

Stratospheric Denitrification in WACCM and Impacts on Polar Chemistry

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+

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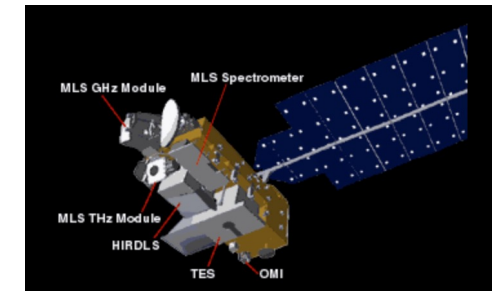
WACCM / Cheyenne



MIPAS ENVISAT



Chemistry



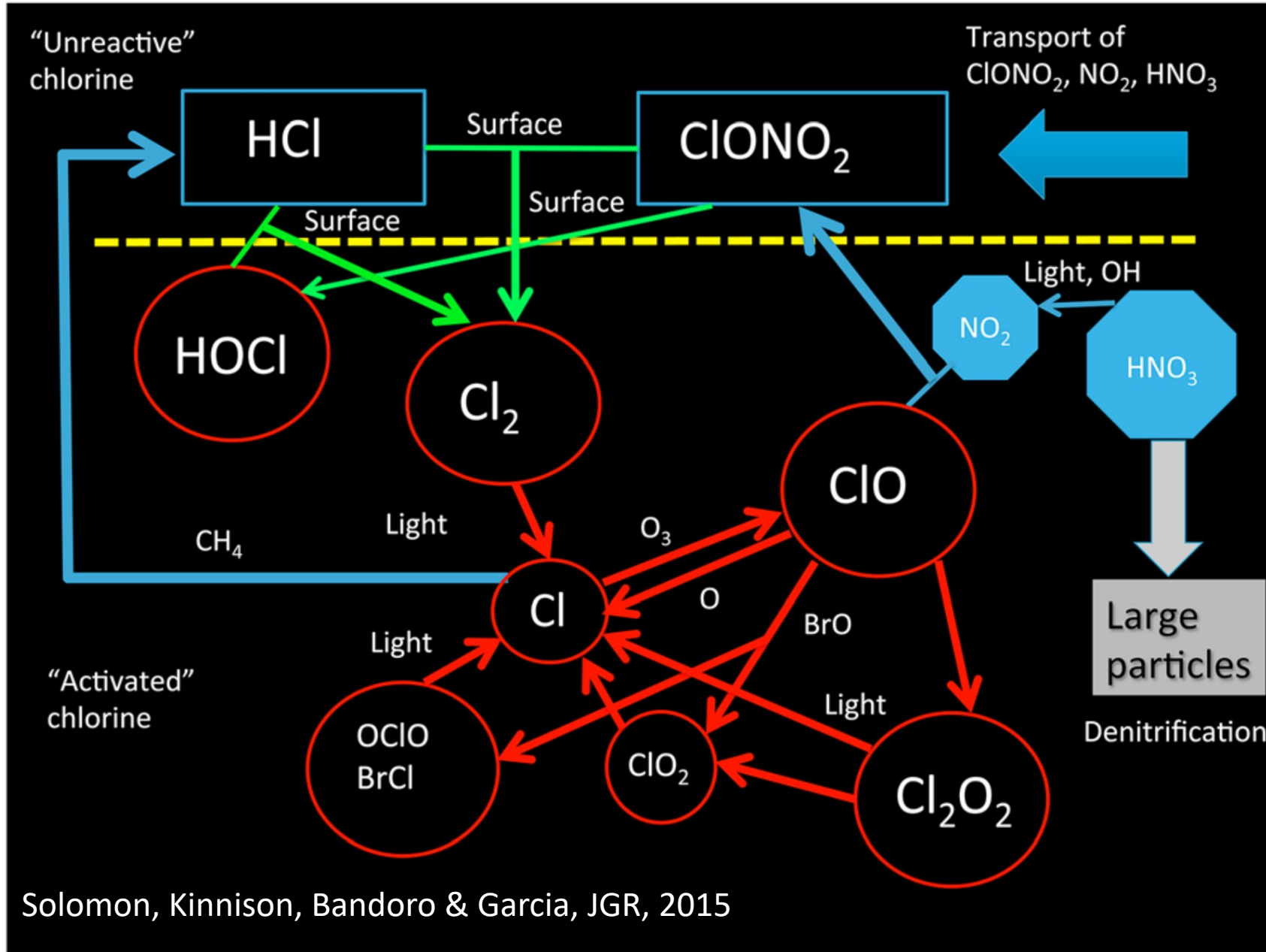
OMI/MLS Aura



Ozonesondes



Whole Atmosphere
Community Climate Model



Why do we care about Denitrification?

- >195K HNO₃ (g), Liquid binary sulfate (LBS)
- ~195K NAT forms deNO_y occurs; larger particles, faster settling rate, more deNO_y
- ~192K Supercooled Ternary Solution forms (STS) – Liquid aerosols that swell and uptake HNO₃ and HCl. Main SAD that activates (inorganic chlorine)

Competition between **activation** and **deactivation** in the polar spring.

Observations and Model version

Satellite Data:

- **HNO₃ (g) *** Microwave Limb Sounder (MLS) (Aura)**, ~4km vertical resolution
 - ~4km vertical res., >0.6ppbv precision
 - Examined only year 2020 (cold NH winter)
- **HNO₃ (g) *** Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), V8**
 - ~3km vertical res., **0.2 ppbv** precision
 - Examined years 2002-2012
- **Total Column Ozone *** Ozone Monitoring Instrument (Aura)**

Ozonesonde

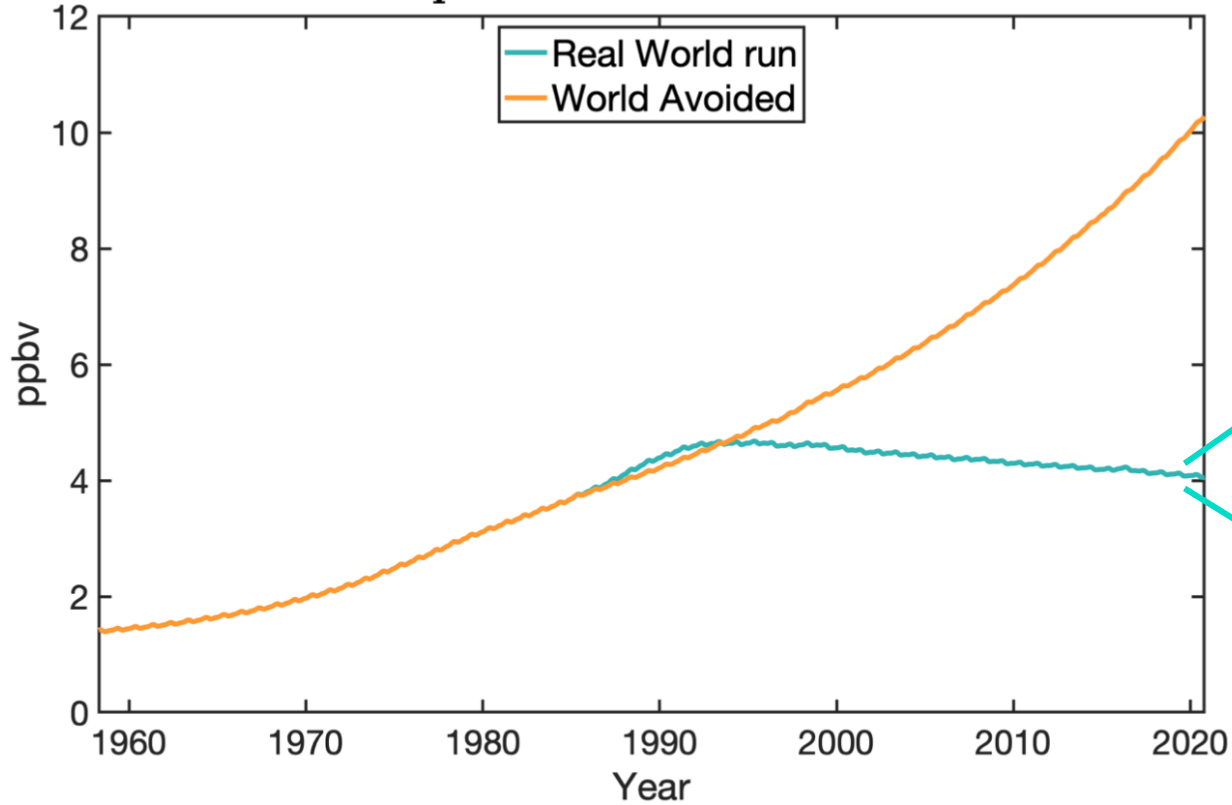
- @Eureka Station (80°N, 86°E)
- Electrochemical conc. cell, precision 3-5%, uncertainty +-10%

Model: CESM2 (WACCM6):

- 2-Deg horizontal, 0-140km
- Meteorological field constraints from MERRA-2– representative temperatures!

Wilka, Solomon, Kinnison, Tarasick, An Arctic ozone hole in 2020 if not for the Montreal Protocol, Atmos. Chem. Phys., 21, 15771–15781, 2021

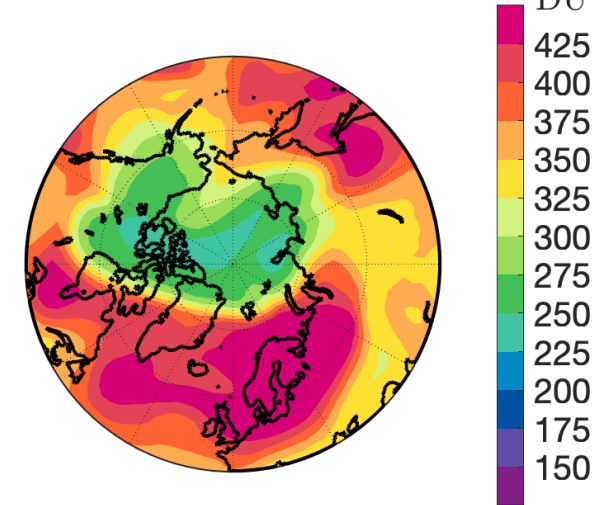
Total Equivalent Effective Chlorine



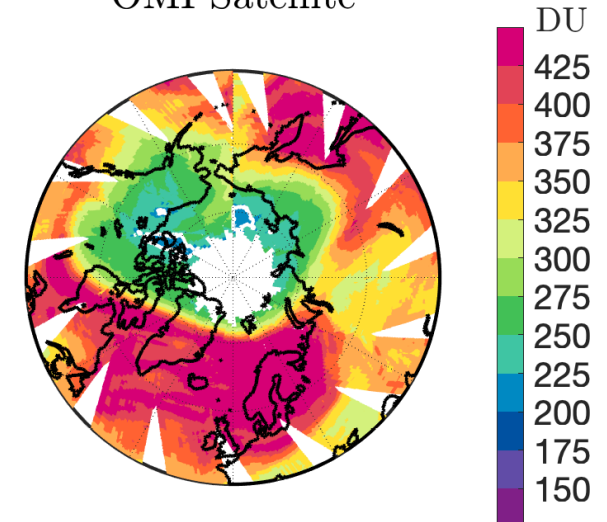
Excellent agreement with OMI.

Total Column Ozone on 13 March 2020

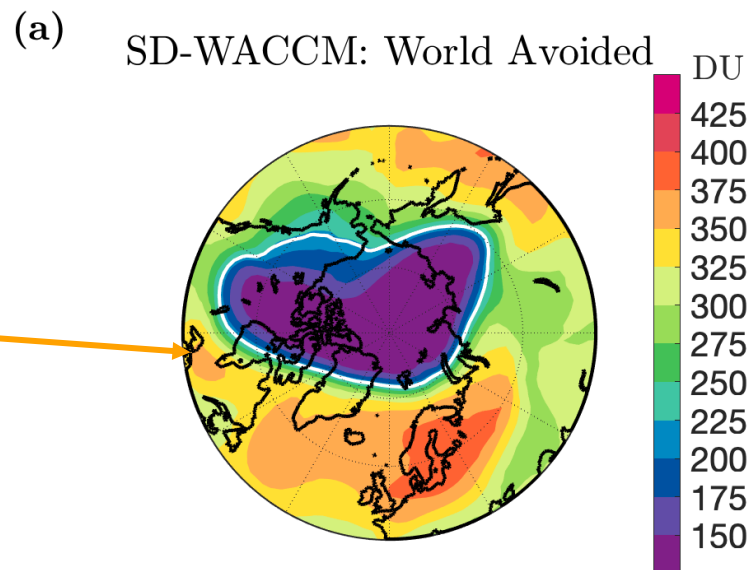
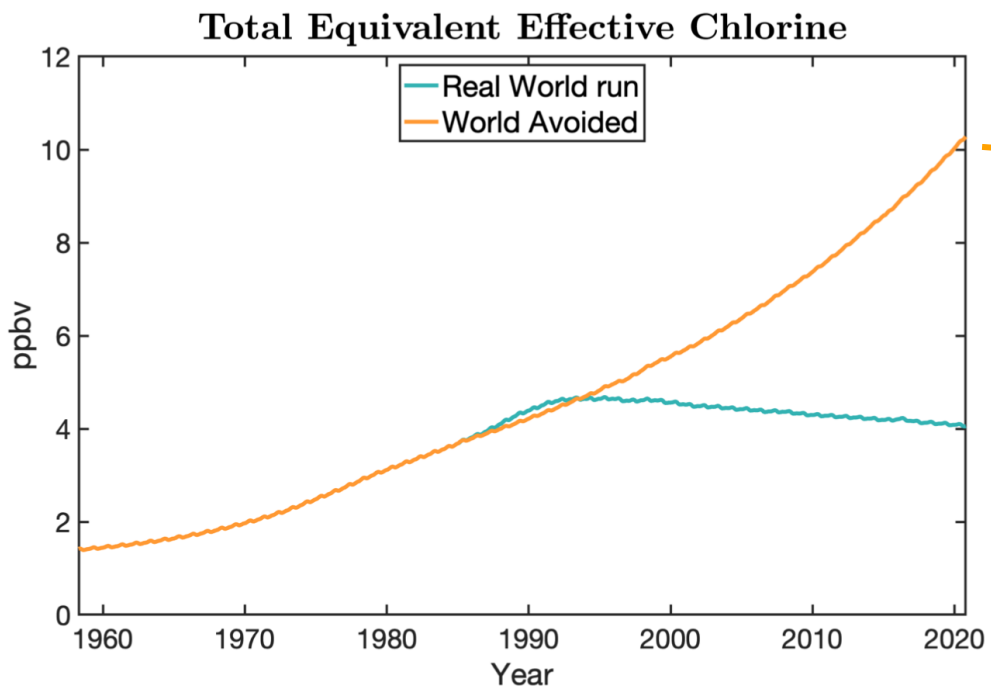
(b) SD-WACCM: Real World



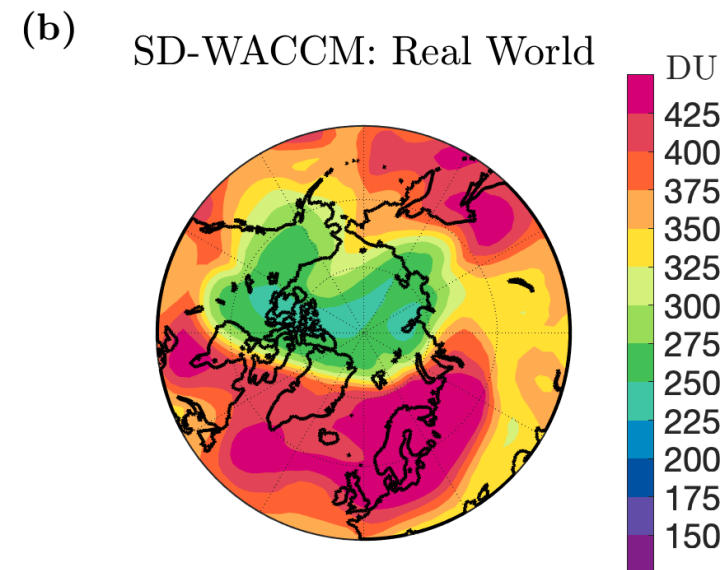
(d) OMI Satellite



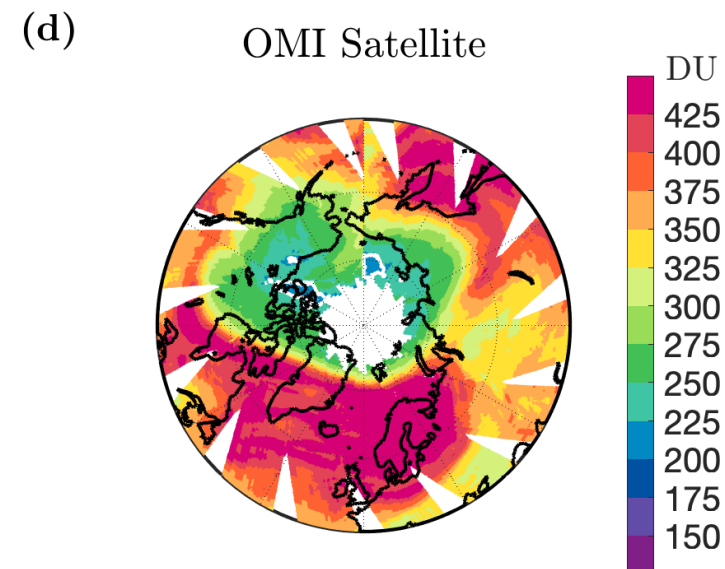
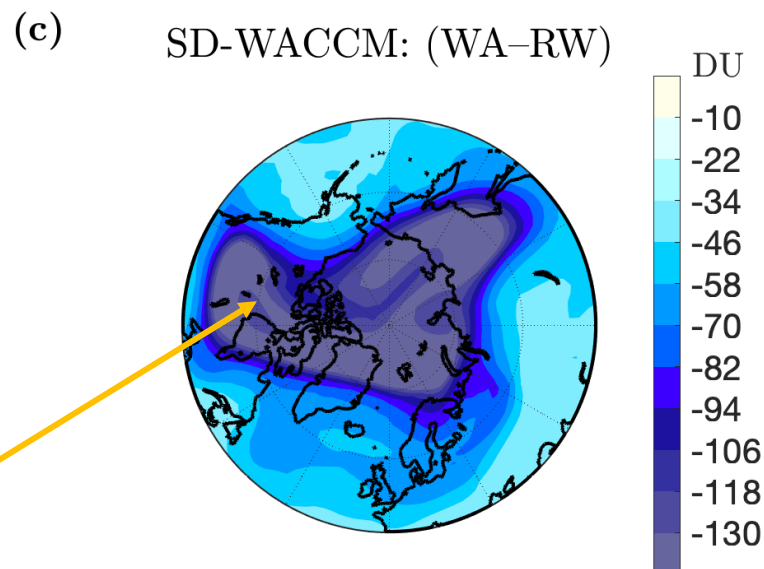
Total Column O₃ on 13 Mar 2020



Area < 220 DU = 19.71 million km²



Area < 220 DU = 0.00 million km²



Area < 220 DU = 0.61 million km²

Difference [World Avoided – Real World]
>130DU depletion and a Northern Hemisphere
 Ozone hole area (~**20 million km²**) similar to
 Southern Hemisphere.

NAT Settling and Denitrification in WACCM

- Derive the **amount of condensed phase HNO₃** (assume supersaturation of 10)
- Assume a **lognormal size distribution**, specify the **width** and **#particles cm⁻³**.
- Derive a Mean **Radius** (deNO_y) and **Surface Area Density** (het. Chemistry)

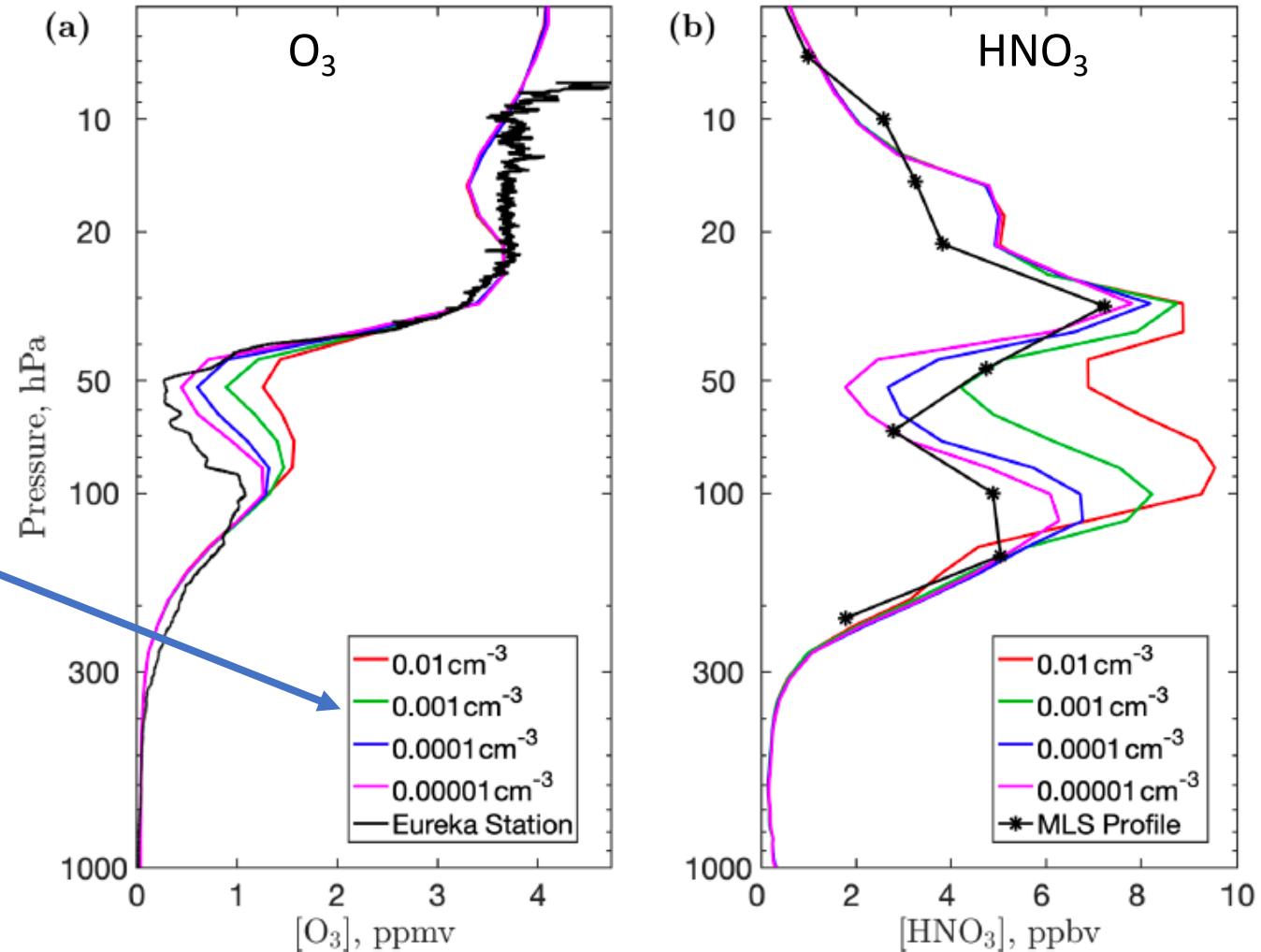
NH observations suggest NAT rocks form (Fahey et al., 2001).

NAT radius is proportional to the amount of **condensed phase HNO₃** and **#particles cm⁻³**.

The **NAT radius** is used to derive the **settling velocity** for NAT.

Use one # global value for #particles per cm⁻³.

Effects of NAT Particle Density Parameterization
on 27 Mar 2020 near Eureka Station



Next Step: We wanted to use a more quantitative approach at deriving the **# NAT particles per cm⁻³** to:

- Better represent HNO₃(g) and
- the amount of HNO₃ available for STS (ozone depletion).

Approach:

- Comparison to MIPAS HNO₃(g)
- Also examined MIPAS O₃ and ClONO₂ (not discussed today).

Analysis Approach Using MIPAS data and Model Output

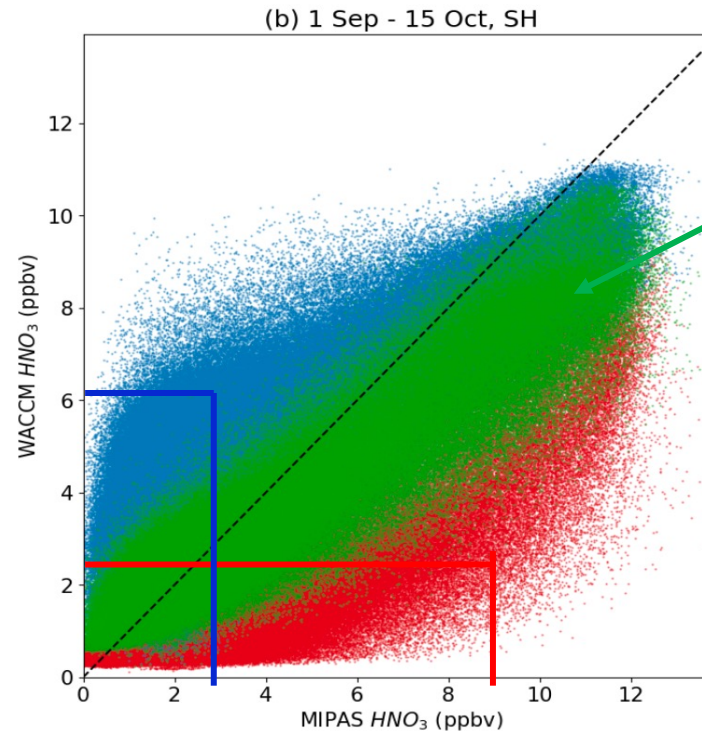
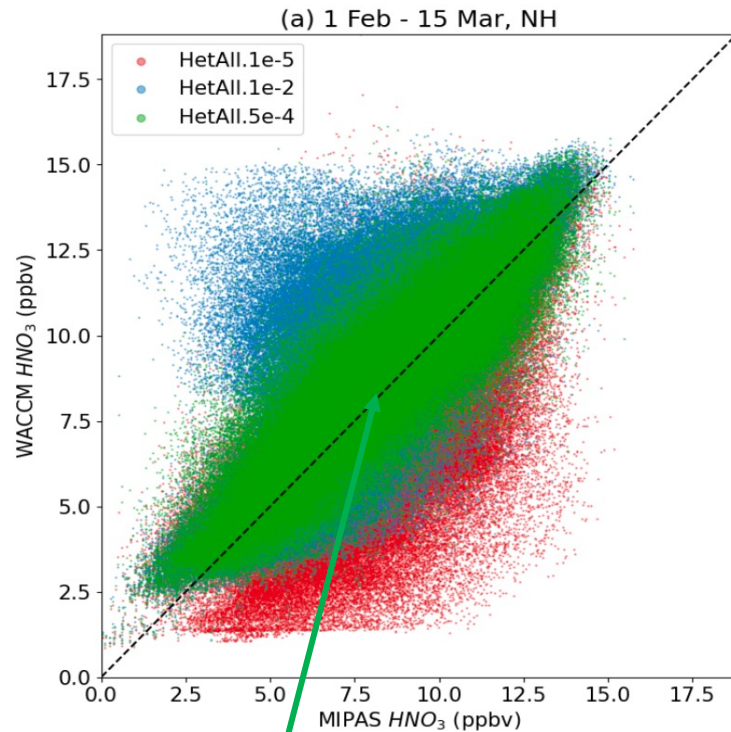
- SD-WACCM6 simulations for the 2002-2012 MIPAS period using multiple NAT particle densities ($1e-5$, $5e-4$, $1e-2$).
- Used coordinate analysis output approach where we output HNO_3 profiles from the model at measurement locations (lat/lon/time)
- Sampled only model and observations within the NH and SH polar vortex (using the Nash Criteria; i.e., Ertel PV max gradient) during Spring.
- Created **Scatter plots** (Obs vs Model sims) – **show today**.
- Created **vortex average time series** for given years and 2002-2012 Average – **show today**.
- Created **PDFs** (Obs vs Model sims) – **show today**
- Created **Cumulative density functions** and derived the maximum difference between obs and model simulation. (Zambri, Kinnison, Solomon, GRL, 2020) – **not shown today**.

Weimer, Kinnison, Wilka, and Solomon, Effects of denitrification on the distribution of trace gas abundances in the polar regions: a model-data comparison, ACP, in press, 2023.

Vortex average 2002-2012: Model uses MIPAS profile coordinates (lat, lon, time)

NAT particle densities: **1e-5**, **5e-4**, **1e-2**.

NH **Spring, 30-150hPa** SH



The SH **5e-4** value does tend to overestimate the denitrification.

NH fits the **5e-4** NAT density really well – consistent with the Wilke et al. study.

Best: NAT particle Density of $5 \times 10^{-4} \text{ cm}^{-3}$

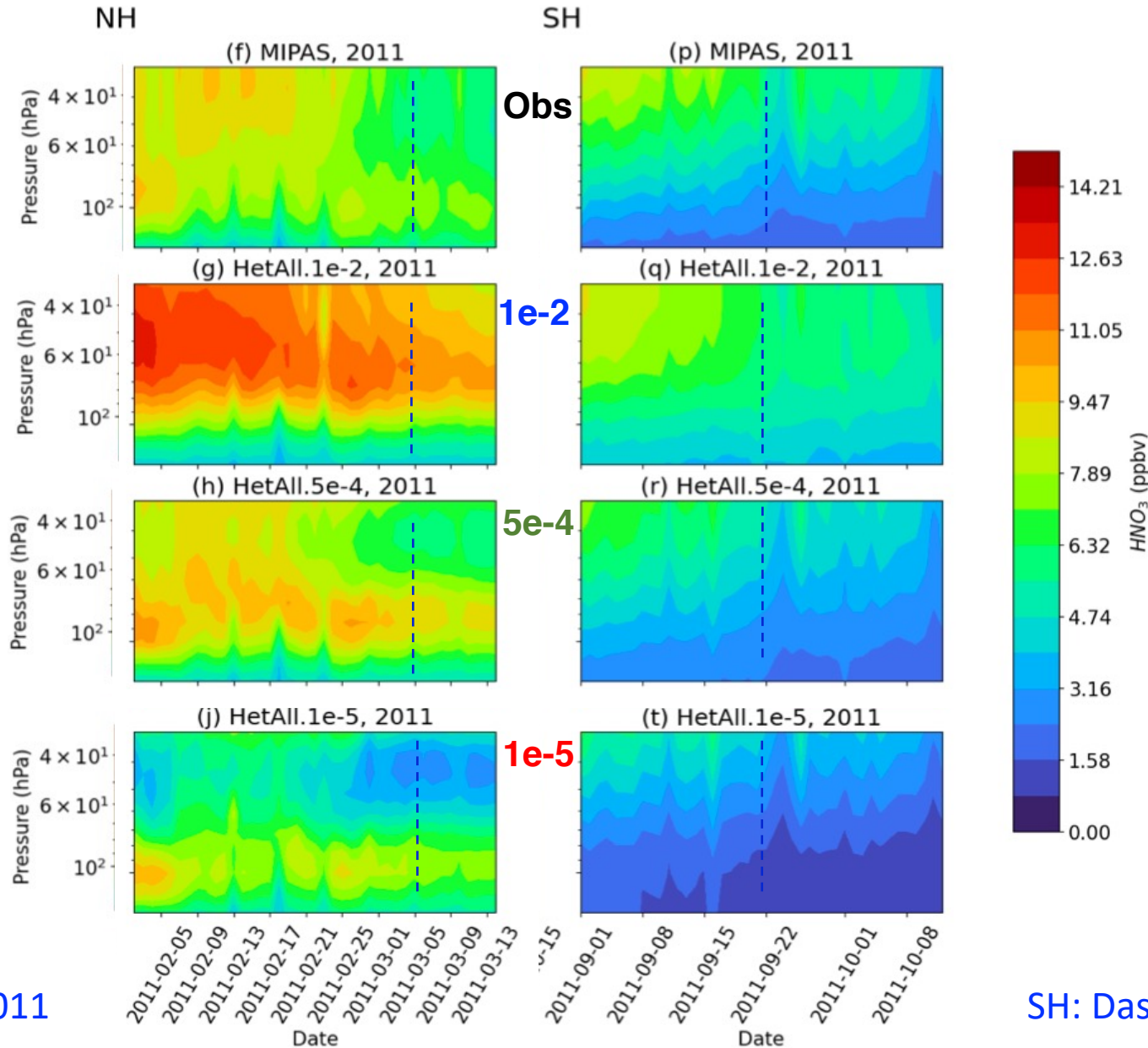
Vortex average time series for year 2011(150-30hPa)

2011 cold NH year, with noticeable deNOY

Very little deNOY with this NAT density

Consistent with Obs.

Too much DeNOy



As the tracer/tracer comparison showed, $5e-4$ NAT density does a good job of representing the deNOY

NH: Dashed line is 3/1/2011

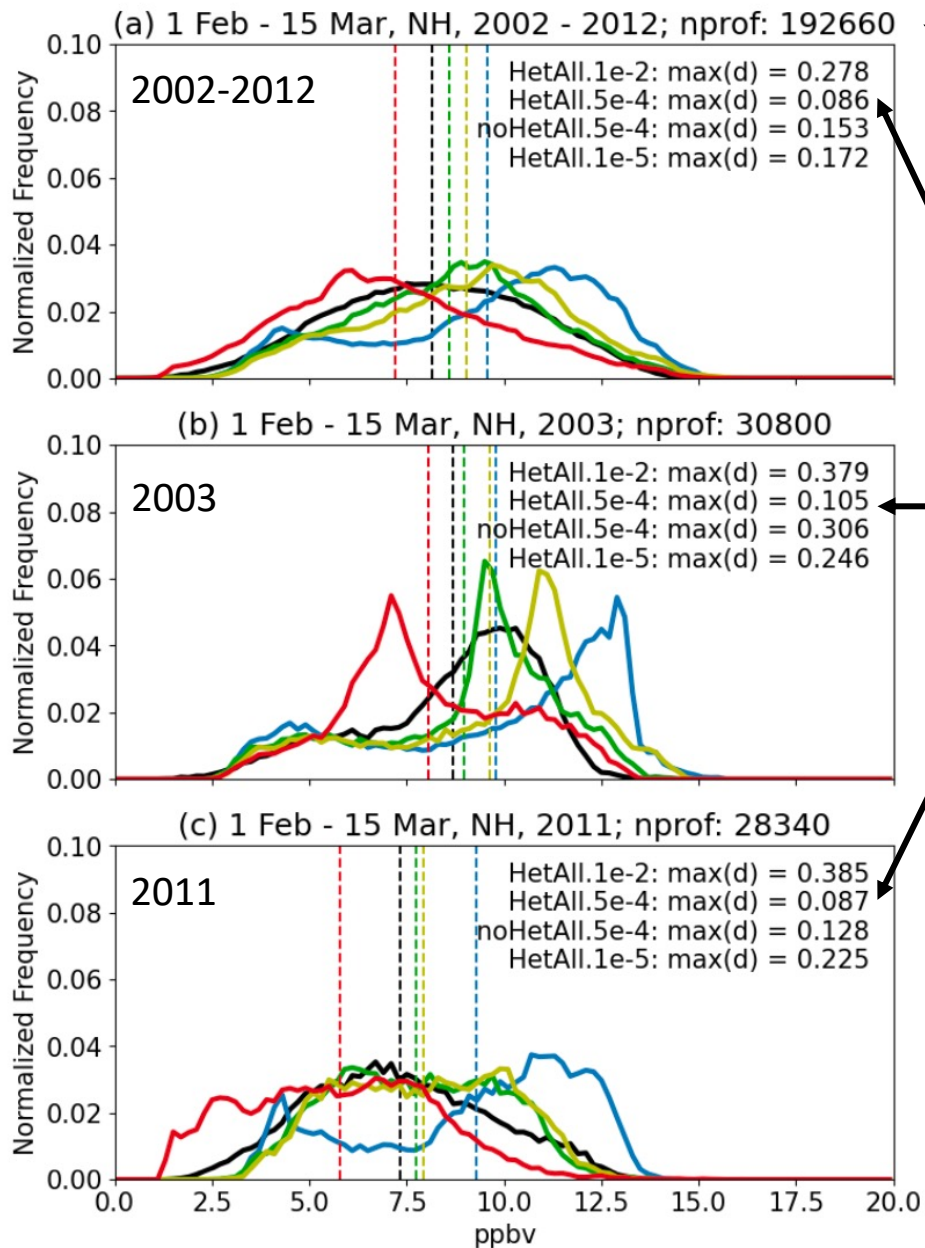
SH: Dashed line is 9/22/2011

Northern Hemisphere

Particle density:

1e-5, 5e-4, 1e-2.

Black line = MIPAS



Note: ~200,000 profiles

The **Max(d)** is the difference in the Cumulative density function. **Smaller the number the more consistent with Obs.** The **5e-4** NAT particle density is the most representative of the observations. See Weimer et al., ACP, for more details.

One can visually see that the **5e-4** NAT particle density agrees better with MIPAS.

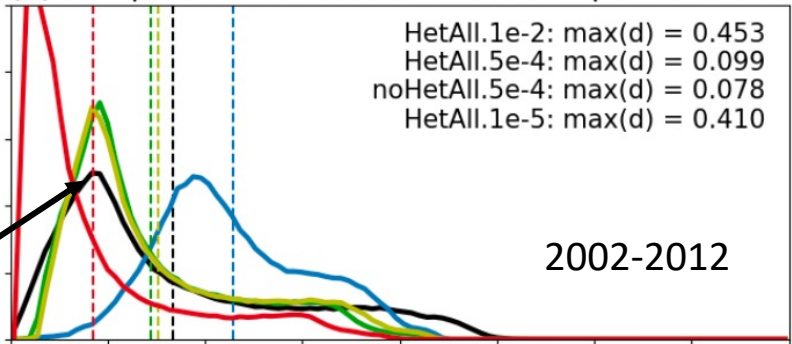
Southern Hemisphere

Particle density:
1e-5, 5e-4, 1e-2.

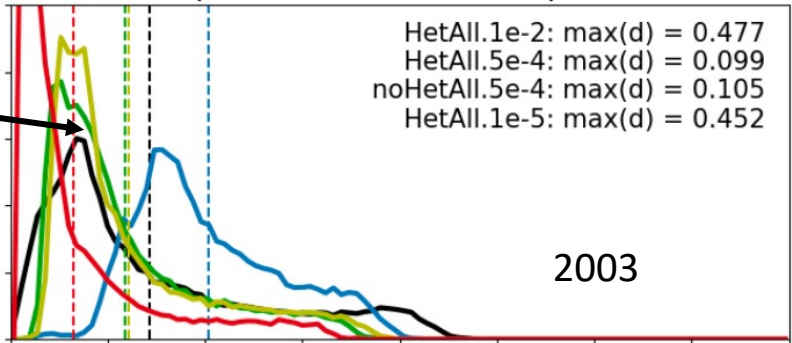
Black line = MIPAS

One can visually see that the 5e-4 NAT particle density agrees better with MIPAS.

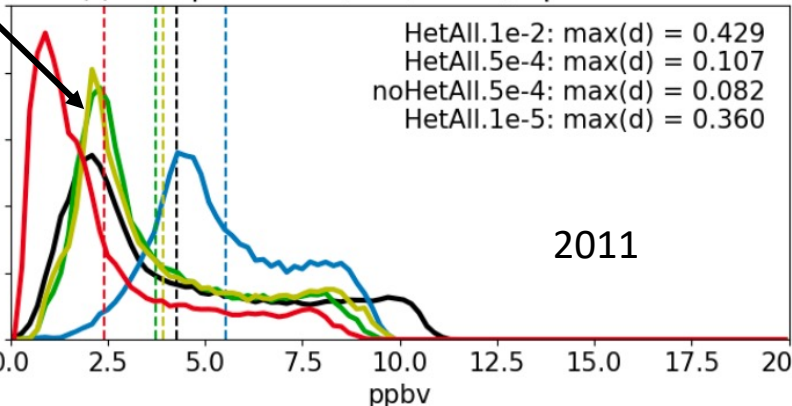
(d) 1 Sep - 15 Oct, SH, 2002 - 2012; nprof: 287100



(e) 1 Sep - 15 Oct, SH, 2003; nprof: 37460



(f) 1 Sep - 15 Oct, SH, 2011; nprof: 36580



Shape and means of the PDF is best represented by NAT particle Density of $5 \times 10^{-4} \text{ cm}^{-3}$

A NAT particle density of $5 \times 10^{-4} \text{ cm}^{-3}$ will be used in all versions of CESM2 “chemistry versions” going forward.

See the Weimer et al. ACP, 2023 paper for more details.

Thank you for your attention!



NCAR



WACCM

*Whole Atmosphere
Community Climate Model*

