m_j} \right\}}{\partial \sigma_{s,l_{m_2}=0,i_{m_2}}^{g'_{m_2} \to g_{m_2}}} = \frac{\partial \left\{ \sum_{g'=1}^{G} \sigma_{s,l=0,i_j}^{g \to g'} \right\}}{\partial \sigma_{s,l_{m_2}=0,i_{m_2}}^{g'_{m_2} \to g_{m_2}}} = \delta_{i_j i_{m_2}} \delta_{g'_{m_2} g'},
\end{aligned} \tag{60}$$

$$\begin{aligned}
&\frac{\partial \Sigma_t^g(\mathbf{t})}{\partial s_{m_2}} = \frac{\partial \left[\sum_{m=1}^{M} \sum_{i=1}^{I} N_{i,m} \sigma_{t,i}^g(\mathbf{t}) \right]}{\partial \sigma_{s,l_{m_2}=0,i_{m_2}}^{g'_{m_2} \to g_{m_2}}} = \frac{\partial \left\{ \sum_{m=1}^{M} \sum_{i=1}^{I} N_{i,m} \left[\sigma_{f,i}^g(\mathbf{f}) + \sigma_{c,i}^g(\mathbf{c}) + \sum_{g'=1}^{G} \sigma_{s,l=0,i}^{g \to g'}(\mathbf{s}) \right] \right\}}{\partial \sigma_{s,l_{m_2}=0,i_{m_2}}^{g'_{m_2} \to g_{m_2}}} \\
&= \frac{\partial \left[\sum_{m=1}^{M} \sum_{i=1}^{I} \sum_{g'=1}^{G} N_{i,m} \sigma_{s,l=0,i}^{g \to g'}(\mathbf{s}) \right]}{\partial \sigma_{s,l_{m_2}=0,i_{m_2}}^{g'_{m_2} \to g_{m_2}}} = \delta_{g'_{m_2} g} N_{i_{m_2},m_{m_2}},
\end{aligned} \tag{61}$$

and inserting the results obtained in Equations (60) and (61) into Equation (59) yields the following expression for Equation (59):

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}} \right)^{(1)}_{(n=N, s=\sigma_{s,l=0})} = -\delta_{i_j i_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega} \psi^{(1),g'_{m_2}}(r, \mathbf{\Omega}) \varphi^{g'_{m_2}}(r, \mathbf{\Omega}) \\
&- N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega} \left[\psi_{1,j}^{(2),g'_{m_2}}(r, \mathbf{\Omega}) \psi^{(1),g'_{m_2}}(r, \mathbf{\Omega}) + \psi_{2,j}^{(2),g'_{m_2}}(r, \mathbf{\Omega}) \varphi^{g'_{m_2}}(r, \mathbf{\Omega}) \right], \\
&for\ j = 1, \ldots, J_n, m_2 = 1, \ldots, J_{\sigma s,l=0}.
\end{aligned} \tag{62}$$

Additional contributions stem from Equation (159) in [5], which takes on the following particular form:

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}} \right)^{(2)}_{(n=N, s=\sigma_{s,l=0})} = \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \psi_{1,j}^{(2),g}(r, \mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \psi^{(1),g'}(r, \mathbf{\Omega}') \frac{\partial \Sigma_s^{g \to g'}(\mathbf{s}; \mathbf{\Omega} \to \mathbf{\Omega}')}{\partial s_{m_2}} \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \psi_{2,j}^{(2),g}(r, \mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r, \mathbf{\Omega}') \frac{\partial \Sigma_s^{g' \to g}(\mathbf{s}; \mathbf{\Omega}' \to \mathbf{\Omega})}{\partial s_{m_2}}, \\
&for\ j = 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma s,l=0}.
\end{aligned} \tag{63}$$

Noting that

$$\begin{aligned}
&\frac{\partial \Sigma_s^{g \to g'}(\mathbf{s}; \mathbf{\Omega} \to \mathbf{\Omega}')}{\partial s_{m_2}} = \frac{\partial \Sigma_s^{g \to g'}(\mathbf{s}; \mathbf{\Omega} \to \mathbf{\Omega}')}{\partial \sigma_{s,l_{m_2},i_{m_2}}^{g'_{m_2} \to g_{m_2}}} = \frac{\partial \left[\sum_{m=1}^{M} \sum_{i=1}^{I} N_{i,m} \sigma_{s,i}^{g \to g'}(\mathbf{s}; \mathbf{\Omega} \to \mathbf{\Omega}') \right]}{\partial \sigma_{s,l_{m_2},i_{m_2}}^{g'_{m_2} \to g_{m_2}}} \\
&= \frac{\partial \left[\sum_{m=1}^{M} \sum_{i=1}^{I} \sum_{l=0}^{ISCT} N_{i,m} (2l+1) \sigma_{s,l,i}^{g \to g'} P_l(\mathbf{\Omega}' \cdot \mathbf{\Omega}) \right]}{\partial \sigma_{s,l_{m_2},i_{m_2}}^{g'_{m_2} \to g_{m_2}}} = \delta_{g'_{m_2} g} \delta_{g_{m_2} g'} N_{i_{m_2},m_{m_2}} (2l_{m_2} + 1) P_{l_{m_2}}(\mathbf{\Omega}' \cdot \mathbf{\Omega}),
\end{aligned} \tag{64}$$

$$\frac{\partial \Sigma_s^{g' \to g}(\mathbf{s}; \mathbf{\Omega}' \to \mathbf{\Omega})}{\partial s_{m_2}} = \frac{\partial \Sigma_s^{g' \to g}(\mathbf{s}; \mathbf{\Omega}' \to \mathbf{\Omega})}{\partial \sigma_{s,l_{m_2},i_{m_2}}^{g'_{m_2} \to g_{m_2}}} = \delta_{g_{m_2} g} \delta_{g'_{m_2} g'} N_{i_{m_2},m_{m_2}} (2l_{m_2} + 1) P_{l_{m_2}}\left(\mathbf{\Omega}' \cdot \mathbf{\Omega} \right), \tag{65}$$

inserting the results obtained in Equations (64) and (65) into Equation (63), using the addition theorem for spherical harmonics in one-dimensional geometry, performing the respective angular integrations, and setting $l_{m_2} = 0$, yields the following expression for Equation (63):

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(2)}_{(n=N,s=\sigma_{s,l=0})} = N_{i_{m_2},m_{m_2}} \int_V dV \left[\xi_0^{(1),g_{m_2}}(r)\xi_{1,j;0}^{(2),g'_{m_2}}(r) + \varphi_0^{g'_{m_2}}(r)\xi_{2,j;0}^{(2),g_{m_2}}(r)\right], \\ &for j = 1,\dots,J_n; m_2 = 1,\dots,J_{\sigma s,l=0}.\end{aligned} \tag{66}$$

Using Equation (167) in [5] in conjunction with the relations $\frac{\partial^2 L}{\partial s_j \partial t_{m_2}} \frac{\partial s_j}{\partial n_j} \frac{\partial t_{m_2}}{\partial s_{m_2}} = \frac{\partial^2 L}{\partial n_j \partial s_{m_2}}$ and $\frac{\partial \Sigma_t{}^g}{\partial t_{m_2}} \frac{\partial t_{m_2}}{\partial s_{m_2}} = \frac{\partial \Sigma_t{}^g}{\partial s_{m_2}}$ yields the following expression:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(3)}_{(n=N,s=\sigma_{s,l=0})} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \left[\theta_{1,j}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega}) + \theta_{2,j}^{(2),g}(r,\mathbf{\Omega})\varphi^g(r,\mathbf{\Omega})\right] \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial s_{m_2}}, \\ &for j = 1,\dots,J_n; m_2 = 1,\dots,J_{\sigma s,l=0},\end{aligned} \tag{67}$$

where the 2nd-level adjoint functions $\theta_{1,j}^{(2),g}$, and $\theta_{2,j}^{(2),g}$, $j = 1,\dots,J_n; g = 1,\dots,G$, are the solutions of the 2nd-Level Adjoint Sensitivity System presented previously in Equations (19)–(22). Inserting the results obtained in Equation (61) into Equation (67), yields the following expression:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(3)}_{(n=N,s=\sigma_{s,l=0})} = -N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega} \left[\theta_{1,j}^{(2),g'_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g'_{m_2}}(r,\mathbf{\Omega}) + \theta_{2,j}^{(2),g'_{m_2}}(r,\mathbf{\Omega})\varphi^{g'_{m_2}}(r,\mathbf{\Omega})\right], \\ &for j = 1,\dots,J_n; m_2 = 1,\dots,J_{\sigma s,l=0}.\end{aligned} \tag{68}$$

The contributions stemming from Equation (168) in [5] to the PERP benchmark are computed as follows:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(4)}_{(n=N,s=\sigma_{s,l=0})} = \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \psi^{(1),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r,\mathbf{\Omega}') \frac{\partial^2 \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial n_j \partial s_{m_2}} \\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{1,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \psi^{(1),g'}(r,\mathbf{\Omega}') \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}} \\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{2,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r,\mathbf{\Omega}') \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial s_{m_2}}, \\ &for\ j = 1,\dots,J_n; m_2 = 1,\dots,J_{\sigma s,l=0}.\end{aligned} \tag{69}$$

Noting that

$$\begin{aligned}&\frac{\partial^2 \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial n_j \partial s_{m_2}} = \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial N_{i_j,m_j} \partial \sigma_{s,l_{m_2},i_{m_2}}^{g'_{m_2}\to g_{m_2}}} = \frac{\partial\left\{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I} N_{i,m}\sigma_{s,i}^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})\right]/\partial N_{i_j,m_j}\right\}}{\partial \sigma_{s,l_{m_2},i_{m_2}}^{g'_{m_2}\to g_{m_2}}} \\ &= \frac{\partial\left\{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I}\sum_{l=0}^{ISCT} N_{i,m}(2l+1)\sigma_{s,l,i}^{g'\to g}P_l(\mathbf{\Omega}'\cdot\mathbf{\Omega})\right]/\partial N_{i_j,m_j}\right\}}{\partial \sigma_{s,l_{m_2},i_{m_2}}^{g'_{m_2}\to g_{m_2}}} = \frac{\partial\left\{\sum_{l=0}^{ISCT}(2l+1)\sigma_{s,l,i_j}^{g'\to g}P_l(\mathbf{\Omega}'\cdot\mathbf{\Omega})\right\}}{\partial \sigma_{s,l_{m_2},i_{m_2}}^{g'_{m_2}\to g_{m_2}}} \\ &= \delta_{i_j i_{m_2}}\delta_{g'_{m_2}g'}\delta_{g_{m_2}g}(2l_{m_2}+1)P_{l_{m_2}}(\mathbf{\Omega}'\cdot\mathbf{\Omega}),\end{aligned} \tag{70}$$

inserting Equations (64), (65) and (70) into Equation (69), using the addition theorem for spherical harmonics in one-dimensional geometry, performing the respective angular integrations, and setting $l_{m_2} = 0$ into the resulting expression yields:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(4)}_{(n=N,s=\sigma_{s,l=0})} = \delta_{i_j i_{m_2}} \xi_0^{(1),g_{m_2}}(r)\varphi_0^{g'_{m_2}}(r) \\ &+N_{i_{m_2},m_{m_2}} \int_V dV \left[\xi_0^{(1),g_{m_2}}(r)\Theta_{1,j;0}^{(2),g'_{m_2}}(r) + \varphi_0^{g'_{m_2}}(r)\Theta_{2,j;0}^{(2),g_{m_2}}(r)\right], for j = 1,\dots,J_n; m_2 = 1,\dots,J_{\sigma s,l=0},\end{aligned} \tag{71}$$

where

$$\Theta_{1,j;0}^{(2),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega}\theta_{1,j}^{(2),g}(r,\mathbf{\Omega}), \quad (72)$$

$$\Theta_{2,j;0}^{(2),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega}\theta_{2,j}^{(2),g}(r,\mathbf{\Omega}). \quad (73)$$

Contributions from the fission cross sections are computed by particularizing Equation (177) in [5], in conjunction with the relations $\frac{\partial^2 L}{\partial f_j \partial t_{m_2}} \frac{\partial f_j}{\partial n_j} \frac{\partial t_{m_2}}{\partial s_{m_2}} = \frac{\partial^2 L}{\partial n_j \partial s_{m_2}}$ and $\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}} \frac{\partial t_{m_2}}{\partial s_{m_2}} = \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial s_{m_2}}$, to obtain:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(5)}_{(n=N,s=\sigma_{s,l=0})} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[u_{1,j}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega}) + u_{2,j}^{(2),g}(r,\mathbf{\Omega})\varphi^{g}(r,\mathbf{\Omega})\right]\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial s_{m_2}},\\ &for \quad j = 1,\dots,J_n; \;\; m_2 = 1,\dots,J_{\sigma s,l=0},\end{aligned} \quad (74)$$

where the 2nd-level adjoint functions $u_{1,j}^{(2),g}$, and $u_{2,j}^{(2),g}$, $j = 1,\dots,J_n; g = 1,\dots,G$, are the solutions of the 2nd-Level Adjoint Sensitivity System presented previously in Equations (27)–(30). Replacing the result obtained in Equation (61) into Equation (74) yields the following expression:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(5)}_{(n=N,s=\sigma_{s,l=0})} = -N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[u_{1,j}^{(2),g'_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g'_{m_2}}(r,\mathbf{\Omega}) + u_{2,j}^{(2),g'_{m_2}}(r,\mathbf{\Omega})\varphi^{g'_{m_2}}(r,\mathbf{\Omega})\right],\\ &for \quad j = 1,\dots,J_n; \;\; m_2 = 1,\dots,J_{\sigma s,l=0}.\end{aligned} \quad (75)$$

Contributions stemming from particularizing Equation (178) in [5] to the PERP benchmark take on the following form:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(6)}_{(n=N,s=\sigma_{s,l=0})} = \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} u_{1,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \psi^{(1),g'}(r,\mathbf{\Omega}') \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\mathbf{\Omega}\to\mathbf{\Omega}')}{\partial s_{m_2}}\\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} u_{2,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(\mathbf{r},\mathbf{\Omega}') \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\mathbf{\Omega}'\to\mathbf{\Omega})}{\partial s_{m_2}},\\ &for\ j = 1,\dots,J_n;\ m_2 = 1,\dots,J_{\sigma s,l=0}.\end{aligned} \quad (76)$$

Inserting the results obtained in Equations (64) and (65) into Equation (76), using the addition theorem for spherical harmonics in one-dimensional geometry, performing the respective angular integrations, and finally setting $l_{m_2} = 0$ in the resulting expression yields the following expression for Equation (76):

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(6)}_{(n=N,s=\sigma_{s,l=0})} = N_{i_{m_2},m_{m_2}} \int_V dV\left[\xi_0^{(1),g_{m_2}}(r) U_{1,j;0}^{(2),g'_{m_2}}(r) + \varphi_0^{g'_{m_2}}(r) U_{2,j;0}^{(2),g_{m_2}}(r)\right],\\ &for j = 1,\dots,J_n; m_2 = 1,\dots,J_{\sigma s,l=0},\end{aligned} \quad (77)$$

where

$$U_{1,j;0}^{(2),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega} u_{1,j}^{(2),g}(r,\mathbf{\Omega}), \quad (78)$$

$$U_{2,j;0}^{(2),g}(r) \triangleq \int_{4\pi} d\mathbf{\Omega} u_{2,j}^{(2),g}(r,\mathbf{\Omega}). \quad (79)$$

Contributions stemming from the source are computed by particularizing Equations (204) and (205) in [5] to the PERP benchmark. Particularizing Equation (204) in [5], in conjunction with the relations $\frac{\partial^2 L}{\partial q_j \partial t_{m_2}} \frac{\partial q_j}{\partial n_j} \frac{\partial t_{m_2}}{\partial s_{m_2}} = \frac{\partial^2 L}{\partial n_j \partial s_{m_2}}$ and $\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}} \frac{\partial t_{m_2}}{\partial s_{m_2}} = \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial s_{m_2}}$, yields:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(7)}_{(n=N,s=\sigma_{s,l=0})} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} g_{1,j}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega}) \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial s_{m_2}},\\ &for \quad j = 1,\dots,J_n; \;\; m_2 = 1,\dots,J_{\sigma s,l=0},\end{aligned} \quad (80)$$

where the 2nd-level adjoint functions $g_{1,j}^{(2),g}, j = 1, \ldots, J_n;\ g = 1, \ldots, G$, are the solutions of the 2nd-Level Adjoint Sensitivity System presented previously in Equations (35) and (36). Inserting the result obtained in Equation (61) into Equation (80) yields the following expression:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(7)}_{(n=N,s=\sigma_{s,l=0})} = -N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} g_{1,j}^{(2),g'{}_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g'{}_{m_2}}(r,\boldsymbol{\Omega}),\\ &for\quad j = 1, \ldots, J_n;\ \ m_2 = 1, \ldots, J_{\sigma s,l=0}.\end{aligned} \tag{81}$$

Particularizing Equation (205) in [5] to the PERP benchmark yields the following contributions:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(8)}_{(n=N,s=\sigma_{s,l=0})} = \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} g_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \psi^{(1),g'}\left(r,\boldsymbol{\Omega}'\right) \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}},\\ &for j = 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma s,l=0}.\end{aligned} \tag{82}$$

Inserting the results obtained in Equations (64) into Equation (82), using the addition theorem for spherical harmonics in one-dimensional geometry, performing the respective angular integrations, and finally setting $l_{m_2} = 0$ in the resulting expression yields the following simplified expression for Equation (82):

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(8)}_{(n=N,s=\sigma_{s,l=0})} = N_{i_{m_2},m_{m_2}} \int_V dV \xi_0^{(1),g_{m_2}}(r) G_{1,j;0}^{(2),g'{}_{m_2}}(r),\\ &for j = 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma s,l=0},\end{aligned} \tag{83}$$

where

$$G_{1,j;0}^{(2),g}(r) \triangleq \int_{4\pi} d\boldsymbol{\Omega} g_{1,j}^{(2),g}(r,\boldsymbol{\Omega}). \tag{84}$$

Collecting the partial contributions obtained in Equations (62), (66), (68), (71), (75), (77), (81) and (83), yields the following result:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)_{(n=N,s=\sigma_{s,l=0})} = \sum_{i=1}^{8}\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)^{(i)}_{(n=N,s=\sigma_{s,l=0})} = -\delta_{i_j i_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} \psi^{(1),g'{}_{m_2}}(r,\boldsymbol{\Omega}) \varphi^{g'{}_{m_2}}(r,\boldsymbol{\Omega})\\ &-N_{i_{m_2},m_{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}} \int_V dV\left[\xi_0^{(1),g_{m_2}}(r) \xi_{1,j;0}^{(2),g'{}_{m_2}}(r) + \varphi_0^{g'{}_{m_2}}(r) \xi_{2,j;0}^{(2),g_{m_2}}(r)\right]\\ &-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega}\left[\theta_{1,j}^{(2),g'{}_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g'{}_{m_2}}(r,\boldsymbol{\Omega}) + \theta_{2,j}^{(2),g'{}_{m_2}}(r,\boldsymbol{\Omega}) \varphi^{g'{}_{m_2}}(r,\boldsymbol{\Omega})\right]\\ &+\delta_{i_j i_{m_2}} \xi_0^{(1),g_{m_2}}(r) \varphi_0^{g'{}_{m_2}}(r) + N_{i_{m_2},m_{m_2}} \int_V dV\left[\xi_0^{(1),g_{m_2}}(r) \Theta_{1,j;0}^{(2),g'{}_{m_2}}(r) + \varphi_0^{g'{}_{m_2}}(r) \Theta_{2,j;0}^{(2),g_{m_2}}(r)\right]\\ &-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega}\left[u_{1,j}^{(2),g'{}_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g'{}_{m_2}}(r,\boldsymbol{\Omega}) + u_{2,j}^{(2),g'{}_{m_2}}(r,\boldsymbol{\Omega}) \varphi^{g'{}_{m_2}}(r,\boldsymbol{\Omega})\right]\\ &+N_{i_{m_2},m_{m_2}} \int_V dV\left[\xi_0^{(1),g_{m_2}}(r) U_{1,j;0}^{(2),g'{}_{m_2}}(r) + \varphi_0^{g'{}_{m_2}}(r) U_{2,j;0}^{(2),g_{m_2}}(r)\right]\\ &-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} g_{1,j}^{(2),g'{}_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g'{}_{m_2}}(r,\boldsymbol{\Omega})\\ &+N_{i_{m_2},m_{m_2}} \int_V dV \xi_0^{(1),g_{m_2}}(r) G_{1,j;0}^{(2),g'{}_{m_2}}(r),\ \ for\ j = 1, \ldots, J_n;\ \ m_2 = 1, \ldots, J_{\sigma s,l=0}\,.\end{aligned} \tag{85}$$

3.1.2. Second-Order Sensitivities $\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)_{(n=N,s=\sigma_{s,l\geq 1})}, j = 1, \ldots, J_n; m_2 = 1, \ldots, \sigma_{s,l\geq 1}$

For the 2nd-order sensitivities $\left(\partial^2 L/\partial n_j \partial s_{m_2}\right)_{(n=N,s=\sigma_{s,l\geq 1})},\ j = 1, \ldots, J_n;\ \ m_2 = 1, \ldots, \sigma_{s,l\geq 1}$, the parameters $n_j \equiv N_{i_j,m_j}$ correspond to the isotopic number densities, and the parameters $s_{m_2} \equiv \sigma_{s,l_{m_2},i_{m_2}}^{g'{}_{m_2}\to g_{m_2}}$ correspond to the l^{th}-order $(l \geq 1)$ scattering cross sections. For this case, the expression for $\left(\partial^2 L/\partial n_j \partial s_{m_2}\right)_{(n=N,s=\sigma_{s,l\geq 1})}$ is obtained by particularizing Equations (159), (168), (178) and (205) in [5] to the PERP benchmark, which yields,

$$
\begin{aligned}
\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)_{(n=N,s=\sigma_{s,l\geq1})} &= \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\psi^{(1),g'}(r,\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}} \\
&+ \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_{m_2}} \\
&+ \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi^{(1),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\frac{\partial^2\Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial n_j\partial s_{m_2}} \\
&+ \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\theta_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\psi^{(1),g'}(r,\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}} \\
&+ \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_{m_2}} \\
&+ \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}u_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\psi^{(1),g'}(r,\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}} \\
&+ \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}u_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(\mathbf{r},\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_{m_2}} \\
&+ \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}g_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\psi^{(1),g'}(r,\boldsymbol{\Omega}')\frac{\partial\Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial s_{m_2}}, \\
&for\ \ j=1,\ldots,J_n;m_2=1,\ldots,J_{\sigma s,l\geq1},
\end{aligned}
\tag{86}
$$

where the 2nd-level adjoint functions $\psi_{1,i}^{(2),g},\psi_{2,i}^{(2),g},\theta_{1,i}^{(2),g},\theta_{2,i}^{(2),g},u_{1,i}^{(2),g},u_{2,i}^{(2),g}$ and $g_{2,j}^{(2),g}$ for $j=1,\ldots,J_n;g=1,\ldots,G$ are the same as those presented in Section 3.1.1, above. Inserting the results obtained in Equations (64), (65) and (70) into Equation (86), using the addition theorem for spherical harmonics in one-dimensional geometry and performing the respective angular integrations, yields the following expression:

$$
\begin{aligned}
\left(\frac{\partial^2 L}{\partial n_j \partial s_{m_2}}\right)_{(n=N,s=\sigma_{s,l\geq1})} &= N_{i_{m_2},m_{m_2}}(2l_{m_2}+1)\int_V dV\left[\xi_{l_{m2}}^{(1),g_{m_2}}(r)\xi_{1,j;l_{m2}}^{(2),g'{}_{m_2}}(r)+\varphi_{l_{m2}}^{g'{}_{m_2}}(r)\xi_{2,j;l_{m2}}^{(2),g_{m_2}}(r)\right] \\
&+\delta_{i_j i_{m_2}}(2l_{m_2}+1)\xi_{l_{m_2}}^{(1),g_{m_2}}(r)\varphi_{l_{m2}}^{g'{}_{m_2}}(r) \\
&+N_{i_{m_2},m_{m_2}}(2l_{m_2}+1)\int_V dV\left[\xi_{l_{m2}}^{(1),g_{m_2}}(r)\Theta_{1,j;l_{m2}}^{(2),g'{}_{m_2}}(r)+\varphi_{l_{m2}}^{g'{}_{m_2}}(r)\Theta_{2,j;l_{m2}}^{(2),g_{m_2}}(r)\right] \\
&+N_{i_{m_2},m_{m_2}}(2l_{m_2}+1)\int_V dV\left[\xi_{l_{m2}}^{(1),g_{m_2}}(r)U_{1,j;l_{m2}}^{(2),g'{}_{m_2}}(r)+\varphi_{l_{m2}}^{g'{}_{m_2}}(r)U_{2,j;l_{m2}}^{(2),g_{m_2}}(r)\right] \\
&+N_{i_{m_2},m_{m_2}}(2l_{m_2}+1)\int_V dV\xi_{l_{m2}}^{(1),g_{m_2}}(r)G_{1,j;l_{m2}}^{(2),g'{}_{m_2}}(r), \\
&for\ j=1,\ldots,J_n;m_2=1,\ldots,J_{\sigma s,l\geq1},
\end{aligned}
\tag{87}
$$

where

$$\Theta_{1,j;l}^{(2),g}(r)\triangleq\int_{4\pi}d\boldsymbol{\Omega}P_l(\boldsymbol{\Omega})\theta_{1,j}^{(2),g}(r,\boldsymbol{\Omega}), \tag{88}$$

$$\Theta_{2,j;l}^{(2),g}(r)\triangleq\int_{4\pi}d\boldsymbol{\Omega}P_l(\boldsymbol{\Omega})\theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega}), \tag{89}$$

$$U_{1,j;l}^{(2),g}(r)\triangleq\int_{4\pi}d\boldsymbol{\Omega}P_l(\boldsymbol{\Omega})u_{1,j}^{(2),g}(r,\boldsymbol{\Omega}), \tag{90}$$

$$U_{2,j;l}^{(2),g}(r)\triangleq\int_{4\pi}d\boldsymbol{\Omega}P_l(\boldsymbol{\Omega})u_{2,j}^{(2),g}(r,\boldsymbol{\Omega}), \tag{91}$$

$$G_{1,j;l}^{(2),g}(r)\triangleq\int_{4\pi}d\boldsymbol{\Omega}P_l(\boldsymbol{\Omega})g_{1,j}^{(2),g}(r,\boldsymbol{\Omega}). \tag{92}$$

3.2. Alternative Path: Computing the Second-Order Sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_s\partial\mathbf{N}$

The results computed using the expressions for $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_s$ obtained in Eqs. (85) and (87) can be verified by obtaining the expressions for the symmetric expression $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_s\partial\mathbf{N}$, the computation of which also requires separate consideration of the zeroth-order scattering cross sections. The two cases involved are as follows:

Case 1: $\left(\frac{\partial^2 L}{\partial s_j \partial n_{m_2}}\right)_{(s=\sigma_{s,l=0},n=N)}$, $j=1,\ldots,J_{\sigma s,l=0}; m_2=1,\ldots,J_n$, where the quantity s_j refers to the parameters underlying the 0th-order scattering cross sections while n_{m_2} refers to the isotopic number densities;

Case 2: $\left(\frac{\partial^2 L}{\partial s_j \partial n_{m_2}}\right)_{(s=\sigma_{s,l\geq 1},n=N)}$, $j=1,\ldots,\sigma_{s,l\geq 1}; m_2=1,\ldots,J_n$, where s_j refers to the parameters underlying the l^{th}-order ($l \geq 1$) scattering cross sections and where n_{m_2} refers to the isotopic number densities.

3.2.1. Second-Order Sensitivities $\left(\partial^2 L/\partial s_j \partial n_{m_2}\right)_{(s=\sigma_{s,l=0},n=N)}$, $j=1,\ldots,J_{\sigma s,l=0}; m_2=1,\ldots,J_n$

The equations needed for deriving the expression of the 2nd-order mixed sensitivities $\left(\partial^2 L/\partial s_j \partial n_{m_2}\right)_{(s=\sigma_{s,l=0},n=N)}$, $j=1,\ldots,J_{\sigma s,l=0}; m_2=1,\ldots,J_n$ are obtained by particularizing Equations (158), (159), (160), (162), (167), (168), (169) and (171) in [5] to the PERP benchmark. This procedure leads to the following expression:

$$
\begin{aligned}
&\left(\frac{\partial^2 L}{\partial s_j \partial n_{m_2}}\right)_{(s=\sigma_{s,l=0},n=N)} = -\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi^{(1),g}(r,\boldsymbol{\Omega})\varphi^{g}(r,\boldsymbol{\Omega})\frac{\partial^2 \Sigma_t{}^{g}(\mathbf{t})}{\partial s_j \partial n_{m_2}} \\
&-\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\left[\psi_{1,i}^{(2),g}(r,\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega})+\psi_{2,i}^{(2),g}(r,\boldsymbol{\Omega})\varphi^{g}(r,\boldsymbol{\Omega})\right]\frac{\partial \Sigma_t{}^{g}(\mathbf{t})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\psi^{(1),g'}(r,\boldsymbol{\Omega}')\frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\varphi^{g'}(r,\boldsymbol{\Omega}')\frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\varphi^{g'}(r,\boldsymbol{\Omega}')\chi^{g}\frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}(\mathbf{f})\right]}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial\left[\left(\nu\Sigma_f\right)^{g}(\mathbf{f})\right]}{\partial n_{m_2}}\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\chi^{g'}\psi^{(1),g'}(r,\boldsymbol{\Omega}') \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial Q^{g}(\mathbf{q};r,\boldsymbol{\Omega})}{\partial n_{m_2}} \\
&-\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\left[\theta_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega})+\theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\varphi^{g}(r,\boldsymbol{\Omega})\right]\frac{\partial \Sigma_t{}^{g}(\mathbf{t})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi^{(1),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\varphi^{g'}(r,\boldsymbol{\Omega}')\frac{\partial^2 \Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_j \partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\theta_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\psi^{(1),g'}(r,\boldsymbol{\Omega}')\frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\varphi^{g'}(r,\boldsymbol{\Omega}')\frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\theta_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial\left[\left(\nu\Sigma_f\right)^{g}(\mathbf{f})\right]}{\partial n_{m_2}}\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\chi^{g'}\psi^{(1),g'}(r,\boldsymbol{\Omega}') \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\,\varphi^{g'}(r,\boldsymbol{\Omega}')\chi^{g}\frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}(\mathbf{f})\right]}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial Q^{g}(\mathbf{q};r,\boldsymbol{\Omega})}{\partial n_{m_2}},\quad for\ j=1,\ldots,J_{\sigma s,l=0}; m_2=1,\ldots,J_n.
\end{aligned}
\tag{93}
$$

In Equation (93), the adjoint functions $\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}$, $j = 1,\ldots,J_{\sigma s,l=0}; g = 1,\ldots,G$ are the solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (30), (32), (36) and (37) of Part II [2], which are reproduced below for convenient reference:

$$B^{g}\left(\boldsymbol{\alpha}^{0}\right)\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = -\delta_{g'_{j}g}N_{i_j,m_j}\varphi^{g}(r,\boldsymbol{\Omega}), j = 1,\ldots,J_{\sigma s,l=0};\ g = 1,\ldots,G, \tag{94}$$

$$\psi_{1,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} < 0; j = 1,\ldots,J_{\sigma s,l=0}; g = 1,\ldots,G, \tag{95}$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^{0}\right)\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) = -\delta_{g'_{j}g}N_{i_j,m_j}\psi^{(1),g}(r,\boldsymbol{\Omega}), j = 1,\ldots,J_{\sigma s,l=0}; g = 1,\ldots,G, \tag{96}$$

$$\psi_{2,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} > 0; j = 1,\ldots,J_{\sigma s,l=0};\ g = 1,\ldots,G. \tag{97}$$

The 2nd-level adjoint functions, $\theta_{1,j}^{(2),g}$ and $\theta_{2,j}^{(2),g}$, $j = 1,\ldots,J_{\sigma s,l=0}; g = 1,\ldots,G$, in Equation (93) are solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (46), (48), (51) and (52) of Part II [2], which are reproduced below for convenient reference:

$$B^{g}\left(\boldsymbol{\alpha}^{0}\right)\theta_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = \delta_{g_j g}N_{i_j,m_j}\left(2l_j+1\right)P_{l_j}(\boldsymbol{\Omega})\phi_{l}^{g'_{j}}(r),\ \ j = 1,\ldots,J_{\sigma s}; g = 1,\ldots,G;\ l = 0,\ldots,ISCT, \tag{98}$$

$$\theta_{1,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} < 0; j = 1,\ldots,J_{\sigma s};\ \ g = 1,\ldots,G, \tag{99}$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^{0}\right)\theta_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) = \delta_{g'_{j}g}N_{i_j,m_j}\left(2l_j+1\right)P_{l_j}(\boldsymbol{\Omega})\xi_{l_j}^{(1),g_j}(r), j = 1,\ldots,J_{\sigma s}; g = 1,\ldots,G;\ l = 0,\ldots,ISCT, \tag{100}$$

$$\theta_{2,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} > 0; j = 1,\ldots,J_{\sigma s}; g = 1,\ldots,G. \tag{101}$$

In Equation (93), the parameters s_j and n_{m_2} correspond to the 0th-order scattering cross sections and the isotopic number densities, denoted as $s_j \equiv \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}$ and $n_{m_2} \equiv N_{i_{m_2},m_{m_2}}$, respectively. Note that:

$$\begin{aligned}\frac{\partial^2 \Sigma_t{}^{g}(\mathbf{t})}{\partial s_j \partial n_{m_2}} &= \frac{\partial^2 \Sigma_t{}^{g}(\mathbf{t})}{\partial \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left\{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I}N_{i,m}\left(\sigma_{f,i}^{g}+\sigma_{c,i}^{g}+\sum\limits_{g'=1}^{G}\sigma_{s,l=0,i}^{g\to g'}\right)\right]/\partial N_{i_{m_2},m_{m_2}}\right\}}{\partial \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}} \\ &= \frac{\partial\left\{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I}\sum\limits_{g'=1}^{G}N_{i,m}\sigma_{s,l=0,i}^{g\to g'}\right]/\partial N_{i_{m_2},m_{m_2}}\right\}}{\partial \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}} = \frac{\partial\left\{\sum\limits_{g'=1}^{G}\sigma_{s,l=0,i_{m_2}}^{g\to g'}\right\}}{\partial \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}} = \delta_{i_j i_{m_2}}\delta_{g'_{j}g},\end{aligned} \tag{102}$$

and

$$\begin{aligned}\frac{\partial^2 \Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial s_j \partial n_{m_2}} &= \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left\{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I}N_{i,m}\sigma_{s,i}^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})\right]/\partial N_{i_{m_2},m_{m_2}}\right\}}{\partial \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}} \\ &= \frac{\partial\left\{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I}\sum\limits_{l=0}^{ISCT}N_{i,m}(2l+1)\sigma_{s,l,i}^{g'\to g}P_l(\boldsymbol{\Omega}'\cdot\boldsymbol{\Omega})\right]/\partial N_{i_{m_2},m_{m_2}}\right\}}{\partial \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}} = \frac{\partial\left\{\sum\limits_{l=0}^{ISCT}(2l+1)\sigma_{s,l,i_{m_2}}^{g'\to g}P_l(\boldsymbol{\Omega}'\cdot\boldsymbol{\Omega})\right\}}{\partial \sigma_{s,l_j=0,i_j}^{g'_{j}\to g_j}} \\ &= \delta_{i_j i_{m_2}}\delta_{g'_{j}g'}\delta_{g_j g}\left(2l_j+1\right)P_{l_j}(\boldsymbol{\Omega}'\cdot\boldsymbol{\Omega}).\end{aligned} \tag{103}$$

Inserting the results obtained in Equations (47)–(52), (102) and (103) into Equation (93) yields the following expression for Equation (93):

$$
\begin{aligned}
&\left(\frac{\partial^2 L}{\partial s_j \partial n_{m_2}}\right)_{(s=\sigma_{s,l=0},n=N)} = -\delta_{i_j i_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega} \psi^{(1),g'_j}(r,\mathbf{\Omega}) \varphi^{g'_j}(r,\mathbf{\Omega}) \\
&- \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \left[\psi_{1,i}^{(2),g}(r,\mathbf{\Omega}) \psi^{(1),g}(r,\mathbf{\Omega}) + \psi_{2,i}^{(2),g}(r,\mathbf{\Omega}) \varphi^{g}(r,\mathbf{\Omega}) \right] \sigma_{t,i_{m_2}}^{g} \\
&+ \sum_{g=1}^{G} \sum_{l=0}^{ISCT} (2l+1) \int_V dV \xi_{1,j;l}^{(2),g}(r) \sum_{g'=1}^{G} \sigma_{s,l,i_{m_2}}^{g \to g'} \xi_{l}^{(1),g'}(r) + \sum_{g=1}^{G} \sum_{l=0}^{ISCT} (2l+1) \int_V dV \xi_{2,j;l}^{(2),g}(r) \sum_{g'=1}^{G} \sigma_{s,l,i_{m_2}}^{g' \to g} \varphi_{l}^{g'}(r) \\
&+ \sum_{g=1}^{G} \int_V dV \chi^{g} \xi_{2,j;0}^{(2),g}(r) \sum_{g'=1}^{G} \nu_{i_{m_2}}^{g'} \sigma_{f,i_{m_2}}^{g'} \varphi_{0}^{g'}(r) + \sum_{g=1}^{G} \int_V dV \nu_{i_{m_2}}^{g} \sigma_{f,i_{m_2}}^{g} \xi_{1,j;0}^{(2),g}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_{0}^{(1),g'}(r) \\
&+ \frac{1}{n_{m_2}} \sum_{g=1}^{G} \int_V dV \xi_{2,j;0}^{(2),g}(r) Q_{SF,i_{m_2}}^{g} - \sum_{g=1}^{G} \int_V dV \sigma_{t,i_{m_2}}^{g} \int_{4\pi} d\mathbf{\Omega} \left[\theta_{1,j}^{(2),g}(r,\mathbf{\Omega}) \psi^{(1),g}(r,\mathbf{\Omega}) + \theta_{2,j}^{(2),g}(r,\mathbf{\Omega}) \varphi^{g}(r,\mathbf{\Omega}) \right] \\
&+ \delta_{i_j i_{m_2}} \left(2l_j + 1\right) \int_V dV \xi_{l_j}^{(1),g_j}(r) \varphi_{l_j}^{g'_j}(r) + \sum_{g=1}^{G} \sum_{l=0}^{ISCT} (2l+1) \int_V dV \Theta_{1,j;l}^{(2),g}(r) \sum_{g'=1}^{G} \sigma_{s,l,i_{m_2}}^{g \to g'} \xi_{l}^{(1),g'}(r) \\
&+ \sum_{g=1}^{G} \sum_{l=0}^{ISCT} (2l+1) \int_V dV \Theta_{2,j;l}^{(2),g}(r) \sum_{g'=1}^{G} \sigma_{s,l,i_{m_2}}^{g' \to g} \varphi_{l}^{g'}(r) + \sum_{g=1}^{G} \int_V dV \nu_{i_{m_2}}^{g} \sigma_{f,i_{m_2}}^{g} \Theta_{1,j;0}^{(2),g}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_{0}^{(1),g'}(r) \\
&+ \sum_{g=1}^{G} \int_V dV \chi^{g} \Theta_{2,j;0}^{(2),g}(r) \sum_{g'=1}^{G} \nu_{i_{m_2}}^{g'} \sigma_{f,i_{m_2}}^{g'} \varphi_{0}^{g'}(r) + \frac{1}{n_{m_2}} \sum_{g=1}^{G} \int_V dV \Theta_{2,j;0}^{(2),g}(r) Q_{SF,i_{m_2}}^{g}, \\
&\mathit{for}\ j = 1, \ldots, J_{\sigma s,l=0}; m_2 = 1, \ldots, J_n.
\end{aligned}
\tag{104}
$$

3.2.2. Second-Order Sensitivities $\left(\partial^2 L / \partial s_j \partial n_{m_2}\right)_{(s=\sigma_{s,l\geq 1},n=N)}, j = 1, \ldots, \sigma_{s,l\geq 1}; m_2 = 1, \ldots, J_n$

For this case, the parameters s_j correspond to the l^{th}-order ($l \geq 1$) scattering cross sections, denoted as $s_j \equiv \sigma_{s,l_j,i_j}^{g_j' \to g_j}$, and the parameters n_{m_2} correspond to the isotopic number densities, denoted as $n_{m_2} \equiv N_{i_{m_2},m_{m_2}}$. Since the l^{th}-order ($l \geq 1$) scattering cross sections are not part of the total cross sections, the expression of $\left(\partial^2 L / \partial s_j \partial n_{m_2}\right)_{(s=\sigma_{s,l\geq 1},n=N)}$ is obtained by particularizing Equations (167), (168), (169) and (171) in [5] to the PERP benchmark, which yields,

$$
\begin{aligned}
&\left(\frac{\partial^2 L}{\partial s_j \partial n_{m_2}}\right)_{(s=\sigma_{s,l\geq 1},n=N)} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \left[\theta_{1,j}^{(2),g}(r,\mathbf{\Omega}) \psi^{(1),g}(r,\mathbf{\Omega}) + \theta_{2,j}^{(2),g}(r,\mathbf{\Omega}) \varphi^{g}(r,\mathbf{\Omega}) \right] \frac{\partial \Sigma_t{}^{g}(\mathbf{t})}{\partial n_{m_2}} \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \psi^{(1),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r,\mathbf{\Omega}') \frac{\partial^2 \Sigma_s^{g' \to g}(\mathbf{s};\mathbf{\Omega}' \to \mathbf{\Omega})}{\partial s_j \partial n_{m_2}} \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{1,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \psi^{(1),g'}(r,\mathbf{\Omega}') \frac{\partial \Sigma_s^{g \to g'}(\mathbf{s};\mathbf{\Omega} \to \mathbf{\Omega}')}{\partial n_{m_2}} \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{2,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r,\mathbf{\Omega}') \frac{\partial \Sigma_s^{g' \to g}(\mathbf{s};\mathbf{\Omega}' \to \mathbf{\Omega})}{\partial n_{m_2}} \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{1,j}^{(2),g}(r,\mathbf{\Omega}) \frac{\partial \left[(\nu \Sigma_f)^{g}(\mathbf{f}) \right]}{\partial n_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \chi^{g'} \psi^{(1),g'}(r,\mathbf{\Omega}') \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{2,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r,\mathbf{\Omega}') \chi^{g} \frac{\partial \left[(\nu \Sigma_f)^{g'}(\mathbf{f}) \right]}{\partial n_{m_2}} \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{2,j}^{(2),g}(r,\mathbf{\Omega}) \frac{\partial Q^{g}(\mathbf{q};r,\mathbf{\Omega})}{\partial n_{m_2}}, \quad \mathit{for}\ j = 1, \ldots, J_{s,l\geq 1}; m_2 = 1, \ldots, J_n,
\end{aligned}
\tag{105}
$$

where the 2nd-level adjoint functions, $\theta_{1,j}^{(2),g}$ and $\theta_{2,j}^{(2),g}$, $j = 1, \ldots, J_{s,l\geq 1}; g = 1, \ldots, G$, are the solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (46), (48), (51) and (52) of Part II [2], which have been reproduced, for convenience, in Equations (98)–(101). Inserting the results obtained in Equations (47)–(52) and (103) into Equation (105), using the addition theorem for spherical harmonics in one-dimensional geometry and performing the respective angular integrations yields the following expression:

$$\left(\frac{\partial^2 L}{\partial s_j \partial n_{m_2}}\right)_{(s=\sigma_{s,l\geq1},n=N)} = -\sum_{g=1}^{G}\int_V dV\sigma^g_{t,i_{m_2}}\int_{4\pi} d\mathbf{\Omega}\left[\theta^{(2),g}_{1,j}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega})+\theta^{(2),g}_{2,j}(r,\mathbf{\Omega})\varphi^g(r,\mathbf{\Omega})\right]$$
$$+\delta_{i_j i_{m_2}}\left(2l_j+1\right)\int_V dV\xi^{(1),g_j}_{l_j}(r)\varphi^{g'_j}_{l_j}(r)+\sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV\Theta^{(2),g}_{1,j;l}(r)\sum_{g'=1}^{G}\sigma^{g\to g'}_{s,l,i_{m_2}}\xi^{(1),g'}_l(r)$$
$$+\sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV\Theta^{(2),g}_{2,j;l}(r)\sum_{g'=1}^{G}\sigma^{g'\to g}_{s,l,i_{m_2}}\varphi^{g'}_l(r)+\sum_{g=1}^{G}\int_V dV\nu^g_{i_{m_2}}\sigma^g_{f,i_{m_2}}\Theta^{(2),g}_{1,j;0}(r)\sum_{g'=1}^{G}\chi^{g'}\xi^{(1),g'}_0(r) \tag{106}$$
$$+\sum_{g=1}^{G}\int_V dV\chi^g\Theta^{(2),g}_{2,j;0}(r)\sum_{g'=1}^{G}\nu^{g'}_{i_{m_2}}\sigma^{g'}_{f,i_{m_2}}\varphi^{g'}_0(r)+\frac{1}{n_{m_2}}\sum_{g=1}^{G}\int_V dV\Theta^{(2),g}_{2,j;0}(r)Q^g_{SF,i_{m_2}},$$
$$for\ j=1,\ldots,J_{s,l\geq1};m_2=1,\ldots,J_n.$$

3.3. Numerical Results for $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_s$

The second-order absolute sensitivities, $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_s$, of the leakage response with respect to the isotopic number densities and the scattering cross sections for all isotopes of the PERP benchmark have been computed using Equations (85) and (87), and have been independently verified by computing the symmetric expression $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_s\partial\mathbf{N}$ using Equations (104) and (106). For the PERP benchmark, computing the second-order absolute sensitivities, $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_s$, using Equations (85) and (87) requires 16 forward and adjoint PARTISN [9] computations to obtain all the required adjoint functions. On the other hand, computing the alternative expression $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_s\partial\mathbf{N}$ using Equations (104) and (106), requires 7101 forward and adjoint PARTISN [9] computations to obtain the needed second level adjoint functions. As have been discussed in Part III [3], the reason for needing "only" 7101, rather than 21,600, PARTISN [9] computations is that all of the up-scattering and some of the down-scattering cross sections are zero for the PERP benchmark. Thus, computing $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_s$ using Equations (85) and (87) is about 450 (≈7101/16) times more efficient than computing $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_s\partial\mathbf{N}$ by using Equations (104) and (106).

The dimensions of the matrix $\partial^2 L/\partial n_j\partial s_{m_2}$, $j=1,\ldots,J_n;m_2=1,\ldots,J_{\sigma s}$ is $J_n\times J_{\sigma s}(=6\times21600)$, where $J_{\sigma s}=G\times G\times(ISCT+1)\times I=30\times30\times4\times6=21600$. The matrix of 2nd-order relative sensitivities corresponding to $\partial^2 L/\partial n_j\partial s_{m_2}$, $j=1,\ldots,J_n;m_2=1,\ldots,J_{\sigma s}$, denoted as $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l,k}\right)$, is defined as follows:

$$\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l,k}\right)\triangleq\frac{\partial^2 L}{\partial N_{i,m}\partial\sigma^{g'\to g}_{s,l,k}}\left(\frac{N_{i,m}\sigma^{g'\to g}_{s,l,k}}{L}\right),\ l=0,\ldots,3;\ \ i,k=1,\ldots,6;m=1,2;\ \ g',g=1,\ldots,30. \tag{107}$$

To facilitate the presentation and interpretation of the numerical results, the $J_n\times J_{\sigma s}(=6\times21600)$ matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l,k}\right)$ has first been partitioned into 4 submatrices, namely, $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=0,k}\right)$, $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=1,k}\right)$, $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=2,k}\right)$ and $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=3,k}\right)$, corresponding to the scattering orders $l=0$, $l=1$, $l=2$, and $l=3$, respectively. Subsequently, each of the 4 submatrices is further partitioned into $I\times I=6\times6$ smaller submatrices, each of dimensions $1\times(G\cdot G)=1\times900$. The results are summarized in Sections 3.3.1–3.3.4, below.

3.3.1. Results for the Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=0,k}\right)$

Table 12 presents the summary of the results for the components of the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=0,k}\right)\triangleq\left(\partial^2 L/\partial N_{i,m}\partial\sigma^{g'\to g}_{s,l=0,k}\right)\left(N_{i,m}\sigma^{g'\to g}_{s,l=0,k}/L\right)$, $i,k=1,\ldots,6;m=1,2;\ \ g',g=1,\ldots,30$, for the 2nd-order relative sensitivities of the leakage response with respect to the isotopic number densities and the 0th-order scattering cross sections for all isotopes. Among the $J_n\times J_{\sigma s,l=0}=6\times5400=32400$ elements in the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=0,k}\right)$, 8844 elements have positive values and 2142 elements have negative values, while the remaining elements are zero. Most of these relative sensitivities are very small. However, 15 elements in the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=0,k}\right)$ have relative sensitivities with absolute values greater than 1.0, as shown in the shaded cells in Table 12. These 15 large values reside in the sub-matrices $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,1}^{g'\to g}\right)$ and $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,6}^{g'\to g}\right)$. The value of the largest element of each of the other sub-matrices is positive, involving the 0th-order self-scattering cross sections for the 12th energy group of isotopes ^{239}Pu, ^{240}Pu, ^{69}Ga, ^{71}G, and C, or (occasionally) the 0th-order out-scattering cross section $\sigma_{s,l=0,k=6}^{16\to 17}$ for isotope ^{1}H. The overall largest value in the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=0,k}^{g'\to g}\right)$ is $S^{(2)}\left(N_{1,1}, \sigma_{s,l=0,k=1}^{12\to 12}\right) = 1.912$.

Table 12. Summary presentation of the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=0,k}^{g'\to g}\right)$, for 2nd-order relative sensitivities of the leakage response with respect to the isotopic number densities and the 0th-order ($l = 0$) scattering cross sections for all isotopes.

	k = 1 (^{239}Pu)	**k = 2 (^{240}Pu)**	**k = 3 (^{69}Ga)**	**k = 4 (^{71}Ga)**	**k = 5 (C)**	**k = 6 (^{1}H)**
$i = 1$ (^{239}Pu)	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,1}^{g'\to g}\right)$ 8 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,2}^{g'\to g}\right)$ Max. value = 1.18 × 10^{-1} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,3}^{g'\to g}\right)$ Max. value = 6.80 × 10^{-3} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,4}^{g'\to g}\right)$ Max. value = 4.36 × 10^{-3} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,5}^{g'\to g}\right)$ Max. value = 8.70 × 10^{-1} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,6}^{g'\to g}\right)$ 7 elements with absolute values >1.0
$i = 2$ (^{240}Pu)	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=0,1}^{g'\to g}\right)$ Max. value = 2.02 × 10^{-1} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=0,2}^{g'\to g}\right)$ Max. value = 2.23 × 10^{-2} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=0,3}^{g'\to g}\right)$ Max. value = 7.74 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=0,4}^{g'\to g}\right)$ Max. value = 4.96 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=0,5}^{g'\to g}\right)$ Max. value = 9.89 × 10^{-2} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=0,6}^{g'\to g}\right)$ Max. value = 1.78 × 10^{-1} g′ = 16, g = 17
$i = 3$ (^{69}Ga)	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=0,1}^{g'\to g}\right)$ Max. value = 6.82 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=0,2}^{g'\to g}\right)$ Max. value = 4.52 × 10^{-5} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=0,3}^{g'\to g}\right)$ Max. value = 5.17 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=0,4}^{g'\to g}\right)$ Max. value = 1.67 × 10^{-6} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=0,5}^{g'\to g}\right)$ Max. value = 3.01 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=0,6}^{g'\to g}\right)$ Max. value = 5.47 × 10^{-4} g′ = 16, g = 17
$i = 4$ (^{71}Ga)	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=0,1}^{g'\to g}\right)$ Max. value = 4.25 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=0,2}^{g'\to g}\right)$ Max. value = 2.82 × 10^{-5} g′ = 13, g = 13	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=0,3}^{g'\to g}\right)$ Max. value = 1.62 × 10^{-6} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=0,4}^{g'\to g}\right)$ Max. value = 3.31 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=0,5}^{g'\to g}\right)$ Max. value = 1.77 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=0,6}^{g'\to g}\right)$ Max. value = 2.93 × 10^{-4} g′ = 16, g = 17
$i = 5$ (C)	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=0,1}^{g'\to g}\right)$ Max. value = 1.70 × 10^{-1} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=0,2}^{g'\to g}\right)$ Max. value = 1.13 × 10^{-2} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=0,3}^{g'\to g}\right)$ Max. value = 6.52 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=0,4}^{g'\to g}\right)$ Max. value = 4.19 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=0,5}^{g'\to g}\right)$ Max. value = 1.59 × 10^{-1} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=0,6}^{g'\to g}\right)$ Max. value = 1.55 × 10^{-1} g′ = 16, g = 17
$i = 6$ (^{1}H)	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=0,1}^{g'\to g}\right)$ Max. value = 2.72 × 10^{-1} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=0,2}^{g'\to g}\right)$ Max. value = 1.80 × 10^{-2} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=0,3}^{g'\to g}\right)$ Max. value = 1.04 × 10^{-3} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=0,4}^{g'\to g}\right)$ Max. value = 6.68 × 10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=0,5}^{g'\to g}\right)$ Max. value = 1.48 × 10^{-1} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=0,6}^{g'\to g}\right)$ Max. value = 3.81 × 10^{-1} g′ = 16, g = 17

Additional information regarding the two submatrices in Table 12 that have elements with absolute values greater than 1.0 is as follows:

1. The submatrix $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,1}^{g' \to g}\right), g', g = 1, \ldots, 30$, comprises the 2nd-order sensitivities of the leakage response with respect to the isotopic number density and to the 0th-order scattering cross sections of ^{239}Pu. Table 13 presents the 8 relative sensitivities in this submatrix that have values greater than 1.0. All of these sensitivities involve the 0th-order self-scattering cross sections for energy groups $g = 7, \ldots, 14$ of isotope ^{239}Pu. The largest value in this submatrix is $S^{(2)}\left(N_{1,1}, \sigma_{s,l=0,k=1}^{12 \to 12}\right) = 1.912$, which involves the 0th-order self-scattering cross sections for the 12th energy group of ^{239}Pu.
2. The submatrix $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,k=6}^{g' \to g}\right), g', g = 1, \ldots, 30$, comprising the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of ^{239}Pu and to the 0th-order scattering cross sections of ^{1}H, includes 7 elements that have values greater than 1.0, as listed in Table 14. Most of these 7 relative sensitivities are with respect to the 0th-order in-scattering or out-scattering cross sections. The largest value in this submatrix is $S^{(2)}\left(N_{1,1}, \sigma_{s,l=0,k=1}^{16 \to 17}\right) = 1.585$, involving the 0th-order out-scattering cross sections for energy groups $g' = 16 \to g = 17$ of isotope ^{239}Pu.

Table 13. Elements of $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,k=1}^{g' \to g}\right), g', g = 1, \ldots, 30$ with absolute values greater than 1.0.

Group	$g' \to g$ 7→7	$g' \to g$ 8→8	$g' \to g$ 9→9	$g' \to g$ 10→10	$g' \to g$ 11→11	$g' \to g$ 12→12	$g' \to g$ 13→13	$g' \to g$ 14→14
values	1.461	1.155	1.206	1.147	1.036	1.912	1.660	1.235

Table 14. Elements of $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,k=6}^{g' \to g}\right), g', g = 1, \ldots, 30$ with absolute values greater than 1.0.

Group	$g' \to g$ 12→13	$g' \to g$ 13→14	$g' \to g$ 14→15	$g' \to g$ 14→16	$g' \to g$ 15→16	$g' \to g$ 16→16	$g' \to g$ 16→17
values	1.386	1.300	1.110	1.146	1.430	1.289	1.585

3.3.2. Results for the Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=1,k}^{g' \to g}\right)$

Table 15 summarizes the results obtained for the elements of the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=1,k}^{g' \to g}\right) \triangleq \left(\partial^2 L / \partial N_{i,m} \partial \sigma_{s,l=1,k}^{g' \to g}\right)\left(N_{i,m} \sigma_{s,l=1,k}^{g' \to g} / L\right), i, k = 1, \ldots, 6; m = 1, 2;\ g', g = 1, \ldots, 30$, which comprises the 2nd-order mixed relative sensitivities of the leakage response with respect to the isotopic number densities and the 1st-order scattering cross sections for all isotopes. Most of these 2nd-order mixed sensitivities are zero, and the non-zero ones are mostly negative. Specifically, the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=1,k}^{g' \to g}\right)$, having dimensions $J_n \times J_{\sigma s,l=1} = 6 \times 5400 = 32400$, comprises 7772 elements with negative values, 2764 elements with positive values, while the remaining elements are zero. Most of the relative 2nd-order mixed relative sensitivities are very small. Only 8 components have large relative sensitivities with absolute values greater than 1.0, as shown in the shaded sub-matrices $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,1}^{g' \to g}\right)$ and $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,6}^{g' \to g}\right)$ in Table 15. Also, in the submatrices which have all their elements with absolute values less than 1.0, the value of the largest element of the respective submatrix is negative and involves the 1st-order self-scattering cross sections for the 7th energy group of isotopes ^{239}Pu, ^{240}Pu, ^{69}Ga and ^{71}Ga, or the 12th energy group of isotope C, or the 1st-order out-scattering cross section $\sigma_{s,l=1,k=6}^{12 \to 13}$ of isotope ^{1}H. The overall most negative element in the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=1,k}^{g' \to g}\right)$ is $S^{(2)}\left(N_{1,1}, \sigma_{s,l=1,k=6}^{12 \to 12}\right) = -1.386$.

Table 15. Summary presentation of the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=1,k}^{g'\to g}\right)$, comprising the 2nd-order relative sensitivities of the leakage response with respect to the isotopic number densities and the 1st-order $(l = 1)$ scattering cross sections for all isotopes.

	k **= 1 (^{239}Pu)**	***k*** **= 2 (^{240}Pu)**	***k*** **= 3 (^{69}Ga)**	***k*** **= 4 (^{71}Ga)**	***k*** **= 5 (C)**	***k*** **= 6 (^{1}H)**
$i = 1$ (^{239}Pu)	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,1}^{g'\to g}\right)$ element with absolute value >1.0	1$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,2}^{g'\to g}\right)$ Min. value = -6.96×10^{-2} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,3}^{g'\to g}\right)$ Min. value = -2.34×10^{-3} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,4}^{g'\to g}\right)$ Min. value = -1.42×10^{-3} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,5}^{g'\to g}\right)$ Min. value = -3.51×10^{-1} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,6}^{g'\to g}\right)$ 6 elements with absolute values >1.0
$i = 2$ (^{240}Pu)	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=1,1}^{g'\to g}\right)$ Min. value = -1.30×10^{-1} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=1,2}^{g'\to g}\right)$ Min. value = -1.32×10^{-2} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=1,3}^{g'\to g}\right)$ Min. value = -2.65×10^{-4} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=1,4}^{g'\to g}\right)$ Min. value = -1.61×10^{-4} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=1,5}^{g'\to g}\right)$ Min. value = -4.00×10^{-2} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=1,6}^{g'\to g}\right)$ Min. value = -1.50×10^{-1} g′ = 12, g = 13
$i = 3$ (^{69}Ga)	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=1,1}^{g'\to g}\right)$ Min. value = -3.42×10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=1,2}^{g'\to g}\right)$ Min. value = -2.14×10^{-5} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=1,3}^{g'\to g}\right)$ Min. value = -1.77×10^{-4} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=1,4}^{g'\to g}\right)$ Min. value = -4.06×10^{-7} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=1,5}^{g'\to g}\right)$ Min. value = -1.22×10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=1,6}^{g'\to g}\right)$ Min. value = -4.56×10^{-4} g′ = 12, g = 13
$i = 4$ (^{71}Ga)	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=1,1}^{g'\to g}\right)$ Min. value = -2.13×10^{-4} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=1,2}^{g'\to g}\right)$ Min. value = -1.33×10^{-5} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=1,3}^{g'\to g}\right)$ Min. value = -4.40×10^{-7} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=1,4}^{g'\to g}\right)$ Min. value = -1.07×10^{-4} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=1,5}^{g'\to g}\right)$ Min. value = -7.23×10^{-5} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=1,6}^{g'\to g}\right)$ Min. value = -2.68×10^{-4} g′ = 12, g = 13
$i = 5$ (C)	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=1,1}^{g'\to g}\right)$ Min. value = -1.12×10^{-1} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=1,2}^{g'\to g}\right)$ Min. value = -6.76×10^{-3} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=1,3}^{g'\to g}\right)$ Min. value = -2.27×10^{-4} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=1,4}^{g'\to g}\right)$ Min. value = -1.38×10^{-4} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=1,5}^{g'\to g}\right)$ Min. value = -6.24×10^{-2} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=1,6}^{g'\to g}\right)$ Min. value = -1.31×10^{-1} g′ = 12, g = 13
$i = 6$ (^{1}H)	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=1,1}^{g'\to g}\right)$ Min. value = -1.77×10^{-1} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=1,2}^{g'\to g}\right)$ Min. value = -1.06×10^{-2} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=1,3}^{g'\to g}\right)$ Min. value = -3.57×10^{-4} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=1,4}^{g'\to g}\right)$ Min. value = -2.17×10^{-4} g′ = 7, g = 7	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=1,5}^{g'\to g}\right)$ Min. value = -5.73×10^{-2} g′ = 12, g = 12	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=1,6}^{g'\to g}\right)$ Min. value = -3.19×10^{-1} g′ = 12, g = 13

Detailed information regarding the two submatrices in Table 15 comprising elements having absolute values greater than 1.0 is as follows:

1. The sensitivity matrix $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,1}^{g'\to g}\right)$, $g', g = 1, \ldots, 30$, comprising the 2nd-order mixed sensitivities of the leakage response with respect to the isotopic number density and the 1st-order scattering cross sections of ^{239}Pu, includes only one element, namely $S^{(2)}\left(N_{1,1}, \sigma_{s,l=1,1}^{7\to 7}\right) = -1.245$,

which has an absolute value greater than 1.0. This element involves the 1st-order self-scattering cross section for the 7th energy group of ^{239}Pu.

2. The sensitivity matrix $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,6}^{g'\to g}\right)$, $g', g = 1, \ldots, 30$, comprising the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of ^{239}Pu and to the 1st-order scattering cross sections of ^{1}H, includes 6 elements that have values greater than 1.0, as listed in Table 16. These 6 large relative sensitivities involve the 1st-order self-scattering or out-scattering cross sections for energy groups $g', g = 12, \ldots, 16$ of isotope ^{1}H, respectively.

Table 16. Elements of $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,k=6}^{g'\to g}\right)$, $g', g = 1, \ldots, 30$ with absolute values greater than 1.0.

Groups	$g' \to g$ 12→12	$g' \to g$ 12→13	$g' \to g$ 13→13	$g' \to g$ 13→14	$g' \to g$ 15→16	$g' \to g$ 16→16
values	−1.103	−1.327	−1.014	−1.162	−1.027	−1.210

3.3.3. Results for the Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=2,k}^{g'\to g}\right)$

The sensitivity results in the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=2,k}^{g'\to g}\right) \triangleq \left(\partial^2 L / \partial N_{i,m} \partial \sigma_{s,l=2,k}^{g'\to g}\right)\left(N_{i,m} \sigma_{s,l=2,k}^{g'\to g} / L\right)$, $i, k = 1, \ldots, 6; m = 1, 2;\ \ g', g = 1, \ldots, 30$, comprising the 2nd-order mixed relative sensitivities of the leakage response with respect to the isotopic number densities and the 2nd-order scattering cross sections for all isotopes, are summarized in Table 17. All of the values in this matrix are smaller than 1.0. This is expected for the 2nd-order sensitivities with respect to higher order scattering cross sections. Of the $J_n \times J_{\sigma s,l=2} = 6 \times 5400 = 32400$ components of $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=1,k}^{g'\to g}\right)$, 6164 elements are positive, 4426 elements are negative, and the remaining elements are zero. As shown in Table 17, most of the largest absolute values in respective submatrices involve either the 2nd-order self-scattering cross sections for the 7th energy group of isotopes ^{239}Pu, ^{240}Pu, ^{69}Ga and ^{71}G and C, or the 12th energy group of ^{1}H. Also, as shown in Table 17, the largest elements in the respective sub-matrix are all positive, and the vast majority of them are very small. The overall largest element in the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=2,k}^{g'\to g}\right)$ is $S^{(2)}\left(N_{1,1}, \sigma_{s,l=2,k=6}^{12\to 12}\right) = 3.50 \times 10^{-1}$.

Table 17. Summary presentation of the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=2,k}\right)$, for 2nd-order relative sensitivities of the leakage response with respect to the isotopic number densities and the 2nd-order $(l=2)$ scattering cross sections for all isotopes.

	k=1 (^{239}Pu)	**k=2 (^{240}Pu)**	**k=3 (^{69}Ga)**	**k=4 (^{71}Pu)**	**k=5 (C)**	**k=6 (^{1}H)**
$i=1$ (^{239}Pu)	$\mathbf{S}^{(2)}\left(N_{1,1},\sigma^{g'\to g}_{s,l=2,1}\right)$ Max. value $= 7.13\times10^{-2}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{1,1},\sigma^{g'\to g}_{s,l=2,2}\right)$ Max. value $= 4.07\times10^{-3}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{1,1},\sigma^{g'\to g}_{s,l=2,3}\right)$ Max. value $= 1.22\times10^{-4}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{1,1},\sigma^{g'\to g}_{s,l=2,4}\right)$ Max. value $= 7.57\times10^{-5}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{1,1},\sigma^{g'\to g}_{s,l=2,5}\right)$ Max. value $= 9.48\times10^{-2}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{1,1},\sigma^{g'\to g}_{s,l=2,6}\right)$ Max. value $= 3.50\times10^{-1}$ $g'=12, g=12$
$i=2$ (^{240}Pu)	$\mathbf{S}^{(2)}\left(N_{2,1},\sigma^{g'\to g}_{s,l=2,1}\right)$ Max. value $= 7.43\times10^{-3}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{2,1},\sigma^{g'\to g}_{s,l=2,2}\right)$ Max. value $= 7.52\times10^{-4}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{2,1},\sigma^{g'\to g}_{s,l=2,3}\right)$ Max. value $= 1.36\times10^{-5}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{2,1},\sigma^{g'\to g}_{s,l=2,4}\right)$ Max. value $= 8.46\times10^{-6}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{2,1},\sigma^{g'\to g}_{s,l=2,5}\right)$ Max. value $= 1.08\times10^{-2}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{2,1},\sigma^{g'\to g}_{s,l=2,6}\right)$ Max. value $= 4.02\times10^{-2}$ $g'=12, g=12$
$i=3$ (^{69}Ga)	$\mathbf{S}^{(2)}\left(N_{3,1},\sigma^{g'\to g}_{s,l=2,1}\right)$ Max. value $= 1.34\times10^{-5}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{3,1},\sigma^{g'\to g}_{s,l=2,2}\right)$ Max. value $= 8.17\times10^{-7}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{3,1},\sigma^{g'\to g}_{s,l=2,3}\right)$ Max. value $= 8.93\times10^{-6}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{3,1},\sigma^{g'\to g}_{s,l=2,4}\right)$ Max. value $= 1.52\times10^{-8}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{3,1},\sigma^{g'\to g}_{s,l=2,5}\right)$ Max. value $= 2.60\times10^{-5}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{3,1},\sigma^{g'\to g}_{s,l=2,6}\right)$ Max. value $= 1.24\times10^{-4}$ $g'=12, g=12$
$i=4$ (^{71}Ga)	$\mathbf{S}^{(2)}\left(N_{4,1},\sigma^{g'\to g}_{s,l=2,1}\right)$ Max. value $= 7.50\times10^{-6}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{4,1},\sigma^{g'\to g}_{s,l=2,2}\right)$ Max. value $= 4.59\times10^{-7}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{4,1},\sigma^{g'\to g}_{s,l=2,3}\right)$ Max. value $= 1.38\times10^{-8}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{4,1},\sigma^{g'\to g}_{s,l=2,4}\right)$ Max. value $= 5.54\times10^{-6}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{4,1},\sigma^{g'\to g}_{s,l=2,5}\right)$ Max. value $= 1.51\times10^{-5}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{4,1},\sigma^{g'\to g}_{s,l=2,6}\right)$ Max. value $= 7.58\times10^{-5}$ $g'=12, g=12$
$i=5$ (C)	$\mathbf{S}^{(2)}\left(N_{5,2},\sigma^{g'\to g}_{s,l=2,1}\right)$ Max. value $= 5.70\times10^{-3}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{5,2},\sigma^{g'\to g}_{s,l=2,2}\right)$ Max. value $= 3.49\times10^{-4}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{5,2},\sigma^{g'\to g}_{s,l=2,3}\right)$ Max. value $= 1.05\times10^{-5}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{5,2},\sigma^{g'\to g}_{s,l=2,4}\right)$ Max. value $= 6.50\times10^{-6}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{5,2},\sigma^{g'\to g}_{s,l=2,5}\right)$ Max. value $= 1.60\times10^{-2}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{5,2},\sigma^{g'\to g}_{s,l=2,6}\right)$ Max. value $= 2.80\times10^{-2}$ $g'=12, g=12$
$i=6$ (^{1}H)	$\mathbf{S}^{(2)}\left(N_{6,2},\sigma^{g'\to g}_{s,l=2,1}\right)$ Max. value $= 7.72\times10^{-3}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{6,2},\sigma^{g'\to g}_{s,l=2,2}\right)$ Max. value $= 4.72\times10^{-4}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{6,2},\sigma^{g'\to g}_{s,l=2,3}\right)$ Max. value $= 1.42\times10^{-5}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{6,2},\sigma^{g'\to g}_{s,l=2,4}\right)$ Max. value $= 8.79\times10^{-6}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{6,2},\sigma^{g'\to g}_{s,l=2,5}\right)$ Max. value $= 1.41\times10^{-2}$ $g'=7, g=7$	$\mathbf{S}^{(2)}\left(N_{6,2},\sigma^{g'\to g}_{s,l=2,6}\right)$ Max. value $= 6.98\times10^{-2}$ $g'=12, g=12$

3.3.4. Results for the Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=3,k}\right)$

Table 18 reports the summary of the results for the 2nd-order mixed relative sensitivities $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=3,k}\right) \triangleq \left(\partial^2 L/\partial N_{i,m}\partial\sigma^{g'\to g}_{s,l=3,k}\right)\left(N_{i,m}\sigma^{g'\to g}_{s,l=3,k}/L\right), i,k = 1,\ldots,6; m = 1,2;\ g',g = 1,\ldots,30$, of the leakage response with respect to the isotopic number densities and the 3rd-order scattering cross sections for all isotopes of the PERP benchmark. Of the $J_n \times J_{\sigma s,l=3} = 6\times5400 = 32400$ components of the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=3,k}\right)$, 5311 elements have negative values and 5183 elements have positive values, while the remaining elements are zero. As in Table 17, most of the largest absolute values in respective submatrices shown in Table 18 involve the 3$^{\text{rd}}$-order self-scattering cross sections for the 7th energy group of isotopes ^{239}Pu, ^{240}Pu, ^{69}Ga and ^{71}G and C, or the 12th energy group of ^{1}H. All of the values presented in Table 18 are very small; the overall largest element in the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^{g'\to g}_{s,l=3,k}\right)$ is $S^{(2)}\left(N_{1,1},\sigma^{12\to12}_{s,l=3,6}\right) = -7.00\times10^{-2}$.

Table 18. Summary presentation of the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{s,l=3,k}^{g'\to g}\right)$, comprising the 2nd-order relative sensitivities of the leakage response with respect to the isotopic number densities and the 3rd-order $(l = 3)$ scattering cross sections for all isotopes.

	$k = 1$ (^{239}Pu)	$k = 2$ (^{240}Pu)	$k = 3$ (^{69}Ga)	$k = 4$ (^{71}Ga)	$k = 5$ (C)	$k = 6$ (^{1}H)
$i = 1$ (^{239}Pu)	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=3,1}^{g'\to g}\right)$ Min. value $= -8.98 \times 10^{-5}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=3,2}^{g'\to g}\right)$ Min. value $= -5.43 \times 10^{-6}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=3,3}^{g'\to g}\right)$ Min. value $= -1.54 \times 10^{-7}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=3,4}^{g'\to g}\right)$ Min. value $= -9.66 \times 10^{-8}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=3,5}^{g'\to g}\right)$ Min. value $= -2.38 \times 10^{-2}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=3,6}^{g'\to g}\right)$ Min. value $= -7.00 \times 10^{-2}$ $g' = 12, g = 12$
$i = 2$ (^{240}Pu)	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=3,1}^{g'\to g}\right)$ Min. value $= -6.38 \times 10^{-6}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=3,2}^{g'\to g}\right)$ Min. value $= -4.90 \times 10^{-7}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=3,3}^{g'\to g}\right)$ Min. value $= -1.11 \times 10^{-8}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=3,4}^{g'\to g}\right)$ Min. value $= -7.62 \times 10^{-9}$ $g' = 6, g=6$	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=3,5}^{g'\to g}\right)$ Min. value $= -2.72 \times 10^{-3}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{2,1}, \sigma_{s,l=3,6}^{g'\to g}\right)$ Min. value $= -8.07 \times 10^{-3}$ $g' = 12, g = 12$
$i = 3$ (^{69}Ga)	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=3,1}^{g'\to g}\right)$ Max. value $= 1.24 \times 10^{-8}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=3,2}^{g'\to g}\right)$ Max. value $= 7.65 \times 10^{-10}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=3,3}^{g'\to g}\right)$ Min. value $= -5.25 \times 10^{-9}$ $g' = 6, g = 6$	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=3,4}^{g'\to g}\right)$ Max. value $= 1.36 \times 10^{-11}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=3,5}^{g'\to g}\right)$ Min. value $= -6.36 \times 10^{-6}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{3,1}, \sigma_{s,l=3,6}^{g'\to g}\right)$ Min. value $= -2.51 \times 10^{-5}$ $g' = 12, g = 12$
$i = 4$ (^{71}Ga)	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=3,1}^{g'\to g}\right)$ Min. value $= -8.45 \times 10^{-9}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=3,2}^{g'\to g}\right)$ Min. value $= -5.20 \times 10^{-10}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=3,3}^{g'\to g}\right)$ Min. value $= -1.48 \times 10^{-11}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=3,4}^{g'\to g}\right)$ Min. value $= -3.61 \times 10^{-9}$ $g' = 6, g = 6$	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=3,5}^{g'\to g}\right)$ Min. value $= -3.69 \times 10^{-6}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{4,1}, \sigma_{s,l=3,6}^{g'\to g}\right)$ Min. value $= -1.58 \times 10^{-5}$ $g' = 12, g = 12$
$i = 5$ (C)	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=3,1}^{g'\to g}\right)$ Max. value $= 6.63 \times 10^{-6}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=3,2}^{g'\to g}\right)$ Max. value $= 4.08 \times 10^{-7}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=3,3}^{g'\to g}\right)$ Max. value $= 1.16 \times 10^{-8}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=3,4}^{g'\to g}\right)$ Max. value $= 7.27 \times 10^{-9}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=3,5}^{g'\to g}\right)$ Min. value $= -3.63 \times 10^{-3}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{s,l=3,6}^{g'\to g}\right)$ Min. value $= -4.15 \times 10^{-3}$ $g' = 12, g = 12$
$i = 6$ (^{1}H)	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=3,1}^{g'\to g}\right)$ Max. value $= 1.41 \times 10^{-5}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=3,2}^{g'\to g}\right)$ Max. value $= 8.65 \times 10^{-7}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=3,3}^{g'\to g}\right)$ Max. value $= 2.46 \times 10^{-8}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=3,4}^{g'\to g}\right)$ Max. value $= 1.54 \times 10^{-8}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=3,5}^{g'\to g}\right)$ Min. value $= -2.73 \times 10^{-3}$ $g' = 7, g = 7$	$\mathbf{S}^{(2)}\left(N_{6,2}, \sigma_{s,l=3,6}^{g'\to g}\right)$ Min. value $= -9.49 \times 10^{-3}$ $g' = 12, g = 12$

4. Mixed Second-Order Sensitivities of the PERP Total Leakage Response with Respect to the Parameters Underlying the Benchmark's Isotopic Number Densities and Fission Cross Sections

This Section presents the computation and analysis of the numerical results for the 2nd-order mixed sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_f$ of the leakage response with respect to the isotopic number densities and group-averaged fission microscopic cross sections of all isotopes of the PERP benchmark. Due to symmetry, these 2nd-order mixed sensitivities can also be computed by using the alternative expression $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_f\partial\mathbf{N}$. These two alternative paths are presented in Sections 4.1 and 4.2, respectively. The numerical results for the 2nd-order mixed sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_f$ have been verified with the results obtained for $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_f\partial\mathbf{N}$, and are presented in Section 4.3.

4.1. Computing the Second-Order Sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N}\partial \boldsymbol{\sigma}_f$

The equations needed for deriving the expressions of the 2nd-order sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N}\partial \boldsymbol{\sigma}_f$ are obtained by particularizing Equations (158), (160), (167), (169), (177), (179), (204) and (206) in [5] to the PERP benchmark. Specifically, using Equation (158) in [5] in conjunction with the relations $\frac{\partial^2 L}{\partial t_j \partial t_{m_2}}\frac{\partial t_j}{\partial n_j}\frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial^2 L}{\partial n_j \partial f_{m_2}}$, $\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}}\frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial f_{m_2}}$ and $\frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial t_j \partial t_{m_2}}\frac{\partial t_j}{\partial n_j}\frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial n_j \partial f_{m_2}}$ yields the following expression:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(1)}_{(n=N,f=\sigma_f)} = -\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi^{(1),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega})\frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial n_j \partial f_{m_2}}\\&-\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\Big[\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega})+\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega})\Big]\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial f_{m_2}},\\&for j=1,\ldots,J_n\,,\ \ m_2=1,\ldots,J_{\sigma f}\,.\end{aligned} \tag{108}$$

The 2nd-level adjoint functions $\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}$, $j=1,\ldots,J_{\sigma f}; g=1,\ldots,G$ in Equation (108) are the solutions of the 2nd-LASS presented in Equations (11)–(14). In Equation (108), the parameters n_j and f_{m_2} correspond to the isotopic number densities and microscopic fission cross sections, respectively, and are denoted as $n_j \equiv N_{i_j,m_j}$ and $f_{m_2} \equiv \sigma_{f,i_{m_2}}^{g_{m_2}}$, where the subscripts i_{m_2} and g_{m_2} refer to the isotope and energy group associated with the parameter f_{m_2}, respectively. Noting that

$$\begin{aligned}\frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial n_j \partial f_{m_2}} &= \frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial N_{i_j,m_j}\partial \sigma_{f,i_{m_2}}^{g_{m_2}}} = \frac{\partial\left\{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I}N_{i,m}\left(\sigma_{f,i}^g+\sigma_{c,i}^g+\sum\limits_{g'=1}^{G}\sigma_{s,l=0,i}^{g\to g'}\right)\right]/\partial N_{i_j,m_j}\right\}}{\partial \sigma_{f,i_{m_2}}^{g_{m_2}}}\\&= \frac{\partial\left\{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I}N_{i,m}\sigma_{f,i}^g\right]/\partial N_{i_j,m_j}\right\}}{\partial \sigma_{f,i_{m_2}}^{g_{m_2}}} = \frac{\partial\left\{\sigma_{f,i_j}^g\right\}}{\partial \sigma_{f,i_{m_2}}^{g_{m_2}}} = \delta_{i_j i_{m_2}}\delta_{g_{m_2}g},\end{aligned} \tag{109}$$

$$\frac{\partial \Sigma_t^g}{\partial f_{m_2}} = \frac{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I}N_{i,m}\sigma_{t,i}^g\right]}{\partial \sigma_{f,i_{m_2}}^{g_{m_2}}} = \delta_{g_{m_2}g}N_{i_{m_2},m_{m_2}}, \tag{110}$$

and inserting the results obtained in Equations (109) and (110) into Equation (108) yields the following expression for Equation (108):

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(1)}_{(n=N,f=\sigma_f)} = -\delta_{i_j i_{m_2}}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega})\varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\\&-N_{i_{m_2},m_{m_2}}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\Big[\psi_{1,i}^{(2),g_{m_2}}(r,\boldsymbol{\Omega})\psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega})+\psi_{2,i}^{(2),g_{m_2}}(r,\boldsymbol{\Omega})\varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\Big],\\&for j=1,\ldots,J_n; m_2=1,\ldots,J_{\sigma f}.\end{aligned} \tag{111}$$

The contributions stemming from Equation (160) in [5] takes on the following particular form:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(2)}_{(n=N,f=\sigma_f)} = \sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\chi^g\frac{\partial\left[(\nu\Sigma_f)^{g'}\right]}{\partial f_{m_2}}\\&+\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\boldsymbol{\Omega}\,\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial\left[(\nu\Sigma_f)^{g}\right]}{\partial f_{m_2}}\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\chi^{g'}\psi^{(1),g'}(r,\boldsymbol{\Omega}'),\\&for\ j=1,\ldots,J_n;\ \ m_2=1,\ldots,J_{\sigma f}\,.\end{aligned} \tag{112}$$

Noting that

$$\frac{\partial\left[\left(\nu\Sigma_f\right)^g\right]}{\partial f_{m_2}} = \frac{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I} N_{i,m}\left(\nu\sigma_f\right)_i^g\right]}{\partial\sigma_{f,i_{m_2}}^{g_{m_2}}} = \frac{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I} N_{i,m}\nu_i^g\sigma_{f,i}^g\right]}{\partial\sigma_{f,i_{m_2}}^{g_{m_2}}} = \delta_{g_{m_2}g}N_{i_{m_2},m_{m_2}}\nu_{i_{m_2}}^g, \tag{113}$$

$$\frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial f_{m_2}} = \frac{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I} N_{i,m}\left(\nu\sigma_f\right)_i^{g'}\right]}{\partial\sigma_{f,i_{m_2}}^{g_{m_2}}} = \frac{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I} N_{i,m}\nu_i^{g'}\sigma_{f,i}^{g'}\right]}{\partial\sigma_{f,i_{m_2}}^{g_{m_2}}} = \delta_{g_{m_2}g'}N_{i_{m_2},m_{m_2}}\nu_{i_{m_2}}^{g'}, \tag{114}$$

and inserting the results obtained in Equations (113) and (114) into Equation (112) yields the following expression for Equation (114):

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j\partial f_{m_2}}\right)^{(2)}_{(n=N,f=\sigma_f)} = N_{i_{m_2},m_{m_2}}\nu_{i_{m_2}}^{g_{m_2}}\int_V dV\left[\xi_{1,j;0}^{(2),g_{m_2}}(r)\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r)\sum_{g=1}^{G}\chi^g\xi_{2,j;0}^{(2),g}(r)\right], \\ for j = 1,\ldots,J_n;\quad m_2 = 1,\ldots,J_{\sigma f}.\end{gathered} \tag{115}$$

Using Equation (167) in [5] in conjunction with the relations $\frac{\partial^2 L}{\partial s_j\partial t_{m_2}}\frac{\partial s_j}{\partial n_j}\frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial^2 L}{\partial n_j\partial f_{m_2}}$ and $\frac{\partial\Sigma_t{}^g}{\partial t_{m_2}}\frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial\Sigma_t{}^g}{\partial f_{m_2}}$ yields the following expression:

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j\partial f_{m_2}}\right)^{(3)}_{(n=N,f=\sigma_f)} = -\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\mathbf{\Omega}\left[\theta_{1,j}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega}) + \theta_{2,j}^{(2),g}(r,\mathbf{\Omega})\varphi^g(r,\mathbf{\Omega})\right]\frac{\partial\Sigma_t{}^g}{\partial f_{m_2}}, \\ for\quad j = 1,\ldots,J_n;\quad m_2 = 1,\ldots,J_{\sigma f},\end{gathered} \tag{116}$$

where the 2nd-level adjoint functions $\theta_{1,j}^{(2),g}$, and $\theta_{2,j}^{(2),g}$, $j = 1,\ldots,J_n; g = 1,\ldots,G$, are the solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (19)–(22). Inserting the results obtained in Equation (110) into Equation (116) yields the following relation:

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j\partial f_{m_2}}\right)^{(3)}_{(n=N,f=\sigma_f)} = -N_{i_{m_2},m_{m_2}}\int_V dV\int_{4\pi} d\mathbf{\Omega}\left[\theta_{1,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\psi^{(1),g_{m_2}}(r,\mathbf{\Omega}) + \theta_{2,j}^{(2),g_{m_2}}(r,\mathbf{\Omega})\varphi^{g_{m_2}}(r,\mathbf{\Omega})\right], \\ for\quad j = 1,\ldots,J_n;\quad m_2 = 1,\ldots,J_{\sigma f}.\end{gathered} \tag{117}$$

The contributions stemming from Equation (169) in [5] to $\partial^2 L/\partial n_j\partial f_{m_2}$ are obtained in the following form:

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j\partial f_{m_2}}\right)^{(4)}_{(n=N,f=\sigma_f)} = \sum_{g=1}^{G}\int_V dV\int_{4\pi} d\mathbf{\Omega}\theta_{1,j}^{(2),g}(r,\mathbf{\Omega})\frac{\partial\left[\left(\nu\Sigma_f\right)^g\right]}{\partial f_{m_2}}\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\chi^{g'}\psi^{(1),g'}(r,\mathbf{\Omega}') \\ + \sum_{g=1}^{G}\int_V dV\int_{4\pi} d\mathbf{\Omega}\theta_{2,j}^{(2),g}(r,\mathbf{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\mathbf{\Omega}'\varphi^{g'}(r,\mathbf{\Omega}')\chi^g\frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial f_{m_2}}, \\ for\ j = 1,\ldots,J_n; m_2 = 1,\ldots,J_{\sigma f}.\end{gathered} \tag{118}$$

Inserting Equations (113) and (114) into Equation (118) yields the following expression for Equation (118):

$$\begin{gathered}\left(\frac{\partial^2 L}{\partial n_j\partial f_{m_2}}\right)^{(4)}_{(n=N,f=\sigma_f)} = N_{i_{m_2},m_{m_2}}\nu_{i_{m_2}}^{g_{m_2}}\int_V dV\left[\Theta_{1,j;0}^{(2),g_{m_2}}(r)\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r)\sum_{g=1}^{G}\chi^g\Theta_{2,j;0}^{(2),g}(r)\right], \\ for j = 1,\ldots,J_n; m_2 = 1,\ldots,J_{\sigma f}.\end{gathered} \tag{119}$$

Further contributions stem from Equation (177) in [5] in conjunction with the relations $\frac{\partial^2 L}{\partial f_j \partial t_{m_2}} \frac{\partial f_j}{\partial n_j} \frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial^2 L}{\partial n_j \partial f_{m_2}}$ and $\frac{\partial \Sigma_t{}^g}{\partial t_{m_2}} \frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial \Sigma_t{}^g}{\partial f_{m_2}}$, as follows:

$$\begin{aligned} \left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}} \right)^{(5)}_{(n=N, f=\sigma_f)} &= -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} \left[u_{1,j}^{(2),g}(r, \boldsymbol{\Omega}) \psi^{(1),g}(r, \boldsymbol{\Omega}) + u_{2,j}^{(2),g}(r, \boldsymbol{\Omega}) \varphi^g(r, \boldsymbol{\Omega}) \right] \frac{\partial \Sigma_t{}^g}{\partial f_{m_2}}, \\ for \quad j &= 1, \ldots, J_n; \;\; m_2 = 1, \ldots, J_{\sigma f}, \end{aligned} \tag{120}$$

where the 2nd-level adjoint functions $u_{1,j}^{(2),g}$, and $u_{2,j}^{(2),g}$, $j = 1, \ldots, J_n; g = 1, \ldots, G$, are the solutions of the 2nd-Level Adjoint Sensitivity System comprising Equations (27)–(30). Replacing the result obtained in Equation (110) into Equation (120) yields the following relation:

$$\begin{aligned} \left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}} \right)^{(5)}_{(n=N, f=\sigma_f)} &= -N_{i_{m_2}, m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} \left[u_{1,j}^{(2),g_{m_2}}(r, \boldsymbol{\Omega}) \psi^{(1),g_{m_2}}(r, \boldsymbol{\Omega}) + u_{2,j}^{(2),g_{m_2}}(r, \boldsymbol{\Omega}) \varphi^{g_{m_2}}(r, \boldsymbol{\Omega}) \right], \\ for \quad j &= 1, \ldots, J_n; \;\; m_2 = 1, \ldots, J_{\sigma f}. \end{aligned} \tag{121}$$

Contributions stemming from Equation (179) in [5] to $\partial^2 L / \partial n_j \partial f_{m_2}$ are given by the following relation:

$$\begin{aligned} \left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}} \right)^{(6)}_{(n=N, f=\sigma_f)} &= \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} \psi^{(1),g}(r, \boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \varphi^{g'}(r, \boldsymbol{\Omega}') \chi^g \frac{\partial^2 \left[(\nu \Sigma_f)^{g'} \right]}{\partial n_j \partial f_{m_2}} \\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} u_{1,j}^{(2),g}(r, \boldsymbol{\Omega}) \frac{\partial \left[(\nu \Sigma_f)^g \right]}{\partial f_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \chi^{g'} \psi^{(1),g'}(r, \boldsymbol{\Omega}') \\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} u_{2,j}^{(2),g}(r, \boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \varphi^{g'}(r, \boldsymbol{\Omega}') \chi^g \frac{\partial \left[(\nu \Sigma_f)^{g'} \right]}{\partial f_{m_2}}, \\ for \quad j &= 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma f}, \end{aligned} \tag{122}$$

where

$$\frac{\partial^2 \left[(\nu \Sigma_f)^{g'} \right]}{\partial n_j \partial f_{m_2}} = \frac{\partial^2 \left[(\nu \Sigma_f)^{g'} \right]}{\partial N_{i_j, m_j} \partial \sigma_{f, i_{m_2}}^{g_{m_2}}} = \frac{\partial \left[\partial \left[\sum_{m=1}^{M} \sum_{i=1}^{I} N_{i,m} \nu_i^{g'} \sigma_{f,i}^{g'} \right] / \partial N_{i_j, m_j} \right]}{\partial \sigma_{f, i_{m_2}}^{g_{m_2}}} = \frac{\partial \left\{ \nu_{i_j}^{g'} \sigma_{f, i_j}^{g'} \right\}}{\partial \sigma_{f, i_{m_2}}^{g_{m_2}}} = \delta_{i_j i_{m_2}} \delta_{g_{m_2} g'} \nu_{i_{m_2}}^{g'}. \tag{123}$$

Inserting the results obtained in Equations (113), (114) and (123) into Equation (122) yields the following expression for Equation (122):

$$\begin{aligned} \left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}} \right)^{(6)}_{(n=N, f=\sigma_f)} &= \delta_{i_j i_{m_2}} \nu_{i_{m_2}}^{g_{m_2}} \int_V dV \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \xi_0^{(1),g}(r) \\ &+ N_{i_{m_2}, m_{m_2}} \nu_{i_{m_2}}^{g_{m_2}} \int_V dV \left[U_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g U_{2,j;0}^{(2),g}(r) \right], \\ for \quad j &= 1, \ldots, J_n; m_2 = 1, \ldots, J_{\sigma f}. \end{aligned} \tag{124}$$

Additional contributions stemming from the sources are computed by particularizing Equations (204) and (206) in [5] to the PERP benchmark. The expression obtained by particularizing Equation (204) in [5], in conjunction with the relations $\frac{\partial^2 L}{\partial q_j \partial t_{m_2}} \frac{\partial q_j}{\partial n_j} \frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial^2 L}{\partial n_j \partial f_{m_2}}$ and $\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial t_{m_2}} \frac{\partial t_{m_2}}{\partial f_{m_2}} = \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial f_{m_2}}$ yields:

$$\begin{aligned} \left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}} \right)^{(7)}_{(n=N, f=\sigma_f)} &= -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} g_{1,j}^{(2),g}(r, \boldsymbol{\Omega}) \psi^{(1),g}(r, \boldsymbol{\Omega}) \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial f_{m_2}}, \\ for \quad j &= 1, \ldots, J_n; \;\; m_2 = 1, \ldots, J_{\sigma f}, \end{aligned} \tag{125}$$

where the 2nd-level adjoint functions $g_{1,j}^{(2),g}$, $j = 1, \ldots, J_n$; $g = 1, \ldots, G$, are the solutions of the 2nd-LASS presented previously in Equations (35) and (36). Inserting the result obtained in Equation (110) into Equation (125) yields the following result:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(7)}_{(n=N,f=\sigma_f)} = -N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} g_{1,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega}),\\ &for \quad j = 1, \ldots, J_n; \; m_2 = 1, \ldots, J_{\sigma f}.\end{aligned} \tag{126}$$

Finally, using Equation (206) in [5] to the PERP benchmark yields the following contributions:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(8)}_{(n=N,f=\sigma_f)} = \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} g_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \frac{\partial\left[(\nu\Sigma_f)^g(\mathbf{f})\right]}{\partial f_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \chi^{g'} \psi^{(1),g'}\left(r,\boldsymbol{\Omega}'\right),\\ &for \quad j = 1, \ldots, J_n; \; m_2 = 1, \ldots, J_{\sigma f}.\end{aligned} \tag{127}$$

Using the result obtained in Equation (113) into Equation (127) yields the following expression for Equation (127):

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(8)}_{(n=N,f=\sigma_f)} = N_{i_{m_2},m_{m_2}} \nu_{i_{m_2}}^{g_{m_2}} \int_V dV G_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r),\\ &for \quad j = 1, \ldots, J_n; \; m_2 = 1, \ldots, J_{\sigma f}.\end{aligned} \tag{128}$$

Collecting the partial contributions obtained in Equations (111), (115), (117), (119), (121), (124), (126) and (128), yields the following result:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)_{(n=N,f=\sigma_f)} = \sum_{i=1}^{8} \left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(i)}_{(n=N,f=\sigma_f)}\\
&= -\delta_{i_j i_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} \psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega}) \varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\\
&-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega}\left[\psi_{1,i}^{(2),g_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega}) + \psi_{2,i}^{(2),g_{m_2}}(r,\boldsymbol{\Omega}) \varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\right]\\
&+N_{i_{m_2},m_{m_2}} \nu_{i_{m_2}}^{g_{m_2}} \int_V dV\left[\xi_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \xi_{2,j;0}^{(2),g}(r)\right]\\
&-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega}\left[\theta_{1,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega}) + \theta_{2,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega}) \varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\right]\\
&+N_{i_{m_2},m_{m_2}} \nu_{i_{m_2}}^{g_{m_2}} \int_V dV\left[\Theta_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \Theta_{2,j;0}^{(2),g}(r)\right]\\
&-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega}\left[u_{1,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega}) + u_{2,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega}) \varphi^{g_{m_2}}(r,\boldsymbol{\Omega})\right]\\
&+\delta_{i_j i_{m_2}} \nu_{i_{m_2}}^{g_{m_2}} \int_V dV \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \xi_0^{(1),g}(r)\\
&+N_{i_{m_2},m_{m_2}} \nu_{i_{m_2}}^{g_{m_2}} \int_V dV\left[U_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g U_{2,j;0}^{(2),g}(r)\right]\\
&-N_{i_{m_2},m_{m_2}} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} g_{1,j}^{(2),g_{m_2}}(r,\boldsymbol{\Omega}) \psi^{(1),g_{m_2}}(r,\boldsymbol{\Omega})\\
&+N_{i_{m_2},m_{m_2}} \nu_{i_{m_2}}^{g_{m_2}} \int_V dV G_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r), \; for \quad j = 1, \ldots, J_n; \; m_2 = 1, \ldots, J_{\sigma f}.\end{aligned} \tag{129}$$

4.2. Alternative Path: Computing the Second-Order Sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial \boldsymbol{\sigma}_f \partial \mathbf{N}$

Due to symmetry of the mixed 2nd-order sensitivities, the results computed using the expression for $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N} \partial \boldsymbol{\sigma}_f$ obtained in Equation (129) can be verified by obtaining and using the expressions for $\partial^2 L(\boldsymbol{\alpha})/\partial \boldsymbol{\sigma}_f \partial \mathbf{N}$. The equations needed for deriving the expression of the 2nd-order mixed sensitivities

$\partial^2 L(\boldsymbol{\alpha})/\partial \boldsymbol{\sigma}_f \partial \mathbf{N}$ are obtained by particularizing Equations (158), (159), (160), (162), (177), (178), (179) and (181) in [5] to the PERP benchmark, which yields the following relation:

$$\begin{aligned}
\left(\frac{\partial^2 L}{\partial f_j \partial n_{m_2}}\right)_{(f=\sigma_f, n=N)} &= -\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi^{(1),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega})\frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial f_j \partial n_{m_2}} \\
&-\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\left[\psi_{1,i}^{(2),g}(r,\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega}) + \psi_{2,i}^{(2),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega})\right]\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\psi^{(1),g'}(r,\boldsymbol{\Omega}')\frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\chi^g\frac{\partial\left[(\nu\Sigma_f)^{g'}(\mathbf{f})\right]}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial\left[(\nu\Sigma_f)^{g}(\mathbf{f})\right]}{\partial n_{m_2}}\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\chi^{g'}\psi^{(1),g'}(r,\boldsymbol{\Omega}') \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial Q^g(\mathbf{q};r,\boldsymbol{\Omega})}{\partial n_{m_2}} \\
&-\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\left[u_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\psi^{(1),g}(r,\boldsymbol{\Omega}) + u_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\varphi^g(r,\boldsymbol{\Omega})\right]\frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}u_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\psi^{(1),g'}(r,\boldsymbol{\Omega}')\frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}u_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi^{(1),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\chi^g\frac{\partial^2\left[(\nu\Sigma_f)^{g'}\right]}{\partial f_j \partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}u_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial\left[(\nu\Sigma_f)^{g}\right]}{\partial n_{m_2}}\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\chi^{g'}\psi^{(1),g'}(r,\boldsymbol{\Omega}') \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}u_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\chi^g\frac{\partial\left[(\nu\Sigma_f)^{g'}\right]}{\partial n_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}u_{2,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial Q^g(\mathbf{q};r,\boldsymbol{\Omega})}{\partial n_{m_2}},\ \ for\ j=1,\ldots,J_{\sigma f}; m_2=1,\ldots,J_n.
\end{aligned} \tag{130}$$

In Equation (130), the adjoint functions $\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}, j = 1,\ldots,J_{\sigma f}; g = 1,\ldots,G$ are the solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (33), (35), (39) and (40) of Part III [3], which are reproduced below for convenient reference:

$$B^g\left(\boldsymbol{\alpha}^0\right)\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = -\delta_{g_j g}N_{i_j,m_j}\varphi^g(r,\boldsymbol{\Omega}), j=1,\ldots,J_{\sigma f};\ g=1,\ldots,G, \tag{131}$$

$$\psi_{1,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} < 0; j=1,\ldots,J_{\sigma f}; g=1,\ldots,G, \tag{132}$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) = -\delta_{g_j g}N_{i_j,m_j}\psi^{(1),g}(r,\boldsymbol{\Omega}), j=1,\ldots,J_{\sigma f}; g=1,\ldots,G, \tag{133}$$

$$\psi_{2,j}^{(2),g}(r_d,\boldsymbol{\Omega}) = 0, \boldsymbol{\Omega}\cdot\mathbf{n} > 0; j=1,\ldots,J_{\sigma f};\ g=1,\ldots,G. \tag{134}$$

Furthermore, the 2nd-level adjoint functions, $u_{1,j}^{(2),g}$ and $u_{2,j}^{(2),g}, j=1,\ldots,J_{\sigma f}; g=1,\ldots,G$, which appear in Equation (130) are the solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (19), (21), (29) and (30) of Part III [3], which are reproduced below for convenient reference:

$$B^g\left(\boldsymbol{\alpha}^0\right)u_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = N_{i_j,m_j}\nu_{i_j}^{g_j}\chi^g\varphi_0^{g_j}(r),\ \ j=1,\ldots,J_{\sigma f}; g=1,\ldots,G, \tag{135}$$

$$u_{1,j}^{(2),g}(r_d,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} < 0; j = 1,\ldots,J_{\sigma f};\;\; g = 1,\ldots,G, \tag{136}$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)u_{2,j}^{(2),g}(r,\mathbf{\Omega}) = \delta_{g_j g} N_{i_j,m_j} \nu_{i_j}^{g} \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r), j = 1,\ldots,J_{\sigma f}; g = 1,\ldots,G, \tag{137}$$

$$u_{2,j}^{(2),g}(r_d,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} > 0; j = 1,\ldots,J_{\sigma f}; g = 1,\ldots,G. \tag{138}$$

In Equation (130), the parameters f_j and n_{m_2} correspond to the fission cross sections and the isotopic number densities, denoted as $f_j \equiv \sigma_{f,i_j}^{g_j}$ and $n_{m_2} \equiv N_{i_{m_2},m_{m_2}}$, respectively. The following results will be used in subsequent derivations:

$$\begin{aligned} \frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial f_j \partial n_{m_2}} &= \frac{\partial^2 \Sigma_t{}^g(\mathbf{t})}{\partial \sigma_{f,i_j}^{g_j} \partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left\{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\left(\sigma_{f,i}^{g}+\sigma_{c,i}^{g}+\sum\limits_{g'=1}^{G}\sigma_{s,l=0,i}^{g\to g'}\right)\right]/\partial\sigma_{f,i_j}^{g_j}\right\}}{\partial N_{i_{m_2},m_{m_2}}} \\ &= \frac{\partial\left\{\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\sigma_{f,i}^{g}\right]/\partial\sigma_{f,i_j}^{g_j}\right\}}{\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left(\delta_{g_j g} N_{i_j,m_j}\right)}{\partial N_{i_{m_2},m_{m_2}}} = \delta_{i_j i_{m_2}}\delta_{g_j g}, \end{aligned} \tag{139}$$

$$\frac{\partial^2\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial f_j \partial n_{m_2}} = \frac{\partial^2\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial \sigma_{f,i_j}^{g_j}\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left[\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\nu_i^{g'}\sigma_{f,i}^{g'}\right]/\partial\sigma_{f,i_j}^{g_j}\right]}{\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left(\delta_{g_j g'} N_{i_j,m_j}\nu_{i_j}^{g'}\right)}{\partial N_{i_{m_2},m_{m_2}}} = \delta_{i_j i_{m_2}}\delta_{g_j g'}\nu_{i_{m_2}}^{g'}. \tag{140}$$

Inserting the results obtained in Equations (47)–(52), (139) and (140) into Equation (130) and performing the respective angular integrations yields the following expression for Equation (130):

$$\begin{aligned} &\left(\tfrac{\partial^2 L}{\partial f_j \partial n_{m_2}}\right)_{(f=\sigma_f,n=N)} = -\delta_{i_j i_{m_2}} \int_V dV \int_{4\pi} d\mathbf{\Omega}\, \psi^{(1),g_j}(r,\mathbf{\Omega})\varphi^{g_j}(r,\mathbf{\Omega}) \\ &- \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega}\left[\psi_{1,i}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega}) + \psi_{2,i}^{(2),g}(r,\mathbf{\Omega})\varphi^{g}(r,\mathbf{\Omega})\right]\sigma_{t,i_{m_2}}^{g} \\ &+ \sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV \xi_{1,j;l}^{(2),g}(r) \sum_{g'=1}^{G}\sigma_{s,l,i_{m_2}}^{g\to g'}\xi_l^{(1),g'}(r) + \sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV \xi_{2,j;l}^{(2),g}(r)\sum_{g'=1}^{G}\sigma_{s,l,i_{m_2}}^{g'\to g}\varphi_l^{g'}(r) \\ &+ \sum_{g=1}^{G}\int_V dV \chi^g \xi_{2,j;0}^{(2),g}(r)\sum_{g'=1}^{G}\nu_{i_{m_2}}^{g'}\sigma_{f,i_{m_2}}^{g'}\varphi_0^{g'}(r) + \sum_{g=1}^{G}\int_V dV \nu_{i_{m_2}}^{g}\sigma_{f,i_{m_2}}^{g}\xi_{1,j;0}^{(2),g}(r)\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r) \\ &+ \tfrac{1}{n_{m_2}}\sum_{g=1}^{G}\int_V dV \xi_{2,j;0}^{(2),g}(r) Q_{SF,i_{m_2}}^{g} - \sum_{g=1}^{G}\int_V dV\int_{4\pi} d\mathbf{\Omega}\left[u_{1,j}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega}) + u_{2,j}^{(2),g}(r,\mathbf{\Omega})\varphi^{g}(r,\mathbf{\Omega})\right]\sigma_{t,i_{m_2}}^{g} \\ &+ \sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV U_{1,j;l}^{(2),g}(r)\sum_{g'=1}^{G}\sigma_{s,l,i_{m_2}}^{g\to g'}\xi_l^{(1),g'}(r) + \sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV U_{2,j;l}^{(2),g}(r)\sum_{g'=1}^{G}\sigma_{s,l,i_{m_2}}^{g'\to g}\varphi_l^{g'}(r) \\ &+ \delta_{i_j i_{m_2}}\nu_{i_{m_2}}^{g_j}\int_V dV \varphi_0^{g_j}(r)\sum_{g=1}^{G}\chi^g\xi_0^{(1),g}(r) + \sum_{g=1}^{G}\int_V dV \nu_{i_{m_2}}^{g}\sigma_{f,i_{m_2}}^{g} U_{1,j;0}^{(2),g}(r)\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r) \\ &+ \sum_{g=1}^{G}\int_V dV \chi^g U_{2,j;0}^{(2),g}(r)\sum_{g'=1}^{G}\nu_{i_{m_2}}^{g'}\sigma_{f,i_{m_2}}^{g'}\varphi_0^{g'}(r) + \tfrac{1}{n_{m_2}}\sum_{g=1}^{G}\int_V dV U_{2,j;0}^{(2),g}(r) Q_{SF,i_{m_2}}^{g}, \\ &for\ j = 1,\ldots,J_{\sigma f}; m_2 = 1,\ldots,J_n. \end{aligned} \tag{141}$$

4.3. Numerical Results for $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_f$

The second-order absolute sensitivities, $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_f$, of the leakage response with respect to the isotopic number densities and the fission cross sections for all isotopes of the PERP benchmark have been computed using Equation (129) and have been independently verified by computing $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_f\partial\mathbf{N}$ using Equation (141). For the PERP benchmark, computing the second-order absolute sensitivities, $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_f$, using Equation (129) requires 16 forward and adjoint PARTISN computations to obtain all the adjoint functions required in Equation (129). On the other hand, computing the alternative expression $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_f\partial\mathbf{N}$ using Equation (141), requires 120 forward and adjoint PARTISN computations to obtain the needed second level adjoint functions required in Equation (141). Thus, computing $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_f$ using Equation (129) is about 8 ($\approx$ 120/16) times more efficient than computing $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\sigma}_f\partial\mathbf{N}$ by using Equation (141).

The matrix $\partial^2 L/\partial n_j \partial f_{m_2}, j = 1,\ldots,J_n; m_2 = 1,\ldots,J_{\sigma f}$ has dimensions $J_n \times J_{\sigma f}(= 6\times 60)$, where $J_{\sigma f} = G \times N_f = 30\times 2$, and where $N_f = 2$ denotes the total number of fissionable isotopes in the PERP benchmark. The matrix of 2nd-order relative sensitivities corresponding to $\partial^2 L/\partial n_j \partial f_{m_2}, j = 1,\ldots,J_n; m_2 = 1,\ldots,J_{\sigma f}$, is denoted as $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^g_{f,k}\right)$ and is defined as follows:

$$\mathbf{S}^{(2)}\left(N_{i,m},\sigma^g_{f,k}\right) \triangleq \frac{\partial^2 L}{\partial N_{i,m}\partial \sigma^g_{f,k}}\left(\frac{N_{i,m}\sigma^g_{f,k}}{L}\right), \quad i = 1,\ldots,6; m = 1,2; k = 1,2;\ g = 1,\ldots,30. \tag{142}$$

Table 19 summarizes the results for the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^g_{f,k}\right), i = 1,\ldots,6; k,m = 1,2;\ \ g = 1,\ldots,30$, which comprises the 2nd-order relative sensitivities of the leakage response with respect to the isotopic number densities and the fission cross sections, for all isotopes. To facilitate the presentation of the numerical results, the $J_n \times J_{\sigma f}(= 6\times 60)$ matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^g_{f,k}\right)$ has been partitioned into $J_n \times N_f(= 6\times 2)$ submatrices, each of dimensions $1\times G = 1\times 30$. The computational results are as follows: (i) all 360 elements of the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^g_{f,k}\right)$ have positive values, and (ii) of the 360 elements, 21 elements have very large relative sensitivities, with absolute values greater than 1.0, as shown in shaded cells in the table. All of these large sensitivities involve the fission cross sections of ^{239}Pu and most of them relate to the isotopic number densities of ^{239}Pu or ^{1}H. Of the sensitivities summarized in Table 19, the single largest relative value is $S^{(2)}\left(N_{1,1},\sigma^{12}_{f,1}\right) = 11.735$. The results in Table 19 also indicate that, when the 2nd-order mixed relative sensitivities $S^{(2)}\left(N_{i,m},\sigma^g_{f,k}\right)$ involve the isotopic number densities of isotopes ^{69}Ga and ^{71}Ga or the microscopic fission cross sections of isotope ^{240}Pu, their absolute values are all smaller than 1.0. The element with the largest value in the respective submatrix is related to the microscopic fission cross sections for the 12th energy group of isotopes ^{239}Pu and ^{240}Pu.

Table 19. Summary presentation of the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma^g_{f,k}\right),\ i = 1,\ldots,6;\ k,m = 1,2; g = 1,\ldots,30$

	***k* = 1** (**^{239}Pu**)	***k* = 2** (**^{240}Pu**)
$i = 1$ (^{239}Pu)	$\mathbf{S}^{(2)}\left(N_{1,1},\sigma^g_{f,1}\right)$ 12 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{1,1},\sigma^g_{f,2}\right)$ Max. value = 5.62×10^{-1} at g = 12
$i = 2$ (^{240}Pu)	$\mathbf{S}^{(2)}\left(N_{2,1},\sigma^g_{f,1}\right)$1 element with absolute value >1.0	$\mathbf{S}^{(2)}\left(N_{2,1},\sigma^g_{f,2}\right)$ Max. value = 1.12×10^{-1} at g = 12
$i = 3$ (^{69}Ga)	$\mathbf{S}^{(2)}\left(N_{3,1},\sigma^g_{f,1}\right)$ Max. value = 4.72×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{3,1},\sigma^g_{f,2}\right)$ Max. value = 2.44×10^{-4} at g = 12
$i = 4$ (^{71}Ga)	$\mathbf{S}^{(2)}\left(N_{4,1},\sigma^g_{f,1}\right)$ Max. value = 2.98×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{4,1},\sigma^g_{f,2}\right)$ Max. value = 1.54×10^{-4} at g = 12
$i = 5$ (C)	$\mathbf{S}^{(2)}\left(N_{5,2},\sigma^g_{f,1}\right)$1 element with absolute value >1.0	$\mathbf{S}^{(2)}\left(N_{5,2},\sigma^g_{f,2}\right)$ Max. value = 6.13×10^{-2} at g = 12
$i = 6$ (^{1}H)	$\mathbf{S}^{(2)}\left(N_{6,2},\sigma^g_{f,1}\right)$ 7 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{6,2},\sigma^g_{f,2}\right)$ Max. value = 9.73×10^{-2} at g = 12

4.3.1. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1},\sigma_{f,1}^{g}\right), g=1,\ldots,30$

The submatrix $\mathbf{S}^{(2)}\left(N_{1,1},\sigma_{f,1}^{g}\right),\ g=1,\ldots,30$, comprises the 2$^{\text{nd}}$-order sensitivities of the leakage response with respect to the isotopic number density and the fission cross sections of ^{239}Pu. The 12 elements of this matrix which have values greater than 1.0 are presented in bold in Table 20. These 12 large 2nd-order mixed relative sensitivities involve the fission cross sections of isotope ^{239}Pu for the energy groups $g=6,\ldots,16$ and $g=30$, respectively. The element having the largest value in this submatrix is $S^{(2)}\left(N_{1,1},\sigma_{f,1}^{12}\right)=11.735$.

Table 20. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1},\sigma_{f,1}^{g}\right),\ g=1,\ldots,30$.

g	Relative Sensitivities	g	Relative Sensitivities
1	0.005	16	**2.654**
2	0.011	17	0.979
3	0.032	18	0.536
4	0.144	19	0.443
5	0.683	20	0.424
6	**1.735**	21	0.380
7	**7.787**	22	0.335
8	**6.470**	23	0.286
9	**7.761**	24	0.267
10	**8.073**	25	0.228
11	**7.521**	26	0.212
12	**11.735**	27	0.194
13	**8.197**	28	0.115
14	**5.313**	29	0.154
15	**3.007**	30	**1.467**

4.3.2. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{2,1},\sigma_{f,1}^{g}\right), g=1,\ldots,30$

The submatrix $\mathbf{S}^{(2)}\left(N_{2,1},\sigma_{f,1}^{g}\right),\ g=1,\ldots,30$, comprising the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of isotope 2 (^{240}Pu) and the fission cross sections of isotope 1 (^{239}Pu), contains a single large element that has an absolute value greater than 1.0, which is $S^{(2)}\left(N_{2,1},\sigma_{f,1}^{g=12}\right)=1.290$.

4.3.3. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{5,2},\sigma_{f,1}^{g}\right), g=1,\ldots,30$

The submatrix $\mathbf{S}^{(2)}\left(N_{5,2},\sigma_{f,1}^{g}\right),\ g=1,\ldots,30$, for the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of isotope 5 (C) and the fission cross sections of isotope 1 (^{239}Pu), also contains a single large element that has an absolute value greater than 1.0, namely, $S^{(2)}\left(N_{5,2},\sigma_{f,1}^{g=12}\right)=1.184$.

4.3.4. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{6,2},\sigma_{f,1}^{g}\right), g=1,\ldots,30$

The submatrix $\mathbf{S}^{(2)}\left(N_{6,2},\sigma_{f,1}^{g}\right),\ g=1,\ldots,30$, comprising the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of isotope 6 (^{1}H) and the fission cross sections of isotope 1 (^{239}Pu), includes 7 elements that have values greater than 1.0, as listed in Table 21. These 7 relative sensitivities are concentrated in the energy groups $g=7,\ldots,13$ of the fission cross sections for isotope ^{239}Pu.

Table 21. Elements of $\mathbf{S}^{(2)}\left(N_{6,2},\sigma_{f,1}^{g}\right)$, $g=1,\ldots,30$ with absolute values greater than 1.0.

Group	$g = 7$	$g = 8$	$g = 9$	$g = 10$	$g = 11$	$g = 12$	$g = 13$
values	1.279	1.061	1.266	1.312	1.217	1.879	1.294

5. Mixed Second-Order Sensitivities of the PERP Total Leakage Response with Respect to the Parameters Underlying the Benchmark's Isotopic Number Densities and Average Number of Neutrons per Fission

This Section presents the computation and analysis of the numerical results for the 2nd-order mixed sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N}\partial \boldsymbol{\nu}$ of the leakage response with respect to the isotopic number densities and the average number of neutrons per fission of all isotopes of the PERP benchmark. These 2nd-order mixed sensitivities can also be computed using the alternative expression $\partial^2 L(\boldsymbol{\alpha})/\partial \boldsymbol{\nu}\partial \mathbf{N}$. These two alternative paths are presented in Sections 5.1 and 5.2, respectively.

5.1. Computing the Second-Order Sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N}\partial \boldsymbol{\nu}$

The equations needed for deriving the expressions of the 2nd-order sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial \mathbf{N}\partial \boldsymbol{\nu}$ are obtained by particularizing Equations (160), (169), (179) and (206) in [5] to the PERP benchmark. Specifically, Equation (160) in [5] takes on the following particular form for the PERP benchmark:

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(1)}_{(n=N,f=\nu)} = \sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\varphi^{g'}(r,\boldsymbol{\Omega}')\chi^{g}\frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial f_{m_2}} \\
&+\sum_{g=1}^{G}\int_V dV \int_{4\pi} d\boldsymbol{\Omega}\psi_{1,j}^{(2),g}(r,\boldsymbol{\Omega})\frac{\partial\left[\left(\nu\Sigma_f\right)^{g}\right]}{\partial f_{m_2}}\sum_{g'=1}^{G}\int_{4\pi} d\boldsymbol{\Omega}'\chi^{g'}\psi^{(1),g'}(r,\boldsymbol{\Omega}'), \\
&for\ j=1,\ldots,J_n;\quad m_2=J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu\,,
\end{aligned} \tag{143}$$

where the 2nd-level adjoint functions $\psi_{1,j}^{(2),g}$ and $\psi_{2,j}^{(2),g}$, $j=1,\ldots,J_{\sigma f}; g=1,\ldots,G$ are the solutions of the 2nd-Level Adjoint Sensitivity System presented previously in Equations (11)–(14). In Equation (143), the parameters n_j correspond to the isotopic number densities, denoted as $n_j \equiv N_{i_j,m_j}$, and the parameters $f_{m_2}, m_2 = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu$, denoted as $f_{m_2} \equiv \nu_{i_{m_2}}^{g_{m_2}}$, correspond to the respective parameter for average number of neutrons per fission in the vector $\boldsymbol{\nu} \triangleq \left[f_{J_{\sigma f}+1},\ldots,f_{J_{\sigma f}+J_\nu}\right]^\dagger \triangleq \left[\nu_{i=1}^{1},\nu_{i=1}^{2},\ldots,\nu_{i=1}^{G},\ldots,\nu_{i}^{g},\ldots,\nu_{i=N_f}^{1},\ldots,\nu_{i=N_f}^{G}\right]^\dagger$, for $i=1,\ldots,N_f$; $g=1,\ldots,G; J_\nu = G\times N_f$, as shown in Part I [1] and Appendix A. Noting that

$$\frac{\partial\left[\left(\nu\Sigma_f\right)^{g}\right]}{\partial f_{m_2}} = \frac{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I}N_{i,m}\left(\nu\sigma_f\right)_i^{g}\right]}{\partial \nu_{i_{m_2}}^{g_{m_2}}} = \frac{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I}N_{i,m}\nu_i^{g}\sigma_{f,i}^{g}\right]}{\partial \nu_{i_{m_2}}^{g_{m_2}}} = \delta_{g_{m_2}g}N_{i_{m_2},m_{m_2}}\sigma_{f,i_{m_2}}^{g}, \tag{144}$$

$$\frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial f_{m_2}} = \frac{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I}N_{i,m}\left(\nu\sigma_f\right)_i^{g'}\right]}{\partial \nu_{i_{m_2}}^{g_{m_2}}} = \frac{\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I}N_{i,m}\nu_i^{g'}\sigma_{f,i}^{g'}\right]}{\partial \nu_{i_{m_2}}^{g_{m_2}}} = \delta_{g_{m_2}g'}N_{i_{m_2},m_{m_2}}\sigma_{f,i_{m_2}}^{g'}, \tag{145}$$

and inserting the results obtained in Equations (144) and (145) into Equation (143), yields the following expression for Equation (143):

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(1)}_{(n=N,f=\nu)} = N_{i_{m_2},m_{m_2}}\sigma_{f,i_{m_2}}^{g_{m_2}}\int_V dV\left[\xi_{1,j;0}^{(2),g_{m_2}}(r)\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r)+\varphi_0^{g_{m_2}}(r)\sum_{g=1}^{G}\chi^{g}\xi_{2,j;0}^{(2),g}(r)\right], \\
&for j=1,\ldots,J_n;\quad m_2=J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu\,.
\end{aligned} \tag{146}$$

The contributions stemming from Equation (169) in [5] to $\partial^2 L/\partial n_j \partial f_{m_2}$ are:

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(2)}_{(n=N,f=\nu)} = \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{1,j}^{(2),g}(r,\mathbf{\Omega}) \frac{\partial\left[\left(\nu\Sigma_f\right)^g\right]}{\partial f_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \chi^{g'} \psi^{(1),g'}(r,\mathbf{\Omega}') \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \theta_{2,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r,\mathbf{\Omega}') \chi^g \frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial f_{m_2}}, \\
&for\ j = 1,\ldots,J_n;\quad m_2 = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu\,,
\end{aligned} \tag{147}$$

where the 2nd-level adjoint functions $\theta_{1,j}^{(2),g}$, and $\theta_{2,j}^{(2),g}$, $j = 1,\ldots,J_n; g = 1,\ldots,G$, are the solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (19)–(22). Inserting Equations (144) and (145) into Equation (147), yields the following expression for Equation (147):

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(2)}_{(n=N,f=\nu)} = N_{i_{m_2},m_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV \left[\Theta_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \Theta_{2,j;0}^{(2),g}(r)\right], \\
&for j = 1,\ldots,J_n;\quad m_2 = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu\,.
\end{aligned} \tag{148}$$

The contributions stemming from Equation (179) in [5] to $\partial^2 L/\partial n_j \partial f_{m_2}$ are given by the following expression:

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(3)}_{(n=N,f=\nu)} = \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} \psi^{(1),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r,\mathbf{\Omega}') \chi^g \frac{\partial^2\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial n_j \partial f_{m_2}} \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} u_{1,j}^{(2),g}(r,\mathbf{\Omega}) \frac{\partial\left[\left(\nu\Sigma_f\right)^g\right]}{\partial f_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \chi^{g'} \psi^{(1),g'}(r,\mathbf{\Omega}') \\
&+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} u_{2,j}^{(2),g}(r,\mathbf{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \varphi^{g'}(r,\mathbf{\Omega}') \chi^g \frac{\partial\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial f_{m_2}}, \\
&for\ j = 1,\ldots,J_n;\quad m_2 = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu\,,
\end{aligned} \tag{149}$$

where the 2nd-level adjoint functions $u_{1,j}^{(2),g}$, and $u_{2,j}^{(2),g}$, $j = 1,\ldots,J_n; g = 1,\ldots,G$, are the solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (27)–(30). Note that the following relations hold:

$$\frac{\partial^2\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial n_j \partial f_{m_2}} = \frac{\partial^2\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial N_{i_j,m_j} \partial \nu_{i_{m_2}}^{g_{m_2}}} = \frac{\partial\left[\partial\left[\sum_{m=1}^{M}\sum_{i=1}^{I} N_{i,m} \nu_i^{g'} \sigma_{f,i}^{g'}\right]/\partial N_{i_j,m_j}\right]}{\partial \nu_{i_{m_2}}^{g_{m_2}}} = \frac{\partial\left\{\nu_{i_j}^{g'} \sigma_{f,i_j}^{g'}\right\}}{\partial \nu_{i_{m_2}}^{g_{m_2}}} = \delta_{i_j i_{m_2}} \delta_{g_{m_2} g'} \sigma_{f,i_{m_2}}^{g'}. \tag{150}$$

Inserting the results obtained in Equations (144), (145) and (150) into Equation (149), yields the following expression for Equation (149):

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(3)}_{(n=N,f=\nu)} = \delta_{i_j i_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \xi_0^{(1),g}(r) \\
&+ N_{i_{m_2},m_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV \left[U_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g U_{2,j;0}^{(2),g}(r)\right], \\
&for\ j = 1,\ldots,J_n;\quad m_2 = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu\,.
\end{aligned} \tag{151}$$

Additional contributions stemming from the sources are computed by particularizing Equation (206) in [5] to the PERP benchmark, which yields the following expression:

$$\begin{aligned}
&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(4)}_{(n=N,f=\nu)} = \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\mathbf{\Omega} g_{1,j}^{(2),g}(r,\mathbf{\Omega}) \frac{\partial\left[\left(\nu\Sigma_f\right)^g(\mathbf{f})\right]}{\partial f_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\mathbf{\Omega}' \chi^{g'} \psi^{(1),g'}\left(r,\mathbf{\Omega}'\right), \\
&for j = 1,\ldots,J_n;\quad m_2 = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu\,,
\end{aligned} \tag{152}$$

where the 2nd-level adjoint functions $g_{1,j}^{(2),g}, j = 1, \ldots, J_n;\ g = 1, \ldots, G$, are the solutions of the 2nd-LASS presented previously in Equations (35) and (36). Inserting the results obtained in Equations (144) into Equation (152) yields the following expression for Equation (152):

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(4)}_{(n=N,f=\nu)} = N_{i_{m_2},m_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV G_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r),\\ &for j = 1, \ldots, J_n;\quad m_2 = J_{\sigma f} + 1, \ldots, J_{\sigma f} + J_\nu\,.\end{aligned} \tag{153}$$

Collecting the partial contributions obtained in Equations (146), (148), (151) and (153), yields the following result:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)_{(n=N,f=\nu)} = \sum_{i=1}^{4} \left(\frac{\partial^2 L}{\partial n_j \partial f_{m_2}}\right)^{(i)}_{(n=N,f=\nu)}\\ &= N_{i_{m_2},m_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV \left[\xi_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \xi_{2,j;0}^{(2),g}(r)\right]\\ &+ N_{i_{m_2},m_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV \left[\Theta_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \Theta_{2,j;0}^{(2),g}(r)\right]\\ &+ \delta_{i_j i_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g \xi_0^{(1),g}(r)\\ &+ N_{i_{m_2},m_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV \left[U_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r) + \varphi_0^{g_{m_2}}(r) \sum_{g=1}^{G} \chi^g U_{2,j;0}^{(2),g}(r)\right]\\ &+ N_{i_{m_2},m_{m_2}} \sigma_{f,i_{m_2}}^{g_{m_2}} \int_V dV G_{1,j;0}^{(2),g_{m_2}}(r) \sum_{g'=1}^{G} \chi^{g'} \xi_0^{(1),g'}(r), for\ j = 1, \ldots, J_n;\quad m_2 = J_{\sigma f} + 1, \ldots, J_{\sigma f} + J_\nu\,.\end{aligned} \tag{154}$$

5.2. Alternative Path: Computing the Second-Order Sensitivities $\partial^2 L(\boldsymbol{\alpha}) / \partial \boldsymbol{\nu} \partial \mathbf{N}$

Due to symmetry of the mixed 2nd-order sensitivities, the results to be computed using the expressions for $\partial^2 L(\boldsymbol{\alpha}) / \partial \mathbf{N} \partial \boldsymbol{\nu}$ obtained in Equation (154) can be verified by obtaining and using the expressions for $\partial^2 L(\boldsymbol{\alpha}) / \partial \boldsymbol{\nu} \partial \mathbf{N}$. The equations needed for deriving the expression of the 2nd-order mixed sensitivities $\partial^2 L(\boldsymbol{\alpha}) / \partial \boldsymbol{\nu} \partial \mathbf{N}$ are obtained by particularizing Equations (177), (178), (179) and (181) in [5] to the PERP benchmark, which yields:

$$\begin{aligned}&\left(\frac{\partial^2 L}{\partial f_j \partial n_{m_2}}\right)_{(f=\nu,n=N)} = -\sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} \left[u_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \psi^{(1),g}(r,\boldsymbol{\Omega}) + u_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) \varphi^g(r,\boldsymbol{\Omega})\right] \frac{\partial \Sigma_t{}^g(\mathbf{t})}{\partial n_{m_2}}\\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} u_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \psi^{(1),g'}(r,\boldsymbol{\Omega}') \frac{\partial \Sigma_s^{g\to g'}(\mathbf{s};\boldsymbol{\Omega}\to\boldsymbol{\Omega}')}{\partial n_{m_2}}\\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} u_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \varphi^{g'}(r,\boldsymbol{\Omega}') \frac{\partial \Sigma_s^{g'\to g}(\mathbf{s};\boldsymbol{\Omega}'\to\boldsymbol{\Omega})}{\partial n_{m_2}}\\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} \psi^{(1),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \varphi^{g'}(r,\boldsymbol{\Omega}') \chi^g \frac{\partial^2 \left[(\nu\Sigma_f)^{g'}\right]}{\partial f_j \partial n_{m_2}}\\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} u_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) \frac{\partial \left[(\nu\Sigma_f)^g\right]}{\partial n_{m_2}} \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \chi^{g'} \psi^{(1),g'}(r,\boldsymbol{\Omega}')\\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} u_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) \sum_{g'=1}^{G} \int_{4\pi} d\boldsymbol{\Omega}' \varphi^{g'}(r,\boldsymbol{\Omega}') \chi^g \frac{\partial \left[(\nu\Sigma_f)^{g'}\right]}{\partial n_{m_2}}\\ &+ \sum_{g=1}^{G} \int_V dV \int_{4\pi} d\boldsymbol{\Omega} u_{2,j}^{(2),g}(r,\boldsymbol{\Omega}) \frac{\partial Q^g(\mathbf{q};r,\boldsymbol{\Omega})}{\partial n_{m_2}},\ for\ j = J_{\sigma f} + 1, \ldots, J_{\sigma f} + J_\nu\,; m_2 = 1, \ldots, J_n,\end{aligned} \tag{155}$$

where the 2nd-level adjoint functions, $u_{1,j}^{(2),g}$ and $u_{2,j}^{(2),g}, j = 1, \ldots, J_{\sigma f}; g = 1, \ldots, G$, are the solutions of the 2nd-Level Adjoint Sensitivity System presented in Equations (116), (118), (124) and (125) of Part III [3], which are reproduced below for convenient reference:

$$B^g\left(\boldsymbol{\alpha}^0\right) u_{1,j}^{(2),g}(r,\boldsymbol{\Omega}) = N_{i_j,m_j} \sigma_{f,i_j}^{g_j} \chi^g \varphi_0^{g_j}(r),\ \ j = J_{\sigma f} + 1, \ldots, J_{\sigma f} + J_\nu; g = 1, \ldots, G, \tag{156}$$

$$u_{1,j}^{(2),g}(r_d,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} < 0; j = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu;\;\; g = 1,\ldots,G, \tag{157}$$

$$A^{(1),g}\left(\boldsymbol{\alpha}^0\right)u_{2,j}^{(2),g}(r,\mathbf{\Omega}) = \delta_{g_j g} N_{i_j,m_j}\sigma_{f,i_j}^{g_j}\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r), j = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu; g = 1,\ldots,G, \tag{158}$$

$$u_{2,j}^{(2),g}(r_d,\mathbf{\Omega}) = 0, \mathbf{\Omega}\cdot\mathbf{n} > 0; j = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu; g = 1,\ldots,G. \tag{159}$$

In Equation (155), the parameters f_j and n_{m_2} correspond to the average number of neutrons per fission and the isotopic number densities, denoted as $f_j \equiv \nu_{i_j}^{g_j}$ and $n_{m_2} \equiv N_{i_{m_2},m_{m_2}}$, respectively. The following relations hold:

$$\frac{\partial^2\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial f_j \partial n_{m_2}} = \frac{\partial^2\left[\left(\nu\Sigma_f\right)^{g'}\right]}{\partial \nu_{i_j}^{g_j}\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left[\partial\left[\sum\limits_{m=1}^{M}\sum\limits_{i=1}^{I} N_{i,m}\nu_i^{g'}\sigma_{f,i}^{g'}\right]/\partial \nu_{i_j}^{g_j}\right]}{\partial N_{i_{m_2},m_{m_2}}} = \frac{\partial\left(\delta_{g_j g'}N_{i_j,m_j}\sigma_{f,i_j}^{g'}\right)}{\partial N_{i_{m_2},m_{m_2}}} = \delta_{i_j i_{m_2}}\delta_{g_j g'}\sigma_{f,i_{m_2}}^{g'}. \tag{160}$$

Inserting the results obtained in Equations (47)–(52) and (160) into Equation (155) and performing the respective angular integrations yields the following expression for Equation (155):

$$\begin{aligned}
\left(\tfrac{\partial^2 L}{\partial f_j \partial n_{m_2}}\right)_{(f=\nu,n=N)} &= -\sum_{g=1}^{G}\int_V dV\int_{4\pi} d\mathbf{\Omega}\left[u_{1,j}^{(2),g}(r,\mathbf{\Omega})\psi^{(1),g}(r,\mathbf{\Omega}) + u_{2,j}^{(2),g}(r,\mathbf{\Omega})\varphi^g(r,\mathbf{\Omega})\right]\sigma_{t,i_{m_2}}^g \\
&+ \sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV U_{1,j;l}^{(2),g}(r)\sum_{g'=1}^{G}\sigma_{s,l,i_{m_2}}^{g\to g'}\xi_l^{(1),g'}(r) + \sum_{g=1}^{G}\sum_{l=0}^{ISCT}(2l+1)\int_V dV U_{2,j;l}^{(2),g}(r)\sum_{g'=1}^{G}\sigma_{s,l,i_{m_2}}^{g'\to g}\varphi_l^{g'}(r) \\
&+\delta_{i_j i_{m_2}}\sigma_{f,i_{m_2}}^{g_j}\int_V dV\varphi_0^{g_j}(r)\sum_{g=1}^{G}\chi^g\xi_0^{(1),g}(r) + \sum_{g=1}^{G}\int_V dV\nu_{i_{m_2}}^g\sigma_{f,i_{m_2}}^g U_{1,j;0}^{(2),g}(r)\sum_{g'=1}^{G}\chi^{g'}\xi_0^{(1),g'}(r) \\
&+ \sum_{g=1}^{G}\int_V dV\chi^g U_{2,j;0}^{(2),g}(r)\sum_{g'=1}^{G}\nu_{i_{m_2}}^{g'}\sigma_{f,i_{m_2}}^{g'}\varphi_0^{g'}(r) + \tfrac{1}{n_{m_2}}\sum_{g=1}^{G}\int_V dV U_{2,j;0}^{(2),g}(r)Q_{SF,i_{m_2}}^g, \\
&for\ j = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu; m_2 = 1,\ldots,J_n.
\end{aligned} \tag{161}$$

5.3. Numerical Results for $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\nu}$

The second-order absolute sensitivities, $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\nu}$, of the leakage response with respect to the isotopic number densities and the average number of neutrons per fission for all isotopes of the PERP benchmark have been computed using Equation (154) and have been independently verified by computing $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\nu}\partial\mathbf{N}$ using Equation (161). Computing the second-order absolute sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\nu}$ using Equation (154) requires 16 forward and adjoint PARTISN computations to obtain all of the required 2nd-level adjoint functions. On the other hand, computing the alternative expression $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\nu}\partial\mathbf{N}$ using Equation (161) requires 60 forward and adjoint PARTISN computations to obtain the second-level adjoint functions required in Equation (161). Thus, computing $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\nu}$ using Equation (154) is about 4 times more efficient than computing $\partial^2 L(\boldsymbol{\alpha})/\partial\boldsymbol{\nu}\partial\mathbf{N}$ by using Equation (161).

The matrix $\partial^2 L/\partial n_j \partial f_{m_2}, j = 1,\ldots,J_n; m_2 = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu$ has dimensions $J_n \times J_\nu (= 6\times 60)$, where $J_\nu = G\times N_f = 30\times 2$. The matrix of 2nd-order relative sensitivities corresponding to $\partial^2 L/\partial n_j \partial f_{m_2}, j = 1,\ldots,J_n; m_2 = J_{\sigma f}+1,\ldots,J_{\sigma f}+J_\nu$, is denoted as $\mathbf{S}^{(2)}\left(N_{i,m},\nu_k^g\right)$ and is defined as follows:

$$\mathbf{S}^{(2)}\left(N_{i,m},\nu_k^g\right) \triangleq \frac{\partial^2 L}{\partial N_{i,m}\partial \nu_k^g}\left(\frac{N_{i,m}\nu_k^g}{L}\right),\quad i = 1,\ldots,6; m = 1,2; k = 1,2;\ g = 1,\ldots,30. \tag{162}$$

Table 22 summarizes the results obtained for the elements of the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\nu_k^g\right), i = 1,\ldots,6; k,m = 1,2;\ g = 1,\ldots,30$, for the 2nd-order relative sensitivities of the leakage response with respect to the isotopic number densities and the average number of neutrons per fission for all isotopes. To facilitate the presentation of the numerical results, the $J_n \times J_\nu (= 6\times 60)$ matrix $\mathbf{S}^{(2)}\left(N_{i,m},\nu_k^g\right)$ has been partitioned into $J_n \times N_f (= 6\times 2)$ submatrices, each of dimensions $1\times G = 1\times 30$. The computational results have shown that the majority (358 out of 360) of the elements in the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma_{f,k}^g\right)$ have positive values; only 2 elements have very small negative values, of the order of 10^{-4} and less. As shown in shaded cells in Table 22, 34 among the 360 components of $\mathbf{S}^{(2)}\left(N_{i,m}, \nu_k^g\right)$ have very large relative sensitivities, with absolute values greater than 1.0. All of these large sensitivities involve the average number of neutrons per fission of isotope ^{239}Pu and relate to the isotopic number densities of isotopes ^{239}Pu, ^{240}Pu, C or ^{1}H, respectively. The overall largest relative value in the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \nu_k^g\right)$ is $S^{(2)}\left(N_{1,1}, \nu_1^{12}\right) = 16.06$. The computed results have also shown that the 2$^{\text{nd}}$-order mixed relative sensitivities $\mathbf{S}^{(2)}\left(N_{i,m}, \nu_k^g\right)$ involving the isotopic number densities of isotopes ^{69}Ga and ^{71}Ga or the average number of neutrons per fission of ^{240}Pu have absolute values smaller than 1.0; the element with the largest value in the respective submatrix is related to the average number of neutrons per fission for the 12th energy group of isotopes ^{239}Pu and ^{240}Pu.

Table 22. Summary presentation of the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \nu_k^g\right),\ i = 1, \ldots, 6;\ k, m = 1, 2; g = 1, \ldots, 30.$

	$k = 1$ (^{239}Pu)	**$k = 2$ (^{240}Pu)**
$i = 1$ (^{239}Pu)	$\mathbf{S}^{(2)}\left(N_{1,1}, \nu_{k=1}^g\right)$ 13 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{1,1}, \nu_{k=2}^g\right)$ Max. value = 7.72×10^{-1} at g = 12
$i = 2$ (^{240}Pu)	$\mathbf{S}^{(2)}\left(N_{2,1}, \nu_{k=1}^g\right)$ 6 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{2,1}, \nu_{k=2}^g\right)$ Max. value = 1.55×10^{-1} at g = 12
$i = 3$ (^{69}Ga)	$\mathbf{S}^{(2)}\left(N_{3,1}, \nu_{k=1}^g\right)$ Max. value = 6.52×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{3,1}, \nu_{k=2}^g\right)$ Max. value = 3.39×10^{-4} at g = 12
$i = 4$ (^{71}Ga)	$\mathbf{S}^{(2)}\left(N_{4,1}, \nu_{k=1}^g\right)$ Max. value = 4.11×10^{-3} at g = 12	$\mathbf{S}^{(2)}\left(N_{4,1}, \nu_{k=2}^g\right)$ Max. value = 2.14×10^{-4} at g = 12
$i = 5$ (C)	$\mathbf{S}^{(2)}\left(N_{5,2}, \nu_{k=1}^g\right)$ 6 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{5,2}, \nu_{k=2}^g\right)$ Max. value = 8.52×10^{-2} at g = 12
$i = 6$ (^{1}H)	$\mathbf{S}^{(2)}\left(N_{6,2}, \nu_{k=1}^g\right)$ 9 elements with absolute values >1.0	$\mathbf{S}^{(2)}\left(N_{6,2}, \nu_{k=2}^g\right)$ Max. value = 1.35×10^{-1} at g = 12

5.3.1. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{1,1}, \nu_{k=1}^g\right), g = 1, \ldots, 30$

The submatrix $\mathbf{S}^{(2)}\left(N_{1,1}, \nu_{k=1}^g\right),\ \ g = 1, \ldots, 30$ comprises the 2nd-order sensitivities of the leakage response with respect to the isotopic number density and the average number of neutrons per fission of ^{239}Pu. Table 23 presents the 13 elements of this submatrix that have values greater than 1.0; these large 2nd-order mixed relative sensitivities are concentrated in energy groups $g = 6, \ldots, 17$ of the average number of neutrons per fission of isotope ^{239}Pu. The largest value in this submatrix is $S^{(2)}\left(N_{1,1}, \nu_1^{12}\right) = 16.06$.

Table 23. Elements of $\mathbf{S}^{(2)}\left(N_{1,1}, \nu_{k=1}^g\right),\ \ g = 1, \ldots, 30$, having absolute values greater than 1.0.

Group	**$g = 6$**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**	**15**	**16**	**17**	**30**
values	2.267	10.10	8.675	10.53	11.07	10.34	16.06	11.30	7.458	4.330	3.987	1.535	5.217

5.3.2. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{2,1}, \nu_{k=1}^g\right), g = 1, \ldots, 30$

The submatrix $\mathbf{S}^{(2)}\left(N_{2,1}, \nu_{k=1}^g\right),\ \ g = 1, \ldots, 30$, comprising the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of isotope 2 (^{240}Pu) and the average number of neutrons per fission of isotope 1 (^{239}Pu), contains 6 large elements that have values greater than 1.0, as listed in Table 24.

Table 24. Elements of $\mathbf{S}^{(2)}\left(N_{2,1}, \nu_{k=1}^{g}\right)$, $g = 1, \ldots, 30$ with absolute values greater than 1.0.

Group	$g = 7$	$g = 9$	$g = 10$	$g = 11$	$g = 12$	$g = 13$
values	1.107	1.162	1.221	1.142	1.772	1.248

5.3.3. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{5,2}, \nu_{k=1}^{g}\right)$, $g = 1, \ldots, 30$

The submatrix $\mathbf{S}^{(2)}\left(N_{5,2}, \sigma_{f,1}^{g}\right)$, $g = 1, \ldots, 30$ comprises the 2nd-order sensitivities of the leakage response with respect to the isotopic number density of isotope C and the average number of neutrons per fission of isotope ^{239}Pu. Table 25 presents the 6 elements of this submatrix which have values greater than 1.0.

Table 25. Elements of $\mathbf{S}^{(2)}\left(N_{5,2}, \nu_{k=1}^{g}\right)$, $g = 1, \ldots, 30$ with absolute values greater than 1.0.

Group	$g = 7$	$g = 9$	$g = 10$	$g = 11$	$g = 12$	$g = 13$
values	1.016	1.083	1.135	1.056	1.638	1.159

5.3.4. Second-Order Relative Sensitivities $\mathbf{S}^{(2)}\left(N_{6,2}, \nu_{k=1}^{g}\right)$, $g = 1, \ldots, 30$

The submatrix $\mathbf{S}^{(2)}\left(N_{6,2}, \nu_{k=1}^{g}\right)$, $g = 1, \ldots, 30$, comprising the 2$^{\text{nd}}$-order sensitivities of the leakage response with respect to the isotopic number density of ^{1}H and to the average number of neutrons per fission of isotope ^{239}Pu, includes 9 elements that have values greater than 1.0, as listed in Table 26. These 9 relative sensitivities are concentrated in the energy groups $g = 7, \ldots, 14$ and $g = 30$ of the average number of neutrons per fission of isotope ^{239}Pu.

Table 26. Elements of $\mathbf{S}^{(2)}\left(N_{6,2}, \nu_{k=1}^{g}\right)$, $g = 1, \ldots, 30$ with absolute values greater than 1.0.

Group	$g = 7$	$g = 8$	$g = 9$	$g = 10$	$g = 11$	$g = 12$	$g = 13$	$g = 14$	$g = 30$
values	1.660	1.424	1.723	1.809	1.687	2.605	1.815	1.188	1.930

6. Discussion and Conclusions

The following conclusions can be drawn from the results for the mixed 2nd-order sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$, $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_s$, $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_f$ and $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\nu}$ reported in this work:

The 2nd-order mixed sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_t$ are mostly negative. Almost all, namely 1072 out of the $J_n \times J_{\sigma t} (= 1080)$ elements in the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{t,k}^{g}\right)$, $i, k = 1, \ldots, 6;\ m = 1, 2; g = 1, \ldots, 30$ of 2nd-order mixed sensitivities have negative values; only 8 elements have very small positive values (e.g., in the order of 10^{-4} or less). Among the 1080 elements in the matrix, 125 elements have very large relative sensitivities, with absolute values greater than 1.0. Majority of those large sensitivities involve the isotopic number densities of isotopes ^{239}Pu or ^{1}H (namely, $N_{1,1}$ and $N_{6,2}$) and/or the microscopic total cross sections of isotopes ^{239}Pu or ^{1}H (namely, $\sigma_{t,1}^{g}$ and $\sigma_{t,6}^{g}$). In the matrix $\mathbf{S}^{(2)}\left(N_{i,m}, \sigma_{t,k}^{g}\right)$, the single largest relative value is $S^{(2)}\left(N_{1,1}, \sigma_{t,6}^{30}\right) = -94.91$. Moreover, the element with the most negative value in each of the submatrices mostly involves the microscopic total cross sections for the 12th energy group or the 30th energy group (i.e., $\sigma_{t,k}^{12}, k = 1, \ldots, 4$ or $\sigma_{t,k}^{30}, k = 5, 6$) of the respective isotopes.

The 2nd-order mixed relative sensitivities corresponding to the $J_n \times J_{\sigma s} = 6 \times 21600$-dimensional matrix $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_s$ are generally very small, with a few exceptions. Specifically, 25 of the $J_n \times J_{\sigma s} = 6 \times 21600$ elements have relative sensitivities with absolute values greater than 1.0. These 25 large elements belong to the submatrices $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,1}^{g' \to g}\right)$, $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=0,6}^{g' \to g}\right)$, $\mathbf{S}^{(2)}\left(N_{1,1}, \sigma_{s,l=1,1}^{g' \to g}\right)$ and

$\mathbf{S}^{(2)}\left(N_{1,1},\sigma_{s,l=1,6}^{g'\to g}\right)$, respectively, and involve the isotopic number density $N_{1,1}$ of ^{239}Pu and the 0th-order or 1st-order scattering cross sections (namely, $\sigma_{s,l=0,k=1}^{g'\to g}, \sigma_{s,l=0,k=6}^{g'\to g}, \sigma_{s,l=1,k=1}^{g'\to g}, \sigma_{s,l=1,k=6}^{g'\to g}$) of ^{239}Pu or ^{1}H. These large sensitivities are positive when involving even-order $(l=0,2)$ scattering cross sections but are negative when involving odd-order $(l=1,3)$ scattering cross sections. Furthermore, the larger the Legendre expansion order $(l=0,\ldots,3)$, the smaller the absolute values of the corresponding mixed 2nd-order relative sensitivities. Noteworthy for the 2nd-order mixed sensitivities $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_s$ is also the observation that for the scattering order $l=0$, most of the largest absolute values of the respective submatrix occur at the 0th-order self-scattering cross sections for the 12th energy group for isotopes ^{239}Pu, ^{240}Pu, ^{69}Ga, ^{71}Ga and C (i.e., $\sigma_{s,l=0,k}^{12\to 12}, k=1,\ldots,5$), or the 0th-order out-scattering cross section between energy groups $g'=12\to g=13$ of isotope ^{1}H (namely, $\sigma_{s,l=0,6}^{12\to 13}$). On the other hand, when the scattering order $l=1,2,3$, these large sensitivities mostly involve the l^{th}-order self-scattering cross sections for the 7th energy group of isotopes ^{239}Pu, ^{240}Pu, ^{69}Ga, ^{71}Ga and C (namely, $\sigma_{s,l,k}^{7\to 7}, l=1,2,3; k=1,\ldots,5$), or 12th energy group of isotope ^{1}H (namely, $\sigma_{s,l,k=6}^{12\to 12}, l=1,2,3$). The overall largest 2nd-order mixed relative sensitivity involving an isotopic number density and a scattering cross section is $S^{(2)}\left(N_{1,1},\sigma_{s,l=0,k=1}^{12\to 12}\right)=1.912$.

All values of the 2nd-order mixed relative sensitivities $\mathbf{S}^{(2)}\left(N_{i,m},\sigma_{f,k}^{g}\right),\ i=1,\ldots,6; k,m=1,2;\ g=1,\ldots,30$ corresponding to the $J_n\times J_{\sigma f}(=6\times 60)$ elements of the matrix $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\sigma}_f$ are positive. The matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma_{f,k}^{g}\right)$ comprises 21 elements which have values greater than 1.0. Most of these elements belong to the submatrices $\mathbf{S}^{(2)}\left(N_{1,1},\sigma_{f,1}^{g}\right)$ and $\mathbf{S}^{(2)}\left(N_{6,2},\sigma_{f,1}^{g}\right)$, involving the fission cross sections of ^{239}Pu and the isotopic number densities of ^{239}Pu or ^{1}H. The 2nd-order mixed relative sensitivities involving the isotopic number densities of ^{240}Pu, ^{69}Ga, ^{71}Ga and C (i.e.,$N_{2,1},N_{3,1},N_{4,1},N_{5,2}$) or the microscopic fission cross sections $\sigma_{f,2}^{g}, g=1,\ldots,30$ of ^{240}Pu are generally smaller than 1.0. The largest element of the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\sigma_{f,k}^{g}\right)$ is $S^{(2)}\left(N_{1,1},\sigma_{f,1}^{12}\right)=11.735$.

The majority of the elements belonging to the $J_n\times J_\nu(=\ 6\times 60)$-dimensional matrix $\mathbf{S}^{(2)}\left(N_{i,m},\nu_k^{g}\right),\ i=1,\ldots,6; k,m=1,2;\ g=1,\ldots,30$, of 2nd-order mixed relative sensitivities corresponding to the matrix $\partial^2 L(\boldsymbol{\alpha})/\partial\mathbf{N}\partial\boldsymbol{\nu}$ have positive values. The matrix $\mathbf{S}^{(2)}\left(N_{i,m},\nu_k^{g}\right)$ comprises 34 elements that have relative sensitivities greater than 1.0. These large sensitivities occur in the submatrices $\mathbf{S}^{(2)}\left(N_{1,1},\nu_{k=1}^{g}\right)$, $\mathbf{S}^{(2)}\left(N_{2,1},\nu_{k=1}^{g}\right)$, $\mathbf{S}^{(2)}\left(N_{5,2},\nu_{k=1}^{g}\right)$ and $\mathbf{S}^{(2)}\left(N_{6,2},\nu_{k=1}^{g}\right)$, which involve the average number of neutrons per fission of isotope ^{239}Pu (i.e., $\nu_{k=1}^{g}$) and the isotopic number densities of isotopes ^{239}Pu, ^{240}Pu, C or ^{1}H (i.e., $N_{1,1},N_{2,1},N_{5,2},N_{6,2}$), respectively. The remaining 2$^{\text{nd}}$-order mixed relative sensitivities in the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\nu_k^{g}\right)$ are all smaller than 1.0. The largest values among these smaller sensitivities involve the average number of neutrons per fission, $\nu_k^{12}, k=1,2$, for the 12th energy group of ^{239}Pu and ^{240}Pu. The overall largest 2nd-order mixed relative sensitivities comprised in the matrix $\mathbf{S}^{(2)}\left(N_{i,m},\nu_k^{g}\right)$ is $S^{(2)}\left(N_{1,1},\nu_1^{12}\right)=16.06$.

Subsequent work will report the values and analyze the effects of the 1st-order and unmixed 2nd-order sensitivities of the PERP's leakage response with respect to the imprecisely known isotopic number densities [13], and the 1st-order sensitivities of the leakage response with respect to the imprecisely known fission spectrum parameters [13]. The overall impact of 1st- and 2nd-order sensitivities on the response uncertainties will also be highlighted [13].

Author Contributions: D.C. conceived and directed the research reported herein, developed the general theory of the second-order comprehensive adjoint sensitivity analysis methodology to compute 1st- and 2nd-order sensitivities of flux functionals in a multiplying system with source, and the uncertainty equations for response moments. R.F. has derived the expressions of the various derivatives with respect to the model parameters to the PERP benchmark and performed all the numerical calculations.

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Appendix A. Definitions of PERP Model Parameters

As presented in Part I [1], the components of the vector of 1st-order sensitivities of the leakage response with respect to the model parameters, denoted as $\mathbf{S}^{(1)}(\boldsymbol{\alpha})$, was defined as follows:

$$\mathbf{S}^{(1)}(\boldsymbol{\alpha}) \triangleq \left[\frac{\partial L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_t}; \frac{\partial L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_s}; \frac{\partial L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_f}; \frac{\partial L(\boldsymbol{\alpha})}{\partial \boldsymbol{\nu}}; \frac{\partial L(\boldsymbol{\alpha})}{\partial \mathbf{p}}; \frac{\partial L(\boldsymbol{\alpha})}{\partial \mathbf{q}}; \frac{\partial L(\boldsymbol{\alpha})}{\partial \mathbf{N}}\right]^{\dagger}. \tag{A1}$$

The symmetric matrix of 2nd-order sensitivities of the leakage response with respect to the model parameters, denoted as $\mathbf{S}^{(2)}(\boldsymbol{\alpha})$, was defined as follows:

$$\mathbf{S}^{(2)}(\boldsymbol{\alpha}) \triangleq \begin{bmatrix}
\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_t \partial \boldsymbol{\sigma}_t} & * & * & * & * & * & * \\
\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_s \partial \boldsymbol{\sigma}_t} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_s \partial \boldsymbol{\sigma}_s} & * & * & * & * & * \\
\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_f \partial \boldsymbol{\sigma}_t} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_f \partial \boldsymbol{\sigma}_s} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\sigma}_f \partial \boldsymbol{\sigma}_f} & * & * & * & * \\
\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\nu} \partial \boldsymbol{\sigma}_t} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\nu} \partial \boldsymbol{\sigma}_s} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\nu} \partial \boldsymbol{\sigma}_f} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \boldsymbol{\nu} \partial \boldsymbol{\nu}} & * & * & * \\
\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{p} \partial \boldsymbol{\sigma}_t} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{p} \partial \boldsymbol{\sigma}_s} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{p} \partial \boldsymbol{\sigma}_f} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{p} \partial \boldsymbol{\nu}} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{p} \partial \mathbf{p}} & * & * \\
\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{q} \partial \boldsymbol{\sigma}_t} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{q} \partial \boldsymbol{\sigma}_s} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{q} \partial \boldsymbol{\sigma}_f} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{q} \partial \boldsymbol{\nu}} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{q} \partial \mathbf{p}} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{q} \partial \mathbf{q}} & * \\
\frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{N} \partial \boldsymbol{\sigma}_t} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{N} \partial \boldsymbol{\sigma}_s} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{N} \partial \boldsymbol{\sigma}_f} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{N} \partial \boldsymbol{\nu}} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{N} \partial \mathbf{p}} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{N} \partial \mathbf{q}} & \frac{\partial^2 L(\boldsymbol{\alpha})}{\partial \mathbf{N} \partial \mathbf{N}}
\end{bmatrix}. \tag{A2}$$

As defined in Equation (1), the vector $\boldsymbol{\alpha} \triangleq \left[\boldsymbol{\sigma}_t; \boldsymbol{\sigma}_s; \boldsymbol{\sigma}_f; \boldsymbol{\nu}; \mathbf{p}; \mathbf{q}; \mathbf{N}\right]^{\dagger}$ denotes the "vector of imprecisely known model parameters", with vector-components $\boldsymbol{\sigma}_t$, $\boldsymbol{\sigma}_s$, $\boldsymbol{\sigma}_f$, $\boldsymbol{\nu}$, $\mathbf{p}$, $\mathbf{q}$ and $\mathbf{N}$, comprising the various model parameters for the microscopic total cross sections, scattering cross sections, fission cross sections, average number of neutrons per fission, fission spectra, sources, and isotopic number densities, which have been described in Part I [1]. For easy referencing, the definitions of these model parameters will be recalled in the remainder of this Appendix A.

The total cross section Σ_t^g for energy group g, $g = 1, \ldots, G$, is computed for the PERP benchmark using the following expression:

$$\Sigma_t^g = \sum_{m=1}^{M=2} \Sigma_{t,m}^g; \;\; \Sigma_{t,m}^g = \sum_i^I N_{i,m} \sigma_{t,i}^g = \sum_i^I N_{i,m} \left[\sigma_{f,i}^g + \sigma_{c,i}^g + \sum_{g'=1}^{G} \sigma_{s,l=0,i}^{g \to g'}\right], \; m = 1, 2, \tag{A3}$$

where m denotes the materials in the PERP benchmark; $\sigma_{f,i}^g$ and $\sigma_{c,i}^g$ 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. As discussed in Part I [1], the total cross section $\Sigma_t^g \to \Sigma_t^g(\mathbf{t})$ will depend on the vector of parameters $\mathbf{t}$, which is defined as follows:

$$\mathbf{t} \triangleq \left[t_1, \ldots, t_{J_t}\right]^{\dagger} \triangleq \left[t_1, \ldots, t_{J_{\sigma t}}; n_1, \ldots, n_{J_n}\right]^{\dagger} \triangleq \left[\boldsymbol{\sigma}_t; \mathbf{N}\right]^{\dagger}, \;\; J_t = J_{\sigma t} + J_n, \tag{A4}$$

where

$$\mathbf{N} \triangleq \left[n_1, \ldots, n_{J_n}\right]^{\dagger} \triangleq \left[N_{1,1}, N_{2,1}, N_{3,1}, N_{4,1}, N_{5,2}, N_{6,2}\right]^{\dagger}, \;\; J_n = 6, \tag{A5}$$

$$\begin{aligned}&\boldsymbol{\sigma}_t \triangleq \left[t_1, \ldots, t_{J_{\sigma t}}\right]^{\dagger} \triangleq \left[\sigma_{t,i=1}^1, \sigma_{t,i=1}^2, \ldots, \sigma_{t,i=1}^G, \ldots, \sigma_{t,i}^g, \ldots, \sigma_{t,i=I}^1, \ldots, \sigma_{t,i=I}^G\right]^{\dagger}, \\ &i = 1, \ldots, I = 6; \; g = 1, \ldots, G = 30; J_{\sigma t} = I \times G.\end{aligned} \tag{A6}$$

In Equations (A4) through (A6), the dagger denotes "transposition," $\sigma_{t,i}^{g}$ denotes the microscopic total cross section for isotope i and energy group g, $N_{i,m}$ denotes the respective isotopic number density, and J_n denotes the total number of isotopic number densities in the model. Thus, the vector **t** comprises a total of $J_t = J_{\sigma t} + J_n = 30 \times 6 + 6 = 186$ imprecisely known "model parameters" as its components.

The scattering transfer cross section $\Sigma_s^{g' \to g}\left(\mathbf{\Omega}' \to \mathbf{\Omega}\right)$ from energy group g', $g' = 1, \ldots, G$ into energy group g, $g = 1, \ldots, G$, is computed using the finite Legendre polynomial expansion of order $ISCT = 3$:

$$\begin{aligned} &\Sigma_s^{g' \to g}\left(\mathbf{\Omega}' \to \mathbf{\Omega}\right) = \sum_{m=1}^{M=2} \Sigma_{s,m}^{g' \to g}\left(\mathbf{\Omega}' \to \mathbf{\Omega}\right), \\ &\Sigma_{s,m}^{g' \to g}\left(\mathbf{\Omega}' \to \mathbf{\Omega}\right) \cong \sum_{i=1}^{I=6} N_{i,m} \sum_{l=0}^{ISCT=3} (2l+1)\, \sigma_{s,l,i}^{g' \to g} P_l\left(\mathbf{\Omega}' \cdot \mathbf{\Omega}\right),\ m = 1, 2, \end{aligned} \tag{A7}$$

where $\sigma_{s,l,i}^{g' \to g}$ denotes the l-th order Legendre-expanded microscopic scattering cross section from energy group g' into energy group g for isotope i. In view of Equation (A7), the scattering cross section $\Sigma_s^{g' \to g}\left(\mathbf{\Omega}' \to \mathbf{\Omega}\right) \to \Sigma_s^{g' \to g}\left(\mathbf{s}; \mathbf{\Omega}' \to \mathbf{\Omega}\right)$ depends on the vector of parameters **s**, which is defined as follows:

$$\mathbf{s} \triangleq \left[s_1, \ldots, s_{J_s}\right]^{\dagger} \triangleq \left[s_1, \ldots, s_{J_{\sigma s}}; n_1, \ldots, n_{J_n}\right]^{\dagger} \triangleq [\boldsymbol{\sigma}_s; \mathbf{N}]^{\dagger},\ J_s = J_{\sigma s} + J_n, \tag{A8}$$

$$\begin{aligned} &\boldsymbol{\sigma}_s \triangleq \left[s_1, \ldots, s_{J_{\sigma s}}\right]^{\dagger} \triangleq \left[\sigma_{s,l=0,i=1}^{g'=1 \to g=1}, \sigma_{s,l=0,i=1}^{g'=2 \to g=1}, \ldots, \sigma_{s,l=0,i=1}^{g'=G \to g=1}, \sigma_{s,l=0,i=1}^{g'=1 \to g=2}, \sigma_{s,l=0,i=1}^{g'=2 \to g=2}, \ldots, \sigma_{s,l,i}^{g' \to g}, \ldots, \sigma_{s,ISCT,i=I}^{G \to G}\right]^{\dagger}, \\ &for\ \ l = 0, \ldots, ISCT;\ i = 1, \ldots, I;\ \ g, g' = 1, \ldots, G;\ \ J_{\sigma s} = (G \times G) \times I \times (ISCT + 1). \end{aligned} \tag{A9}$$

The expressions in Equations (A7) and (A3) 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. Therefore, the total number of zeroth-order scattering cross section comprise in $\boldsymbol{\sigma}_s$ is denoted as $J_{\sigma s,l=0}$, where $J_{\sigma s,l=0} = G \times G \times I$; and the total number of higher order (i.e., $l \geq 1$) scattering cross sections comprised in $\boldsymbol{\sigma}_s$ is denoted as $J_{\sigma s,l\geq 1}$, where $J_{\sigma s,l\geq 1} = G \times G \times I \times ISCT$, with $J_{\sigma s,l=0} + J_{\sigma s,l\geq 1} = J_{\sigma s}$. Thus, the vector **s** comprises a total of $J_{\sigma s} + J_n = 30 \times 30 \times 6 \times (3+1) + 6 = 21606$ imprecisely known components ("model parameters").

The transport code PARTISN [9] computes the quantity $\left(\nu\Sigma_f\right)^g$ using directly the quantities $(\nu\sigma)_{f,i}^{g}$, which are provided in data files for each isotope i, and energy group g, as follows

$$\left(\nu\Sigma_f\right)^g = \sum_{m=1}^{M=2} \left(\nu\Sigma_f\right)_m^g;\quad \left(\nu\Sigma_f\right)_m^g = \sum_{i=1}^{I=6} N_{i,m}\left(\nu\sigma_f\right)_i^g,\ \ m = 1, 2. \tag{A10}$$

In view of Equation (A10), the quantity $\left(\nu\Sigma_f\right)^g \to \left(\nu\Sigma_f\right)^g(\mathbf{f}; r)$ depends on the vector of parameters **f**, which is defined as follows:

$$\mathbf{f} \triangleq \left[f_1, \ldots, f_{J_{\sigma f}}; f_{J_{\sigma f}+1}, \ldots, f_{J_{\sigma f}+J_\nu}; f_{J_{\sigma f}+J_\nu+1}, \ldots, f_{J_f}\right]^{\dagger} \triangleq \left[\boldsymbol{\sigma}_f; \boldsymbol{\nu}; \mathbf{N}\right]^{\dagger},\ J_f = J_{\sigma f} + J_\nu + J_n, \tag{A11}$$

where

$$\begin{aligned} &\boldsymbol{\sigma}_f \triangleq \left[\sigma_{f,i=1}^{1}, \sigma_{f,i=1}^{2}, \ldots, \sigma_{f,i=1}^{G}, \ldots, \sigma_{f,i}^{g}, \ldots, \sigma_{f,i=N_f}^{1}, \ldots, \sigma_{f,i=N_f}^{G}\right]^{\dagger} \triangleq \left[f_1, \ldots, f_{J_{\sigma f}}\right]^{\dagger}, \\ &i = 1, \ldots, N_f;\ g = 1, \ldots, G; J_{\sigma f} = G \times N_f, \end{aligned} \tag{A12}$$

$$\begin{aligned} &\boldsymbol{\nu} \triangleq \left[\nu_{i=1}^{1}, \nu_{i=1}^{2}, \ldots, \nu_{i=1}^{G}, \ldots, \nu_i^g, \ldots, \nu_{i=N_f}^{1}, \ldots, \nu_{i=N_f}^{G}\right]^{\dagger} \triangleq \left[f_{J_{\sigma f}+1}, \ldots, f_{J_{\sigma f}+J_\nu}\right]^{\dagger}, \\ &i = 1, \ldots, N_f;\ g = 1, \ldots, G; J_\nu = G \times N_f, \end{aligned} \tag{A13}$$

and where $\sigma_{f,i}^{g}$ denotes the microscopic fission cross section for isotope i and energy group g, ν_i^g denotes the average number of neutrons per fission for isotope i and energy group g, and N_f denotes the total number of fissionable isotopes. For the purposes of sensitivity analysis, the quantity ν_i^g, can be obtained by using the relation $\nu_{f,i}^g = (\nu\sigma)_{f,i}^g / \sigma_{f,i}^g$, where the isotopic fission cross sections $\sigma_{f,i}^g$ are available in data files for computing reaction rates.

The quantity χ^g in Equation (3) quantifies the material fission spectrum in energy group g, and is defined in PARTISN [9] as follows:

$$\chi^g \triangleq \frac{\sum_{i=1}^{N_f} \chi_i^g N_{i,m} \sum_{g'=1}^{G} \left(\nu\sigma_f\right)_i^{g'} f_i^{g'}}{\sum_{i=1}^{N_f} N_{i,m} \sum_{g'=1}^{G} \left(\nu\sigma_f\right)_i^{g'} f_i^{g'}}, \quad with \sum_{g=1}^{G} \chi_i^g = 1, \tag{A14}$$

where the quantity χ_i^g denotes the isotopic fission spectrum in energy group g, while the quantity f_i^g denotes the corresponding spectrum weighting function.

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