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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

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PhD thesis + 2 Manuscripts *

StratoClim

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ALFRED-WEGENER-INSTITUT
HELMHOLTZ-ZENTRUM FÜR POLAR- UND MEERESFORSCHUNG

NAVIGATION

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(I) Motivation

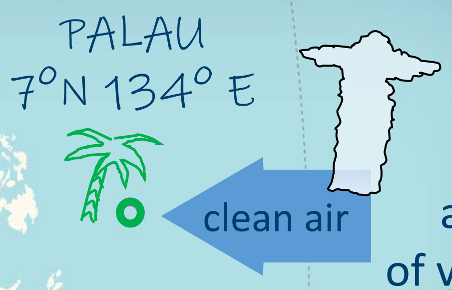
Why the Tropical West Pacific (TWP)?

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)*

Persistent tropospheric **Ozone minimum**

Corresponding **OH minimum** and prolonged life times of various chemical species



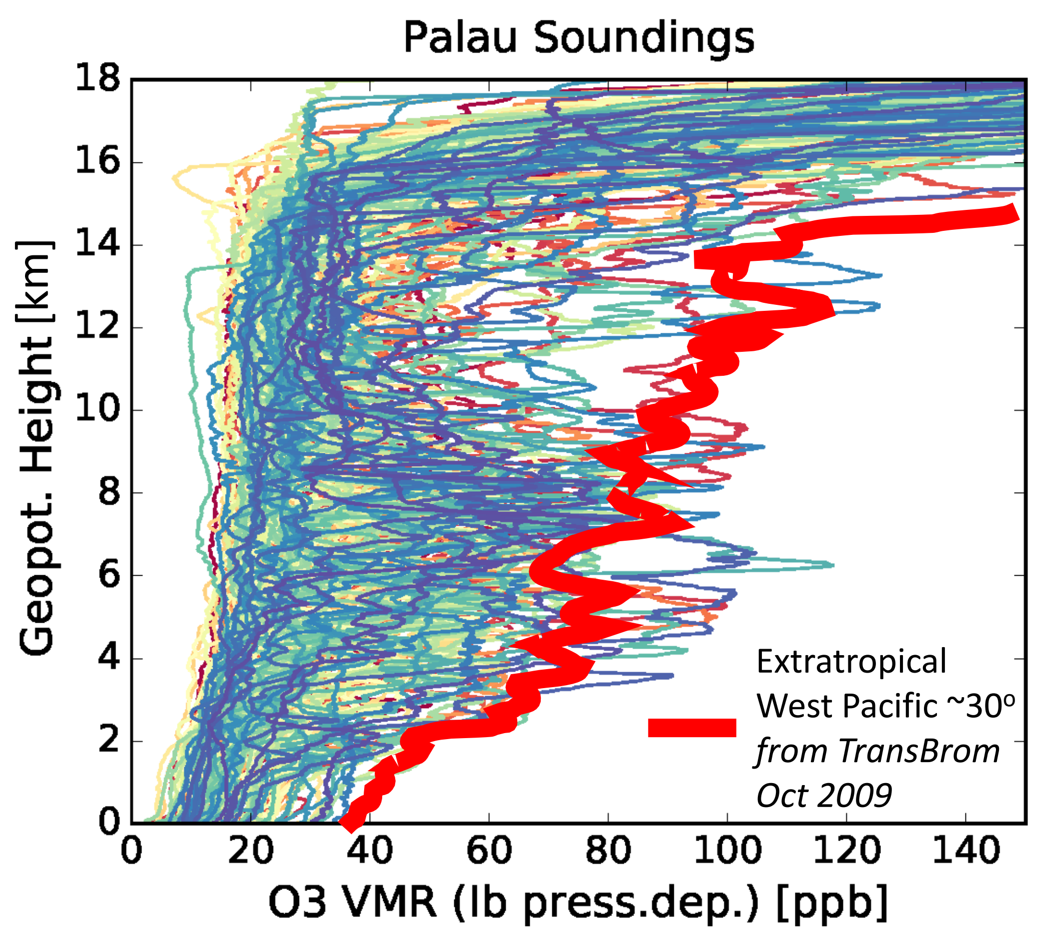
Important region for supply of chemical species to the stratosphere *

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

Key feature of the clean TWP troposphere: close coupling of the O_3 concentration and oxidizing capacity (OH), influencing overall transport of chemical species to the stratosphere.

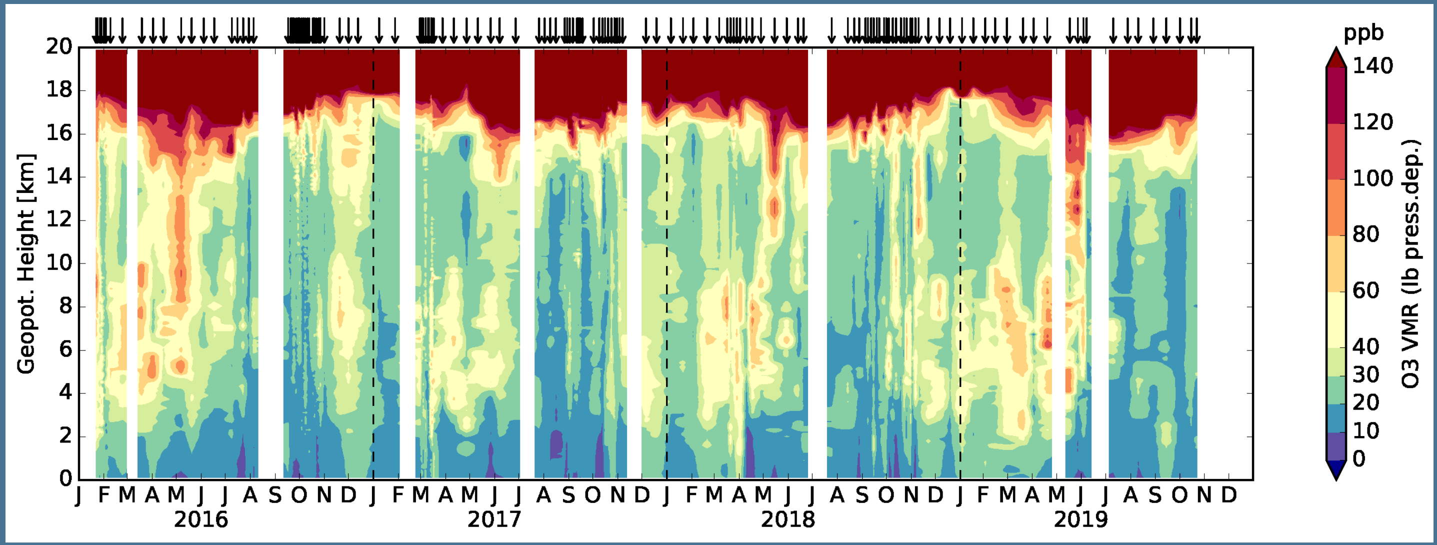
To improve the limited availability of tropospheric O_3 observations from this key region, the **Palau Atmospheric Observatory** * was established in 2016 as part of the EU-project StratoClim.

(I) O_3 Dataset



First characterization of tropospheric O_3 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 O3 observations in the TWP *(e.g. Voemel and Diaz 2010, Rex et al. 2014, Thompson et al. 2019)*



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

Time-height-cross-section

(I) Why O_3? As a chemical tracer…

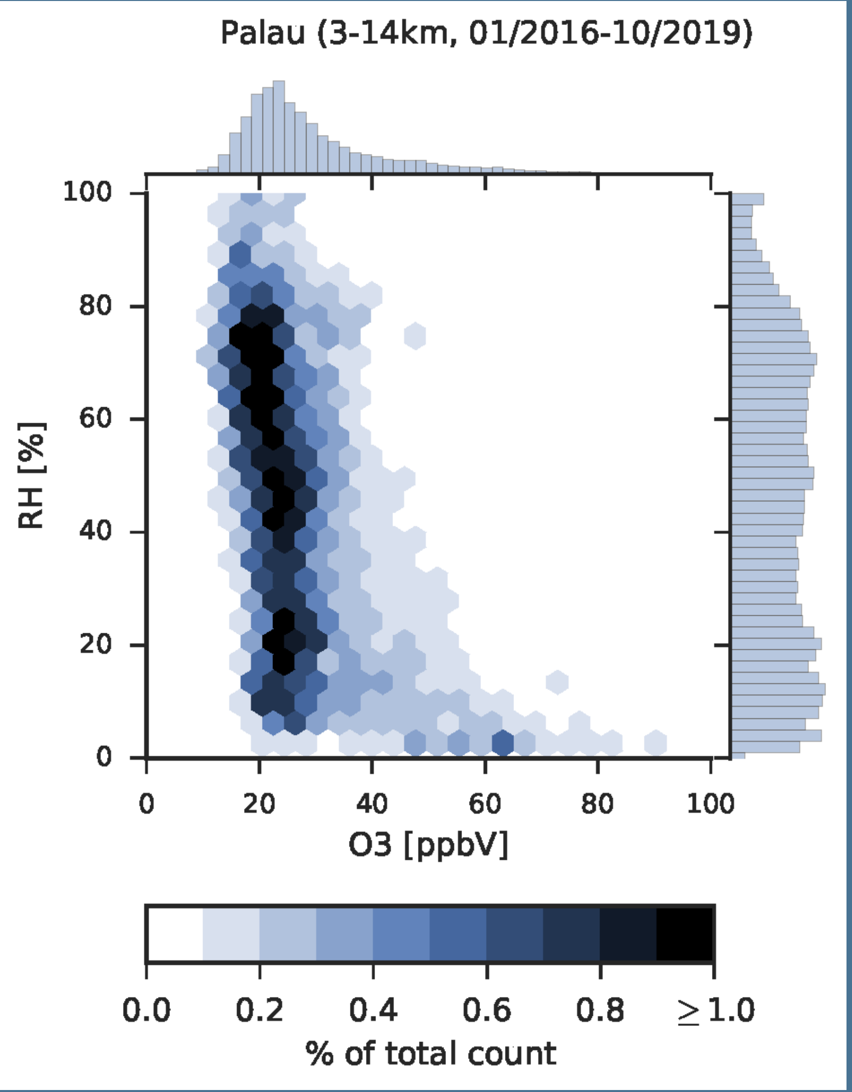
…for **local convective activity** in clean maritime air: **"low" O_3** (*e.g. Folkins, 2002; Folkins et al., 1999; Kley et al., 1996; Paulik and Birner, 2012; Solomon et al., 2005*) and

… for **long range transport** processes to the region, either related to air pollution or stratospheric intrusions: **"high" O_3** (*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*)

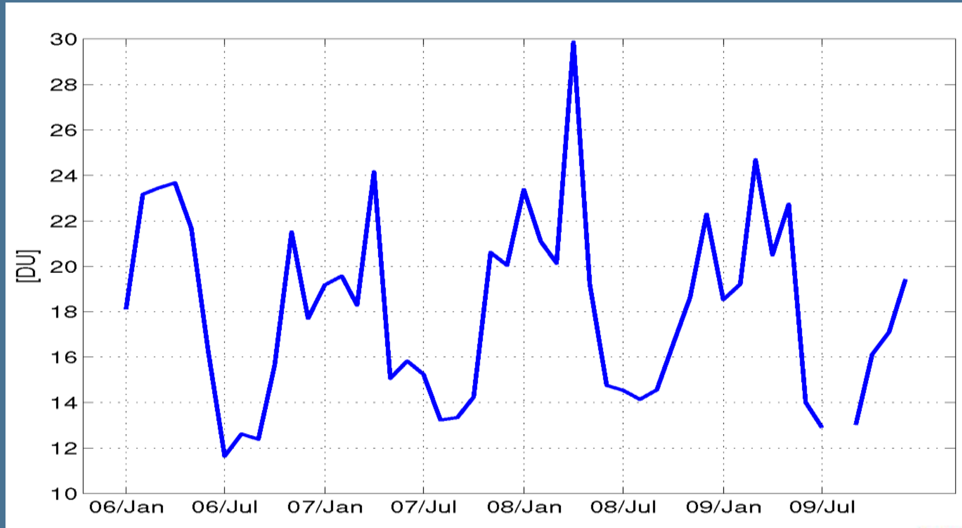
Central Question:
Can we identify air mass origin and its seasonality with the observed O_3/RH relation?

Free-tropospheric O_3/RH distribution of all observations



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

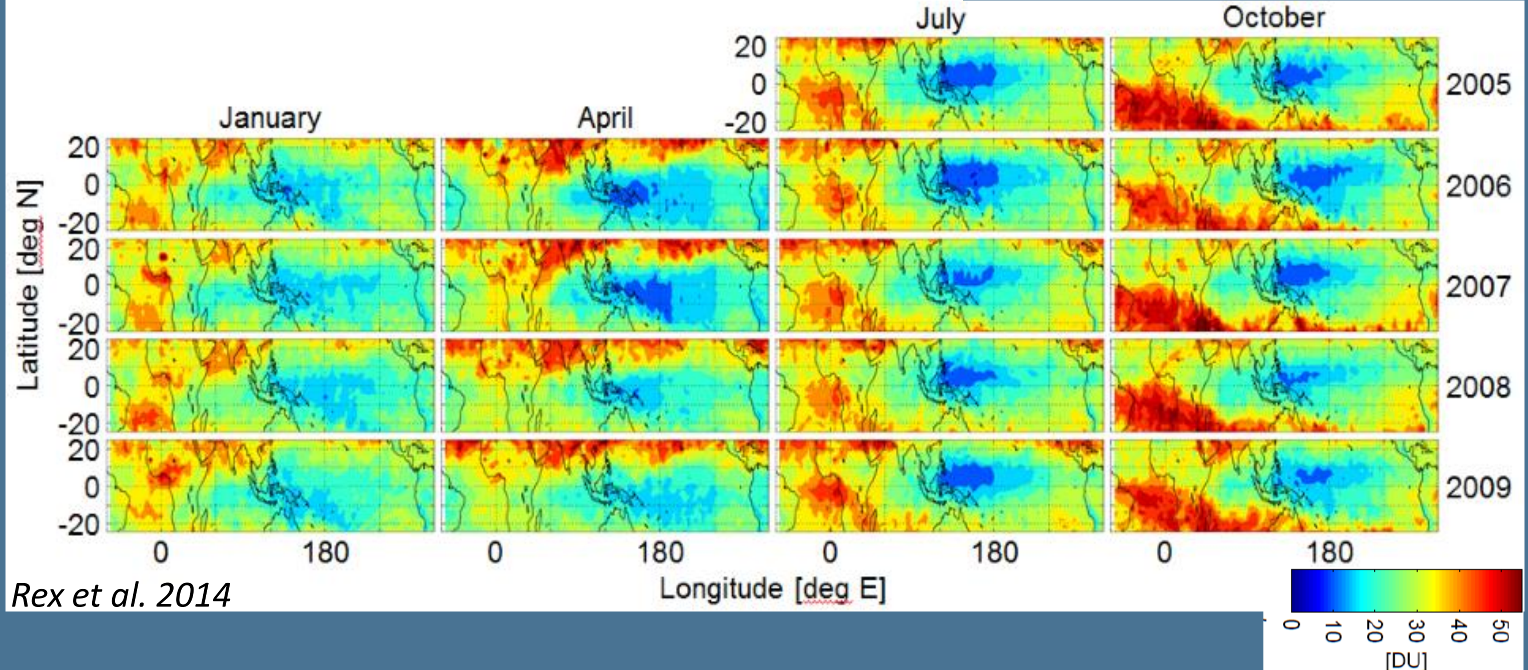
(II) Tropospheric O_3 variability



Tropospheric O_3 column from TES (on Aura Satellite)

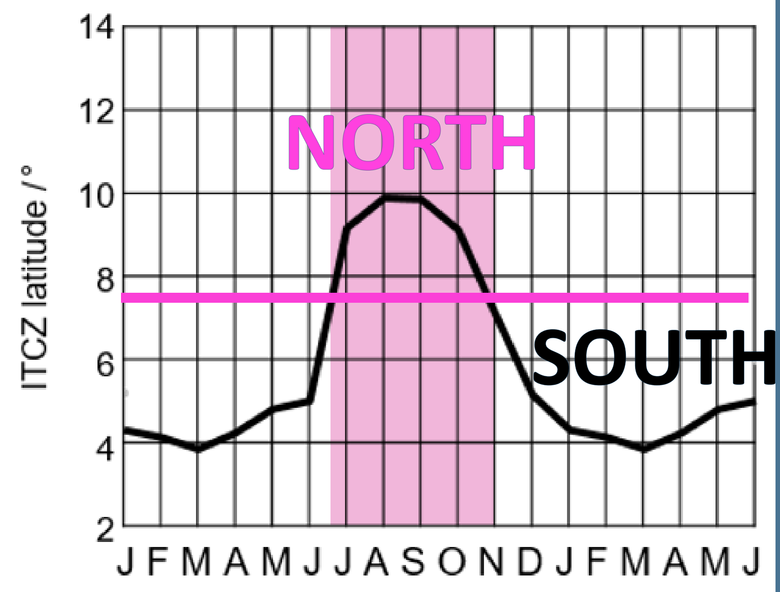
@ Palau

and in the tropics



Rex et al. 2014

- **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

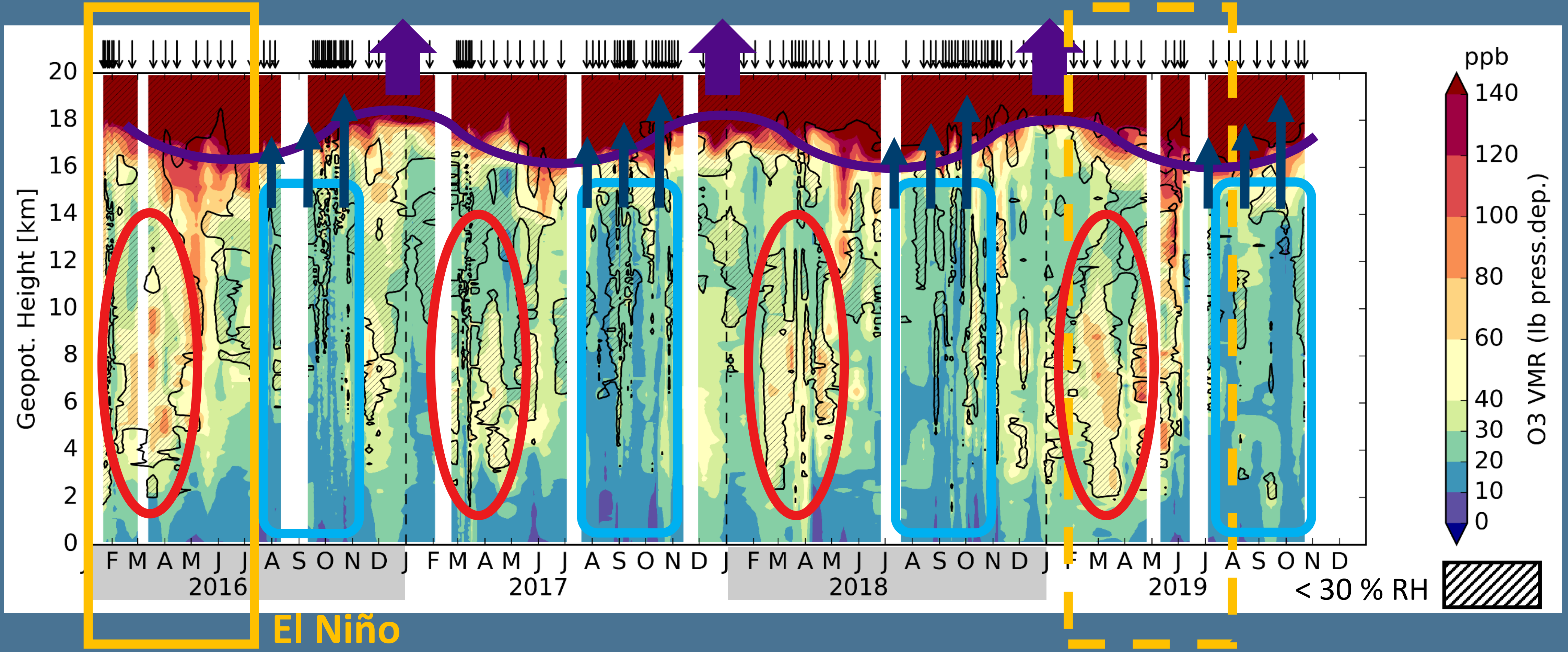


TRMM (1998-2009) average for 125-175° E, 0-20°N, adapted from *Shonk et al. 2018*

Movement of the ITCZ

(II) Tropospheric O_3 variability

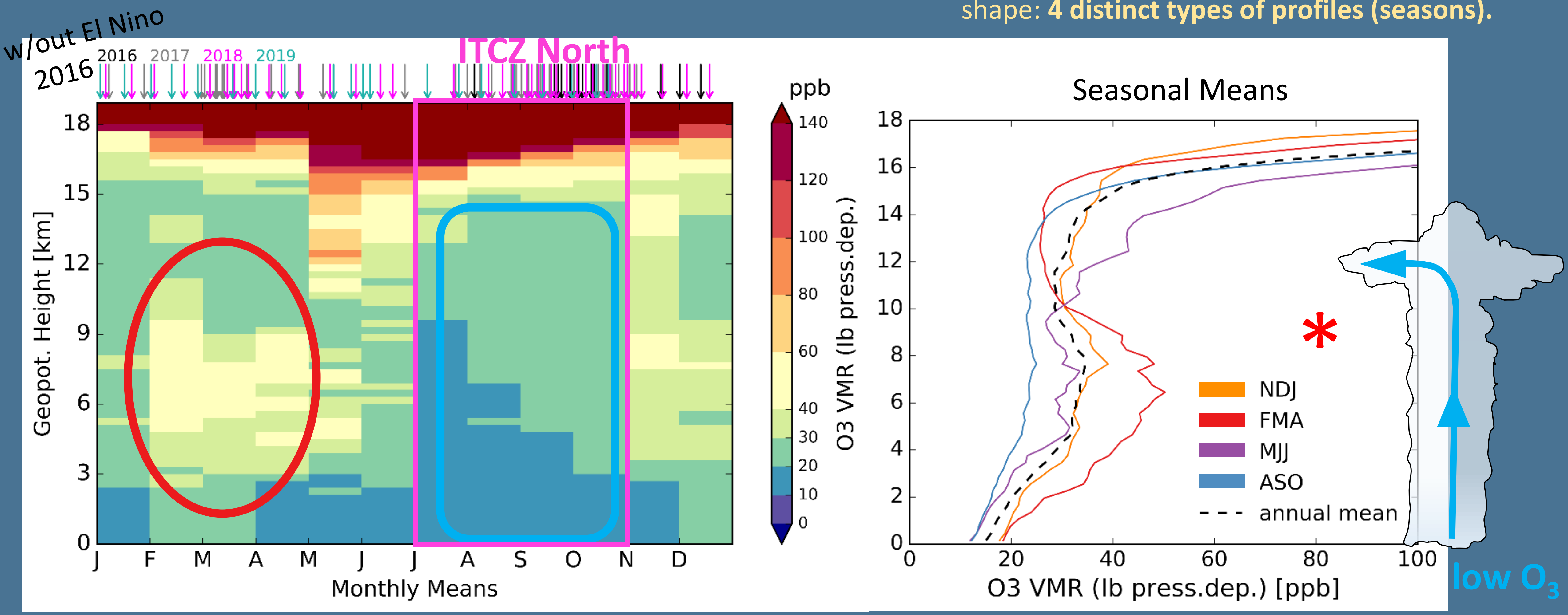
Annual TTL cycle: Brewer Dobson circulation
+ enhanced high altitude **convective outflow**
Onset of transport to the stratosphere and the **tropospheric O_3 minimum** occur simultaneously



Mid-troposphere: **O_3 minimum** from July-October, layers **of enhanced O_3** from February-April, often **anti-correlated with RH**

(II) Tropospheric O_3 variability

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

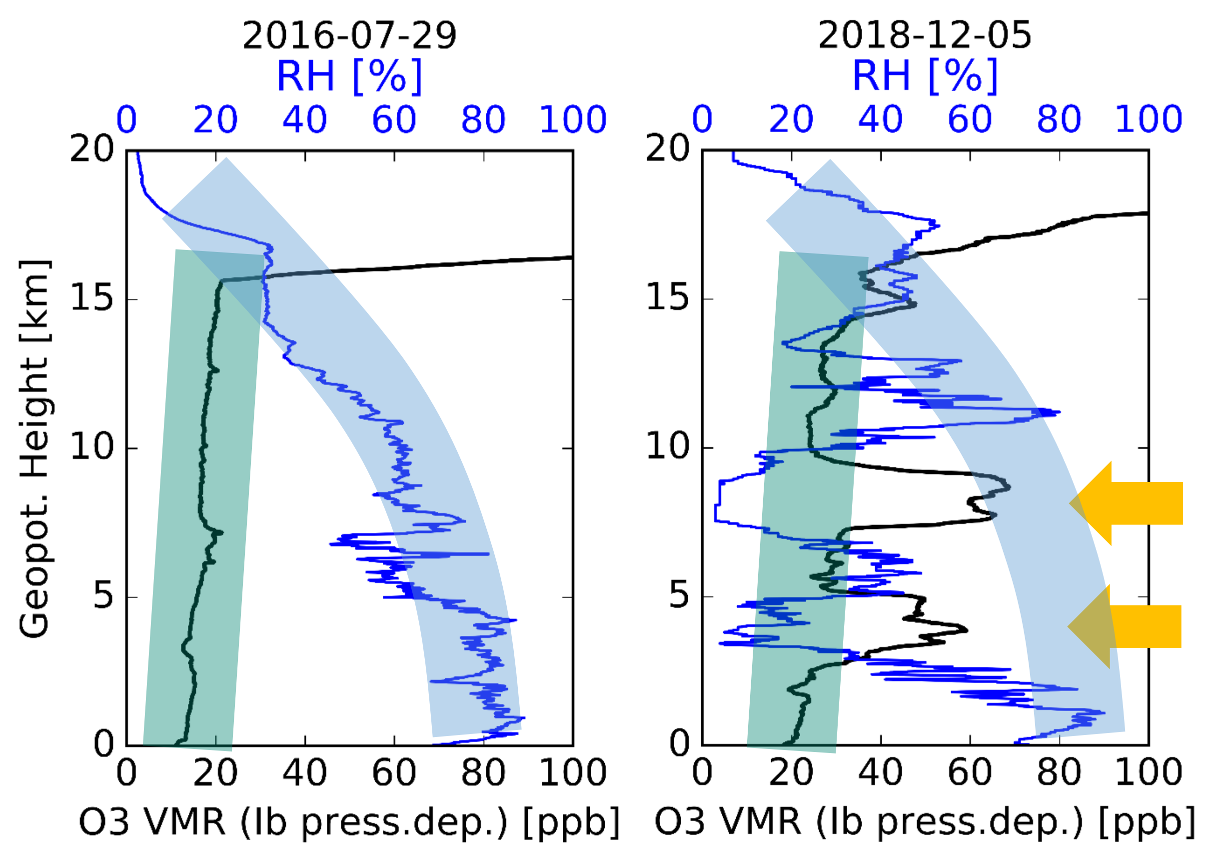


Monthly means highlight annual cycles, O_3 **minimum** corresponds with the **ITCZ located North** of Palau.

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.

(III) Air mass definition

Example tropospheric O_3 and RH profiles



convectively controlled, well-mixed **background:**

LOCAL mode

interruptive layers, controlled by transport:

NON LOCAL mode

Underlying processes:

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

LOCAL mode

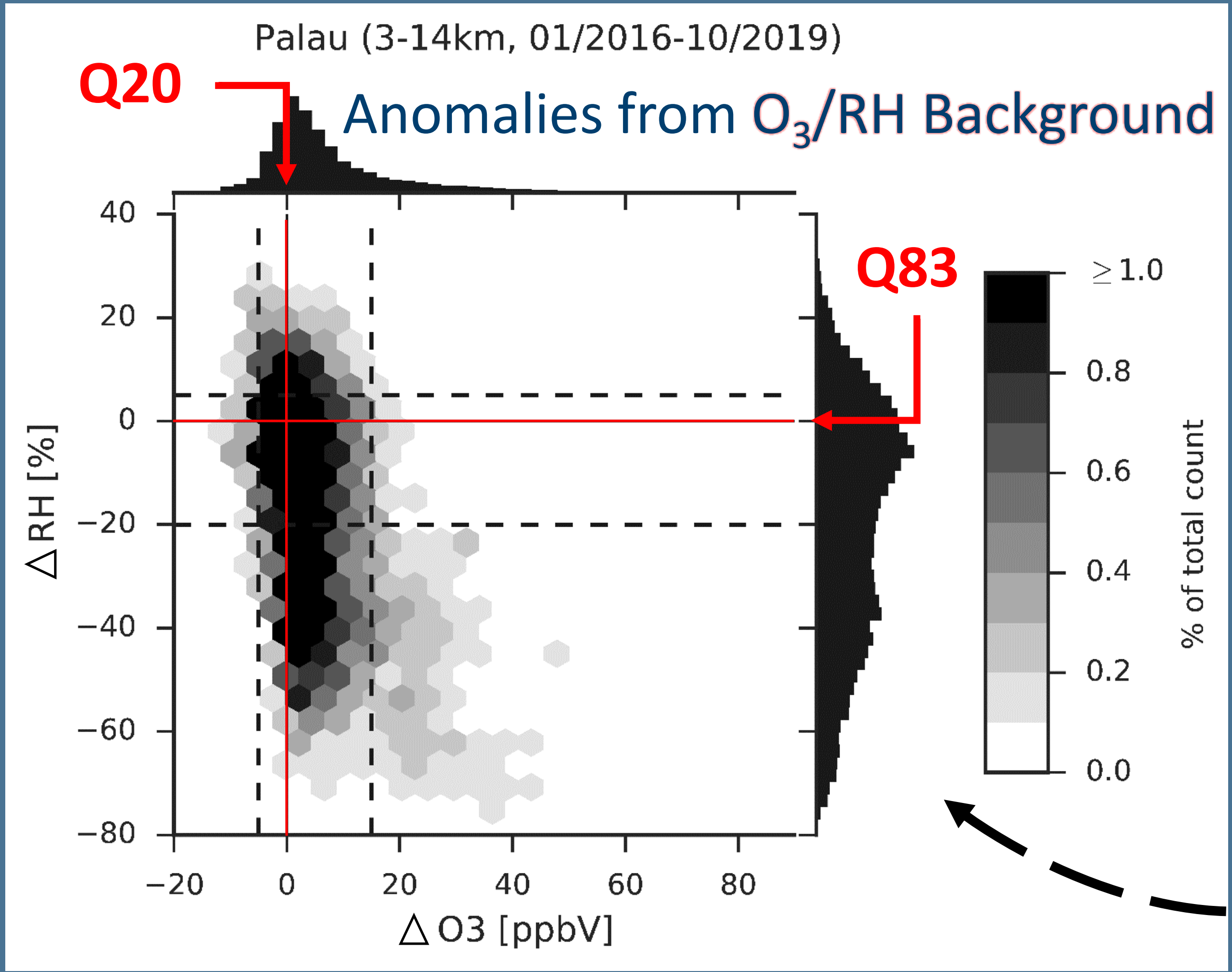
No known mechanism for in situ production of **high O_3** or **dehydration** in the mid-troposphere - origin either transport from the (extratropical) stratosphere or non-local ground pollution, lifted convectively in the area of origin then undergoing dehydration during transport, e.g. via large-scale descent and radiative cooling. (*compare Dessler and Minschwaner, 2007; Andersen et al., 2016*)

NON LOCAL mode

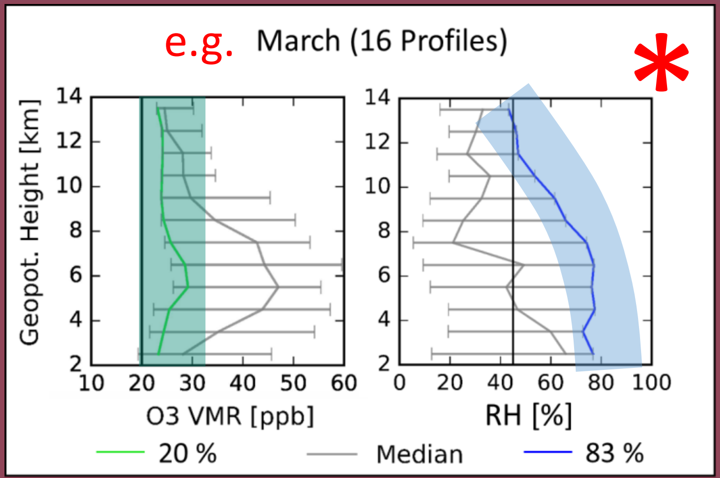
Layered structures and respective background are hidden in **the belly of the „S“** of mean profiles!

(III) Air mass definition

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

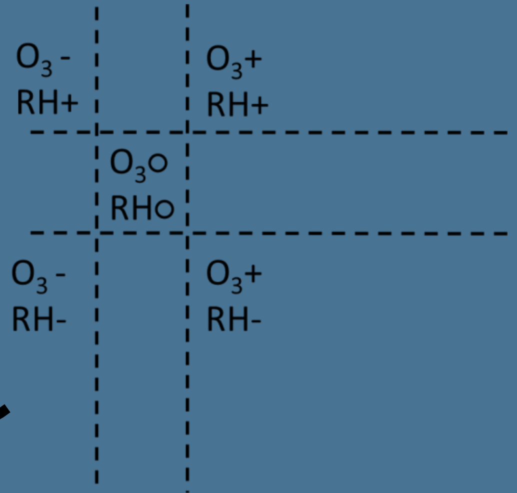


Background profiles:
the monthly 20th (O_3) and 83rd (RH) quantile,
Q20 and Q83
(altitude dependent)



- Third step: **bimodality in RH anomalies** motivates classification in O_3RH groups

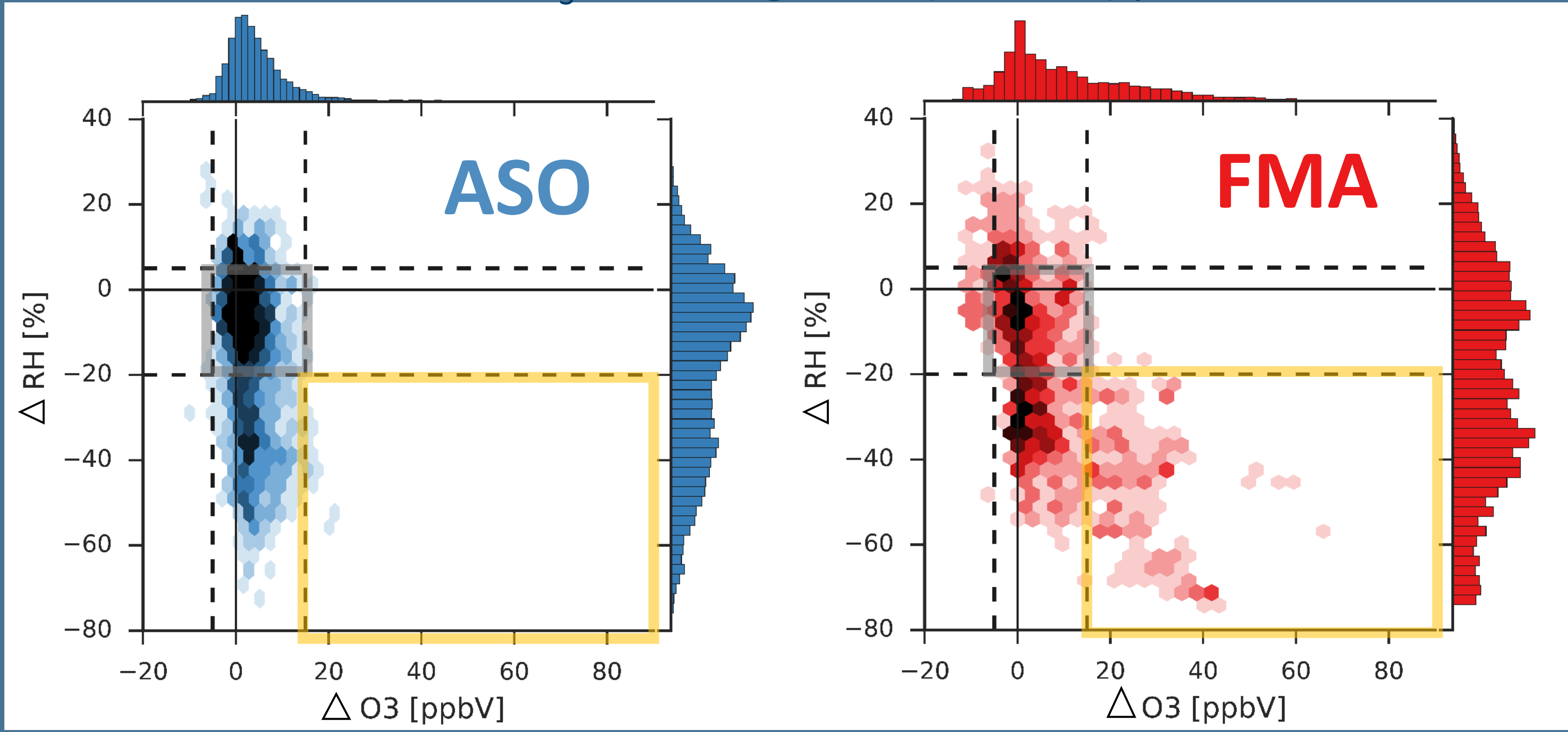
→ **air mass definition**



Classification in a 3x3 grid (dashed lines), with respect to the distributions,
focus in the following on **central background group** (O_3○ RH○) and **dry O_3-rich anomaly** (O_3+RH-)

(III) Seasonal Air Mass Occurence

Anomalies from O_3/RH Background (3-14 km) per season



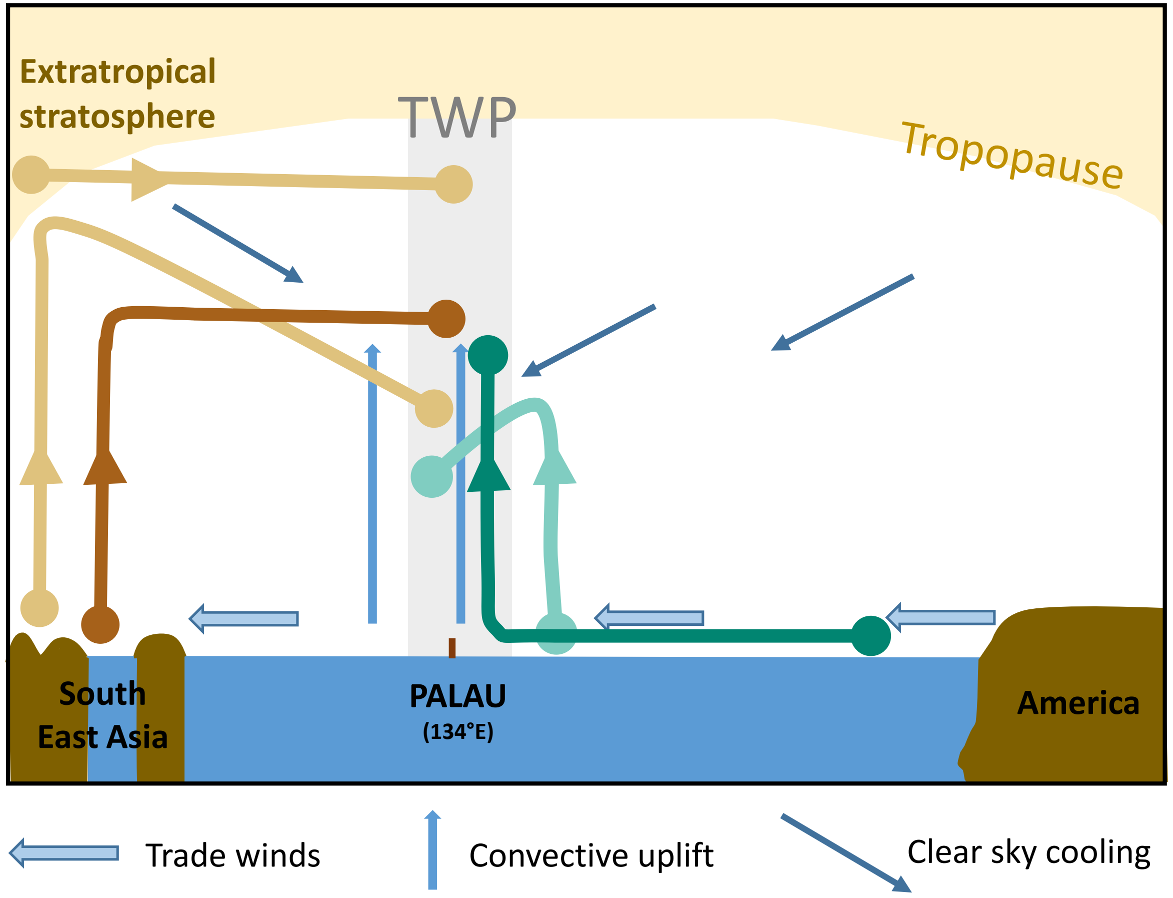
O_3oRHo:
humid, O_3-poor background, present year-round (!), but dominates **ASO**

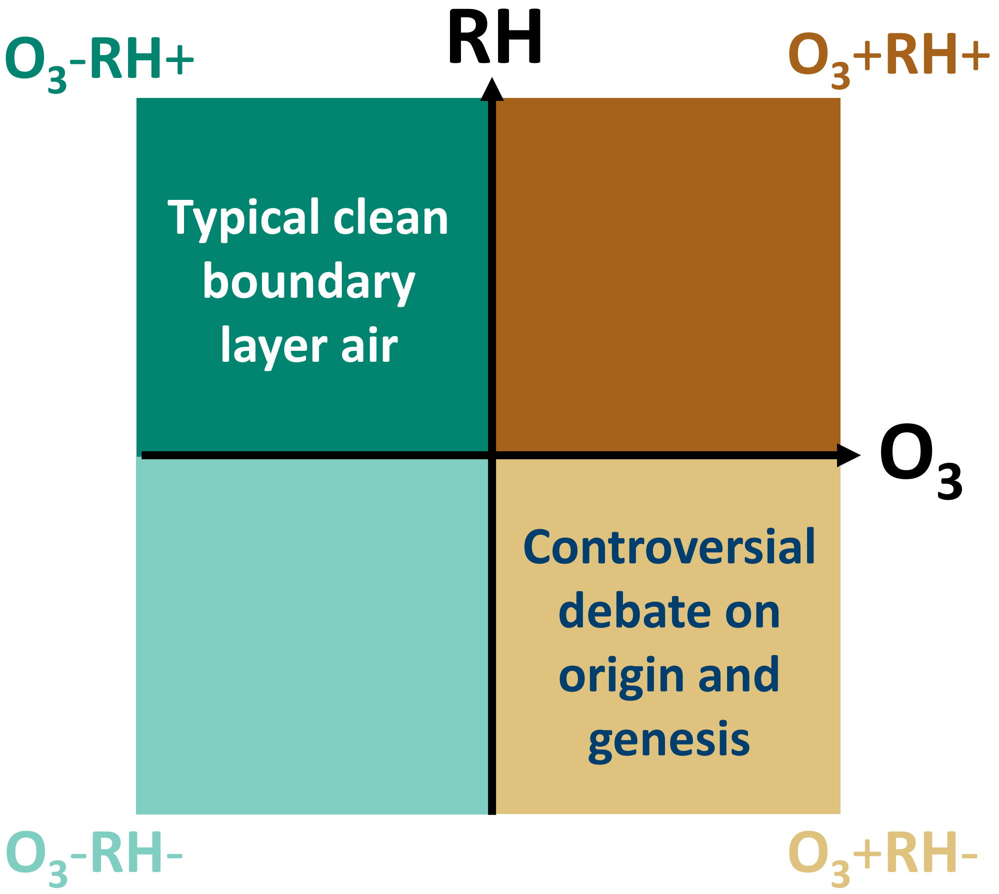
O_3+RH- :
dry, O_3-rich, most frequent in **FMA**

For more detailed statistics on all O3/RH anomaly groups, **click HERE** *

(IV) Transport Pathways and Processes

With our process understanding, we identified major transport pathways related to the O_3RH relation observed in Palau:





(IV) Backward Trajectories

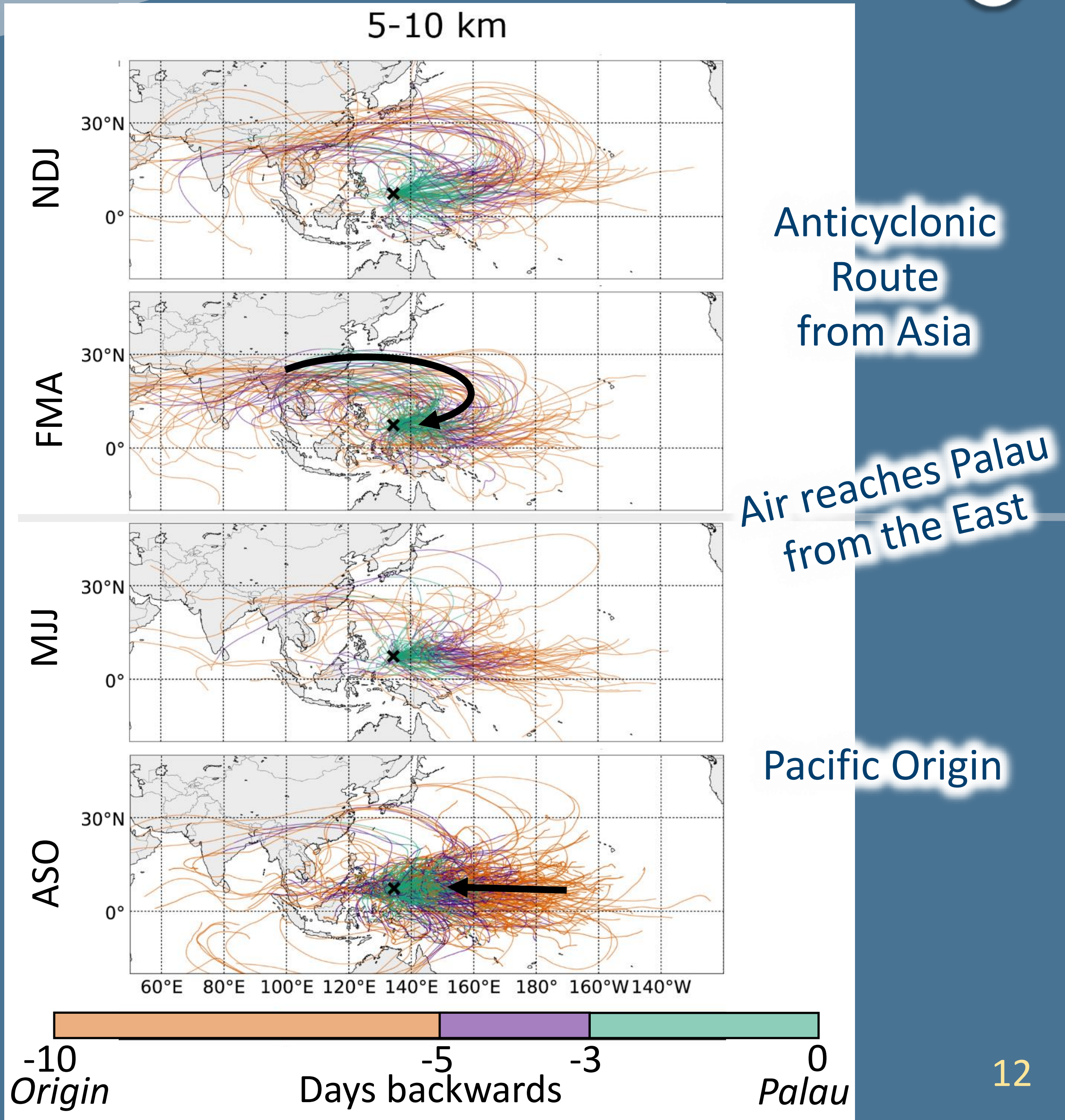
Transport module of Langrangian Chemistry and Transport Model ATLAS (Wohltmann et al. 2010)

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**

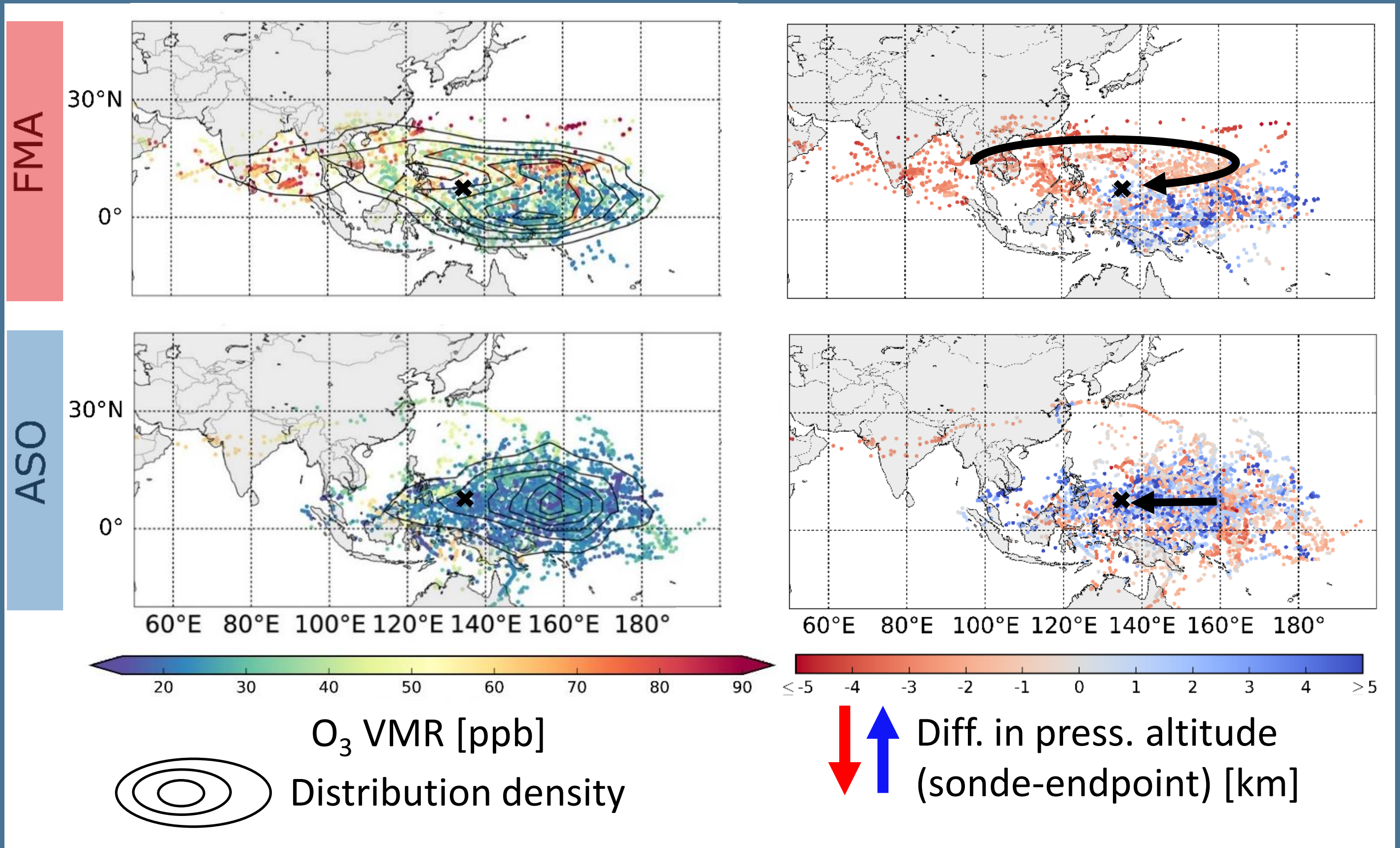
Assumptions:

- 10-day-backtrajectories for dynamical footprint
- Due to typical lifetime of marine boundary layer O_3: **5-day**-backtrajectory ending points = origin of air mass composition



(IV) Origin of Air Masses

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



All Observations per season:

O_3 VMR distributions:

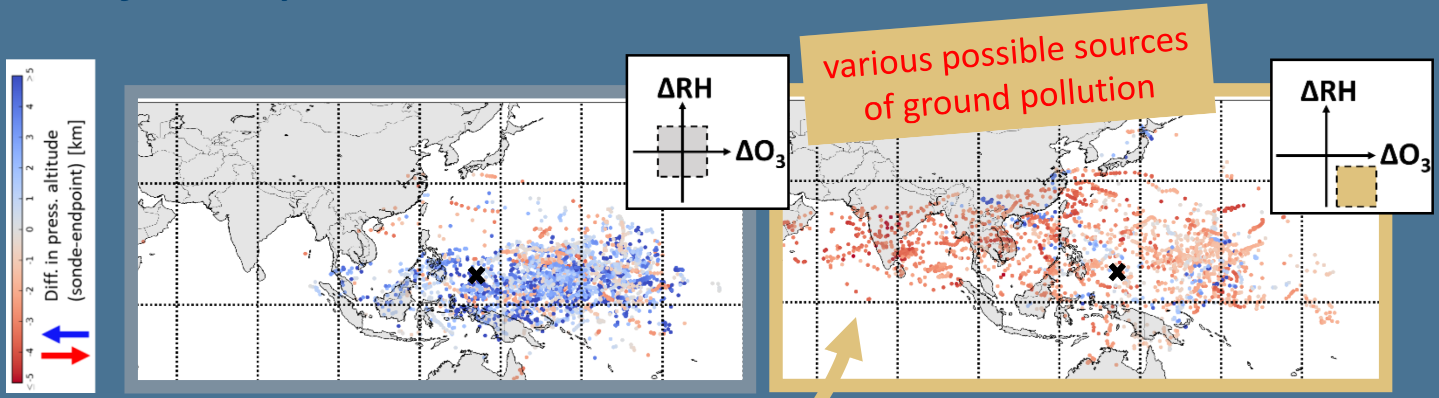
- Center of low O_3 in both seasons, FMA and ASO, East of Palau
- Secondary center of **enhanced O_3** in FMA, North of Palau from India to East China

Vertical displacement:

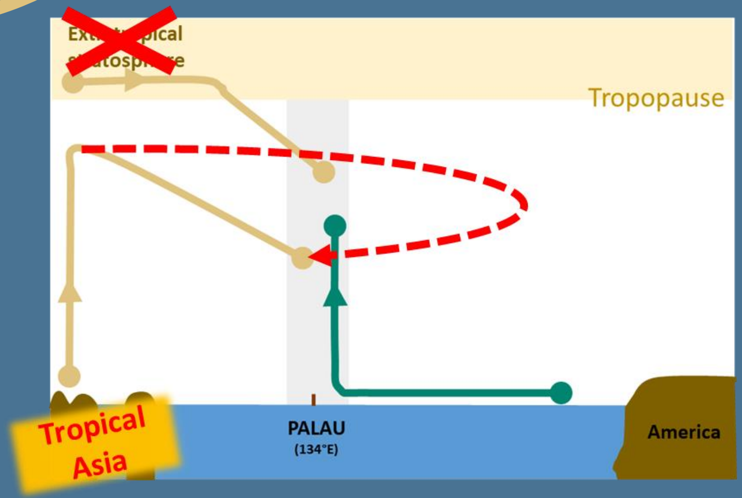
- Mainly in FMA, North of Palau air masses **descend** towards Palau (**anti-cyclonic route**), consistent with large-scale descent within the Hadley circulation and subsequent dehydration
- **Ascent** dominates ASO air masses (**Pacific origin**), corresponding well with the dominance of convective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

(IV) Origin of Air Masses

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



Origin of **dry O_3-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_3/RH group* :

Selection of trajectories for air masses identified as **humid, O_3-poor background (O_3○ RH○)** or **dry O_3-rich (O_3+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.

V Take home messages

- ✓ **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_3-poor	Dry, O_3-rich
Processes	Convective background	Large scale descent, pollution
Origin	Pacific or local	Tropical Asia (anticyclonic route)
Frequency	Year-round, dominates Aug-Oct	Most frequent in Feb-Apr

- ✓ **Watch out** for the upcoming publications!

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K. Müller et al. (*2021*): The Palau Atmospheric Observatory – Introducing the comprehensive setup and the ECC ozone sondes time series, in prep.

This PICO is based on an earlier conference submission:

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

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*EXTRA SLIDES

Palau Atmospheric Observatory

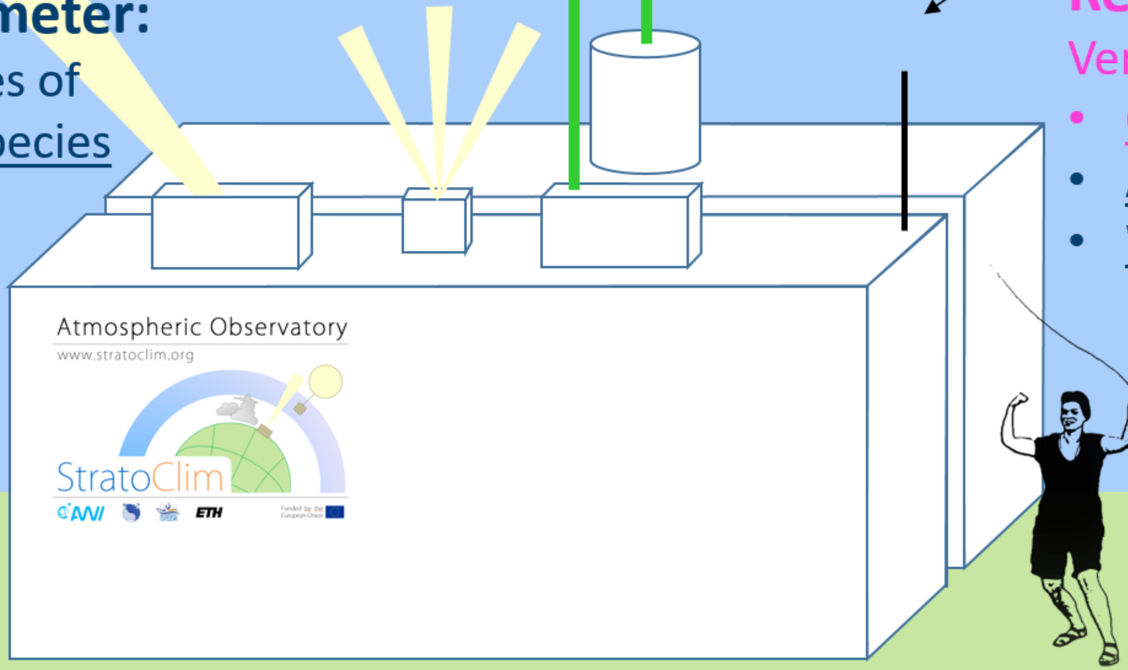
MaxDOAS:
Pandora2S –
Pandonia Network
O_3, NO_2, AOD
(, H_2O, SO_2, …)

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

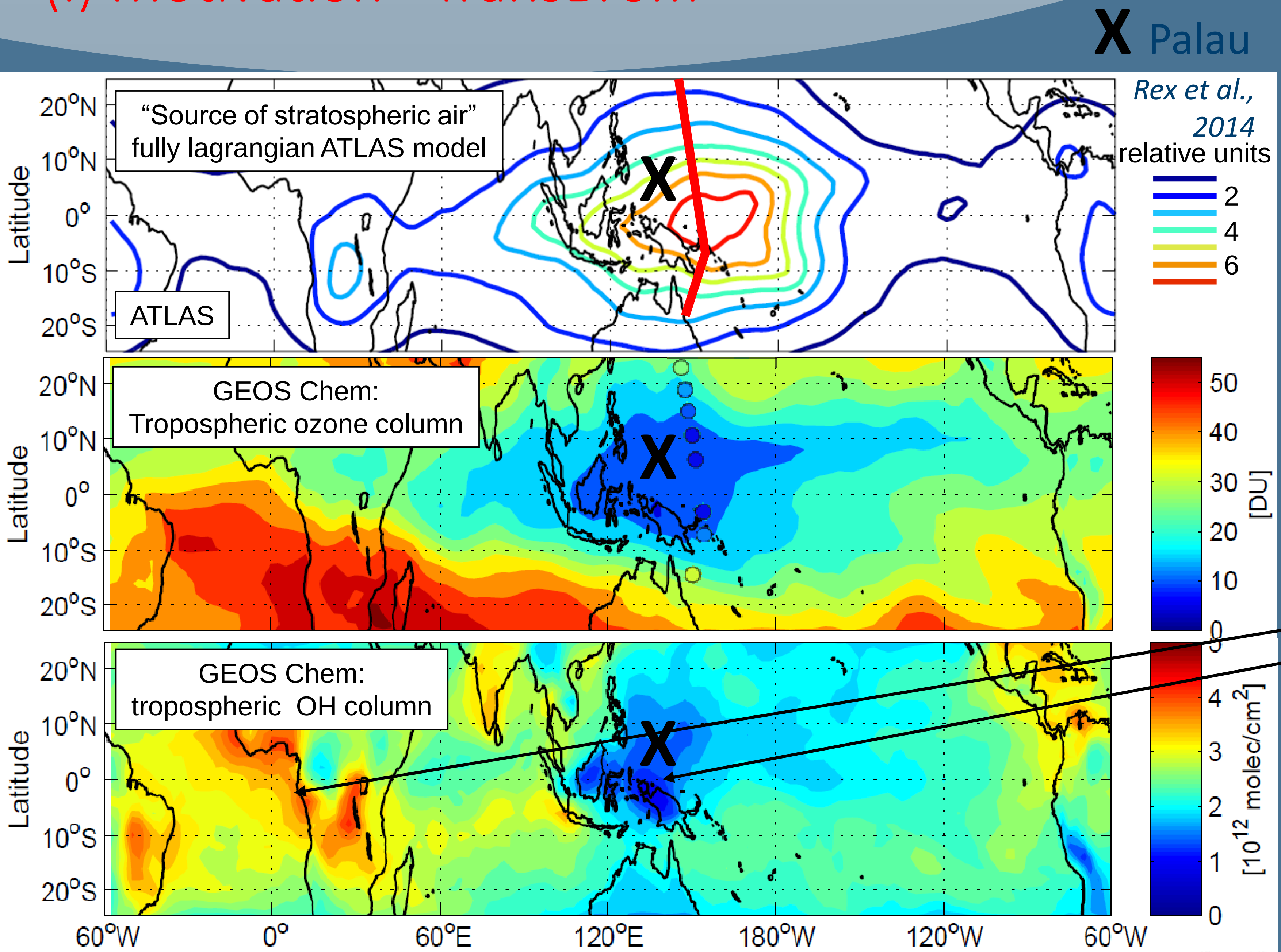
Research balloons:
Vertical profiles of
- Ozone
- Aerosol
- Water vapour

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

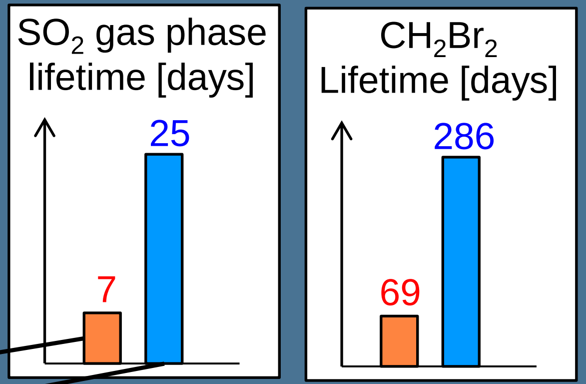


Müller 2020

(I) Motivation - TransBrom



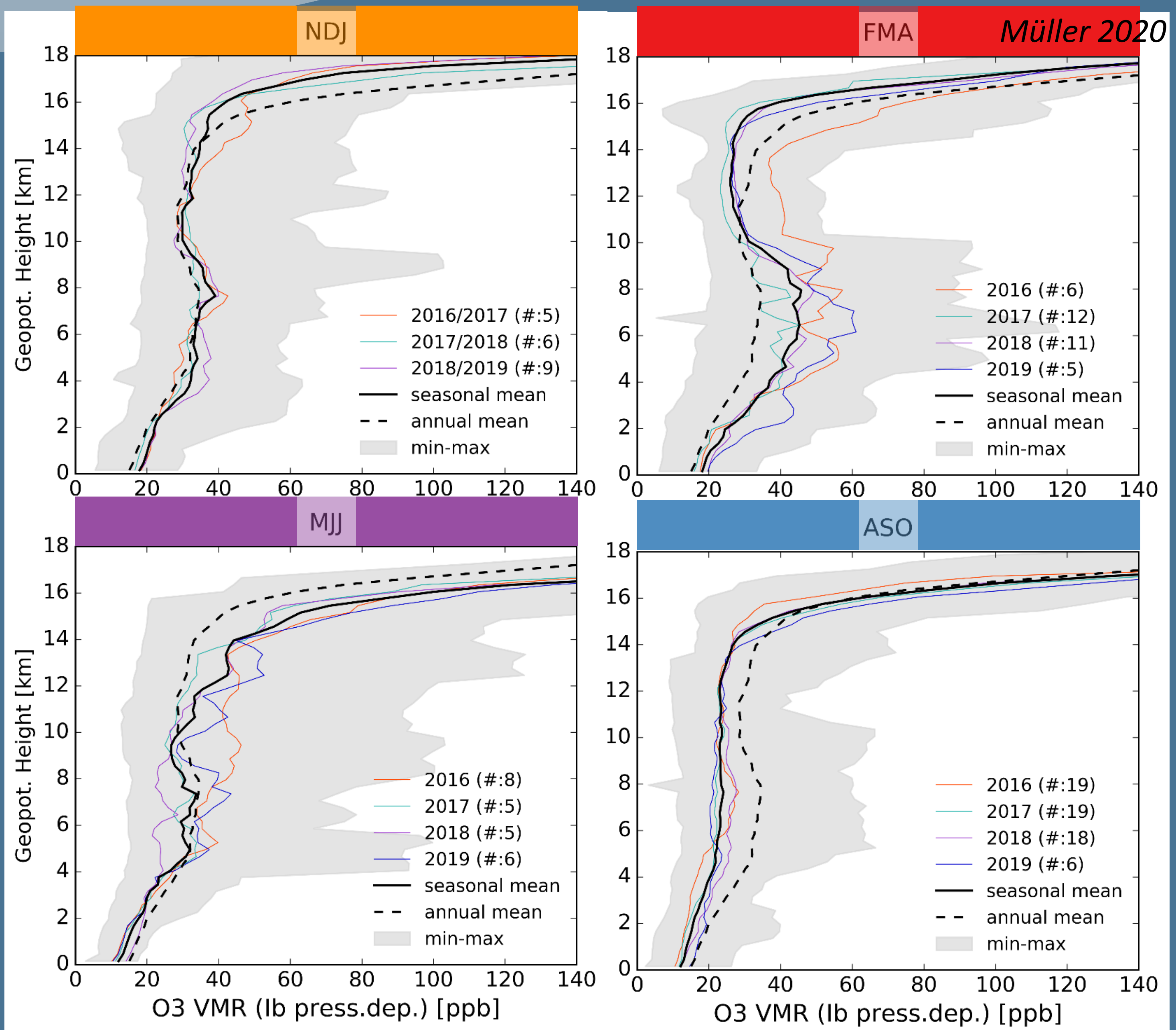
Density distribution function of the horizontal positions of the trajectories between boundary layer and Lagrangian Cold Points; **red thick line:** TransBrom Cruise 2009; filled circles: from ozonesonde measurements



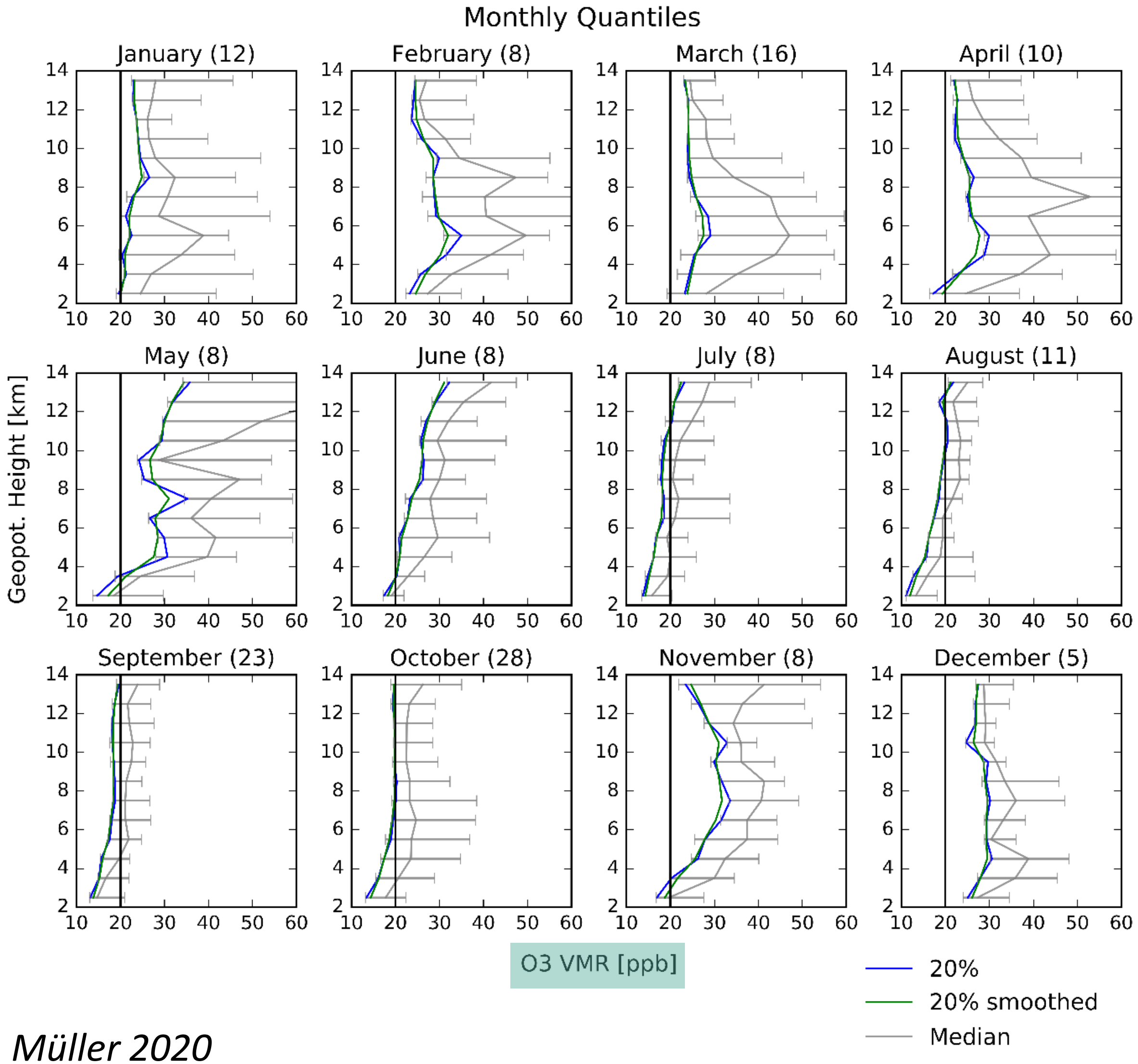
Lifetime comparison for tropical Atlantic and West Pacific (values for mid-troposphere- 500 hPa at the equator for typical conditions)

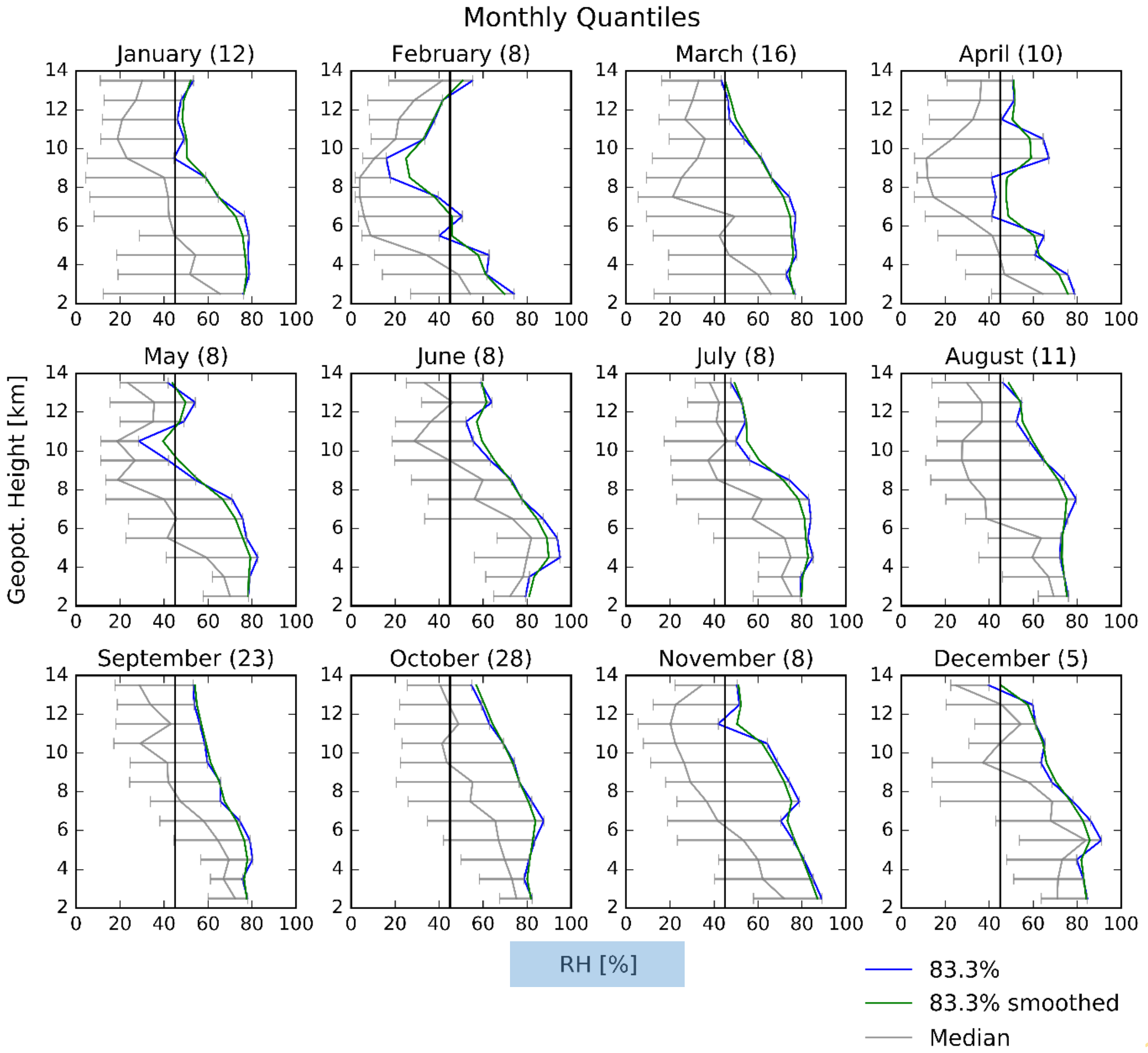
(II) Seasons

- November-January: NDJ, February-April: FMA, …, chosen due to similar profile shapes
- Low O_3 (<25ppb) and enhanced mid-trop. O_3 (>50 ppb) observed in all seasons
- greatest anomalies from the annual mean: **FMA** & **ASO**
- Air masses deviating from a low O_3 background signal occur as **filaments or layers**, predominantly in the **5-10 km** layer, disguised in the averaged belly of the ‚S'



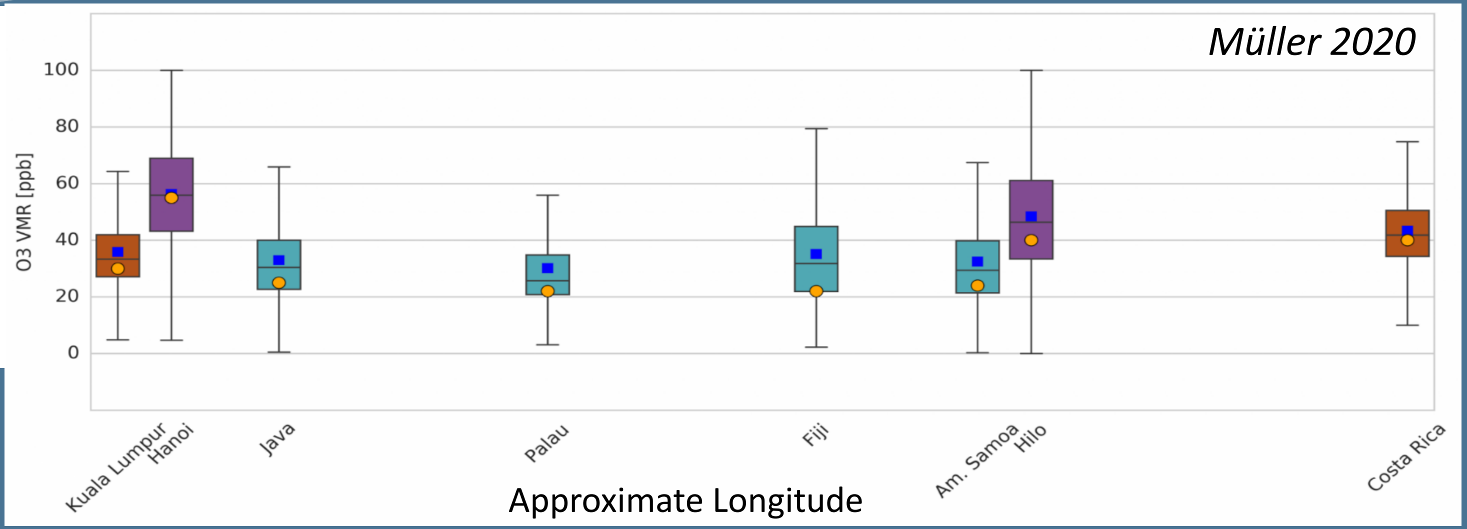
Background Definition



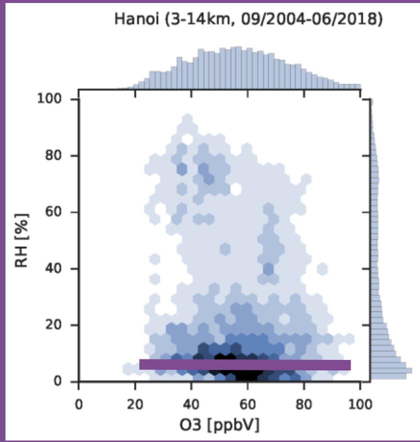


Müller 2020

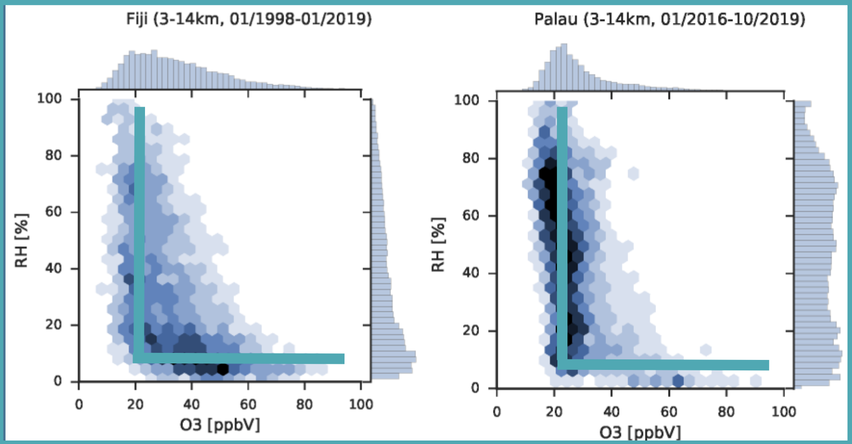
O_3/RH Comparison with SHADOZ



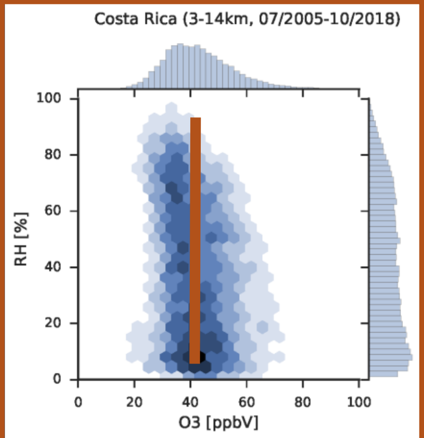
Analyses of 7 selected SHADOZ stations reveals **3 types of free-tropospheric O_3/RH distributions** (see Müller 2020). Seasonal distributions (not shown here) highlight uniqueness of Palau



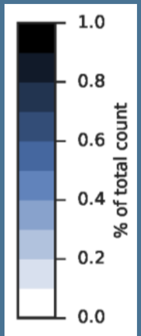
Predominantly dry air over a wider range of O_3 VMR



„L"-shaped: low O_3 over whole RH range + tail towards higher O_3 corresponding to low RH



Gaussian distribution of O_3 + evenly distributed RH



Seasonal Occurrence of O_3/RH groups

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

	O_3 -			O_3 o			O_3 +		
	O3-RH-	O3-RHo	O3-RH+	O3oRH-	O3oRHo	O3oRH+	O3+RH-	O3+RHo	O3+RH+
NDJ (#24)	8	12	12	79	79	54	62	25	12
FMA (#31)	0	19	23	71	74	48	71	42	26
MJJ (#24)	8	17	21	71	88	46	54	50	21
ASO (#59)	5	5	3	86	88	54	25	10	3

0 10 20 30 40 50 60 70 80

seasonal occurence [% of (total # of seasonal profiles)]

Müller 2020

	O_3 -			O_3 o			O_3 +		
	O3-RH-	O3-RHo	O3-RH+	O3oRH-	O3oRHo	O3oRH+	O3+RH-	O3+RHo	O3+RH+
NDJ (#2375)	4	1	1	28	24	16	24	1	1
FMA (#3022)	0	3	4	23	16	8	40	4	3
MJJ (#2438)	0	2	2	32	29	12	16	5	3
ASO (#5728)	1	1	1	40	34	14	9	1	0

0 4 8 12 16 20 24 28 32 36 40

seasonal occurence [% of (total # of seasonal datapoints)]

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.