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Modelling seasonal cycle of atmospheric δ13C-CH_4 and their evaluations with δ13C-CH_4 observations

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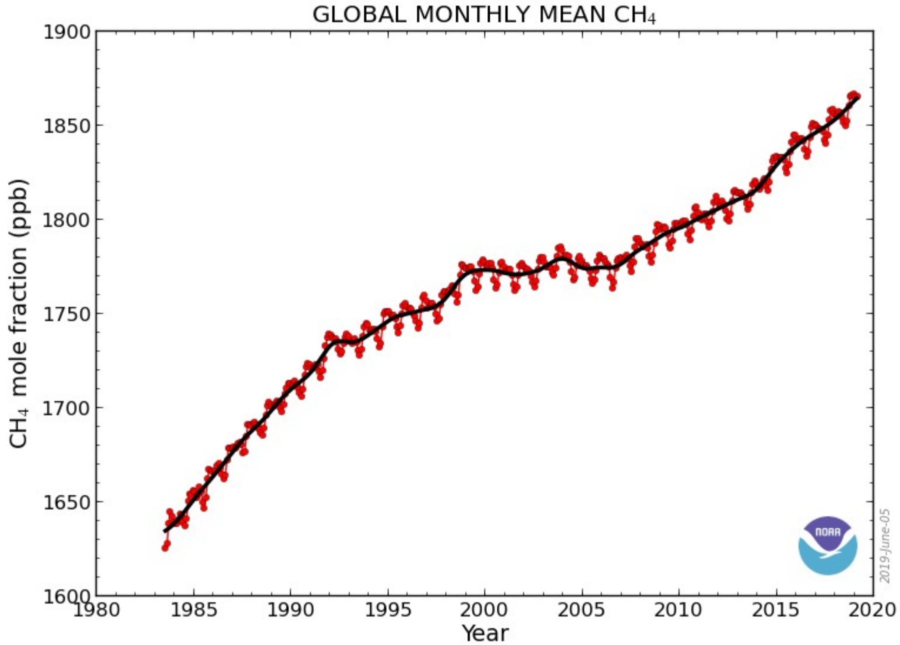
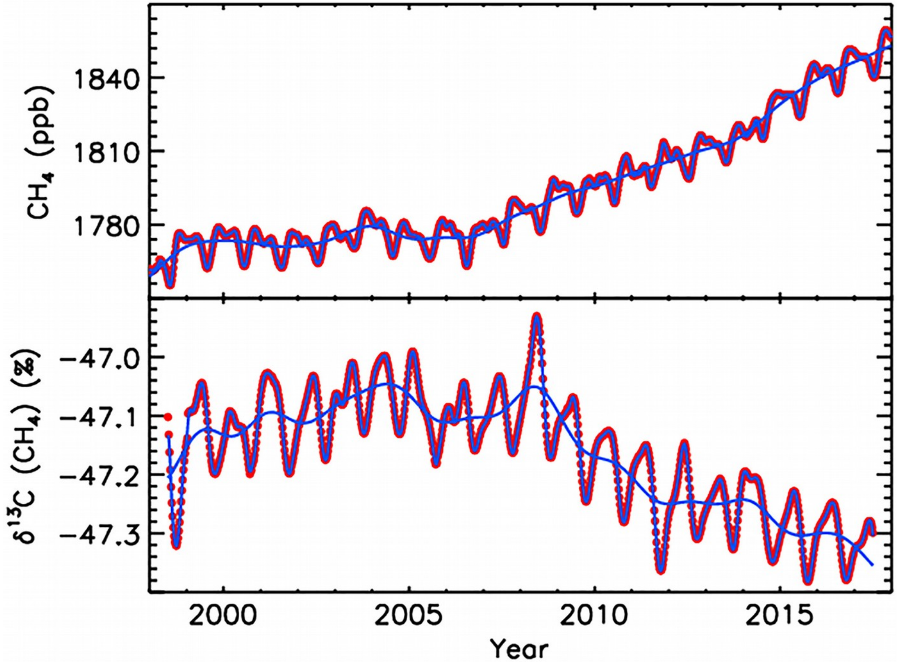
The work is also part of VERIFY-project WP4 (lead by Rona Thompson)

Background





- CH_4 increased until 2000, but during years 2000-2006 the atmospheric concentrations stayed constant after that the concentrations started to increase again (Figure in left). In 2006 when the atmospheric CH_4 started to increase the became more negative i.e. atmosphere is less enriched with $^{13}CH_4$.

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Background

- Stable isotopes $^{12}CH_4$ and $^{13}CH_4$
  - isotopic separation due to different masses
- Each CH_4 source have process specific isotopic signature

$$\delta^{13}C-CH_4=[\frac{(^{13}CH_4/^{12}CH_4)_{sample}}{(^{13}CH_4/^{12}CH_4)_{standard}}-1]1000\text{‰}$$

Background

- Hein et al. (1997): briefly showed that with inversion, we could estimate $\delta^{13}C$-CH_4 well.
  - Comparison to observations, but limited number of observations at that time.
- Tyler et al. (2007) showed weak anti-correlation and r^2 of $\delta^{13}C$-CH_4 to CH_4 at NWT and even worse for MDO (from measurements).
  - Strong source driven component + shift in phases (3 - 6 months)
  - SH should be better correlated?

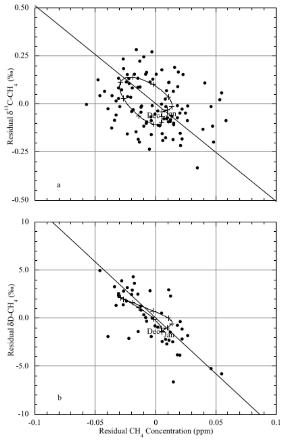


Figure 5. Phase ellipses for CH_4 at Niwot Ridge, Colorado, made by plotting (a) residual $\delta^{13}C$-CH_4 versus residual CH_4 mixing ratio and (b) residual δD-CH_4 versus residual CH_4 mixing ratio. The slopes of the lines result in kinetic isotope effects for atmospheric loss processes of 1.0090 ± 0.003 and 1.30 ± 0.03 for k_{12}/k_{13} and k_h/k_D, respectively.

HEIN ET AL.: THE GLOBAL ATMOSPHERIC METHANE CYCLE 67

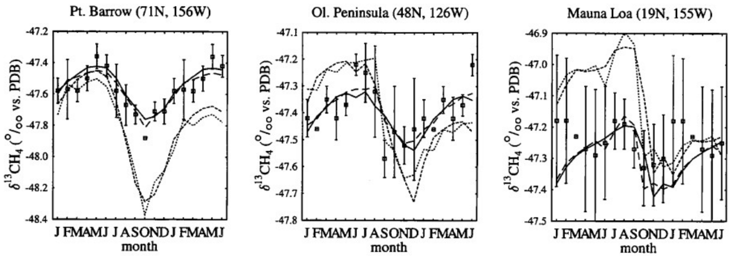
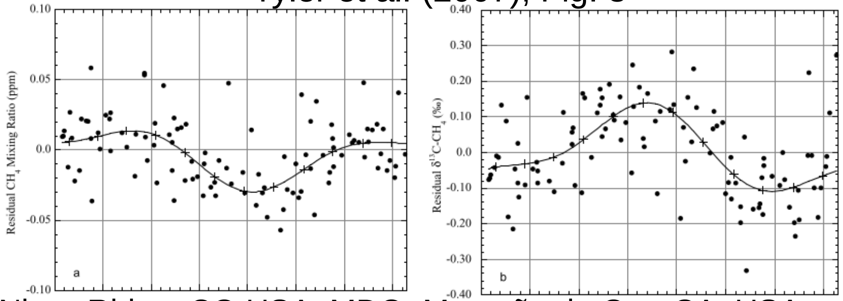


Figure 11. Simulated (dotted and solid lines, a priori scenario and scenario S_1 using 1987 wind data; short-dashed and long-dashed lines, a priori scenario and scenario S_3 using 1986 wind data in the transport model) and observed (squares with 1σ error bars) $\delta^{13}CH_4$ isotope ratios at the three available observational sites [*Quay et al.*, 1991]. The months of January to June are displayed twice in order to reveal the seasonal cycle more clearly.

Tyler et al. (2007), Fig. 3



NWT=Niwot Ridge, CO,USA; MDO=Montaña de Oro, CA, USA

Background

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  - Comparison to observations, but limited number of observations at that time.
- Tyler et al. (2007) showed weak anti-correlation and r^2 of $\delta^{13}C$-CH_4 to CH_4 at NWT and even worse for MDO (from measurements).
  - → strong source driven component + shift in phases (3 - 6 months)
  - → SH should be better correlated?
- Which source component drives the seasonal cycle of $\delta^{13}C$-CH_4 and anti-correlation of $\delta^{13}C$-CH_4 to CH_4?
- EDGAR v4.3.2 introduced seasonal cycle of the emission sources for the first time, but significant update in v5.0. Can we say which is more reasonable?
- How does different CH_4 sources and sinks affect the CH_4 and $\delta^{13}C$-CH_4 seasonal cycle?

Methods

- TM5 atmospheric chemistry model
  - Resolution 1º x 1º over Europe, elsewhere 6º x 4º
  - 25 vertical layers
  - Includes atmospheric loss i.e. OH, Cl+O(^{1}D) sinks, soil sink
    - the sinks enrich the atmosphere in $^{13}CH_4$
  - Simulation years: 2000-2012
- TM5 spin-up: repeat year 2000 40 times
  - isotopic signatures (Table later) multiplied by 1.095
- Simulations
  - Starting from a well mixed fields obtained from a spin-up
    - **EDGAR v4.3.5**
    - **EDGAR v5**
    - **EDGAR v4.3.2 + entferns** (no seasonal cycle)
    - **Bions** (no seasonal cycle)

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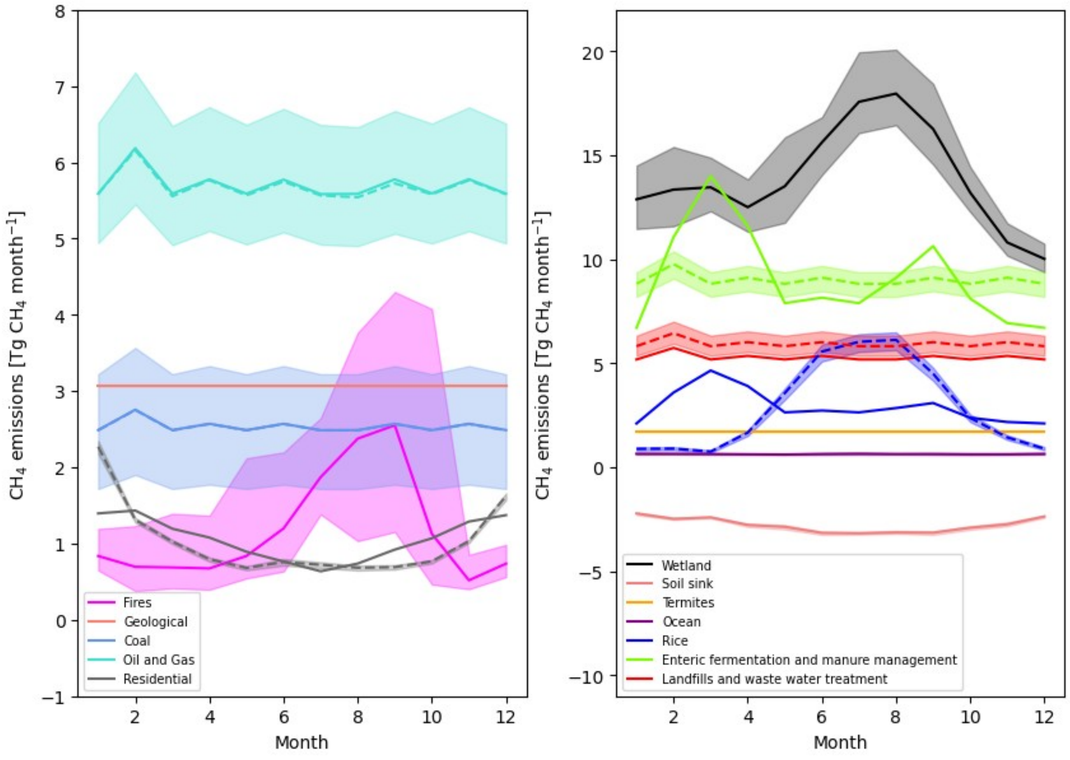
Methods

- Emission fields
  - Anthropogenic (monthly): EDGAR v4.3.2, v5.0
    - Rice
    - Enteric Fermentation and Manure Management (Livestock)
    - Landfills and waste water treatment
    - Coal
    - Oil and gas
    - Residential
  - Wetlands (monthly): LPX-Bern DYPTOP v1.4 (Lienert & Joos 2018)
    - Wetland/peatland source, Mineral soil (source)
    - Mineral soil (sink)
  - Fire (monthly; GFED v4.1s), Geological (annual; Etiope et al., 2019), Termites (annual: Ito and Inatomi 2012), Ocean (monthly; FMI, Tsuruta et al., 2017)

Methods

- Emission fields
  - Global total (for 2000, Tg CH_4 yr^{-1}):
    - EDGAR v4.3.2: 292.17
    - EDGAR v5.0: 299.05
  - Emissions from biogenic sources are higher than those from fossil sources, and wetland source (bio) being the highest
  - Seasonal cycle amplitude (annual max - min) is high for wetland source, enteric fermentation and manure management (entFer_manMan) v4.3.2 (appox. 7 Tg CH_4)
  - Amplitude and shape of seasonal cycle is very different between EDGAR v4.3.2 and v5.0 in rice, and enteric fermentation and manure management

Monthly CH_4 emissions



Solid line: EDGAR v4.3.2, Dashed line: EDGAR v5.0

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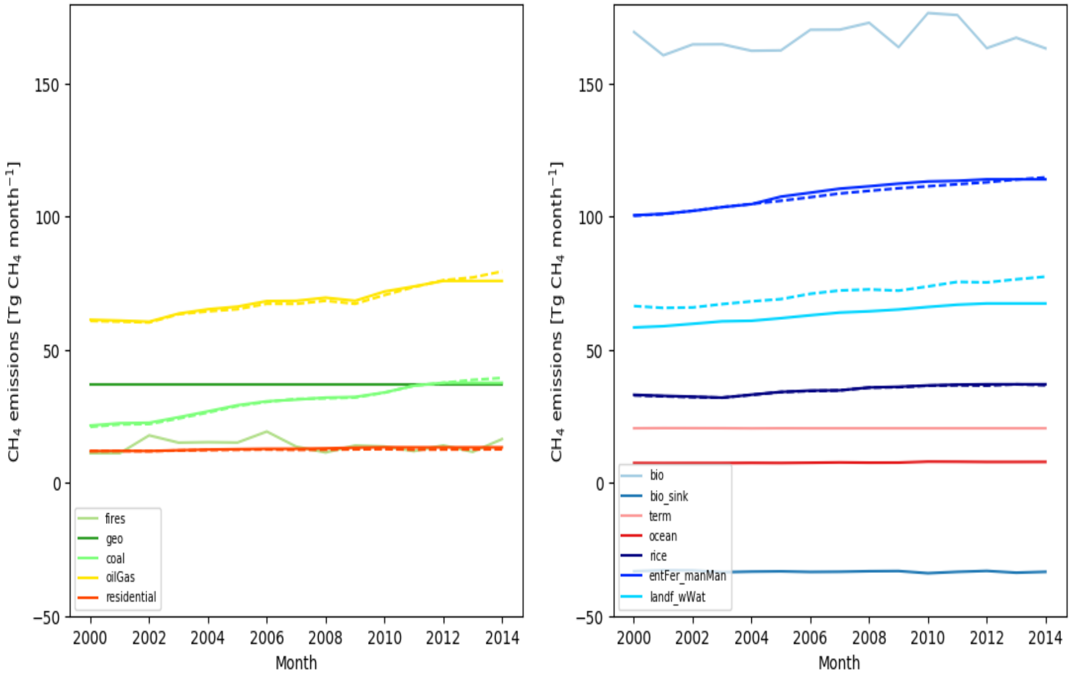
Methods

- Emission fields
  - Trends in two versions of EDGAR is similar.
  - Clear increasing trends in oil and gas, coal, enteric fermentation and manure management, and landfill and waste water sectors
  - Shifts? in rice emission levels before 2002 and after 2006
  - Wetland source has largest year-to-year variations

Annual CH4 emissions



Solid line: EDGAR v4.3.2, Dashed line: EDGAR v5.0

Methods

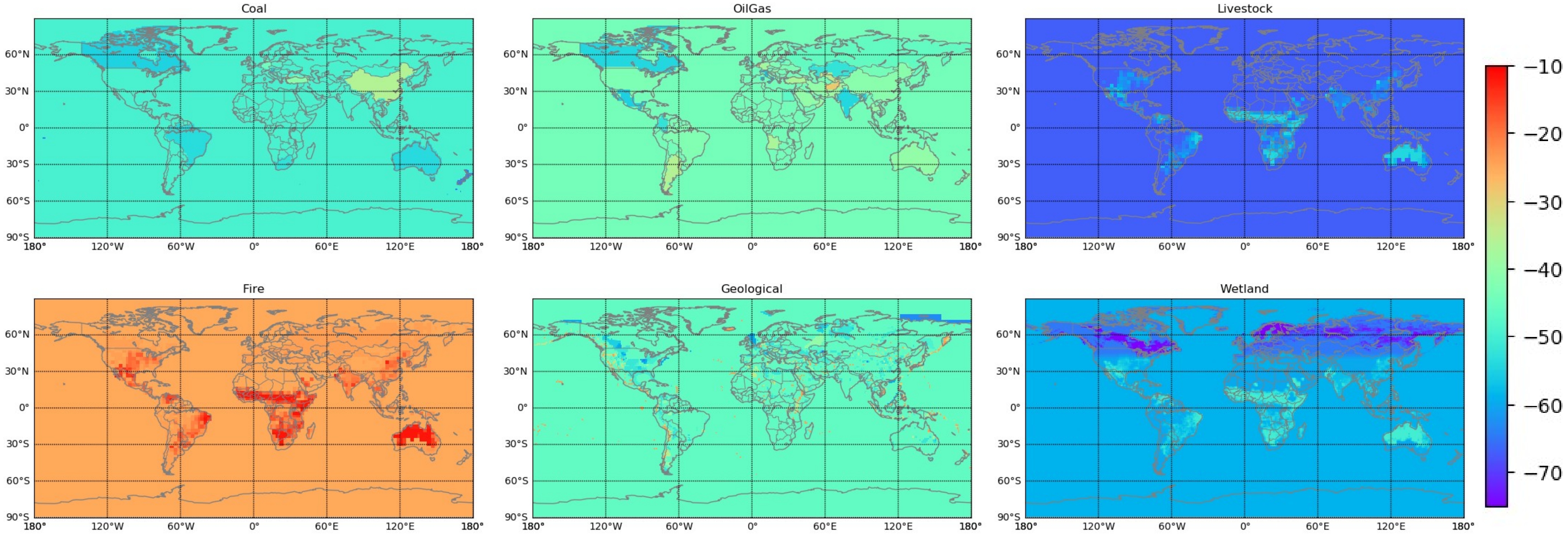
- Isotopic signatures
  - $^{13}CH_4$ fields are converted using isotope signature
  - Isotopic signature maps are used if available otherwise single value globally
  - Else, based on Monteil et al.,2011 (Table below)
  - Multiplied by 1.095* to converge to observed $\delta^{13}C\text{-}CH_4$ level

Source (Database)	$\delta^{13}CH_4$ (‰)	Source (Database)	$\delta^{13}CH_4$ (‰)
Rice agriculture(EDGAR)	-63[1]	Wetlands, mineral soils as source (LPX-Bern DYPTOP)	[-74.9, -50][5]
Enteric Fermentation and Manure Management (EDGAR)	[-67, -54][2]	Mineral soils, sinks (LPX-Bern DYPTOP)	-68.53
Landfills and waste water treatment (EDGAR)	-55[1]	Fire (GFED v4.1s)	[-25, -12][2]
Coal (EDGAR)	[-64, -36][3]	Geological (Etiope et al., 2019)	[-68, -24.3][4]
Oil and gas (EDGAR)	[-56, -29][2]	Termites (Ito and Inatomi, 2012)	-57[1]
Residential (EDGAR)	-38[1]	Ocean (FMI)	-59[1]

[1]Monteil et al., 2011
[2]Feinberg et al., 2017
[3]Sherwood et al., 2017
[4]Etiope et al., 2019
[5]Ganesan et al., 2018

Methods

- Isotopic signature maps (‰)



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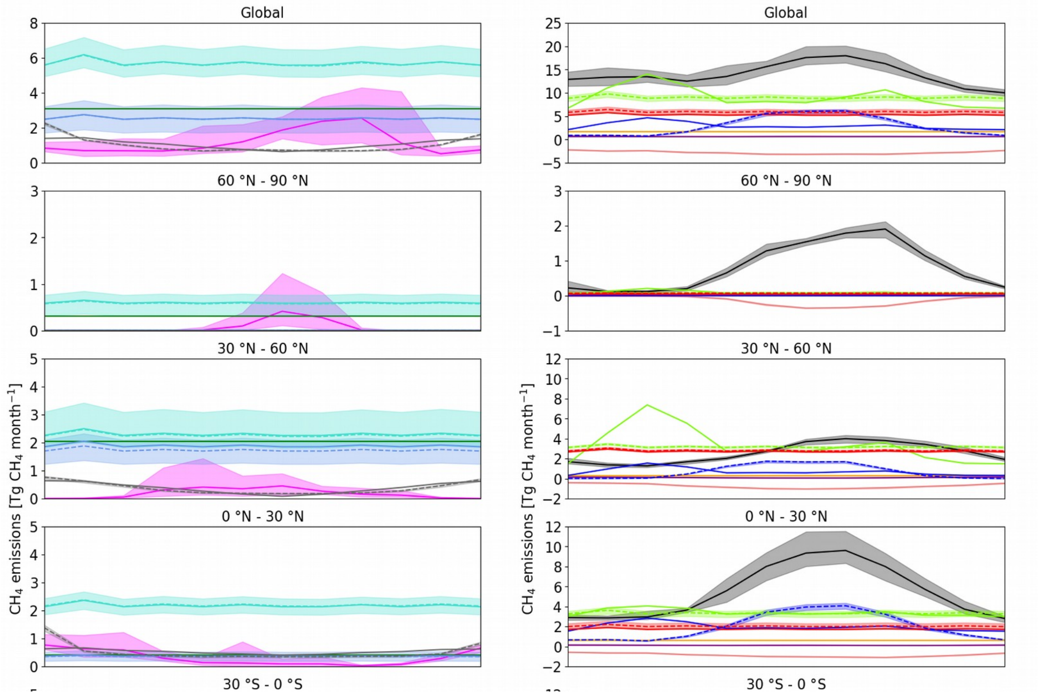
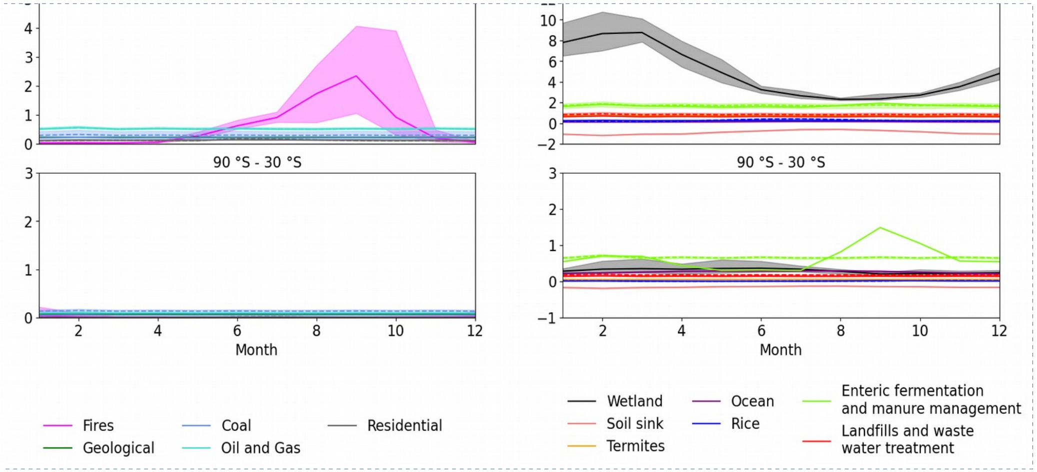
Emissions in different latitudes





- Emission sources vary in magnitude in different latitudes
- SH emissions are small

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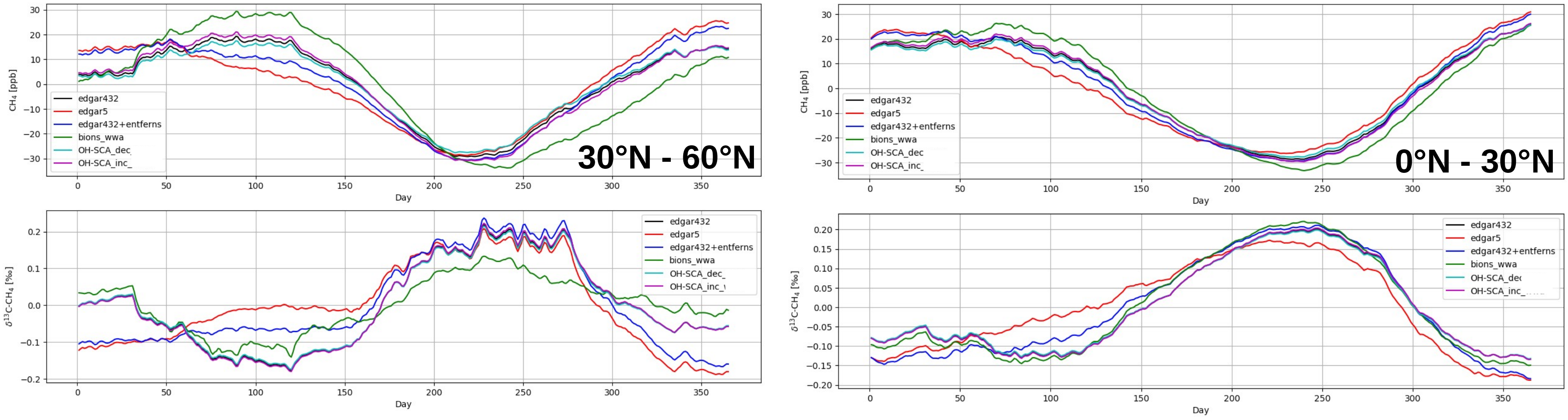
Results: Years 2000-2012

- Spring:
  - EDGAR v4.3.2 show depletion, while EDGAR v5.0 show increase in delta values in Tropics and Temperate latitudes in NH. Differences of more than 0.15 ‰. → effect of biogenic source in EDGAR v4.3.2 which increases in spring.
  - Effects of enteric fermentation and manure management emissions are stronger in Temperate region than in the Tropics.

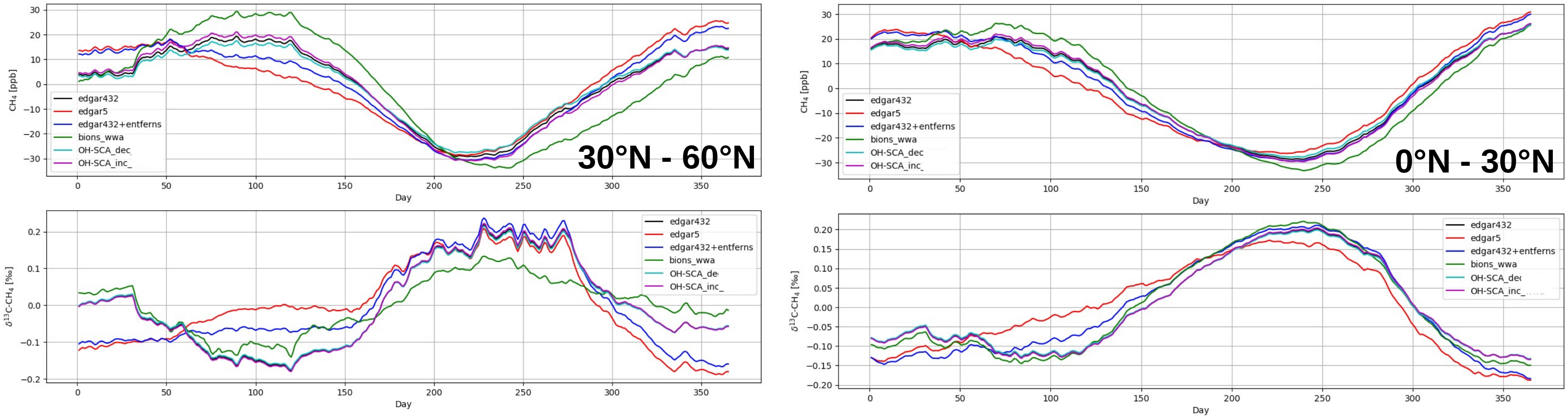
2002-2012 average seasonal cycle lower troposphere (model level 1-5), detrended



Results: Years 2000-2012

- Summer:
  - Differences in the seasonal cycle of rice field emissions is little – we expected to see more depletion
- Winter:
  - More depleted methane in EDGAR v5.0 and enferns simulations. → EDGAR v4.3.2 has lower enteric fermentation and manure management emissions (only a few Tg per month)

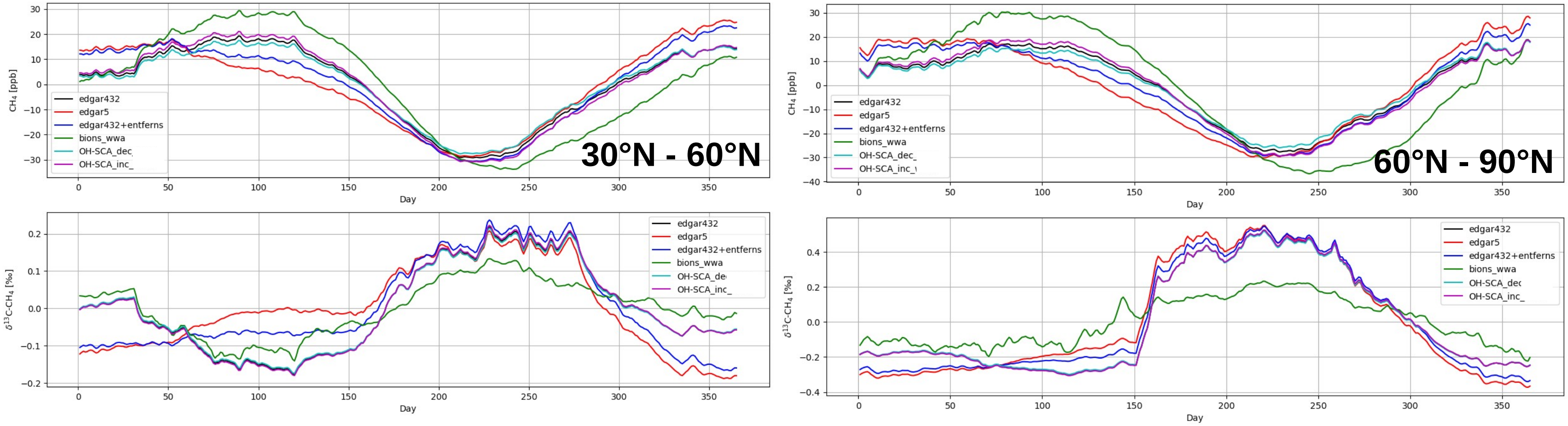
2002-2012 average seasonal cycle lower troposphere (model level 1-5), detrended



Results: Years 2000-2012

- Without seasonal cycle in wetland source
  - Delta values are mostly affected in the NHL, where wetlands is dominant biogenic source.
  - SCA in the NHL without wetland seasonal cycle is less than two times smaller than those with.
  - Still some differences in summer and winter, probably driven mostly by OH?

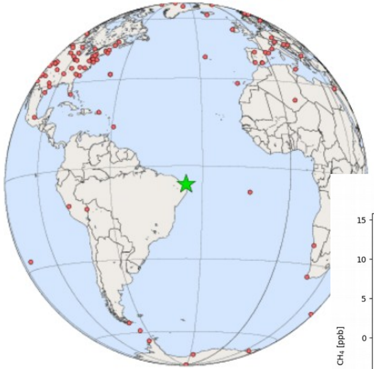
2002-2012 average seasonal cycle lower troposphere (model level 1-5), detrended



2000-2012 runs

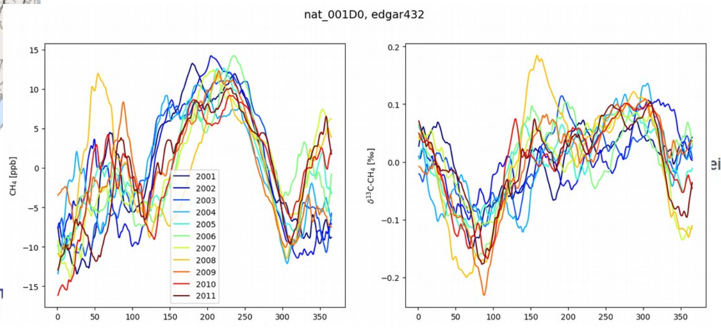
- Variations between years
  - NAT: significantly higher delta value in summer 2008 → could not find obvious decrease in wetland sources or increase in fossil fuel sources around the sites

Farol De Mae Luiza Lighthouse, Brazil [NAT]



Location

- Country: Brazil
- Latitude: 5.7952° South
- Longitude: 35.1853° West
- Elevation: 50.00 masl
- Time Zone: Local Standard Time + 3.0 hour(s) = UTC



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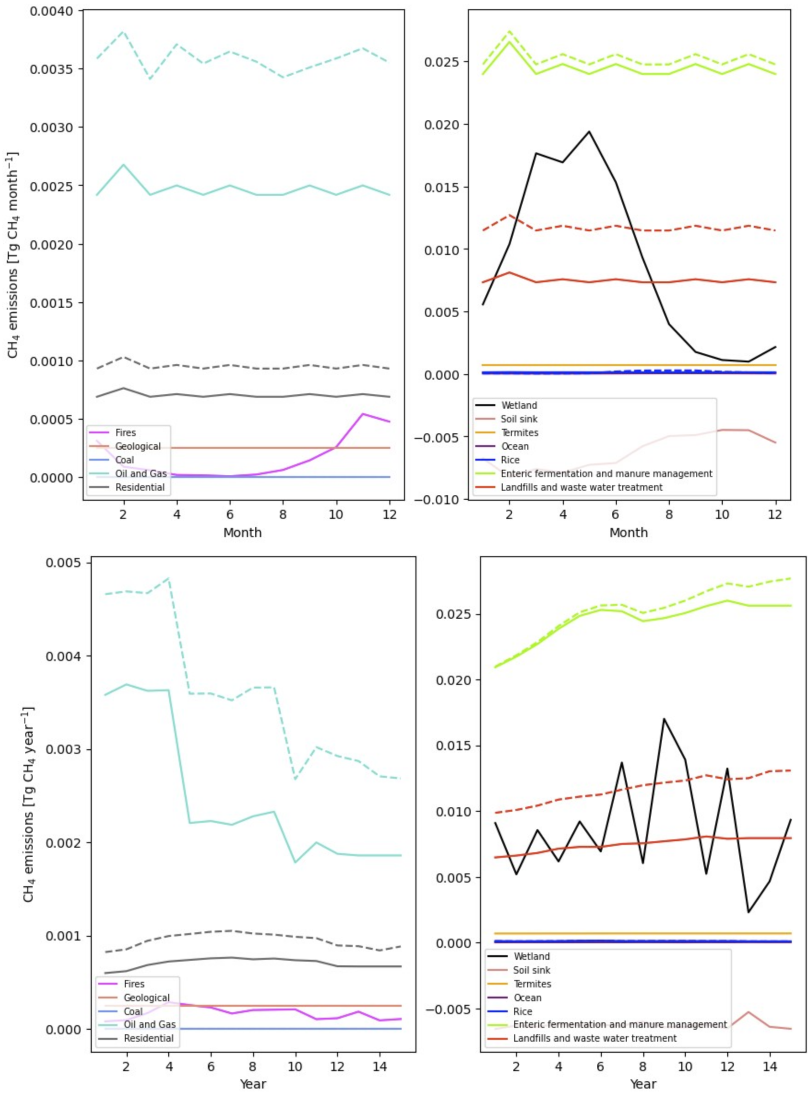
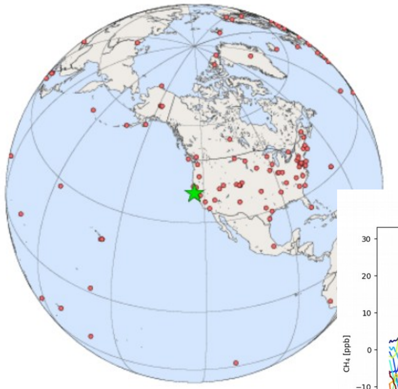
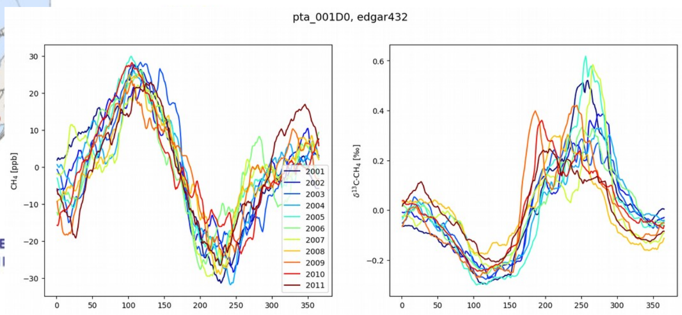
Emissions at NAT



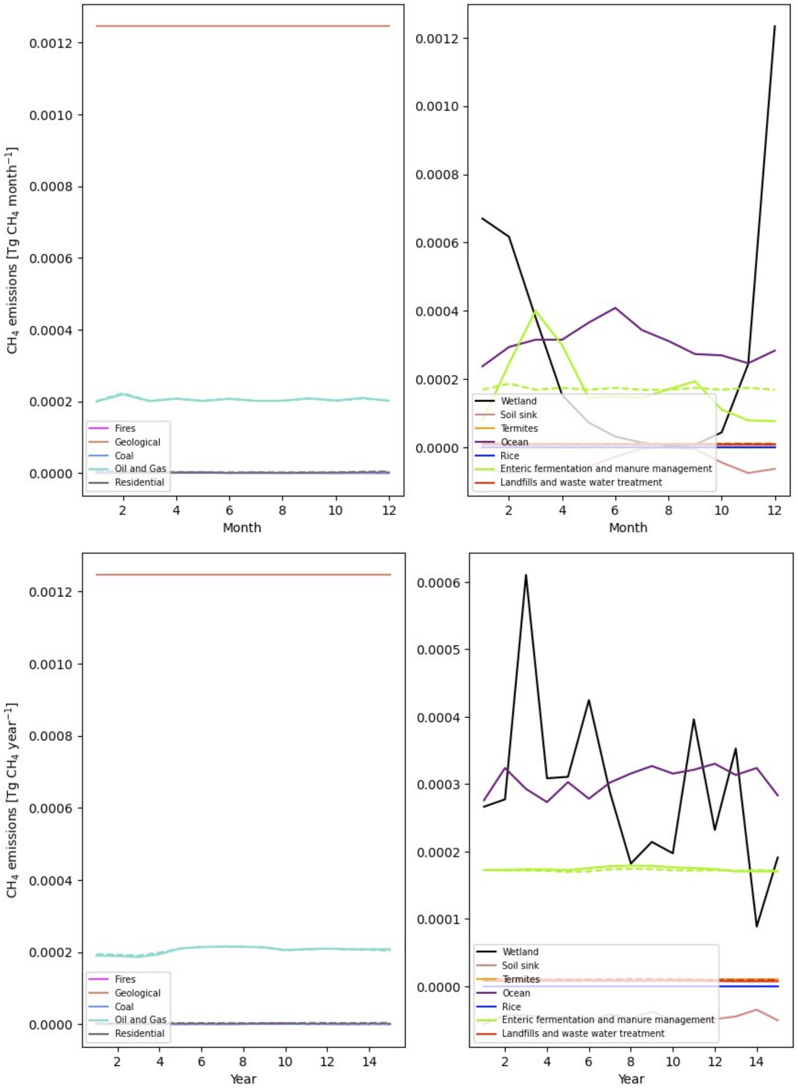
2000-2012 runs

- Variations between years
  - PTA: earlier increase in delta values (i.e. shift of summer maximum in late years) → decrease in wetland emissions?



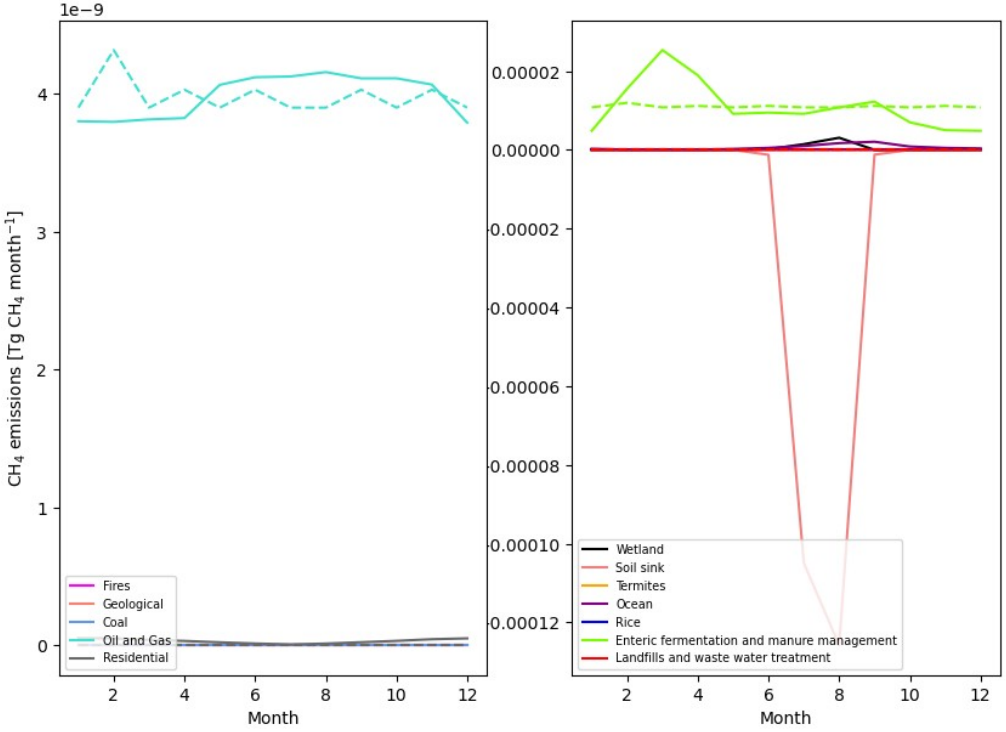


Emissions at PTA

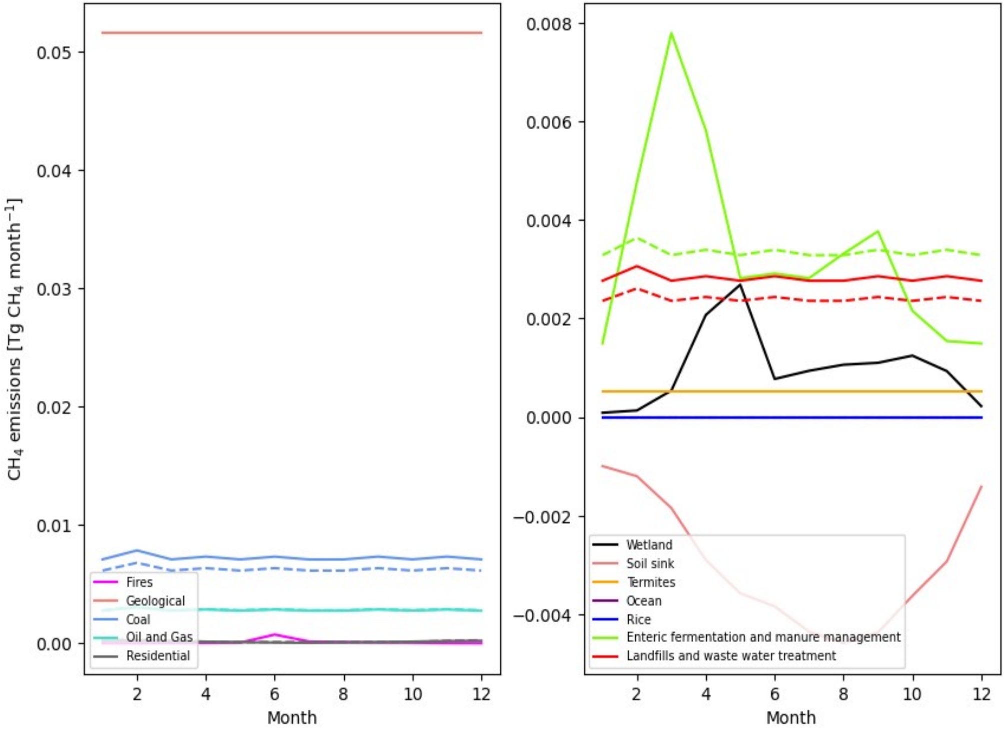


Difference in emissions in two stations

alt_001D0



nwr_001D0



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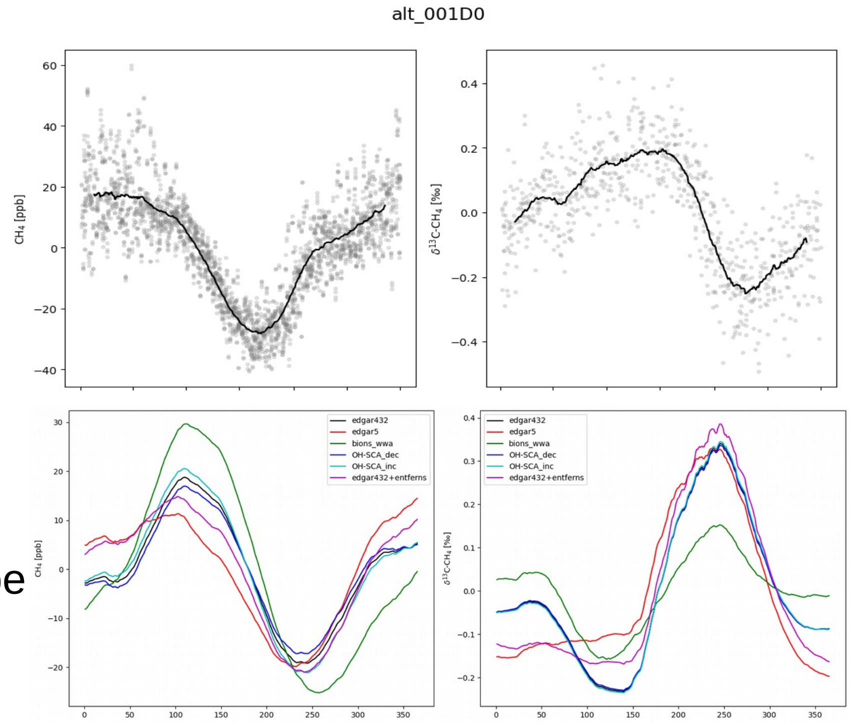
Comparison to observations

- Due to limited number of observations, it’s difficult to evaluate seasonal cycle of the observations year-by-year.
  - All available observations put into one year
  - Trends removed from obs by simply taking annual averages

- ALT:
  - Modelled CH_4 and delta values show later annual min. and max., respectively.
  - Obs. show no depletion of CH_4 in spring - EDGAR v5.0, and with wetland source seasonal cycle may be more reasonable
  - No simulation was able to reproduce depletion in autumn



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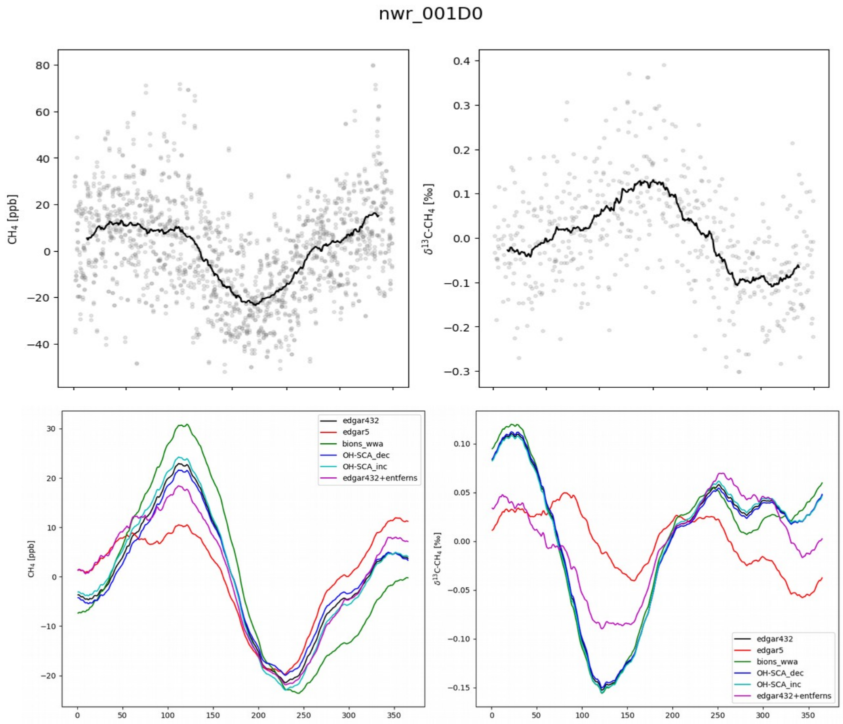
Comparison to observations

- Due to limited number of observations, it's difficult to evaluate seasonal cycle of the observations year-by-year.
  - All available observations put into one year
  - Trends removed from obs by simply taking annual averages
- NWR:
  - Very strong depletion in spring with EDGAR v4.3.2, which is not shown in the observations.
  - All simulations show depletion in spring to some extent, but those with EDGAR v5.0 may be the closest to the observations



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Summary and Future Work

- Seasonal cycle at lower atmosphere
  - It is generally an inverse of CH_4 (e.g. for NH, high in summer and low in winter)
  - However, not perfectly anticorrelated with CH_4, indicating effect of sources
- We can possibly learn about source distribution not only from the absolute delta values, but also from the seasonal cycle
  - Effects of changing sources were seen at latitudinal zonal mean and site-level estimates
  - Changing EDGAR versions have a large impact in some locations; in general v5.0 was better
- Future work:
  - Check anti-correlation to CH_4, and spatial distributions of emissions more in detail
  - More comprehensive comparison to observations
  - Investigate the effect of atmospheric sinks (including stratosphere)
  - Inversion runs with CarbonTracker-Europe $^{13}CH_4$

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Thank You!

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