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Modelling atmospheric CH_4 and $\delta^{13}C$-CH_4 with TM5

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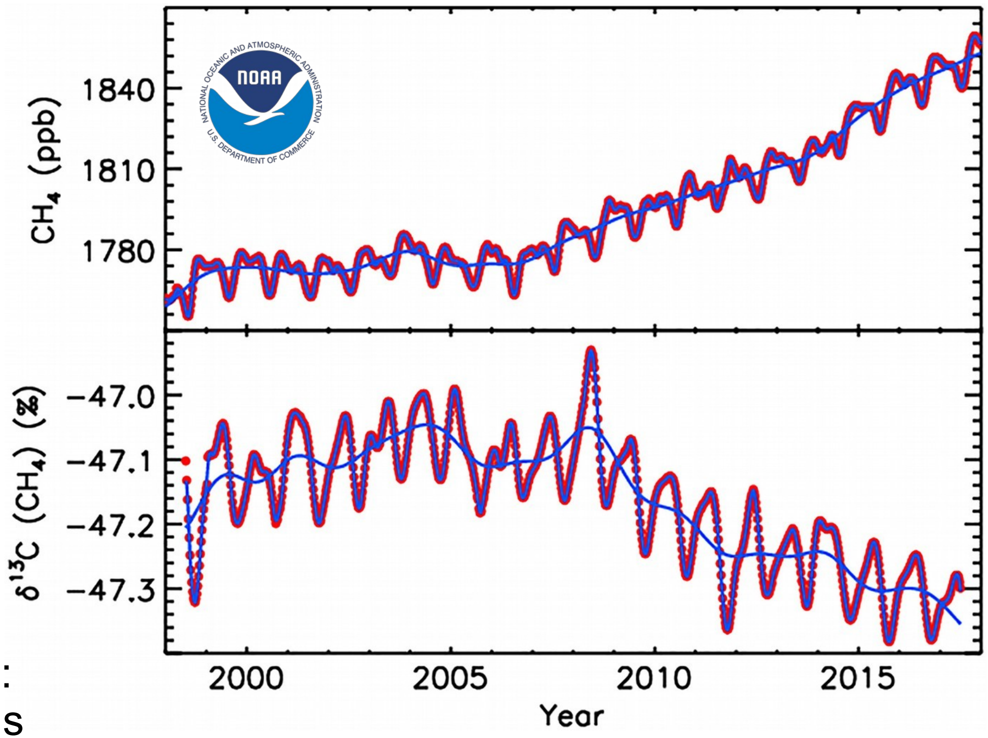
Background

- 21st century atmospheric CH_4 started from a near-equilibrium state, without much increase until around 2007.
- After 2007, atmospheric CH_4 is increasing at rate of 5 – 14 ppb/yr
- $\delta^{13}C$-CH_4 is decreasing significantly also after 2007.
- Those indicate:
  - Increase in CH_4 emissions after 2007
  - Changes in emissions source components: less thermogenic (e.g. fossil fuel) emissions and/or more biogenic sources.



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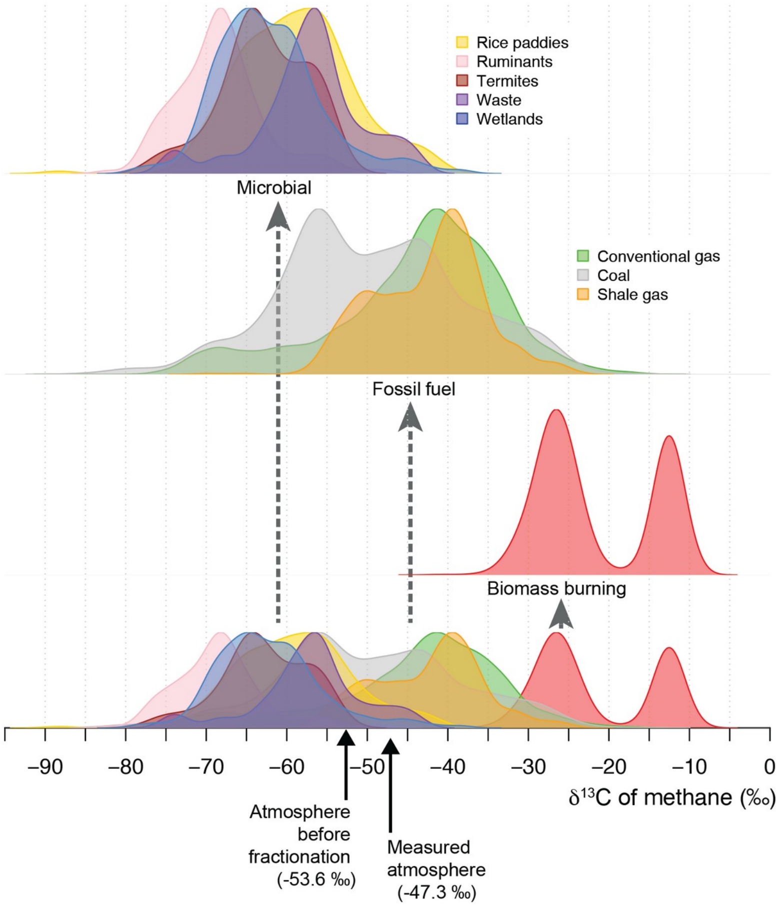
Background

- $\delta^{13}C\text{-}CH_4$ (‰) is ratio of ^{13}C and ^{12}C in CH_4, defined by:

$$\delta^{13}\mathrm{C{-}CH_4} = \left(\frac{(^{13}\mathrm{C}/^{12}\mathrm{C})}{R_{\mathrm{std}}} - 1\right) \times 1000.$$

- $\delta^{13}C\text{-}CH_4$ in atmosphere is about -47.3 ‰
- $\delta^{13}C\text{-}CH_4$ is higher for biomass burning (enriched in ^{13}C)
- $\delta^{13}C\text{-}CH_4$ is lower in microbial sources compared to fossil fuel emissions (depleted in ^{13}C)

Normalized pdfs for $\delta^{13}C\text{-}CH_4$



Sherwood et al., 2017, ESSD

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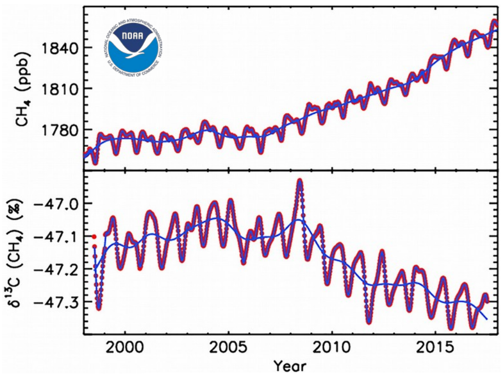
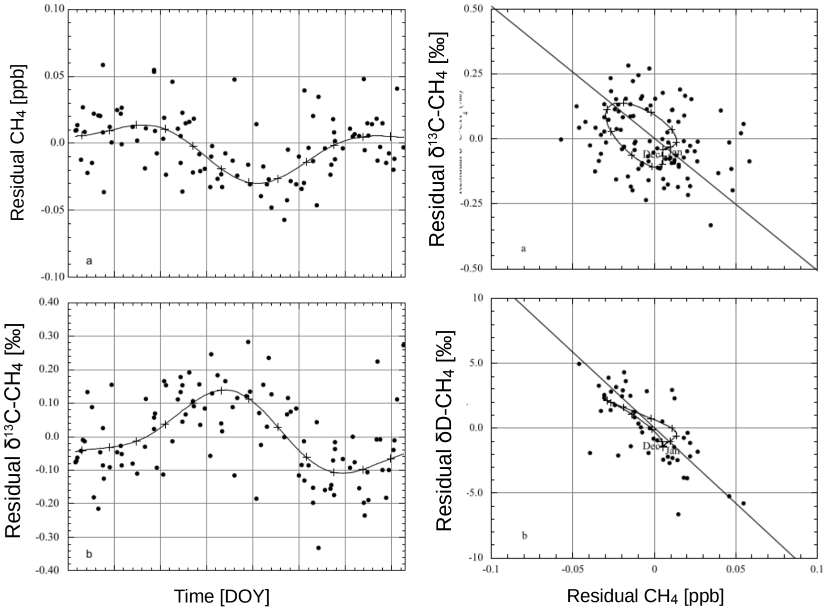
Background

- Changes in δ^{13}C-CH_4 from pre-2007 to the latest years are approx. 0.2 ‰.
- Seasonal cycle amplitude in δ^{13}C-CH_4 is 0.1 – 0.2 ‰.
- δ^{13}C-CH_4 seasonal cycle is strongly driven by emission sources – weak correlation in CH_4:δ^{13}C.
- Research questions:
  - Can we detect the seasonal cycle using a transport model?
  - What emission sources contribute to the seasonal cycle of δ^{13}C-CH_4?





Measured CH_4 (TL) and δ^{13}C (BL) seasonal cycle at Niwot Ridge, and residuals of isotopes against of residuals of CH4 (δ^{13}C on LR and δD on BR). Tyler et al., 2007, JGR

Methods

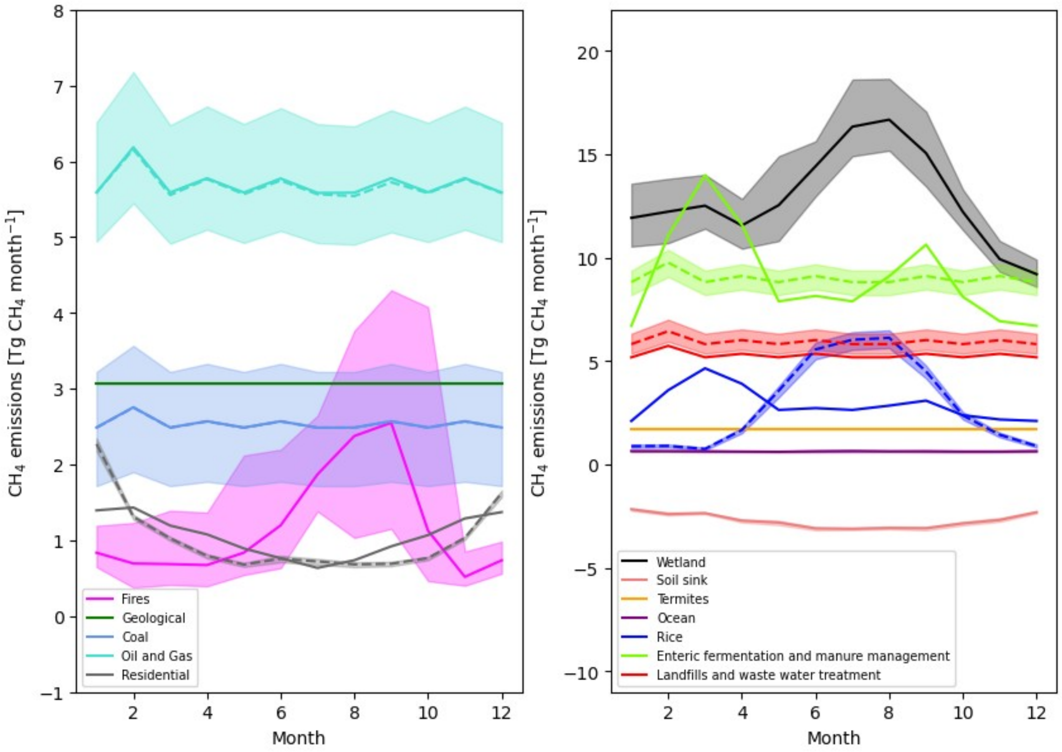
- TM5 atmospheric chemistry model
  - Resolution 1º x 1º over Europe, elsewhere global 6º x 4º
  - 25 vertical layers
  - Includes off-line atmospheric loss i.e. OH, Cl, $O(^1D)$ sinks
  - Simulation years: 2000-2012
- Flux fields from
  - Anthropogenic (monthly): EDGAR v4.3.2, v5.0
  - Wetlands and soil sink (monthly): LPX-Bern v1.4
  - 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

Seasonal cycle of flux fields

- Emissions and seasonal cycle amplitude from biogenic sources are higher than those from thermogenic sources
- Seasonal cycle amplitude (monthly max – min) is high for wetland source
- Amplitude and shape of seasonal cycle is very different between EDGAR v4.3.2 and v5.0
- Amplitude is highest in emissions from enteric fermentation and manure management (EFMM) in v4.3.2 (≈ 7 Tg CH_4), and from Rice in v5.0 (≈ 6 Tg CH_4)
- Global total do not differ much between the two EDGAR versions (v4.3.2: 292.17, v5.0: 299.05 Tg CH_4 yr^{-1})

Global monthly CH_4 fluxes



Solid line: EDGAR v4.3.2, Dashed line: EDGAR v5.0
Shaded areas show annual variations.

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Methods

Isotopic signatures

- CH_4 emission fields are converted to $^{13}CH_4$ fields using the isotope signatures (constant over time).
- Spatial distributions are taken into account if available (filled-value from Thompson et al., 2018). Otherwise a single value globally.

Source (Database)	$\delta^{13}CH_4$ (‰)	Source (Database)	$\delta^{13}CH_4$ (‰)
Enteric Fermentation and Manure Management (EDGAR)	[-67.9, -54.5][2]	Landfills and waste water treatment (EDGAR)	-55.6[1]
Wetlands, mineral soils as source (LPX-Bern v1.4)	[-74.9, -50.0][4]	Rice agriculture (EDGAR)	-62.1[1]
Coal (EDGAR)	[-64.1, -36.1][2]	Termites (Ito and Inatomi, 2012)	-65.2[1]
Oil and gas (EDGAR)	[-56.6, -29.1][2]	Residential (EDGAR)	-40[1]
Fire (GFED v4.1s)	[-25.0, -12.0][2]	Mineral soils, sinks (LPX-Bern v1.4)	-19.6[1]
Geological (Etiope et al., 2019)	[-68.0, -24.3][3]	Ocean (FMI)	-47[1]

Blue: biogenic anthropogenic Green: biogenic natural Orange: thermogenic anthropogenic Gray: thermogenic natural

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

Methods

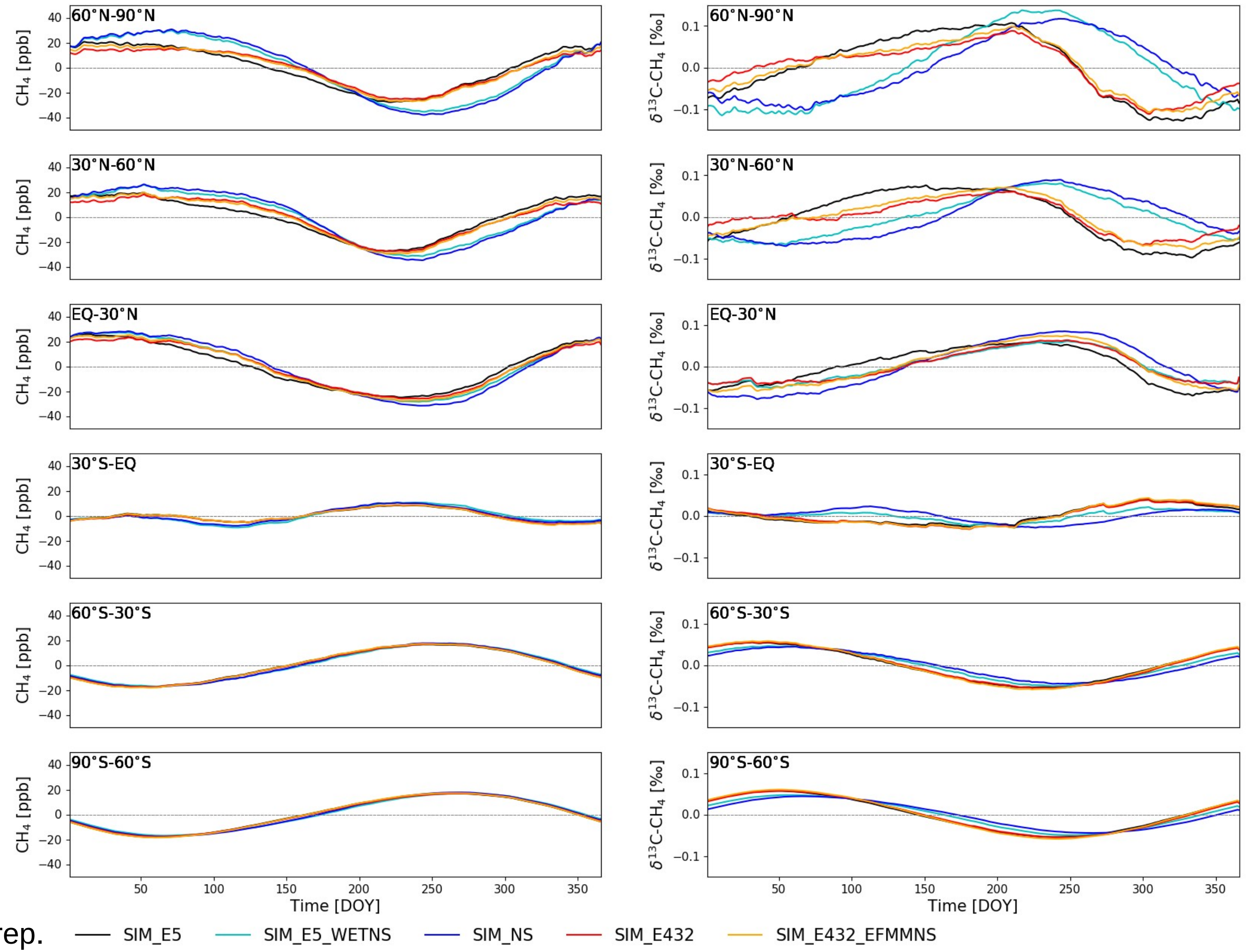
- Simulations using different emission fields
  - EDGAR v5.0 (E5)
  - Remove seasonal cycle of wetland source (E5_WETNS)
  - EDGAR v4.3.2 (E432)
  - EDGAR v4.3.2, but remove seasonal cycle of enteric fermentation and manure management (E432_EFMMNS)
  - No seasonal cycle in emission fields (NS)

Results

Zonal means

- $\delta^{13}C$-CH_4 cycle is generally an inverse of CH_4 cycle
  - KIE effect of atmospheric sinks
- The model is able to reproduce $\delta^{13}C$-CH_4 amplitude of ≈ 0.1-0.2 ‰

Detrended daily averaged zonal mean at L1-5



Kangasaho et al., in prep.

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Results

Zonal mean δ^{13}C-CH_4 cycles

- Affected by emissions mostly in the NH.
- Wetland emission is a key to determine the shape of δ^{13}C-CH_4 cycle
  - Timing of minimum in 30°N> is 50-80 days later without wetland seasonal cycle
- Changing EDGAR versions affects both amplitude and shape
  - Amplitude is smaller with v4.3.2 EFMM cycle
  - More gradual increase in δ^{13}C-CH_4 in the beginning of the year using v4.3.2
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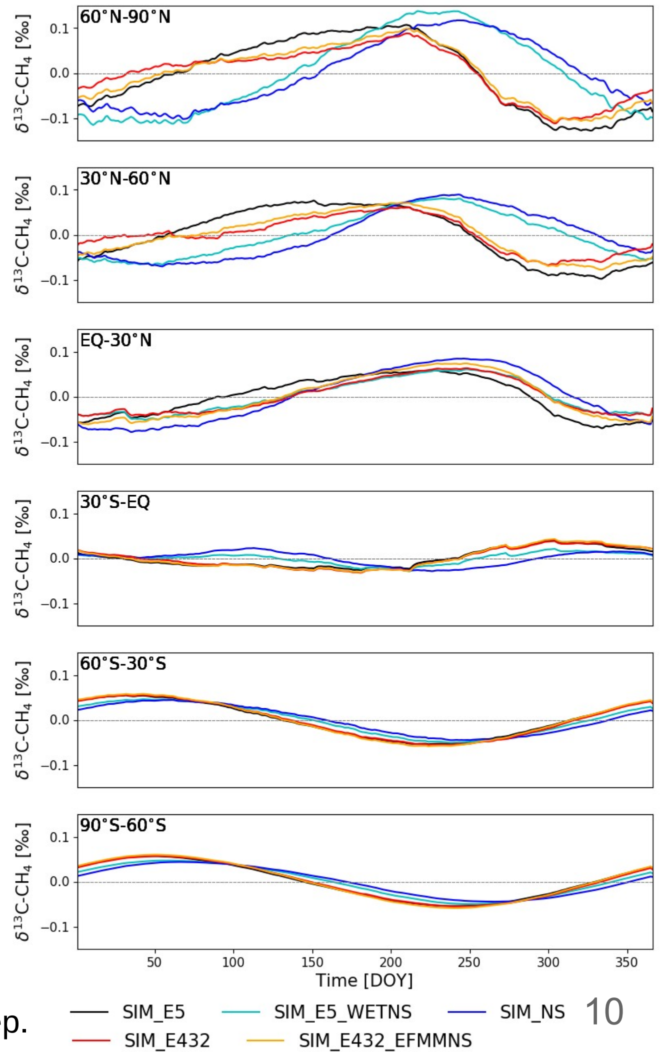
Detrended zonal mean seasonal cycles



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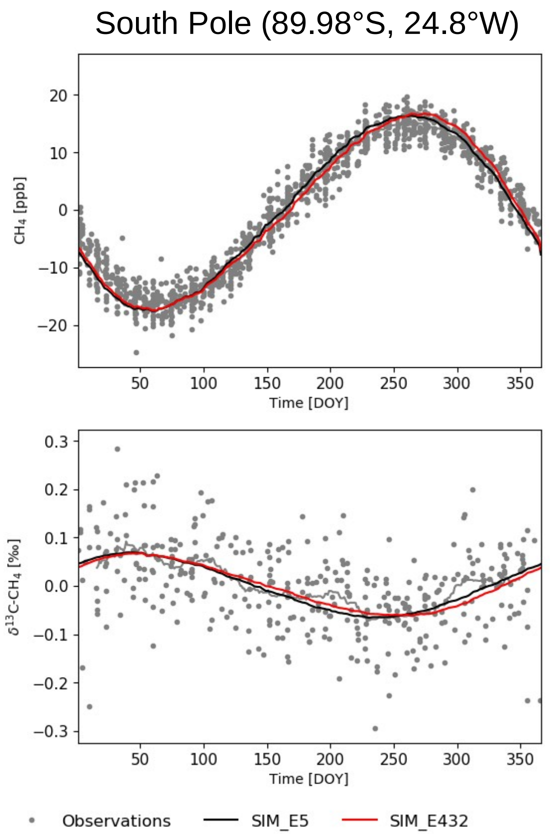
Comparison to observations

- δ^{13}C-CH_4 observations generally have high deviations
- South Pole: model captured seasonal cycle well



Comparison of seasonal cycles between observations (<2006) and model results. Kangasaho et al, in prep.

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Comparison to observations

- $\delta^{13}C\text{-}CH_4$ observations generally have high deviations
- Mace Head:
  - E5 captures better the increase in $\delta^{13}C\text{-}CH_4$ in the beginning of the year; E432 show depletion.
  - Both versions failed to capture strong decrease in $\delta^{13}C\text{-}CH_4$ in autumn; similar feature is found at e.g Alert. → possibly due to wetland emission cycle?
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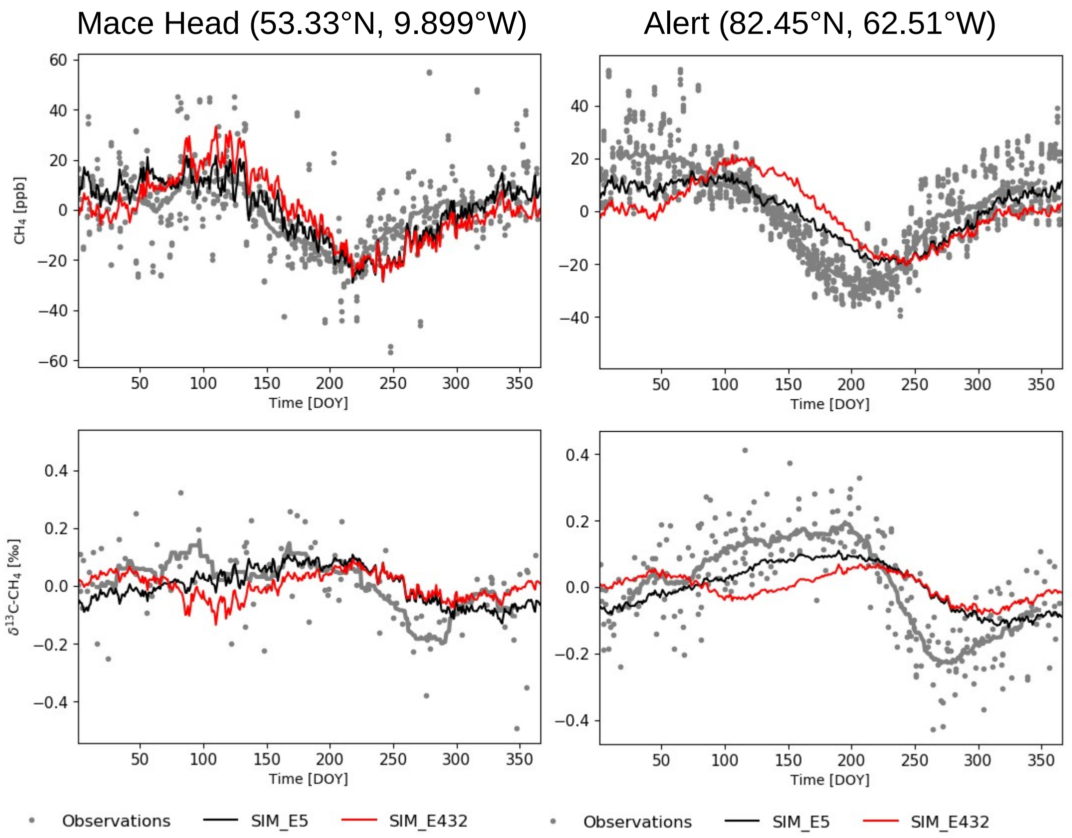

Comparison of seasonal cycles between observations (<2006) and model results. Kangasaho et al, in prep.

Summary/Conclusion

- We examined seasonal cycle of $\delta^{13}C$-CH_4 in the 21st century using TM5 transport model
- Model showed zonal mean $\delta^{13}C$-CH_4 amplitude of ≈ 0.1-0.2 ‰, similarly to those measured
- Wetland is a key source to determine the shape of $\delta^{13}C$-CH_4 cycle
  - Seasonal cycle in wetland emissions is strongest
  - Lead to depleting $\delta^{13}C$-CH_4 in autumn in the NH
  - Improvement in emission seasonal cycle maybe needed – could use inversion results
- Comparison to observations show EDGAR v5.0 seasonal cycle is probably more realistic than v4.3.2

Methods

- Isotopic signature maps (‰)