Assessing the contribution of residential emission sources to carbonaceous aerosols concentrations in the western Mediterranean basin

Hector Navarro[1], Marco Pandolfi[2], Marc Guevara[1], Santiago Enciso[1], Carles Tena[1], Marta Via[2], Jesús Yus-Díes[2], Cristina Reche[2], Noemi Pérez[2] and <u>Oriol Jorba</u>[1]

[1] Barcelona Supercomputing Center, Barcelona, Spain

[2] Institute of Environmental Analysis and Water Research (IDAEA-CSIC), Barcelona, Spain

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Motivation

- Recent studies suggest that residential wood combustion (RWC) contributes to a significant fraction of carbonaceous aerosols (CAs) not only in urban areas but also in rural sites across Europe, mainly during cold seasons (Denier Van Der Gon et al., 2015).

- Condensable compounds refer to the portion of material emitted into the atmosphere as gas that rapidly condenses into particulate matter without undergoing chemical reactions.

- The inclusion or exclusion of this condensable fraction in emission inventories is one of the major uncertainties regarding CAs combustion sources, mainly due to the particulate matter measurement techniques used to derive emission factors (filter or dilution tunnel measurements).
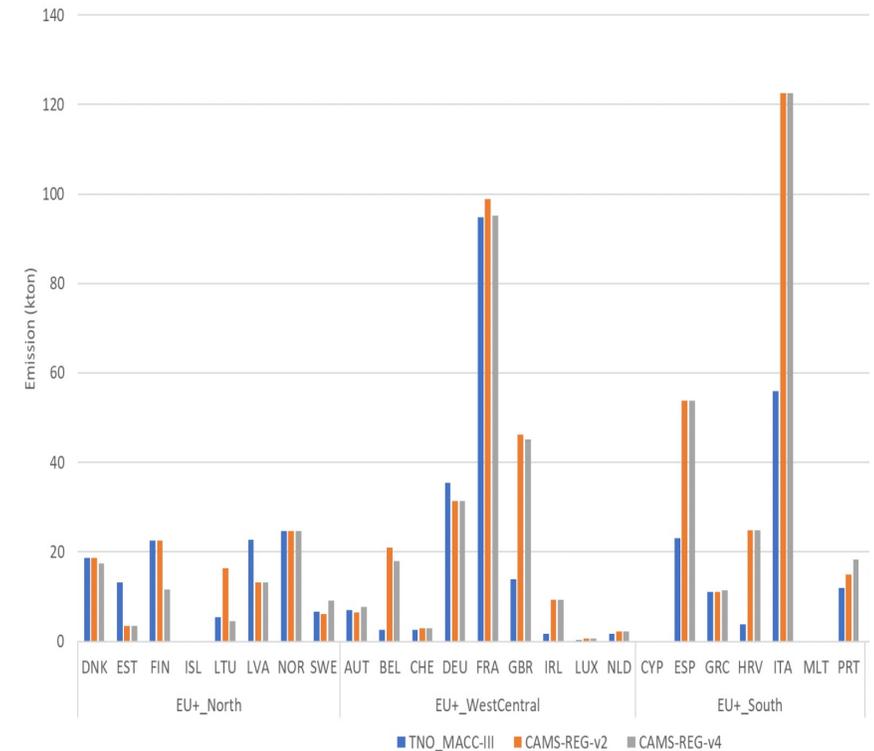
PM2.5 emissions from small combustion (GNFR_C) year 2010 (Kuenen et al., 2022)

Objective

- To quantify the **role of RWC residential emissions** and their characterization (CAs speciation, spatial distribution, and treatment of condensable fraction) on the concentrations of CAs, **focusing on black carbon (BC)**, **observed in northeast Spain**.

- To evaluate the main uncertainties associated with the description of CAs in NE Spain by comparing results obtained using **three different anthropogenic emission inventories** (Copernicus CAMS-REG and HERMESv3 bottom-up).

Methods

Domain of study and observational datasets
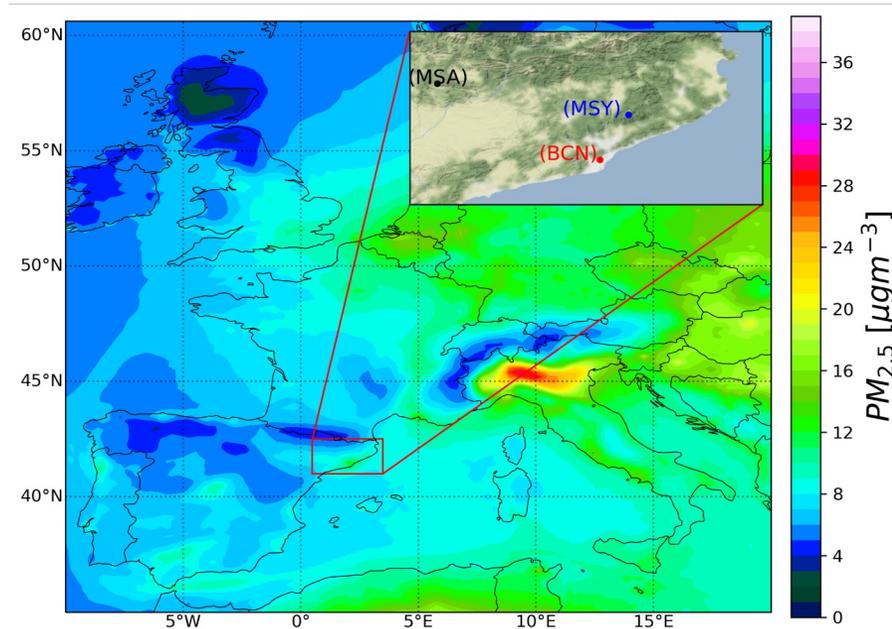
Station	Variable	Instrument
BCN	OC/EC	SUNSET analyzer
	OA	ACSM
	BC/Abs	MAAP/AE33
MSY	OC/EC	SUNSET analyzer
	BC/Abs	MAAP/AE33
MSA	OC/EC	SUNSET analyzer
	BC/Abs	MAAP/AE33

[a] Abs: absorption.

- Three atmospheric research supersites run by IDAEA-CSIC:
  - Urban background site in the city of Barcelona (BCN – 67 m asl)
  - Regional background site in Montseny (MSY – 720 m asl)
  - Remote background site in Montsec (MSA 1500 m asl)
- Online analysis: **Aethalomenter/Multi Angle Absorption Photometer (BC)**.

Source contribution derived from measurements: BC fossil/non-fossil

- Determination of fossil and non-fossil relative contributions to the measured concentrations of BC particles in BCN, MSY and MSA sites.

- The Sandradewi model (Sandradewi et al., 2008):
  - non-fossil sources of BC emit OA with absorption properties (BrC) that enhance the absorption of light in the ultraviolet wavelength range compared to that in the near-infrared range, where BC dominates the absorption.
  - Light absorption Multi-wavelength Aethalometer measurements can be used to assess the source contribution of non-fossil (e.g. wood burning) versus fossil sources (e.g. traffic, shipping, industry, etc) to BC concentrations.

- Source contribution uncertainty not less than 20% due to specific assumptions in the model (e.g., chosen Absorption Angstrom Exponents or equal Mass Absoprtion Cross section of BC emitted from fossil and non-fossil sources).

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Modelling tools

MONARCH AQ model
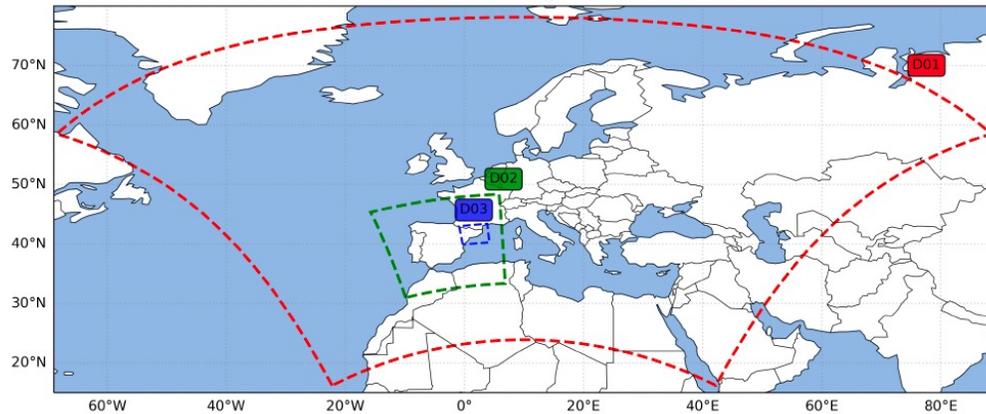
- *Online atmosphere-chemistry model*
- *Chemistry: CB05 + BSC aerosol scheme*
- *3 domains: **20**-**5**-1 km resolution, 24 layers (top 50 hPa)*
- *Emissions: CAMS-REG/HERMESv3, MEGAN, GFAS*
- *Period: 2018*

HERMESv3 emission model

A **python-based, open source, parallel and multiscale** emission model
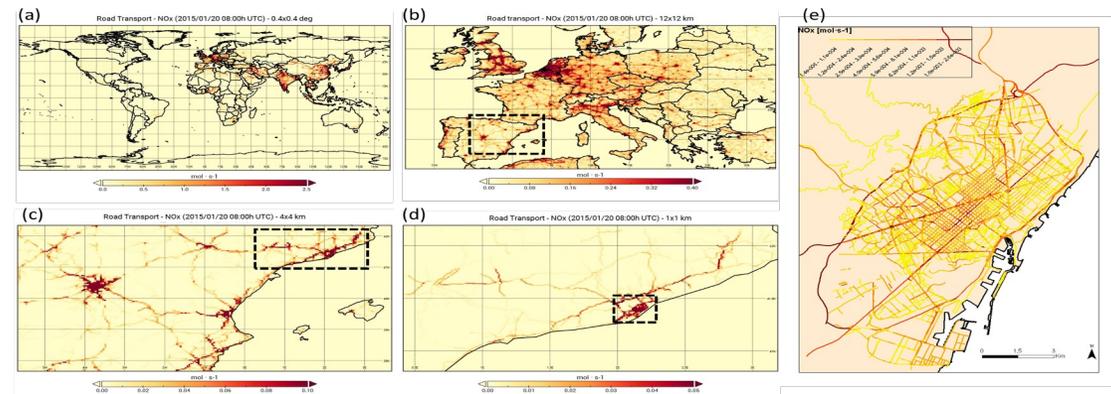
- *Global-regional module: processing existing gridded emission inventories*
- *Bottom-up module: computes emissions for Spain at the source level*

Emission inventories

	CAMS-REGv4.2	CAMS-REGv4.2_Ref2	HERMESv3_BU
Air pollutants	NOx, SO_2, NMVOC, NH_3, CO, PM_{10}, $PM_{2.5}$	NOx, SO_2, NMVOC, NH_3, CO, PM_{10}, $PM_{2.5}$	NOx, SO_2, NMVOC, NH_3, CO, PM_{10}, $PM_{2.5}$
Reference year	2017	2017	2018
Domain	Europe	Europe	Spain
Spatial resolution	0.1x0.05 deg	0.1x0.05 deg	User configurable (5-1km)
Temporal resolution	Yearly	Yearly	Hourly
condensable fraction RWC	Not consistenly	Yes	Yes
EC fraction RWC[a]	0.4866	0.0989	0.07
OC fraction RWC[a]	0.4068	0.8778	0.75[b]
Reference	Kuenen et al. (2022)	Denier Van Der Gon et al. (2015); Kuenen et al. (2022)	Guevara et al. (2020)

- Two versions of the European-scale Copernicus CAMS-REG
- Spanish bottom-up inventory HERMESv3_BU
- *Treatment of condensable fraction RWC in emissions*

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Results

Spatial distribution of Residential emissions

- PM2.5 **Residential emissions** in Spain and around Barcelona city (5km resolution)
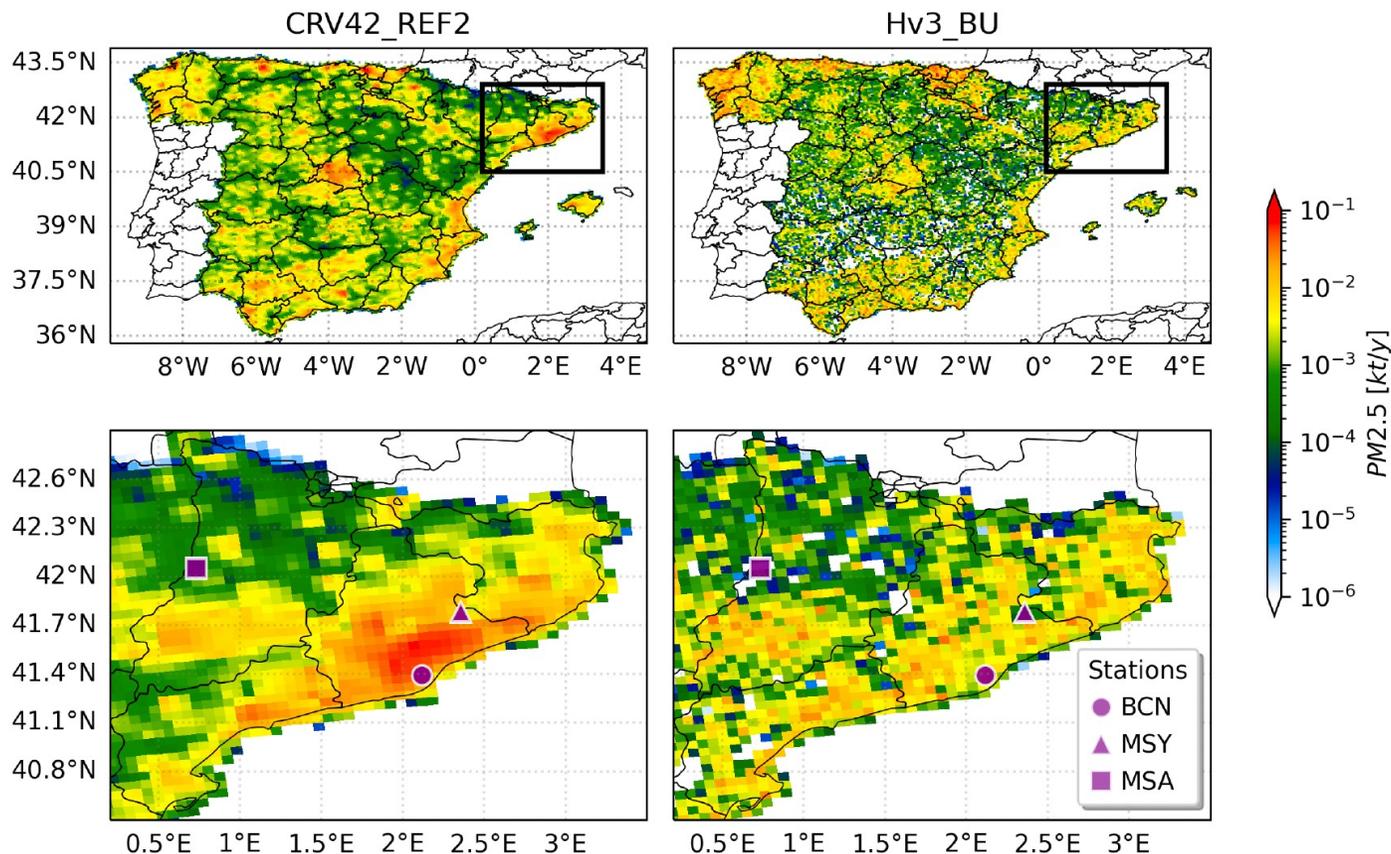
- Consistent total PM2.5 residential emissions in Spain (68-63 kton yr-1)

- Different spatial distribution proxy: population density + proximity to wood (CAMS-REG) vs rural population (HERMESv3)

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Temporal distribution and speciation (inclusion of condensables)

(c) Spain — EC [kt]

(d) Barcelona — EC [kt]

(e) Spain — OC [kt]

(f) Barcelona — OC [kt]
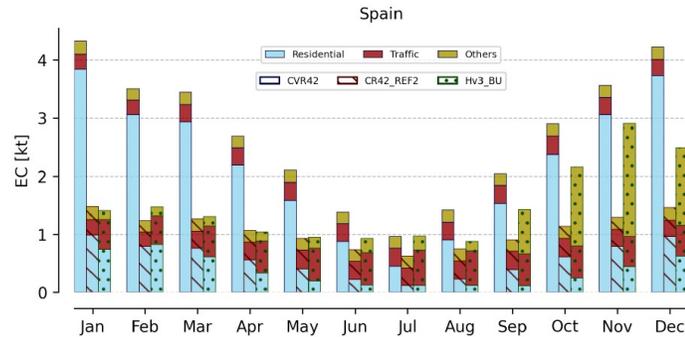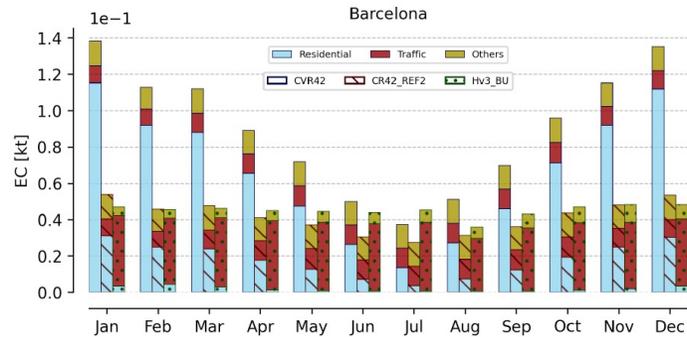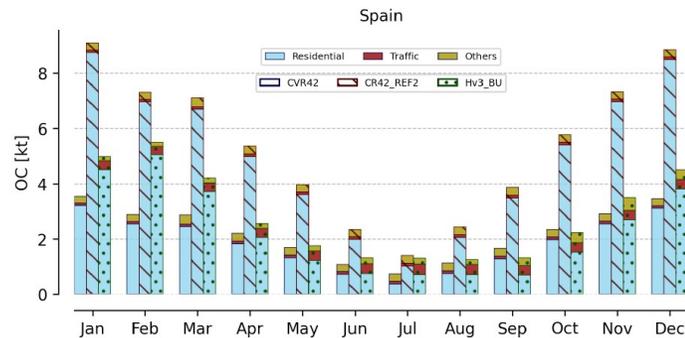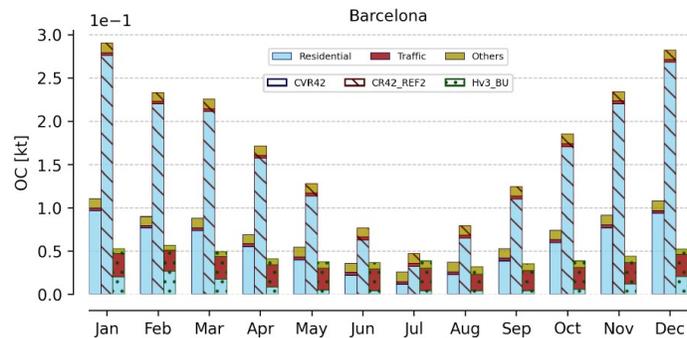
- Temporal distribution:
  - CAMS-REG V-shape dominated by RWC
  - HERMESv3 uses meteorology-dependent profiles (peak in February RWC)
- Major speciation change when considering condensables

	CRV42	CRV42_REF2	Hv3_BU
EC fraction RWC[a]	0.4866	0.0989	0.07
OC fraction RWC[a]	0.4068	0.8778	0.75[b]
Reference	Kuenen et al. (2022)	Denier Van Der Gon et al. (2015); Kuenen et al. (2022)	Guevara et al. (2020)

[a] RWC EC/OC speciation for Spain as reported by Kuenen et al. (2022)

[b] Emissions compute organic mass and assumes OM:OC of 1.8 for RWC and 1.4 for other sources, as suggested by Klimont et al. (2017)

Impact of emission uncertainties on BC concentrations

- Investigating the BC variability in Barcelona city (Palau Reial monitoring site)
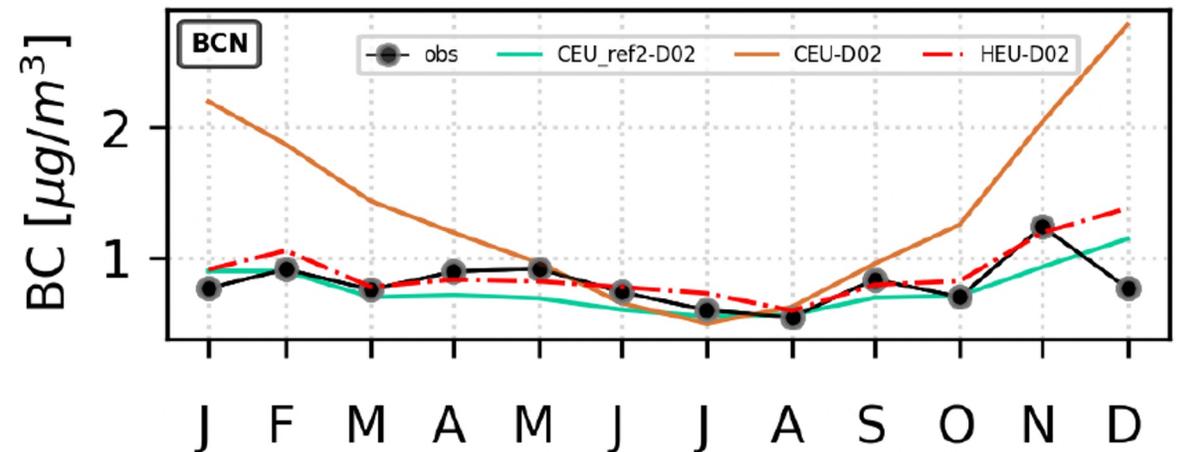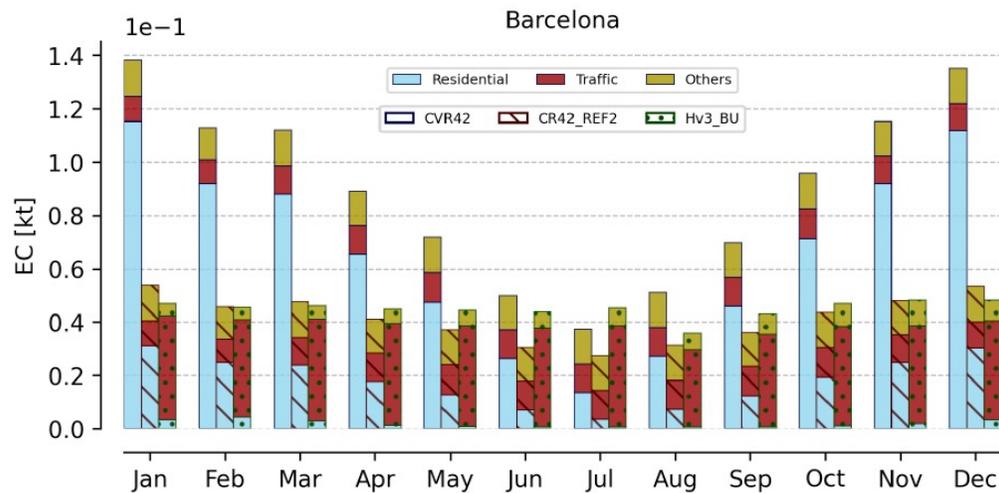
CAMS-REGv4/CAMS-REGv4_Ref2/HERMESv3_BU
emissions over Barcelona urban region

MONARCH (5km) BC concentrations at BCN site
Obs. (MAC 9.6m²/g)

Large differences in source contribution in emissions, but rather consistent total BC concentrations (CAMS-REGv4.2_REF2 and HERMESv3)

Source contribution based on observations and model: *January*

- Tagged sectors: traffic, shipping, residential, biomass, others
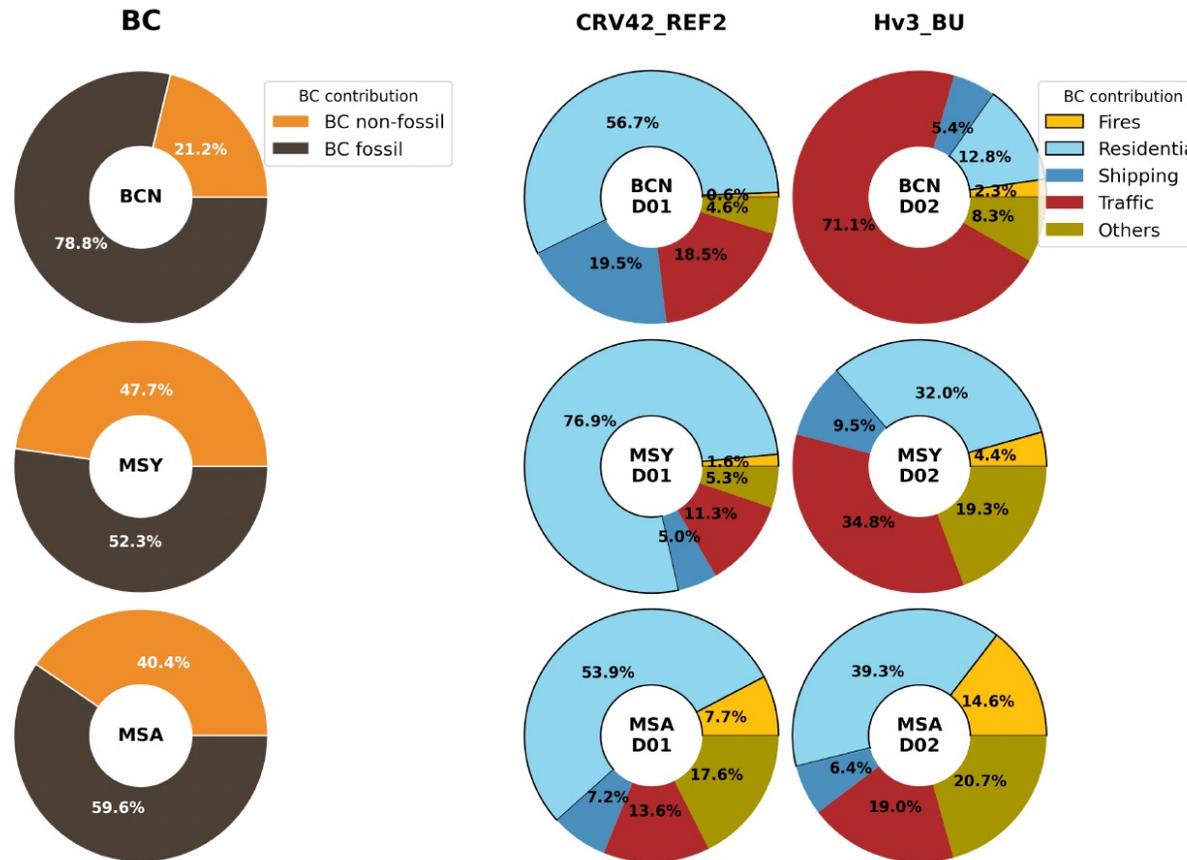
BC

CRV42_REF2   **Hv3_BU**

WP1 obs. Aethalometer model
(Sandradewi et al., 2008)

MONARCH - perturbation method
D01(20km), D02(5km)

CAMS-REG emissions:
- Mismatch between fossil and non-fossil
- More weight in the **residential**/shipping compared with traffic in **January**/July

HERMESv3 bottom-up:
- Traffic is the dominant sector
- Good match with source contribution derived from observations

Very good agreement in remote background (MSA)

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Source contribution based on observations and model: *July*

- Tagged sectors: traffic, shipping, residential, biomass, others
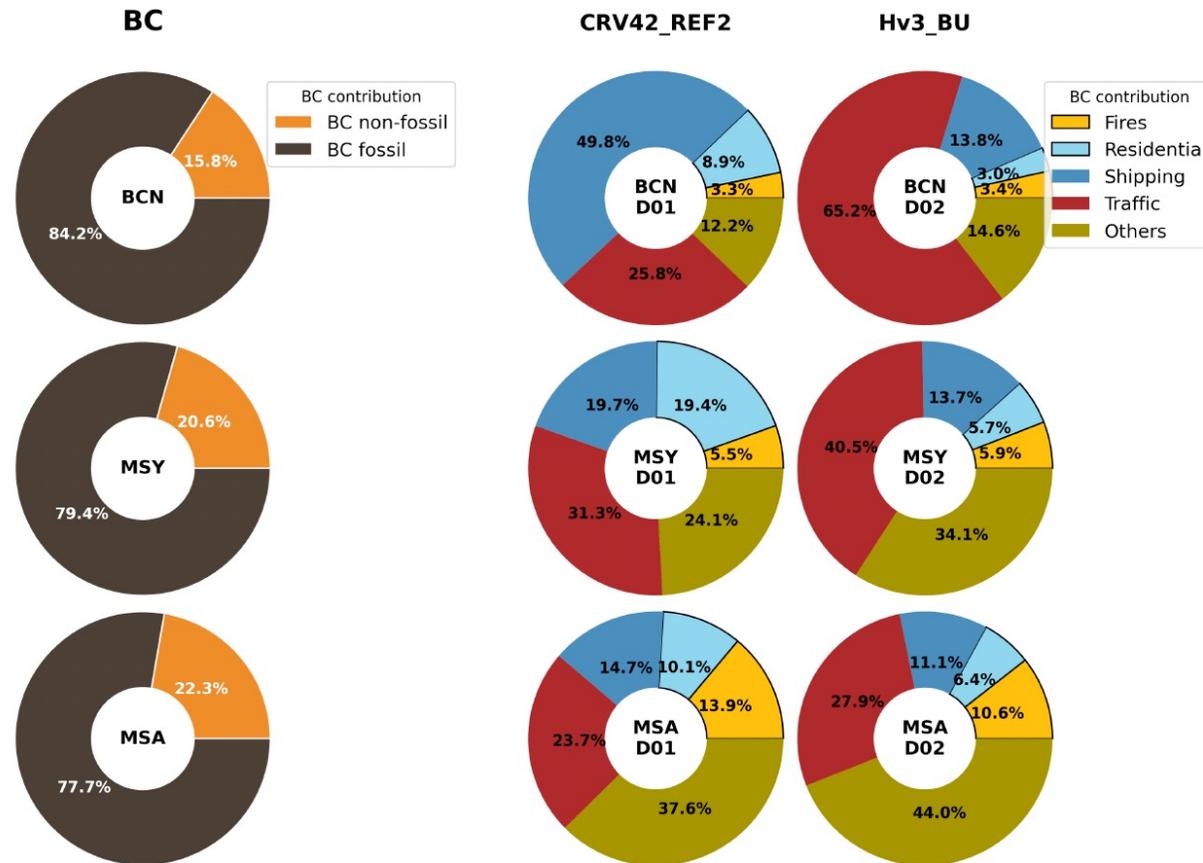
WP1 obs. Aethalometer model
(Sandradewi et al., 2008)

MONARCH - perturbation method
D01(20km), D02(5km)

CAMS-REG emissions:
- Mismatch between fossil and non-fossil
- More weight in the residential/**shipping** compared with traffic in January/**July**

HERMESv3 bottom-up:
- Traffic is the dominant sector
- Good match with source contribution derived from observations

Very good agreement in remote background (MSA)

Modeling source contribution of Black carbon

January
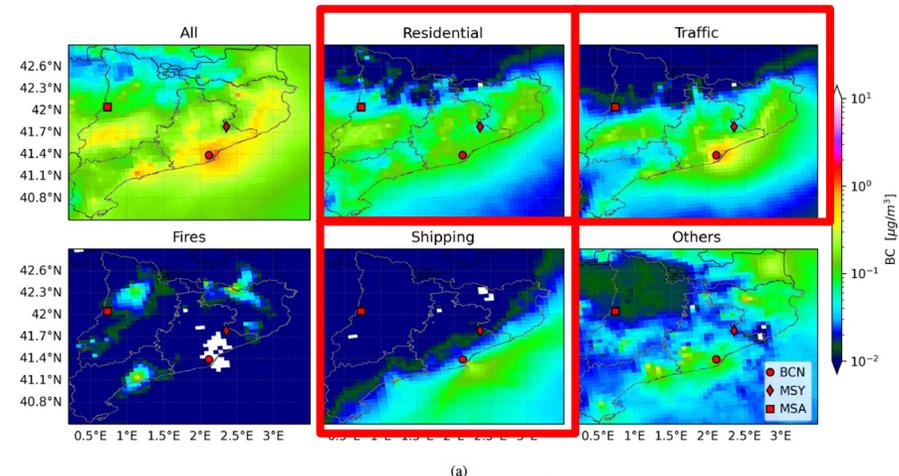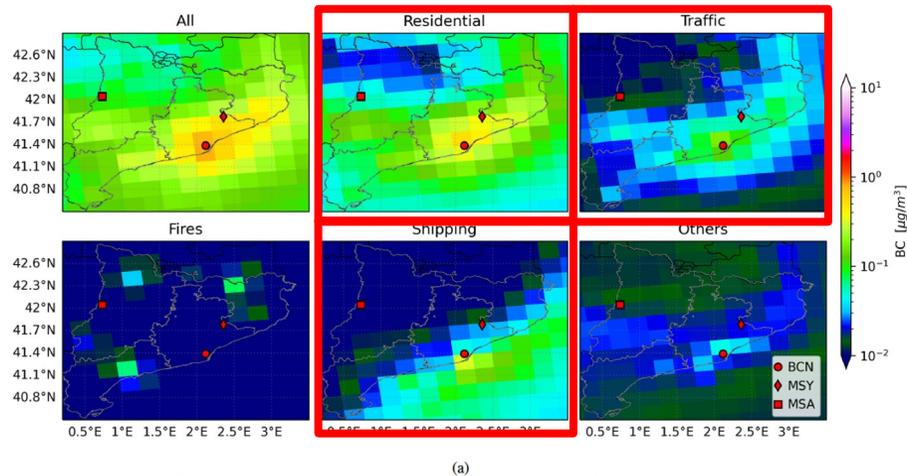
July
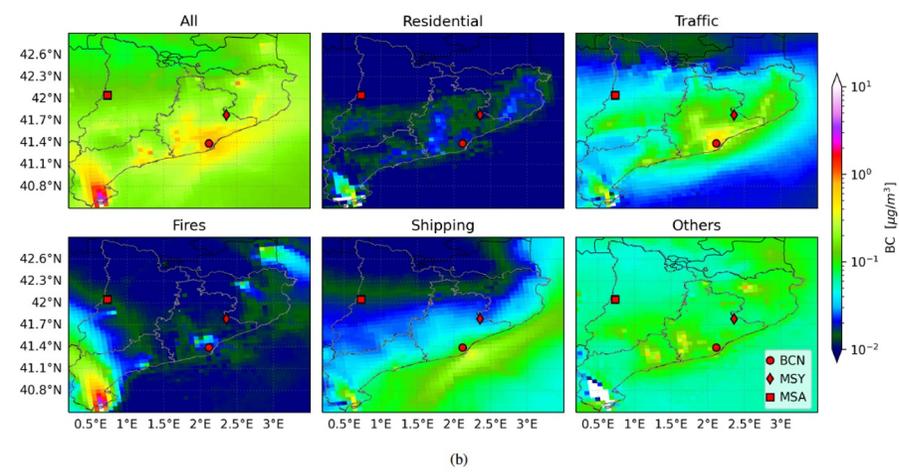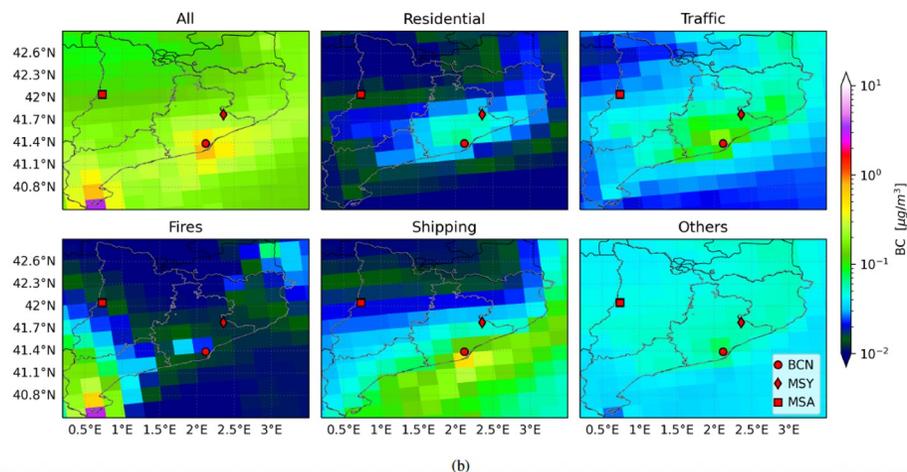
MONARCH - CAMS-REGv4.2_Ref2 emissions

MONARCH - HERMESv3 emissions

Very different representation of Traffic/Residential/Shipping contributions

Conclusions

Conclusions

- We investigated the uncertainties in CAs simulation and source allocation (from traffic, RWC, shipping, fires and others) in Northeast Spain.

- Three emission inventories (CAMS-REGv4 official and science version and HERMESv3 bottom-up) were intercompared with similar total particulate matter emissions

- Large discrepancies found between them mainly related to the sector representation, the spatio-temporal distribution of emissions and the speciation profiles of primary elemental carbon/organic carbon (EC/OC) fractions, particularly relevant for RWC sources.

- Observation-based source contribution helps to disentangle some model biases.

- Main source contribution to BC concentrations in the urban site BCN is traffic, accounting for 71.1%/65.2% (January/July) in close agreement with the fossil contribution derived from observations (78.8%/84.2%), followed by RWC (12.8%/3%) and shipping emissions (5.4%/13.8%).

- CAMS-REG_v4 over-represents BC RWC/Shipping contribution in the urban site in January/July.

- Very good agreement in the remote background site MSA between CAMS-REGv4_Ref2 and HERMESv3

Thank you!

oriol.jorba@bsc.es