A two-dimensional global model to quantify halocarbon emissions

Luke Western

Global Monitoring Lab, NOAA; University of Bristol

luke.western@noaa.gov

Scott Bachman (NCAR), Matt Rigby (Univ. of Bristol), Stephen Montzka (NOAA)

Background

- Global emissions were estimated for nearly 50 non-CO_2 greenhouse gases and ozone depleting substances for the 2022 Scientific Assessment of Ozone Depletion
- Ozone assessment is every 4 years, plus many additional scientific papers
- Emissions are derived using 2 global networks of surface mole fraction measurements and a model of atmospheric transport
- Routine emissions updates are performed by ~3 people globally

Controlled production

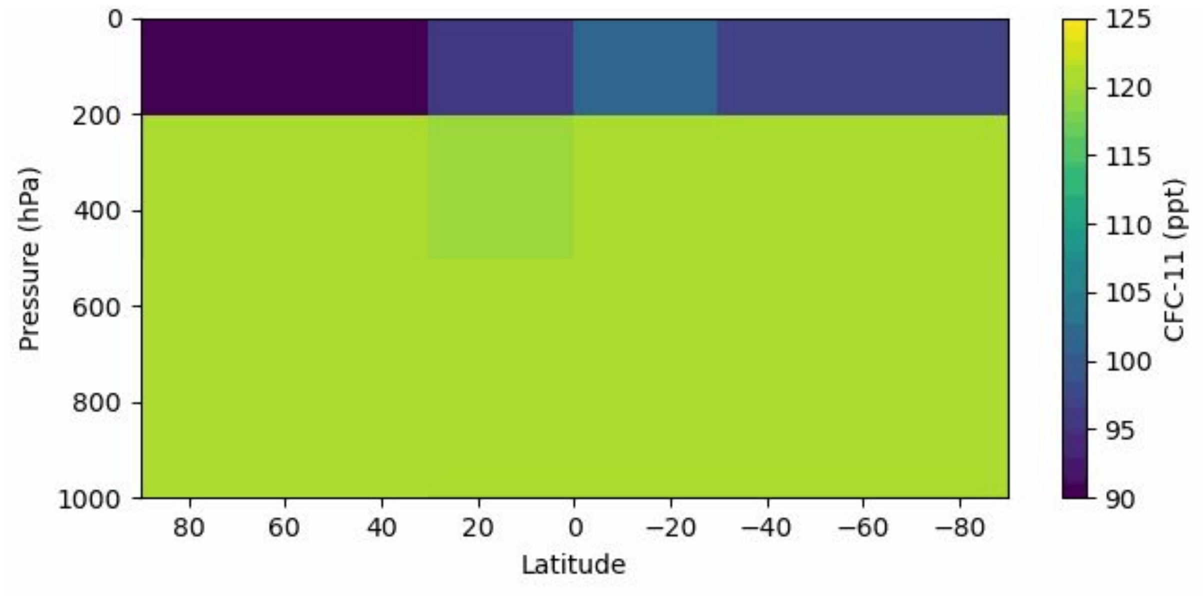
CFC-11
CFC-12
CFC-13
CFC-113
CFC-114
CFC-115
HCFC-124
HCFC-133a
HCFC-132b
HCFC-141b
HCFC-142b
HCFC-22
HCFC-31
H-1211
H-1301
H-2402
CH_3CCl_3
CCl_4

HFC-125
HFC-227ea
HFC-32
HFC-134a
HFC-23
HFC-365mfc
HFC-143a
HFC-236fa
HFC-4310mee
HFC-152a
HFC-245fa

$CHCl_3$
CH3Cl
$CH2Cl_2$
PCE
CH_3I
CH_3Br
Desflurane
SO_2F_2
SF_5CF_3
NF_3
CF_4
C_2F_6
C_3F_8
c-C_4F_8
C_6F_{14}
CH_4
N_2O
SF_6

Current practice

- Box models generally provide models of atmospheric transport
- Combined with surface measurements and inversion framework to derive emissions
- Annually repeating parameterised dynamics
- Lack any large-scale variability
- Models consist of 1-12 boxes
- Limited representation in space and time

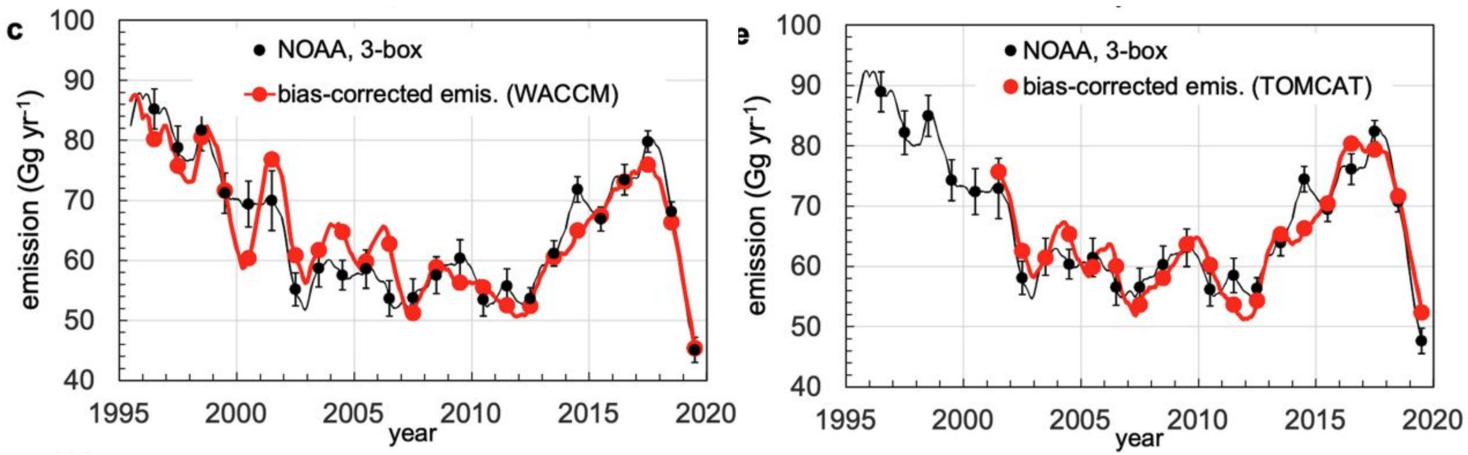


Way forward

- Need a model that:
  - better represents large-scale dynamics
  - has better spatial and temporal resolution
  - is fast running (inferring 10° latitudinal emissions over 50-year period requires running for >20,000 model years)

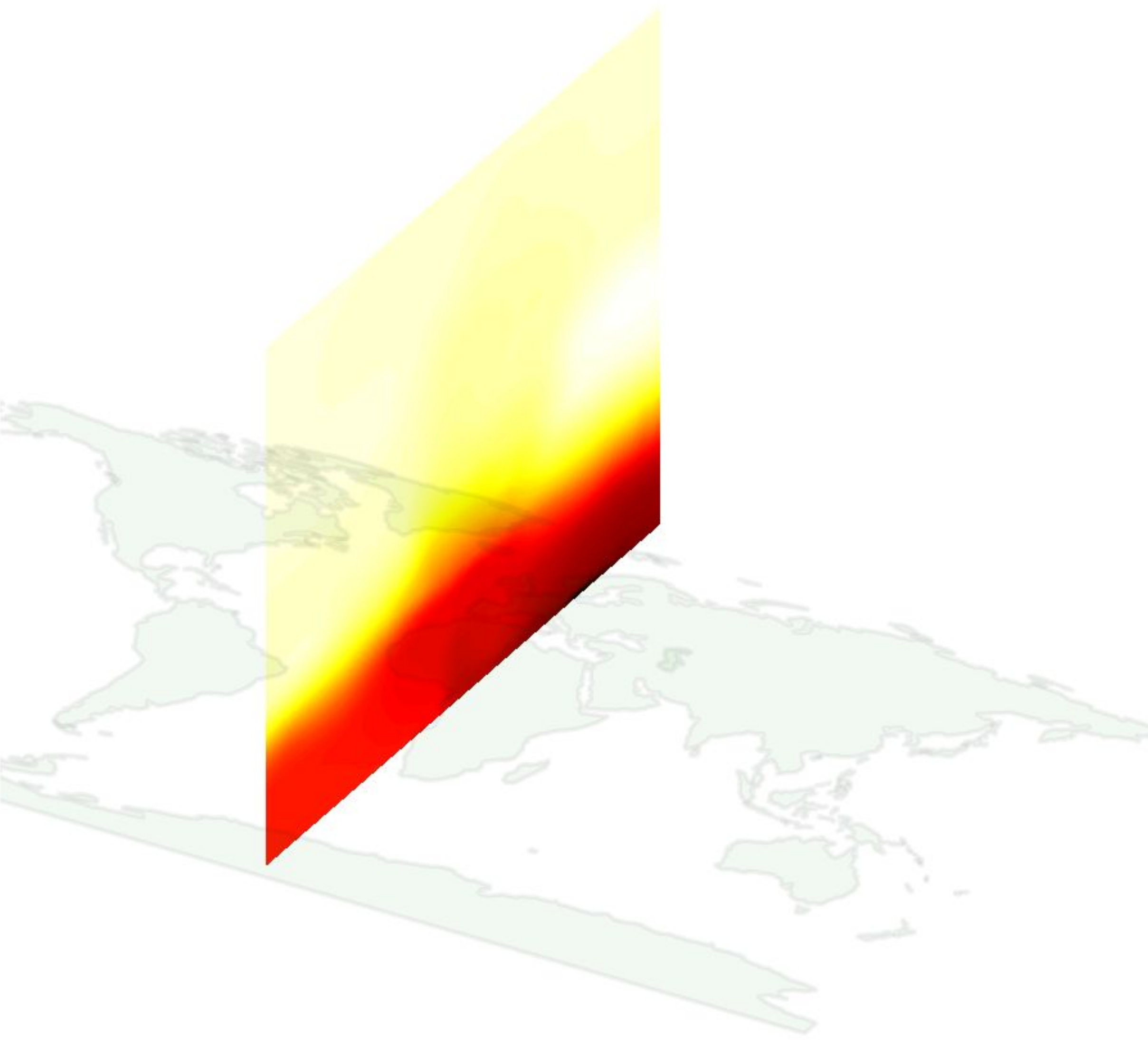
The influence of the stratospheric Quasi-Biennial Oscillation on trace gas levels at the Earth's surface

How Atmospheric Chemistry and Transport Drive Surface Variability of N_2O and CFC-11



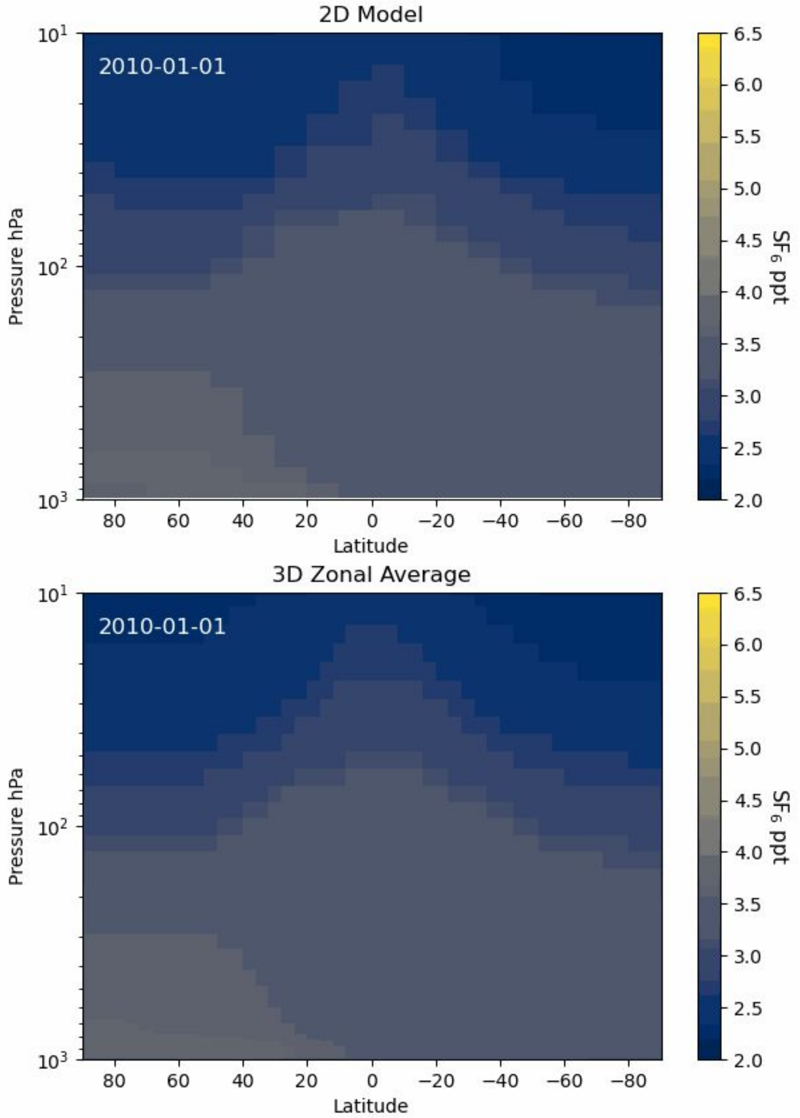
Model description

- Two-dimensional (zonal mean) model of atmospheric transport (10° x ~1.2km)
- Driven by MERRA reanalysis fields
- Monthly varying transport
- Eddy transport processes derived from tracer experiments in GEOS-Chem
- Progress with 2D atmospheric models stagnated in 90s; progress has been made in ocean literature
- Offline chemistry



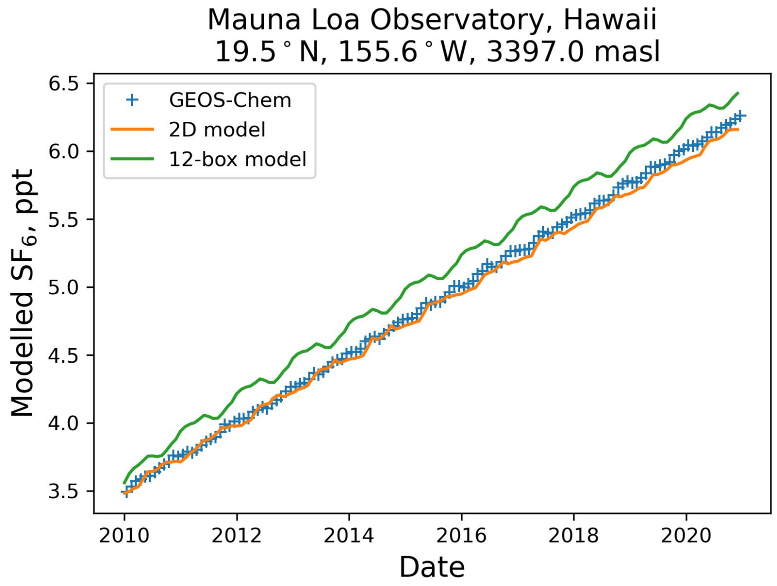
Derivation of model parameters

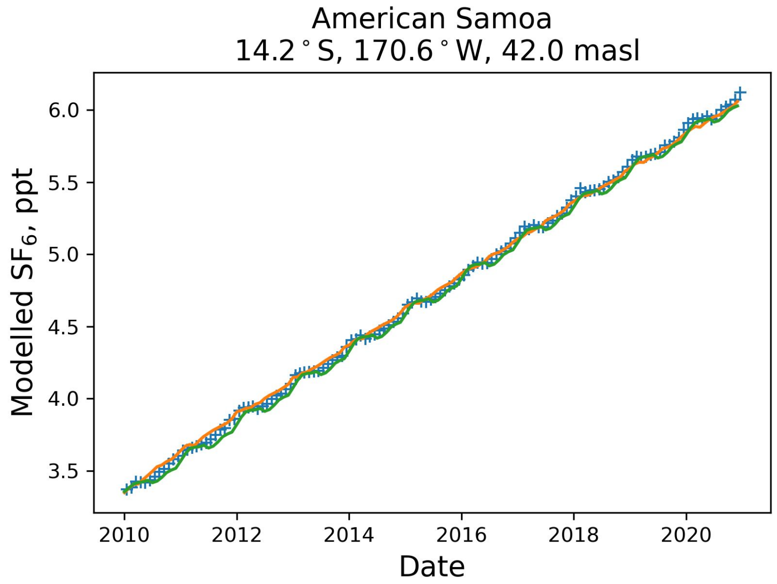
- Eddy flux tensor derived from ensemble of orthogonal tracer experiments
- Residual velocities and diffusion derived from this
- 2D model has mixed derivative diffusive term $\left(D_{yz}\frac{\partial^2 q}{\partial y \partial z}\right)$, which has generally been neglected or unphysical in 2D models
- A new positivity, mass and concentration-preserving mixed derivate diffusion implemented
- Offline losses taken from literature



Validation: Comparison of modelled surface mole fractions

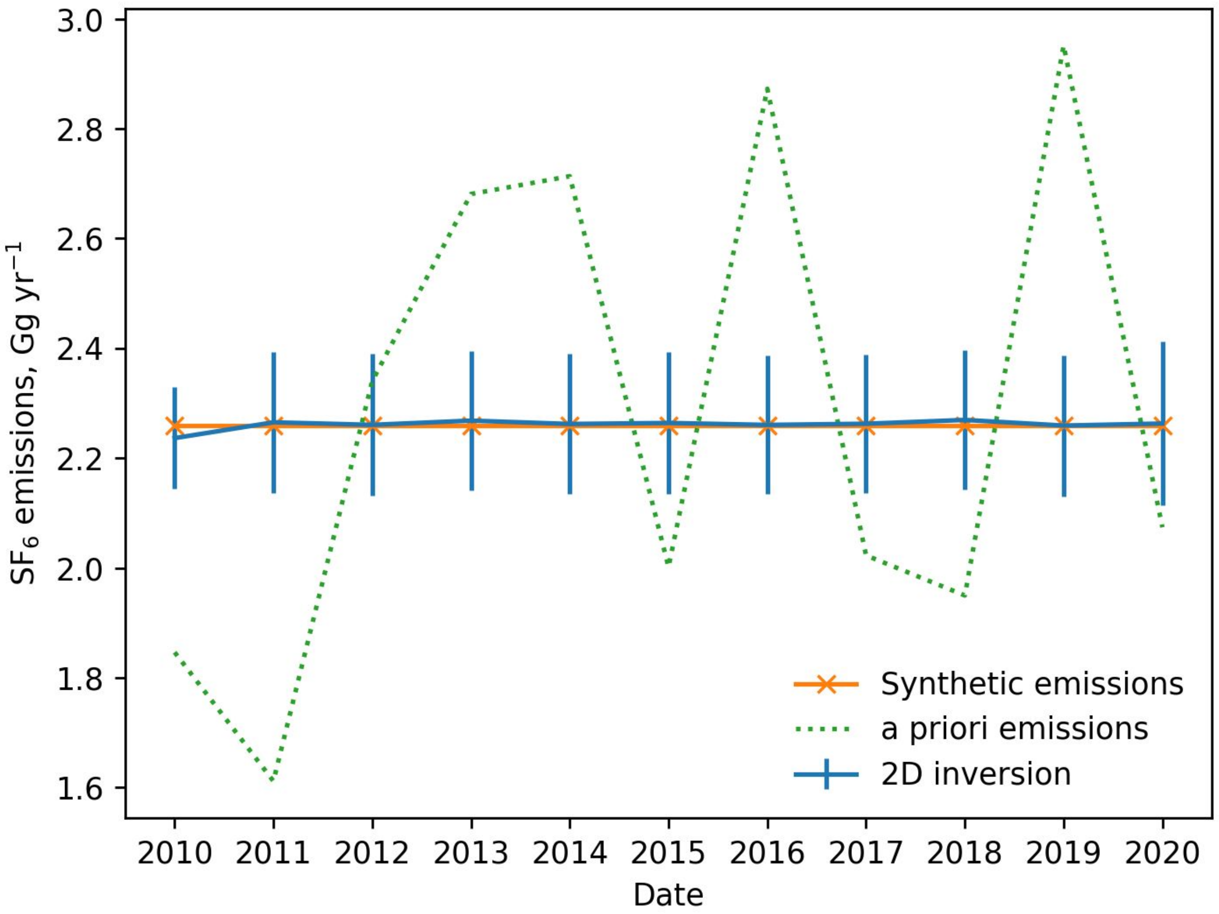
- Comparison of surface mole fractions at NOAA measurement site locations
- All models using same emissions
- 12-box model generally performs well when measurement site at sea level
- Large improvement with 2D model when modelling measurements made at elevation
- Down to poor vertical resolution in box models which may bias emissions estimates





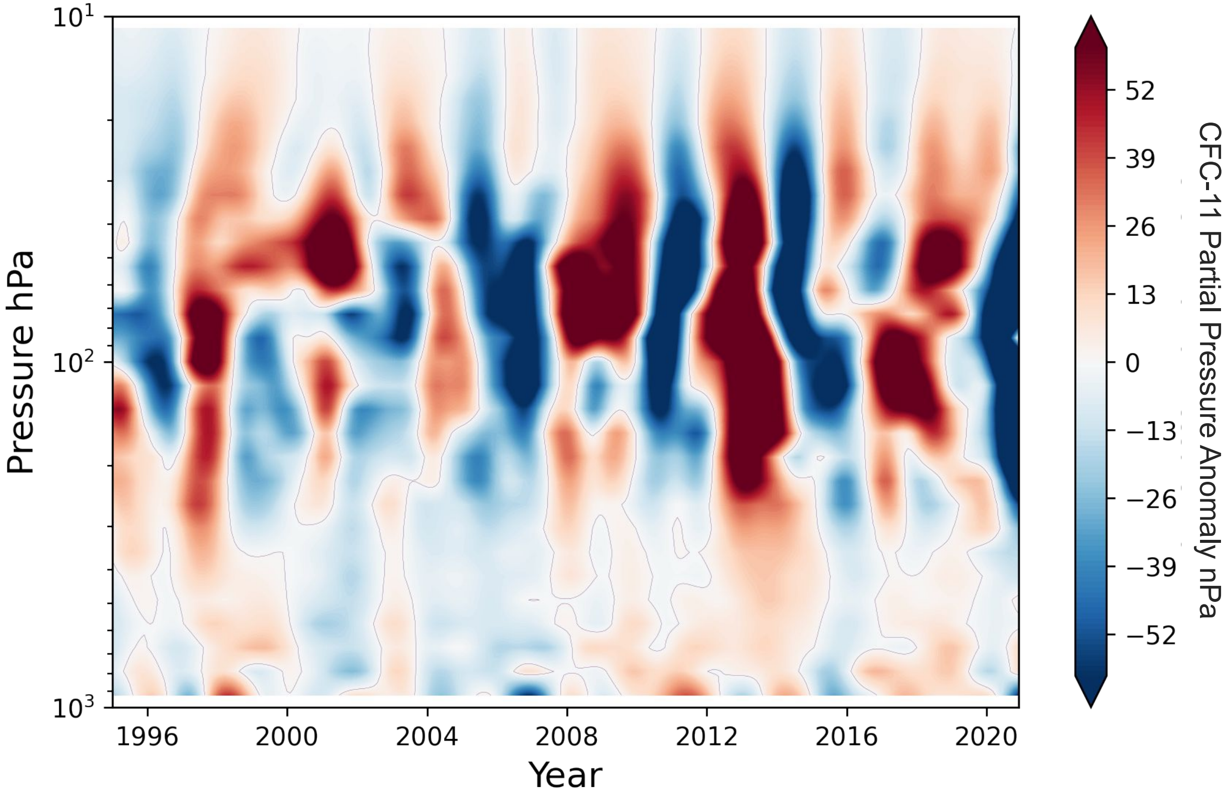
Validation: SF_6 emissions estimates

- Use "surface concentrations" from 3D model to estimate emissions
- Emissions kept constant in time to produce synthetic dataset
- Uses simple analytical Gaussian inference
- A priori emissions are a random perturbation of true emissions (large uncertainty)
- One model year on laptop takes ~2s



Representation of the Quasi Biennial Oscillation

- Question whether representation of large-scale dynamics (e.g., zonally varying winds of QBO) would be captured in a zonally averaged model
- Whole atmosphere partial pressure anomalies of CFC-11 show some signal propagating to the surface, impacting surface concentrations of trace gases
- It’s messy! But driving reanalysis meteorology will itself have its limitations when representing QBO



Future direction

- Implement sinks for more trace gases
- Derive emissions using real measurements
- Compare upper atmosphere concentrations to, e.g., ACE
- Publish openly and accessibly
- Explore more applications