I thank the authors for their patience with my previous review, which was mostly incoherent due to a formatting problem. Prior to submitting that, I submitted a referee report at the initial stage of the review process. Perhaps I submitted that prematurely, and the authors may not have seen it. That report contained the bulk of my "General and Specific Comments." Both of the old reports are included in this document in case they can still be useful to the authors, but a line-by-line response is not requested, and many of the comments have already been addressed. I am very sorry to have caused the authors so much confusion.

#### June 2021 Referee Report

#### **General Comments**

The manuscript is improved, particularly by clarifying the language around key points throughout, and by adding content to the summary and conclusions. I think it is fit for publication as is (so long as the technical corrections are made).

#### **Specific Comments**

Section 1.2: Just a suggestion, but I think a hint at what strategy you will use for weighting ensemble members is appropriate here. The reader cannot tell whether you will do something ordinary or novel.

Line 117: Your argument against a Gaussian likelihood is still a little bit wanting, in my opinion. When you say that "We think that the whole measurement set should bring information" what you are really saying is that it is more important to learn from a larger count of measurements than it is to learn from the smaller count of the largest errors. Or that relative error is what matters. Those are not self-evidently true, to me. So I think you should provide some concise statement about the benefits of learning from a larger number of observations (less sensitive to outliers?), or just say that it is worth testing.

#### **Technical Corrections**

Line 10: "suitable" instead of suited Line 372: Correct typo and clarify what you mean Line 441: Indent and expand this paragraph? Or combine it with the following paragraphs.

#### Previous report (December 2020)

This paper does exactly what it says it will do: source term estimation for an emission of Ruthenium using observations, an atmospheric model, and Bayesian inversion. It excels at explaining the concepts involved, making it especially accessible to someone who has not done this exact type of problem before. However, many corrections are needed to the wording, particularly in section 2, and the organization, particularly in section 3.

#### Comments:

37. In the climate modeling community we would refer to an initial conditions ensemble of the same atmospheric model as a "single-model ensemble" rather than a "multi-model ensemble."

#### 39. Missing "a"

60-63. Comment: I like this concise explanation of how a model, source, observations, and likelihood fit together.

89. Suggested "These three sources of uncertainty are explored in an application of source term estimation for the  $^{106}$ Ru release..."

#### 90. state of the art of

89-94. Can you rephrase this so that it flows monotonically, i.e. reference section 2 before section 3?

103. The math is correct but the wording is not quite right. I think you mean that *r* is a positive coefficient and R (and *r*I) is a positive diagonal matrix; *r* itself is not a "positive diagonal coefficient."

#### 106. Of the problem

111-112. Suggest something like...choosing Gaussian likelihood penalizes the largest errors to an extent that smaller errors are negligible.

115. Rephrase. Consecutive sentences starting with "in other words."

116. I think you should delete the sentence starting with ``Every " as the wording is confusing. Your example (100,120) vs (10,12) has already made this point.

120. Bracket typo.

126. What do you mean by mitigated here? I think you can say "should be 1" or "should be close to 1."

131. Missing a word here, which obscures the meaning of the sentence.

144. It may be helpful for the reader if you reference the section in which the threshold is discussed.

190. If the observation sorting algorithm is the division into r and  $r_{nd}$ , then you should not start a new paragraph for sentence 191.

268, 274. "22<sup>nd</sup>"

305. This summary section should be clarified if possible. For uniformity, I recommend starting each bullet point with a section number, e.g.

- Section 3.3.2 is an application of the observation sorting algorithm...;
- Section 3.3.3 is an application of the different likelihood functions and spatial clustering ...;
- Section 3.3.4 is an application of the perturbed dispersion parameters and enhanced ensemble...

Secondly, the section heading "Summary" section seems out of place, especially since you have a summary section later. I would suggest renaming 3.3 "Results" and renaming 3.3.1 "Overview."

345. "Probable sources"

355. "which is not justifiable."

440. Explain when and where this accident took place, and maybe ad some thoughts about how this might compare to what you just did.

436-440. I think more discussion would be helpful for the reader. Remember, many readers skim the paper until they get to the conclusions.

## Initial Report (to determine whether manuscript should proceed to review and discussion stage)

Initial manuscript evaluation of JDLB et al., 2020.

I very much enjoyed reading this paper. The author excels at explaining the concepts involved, which is refreshing. For example, the intuition of how the likelihood functions affect posteriors, etc., is well-done. The results are presented clearly.

There are a handful of language errors that should be addressed before a preprint is posted on the web. I have documented some of them here but would strongly recommend another proofread. **Any comment that should addressed before preprint is in bold.** General comments more suited to a full review are also included, but those do not need to be addressed at this stage.

### Typo: "a an" -> an Language: "fruitful in the recent years."

61. Comment: I particularly like this explanation of how a model is used as a tool to calculate the likelihood.

Equation 2. Why the '...'?

## 105. Language: When I read "Inverse" I think of a matrix inverse or ^(-1). I believe the author means "negative" in this context. If so, this is very important to fix.

109. Comment: In the case where the observations and the predictions are all equal, is equation 4, the cost, actually equal to zero? To me it looks like the answer is no.

112. The cost of couple 1 is 100x larger than couple 2 only if couple 1 and couple 2 are assumed to have the same variance, so this isn't necessarily a property of the Gaussian distribution itself.

179-186. Comment: This paragraph is generally well-written and the concept is clear.

# 273. I do not understand the meaning of "impulsions" in this context. If this is a technical term then I apologize. If this is a language error, I think the author is referring to the daily average release rate, so "releases" or "emissions" would suffice.

298. The variances of these transition parameters is listed here, but what about the other parameter for the folded normal distribution? Is there not a "location parameter?" I cannot tell if that is specified somewhere. Perhaps I am misunderstanding.

Fig 4 Caption: I think this caption could be rewritten to be more clear, even if it takes more text.

Fig8. Comment: Remove the fill here so we can see more clearly?

Fig. B1. Is it possible to overlay where your ensembles fit on this plot? Otherwise it is a little eyebrow-raising to say that the criteria "totally justify" using this method, as you do on line 470. Also, what are the implications of appendix B for using three spatial clusters? This seems worth a comment.

474. Could you clarify what is a hit and what is a false occurrence? This may be obvious to some readers but not to me. Is a "hit" correctly identifying the bin of the observation?