<u>Response to the Anonymous Referee #2</u>

Ref.: GMD-2023-9 | Model evaluation paper

Inverse Modeling of CH_4 emissions over Europe, Part I: Forward Modeling Evaluation against Near-Surface and Satellite Data

First of all, we want to thank the anonymous referee for the appropriate and constructive comments which helped to clarify some points regarding methodological descriptions on the study period definition, model sensitivity tests and how emission fluxes are converted into atmospheric concentrations. In order to facilitate the identification of actions for each reviewer's comments, we copied the comments in *italics* and set the answers in **bold**. All the manuscript modifications are indicated in **blue**.

<u>General comment:</u>

The manuscript describes the first part of the AUMIA system, which focuses on the forward modelling with WRF-GHG and its evaluation using TROPOMI and ICOS observations. The major concern is that without the inverse modelling part of the work, this first paper does not include much of a model development but focuses on forward modelling evaluation. In addition, there are several methodological descriptions missing, that should be clarified, that I listed below. Other than these aspects, the manuscript is well-written and easy to follow and understand. However, before being suitable for publishing in GMD, the below comments need to be addressed and implemented.

We understand the reviewer's concern that without the inverse modeling part not much of a model development has been exhibited so far, especially considering that the

inversion part by itself captures much more attention than forward simulations; however, we do think that gradual dissemination of findings and contributions can establish a foundation for subsequent parts. Submitting a forward modeling evaluation paper first will allow us to receive timely feedback and suggestions that can inform any necessary improvements or modifications that need to be made before delving into further aspects of the backward modeling part (e.g. by identifying the problem with not including the top layer in the integration, which the inversion part would likely just have solved by adjusting the state vector). Additional methodological descriptions on the study period definition, model sensitivity tests and how emission fluxes are converted into atmospheric concentrations, have now been included. Also, a better model description on how CH_4 emissions from biogenic sources are calculated, has been accomplished (RC1 suggestion).

Specific comments:

Lines 34-36. This last sentence sounds like an overstatement as there are previous studies using TROPOMI observations.

The first part of the sentence "The results found in this study contribute with a new model evaluation of methane concentrations over Europe" is true in the sense that new EDGAR emission estimates (Ferrario et al., 2021) together with improved TROPOMI observations (Lorente et al., 2022) were used to evaluate the methane concentrations over Europe. Regarding the second part "and demonstrate a huge and under explored potential for methane inverse modeling using improved TROPOMI products in large-scale applications", it can be supported by the domain-wide statistical metrics which are in line with previous studies conducted over specific locations in Central Europe (e.g. Zhao et al., 2019, 2022; Tsuruta et al., 2023). The more satellite observations are

improved, the more accurate the inversion estimates tend to be. Anyway, the term "and under explored" has been removed to not insinuate the TROPOMI data we used here have been available for so long and that even so it's been under explored. Also, the proper EDGARv6.0 reference is now included in the manuscript.

"Anthropogenic fluxes of CH_4 (not including biomass burning sources) are externally prepared based on the Emissions Database for Global Atmospheric Research (EDGAR) version 6 Greenhouse Gas Emissions (Crippa et al., 2021). EDGAR has been widely used..." ➔ "Anthropogenic fluxes of CH_4 (not including biomass burning sources) are externally prepared based on the Emissions Database for Global Atmospheric Research (EDGAR) version 6 Greenhouse Gas Emissions (Ferrario et al., 2021). EDGAR has been widely used..."

Ferrario, M. et al.: EDGAR v6.0 Greenhouse Gas Emissions. European Commision, Joint Research Centre (JCR) [Dataset] PID: http://data.europa.eu/89h/97a67d67-c62e-4826-b873-9d972c4f670b

Lorente, A. et al.: Evaluation of the methane full-physics retrieval applied to TROPOMI ocean sun glint measurements, Atmos. Meas. Tech., 15, 6585-6603, 2022.

Tsuruta, A. et al.: CH_4 Fluxes Derived from Assimilation of TROPOMI XCH_4 in CarbonTracker Europe-CH_4: Evaluation of Seasonality and Spatial Distribution in the Northern High Latitudes, Remote Sensing, 15, 1620, doi:10.3390/rs15061620, 2023.

Zhao, X. et al.: Understanding greenhouse gas (GHG) column concentrations in Munich using WRF, Atmos. Chem. Phys., doi:10.5194/acp-2022-281, 2022.

Zhao, X. et al.: Analysis of total column CO_2 and CH_4 measurements in Berlin with WRF-GHG, Atmos. Chem. Phys., 19, 11279-11302, 2019.

Line 98-99: How are these periods selected? This should be better described.

The main criteria for selecting the two-week periods for model sensitivity tests was to have at least 75% of days with TROPOMI data covering large portions of Europe. Large numbers of observation/model pairs (that spread out across the modeling domain) allow to perform a more representative domain-wide statistical evaluation. The one-year period from April 01, 2018 to March 31, 2019 was selected because of the following reasons: 1) availability of TROPOMI operational data and the improved TROPOMI data from March 2018 onwards; 2) evaluate the most recent EDGARv6.0 emissions for methane (2018); and 3) avoid sustained irregular scenarios in terms of emissions, e.g., fire outbreaks in most part of 2019 and emission reductions associated with COVID-19 lockdowns in 2020 and 2021, both at global scale. The 2018 summer was particularly interesting to focus on because of the larger than average number of cloud-free days over Denmark and most part of Europe. Section 2.3 Experimental design was rewritten and now it includes the criteria for selecting the study periods for the model sensitivity tests

"Initially, a model sensitivity analysis for evaluation of model parameterizations such as planetary boundary layer and cumulus clouds, as well as global forcings for CH_4 concentration, was carried out over several two-week periods in 2018 and 2019. Then, based on the model configuration that best fit the satellite data, one-year simulation period from..." ➔ "Initially, a model sensitivity analysis for evaluating physics schemes such as planetary boundary layer and cumulus clouds, and global forcings for meteorological fields and CH_4 concentration, was carried out over several two-week periods in 2018 and 2019. Each of these two-week periods were previously examined to have at least 75% of days with TROPOMI XCH_4 data covering large portions of

Europe. As a result, the physics schemes Yonsei University (YSU) for planetary boundary layer and Kain-Fritsch scheme for cumulus clouds, together with initial and boundary conditions from the European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis v5 (ERA5) model (Hersbach et al., 2020), for meteorological processes, and from the NCAR Community Atmosphere Model with Chemistry (CAM-chem) (Lamarque et al., 2012; Emmons, et al., 2020), for background concentrations of CH_4, were selected and then used to perform a one-year simulation period from…"

And for the one-year simulation period

"This period was defined based on the following criteria: i) availability of TROPOMI XCH_4 data, ii) latest available year data of EDGARv6.0 emissions for CH_4, and iii) no occurrence of sustained and irregular scenarios in terms of emissions (e.g., large-scale fire outbreaks and emission reductions associated with COVID-19 lockdowns)."

Line 119: What are these flux models and how do they work? More information is needed here.

Basically, since each gas occupies the same volume under the same atmospheric pressure and temperature, all gas species can be converted from mol/km^2/h to Δ[ppmv] (response to changes in pressure and temperature) using the same approach. Mathematically, the two-dimensional flux variable of a gas specie (emis_ant) is multiplied by a conversion factor (conv_rho) and then added to the first layer of the three-dimensional tracer variable of that gas specie (chem).

chem = chem + conv_rho*emis_ant

conv_rho = 8.0461e-6*(1/rho_phy*dtstep/dz8w)

where rho_phy, dtstep and dz8w denote the air density [kg/m^3], model time step [s] and the thickness of the first model layer [m], respectively. 8.0461e-6 is the molar mass of air per second [g/mol/s]. The file module_ghg_fluxes.F in /chem contains all subroutines for adding the emissions of CH_4, CO_2, and CO calculated per time step to the corresponding atmospheric concentrations. An additional sentence on how emission fluxes are converted into atmospheric concentrations is now included in the manuscript.

"...are converted into atmospheric concentrations based on flux models. On the other hand, online calculations comprise CH_4 emissions from wetlands and termites, and CH_4 uptake by soil. CH_4 contributions from anthropogenic..." ➔ "...are converted into atmospheric concentrations based on an incremental approach. The CH_4 concentration changes are calculated as the CH_4 emission multiplied by a conversion factor that depends on the air density and thickness of the first model layer. On the other hand, CH_4 emission fluxes from wetlands and termites, as well as CH_4 uptake by soil, are all calculated online in the simulations (see Section 2.2.2 for further details). CH_4 contributions from anthropogenic..."

Table 1 can be considered to be moved to the supplement.

As Table 1 shows attributes that are not described in Section 2.1.1 Grid configuration, we do think it should be kept on the manuscript.

Lines 235-237: More information is needed for these sensitivity simulations.

Section 2.3 Experimental design was rewritten and now it better describes how the model sensitivity tests were conducted. Also, sentences explaining the criteria for selecting the study periods are included.

"Initially, a model sensitivity analysis for evaluating physics schemes such as planetary boundary layer and cumulus clouds, and global forcings for meteorological fields and CH_4 concentration, was carried out over several two-week periods in 2018 and 2019. Each of these two-week periods were previously examined to have at least 75% of days with TROPOMI XCH_4 data covering large portions of Europe. As a result, the physics schemes Yonsei University (YSU) for planetary boundary layer and Kain-Fritsch scheme for cumulus clouds, together with initial and boundary conditions from the European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis v5 (ERA5) model (Hersbach et al., 2020), for meteorological processes, and from the NCAR Community Atmosphere Model with Chemistry (CAM-chem) (Lamarque et al., 2012; Emmons, et al., 2020), for background concentrations of CH_4, were selected and then used to perform a one-year simulation period from April 01, 2018 to March 31, 2019. This period was defined based on the following criteria: i) availability of TROPOMI XCH_4 data, ii) latest available year data of EDGARv6.0 emissions for CH_4, and iii) no occurrence of sustained and irregular scenarios in terms of emissions (e.g., large-scale fire outbreaks and emission reductions associated with COVID-19 lockdowns). Tables 2 lists the physics and emissions schemes used in the simulations, with physics schemes other than planetary boundary layer and cumulus clouds being selected based on Beck et al. (2011). A schematic of the model running process is depicted in Appendix A. Off-line initial and boundary conditions derived from the simulations at 30 km are used as input to feed the simulations at 10 km. Model results and discussion for the nested domain are under development and will be described in a forthcoming paper."

Beck, V. et al.: The WRF Greenhouse Gas Model (WRF-GHG), Technical Report No. 25, Max Planck Institute for Biogeochemistry, Jena, Germany, 2011.

Lines 244-250: This section and Table 2 are identical, just keep one of them.

Section 2.3 Experimental design was rewritten and Table 2 kept.

ICOS stations in Figures 3-5 seem to not change, should be double-checked.

Yes, the CH_4 concentrations from ICOS stations did not change significantly near the surface (0 to 100 m) during the study period. This can be also observed in Figure S1 in the Supplement, with CH_4 concentrations ranging roughly from 1970 to 2030 ppb.

<u>Editorial comments:</u>

Line 15: Remove "a" before powerful tools.

Corrected.

Line 28: Remove "otherwise" and add "On the other hand" in the beginning of the sentence.

Corrected. A part of the sentence has been rewritten.

"…respectively. For winter months, otherwise, model-observation discrepancies show a significant..." ➔ "...respectively. On the other hand, model-observation discrepancies for winter months show a significant..."

Line 39: Add a reference after the first sentence.

A reference has been included to support the sentence

"Atmospheric methane (CH_4) has more than doubled since the pre-industrial." ➔ "Atmospheric methane (CH_4) has more than doubled since the pre-industrial era (Meinshausen et al., 2017)"

Meinshausen, M. et al.: Historical greenhouse gas concentrations for climate modelling (CMIP6), Geosci. Model Dev., 10(5), 2057-2116, 2017.

Figures 3-5. Units are missing in the figures and/or the figure caption.

Figures 3, 4 and 5 and their captions now include the units and also a single colorbar.