Stratospheric chemistry and aerosols

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Stratospheric chemistry

Gas phase chemistry

1. Chapman Cycle
2. Catalytic cycles
    1. Hydrogen radicals (HO_x)

$$OH + O \rightarrow H + O_2$$
$$H + O_3 \rightarrow OH + O_2$$
$$\text{Net: } O + O_3 \rightarrow 2\,O_2$$
$$OH + O \rightarrow H + O_2$$
$$H + O_2 + M \rightarrow HO_2 + M$$
$$HO_2 + O \rightarrow OH + O_2$$
$$\text{Net: } O + O_3 \rightarrow O_2$$
$$HO_2 + O \rightarrow OH + O_2$$
$$OH + O_3 \rightarrow HO_2 + O_2$$
$$\text{Net: } O + O_3 \rightarrow 2\,O_2$$

Hydrogen Source Gases: H_2O, CH_4

- Long term trends
- HO_x chemistry in the upper stratosphere and mesosphere

Stratospheric chemistry

2. Nitrogen radicals (NO_y)

$$NO_2 + O \rightarrow NO + O_2$$
$$NO + O_3 \rightarrow NO_2 + O_2$$
$$\text{Net:} \quad O + O_3 \rightarrow 2\,O_2$$

Nitrogen Source Gas: N_2O (and …)

- Long term trends
- NO_y partitioning (in the lower stratosphere: aerosols)

3. Chlorine radicals (Cl_y)

$$ClO + O \rightarrow Cl + O_2$$
$$Cl + O_3 \rightarrow ClO + O_2$$
$$\text{Net:} \quad O + O_3 \rightarrow 2\,O_2$$

Chlorine Source Gases: Organic Chlorine

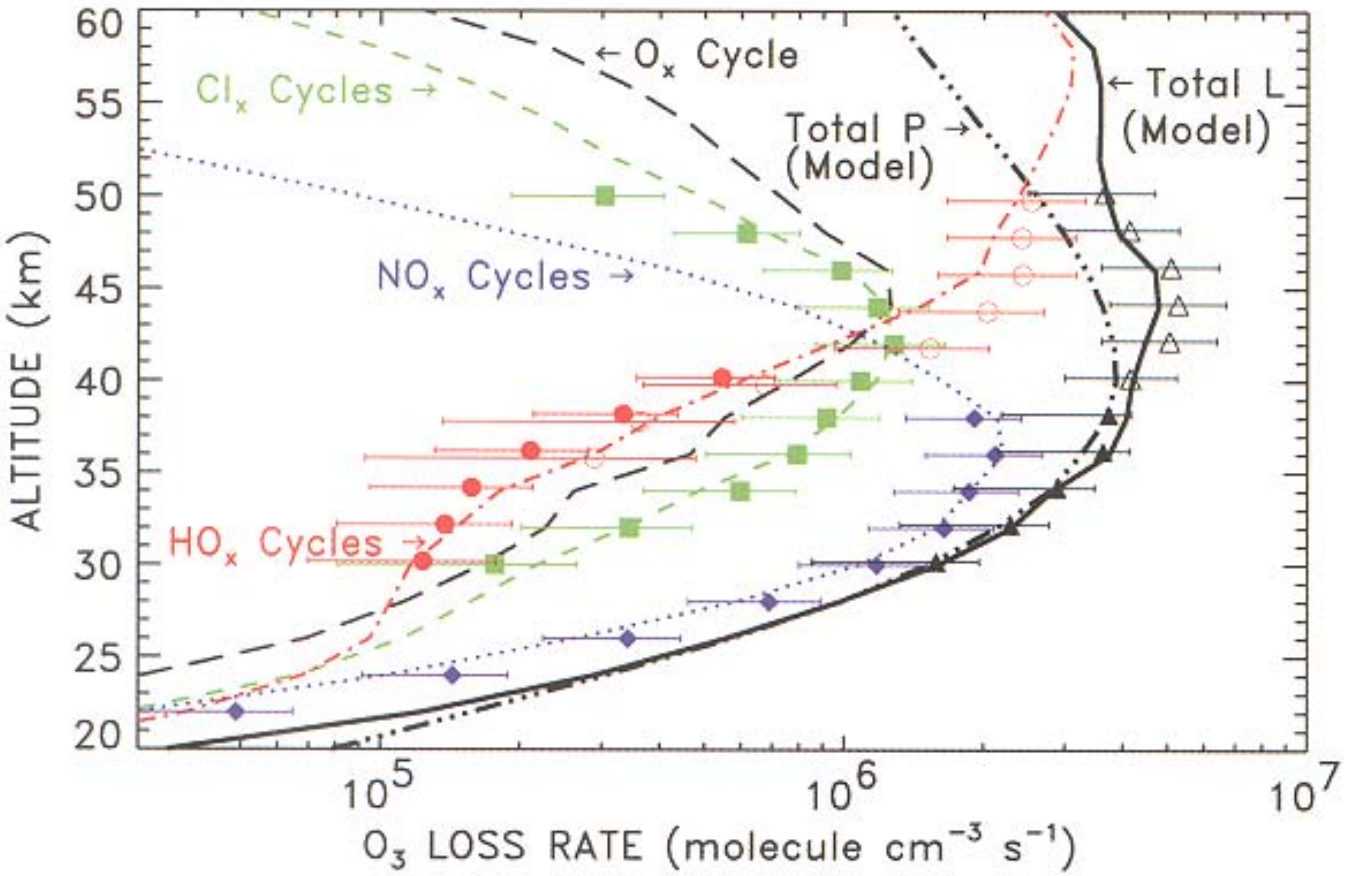
- Long term trends
- Cl_y partitioning (in the lower stratosphere: aerosols)

Ozone loss

35°N,September 1993

24 hour average

WMO, 1998



Lower Stratosphere

Gas phase chemistry triggered by heterogeneous processing

$ClO + ClO + M \rightarrow ClOOCl + M$

$ClOOCl + h\nu \rightarrow 2\,Cl + O_2$

$2\,(\,Cl + O_3 \rightarrow ClO + O_2\,)$

Net: $2\,O_3 \rightarrow 3\,O_2$

$BrO + ClO \rightarrow Br + Cl + O_2$

$\rightarrow BrCl + O_2$

$BrCl + h\nu \rightarrow Br + Cl$

$Br + O_3 \rightarrow BrO + O_2$

$Cl + O_3 \rightarrow ClO + O_2$

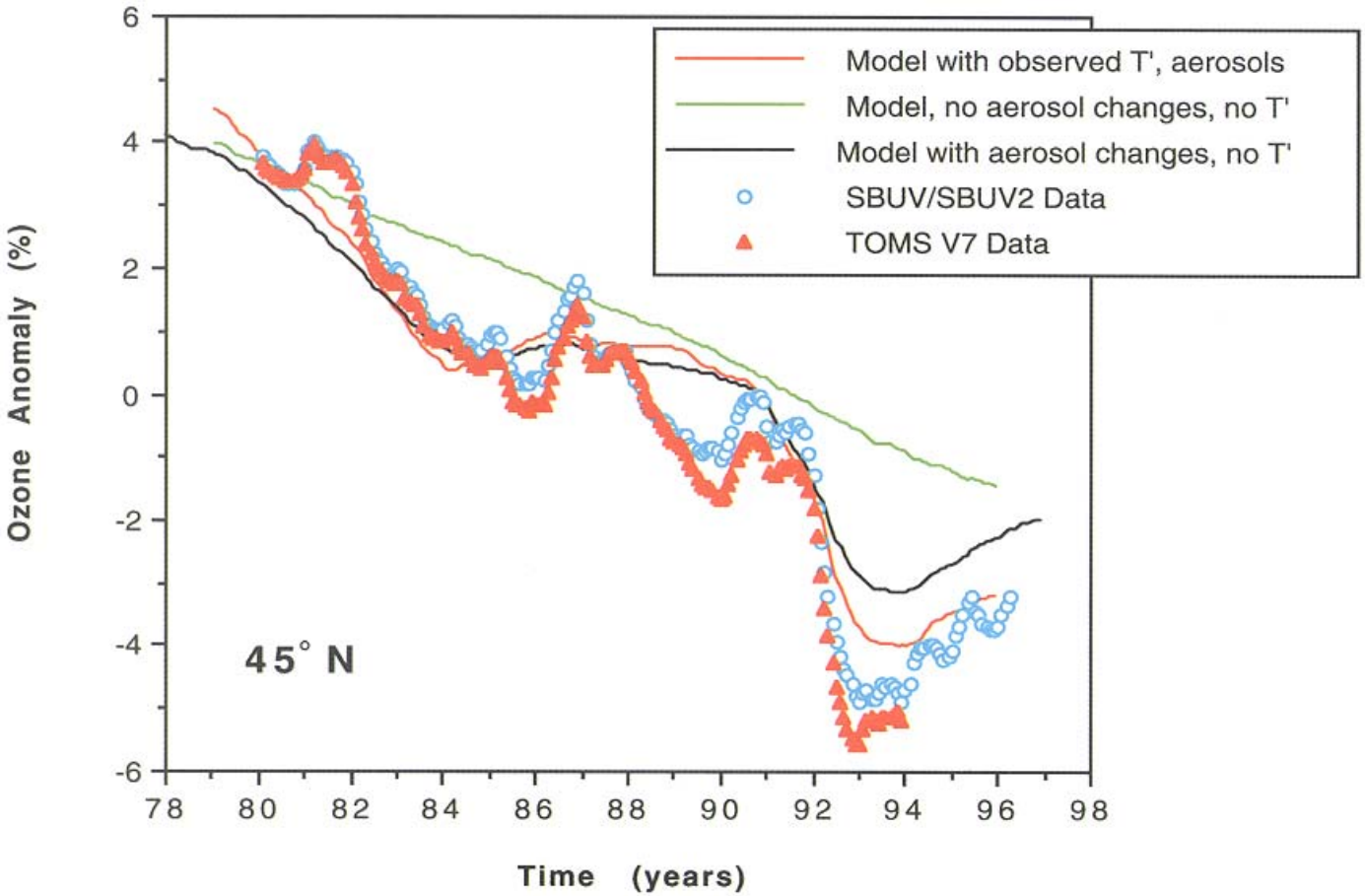
Net: $2\,O_3 \rightarrow 3\,O_2$

Polar Stratospheric Clouds

Aerosols (Pinatubo)

ClO_x/Cl_y & NO_x/NO_y

Lower Stratosphere Ozone Loss



What determines Ozone

- Source budgets: determines radicals budget
  (emissions, transport, decay)
- Family budgets: determines radicals budget
  (source decay, transport)
- Repartitioning: determines radicals budget
  (chemistry, particle budgets)
- Particle budgets: determines radicals budget
  (emission, microphysics, volcanic eruptions, …)

(temperature, illumination, …)

Nitrogen Source, N_2O

Data Assimilation: MIPAS constrained N_2O budget

↓

Chemical forecast (same dynamical fields)

↓

Assessment of the evolution of N_2O

Case study: Sept., 2002

Forecast initiated @ Sept. 2, 2002

Reminder non-operational MIPAS

Assimilation system

4D-var: (Talagrand & Courtier, 1987)

Model grid: horizontal: 3°.75 *x* 3°.75 (96 *x* 49 pts)
vertical: 37 pressure levels, surface → 0.1 hPa
(subset of ECMWF hybrid levels, keeping stratospheric levs)

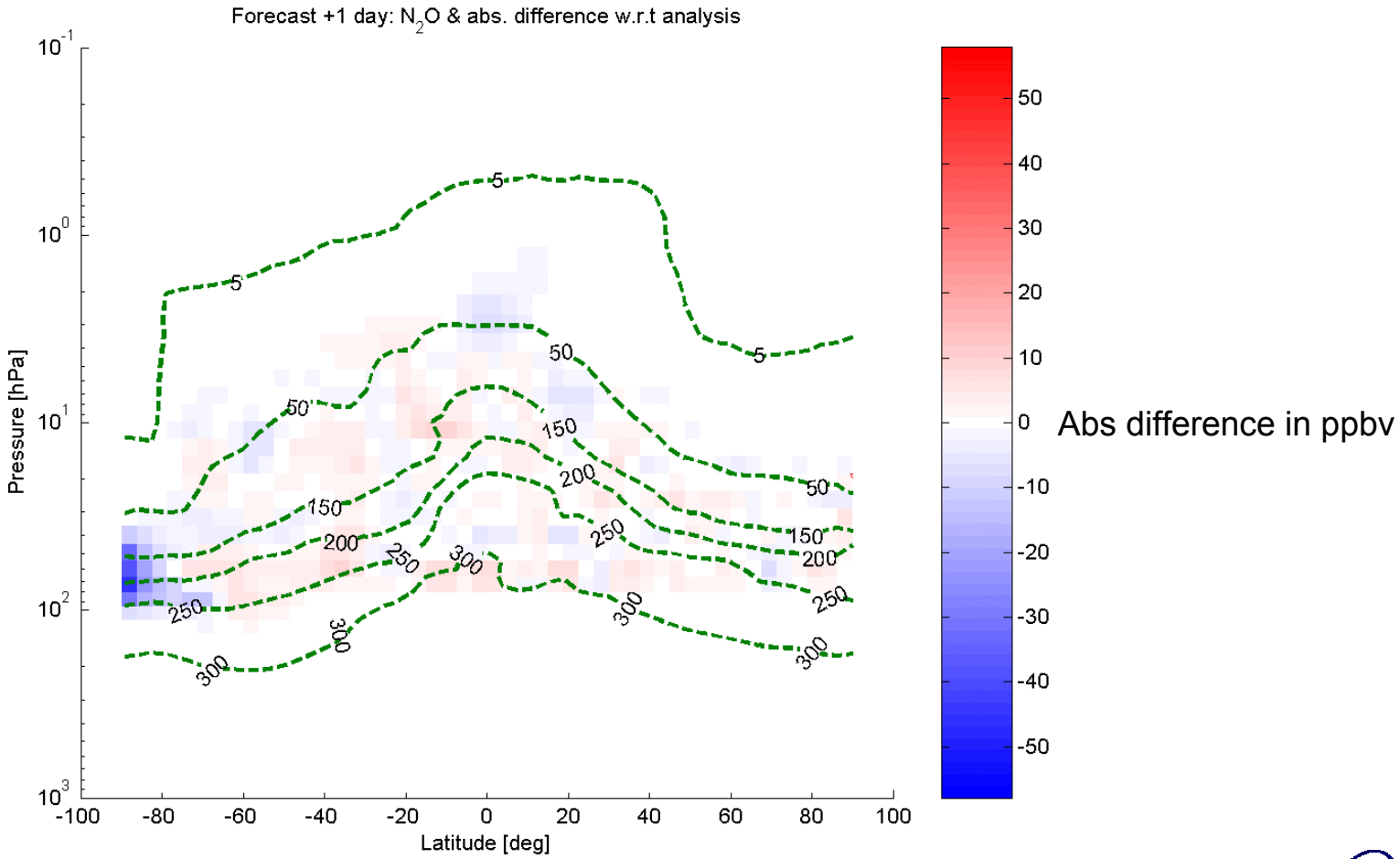
Chemistry: 57 chemical species

PSC: 4 types of PSC particles (36 size bins)

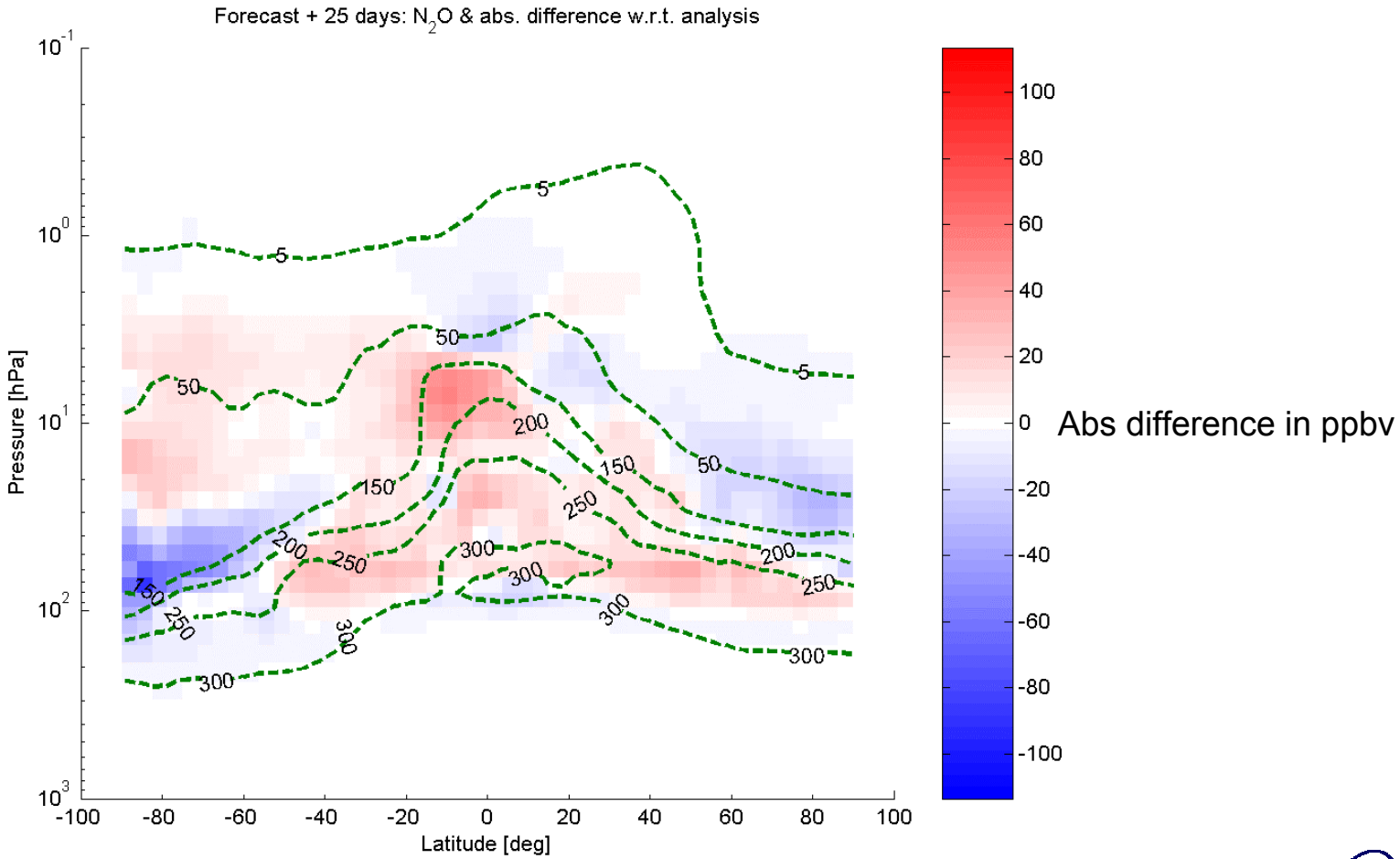
Dynamics: Eulerian

driven by ECMWF 6h analyses
advection by Lin & Rood (1996) with 30' time step

Forecast vs analysis



Forecast vs analysis



Forecast vs analysis ?

Cause of difference

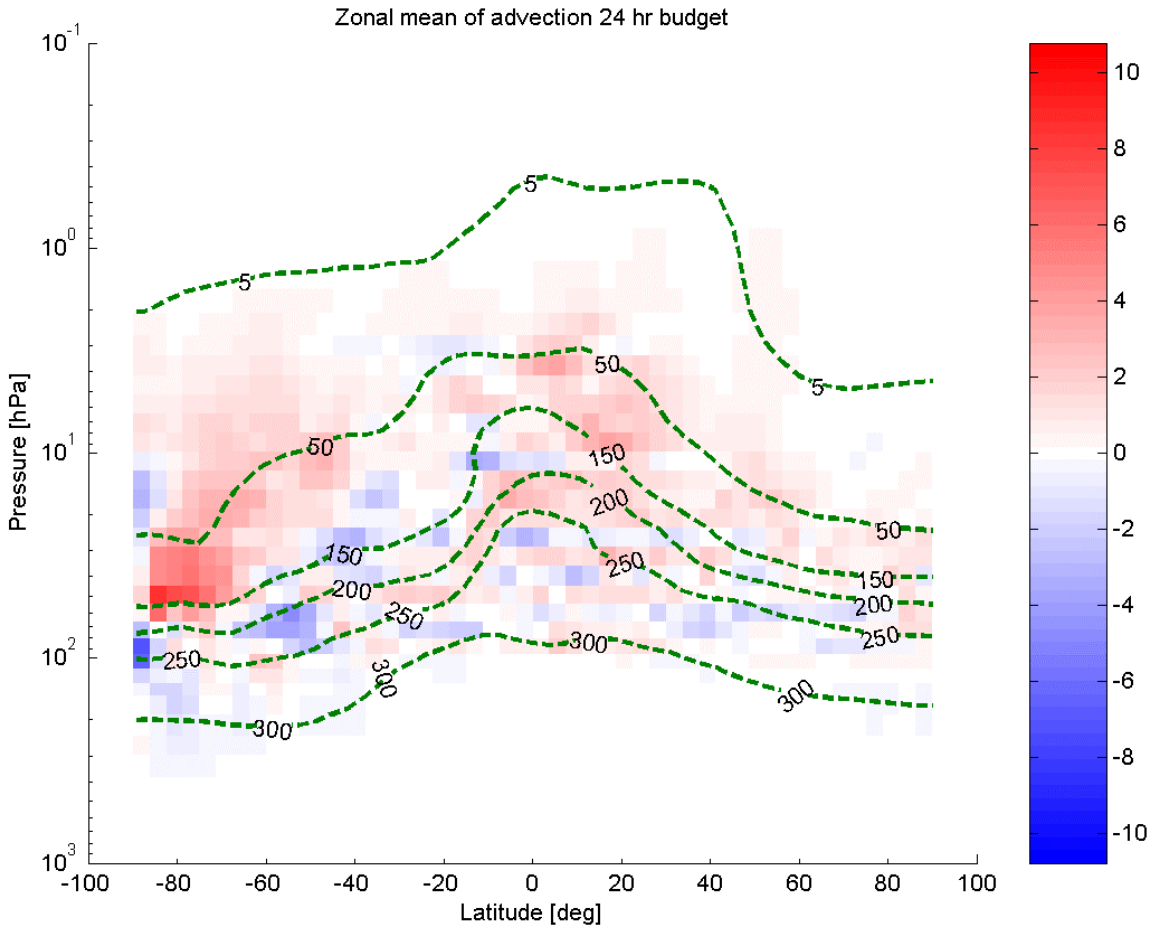
↓ (4D – VAR, history)

Local advection / chemistry tendency
(24 hr integrated tendency)

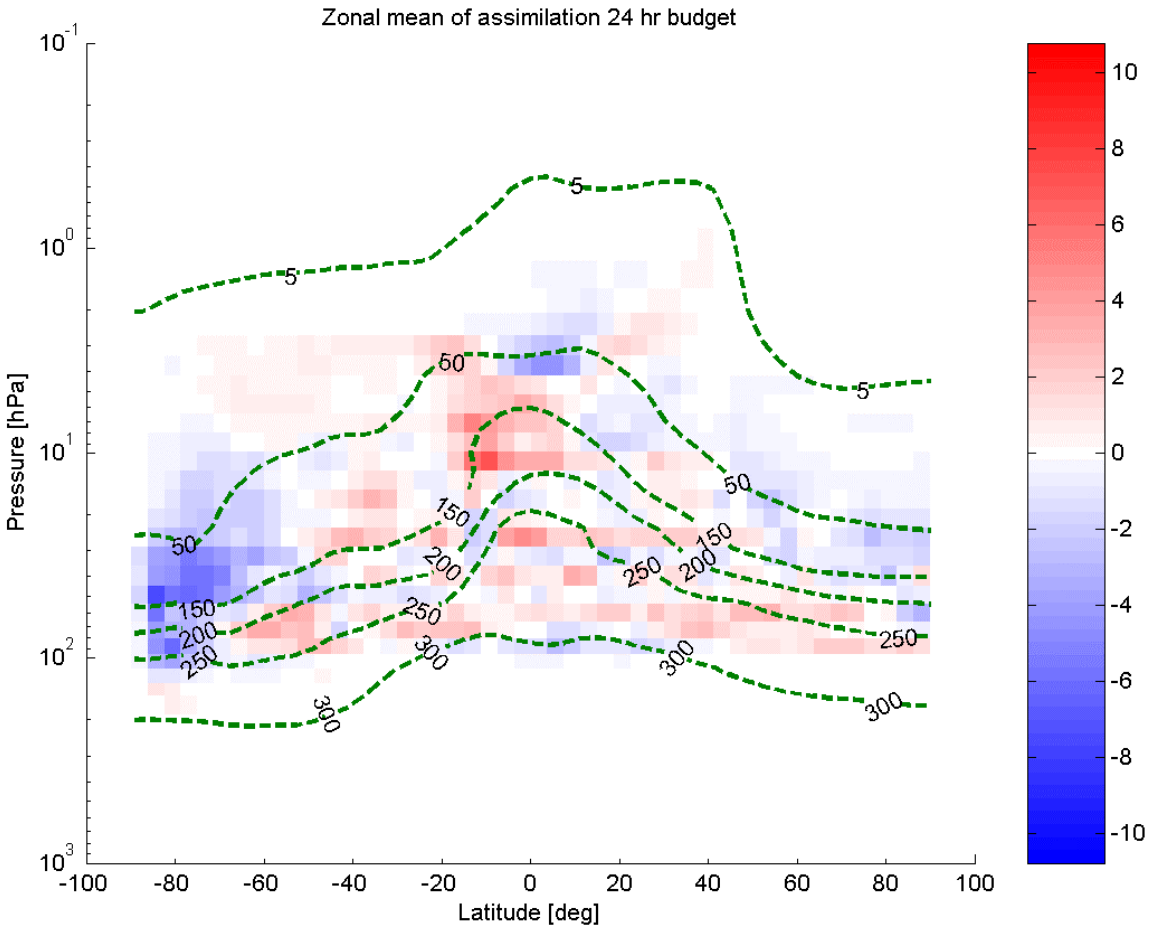
↓

Comparison with local assimilation "tendency": $(\text{analysis} - \text{first guess})_{t0}$
September mean

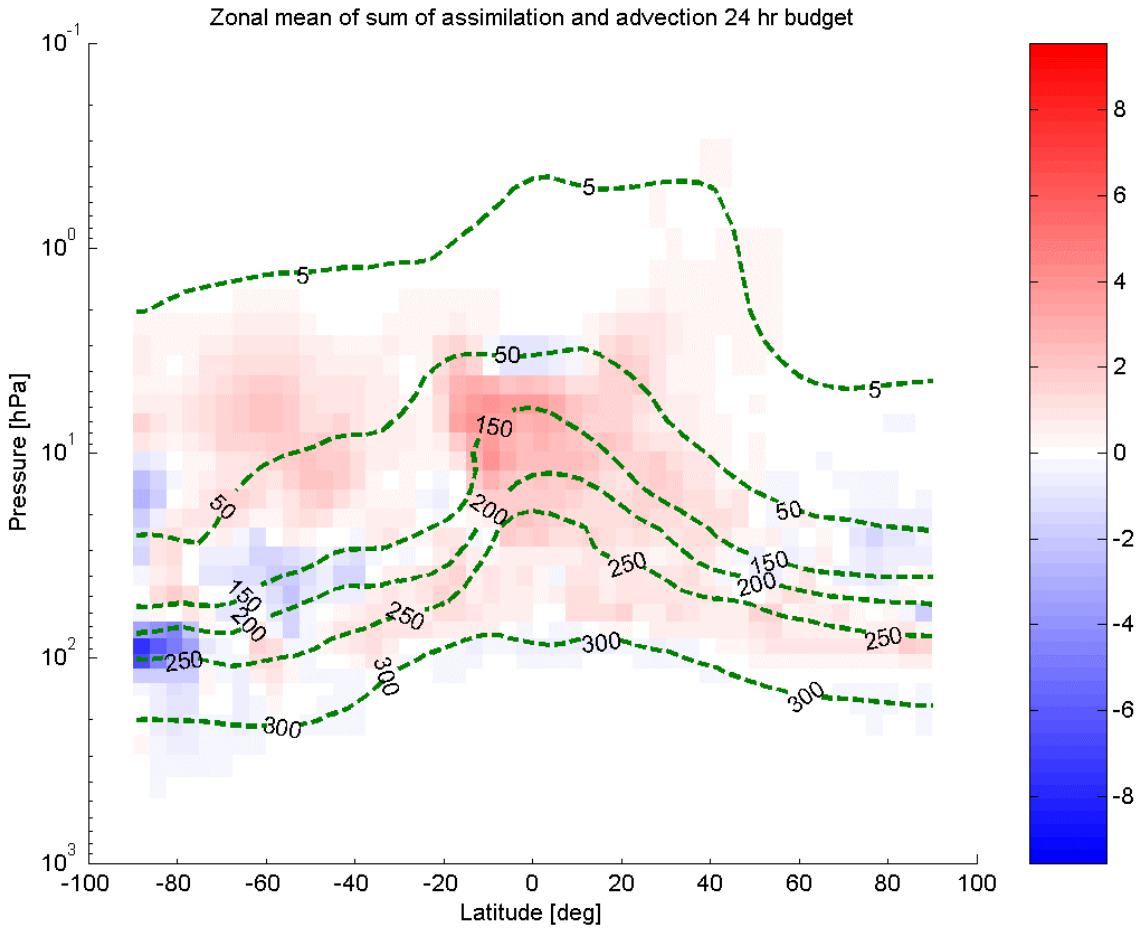
24 hr tendencies



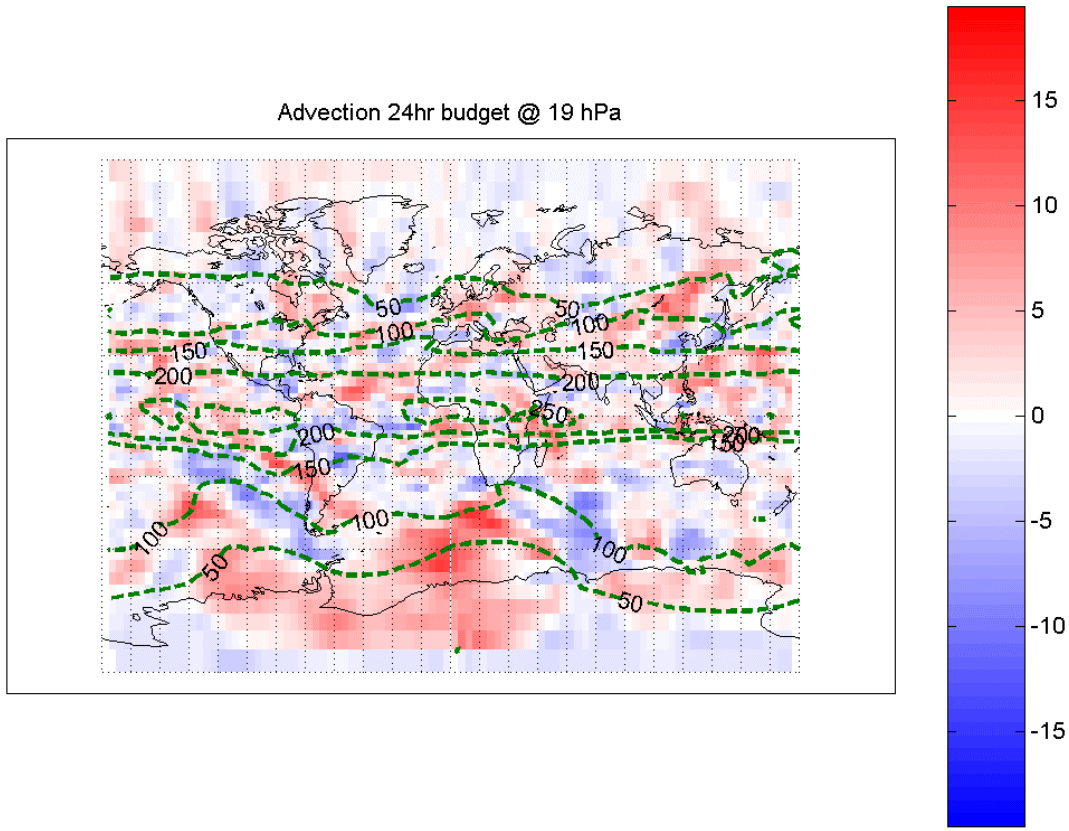
24 hr tendencies



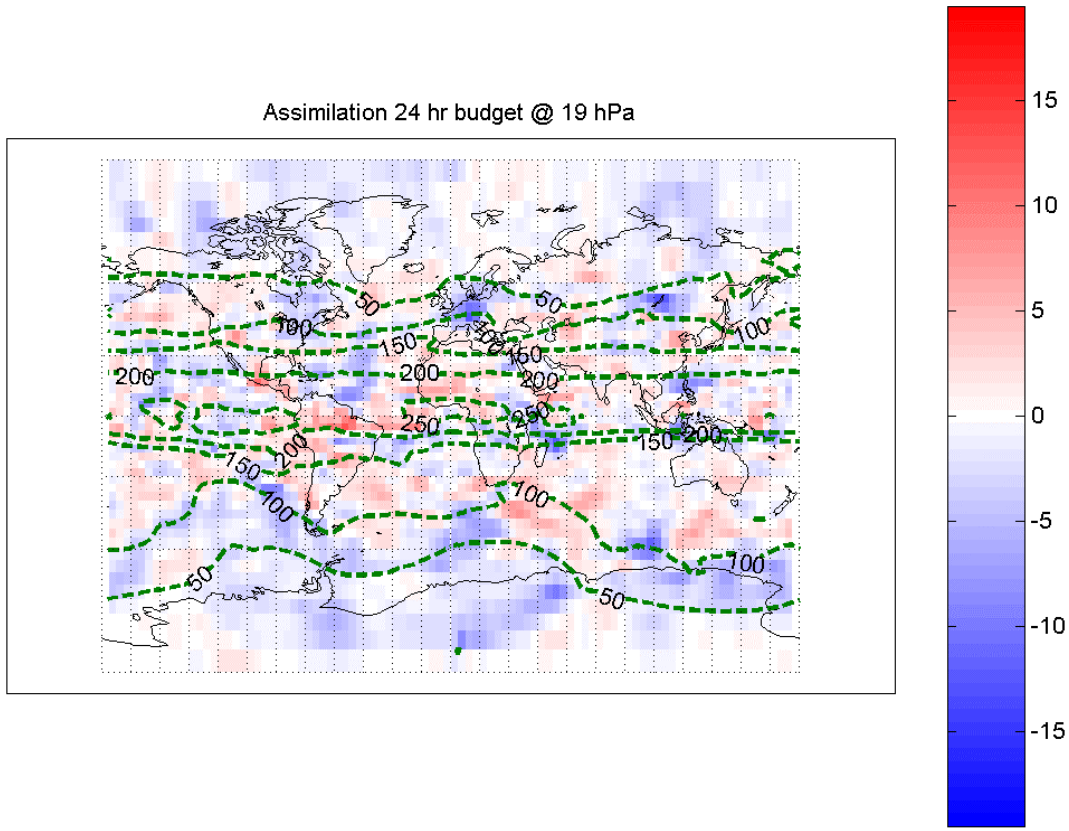
24 hr tendencies



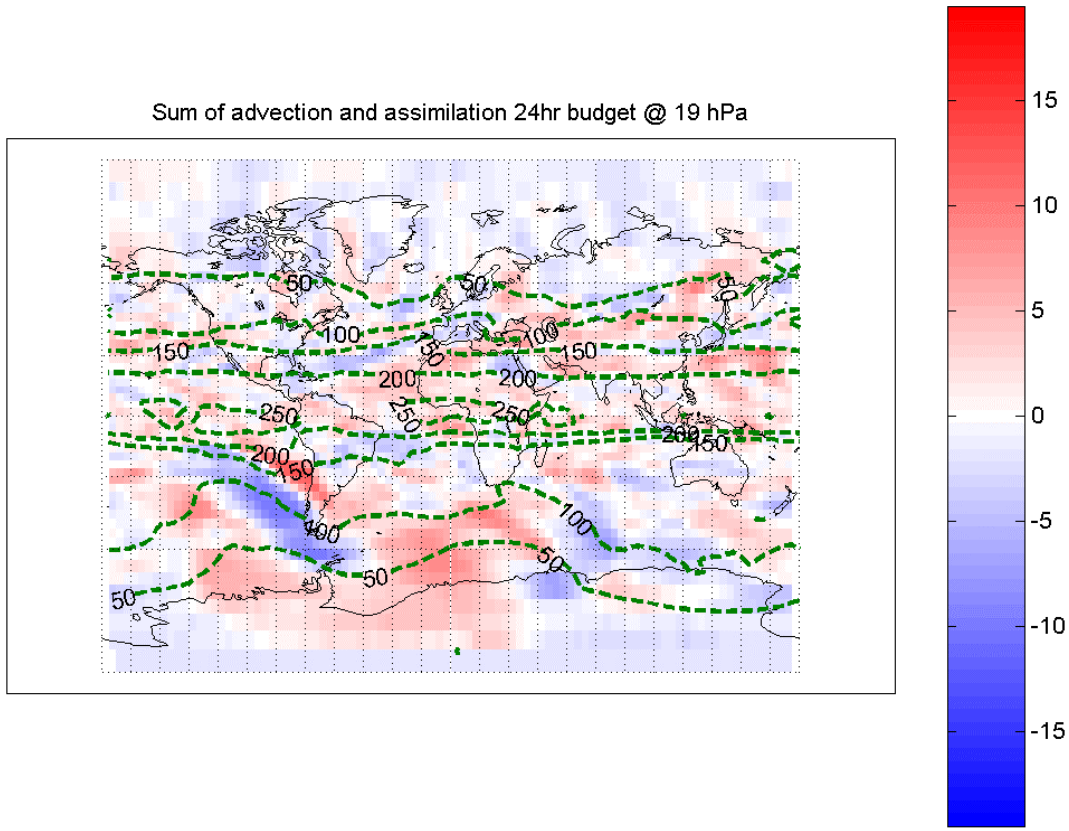
24 hr tendencies



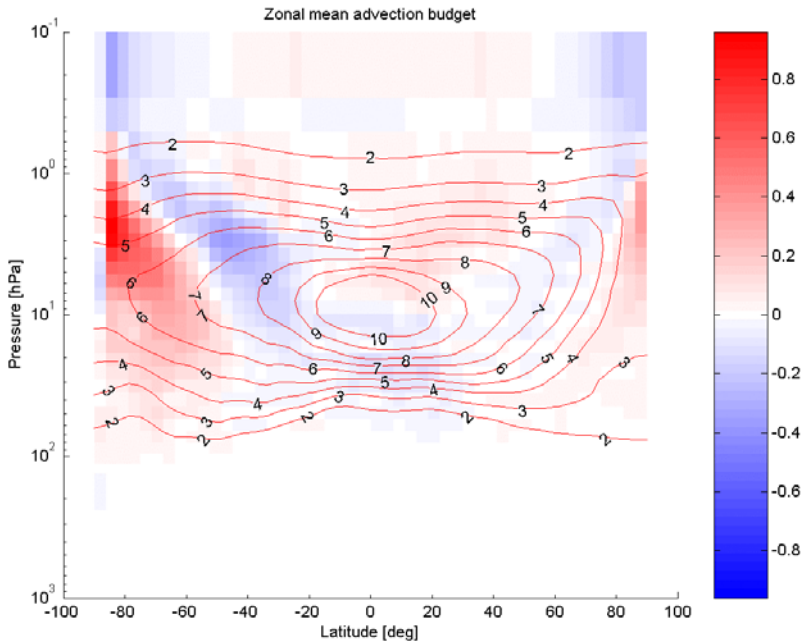
24 hr tendencies

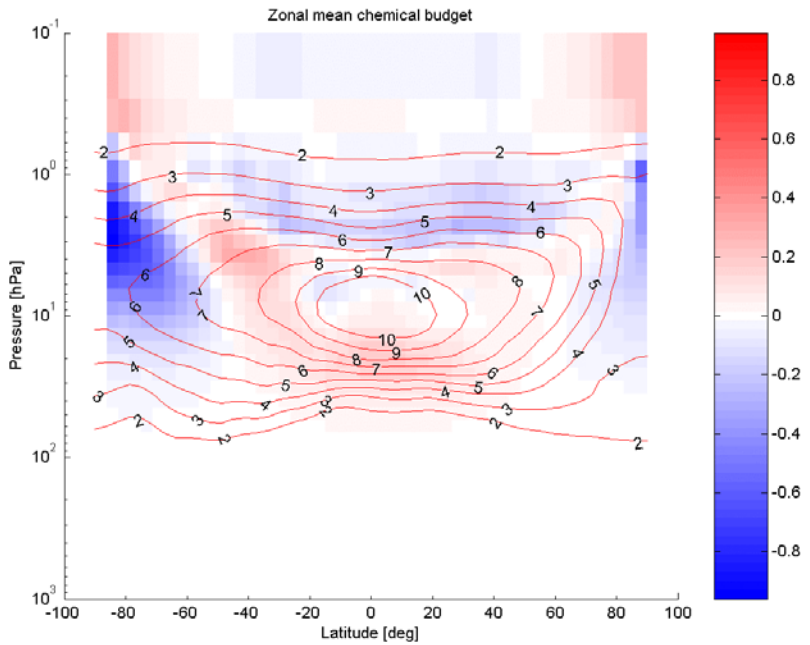


24 hr tendencies

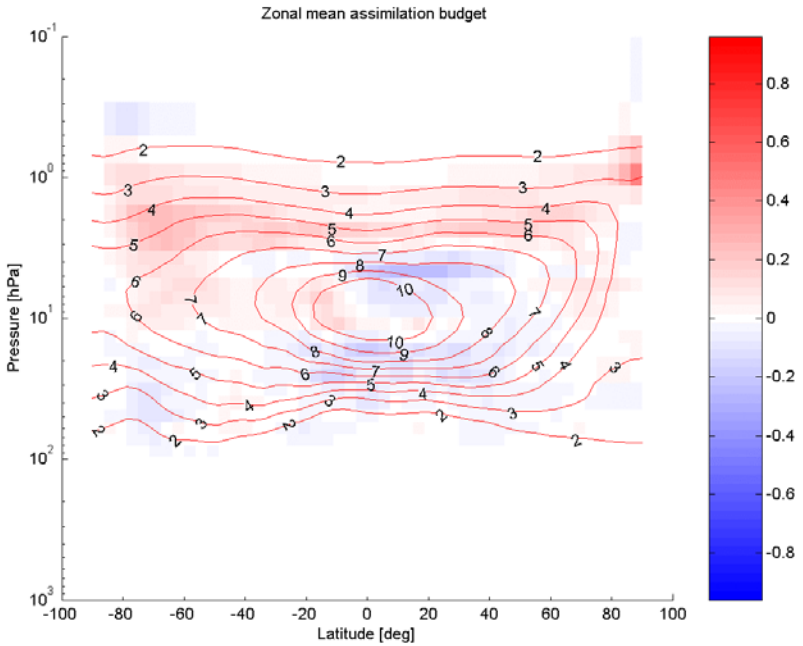


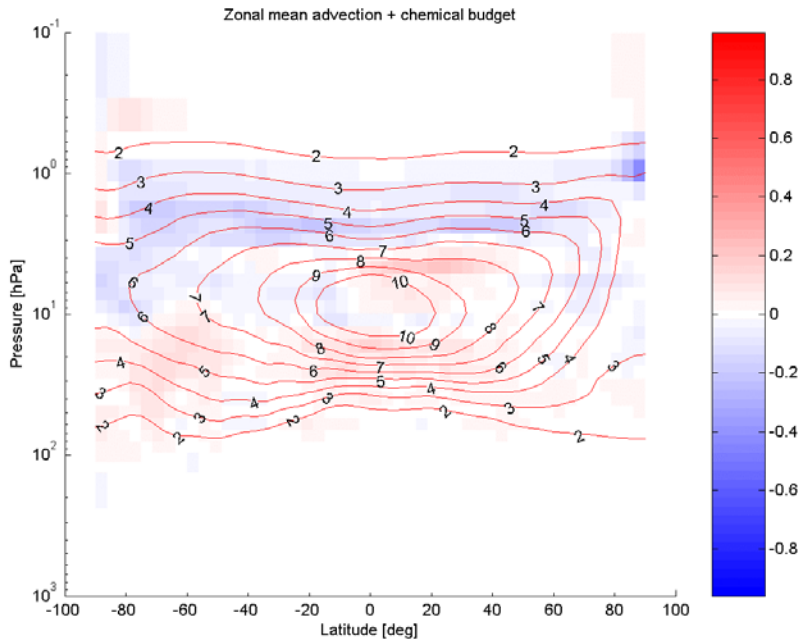
24 hr ozone tendencies



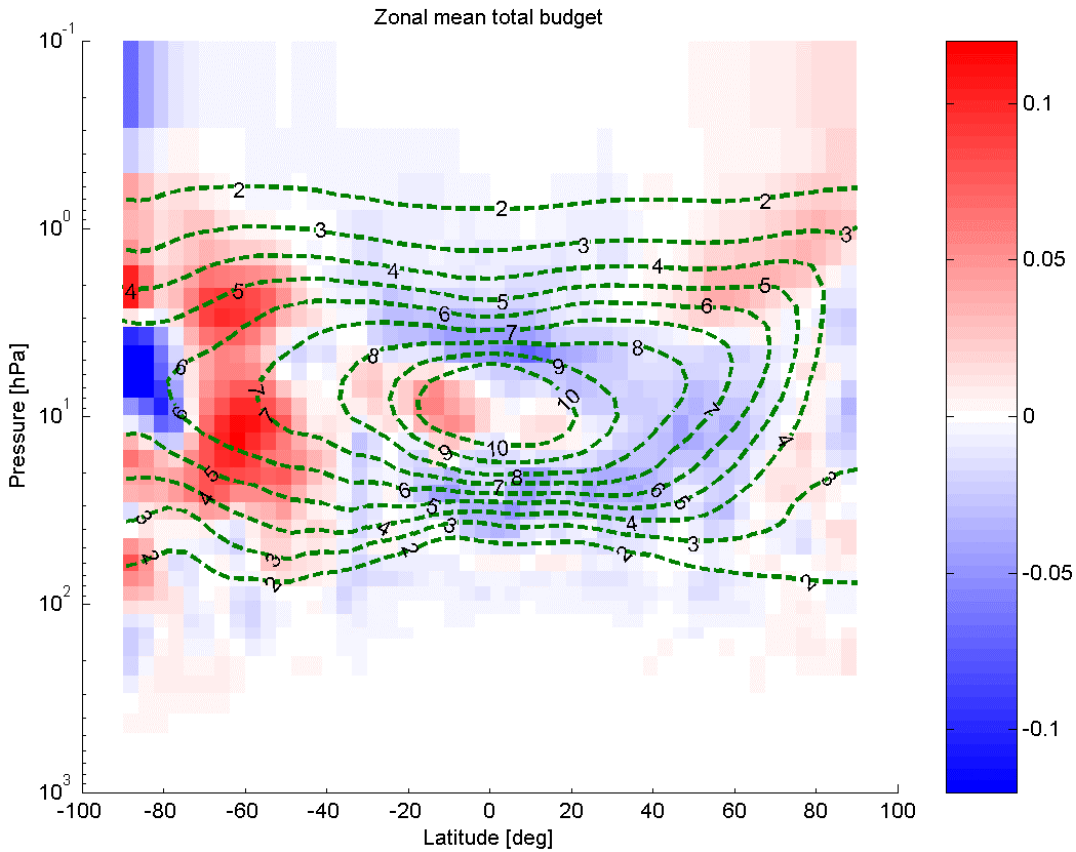


24 hr ozone tendencies





24 hr ozone tendencies



Anymore uncertainties