Reply on RC1:

Thank you for your constructive and insightful comments. We have carefully addressed all comments and suggestions. Below are our point-by-point responses to the reviewer's concerns.

MAIN COMMENTS:

Comment 1: The authors criticise previous RFari uncertainty attribution work for their lack of account for covariance between uncertain factors and their inability to quantify contributions to total uncertainty. But their literature review stops in 2018, and studies published since then have addressed many of the authors' concerns, including using methods very similar to their proposed CP method. Thorsen et al. (2020, 2021) https://doi.org/10.1175/Jcli-D-19-0669.1 https://doi.org/10.1175/Jcli-D-19-1009.1 and Elsey et al. (2024) https://doi.org/10.5194/acp-24-4065-2024 (and references therein) are particularly relevant. The authors need to place the motivation and results of their study in the current state of the art.

Response: We sincerely appreciate the reviewer's insightful comment. We agree that recent advancements in uncertainty analysis methods should be thoroughly discussed to better contextualize our work. In response, we have revised the **Introduction** section to incorporate the following key studies:

- Thorsen et al., 2020, 2021: These two studies decomposed the factor uncertainty contribution analysis by calculating the product of the radiation kernel and the factor observation uncertainty while discarding the less accurate aerosol characteristic data from satellite remote sensing observations and instead utilizing the more precise AERONET ground-based observation data. They improved the factor uncertainty contribution analysis in both methodology and data quality, a concept similar to the improvements proposed in this work, further demonstrating the advantages of the CP method.
- Elsey et al., 2024: This paper employs Monte Carlo simulation to analyze the impact of factor uncertainty disturbances on RF_{ari}, highlighting the contribution of factor uncertainty. This analytical approach more effectively captures the impact of interactions between factors on the results, similar to the CP method. Building on this approach, this work proposes a further discussion on sensitivity quantification and conducts an in-depth analysis of the advantages and differences between the Monte Carlo simulation and the OAT method results.

Compared to these studies, we provide a more comprehensive discussion of the quantitative analysis of the results. We have conducted detailed mathematical calculations on the factor sensitivity analysis results obtained using the CP method, explored the physical interpretation of the results, and examined the differences with the OAT method. Integrating the key analytical concepts from these studies has helped inform and strengthen the development of the CP method in this work.

Modification:

- New modifications and more information have been added to the Introduction section to describe the improvements in factor uncertainty contribution analysis methods in these studies (Page 2, lines 45-49 in the revised manuscript): "*Several studies have enhanced*

evaluation methods to improve the credibility of results. These improvements include replacing satellite remote sensing data with more accurate AERONET global ground-based observation data (Thorsen et al., 2021), and utilizing Monte Carlo simulations to assess the impact of disturbances due to factor uncertainty on system output (Lee et al., 2016; Elsey et al., 2024). Proposing a new method that integrates these improvements to overcome the limitations of OAT analysis could lead to a more accurate understanding of ARI."

- References to these studies have been incorporated into the Introduction section, which discusses the current state of research on sensitivity analysis methods (Page 2, lines 47, 48 in the revised manuscript)：
    1. Thorsen, T. J., Ferrare, R. A., Kato, S., and Winker, D. M.: Aerosol direct radiative effect sensitivity analysis, Journal of Climate, 33, 6119-6139, 2020.
    2. Thorsen, T. J., Winker, D. M., and Ferrare, R. A.: Uncertainty in observational estimates of the aerosol direct radiative effect and forcing, Journal of Climate, 34, 195-214, 2021.
    3. Elsey, J., Bellouin, N., and Ryder, C.: Sensitivity of global direct aerosol shortwave radiative forcing to uncertainties in aerosol optical properties, Atmospheric Chemistry and Physics, 24, 4065-4081, 2024.

Comment 2: The list of challenges in Section 2.2 is interesting, but it is often unclear why they really challenge uncertainty attribution. Challenges 1 and 5 are real issues but would pose a problem for any uncertainty attribution methodology, including CP. Challenges 2 and 3 are real, but temporal variations do not necessarily translate to variations in uncertainty contributions. Challenges 4 on covariances is real, but crucially depends on which factors are considered and the size of the covariances. So the relevance of each challenge to uncertainty attribution needs to be clarified by extending the discussion.

Response: We thank the reviewer for this constructive comment. We have substantially expanded Section 2.2 to explicitly link each challenge to its specific implications for uncertainty attribution, particularly highlighting how our CP method addresses or mitigates these issues. For each of the challenges listed, our responses are as follows:

- Challenge 1: For the OAT method, a central value that represents the overall data level is required for sensitivity analysis. When the number of observation samples is small or the instrument system error is large, the resulting data distribution may not follow a natural distribution. In such cases, the central value calculated using the statistical average may not accurately reflect the actual aerosol characteristics, meaning the A_{cal} value and the actual A_{real} value could differ significantly. As discussed in Section 3.3.2, even a small difference between these values can lead to substantial errors in the evaluation results, which is why this is considered a challenge. In contrast, the CP method proposed in this work uses the overall distribution of the input data, regardless of whether it follows a natural distribution. The sensitivity analysis results obtained with this method can capture the effects of the data distribution. The only limitation is the accuracy of the observations, and the method itself does not introduce additional uncertainty into the evaluation results. Therefore, this is one of the key improvements of the CP method over the OAT method.

We have revised the expression in this section to make the purpose clearer.

- Challenge 5: This challenge is indeed related to the limitations of the model itself and is not specifically tied to the OAT or CP methods. To improve clarity, we have removed this challenge from the discussion in Section 2.2.
- Challenges 2&3: These two challenges address the impact of temporal and spatial changes on observed aerosol characteristics data, which may exhibit trends. For the OAT method, the statistical average may not accurately represent the characteristics of such data, potentially introducing new sources of error. If trends in the data are not addressed, the OAT method may inadvertently introduce additional uncertainties, rather than the trends themselves. Since there was a duplication in the original manuscript regarding these two challenges, they have been merged, and the expressions have been modified for clarity.
- Challenge 4: Regarding the discussion of covariance in this challenge, the relative size of the covariance is indeed a crucial factor as it determines the interaction relationship between the factors. We have added this description to clarify the importance of covariance in the analysis of factor interactions.

Modification:

- Modified the text of Section 2.2 on the challenges of the OAT method in analyzing aerosol radiation interactions to highlight the advantages of the CP method (Pages 4-5, lines 119-144 in the revised manuscript)**:**
    (1) In aerosol observations, inaccuracies in certain instruments or uncertainties arising from the inversion process of the joint observation system can lead to significant uncertainties in the generated observation data. As a result, the statistical average of the measurement results may not accurately reflect the true properties of aerosols. This discrepancy means that there can be a considerable deviation between the calculated A value A_{cal} *and the actual A value* A_{real}*, resulting in obtained results that are not entirely accurate.*
    (2) Due to the influence of multiple factors, such as aerosol sources, aging processes, and changes in the atmospheric environment, it is essential to consider that the observed values of the physical and chemical properties of aerosols exhibit significant variability over time (e.g., diurnal, seasonal, and interannual changes) and space (e.g., coastal vs. inland, urban vs. rural, boundary layer vs. free atmosphere). This variability can lead to inhomogeneity in aerosol optical properties, resulting in trends or patterns in the observed data rather than a strict normal distribution. When using the OAT method for sensitivity analysis, the statistical mean may not accurately represent the data, thus introducing additional sources of error. To address this, it is necessary to first eliminate the influence of trends in the data. Overcoming this challenge requires analyzing the actual physical environment, which may involve additional time and computational costs.
    (3) For ARI, the interactions between factors are significant, and it is not feasible to assume that all the factors discussed are strictly independent. The covariance between the observed data may not be zero. When using the OAT method to analyze factor sensitivity, it is crucial to consider the relative magnitude of the covariance between the observed data of each factor. When the covariance between the data

is large, applying a disturbance to one factor while holding the others constant does not reflect the actual physical situation, and thus fails to satisfy the strict conditions required for the OAT method. As a result, the final analysis may contain errors in the factor interaction terms.

(4) *The outcome of sensitivity analysis is to ascertain the uncertainty contribution of various factors. It is essential to perform separate statistical analyses on the sensitivity of these factors and observational uncertainty. To address the challenge of high uncertainty in the evaluation results of* RF_{ari}*, a more practical discussion should focus on how constraints imposed by observations, or improvements in observational accuracy, can enhance the reliability of the results. While sensitivity analysis does not directly answer this question, it offers valuable insights into the significance of each factor from a different perspective, guiding future efforts to reduce uncertainty.*

Comment 3: Section 3.2 is incomplete and would not allow to reproduce the authors' calculations. First, it is unclear what the central values given in Table 1 are supposed to represent, and therefore which aerosols the uncertainty analysis is relevant to. A typical industrial aerosol? A typical Chinese aerosol? In addition, the uncertainty ranges (the sigmas) are missing for all parameters, but are obviously crucial to apply both the OAT and CP methods. They must be added to Table 1.

Response: We thank the reviewer for highlighting these critical suggestions. To address the concerns regarding reproducibility and clarity, we have thoroughly revised **Table 1** as follows:

1. Clarification of aerosol types and central values. The central values in Table 1 now explicitly represent **urban-industrial aerosols** observed in **East Asia**.
2. Addition of uncertainty ranges (σ). The σ is based on the combined result of random observation errors and systematic instrument errors.

Modification:

- Revised Table 1 with full uncertainty ranges and geographic/aerosol-type annotations (Page 10, lines 271-272 in the revised manuscript) as follows:

• **Properties**	**Values**	**Data Source**
CRI_{shell}	$(1.58 \pm 0.2) + 1e^{-3}j$	Peking University Observation Site
CRI_{LAC}	$(1.67 \pm 0.36) + (0.67 \pm 0.35)j$	Zhao et al., 2020
CRI_{EC}	$2.26 + 1.26j$	Taylor et al., 2015
CRI_{air}	$1 + 1e^{-3}j$	Zhao et al., 2020
ρ_{EC}	1.8	Bond and Bergstrom, 2006
ρ_{LAC}	0.95 ± 0.3	Zhao et al., 2019
$Kappa$	0.22 ± 0.2	Peking University

		Observation Site
MS	0.7 ± 0.3	Gong et al., 2016
$PNSD_{dry}$	/	Taizhou Observation Site
$LACPNSD$	/	Taizhou Observation Site
VD	/	Liu et al., 2009
RH	/	ERA5 Data
$albedo$	/	MODIS Data
Location Settings	116° W, 40° N	/
Date Settings	April 1	/
Time Settings	12:00 local time	/

Table.1 Data sources and SBDART environment parameter settings. The aerosol optical properties represent urban-industrial aerosols observed in East Asia.

OTHER COMMENTS:

Comment 1: Abstract, lines 11-12: Need to make the description of the CP method clearer. The current description could apply equally to the OAT method.

Response: We thank the reviewer for identifying this ambiguity. To better distinguish the CP method from traditional OAT approaches, we have revised the abstract as follows (Page 1, line 11 in revised manuscript): "*This method constrains the uncertainty of a single factor* ***between two Monte Carlo simulations*** *and evaluates its sensitivity by analyzing how this change affects output uncertainty.*"

Comment 2: Line 25: RFaci is missing from the list of strong radiative forcing. It is probably much stronger than RHari.

Response: In fact, the uncertainty associated with aerosol-cloud interactions is higher than that of aerosol-radiation interactions. In this study, we aim to obtain more precise aerosol characteristic data through laboratory observations to enable a more accurate quantitative analysis of the uncertainty contribution of RF_{ari}. However, due to the absence of direct observation methods for RF_{aci}, it is not addressed in this work. To avoid ambiguity, RF_{aci} is not discussed in the introduction.

Comment 3: Line 27: Do you mean "has *not* undergone substantial revisions"? Otherwise, the "Despite significant advances" does not work with the sentence.

Response: In this introduction, we aim to convey that although methods for evaluating the contribution of factor uncertainty are continually improving, significant uncertainties remain in the evaluation of aerosol-radiation interactions due to model limitations, observational data accuracy, and other factors. The IPCC has revised these results multiple times in previous assessment reports, with each revision showing substantial changes, often with high values and high uncertainty. This indicates that a consensus on this result has yet to be reached. This part has been revised in the

introduction to clarify the statement (Page 1, lines 26-28 in revised manuscript): "*Despite the estimated RF_{ari} has undergone substantial revisions across successive IPCC reports, and its uncertainty has not notably decreased (Houghton, 1996; Houghton, 2001; Solomon, 2007; Stocker, 2014; Forster et al., 2021).*"

Comment 4: Lines 28-31: Are those model-observation differences still actual? I think the latest IPCC report concluded those disagreements were mostly resolved now.
Response: There has been considerable work on the model assessment and actual observation of aerosol-radiation interactions; however, a significant discrepancy between the two persists. A typical example of this is the black carbon absorption enhancement phenomenon, where the surface of black carbon particles in the atmosphere is coated with a material, leading to an increased absorption capacity. Several factors contribute to this phenomenon, including the lens effect of the coating, particle morphology, and seasonal variations. Although most studies attribute the enhancement of black carbon absorption to the lens effect (Fuller et al., 1999, https://doi.org/10.1029/1998JD100069; Bond et al., 2006, ttps://doi.org/10.1080/02786820500421521), recent research suggests that the lens effect alone cannot fully explain the observed differences in black carbon absorption, indicating that further discussion is needed to address this issue (Fierce et al., 2016, https://doi.org/10.1038/ncomms12361; Gong et al., 2016, https://doi.org/10.5194/acp-16-5399-2016; Liu et al., 2017, https://doi.org/10.1038/ngeo2901; Zhao et al., 2021, https://doi.org/10.5194/acp-21-18055-2021; Zhai et al., 2022, https://doi.org/10.1021/acs.est.1c06180).

Comment 5: Line 25: The list of previous studies is missing recent work that already addresses the shortcomings of older work.
Response: Thank you for the comment. This has been addressed in response to main comment 1. Regarding the factor sensitivity analysis method, a description of the improvements made in the latest work has been added to the introduction (Page 2, lines 45-49 in the revised manuscript): "*Several studies have enhanced evaluation methods to improve the credibility of results. These improvements include replacing satellite remote sensing data with more accurate AERONET global ground-based observation data (Thorsen et al., 2021), and utilizing Monte Carlo simulations to assess the impact of disturbances due to factor uncertainty on system output (Lee et al., 2016; Elsey et al., 2024). Proposing a new method that integrates these improvements to overcome the limitations of OAT analysis could lead to a more accurate understanding of ARI.*"

Comment 6: Section 2.1 is difficult to understand because many variables are not defined: the exponent T, function g, A, the arrow operator, …
Response: Thank you for pointing out the omission in the description of the formula. Section 2.1 has now been revised to include detailed descriptions of all letters and symbols used.

Comment 7: Line 118: Why "linear trend" specifically?
Response: Thank you for your comment. We were not clear enough in our expression. We have revised the text to make it more accurate. What we intend to convey is that due to temporal and spatial variations, there may be trends (patterns) in the observation data of aerosol properties, which could lead to observed aerosol properties that do not align with their physical characteristics. When

applying the OAT method, it is necessary to use averaged aerosol property data. However, when using a dataset with a trend, simple statistical averaging may result in data that lacks practical significance. Therefore, before using the data, it is essential to process it to remove the impact of such trends. This point has been revised in the article to improve accuracy (Page 5, lines 124-132): "*Due to the influence of multiple factors, such as aerosol sources, aging processes, and changes in the atmospheric environment, it is essential to consider that the observed values of the physical and chemical properties of aerosols exhibit significant variability over time (e.g., diurnal, seasonal, and interannual changes) and space (e.g., coastal vs. inland, urban vs. rural, boundary layer vs. free atmosphere). This variability can lead to inhomogeneity in aerosol optical properties, resulting in trends or patterns in the observed data rather than a strict normal distribution. When using the OAT method for sensitivity analysis, the statistical mean may not accurately represent the data, thus introducing additional sources of error. To address this, it is necessary to first eliminate the influence of trends in the data. Overcoming this challenge requires analyzing the actual physical environment, which may involve additional time and computational costs.*"

Comment 8: Caption of Table 1: I suggest deleting "Observation" in the caption, since many data sources are either not from observations or not directly relevant to the observation site.
Response: Thank you for comment. The title of the table has been revised to remove the word "Observation." (Page 5, lines 271-272 in the revised manuscript): "*Table.1 Data sources and SBDART environment parameter settings. The aerosol optical properties represent urban-industrial aerosols observed in East Asia.*"

Comment 9: Lines 251-253: It would be interesting to also look at the sensitivity of RFari to AOD, SSA and g, to compare with similar studies.
Response: The sensitivity analysis of RF_{ari} to aerosol optical parameters, specifically AOD, SSA, and g, is crucial. As mentioned in the introduction, aerosol optical parameters are the most direct factors affecting RF_{ari} and serve as the primary inputs for most models. A correct analysis of this sensitivity will enhance our understanding of the impact of aerosol characteristics on the environment and climate. However, as noted in the introduction, significant work has already been done to evaluate this aspect, with many studies using the same radiation model, SBDART, employed in this work to assess the sensitivity of aerosol optical parameters to RF_{ari} (McComiskey et al., 2008; Loeb and Su, 2010). This study aims to focus on the quantitative ranking of uncertainty contributions from aerosol and environmental parameters to RF_{ari} based on their influence on aerosol optical parameters, an approach that is rarely explored. The goal is to identify the most sensitive factors affecting aerosol optical parameters and RF_{ari}, as well as factors whose importance may be underestimated, thereby aiding in the improvement of observations and model settings. Consequently, this study does not separately discuss the sensitivity analysis of AOD, SSA, and g to RF_{ari}.

Comment 10: Section 3.3.2 would work more logically before Section 3.3.1. Describe the physical meaning of the results before the OAT/CP differences. Overall the ranking of sensitivity follow expectations, given that Mie theory is used, but I am surprised that coating thickness is important to sensitivity, but kappa is not despite directly affecting aerosol size. Why is that?
Response: Thank you for the suggestion on the logical issues in the presentation of the results. To

improve the clarity of the article, we have rearranged the order of Sections 3.3.2 and 3.3.1. Regarding the concerns you raised about the calculation results, our findings show that kappa is more sensitive to g but less sensitive to AOD and SSA. The coating thickness parameter is highly sensitive to both SSA and g, but less sensitive to AOD. Kappa exhibits lower sensitivity to RF_{ari}, which can be explained by the following points:

(1) SSA is the aerosol optical parameter with the strongest sensitivity to RF_{ari} (Thorsen et al., 2020), while kappa is less sensitive to SSA and more responsive to the coating thickness factor, leading to the observed differences in sensitivity between the two to RF_{ari}.

(2) The effect of kappa on particle size is influenced by ambient humidity. In the environment set in this study (noon on the summer solstice in the Chinese background, using ERA5 reanalysis data), the boundary layer aerosol was in a medium-range relative humidity (50%–60%), which limited the hygroscopic growth of aerosol particle size. Compared to the direct effect of the coating thickness factor on particle size, the effect of kappa on particle size is somewhat diminished.

Reply on RC2:

Thank you for your constructive and insightful comment. We are grateful for your recognition of the CP method proposed in this work. We have carefully addressed all comments and suggestions. Below are our responses and modifications to the reviewer's concerns.

Response:

In this review, the reviewer expressed concerns about the data and results, suggesting a comparison with findings from other literature. This work aims to provide a more detailed assessment of the contribution of factor uncertainty to aerosol-radiation interactions through an in-depth factor analysis. Therefore, we utilize a large dataset of near-ground aerosol property observations, including the complex refractive index, hygroscopicity, density, and aerosol particle size distribution. All data are derived from field observations conducted by our research group in North China using self-built observational instrument systems, as described in Table 1 of the manuscript.

Field experiments to gather such extensive aerosol property data in the same region require considerable time and instrument resources. In contrast, previous studies or standard observation sites rarely provide sufficient data to match the scope of our work, which we identified during our literature and data research. As a result, we did not incorporate additional verified data into the experiments. While this limits the robustness of our results to some extent, as discussed in Section 3.1, our objective is to apply the CP method within the factor analysis framework we have developed and compare it with the OAT method to verify the characteristics of the CP method. As the reviewer noted, the results obtained in this study are not universally applicable but rather specific to the environmental settings in this work.

The results produced by the CP method depend critically on the performance of the model used. In other words, they are an evaluation of the model. The closer the model's description of real physical processes, the more accurate the results obtained by the CP method will be. With this in mind, we employed the Mie theory model to accurately describe the single-particle radiation process and the SBDART radiation transfer model to focus on the radiation transfer process, aiming to minimize model-induced uncertainty. The use of the Mie theory model also allows for some of the conclusions in this work to be directly verified, such as the identification of the aerosol complex refractive index and coating thickness as highly sensitive factors.

One important consideration is that the conclusions obtained by the CP method are also influenced by the distribution of the input aerosol property data. Using the same model framework with aerosol property data from typical urban pollution aerosols in regions outside North China for sensitivity analysis may yield significantly different results, as discussed in Section 2.3. In summary, obtaining universal conclusions requires more accurate models and representative, detailed aerosol property data. We plan to address these aspects in our future work. Additionally, we hope to use our CP method for a detailed factor uncertainty contribution analysis of the black carbon absorption enhancement phenomenon to resolve the current discrepancies between theory and observation regarding this phenomenon.

We will organize the above discussion and incorporate it into the **Conclusions and Discussions** section of the manuscript.

Modification:

In the Conclusions and Discussions section, add new paragraphs to answer concerns about the robustness of the conclusions reached in this work (Page 15, lines 398-406 in revised manuscript): "*The quantitative ranking results of factor uncertainty obtained in this study using the CP method are highly dependent on the model employed and the distribution of input aerosol optical property data. The more accurately the model describes the actual physical processes and the higher the precision of the aerosol optical property data, the closer the evaluation results from the CP method will align with observed values. Therefore, this study utilizes the Mie theoretical model and the SBDART radiation mechanism model, which provide a more accurate representation of the single-particle radiation process, along with field experimental data from direct observations of aerosol optical properties. This approach aims to minimize the uncertainty introduced by the model and data accuracy. Using different model frameworks or aerosol optical property data from polluted aerosols in cities outside of North China to conduct the same factor sensitivity analysis may yield significantly different results.*"