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

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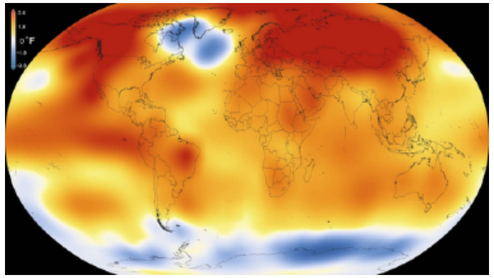
Why tropospheric Ozone?

- 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.

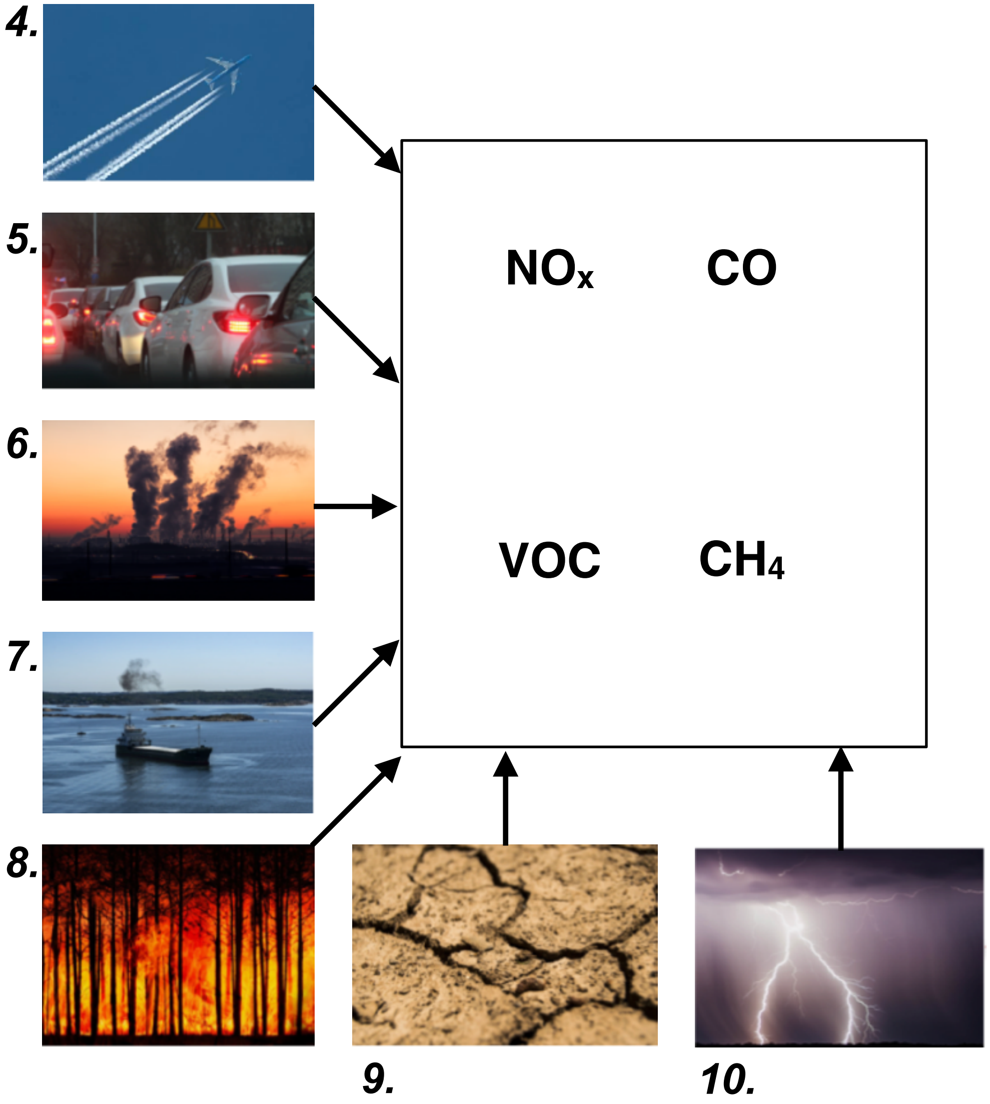
Sources:

1. NASA

2. Environmental Agency of Zambia

3. University of Florida

Tropospheric Ozone formation



*$OH + CO \rightarrow HOCO$

$HOCO + O_2 \rightarrow HO_2 + CO_2$

*oxidation of CO by OH radical

**$HO_2 + NO \rightarrow OH + NO_2$

$NO_2 + h\nu \rightarrow NO + O(^3P)$, $\lambda < 400$ nm

$O(^3P) + O_2 \rightarrow O_3$

**note that these three reactions are what forms the ozone molecule, and will occur the same way in the oxidation of CO or VOCs case.

- Anthropogenic non-traffic (i.e. households, industry, etc.) and land transport emissions are important precursors of tropospheric O_3.
- Hot spot regions of NO_x emissions are Central Europe, parts of China, Southern part of Africa and North America.

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

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 and Setup

- **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 emission sectors and different source regions, because formation of tropospheric O_3 by precursor emissions is highly non-linear.

Simulation period for MECO(2):

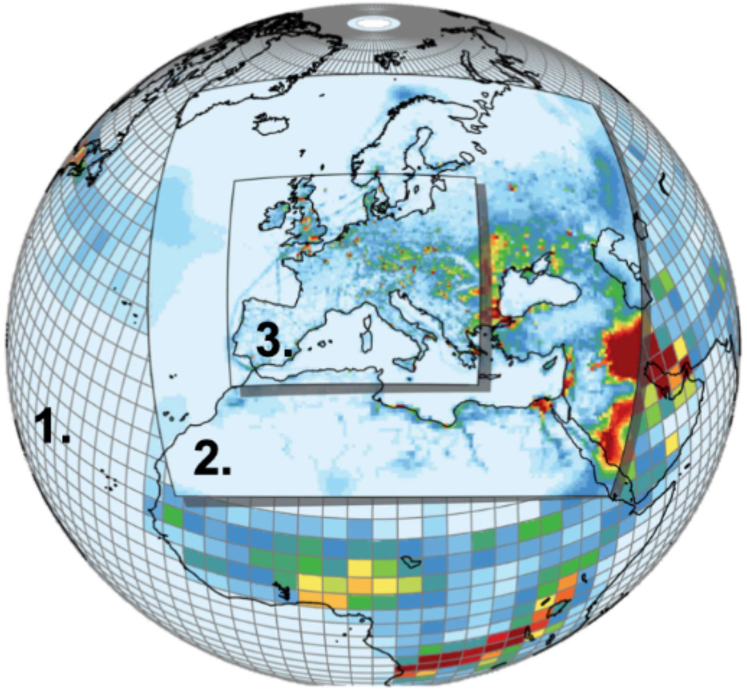
- 01.12.2016 - 31.01.2019 (1st month spin-up)

Emission inventories:

- EDGAR 5.0 (2015)
- GFAS 1.2 (Bioburn)
- CCMI (GHG)

Study Areas:

- Focus on the Benelux region, the Po Valley and Ireland

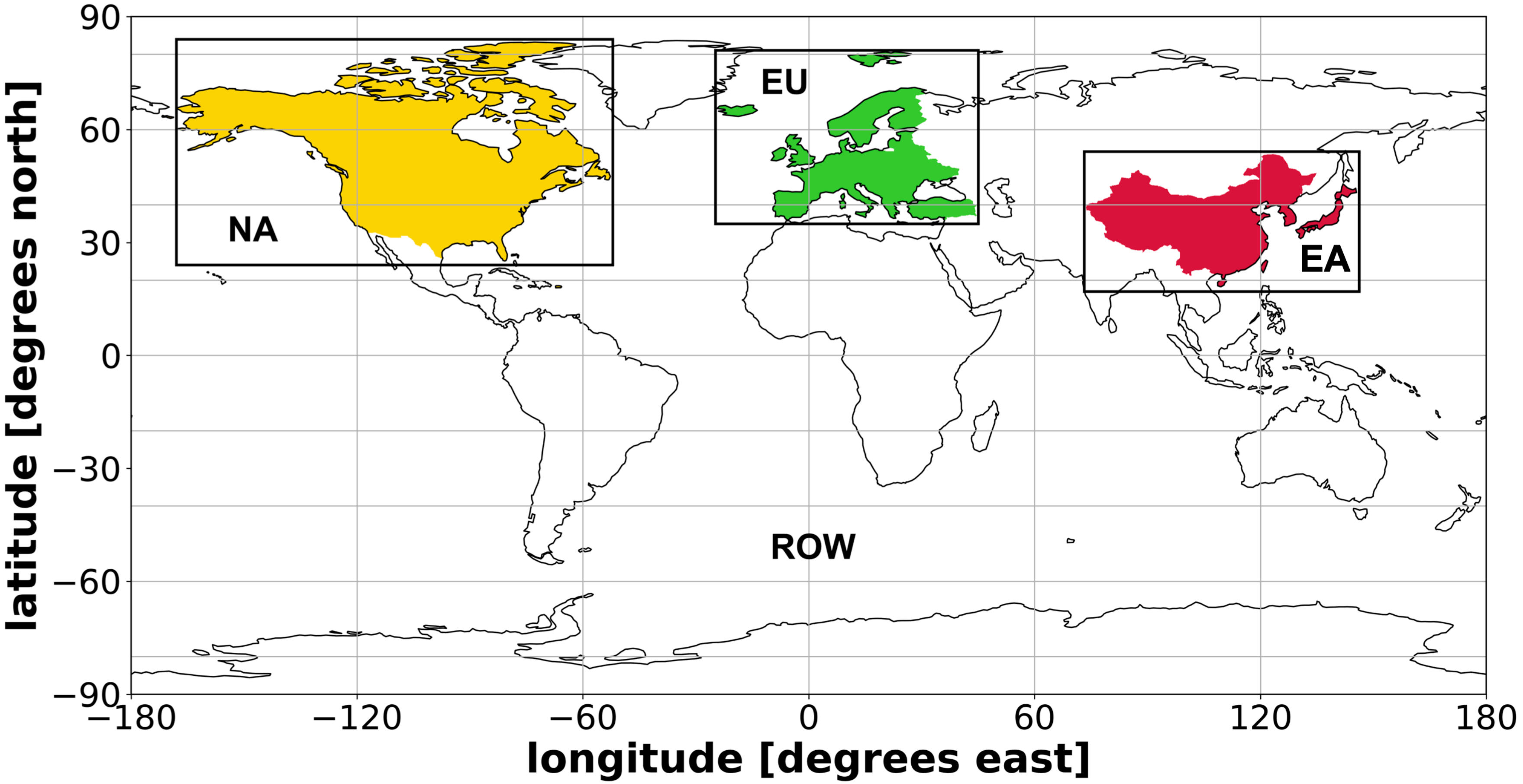


Colour coded: NOx emissions

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

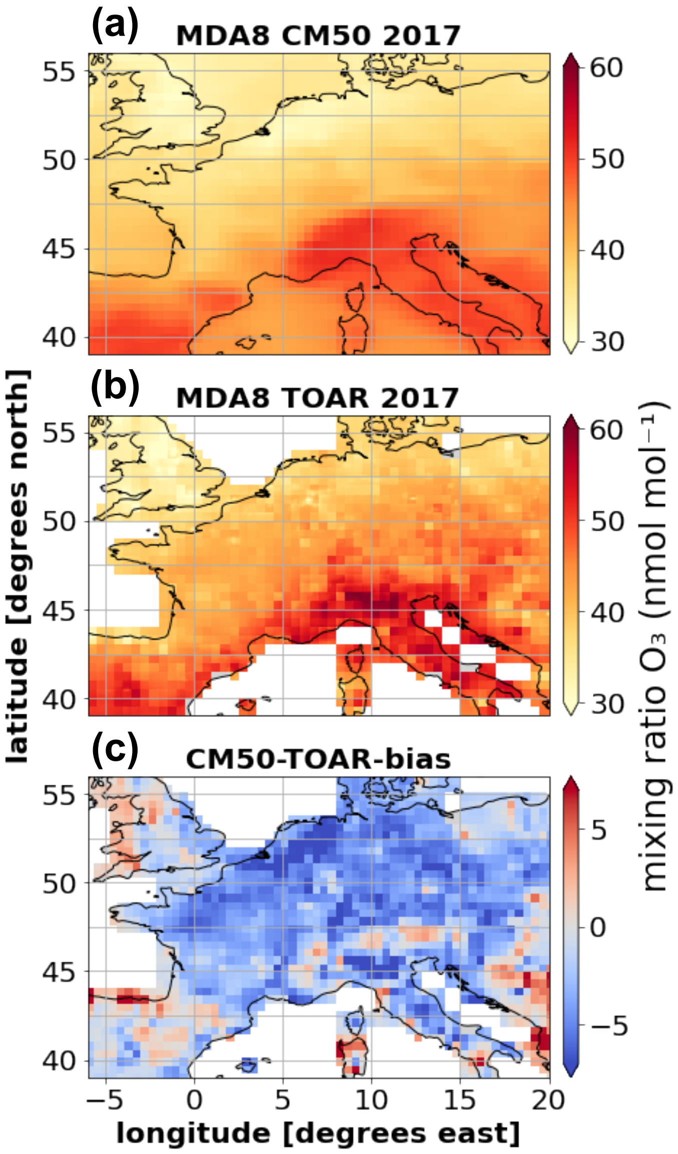
Source attribution by tagging regions

- We define three tagging regions: Europe (EU), North America (NA) and East Asia (EA)
- 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 area mean bias of 15 nmol mol^{-1}) ground-level O_3 seasonal daily maximum 8 h mixing ratio (OSDMA8) in nmol mol^{-1} as simulated by CM50
- **(b)** shows the OSDMA8 of the DeLang et al. (2019) dataset
- **(c)** difference of D21-CM50-bias
- geographical distribution of ozone over Europe is well represented in MECO(n)
- O_3 systematically overestimated in rural regions and underestimated in urban areas



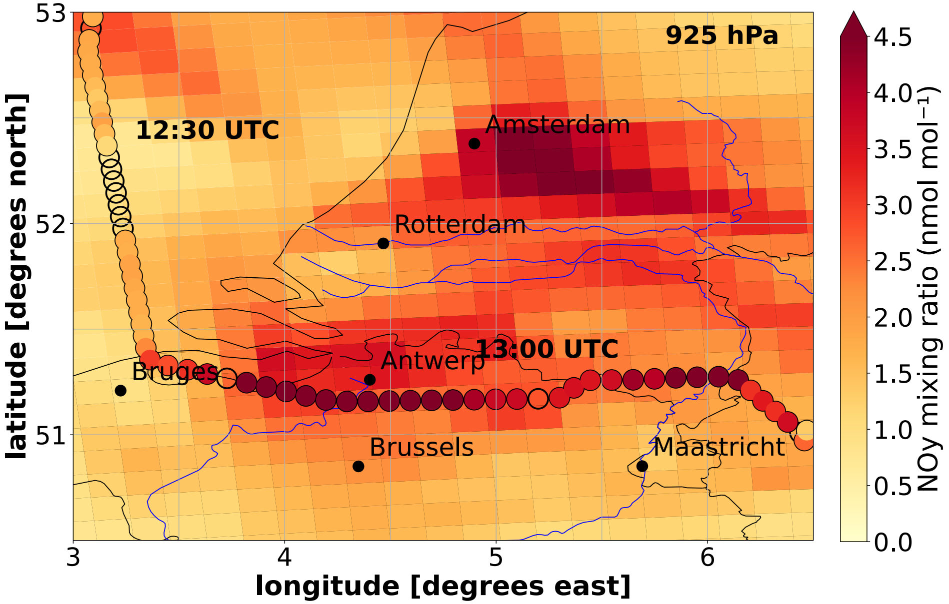
Evaluation with in situ data (EMeRGe)

Benelux region: 26.07.2017

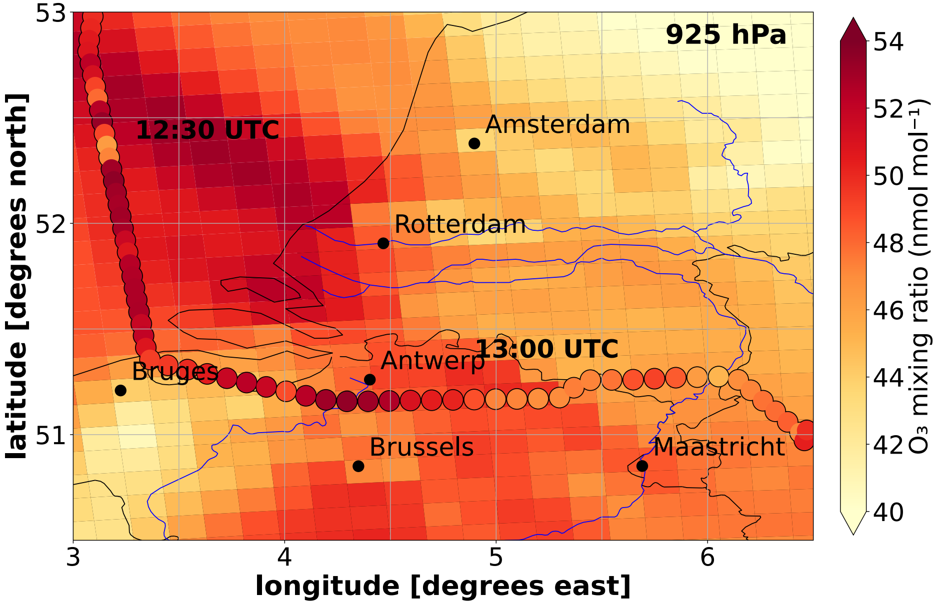
Comparison between model results of CM12 (background color) and in situ measurements for the Benelux region (filled circles) at 925 hPa:

- 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)
- O_3 is mostly well represented outside city plumes, but underestimated within city plumes (e.g. between Bruges and Antwerp).

CM12 NO_y



CM12 O_3



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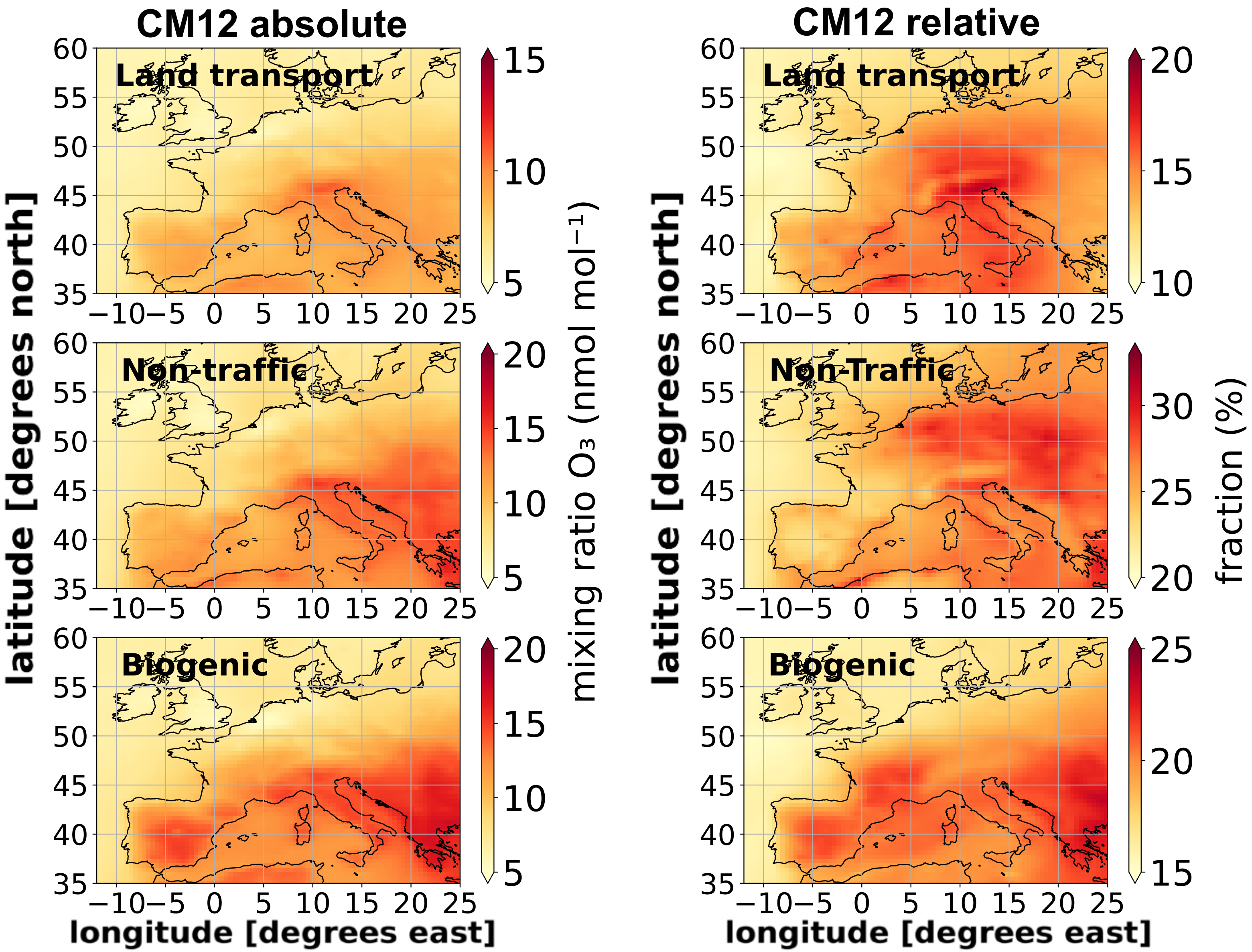
Ozone Contributions JJA 2017

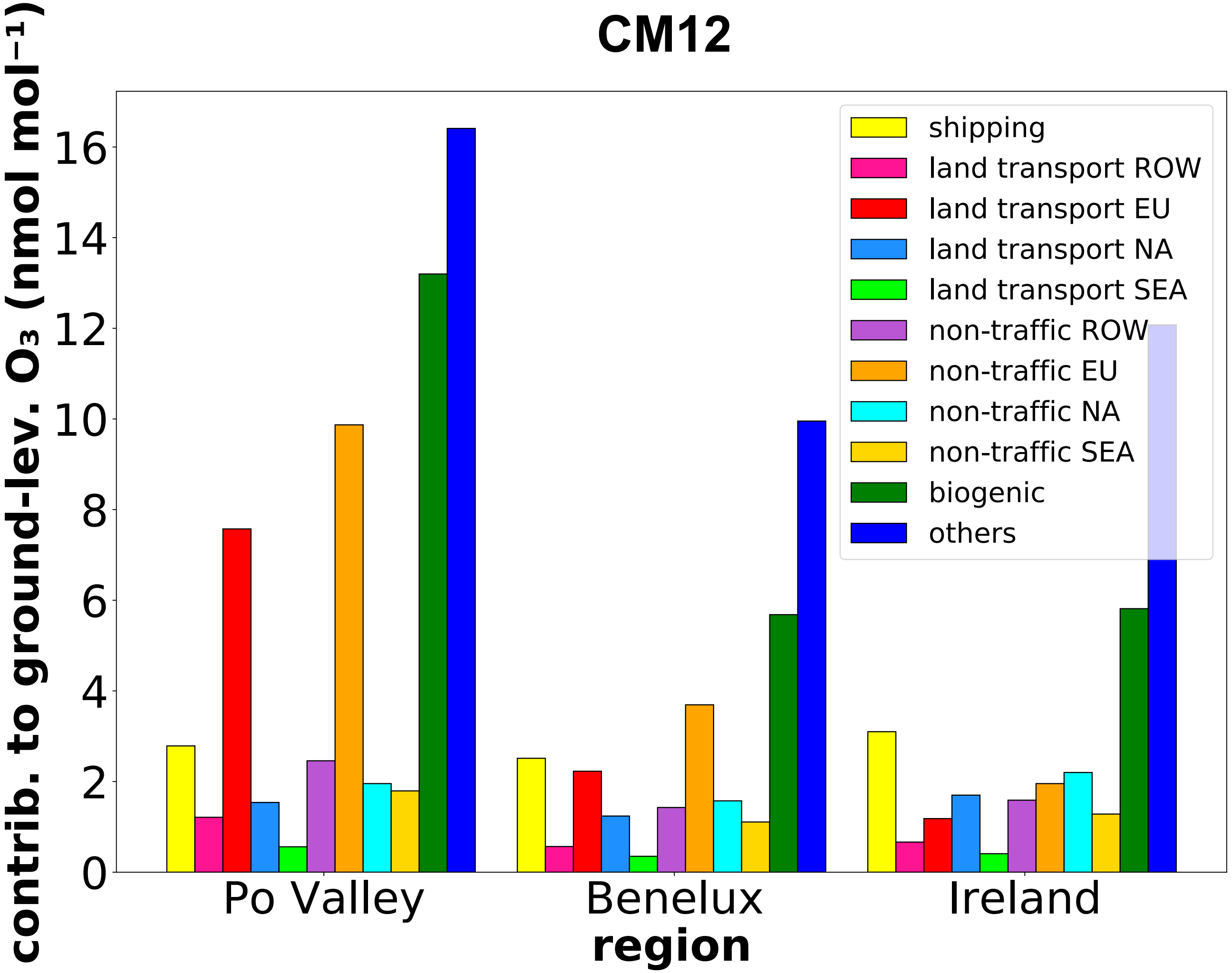
- positive gradient of absolute O_3 contributions (left) in North-South direction
- Anth. non-traffic sector is largest contributor to ground-level O_3 in Europe with up to 35 % during summer 2017.
- Biogenic sector is also important contributor to ground-level O_3 with up to 25 % especially in South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



Ozone Contributions

Shown are absolute O_3 contributions (nmol mol^{-1}) as monthly mean for July 2017:

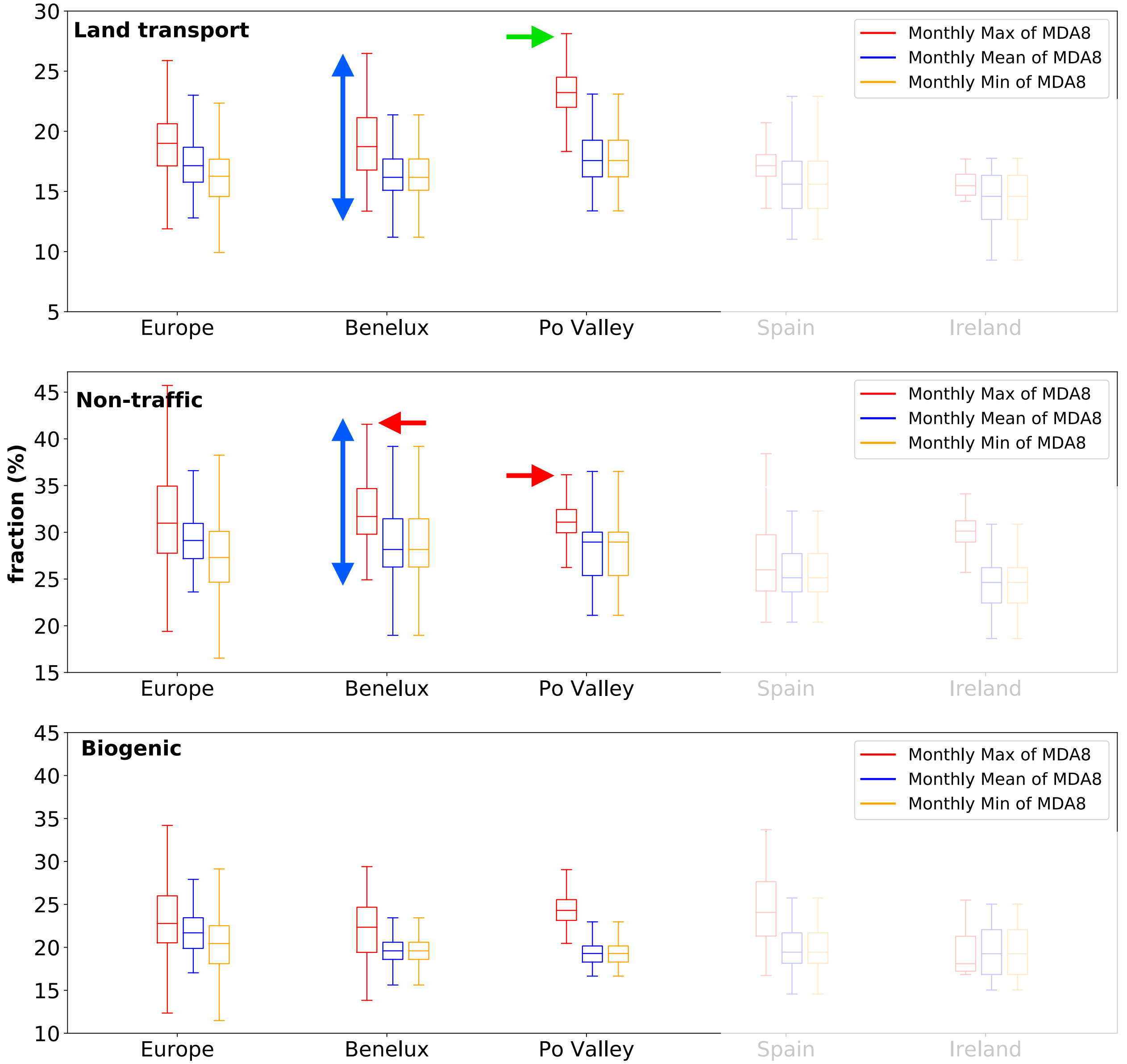
- In the Po Valley absolute O_3 contributions from European land transport and anth. non-traffic emissions are twice as large as in the Benelux region.
- Anthropogenic O_3 contributions from long-range transported precursors have the same order of magnitude in all three regions.



Ozone contributions during extreme ozone events

Shown are monthly maximum MDA8 O3 contributions for land transport, anthropogenic, and biogenic emissions to ground-level ozone as simulated by CM12 for July 2017:

- Benelux region has the largest geographical spread of the O3 contributions from anth. emissions
- The largest O3 contributions (during max. MDA8) from anth. non-traffic emissions are up to 36 % in the Po Valley and 42 % in the Benelux region.
- The largest O3 contributions from land transport emissions arises in the Po Valley to maximum of MDA8 ozone.



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?

- Po Valley: O_3 contributions from European anthropogenic emissions twice as large as in Benelux region
- Biogenic emissions: contribute twice as much as in the Benelux region
- extreme ozone events are dominated by anthropogenic emissions and are largest in the Po Valley

Tagging Sector	Benelux region	Po Valley
Land transport	4 nmol $nmol^{-1}$(14 %)	11 nmol $nmol^{-1}$(18 %)
Anth. Non-traffic	9 nmol $nmol^{-1}$(25 %)	17 nmol $nmol^{-1}$(26 %)
Biogenic	6 nmol $nmol^{-1}$(17 %)	13 nmol $nmol^{-1}$(22 %)

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?

- Po Valley: O_3 chemistry is dominated by European anthropogenic emissions and less by long-range transported sources.
- Benelux: In-situ production and long-range transport are both important

Tagging Sector	Benelux region	Po Valley
Land transport EU	2 nmol nmol^{-1}(8 %)	8 nmol nmol^{-1}(13 %)
Land transport LRT	2 nmol nmol^{-1}(6 %)	3 nmol nmol^{-1}(5 %)
Anth. Non-traffic EU	4 nmol nmol^{-1}(15 %)	10 nmol nmol^{-1}(17 %)
Anth. Non-traffic LRT	4 nmol nmol^{-1}(10 %)	6 nmol nmol^{-1}(9 %)

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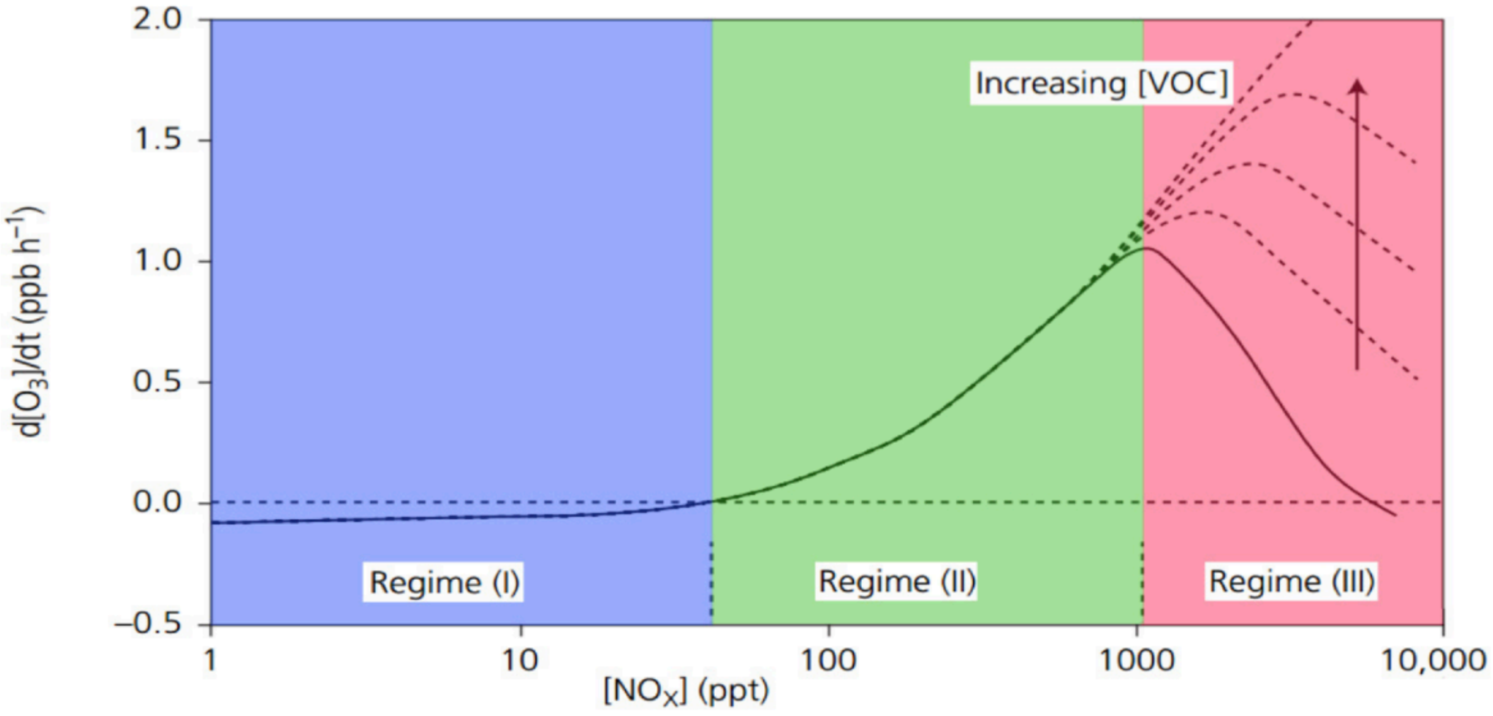
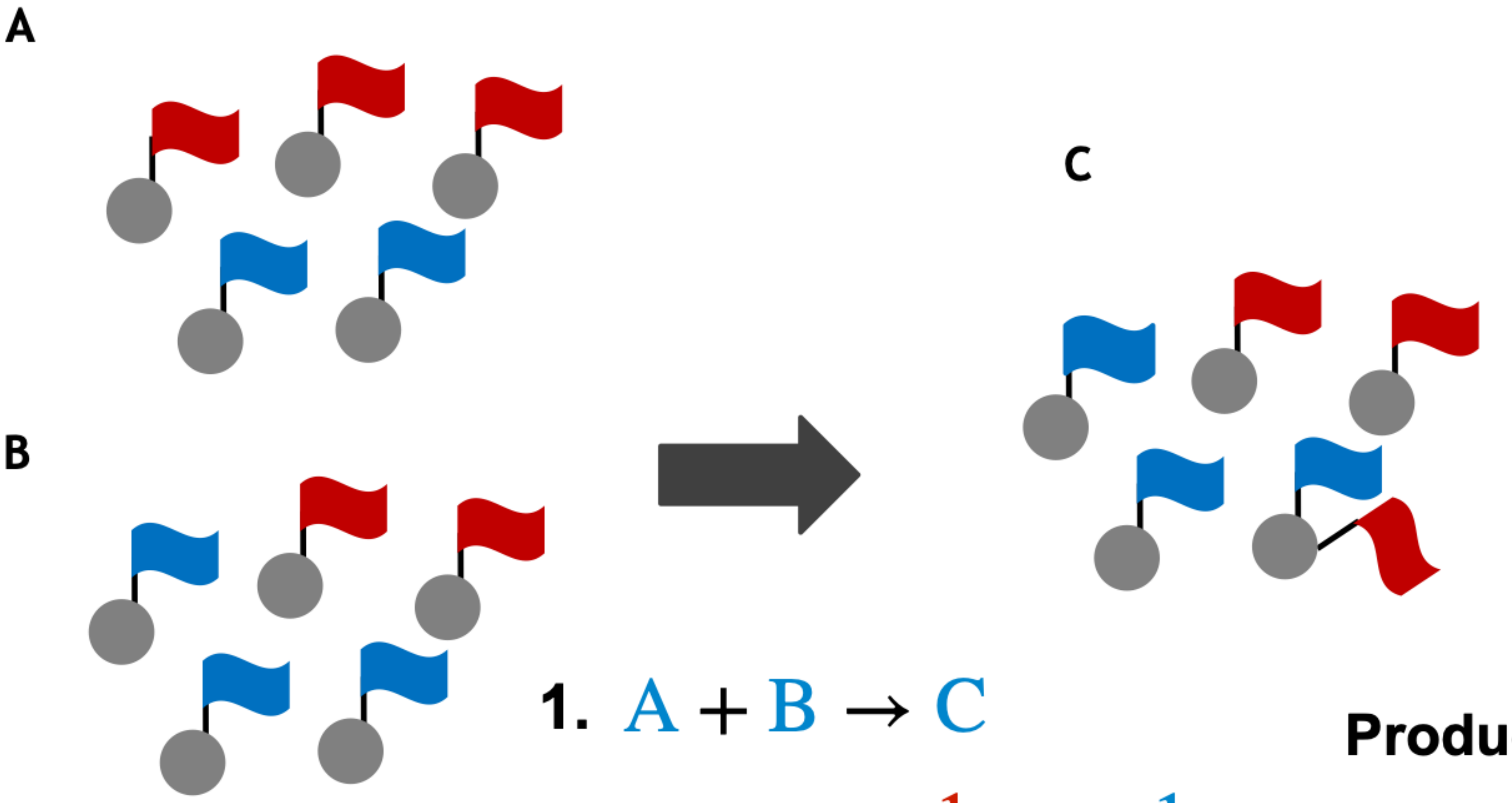
Nonlinearity of tropospheric O_3 formation

- 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)

Tagging Method



emission sectors:
landtransport
industry

1. $A + B \rightarrow C$

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

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

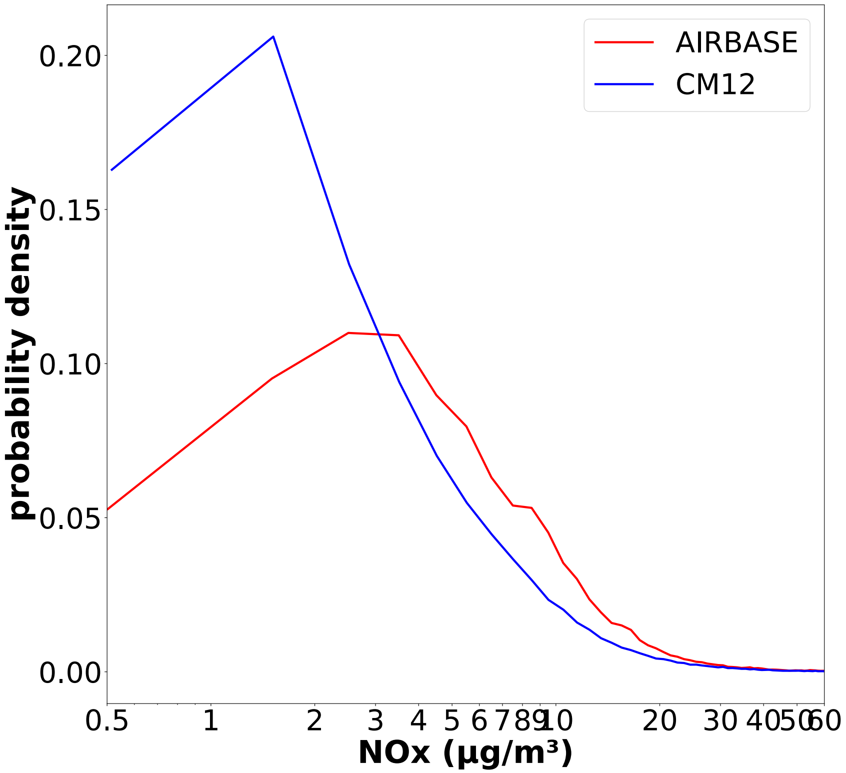
Production

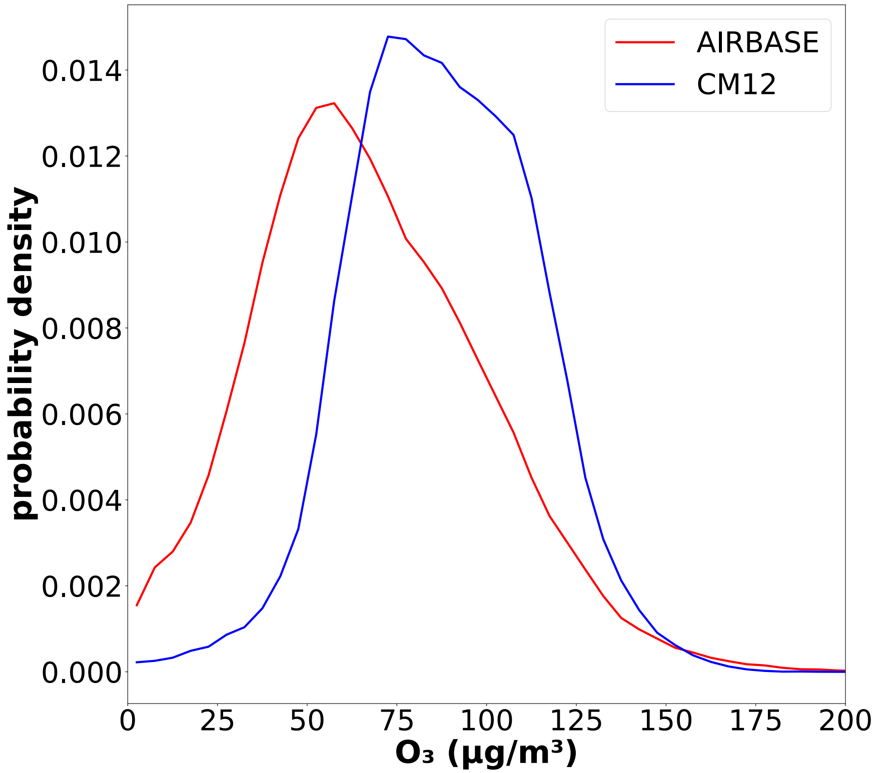
$$P(C^{tra}) = \frac{1}{2} k \; A \; B \left(\frac{A^{tra}}{A} + \frac{B^{tra}}{B} \right)$$

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

Evaluation with AIRBASE stations

- Comparison of the PDFs of ground-level NO_x and O_3 concentrations from CM12 with air quality stations.
- 419 rural stations throughout Europe were selected





- 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)

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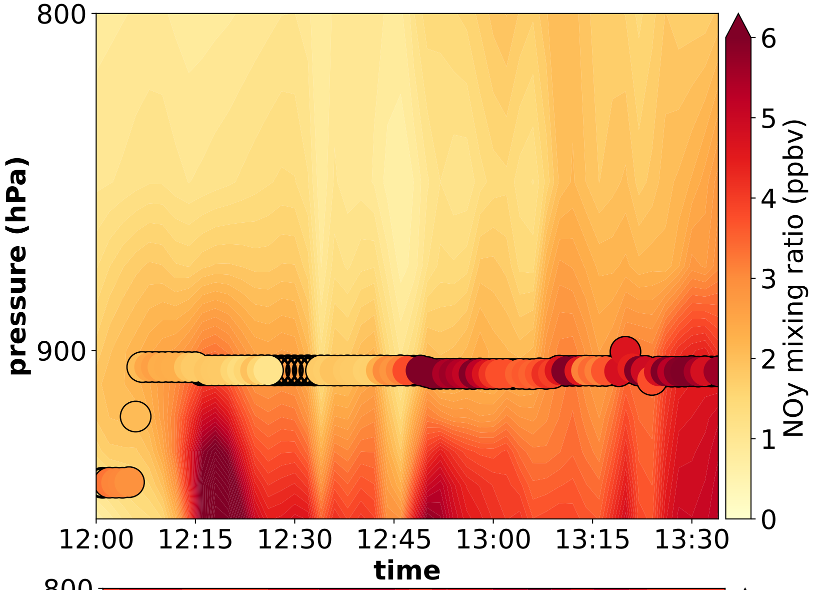
Evaluation with in situ data (EMeRGe)

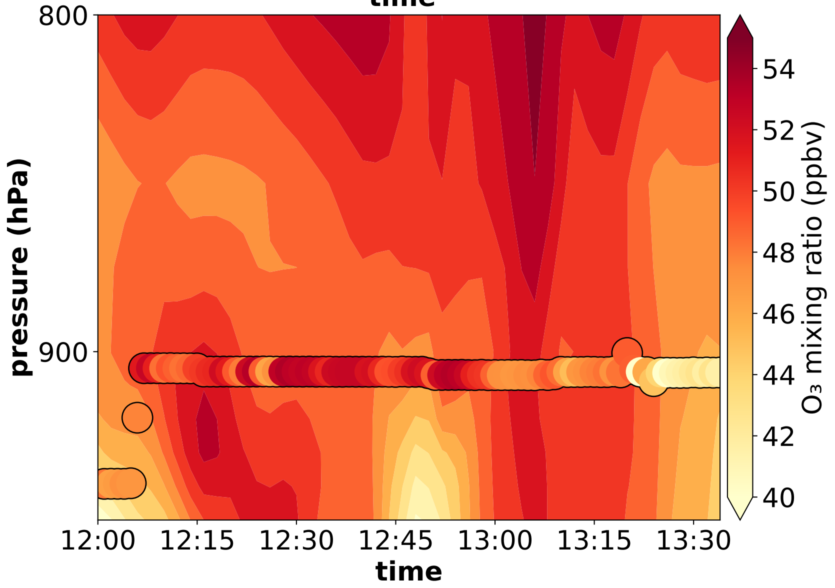
Benelux region: 26.07.2017

- 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



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).