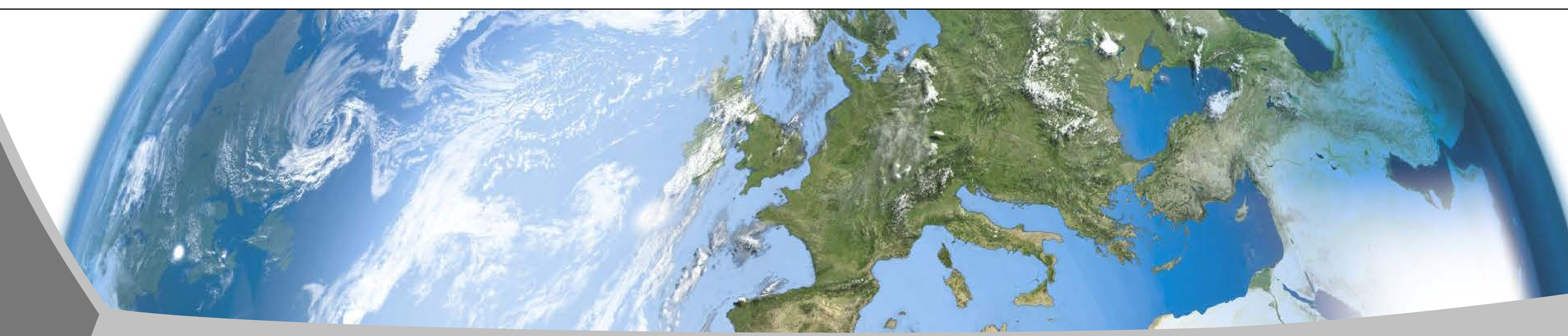


DLR

# DKRZ – project 0783 - AEROTROP

# Reactive Nitrogen in a CGCM: Impact of model uncertainties on trace gas mixing ratios and aviation $NO_x$ effects

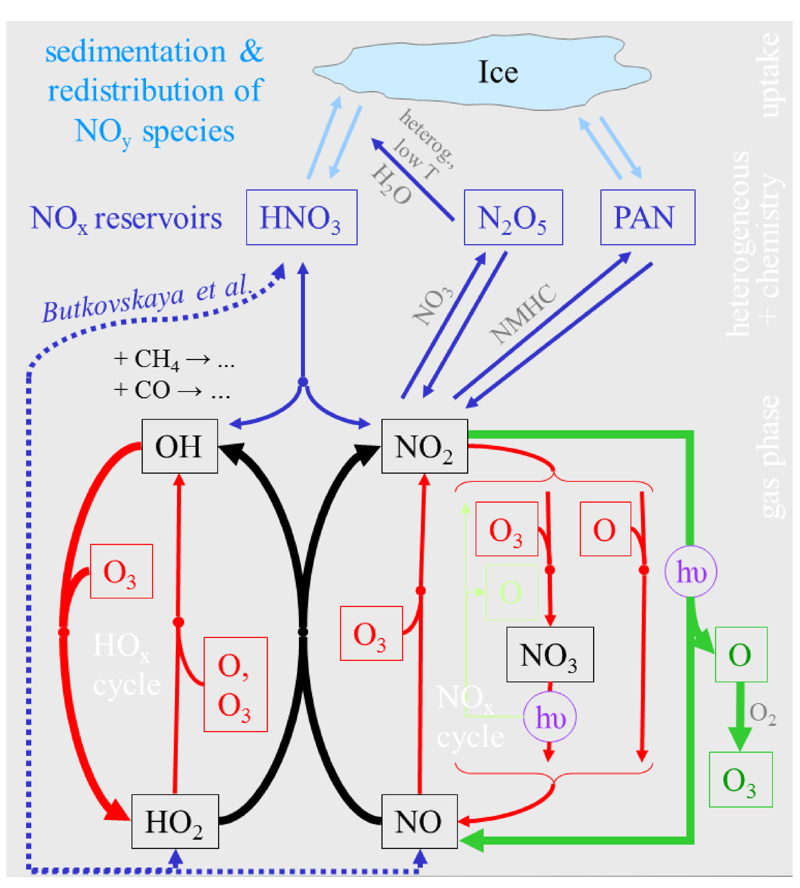
K. Gottschaldt and C. Voigt


## Abstract

Reactive nitrogen ($NO_y$) species are involved in the chemical processes affecting the atmospheric mixing ratios of the greenhouse gases ozone and methane.
Here we investigate three uncertainties related to the representation of $NO_y$ in the EMAC-CGCM:

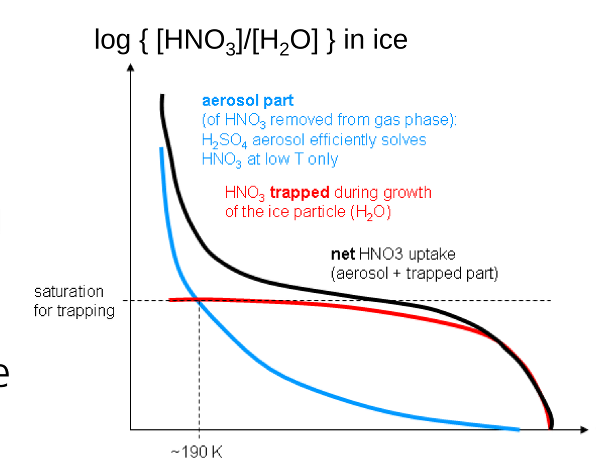
- Impact of the reaction $HO_2 + NO \rightarrow HNO_3$ on atmospheric oxidizing capacity and methane lifetime. The reaction is not included in standard chemical mechanisms yet.
- Global sensitivity of aviation $NO_x$ effects to the above reaction
- Impact of different parameterizations for $HNO_3$ scavenging by cirrus ice particles on the $NO_y$ budget in the tropopause region


## Introduction

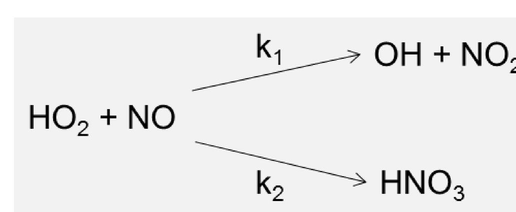



**Options for $HNO_3$ uptake on ice:**

- fixed scavenging value
- Henry's law
- Langmuir uptake
- Trapping (/7/)




**Reaction channels of $HO_2$ + NO:**




Less than 2% go into $HO_2 + NO \rightarrow HNO_3$, but during each iteration of the catalytic $HO_x$ & $NO_x$ cycles. The rate coefficient $k_2$ depends on pressure, temperature, or additionally on humidity (/1/).

## Methodology

- EMAC 1&2 (/5,6/), T42/L90MA resolution (up to 1 Pa)
- EVAL Chemistry (~ 270 gas phase reactions, 13 het, 41 aq)
- Dynamics nudged towards ECMWF data
- QCTM: constant feedback from chemistry on dynamics (/2/)
- evaluate signal of chemical **perturbations** with respect to a **control simulation**.

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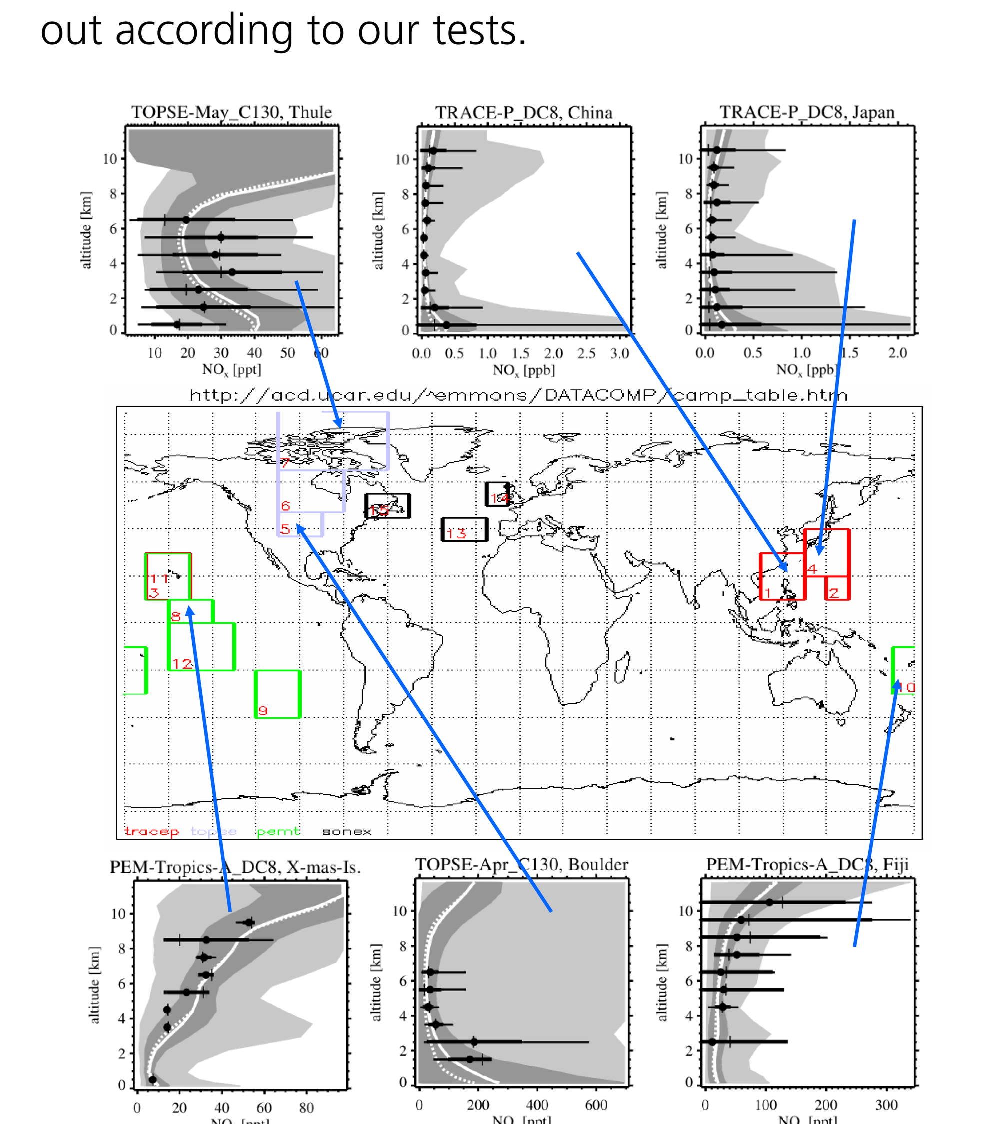
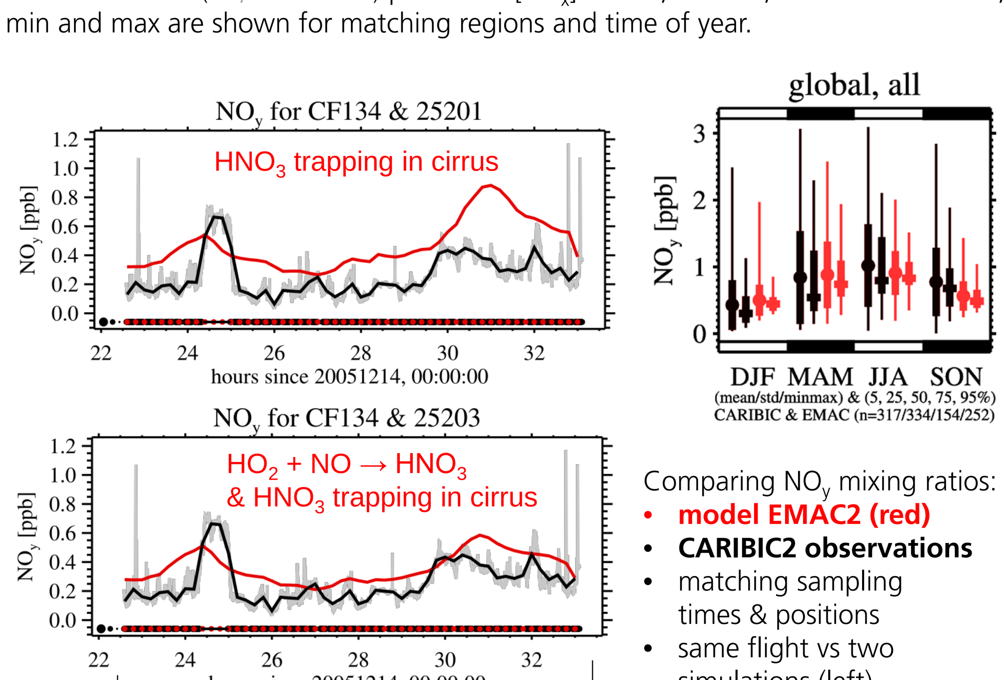
## Comparisons to observations

Simulated mixing trace gas ratios are compared to observations in an attempt to check if the results are reasonable for all model modifications. None of the simulations described here could be ruled out according to our tests.




Examples for the climatological comparison of modelled (EMAC1, grey background) versus observed (/3/, black bars) profiles of $[NO_x]$. Mean, median, standard deviation, min and max are shown for matching regions and time of year.




Frankfurt → São Paulo

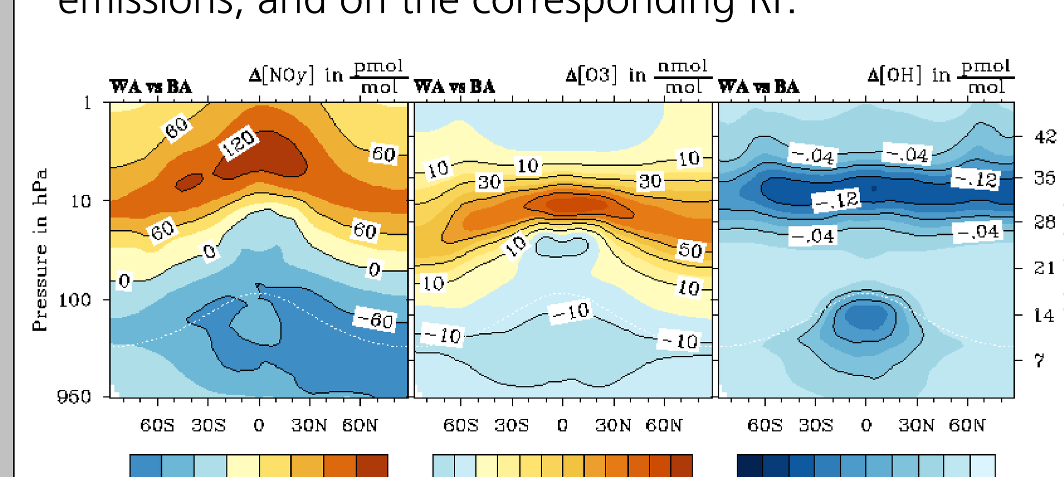
Comparing $NO_y$ mixing ratios:

- model EMAC2 (red)
- CARIBIC2 observations
- matching sampling times & positions
- same flight vs two simulations (left)
- all CARIBIC flights of 1 year vs top left simulation (right)

## Effects on background chemistry

| | | | |
|---|---|---|---|
| **BA** | • $k_2 = 0$ • Aviation $NO_x$ | **B0** | • $k_2 = 0$ • no Aviation $NO_x$ |
| **WA** | • $k_2$ (p, T, $c_{H2O}$) • Aviation $NO_x$ | **W0** | • $k_2$ (p, T, $c_{H2O}$) • no Aviation $NO_x$ |

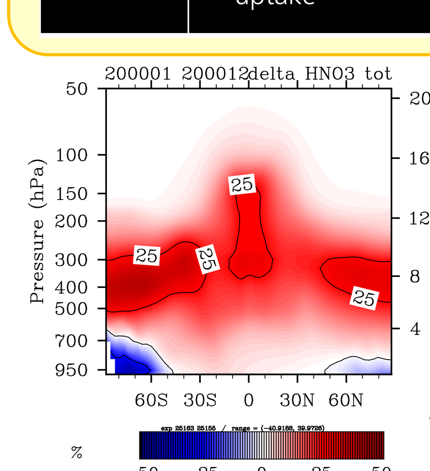
Including $HO_2 + NO \rightarrow HNO_3$ in the chemical mechanism decreases $[NO_x]$, $[NO_y]$ and $[O_3]$ in the tropopause region. The total ozone column decreases by about 2%. Lower OH mixing ratios throughout the atmosphere increase methane lifetime (for oxidation $CH_4$+OH) by about 50%. The uncertainties associated with the $HNO_3$-forming channel thus propagate considerable additional uncertainties on methane lifetime estimates, on predictions of future methane abundances, on estimates of lifetime changes due to anthropogenic emissions, and on the corresponding RF.




Annual zonal mean mixing ratios: Difference between a simulation with $HO_2 + NO \rightarrow HNO_3$ and a simulation without it (/4/).

| | |
|---|---|
| **BA** | • Langmuir $HNO_3$ ice uptake |
| **nLA** | • no $HNO_3$ ice uptake |

$[HNO_3]$ in the UTLS would be about 30% higher without uptake of $HNO_3$ on cirrus ice particles and subsequent removal by sedimentation. The impact of different mechanisms for $HNO_3$ ice uptake on $HO_x$-$NO_x$ chemistry is currently being evaluated.

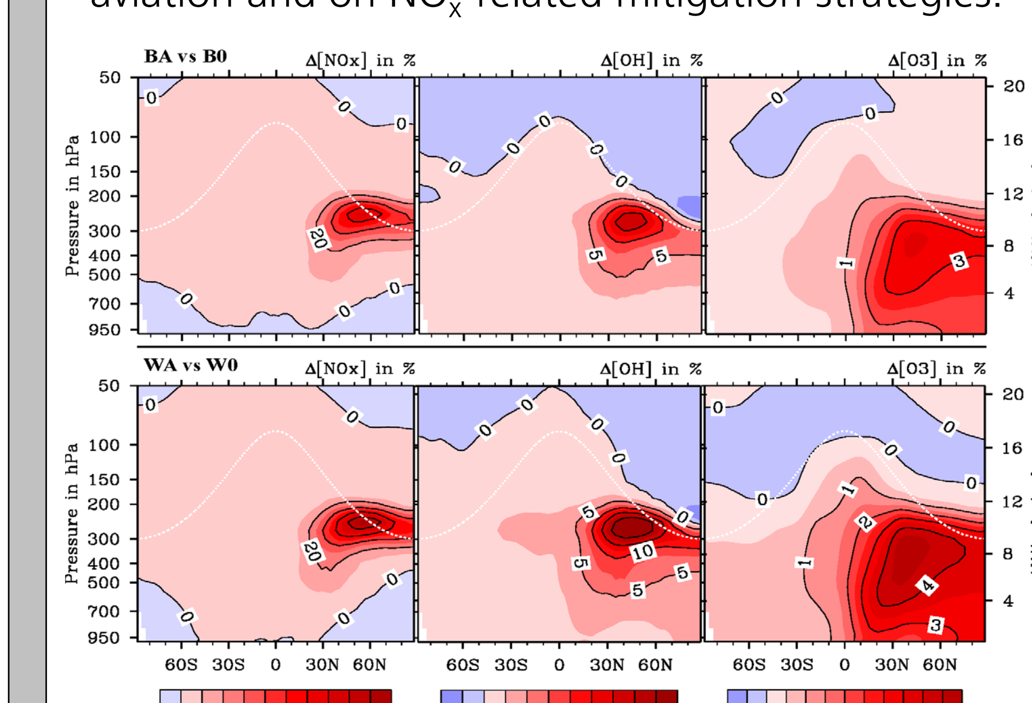



Annual zonal mean $HNO_3$ mixing ratios: Relative deviation between a simulation without uptake on ice (nLA) and a simulation with Langmuir uptake.

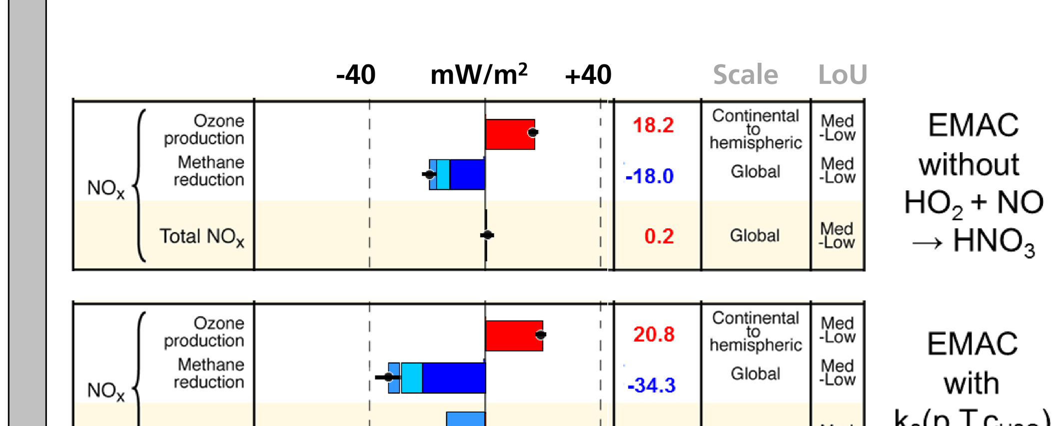
## Global sensitivity of aviation NOx effects to $HO_2 + NO \rightarrow HNO_3$

| | | | |
|---|---|---|---|
| **BA** | • $k_2 = 0$ • Aviation $NO_x$ | **B0** | • $k_2 = 0$ • no Aviation $NO_x$ |
| **WA** | • $k_2$ (p, T, $c_{H2O}$) • Aviation $NO_x$ | **W0** | • $k_2$ (p, T, $c_{H2O}$) • no Aviation $NO_x$ |

Warming and cooling effects of aircraft $NO_x$ emissions are both enhanced when considering the $HNO_3$-forming channel, but the sum is shifted towards negative radiative forcing. Uncertainties associated with the inclusion of the $HO_2 + NO \rightarrow HNO_3$ reaction and with its corresponding rate coefficient propagate a considerable additional uncertainty on estimates of the climate impact of aviation and on $NO_x$-related mitigation strategies.




Relative deviations (e.g. $100 \cdot \frac{BA - B0}{B0}$) of annual zonal mean mixing ratios of $NO_x$, OH and $O_3$ in a simulation with aviation $NO_x$ from one without (/4/).




Long-term Radiative Forcing (RF) components from sustained aircraft $NO_x$ emissions of the year 2000. The planet might be warmer without aviation $NO_x$.