Comparison of the O_3 chemistry in the Po Valley with that in the Benelux region as simulated with MECO(n)

EGU 2022

Markus Kilian[1], Volker Grewe[1,3] , Patrick Jöckel[1], Astrid Kerkweg[2], and Mariano Mertens[1]

[1]Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre, Oberpfaffenhofen, Germany

[2]Institute of Energy and Climate Research, IEK-8: Troposphere, Forschungszentrum Jülich, Jülich, Germany

[3]Delft University of Technology, Faculty of Aerospace Engineering, Section Aircraft Noise and Climate Effects, Delft, The Netherlands

DLR-Institut for Atmospheric Physics,

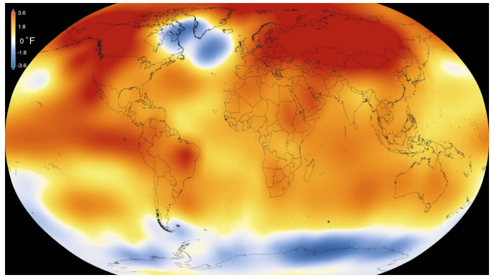
Oberpfaffenhofen

Table of Contents

Why tropospheric Ozone?

- Tropospheric O_3 affects air quality and contributes to global warming.
- O_3 is harmful to human health especially for the respiratory system.
- Tropospheric O_3 damages plants and affects agricultural production.

1.



2.

3.

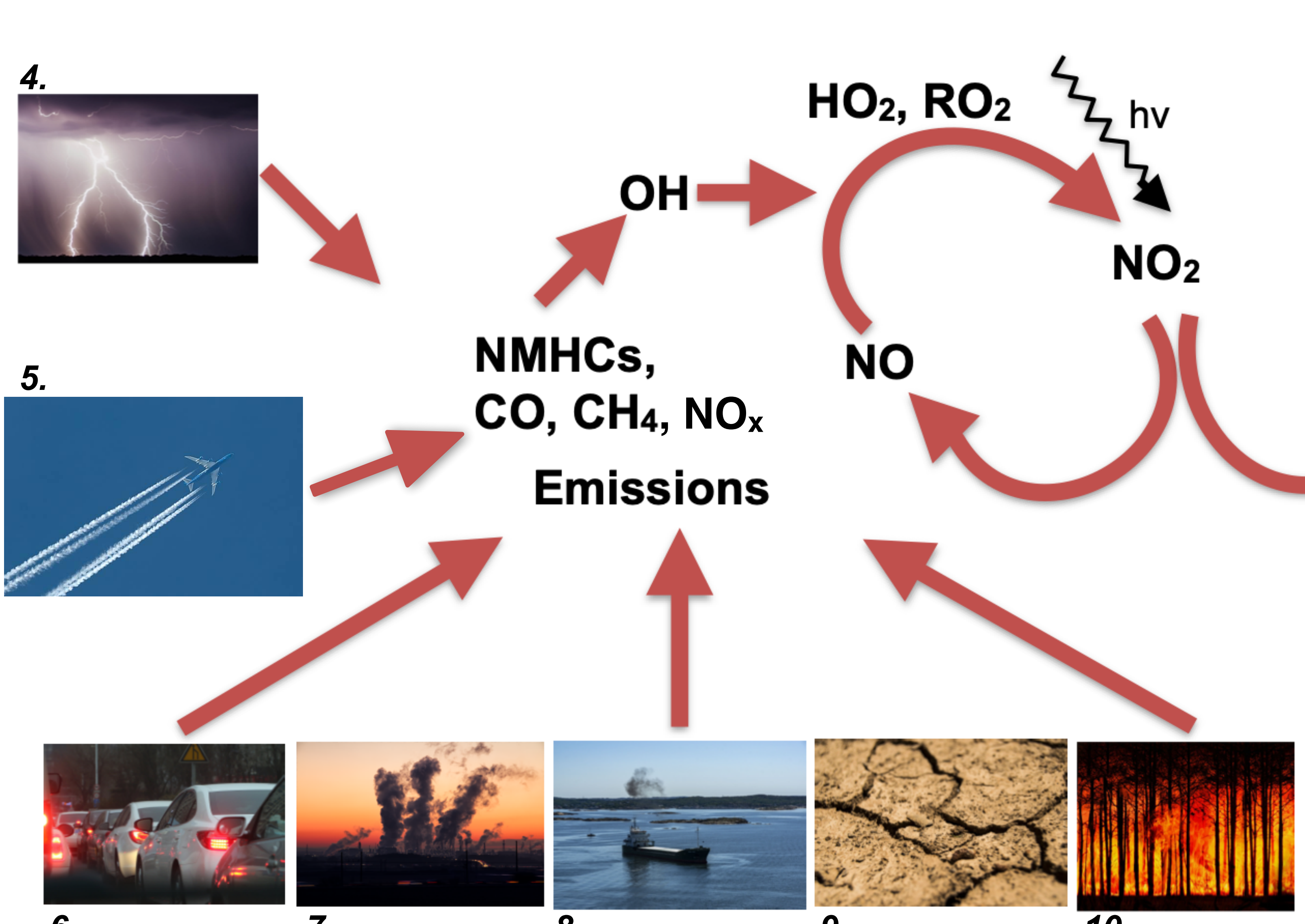
Sources:

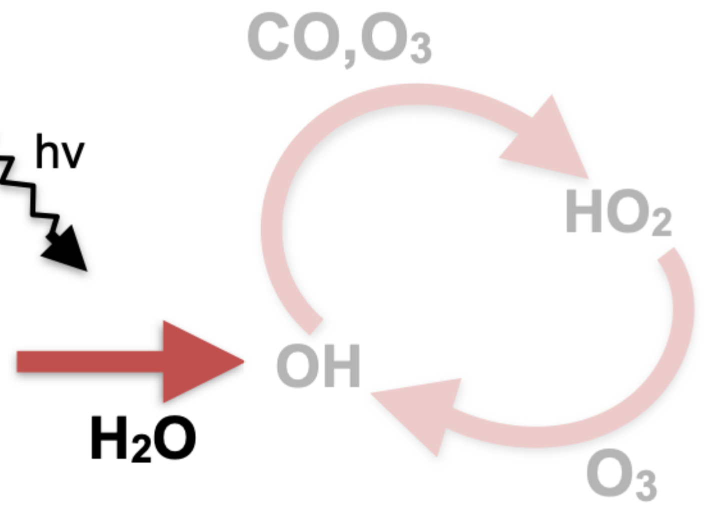
1. NASA

2. Environmental Agency of Zambia

3. University of Florida

Formation of tropospheric O_3





- Anthropogenic non-traffic (i.e. households, industry, etc.) and land transport emissions are important precursors of tropospheric O_3.
- Shown are the most important reactions

Sources: *4.-10. Open Source Stock Pictures*

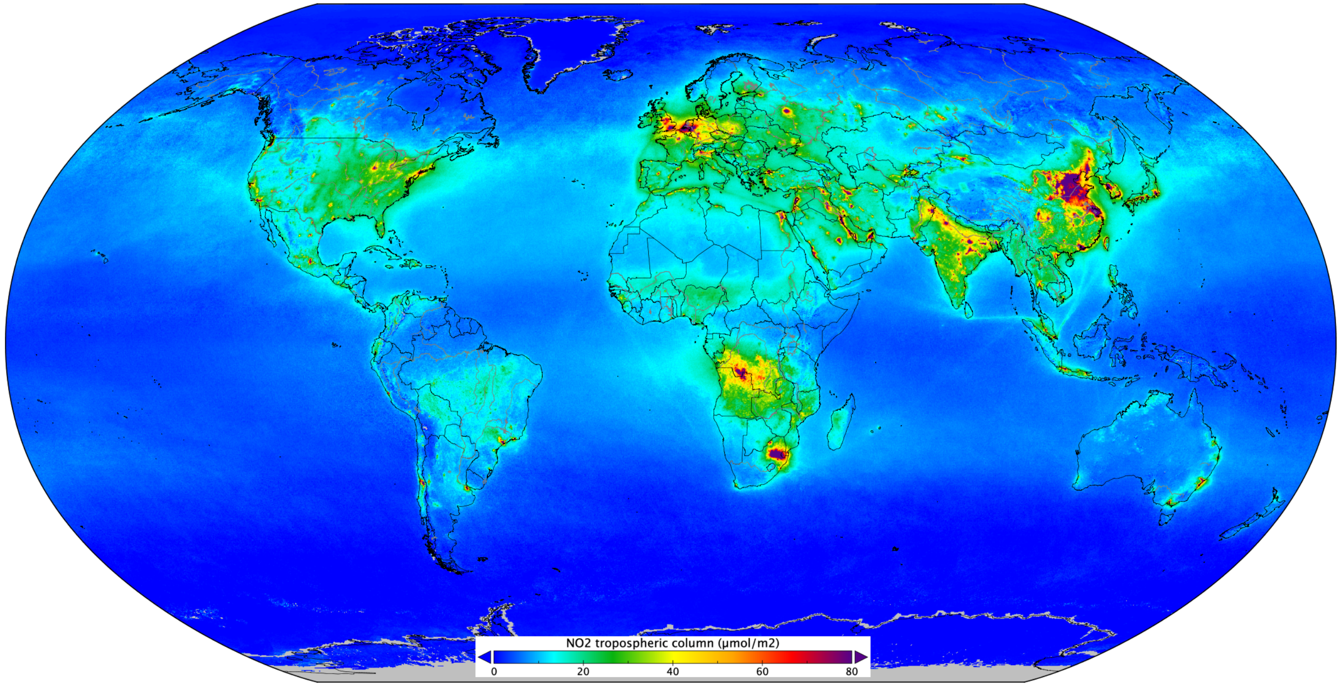
Where does NO_x come from?

- **$NO_x = NO + NO_2$**

Table 1: Estimate of Global Tropospheric NO_x Emissions in Tg N yr^{-1} for Year 2000

Sources	Emissions, Tg N yr^{-1}
Fossil Fuel Combustion	33.0
Aircraft	0.7
Biomass Burning	7.1
Soils	5.6
NH_3 Oxidation	--
Lightning	5.0
Stratosphere	<u><</u>0.5
Total	51.9

Reed et al. (2012)

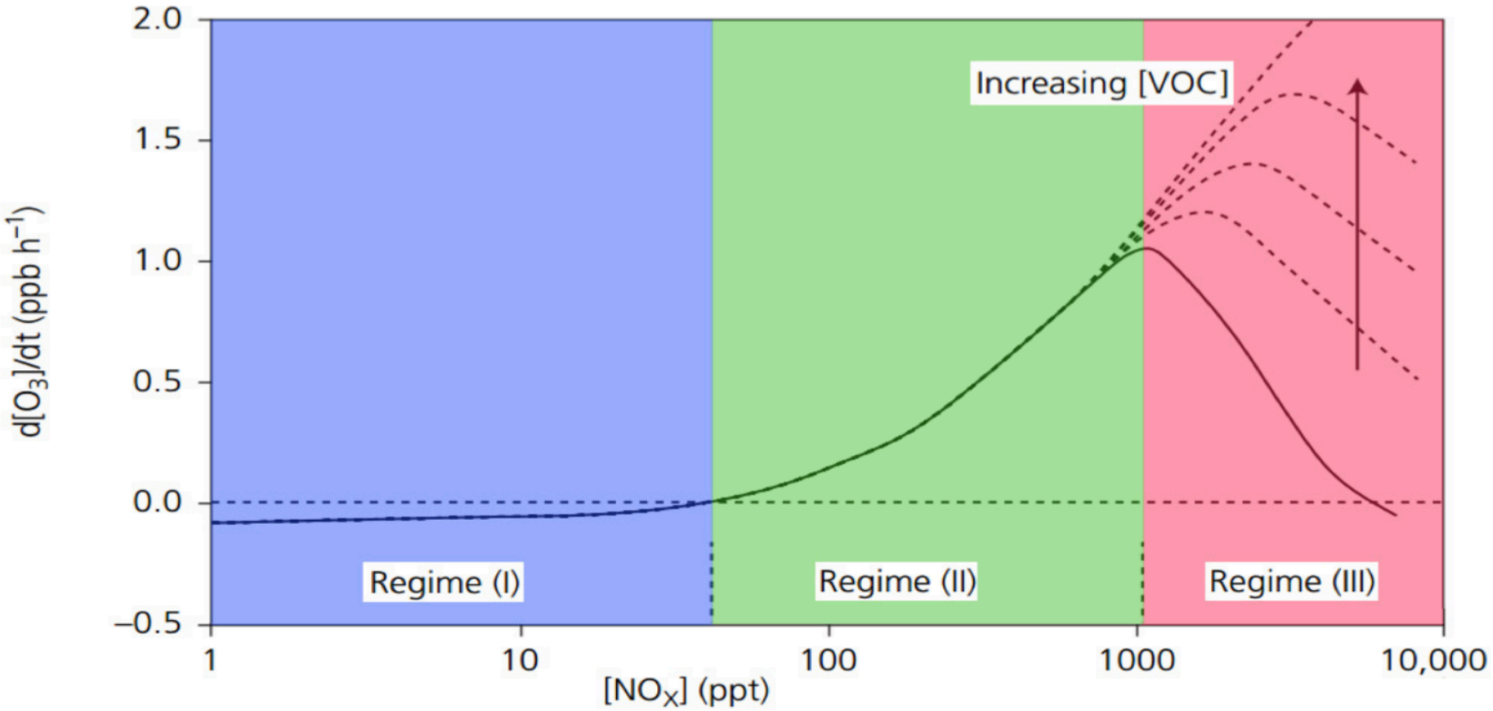


Sentinel 5P NO2 tropospheric column ESA (2019)

- Anthropogenic emissions have the largest share of the global NO_x totals.
- Hot spot regions of NO_x emissions are Central Europe, parts of China, Southern part of Africa and North America.

Nonlinearity of tropospheric O_3 formation

- Formation of tropospheric O_3 by precursor emissions is highly non-linear.
- Increasing NO_x emissions increase O_3 formation to certain threshold (NO_x limited regime; green area).
- Above threshold only VOC emissions can further increase the O_3 formation (VOC limited regime; red area).
- Source attribution methods to diagnose O_3 contributions are required (Grewe et al., 2010).



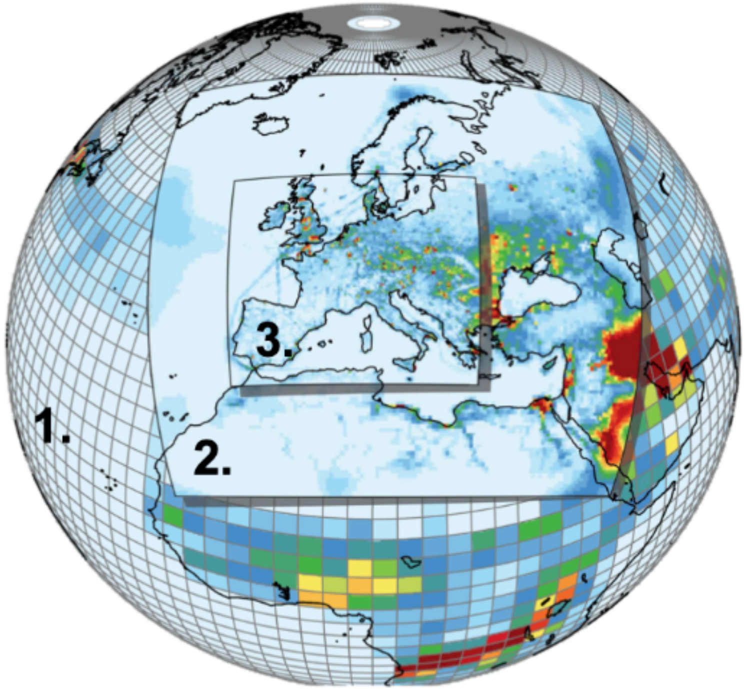
Non-linearity of the ozone formation (edited by Royal Society, 2008)

Scientific Questions

- How do the various emission sectors contribute to NO_y and O_3 in the Po Valley, and how does this differ in comparison with the Benelux region?
- How large are the contributions from European emissions compared to the contributions from long-range transported emissions to ground-level O_3?

MECO(n) model system

- **MECO(n):** „MESSy-fied ECHAM and COSMO nested n-times“
- online coupling of the global EMAC model with regional model COSMO/ MESSy
- Allows zooming in specific regions with fine resolution
- Applied source attribution to diagnose O_3 contributions of different sectors and different source regions.



**personal communication: Mariano Mertens*
Colour coded: NOx emissions

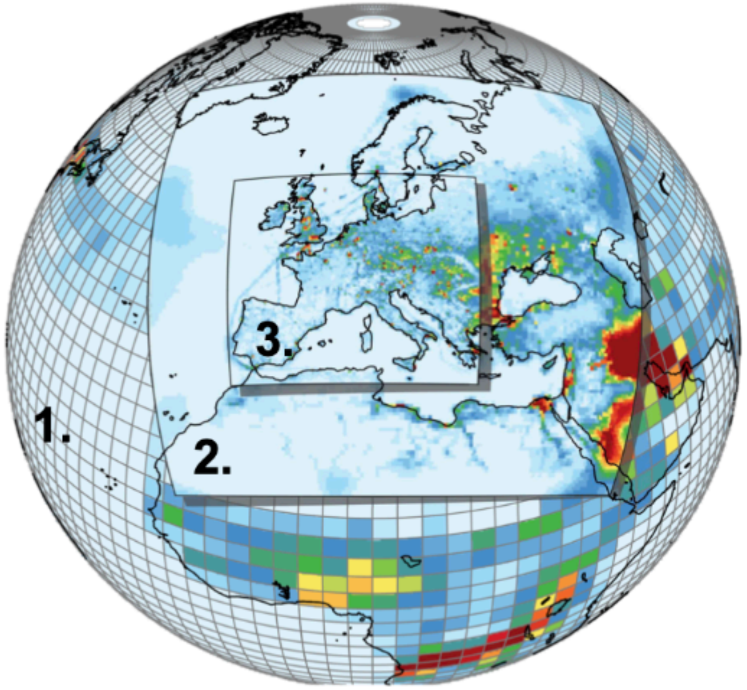
Model Setup MECO(2)

Setup:
- EMAC T42L90MA (ECHAM/MESSy Atmospheric Chemistry: ECHAM5, version 5.3.02; MESSy, version 2.55.2)
- Nudged against ERA Interim data

Simulation period:
- 01.12.2016-31.01.2019 (1st month spin-up)

Emission inventories:
- EDGAR 5.0 (2015)
- GFAS 1.2 (Bioburn)
- CCMI (GHG)

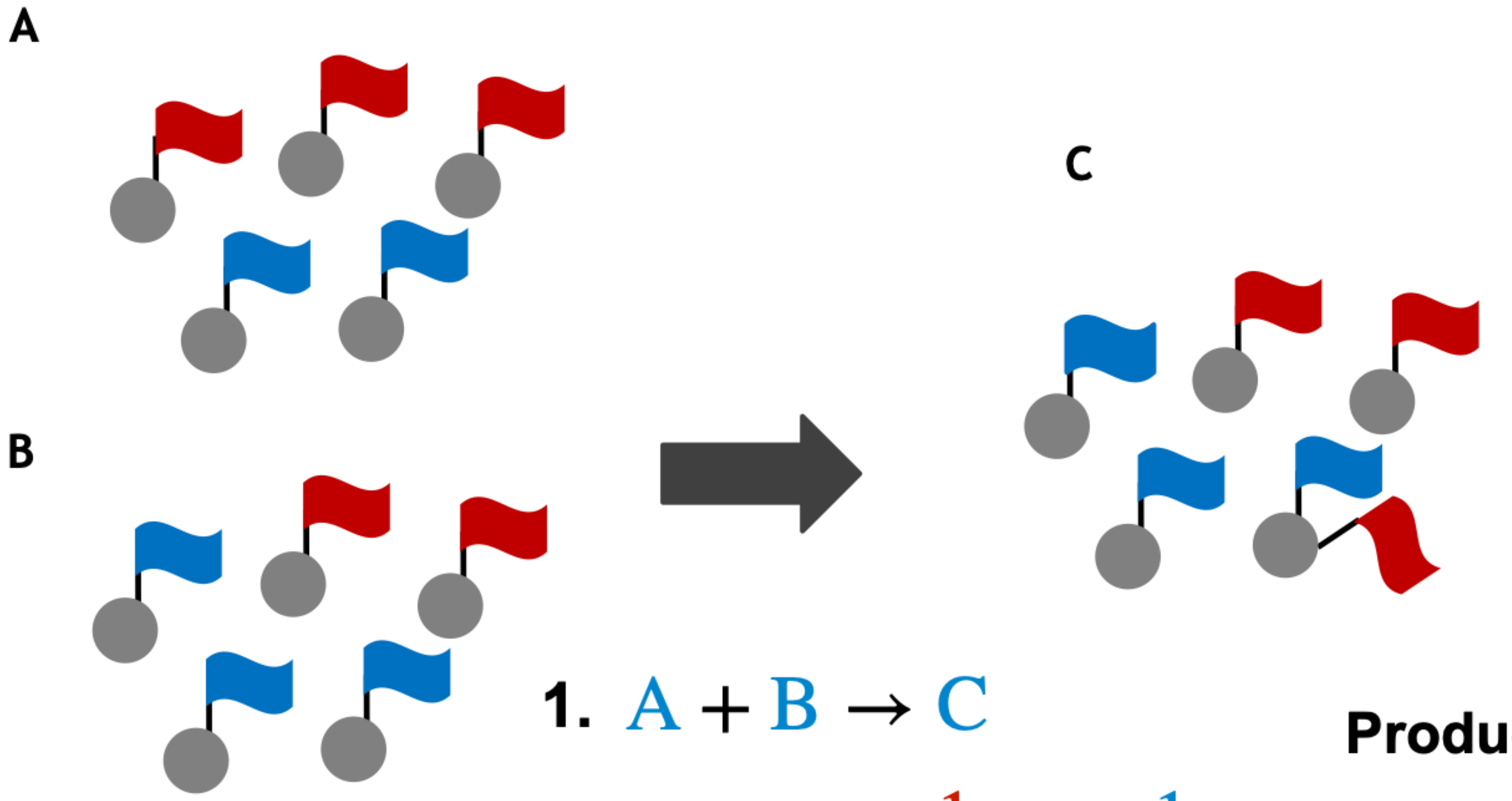
Focus:
- results are only shown for CM12



**personal communication: Mariano Mertens*
Colour coded: NOx emissions

1. EMAC T42L90MA
2. CM50 EU 0.44° x 0.44° (50km)
3. CM12 EU 0.11° x 0.11° (12km)

Tagging Method



emission sectors:
landtransport
industry

1. $A + B \rightarrow C$
2. $A + B \rightarrow \frac{1}{2} C + \frac{1}{2} C$
3. $A + B \rightarrow \frac{1}{2} C + \frac{1}{2} C$

Production

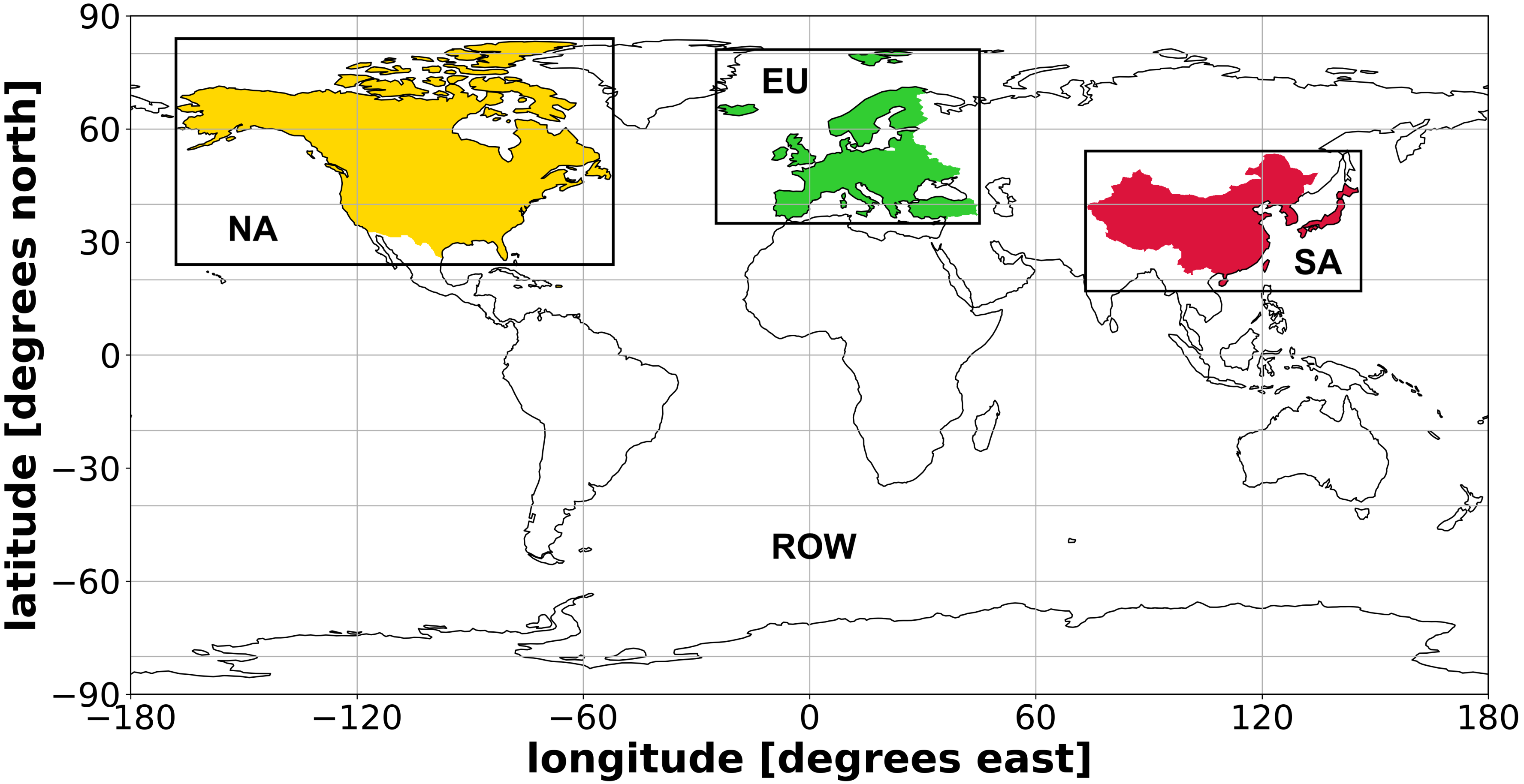
$$P(C^{tra}) = \frac{1}{2} k\ A\ B \left(\frac{A^{tra}}{A} + \frac{B^{tra}}{B} \right)$$

Tsati, 2014; Grewe et al., 2010, 2017 personal communication, Mertens and Rieger

DLR

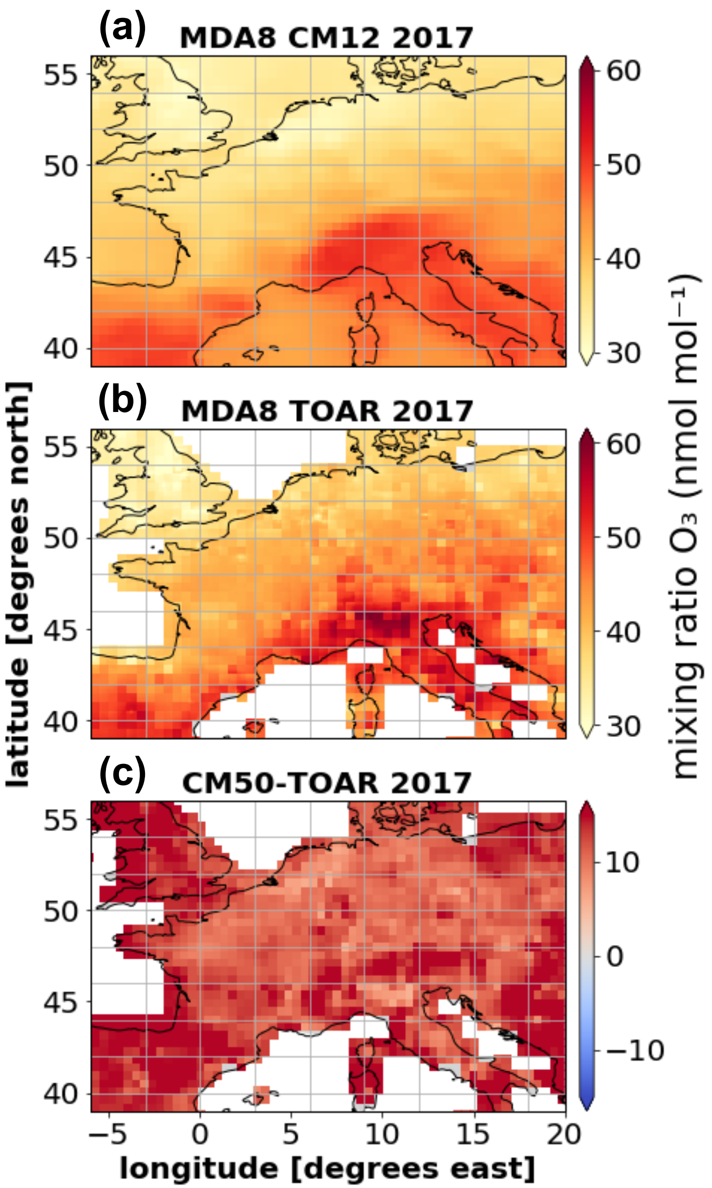
Source attribution by tagging regions

- We define three tagging regions: Europe (EU), North America (NA) and Southeast Asia (SA)
- The rest of the world (ROW) combines all remaining regions including the ocean.
- Enables the attribution by regional sources (i.e. same continent) and by sources from long-range transport.



Evaluation with TOAR/model dataset (D21)

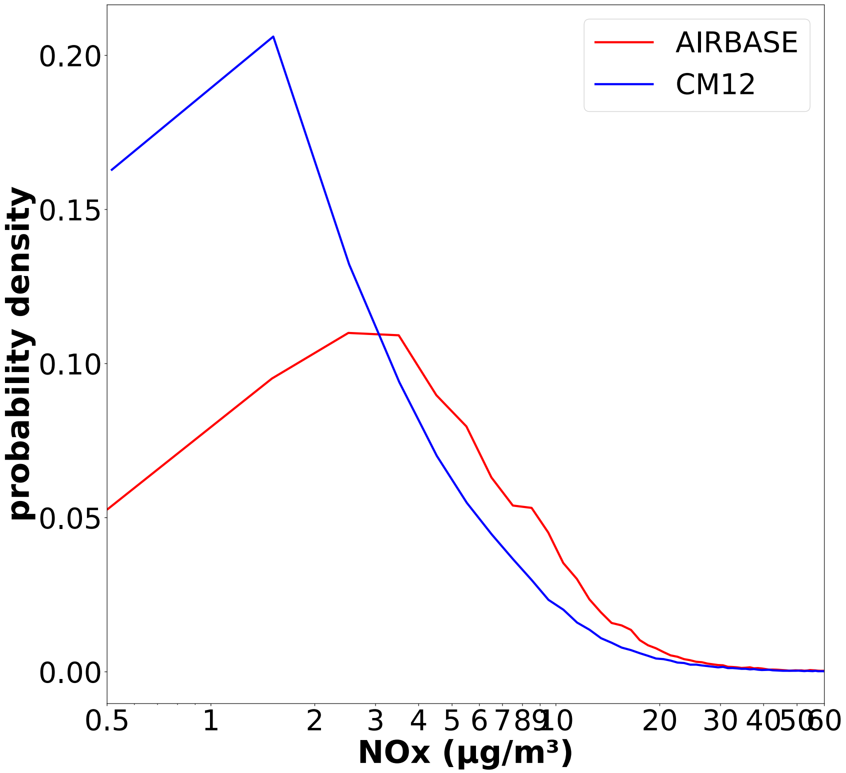
- D21 dataset based on TOAR and model data DeLang et al. (2021)
- **(a)** shows the de-biased (by 15 nmol mol^{-1}) ground-level O_3 seasonal daily maximum 8 h mixing ratio (OSDMA8) in nmol mol^{-1} as simulated by CM12
- **(b)** shows the OSDMA8 of the DeLang et al. (2019) dataset
- **(c)** difference of D21-CM12
- geographical distribution of ozone over Europe is well represented in MECO(n)
- O_3 systematically overestimated (bias ≈ 15 nmol mol^{-1})

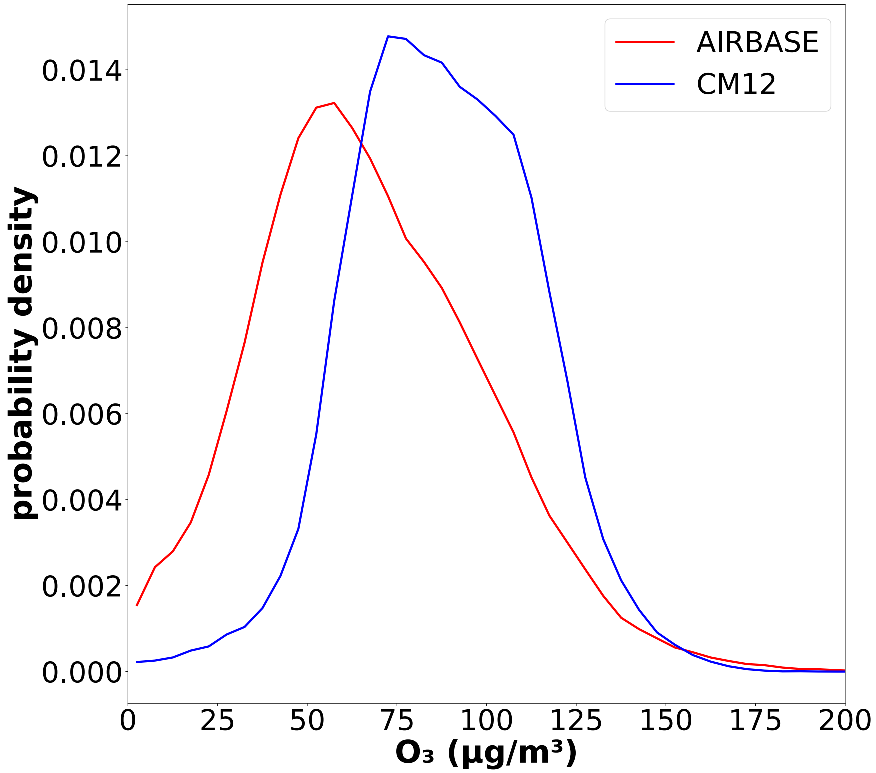


Evaluation with AIRBASE stations

- Comparison of the PDFs of ground-level NO_x and O_3 concentrations from CM12 with air quality stations.

- Selection of 419 rural stations throughout Europe.





- CM12 overestimates NO_x for small concentrations below 4 µg m^{-3}

- Underestimation for large NO_x concentrations

- CM12 overestimates O_3 throughout Europe

- Confirms O_3 bias of 20-25 µg m^{-3} (see slide 12)

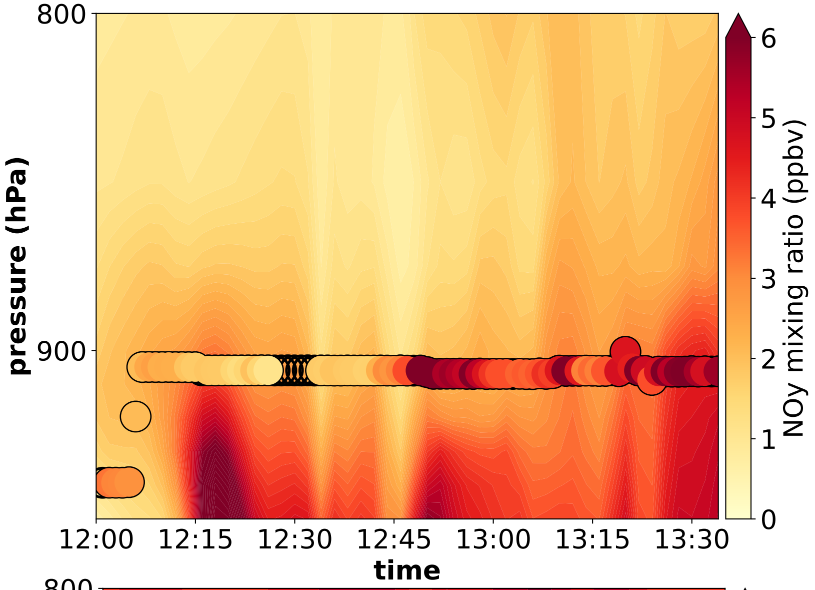
DLR

Evaluation with in situ data (EMeRGe)

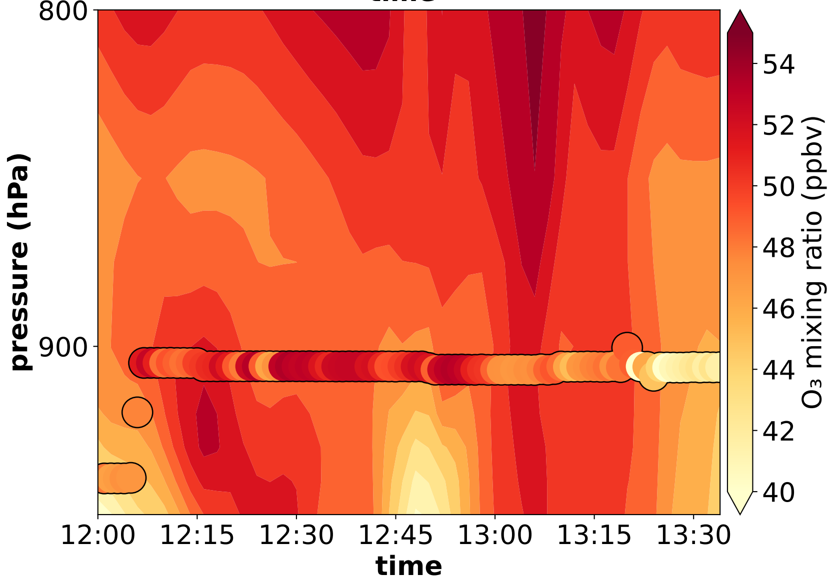
Benelux region: 26.07.2017

- Comparison between model results sampled along flight path of CM12 (background color) and in situ measurements for the Benelux-region (filled circles).
- NO_y mixing ratios in CM12 are underestimated within city plumes.
- Outside of major polluted areas NO_y is well represented.
- O_3 is well represented within city plumes but overestimated in more rural regions.

NO_y



O_3



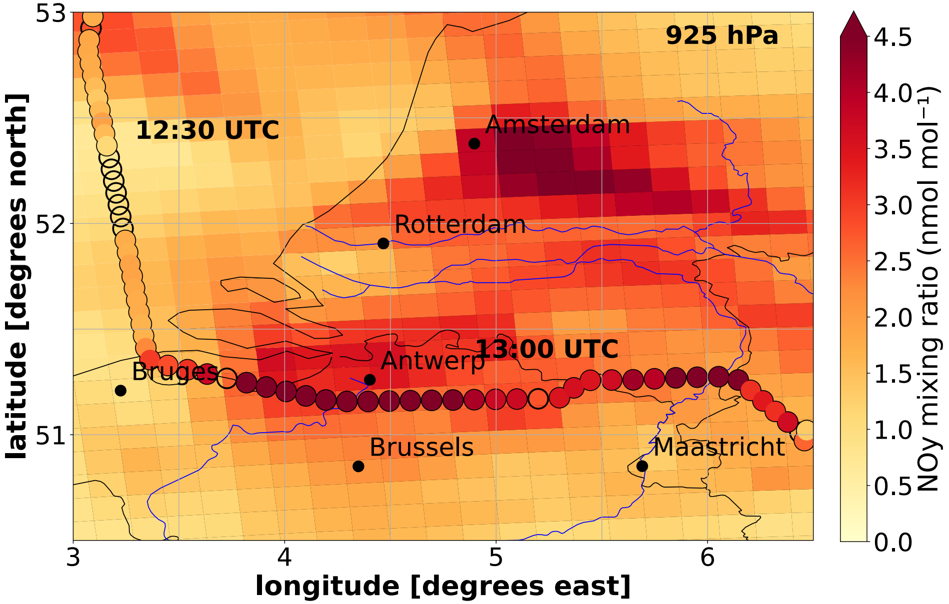
DLR

Evaluation with in situ data (EMeRGe)

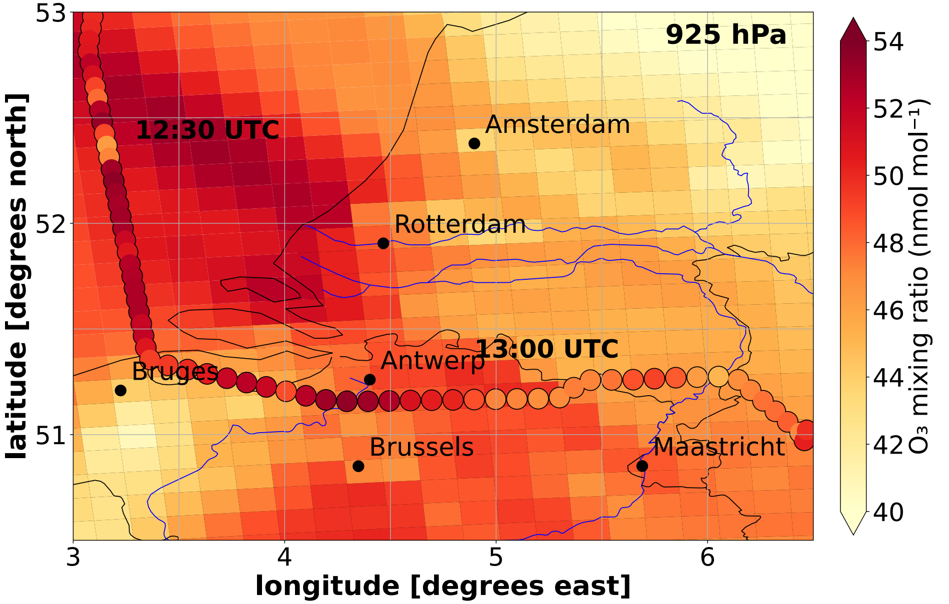
Benelux region: 26.07.2017

- Comparison between model results of CM12 (background color) and in situ measurements for the Benelux region(filled circles) at 925 hPa.
- Confirms the underestimation of NO_y mixing ratios in CM12 within city plumes (e.g. Antwerp).
- NO_y is well represented outside polluted areas (e.g. English Channel, see comparison slide 14).
- O_3 is mostly well represented outside city plumes, but underestimated within city plumes (e.g. between Bruges and Antwerp).

NO_y



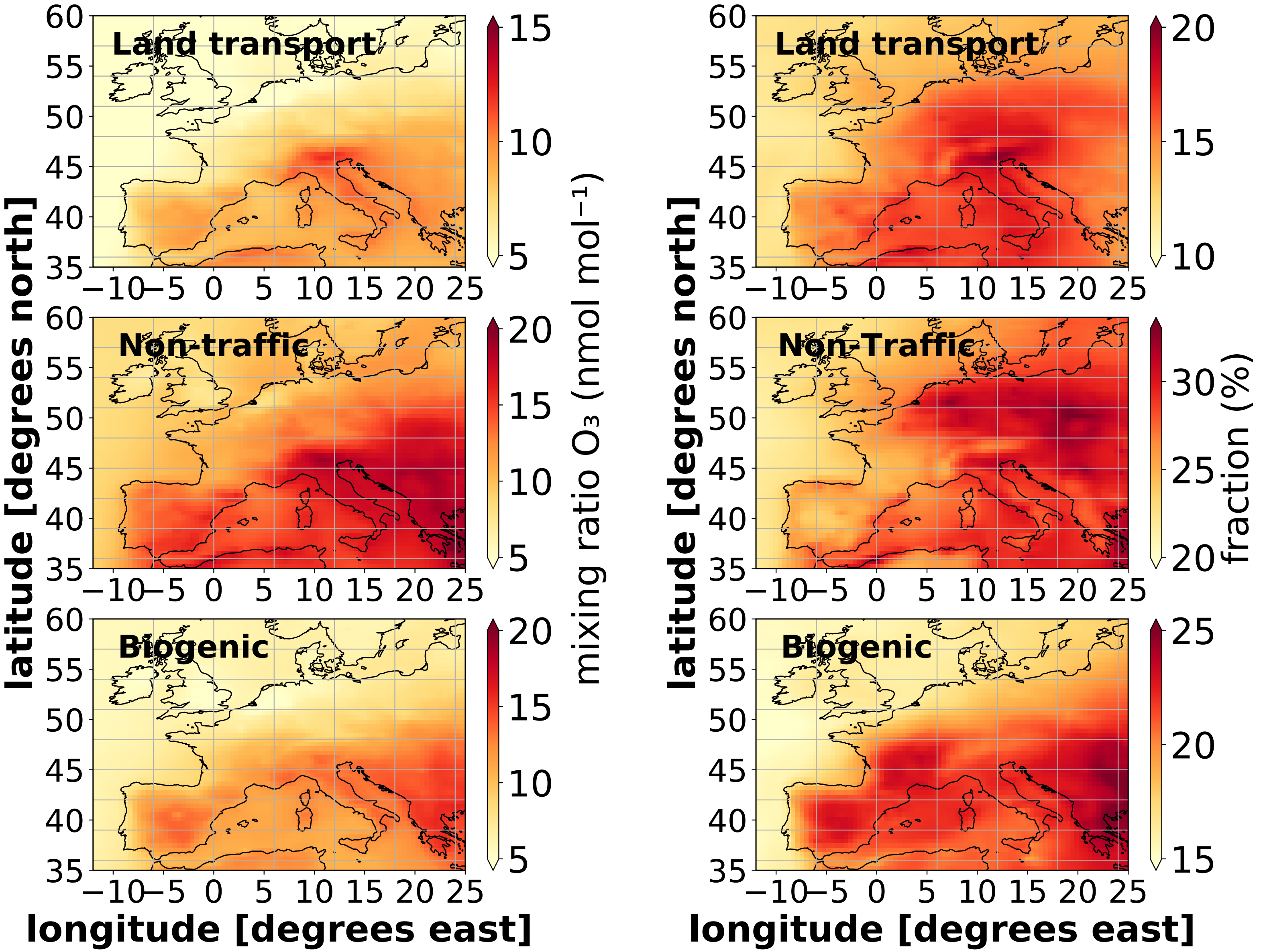
O_3



DLR

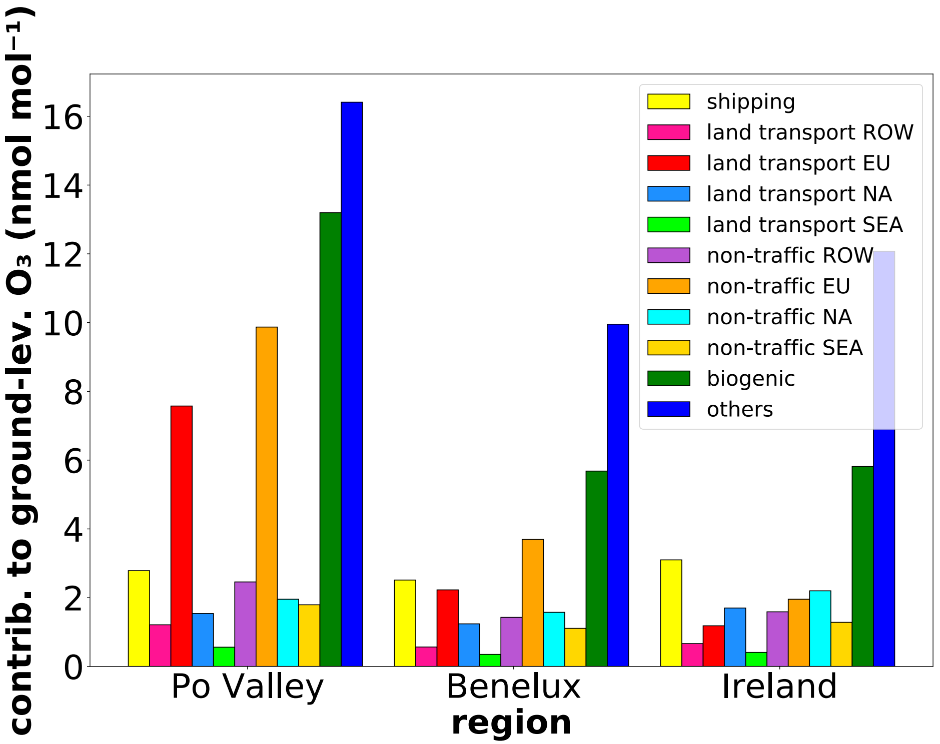
Ozone Contributions JJA 2017

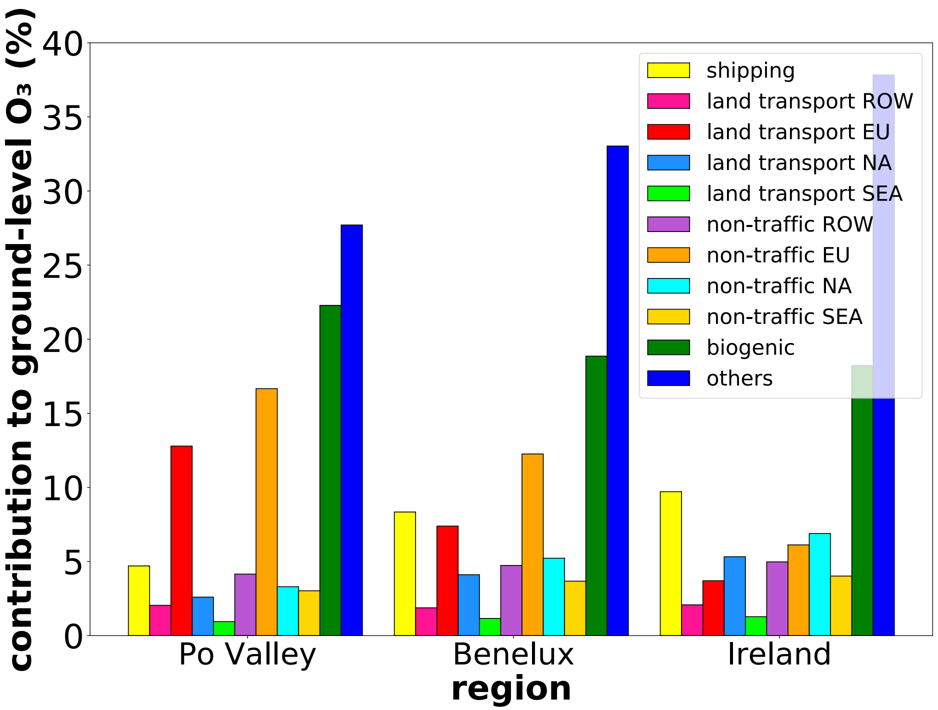
- Absolute O_3 contributions (left) have positive gradient in North-South direction.
- Anth. non-traffic sector is largest contributor to ground-level O_3 in Europe with up to 35 % during summer 2017.
- Biogenic sector is also important contributor to ground-level O_3 with up to 25 % especially in South Europe.



DLR

Ozone Contributions



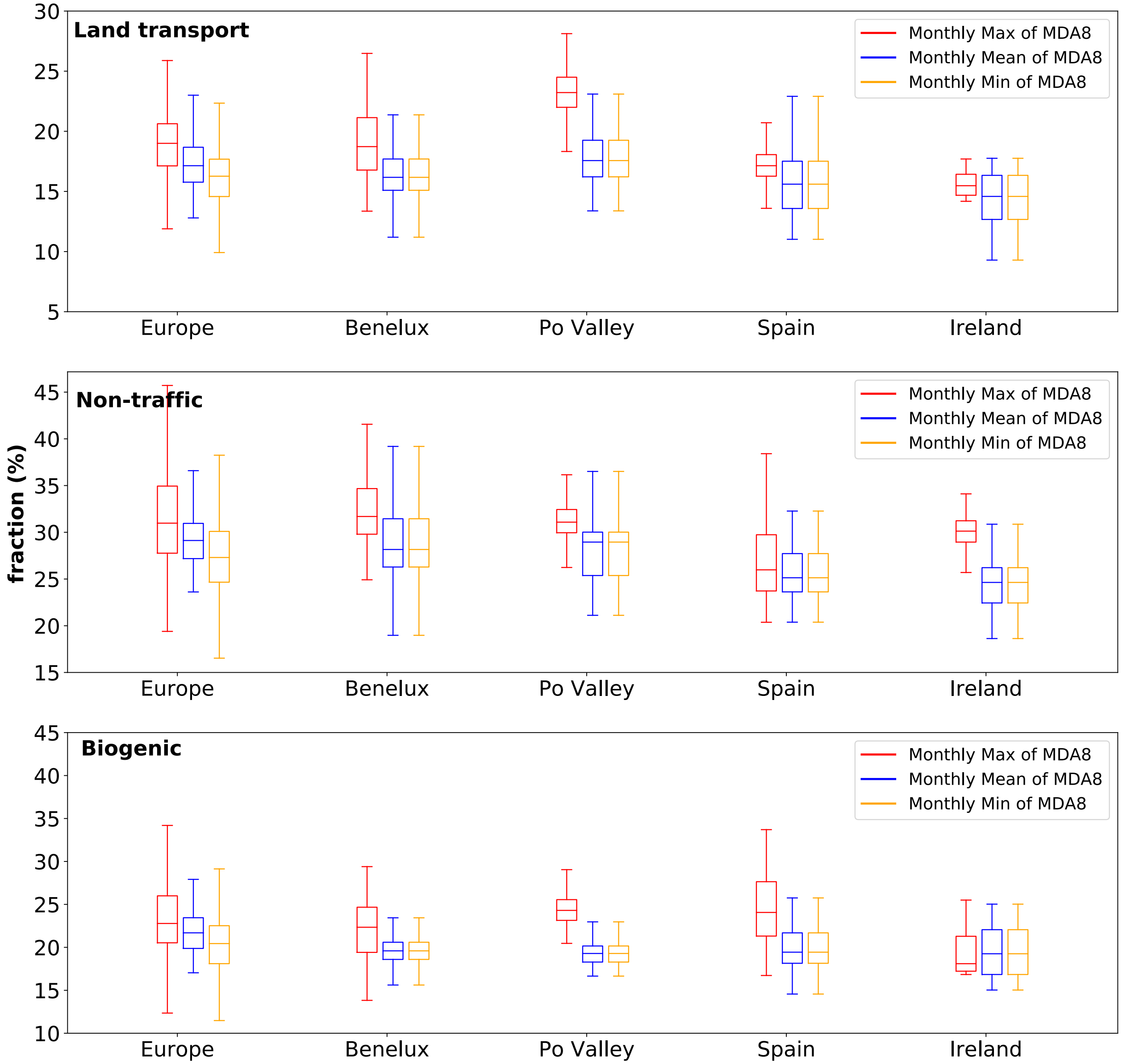


- Shown are absolute O_3 contributions (nmol mol^{-1}) as monthly mean for July 2017.
- In the Po Valley absolute O_3 contributions from European land transport and anth. non-traffic emissions are twice as large as in the Benelux region.

- Shown are relative O_3 contributions (%) as monthly mean for July 2017.
- Land transport and anth. non-traffic sector are the largest relative contributors to ground-level O_3 in the Po Valley and the Benelux region.

Ozone contributions during extreme ozone events

- Box-whisker plot showing the relative contributions of the most important emission sources of ozone as simulated by CM12 for July 2017.
- Shown are O_3 contributions for land transport, anthropogenic, and biogenic emissions to ground-level ozone during the monthly maximum (monthly mean and minimum) of the maximum daily 8-h average (MDA8), based on 1-hourly model output.
- During extreme ozone events (MDA8) O_3 contributions from anth. non-traffic emissions are up to 36 % in the Po Valley and 42 % in the Benelux region.
- The land transport sector contributes 26 % in the Benelux region and 28 % in the Po Valley to ground-level MDA8 O_3.



Summary: Po Valley vs. Benelux region

How do the various emission sectors contribute to NO_y and O_3 in the Po Valley, and how does this differ in comparison with the Benelux region?

- In the Po Valley abs. O_3 contributions from European land transport and anth. non-traffic emissions are twice as large as in the Benelux region.
- Land transport and anth. non-traffic sector contribute 13 % and 17 % to ground-level O_3 in the Po Valley; in the Benelux region 7 % and 12 %, respectively.
- The biogenic sector in the Po Valley contributes absolutely twice as much as in the Benelux region to ground-level O_3.
- Monthly mean O_3 contributions during extreme ozone events from anthropogenic emissions are larger in the Po Valley than in the Benelux region.

Summary: European vs. long-range transported

How large are the contributions from European emissions compared to the contributions from long-range transported emissions to ground-level O_3?

- In the Po Valley O_3 contributions from European anthropogenic emissions are significant smaller than from long-range transported sources.
- In the Benelux region this difference is rather small, caused by the vicinity to the direct inflow of emissions favored by westerly winds.
- Relative O_3 contributions from European land transport and anth. non-traffic emissions are 12.5 % and 17 % in the Po valley and 8 % and 12 % in the Benelux region, respectively.

Outlook

- Comparison of O_3 contribution between Europe and Southeast Asia is planned.
- Source regions for tagging could be defined with a finer resolution, e.g. country-by-country.
- Analyses of uncertainties due to natural emissions are ongoing.
- Publication is ready for submission to Atmospheric Chemistry and Physics (ACP).

References

Bieser, Johannes & Aulinger, A. & Matthias, Volker & Quante, M. & Builtjes, P.J.H.. (2010). SMOKE for Europe – adaptation, modification and evaluation of a comprehensive emission model for Europe. Geoscientific Model Development Discussions. 3. 10.5194/gmdd-3-949-2010.

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys., 6, 4321–4344, doi:10.5194/acp-6-4321-2006, 2006.

Grewe, V., Tsati, E., and Hoor, P.: On the attribution of contributions of atmospheric trace gases to emissions in atmospheric model applications, Geosci. Model Dev., 3, 487–499, https://doi.org/10.5194/gmd-3-487-2010, 2010.

Grewe, V. & Dahlmann, Katrin & Matthes, Sigrun & Steinbrecht, Wolfgang. (2012). Attributing ozone to NOx emissions: Implications for climate mitigation measures. Atmospheric Environment. 59. 102–107. 10.1016/j.atmosenv.2012.05.002.

Grewe, V., Tsati, E., Mertens, M., Frömming, C., and Jöckel, P., Contribution of emissions to concentrations: The TAGGING 1.0 submodel based on the Modular Earth Submodel System (MESSy 2.52), Geosci. Model Dev. 10, 2615-2633, doi:10.5194/gmd-2016-298, 2017.

Kerkweg, Astrid und Jöckel, Patrick (2012) *The 1-way on-line coupled atmospheric chemistry model system MECO(n) Part 1: Description of the limited-area atmospheric chemistry model COSMO/MESSy.* Geoscientific Model Development, 5, Seiten 87-110. Copernicus Publications. DOI: 10.5194/gmd-5-87-2012.

Reed, Andra J.. “A Comparison of Satellite and Ground-based Retrievals of Total Column Ozone and Nitrogen Dioxide, During DISCOVER-AQ.” (2012).

Tsati, Eleni-Eugenia (2014): Investigation of the impacts of emissions on the trace gas budgets in the troposphere by using global climate chemistry model simulations. Dissertation, LMU München: Fakultät für Physik

Vinken, Geert & Boersma, Klaas & D. Maasakkers, J & Adon, Marcellin & Martin, Randall. (2014). Worldwide biogenic soil NOx emissions inferred from OMI NO2 observations. Atmospheric Chemistry and Physics Discussions. 14. 14683-14724. 10.5194/acpd-14-14683-2014.

DLR