



# Challenges of detecting free tropospheric ozone trends in a sparsely sampled environment

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October 24 to 27, 2023

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## Evaluating satellite-detected tropospheric ozone trends

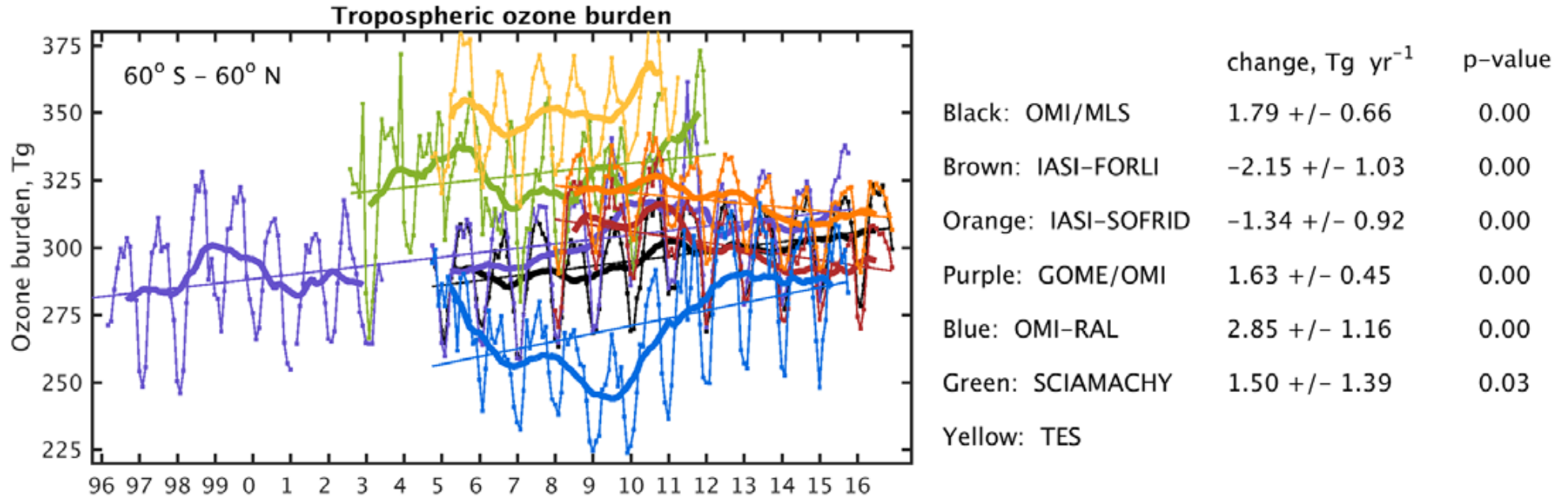


Figure 26. from *TOAR-Climate* (Gaudel et al., 2018)

Gaudel, A., et al. (2018), *Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation*, *Elem. Sci. Anth.*, 6(1):39, DOI: <https://doi.org/10.1525/elementa.291>

# Ozone trends have high regional variability among satellite products

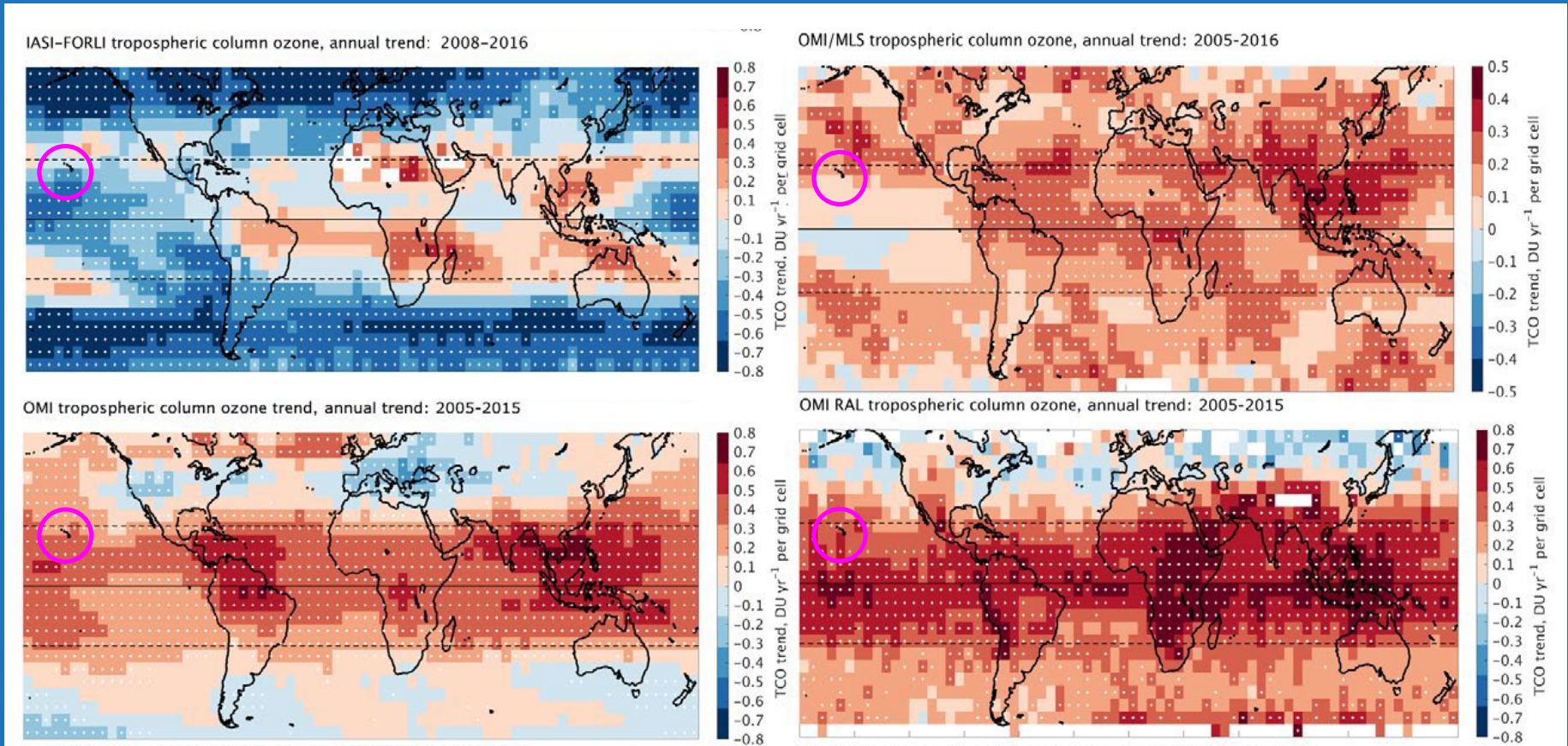


Figure 24. from *TOAR-Climate* (Gaudel et al., 2018)

Gaudel, A., et al. (2018), *Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation*, *Elem. Sci. Anth.*, 6(1):39, DOI: <https://doi.org/10.1525/elementa.291>

# What sampling rate (*in situ*) is required to detect an ozone trend in the free troposphere?

Sites with sampling rates  
> 1 per week

Frankfurt (IAGOS): daily

JPL Table Mountain lidar:  
4-5 per week since 2018

3 per week since 1960s  
- Uccle, Belgium  
- Payerne, Switzerland  
- Hohenpeissenberg, Germany

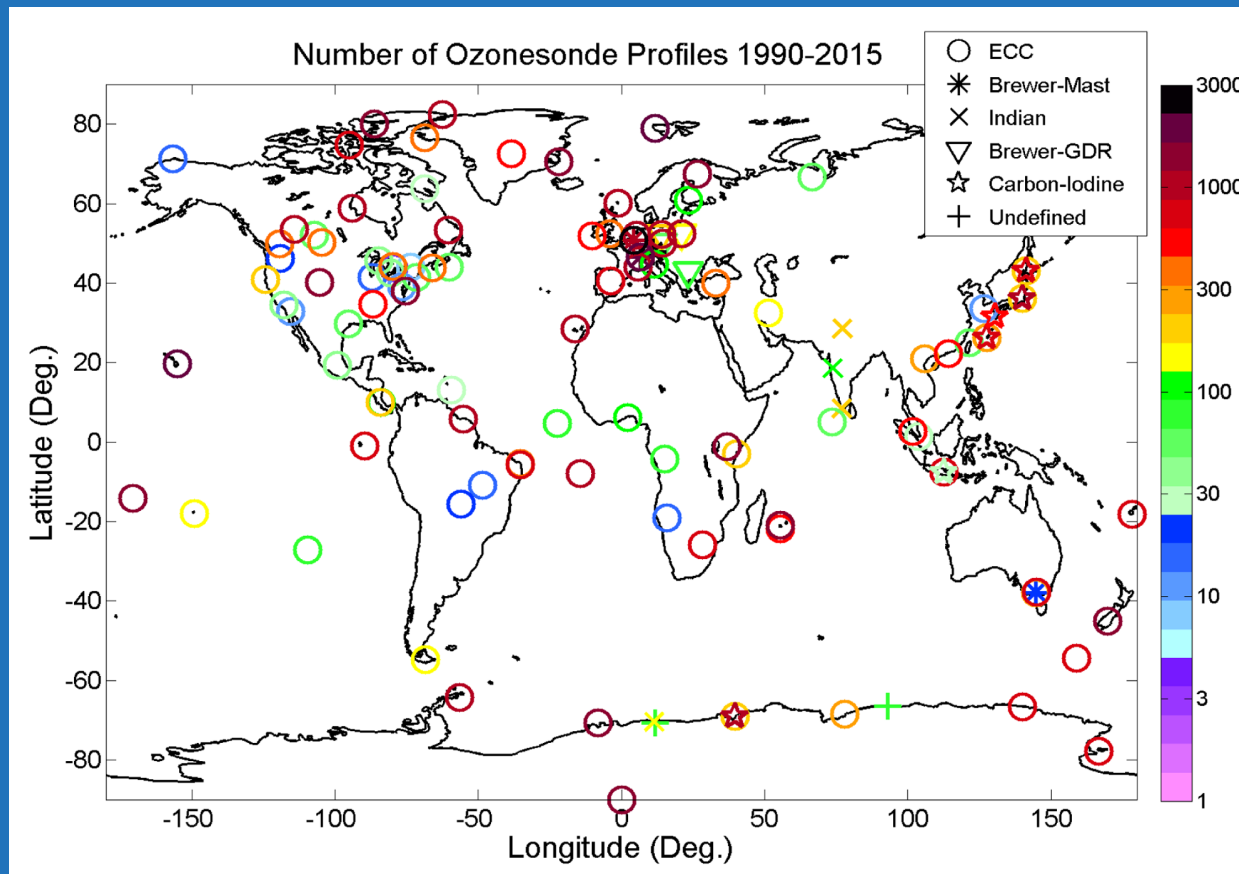


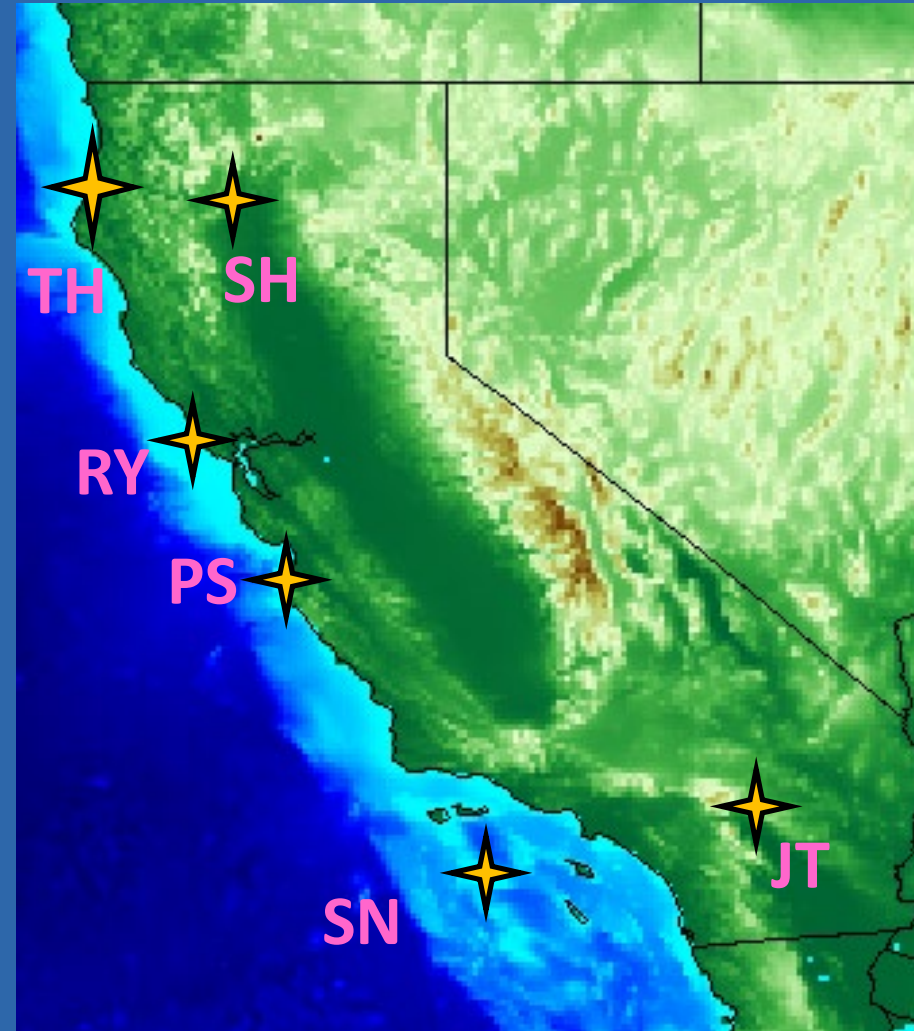
Figure 16. from *TOAR-Observations* (Tarasick and Galbally et al., 2019)

Tarasick, D. W., I. E. Galbally (2019), *Tropospheric Ozone Assessment Report: Tropospheric ozone from 1877 to 2016, observed levels, trends and uncertainties*. *Elem Sci Anth*, 7(1), DOI: <http://doi.org/10.1525/elementa.376>

# CALNEX 2010 ozonesonde network: May 10 – June 19, 2010

The experiment yielded:

- a total of 130 coastal ozone profiles
- from four coastal sites over a six week period
- the most detailed set of ozone profiles ever collected along the US west coast

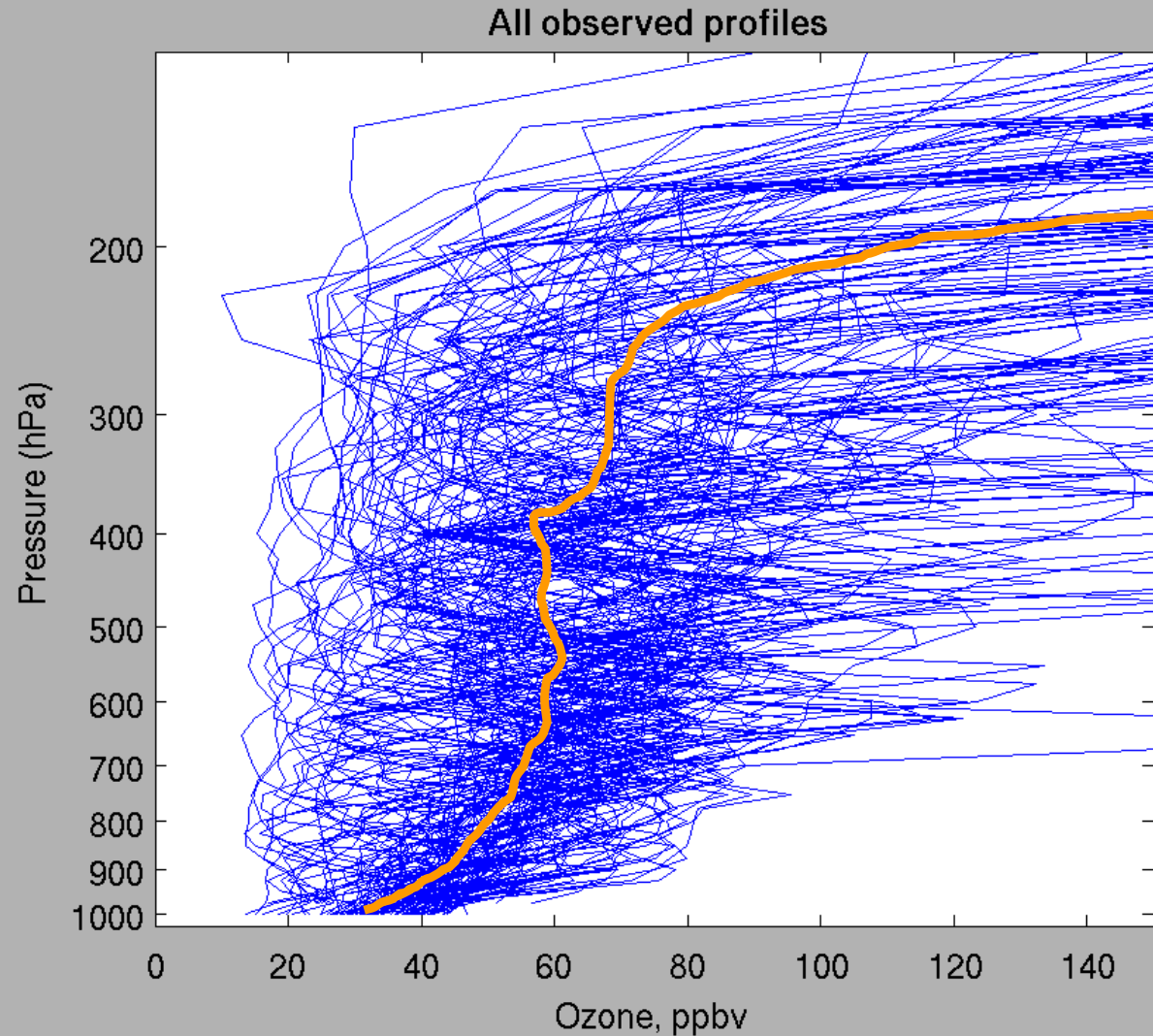


Site	Latitude	Longitude	Elevation	Number of Profiles
<i>IONS-2010 Ozonesonde Sites</i>				
Trinidad Head (TH)	41.06	-124.16	20 m	37
Point Reyes (RY)	38.09	-122.95	29 m	35
Point Sur (PS)	36.30	-121.89	12 m	37
San Nicolas (SN)	33.26	-119.49	14 m	26
Joshua Tree (JT)	34.08	-116.39	1216 m	36
Shasta (SH)	40.60	-122.49	314 m	34

Cooper, O. R., et al. (2011), Measurement of western U.S. baseline ozone from the surface to the tropopause and assessment of downwind impact regions, J. Geophys. Res., 116, D00V03, doi:10.1029/2011JD016095

Free tropospheric ozone trends are typically calculated from monthly or seasonal means.

What is the smallest sample size required to capture the monthly mean profile?



## Past research on ozone profile sampling rate

Since 1999, at least 4 papers have discussed the challenges of quantifying ozone monthly means and/or ozone trends based on sparsely sampled *in situ* data sets.

In 1999, Jennifer Logan concluded that for mid-latitudes a minimum of 20 profiles per month (~5 per week) is required to ensure that the monthly mean value is reliable to  $\pm 15\%$  for 800-500 hPa.

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 104, NO. D13, PAGES 16,115–16,149, JULY 20, 1999

### **An analysis of ozonesonde data for the troposphere: Recommendations for testing 3-D models and development of a gridded climatology for tropospheric ozone**

Jennifer A. Logan

Department of Earth and Planetary Sciences and Division of Engineering and Applied Sciences,  
Harvard University, Cambridge, Massachusetts

GEOPHYSICAL RESEARCH LETTERS, VOL. 26, NO. 14, PAGES 2175-2178, JULY 15, 1999

### **Effect of rising Asian emissions on surface ozone in the United States**

Daniel J. Jacob, Jennifer A. Logan and Prashant P. Murti

Division of Engineering and Applied Science, and Department of Earth and Planetary Sciences,  
Harvard University

## Past research on ozone profile sampling rate

In 2010, *Cooper et al.* merged all April-May ozone profiles above western North America to show that ozone had increased in the free troposphere from 1995 to 2008.

They determined that 50 profiles per April-May season (or 25 profiles per month, or ~6 per week) are required to produce a regional mean value within  $\pm 2\%$  of the true mean value.

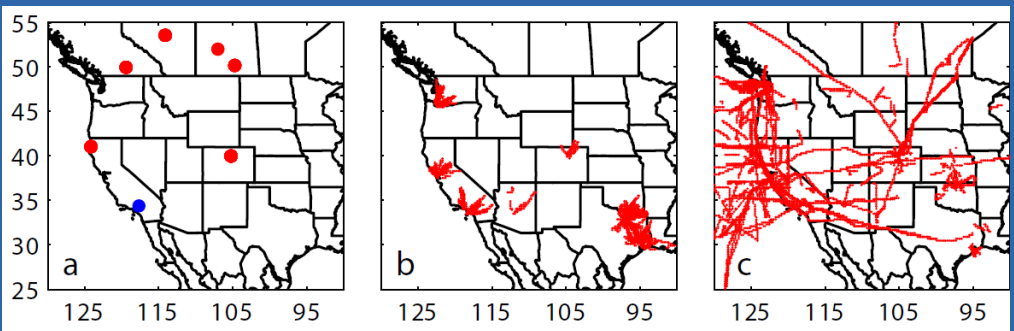
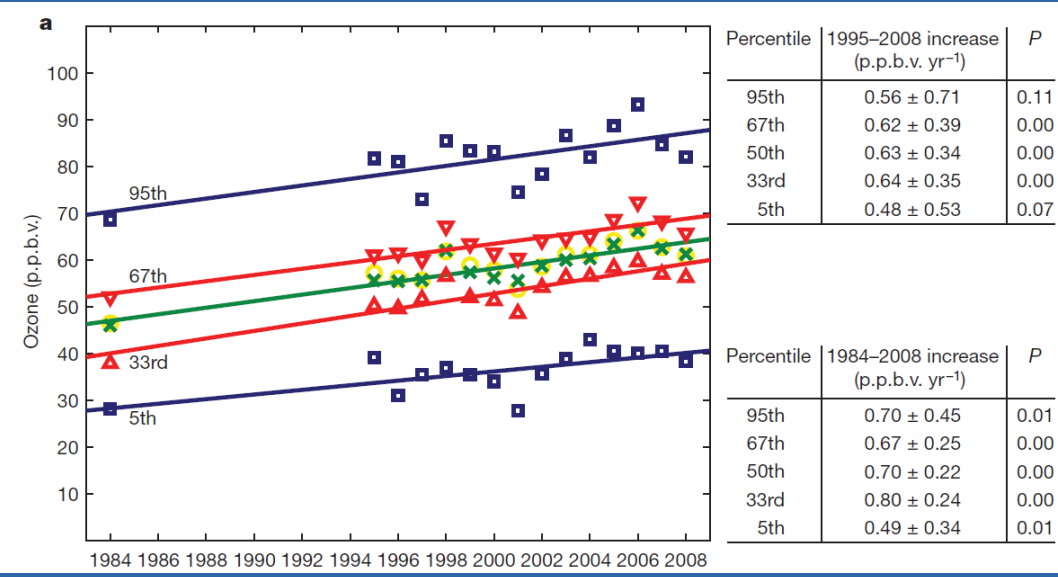


Figure S1. Maps showing the study region and the locations of a) ozonesonde (red) and lidar (blue) stations, b) MOZAIC aircraft profiles between 3-8 km, and c) research aircraft flight tracks between 3-8 km.

# LETTERS

## Increasing springtime ozone mixing ratios in the free troposphere over western North America

O. R. Cooper<sup>1,2</sup>, D. D. Parrish<sup>2</sup>, A. Stohl<sup>3</sup>, M. Trainer<sup>2</sup>, P. Nédélec<sup>4</sup>, V. Thouret<sup>4</sup>, J. P. Cammas<sup>4</sup>, S. J. Oltmans<sup>2</sup>, B. J. Johnson<sup>2</sup>, D. Tarasick<sup>5</sup>, T. Leblanc<sup>6</sup>, I. S. McDermid<sup>6</sup>, D. Jaffe<sup>7</sup>, R. Gao<sup>2</sup>, J. Stith<sup>8</sup>, T. Ryerson<sup>2</sup>, K. Aikin<sup>1,2</sup>, T. Campos<sup>9</sup>, A. Weinheimer<sup>9</sup> & M. A. Avery<sup>10</sup>





## Past research on ozone profile sampling rate

In 2012, *Saunois et al.* sub-sampled the daily IAGOS ozone profiles above Frankfurt, Germany.

A sampling strategy of only 4 profiles per month (or 1 profile per week) results in an uncertainty of seasonal mean ozone that is typically greater than 10% in the free troposphere.

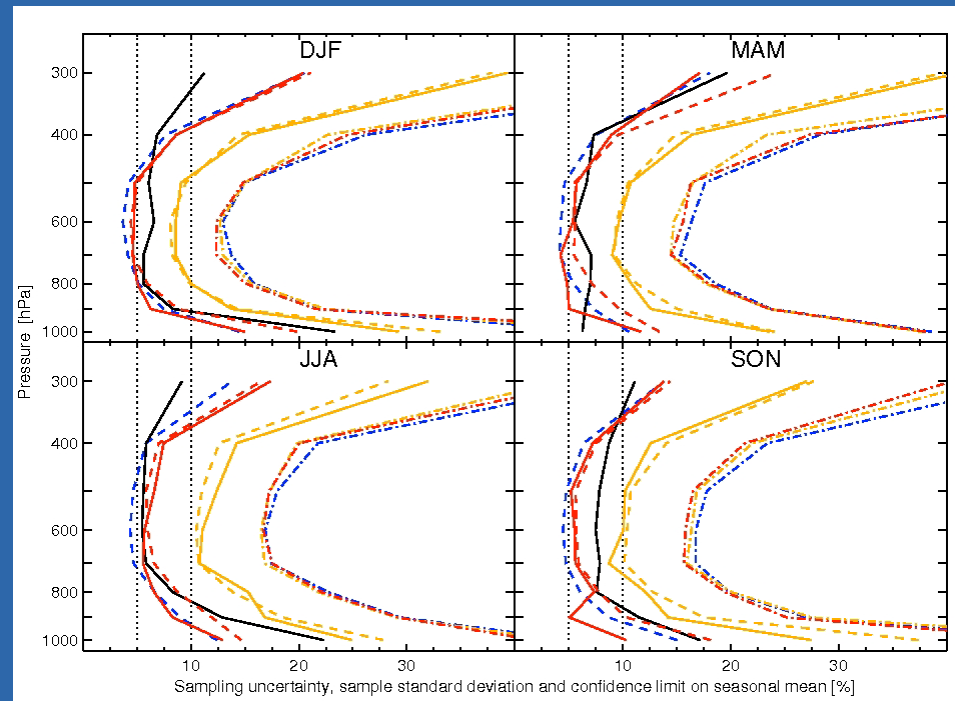
But if the sampling strategy is increased to 12 profiles per month (3 per week), the uncertainty is only about 5%.

Atmos. Chem. Phys., 12, 6757–6773, 2012  
www.atmos-chem-phys.net/12/6757/2012/  
doi:10.5194/acp-12-6757-2012  
© Author(s) 2012. CC Attribution 3.0 License.



## Impact of sampling frequency in the analysis of tropospheric ozone observations

M. Saunois<sup>1,\*</sup>, L. Emmons<sup>1</sup>, J.-F. Lamarque<sup>1</sup>, S. Tilmes<sup>1</sup>, C. Wespes<sup>1</sup>, V. Thouret<sup>2,3</sup>, and M. Schultz<sup>4</sup>



# Past research on ozone profile sampling rate

In response to these earlier studies, *Chang et al.* (2020) developed an improved methodology for detecting trends based on sparse ozonesonde profiles.

Using a standard trend detection method, 8 profiles per month are required for detecting the signal of the trend at a 2-sigma confidence level, and 18 profiles per month are required for the bias of the trend to be less than 5%.

Using the improved trend-detection method, 4 profiles per month are required for basic trend detection and 14 profiles per month for accurate trend quantification.

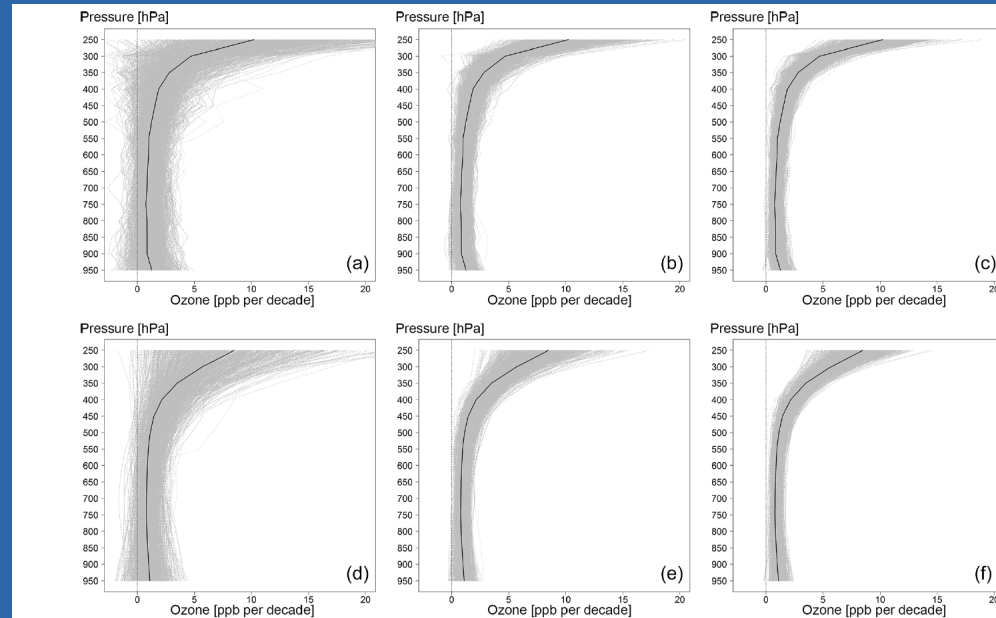
“While our method improves trend detection from sparse datasets, the key to substantially reducing the uncertainty is to increase the sampling frequency.”

Atmos. Chem. Phys., 20, 9915–9938, 2020  
<https://doi.org/10.5194/acp-20-9915-2020>  
© Author(s) 2020. This work is distributed under the Creative Commons Attribution 4.0 License.



## Statistical regularization for trend detection: an integrated approach for detecting long-term trends from sparse tropospheric ozone profiles

Kai-Lan Chang<sup>1,2</sup>, Owen R. Cooper<sup>1,2</sup>, Audrey Gaudel<sup>1,2</sup>, Irina Petropavlovskikh<sup>1,3</sup>, and Valérie Thouret<sup>4</sup>



**Figure 7.** Sensitivity analysis for one (a, d), five (b, e) and nine (c, f) profiles per month based on 1000 random samples for each of the 15 vertical layers above western Europe. The analysis was conducted using the separated fit (a, b, c) and the integrated fit (d, e, f). Black curves represent the vertical distribution of the true trends based on the full IAGOS dataset.

## Past research on ozone profile sampling rate

Detection of ozone trends at remote locations or in the free troposphere is challenging due to the influence of climate variability (e.g. ENSO) that causes large fluctuations in ozone on annual or decadal time scales.

Modeling studies by *Barnes et al.* (2016) and *Fiore et al.* (2022) show 20 or more years of ozone observations are typically required in order to detect a trend above the noise of climate variability.

## Journal of Geophysical Research: Atmospheres

### RESEARCH ARTICLE

10.1002/2015JD024397

#### Key Points:

- Climate variability induces significant uncertainty in multidecadal trends of atmospheric constituents
- Future trends in surface ozone are mainly driven by internal variability and emissions changes

### Detection of trends in surface ozone in the presence of climate variability

Elizabeth A. Barnes<sup>1</sup>, Arlene M. Fiore<sup>2</sup>, and Larry W. Horowitz<sup>3</sup>

<sup>1</sup>Department of Atmospheric Science, Colorado State University, Fort Collins, Colorado, USA, <sup>2</sup>Department of Earth and Environmental Sciences and Lamont-Doherty Earth Observatory of Columbia University, New York, New York, USA,

<sup>3</sup>NOAA Geophysical Fluid Dynamics Laboratory, Princeton, New Jersey, USA

IOP Publishing

*Environ. Res.: Climate* **1** (2022) 025008

<https://doi.org/10.1088/2752-5295/ac9cc2>

## ENVIRONMENTAL RESEARCH CLIMATE



### PAPER



## Understanding recent tropospheric ozone trends in the context of large internal variability: a new perspective from chemistry-climate model ensembles

OPEN ACCESS

RECEIVED  
9 July 2022

REVISED  
7 September 2022

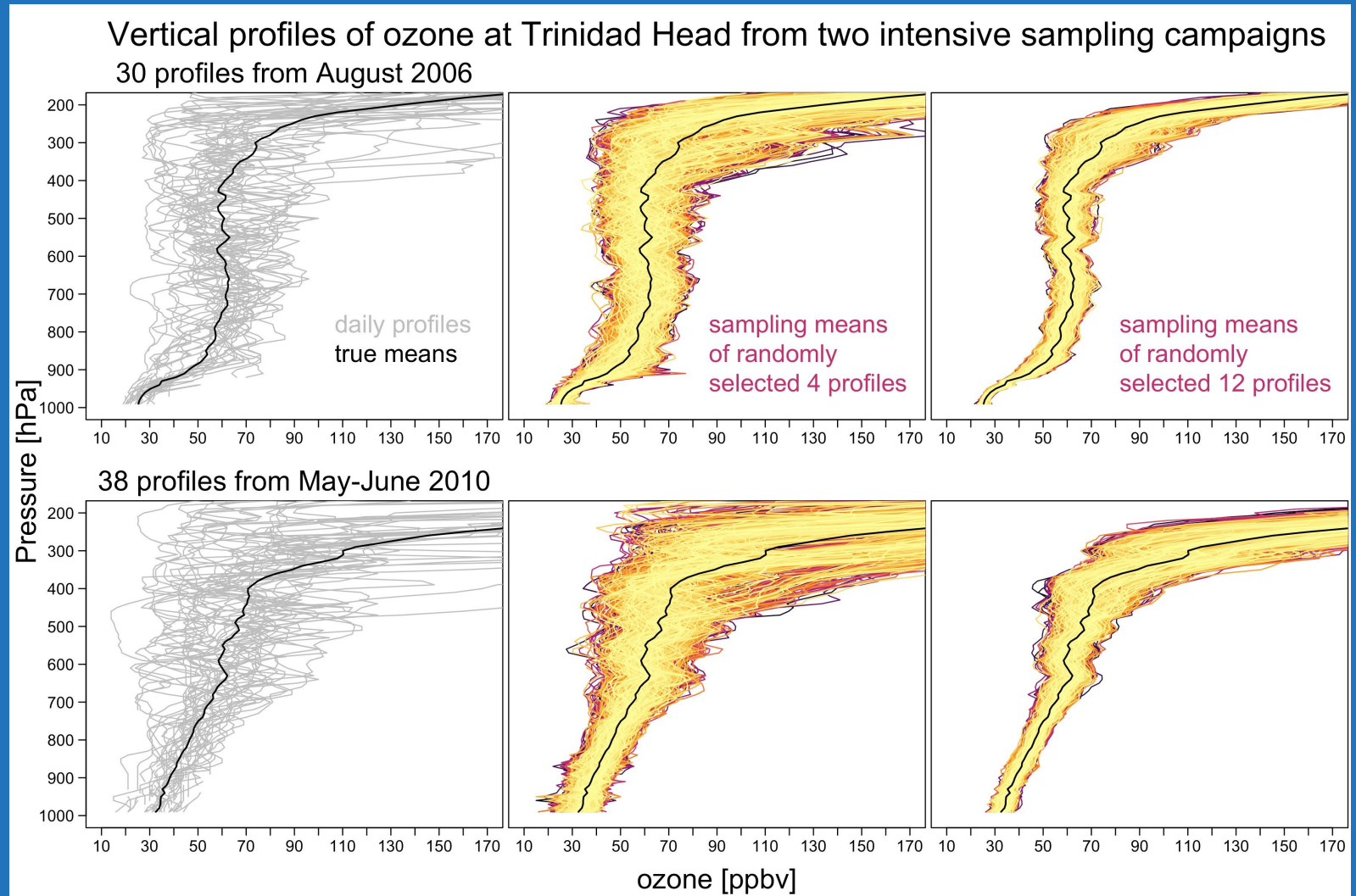
ACCEPTED FOR PUBLICATION  
21 October 2022

Arlene M Fiore<sup>1,2,\*</sup> , Sarah E Hancock<sup>3,12</sup>, Jean-François Lamarque<sup>4</sup>, Gustavo P Correa<sup>2</sup>, Kai-Lan Chang<sup>5,6</sup>, Muye Ru<sup>7,13</sup>, Owen Cooper<sup>5,6</sup>, Audrey Gaudel<sup>5,6</sup>, Lorenzo M Polvani<sup>2,8</sup> , Bastien Sauvage<sup>9</sup> and Jerry R Ziemke<sup>10,11</sup>

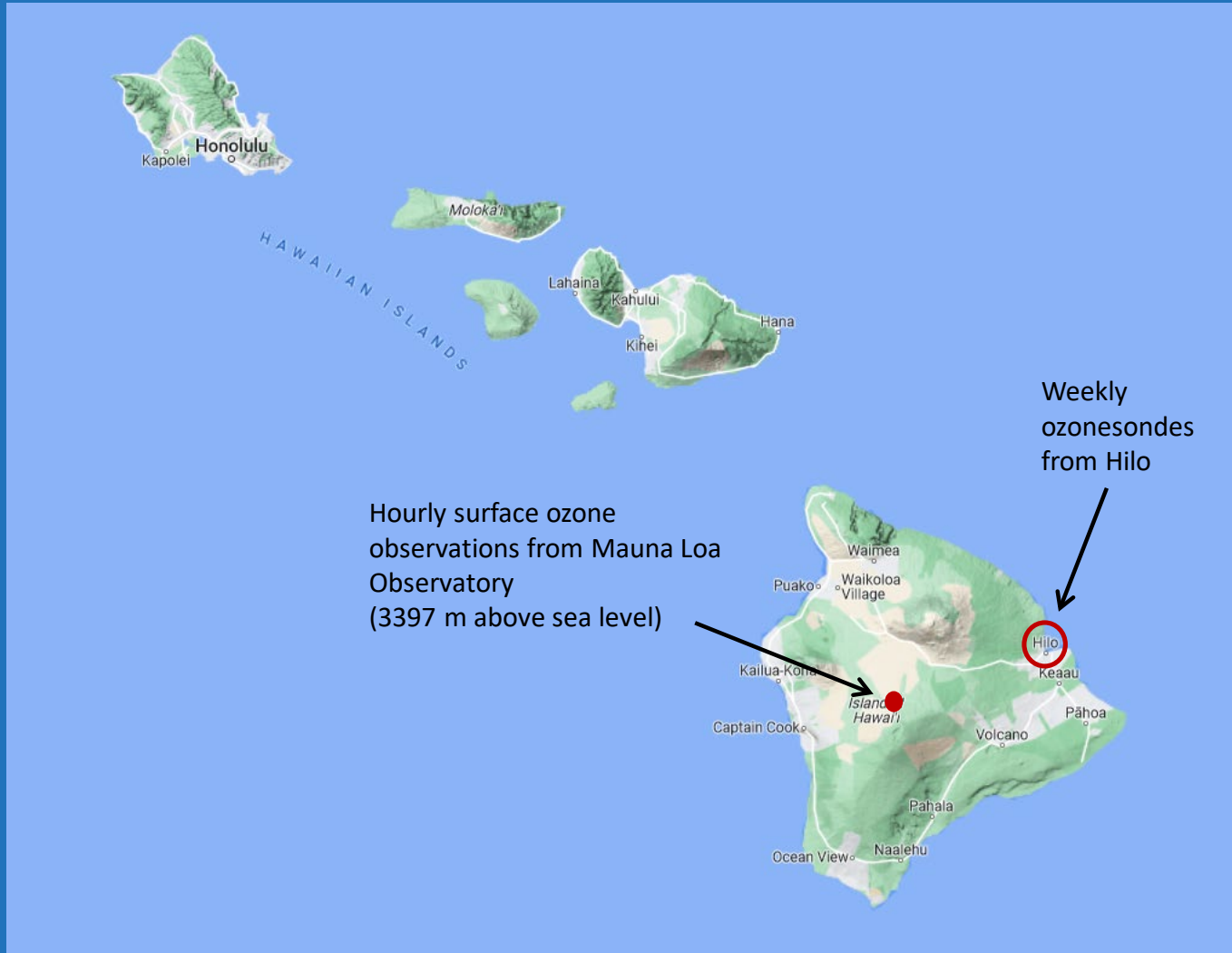
## Trinidad Head case studies

A sampling rate of once per week often misses the monthly mean by 10-20 ppbv.

A sampling rate of 3 times per week greatly reduces the uncertainty.



# NOAA GML ozone observations on the Big Island of Hawaii



# The Mauna Loa ozone record is ideal for developing free tropospheric sampling strategies

60 years ago scientists at Mauna Loa Observatory (MLO) determined that nighttime observations are representative of the lower free troposphere (*Price and Pales, 1963*)

OCTOBER–DECEMBER 1963

MONTHLY WEATHER REVIEW

## MAUNA LOA OBSERVATORY: THE FIRST FIVE YEARS

SAUL PRICE

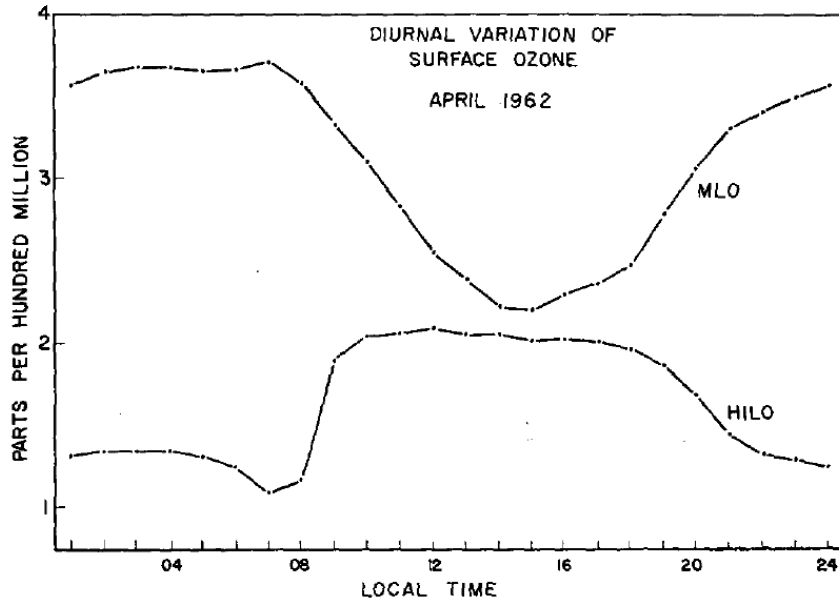
Pacific Supervisory Office, U.S. Weather Bureau, Honolulu, Hawaii

and

JACK C. PALES

Mauna Loa Observatory, U.S. Weather Bureau, Hawaii

[Manuscript received July 5, 1963; revised September 23, 1963]



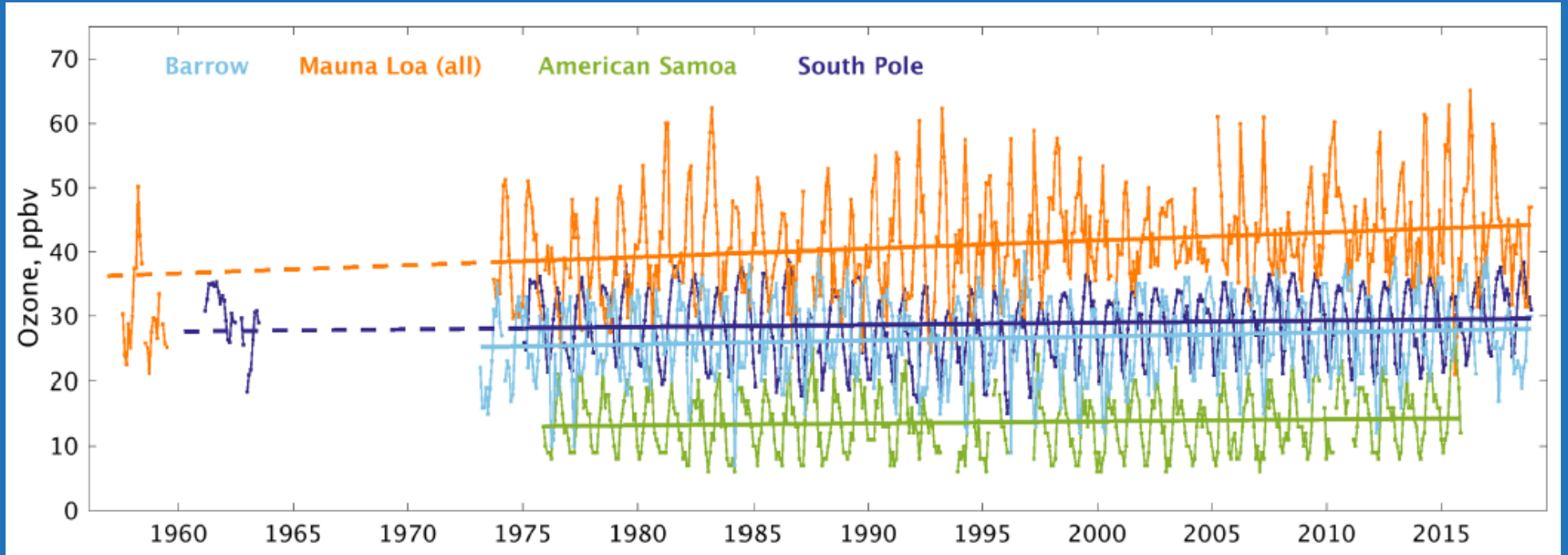
Ever since, ozone trends at MLO have been based on nighttime observations (*Oltmans et al., 2013, Tarasick and Galbally et al., 2019*).

Oltmans, S.J., et al., 2013. Recent tropospheric ozone changes—A pattern dominated by slow or no growth. *Atmospheric Environment*, 67, pp.331-351.

Tarasick, D. W., I. E. Galbally et al. (2019), Tropospheric Ozone Assessment Report: Tropospheric ozone from 1877 to 2016, observed levels, trends and uncertainties. *Elem Sci Anth*, 7(1), DOI: <http://doi.org/10.1525/elementa.376>

## Continuous surface ozone observations began at Mauna Loa in 1973

Additional reliable observations are available from the late 1950s



Nighttime ozone at MLO has increased by 16% since 1973 (~ 50% since the late 1950s)

Cooper, O. R., M. G. Schultz, S. Schröder, K.-L. Chang, A. Gaudel, G. Carbajal Benítez, E. Cuevas, M. Fröhlich, I. E. Galbally, D. Kubistin, X. Lu, A. McClure-Begley, S. Molloy, P. Nédélec, J. O'Brien, S. J. Oltmans, I. Petropavlovskikh, L. Ries, I. Senik, K. Sjöberg, S. Solberg, T. G. Spain, W. Spangl, M. Steinbacher, D. Tarasick, V. Thouret, X. Xu (2020), Multi-decadal surface ozone trends at globally distributed remote locations, *Elem Sci Anth*, 8(1), p.23. DOI: <http://doi.org/10.1525/elementa.420>

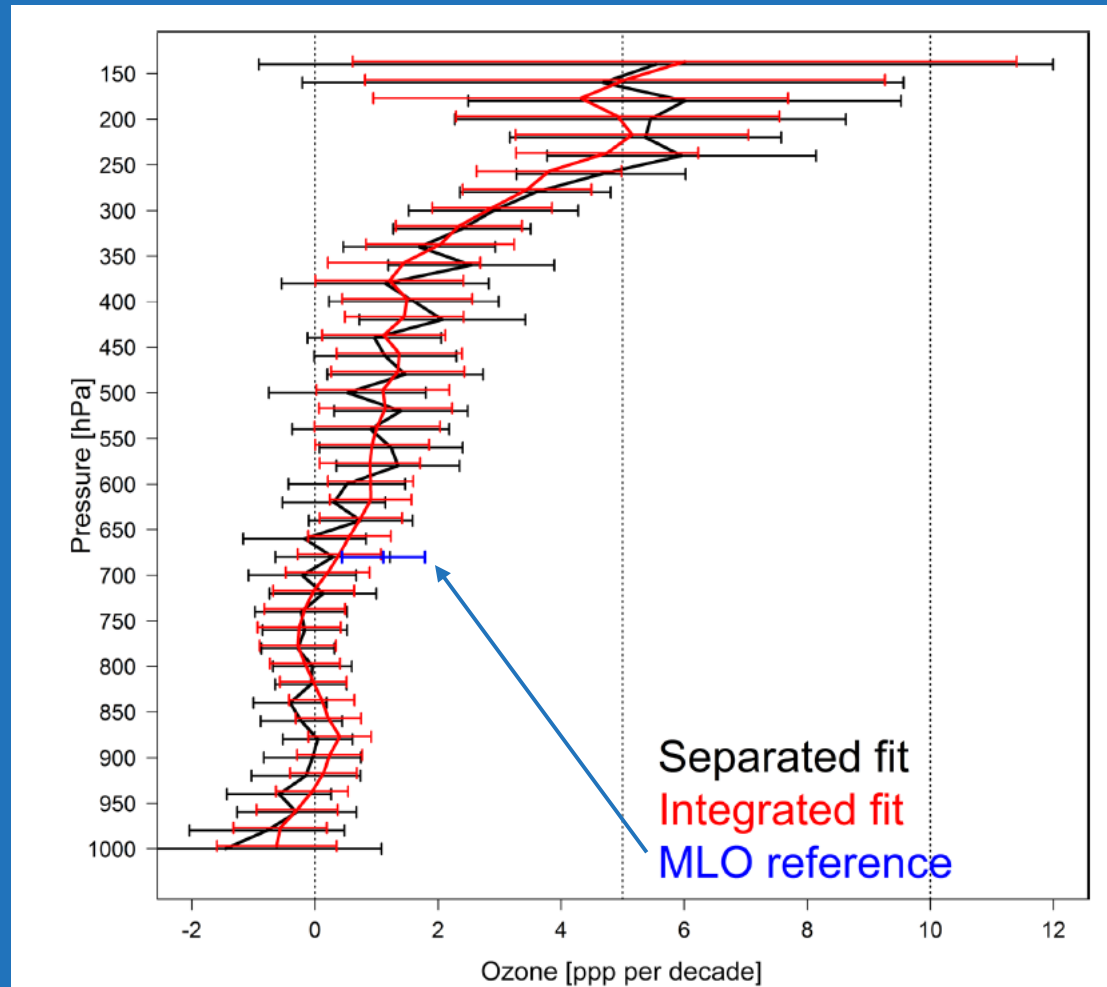
Weekly ozonesondes are launched from Hilo, Hawaii, ~60 km northeast of MLO.

This figure shows the ozone trends every 20 hPa above Hilo for the period 1982-2018 (*Chang et al., 2020*).

Trends are positive above 650 hPa.

A simple linear fit to the ozonesonde observations at 680 hPa shows almost no increase of ozone. In contrast the MLO surface ozone observations at the same altitude\* show a clear positive trend of  $1.2 \pm 0.6$  ppbv decade<sup>-1</sup>.

Given that the sampling frequency at MLO is 7 times greater than the sampling frequency at Hilo, we would expect the MLO trend to be more reliable.





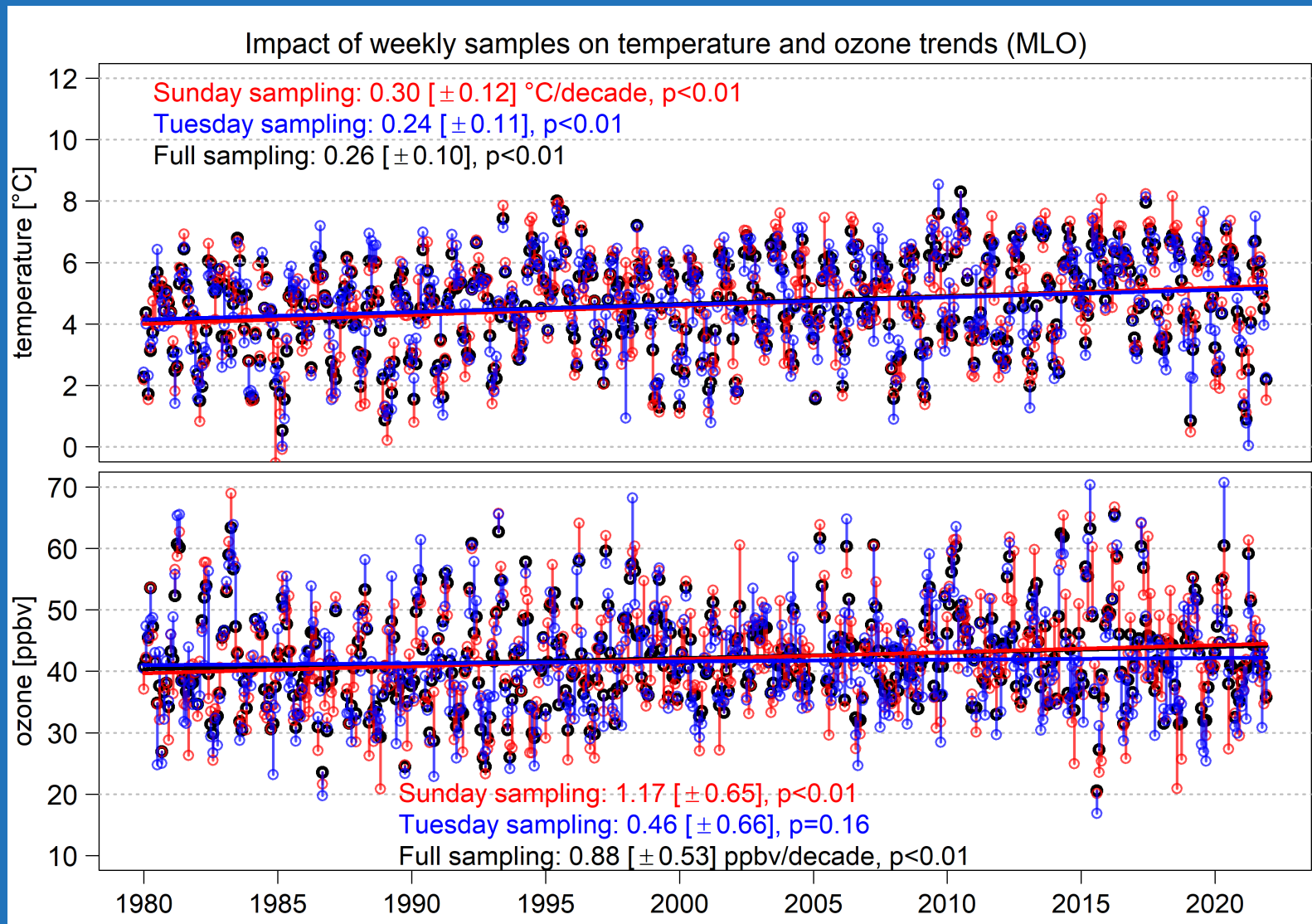
Ozone is much more variable than temperature at Mauna Loa

O<sub>3</sub> sampling strategy:

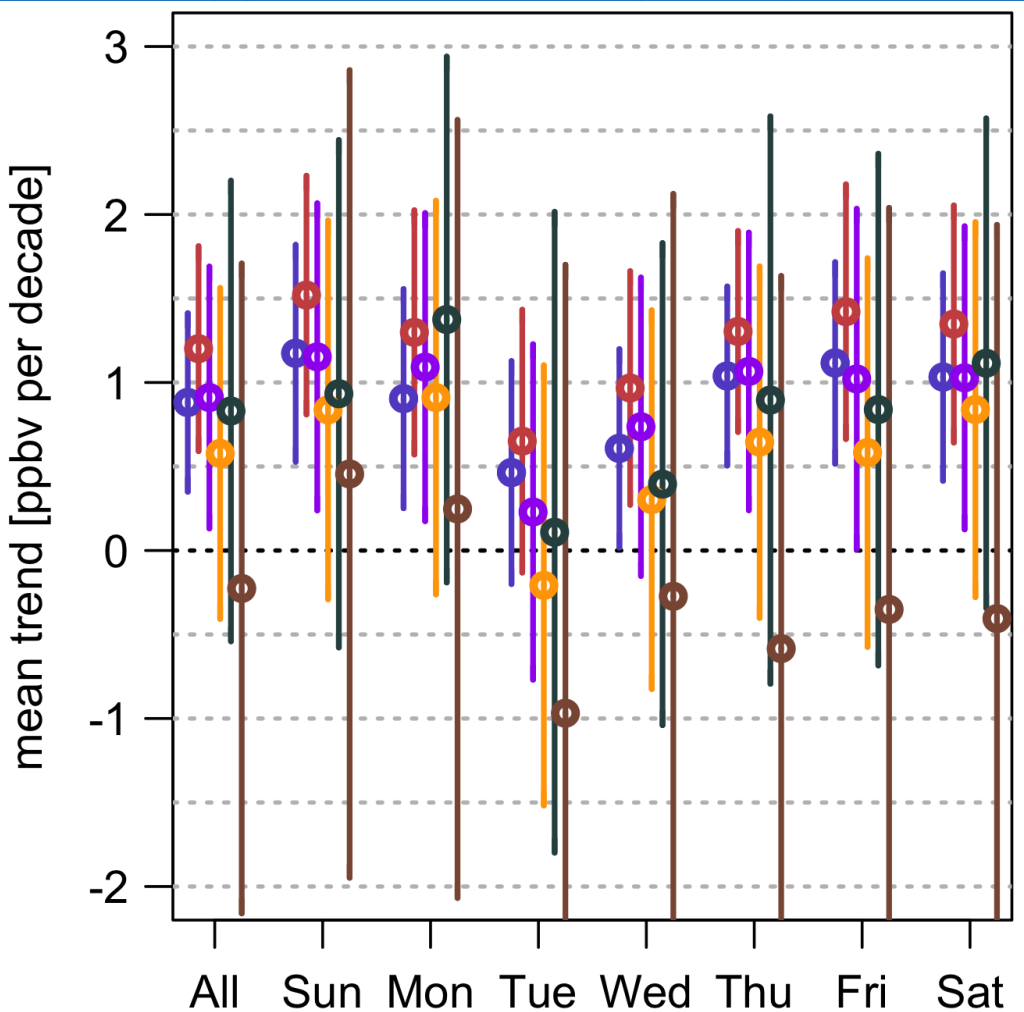
All days:  
 $0.9 \pm 0.5$  ppb/decade

Sundays:  
 $1.2 \pm 0.7$  ppb/decade

Tuesdays:  
 $0.5 \pm 0.7$  ppb/decade



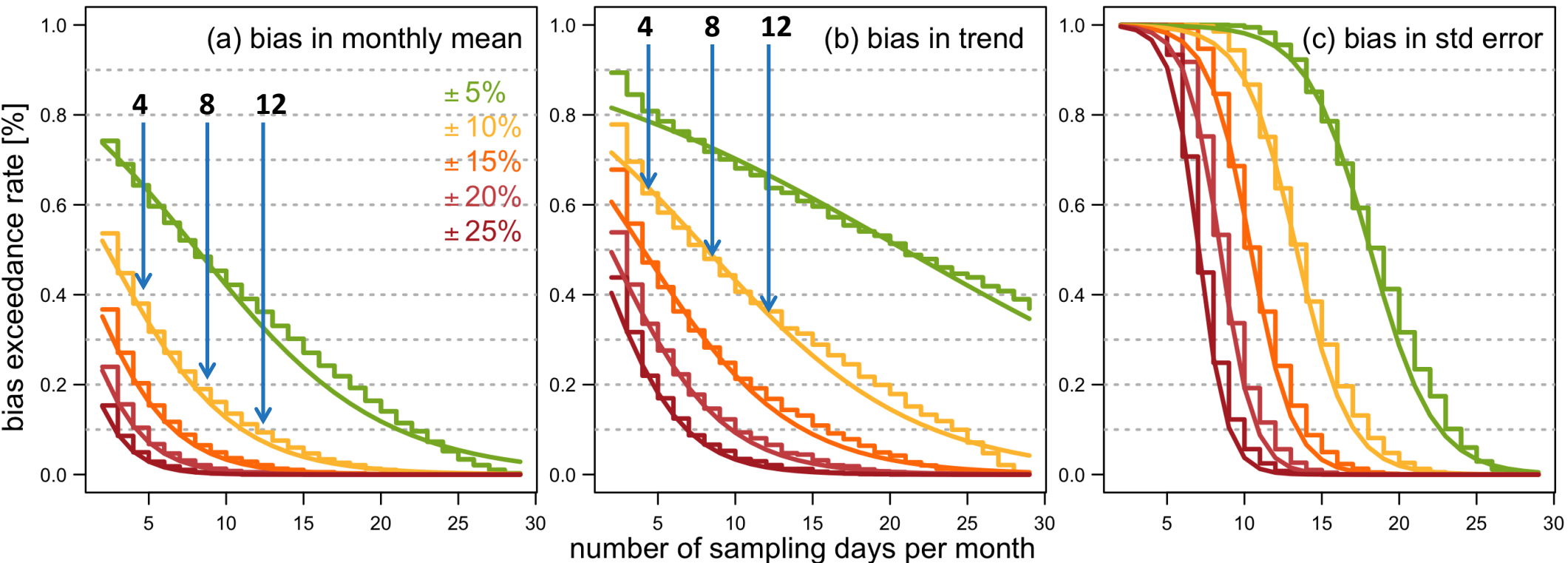
# Ozone trends at Mauna Loa by day of the week



1980-2021 1990-2021 2000-2021  
1985-2021 1995-2021 2005-2021

# Any increase in sampling frequency will improve trend detection

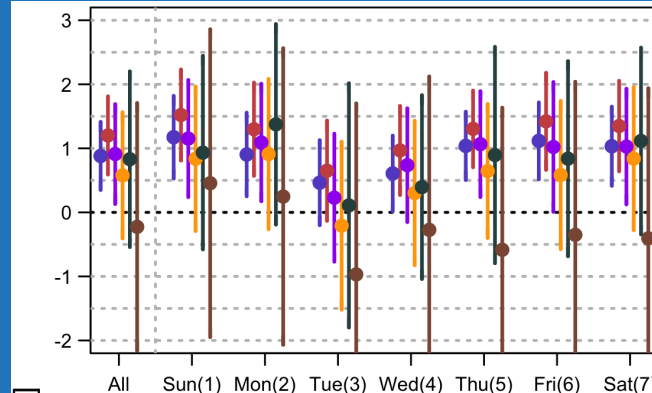
## Reduction of bias exceedance rate based on different thresholds



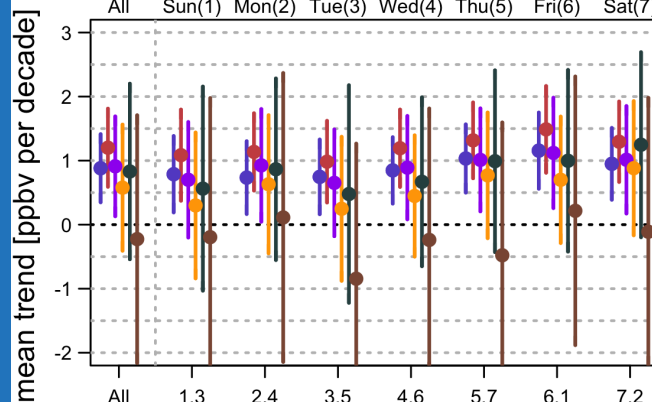
# Ozone trends at Mauna Loa by day of the week

Increased sampling reduces trend uncertainty

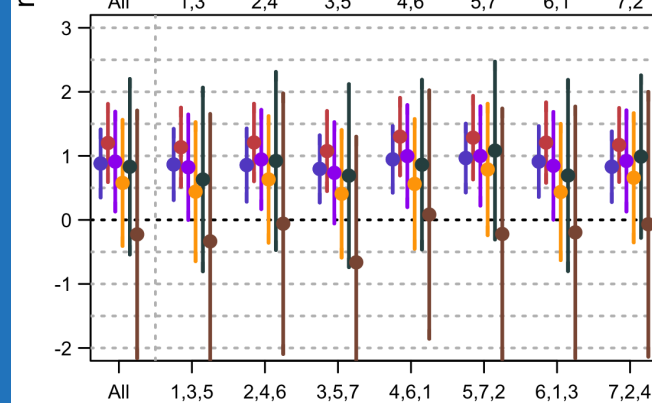
1980-2021 1990-2021 2000-2021  
1985-2021 1995-2021 2005-2021



1 profile per week



2 profiles per week



3 profiles per week

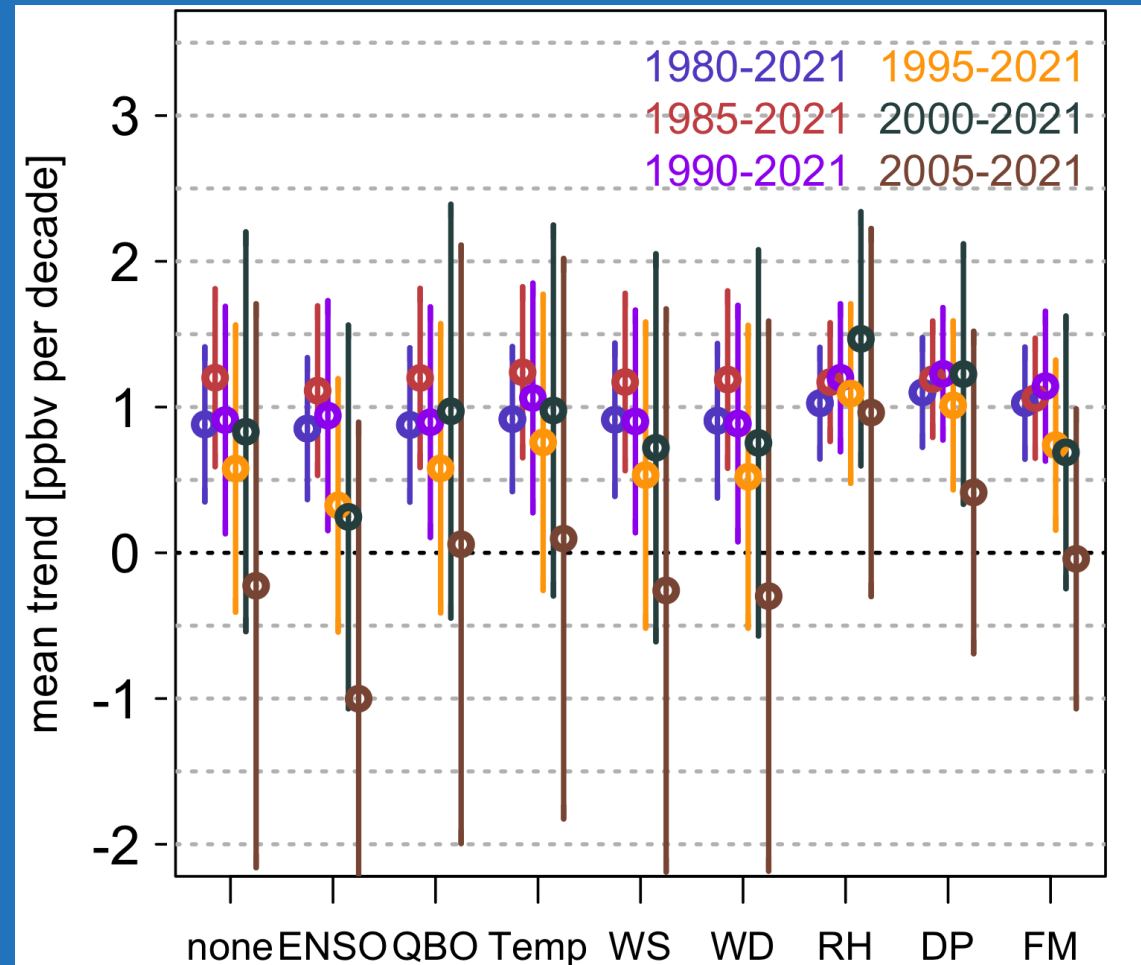
## Meteorological adjustments can reduce trend uncertainty

Multiple linear regression can remove interannual variability associated with:

1) Climate variability indicators such as ENSO or QBO

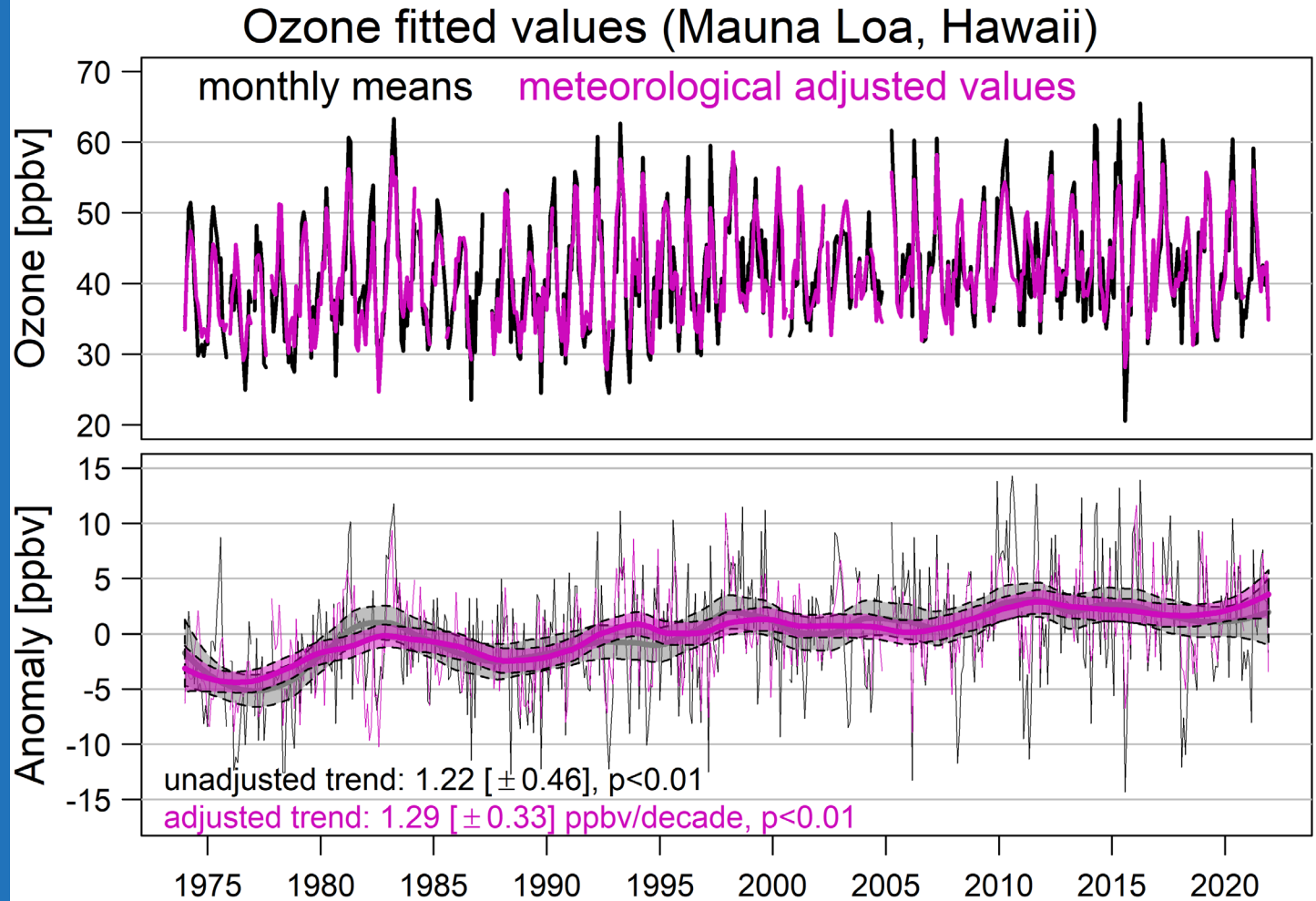
2) Daily/hourly variability of temperature, relative humidity, dewpoint, wind direction, wind speed

At Mauna Loa the only variables that improve the trend estimation are relative humidity and dewpoint



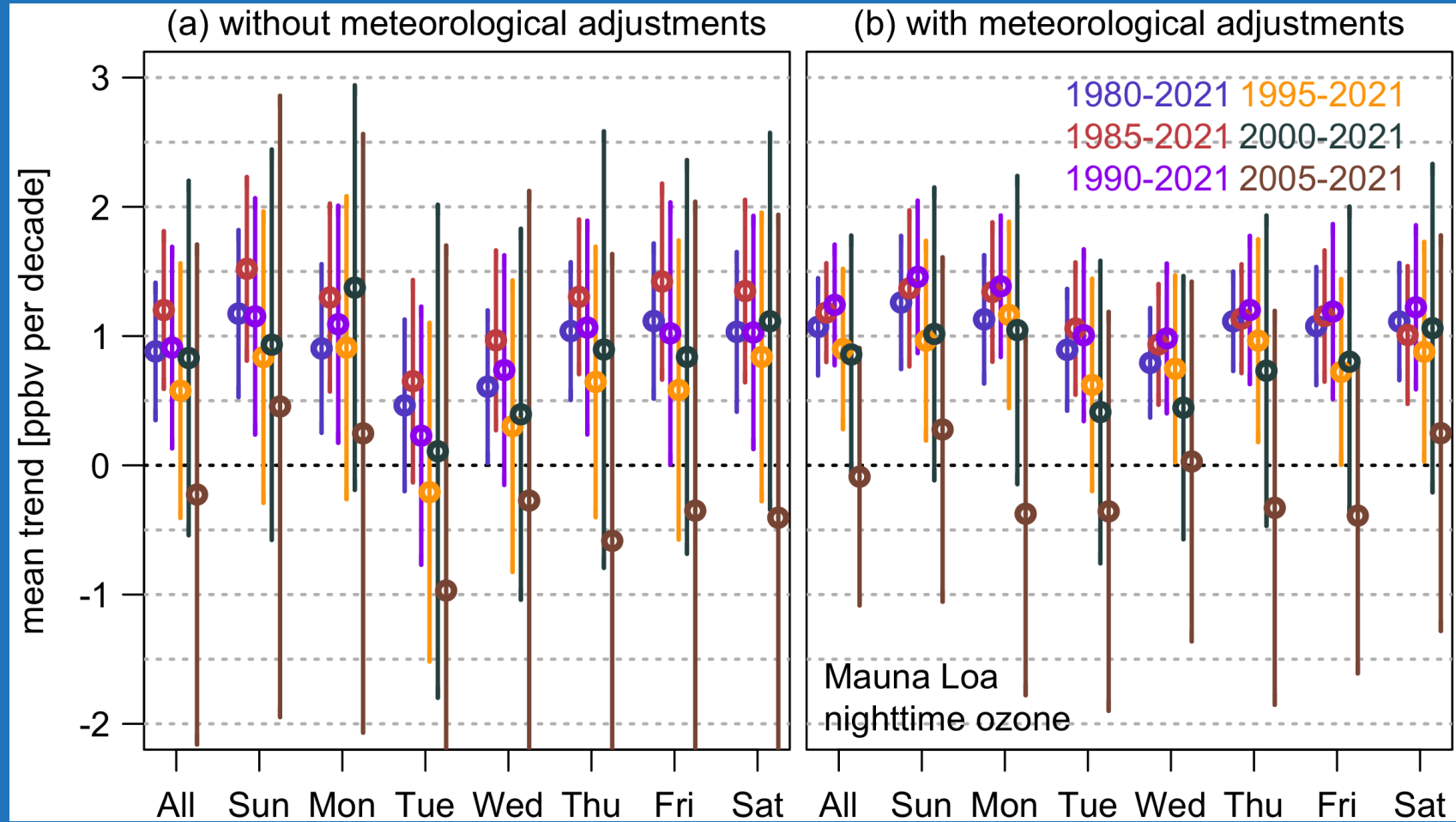
## Meteorological adjustments can reduce trend uncertainty

The meteorologically adjusted ozone trend shows the highest ozone values at Mauna Loa are found in the most recent year (2021).

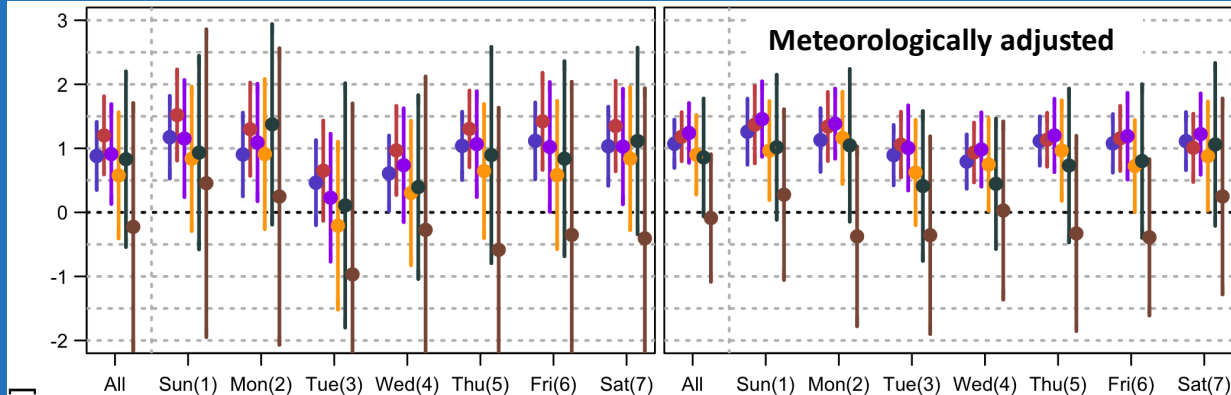


# Meteorological adjustments can reduce trend uncertainty

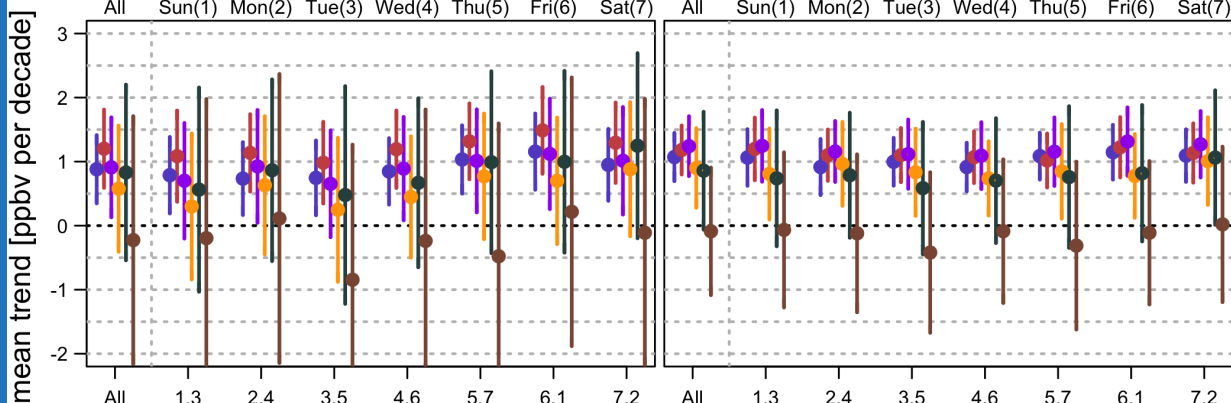
The accuracy of trends based on once per week sampling is improved when meteorology is accounted for.



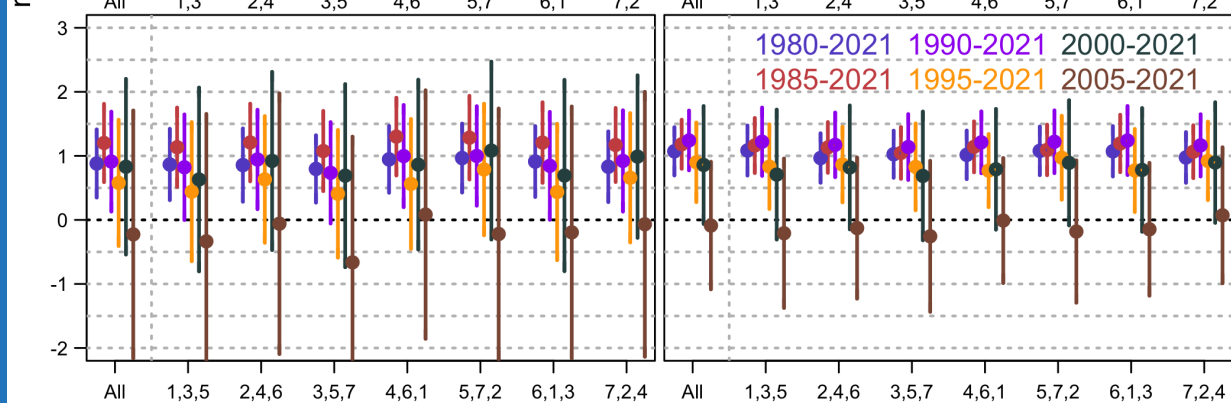
1 profile per week



2 profiles per week



3 profiles per week

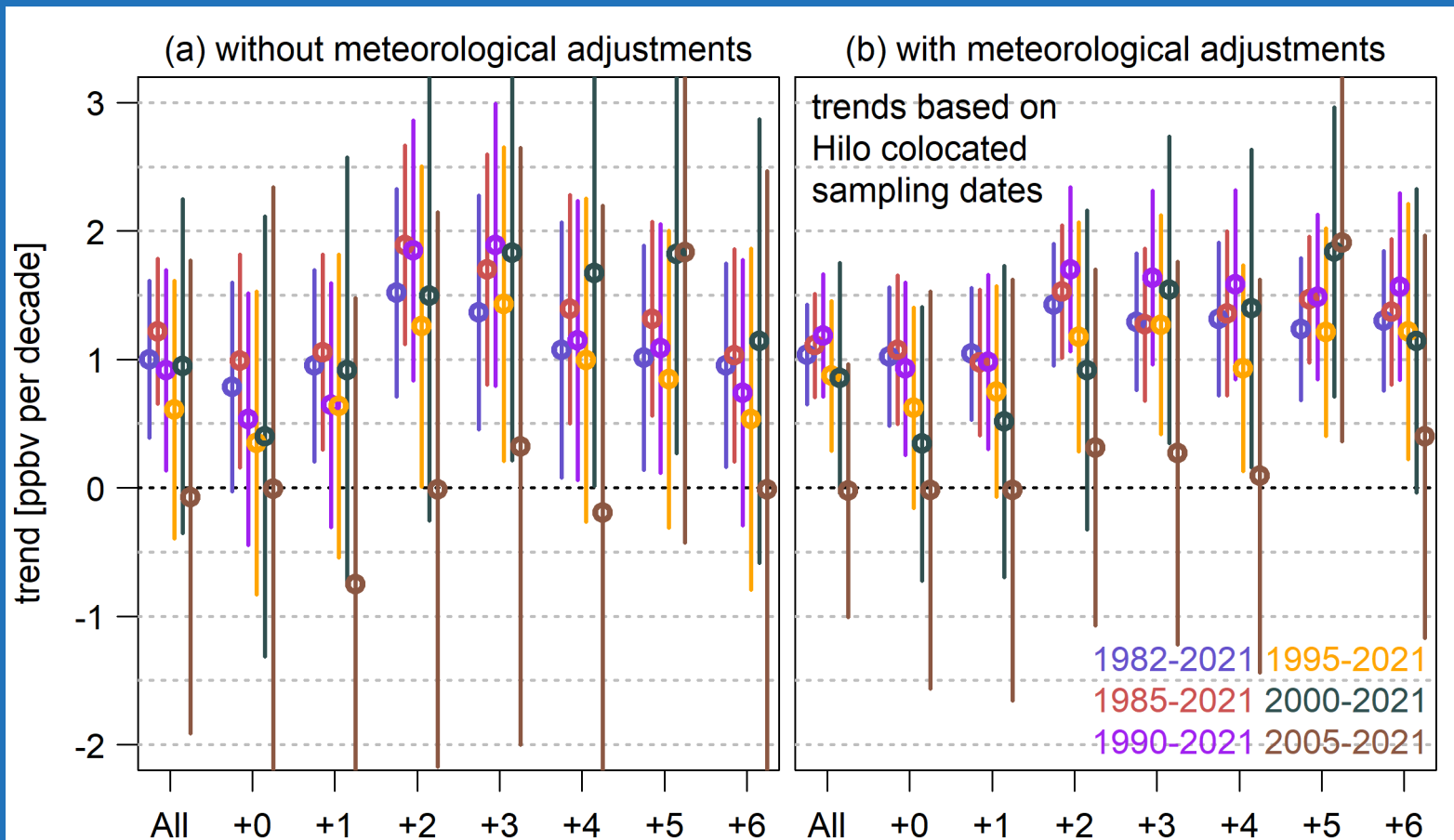




## Conclusions

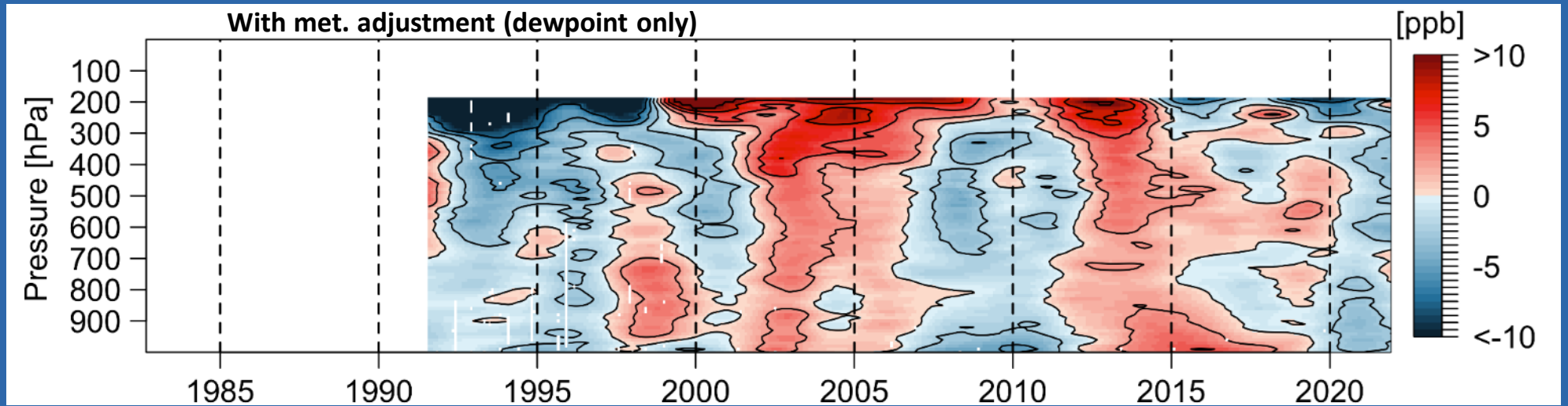
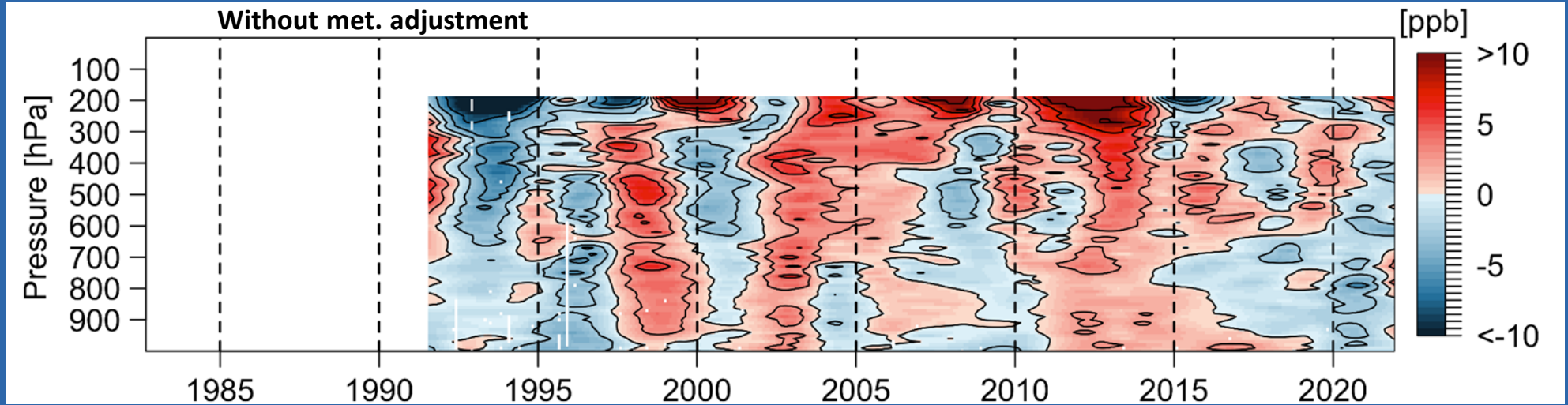
- 1) Any increase in sampling frequency will improve ozone trend detection in the free troposphere
- 2) Multiple linear regression further reduces trend uncertainty (co-located meteorological observations are particularly helpful)
- 3) Scientists and agencies monitoring free tropospheric ozone with ground based instruments need support if they are to enhance their sampling frequency





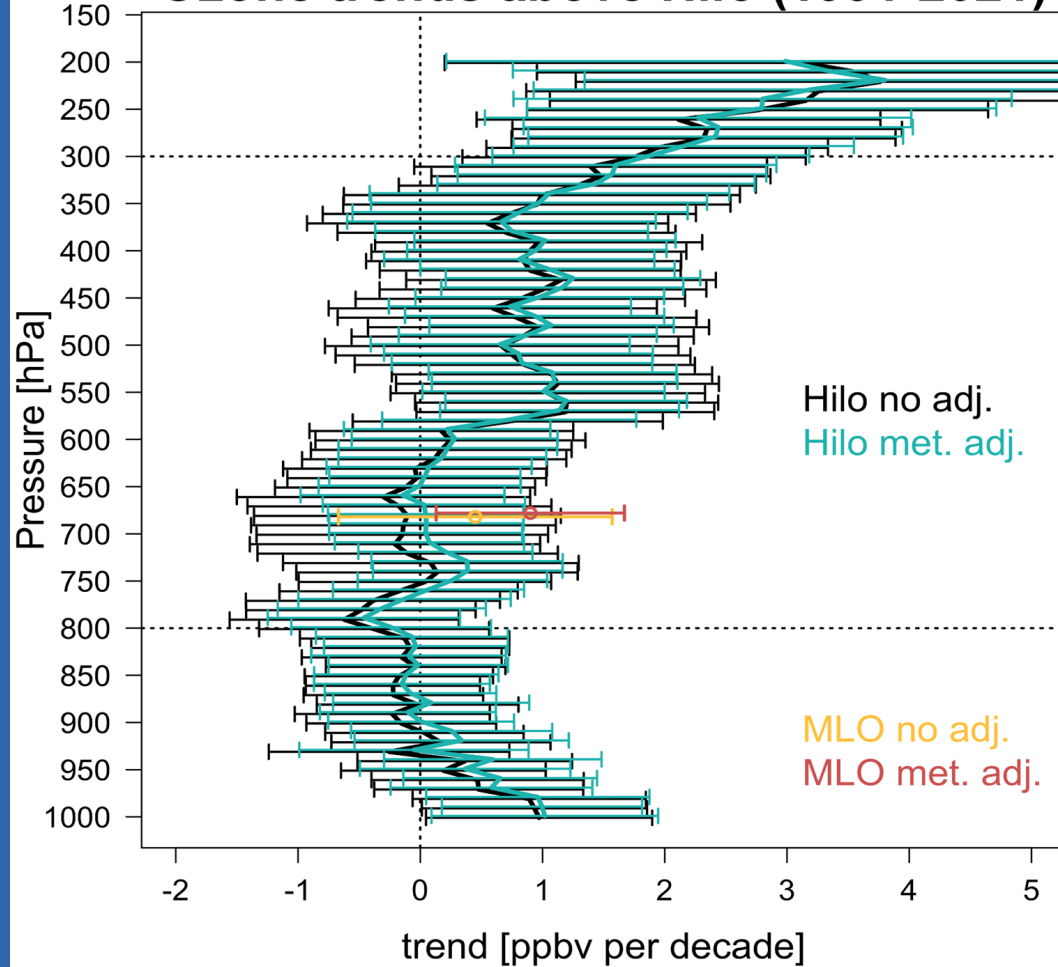
MLO ozone trends based on Hilo ozonesonde sampling dates (labeled as +0), where +1 indicates the trends based on data taken from one day after Hilo ozonesonde sampling dates, and so on.

# Ozone anomalies at Hilo

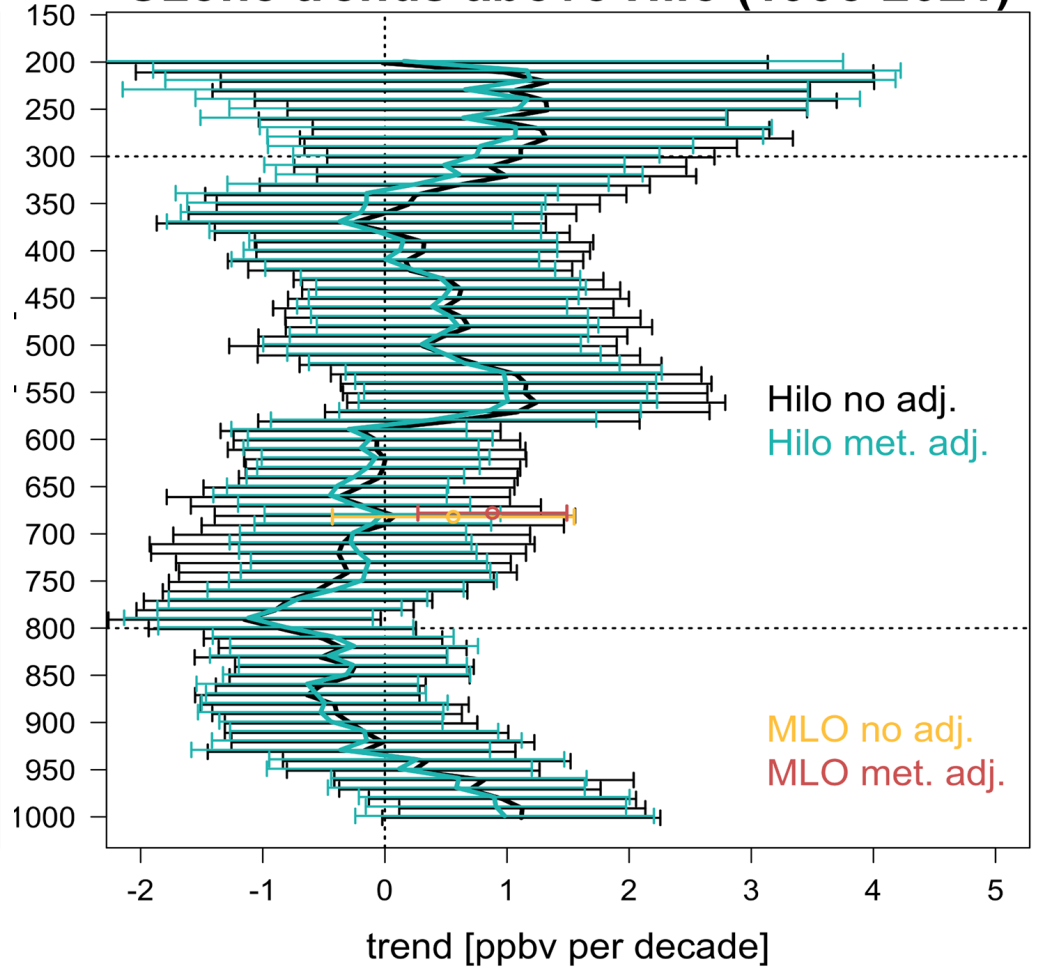


# Vertical ozone trend distribution above Hilo

## Ozone trends above Hilo (1991-2021)



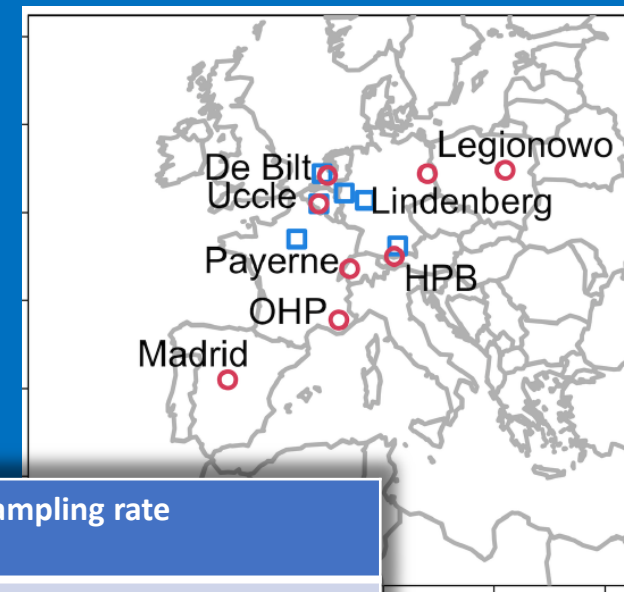
## Ozone trends above Hilo (1995-2021)



Chang et al. (2022) produced a regional-scale ozone trend for Western Europe based on IAGOS aircraft profiles and ozonesondes from several long-term monitoring sites.

The following slides illustrate the variability of ozone trends across five individual sites within an area of only 5x7 degrees.

This table shows that the trends range widely from  $+2.26 \pm 1.04$  ppbv decade<sup>-1</sup> at De Bilt, to  $-1.56 \pm 0.85$  ppbv decade<sup>-1</sup> at Payerne. The IAGOS trend falls in the middle of the range.



Site	Mid-tropospheric ozone trend (700-300 hPa) 1994-2019	Sampling rate
IAGOS (Frankfurt/Paris/Munich)	$1.16 \pm 0.77$ ppbv decade <sup>-1</sup> (p <0.01)	Daily aircraft profiles
Payerne, Switzerland	$-1.56 \pm 0.85$ ppbv decade <sup>-1</sup> (p <0.01)	3 sondes per week
Uccle, Belgium	$1.49 \pm 0.89$ ppbv decade <sup>-1</sup> (p <0.01)	3 sondes per week
Hohenpeissenberg, Germany	$-0.17 \pm 0.73$ ppbv decade <sup>-1</sup> (p =0.63)	3 sondes per week
De Bilt, The Netherlands	$2.26 \pm 1.04$ ppbv decade <sup>-1</sup> (p <0.01)	1 sonde per week

As reported in Table S-2 in the Supplement of Chang et al., 2022

Chang, K.-L., O. R. Cooper, A. Gaudel, M. Allaart, G. Ancellet, H. Clark, S. Godin-Beekmann, T. Leblanc, R. Van Malderen, P. Nédélec, I. Petropavlovskikh, W. Steinbrecht, R. Stübi, D. W. Tarasick, C. Torres (2022), Impact of the COVID-19 economic downturn on tropospheric ozone trends: an uncertainty weighted data synthesis for quantifying regional anomalies above western North America and Europe, AGU Advances, 3, e2021AV000542. <https://doi.org/10.1029/2021AV000542>

These curtain plots show the ozone variability above the 5 monitoring sites from 1994 to 2020 (from Figure S-5 of Chang et al., 2022). Each site has unique features in the mid-troposphere (700-300 hPa) that are not seen at the other sites (indicated by green boxes):

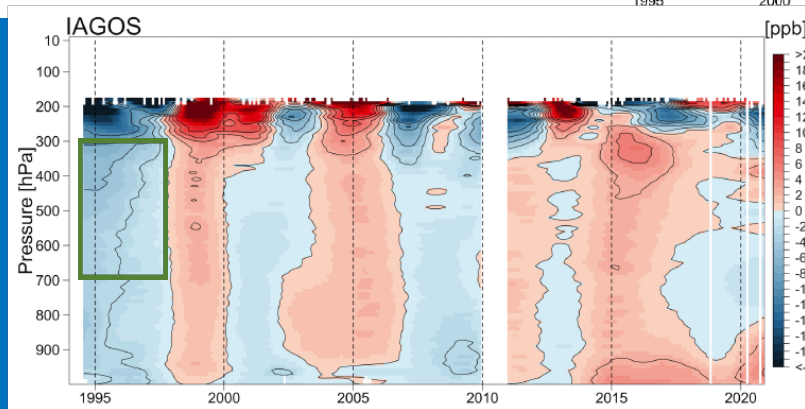
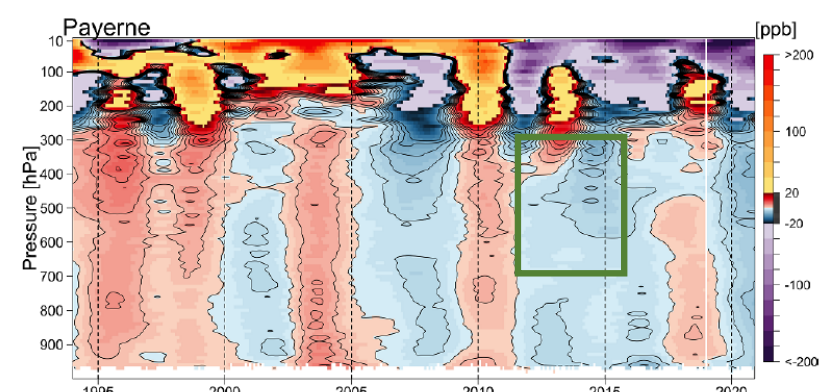
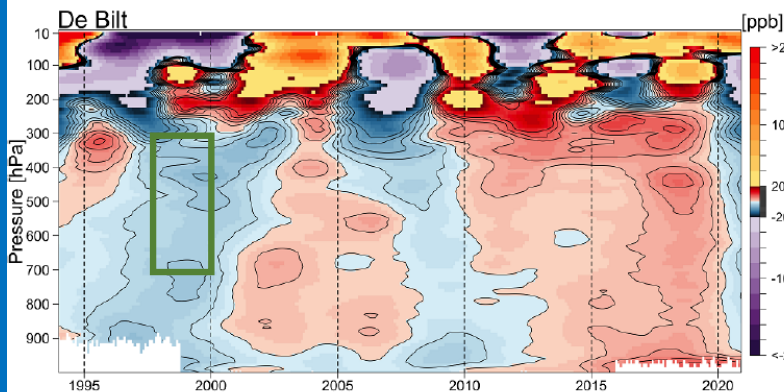
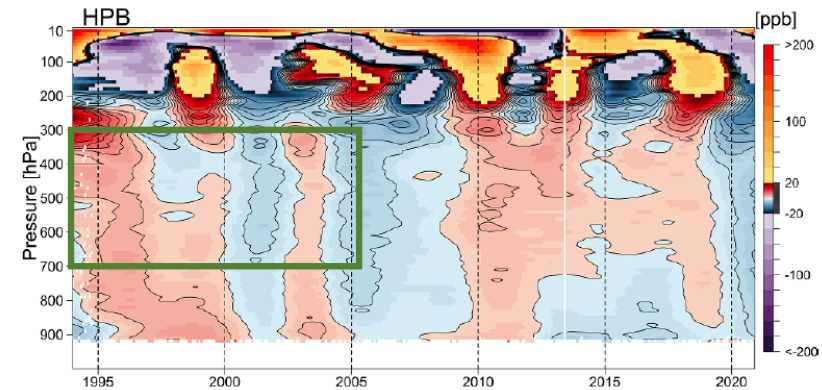
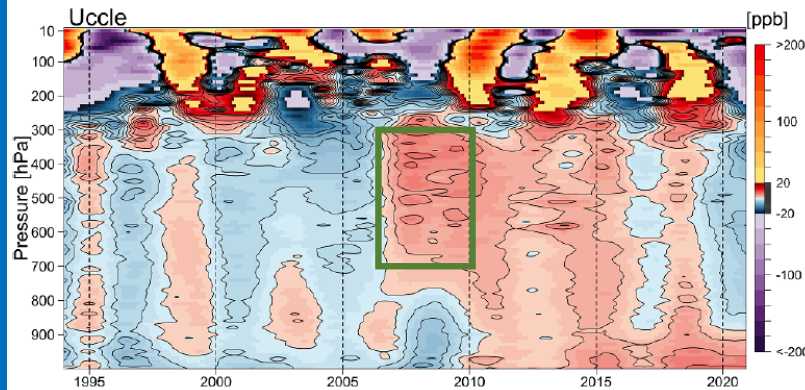
**Uccle:** strong enhancements in 2007-2009

**De Bilt:** most sites in the N. Hemisphere saw an ozone enhancement in 1998, but it does not appear at De Bilt

**IAGOS:** ozone is lower in 1994-1997 compared to HPB and Payerne

**Payerne:** ozone is much lower in 2012-2015 compared to the other sites

**HPB:** ozone decreased from 1994 to 2005, at a rate not seen at the other sites

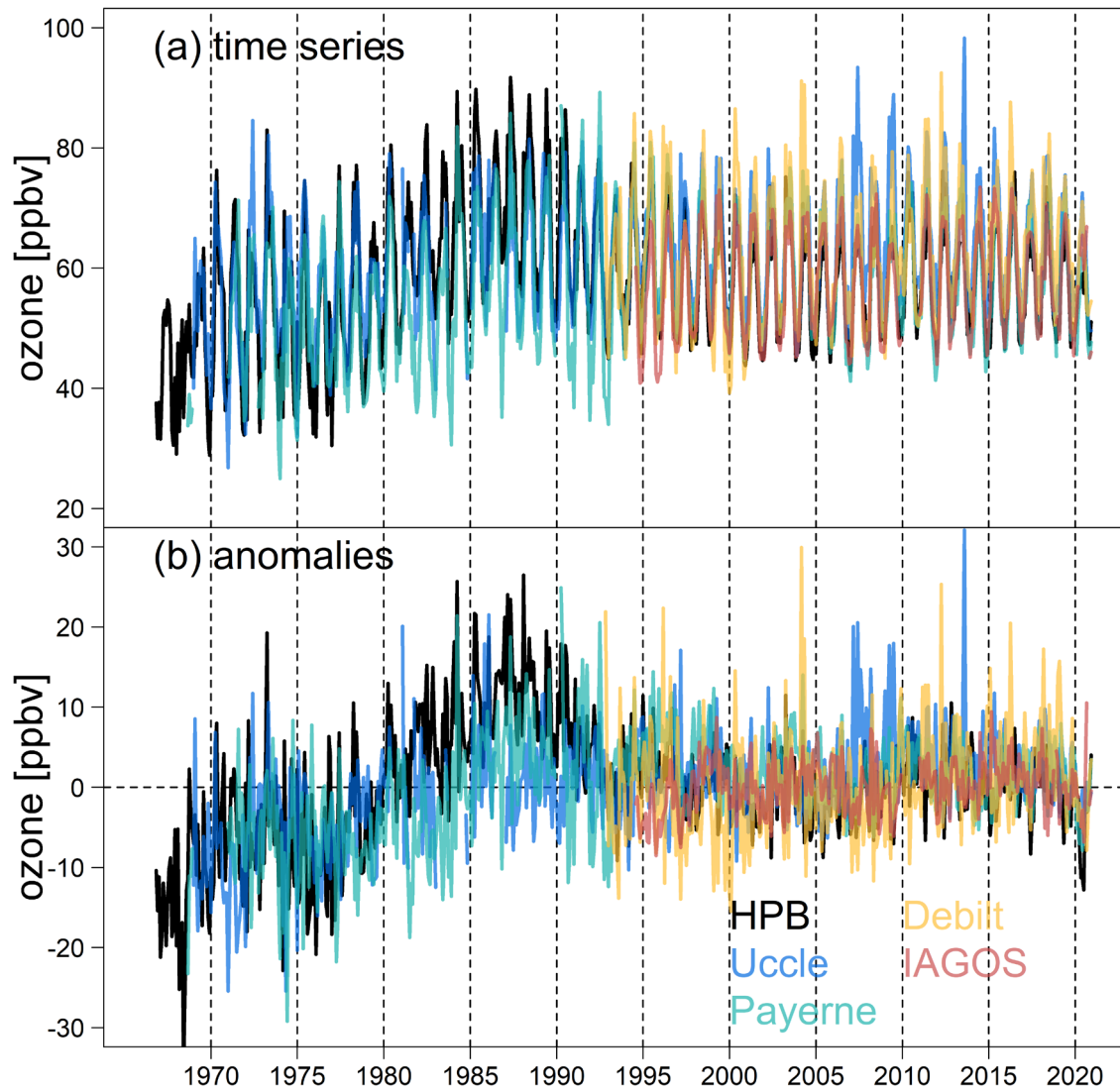


## Ozone variability across Western Europe

This figure shows every monthly mean ozone value in the mid-troposphere (700-300 hPa) for five ozone monitoring locations in western Europe, from the late 1960s through the end of 2020, when ozone dropped suddenly due to the COVID-19 economic slowdown.

Even though these locations are fairly close together (within 5x7 degrees) the ozone variability is large for any given month.

## Free tropospheric ozone above Europe

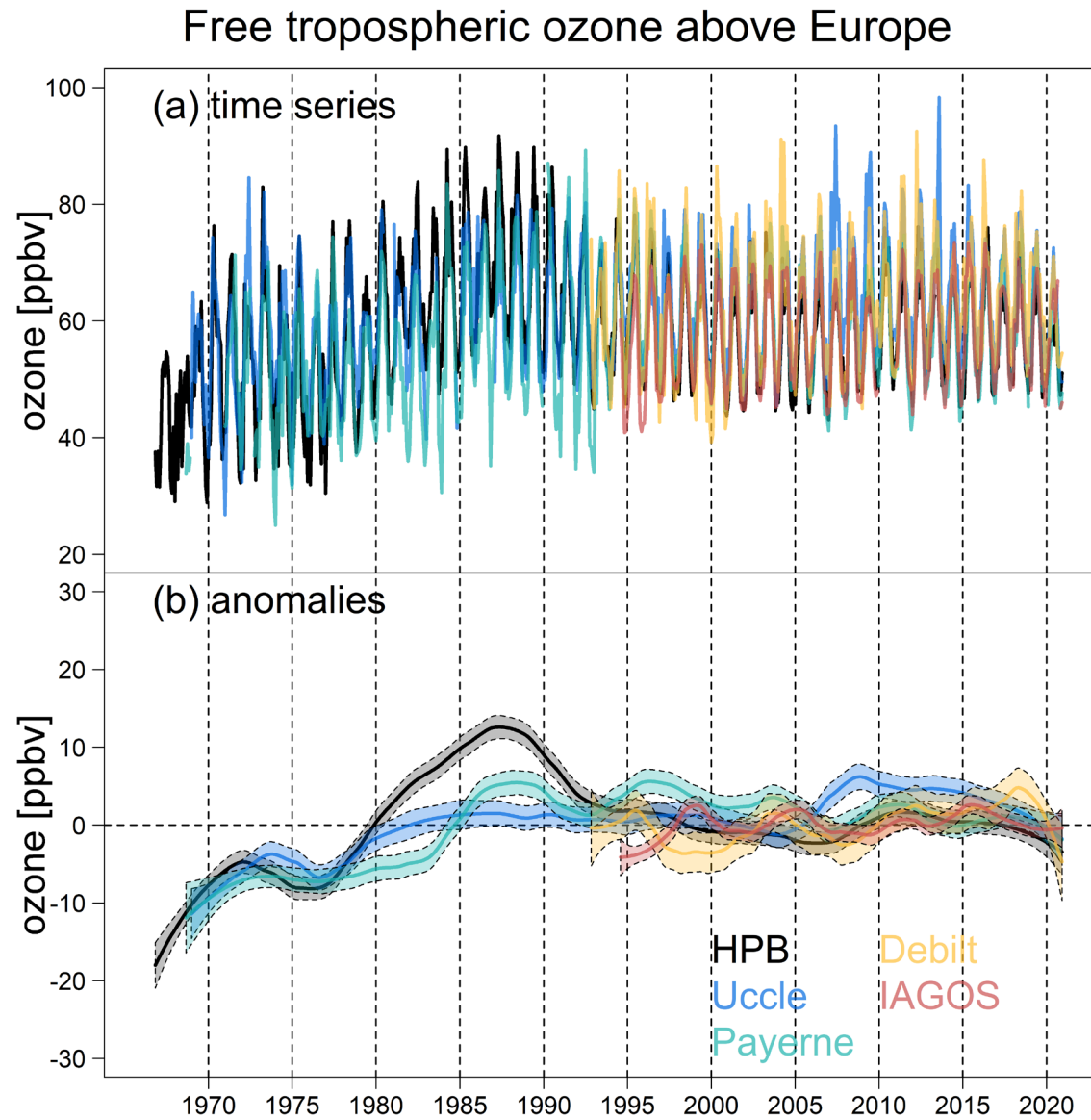




## Ozone variability across Western Europe

This figure shows every monthly mean ozone value in the mid-troposphere (700-300 hPa) for five ozone monitoring locations in western Europe, from the late 1960s through the end of 2020, when ozone dropped suddenly due to the COVID-19 economic slowdown.

Applying the Loess smoother to the monthly ozone anomalies allows us to see anomalies on longer timescales of 1-2 years. There are very few periods where all time series converge, and this plot illustrates the variability between the monitoring locations despite their proximity.



## Ozone variability across Western Europe

This is Chang et al.'s (2022) best estimate of ozone variability and trends above Western Europe based on all IAGOS and ozonesonde records (45,700 profiles):

$0.65 \pm 0.19$  ppbv decade<sup>-1</sup> ( $p < 0.01$ ), 1994-2019

$0.36 \pm 0.20$  ppbv decade<sup>-1</sup> ( $p < 0.01$ ), 1994-2020

