Origin of Tropospheric Air Masses in the Tropical West Pacific identified by Balloon-borne Ozone and Water Vapor Measurements from Palau

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Abstract

Motivated by previous measurements of very low tropospheric ozone concentrations in the Tropical West Pacific (TWP) and the implied low oxidizing capacity of this key region for transport into the stratosphere (e.g. Rex et al. 2014), we set up an atmospheric research station in Palau (7° N 134° E). Our analysis of regular balloon-borne tropospheric ozone observations at Palau from 01/2016-10/2019 confirms the year-round dominance of a low ozone background in the mid-troposphere. Layers of enhanced ozone are often anti-correlated with water vapor and occur frequently. Moreover, the occurrence of respective layers shows a strong seasonality. Dry and ozone-rich air masses between 5 and 10 km altitude were observed in 71 % of the profiles from February until April compared to 25 % from August until October. By defining monthly atmospheric background profiles for ozone and relative humidity based on observed statistics, we found that the deviations from this background reveal a bimodal distribution of RH anomalies. A previously proposed universal bimodal structure of free tropospheric ozone in the TWP could not be verified (Pan et al. 2015). Back trajectory calculations confirm that throughout the year the mid-tropospheric background is controlled by local convective processes and the origin of air masses is thus close to or East of Palau in the Pacific Ocean. Dry and ozone-rich air originates in tropical Asia and reaches Palau in anticyclonic conditions over an area stretching from India to the Philippines. This supports the hypothesis of several studies which attribute ozone enhancement against the low ozone background to remote pollution events on the ground such as biomass burning (e.g. Andersen et al. 2016). A potential vorticity analysis revealed no stratospheric influence and we thus propose large-scale descent within the tropical troposphere as responsible for dehydration of air masses on their way to Palau.

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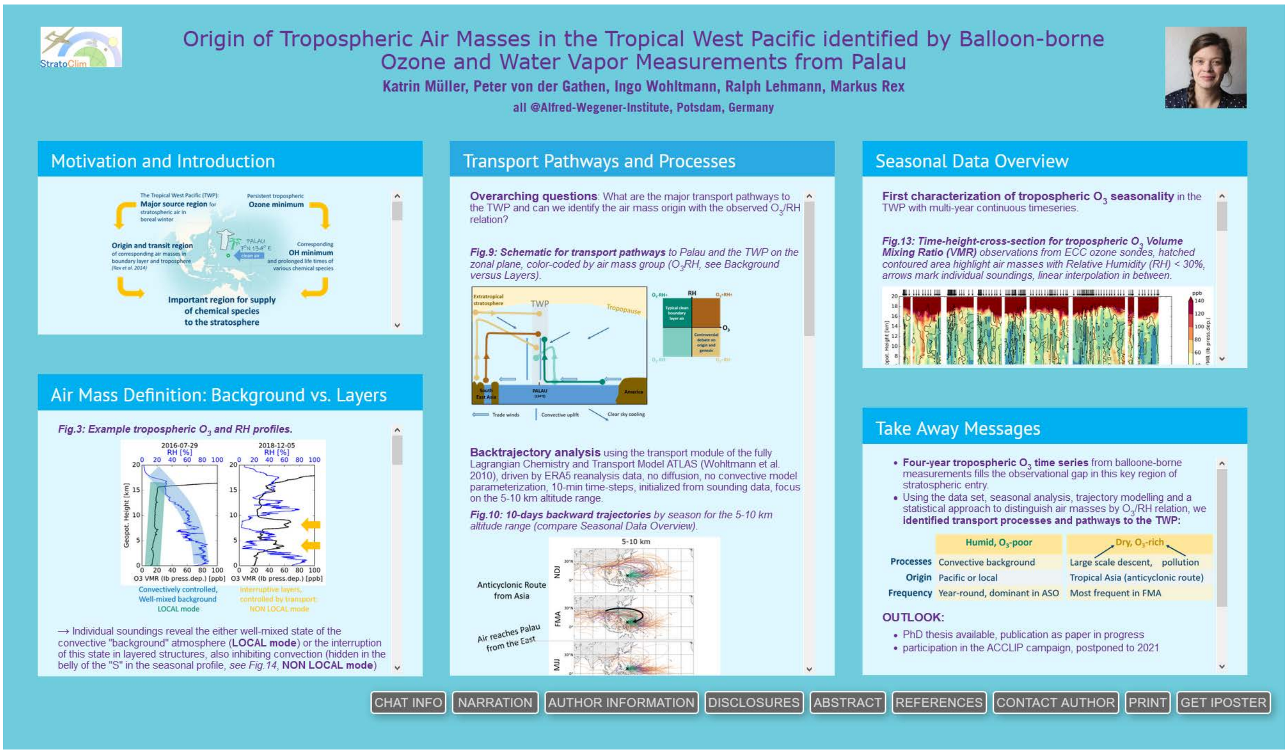

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PRESENTED AT:

MOTIVATION AND INTRODUCTION

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:

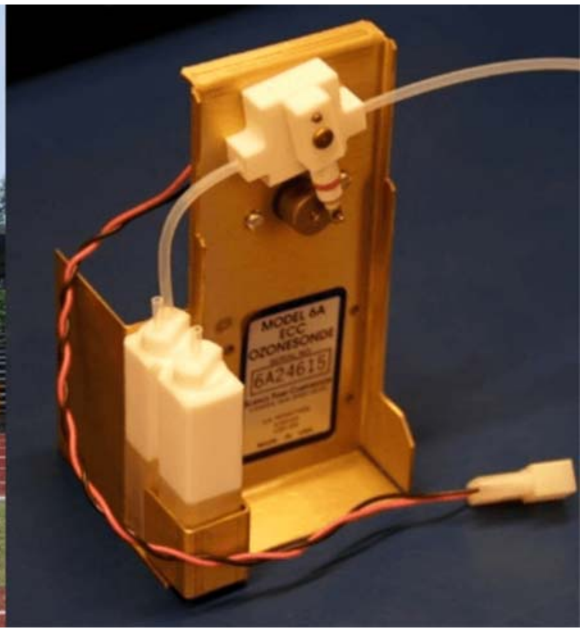
$$O_3 + h\nu \rightarrow O(^1D) + O_2$$
$$O(^1D) + H_2O \rightarrow OH + OH$$

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.

Fig.1: Palau Atmospheric Observatory *(Photo: Ingo Beninga).*

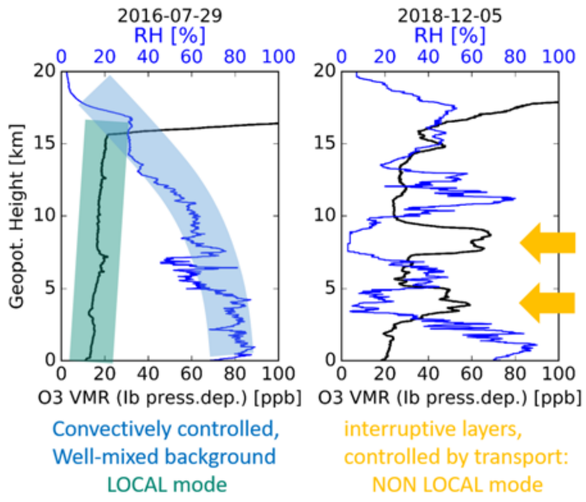
The data set presented here consists of 145 soundings from ECC (Electrochemical Concentration Cell) ozone- and radiosondes (SPC model 6A, Vaisala RS92/41), from 01/2016 until 10/2019, obtained in bi-weekly frequency or intensive measurement campaigns.

Fig.2: Weather balloon launch with ECC ozonesonde
(Photos: Jürgen Graeser, Katrin Müller).


MODEL 6A
ECC
OZONESONDE
6A24615

AIR MASS DEFINITION: BACKGROUND VS. LAYERS

Fig.3: Example tropospheric O_3 and RH profiles.

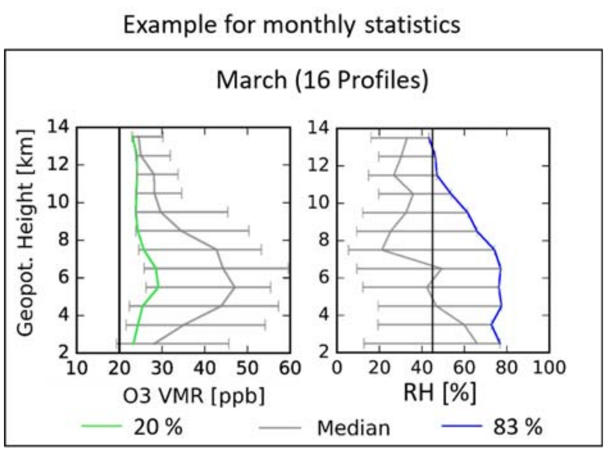


→ Individual soundings reveal the either well-mixed state of the convective "background" atmosphere (**LOCAL mode**) or the interruption of this state in layered structures, also inhibiting convection (hidden in the belly of the "S" in the seasonal profile, *see Fig.14*, **NON LOCAL mode**)

Underlying processes:

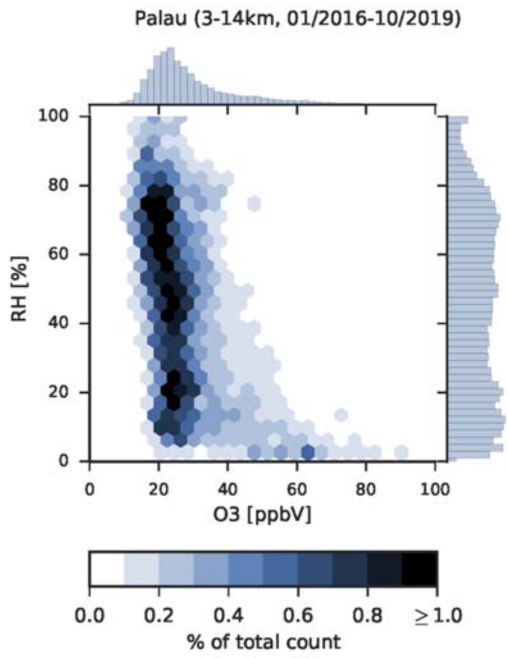
- Local boundary layer air masses lacking pollution and therefore **low in O_3** are lifted locally by convection.
- No known mechanism for in situ production of **high O_3** or dehydration in the mid-troposphere - origin either transport from the (extratropical) stratosphere or non-local ground pollution, lifted convectively in the area of origin then undergoing dehydration during transport, e.g. via large-scale descent and radiative cooling (*see Fig.9, compare Dessler and Minschwaner, 2007; Anderson et al. 2016*).

***Fig.4: Example of the statistical approach** to determine a background profile and identify deviating layers from the entire dataset.*



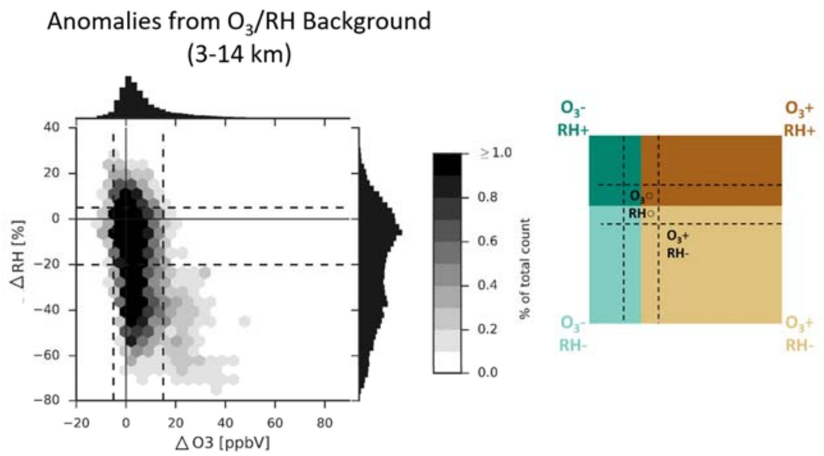
→ **Background definition:** the monthly 20^{th} (O_3) and 83^{rd} (RH) quantile, altitude-dependent.

***Fig.5: Free-tropospheric (3-14 km) O_3/RH relation** for the full data set.*



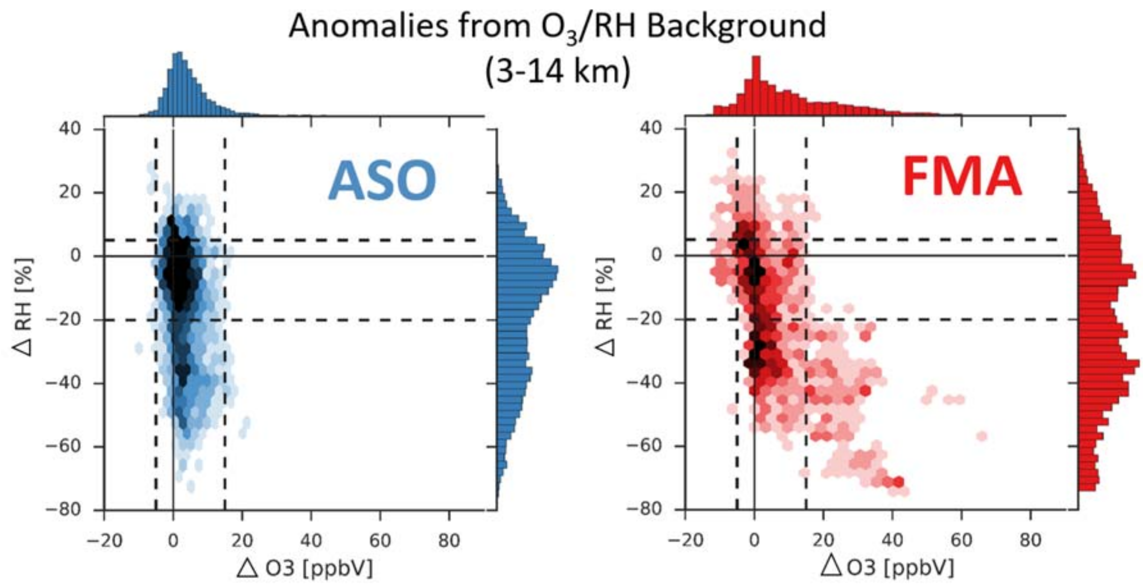
→ **Unique for Palau** compared to other SHADOZ observations, but offers no ideal separation of background and layers. Additional analysis similar to Pan et al. 2015 was also unsuccessful.

Fig.6: O_3 and RH anomalies *from the above defined background and classification of air masses in a 3x3 grid.*



→ **Bimodality in RH anomalies:** motivating classification in O_3RH groups and thus constituting our air mass definition in the following.

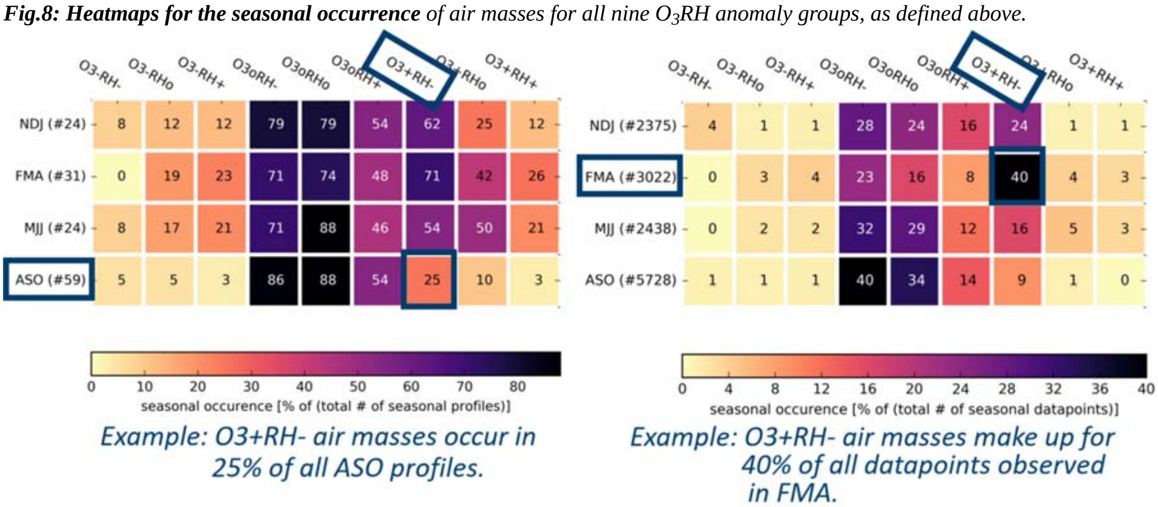
Fig.7: Seasonal differences *of air mass occurence (compare with "Seasonal Data Overview").*



→ **O_3oRHo:** humid, O_3-poor background occurs year-round, but dominates ASO.

→ **O_3+RH-:** dry, O_3-rich air masses are most frequent in FMA.

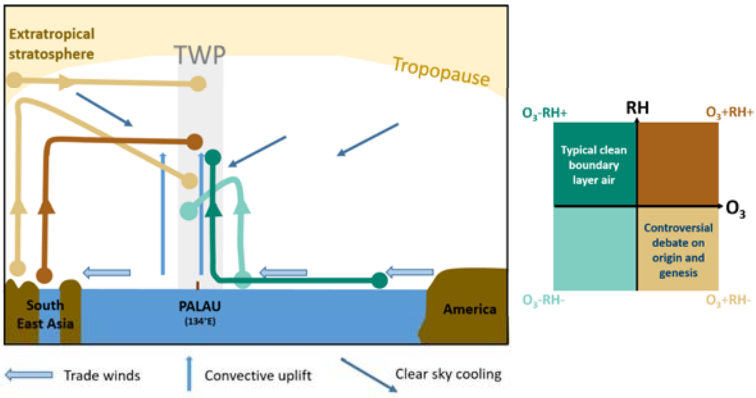
Fig.8: Heatmaps for the seasonal occurrence *of air masses for all nine O_3RH anomaly groups, as defined above.*



TRANSPORT PATHWAYS AND PROCESSES

Overarching questions: What are the major transport pathways to the TWP and can we identify the air mass origin with the observed O_3/RH relation?

Fig.9: Schematic for transport pathways *to Palau and the TWP on the zonal plane, color-coded by air mass group (O_3RH, see Background versus Layers).*



Backtrajectory analysis using the transport module of the fully Lagrangian Chemistry and Transport Model ATLAS (Wohltmann et al. 2010), driven by ERA5 reanalysis data, no diffusion, no convective model parameterization, 10-min time-steps, initialized from sounding data, focus on the 5-10 km altitude range.

Fig.10: 10-days backward trajectories *by season for the 5-10 km altitude range (compare Seasonal Data Overview).*

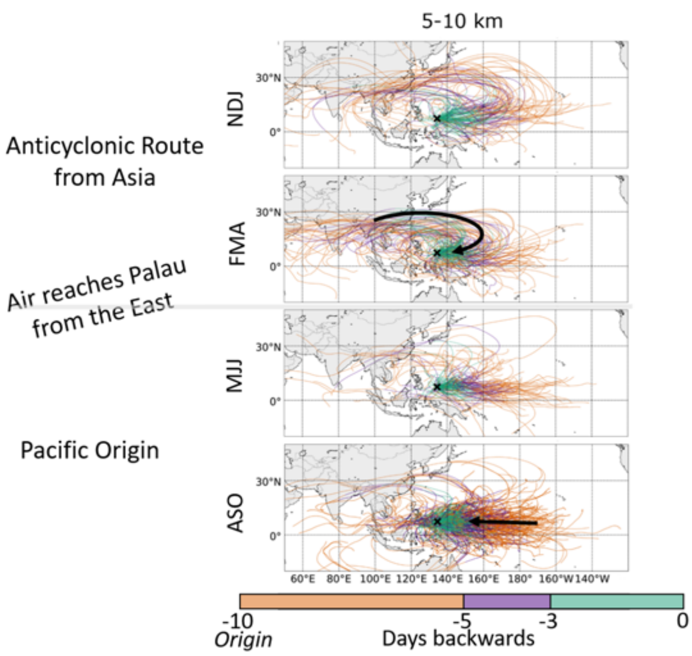
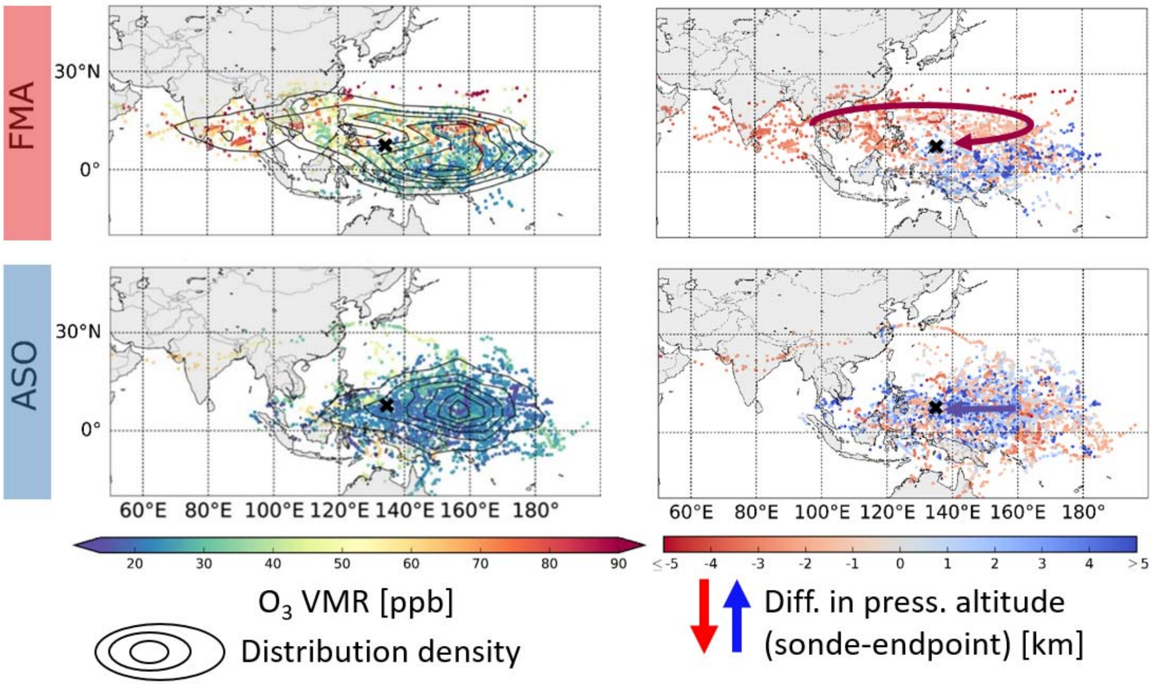


Fig.11: 5-days backward trajectory ending points *(5-10 km), defined here as origin of air masses (due to lifetime of marine boundary layer O_3)* ***by season****, color-coded by either O_3 or difference in pressure altitude.*

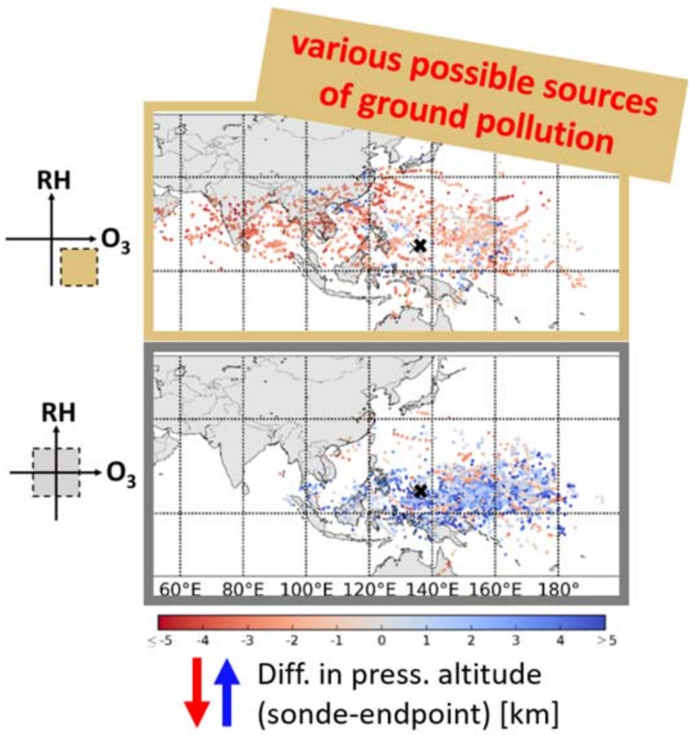


→ **Center of** **low O_3** in both seasons East of Palau, **secondary center of** **enhanced O_3** in FMA, North of Palau from India to East China

→ **Vertical displacement of air masses:**

- mainly in FMA, North of Palau, air masses **descend** towards the islands **(anti-cyclonic route)**, consistent with the large-scale descent within the Hadley circulation and subsequent dehydration
- **ascent** dominates ASO air masses, corresponding well with the proposed convective uplift.

Fig.12: 5-days backward trajectory ending points *(5-10 km) selected* ***by air mass group*** *(see Fig.6 for definition).*



→ **Selection of trajectories for** air masses identified as humid,O_3-poor background (**O_3oRHo**) or dry, O_3-rich (**O_3+RH-**) anomaly from the background (see Air Mass Definition) separates air masses according to the processes controlling RH (convective uplift in blue, ASO, and dehydration during descent in red, FMA) and locates spacially separate source regions (see below).

- **No** indication for significant contribution of **stratospheric air**: Potential Vorticity analysis for all profiles (#138, 5-10 km) revealed essentially no air mass crossing the 1.5 PVU threshold for more than a day during 10 days backwards.
- Origin of air masses are areas of increased air pollution on the ground from industry or bio mass burning, speaking in favor for the **pollution based origin.**

Summarized results:

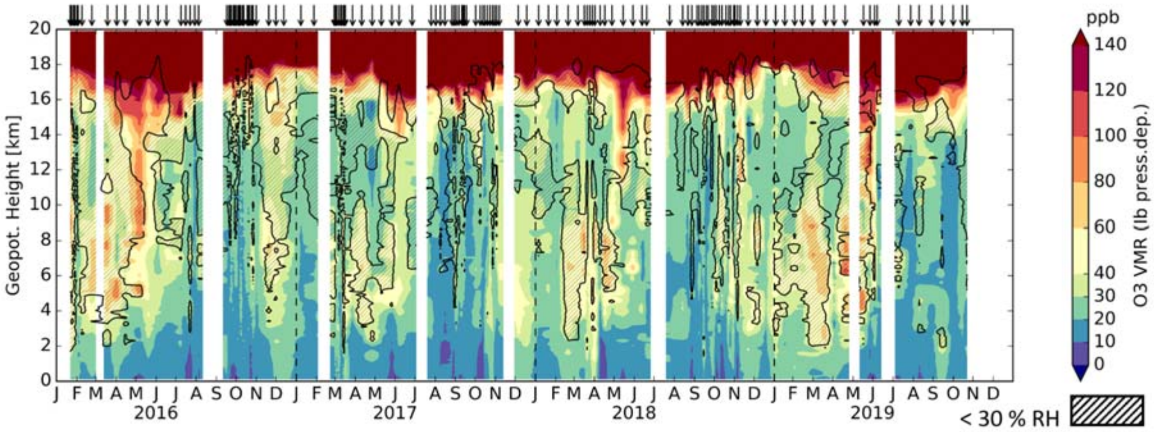
Dry, O_3-rich: most frequent in **FMA,** ↓, tropical ASIA and Pacific

Humid, O_3-poor: year-round, but dominant in **ASO,** ↑ , PACIFIC

SEASONAL DATA OVERVIEW

First characterization of tropospheric O_3 seasonality in the TWP with multi-year continuous timeseries.

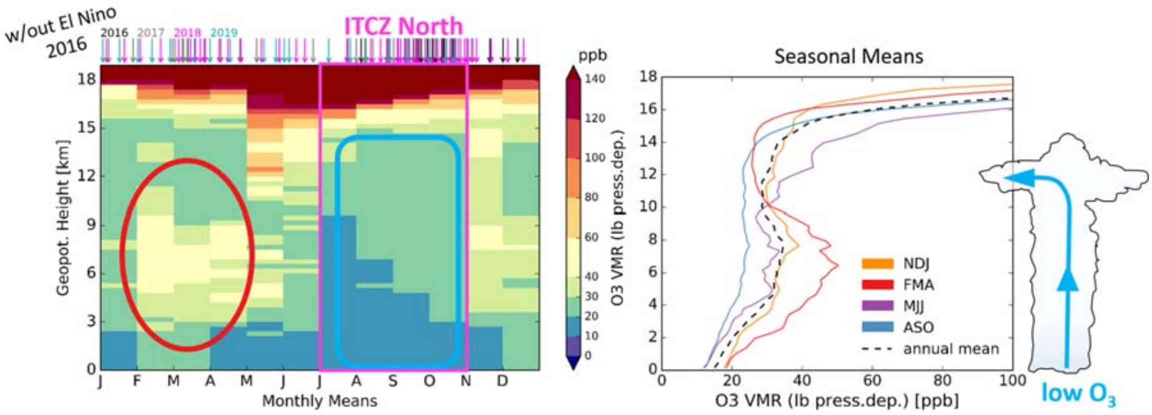
***Fig.13: Time-height-cross-section for tropospheric O_3 Volume Mixing Ratio (VMR)** observations from ECC ozone sondes, hatched contoured area highlight air masses with Relative Humidity (RH) < 30%, arrows mark individual soundings, linear interpolation in between.*



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

→ **TTL cycle:** fits tropopause height and temperature variations related to the seasonality of the Brewer Dobson circulation and enhanced high altitude convective outflow (Randel et al. 2007, Folkins et al. 2006).

Fig.14: Time-height-cross-section for monthly mean O_3 VMR and seasonal mean profiles.



→ **Monthly means** highlight the annual cycles: O_3 minimum corresponds with the Intertropical Convergence Zone (ITCZ) located North of Palau.

→ **Annual mean:** typical (tropical) "S-shape"

→ Monthly means grouped according to similar shape:
4 distinct types of profiles (seasons):

NDJ -- November-December-January

FMA -- February-March-April

MJJ -- March-June-July

ASO -- August-September-October

→ 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** (background) and **FMA** (deviating layers, *compare Fig.7,8*).

TAKE AWAY MESSAGES

- **Four-year tropospheric O_3 time series** from balloone-borne measurements fills the observational gap in this key region of stratospheric entry.
- Using the data set, 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, dominant in ASO	Most frequent in FMA

OUTLOOK:

- PhD thesis available, publication as paper in progress
- participation in the ACCLIP campaign, postponed to 2021

DISCLOSURES

Major part of this work was funded under the EU-project StratoClim (www.stratoclim.org), additional funding was provided by the Alfred-Wegener-Institute. All data and code are available upon request.

We used ECMWF ERA5 data provided by the Copernicus Climate Change Service Climate Data Store for the trajectory analysis.

I want to acknowledge our logistical and scientific service partners in Palau, namely CTSI Logistics and CRRF, including our main local operator Sharon Patris, the Palau Community College and principal Patrick Tellei for the provision of space, but also technical "inhouse" support from the AWI mainly by Jürgen Graeser, but also impres GmbH with Ingo Beninga and Wilfried Ruhe, majorly involved in setting up and maintaining the station.

AUTHOR INFORMATION

I am a Postdoc at the Alfred-Wegener-Institute in Potsdam, Germany, and just recently defended my PhD thesis, from which I present major results on my poster.

Feel free to contact me, I am happy and curious for new collaborations!

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Motivated by previous measurements of very low tropospheric ozone concentrations in the Tropical West Pacific (TWP) and the implied low oxidizing capacity of this key region for transport into the stratosphere (e.g. Rex et al. 2014), we set up an atmospheric research station in Palau (7°N 134°E). Our analysis of regular balloon-borne tropospheric ozone observations at Palau from 01/2016-10/2019 confirms the year-round dominance of a low ozone background in the mid-troposphere. Layers of enhanced ozone are often anti-correlated with water vapor and occur frequently. Moreover, the occurrence of respective layers shows a strong seasonality. Dry and ozone-rich air masses between 5 and 10 km altitude were observed in 71 % of the profiles from February until April compared to 25 % from August until October. By defining monthly atmospheric background profiles for ozone and relative humidity based on observed statistics, we found that the deviations from this background reveal a bimodal distribution of RH anomalies. A previously proposed universal bimodal structure of free tropospheric ozone in the TWP could not be verified (Pan et al. 2015).

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