N_2O behavior between the lower stratosphere and the surface suggested by aircraft observation and model

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Introduction

- ➢ Estimation of temporal-spatial changes of nitrous oxide (N2O) fluxes are still very uncertain on the globe (IPCC, 2008)
- ➢ A few N2O inverse modeling researches have recently estimated global N2O fluxes (Hirsch et al [2006], Huang et al [2008])

Low-resolution flux estimation:
12 regions on the globe, 4-yrs average,
no seasonal/interannual changes

reason

- **Scale of temporal-spatial variability of N2O concentration are small compared to the measurement precision (observation)**
- **Number of observation sites is insufficient (observation)**
- **There is a large uncertainty in estimation of influence of the stratosphere-troposphere exchange (STE) on tropospheric N2O concentration (model)**

Purpose

Hirsch et al. [2008] concludes

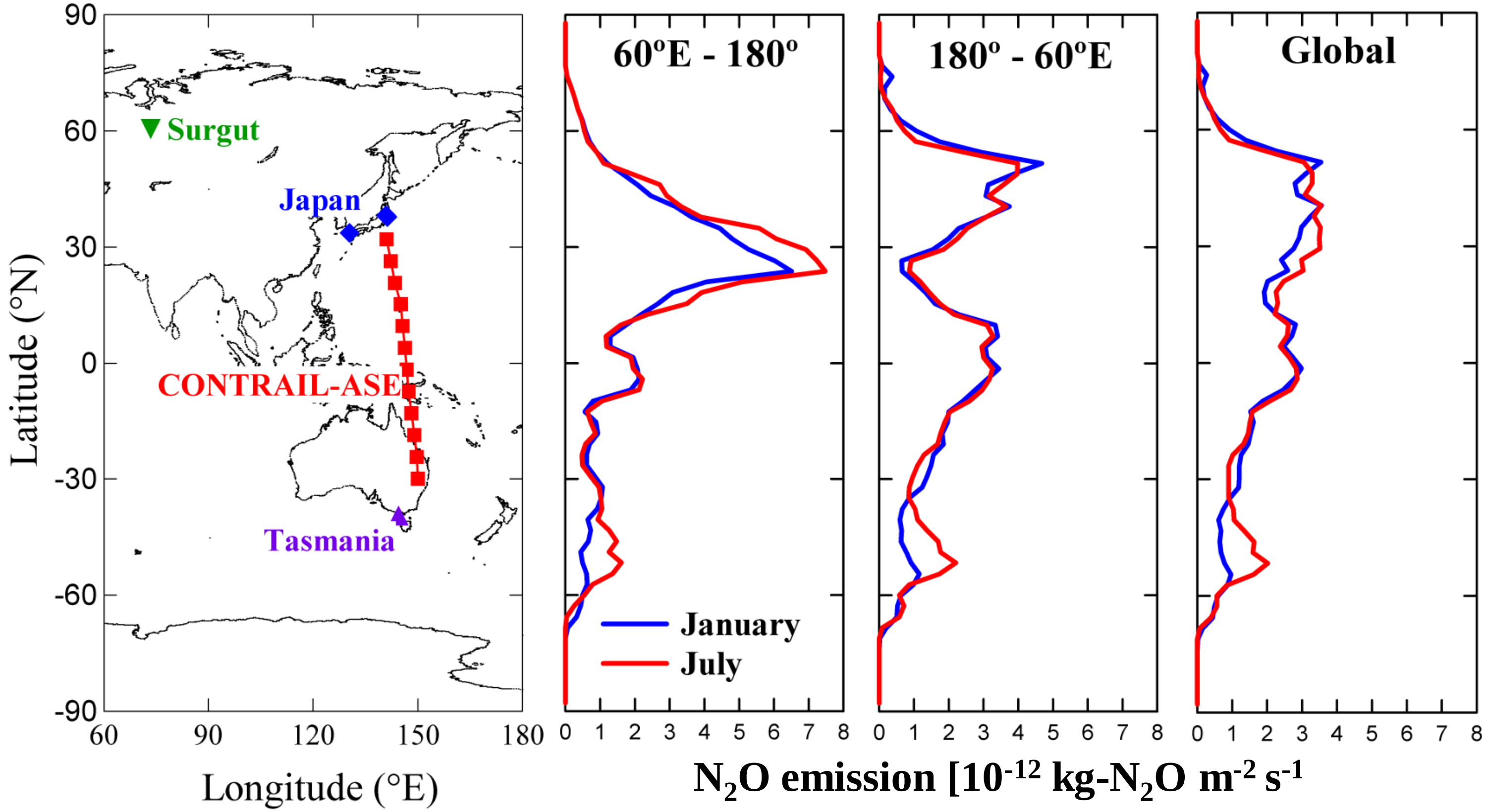
More measurements of N_2O and that kind of tracers in the stratosphere and upper troposphere could be useful to improve the model simulations of the seasonal and interannual changes of STE, and enable more accurate estimation of surface N_2O fluxes, including the seasonal and interannual variations.

We validate our model for N_2O concentration in the upper troposphere and lower stratosphere (UT/LS), and estimate the stratospheric influence on the seasonal cycle of the tropospheric N_2O concentration, using aircraft observation data

Observation

Observation	Category	Agency	Location	Region	Period
Aura-MLS (microwave limb sounder)	Satellite	NASA *U.S.*	80S-80N 100-1 hPa	stratosphere	Dec 05–Dec 07
CONTRAIL-ASE	Aircraft	NIES/MRI *Japan*	32N,141E - 30S,151E 9 - 12 km	upper troposphere	Dec 05–Jan 08
over **Surgut**	Stationary Aircraft	NIES *Japan*	61N, 73E 0.5 - 7.0 km	troposphere	Apr 01–Feb 05
over **Japan**	Stationary Aircraft	Tohoku Univ. *Japan*	34-38N, 130-141E 0.2 - 11 km	lower to upper troposphere	Jun 01–Jan 08
over **Tasmania**	Stationary Aircraft	CSIRO *Australia*	40S, 144E 0.2 – 8.0 km	troposphere	Sep 92–Sep 00

Location of aircraft observation and latitudinal distribution of N2O emission in model

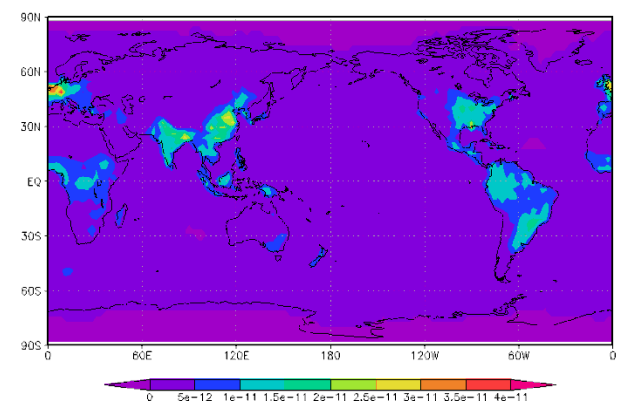


Three N2O emission categories combined in ACTM

Natural soil: constant natural soil flux by EDGAR2 1990 x 1.1 (Bouwman et al., 1993)

Ocean: monthly varying fluxes by Nevison et al. [1995]

Anthropogenic : annual fluxes from EDGAR 32FT2000 (Olivier et al., 2005)

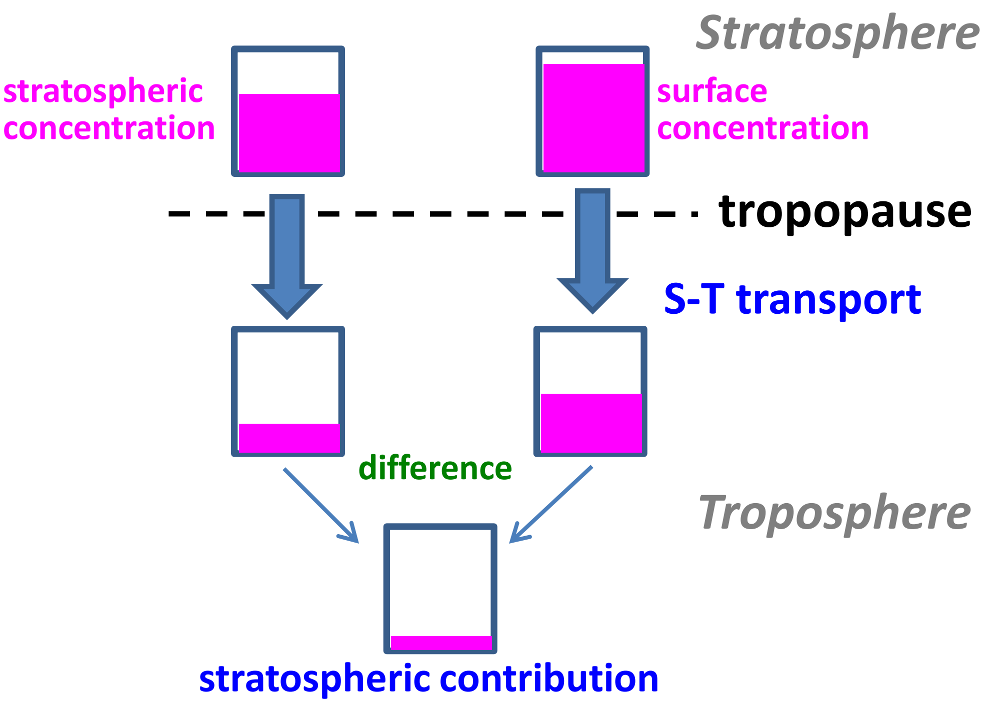


Model

Model	CCSR/NIES/FRCGC AGCM (5.7b) **(ACTM)**
Resolution	Horizontal: T42 Vertical : 67 layers (0~90 km)
Transport	Grid scale: flux-form semi-Lagrangian Sub-grid scale: convection, vertical diffusion
Nudging	NCEP/DOE AMIP-II Reanalysis (NCEP2) (1987~2008), JRA (2006-2007)
Chemistry	$O + O_2 + M \rightarrow O_3 + M$ $O(^1D) + O_2 \rightarrow O + O_2$ $O + O_3 \rightarrow O_2 + O_2$ $O(^1D) + N_2 \rightarrow O + N_2$ $O + O + M \rightarrow O_2 + M$ $N_2O + UV \rightarrow N_2 + O(^1D)$ $N_2O + O(^1D) \rightarrow 2NO$ $N_2O + O(^1D) \rightarrow N_2 + O_2$
Spin-up	Model-runs were started with a realistic concentration distribution from 1987 and spun-up for 5 years to stabilize chemical reactions and atmospheric transport

Tagged tracer

To distinguish the stratospheric contribution in the tropospheric N2O, the stratospheric tracers tagged above the tropopause were calculated in the model.



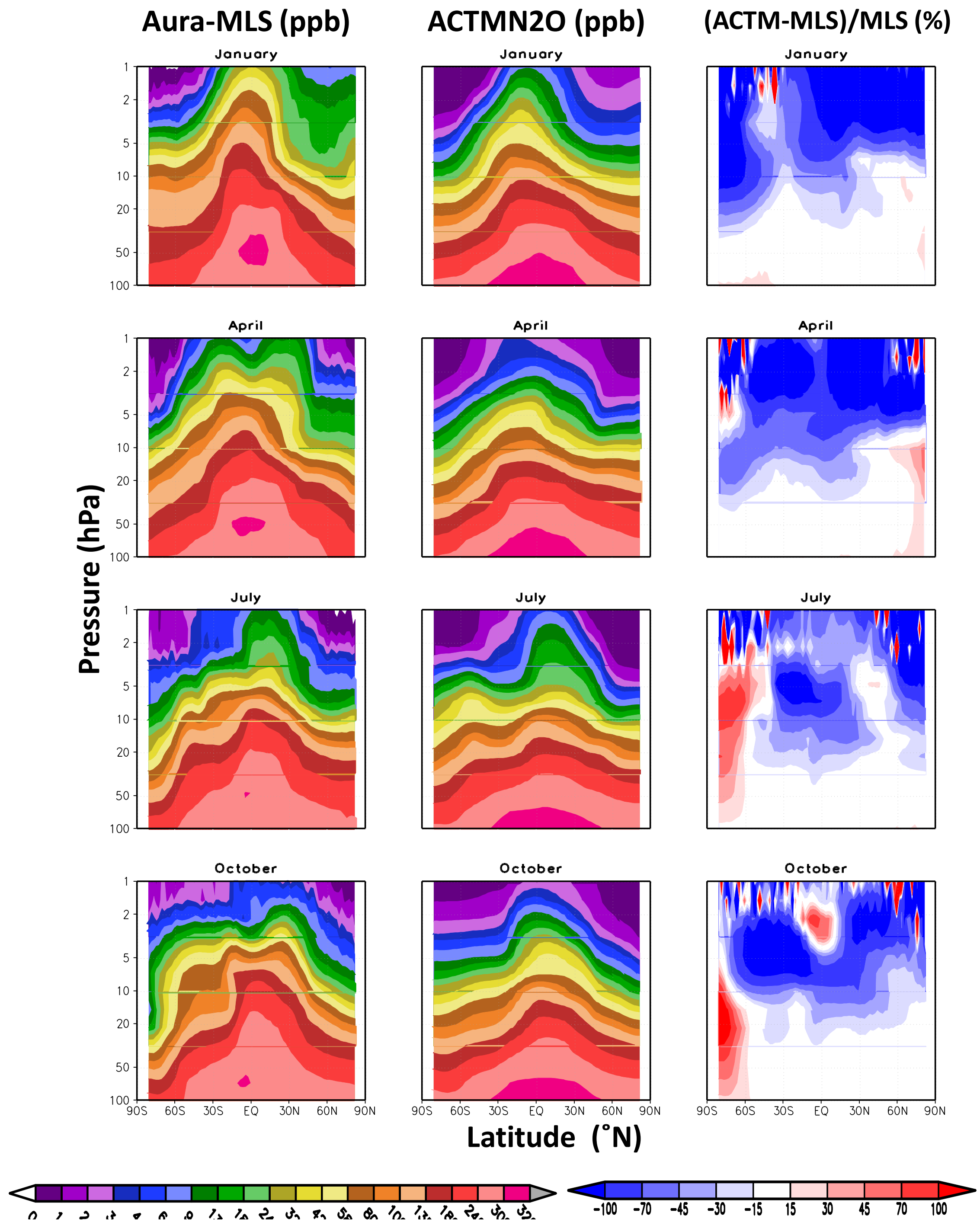
The stratospheric contribution is defined as difference between the two kinds of stratospheric tracers in the troposphere.

Results

Comparison with satellite observation in the stratosphere

- MLS and ACTM similarly show decreasing concentration gradient from low latitudes and pressure levels to high latitudes and pressure levels, as well as enhanced upwelling by convection at tropics.

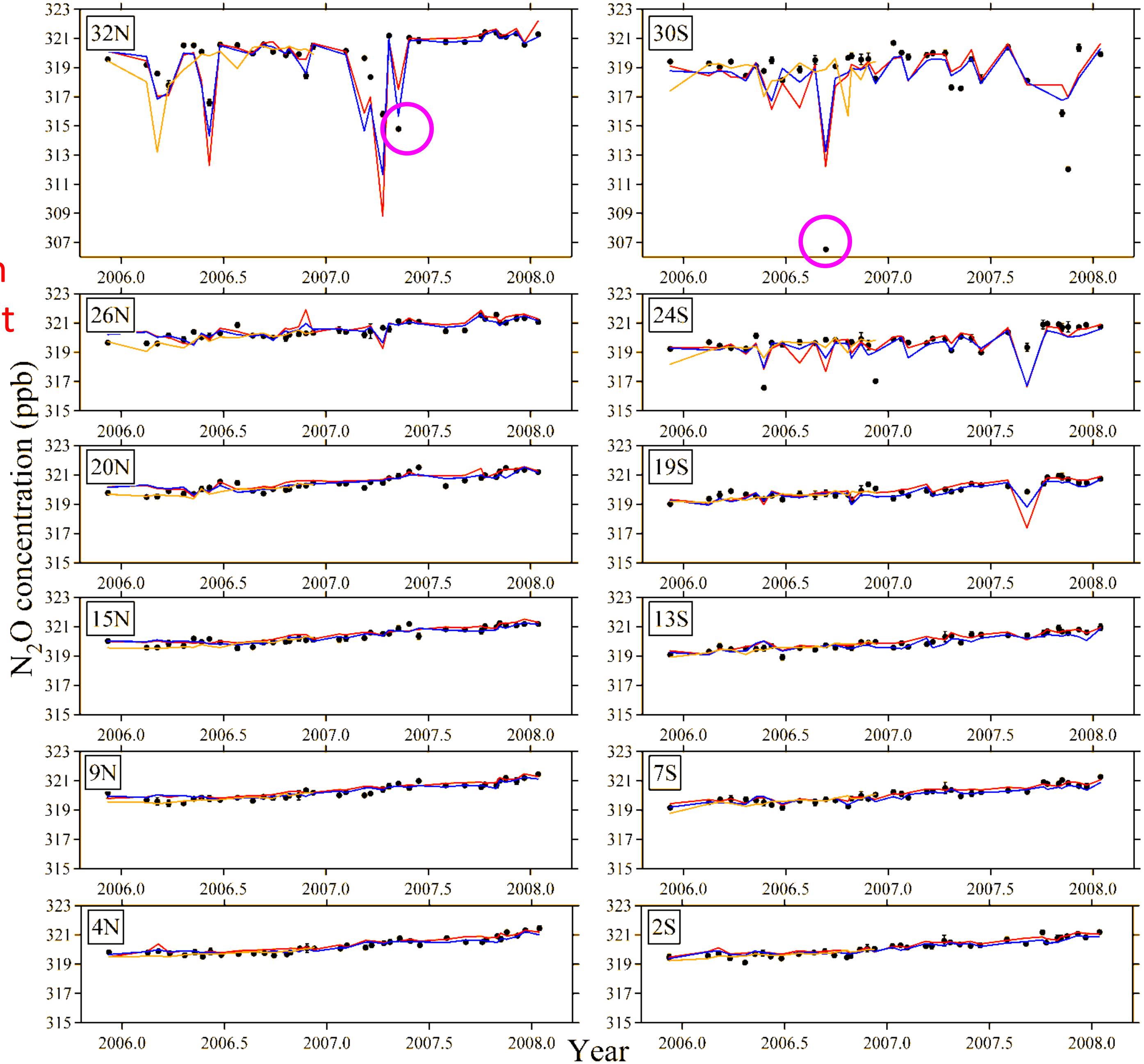
- ACTM tends to overestimate in polar regions, especially over Antarctica.



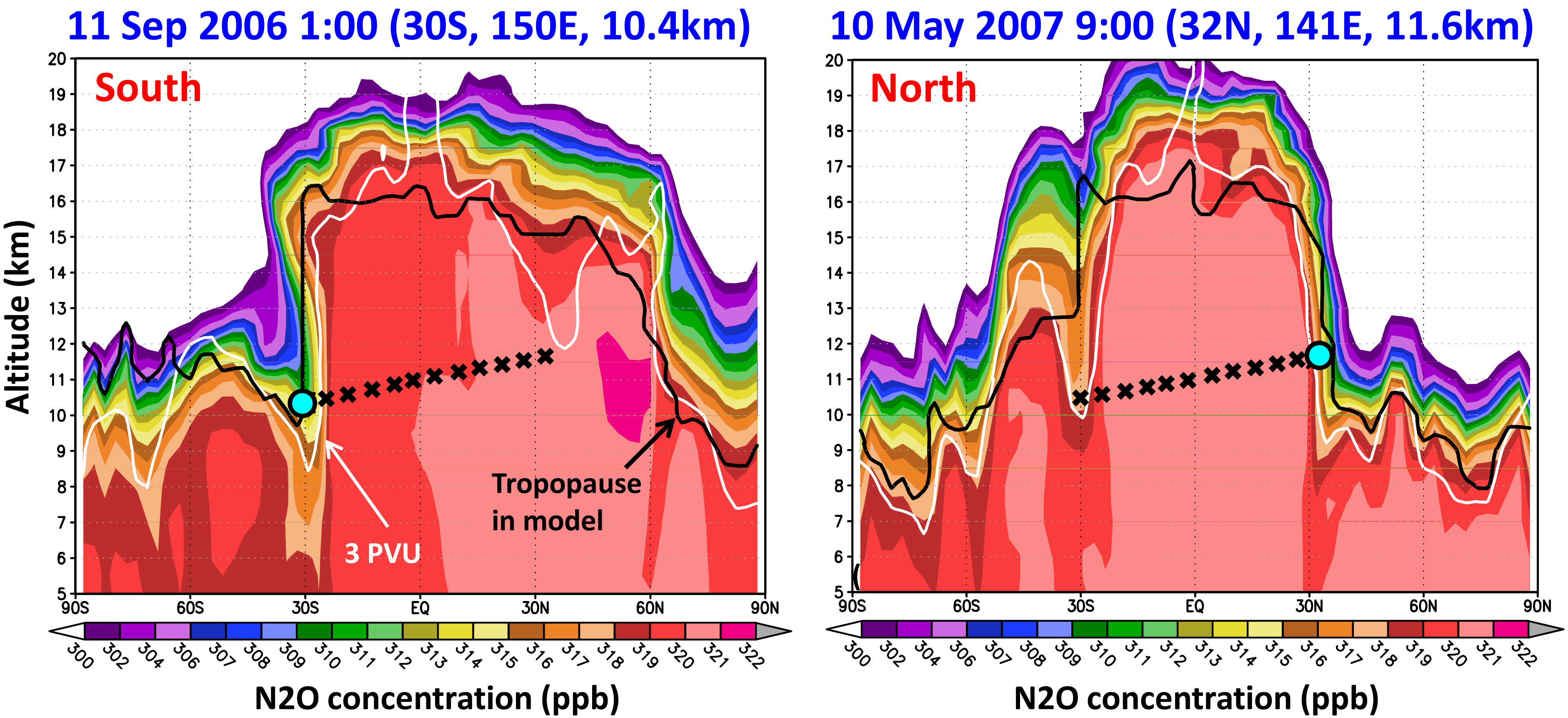
Comparison between CONTRAIL-ASE and ACTM

- Model well simulates N2O trend, and low concentrations at 32N and 30S in spring.
- The low concentration values are more frequent at 32˚N than at 30˚S.
- The causes are differences in dynamics and of air sampling positions:

Latitude	Altitude
32˚N	~ 11.5 km
30˚S	~ 10.5 km



N_2O cross section in the case of the lowest concentration observed in each hemisphere

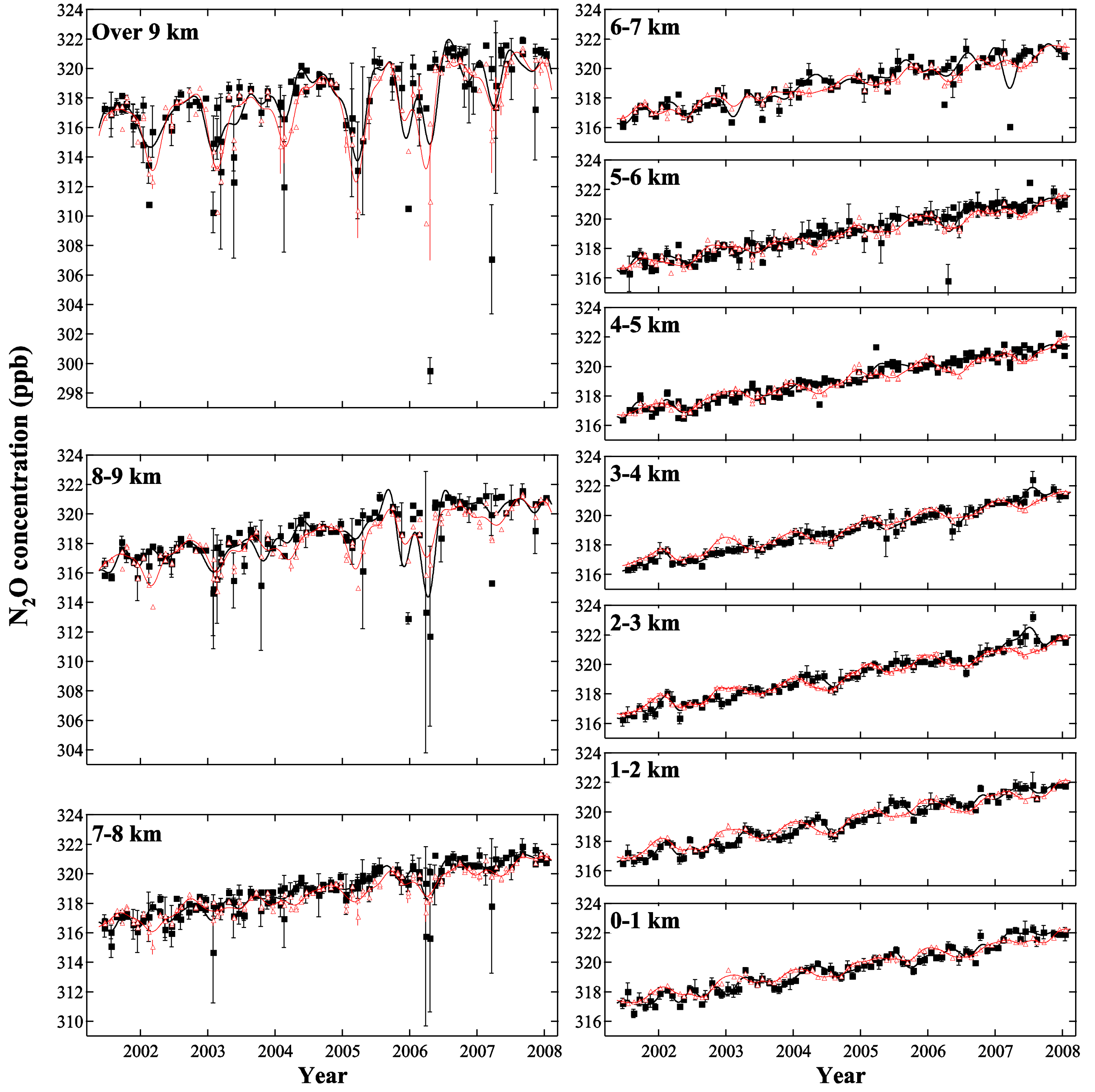


South : A deep tropopause folding occurred, so the sampling point at 30˚S was almost in the stratospheric air with low N2O concentration.

North : No folding occurred, but the aircraft was flying at the northern edge of the extratropical surf zone.

N2O concentration over Japan

- Trends at all heights, and very low concentrations over 7km are well simulated by ACTM.
- This region is highly affected by the stratosphere due to vicinity to the stratosphere, and tropopause folding frequently happening, exited by subtropical jet (and polar jet) in winter-spring.



Extraction of seasonal cycle

Component of seasonal variation are extracted, by applying a digital-filtering technique (Nakazawa et al., 1997) to time-series data of both observation and model for stationary aircraft observation over Tasmania, Japan and Surgut

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Seasonal variation **: composite of 3 harmonics with respective periods of 4, 6 and 12 months**

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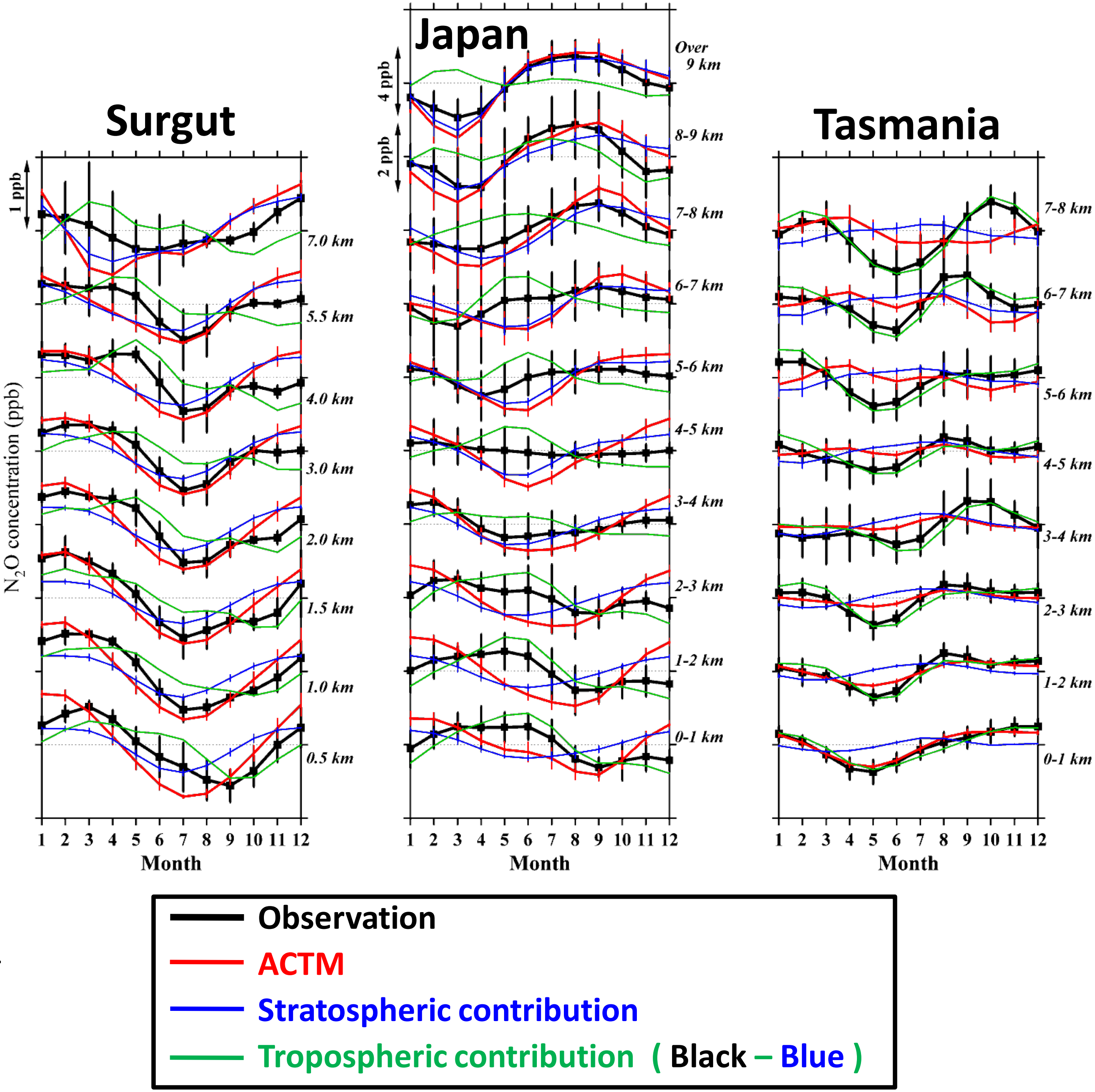
Seasonal cycle is defined by average of the seasonal variation for 3-6 years

N2O seasonal cycle over stationary aircraft observation sites

Surgut : Minima in summer due to the stratosphere are well reproduced at 1 - 5.5km. ACTM possibly overestimate the stratospheric effect at 7km.

Japan : Strong stratospheric effects over 7km are well captured by ACTM. Maximum of surface flux in early-summer is indicated by tropospheric contribution near surface.

Tasmania : The stratospheric effect is small. Model fluxes seem to be reasonable from results at 0-1km.

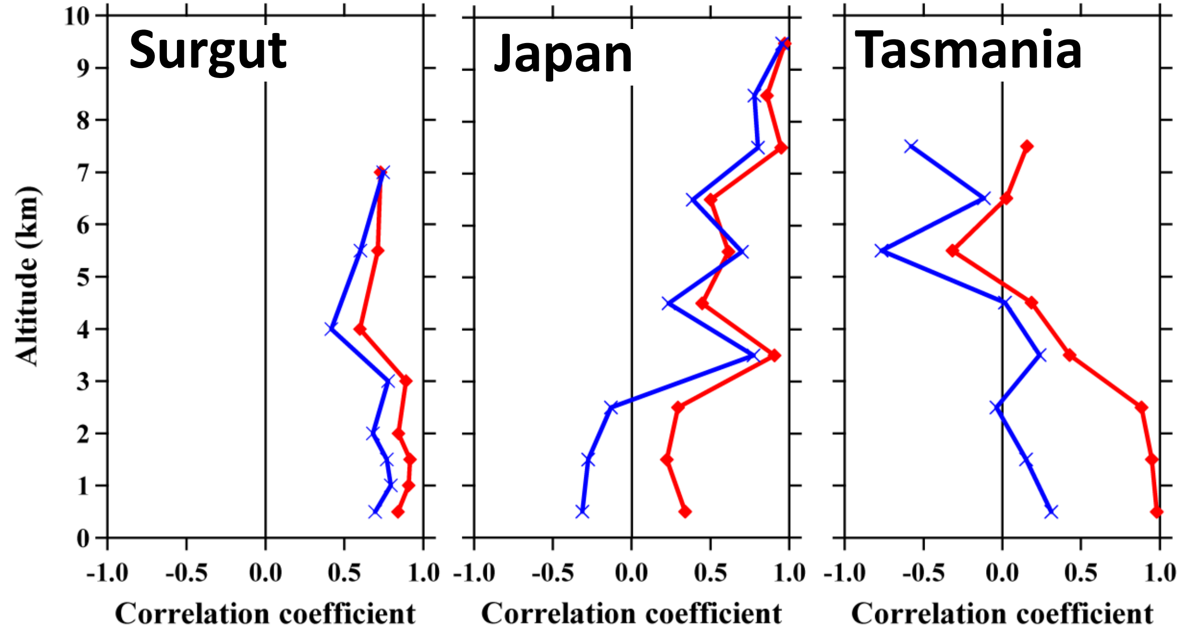


Surgut : The stratospheric signal is dominant for the seasonal cycles at all altitudes

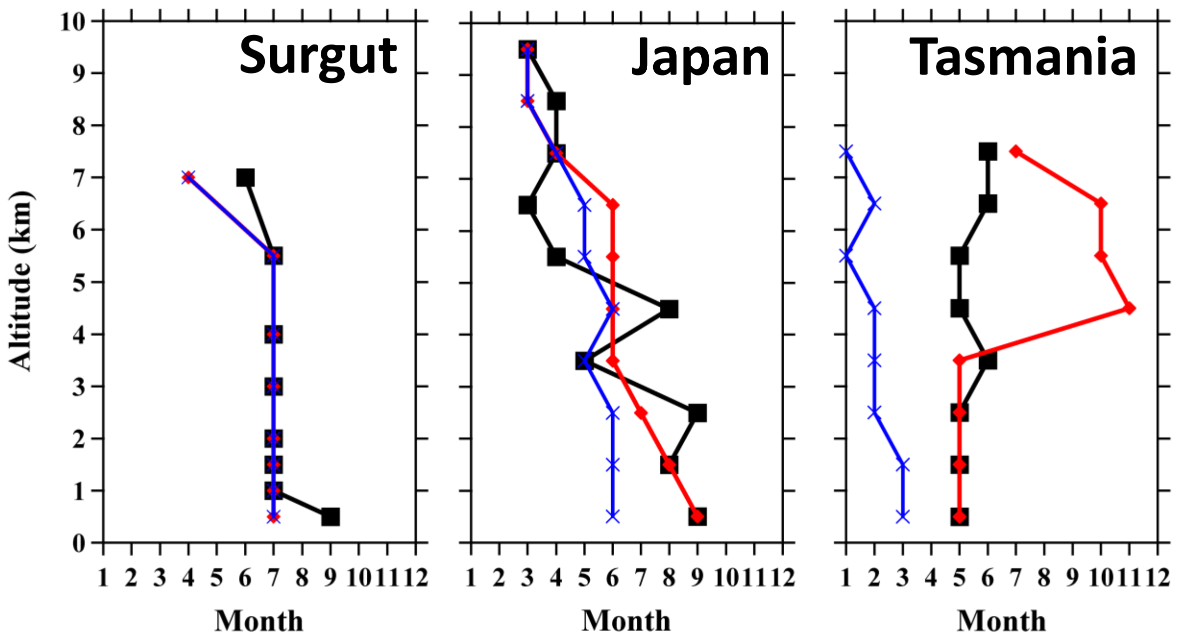
Japan : The stratospheric signal is dominant in the free troposphere, but influences of surface fluxes seem to be strong near surface.

Tasmania : The stratospheric effect is small, and the tropospheric contribution seems to be strong below 3km.

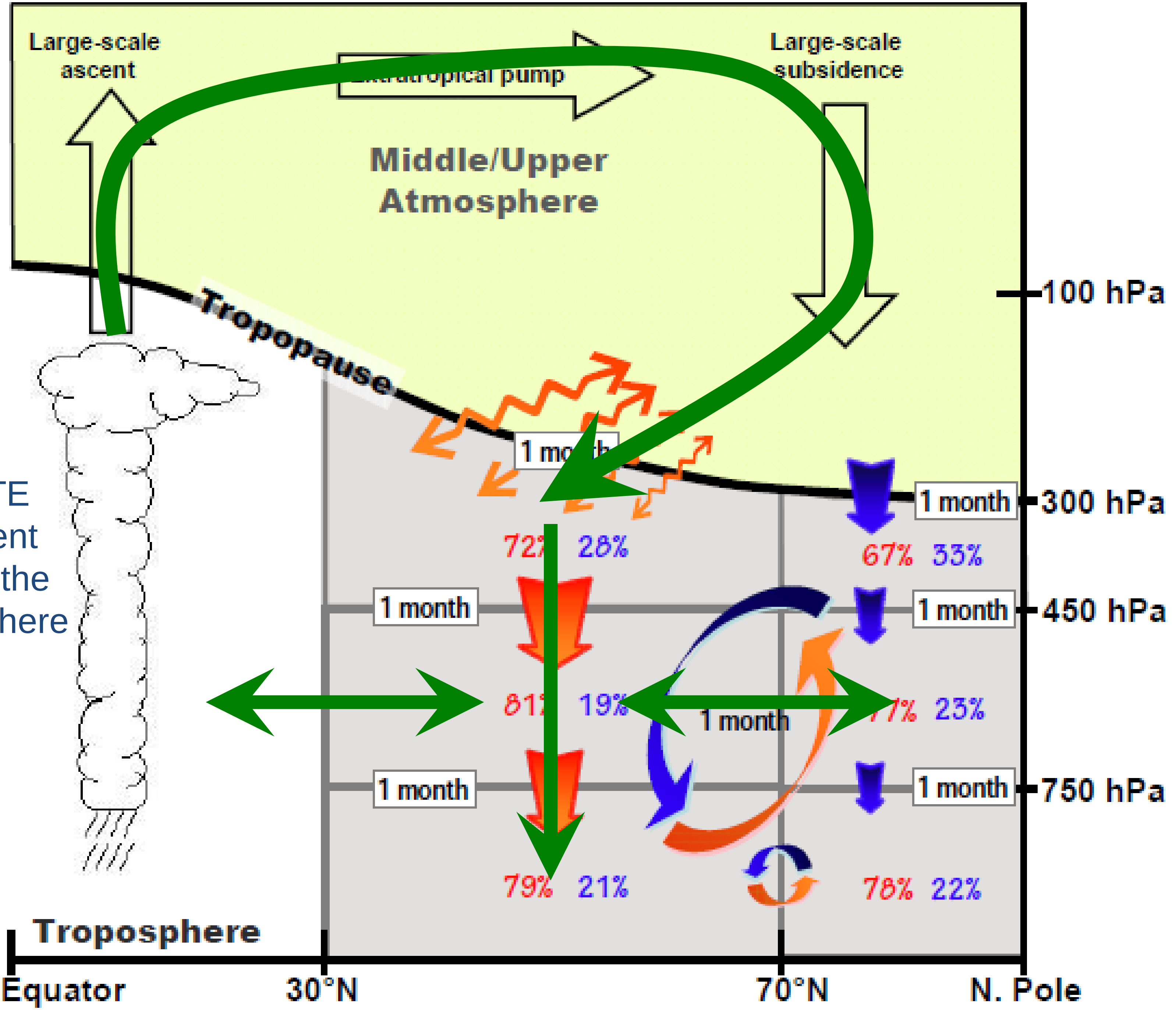
Correlation coefficient



Month of seasonal minimum



A schematic diagram of STE and subsequent transport into the lower troposphere



Holton et al (1995), but modified by Liang et al. (2009)

Summary

- ACTM reasonably reproduced AURA-MLS satellite observation in the stratosphere and CONTRAIL-ASE observation in the upper troposphere .

- N2O seasonal cycle over stationary aircraft observation site showed different feature in terms of large-scale transport and the surface flux in different site

Surgut : transport of the stratospheric signal
Japan : stratospheric and surface-flux signal
Tasmania : the tropospheric or surface-flux signal

- Surface fluxes around Japan are indicated to be maxima in early summer

Acknowledgment

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