Author's response to the editor remarks on version 3 of GI-2021-28

Knut Ola Dølven[1], Juha Vierinen[2], Roberto Grilli[3], Jack Triest[4], and Bénédicte Ferré[1]

[1]Centre for Arctic Gas Hydrate, Environment, and Climate,UiT The Arctic University of Norway, Tromsø, Norway
[2]Institute for Physics and Technology, UiT The Arctic University of Norway, Tromsø, Norway
[3]CNRS, University of Grenoble Alpes, IRD, Grenoble INP, 38000 Grenoble, France
[4]4H-JENA engineering GmbH Wischhofstrasse 1-3, 24148 Kiel, Germany

Correspondence: Knut Ola Dølven (knut.o.dolven@uit.no)

We would again like to express our thanks for the impeccable handling and review of our manuscript by the editor and his team and complement the comments which have really helped improving this manuscript. We hope we have interpreted the comments correctly and addressed them appropriately herein and in the revised version of the manuscript.

Text from "An editor's review" of version 3 of GI-2021-28 are in grey italic font and our responses are in black normal font. Text added to the manuscript is written in emboldened font and are in quotation marks. Line numbers referred to in this document might be slightly off due to changes in document length in the review process.

1 Minor comments

- *L. 107: in the in-line equation of γ, one representative σ is used but how it is obtained from σ_j is not explained (although I would guess the largest of σ_j).* To ensure that γ is large, we have used the smallest σ_j. The reason for this is that artifacts more likely occur when decreasing γ compared to increasing it by a similar amount. See also reply to the major comment below. We changed the text inside the paranthesis to **"we used $\gamma = 2\Delta t \cdot 10^5 \sigma_s^{-1}$, where σ_s is the smallest σ in m'"**

- *L. 145: Eq. (13) is identical with Eq. (3). Avoid repeating the same equation to appear in different places with different numbers (this is not a lengthy paper) but just mention here "The simulated measurements m_j are obtained according to Eq. (3)" or alike.* We removed the equation and changed the sentence introducing the equation to: **"A simulated set of measurements m_j was then obtained by sampling from $u_m(t)$ and adding Gaussian noise $\xi_j \sim \mathcal{N}(0, \sigma_j^2)$ (see Eq. 3"**.

- *L. 182-183: "The error estimate is obtained from Eq. 11" needs to be explained more explicitly. My understanding is that the diagonal elements of the upper left quadrant (N x N) of Σ_{MAP} (the covariance matrix) are used, right? Explain in the text.* That's right. We have added an appendix showing the Σ_{MAP} of from the field experiment - see response to major comment below. We also changed the referenced sentence to: **"The error estimate, given as a 95% confidence interval (2σ uncertainty), is obtained from the the diagonal terms of the upper left quadrant of the covariance matrix (the quadrant concerning $[a, a]$, see Eq. 11) using the simulated measurement uncertainties as input variance (which**

are proportional to $u_m(t)$ except an offset σ_0, Eq. 13)." We also expanded on the sentence following Eq. 11 (∼L. 130) such that it now reads: **"This essentially achieves a mapping of data errors into model errors, including the prior assumption of smoothness."**

– *L. 184 add "except an offset σ_0" after "proportional to $u_m(t)$"* Yes, added this.

– *L. 232 not very clear why re-definition of s_j is needed instead of using that in Eq. (14). Please clarify.* Added and re-wrote the middle part of this section to make the reasoning behind this clearer. It is unnecessary to use the wide error margins provided in the sensor data sheet (which takes into account a wide range of potential measurement errors) in a controlled environment with a freshly calibrated sensor. Furthermore, it illustrates the possibility of calculating sensor noise directly from the sensor signal. The passage now reads (included the sentence prior to and after the changes for context): **"The RT of the sensor was determined to 23 minutes at 22°C (k=0.000725s^{-1}) prior to the experiment following standard calibration procedure of the sensor, and parameters affecting membrane permeation was controlled (i.e. water flow rate over membrane surface and water temperature). Running a calibrated sensor in a controlled environment assuming random errors, we chose to use signal noise as basis for measurement uncertainty (rather than as stated in the instrument data sheet). This was estimated using the standard deviation of the single point finite differences in the measurement data, calculated as**

$$\sigma_{j=1,2,3,\ldots M} = \sqrt{\frac{1}{M-1}\sum_{j=1}^{M-1}(m_{j+1}-m_j-\mu)^2}, \quad \text{where} \quad \mu = \frac{m_{M-1}-m_1}{M-1} \tag{1}$$

The first 2 hours had lower noise due to the reduced sampling rate and hence longer internal averaging period, but noise was otherwise constant and unrelated to the measured concentration. We therefore used two separate σs (measurement uncertainty); one for the first 2 hours and one for the latter part of the measurement period. Δt was determined to 179 s using the automatic Δt selection based on the L-curve criterion (see Appendix)."

– *L. 232 I believe this (equation about μ) reduces to $\mu = \frac{1}{M-1}(m_M - m_1)$* It indeed does - we simplified accordingly.

– *L. 246 "with process 2 inverted" may not be appropriate as processes 2 and 3 are no longer discernible (or, in other word, the second derivative of the curve never becomes null) in the step decrease phases after ∼2 x 104 s (Fig. 4c). Better rewording this.* Agree. We changed the last sentence in this paragraph to: **"The step decreases behave consistent with the step increases, although process 2 and 3 becomes indiscernible since they both act towards decreasing the concentration."**

2 Major comment

L. 313-326 and Figure 6: I have one major concern here about the uncertainties estimated for $u_a(t)$ displayed in Fig. 6d. As the authors noted that "lower for increasing concentrations … at 00:15", the behavior of uncertainties in Fig. 6d (almost *proportional to instantaneous values of "already convolved" measurements, m_j, in Fig. 6a) is far from one would expect. Since the response time of the EB sensor is longer than 2000 s (as in L. 265-266, for the temperature range of field experiment), reconstruction of $u_a(t)$ at a time actually refers all subsequent measurements in next thousands of seconds (wouldn't this appear in the covariance matrix?). How come UPs and DOWNs of $u_a(t)$ in last 10 or so minutes (after ∼07:15) be reconstructed this good as in Fig. 6a? This also implies that uncertainties of the reconstructed signals should behave like weighted integrals of the measurement uncertainties in the same length of time. In addition, the RT-corrected EB data deviate from the DRB sensor measurements by the order of tens of ppm at many occasions while the estimated errors of the former are mostly smaller than 3 ppm except 00:15 – 00:45 after the largest peaks of methane concentrations. To my eyes, attributing ALL of these discrepancies to the different characteristics of the EB sensor and the DRB sensor would not be appropriate but (at least) some should be to suspected under-estimate of the uncertainties of $u_a(t)$. Although I'm not 100 % sure how this could happen, one possible area may be the authors' choice of numerically large weighting constant (γ) for the growth-law equation, the upper part in Eq. (6). It seems that a factor 10^5 in γ has rather arbitrarily been chosen. What if this factor is much smaller, say 10^3, 10^2 or even unity? I'm very curious whether or not smaller factors could yield "more understandable" behavior of uncertainties for $u_a(t)$ which may make me (and readers) feel probably more comfortable.*

Firstly we would like to comment on that we discovered an error in Figure 6d during the work with this comment. The errors shown in this figure did not previously show 2 standard deviations but only 1 standard deviation. This is now fixed (the error bars and errors reported in the text should be correct). This is of course relevant to the comment above and this discussion in general.

We believe there are several concerns here and we will try to respond to these one by one.

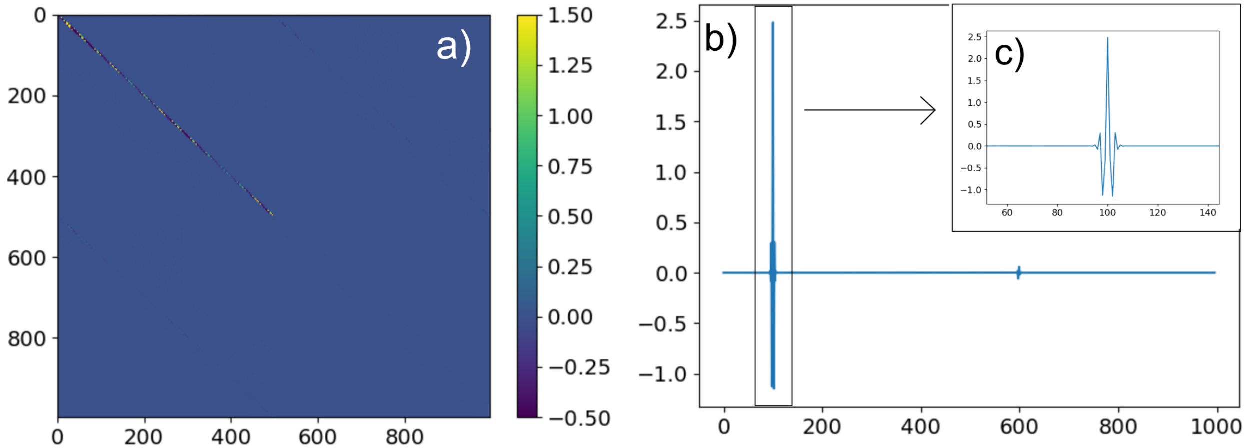
1. The errors we model in our method is confined to the errors in measured concentration within the measuring chamber. This accuracy is given as a percentage (with an offset) of the measured concentration within the measuring chamber. We therefore expect that the model errors to follow and have the same smoothened characteristics as the measured concentration within the measuring chamber (the untreated EB data). To be a bit clearer on this, we added a sentence in the last paragraph of section 4.1 that reads **"It is worth noting that this uncertainty regards the gas detection occurring within the measurement chamber and not potential errors caused by other factors such as imperfections with the e.g. membrane or pump operation. "**

   As the editor suggests, there are indeed non-diagonal elements in the covariance matrix and to a relatively small degree some temporal relationship within model errors. Figure D1 show the covariance matrix from the field experiment (Σ_{MAP} as calculated by Eq. 11 in the manuscript). The upper left quadrant of Figure D1a) show the $(\boldsymbol{a}, \boldsymbol{a})$ covariance and the diagonal elements gives the model uncertainty estimates. The fluctuations on each side of the diagonal elements (non-diagonal elements, see Figure D1b and c) show that there is auto-correlation in the error estimates of $\boldsymbol{a}$. This is expected since the elements in $\boldsymbol{a}$ are constructed from a set of linear combinations of the elements of $\boldsymbol{m'}$ (and $\boldsymbol{G}$, see e.g. chapter 2 of Aster et al., (2018)). In our case, the auto-correlation is quite narrowly confined in time, indicating a relatively short impulse response on our estimate. To explain this (despite a RT of τ_{63} >2000 s), consider that even though the response time is slow, the effect of a change in the property represented by $\boldsymbol{a}$ is still in principle recorded at the immediate subsequent measurement time-step by producing a change in the shape of $\boldsymbol{m'}$ at that point in time. Knowing the forward operator (the growth-law) makes it possible to interpret this accordingly. Practically speaking, the auto-correlation in the covariance matrix can be viewed as occurring due to the uncertainty in the exact point where the model interprets changes in $\boldsymbol{m'}$ to be a factual change in the property represented by $\boldsymbol{a}$, rather than just caused by a random error. The large increase in estimated uncertainty at each side of the time-series due to lack of measurements (on each side of the time-series) also illustrates the length of the uncertainty estimate impulse response (being relatively short compared to the total time-series).

We have extended and modified the sentence describing how we calculated the uncertainty of the RT-corrected signal in the field experiment (L.314) to read: **"Using the framework of inverse theory allows us to model error behavior by calculating the covariance matrix Σ_{MAP} (see Eq. 11 and Appendix D),"** We also added an appendix (Appendix D) on the covariance matrix from the field experiment showing Figure 1 and a short, slightly modified version of the text above: **"Figure 1 show the covariance matrix from the field experiment (Σ_{MAP} as calculated by Eq. 11). The upper left quadrant of Figure 1 concerns the $[\boldsymbol{a}, \boldsymbol{a}]$ covariances and the diagonal elements are the model uncertainty estimates. The other quadrants concerns the $[\boldsymbol{a}, \boldsymbol{u}]$, $[\boldsymbol{u}, \boldsymbol{a}]$, and $[\boldsymbol{u}, \boldsymbol{u}]$ covariances. Note that the color-scale in Figure 1 are set to be small to increase the contrast of the plot. The fluctuations on each side of the diagonal elements (non-diagonal elements) in row 100 (see Figure 1b and c) show that there is auto-correlation in the error estimates of $\boldsymbol{a}$. This is expected since the elements in $\boldsymbol{a}$ are constructed from a set of linear combinations of the elements of $\boldsymbol{m'}$ and also, being well confined in time, of little concern (see e.g. Aster et al., (2018))."**

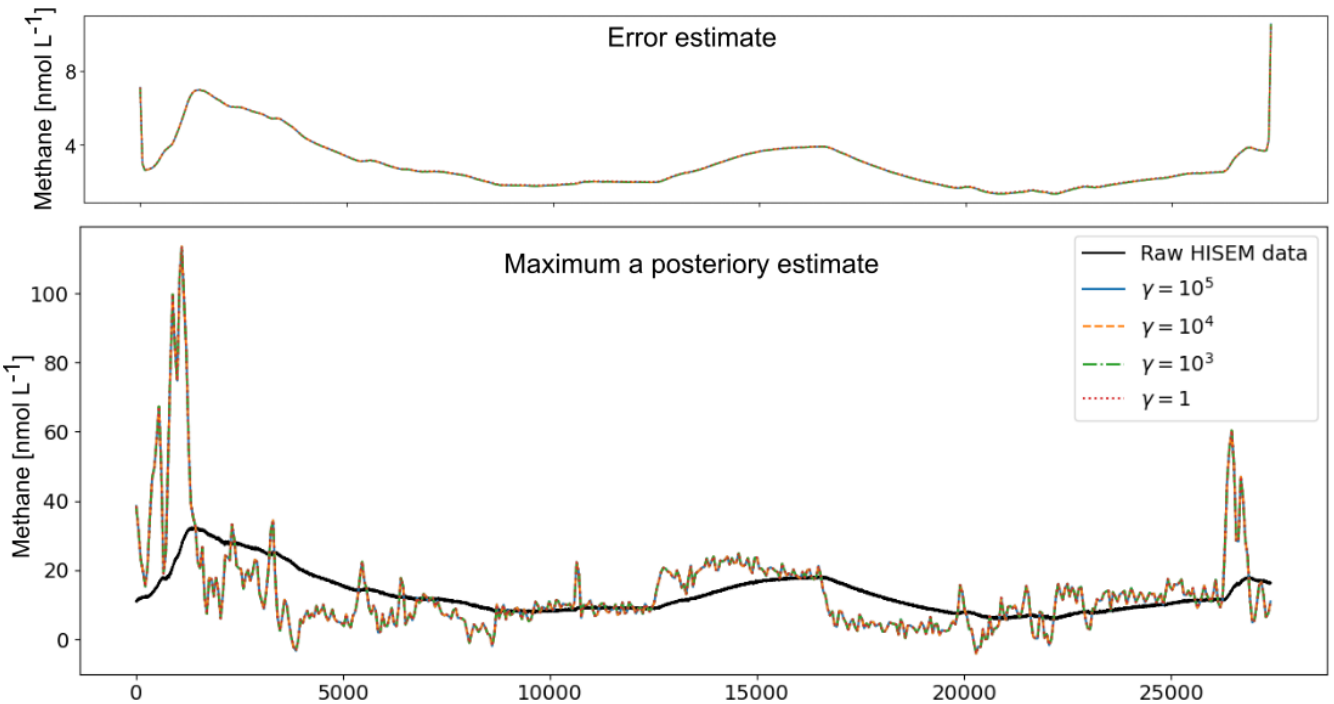
2. We are very glad the editor pointed this out, since there was indeed a factor 2 error in Figure 6d. This is now fixed. Other than that, the difference between the RT-corrected EB data and DRB sensor exceeds the error margins 8% of the time (this is pointed out in the manuscript) and this should be correct. The reason for this can be many, e.g. imperfections in one or both of the sensors or associated components (e.g. pumps). We comment on this in L. 315 in the manuscript, but we believe our addition in the last paragraph of section 4.1 presented above (in 1.) should help here as well.

3. The choice of weighing constant γ can be chosen in this manner because the estimates are very insensitive to the choice of γ. This is illustrated by Figure 2 which show the reconstruction and error estimate of RT-corrected field experiment data (essentially similar to Figure 6 in the manuscript) using a wide range of γ values. Even though the result is very insensitive to γ, this weighing is crucial to safeguard against violation of our theory assumption the growth law equation (which would essentially render the result worthless). Without the γ constant, the least squares solution might end up prioritize a perfect mapping of u_m to m' rather than fully adhere to the growth-law. In practice, this constant cannot be too large, because the least squares solver will not work if the constant is too large. It also must not be too small, because we then allow large errors in the growth law equation solution. However, as long as γ is large (relatively speaking) and to some degree balanced by the measurement uncertainty (in our case we define $\gamma = 2\Delta t 10^5 \sigma^{-1}$, i.e. large uncertainty in measurements allows larger residuals in the rows with γ in $\boldsymbol{G}$, see Eq. 6 in the manuscript) and the least squares solver



AR 1. a) Covariance matrix from response time correction of field experiment data. The $[\boldsymbol{a}, \boldsymbol{a}]$ covariance is shown in upper left quadrant and the lower left and upper right quadrants with very faint but visible diagonals show the $[\boldsymbol{a}, \boldsymbol{u}]$ and $[\boldsymbol{u}, \boldsymbol{a}]$ covariance matrices b) Row 100 of the covariance matrix and c) zoomed in excerpt of row 100 of the covariance matrix (approximate region indicated by the square in c).)

works, the matrix equation should be appropriately weighted. We added a sentence in ∼L. 110 (where γ is discussed) that reads: **"Estimates are insensitive to the exact choice of γ as long as it is large enough to disallow large errors in the growth law equation solution (which is why it is balanced by σ)."**



AR 2. Concentration and error estimates done with different γ values. The dashed/different colored lines (blue, orange, green, and pink) are overlapping, creating a mixed colored graph.

References

Aster, R. C., Borchers, B., and Thurber, C. H.: Parameter estimation and inverse problems, Elsevier, third edn., 2019.