Barcelona Supercomputing Center
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EGAR ENVIRONMENTAL GEOCHEMISTRY AND ATMOSPHERIC RESEARCH

"Experimentally-constrained organic aerosol chemical and absorption properties in the atmospheric chemistry MONARCH model"

Hector Navarro-Barboza

Directors:

Oriol Jorba[1]
Marco Pandolfi[2]

28/09/2021

[1]Barcelona Supercomputing Center (BSC), Earth Sciences, Barcelona, Spain
[2]Institute of Environmental Assessment and Water Research (IDAEA-CSIC), Spain

Outline

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- *Objectives*
- *Methods*
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  - *Simulation setup*
- *Results*
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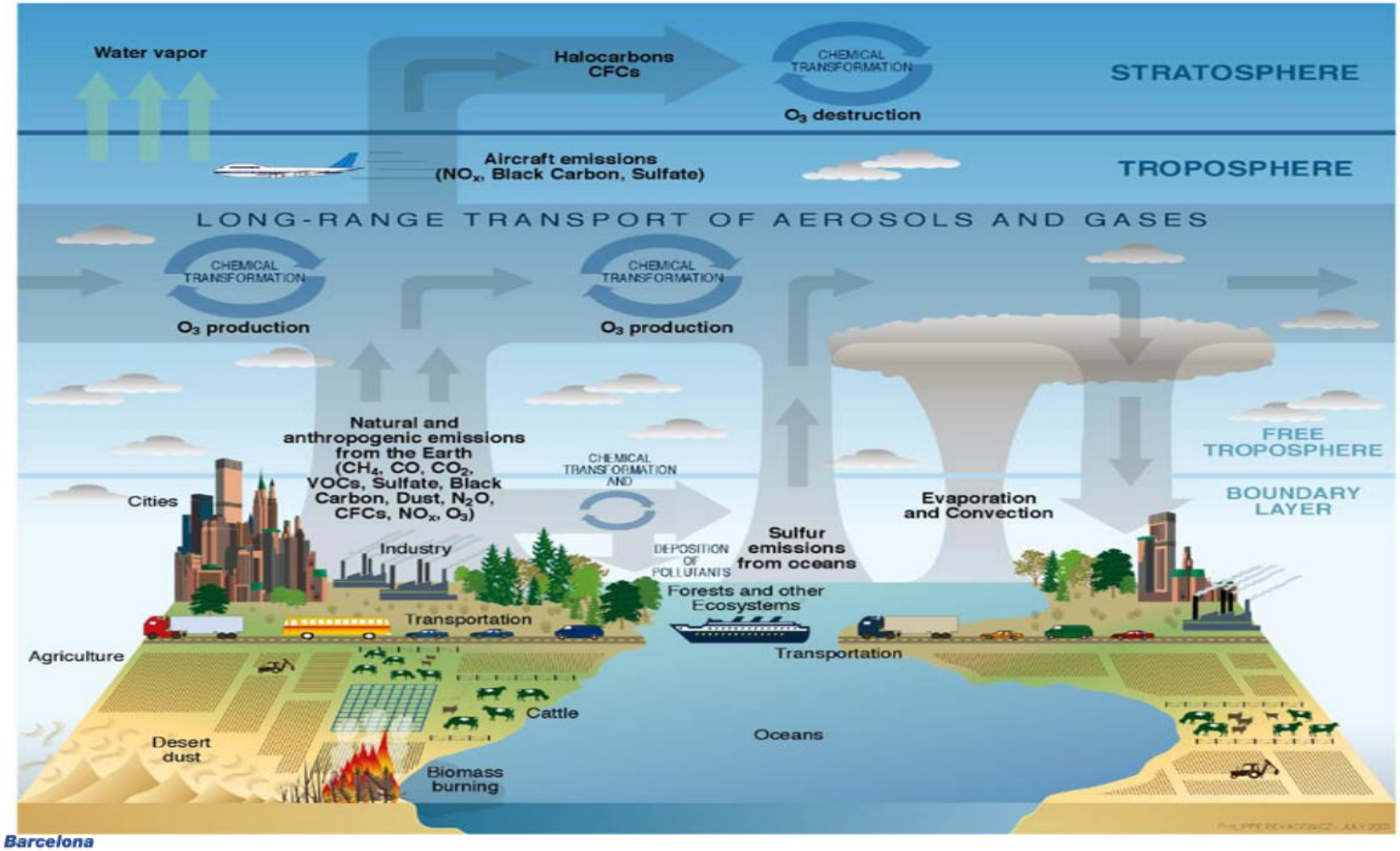
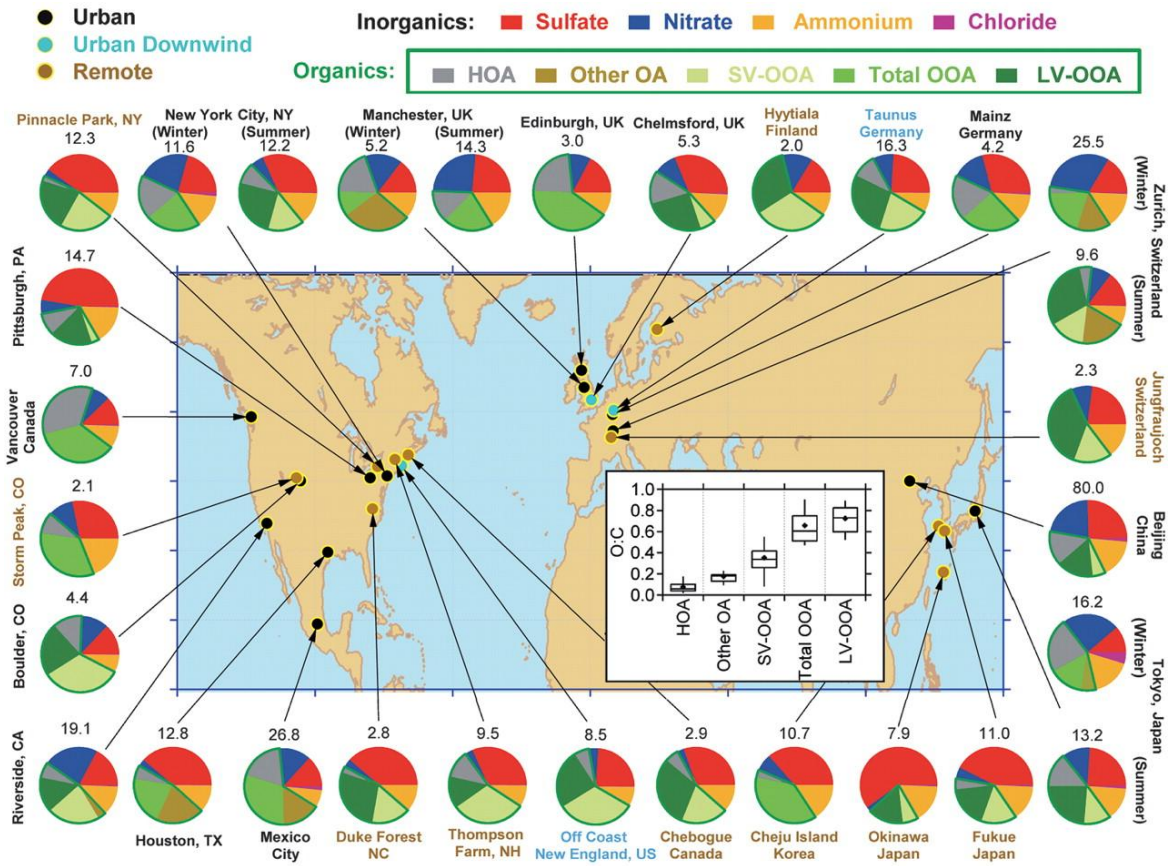
Introduction: Atmospheric aerosols

Atmospheric aerosols: Solid or liquid particles suspended in the atmosphere to the exception of all hydrometeors (cloud droplets, ice crystals, raindrops, snowflakes, and graupel); their aerodynamic radius can ranges from $0.002\mu m$ to about $100\mu m$..



Motivation

Atmospheric aerosols can alter the balance of energy in the atmosphere. Depending on their chemical composition, aerosols cool or warm the atmosphere. From all aerosol types, organic aerosols (OA) represent a significant fraction.



Aerosol chemical composition (Jimenez et al., 2009)

Motivation

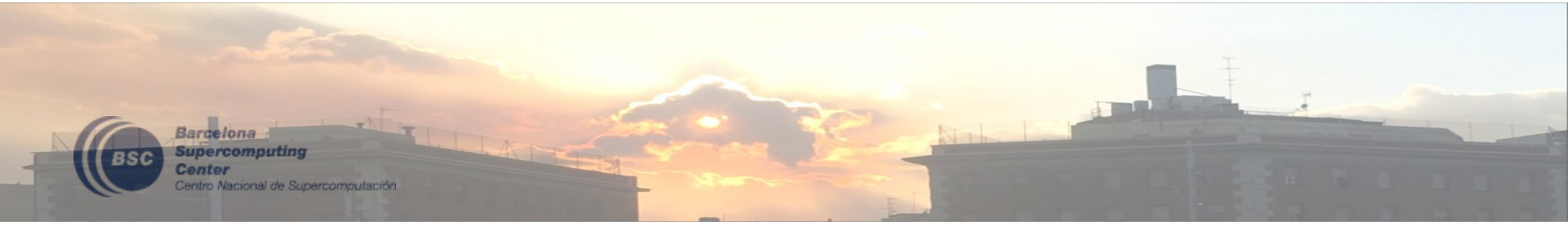
OA has been classically treated in atmospheric models as mainly scattering particles; however, the findings of researchers through laboratory experiments have demonstrated that a fraction of OA is absorbent, even recent research has found certain compounds of secondary origin (SOA) tend to change their color from white to brown while are aged in the atmosphere. The term “brown carbon” (BrC) has emerged to describe this type of OA characterized by an absorption spectrum that increases from visible to UV wavelengths.

Effect of mixture changes in filter appearance (Jaoui et al., 2008)

Objectives

- Ongoing work to experimentally constrain the absorption property of OA simulated by the Multiscale Online Nonhydrostatic Atmosphere Chemistry (MONARCH) model.

- Evaluation of the chemical composition of aerosols and OA optical properties at three in-situ super-sites in northeast Spain.



Methods: observational dataset

IDAEA-CSIC gas-phase and aerosols measurements:

Measurements has been made by IDAEA-CSIC group and still continues.

1. Two techniques are used to get the data: Off-line and On-line.
2. Measuring sites are Barcelona (BCN), Montseny (MSY), Montsec (MSA).

- Offline filter analysis of PM chemical composition: PM1, PM2.5, PM10.
- Online analysis: Aethalometer, Multi Angle Absorption Photometer, Aerosol Chemical Speciation Monitor.

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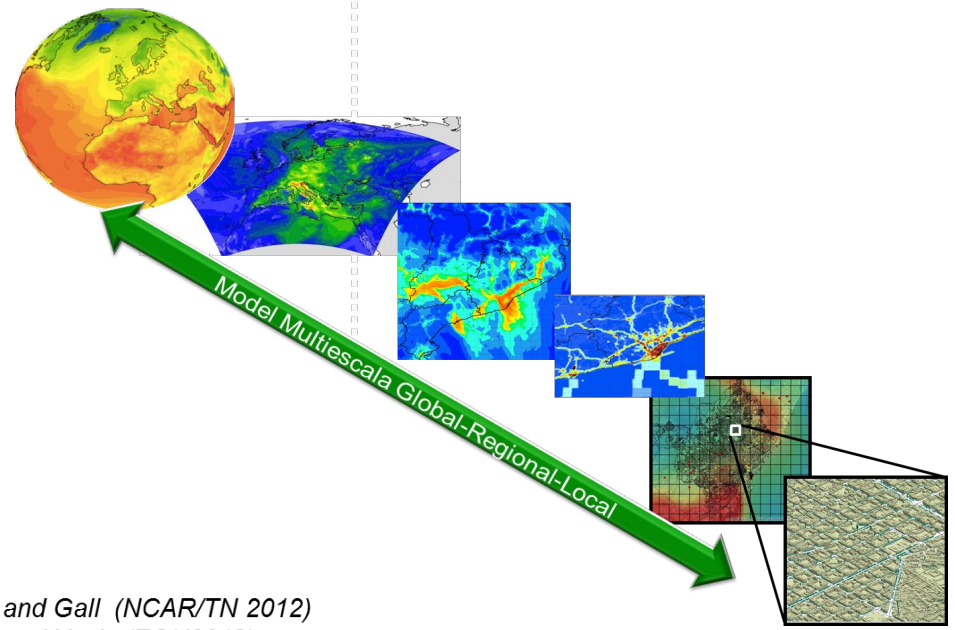
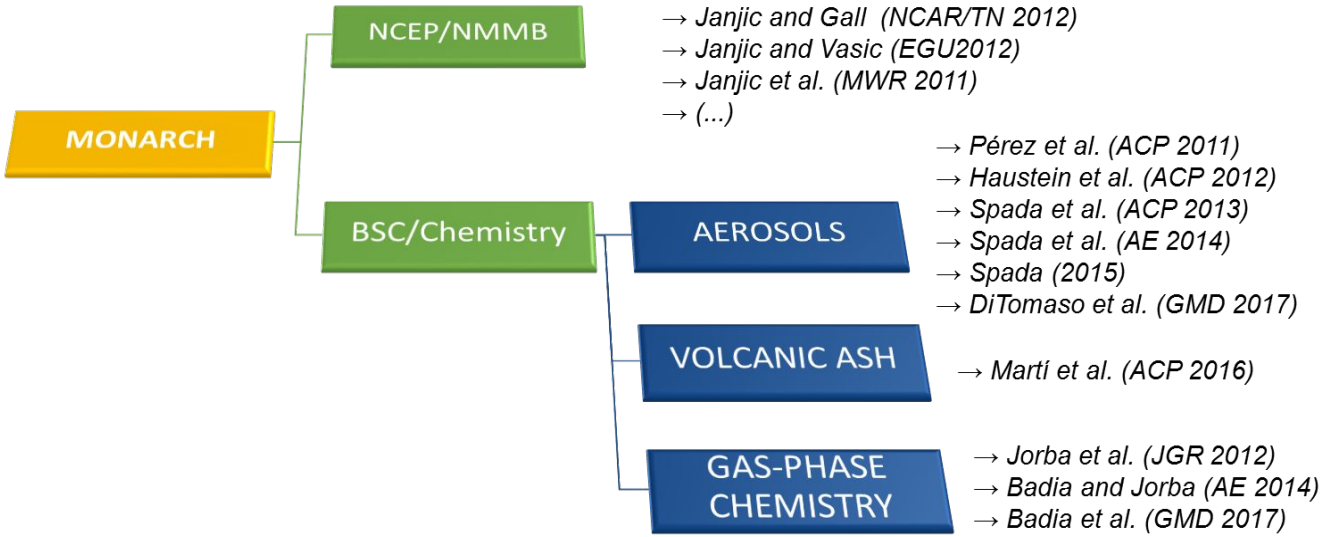
Methods: MONARCH model

- Fully **on-line** coupling: weather-chemistry feedback processes
- **Multiscale**: global to regional (up to 1km) scales (**nesting capabilities**)
- **Data assimilation** system
- Ready to be coupled with **Urban model**





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Multiscale Online AtmospheRe CHemistry (MONARCH) model

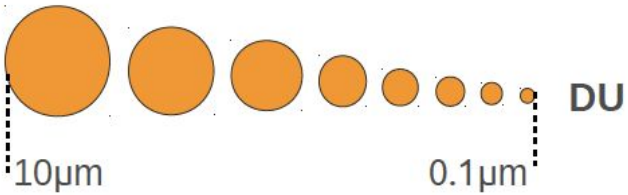
Methods: MONARCH aerosol representation

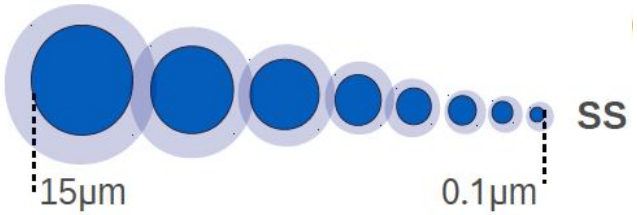
Mass-based scheme described in Spada (2015)

Sectional

dust (DU)

sea-salt (SS)





Bulk

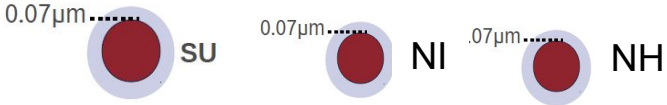
Black Carbon (BC) hydrophobic/hydrophilic modes

Organic Aerosols (OA)

Primary Organic Aerosols (POA) hydrophobic/hydrophilic modes

Secondary organic aerosols (SOA)

Sulfate (SU), Nitrate (NO3) and Ammonium (NH4)



Period simulation: 2018

Methods: sensitivity tests

- Perturbing BrC/OA fraction and imaginary refractive index (ni)

	Mass fraction	Imaginary refractive index (ni)		
	BrC/OA %	Hydrophobic Primary-BrC	Hydrophilic Primary-BrC	SOA-BrC anthro./biomass
Case 1 - no BrC	0.	---	---	---
Case 2 - Strongly P and S BrC absorption	100./50./20.	1e-1	1e-1	1e-2
Case 3 - Moderately P and S BrC absorption	100./50./20.	1e-1	1e-2	1e-2
Case 4 - Moderately P and weakly S BrC abs.	100./50./20.	1e-1	1e-2	1e-3
Case 5 - Moderately P and very weakly S BrC abs.	100./50./20.	1e-1	1e-2	1e-4
Case 6 - Weakly P and very weakly S BrC abs.	100./50./20.	1e-2	1e-3	1e-4

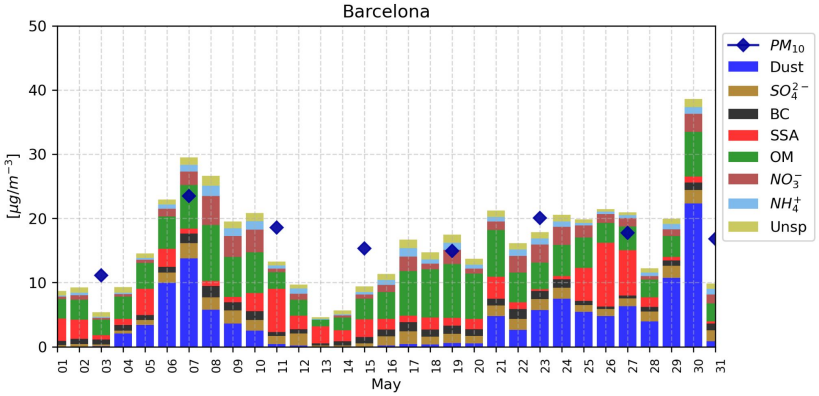
P: primary; S: secondary

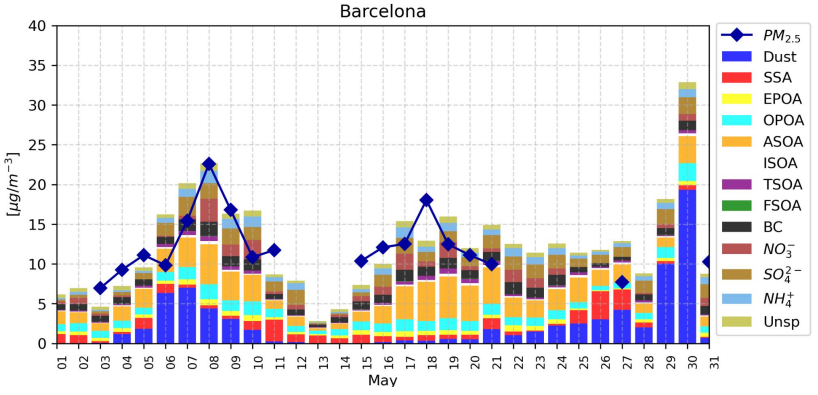
Results

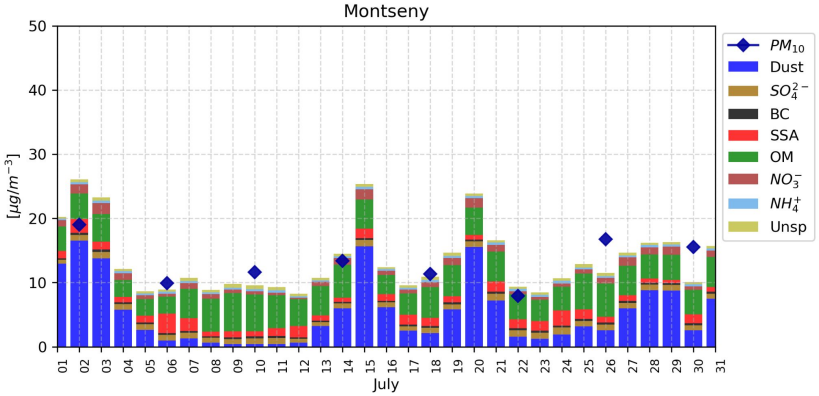
- Next slides show results for May 2018 (Barcelona site) and July 2018 (Montseny and Montsec)
  - Months with good agreement with BC observations
  - BC model biases in other periods attributed to uncertainties on emissions and lifetime
  - Allows isolating the effect of BrC

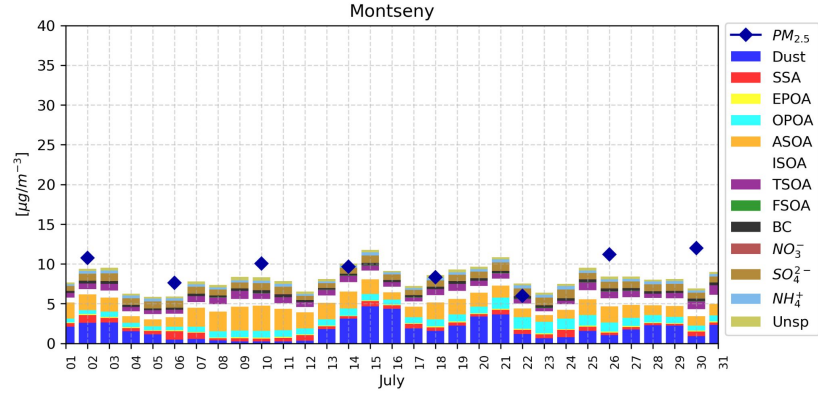
Results: PM chemical composition

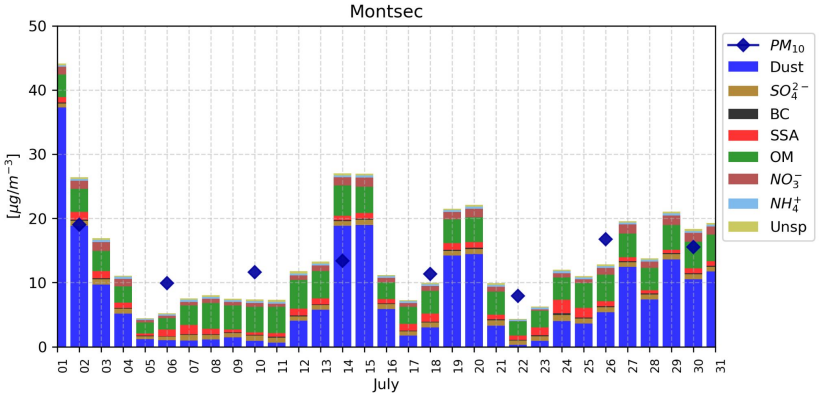
PM10, PM2.5 model composition and sum of off-line PM chemistry observation

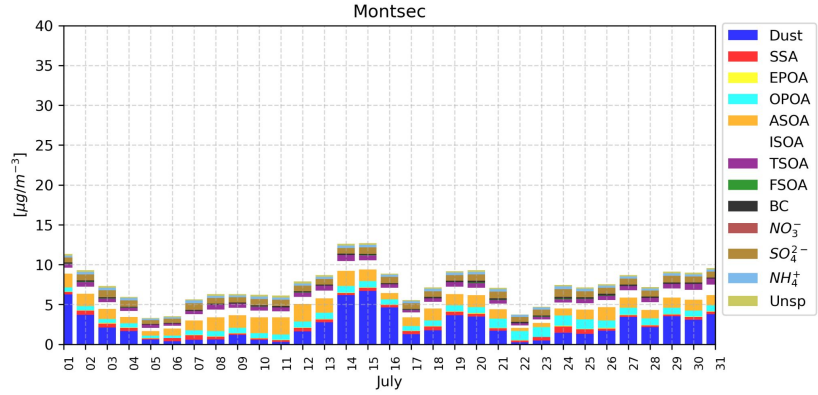




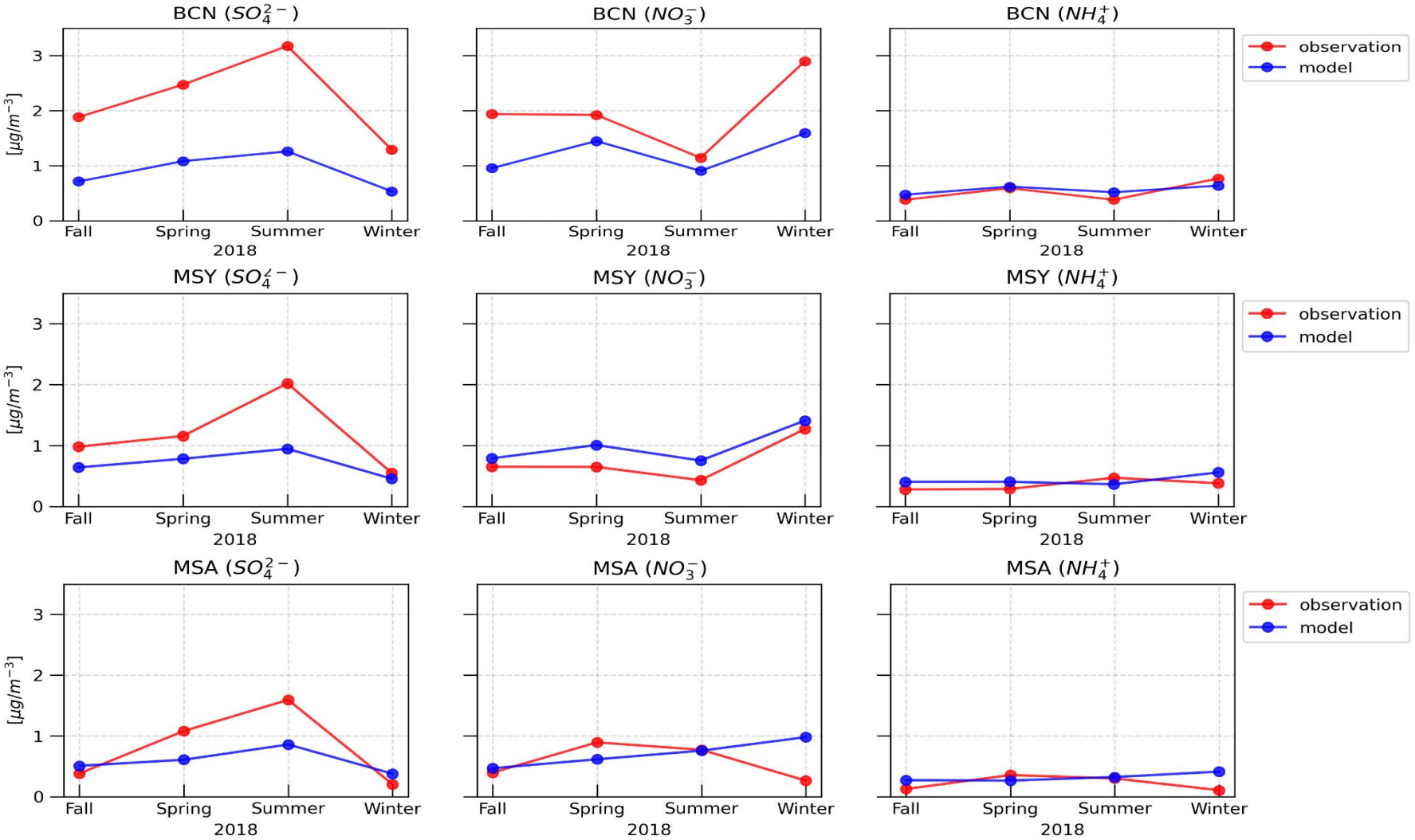






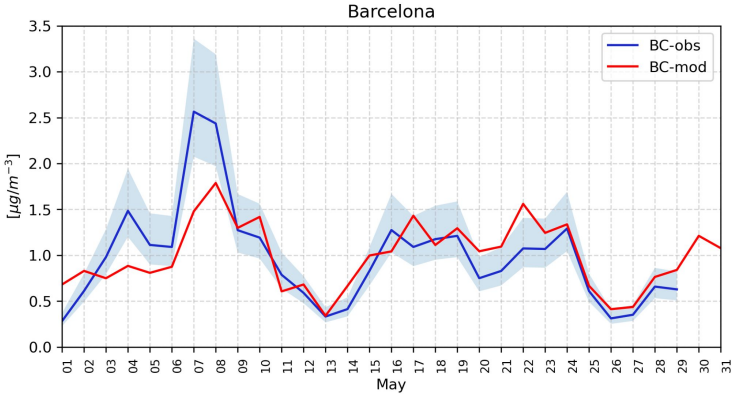


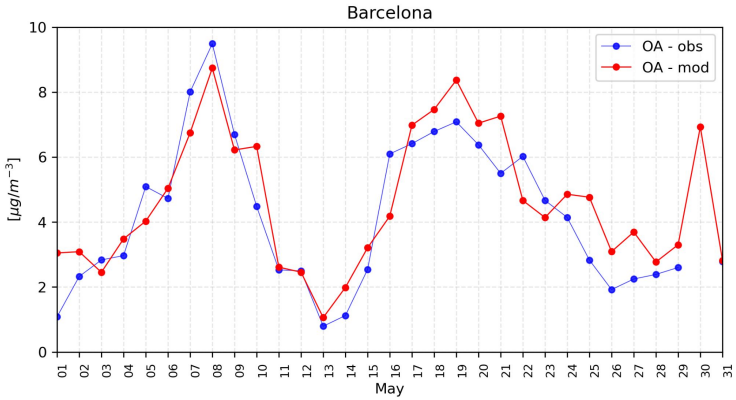
Results: Sulfate, nitrate, and ammonium

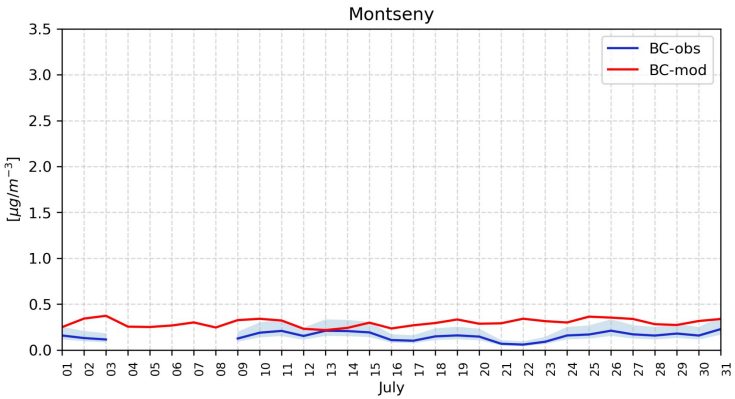


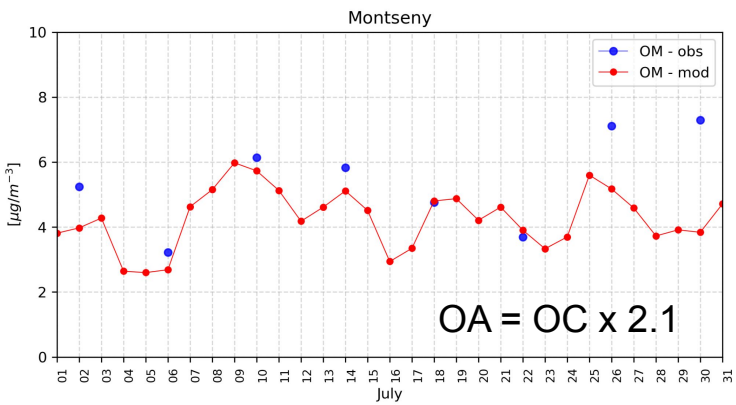
Results: BC and OA mass

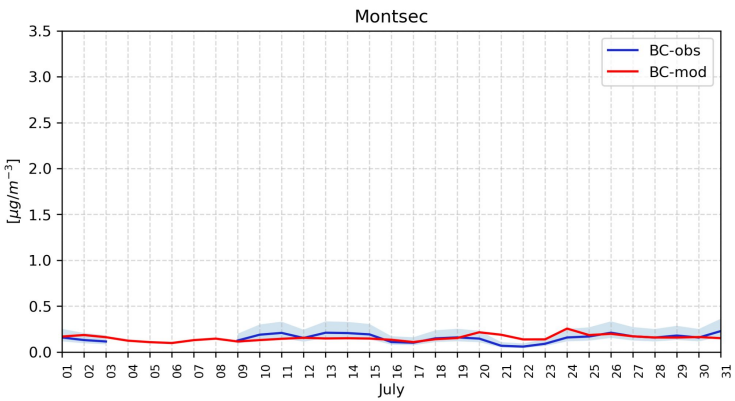
Observations: BC derived from MAAP, OA (ACSM BCN, filter MSY/MSA)

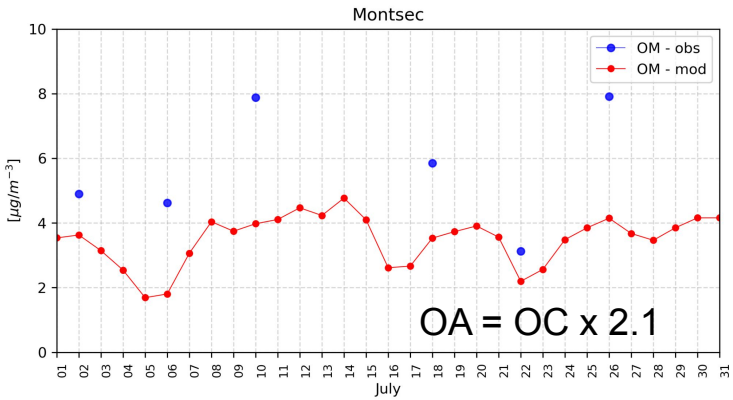






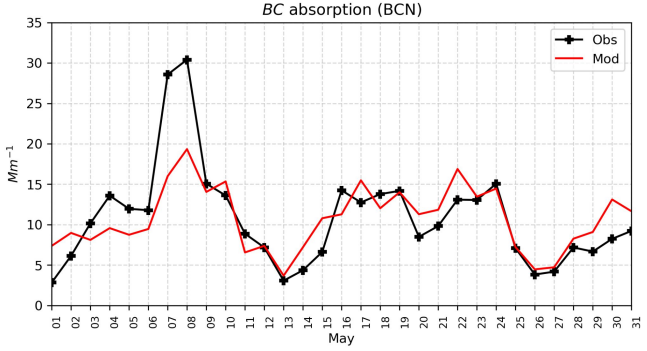


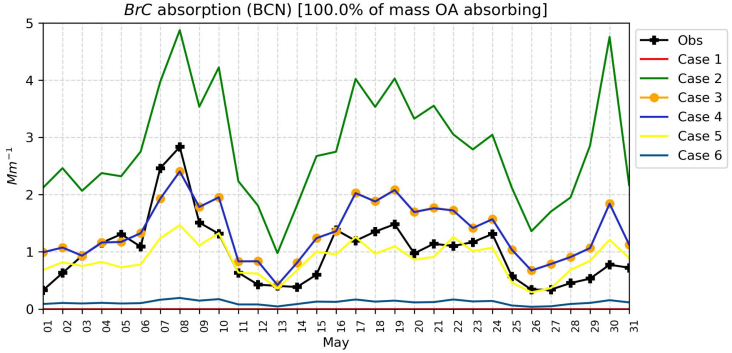


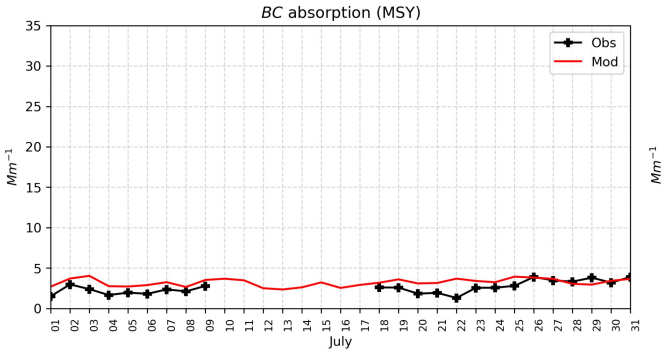


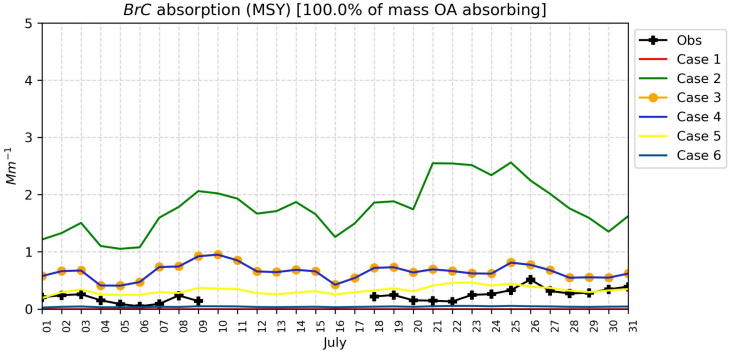
Results: BC and BrC absorption 520nm

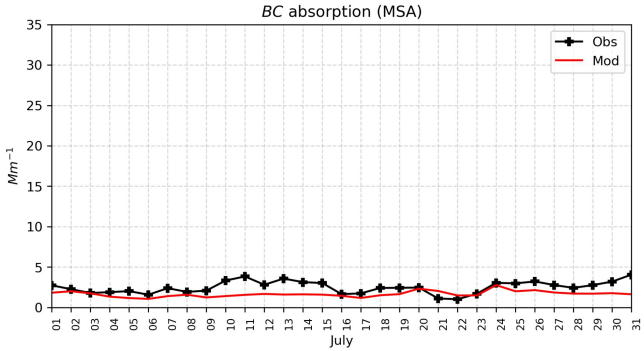
Observations: BC and BrC (derived from aethalometer)

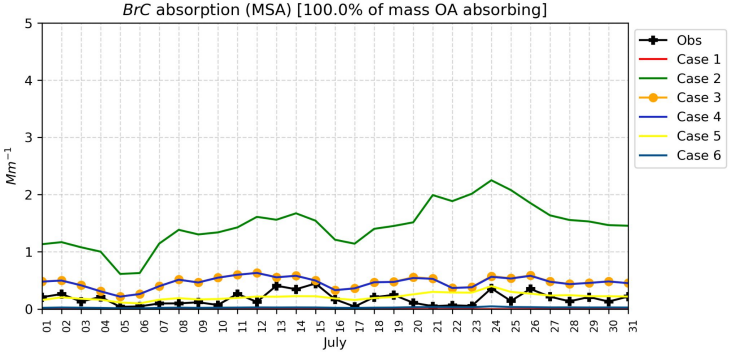












Absorption explained by large abundance of BrC with a mix of primary strongly and secondary weakly absorbing aerosol in urban site

Regional (MSY) and remote (MSA) sites likely not dominated by a large abundance of BrC

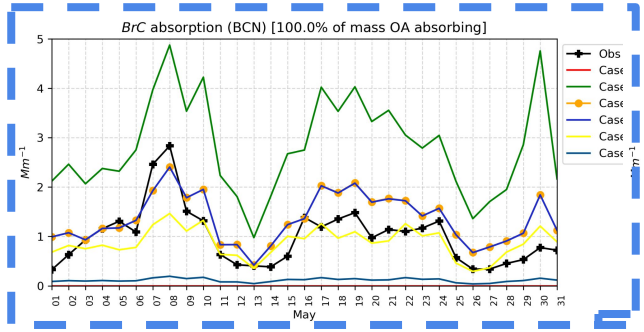
Results: sensitivity BrC

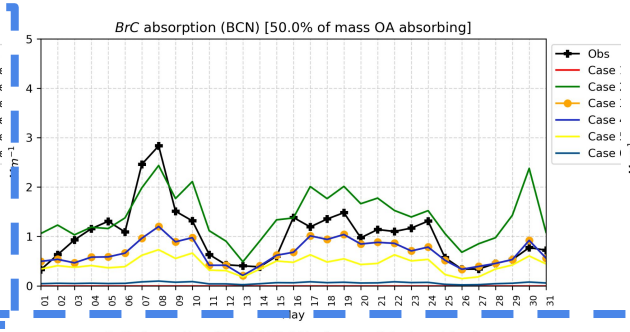
Perturbing BrC/OA fraction and imaginary refractive index

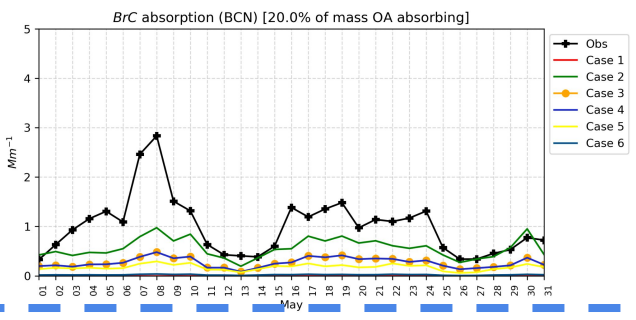
100% BrC/OA

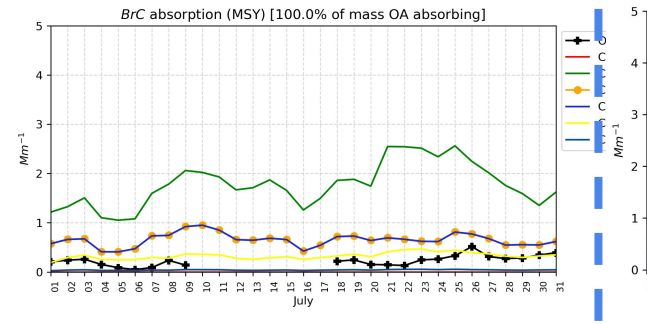
50% BrC/OA

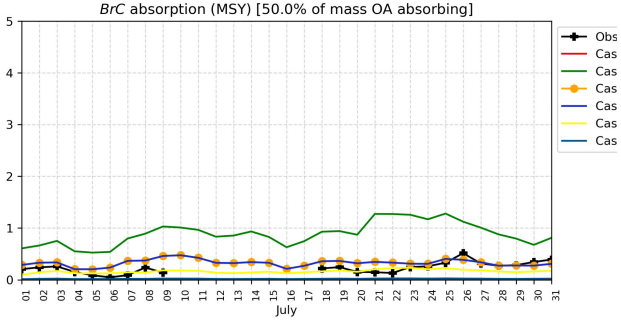
20% BrC/OA

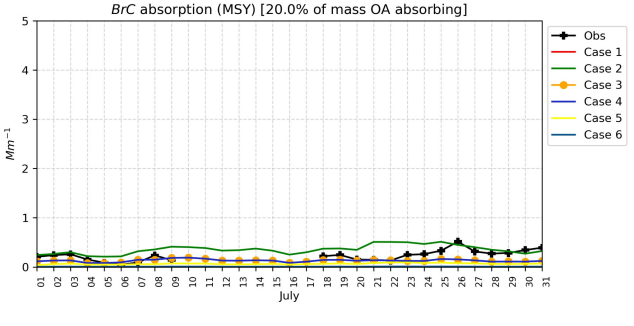


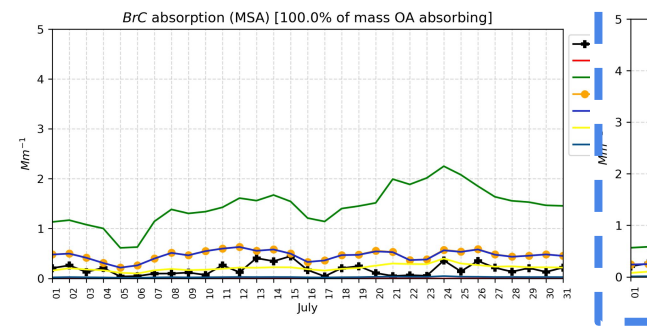


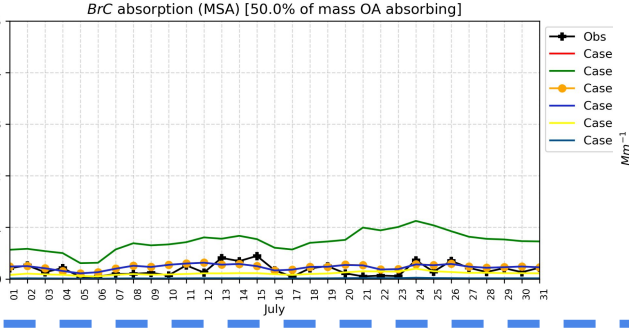


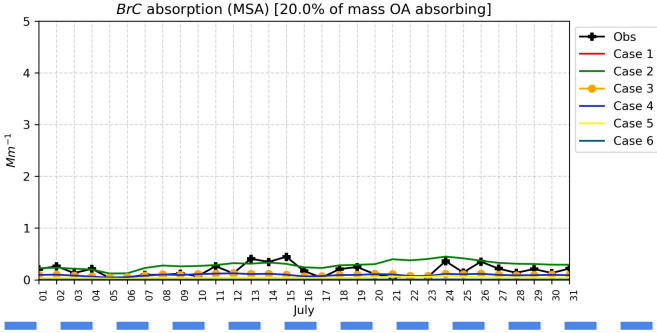












Summary

- Exploring different treatments of BrC in an atmospheric chemistry model
- Evaluation of the chemical composition and optical properties in three super-sites in northeast Spain
- Periods with a good characterization of BC and OA mass result with good agreement of absorption assuming a large BrC/OA fraction (urban site) and lower fraction (regional/remote site) with mixtures of weakly to strongly absorbing BrC
- The combination of different types of measurements allows identifying which components deserve further refinement in the model: secondary inorganic aerosols (sulfate, nitrate)
- The Organic Aerosol scheme in MONARCH performs very well
- Future work will extend the analysis to other European sites and global scale.

Thank you!
Any question?

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Acknowledgments

- Severo Ochoa Program, awarded by the Spanish Government [grant: SEV-2015-0493-18-I]
- Spanish Ministry of Economy and Competitiveness [grant: RTI2018-099894-B-I00]
- H2020 FORCES [grant: 821205]
- 'FPI SO' grant: PRE2018-084988]
- High Performance Computer resources of the "Red Española de Supercomputación" (AECT-2020-1-0007, AECT-2021-1-0027).

06/07/2021