PAL MOD | GERMAN CLIMATE MODELING INITIATIVE

Factors controlling atmospheric oxidative capacity and CH_4 lifetime during the LGM

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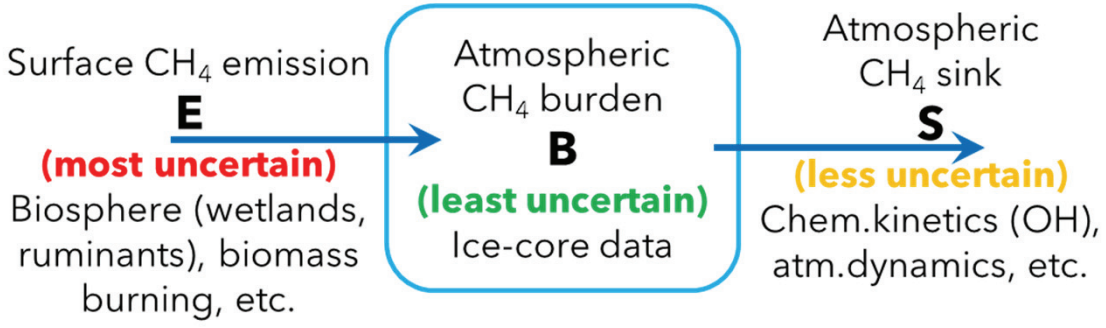
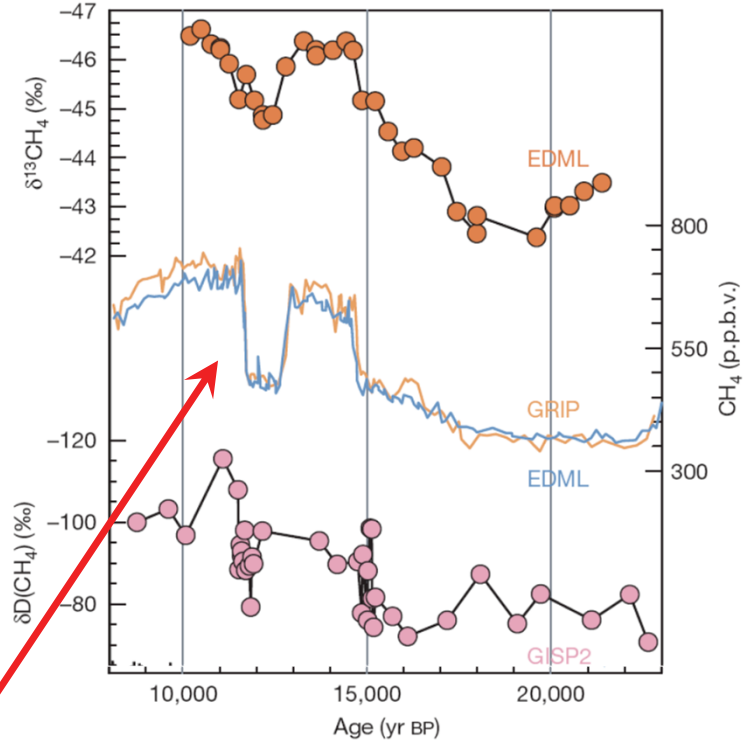
Past atmospheric CH_4 variations

- CH_4 provides additional insight on the cycle of C and biosphere changes in the past
  - Climate transitions –> triggering of different (natural) sources

- Inferring sources strengths requires/can be done using adequate estimates of the sinks
  - Which component can be estimated with greater certainty?





[Fischer et al., Nat#425, 2008]

Shutdown of sources?
Changes in sinks?

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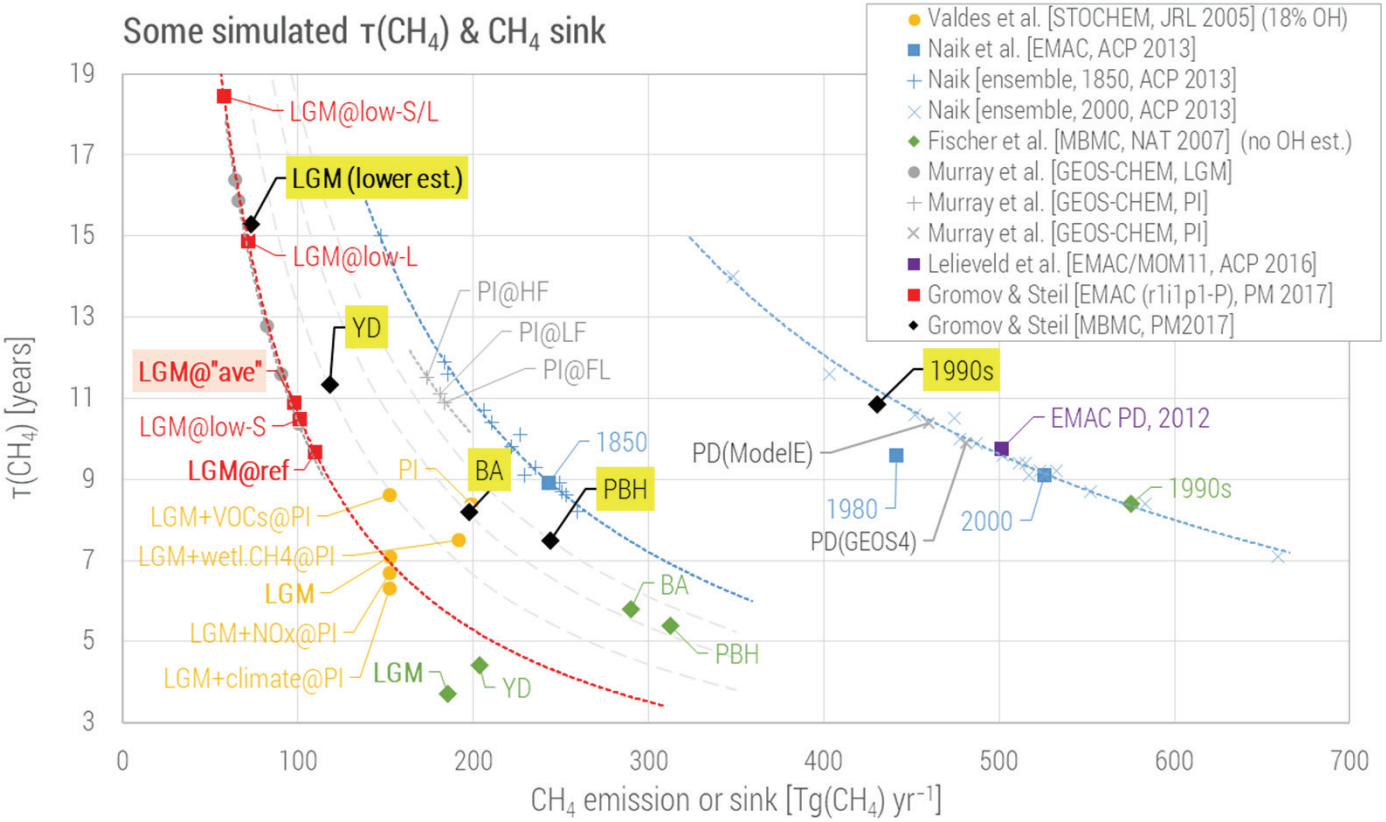
CH_4 lifetime concept



- **S** *vs.* **τ** found at CH_4 burden isopleths in different climates
  - **As lifetime gets longer, less uncertainty is expected in emissions**

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Model Setup | EMAC

EMAC = ECHAM5/MESSy Atmospheric Chemistry model [Jöckel *et al.*, 2006, 2010]

- LGM-chemistry version of AC-GCM (MESSy/EMAC v2.52) :
  + Climate state from **MPI-ESM-P model CMIP5/r1i1p1-P exp.** [Giorgetta *et al.*, 2013]:
    – Vegetation state and other boundary conditions (JSBACH)
    – Boundary conditions and atmospheric dynamics (optional)
  + **On-line calculation of trace gas emissions** (EMAC submodels):
    – Biosphere (land/ocean), biomass burning, lightning
  + Orbital parameters, GHGs mixing ratios, dust, etc. prescribed (PMIP3)
  + Comprehensive **MIM1 chemistry: latest JPL'15, 300+ reactions, ~200 species** (O_X, HO_X, NO_X, VOCs, Cl, S, halogens, isotope C/H/O)

⇒ **Time-slice simulations for LGM at T63L47 resolution** (top at ~80 km)

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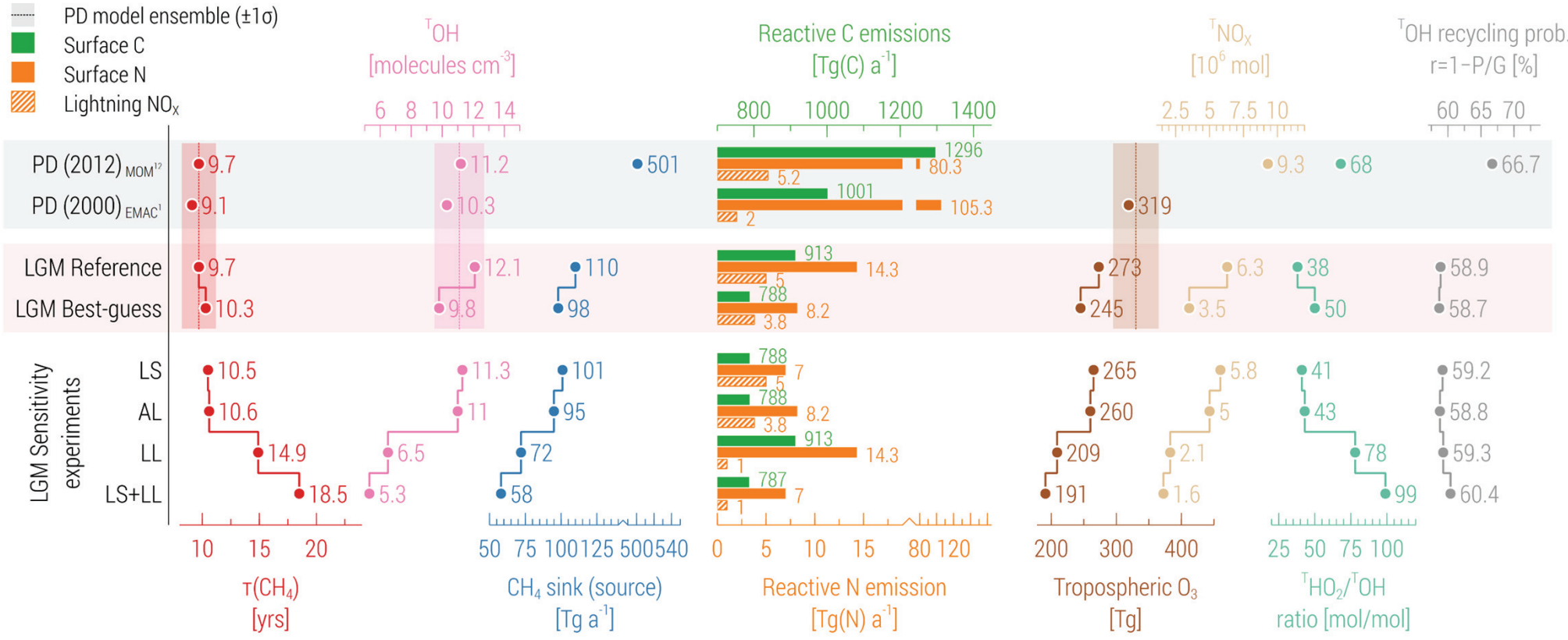
Overview of Model Experiments



Typical uncertainties: Trace gas emissions: ±20%, up to > ±50% (some VOCs)

Gas-phase kinetics: ±(15–20)‰

Wet/dry deposition: up to ±50%

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Results from EMAC

- Sensitivity of k(CH_4+OH) to T is low:
  at ΔT_{LGM-PD} = −3.1K (troposphere, mass-weighted!) => $\Delta\tau(CH_4)$ is **+0.5 yrs**

- Emissions probed in sensitivity experiments (compared to PD estimates)

RN:	Reactive nitrogen, 8−19 TgN/yr	(9−23)%
	from lightning NO_X	**(20−100)‰**
RC:	Reactive carbon, 770−910 TgC/yr	(61−71)%

- Resulting tropospheric burdens

O_3:	191−265 Tg	(60−85)%
OH:	$(5-12)\cdot10^5$ molecules/cm^3	(50−120)%

- Lifetime ranges **9−11 yrs** (up to 18 yrs in sensitivity simulations)
- Changes are driven mostly by **RN from lightning NO_X emissions (LNO_X)**
  - Most of CH_4 sink occurs via OH in the free troposphere
  - Minor players are surface RN and RC emissions (vegetation, biomass burning in the BL)

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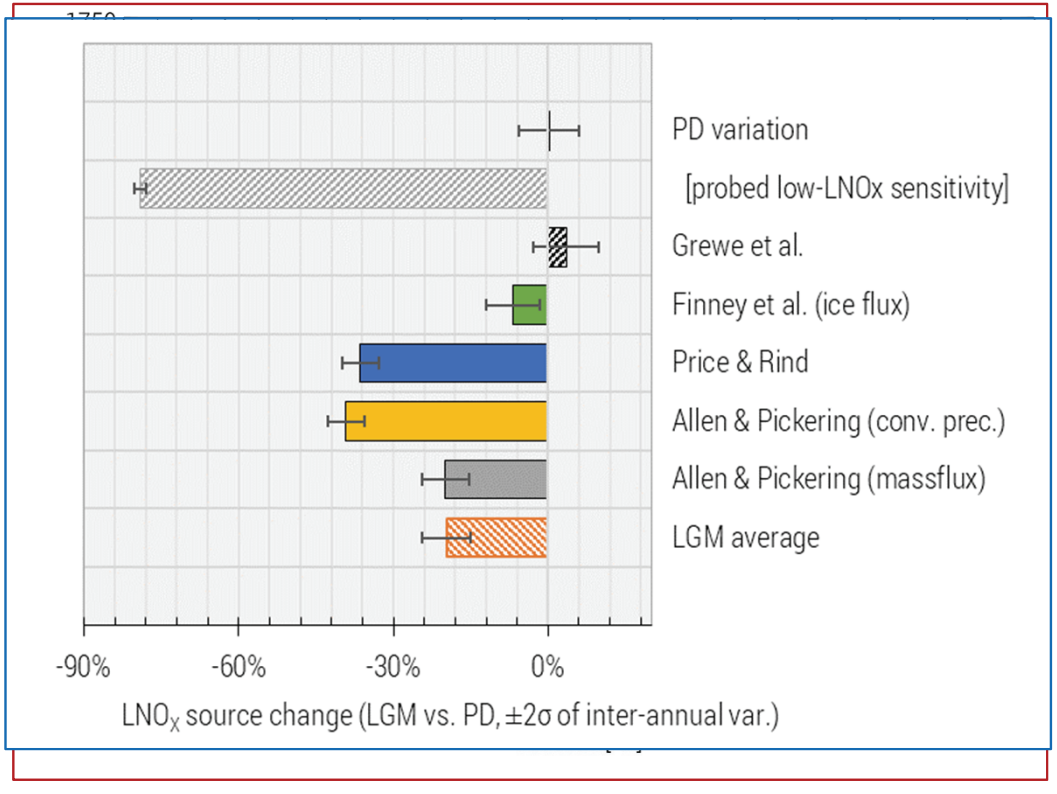
Lightning NO_X source is buffered

- **LNO_X is directly coupled to meteorology**
- **LGM–PD changes are buffered**
  - Shift of high-latitude LNO_X into tropics
- EMAC implements several parameterisations
  - Earlier schemes suggest LNO_X drop by up to 40%
  - Recent schemes: similar to PD
  - Most sensible: Grewe *et al.* & Finney *et al.*
  - Injection latitude is unimportant for $\tau(CH_4)$

LGM LNO_X source is (75–100)% of PD,
or (3.8–5) TgN/yr => $\tau(CH_4)$ is ~(10.5±1) yrs



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EMAC Results

- Analysis of the sensitivity experiments

  $\tau_{fit}(CH_4) = a \cdot (\mathbf{LNO_X} + f_{RNS} \cdot \mathbf{RN_S} + f_{RCS} \cdot \mathbf{RC_S})^p \cdot [B/B_0]$

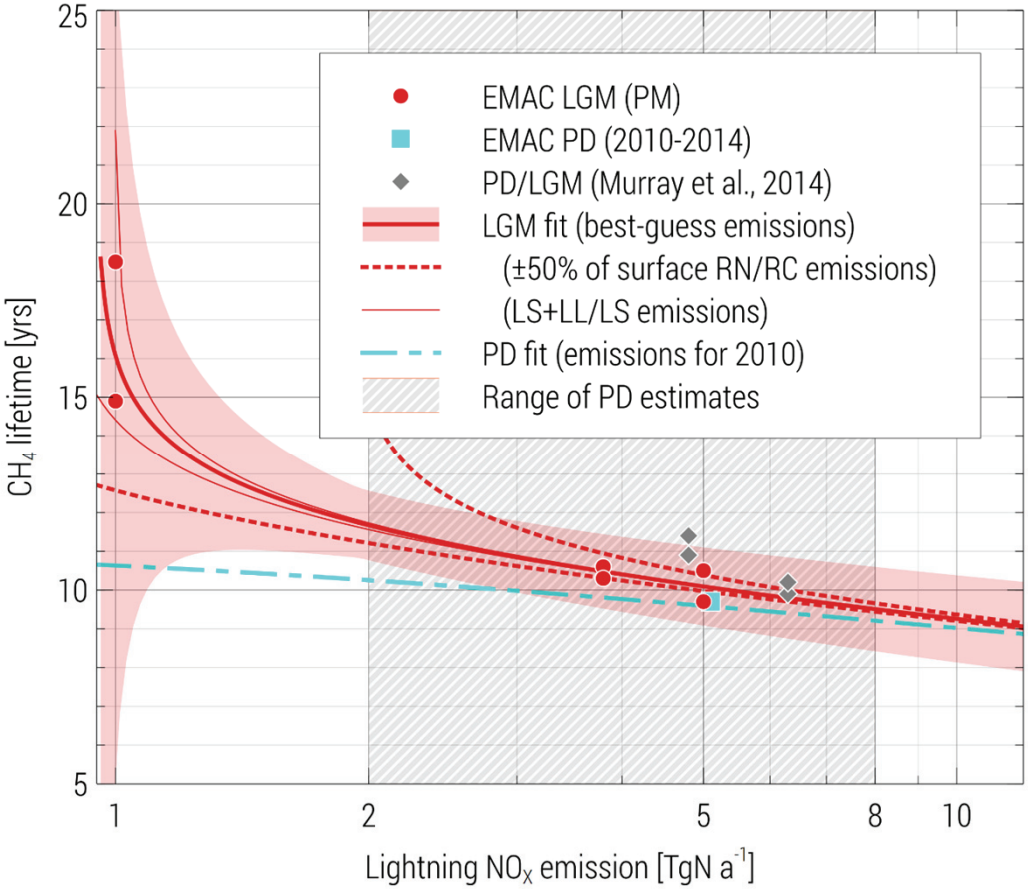
  LNO_X: lightning NO_X emission flux
  RN_S / **RC_S**: surface reactive N and C emission fluxes
  B / B_0 = 1062 Tg(CH_4): CH_4 burdens

- Dependence: $\tau(CH_4) \sim OH^{-1} \sim LNO_X$

  f_{RNS}=+4.2% : OH recycling due to surface RN emis.
  f_{RCS}=−0.16% : OH reduction due VOCs

- Surface RC and RN emissions are influential for $\tau(CH_4)$ only at low LNOx emissions (<2 Tg/yr)

- Fit agrees with the PD conditions



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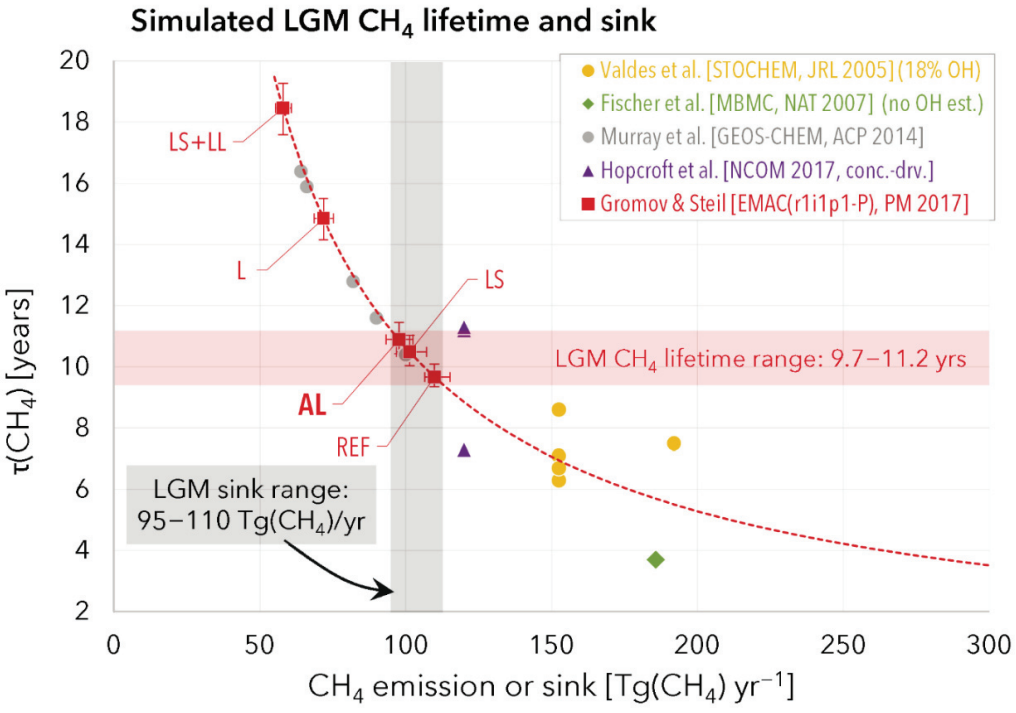
Conclusions

- New results for LGM with comprehensive atmospheric chemistry kinetics in EMAC model
- **Atmospheric oxidative capacity is well buffered by merely one player – lighting NO_X => LGM CH_4 lifetimes are similar to PD**
- Quantification of uncertainties of the total LGM CH_4 source: 95–110 Tg/yr (large shutdown)
- Fast LNO_X calculation => efficient $\tau(CH_4)$ estimates for transient runs



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… and you for your attention!

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