T3.3. Extending AQ modelling to health and policy relevant indicators down to urban scale (M02-M30)

Lead: Spyros Pandis, FORTH; Augustin Colette, INERIS;

Inv.: FMI, BSC, MET Norway, TNO, CNRS (CEREA/LISA)

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Main scope of Task 3.3

➢ Design & Develop health and policy relevant indicators at urban scale

- What?
  - Improving the modelling of EC/BC, UFP, OP, VOC (taking stock of WP1/WP2)
  - Source apportionment (health & policy indicators)

- How ?
  - Collaboration between modelling groups (to desing generic approaches)
  - Collaboration between models/emissions/observations experts

- For Whom?
  - 3.4 Implement in models supporting policy
  - 3.5 Upscale as Service Tools

UFP Modelling

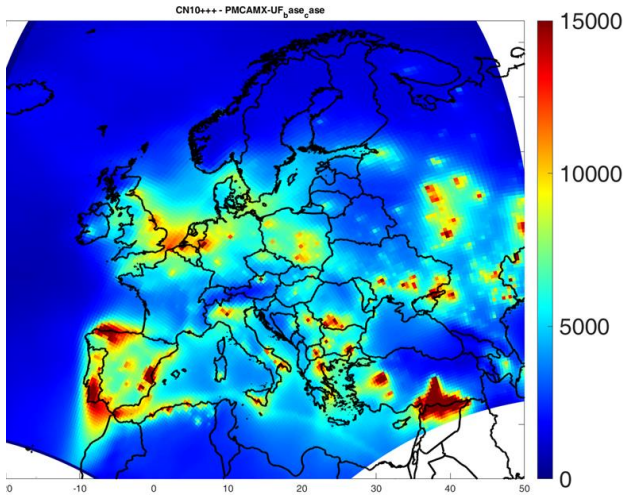
Simulations of Ultrafine Particles-PMCAMx-UF

- Simulates both aerosol number and mass distributions using 42 aerosol size sections (from 1 nm to 10 μm)
  - TOMAS algorithm
- Complete organic aerosol simulation using the Volatility Basis Set including ELVOCs, LVOCs, SVOCs, IVOCs and VOCs.
- Uses TNO inventory (EUCAARI version)
  - Easy to adapt to new version of inventory
  - Has been extended to include traffic emissions down to 1 nm (Olin et al., ACP, 2022).
- Includes several nucleation parameterizations with sulfuric acid, ammonia, amines, oxidized biogenic organics.
- Runs at 36x36 km for Europe and zooms gradually to 1x1 km over selected cities using the emissions at the corresponding scale.
- Number source apportionment using the technique of Posner and Pandis (Atmos. Environ., 2015).

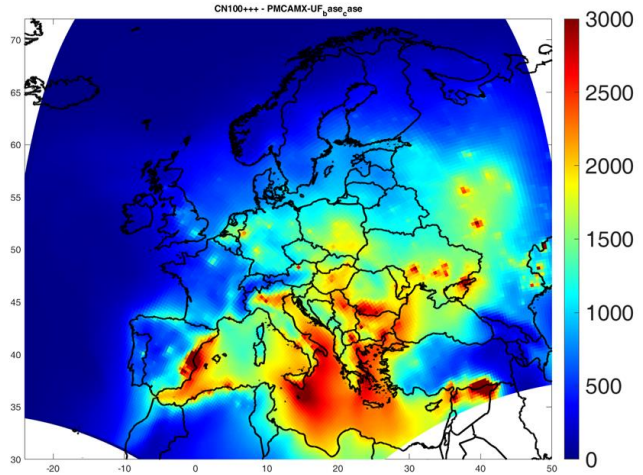
PMCAMx-UF Predictions
PEGASOS Campaign Summer 2012

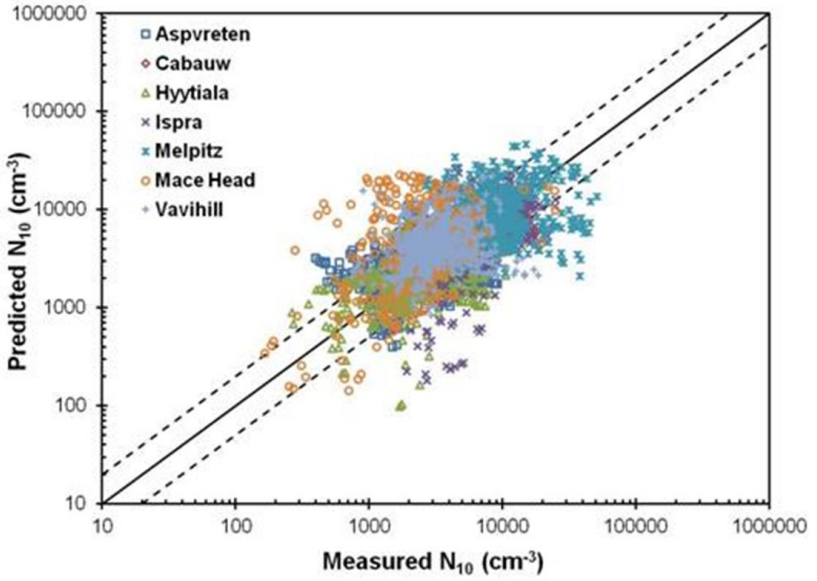
Spyros Pandis, FORTH

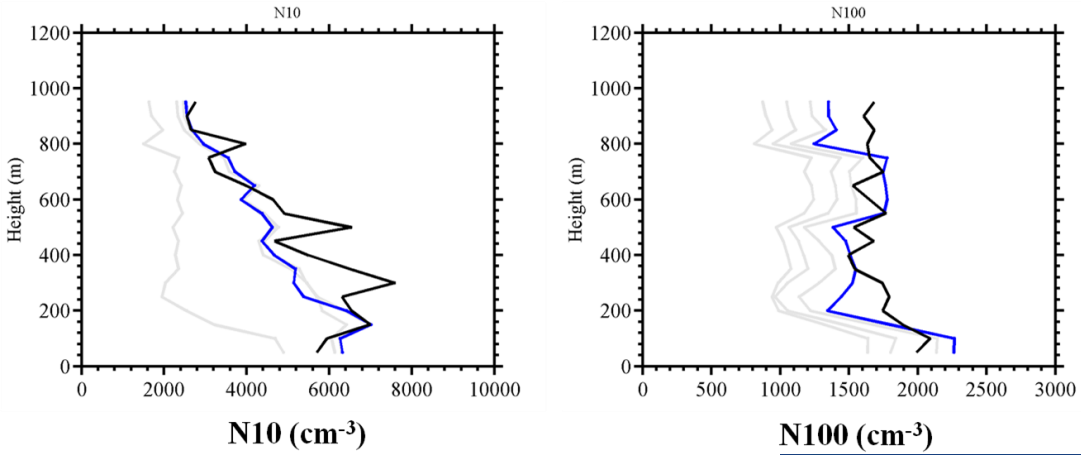
N_{10}



N_{100}







Measured
Predicted
(15 flights over Po Valley)

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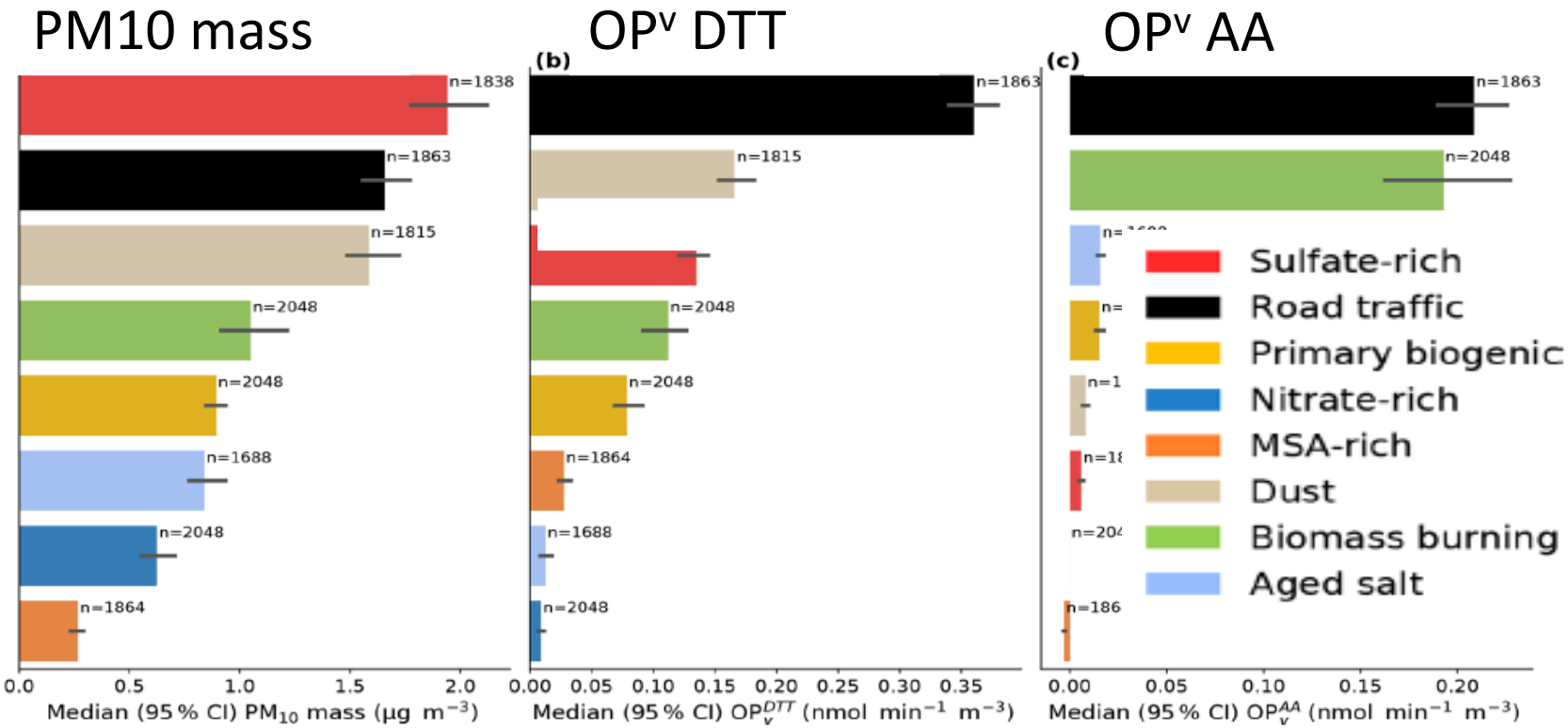
PM Oxidative Potential Modelling

Oxidative Potential modelling

Gilles Foret, LISA
Renske Timmermans, TNO

- **OP** is a measure of **oxidative stress**, a process linked to many **health impacts**
- *In situ* **measurements** are possible → attribution of **OP (intrinsic) by sources** with source attr. (**PMF**)



Weber et al, 2021

- Model the OP for mapping, forecasting and define future policies
- Implementation in operational tools - regional CTM:
  - CHIMERE, Lotos-Euros (uEMEP planned)

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OP modelling: Methods

Gilles Foret, LISA
Renske Timmermans, TNO

- Modelling PMF sources with modelled sources from tagging PSAT approach

Simulated OP → $OP_t^v = \sum_{t,s} (OP_s^i * C_{t,s}^{mod})$ ← [PM] of PMF sources simulated with PSAT

t= time

s= sources

Intrisic OP of PMF sources

Example of combination of PSAT sources to simulate PMF

PMF (Weber et al, 2021)	AA (nmol.min^{-1}.µg^{-1})	DTT (nmol.min^{-1}.µg^{-1})	PSAT
Biomass burning	0.17	0.13	Residential combustion (S2) + waste management (S9) - primary species
Primary traffic	0.14	0.21	Energy production (S1) + industrial comb. (S3) + road transport (S7) + other transports (S8) - primary species
Dust	0.01	0.11	Prod. Processes (S4) + agriculture (S10) + dust (model) - primary species
Primary Biogenic	0.02	0.13	-- not in CHIMERE yet ---
Aged sea salt	0.01	0.04	Sea salt (model) + other transports (S8) - primary + secondary
Nitrate rich	0.01	0.04	Road transport (S7) + agriculture (S10) + 40% NO_3^- (model) - secondary
Sulfate rich	0.01	0.09	Energy prod. (S1)+ Residential comb. (S2) + Indus. Comb. (S3) + Prod proc. (S4) waste management (S9) traitement des déchets - secondary
MSA rich	0.02	0.12	--- not used ---

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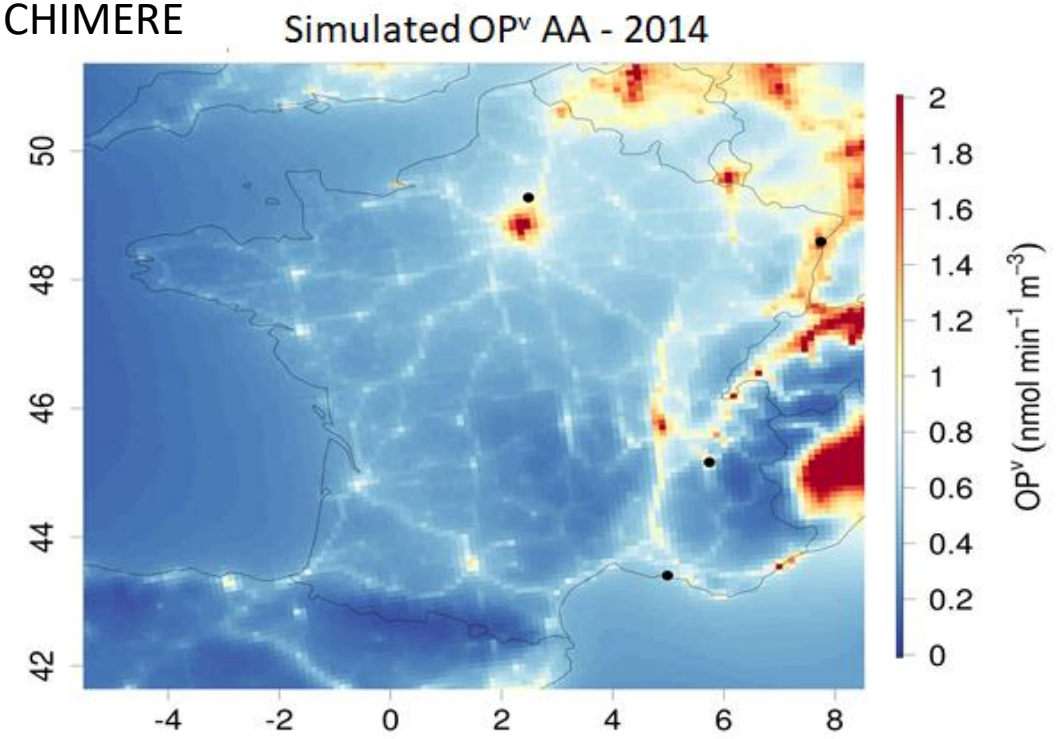
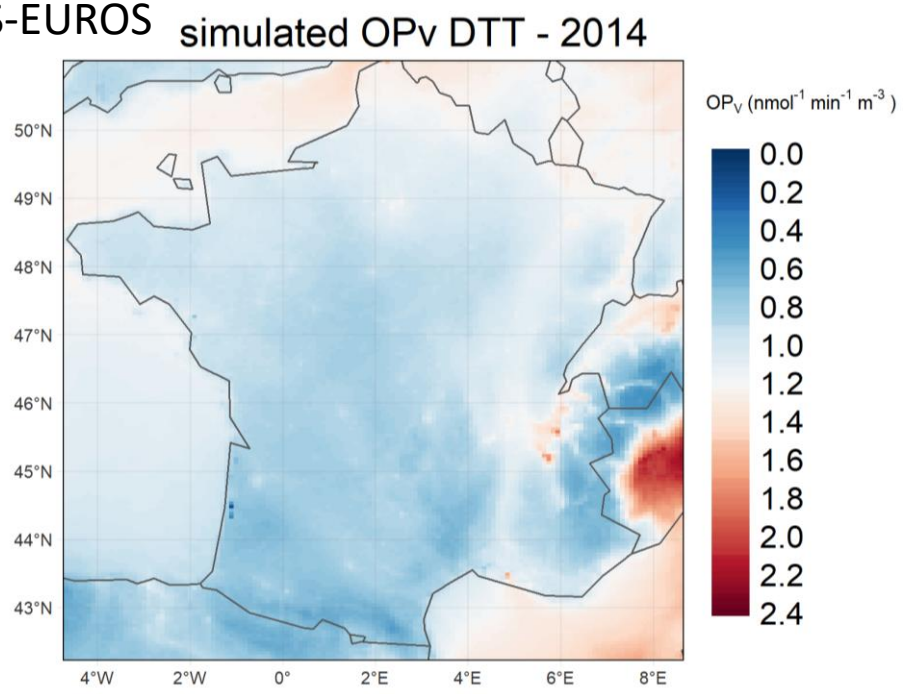
OP Modelling : Results

Gilles Foret, LISA
Renske Timmermans, TNO

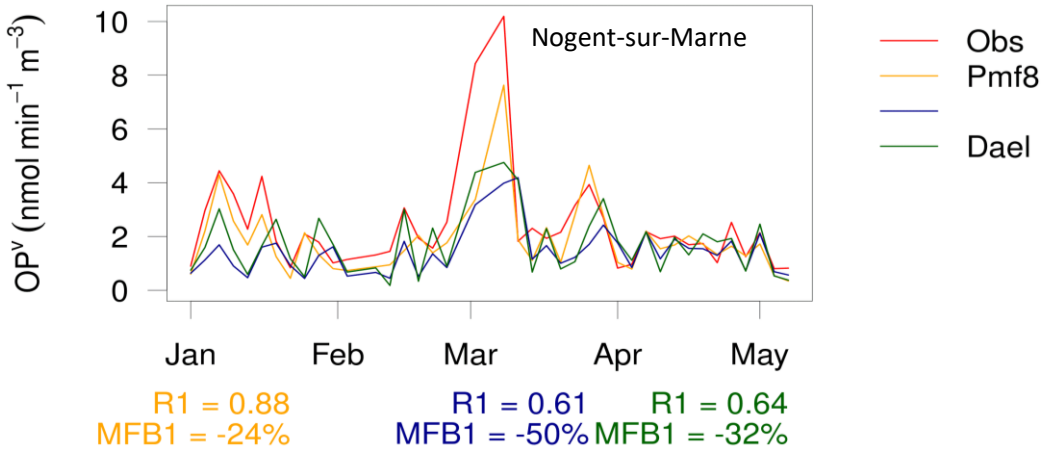
CHIMERE



LOTOS-EUROS



Model-Obs comparisons



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OP Modelling: Perspectives

Gilles Foret, LISA
Renske Timmermans, TNO

- Intercomparisons of source apportionment and PO modelling methods between LISA and TNO for the year 2018
- In collaboration with IGE investigate of better mapping potential between observed OP and sources in models (e.g. derive an OP for agriculture based on observations)
- Further develop an alternative method consisting in fitting of OP from observations to modelled source profiles to derive intrinsic OP from model
- Cross-comparisons exercices with other European modelling tools (LISA, TNO, FORTH, MetNO/EMEP …) → to be discussed

EC/eBC/BC

EC/BC/eBC

- Comparison between modelled and measured EC/eBC/BC

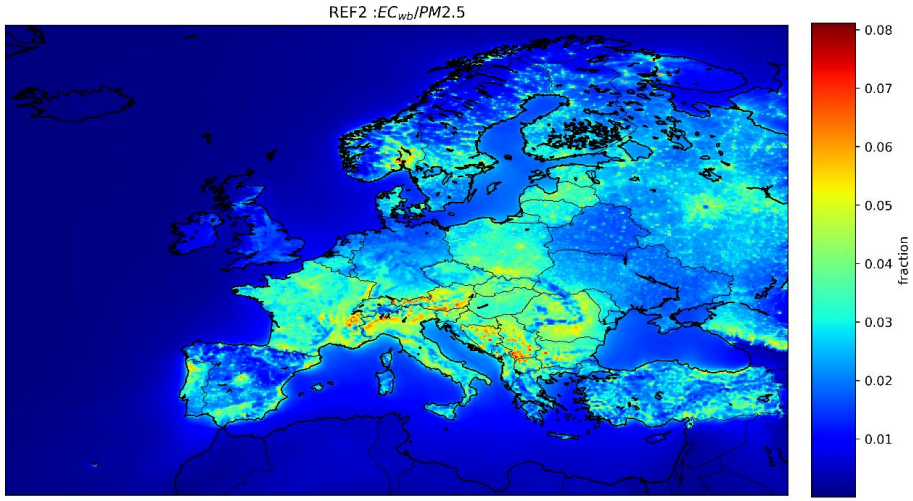
- Status
  - WP1 & ACTRIS working on measurement coordination
  - CAMS already producing EC in European AQ Forecasts (inc. Source app)

- Current activities
  - Clarify the model/observation procedures (EC/BC/eBC)

- Way forward:
  - Paving the way to NRT source apportionment validation (Service Tool)
  - First priority EC ff/wb
  - Before engaging in more challenging secondary PM Source App (ACSM)

  - Guide model development/improvement:
  - sensitivity to emission factors of BC (inc non-exhaust)
  - Trends (policy effectiveness)

Multi-Model ECwb
(CAMS/Eurodelta)

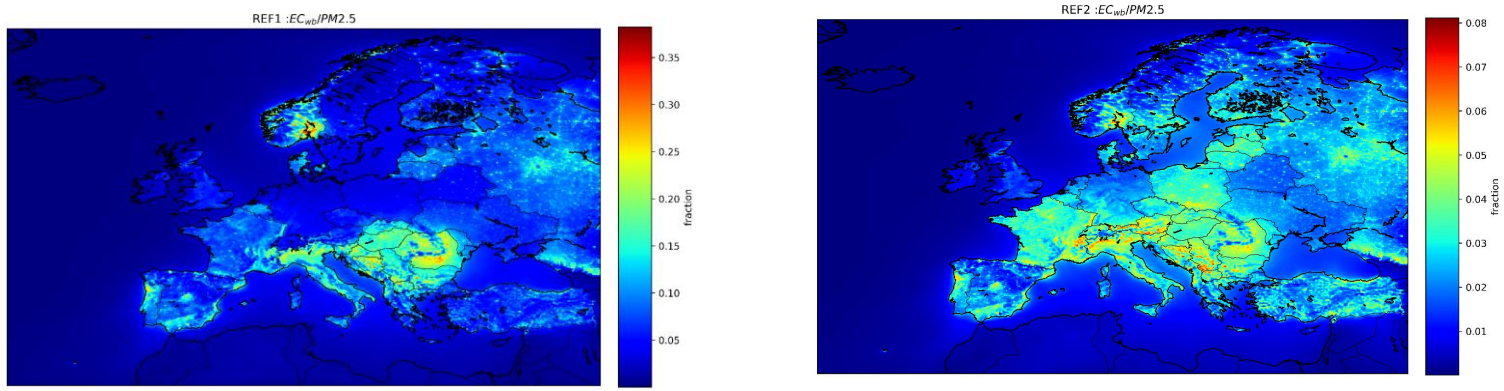


Augustin Colette, INERIS
Renske Timmermans, TNO

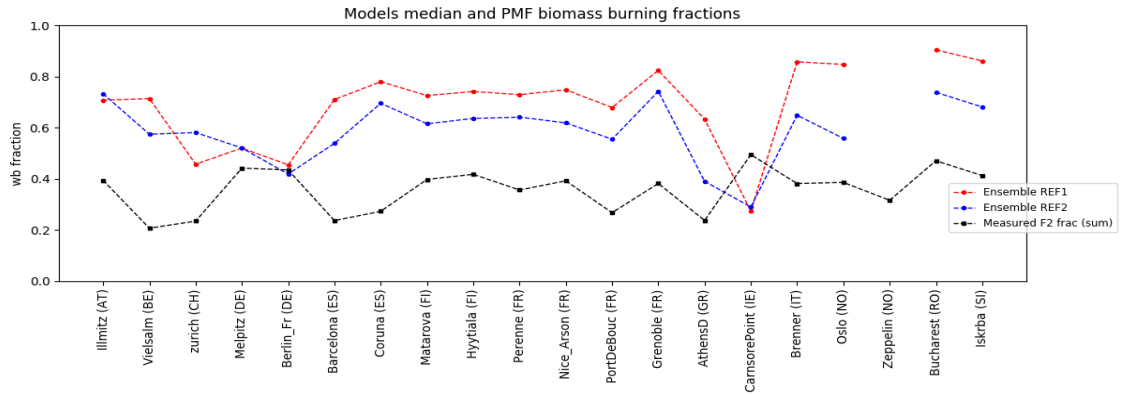
EC/BC/eBC: motivation (Source App)

- Eurodelta-Carb
  - 10 CTM intercomparison
  - COLOSSAL/ACTRIS/EMEP Winter 2017-2018 campaign
- Including condensables
  - Improve PM10 bias 20%
  - Bring model tagging of wb closer to observations
- A typical example of how multi-model & observation source apportionment can feed back on inventory improvement

Fraction of wb in EC (REF / REF2=inc condensables), courtesy J. Tokaya



Share of wb in EC from PMF, courtesy S. Platt, NILU



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Antoine Guion, INERIS

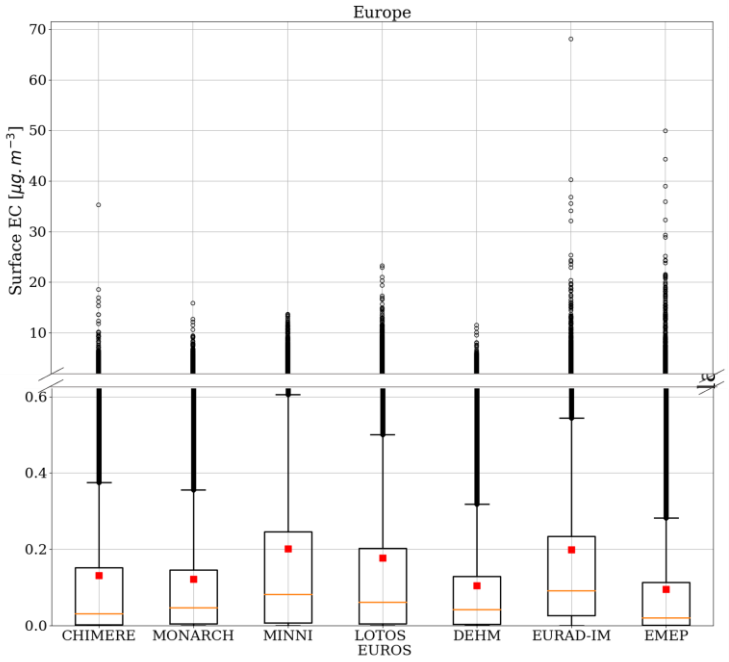
EC/BC/eBC Models in CAMS & Observations

- Regional models have some spread in modelling EC
  - CAMS_61 consistent multi-model re-run for 2018 as data set
  - diversity in CTM formulation
  - diversity in PM split in the inventory

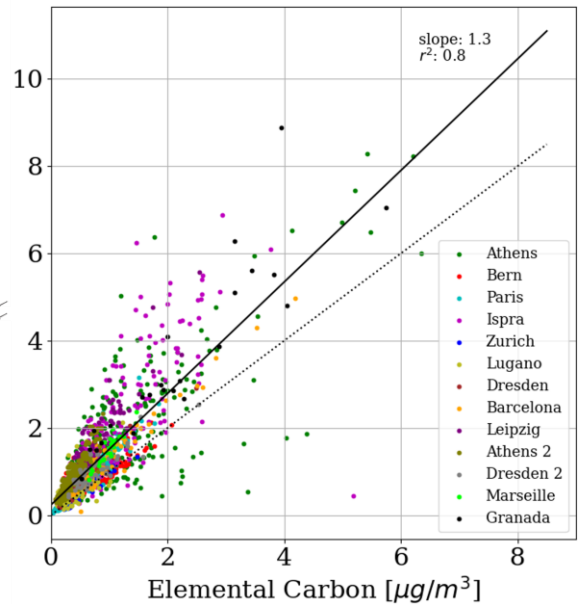
- Observations too exhibit a spread between EC and eBC
  - measured mass for EC (thermo-optical method) .vs. approximated mass for eBC (light attenuation method)
  - different instruments and Mass Absorption Cross section coefficients for eBC (e.g. AE22, AE33, MAAP)
  - different wavelengths and absorption bands among aethalometers for eBC

- How to factor the 30% difference between observed EC & eBC when comparing to models?

Simulated EC concentrations from CAMS models



EC measurements and harmonized eBC at 13 stations



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Antoine Guion, INERIS

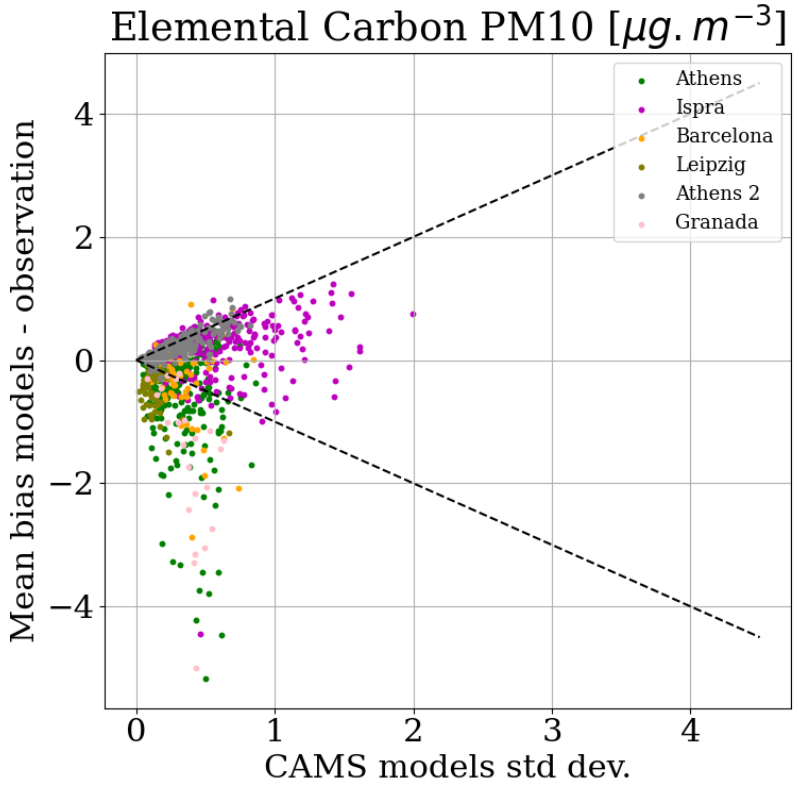
EC/BC/eBC Models in CAMS & Observations

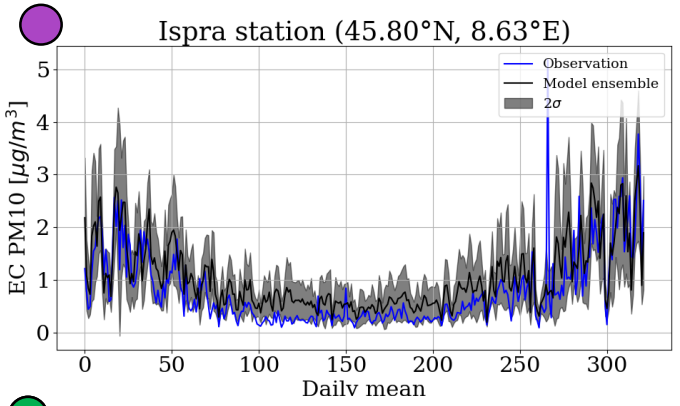
Findings :

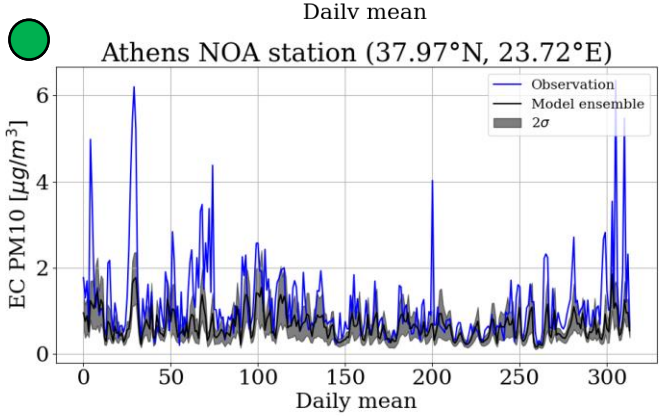
- 48.6% of EC measurements (6 stations)
  - mean bias > model std dev.
  - i.e. observations fall outsite model spread
- Could be due to the EC inventory (with a mix of EC/BC Emission Factors), or model representativity,
- But appears to be site specific => points toward measurement homogeneisation

Beneficial developments for modellers :

- corrected and harmonized eBC data that would be “model-ready”
- measured data with corresponding uncertainty
- One possibility would be to compare models to both site-specific & median MAC (same as for WP2/Health)







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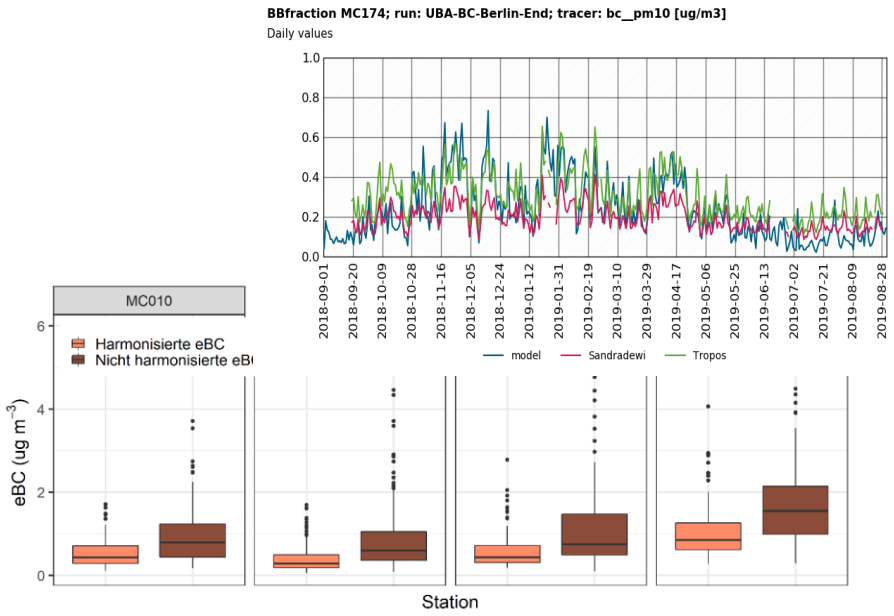
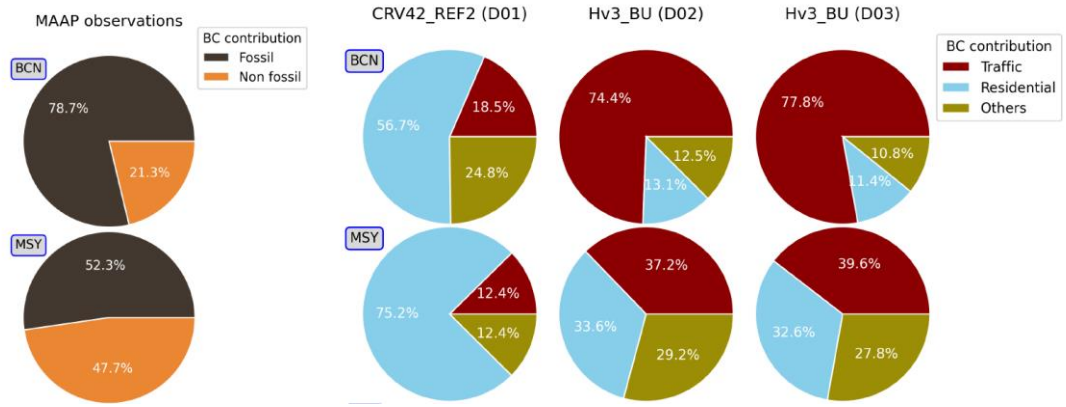
Renske Timmermans, TNO
Oriol Jorba, BSC

EC/BC/eBC towards source apportionment

TNO/TROPOS:
BC measurement harmonisation in Germany
compared to Lotos-Euros

BSC/CSIC:
BC ff/wb compared to
top-down or bottom-up emissions in MONARCH





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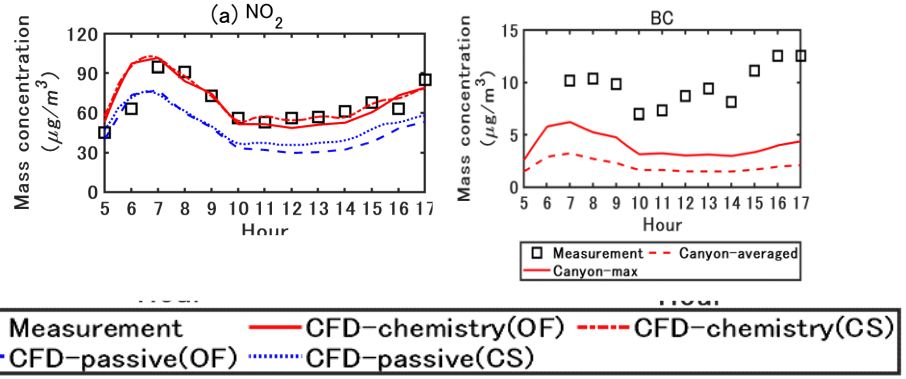
Karine Sartelet, CEREA

EC/BC/eBC/OC model development (1)

- Both street-in-grid (MUNICH) and CFD exhibit low BC bias which is not seen for NO2/PM10
- Investigations using a variety of emission factor methodologies point toward a potential flaw in non-exhaust BC emissions
- Uncertainties in exhaust emissions: not the main cause of black carbon (Sim. 6 x Sim. 4)
- => Further comparisons to be performed in Paris for more recent years and using more measurement stations (link to WP4.3)



	o [μg.m^{-3}]	s [μg.m^{-3}]
1	6.07	1.37
2	6.07	1.74
3	6.07	1.62
4	6.07	4.91
5	6.07	4.92
6	6.07	5.12

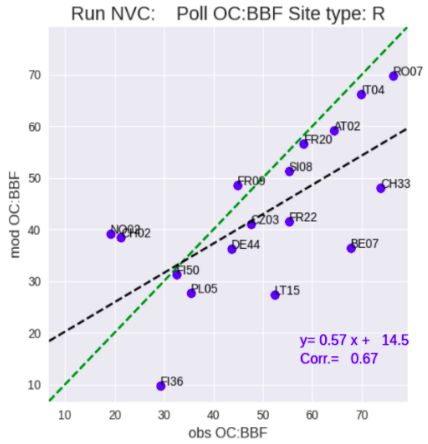
Lugon et al. GMD (2021)

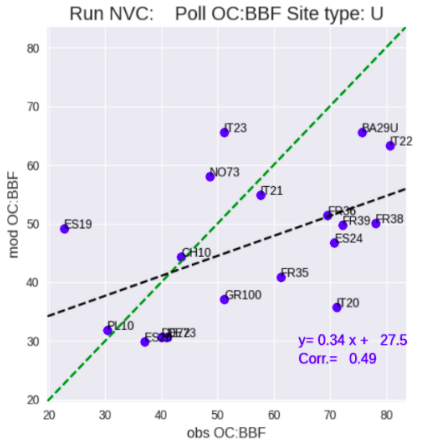
Sim. 1 – Only exhaust emissions
Sim. 2 – European guidelines (EMEP)
Sim. 3 – NORTRIP model (2013)
Sim 4 – Boulter (2005), Luhana et al. (2004)
Sim. 5 – Sim. 4 without particle deposition (representing maximal resuspension)
Sim. 6 – Sim. 4 considering the uncertainties of black carbon exhaust emissions (EMEP)

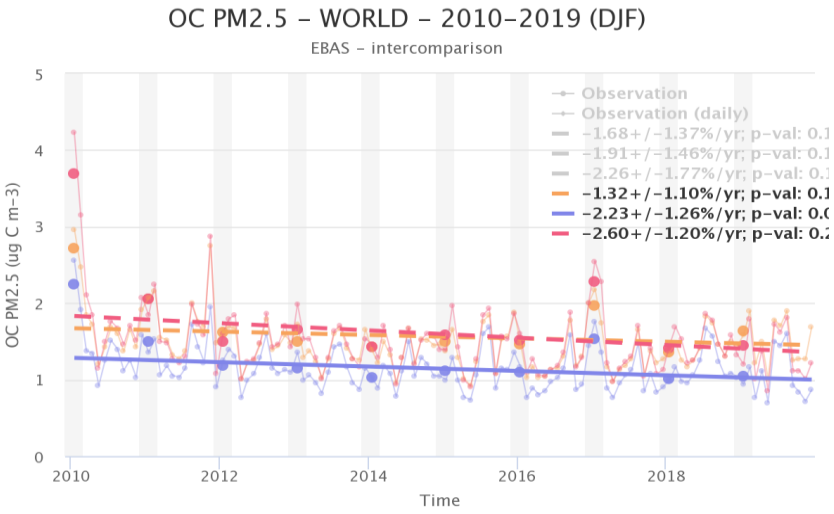
Hilde Fagerli, MetNo

EC/BC/eBC/OC model development (2)

- Recent update of Condensable (REF2) inventory in NMR Project
- Model compared to OC, and wb fraction of OC (based on levo)
- OC trends (DJF) 2000-2019 using TNO ‘typical’ emission data (NVC)
- The SVOC&IVOC model setup tend to show the largest negative trends, with better comparison to observed trends.
- The ability of models to capture trends has very important implications for policy effectiveness



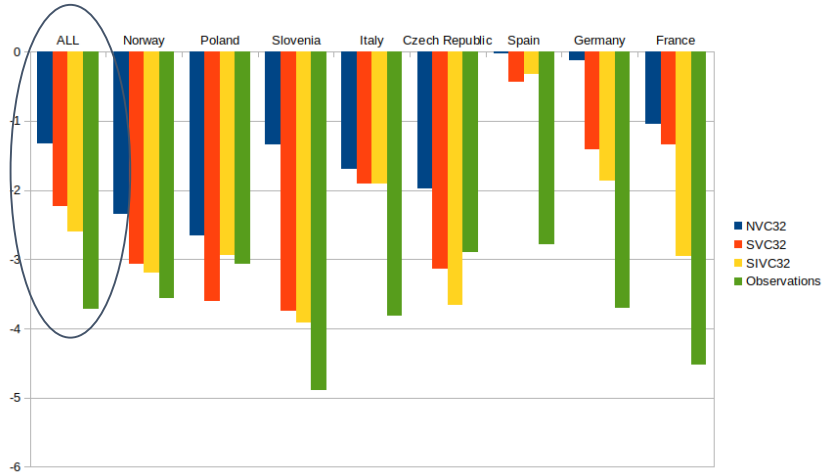




NVC32: inert PPM

SVC32: semivolatile OA allowed to evaporate (uses 1-5D VBS)

SIVC32: as SVC32 , but with added intermediate volatility compounds (IVOC is assumed to be 4.75 * mass of POA, following Ciarelli)



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VOC Modelling

VOC Model/Observations

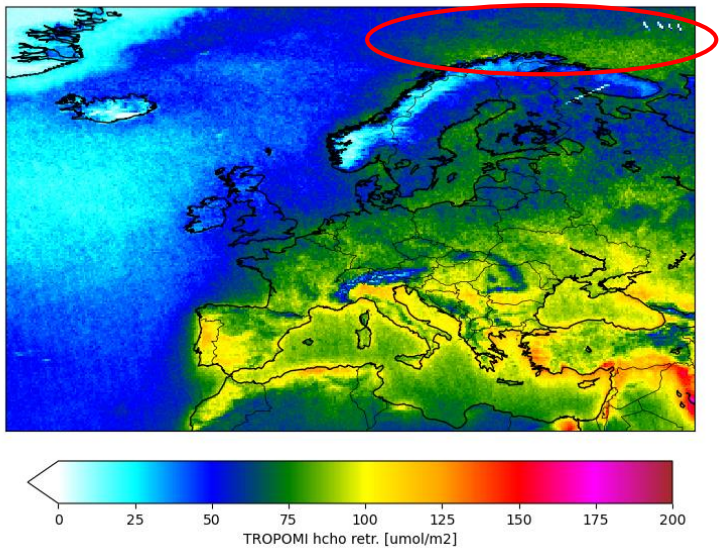
- Problem:
  - VOC speciation used in models is based upon very old data. It is unclear how realistic it is.
  - Evaluation of NMVOC species tricky since models use surrogate compounds
    - e.g. EMEP model's n-butane represents all heavier alkanes – cannot be directly compared to measurements.
- Solution:
  - "tracers" can be added to the chemical schemes to allow tracking of n-butane, i-butane, n-pentane, etc., and comparison with observations.
  - Updated emission speciation become available (CAMS, UK NAI)
  - Some CTMs have a variety of build-in chemical schemes and speciations
- Aims:
  - To evaluate emissions (as far as possible: there are many uncertainties).
  - To re-assess importance of different VOC species for both SOA and VOC
  - Evaluate POCP/MIR assumptions in realistic conditions
- Opportunities
  - WP1 link with VOC monitoring, including in urban areas
  - Emerging satellite data for evaluation in addition to the surface data

HCHO: model/in-situ/satellite

- Substantial noise in retrievals, including negatives; requires averaging over area and/or time
- Pixels gridded on 0.2x0.2 deg grid (~20x10 km), weighted by overlap
- Temporal means over 2020
- Can be compared to EMEP in-situ

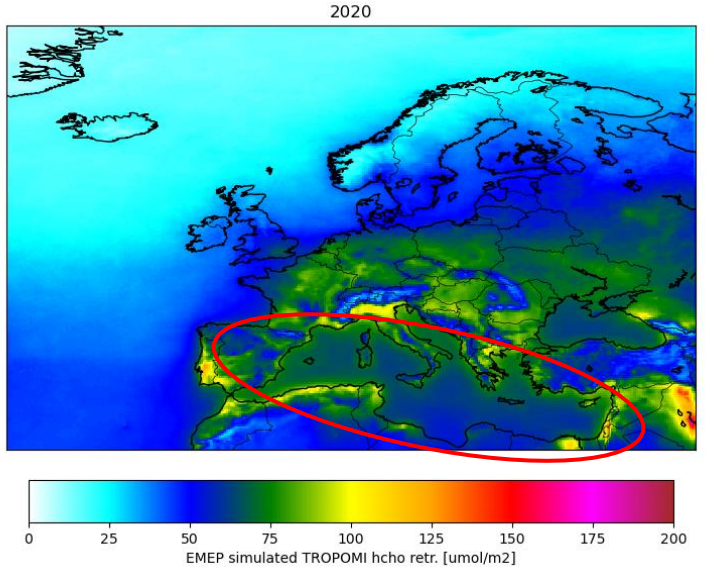
S5p/HCHO over 2020

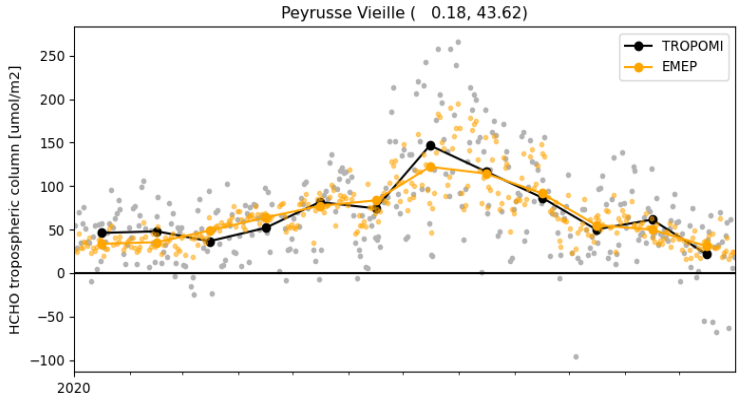
- accurate over Nordic sea?

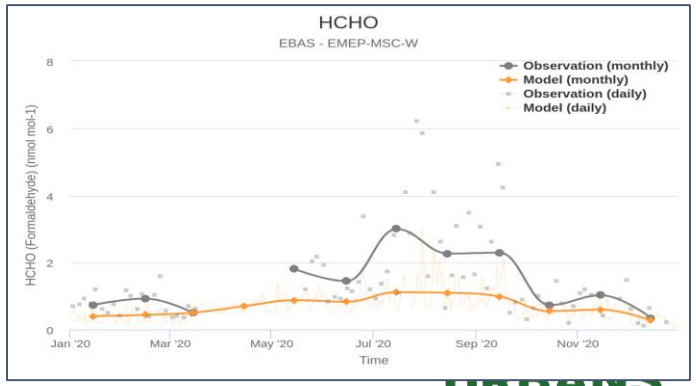


EMEP simulated S5p/HCHO over 2020

- Lower values, especially over water







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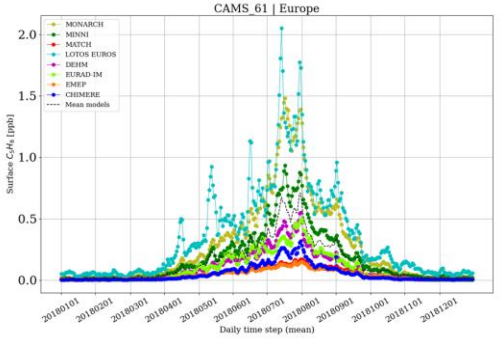
VOC Model/Observations

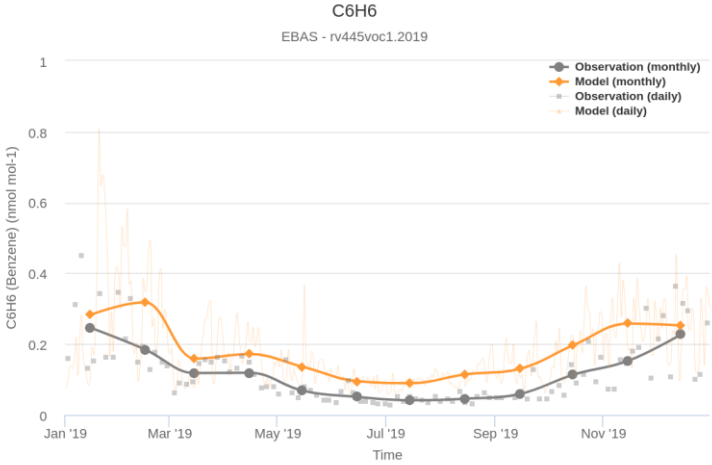
- Single model comparison
  - Implement new species/tracers
  - EMEP Model to background sites observations (eg Kosetice)
- Multi-model comparison
  - Variety of scheme / upscaling
  - Comparing CAMS ensemble for BVOC (C5H8)
- => Needs to be further discussed in light of RI-Urbans WP1 VOC measurements



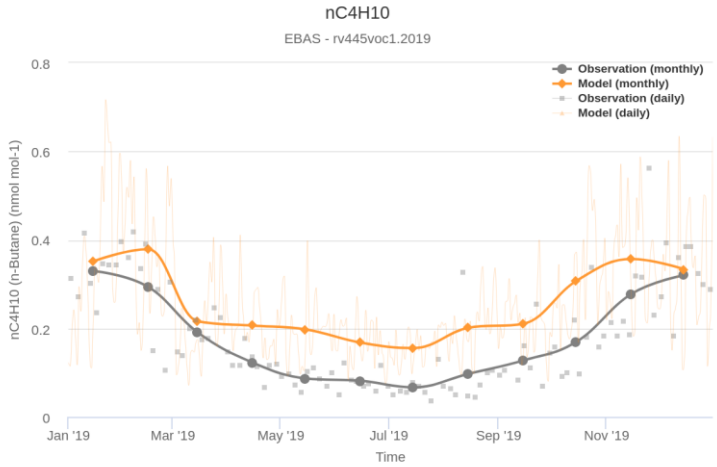
Validation scores over summer 2018 (mean measured concentrations of 0.42 ppb)

Models	*Concentration [ppb]*	*Bias (mod.-obs.)*	*RMSE*	*Pearson correlation (R)*
MONARCH	1.23	0.81	1.27	0.65
MINNI	0.81	0.38	1.08	0.60
MATCH	0.36	-0.06	0.50	0.57
LOTOS-EUROS	0.55	0.32	0.81	0.53
DEHM	0.43	0.01	0.82	0.56
EURAD-IM	0.39	-0.03	0.64	0.57
EMEP	0.12	-0.33	0.45	0.65
CHIMERE	0.49	0.07	0.86	0.66

benzene (ppb), Kosetice, 2019



n-butane (ppb), Kosetice, 2019



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Concluding Remarks and Way forward

- Ongoing activities
  - Model development
  - Improve links between model & observations & emissions
  - Topics: UFP, EC/BC, VOC, OP

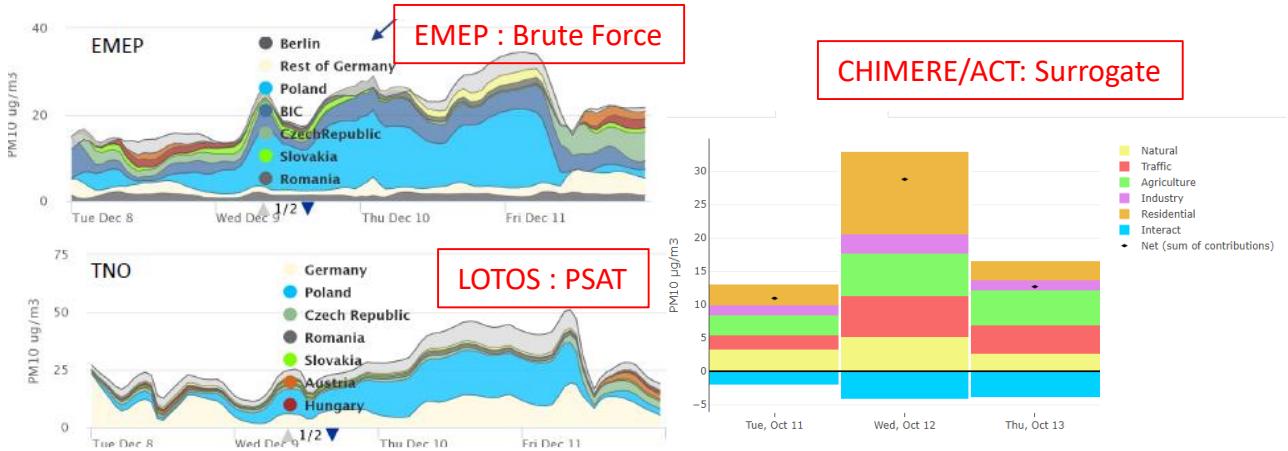
- Forthcoming, still with strong model/obs synergy
  - Policy-relevant indicators: Source apportionment
  - Health exposure: PM metrics of urban background concern

- Links to SP2&3 (demonstration & service tools)
  - Deploy over pilots
  - Nesting and spatial downscaling at urban scale (uEMEP)
  - Implement in policy support tools
  - Design Service Tools with operational links
    - Methodology UFP/OP
    - NRT Model QA/QC (Source app, EC/BC/eBC, VOC)

https://fairmode.jrc.ec.europa.eu/document/fairmode/WG3/European%20guide%20SA_3.1_online.pdf

Source allocation in CAMS Policy Service

https://policy.atmosphere.copernicus.eu/



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