Reply to Anonymous Referee #1

Dear Reviewer 1,

We would like to thank you for your review. We believe that your comments and suggestions will help us to improve our manuscript. Please find below a step-by-step reply to your comments and suggestions.

Yours faithfully, The authors

> The proposed manuscript presents important question regarding uncertainties associated with inverse modelling problems for unknown atmospheric releases. It is used ensemble approach which is applayed to the ruthenium 106 case from 2017. The study is interesting and clear to read, but more questions remain in conection with the selected case and its suitability for the cause. Although the goal of the paper is not to find the origin of the ruthenium in 2017, the estimated probability regions of the release far from the actual release site need more discussions. It is difficult to draw and to follow conclusions based on results inconsistent with state-of-the-art knowledge about the ruthenium case. Perhaps, wider ruthenium dataset or another case with known location and known release profile could be more appropriate for this type of study.

Specific comments:

Although there is dataset with hudreds positive measurements regarding Ru-106 case in 2017 available (Masson et al., 2019), the authors choose data from 5 locations with12 positive measurements. This is quite suprising and authors should comment this. Moreover, the choice of CTBT stations seems problematic in this case since the main activities have been observed around Mayak and then south-west-wind in Ukraine, Romania etc. (Masson et al., 2019). Hence, my opinion is that the used data can contain rather fractions of information about the release and the results are dominated by the fact that the release period is preselected in the algorithm. This is probably closely related also to the fact that the probable location, Mayak, is not estimated within the probability region in any case (in fact, Dimitrovgrad is much more probable in all cases). This should be discussed in the paper.

Reply:

There are several reasons for using only a small subset of all the available data:

1. Many observations have redundant information so that including these additional observations would not significantly affect the results.

2. If we would have used observations taken nearby the Mayak facility, indeed the source location could be much more confined. However, the meteorological data and the EDA ensemble that were used in this study are not appropriate for use on local scales, as the ensemble does not capture well local scale uncertainties.

3. In the CTBTO verification context, we typically have available sparse observations around the world at receptor locations from the International Monitoring System that are about O(1,000 km)

apart from each other. Hence, we are dealing with long-range inverse atmospheric transport problems. As this work was carried out in the context of the CTBT, we have chosen to use (a subset of) observations from the IMS.

4. Furthermore, our results are in agreement with other inverse long-range atmospheric transport modelling studies as it shows a broad region in the Southern Urals as possible source location (see for instance Fig 1 of Saunier et al 2019).

5. Another point to take into account is the computational effort associated to ensemble atmospheric transport modelling. If we would use 1,000 observations, this would require 51,000 Flexpart simulations if we assume that the source location is unknown.

We thank you for your comment since indeed we did not discuss these reasons in our original manuscript. We will add the above discussion in the revised manuscript.

p. 6, l. 131: The authors claimed that "the release rate is assumed constant during the release period". This assumption seems to be quite strong since the release rate may vary and, in this particular case of Ru-106 release, did vary during the time as estimated by e.g. (Saunier et al., 2019). Is this assumption necessary and what is the impact of it?

Reply:

It is important to distinguish between different geotemporal scales. While time-varying emissions can have a huge impact nearby the source, these effects are less significant further away from the source due to the atmospheric transport and dispersion processes (and the atmospheric transport model, which filters such information out). Hence, we expect a constant release within release parameters t_start and t_stop to be appropriate to describe the Ru-106 source. In the version of the FREARtool that is described here, the source is parameterised as a fixed release during a certain period. A more recent version of the FREARtool can also deal with time-varying emissions.

p. 6, l. 141: The authors assume that "the release is assumed to have occurred between 25 September 2017 0000 UTC and 28 September 2017 0000 UTC", however, the release was estimated before e.g. in (Saunier et al., 2019; Western et al., 2020). Could you, please, comment this choice?

Reply:

According to Figure 2 of Saunier et al 2019, the release that occurred between 25 and 28 September 2017 is about three orders of magnitude larger than the release outside that time period. Since our source parameterisation deals with a fixed release rate between a certain period, and we do not consider local observations which might be affected by smaller releases that potentially took place before 25 September, we believe it is appropriate to neglect smaller emissions that potentially occurred earlier (or later).

p. 6, l. 154: The Currie critical threshold, LC, is used extensively in the paper. Could you please briefly explain basics about this value?

Reply:

We will add to the revised manuscript:

"Currie (1968) defined a critical level L_C above which a net signal (which is the detected signal from which the effects of background radiation are subtracted) should be in order to declare the net signal to be a detection."

Note that, without statistical fluctuations, one could simply use $L_C = 0$, since any positive net signal would be a detection. However, as this is not the case, we set $L_C > 0$ in order to reduce the number of false positives.

p. 9, l. 203: oobs seems to be fixed in your scenario. Do you have uncertainties associated with measurements? How are these uncertainties related to this oobs value?

Reply:

 σ_{obs} is specific for each observation and is the measurement uncertainty reported by the International Monitoring System. However, there was an inaccuracy in L 203 of the original manuscript, where we wrote:

"The detected activity concentration c_det is assumed to be Gaussian distributed around the true activity concentration c_true , with a standard deviation $\sigma_{obs} = L_C / k_\alpha$ as in Eq. 14."

This is only valid in case of a true non-detection or a false alarm. We have rewritten that sentence as follows:

"The detected activity concentration c_det is assumed to be Gaussian distributed around the true activity concentration c_true, with a standard deviation σ_{obs} equal to the reported observation error."

The latter makes it also more clear that σ _obs is not a fixed value.

Technical corrections:

It is not necessary to have new paragraph after each equation.

Reply:

Thank you for pointing this out.

The abraviation FREAR(in title) is not used and define in the manuscript.

Reply:

Thank you for pointing this out. We will add to the revised manuscript: "FREAR stands for Forensic Radionuclide Event Analysis and Reconstruction"

p. 8, l. 189: consider to remove"used".

Reply:

Thank you for your suggestion.

p. 10, l. 219: there is no si in Eq. (13), please, clarify.

Reply:

Thank you for pointing this out. Eq. (13) is simply Eq. (5) but with c_true replaced by c_det,i. We will also add a reference to Eq. (5) in the manuscript for clarity.

Sec. 4.1: you should specify that this is related to the Fig. 3, LEFT

Reply:

Indeed, thank you for this remark. We will specify this in the revised manuscript.