Modelling the abundance of $^{18}O^{18}O$ in the atmosphere and its sensitivity to temperature and O_3 photochemistry

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Modular Earth Submodel System

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Abstract

Atmospheric temperature and ozone photochemistry are recognised to play dominant roles in setting the abundance of $^{18}O^{18}O$ isotopologues (expressed via Δ_{36}) of atmospheric oxygen. Here, we use the AC-GCM EMAC to simulate the abundance of atmospheric $^{18}O^{18}O$ in a most consistent to date kinetic chemistry modelling framework.

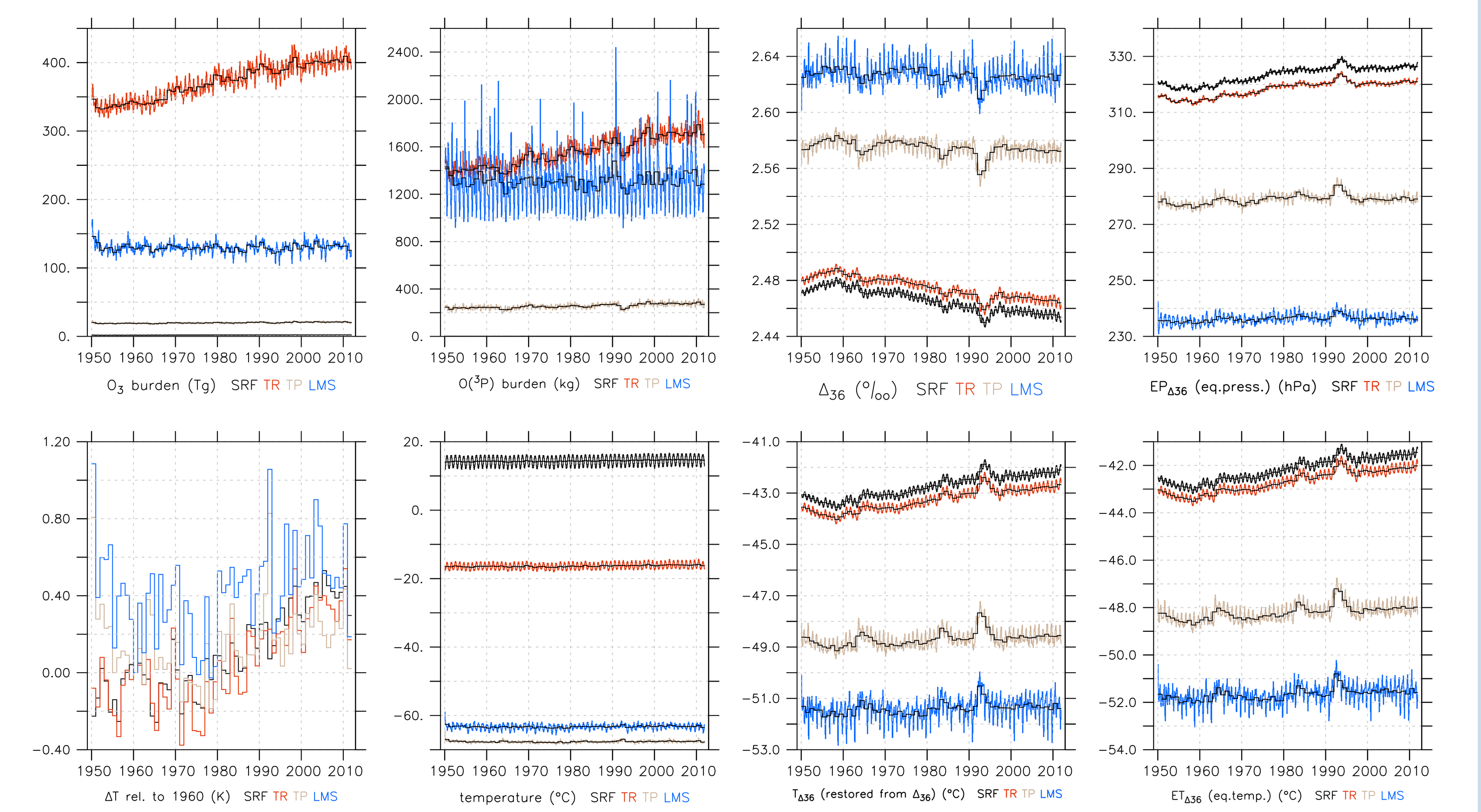
Extensive model diagnostics allow us quantifying contribution of various factors into changes in Δ_{36} since the last 60 years. It is shown that atmospheric dynamics is another fundamental ingredient of atmospheric Δ_{36} distribution.

We discuss potential applications of clumped O_2 composition for quantifying various atmospheric processes like decadal changes in tropospheric O_3 abundance or tropopause warming due to volcanism.

EMAC model / Setup and sensitivity experiments

We use the ECHAM/MESSy Atmospheric Chemistry (EMAC) model [1, v.2.52e]

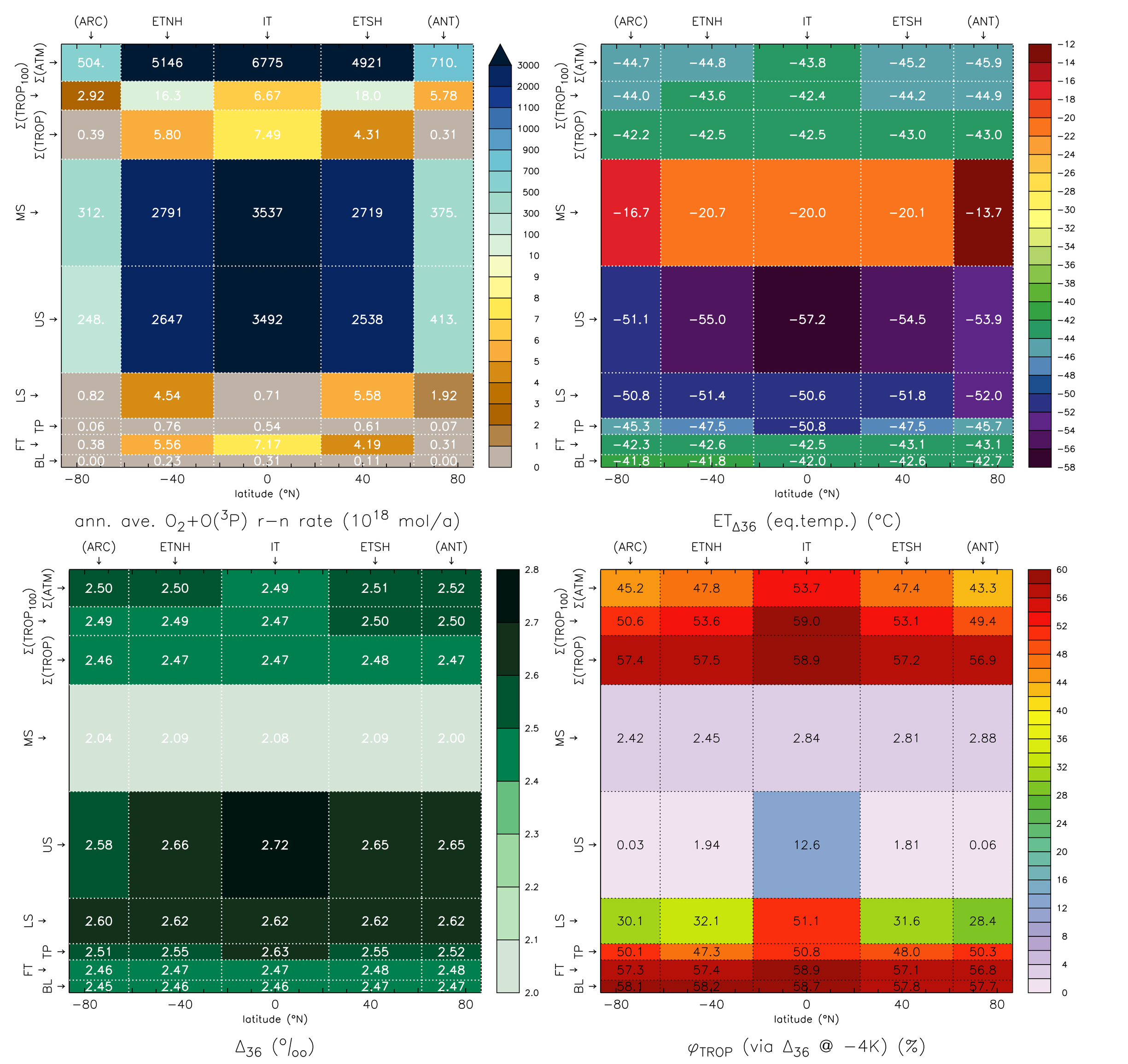
- Based on ESCiMO CCMI setup [2] + output of the **RC1-base-07** experiment
  1960–2011, full chemistry, RCP**6.0**, no specified dynamics (nudging), assimilated SST/SIC, **T42L90MA** up to 80 km
- $^{18}O^{18}O$ and $^{17}O^{17}O$ isotopologues are explicitly simulated (advection + T-dependent equilibrium kinetics in MECCA-TAG submodel [3,4] verified against the fully resolved kinetic scheme of [5])
- Eight $^{18}O^{18}O$ counterparts are added to test the sensitivity of Δ_{36} to changes in temperature (T) and QQ+O(^{3}P) exchange rate (k)
  T: ±4K globally (2x), −4K only in troposphere/overworld (2x)
  k: ±10% globally (2x), −10‰ only in troposphere/overworld (2x)
  Overworld criterion: O_3 >145 ppbv at pressures <500 hPa
- Six diagnostic tracers recording equilibration temperature/pressure (ET/EP, glob./trop./overworld) weighted by the QQ+O(^{3}P) rate
- One $^{18}O^{18}O$ counterpart tracer $^C\Delta_{36}$ reacting with average 1950–1960 "climatological" O(^{3}P) to test for changes induced by growing O_3



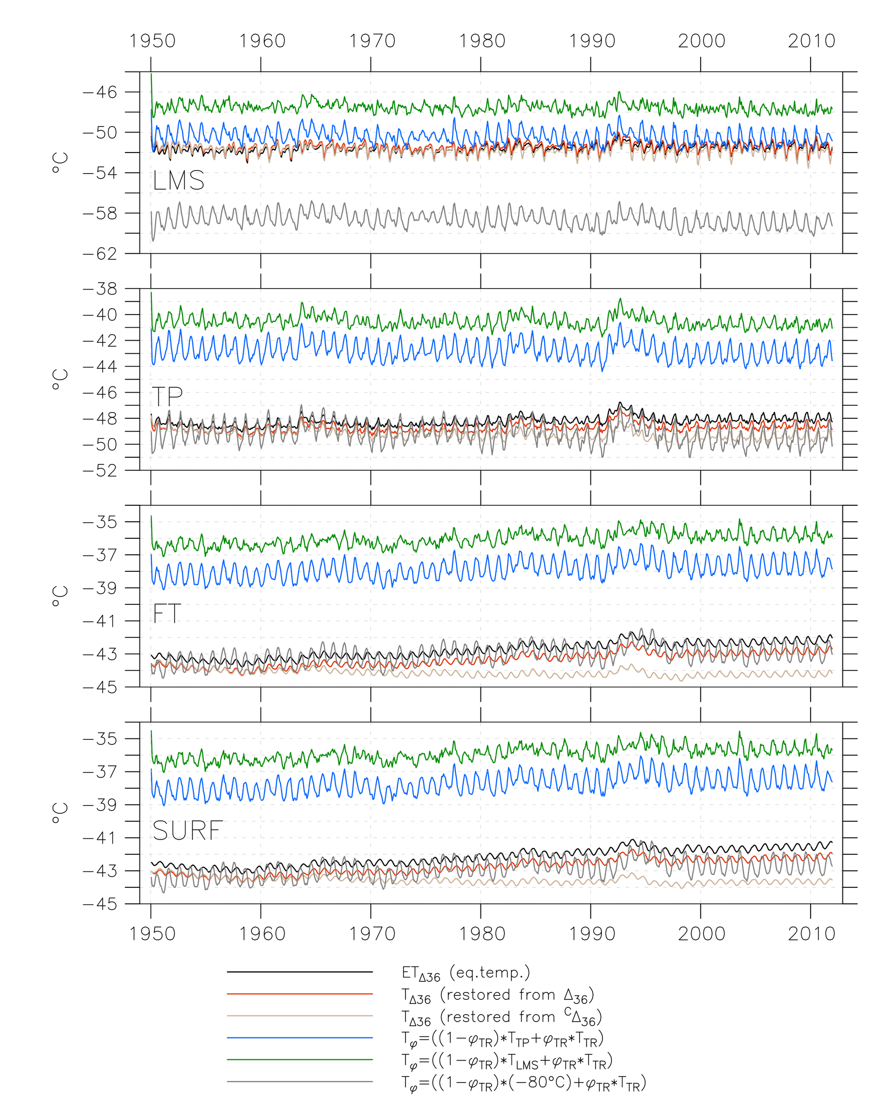
Atmospheric integrals of relevant parameters/burdens simulated in EMAC in various domains (SRF: surface, TR: troposphere, TP: tropopause, LMS: lowermost stratosphere)

Δ_{36} distribution: kinetics vs. transport

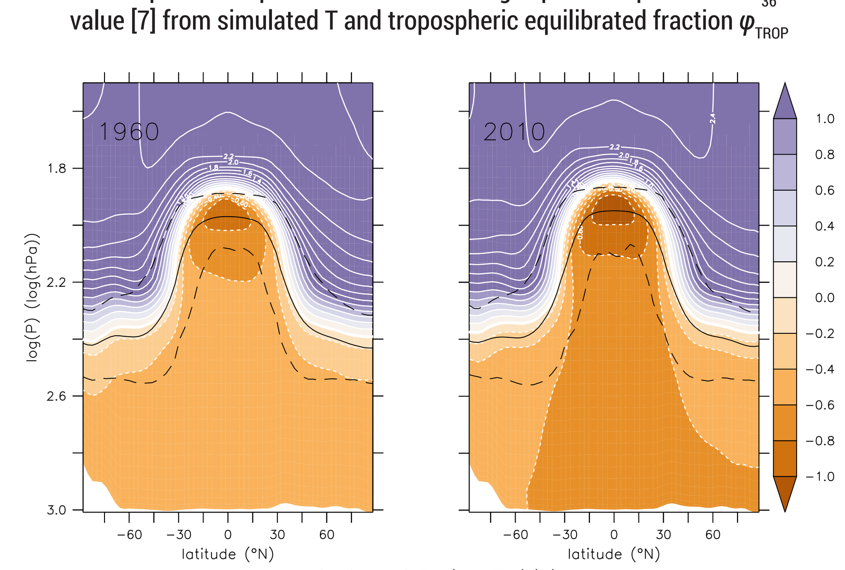
- Most of equilibration (absolute) occurs in the overworld (LMS, MS)
- Fraction of LMS-equilibrated O_2 exchanges with troposphere and vice versa
- Troposphere/overworld-only −4K sensitivity tracers allow deriving the fraction of Δ_{36} reset in/advected to respective domains (φ_{TROP} and φ_{OW})
- Simulated Δ_{36} and equilibration temperature correlate in the upper/middle stratosphere, but not in the LMS and troposphere due to transport
  => Estimates based on static T/rate distributions (e.g. [6]) are unrealistic!



Annual averages of the zonal/vertical integrals related to Δ_{36} simulated in EMAC. Σ denotes sum over the given domain. $TROP_{100}$ is troposphere under 100 hPa.



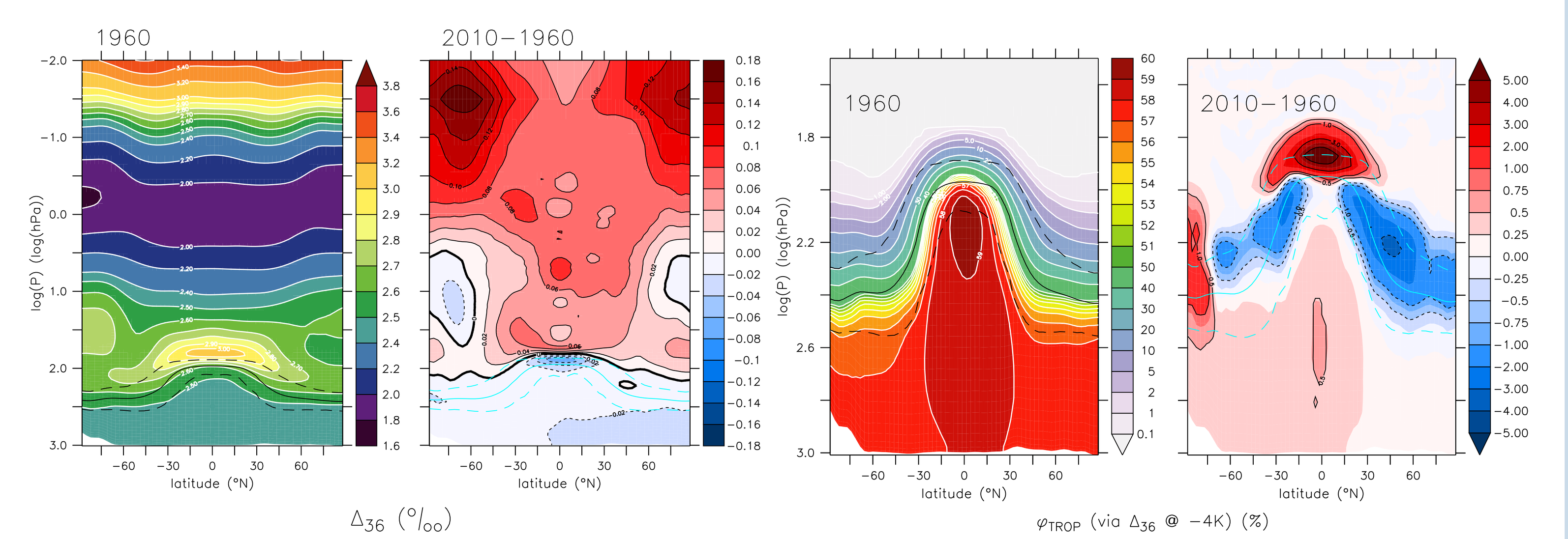
Atmospheric temperatures restored using expected equilibrated Δ_{36} value [7] from simulated T and tropospheric equilibrated fraction φ_{TROP}



Annual zonal average of the non-linearity error in T restored from simulated Δ_{36} in 1960 (left) and 2010 (right), diagnosed using the equilibration temperature (ET) tracer

Recovering temperatures from Δ_{36} signal?

- Δ_{36} is a composite signal of O_2 equilibrated in troposphere and overworld; input proportions depend on the domain (see Fig. on the left)
- T restored from simulated Δ_{36} exhibits mixing effects (due to non-linear T-dependent equilibration kinetics, see [7])
  - underestimation <1K in the troposphere (largest at tropical tropopause)
  - overestimation >2K in the overworld
  - effects are smallest in the LMS
- Tropospheric temperature can be roughly derived taking average "overworld" temperature of about −80°C, but not that of LMS or tropopause
- Long-term changes in restored T are seen only in troposphere



Annual zonal averages of the local Δ_{36} value (left) and its fraction equilibrated in the troposphere (right). Left and right panels present the 1960 values and 2010-1960 absolute changes, respectively.

Δ_{36} sensitivities & short- and long-term excursions

Using the ensemble of counterpart tracers allows to test sensitivity of Δ_{36} to changes in atmospheric temperature and equilibration rate

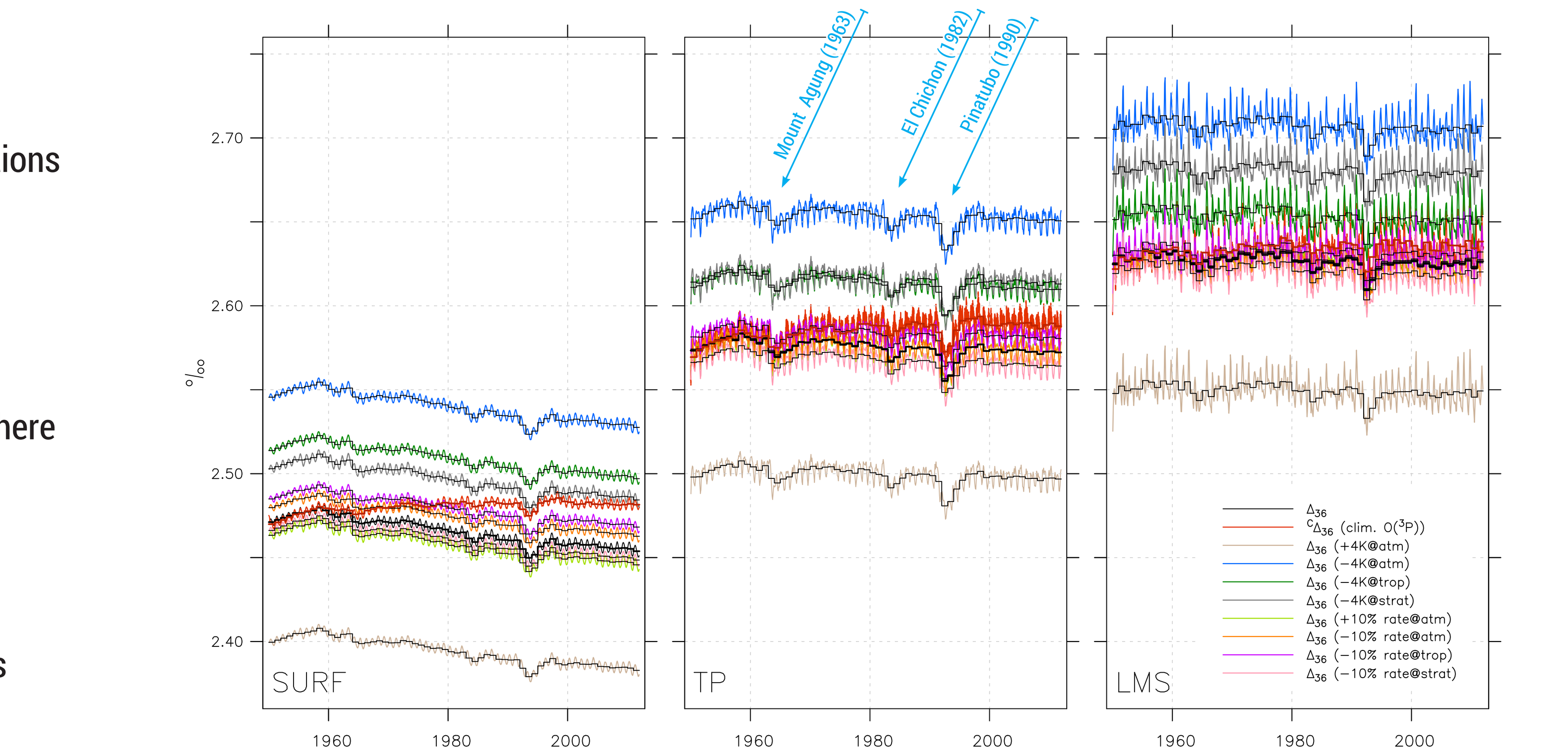
- Strong T sensitivity (−0.02 ‰/+1K) compared to that for rate (−0.00125‰/+1%)
- Sensitivities are similar in all domains (stronger only for -4K@atm in LMS)
- Troposphere- and overworld-only sensitivities are additive => allows studying contributions of different vertical/zonal domains
- Annual variation in Δ_{36} increases with altitude <= dampened equilibration rates and increased mixing in the troposphere

Short-term (several years) lowering in Δ_{36} coincide with large eruptions

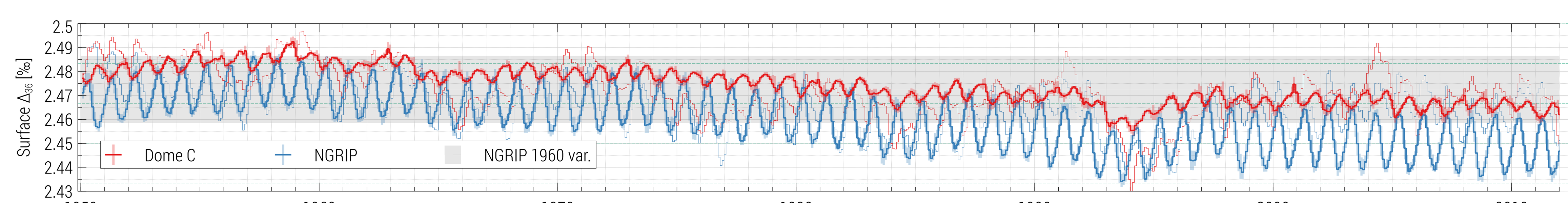
- Recorded T signal (via historical SST forcing) is most pronounced in the upper troposphere
- Largest Δ decrease up to 0.03‰ in the TP seen after Mt. Pinatubo (1990) eruption
- Local signals are stronger than those shown for domain integrals

Long-term (1950–2011) change in Δ_{36} is of O_3 origin

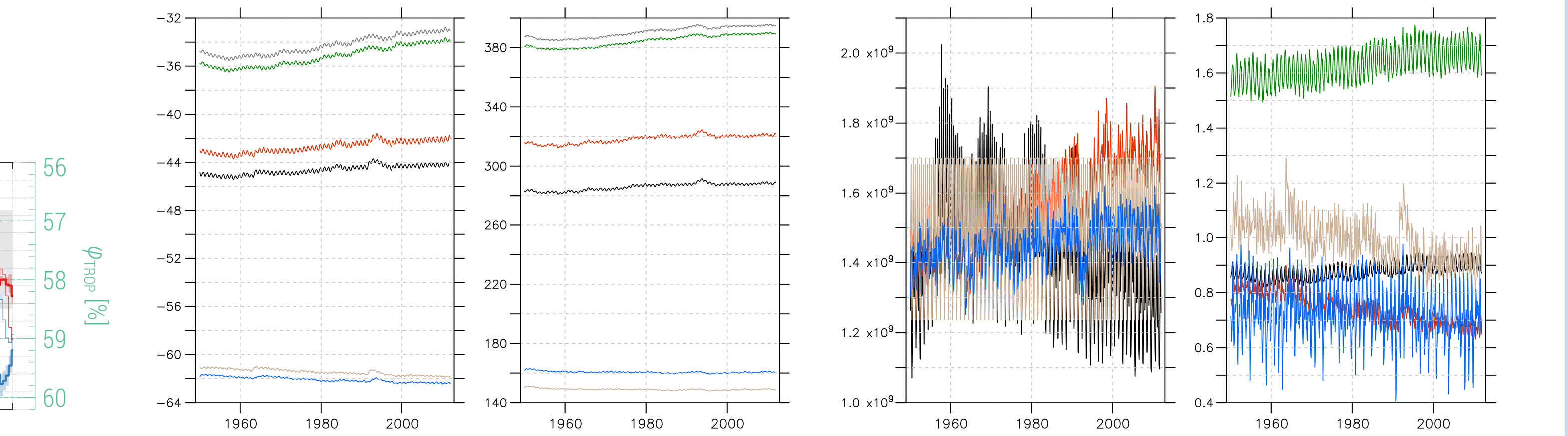
- Decadal trend (−0.03‰/60 yrs) in Δ_{36} is seen only in tropospheric domain and coincides with increase in exchange rate / tropospheric O(^{3}P) (~+7%) and O_3 (~+15%)
- T- and P-equilibration tracers indicate the shift of O_2 equilibration into the troposphere (warming for tropospheric share of Δ_{36} and cooling for the overworld)
- LMS and whole-atmosphere (ATM) O_2 equilibration rate, however, decreases (?)
- Surface polar Δ_{36} variation and trend is larger in the NH (NGRIP) than in SH (Dome C)



Ensemble of Δ_{36} sensitivity values simulated for perturbed T and QQ+O(^{3}P) rate conditions globally, in the troposphere and overworld. Results for reference and climatological O(^{3}P) conditions are shown in black and red, respectively.



Surface Δ_{36} values simulated for 1950–2011 with EMAC at NGRIP and Dome C locations (thick lines, left axis, monthly averages, error bars denote monthly variation) and tropospheric share of Δ_{36} value (thin lines, right axis, respectively). Shaded area denotes variation at NGRIP in 1960.



Equilibration temperature and pressure (ET/EP tracers, left panels), O(^{3}P) burden and isotope exchange turnover time (τ) over selected domains simulated in EMAC. Superscripts S,T,C denote sensitivities in troposphere, overworld and to climatological O(^{3}P).

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