# Tropospheric ozone trends from harmonized ground-based measurements in TOAR-II & implications for stratospheric ozone trends assessment

## Roeland Van Malderen

Royal Meteorological Institute of Belgium, Brussels, Belgium



... but with contributions from many, many colleagues





## **Outline**

- Introduction: (tropospheric) ozone
- From TOAR-I to TOAR-II
- Harmonized ground-based measurements (HEGIFTOM)
- Tropospheric ozone column distribution (+ impact of COVID-19)
- Tropospheric ozone column trends
  - ✓ Individual site trends
  - ✓ Regionalized trends(+ impact of COVID-19)
- Conclusions and outlook
  - ✓ Tropospheric ozone trends
  - ✓ Implications for stratospheric ozone trends assessment



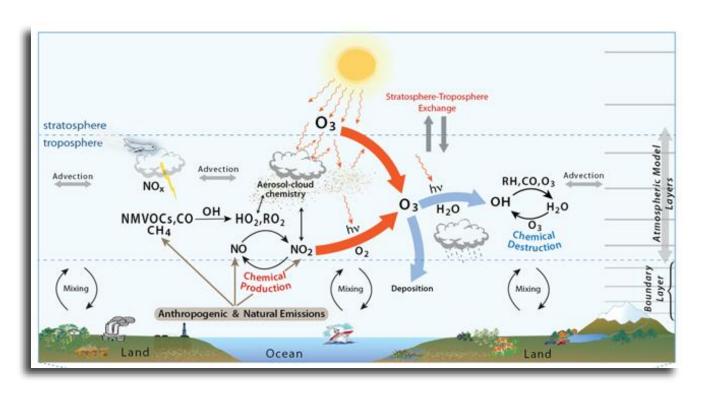




## **Tropospheric Ozone**



- 10% of atmospheric ozone
- harmful impact: toxic effects on humans (health), ecosystems and crops
- greenhouse gas (→ climate)
- formation/destruction of tropospheric ozone by
  - ✓ stratosphere-troposphere exchange
  - ✓ photochemical formation: sun + precursors (NO<sub>x</sub>, CO, CH<sub>4</sub> and VOC) coming from vehicule exhaust and industrial emissions
  - ✓ photochemical destruction in low NO<sub>x</sub> conditions (OH-HO<sub>2</sub> cycle)
  - ✓ dry deposition on the ground



From Young et al., Elementa, 2018



## **TOAR-I 2014-2019**



# **Tropospheric Ozone Assessment Report**

co-chairs: Owen Cooper (NOAA) & Martin Schultz (FZJ)

#### **Mission:**

To provide the research community with an up-to-date scientific assessment of tropospheric ozone's global distribution and trends from the surface to the tropopause.

#### **Deliverables:**

 The first tropospheric ozone assessment report based on all available surface observations, the peer-reviewed literature and new analyses.

#### **Stakeholders:**











Task Force on Hemispheric
Transport of Air Pollution



## **TOAR-I Publications**

# https:// collections.elementascience.org/toar



Young, PJ, et al. 2018 Tropospheric Ozone Assessment Report: Assessment of global-scale model performance for global and regional ozone distributions, variability, and trends. Elem Sci. Arth, 6: 10. DOI: https://doi.org/10.1525/elementa.265

#### REVIEW

Tropospheric Ozone Assessment Report: Assessment of global-scale model performance for global and regional ozone distributions, variability, and trends

P. J. Young\*\*\*, V. Naik\*, A. M. Fiorel\*\*, A. Gaudel\*\*\*†, J. Guo!, M. Y. Lins\*\*, J. L. Neu\*\*, D. D. Parrish\*\*\*\*, H. E. Rieder\*\*\*, J. L. Schnel!\*\*, S. Tilmes\*\*\*, O. Wild\*, L. Zhang\*\*\*, J. Ziemke\*\*\*\*, Brandt\*\*\*, A. Delcloof\*\*\*, R. M. Doherty\*\*\*\*, C. Geels\text{\text{M}}, M. I. Hegglin\*\*\*\*, L. Hu\text{\text{H}}\*\*, U. Im\text{\text{I}}\*\*, A. Luhar\text{\text{I}}\*\*, L. Murray\text{\text{\text{M}}\*\*}, D. Plummer\*\*\*\*\*, J. Rodriguez\text{\text{\text{R}}\*\*}, A. Saiz-Lopez\text{\text{\text{I}}\*\*}, M. G. Schultz\text{\text{\text{\text{I}}\*\*}\*\*, M. T. Woodhouse\text{\text{\text{I}}\*\*} and G. Zeng\text{\tex



Schultz, MG, et al 2017 Tropospheric Ozone Assessment Report: Database and metrics data of global surface ozone observations. *Elem Sci Anth*, 5: 58. DOI: https://doi.org/10.1525/elementa\_244

#### RESEARCH ARTICLE

Tropospheric Ozone Assessment Report: Database and metrics data of global surface ozone observations

Martin G. Schultz<sup>1,82</sup>, Sabine Schröder<sup>1</sup>, Olga Lyapina<sup>1</sup>, Owen R. Cooper<sup>2,3</sup>, Ian Galbally<sup>4</sup>, Irina Petropavlovskikh<sup>2,3</sup>, Erika von Schneidemesser<sup>5</sup>, Hiroshi Tanimoto<sup>6</sup>, Yasin Elshorbany<sup>7,8</sup>, Manish Naja<sup>9</sup>, Rodrigo J. Seguel<sup>10</sup>, Ute Dauert<sup>11</sup>, Paul Eckhardt<sup>12</sup>, Stefan Feigenspan<sup>11</sup>, Markus Fiebig<sup>12</sup>, Anne-Gunn Hjellbrekke<sup>12</sup>, You-Deog Hong<sup>13</sup>, Peter Christian Kjeld<sup>14</sup>, Hiroshi Koide<sup>15</sup>, Gary Lear<sup>16</sup>, David Tarasick<sup>17</sup>, Mikio Ueno<sup>15</sup>, Markus Wallaschi<sup>8</sup>, Darrel Baumgardner<sup>19</sup>, Ming-Tung Chuang<sup>20</sup>, Robert Gillett<sup>4</sup>, Meehye Lee<sup>21</sup>, Suzie Molloy<sup>4</sup>, Raeesa Moolla<sup>22</sup>, Tao Wang<sup>23</sup>, Katrina Sharps<sup>24</sup>, Jose A. Adame<sup>25</sup>, Gerard Ancellet<sup>26</sup>, Francesco Apadula<sup>27</sup>, Paulo Artaxo<sup>28</sup>, Maria E. Barlasina<sup>29</sup>, Magdalena Bogucka<sup>30</sup>, Paolo Bonasoni<sup>31</sup>, Limseok Chang<sup>32</sup>,



Archibald, A. T., et al. 2020. Tropospheric Ozone Assessment Report: A critical review of changes in the tropospheric ozone burden and budget from 1850 to 2100. Elem Sci Anth, 8: 1. DOI: https://doi.org/10.1525/elementa.2020.034

#### RESEARCH ARTICLE

Tropospheric Ozone Assessment Report: A critical review of changes in the tropospheric ozone burden and budget from 1850 to 2100

A. T. Archibald<sup>1,2,e</sup>, J. L. Neu<sup>3</sup>, Y. F. Elshorbany<sup>4</sup>, O. R. Cooper<sup>5,6</sup>, P. J. Young<sup>7,8,9</sup>, H. Akiyoshi<sup>10</sup>, R. A. Coxi, M. Coyle<sup>11,12</sup>, R. G. Derwent<sup>13</sup>, M. Deushi<sup>14</sup>, A. Finco<sup>15</sup>, G. J. Frost<sup>6</sup>, I. E. Galbally<sup>16,17</sup>, G. Gerosa<sup>15</sup>, C. Granier<sup>5,6,18</sup>, P. T. Griffiths<sup>1,2</sup>, R. Hossaini<sup>7,8</sup>, L. Hu<sup>19</sup>, P. Jöckel<sup>20</sup>, B. Josse<sup>21</sup>, M. Y. Lin<sup>22</sup>, M. Mertens<sup>20</sup>, O. Morgenstern<sup>23</sup>, M. Naja<sup>24</sup>, V. Naik<sup>25</sup>, S. Oltmans<sup>26</sup> D. A. Plummer<sup>27</sup>, L. E. Revell<sup>28</sup>, A. Saiz-Lopez<sup>29</sup>, P. Saxena<sup>30</sup>, Y. M. Shin<sup>1</sup>, I. Shahid<sup>31</sup>, D. Shallcross<sup>32</sup>, S. Tilmes<sup>33</sup>, T. Trickl<sup>34</sup>, T. J. Wallington<sup>35</sup>, T. Wang<sup>36</sup>, H. M. Worden<sup>33</sup>, and G. Zeng<sup>23</sup>



Lefohn, AS, et al. 2018 Tropospheric ozone assessment report: Global metrics for climate change, human health, and crop/ecosystem research Sci Anth, 6: 28. DOI: https://doi.org/10.1525/elementa.279

#### RESEARCH ARTICLE

Tropospheric ozone assessment report: Global ozone metrics for climate change, human health, and crop/ecosystem research

Allen S. Lefohn\*, Christopher S. Malley¹.‡.§, Luther Smithl\*, Benjamin Wells\*, Milan Hazucha\*\*, Heather Simon\*, Vaishali Naikt\*, Gina Mills\*†, Martin G. Schultzs\*§, Elena Paolettill\*, Alessandra De Marco\*\*, Xiaobin Xu\*\*\*, Li Zhang\*\*†, Tao Wang\*\*†, Howard S. Neufeld\*\*\*, Robert C. Musselmans\*§, David Tarasicklill\*, Michael Brauer\*\*\*, Zhaozhong Feng\*\*\*\*, Haoye Tang\*\*†\*, Kazuhiko Kobayashi\*\*\*\*, Pierre Sicard\*\*\*, Sverre Solberg\*\*\*, Sverre Solberg\*\*\*\*, Sverre Solberg\*\*\*, Sverre Solberg\*\*\*,



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

#### RESEARCH ARTICLE

Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation

A. Gaudel<sup>1,2</sup>, O. R. Cooper<sup>1,2</sup>, G. Ancellet<sup>3</sup>, B. Barret<sup>4</sup>, A. Boynard<sup>3,5</sup>, J. P. Burrows<sup>6</sup>, C. Clerbaux<sup>3</sup>, P.-F. Coheur<sup>7</sup>, J. Cuesta<sup>8</sup>, E. Cuevas<sup>9</sup>, S. Doniki<sup>7</sup>, G. Dufour<sup>8</sup>, F. Ebojie<sup>10</sup>, G. Foret<sup>8</sup>, O. Garcia<sup>11</sup>, M. J. Granados-Muñoz<sup>12,13</sup>, J. W. Hannigan<sup>14</sup>, F. Hase<sup>15</sup>, B. Hassler<sup>1,2,16</sup>, G. Huang<sup>17</sup>, D. Hurtmans<sup>7</sup>, D. Jaffe<sup>18,19</sup>, N. Jones<sup>20</sup>, P. Kalabokas<sup>21</sup>, B. Kerridge<sup>22</sup>, S. Kulawik<sup>2,3,24</sup>, B. Latter<sup>22</sup>, T. Leblanc<sup>12</sup>, E. Le Flochmoën<sup>4</sup>, W. Lin<sup>25</sup>, J. Liu<sup>26,27</sup>, X. Liu<sup>17</sup>, F. Mahieu<sup>27</sup>, A. McClure-Begley<sup>1,2</sup>, J. L. Neu<sup>23</sup>, M. Osman<sup>29</sup>, M. Palm<sup>6</sup>, H. Petetin<sup>4</sup>, I. Petropavlovskikh<sup>1,2</sup>, R. Querel<sup>28</sup>, N. Rahpoe<sup>23</sup>, A. Rozanov<sup>23</sup>, M. G. Schult<sup>23,13</sup>, J. Schwab<sup>3</sup>, R. Siddans<sup>22</sup>, D. Smale<sup>20</sup>, M. Steinbacher<sup>34</sup>, H. Tanimoto<sup>35</sup>, D. W. Tarasick<sup>36</sup>, V. Thouret<sup>4</sup>, A. M. Thompson<sup>37</sup>, T. Trickl<sup>38</sup>, E. Weatherhead<sup>1,2</sup>, C. Wespes<sup>39</sup>, H. M. Worden<sup>40</sup>, C. Vigouroux<sup>40</sup>, X. Xu<sup>41</sup>, G. Zeng<sup>20</sup>, J. Ziemke<sup>22</sup>



Tarasick, D, et al. 2019. Tropospheric Ozone Assessment Report: Troposone from 1877 to 2016, observed levels, trends and uncertainties. Anth, 7: 39. DOI: https://doi.org/10.1525/elementa.376

#### **REVIEW**

Tropospheric Ozone Assessment Report: Tropospheric ozone from 1877 to 2016, observed levels, trends and uncertainties

David Tarasick\*, Ian E. Galbally<sup>†,‡</sup>, Owen R. Cooper<sup>§,|</sup>, Martin G. Schultz<sup>†,</sup> Gerard Ancellet\*, Thierry Leblanc<sup>††,</sup> Timothy J. Wallington<sup>‡‡</sup>, Jerry Ziemke<sup>§§</sup>, Xiong L Martin Steinbacher<sup>††</sup>, Johannes Staehelin<sup>††</sup>, Corinne Vigouroux<sup>††,†</sup> James W. Hanniga Omaira García<sup>§§</sup>, Gilles Foret<sup>†</sup>, Prodromos Zanis<sup>††</sup>, Elizabeth Weatherhead<sup>§,|</sup>, Irina Petropavlovskikh<sup>§,|</sup>, Helen Worden<sup>‡†</sup>, Mohammed Osman<sup>\*\*\*†††,†††</sup>, Jane Liu<sup>§§§</sup>, Jahre Gaudel<sup>§,|</sup>, Meiyun Lin<sup>††</sup>, Waria Granados-Muñoz<sup>††††</sup>, Anne M. Thompson<sup>§§</sup>, Samuel J. Oltmans<sup>‡†††</sup>, Juan Cuesta<sup>|||</sup>, Gaelle Dufour<sup>|||</sup>, Valerie Thouret<sup>§§§§</sup>, Birgit Hassler<sup>|||||||</sup>, Thomas Trick<sup>††</sup>, Thomas Trick<sup>††</sup>, Neu<sup>†††</sup>



Fleming, ZL, et al. 2018 Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health. *Elem Sci Anth*, 6: 12. DOI: https://doi.org/10.1525/elementa.273



#### RESEARCH ARTICLE

## Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health

Zoë L. Fleming\*, Ruth M. Doherty¹, Erika von Schneidemesser‡, Christopher S. Malley⁵\*\*\*\*\*\*\*\*\*\*\*\*, Owen R. Cooper³³³³³³\*, Soeph P. Pinto¹³, Augustin Colette\*\*, Xiaobin Xu¹¹, David Simpson⁴\*\*\*\*\*\*, Martin G. Schultz⁵³³³³, Allen S. Lefohn⁵¹¹, Samera Hamad\*\*\*\*, Raeesa Moolla¹¹¹, Sverre Solberg‡‡ and Zhaozhong Feng<sup>666</sup>



Mills, G, et al. 2018. Tropospheric Ozone Assessment Report: Present-day tropospheric ozone distribution and trends relevant to vegetation. *Elem Sci Anth*, 6: 47. DOI: https://doi.org/10.1525/elementa.302

#### RESEARCH ARTICLE

# Tropospheric Ozone Assessment Report: Present-day tropospheric ozone distribution and trends relevant to vegetation

Gina Mills\*\*, Håkan Pleijel\*, Christopher S. Malley\*5.1, Baerbel Sinha\*, Owen R. Cooper\*\*, Martin G. Schultz\*†, Howard S. Neufeld\*†, David Simpson<sup>§5.1</sup>1, Katrina Sharps\*, Zhaozhong Feng\*\*1, Giacomo Gerosa\*\*\*, Harry Harmens\*, Kazuhiko Kobayashi\*\*1, Pallavi Saxena\*\*1, Elena Paoletti®, Vinayak Sinha\* and Xiaobin Xu<sup>|||||</sup>



Chang, K-L, et al 2017 Regional trend analysis of surface ozone observations from monitoring networks in eastern North America, Europe and East Asia. *Elem Sci Anth.* 5: 50, DOI: https://doi.org/10.1525/elementa.243

#### RESEARCH ARTICLE

Regional trend analysis of surface ozone observations from monitoring networks in eastern North America, Europe and East Asia

Kai-Lan Chang\*, Irina Petropavlovskikh\*†, Owen R. Cooper\*†, Martin G. Schultz† and Tao Wang§

Surface ozone is a greenhouse gas and pollutant detrimental to human health and crop and ecosystem productivity. The Tropospheric Ozone Assessment Report (TOAR) is designed to provide the research community with an up-to-date observation-based overview of tropospheric ozone's global distribution and trends. The TOAR Surface Ozone Database contains ozone metrics at thousands of monitoring sites



Xu, X, et al. 2020. Long-term changes of regional ozone in China: implications for human health and ecosystem impacts. *Elem Sci Anth*, 8: 13. DOI: https://doi.org/10.1525/elementa.409

#### RESEARCH ARTICLE

Long-term changes of regional ozone in China: implications for human health and ecosystem impacts

Xiaobin Xu\*, Weili Lint\*, Wanyun Xu\*, Junli Jint, Ying Wang\*, Gen Zhang\*, Xiaochun Zhangt, Zhiqiang Ma§, Yuanzhen Dongl, Qianli Ma¶, Dajiang Yu\*\*, Zou Litt, Dingding Wangtt and Huarong Zhao<sup>§§</sup>



## **TOAR-I 2014-2019**



# **Tropospheric Ozone Assessment Report**

co-chairs: Owen Cooper (NOAA) & Martin Schultz (FZJ)

#### **Mission:**

To provide the research community with an up-to-date scientific assessment of tropospheric ozone's global distribution and trends from the surface to the tropopause.

#### **Deliverables:**

- The first tropospheric ozone assessment report based on all available surface observations, the peer-reviewed literature and new analyses.
- 2) A database containing ozone exposure metrics at thousands of measurement sites around the world, freely accessible for research on the global-scale impact of ozone on climate, human health and crop/ecosystem productivity: https://toar-data.org/ → hosted by Jülich Supercomputing Center at FZJ!

#### **Stakeholders:**











Task Force on Hemispheric
Transport of Air Pollution



## **TOAR-I** Database key results



The first global-scale view of all available surface ozone observations

98<sup>th</sup> percentile

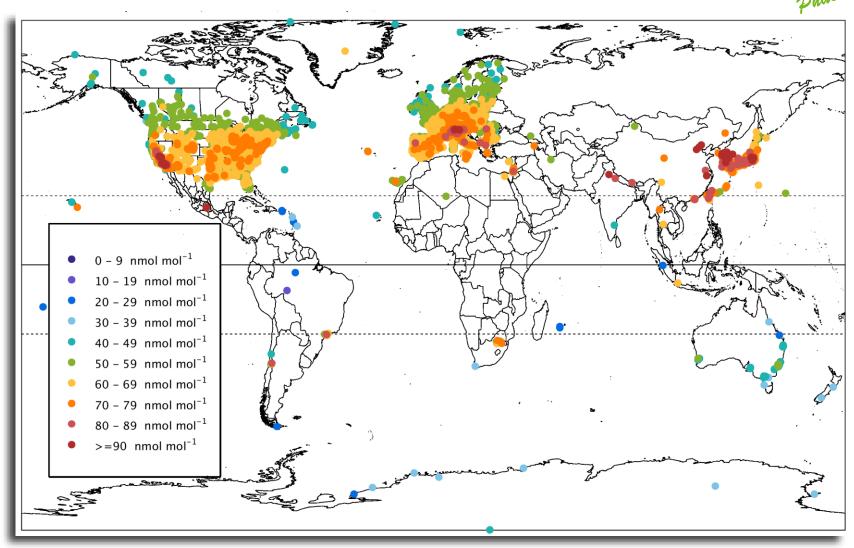
5-year average (2010-2014)

Summertime months

→ surface O<sub>3</sub> data harmonization: world's largest database of surface ozone observations, with ozone metrics and trends calculated consistently for all time series

https://toar-data.org/

https://toar-data.fz-juelich.de/



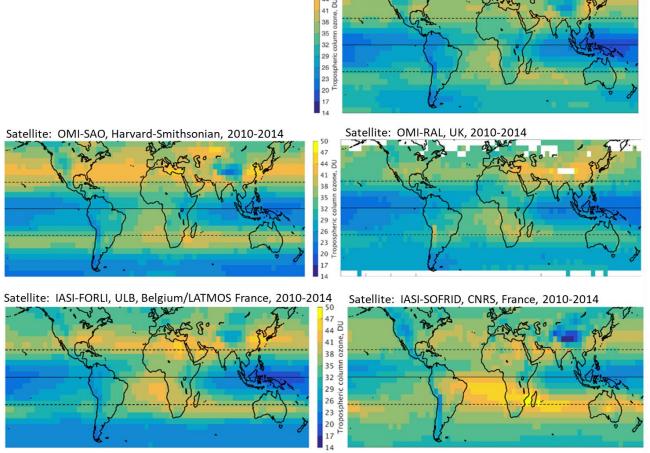


# **TOAR-I** key results



#### The first intercomparison of satellite ozone products

Satellite products generally agree regarding global tropospheric ozone hotspots.



Satellite: OMI/MLS, NASA, 2010-2014



## **TOAR-I** key results



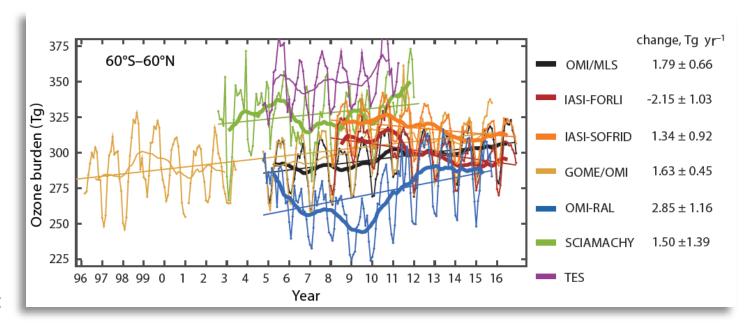
#### The first intercomparison of satellite ozone products

Satellite products generally agree regarding global tropospheric ozone hotspots.

Satellites and IPCC models report similar values for the tropospheric ozone burden.

However, the satellites disagree regarding trends over the past decade (2008-2016).

→ TOAR-I identified major discrepancies among the ozone trends reported by different satellite products: TOAR-II Satellite Ozone working group.



→ Tropospheric ozone trends from ground-based and in-situ techniques? TOAR-II GB working group



## **TOAR-II 2020-2026: Primary goals**



# Tropospheric Ozone Assessment Report, Phase II

co-chairs: Helen Worden (NCAR) & Martin Schultz (FZJ)

**TOAR Database:** Updated with all recent ozone observations worldwide; add ozone precursors and meteorological data.

**Final Product**: An observation-based assessment of tropospheric ozone's distribution and trends on regional, hemispheric and global scales (modelled after IPCC Working Group I)



**Impact studies**: will quantify the *impacts* of ozone on human health, vegetation and climate

(modelled after IPCC Working Group II)



## **TOAR-II Focus Working Groups**



New research is organized in independent Focus Working Groups:

**Africa** Focus Working Group

**Chemical Reanalysis** Focus Working Group

**East Asia** Focus Working Group

**Global and Regional Models** Focus Working Group

**HEGIFTOM** Focus Working Group

**Human Health Impacts of Ozone** Focus Working Group

**Machine Learning for Tropospheric Ozone** 

Ozone over the Oceans Focus Working Group

Ozone and Precursors in the Tropics (OPT) Focus Working Group

**Ozone Deposition** Focus Working Group

**Radiative Forcing Focus Working Group** 

**ROSTEES** Focus Working Group

Satellite Ozone Focus Working Group

**South Asia** Focus Working Group

**Statistics** Focus Working Group

Tropospheric Ozone Precursors (TOP) Focus Working Group

**Urban Ozone** Focus Working Group



# **TOAR-II Community Special Issue**

Focus Working Group findings submitted to the
 Community Special Issue before 30 Nov 2024
 = inter-journal special issue hosted by Copernicus
 https://acp.copernicus.org/articles/special issue1256.html

 Papers from this community SI (and others) will feed the TOAR-II Assessment Papers:

Health, Climate, Vegetation, STE,

Satellite, S. America, Africa,

Oceans

Geoscientific
Model Development

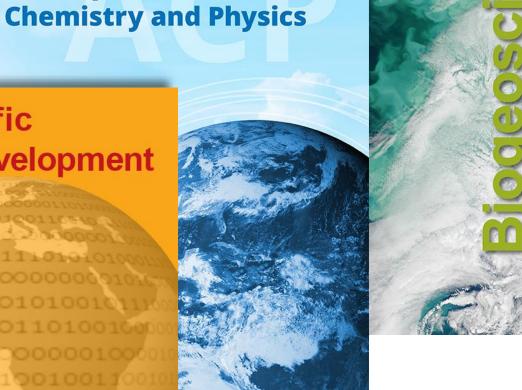
**Atmospheric** 

Atmospheric
Measurement
Techniques

Earth System Science

Data

The Data Publishing Journa



Copernicus Publications



## **TOAR-II Focus Working Groups**



New research is organized in 17 independent Focus Working Groups:

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## **TOAR-II Focus Working Group HEGIFTOM**



## Harmonization and Evaluation of Ground-based Instruments for Free Tropospheric

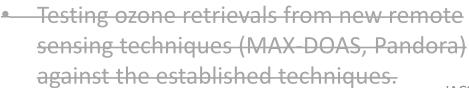
Ozone Measurements, chairs: Herman Smit & Roeland Van Malderen

#### **Key Objective:**

Evaluation and harmonization of the different free tropospheric ozone profiling datasets of the established measuring platforms (in-service aircraft, ozonesondes, Brewer/Dobson Umkehr, FTIR, Lidar).

#### **Major Deliverables:**

- Quality assessed ozone data sets, whereby each measurement gets also an <u>uncertainty</u> and a <u>quality flag</u>.
- Thereby, <u>representativeness</u> and <u>instrumen-</u> <u>tal drifts</u> will be characterized and evaluated.
- Assessment of tropospheric ozone trends.









Ozonesondes

Brewer/Dobson Umkehr







Lidar

MAX-DOAS & Pandora

http://hegiftom.meteo.be/datasets



## **HEGIFTOM:** Homogenized datasets - internal consistency



## **Achievements and updates:**

- IAGOS:
  - internal consistency paper published in AMT (Blot et al., <a href="https://doi.org/10.5194/amt-14-3935-2021">https://doi.org/10.5194/amt-14-3935-2021</a>),
  - o simulation chamber comparison of IAGOS-CORE UV-photometer and reference photometer for ozonesondes (Smit et al., 2025, AMT, TOAR-II SI, <a href="https://doi.org/10.5194/egusphere-2024-3760">https://doi.org/10.5194/egusphere-2024-3760</a>)
- Lidar: TMF data has been updated with new data processor, OHP will follow
- FTIR: flagging applied to the NDACC data
- Brewer/Dobson Umkehr:
  - 6 Dobson Umkehr sites have been homogenized (Petropavlovskikh et al., <a href="https://doi.org/10.5194/amt-15-1849-2022">https://doi.org/10.5194/amt-15-1849-2022</a>)
  - Updated uncertainty estimation of the retrievals.
- ozonesondes:
  - 12 more sites homogenized, e.g. OHP, Lauder, Arctic sites (10-15/55 remaining)
  - homogenized data available on ftp-server



# **HEGIFTOM:** Homogenized datasets



**Deliverable:** Homogenized free tropospheric ozone profile data, described at HEGIFTOM website, with same template for each dataset:

## **Availability**

location (ftp, data archive, website, doi, e-mail address contact person, etc.).

#### **Data field description**

Measured data fields (and their units), incl. auxiliary data fields, available metadata. Data format

### **Description of homogenization procedure**

short description of the steps taken to make the dataset (more) homogeneous within the network.

#### **Data management**

- Flagging
- Uncertainties
- Traceability
- Internal consistency
- External consistency
- Data quality indicators
- List of homogenized sites (name, geographical location, period of observations)

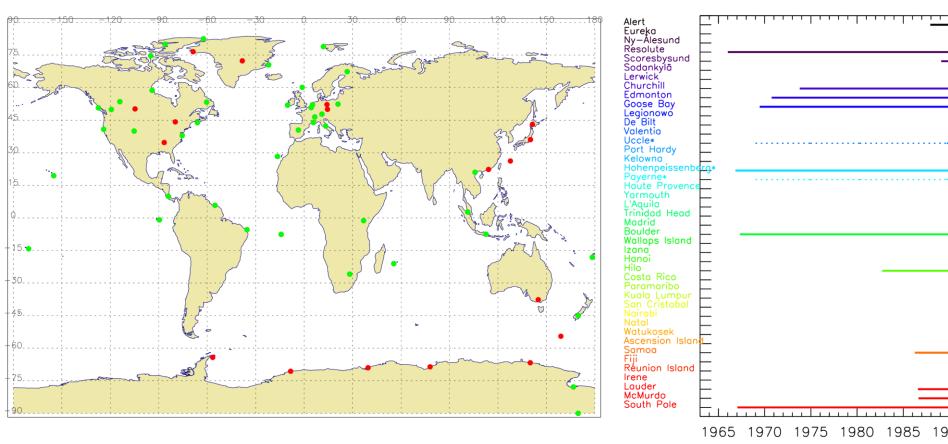
https://hegiftom.meteo.be/datasets

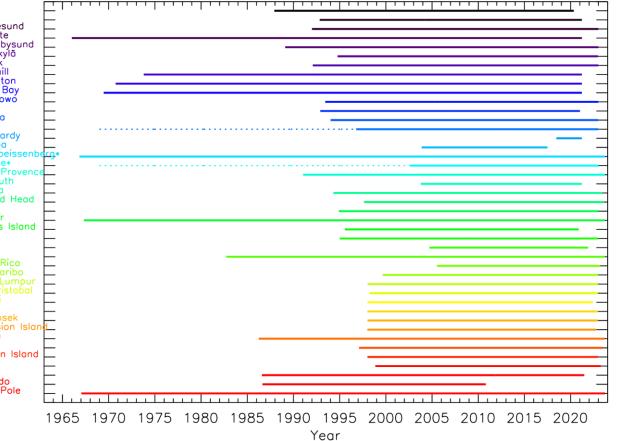


## **HEGIFTOM:** Homogenized datasets



## **Ozonesondes**





- 43 sites (green dots) with homogenized ozone profile data
- Profile data available at ftp-server

https://hegiftom.meteo.be/datasets/ozonesondes



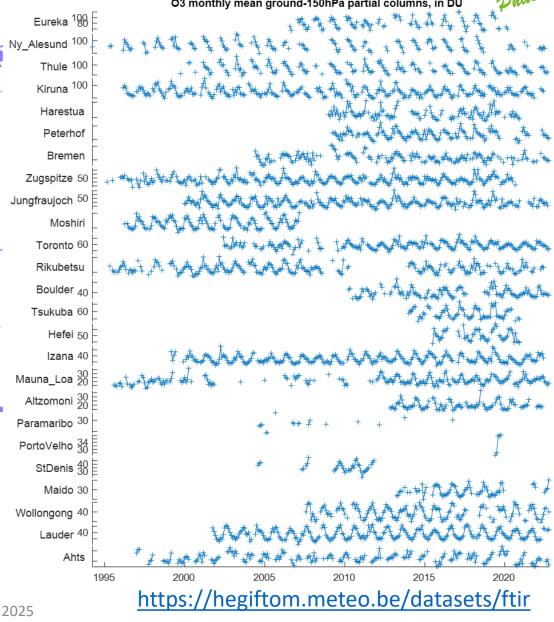
## **HEGIFTOM:** Homogenized datasets







- Oldest date back to the mid 90s, most since mid 2000s
- Those sites also provide CO/HCHO

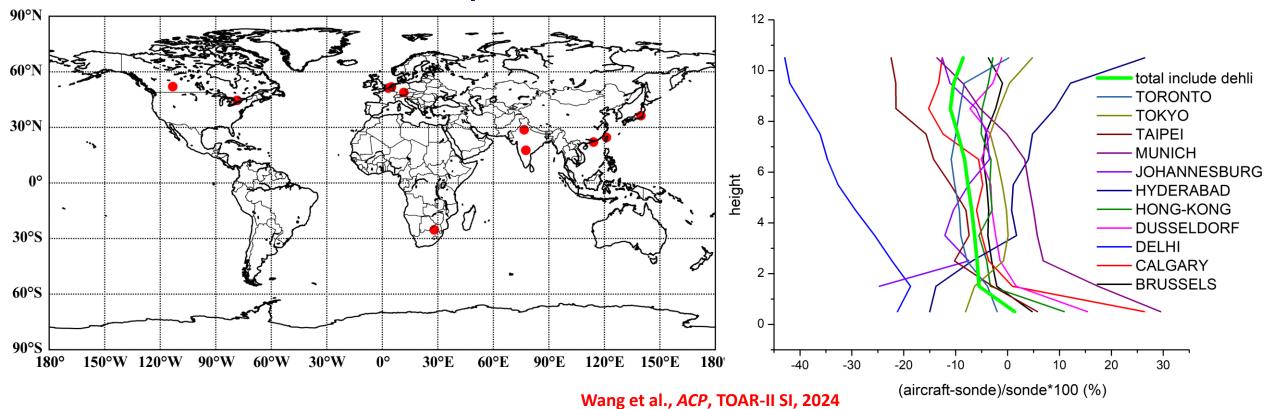




# **HEGIFTOM:** Intercomparisons (external consistency)



- Deliverable: TOAR-II Intercomparison Guidelines for Observations of Tropospheric Column
  Ozone and Tropospheric Ozone Profiles (<a href="https://igacproject.org/sites/default/files/2022-03/TOAR-">https://igacproject.org/sites/default/files/2022-03/TOAR-</a>
  <a href="https://igacproject.org/sites/default/files/2022-03/TOAR-">II Guidelines for TCO and Profile Intercomparisons.pdf</a>)
- IAGOS aircraft vs. ozonesonde profiles at 11 stations



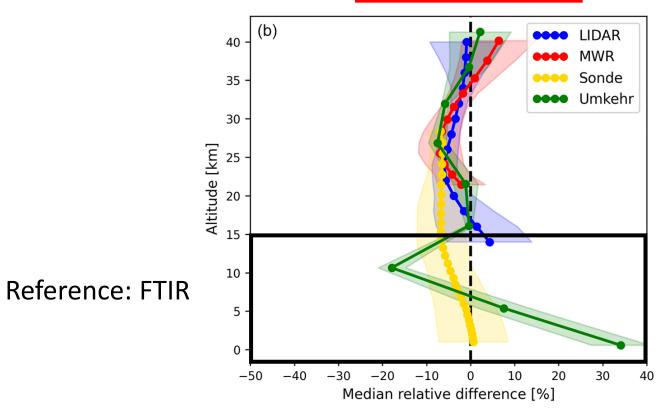


# **HEGIFTOM:** Intercomparisons (external consistency)



**Intercomparisons:** comparison of (tropospheric) ozone retrievals from different ground-based instruments at dedicated sites

Lauder (New Zealand)



Björklund et al., AMT, TOAR-II SI, 2024



## **HEGIFTOM:** Tropospheric ozone columns (TrOC)



the 2 recommended

TOAR-II tropospheric

ozone column definitions

Deliverable: time series of different (partial) tropospheric ozone column amounts

- 1.  $P > P_TP (WMO)$
- 2. P > P (lat) (e.g. 150 hPa @ tropics, 400 hPa in polar regions)
- 3. P > 300 hPa
- 4. FT: 4 < h < 8 km AND 700 hPa > P > 300 hPa
- 5. LT: h < 4 km AND P > 700 hPa
- 6. BL: h < 2 km
- for all sites/techniques, if feasible
- provided for all measurements (L1), together with daily means (L2) and monthly means (L3)
- available in DU or ppb
- uncertainties included (random, systematic, total, statistical)
- simple csv files, with readme files per technique

https://hegiftom.meteo.be/datasets/tropospheric-ozone-columns-trocs



## **HEGIFTOM:** Tropospheric ozone columns (TrOC)



the 2 recommended

TOAR-II tropospheric

ozone column definitions

Deliverable: time series of different (partial) tropospheric ozone column amounts

- 1.  $P > P_TP (WMO)$
- 2. P > P (lat) (e.g. 150 hPa @ tropics, 400 hPa in polar regions)
- 3. P > 300 hPa **HERE!**
- 4. FT: 4 < h < 8 km AND 700 hPa > P > 300 hPa
- 5. LT: h < 4 km AND P > 700 hPa
- 6. BL: h < 2 km
- for all sites/techniques, if feasible
- provided for all measurements (L1), together with daily means (L2) and monthly means (L3)
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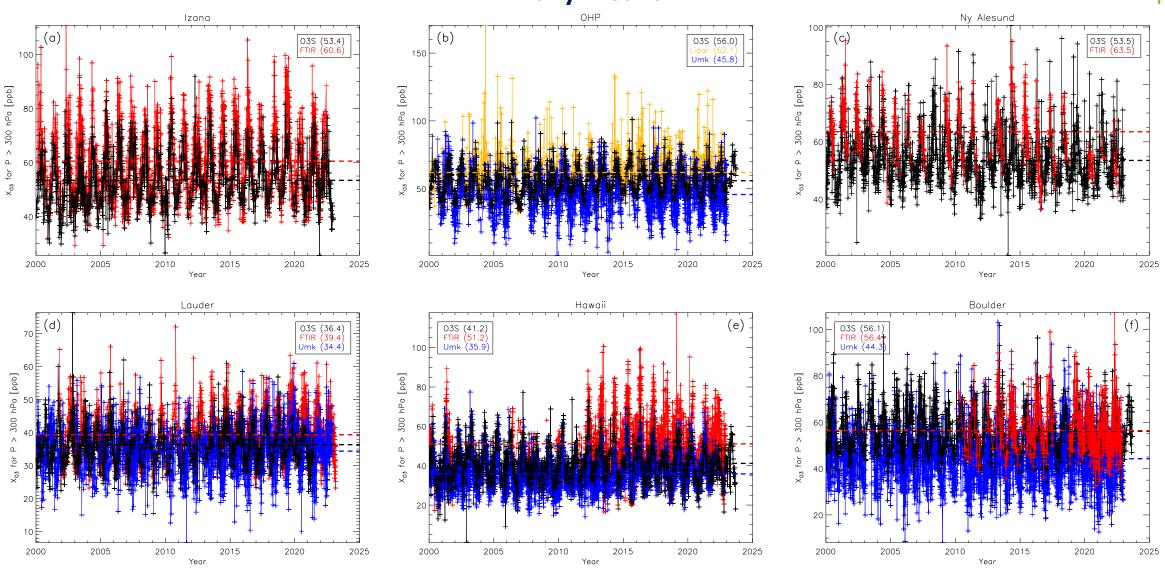
https://hegiftom.meteo.be/datasets/tropospheric-ozone-columns-trocs



# **HEGIFTOM:** TrOC intercomparisons at collocated sites



## **Daily means**

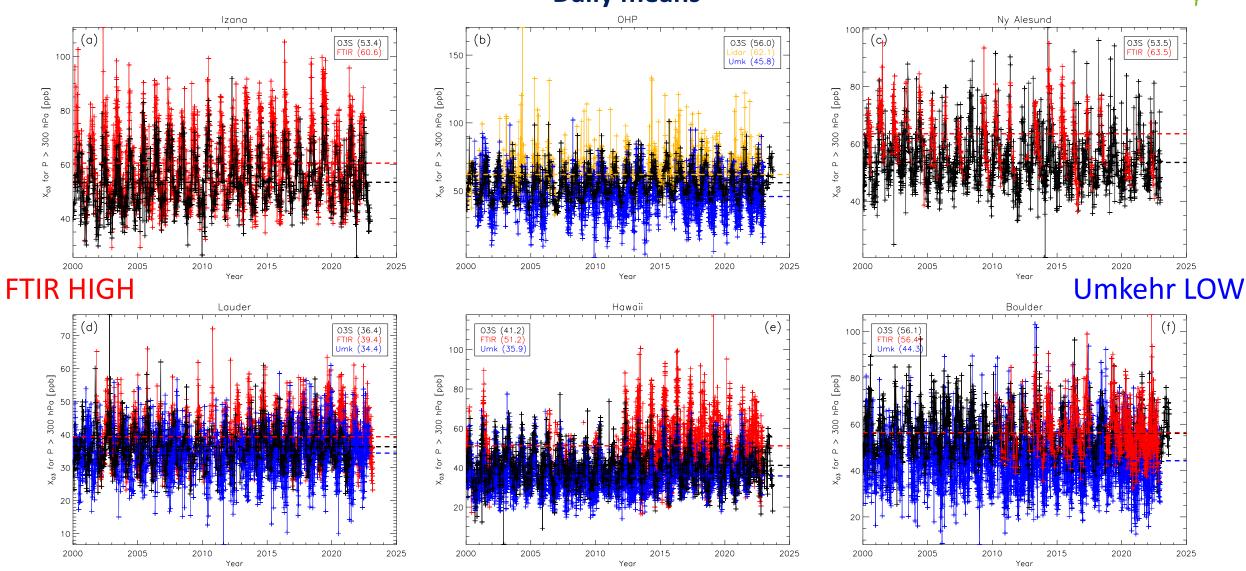




# **HEGIFTOM:** TrOC intercomparisons at collocated sites









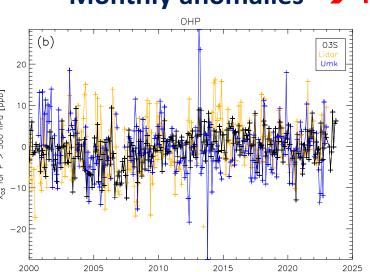
10

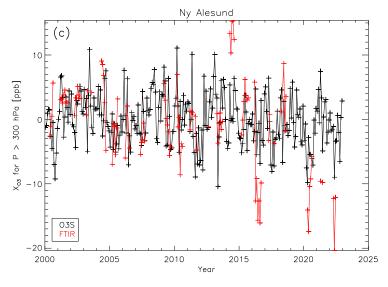
2005

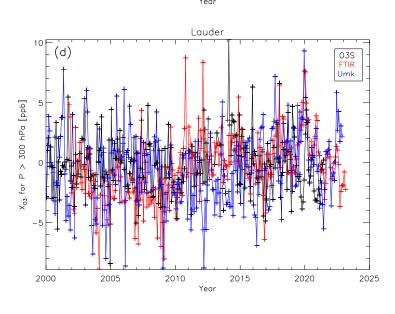
# **HEGIFTOM:** TrOC intercomparisons at collocated sites



## Monthly anomalies → TRENDS







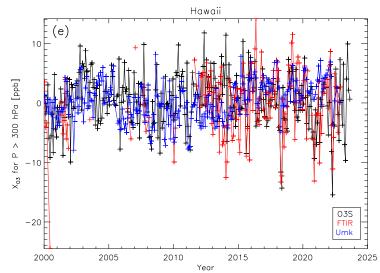
2010

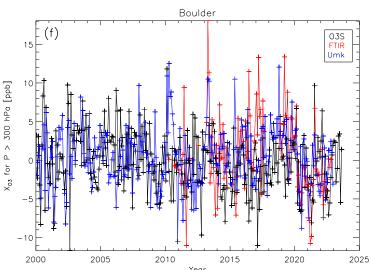
2015

2020

2025

Izana







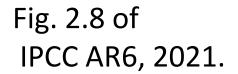
## **HEGIFTOM:** Tropospheric ozone column trends

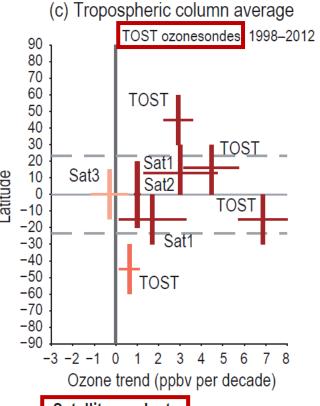


- TOAR-II: tropospheric ozone trends assessment
- In literature:

TOST = Trajectorymapped Ozonesonde dataset for the Stratosphere and Troposphere

(Wang et al., ACP, TOAR-II SI, 2024)





#### Satellite products:

Sat1 1979-2016 (TOMS, OMI/MLS)
Sat2 1995-2015 (GOME, SCIAMACHY, OMI,
GOME-2A, GOME-2B)
Sat3 1995-2015 (GOME, SCIAMACHY, GOME-II)

- ✓ Here: focus on high-quality ground-based and in-situ measurements (individual sites + "merged")
- ✓ Consistency in tropospheric ozone column metric (<a href="https://www.netricolumn.com/here">here</a>: surface to 300 hPa)
- ✓ Consistency in used trend estimation tools (QR vs. MLR)
- ✓ Consistency in time ranges (here: 2000-2002 till 2019-2022)
- √ Consistency in units (ppbv/dec)
- ✓ Not only as function of latitude!



## **HEGIFTOM:** Tropospheric ozone column trends

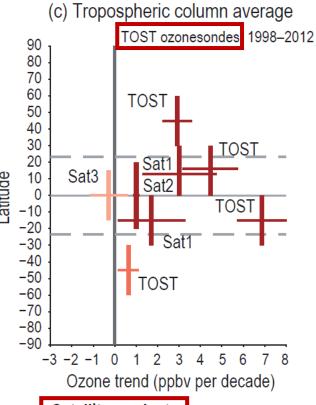


- TOAR-II: tropospheric ozone **trends** assessment
- In literature:

TOST = Trajectorymapped Ozonesonde dataset for the Stratosphere and Troposphere

(Wang et al., ACP, TOAR-II SI, 2024)

Fig. 2.8 of IPCC AR6, 2021.



#### Satellite products:

Sat1 1979–2016 (TOMS, OMI/MLS)
Sat2 1995–2015 (GOME, SCIAMACHY, OMI,
GOME-2A, GOME-2B)
Sat3 1995–2015 (GOME, SCIAMACHY, GOME-II)

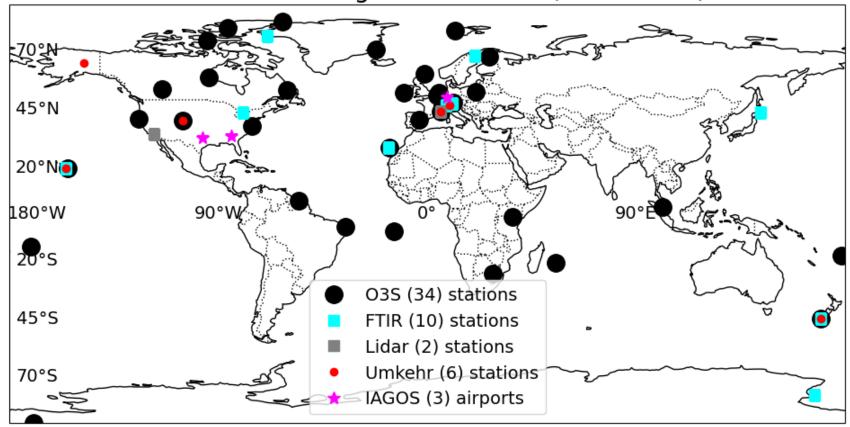
- ✓ Here: focus on high-quality ground-based and in-situ measurements
- (individual sites + "merged")
  - ✓ Consistency in tropospheric ozone column metric (here: surface to 300 hPa)
  - ✓ Consistency in used trend estimation tools (QR vs. MLR)
  - ✓ Consistency in time ranges (here: 2000-2002 till 2019-2022)
  - √ Consistency in units (ppbv/dec)
  - ✓ Not only as function of latitude!



## Individual site trends sample



## Global Sites Contributing to HEGIFTOM (55 L1 Data) Trends

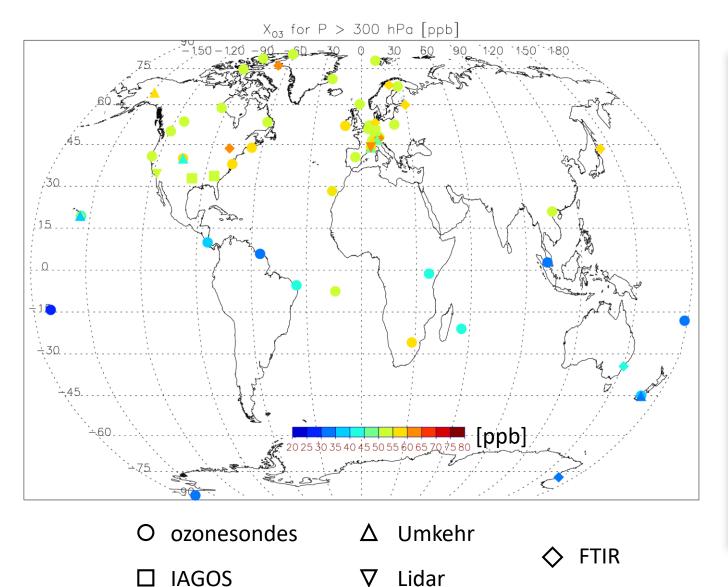


- Sampling (> 120months of data) andgaps (2000+) putconstraints
- 55 sites
- Some sites with different techniques (Boulder, Hawaii, Lauder, OHP, Ny Ålesund, Izaña, ...)
  - → intercomparisons



## Tropospheric ozone column distribution





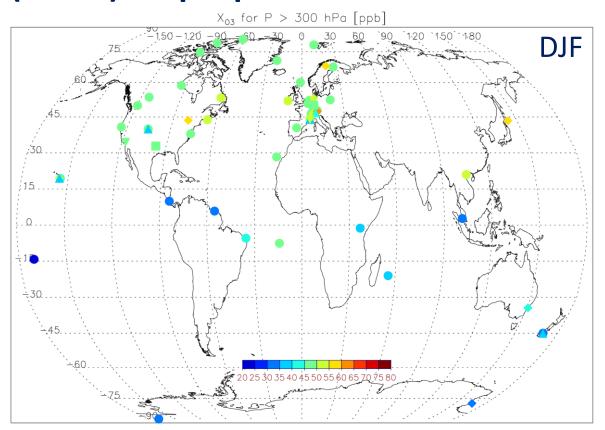
- Mean column-averaged tropospheric ozone distribution (TrOC) from surface – 300 hPa for 2000-2022
- <u>Lowest</u>: tropics (< ±15°) and SH;</li>
   <u>Highest</u>: NH (spring & summer!)
- Reason: ozone production from enhanced anthropogenic emissions in the NH and higher rates of stratospheric downwelling

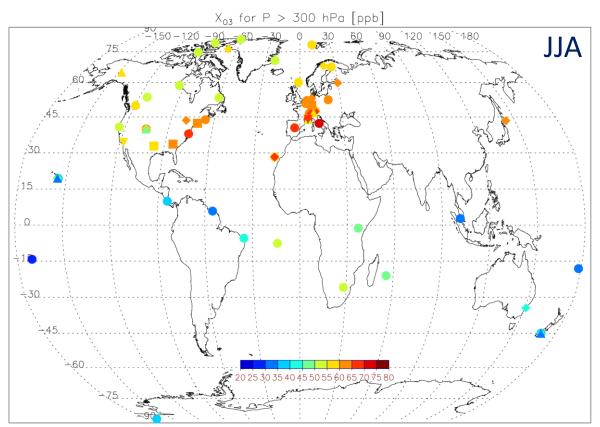


## **Tropospheric ozone column distribution**



## (mean) Tropospheric ozone column distribution: DJF vs. JJA



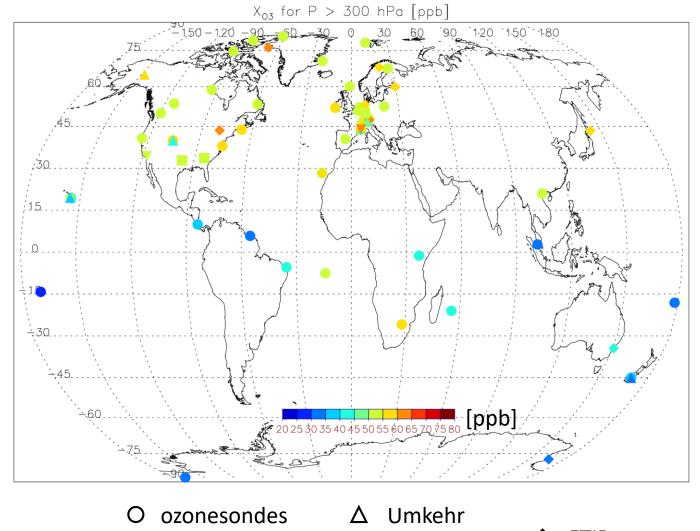


- Highest values in JJA in NH: peak photochemical production & summertime emission max of biogenic VOC ozone precursors
- >< SH (SON): STE & biomass burning</li>



## Tropospheric ozone column distribution



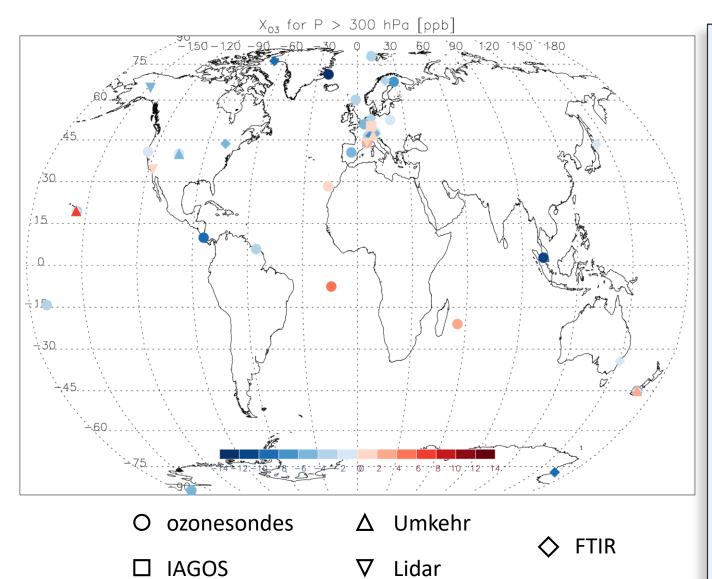


- Mean column-averaged tropospheric ozone distribution (TrOC) from surface – 300 hPa for 2000-2022
- Now compare mean TrOC values for 2000 – 2019 vs. 2020 – 2022 (COVID-19 period)



## **Tropospheric ozone column distribution: COVID impact**





• Relative change of mean TrOC for the time period 2020-2022 vs. 2000-2019

Blue: 2020-2022 < 2000-2019

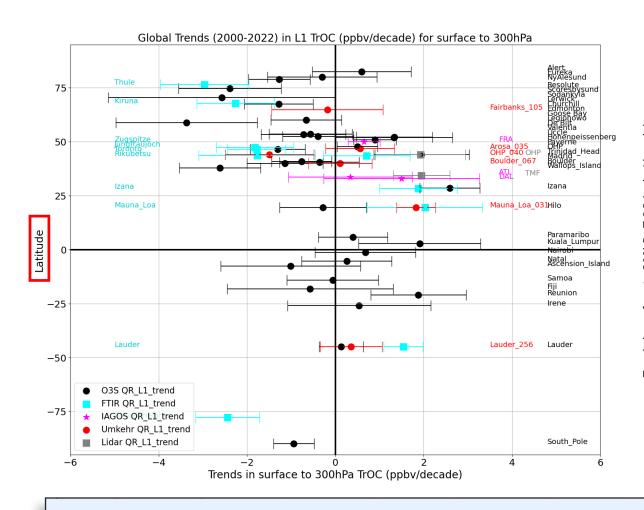
Red: 2020-2022 > 2000-2019

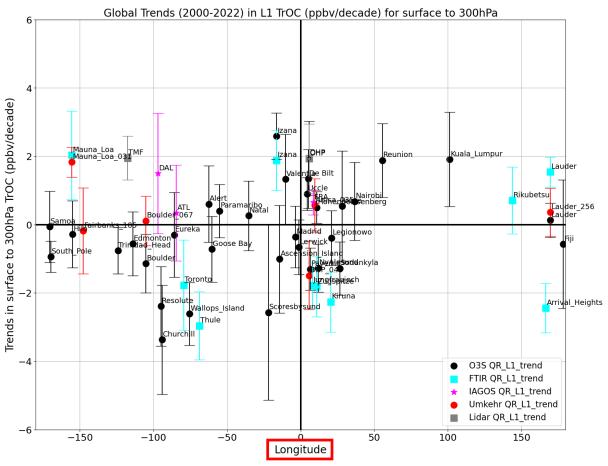
- Decline in 75% of the sites, on average -2.5% prominent in NH (spring + summer)
- Reason: decreased emissions of ozone precursors (e.g. NO<sub>2</sub>) due to COVID-19 lockdown restrictions
- Impact on trends!



## Individual site trends: QR median trends





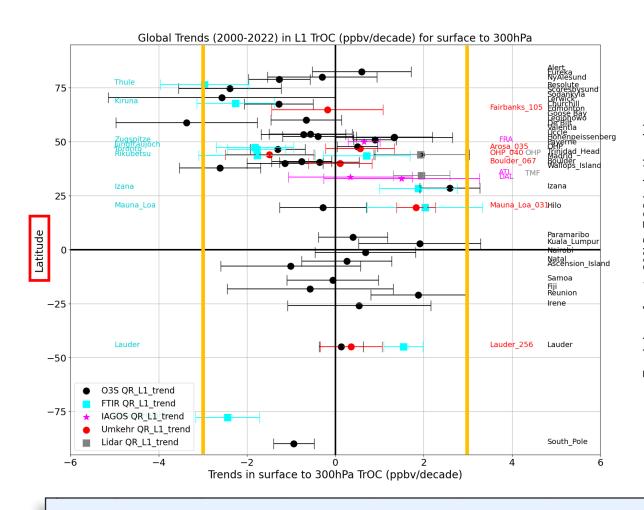


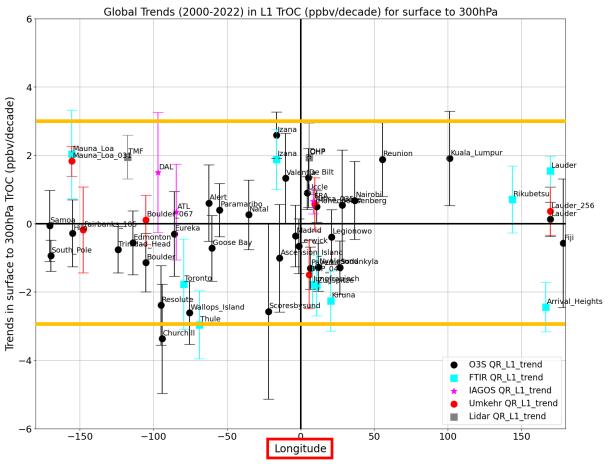
- same number of positive and negative trends, 42% of the sites with non-significant trends
- mostly within ± 3 ppbv/decade → constraints for satellite and model products



## Individual site trends: QR median trends





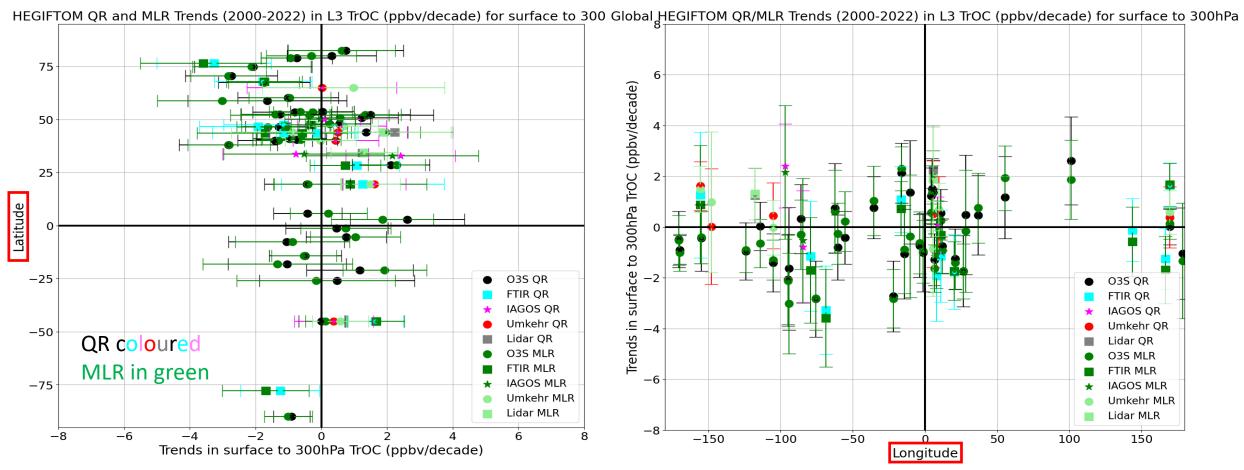


- same number of positive and negative trends, 42% of the sites with non-significant trends
- mostly within ± 3 ppbv/decade → constraints for satellite and model products



## **Individual site trends: QR median + MLR trends**



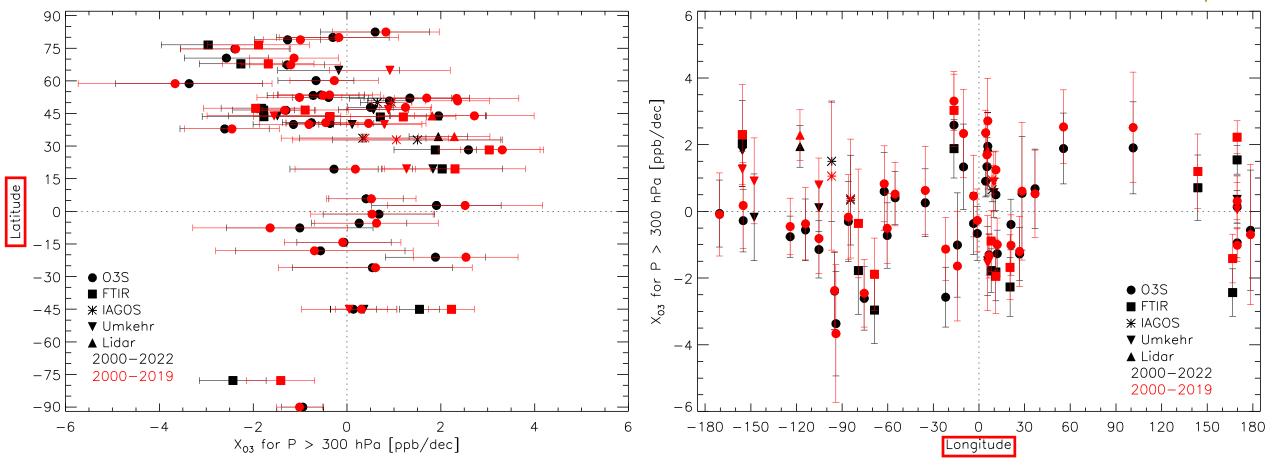


→ Estimates and conclusions fairly independent of used statistical trend estimation tool



# Individual site trends: post-COVID impact

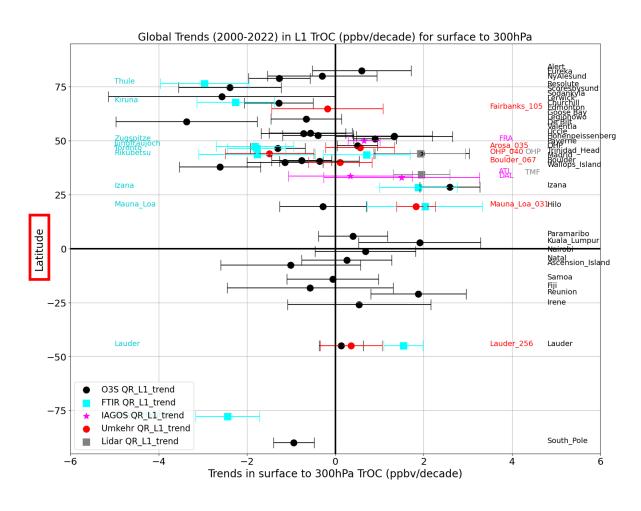


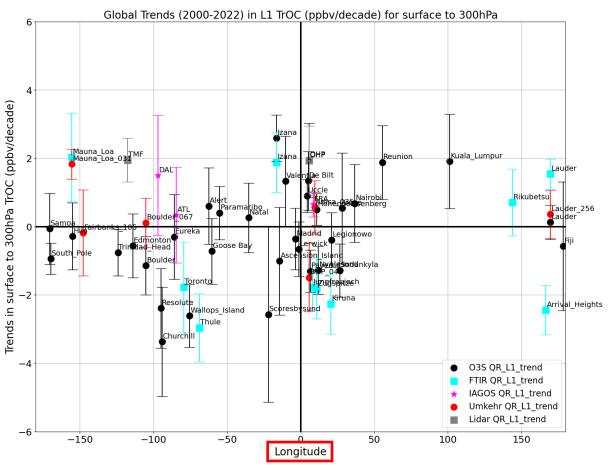


For 75% of sites: **trend reduction** in 2000-2022 w.r.t. 2000-2019 period! (-0.34 ppbv/dec for entire sample)





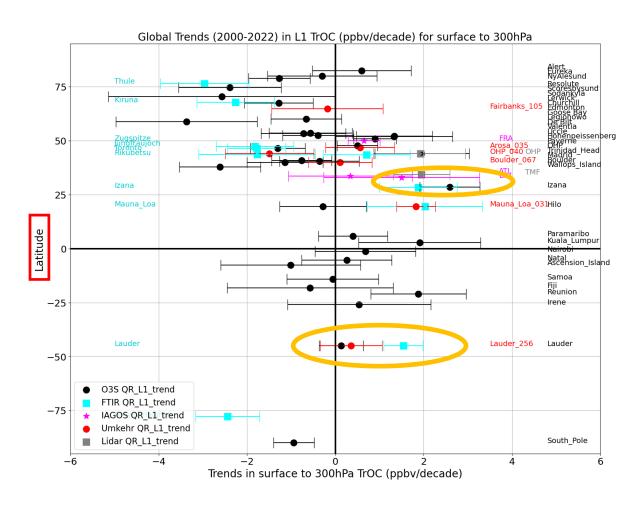


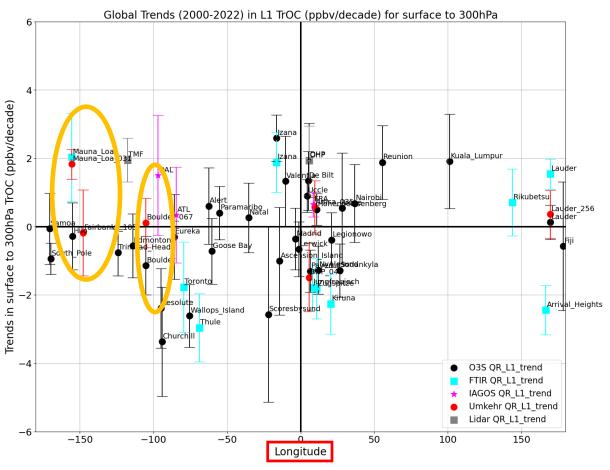


- Trend differences at multi-instrument sites?
- Negative trends at high (polar) latitudes?





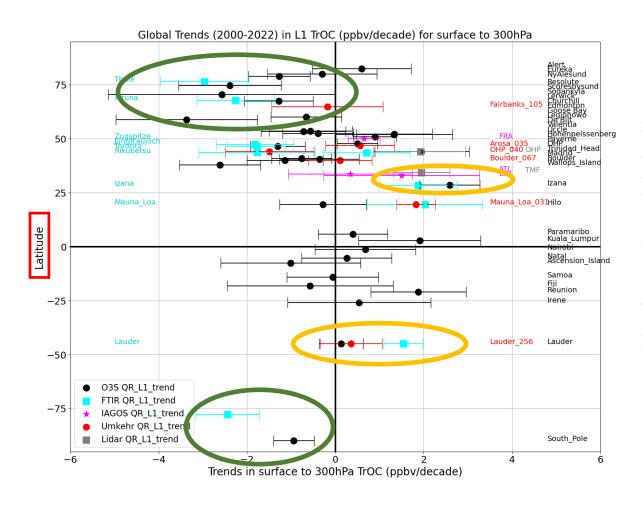


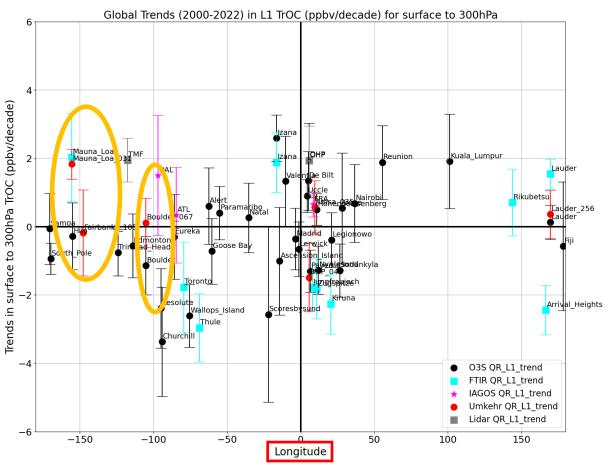


- Trend differences at multi-instrument sites?
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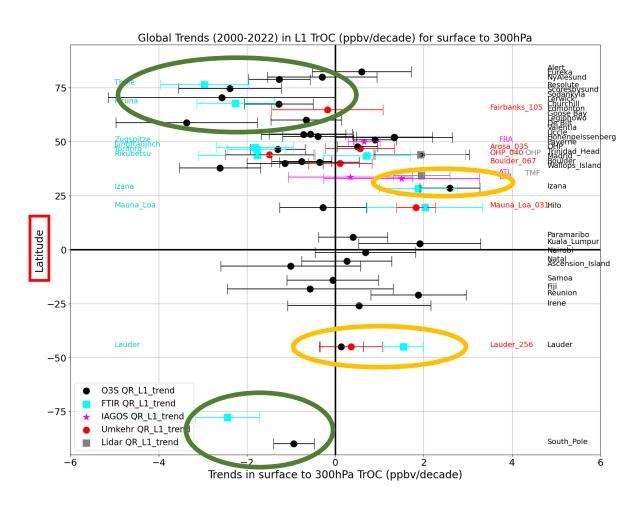


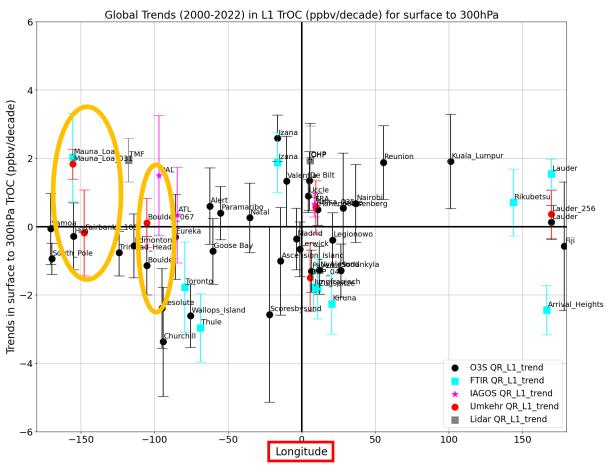


- Trend differences at multi-instrument sites?
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- Trend differences at multi-instrument sites?
- Negative trends at high (polar) latitudes?



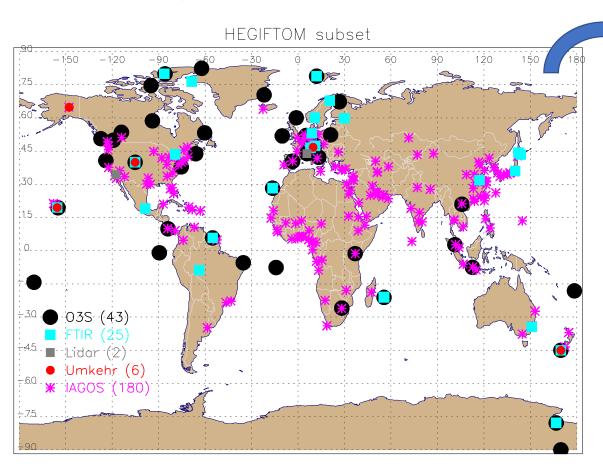
Regional trend consistency? Merging? Spatial Gap filling?



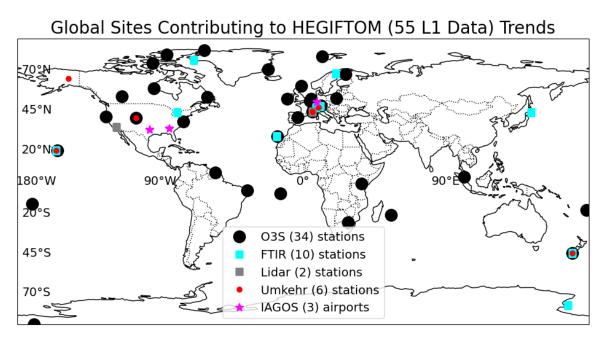
#### "Regionalized" trends



#### **Gap filling?**



Sites with data in 2000-2022 period



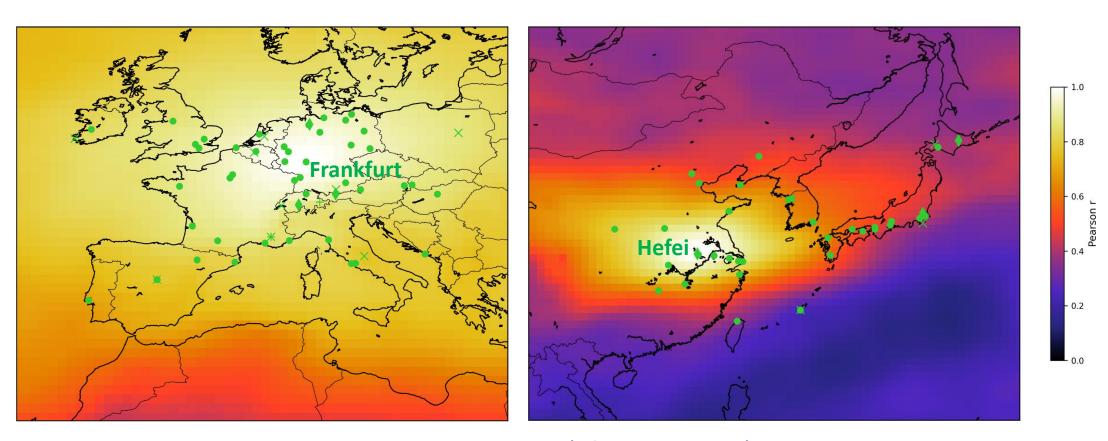
Sites used for trend calculation for 2000-2022 period



# **Strategy for regionalized trends**



#### Which regions/sites?



- Correlation maps between CAMS TrOC (sfc 300 hPa) monthly anomalies at HEGIFTOM sites (here: Frankfurt, IAGOS & Hefei, FTIR)
- r > 0.7!

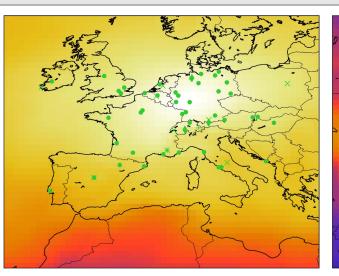


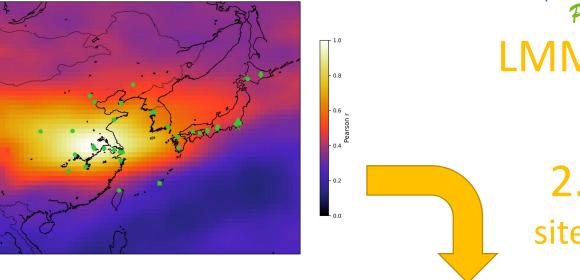
#### 2 strategies for regionalized trends



# **TOST**







Trends in defined regions with TOST

(Trajectory-mapped

Ozonesonde dataset

for the Stratosphere and

Troposphere):

ozonesondes only!

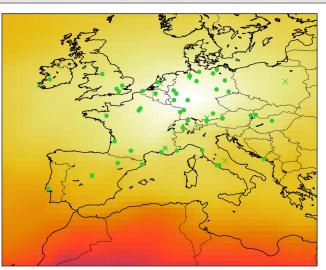
Statistical method (linear mixed-effects modelling, LMM) for calculating synthetized trends from well-correlated individual time series for all instruments, allowing an intercept and a slope to adjust the difference from each individual trend against the overall trends.

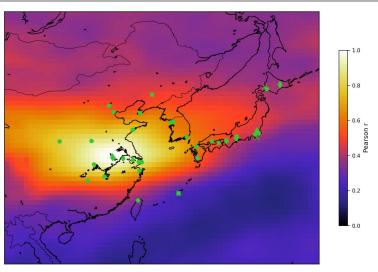




# **TOST**







Trends in defined regions with TOST

(Trajectory-mapped

Ozonesonde dataset

for the Stratosphere and

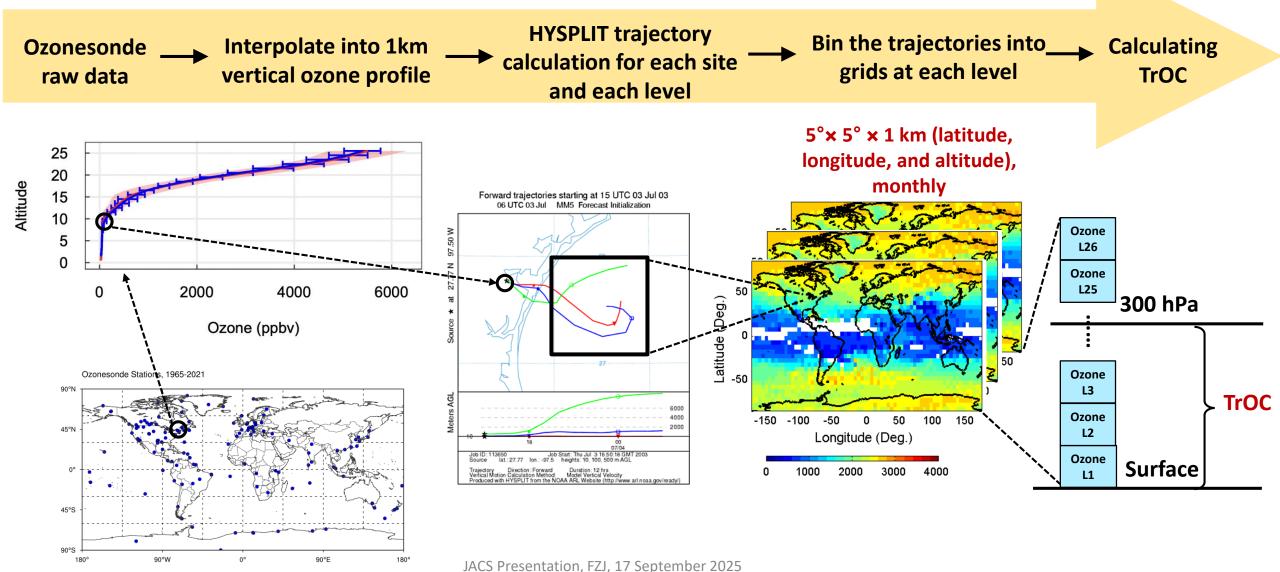
Troposphere):

ozonesondes only!





#### **TOST**





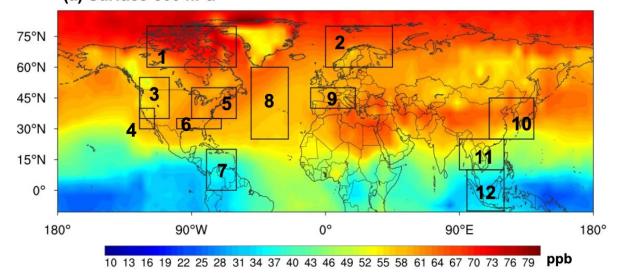


# Regions

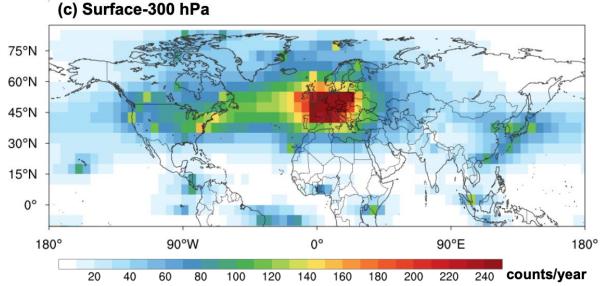
T. regions

- 12 regions, mainly NH (highest TrOC amounts)
- Based on the density of <u>ozonesonde</u> observations in well-correlated regions
- 2 periods:1995-2021, 2000-2021

# Mean ozone concentrations, 1995-2021 (a) Surface-300 hPa

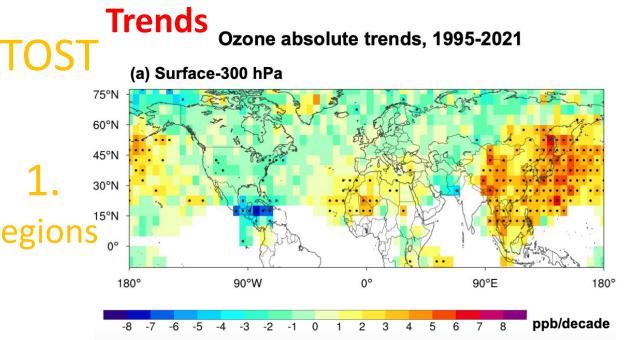


#### Mean ozone independent samples, 1995-2021

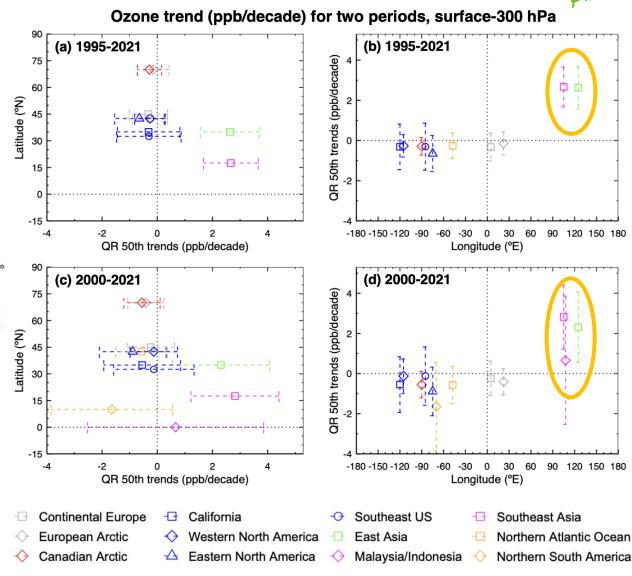








- **positive trends** in (**South)East Asia**, negative elsewhere (*except around some individual sites in Africa*)
- **trend differences** between two different periods (1995+ vs. 2000+) **insignificant**







#### **COVID** impact?

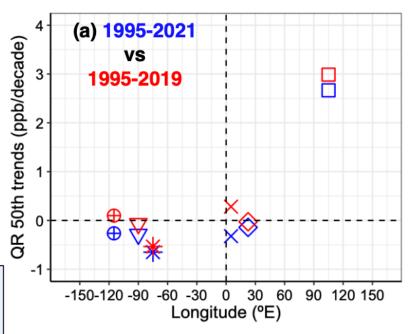
**TOST** 

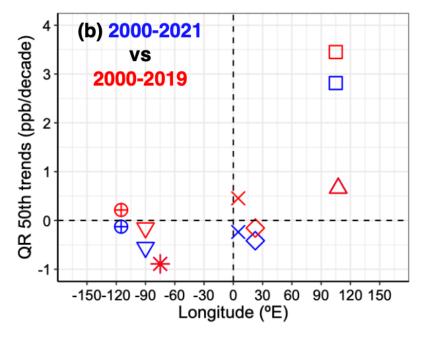
1. regions

for all regions and two periods (1995+, 2000+):

pre-COVID trends > post-COVID trends

# Ozone trends (ppb/decade) comparison, post-COVID and pre-COVID of surface-300 hPa





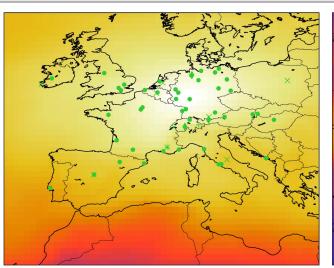


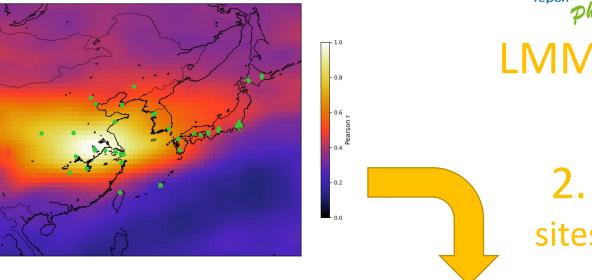
#### 2 strategies for regionalized trends



# **TOST**







Trends in defined regions with TOST

(Trajectory-mapped

Ozonesonde dataset

for the Stratosphere and

Troposphere):

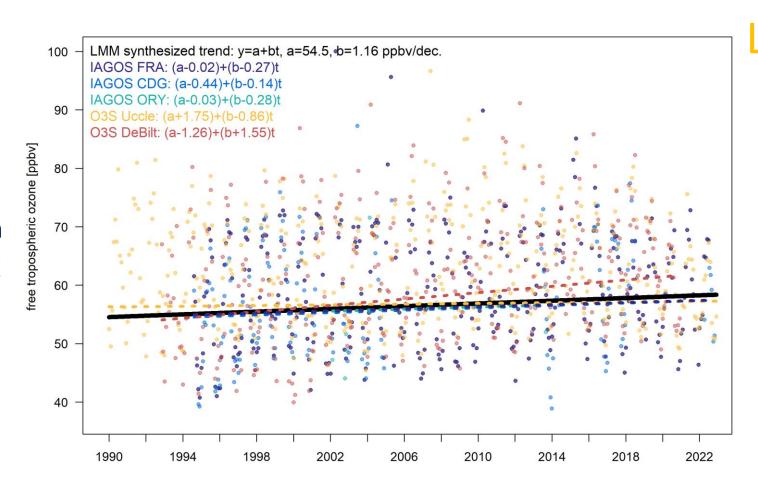
ozonesondes only!

Statistical method (linear mixed-effects modelling, LMM) for calculating synthetized trends from well-correlated individual time series for all instruments, allowing an intercept and a slope to adjust the difference from each individual trend against the overall trends.





- the individual trends are adjusted to calculate a synthetized trend for the combined time series
- example: monthly means, in practice: all measurements!
- different weights for different techniques (uncertainties) to counterbalance weight given to higher number of measurements



MM<sub>-</sub>

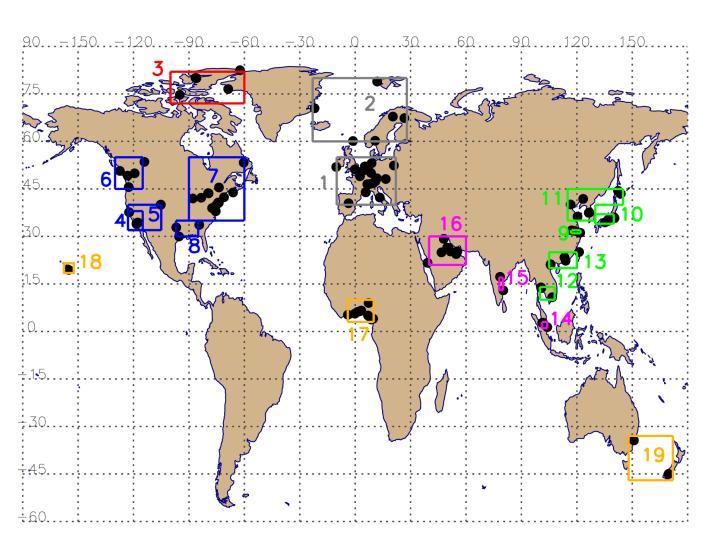
2. sites





#### Regions





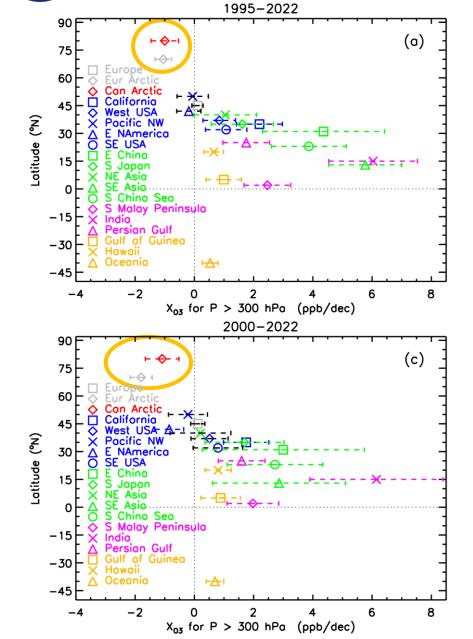
• 19 regions! Mostly NH.

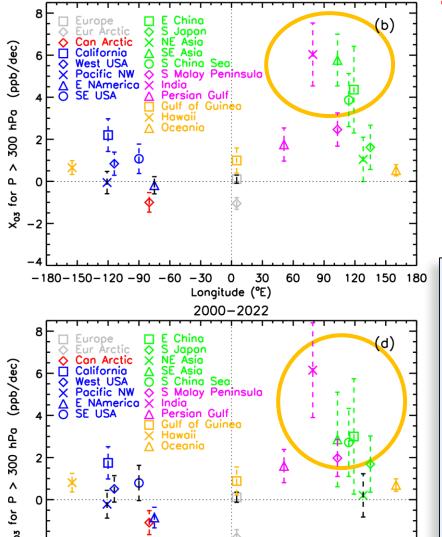
sites

- at least two sites/techniques (Hawaii)
- only time series which are long enough (>60 months of data) are included
- 2 periods:1995-2022 & 2000-2022









Longitude (°E)

90 120 150

#### **Trends**

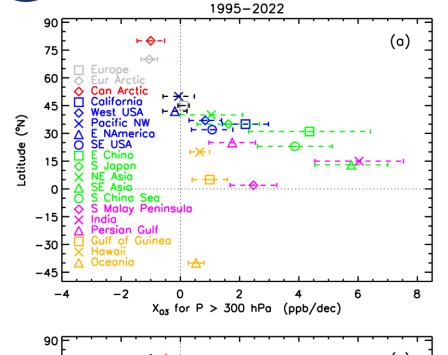
**LMM** 

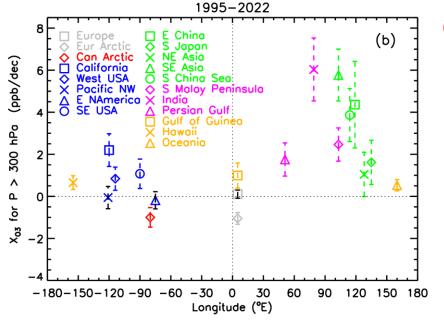
2. sites

- mostly pos. trends
   (Asia!), except in Arctic
   regions
- trend diff. between two different periods (1995+ vs. 2000+) mostly insignificant (! East China + SE Asia)





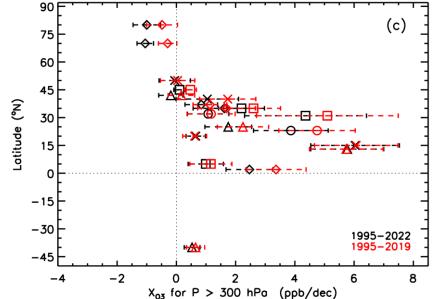


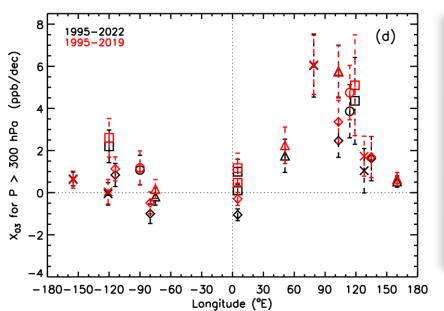


COVID Impact

LMM

2. sites





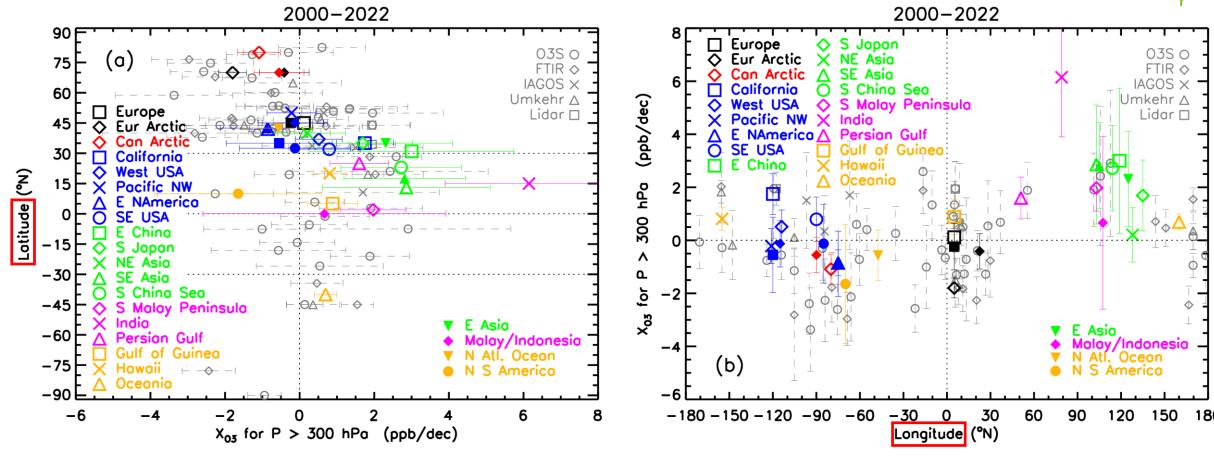
for all regions and two periods (1995+, 2000+):

pre-COVID trends >
post-COVID trends



#### All trends: 2000-2022





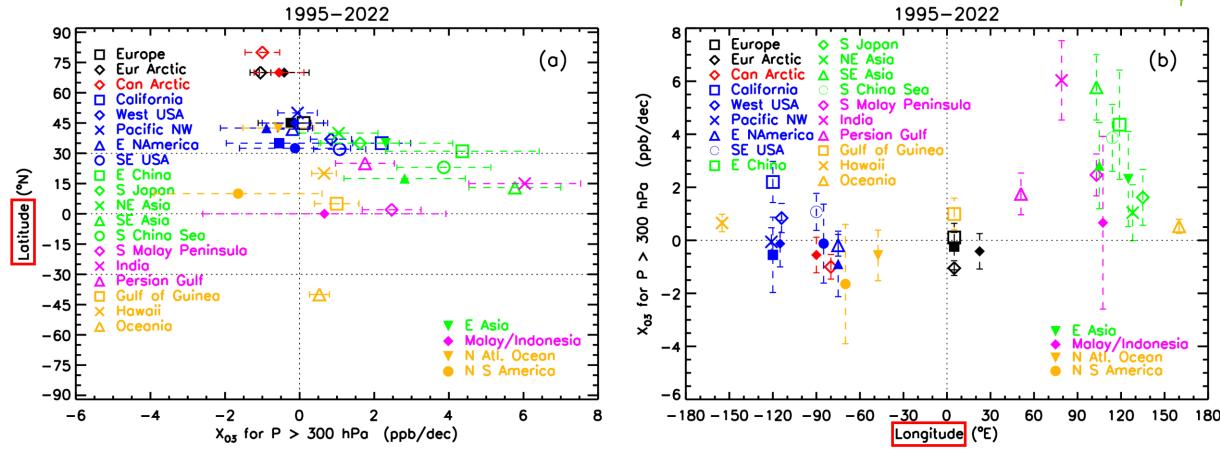
background grey = individual site trends different colors = different regions open symbols = synthesized LMM trends filled symbols = TOST regional trends

- Regional trends "summarize" individual trend estimates
- No large trend diff. between similar regions for 2 approaches



#### All trends: 1995-2022





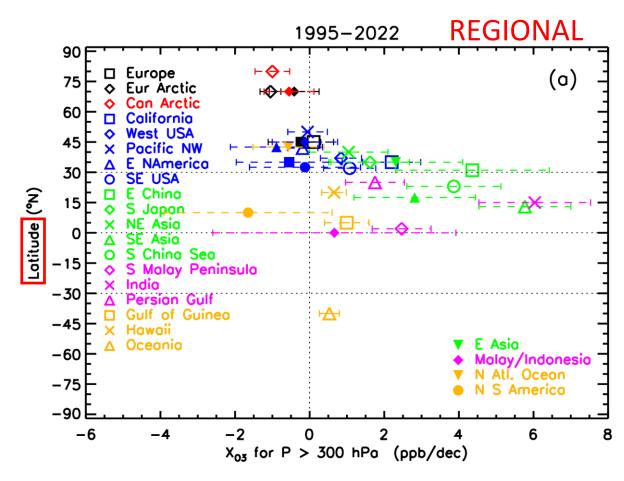
different colors = different regions open symbols = synthesized LMM trends filled symbols = TOST regional trends

- No large trend diff. between similar regions for 2 approaches
- TOST trends closer to zero than LMM trends

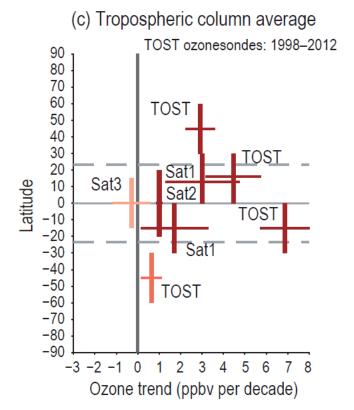


#### All trends





different colors = different regions open symbols = synthesized trends filled symbols = TOST regional trends



**LATITUDINAL** 

#### Satellite products:

Sat1 1979-2016 (TOMS, OMI/MLS)

Sat2 1995-2015 (GOME, SCIAMACHY, OMI,

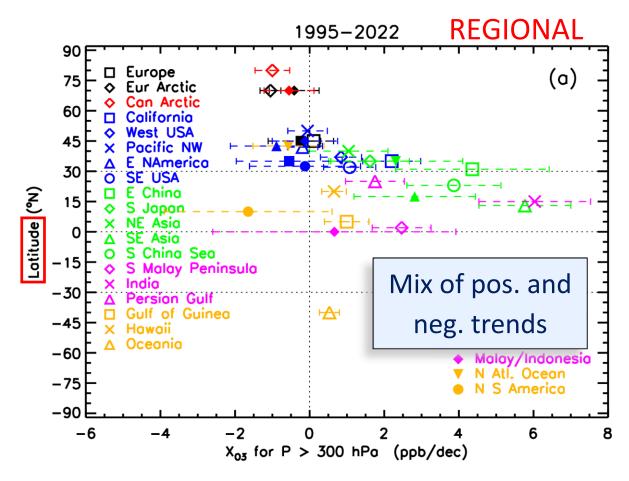
GOME-2A, GOME-2B)

Sat3 1995–2015 (GOME, SCIAMACHY, GOME-II)

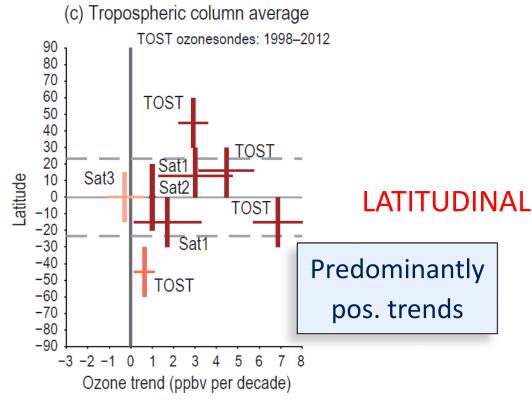


#### All trends





different colors = different regions open symbols = synthesized trends filled symbols = TOST regional trends



#### Satellite products:

Sat1 1979–2016 (TOMS, OMI/MLS)
Sat2 1995–2015 (GOME, SCIAMACHY, OMI,
GOME-2A, GOME-2B)
Sat3 1995–2015 (GOME, SCIAMACHY, GOME-II)



#### **Conclusions and outlook**



- Homogenized ground-based measurements provide TrOC (sfc- 300 hPa)
   trend estimates within ± 3 ppb/dec, for TrOC ranging between 20 (SH, DJF)
   and 80 ppb (NH, MAM & JJA, especially East USA, South EU, East Asia)
- Mixture of positive and negative trends worldwide, but consistently negative in Arctic (?) and positive in East Asia (continuing increase of ozone precursor emissions)
- **COVID-19 restrictions** led to less ozone precursor emissions and **decreasing TrOC amounts**, impacting present-day (i.e. post-COVID) trends



# **Further reading and outlook**



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https://doi.org/10.5194/acp-25-7187-2025

Global ground-based tropospheric ozone measurements: reference data and individual site trends (2000–2022) from the TOAR-II/HEGIFTOM project

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Ground-based tropospheric ozone measurements: regional tropospheric ozone column trends from the TOAR-II/HEGIFTOM homogenized datasets

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Jane Liu², Eliane Maillard Barras⁶, Corinne Vigouroux⁵, Irina Petropavlovskikh³,7, Thierry Leblanc⁶,
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Deniz Poyraz¹, Gérard Ancellet¹⁶, Marie-Renée De Backer¹७, Matthias M. Frey¹⁶,
James W. Hannigan¹ゥ, José L. Hernandez²₀, Bryan J. Johnsonⁿ, Nicholas Jones²¹, Rigel Kivi²²,
Emmanuel Mahieu²³, Isamu Morino²⁴, Glen McConville⁷, Katrin Müller²⁵, Isao Murata²⁶,
Justus Notholt²⊓, Ankie Piters²⁶, Maxime Prignon²ゥ, Richard Querel³₀, Vincenzo Rizi³¹, Dan Smale³₀,
Wolfgang Steinbrecht³², Kimberly Strong³³, and Ralf Sussmann³⁴



Those contain additionally detailed TrOC intercomparisons at nearby/collocated sites,
 1990/1995/2000 – 2022 trend comparisons, relative contribution of lower+free-tropospheric ozone column trends to entire tropospheric ozone column trends, TrOC seasonal cycle change, etc.

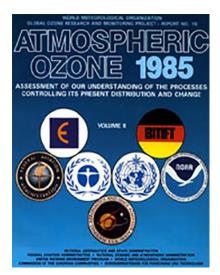


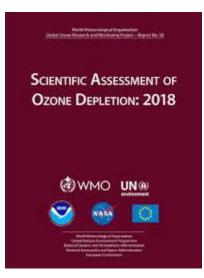
Those do not contain: variation of seasonal or low/high percentile trends!

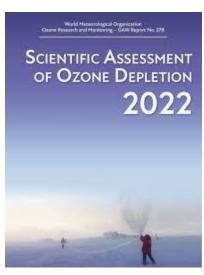




Stratospheric ozone trends in "Scientific Assessment of Ozone Depletion" (every 4 years)







For last two assessments: APARC LOTUS (Long-term Ozone Trends and Uncertainties in the Stratosphere)

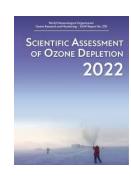


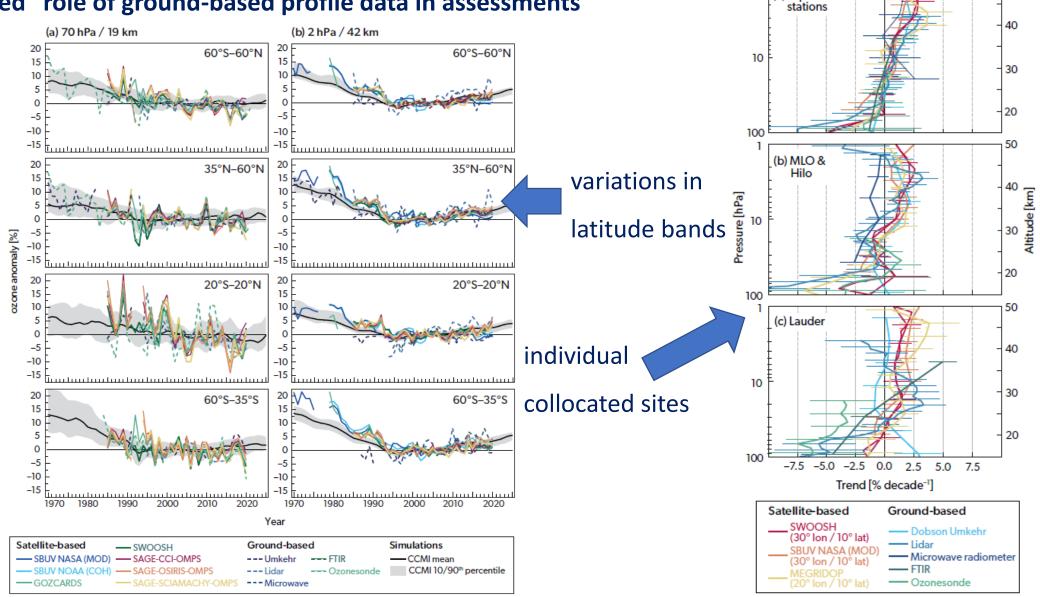




(a) Alpine

#### "Limited" role of ground-based profile data in assessments

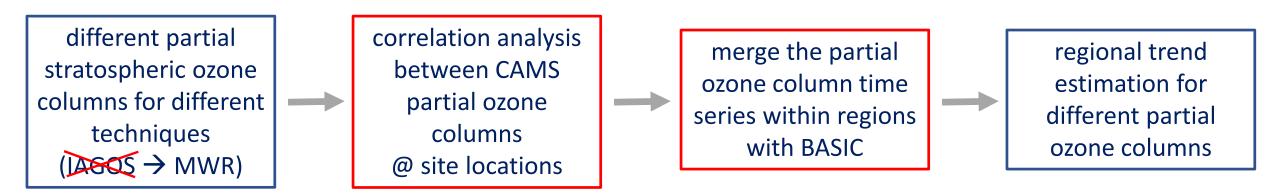








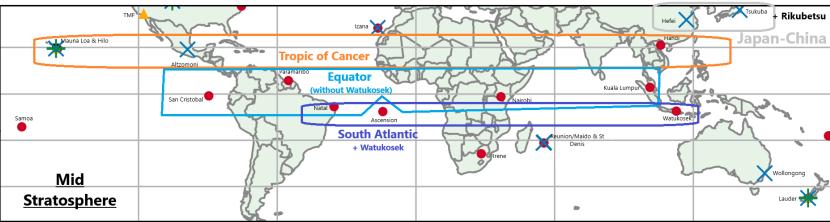
• For next assessment, within LOTUS, "mimic" HEGIFTOM regional trends approach

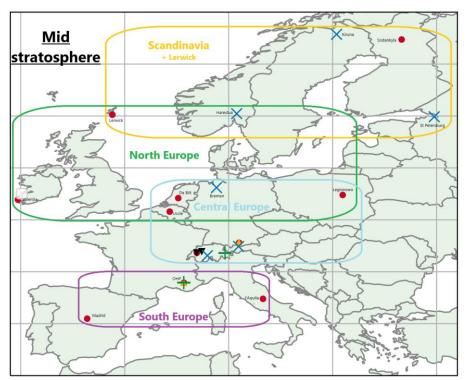


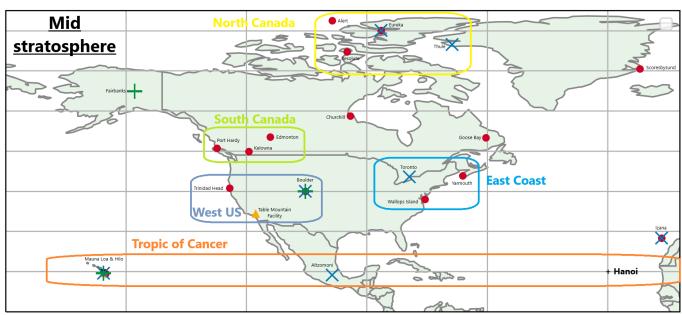




correlation analysis
between CAMS
partial ozone
columns
@ site locations







Courtesy Caroline Jonas & Corinne Vigouroux (BIRA)

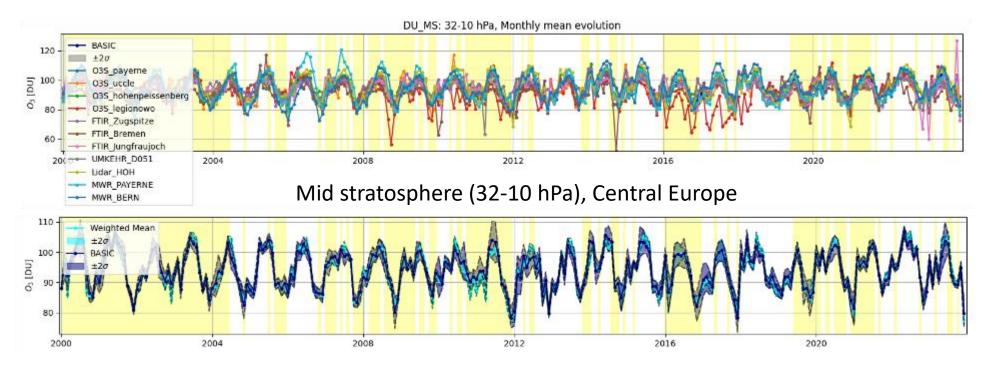




merge the partial ozone column time series within regions with BASIC

Use Bayesian method to "automatically correct" the ozone composites:

- 1. Construct uncertainty estimates (on each monthly mean) and include all additional prior information that we have available
- 2. Form a Gaussian-mixture likelihood (Probabilistic model of the data)
- 3. Develop transition priors to estimate how ozone is expected to vary on monthly timescales (month-to-month climatology)
- 4. Multiply the transition prior with the likelihood and obtain the posterior distribution.

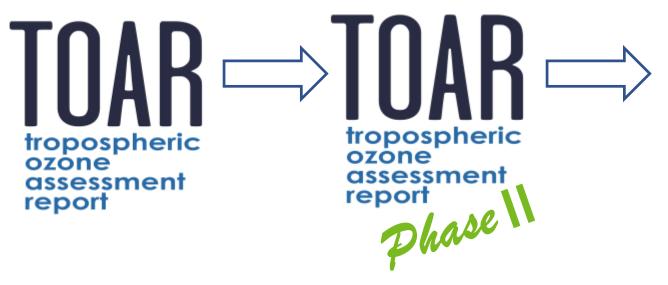


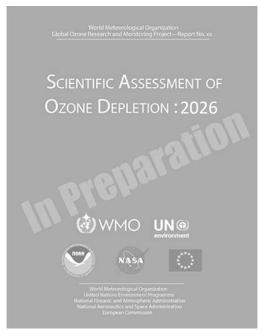
Courtesy Louis Mirallie & Eliane Maillard Barras (MeteoSwiss)



#### This talk:







... with focus on ground-based measurements!



TANOS PHERIC CHEMIS





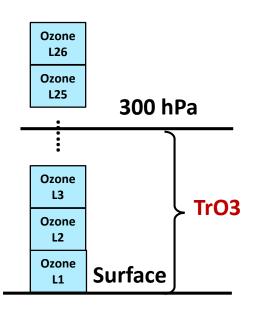
# Thank you for your attention!

**Questions?** 









- 1. Convert TOST from altitude level to pressure level: using the MERRA-2 geopotential height at each pressure level.
- 2. Monthly TOST **gap filling vertically**: linearly interpolate the profile when there are >80% available data between the selected altitude range (>300hPa/4-8km)
- 3. Monthly TOST **gap filling horizontally**: if there's a gap, averaging the closest surrounding (<1000km) gridpoints with available data, weighted by distance [details see Liu et al. (2022)].
- 4. Calculate **seasonal mean TOST** in each year: at least one available value for each season.
- 5. Calculate **seasonal mean TrO3** in each year.
- 6. Calculate the annual mean TrO3: at least one available value for each season.
- 7. Calculate **the regional mean TrO3**: average the TrO3 over the selected region weighted by grid area [\*cosine(lat)].
- 8. Calculate the **Quantile Regression trend** using the annual and regional mean TrO3

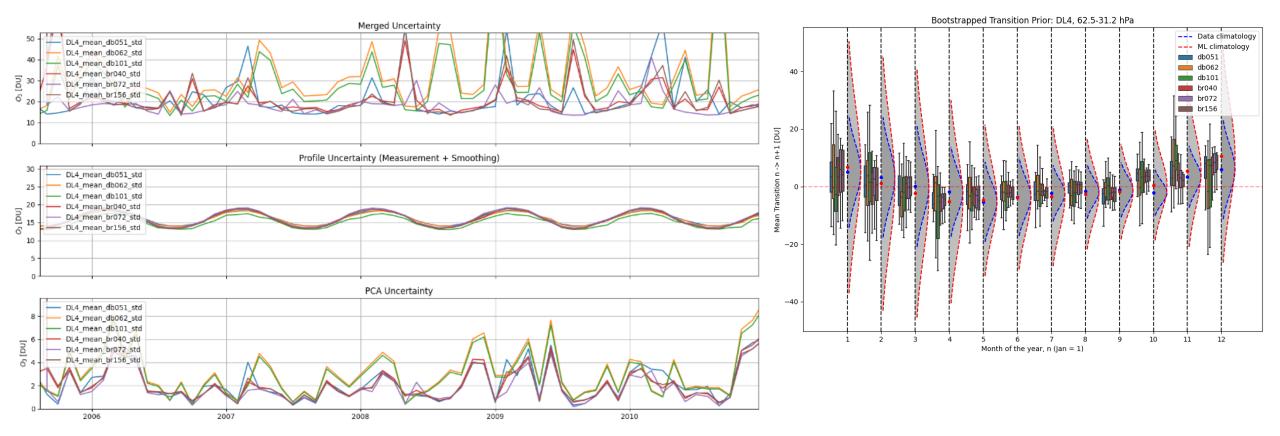


#### **BASIC**



Use Bayesian method to "automatically correct" the ozone composites:

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- 2. Form a Gaussian-mixture likelihood (Probabilistic model of the data)
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- 4. Multiply the transition prior with the likelihood and obtain the posterior distribution.



JACS Presentation, FZJ, 17 September 2025