Modelling SOA, EC and OC in Europe
Comparison with measurements

David Simpson

EMEP MSC-W

Norwegian Meteorological Institute (Location: Chalmers, Sweden)

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- Includes primary OC (POC) and EC
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- SOA is a 'research' module

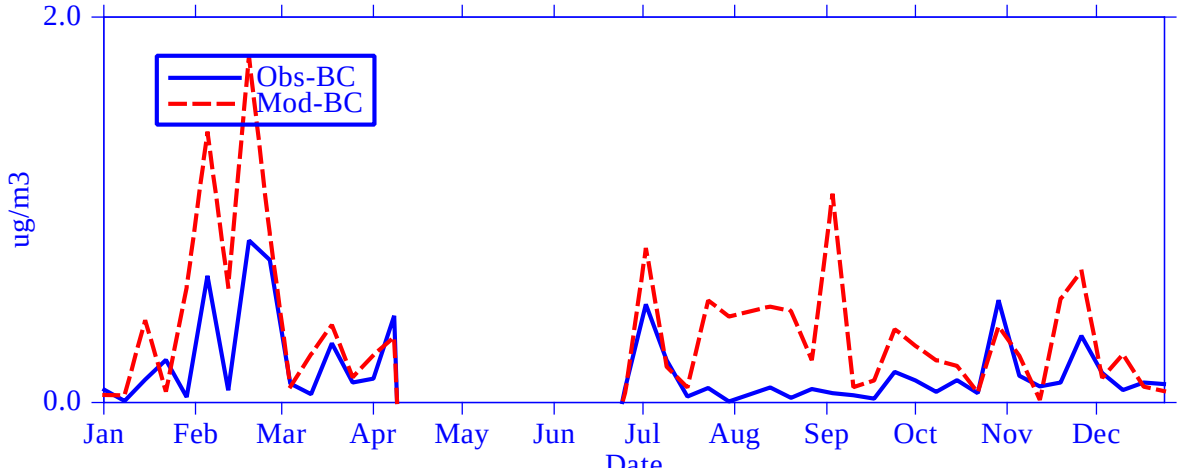
Measurements?

Sources of Data:

- NILU EC/OC Campaign, 2002-2003 - 24h sample of EC/OC once per week
- CARBOSOL (EU Project) - weekly EC/OC + chemical composition (e.g. OC1, OC2, WSOC, HULIS, levoglucosan, etc.)
- Austrian AUPHEP sites - daily EC/OC

BC? (NILU EC/OC data)

NO:



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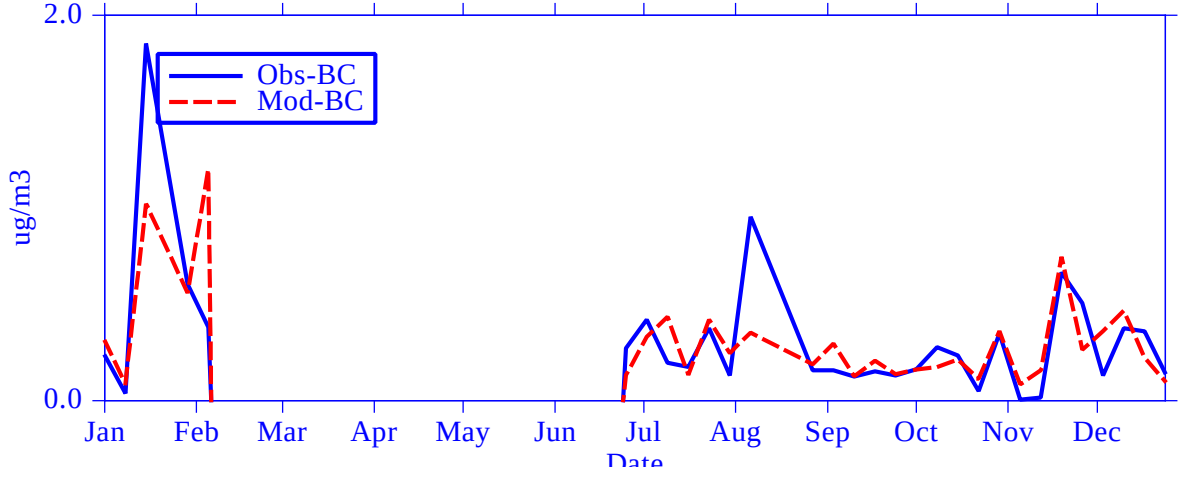

BC cont.

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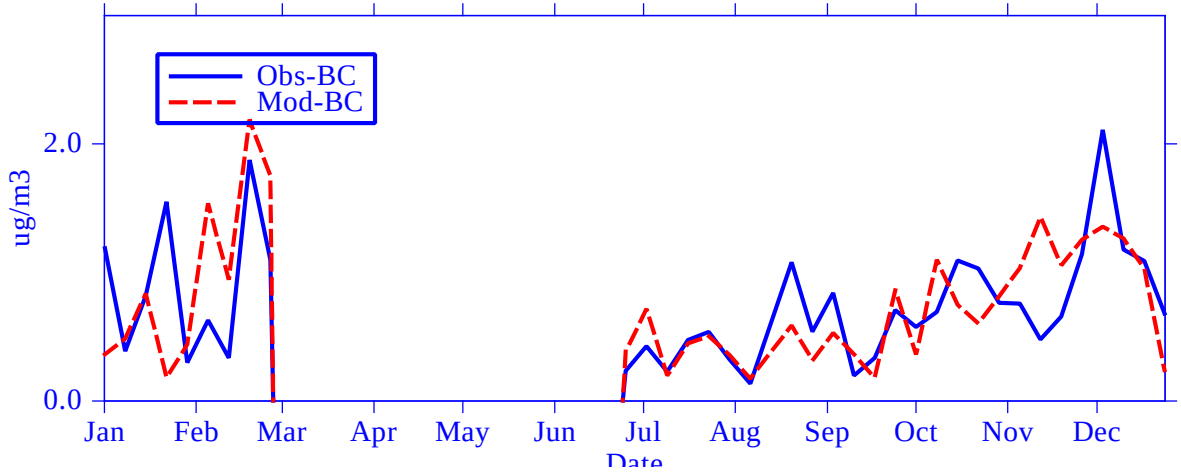
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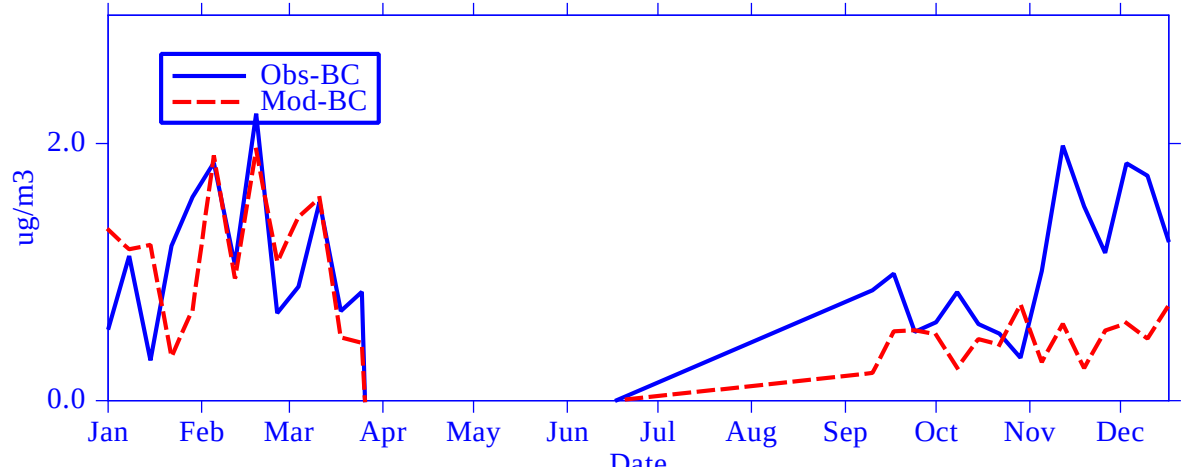
HU: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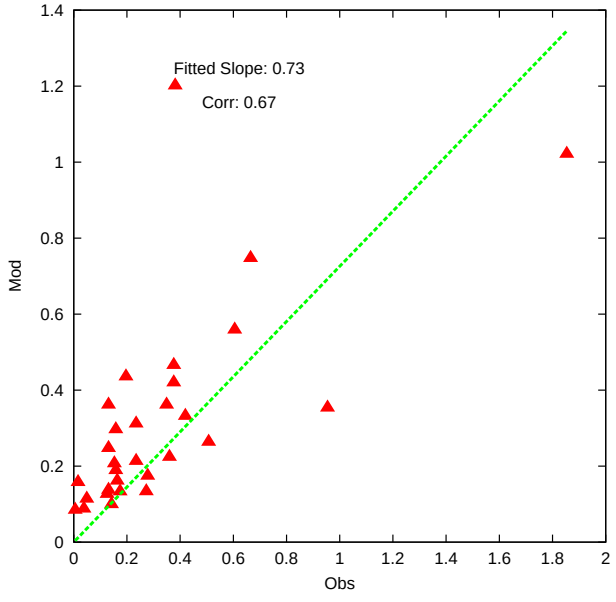
BC cont.

Sweden



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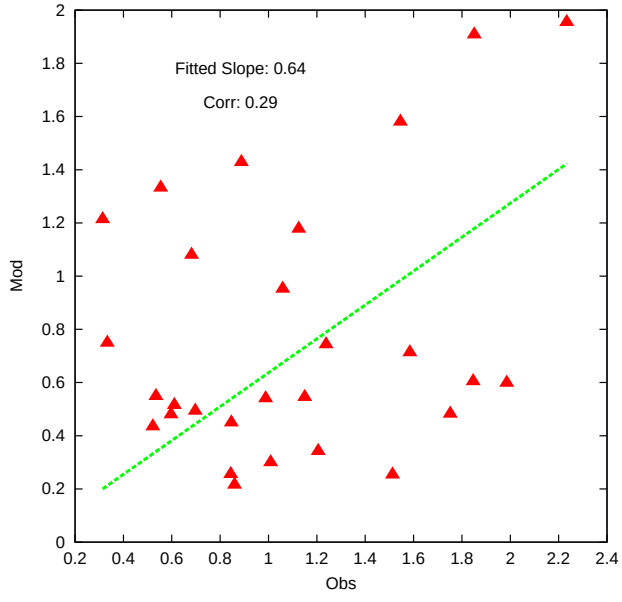
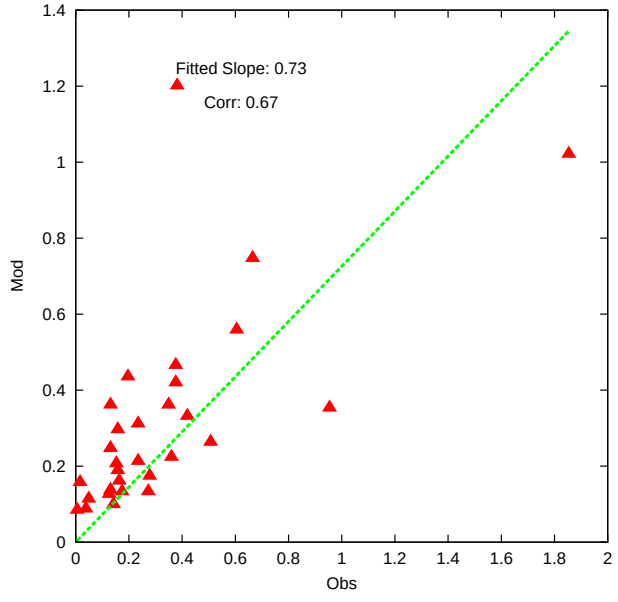


BC cont.

Sweden



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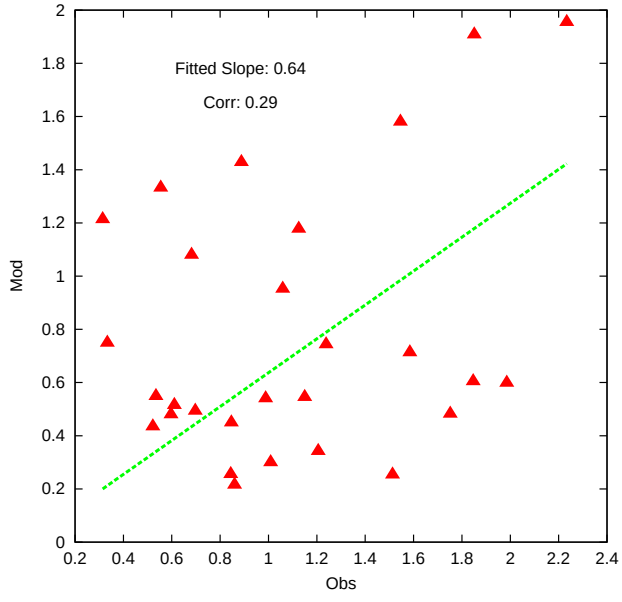
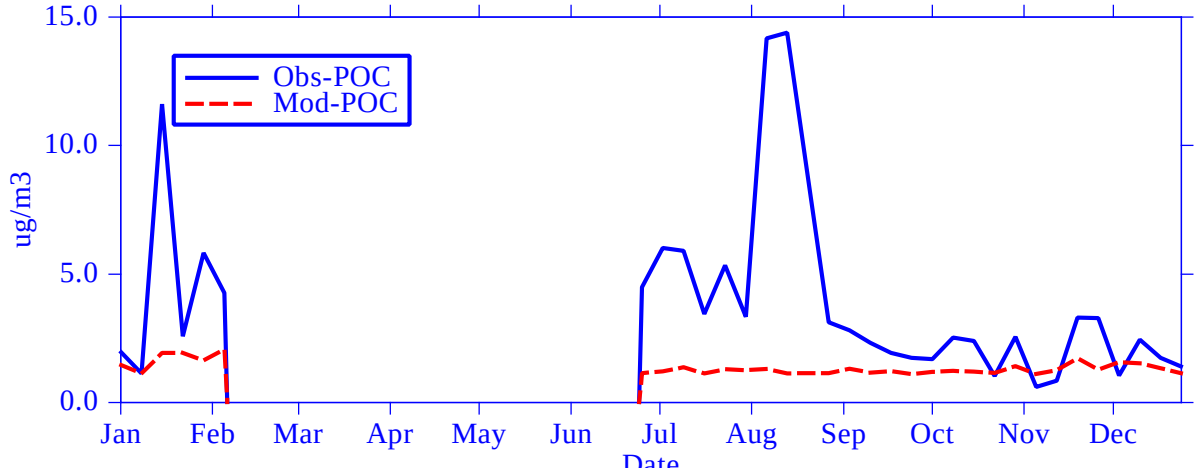


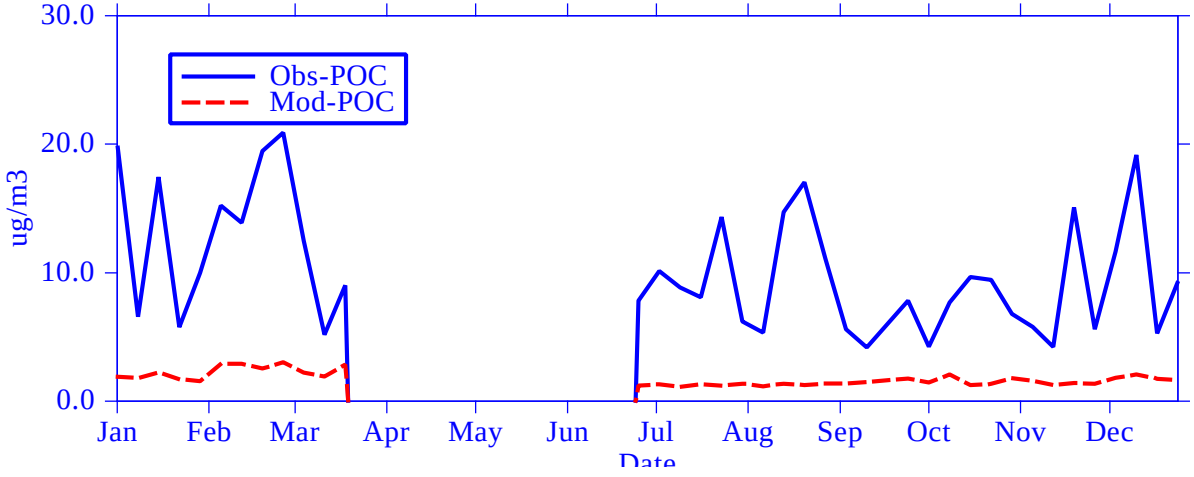
- Conclusion? Not too bad, considering. . .

POC+BGND vs OC?

Sweden:



Austria:



What is the missing OC?

- Could this be SOA?

What is the missing OC?

- Could this be SOA?
- Or, missing primary emissions?

What is the missing OC?

- Could this be SOA?
- Or, missing primary emissions?
- Use model to compare

EMEP SOA model

- Emissions characterised as:
  - ‘Oil’: Fossil-fuel combustion emissions
  - ‘Wood’: Residential combustion

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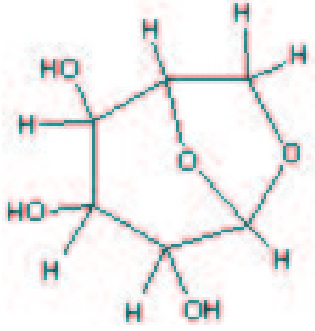
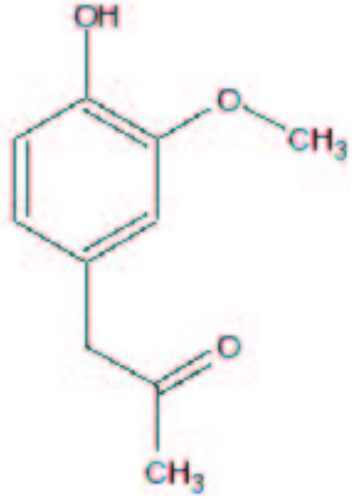
  - ‘Oil’: Fossil-fuel combustion emissions
  - ‘Wood’: Residential combustion
- Explicit surrogate, e.g. from wood-combustion

Levoglucosan Guaiacyl acetone





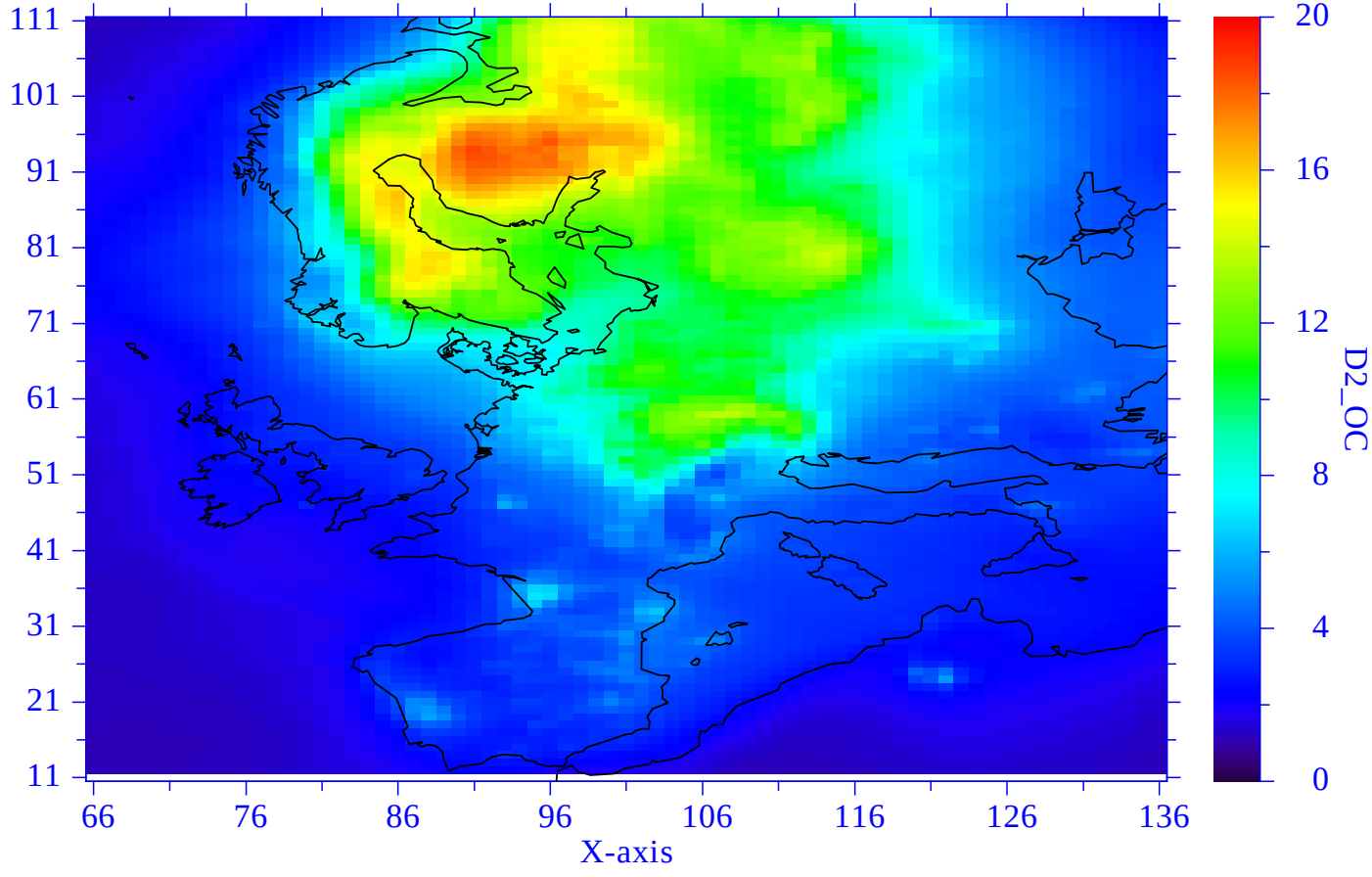
(+benzoic acid, palimitic acid)

SOA model cont.

- Components:
  - POC: Primary emissions (wood + oil)
  - ASOA: Anthropogenic SOA (from aromatics)
  - BSOA: Biogenic SOA (from terpenes)
  - BGND: Background OC (mix of oil/wood/BSOA)
- Gas/Particle partitioning (Pankow/Kamens-type approach. Lee-Kessler for vapour pressure (Makar))
- Detailed α-pinene scheme (Kamens et. al, 1999, Andersson-Sköld and Simpson, 2001).
- 2-product scheme for aromatics (3-methyl-2,5-furandione, tolualdehyde, c.f. Ansari+Pandis, 2000).

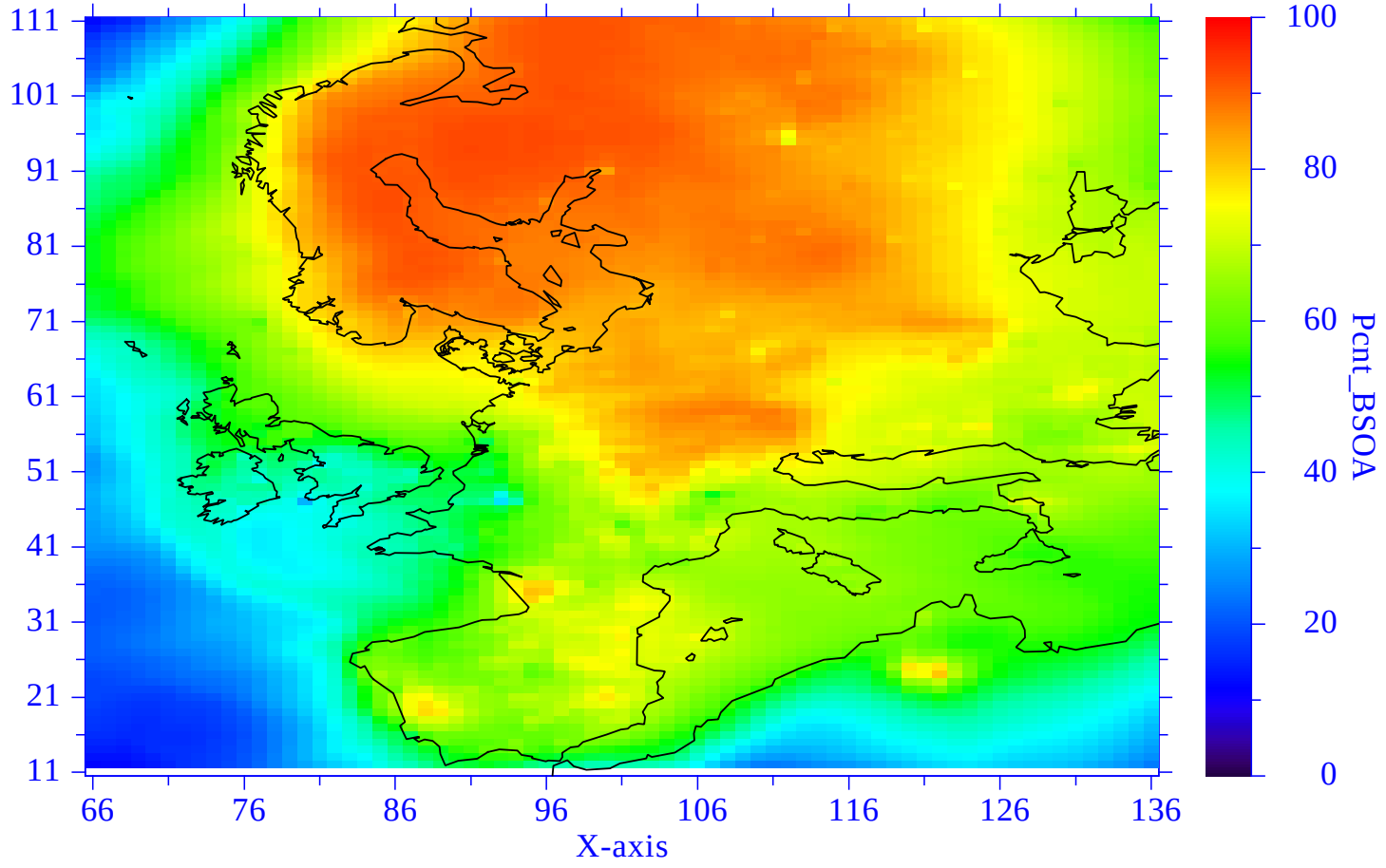
EMEP SOA Model:

Results: Annual Average OC, year 2002 (ug/m3)



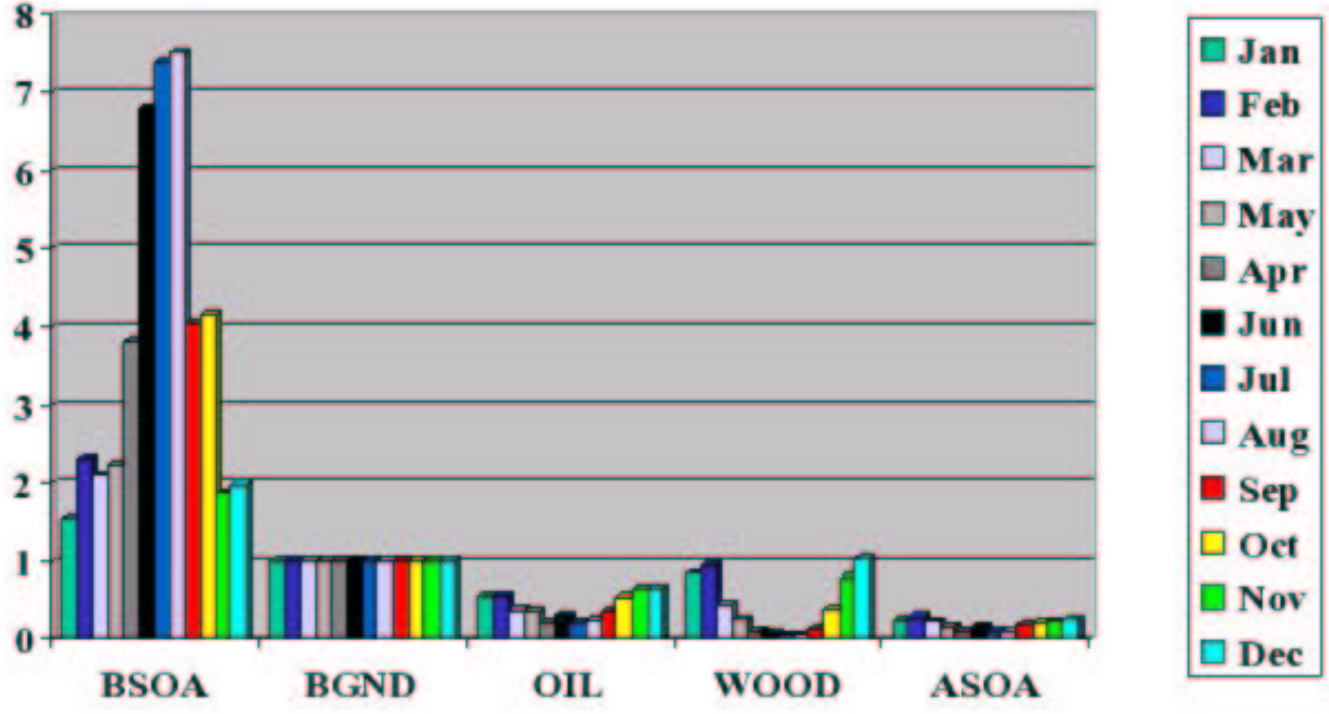
BSOA contribution

BSOA/OC (%)



Seasonal contributions:

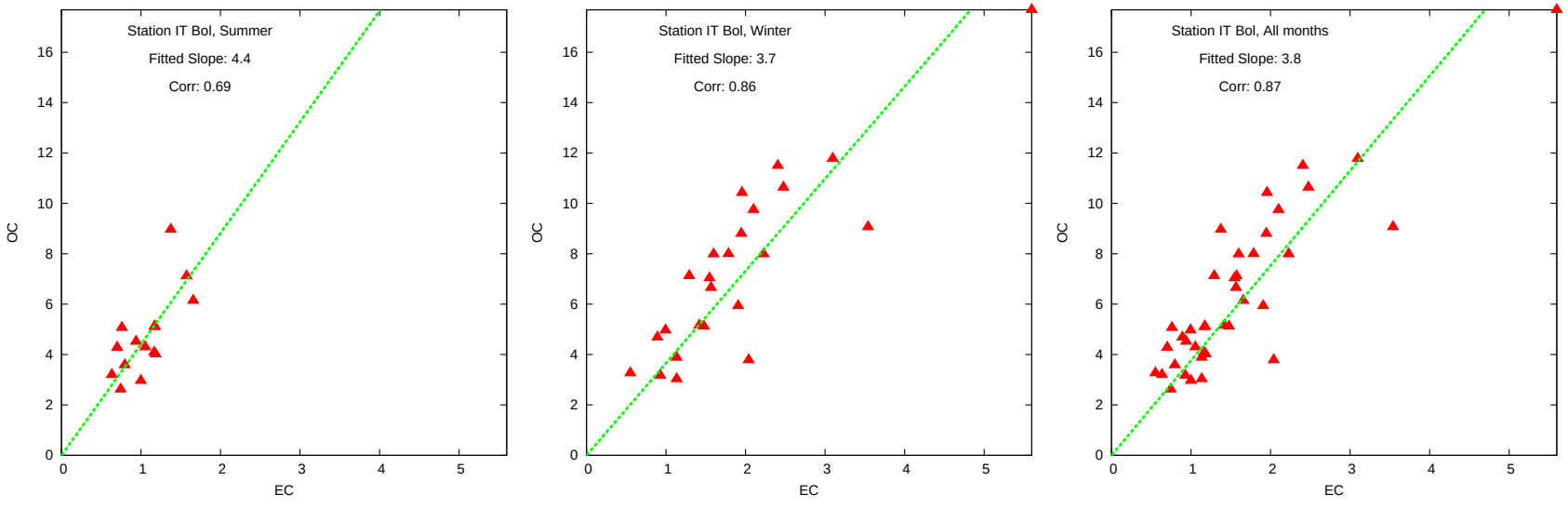
K-Puzsta, Hungary:



Note: The BGND contribution is held fixed in the model.

Summer max in Obs.?

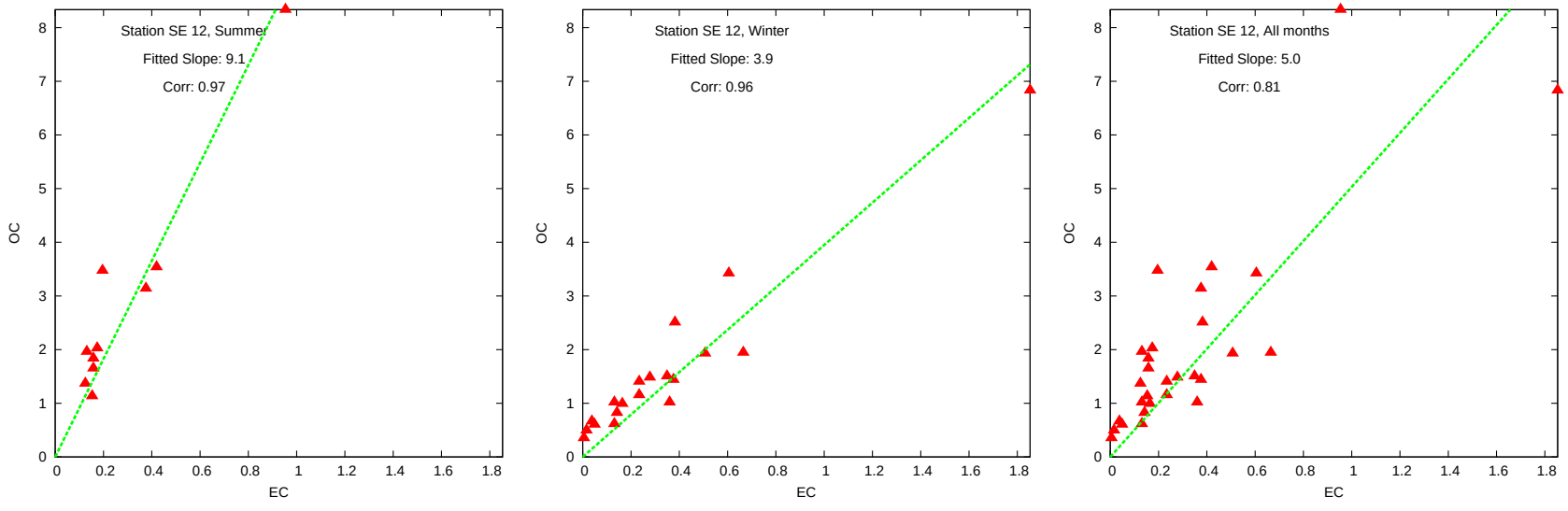
Belogna, Italy:



Not here! Slope very similar in all seasons.

summer max, cont.

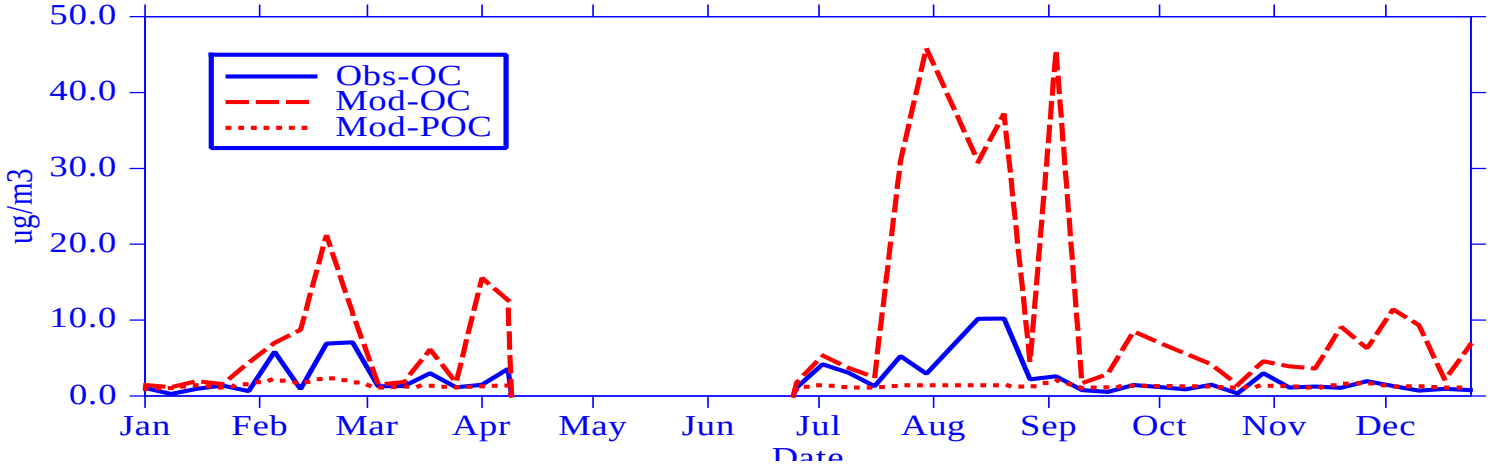
Aspvreten, Sweden:



But here! Slope very different! Why?

OC? (NILU EC/OC data)

NO:



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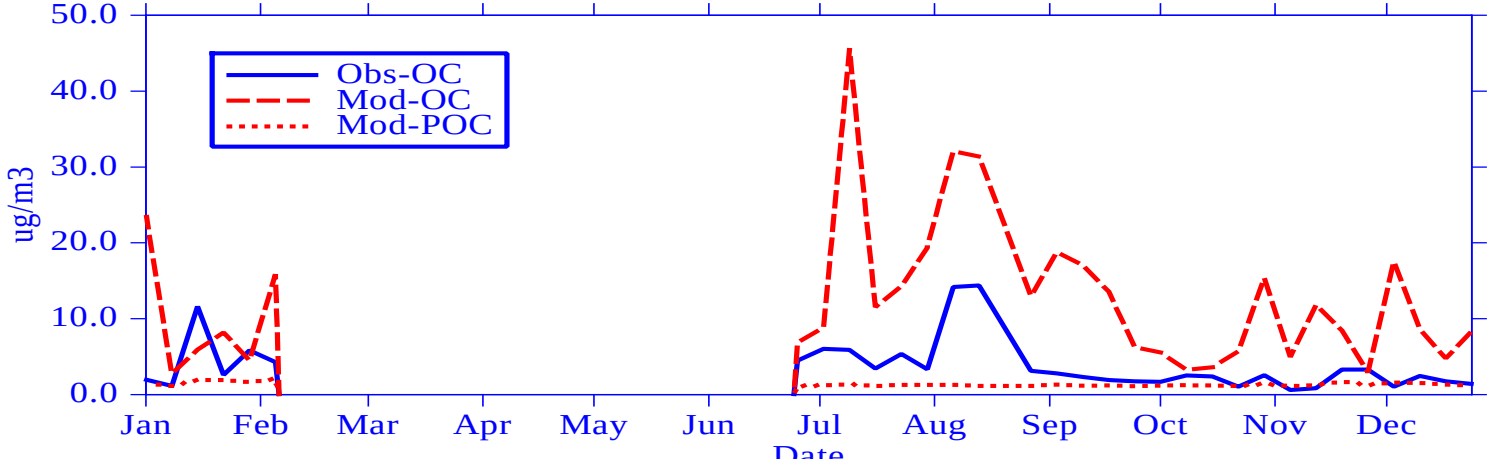
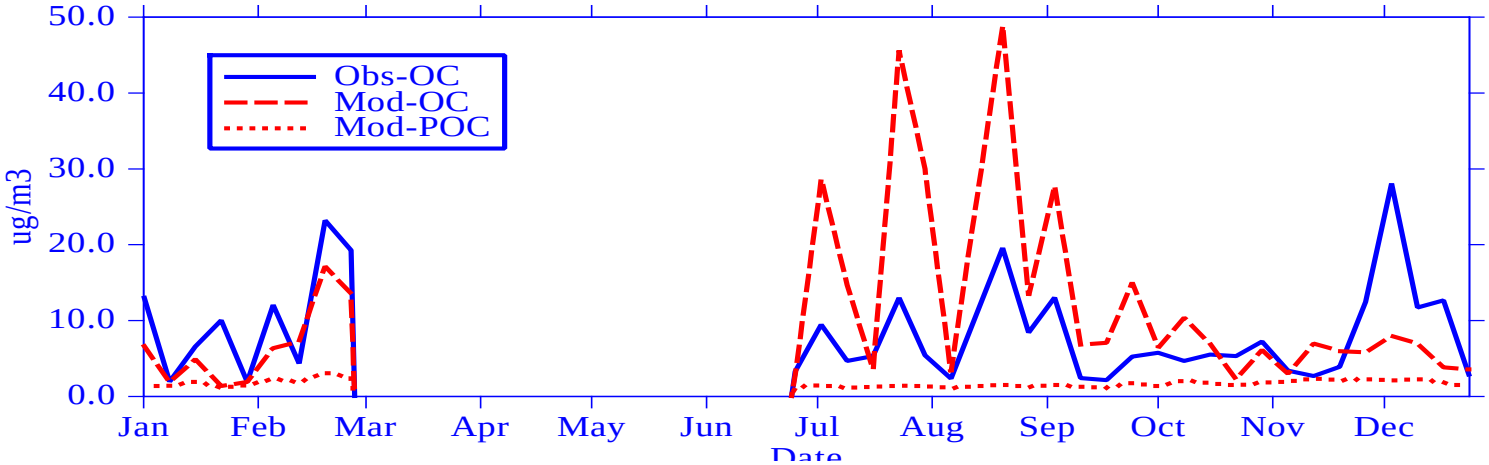


OC cont.

DE:


Obs-OC
Mod-OC
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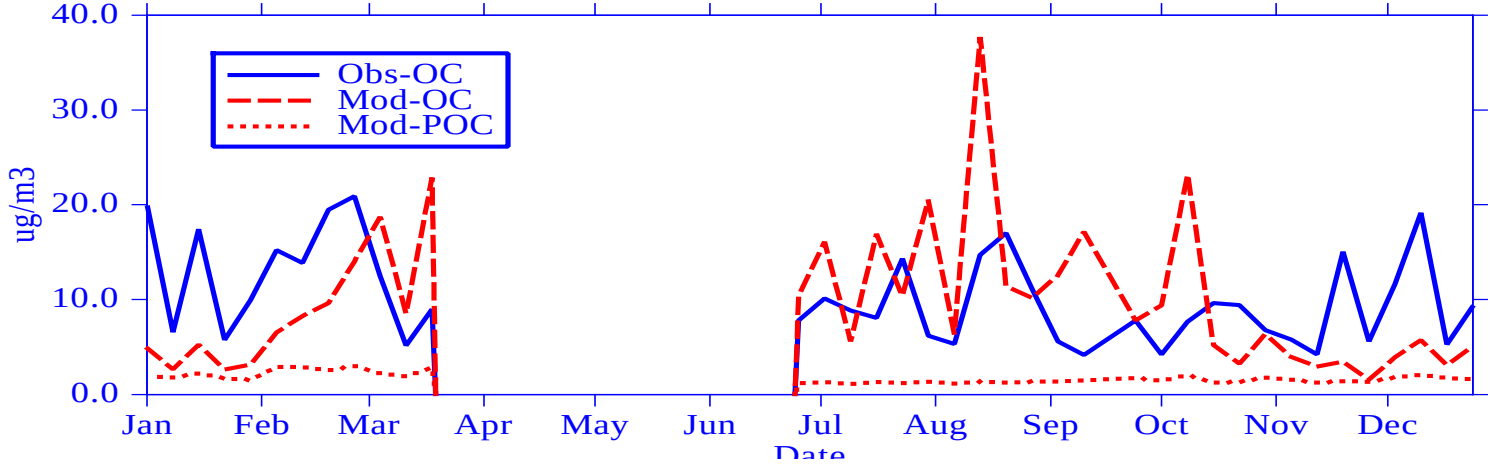
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Levoglucosan

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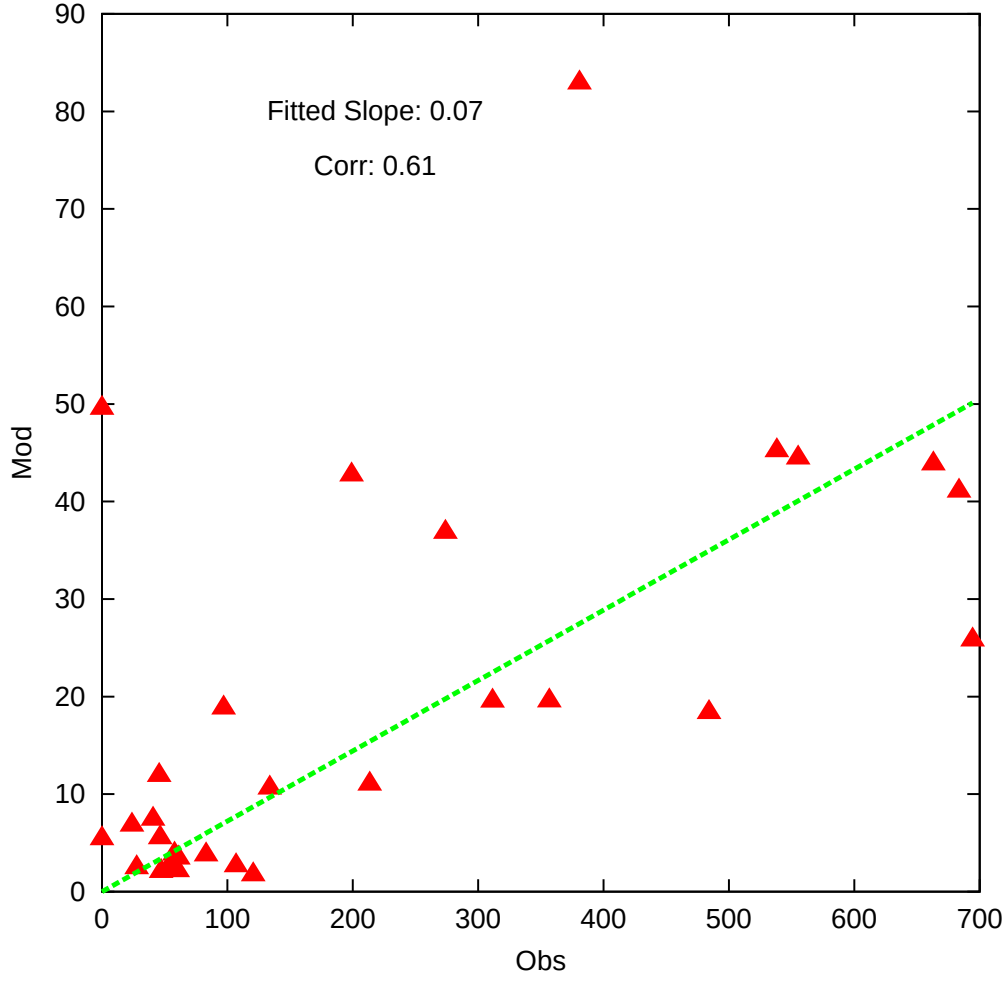
- Known problem - not all residential emissions are reported

LEV, K-Puszta

(Units: ng m^{-3})

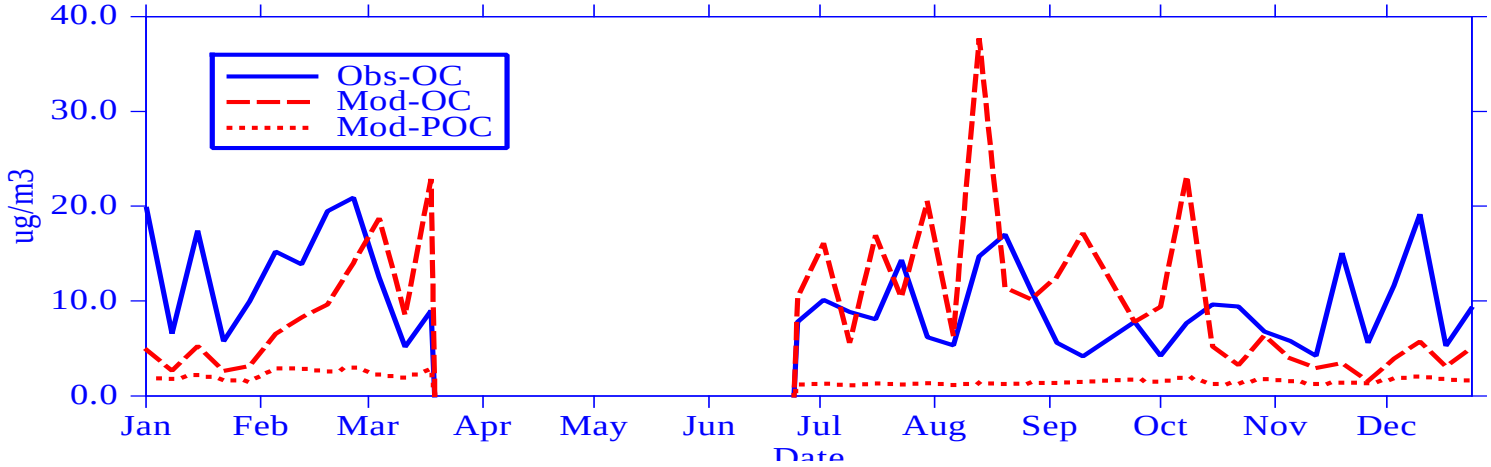


LEV - conclusions

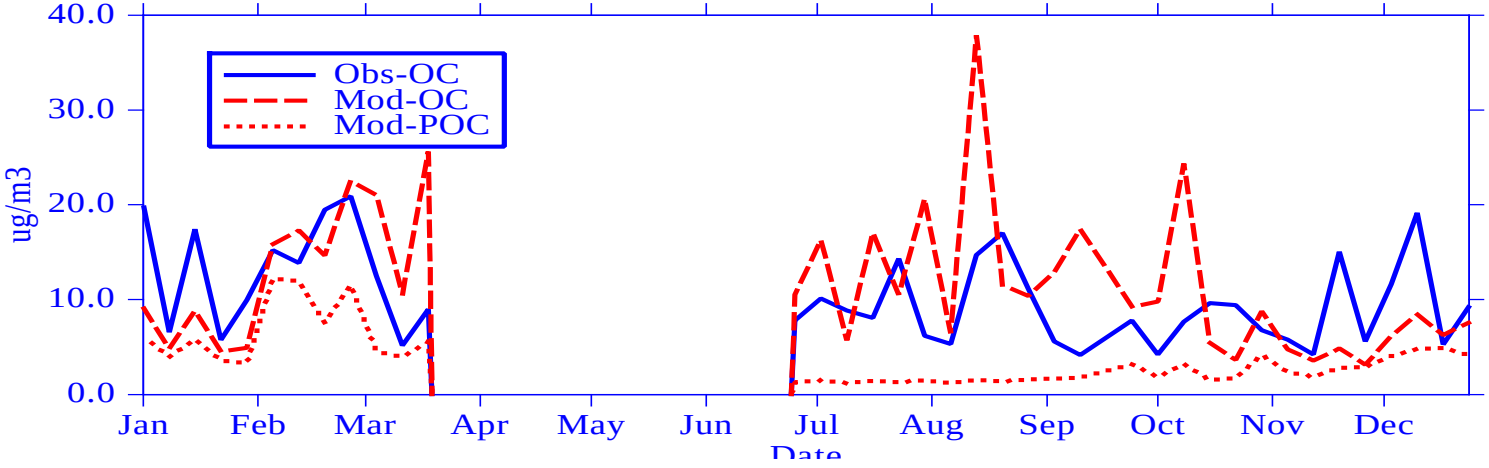
- Results from ’first-estimate’ suggest that we undepredict POC from wood by a factor of 10.
- Likely much more, since our LEV is a surrogate! Levoglucosan $\sim$ 10% POC-WOOD more likely for ’real’ emissions.
- Also very likely that emissions of POC-WOOD not reported properly.
- Experiment: ’correct’ POC-WOOD by factor 40. . . (!)

Experiment:

Orig.:



After:



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- Correction for this might help explain a significant fraction of missing OC in wintertime, but not summer
- Seems likely that the missing OC results from both SOA and missing POC.

State of SOA Modelling

- Model theories change every year!
- Over last 10 years we have seen:
  - Fixed-yield theories
  - Need to exceed P^{sat}
  - Gas-Particle partioning (α-K) - succesful for smog-chambers
- Possible reactions within the aerosol complicate most current theories!
- Increasing evidence for polymerisation and other reactions within aerosol

Conclusions cont.

- Models can provide almost any number required for OC!
- SOA theories are too immature for application within EMEP policy framework
- Measurements are required to constrain models and validate emissions
- Needs chemical speciation, tracers, many locations
- Main wishes:
  - Primary versus secondary contributions
  - Anthropogenic versus biogenic

Acknowledgements

- NILU - EC/OC data
- Paul Makar/AE Canada - Vapour pressure + UNIFAC
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  - CARBOSOL
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