CRI-Strat: a new chemistry mechanism for UKESM

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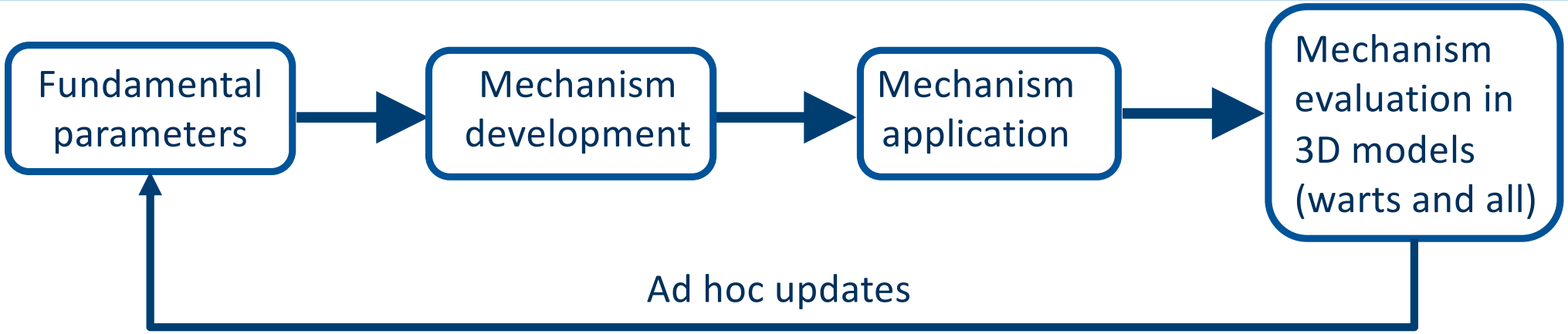
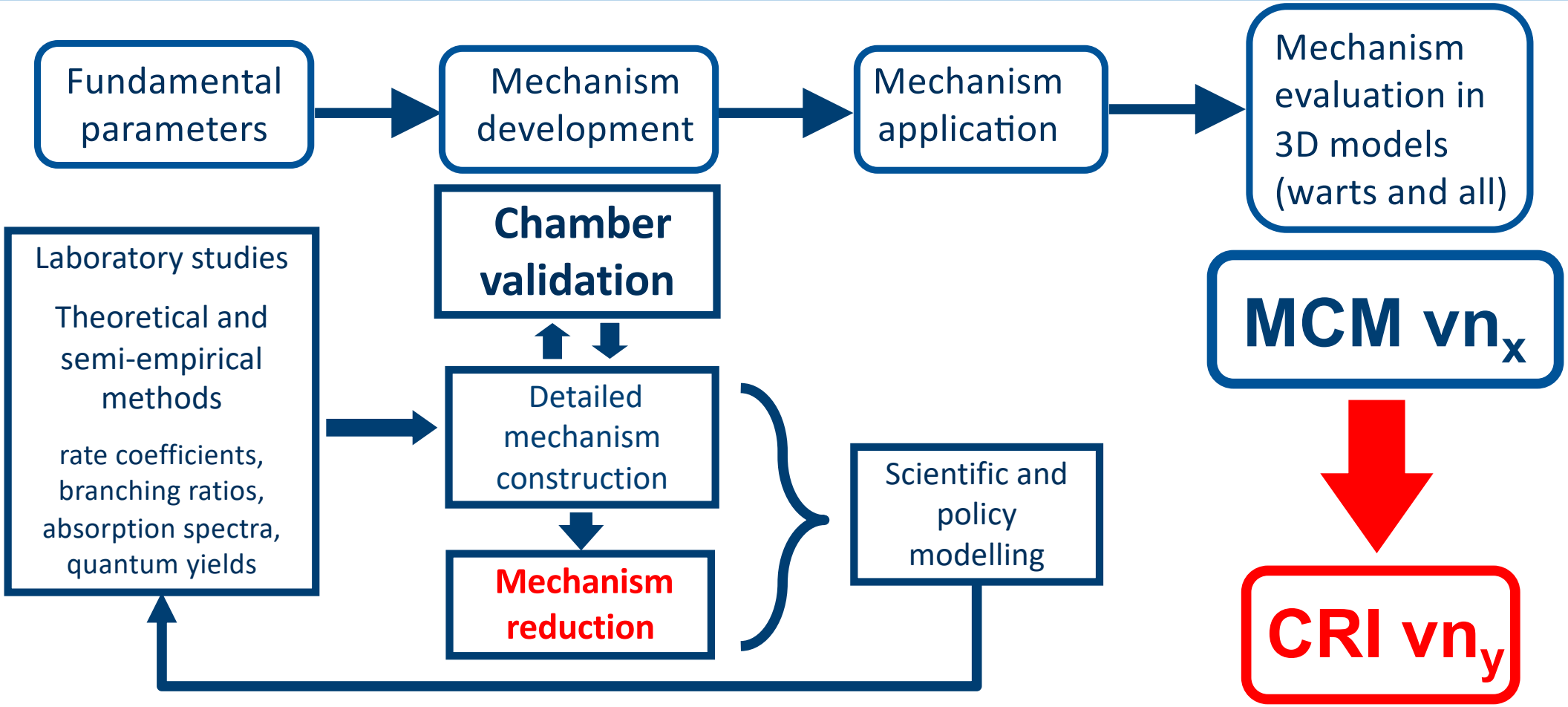
“Traditional” Chemical mechanism development (e.g. for StratTrop)



Issues

- How can we be sure the most “important” reactions are captured?
- Impossible to separate biases due to mechanism from other model errors.
- Risk of compounding errors as mechanism is expanded - different levels of complexity or sources of kinetic data for different aspects of chemistry.
- No **traceability** to explicit mechanisms.

The Common Representative Intermediates Approach



CRI Implementation in UKCA

Common Representative Intermediates (CRIv2.1-R5), extended with Stratospheric & aerosol chemistry to make CRI-Strat+GLOMAP.

Traceable to the Master Chemical Mechanism (MCMv3.2).

More than 2x as complicated, runs 80% slower – UKESM benchmark simulations doable

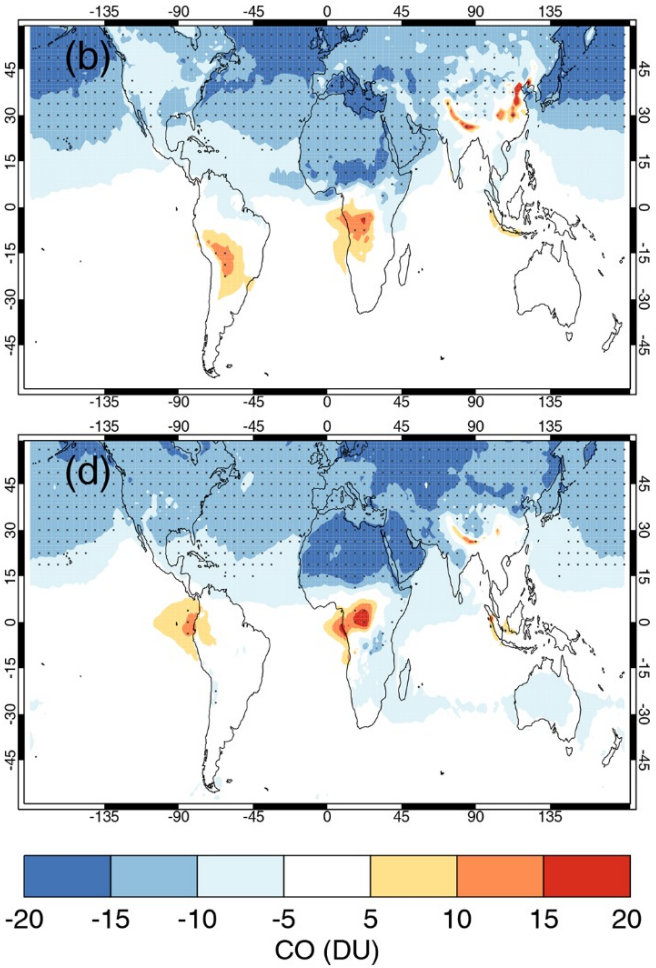
Ready to use from UM version 11.8

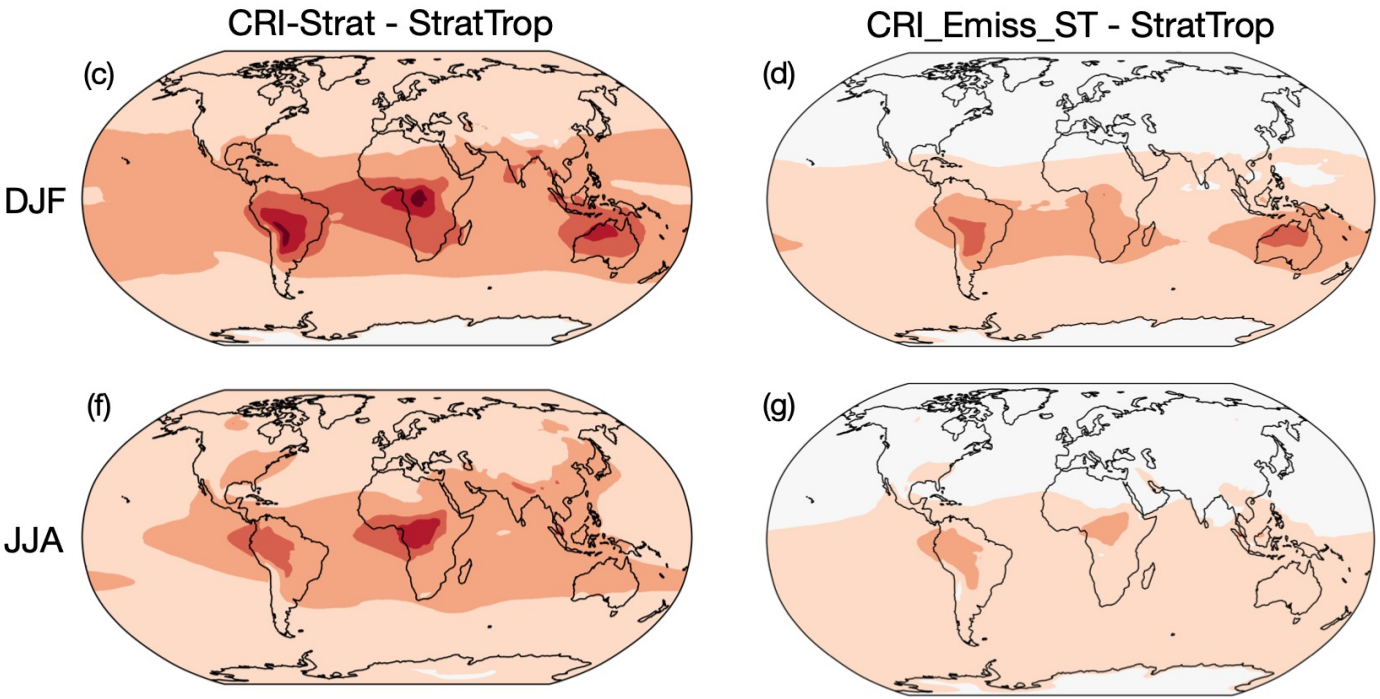
Additional chemistry to StratTrop:

- Alkanes & alkenes up to C4
- Aromatics (benzene, toluene & o-xylene).
- Detailed BVOC chemistry.
- Organic nitrates (RO2+NO → RONO2) for all relevant species.
- Self- and cross- reactions parameterized for all 47 RO2 species.
- **All** available VOC emissions used.

Carbon Monoxide & methane lifetime

StratTrop – MOPITT, from Archibald et al., (2020)





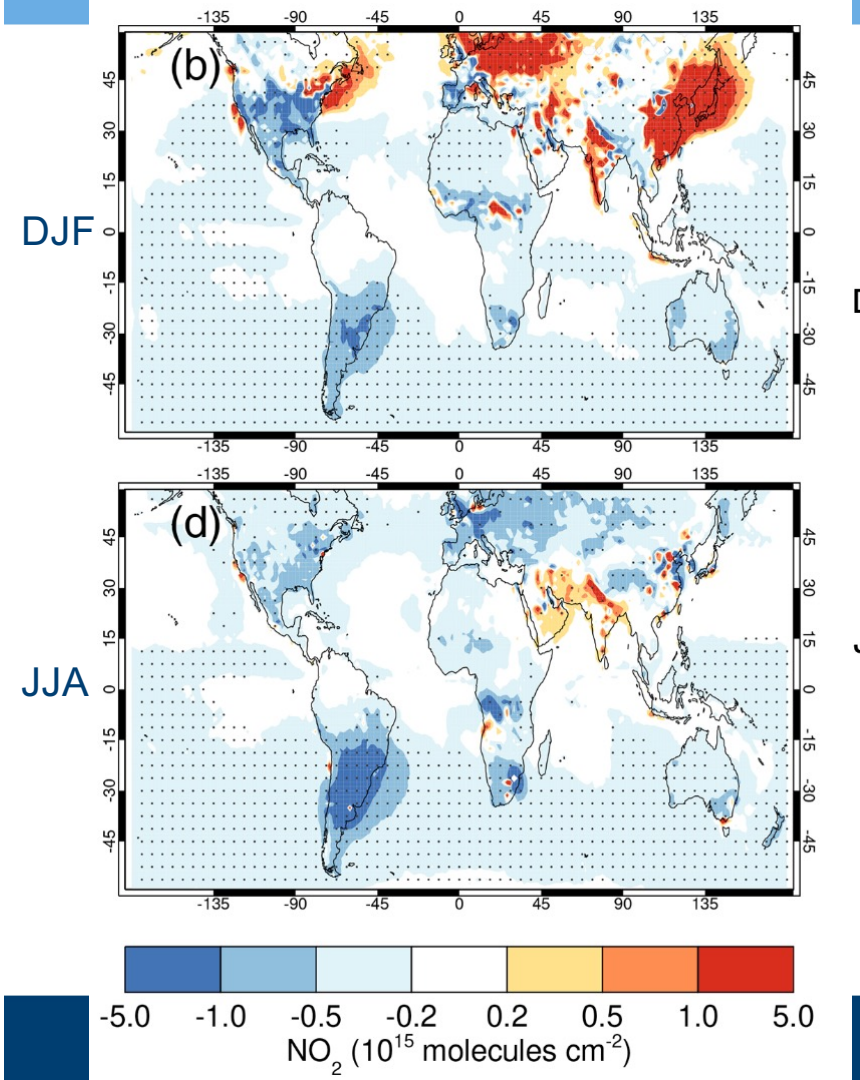
Greater CO in CRI-Strat due to secondary production, particularly from more explicit isoprene and monoterpene chemistry.

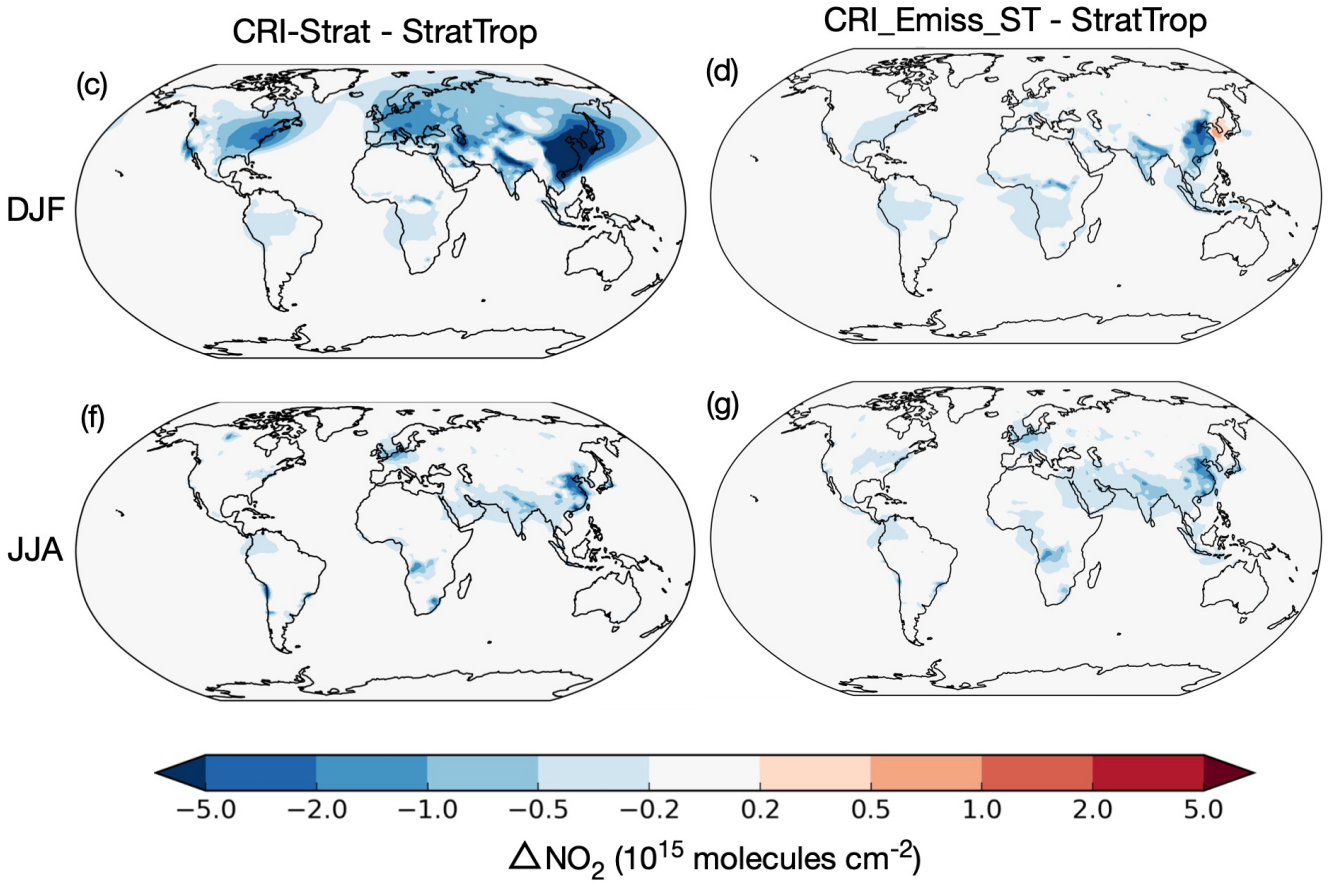
Important for oxidant budget

	CRI-Strat	StratTrop
[OH] (10^6 molecules cm^{-3})	1.363	1.339
OH NH:SH ratio	1.38	1.35
[HO_2] (pptv)	6.26	5.90
OH : HO_2 ratio (%)	1.56	1.67
CH_4 lifetime W.R.T. OH (years)	7.65	8.13
$HO_2 + HO_2$ flux (P mole $year^{-1}$)	41.9	32.2

Tropospheric Column NO2

StratTrop – OMI, from Archibald et al., (2020)



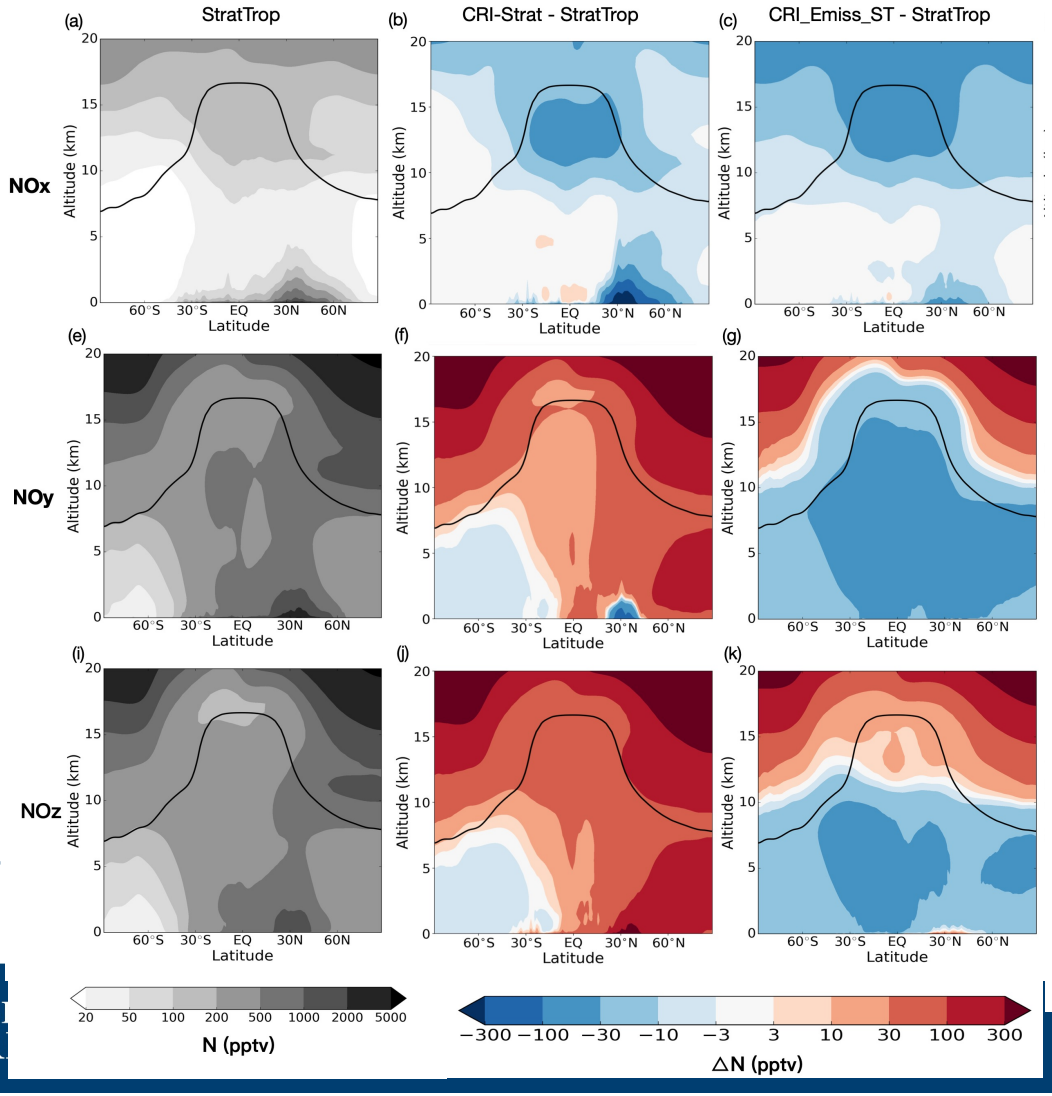


NOy burden

NOx = NO+NO2

NOy = NOx+NOz

NOz = NO_3 + $2N_2O_5$ + HNO_3 + PAN + RNO_3 + …



In CRI-Strat: extra NMVOCs & RONO2 mean more NOx stored in long-lived reservoirs => less NOx, more NOz

If CRI-Strat use identical emissions to StratTrop, HNO3 formed faster => Greater deposition in polluted regions leads to net loss of NOy

Nox burden depends on chemistry **and** VOC emissions, not just NOx emissions

Summary and Conclusions

- Comprehensive CRI-Strat mechanism, with detailed NMVOC chemistry built from CRIv2.1-R5, implemented in UKCA.
- Paradigm shift in mechanism development - ozone production **traceable** to MCMv3.2.
- Improved aromatic and BVOC oxidation chemistry
  - Can be used to improve links between **chemistry**, **oxidants** and **aerosol formation**
- Much more detailed representation of **NMVOCs**, full use of emission inventory data
  - We know these NMVOC emissions are important for **O3, NOx, CO & HOx** => therefore **climate**
- Slower and more complex to run/analyse, runs ~1.8x slower. However, long-term and high-res runs possible
  - **Benchmarking** => use for one or two key simulations. Are radiative forcing and climate sensitivity dependent on chemical mechanism?
- Being at the forefront the field – most comprehensive chemistry of all CMIP models opens up science questions:
  - **Global air quality and future change, biosphere/atmosphere interactions, SOA formation**

CRI-Strat (v2.1) is useable in version UM from version 11.7, first committed in ticket #4231, bug fix #5523.

See Archer-Nicholls et al., JAMES, 2021 for details (*in review*, preprint: doi:10.1002/essoar.10505092.1)

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EXTRA SLIDES

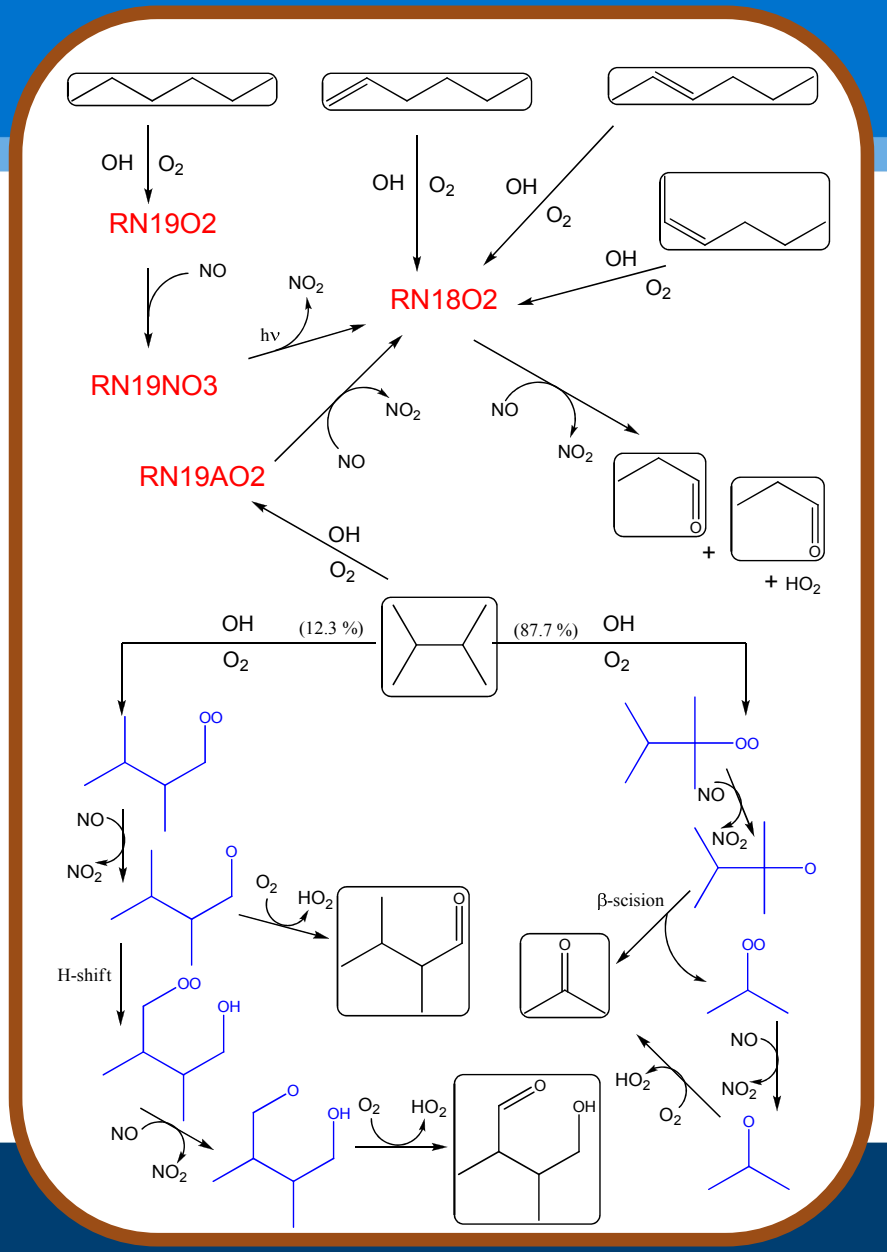
Common Representative Intermediates

Master Chemical Mechanism (MCM) chemistry highlights the complex oxidation of one C_6 alkane.

The CRI approach is one which conserves the O_3 forming potential of VOCs. Lumping the intermediate reactive products (RO_2 and their daughters).

Each version of CRI fully **traceable** to a version of the MCM

Further reductions: primary VOCs are lumped only if ozone production is conserved, as tested by rigorous evaluation against MCM.



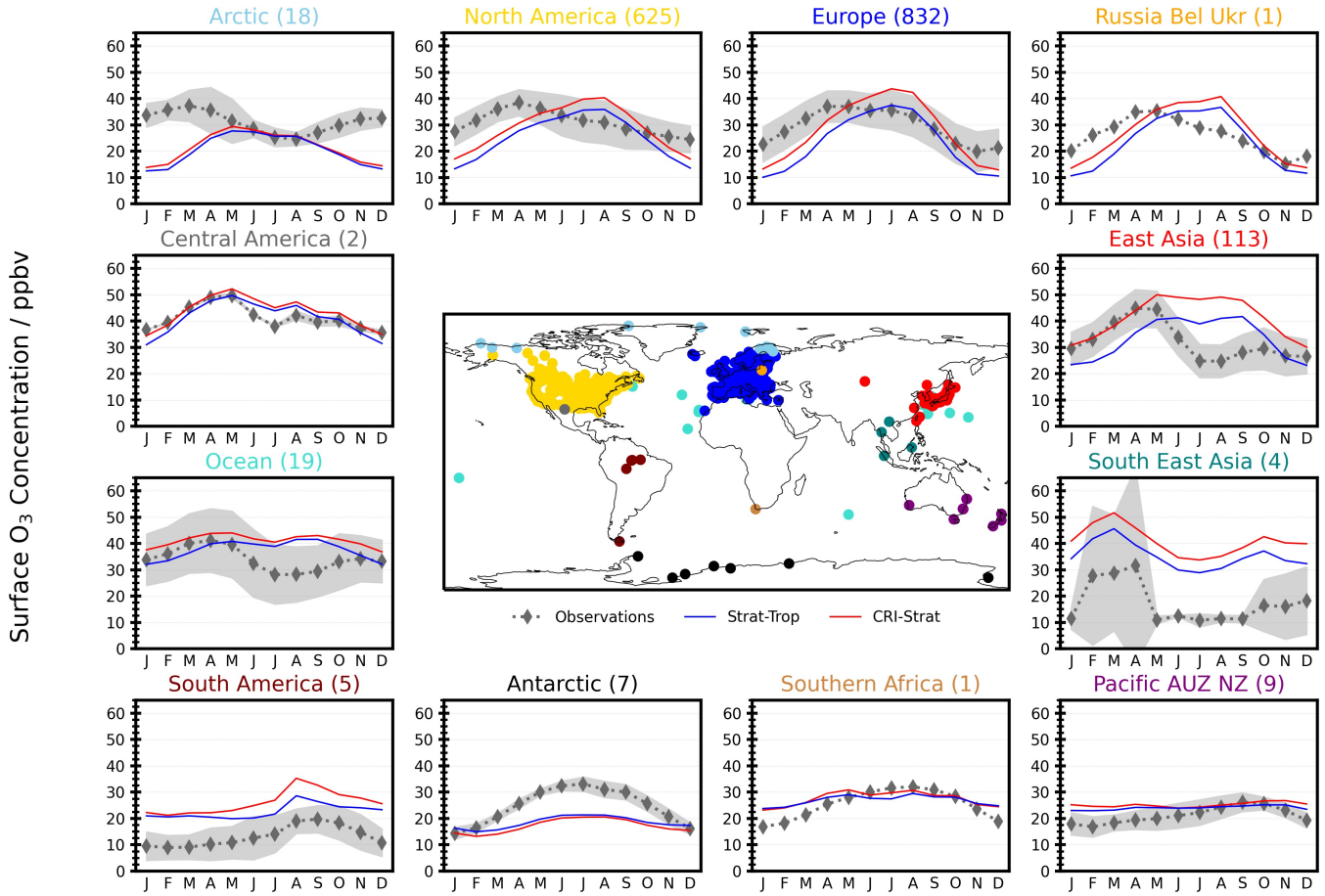
Experiments

- Comparing StratTrop and CRIv2-R5+Stratosphere (CRI-Strat) simulations, both with GLOMAP aerosol at UM vn10.9.
- N96 (1.875˚ x 1.25˚) 85 vertical levels.
- Nudged meteorology, 2009-2019.
- 2014 CEDS emissions, as used for CMIP6.
- All NMVOC emission classes mapped in CRI-Strat vs StratTrop:
  - **70.5 vs 27.9 GgC/yr anthropogenic.**
  - **40.6 vs 23.9 GgC/yr biomass burning.**
  - **900.6 vs 710.6 GgC/yr biogenic.**

Scenario	Mechanism	NMVOC Emissions (Tg C yr^{-1})
CRI-Strat	CRI-Strat	1012 - CRI speciation
StratTrop	StratTrop	762 - ST speciation
CRI_Emiss_ST	CRI-Strat	762 - ST speciation
CRI_Emiss_C2C3	CRI-Strat	762 - CRI speciation

Use additional runs to differentiate impact of chemistry vs NMVOC emissions

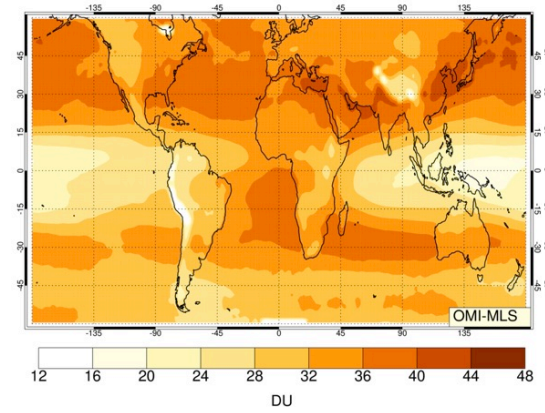
TOAR rural comparison

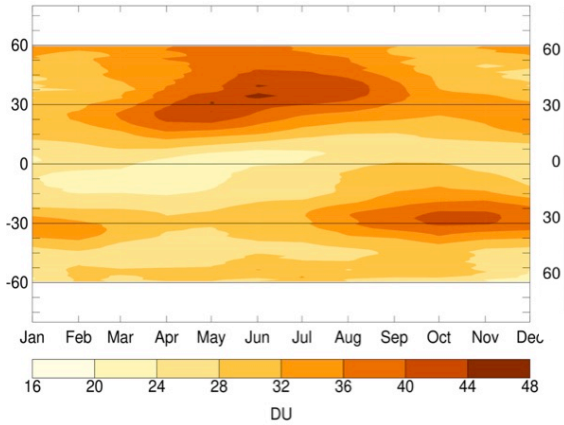


- Year-round increase in surface ozone in most regions
- Reduces winter low bias but increases summer high bias
- Production *and* loss of ozone increase in CRI-Strat

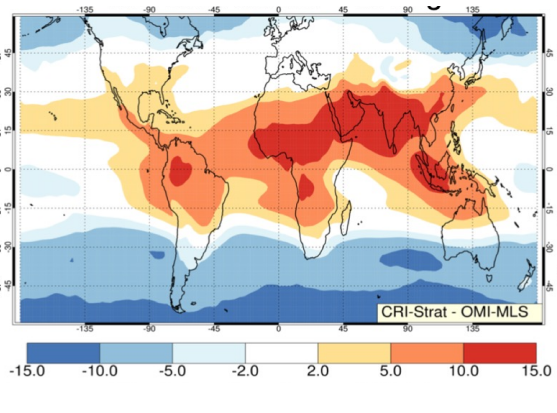
Tropospheric ozone column vs OMI-MLS

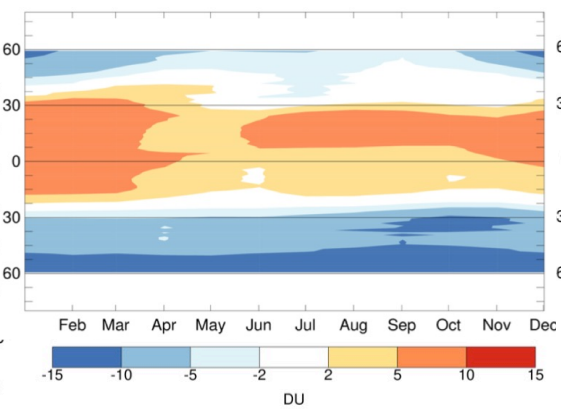
OMI-MLS
60N:60S Burden = 301 Tg



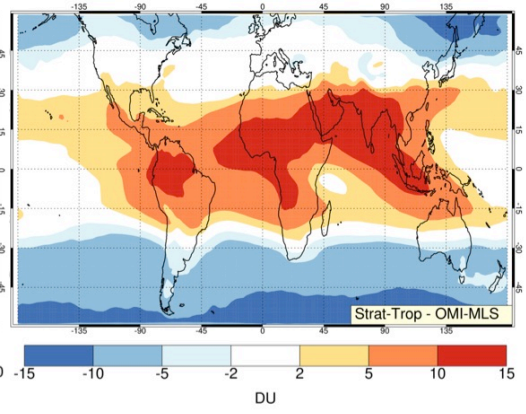


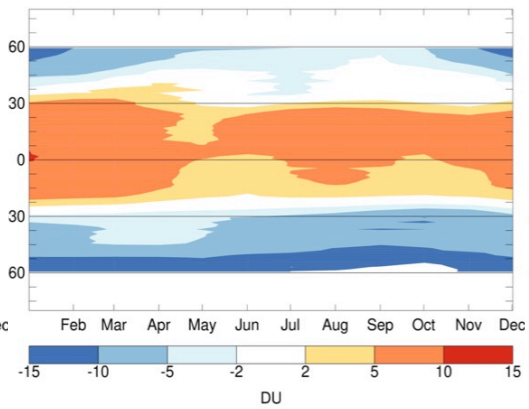
CRI-Strat – OMI-MLS
CS Burden = 303 Tg





StratTrop – OMI-MLS
ST Burden = 308 Tg





CRI-Strat - StratTrop