High-resolution inverse modelling of European CH4 emissions using novel FLEXPART-COSMO TM5 4DVAR inverse modelling system

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## Overview

This paper describes an improved, higher resolution, inversion technique, FLEXVAR and compares it to three other related inversion systems. The method is applied to estimating CH4 emissions over Europe. Many sensitivity studies are presented, exploring various setup parameter choices that need to be made. The paper is well written and is very thorough. The presentation of the figures could be improved I believe, they contain too much cramped information leading to a lack of clarity. I have made several scientific points and questions and some minor text corrections.

## Comments

P2 L42: "of the derived inversion increments" - please clarify what this means

P6 L189: "ignores any error correlation between different observations" – Please briefly discuss the impact of making this assumption between different 3hr periods. It is highly likely that a measurement is strongly correlated with the 3hr measurements either side. How can this effect be minimised? Please explain why this assumption is necessary.

P6 L196 Eq10: Please clarify the equation with the use of brackets especially where items are divided. Please also provide the units of e (I assumed g/m2/s), it would be useful to have this stated.

P10 L301: "The matrix was scaled" – Please be more specific, which matrix? I assumed **B** the error covariance matrix? Fixing this to 20%, how did this compare to the other inversion setups?

P10 L312: "using 25 vertical layers" – I assume these are concentrated near to the surface? Please add a brief sentence describing this selection.

P10 L319: Please describe the rational behind the choice of the temporal correlation time scales.

P10 L321: "function of local emissions" - Are these the prior emissions? What distance is 'local'?

P10 L323 – 325: This last sentence seems out of place to me. How does it relate to the actual "TM5-4DVAR" inversion being discussed in this section?

P12 L369: Please specify which stations are classed as "mountain stations" as some are obvious others are less so e.g. Ochsenkopf, Beromunster etc? Also please describe which stations have the >200m difference between the model and actual orography imposed and what these release heights actually are, maybe simply add extra columns in Table 1 describing this height and class of station.

P12 Tab1: Why are Tacolneston 100m data used? In 2018, the 185m inlet samples much more frequently and is obviously higher and better able to be simulated?

P13 L373: "measurement uncertainty is set to 3 ppb" – How has this been derived? Most if not all observations come with an understanding of this quantity and this can vary between sites and over time. For instance why not use the variability in the CH4 observations across the 3hr period, the data are reported at up to 1 minute resolution? Also there are repeated measurements against standards, the repeatability of these observations also indicate how uncertain the measurements are.

P13 L391: "Natural CH<sub>4</sub> emissions were generally used" – When were these not used? The word 'generally' implies that in some instance they were not used, when they are not used, what was used? Table 2 implies they are always used.

P15 L415: "Offshore emissions over the sea are not included in the country totals" – Please explain the impact of this decision? The UK, Netherlands, Norway have significant emissions offshore in the North Sea. In the prior inventories how significant are these, and how does this impact on the conclusions that the UK+Ire totals are similar to what is reported given that the reported totals include these emissions?

P16 L455: Figure 5 is referred to here but is not shown until page 24. I think it should appear earlier in the document.

P20 L530: Maybe a similar comment also could be made about NW France which always appears to have very enhanced emissions but is relatively far from emissions? Or do you think these are real?

P22 L575: "INV-E1-O2 compared to INV-E1-O1" – Please provide the actual values for both simulations as well, along with the +- uncertainties, Figure 5 is too crowded to really extract values.

P24 Fig5: These figures are just too crowded and the different colours are impossible to discern e.g. inv-E1-O1-S2.1 and inv-E1-O1-S1 are indistinguishable. There is just too much information on each plot. On the RHS plots why are the E1 data repeated multiple times? The text for the range lines are blurred onto the lines. Please can this plot be improved?

P25 L606: Please mention that Fig6 resolution has been downgraded compared to earlier, I assume to match TM5?

P25 L613: Please provide the actual emission numbers rather than just the % change.

P27 Fig7: Similar comment to Fig5, it is hard to read the words/numbers in the RH plots, the plots themselves obscure the letters. Please can these be made clearer in some way? Although it is useful to see different inversions compared.

P29 L688: Summarising the results for each country grouping in a table would be very useful here.

P29 L710: "emissions in September (Fig. 5)." - I found this impossible to see as there are too many lines.

## **Minor Text Comments**

- P1 L35: add "CH4" emissions
- P2 L55: "effective radiate forcing (ERF)" should be effective radiative forcing
- P2 L56: "preindustrial levels 1750" preindustrial levels in 1750
- P2 L60: "especially on the near-term" especially in the near-term
- P2 L61: "due to the relatively short" due to CH4's relatively short
- P2 L70: "is particularly challenging" consider removing the word "challenging", when compared to CO2
- P3 L86: "which became available" which have become available
- P3 L94: "system is currently developed" system has been developed

- P4 L105: "As alternative" As an alternative
- P4 L106: "applied also the" also applied the
- P4 L111: "which allows to optimize a much" which allows the optimization of a much
- P4 L114: "emissions of individual" emissions from individual
- P5 L155: "allows to optimize emissions" allows the optimization of emissions
- P8 L254: "as function of" insert an "a"
- P10 L306: "which allows to zoom" replace with 'allows the system to zoom'
- P10 L307: "-18°...  $42^{\circ}$ " -18° to  $42^{\circ}$
- P10 L308: "while the global domain" while the remaining global domain
- P13 L375: "as observational base data set" "as the observational base data set"
- P22 L555: "in the country region" "in the prior country region"
- P22 L582: "the emission data" "the prior emission data"
- P24 Fig5 Caption: "and error bars the 2-sigma" "and the error bars are the 2-sigma"
- P25 L601: "Similar as for observation data set O1" "In a similar way, as shown with observation data set O1"
- P25 L608: "of major" "of the major"
- P28 L654: "is only relatively" "is relatively"
- P28 L658: "emission data set E3" "prior emission data set E3"
- P28 L676: "requires to include estimates" "requires the inclusion of estimates"
- P30 L714: "that it allows to constrain a" "that it constrains a"

P30 L718: "emissions 2018 using 24 stations" - "emissions in 2018 using 24 stations"

P30 L725: "derive over large parts of the domain somewhat" – "derive, over large parts of the domain, somewhat"

P30 L733: "in the surroundings of these sites" - "in the vicinity of these sites"

P31 L745: "allow to better reproduce the observations than the" – "allow a better the reproduction of the observations compared to the"

P31 L747: "freedom to optimize" - "freedom used to optimize"

P31 L748: "differences of the inversions, as e.g., the" - " differences in the inversions, e.g., the"