# QOS 2021

October 3 (Sunday) - 9 (Saturday), 2021



[C\_226]

Origin of Tropospheric Air Masses
in the Tropical West Pacific
and related transport processes
inferred from balloon-borne Ozone and
Water Vapour observations from Palau

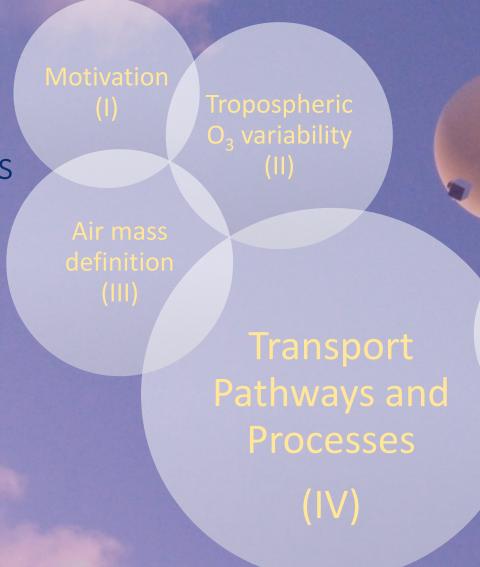


**Katrin Müller**, Ingo Wohltmann, Peter von der Gathen, Ralph Lehmann, and Markus Rex

Contact: Katrin.Mueller@awi.de
PhD thesis + 2 Manuscripts

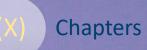












Apters \*/ More Info /Back

# (I) Motivation

#### QUADRENNIAL **OZONE SYMPOSIUM**

Key feature of the clean TWP troposphere: close coupling of the O<sub>3</sub> concentration and oxidizing capacity (OH), influencing overall transport of chemical species to the stratosphere.

To improve the limited availability of tropospheric O<sub>3</sub> observations from this key region, the Palau 🍁 was established in 2016 as part of the EU-project StratoClim.



#### Why the Tropical West Pacific (TWP)?

Major source region for stratospheric air in

boreal winter

Origin and transit region of corresponding air masses in

boundary layer and troposphere (Rex et al. 2014)



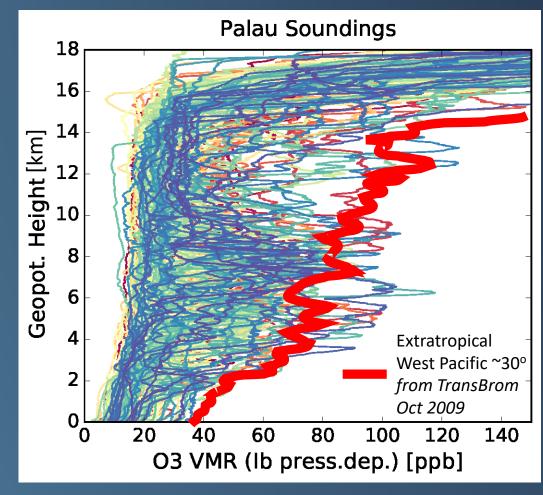
**Important region for supply** of chemical species

to the stratosphere

Need for monitoring of air composition and understanding of underlying processes and transport pathways to TWP



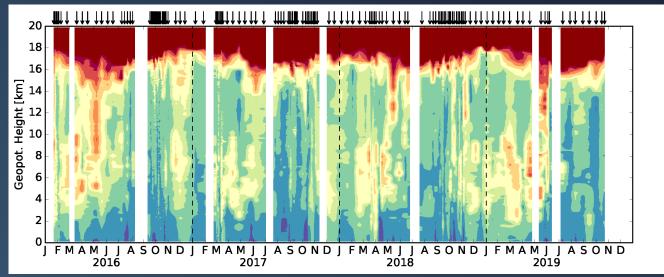


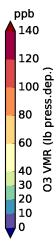


Tropospheric profiles with altitude: 145 sondes, 01/2016-10/2019

First characterization of tropospheric O<sub>3</sub> seasonality in the TWP with a multi-year continuous time series from ECC ozonesonde measurements every two weeks or in intensive campaigns (SPC 6A, Vaisala RS92/41). → Müller 2020

Special focus on quality issues of tropical soundings due to controversy around near-zero  $O_3$  observations in the TWP (e.g. Voemel and Diaz 2010, Rex et al. 2014, Thompson et al. 2019)





## (I) Why $O_3$ ? As a chemical tracer...





...for **local convective activity** in clean maritime air: "low" O<sub>3</sub> (e.g. Folkins, 2002; Folkins et al., 1999; Kley et al., 1996; Paulik and Birner, 2012; Solomon et al., 2005) and

... for **long range transport** processes to the region, either related to air pollution or stratospheric intrusions: **"high"** O<sub>3</sub> (e.g. Andersen et al., 2016; Browell et al., 2001; Randel et al., 2016; Tao et al. 2018; Thouret et al., 2000; Pan et al. 2015).

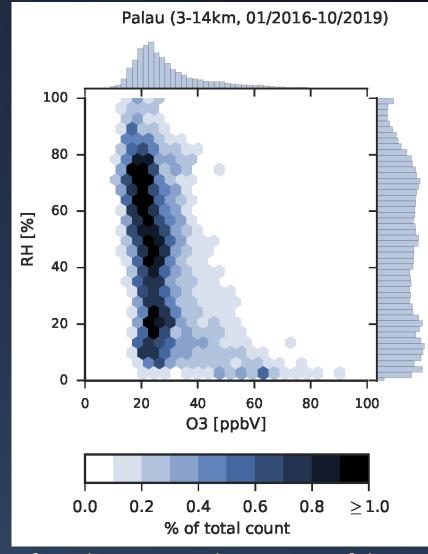
RH as a tracer for vertical displacement

High humidity due to convection, dryness due to large scale descent (e.g. Hayashi et al., 2008; Andersen et al., 2016; Cau et al., 2007; Dessler and Minschwaner, 2007)

#### **Central Question:**

Can we identify air mass origin and its seasonality with the observed O<sub>3</sub>/RH relation?

#### Free-tropospheric O<sub>3</sub>/RH distribution



unique for Palau compared to stations of the tropical SHADOZ ozonesonde network

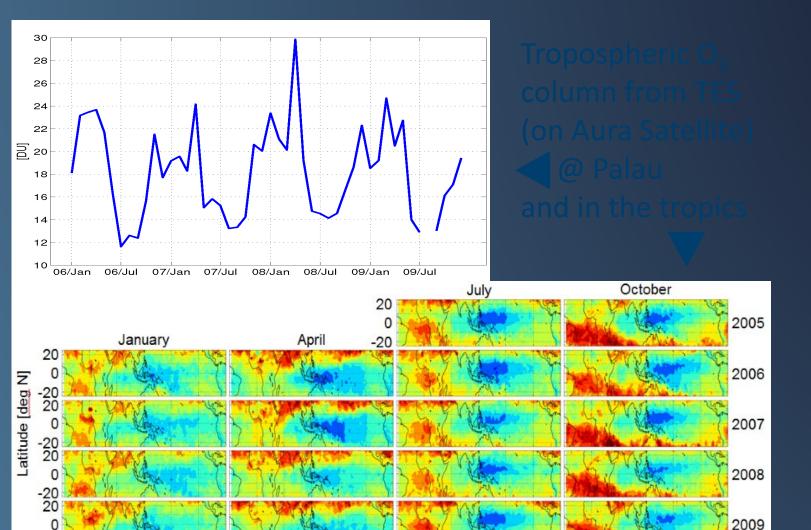
# (II) Tropospheric O<sub>3</sub> variability

180

Rex et al. 2014





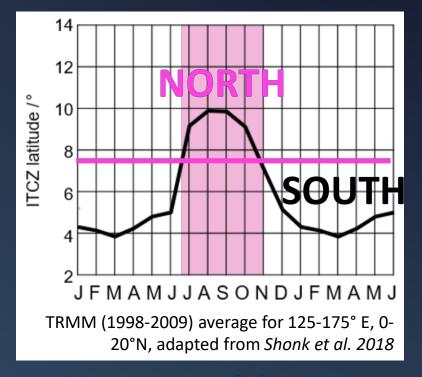


Longitude [deg E]

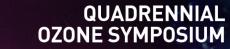
180

20 [DU]

- Seasonal drivers: circulation (Walker, Hadley, West Pacific Monsoon, Brewer-Dobson)
   → modulated by Inter Tropical Convergence Zone (ITCZ)
- Hot, humid & wet climate all year: high convective activity
- Important Variation: ENSO



# (II) Tropospheric O<sub>3</sub> variability

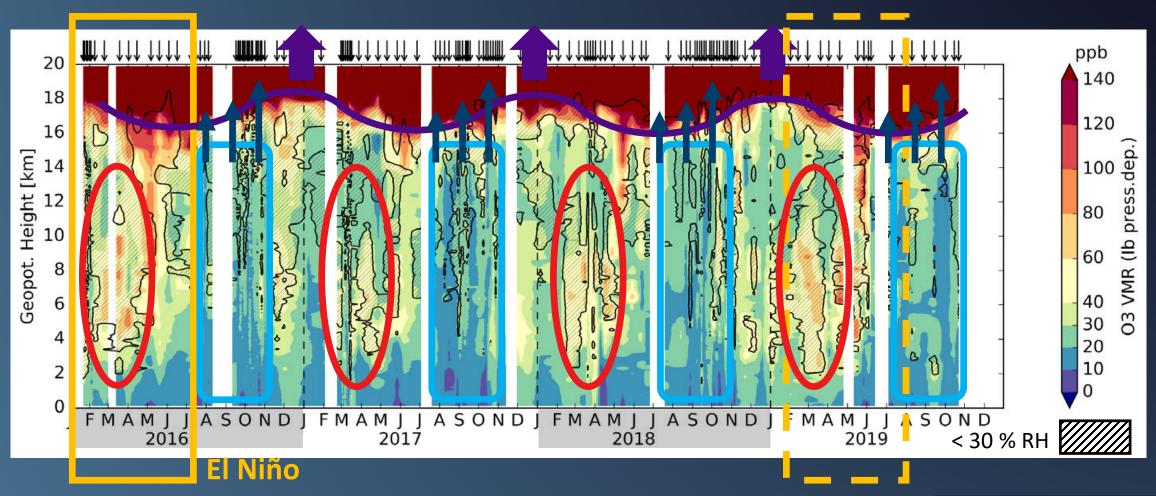




Annual TTL cycle: Brewer Dobson circulation

+ enhanced high altitude comments and the management of the comments and the comments are the comments and the comments are t

Onset of transport to the stratosphere and the tropospheric O<sub>3</sub> minimum occur simultaneously

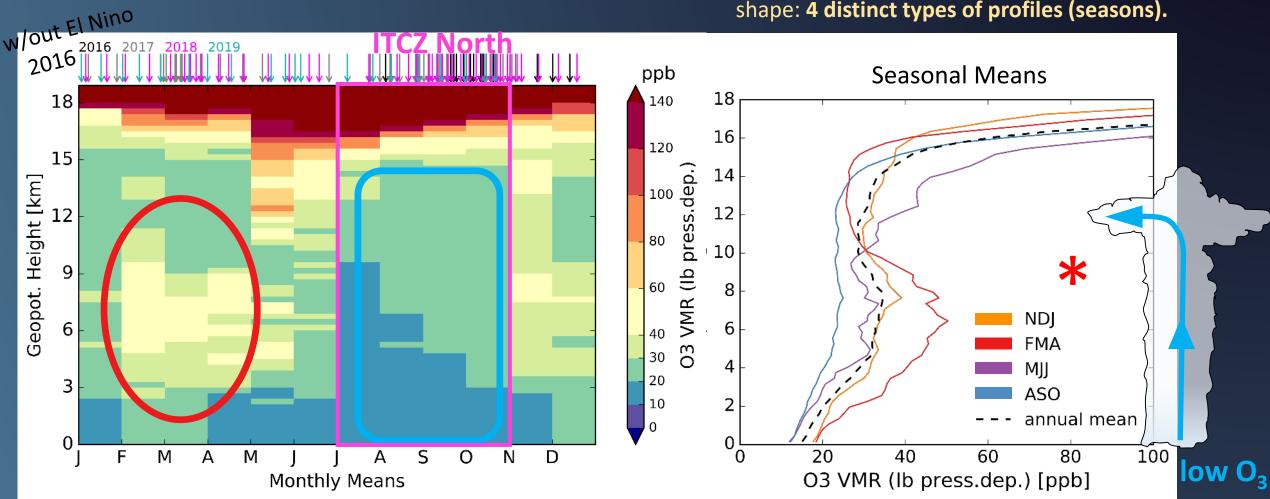


# (II) Tropospheric O<sub>3</sub> variability

# QUADRENNIAL OZONE SYMPOSIUM



Annual mean: typical (tropical) "S-Shape", monthly means grouped according to similar shape: 4 distinct types of profiles (seasons).



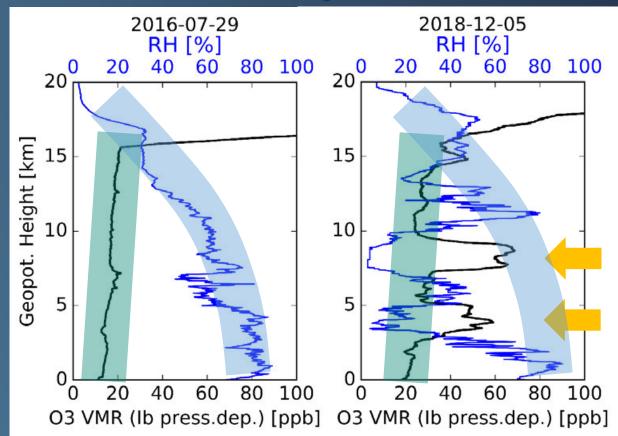
Monthly means highlight annual cycles,

O<sub>3</sub> minimum corresponds with
the ITCZ located North of Palau.

Deep convective detrainment can explain upper dent in the "S" (10-14 km); between 5-10 km or the belly of the "S": weak cloud-mass divergence, greatest anomalies from annual mean in ASO and FMA.

# NON NON

#### Example tropospheric O<sub>2</sub> and RH profile



convectively controlled, well-mixed background:

LOCAL mode

interruptive layers, controlled by transport:

**NON LOCAL mode** 

#### **Underlying processes:**

Local boundary layer air masses lacking pollution  $(\rightarrow low in O_3)$  are lifted locally by convection (humid), creating a uniform profile.

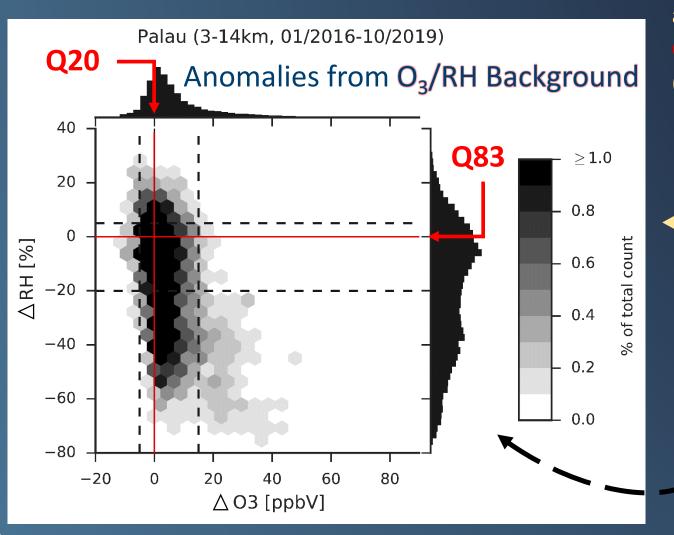
No known mechanism for in situ production of high O<sub>3</sub> or dehydration in the mid-troposphere origin either transport from the (extratropical) stratosphere or non-local ground pollution, lifted convectively in the area of origin then undergoing dehydration during transport, e.g. via large-scale descent and radiative cooling.

(compare Dessler and Minschwaner, 2007; Andersen et al., 2016)

Layered structures and respective background are hidden in the belly of the "S" of mean profiles!



- First step: define **background profiles** for both tracers
- Second step: determine anomalies against this background

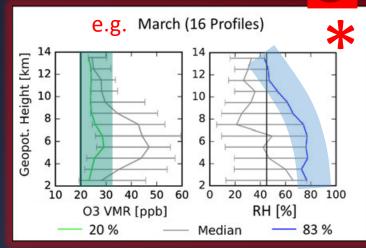


Background profiles: the monthly 20th (O<sub>3</sub>) and 83rd (RH) quantile,

Q20 and Q83

RH-

(altitude dependent)



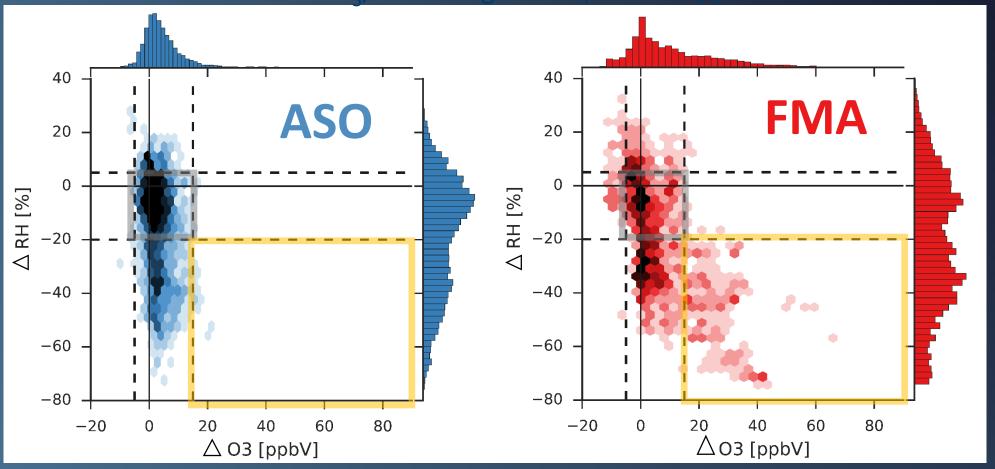
Third step: **bimodality in RH anomalies**motivates classification in O<sub>3</sub>RH groups

→ air mass definition

Classification in a 3x3 grid (dashed lines), with respect to the distributions, focus in the following on central background group (O<sub>3</sub>O RHO) and dry O<sub>3</sub>-rich anomaly (O<sub>3</sub>+RH-)



#### Anomalies from O<sub>3</sub>/RH Background (3-14 km) per season



#### O<sub>3</sub>oRHo:

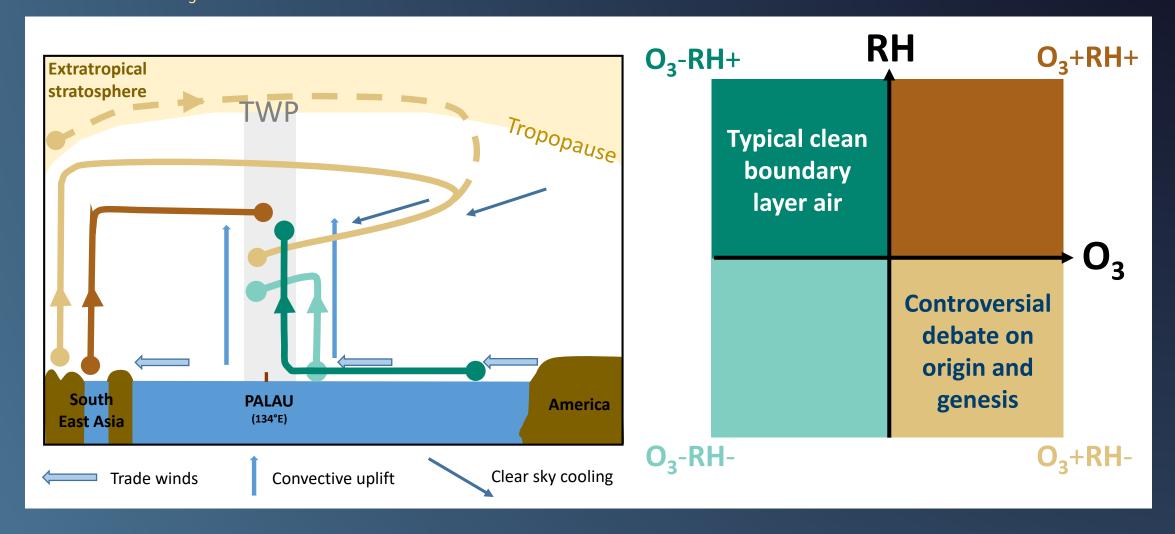
humid, O<sub>3</sub>-poor background, present year-round (!), but dominates **ASO** 

#### O<sub>3</sub>+RH-: dry, O<sub>3</sub>-rich, most frequent in **FMA**





With our process understanding, we identified major transport pathways related to the O<sub>3</sub>RH relation observed in Palau:







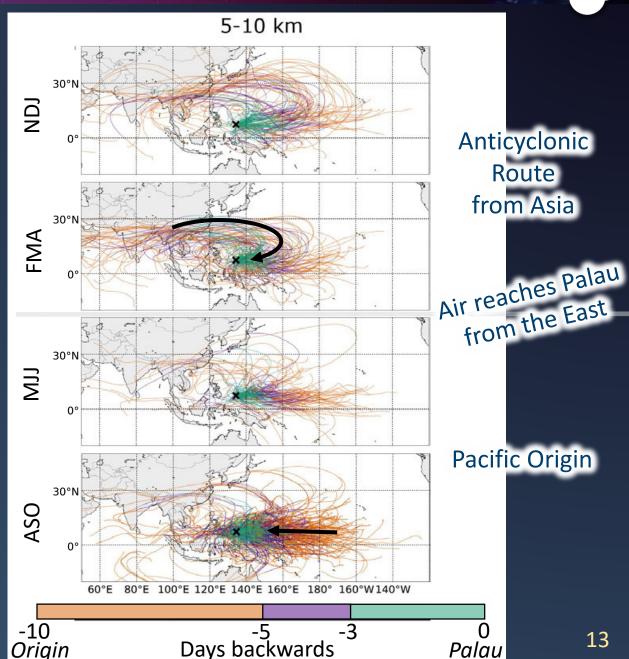
#### Transport module of Langrangian Chemistry and Transport Model ATLAS (Wohltmann et al. 2010)

#### Setup:

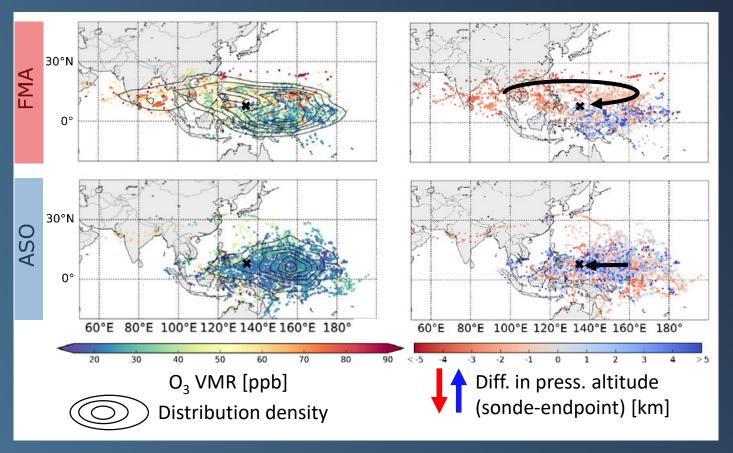
- driven by ERA5 reanalysis data, no diffusion, no convective model parameterization, 10-min time steps
- initialized from ozone sounding data, 01/2016-10/2019,
   2-14 km, every 10th measurement
  - → focus on 5-10 km altitude range

#### **Assumptions:**

- 10-day-backtrajectories for dynamical footprint
- Due to typical lifetime of marine boundary layer O<sub>3</sub>:
   5-day-backtrajectory ending points
   = origin of air mass composition



# 5-days-back trajectory ending points ≡ origin, trajectory start @ 5-10 km in Palau x



#### All Observations per season:

#### O<sub>3</sub> VMR distributions:

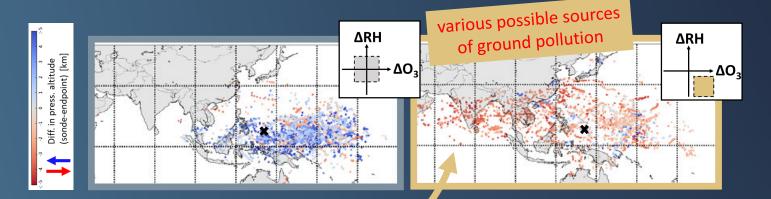
- Center of **low O<sub>3</sub>** in both seasons, FMA and ASO, East of Palau
- Secondary center of enhanced O<sub>3</sub> in FMA,
   North of Palau from India to East China

#### **Vertical displacement:**

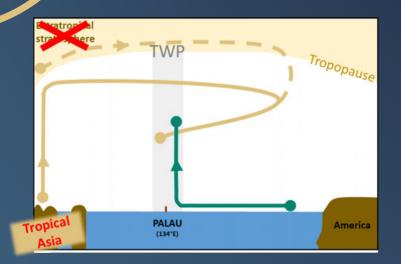
- Mainly in FMA, North of Palau air masses
   descend towards Palau (anti-cyclonic route),
   consistent with large-scale descent within the
   Hadley circulation and subsequent
   dehydration
- Ascent dominates ASO air masses
   (Pacific origin), corresponding well with the
   dominance of convective uplift



# 5-days-back trajectory ending points ≡ origin trajectory start @ 5-10 km in Palau x



Origin of dry O<sub>3</sub>-rich air masses in areas of increased air pollution on the ground from industry or bio mass burning, speaking in favor for a pollution based origin



### All Observations per O<sub>3</sub>/RH group:

Selection of trajectories for air masses identified as humid, O<sub>3</sub>-poor background (O<sub>3</sub>O RHO) or dry O<sub>3</sub>-rich (O<sub>3</sub>+RH-) anomaly from the background separates air masses according to the processes controlling RH (Convective uplift, ASO; dehydration during descent, FMA) and locates spatially separate source regions

No indication for significant contribution of stratospheric air:

Potential Vorticitiy analysis for all trajectories (from 4 years, 138 profiles, 5-10 km) revealed essentially **no air mass crossing the 1.5 PVU threshold** for more than a day during 10 days backwards.

-

- $\checkmark$  Palau's four-year tropospheric  $O_3$  time series fills the observational gap in this key region of stratospheric entry.
- ✓ Using the ECC  $O_3$  sounding data set (01/2016-10/1019), seasonal analysis, trajectory modelling and a statistical approach to distinguish air masses by  $O_3$ /RH relation, we **identified transport processes and pathways to the TWP:**

	Humid, O <sub>3</sub> -poor	
Processes	Convective background	Large
Origin	Pacific or local	Tropica
Frequency	Year-round, dominates Aug-Oct	Most f

Dry, O<sub>3</sub>-rich

Large scale descent, pollution

Tropical Asia (anticyclonic route)

Most frequent in Feb-Apr

✓ Watch out for the upcoming publications!

#### References

Katrin Müller, 2020: Characterization of Ozone and the Oxidizing Capacity of the Tropical West Pacific Troposphere, **PhD thesis**, https://doi.org/10.26092/elib/463.

#### WATCH OUT: two papers to be submitted this fall:

K. Müller, Ingo Wohltmann, Peter von der Gathen and Markus Rex (2021): Air Mass Transport to the Tropical West Pacific Troposphere inferred from Ozone and Relative Humidity Balloon Observations above Palau, in prep.

K. Müller, Jordis Tradowsky, Christoph Ritter, Justus Notholt, Ingo Beninga, Wilfried Ruhe, Winfried Markert, Juergen Graeser,

Sharon Patris and Markus Rex (2021): The Palau Atmospheric Observatory – continuous monitoring of tropospheric composition in the Tropical West Pacific, in prep.

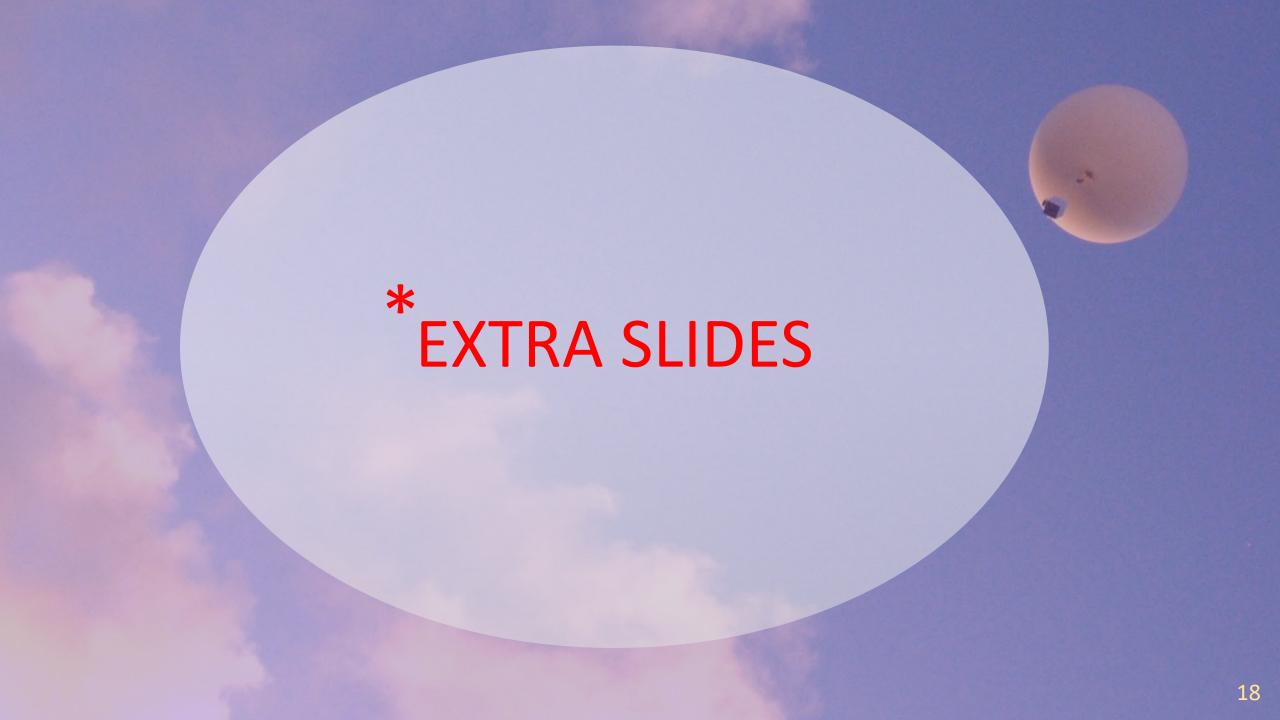


K. Müller. et al. (2020): AGU presentation, Origin of Tropospheric Air Masses in the Tropical West Pacific identified by Balloon-borne Ozone and Water Vapor Measurements from Palau, https://doi.org/10.1002/essoar.10505805.1

K. Müller. et al. (2021): EGU presentation, Origin of Tropospheric Air Masses in the Tropical West Pacific identified by Balloon-borne Ozone and Water Vapor Measurements from Palau

- D. C. Anderson et al. A pervasive role for biomass burning in tropical high ozone/low water structures. Nature Communications, 7(1):10267, April 2016. ISSN 041-1723. doi: 10.1038/ncomms10267.
- E. V. Browell et al. Large-scale air mass characteristics observed over the remote tropical Pacific Ocean during March-April 1999: Results from PEM-Tropics B field experiment. Journal of Geophysical Research: Atmospheres, 106(D23):32481–32501, December 2001. doi: 10.1029/2001JD900001.
- P. Cau, J. Methven, and B. Hoskins. Origins of Dry Air in the Tropics and Subtropics. Journal of Climate, 20(12):2745–2759, June 2007. doi: 10.1175/JCLI4176.1.
- A. E. Dessler and K. Minschwaner. An analysis of the regulation of tropical tropospheric water vapor. Journal of Geophysical Research: Atmospheres, 112(D10), May 2007. doi: 10.1029/2006JD007683.
- I. Folkins. Tropical ozone as an indicator of deep convection. Journal of Geophysical Research, 107(D13):4184, 2002. ISSN 0148-0227. doi: 10.1029/2001JD001178.
- I. Folkins, M. Loewenstein, J. Podolske, S. J. Oltmans, and M. Proffitt. A barrier to vertical mixing at 14 km in the tropics: Evidence from ozonesondes and aircraft measurements. Journal of Geophysical Research: Atmospheres, 104(D18): 22095–22102, 1999. doi: 10.1029/1999JD900404
- I. Folkins, P. Bernath, C. Boone, G. Lesins, N. Livesey, A. M. Thompson, K. Walker, and J. C. Witte. Seasonal cycles of O3, CO, and convective outflow at the tropical tropopause. Geophysical Research Letters, 33(16):L16802, 2006. doi: 10.1029/2006GL026602.
- H. Hayashi, K. Kita, and S. Taguchi. Ozone-enhanced layers in the troposphere over the equatorial Paci c Ocean and the influence of transport of midlatitude UT/LS air. Atmospheric Chemistry and Physics, page 14, 2008.
- D. Kley, P. J. Crutzen, H. G. J. Smit, H. Vomel, S. J. Oltmans, H. Grassl, and V. Ramanathan. Observations of Near-Zero Ozone Concentrations Over the Convective Pacific: Effects on Air Chemistry. Science, 274(5285):230–233, October 1996. doi: 10.1126/science.274.5285.230.
- L. L. Pan et al. Bimodal distribution of free tropospheric ozone over the tropical western Pacific revealed by airborne observations. Geophysical Research Letters, 42(18):7844–7851, September 2015. doi: 10.1002/2015GL065562.
- L. C. Paulik and T. Birner. Quantifying the deep convective temperature signal within the tropical tropopause layer (TTL). Atmospheric Chemistry and Physics, 12(24): 12183–12195, December 2012. doi: 10.5194/acp-12-12183-2012.
- W. J. Randel, M. Park, F. Wu, and N. Livesey. A Large Annual Cycle in Ozone above the Tropical Tropopause Linked to the Brewe-Dobson Circulation. Journal of the Atmospheric Sciences, 64(12):4479{4488, December 2007. doi: 10.1175/2007JAS2409.1.
- M. Rex, I. Wohltmann, T. Ridder, R. Lehmann, K. Rosenlof, P. Wennberg, D. Weisenstein, J. Notholt, K. Kruger, V. Mohr, and S. Tegtmeier. A tropical West Pacific OH minimum and implications for stratospheric composition. Atmospheric Chemistry and Physics, 14(9):4827{4841, May 2014 doi: 10.5194/acp-14-4827-2014.
- J. K. P. Shonk, E. Guilyardi, T. Toniazzo, S. J. Woolnough, and T. Stockdale. Identifying causes of Western Pacific ITCZ drift in ECMWF System 4 hindcasts. Climate Dynamics, 50(3-4):939-954, February 2018. doi: 10.1007/s00382-017-3650-9.
- S. Solomon, D. W. J. Thompson, R. W. Portmann, S. J. Oltmans, and A. M. Thompson. On the distribution and variability of ozone in the tropical upper troposphere: Implications for tropical deep convection and chemical-dynamical coupling. Geophysical Research Letters, 32(23):L23813, 2005. doi: 10.1029/2005GL024323.
- M. Tao, L. L. Pan, P. Konopka, S. B. Honomichl, D. E. Kinnison, and E. C. Apel. A Lagrangian Model Diagnosis of Stratospheric Contributions to Tropical Midtropospheric Air. Journal of Geophysical Research: Atmospheres, 123(17):9764–9785, September 2018. doi: 10.1029/2018JD028696

  A. M. Thompson et al. Ozonesonde Quality Assurance: The JOSIE–SHADOZ (2017) Experience. Bulletin of the American Meteorological Society, 100(1):155–171, January 2019. doi: 10.1175/BAMS-D-17-0311.1.
- V. Thouret, J. Y. N. Cho, R. E. Newell, A. Marenco, and H. G. J. Smit. General characteristics of tropospheric trace constituent layers observed in the MOZAIC program. Journal of Geophysical Research: Atmospheres, 105(D13):17379–17392, 2000. doi: 10.1029/2000JD900238.
- H. Voemel and K. Diaz. Ozone sonde cell current measurements and implications for observations of near-zero ozone concentrations in the tropical upper troposphere. Atmospheric Measurement Techniques, 3(2):495–505, April 2010. doi: 10.5194/amt-3-495-2010.
- I. Wohltmann, R. Lehmann, and M. Rex. The Lagrangian chemistry and transport model ATLAS: simulation and validation of stratospheric chemistry and ozone loss in the winter 1999/2000. Geoscienti c Model Development, 3(2):585(601, November 2010. doi: 10.5194/gmd-3-585-2010



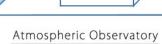
# Palau Atmospheric Observatory

#### **MaxDOAS:**

Pandora2S –
Pandonia Network
O<sub>3</sub>, NO<sub>2</sub>, AOD
(, H<sub>2</sub>O, SO<sub>2</sub>, ...)

#### FTIR Spectrometer:

Total abundances of ~ 20 chemical species





#### Lidar:

Vertical profiles of aerosol properties
Since 2018: multi-λ cloud and aerosol lidar ComCAL in new lab

#### **Research balloons:**

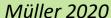
Vertical profiles of

- Ozone
- Aerosol
- Water vapour

#### MICA:

Ground sampling of OCS, CO, CO2, H2O Temp. @ CRRF site



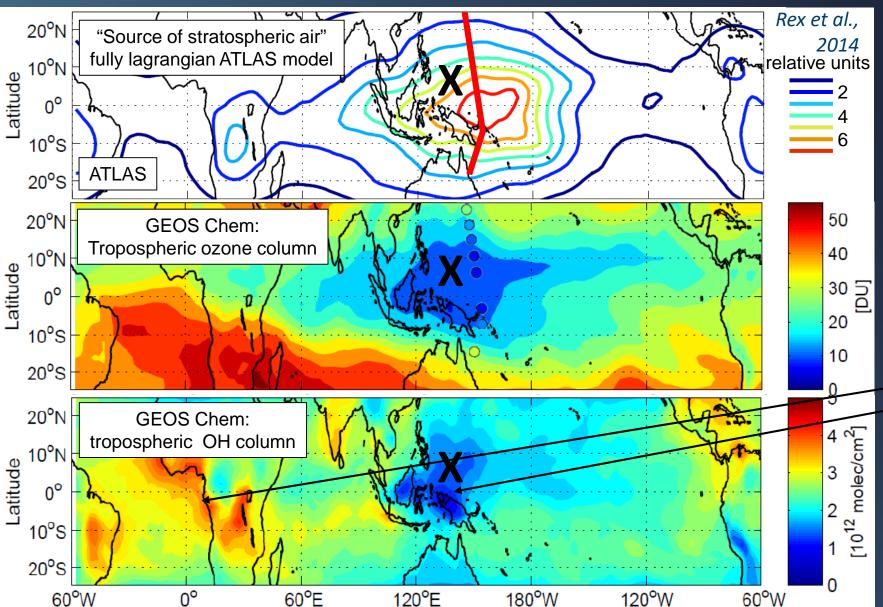


## (I) Motivation - TransBrom

# QUADRENNIAL OZONE SYMPOSIUM

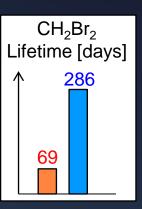






Density distribution
function of the horizontal
positions of the trajectories
between boundary layer and
Lagrangian Cold Points;
red thick line: TransBrom
Cruise 2009; filled circles: from
ozonesonde measurements



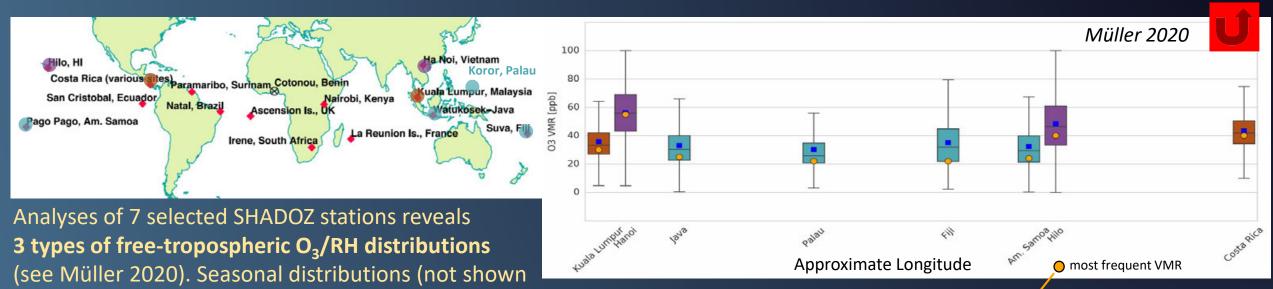


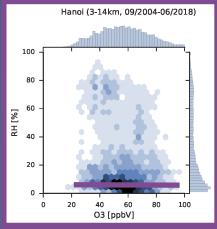
Lifetime comparison for tropical Atlantic and West Pacific (values for midtroposphere- 500 hPa at the equator for typical conditions)

## (I) O<sub>3</sub>/RH Comparison with SHADOZ



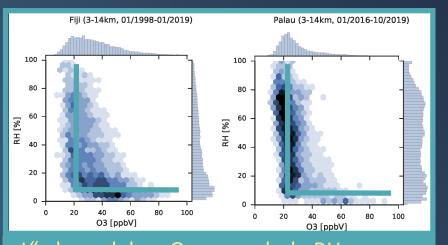




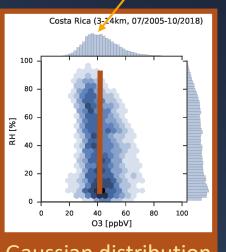


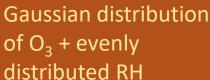
here) highlight uniqueness of Palau

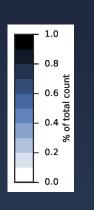
Predominantly dry air over a wider range of O<sub>3</sub> VMR



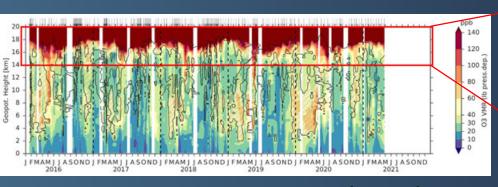
"L"-shaped: low O<sub>3</sub> over whole RH range + tail towards higher O<sub>3</sub> corresponding to low RH

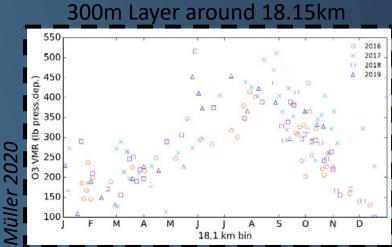




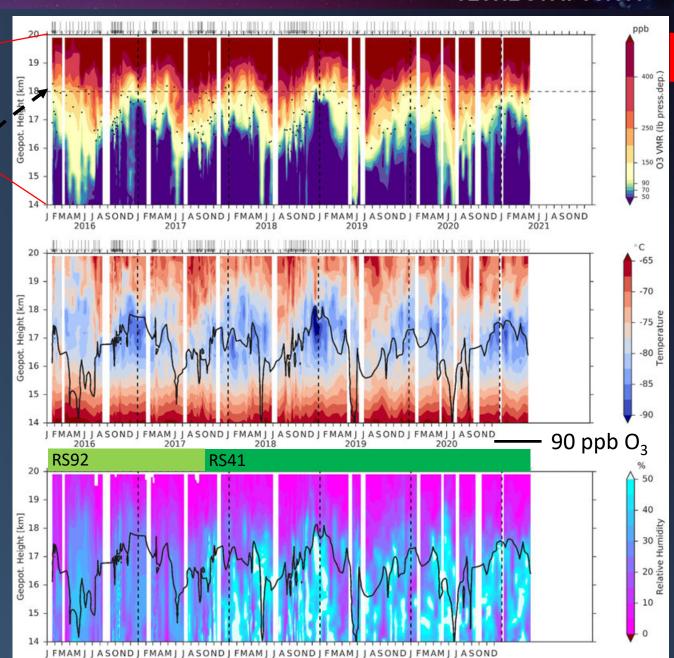


# QUADRENNIAL OZONE SYMPOSIUM





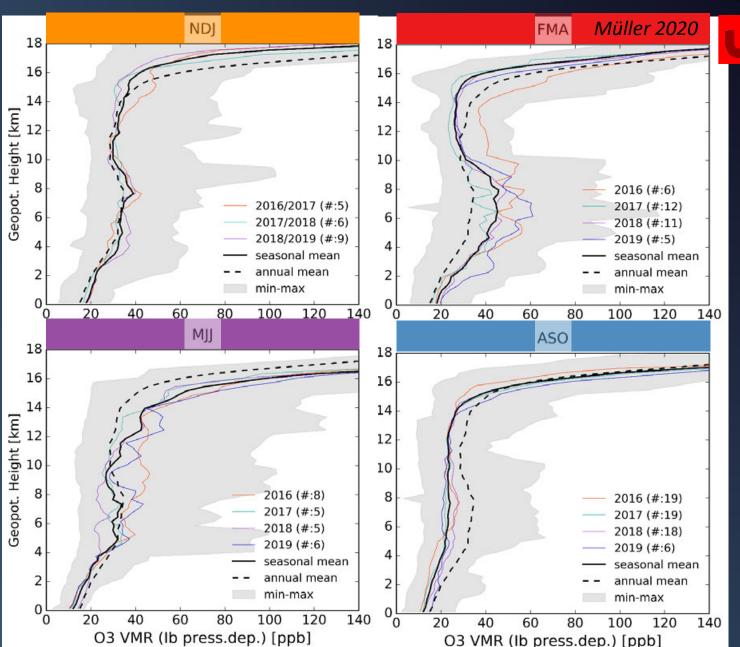
- Greatest amplitude of the TTL O<sub>3</sub> cycle occurs around 18 km
- Feedback during El Nino 2016 in the UT: suppressed convection with less uplift of ozone-poor air from the ground or wash-out of O<sub>3</sub> precursors



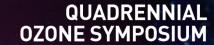




- November-January: NDJ,
   February-April: FMA, ..., chosen
   due to similar profile shapes
- Low O<sub>3</sub> (<25ppb) and enhanced mid-trop. O<sub>3</sub> (>50 ppb) observed in all seasons
- greatest anomalies from the annual mean: FMA & ASO
- Air masses deviating from a low O<sub>3</sub> background signal occur as filaments or layers, predominantly in the 5-10 km layer, disguised in the averaged belly of the ,S'

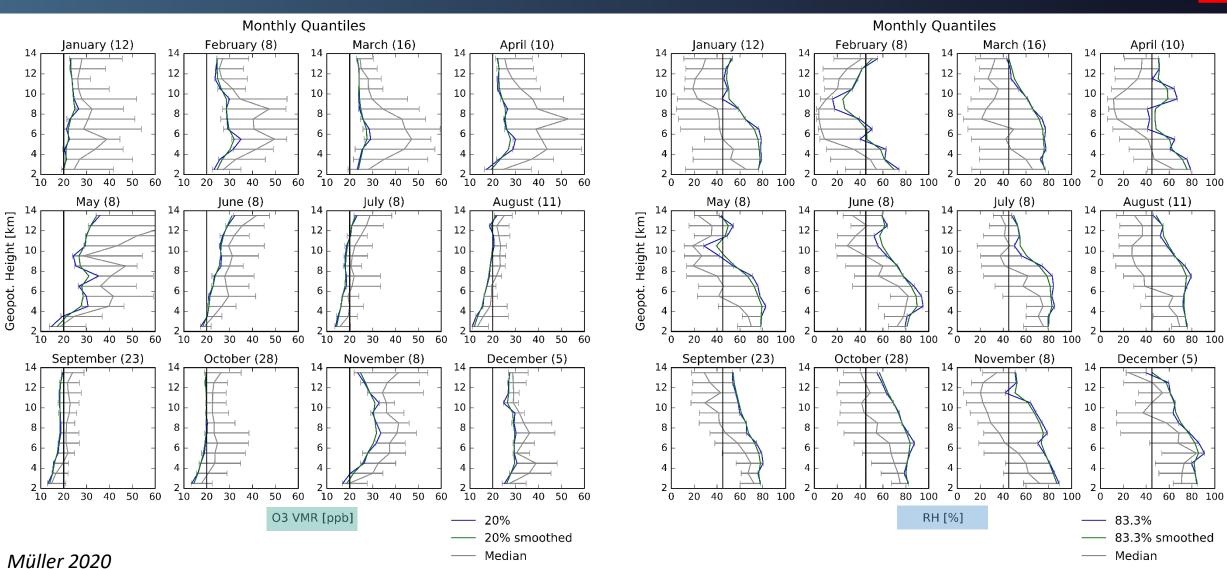


# (III) Background Definition

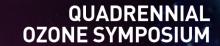








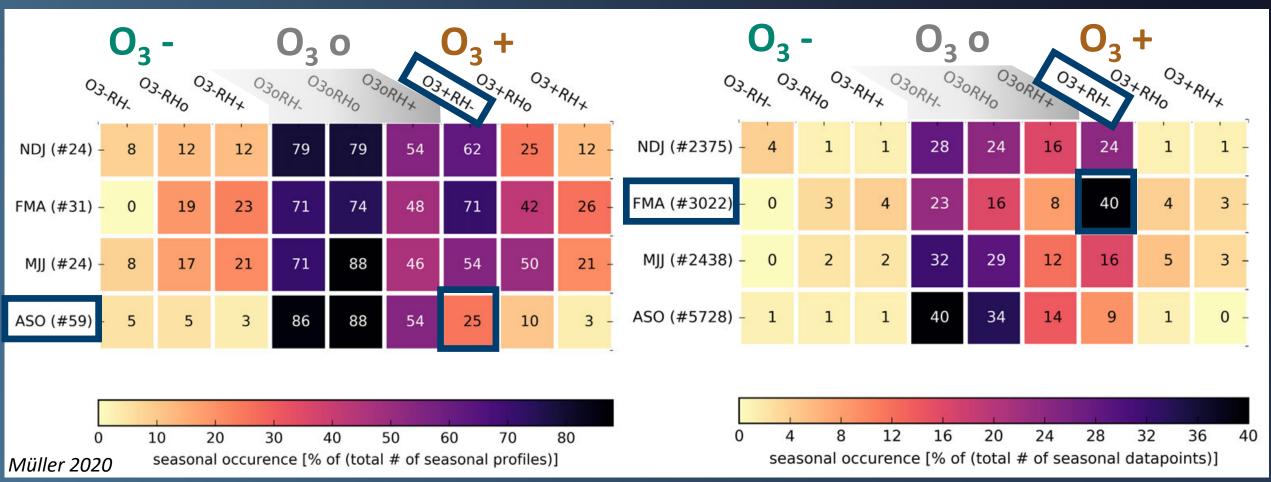
# (III) Seasonal Occurrence of O<sub>3</sub>/RH groups





Ú

Heatmaps for the seasonal occurrence of air masses for all nine anomaly groups, full time series, between **5-10 km** altitude



Example: O3+RH- air masses occur in 25% of all ASO profiles.

Example: O3+RH- air masses make up for 40% of all datapoints observed in FMA.