Evaluation of organic aerosol in CTMs by comparison with positive matrix factorization outputs throughout Europe

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OA PMF

- Inside the European project (lead by X. Querol), PMF on Organic Aerosol (OA) have been performed over 31 sites for the 2017-2019 period with a harmonized methodology

- Provide information on:

  - HOA : Hydrocarbon-like Organic Aerosol

  - BBOA : Biomass Burning Organic Aerosol

  - LOOA : Lowly Oxidized Organic Aerosol

  - MOOA: More Oxidized Organic Aerosol

  - On some stations some other factors can appear in the PMF. Especially COA (Cooking Organic Aerosol)

Comparison between CTM and PMF

- Objectives: Propose a methodology to compare results of CTM to PMF contributions
- Difficulties:

  - SOA mechanisms do not have the same precision (e.g. some mechanisms may treat POA as nonvolatile without aging). Some mechanisms may account for the degree of oxidation, other do not.

  - The comparison to L-OOA (low oxidized) and M-OOA (more oxidized) is not straightforward. No clear criteria on O/C to distinguish compounds in L-OOA to M-OOA

  - Possibility that the PMF allocate part of primary compounds into L-OOA

	CHIMERE	CAMx	EMEP	LOTOS-EUROS
SOA scheme	Wang et al. (2024) Mechanism reduced from explicit MCM+PRAM mechanism for monoterpenes and sesquiterpenes.	VBS-1d (Jiang et al., 2019), which describes the evolution of OA in the 2-D space of oxidation state and volatility.	Standard VBS approach: "NPAS" scheme of Simpson et al. (2012), see also Bergström et al. (2012)	Standard VBS approach: Sturm et al. (2023)
POA treatment	Semivolatile. According to the volatility distributions, at 298K and 2 μg/m³, 27% and 17.9% of primary SVOC are in the particle for biomass burning and for other sources, respectively.	Nonvolatile	Nonvolatile	According to the volatility distributions, at 298K and 2 μg/m³, 56% of primary SVOC are in the particle for all sources.
Aging of SOA	Aging represented explicitly for monoterpenes and sesquiterpenes	No aging of SOA except for SOA from biomass burning	Aging for all SOA.	Aging for anthropogenic SOA. No aging for biogenic SOA
Aging of POA	A single oxidation step of aging.	No aging as POA is nonvolatile	No aging as POA is nonvolatile	As aging of anthropogenic SOA
IVOC	No IVOC emissions	No IVOC emissions	No IVOC emissions	IVOC/POA=1.5. IVOC are distributed for C*>100 ug/ m³.

VBS for EMEP, CAMX and LOTOS-EUROS
Mechanism reduced from explicit mechanisms for CHIMERE

Nonvolatile POA: EMEP, CAMX
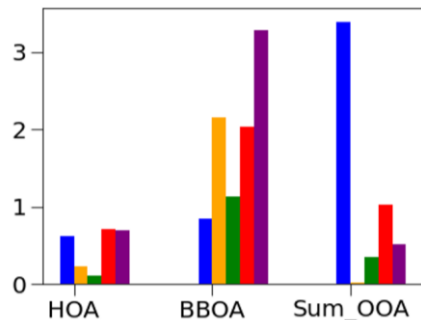Semivolatile POA with LOTOS and CHIMERE (but with very different volatility distribution)

Aging in all models but treated differently
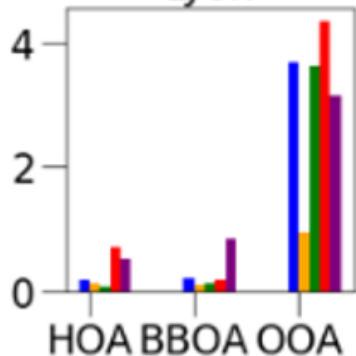
IVOC emissions only for LOTOS-EUROS

Typical results

- Very different results between stations but main conclusions can be illustrated by the ATOLL station

ATOLL station (near Lille)
Winter

Lyon station
Summer

Legend: Observations, Lotos, Chimere, CAMx, EMEP

HOA strongly underestimated by models with semivolatile POA (LOTOS and CHIMERE). Generally good results with EMEP and CAMX.

BBOA overestimated by all models but to a different degree. CHIMERE is generally close to observations in winter

OOA: generally (except for a few stations) all models strongly underestimate OOA in winter
In summer, CHIMERE, CAMX and EMEP are generally around the same level of concentrations and are not so far from observations at some stations.
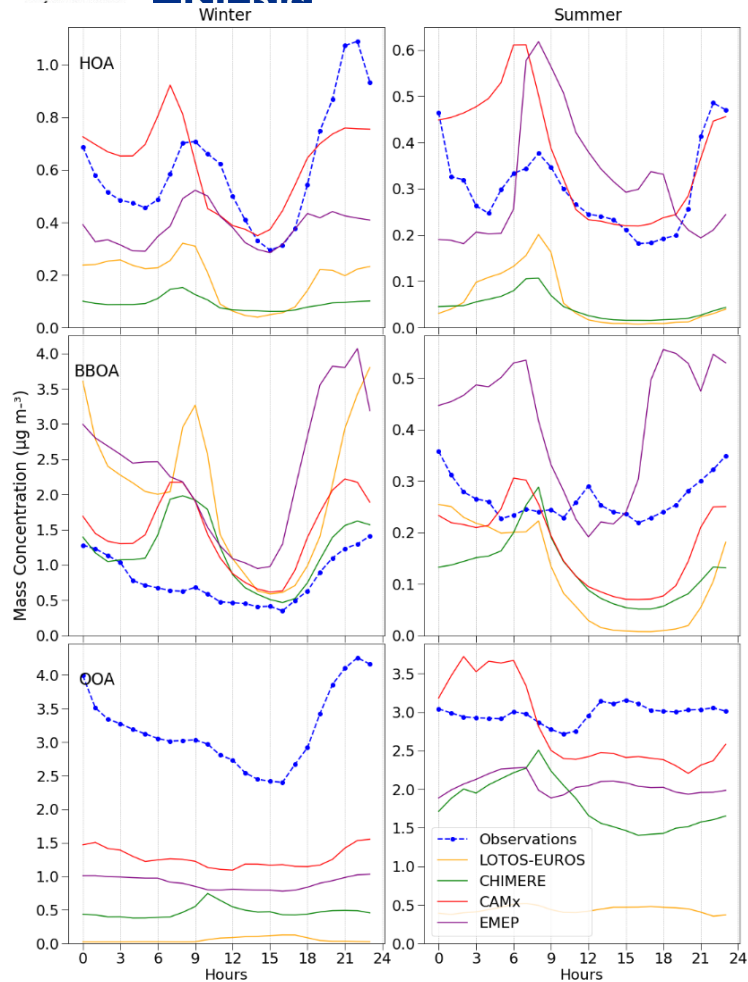
Comparison of diurnal profiles:

- No model quite agree with the diurnal profiles from the PMF

- HOA peak in the evening in missing in all models in summer. This peak appear in some models in winter but seems underestimated

- BBOA: all models (except EMEP) predict a peak in the morning that do not appear in the PMF results

An issue has been found in the emission temporalization with EMEP. Results need to be updated.

Should the matching be revised?

- How to improve results:
  - Semivolatile POA seem to improve the simulation of BBOA and to degrade HOA
  - Could some of the aged POA still be considered as HOA?
  - BBOA really all OA from biomass burning

- Alternative methodology:
  - BBOA linked to m/z=60 in the ACSM. Should be related to sugars and anhydrosugars (levoglucosan). BBOA="Sugar liked OA"
  - HOA should be related to almost non-oxidized POA and SOA. In some studies, compounds with O/C<0.2 should be regarded as HOA (may included some aged POA)
  - LOOA and MOOA could be defined as compounds with O/C < 0,7 and O/C>0,7

Sensitivity simulation with CHIMERE

- Revision of the CHIMERE simulation:
  - Add IVOC emissions, update of the toluene mechanism, add several aging steps
  - Add aging with NO_3 for wood burning SVOC (following Kodros et al., 20202)
  - Use of CAMS-TEMPO temporal profiles
  - HOA defined as O/C<0,2
- Now good temporal profile. Due to aging with NO_3 and use of CAMS-TEMPO
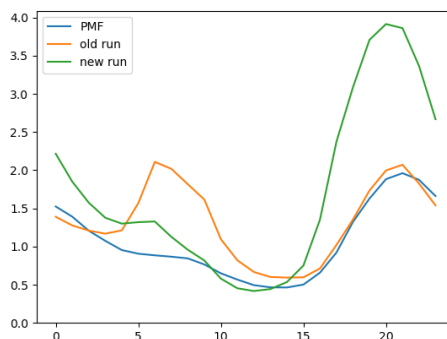
Diurnal profile of OM in winter
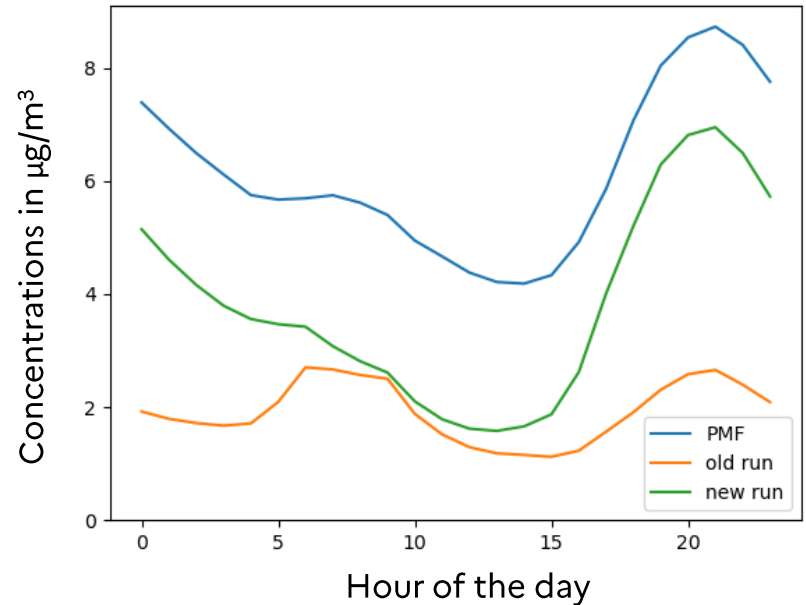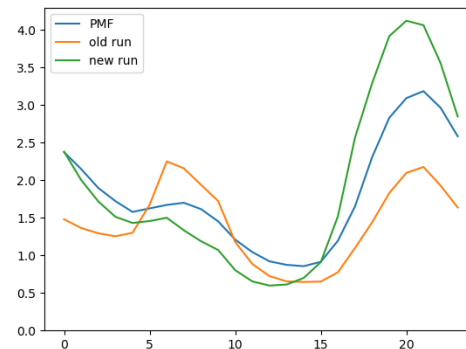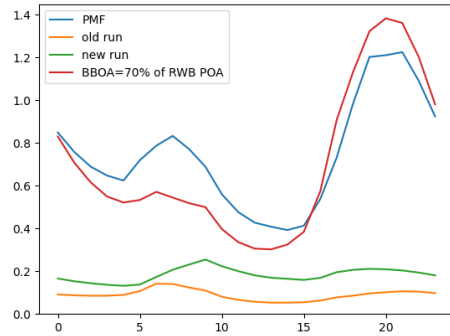
Sensitivity simulation with CHIMERE (winter)
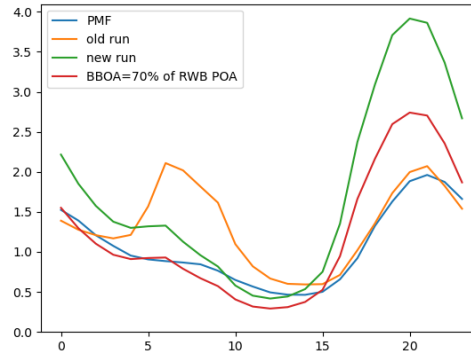
HOA
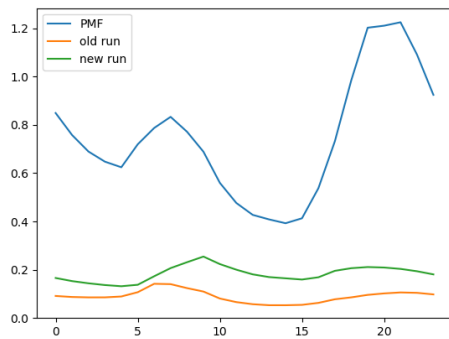
BBOA

HOA+BBOA

Good amount of primary (HOA+BBOA) but issues with HOA and BBOA. Issues with the split between HOA and BBOA
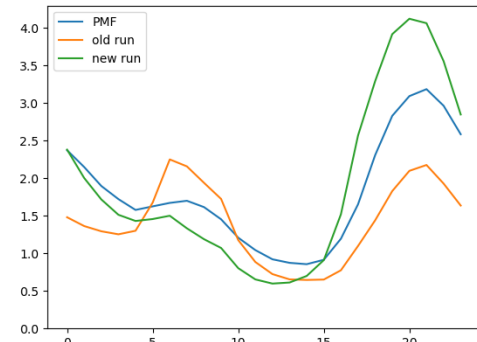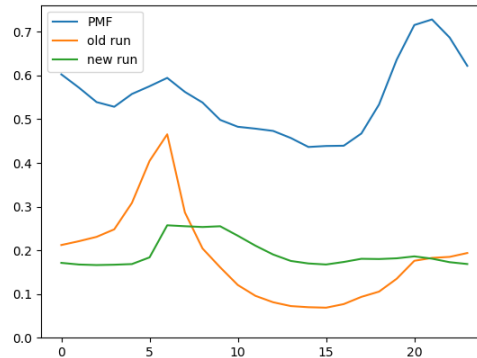
Sensitivity simulation with CHIMERE (winter)

HOA

BBOA

HOA+BBOA

Red curve. 70% of RWB POA in BBOA, the rest in HOA

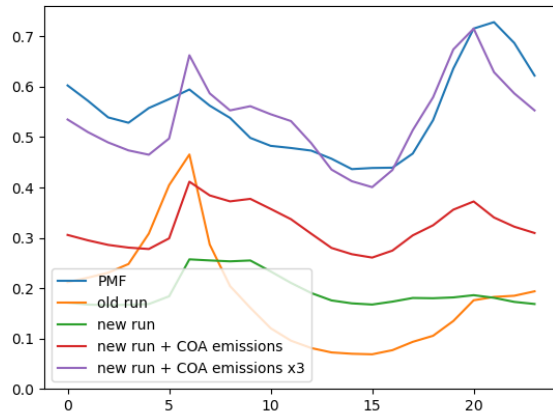# Sensitivity simulation with CHIMERE (summer)

HOA+BBOA

Lack of primary PM. Still of late night of OA that is not represented in the model.

Sensitivity simulation with CHIMERE (summer)

Ots et al. (2016) propose to add emissions based on population density.

Assuming nonvolatile COA, they determine COA emissions = 320 mg person^{-1} day^{-1}
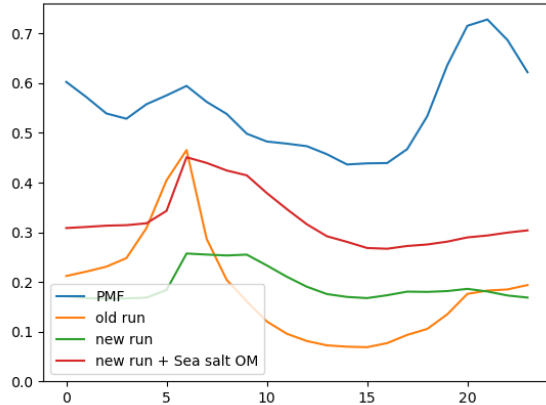
HOA+BBOA

Effect of adding COA emissions (red curve) with the volatility profile of RWB OA

Purple curve. Emissions multiplied by 3 (to account that around 1/3 of primary SVOC should be in the particle)

Sensitivity simulation with CHIMERE (summer)

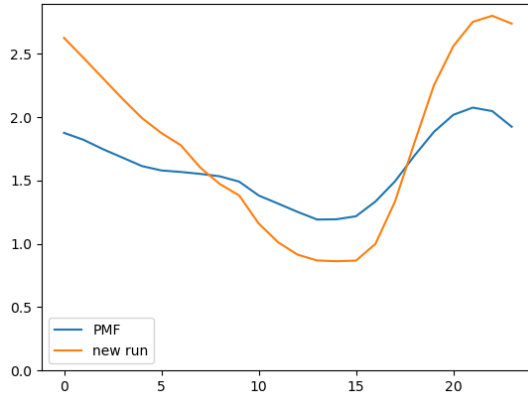Another possible contribution is the OM from sea salt

HOA+BBOA

Red curve: add of sea salt OM
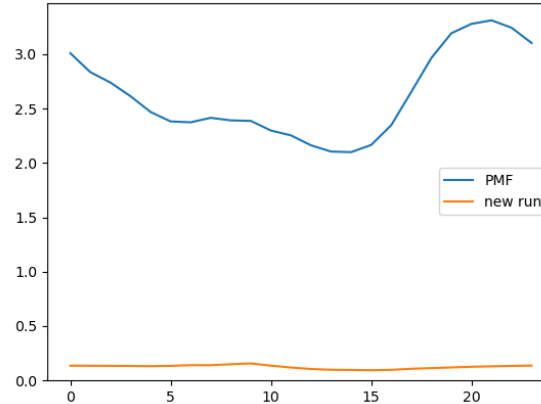based on Gantt et al. (2012)

LOOA and MOOA

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RÉPUBLIQUE FRANÇAISE
Liberté
Égalité
Fraternité
maîtriser le risque
pour un développement durable

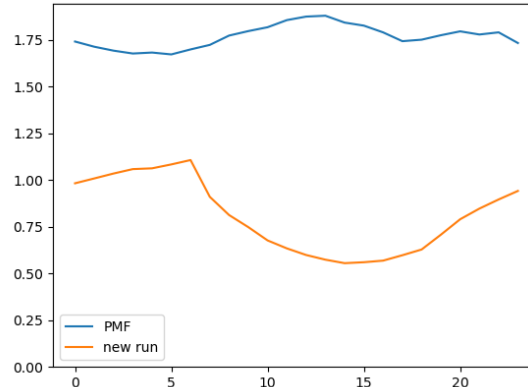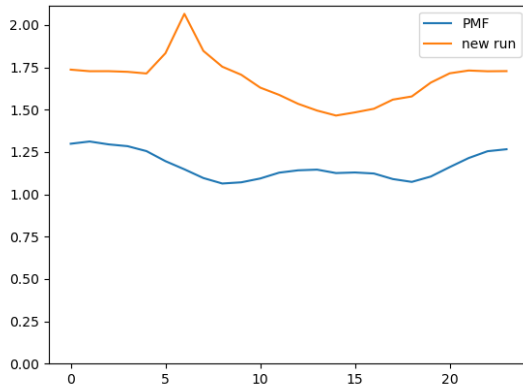Assumes that RWB aging SOA are LOOA

LOOA

MOOA

Winter

Summer

Winter MOOA missing
Probably a part of RWB aging SOA are MOOA

Too much LOOA and lack of MOOA
Perhaps due to the aging mechanism

Conclusions

Comparing CTM with PMF results is not straightforward and not easy with a lot of methodological issues. However, trying to compare the two of them could help to:

- Better represent organic aerosols in CTMs
- Better understand the PMF results and the meaning of the different factors

Some important conclusions:

- Representing the semivolatile aspect of POA in the models may not help to improve the OM results at first but the tests done with CHIMERE seem to indicate that it could improve the simulation of total POA (HOA+BBOA) in winter.
- POA in summer could originate from sources generally not taken into account in models (COA, primary sea salt)
- The aging mechanism is important. Probably an effect of nighttime oxidation. Probably a need for mechanisms that better represent HOA -> LOOA -> MOOA