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EXCELENCIA SEVERO OCHOA

A source apportionment assessment of O_3 in peak summer events over southwestern Europe

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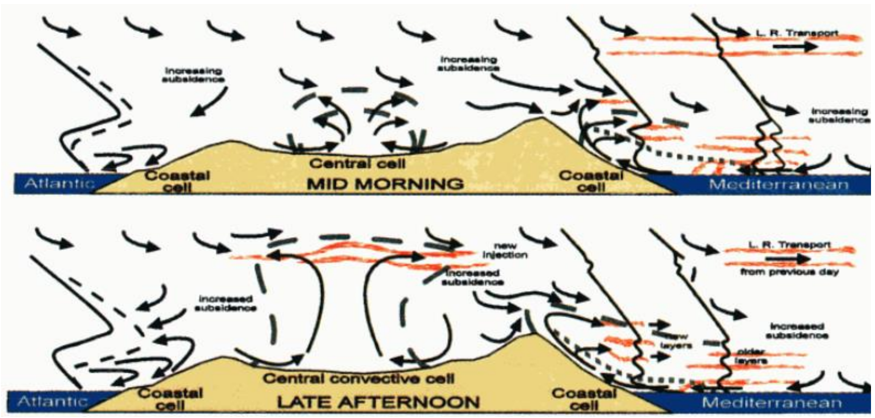
11th Air Quality Conference - Barcelona (Spain)

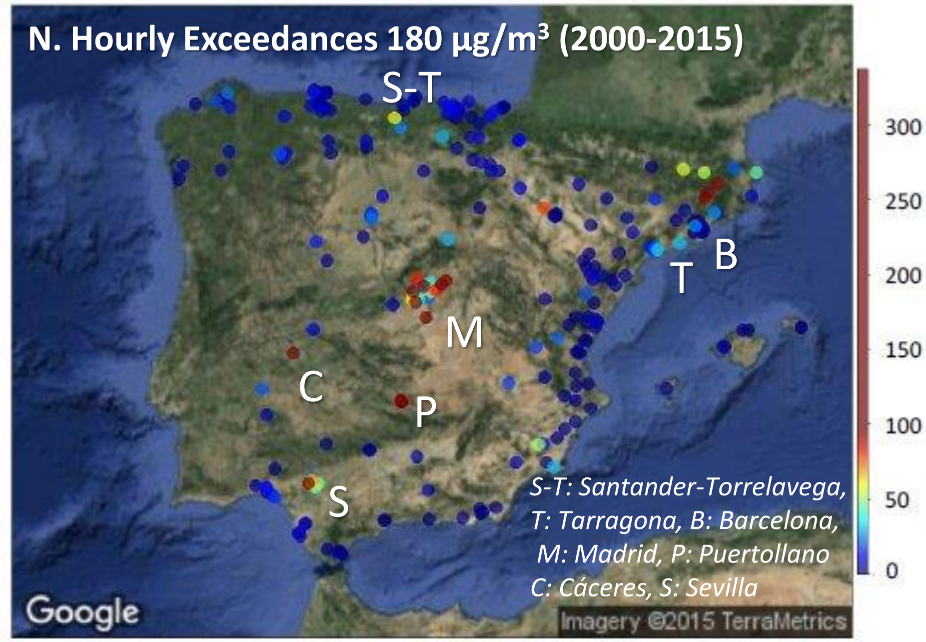
Background and Motivation

O_3 dynamic



Sources: Millán et al., 1997, 2000, 2014; Gangoiti et al, 2001, 2002, 2006; Toll and Baldasano, 2000

O_3 Trends and exceedances



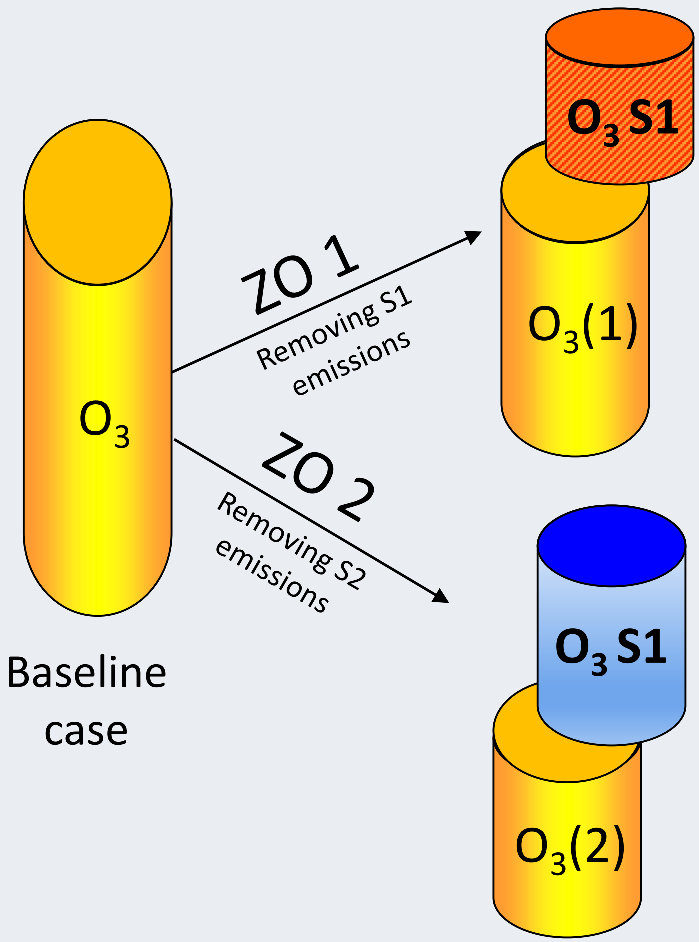
Source: Querol et al. (2016).

Open questions:

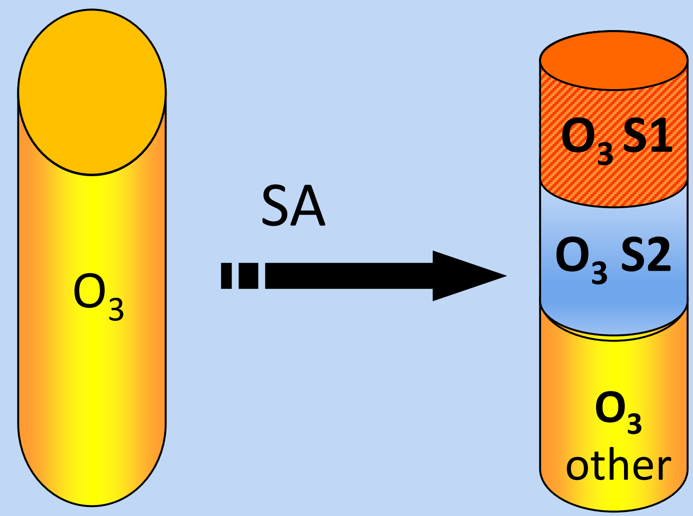
- What are the sources responsible for the high O_3 concentration in Spain?
- Can administrations implement control strategies that are effective to reduce high O_3 concentration?

Source Apportionment (SA) methods in Air Quality Models (AQM)

Zero-Out (ZO)



SA within the AQM



Advantages

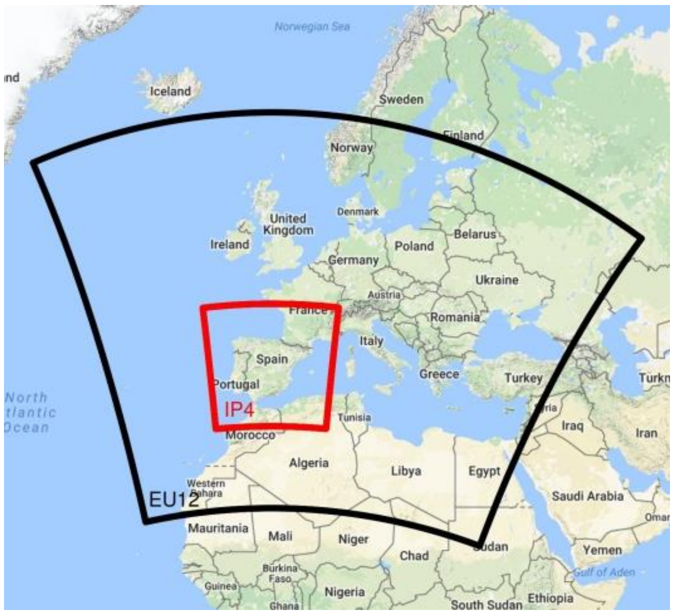
- Time saving (one simulation)
- Mass consistency
- Real atmospheric conditions
- Fully traceable

Objective

Unraveling the origin of the high surface O_3 concentrations in the Spanish Iberian Peninsula (IP)

- Quantifying the contribution from:
  - the **main NO_x emission sectors within the IP**
  - the **external contribution** (O_3 produced outside of the IP)
- Using the Integrated Source Apportionment Method (ISAM) in the CALIOPE air quality modelling System at high resolution over the IP.

The CALIOPE System



Set-up for the Source Apportionment

METEO

- **WRF-ARWv3.5** (RRTM/WSM3/YSU/NoahLSM)
- Ver. Res.: 37σ / 50hPa (top)
- Hor. **Res: 12 km (EU12) 4 km (IP4)**
- **IC/BC (EU12/IP4): GFS (NCEP)** / nesting EU12

EMIS

- **HERMESv2.0**
- EU12: HERMES-DIS (EMEP data 2009)
- **IP4: HERMES-BOUP (Spain) + HERMES-DIS(Europe)**
- Biogenic emission MEGANv2.0.4

CHEM

- **CMAQv5.0.2** (**ISAM**, CB05TUCL, AERO6)
- Ver. Res: 37σ / 50hPa (top)
- Hor. Res: 12 km (EU) 4 km (IP)
- BC (EU12/IP4): MOZART4-GEOS-5/nesting EU12
- MCIPv4.0

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O_3 Integrated Source Apportionment Method (ISAM)

- Augmented version of **CMAQv5.0.2** (AERO6, CB05TUCL) (*Kwok et al., 2013; 2014*)
- **Mass conservative**: the bulk O_3 concentration in each model grid cell (P_{bulk}) is equal to the sum of O_3 tracers that were produced in either NO_x or VOC-limited conditions.

$$P_{bulk} = \sum_{tag} P_{tag} = \sum_{tag} P_{tag}^{N} + \sum_{tag} P_{tag}^{V}$$

- **O_3 formation regime** (NOx- or VOC-limited conditions): ratio H_2O_2/HNO_3 (*Zhang et al., 2009*).

$$P_{tag}^{N,new} = P_{tag}^{N,old} + P_{bulk}^{new} \frac{\sum_x NO_{x,tag}}{\sum_{tag} \sum_x NO_{x,tag}}$$

NO_x-limited conditions - $Ratio_{H2O2/HNO3} > 0.35$

$$P_{tag}^{V,new} = P_{tag}^{V,old} + P_{bulk}^{new} \frac{\sum_y VOC_{y,tag} \times MIR_y}{\sum_{tag} \sum_y VOC_{y,tag} \times MIR_y}$$

VOC-limited conditions - $Ratio_{H2O2/HNO3} < 0.35$

NOx,tag: concentrations of the nitrogen species in CB05 (9 species)
VOCj,tag: concentrations of the VOC species in CB05 (14 species)
MIRy: Maximum Incremental reactivity factor of the VOC species y, corresponding to the O_3 generating potential of each single VOC specie.

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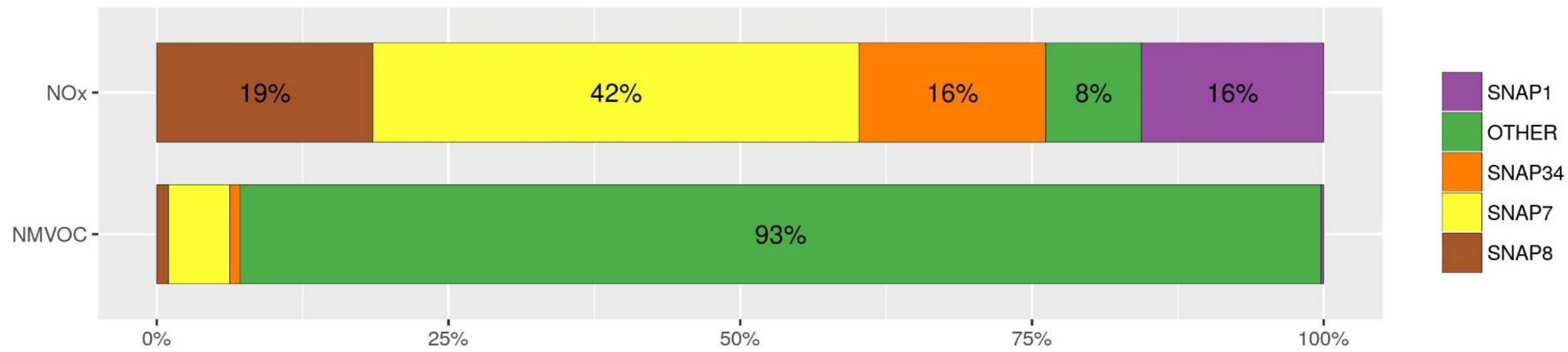
O_3 tagged sources in this study

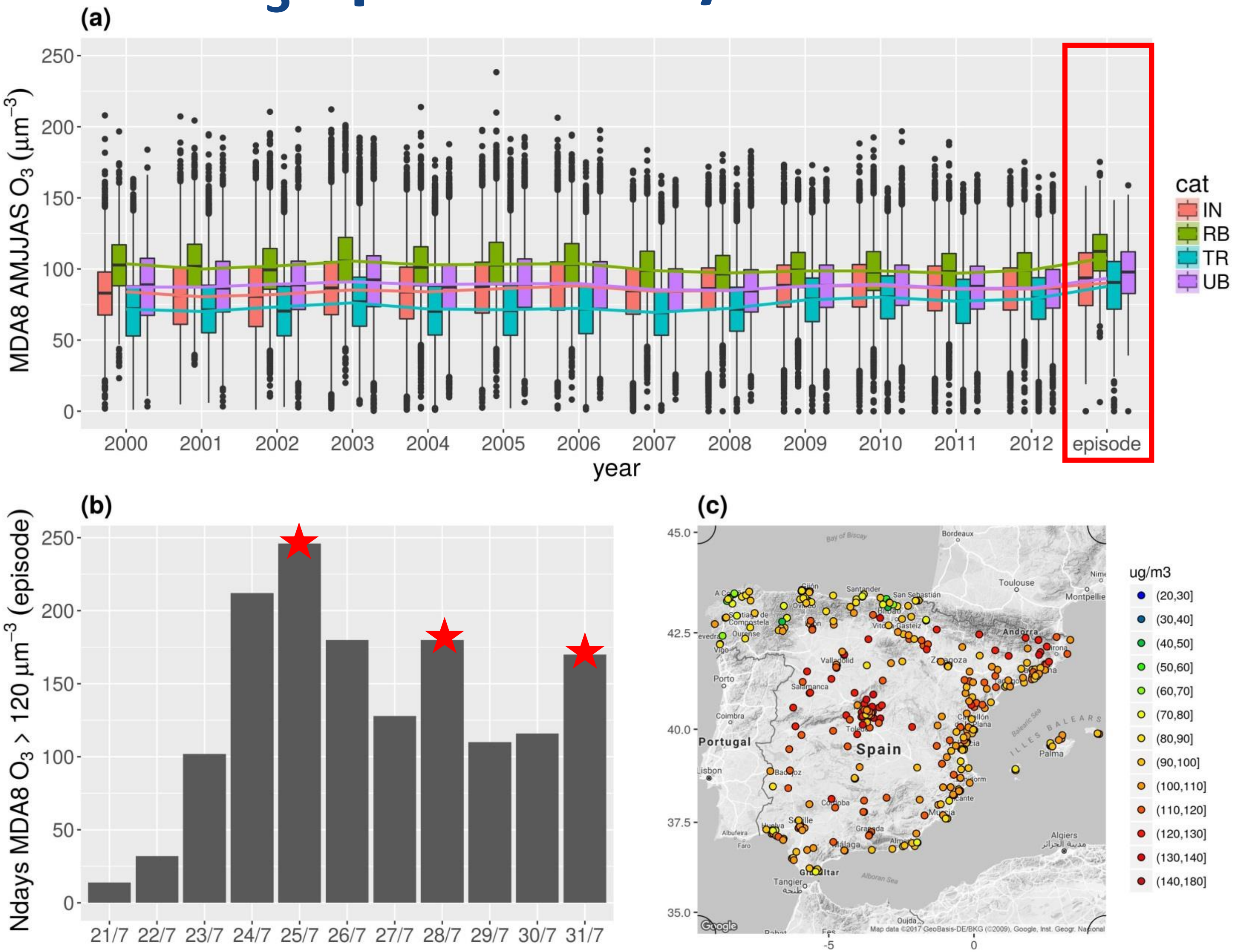
Annual emissions HERMESv2.0 in Spain 2009



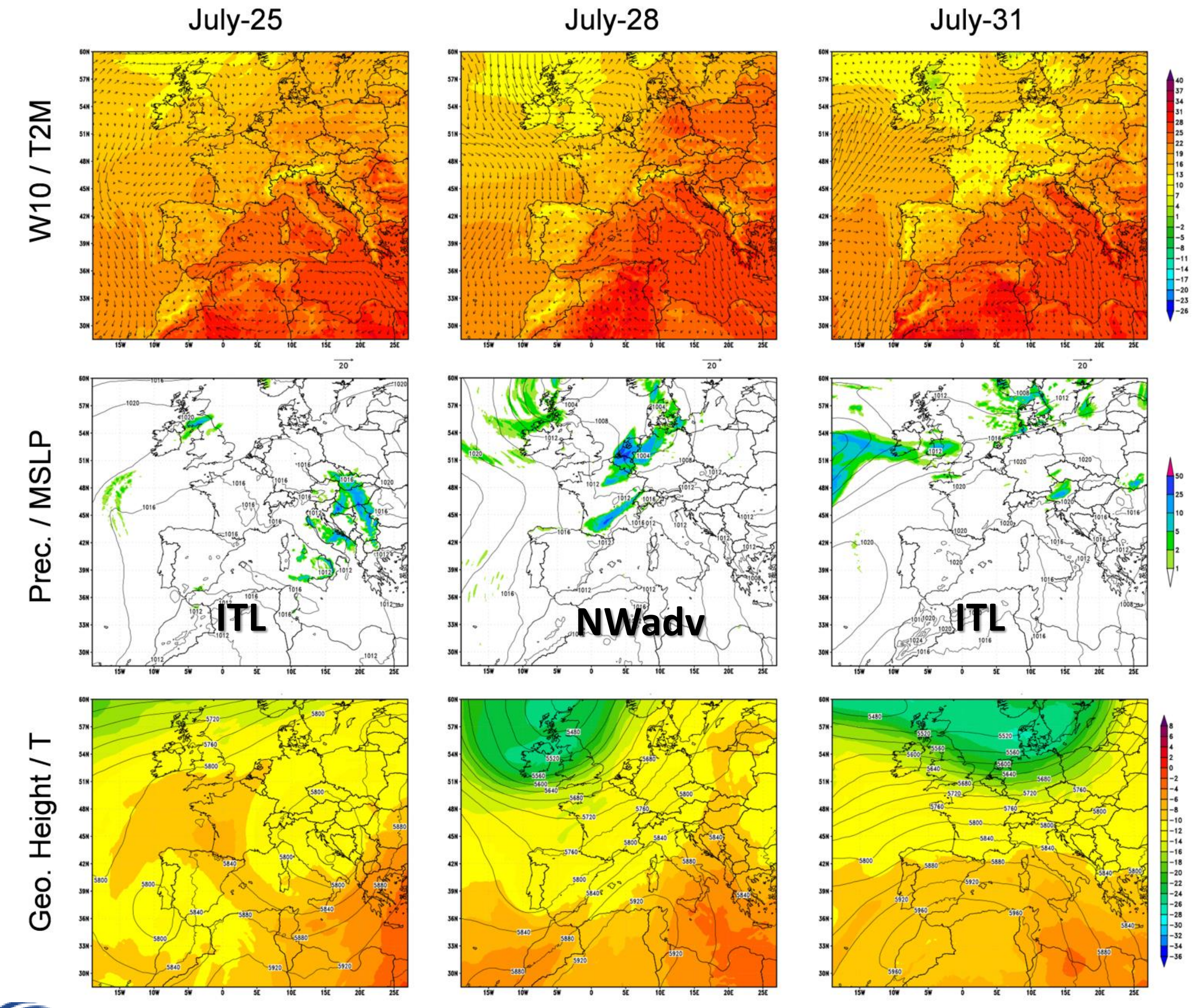
ISAM tag*	Emission by SNAP category	Description
SNAP1	SNAP1	SNAP1: Energy industry
SNAP34	SNAP34	SNAP34: Industry (combustion and processes)
SNAP7	SNAP7	SNAP7: Road transport, exhaust and non-exhaust
SNAP8	SNAP8	SNAP8: Non-road transport (international shipping)
OTHR	SNAP2 + SNAP5 + SNAP6 + SNAP9 + SNAP10 + SNAP11	SNAP2: residential and commercial/institutional combustion SNAP5: Fugitive emissions from fuels SNAP6: Product use including solvents SNAP9: waste management SNAP10: Agriculture SNAP11: Other sinks
BCON	-	Chemical boundary conditions to IP4 domain from the EU12 simulation which includes the contribution from Europe and international contribution from MOZART-4
ICON	-	Initial chemical condition of the domain IP4

<1 %

O_3 episode: July 21-31 2012

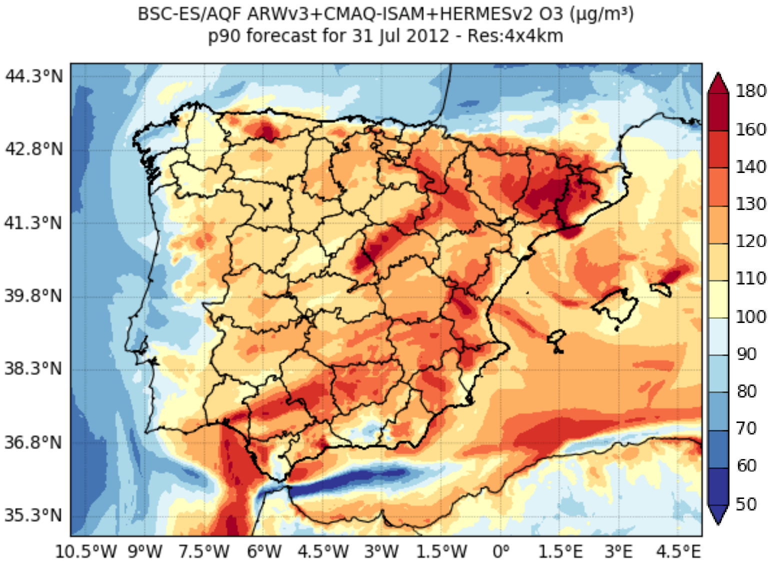


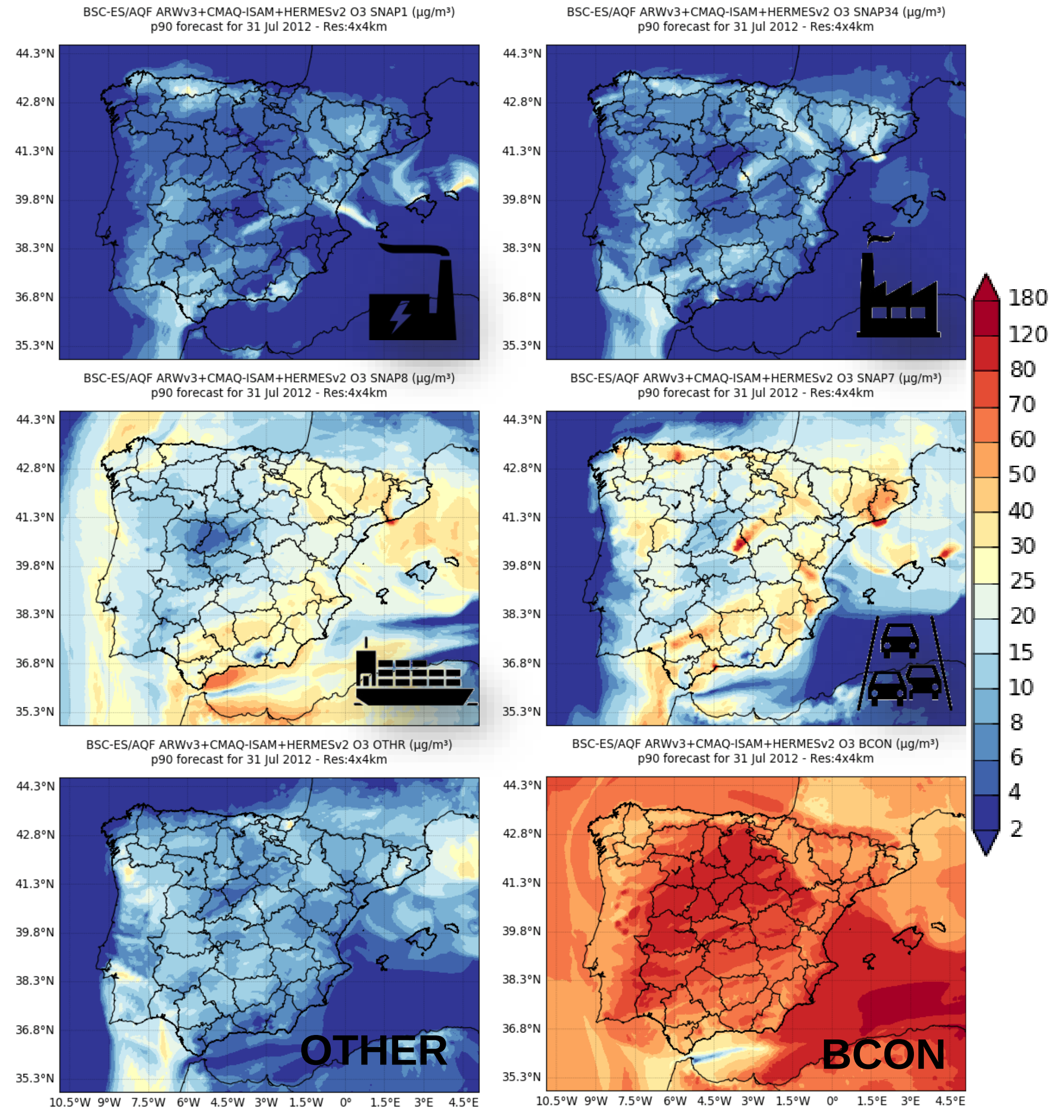
Meteorological context (6 UTC)



ITL and **NWadv** represent 44% of the days in the IP both taking place in summer (*Valverde et al., 2015*)

Source-sector contribution during peaks
ITL – July





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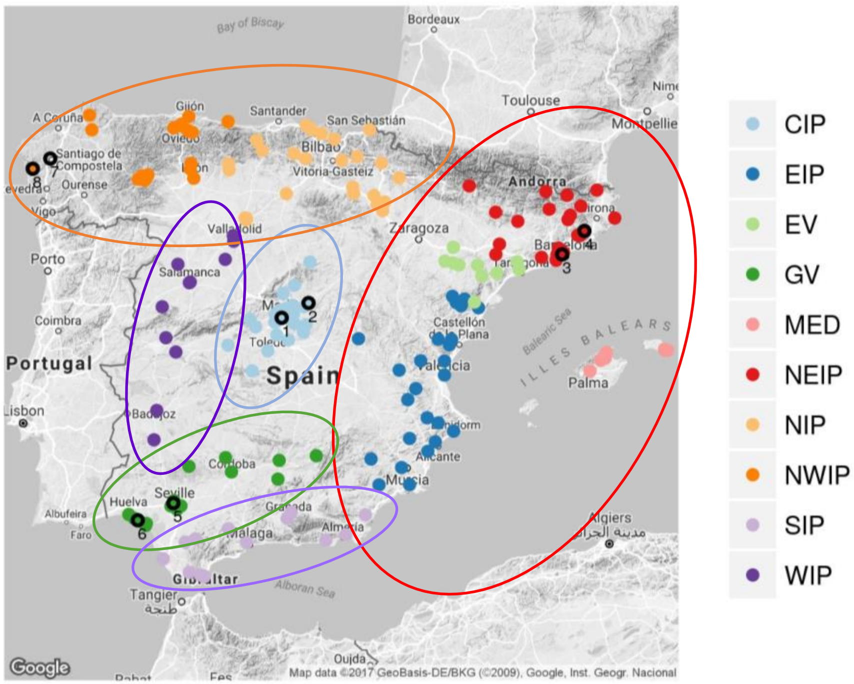
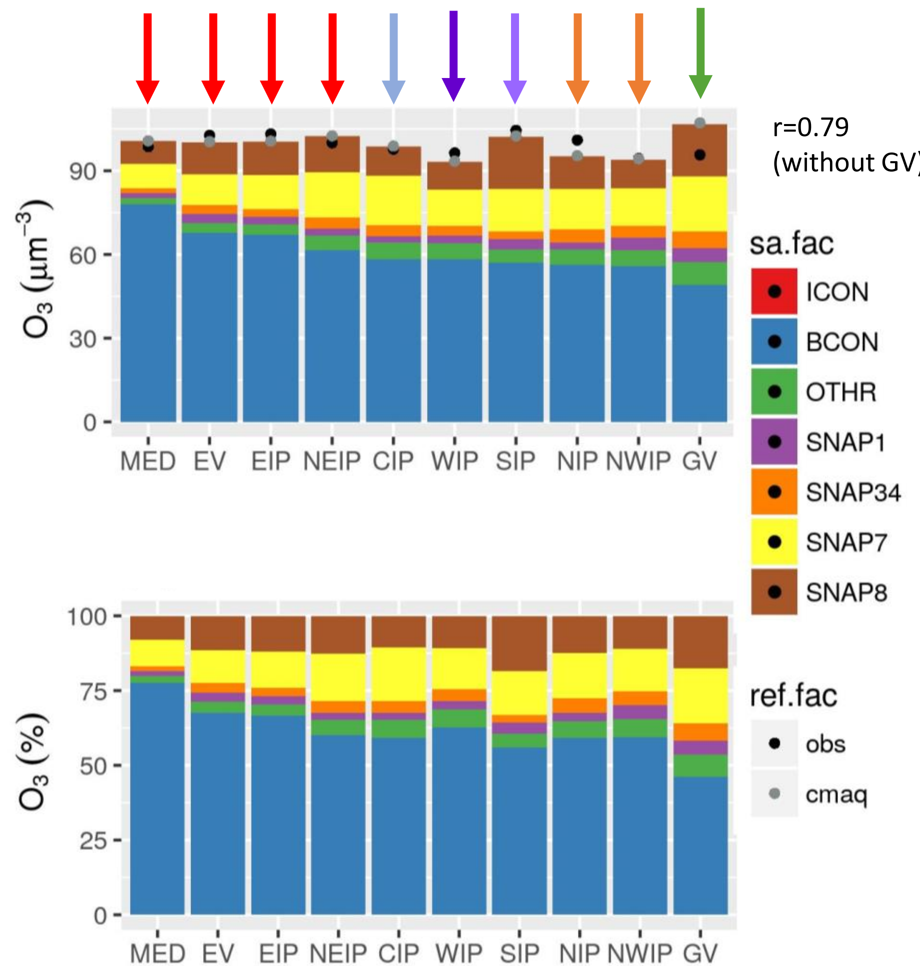
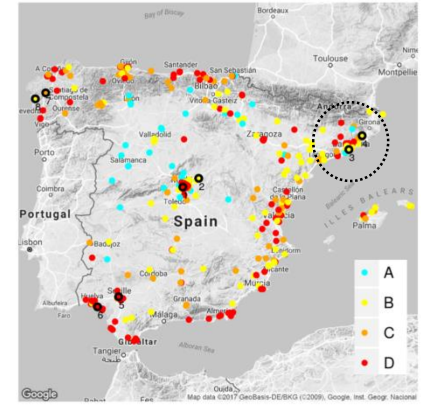
Regionalization of source-sector contribution



Daily mean contribution during DMA8 > 120 µ/m³

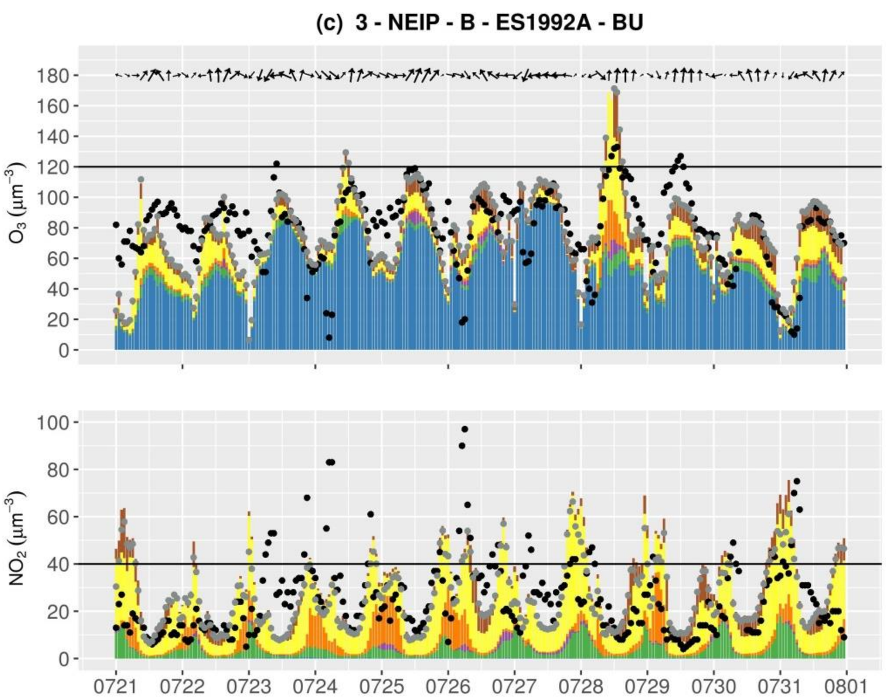


Northeastern Iberian Peninsula (NEIP)

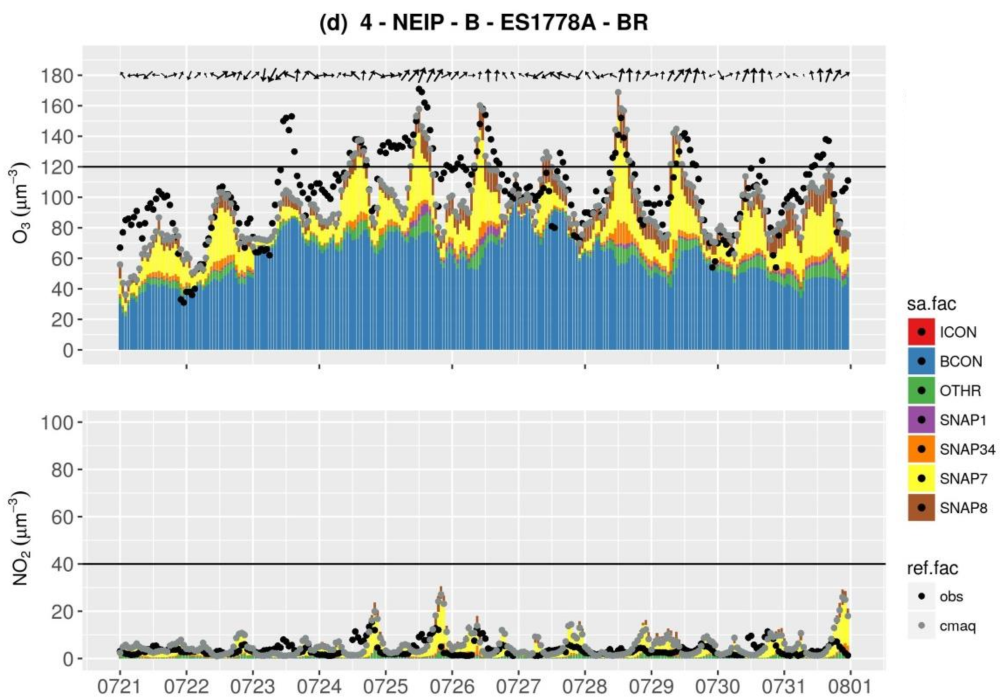


→ On-road traffic influence

Urban station



Rural station



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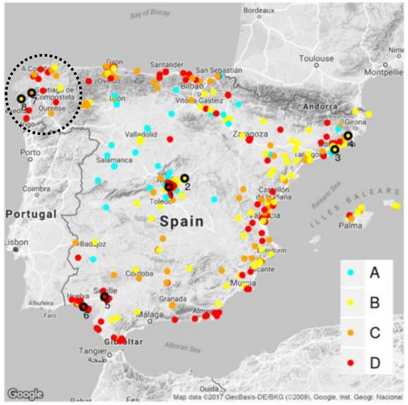
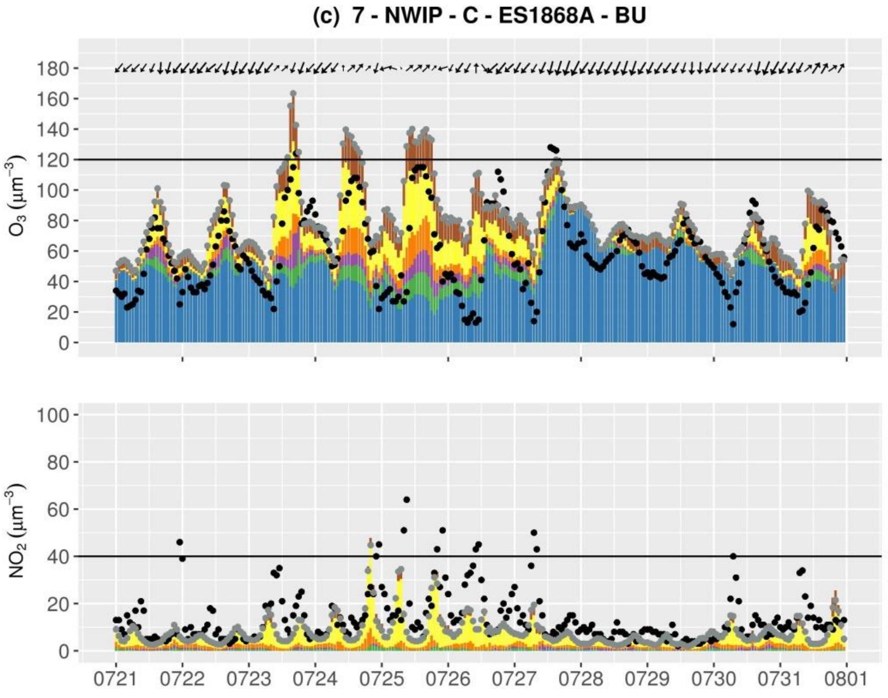
North Western Iberian Peninsula (NWIP)

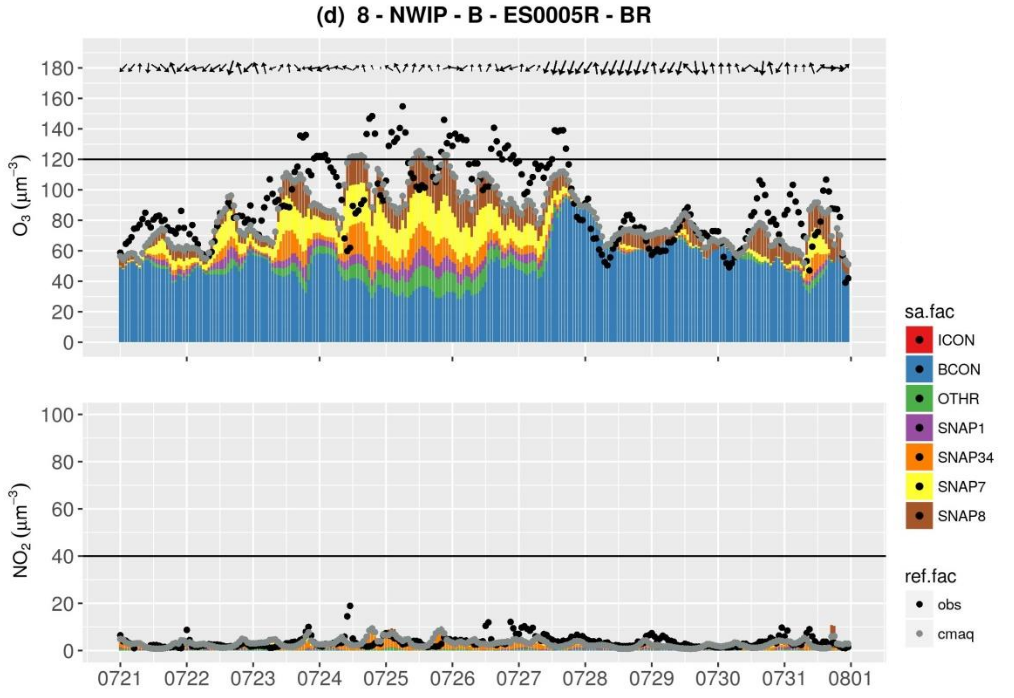


→ Industrial influence

Urban station

Rural station





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Conclusions

- Estimation of the **origin of surface O_3 over main receptors regions in Spain**.
- The O_3 problem is local, regional and hemispheric:
  - The **external contribution** accounted for 46% to 68% of the daily mean O_3 under exceedances of the 120 µg m^{-3} threshold for the DMA8 O_3, depending on the region.
  - **Contribution from local/regional sources** is significant in O_3 peaks downwind of NO_x hotspots.
    - Central and NE IP (big cities in Spain): the highest **road transport** contribution to O_3 (up to 40% in daily peak during events).
    - Industrial regions (N and NW IP and Guadalquivir Valley): **energy generation and industrial processes** contribute to O_3 up to 11%.
- The **non-road transport** is a contributor as significant as the road transport in all sub-regions (10-19%).
- **ISAM-CALIOPE** useful tool:
  - Identification of potential errors in emission estimates
  - Design more cost-efficient mitigation plans (together with source sensitivity).
- **Future work**: expand the study to a longer period and different ozone episodes.

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THANK YOU!

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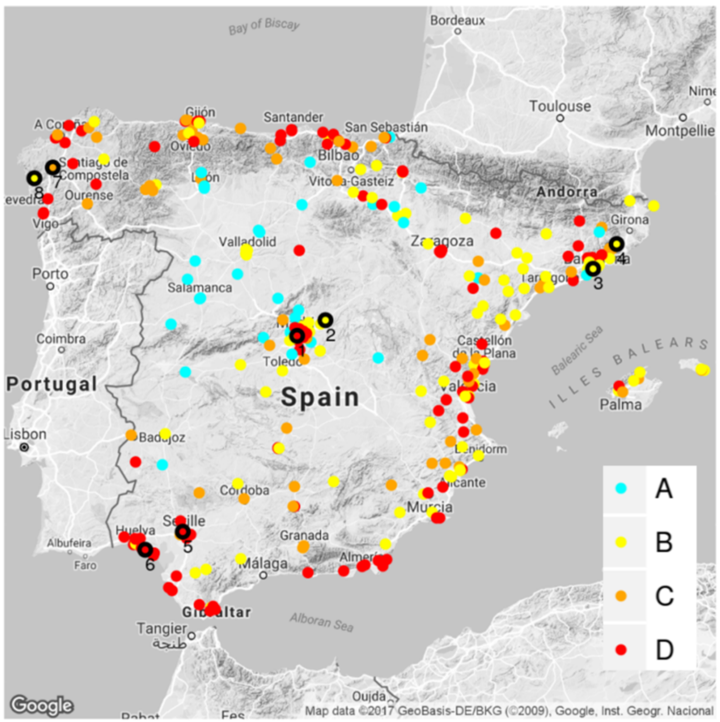
Evaluation

EIONET Spanish monitoring stations: 347 O_3 and 357 NO_2 (85% temporal coverage in the episode)

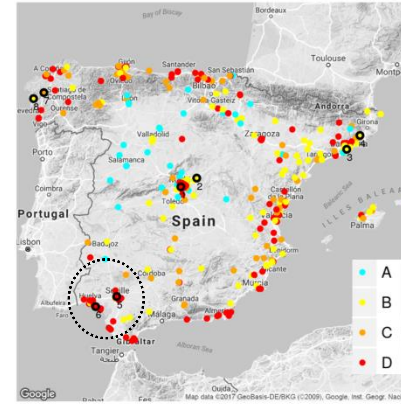
- O_3 at RB (±4 µg/m^3) and r > 0.6 (50% stations).
- NO_2 highest underestimation at TR (-7 µg/m^3) and r >0.6 (25 % stations.)

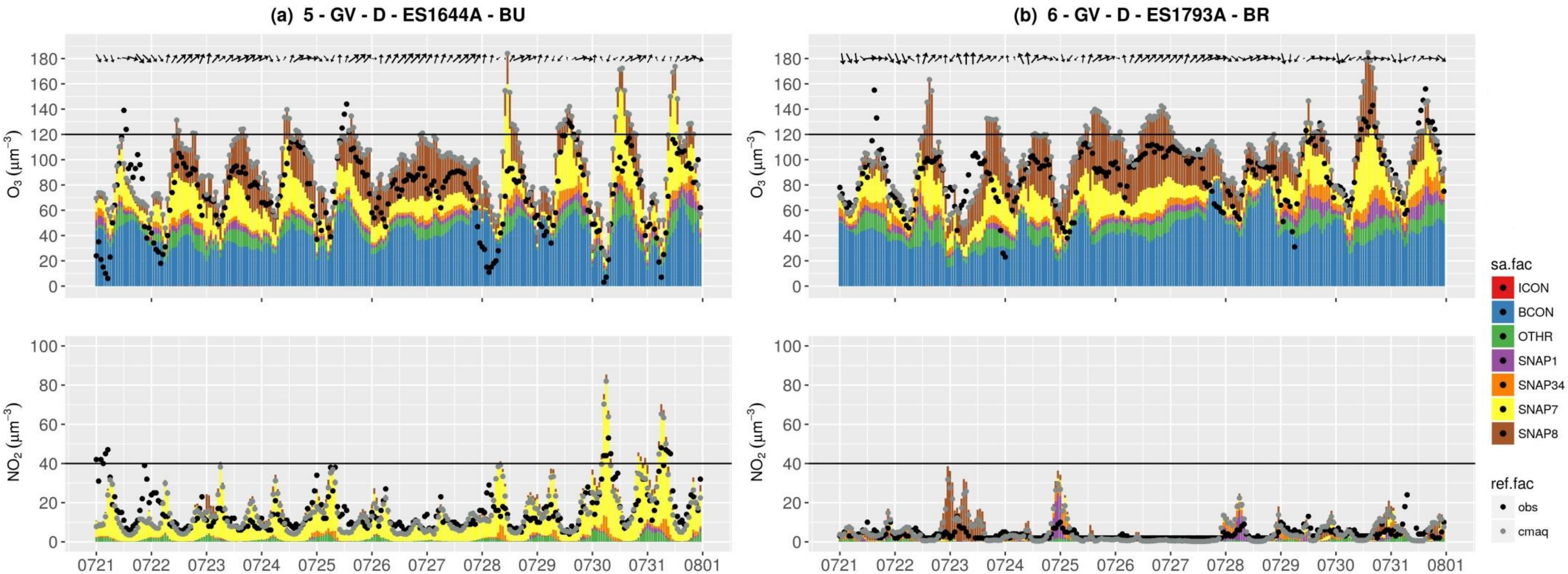


MB (µg/m³)		O₃ DMA8				
		<-40	(-40,-10]	(-10,10]	(10,40]	> 40
O_3 HL	<-40	0	0	0	0	0
	(-40,-10]	0	7 (2%)	0	0	0
	(-10,10]	0	A 35 (10%)	B 94 (28%)	5 (2%)	0
	(10,40]	0	3 (1%)	C 65 (19%)	D 122 (36%)	5 (2%)
	>40	0	0	0	1 (<1%)	5 (2%)

1 - CIP - D
2 - CIP - B
3 - NEIP - B
4 - NEIP - B
5 - GV - D
6 - GV - D
7 - NWIP - D
8 - NWIP - B

Gualdaquivir Valley (GV)





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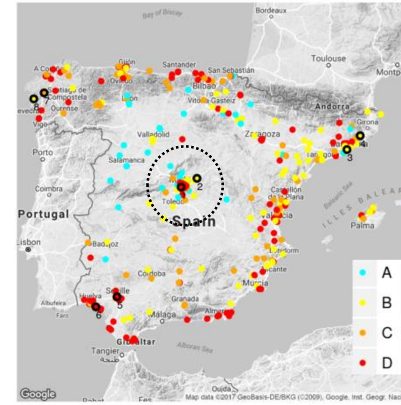
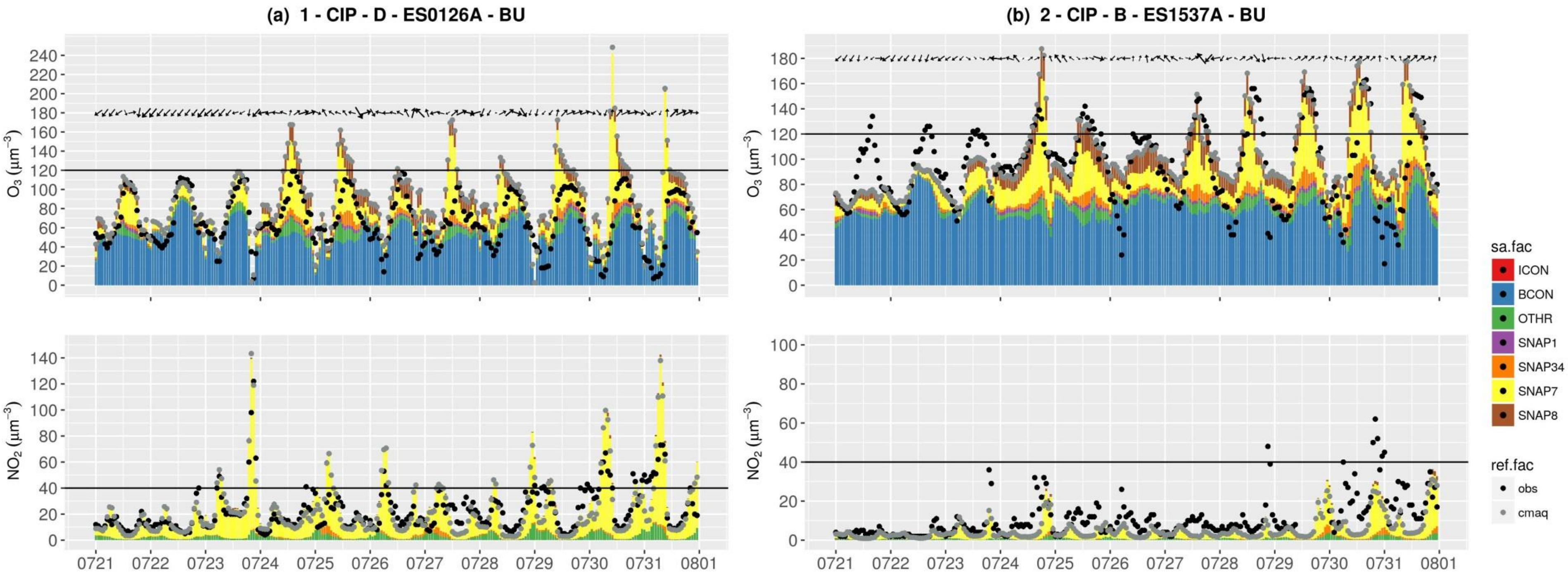
Central Iberian Peninsula (CIP)





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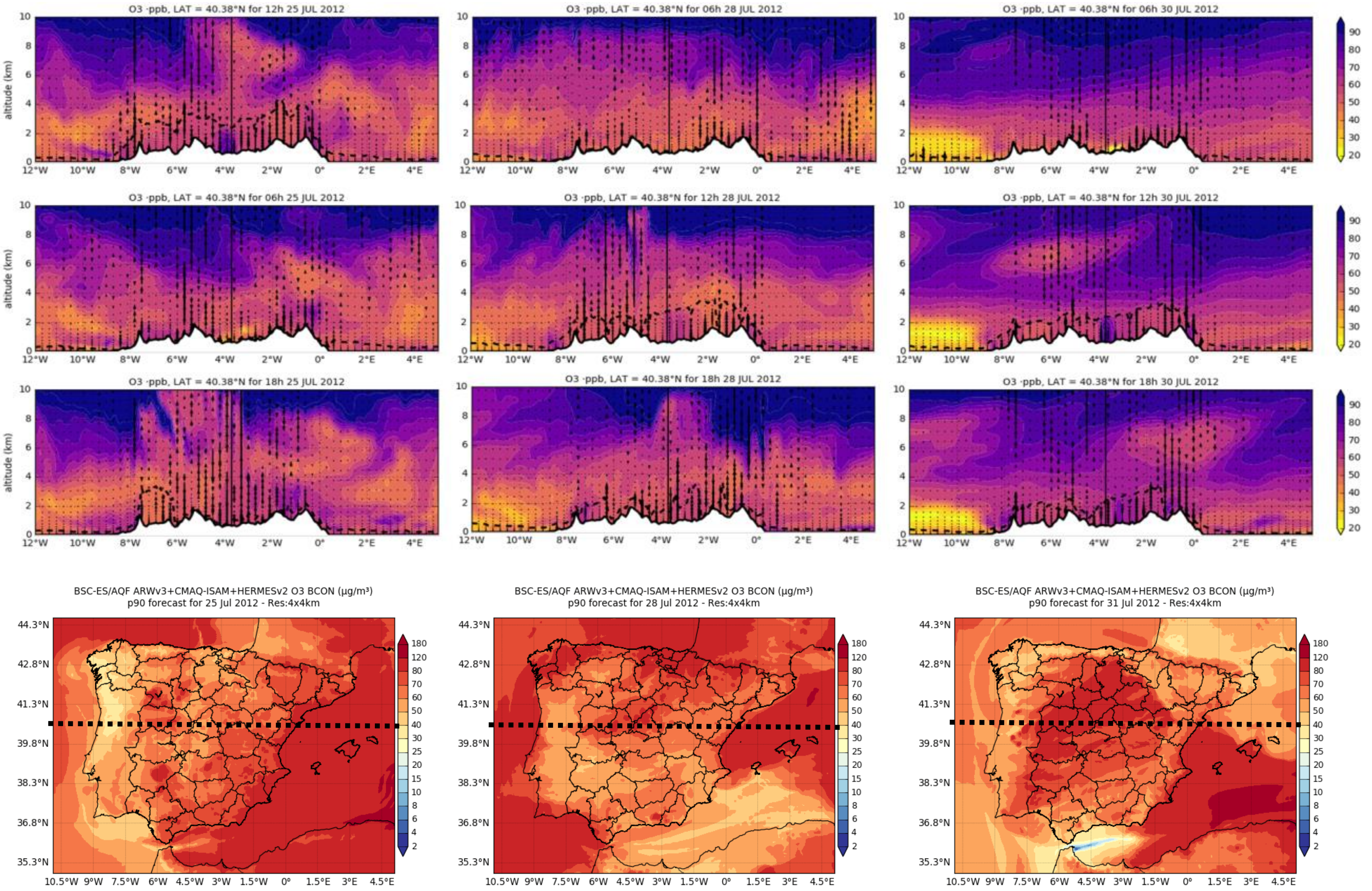
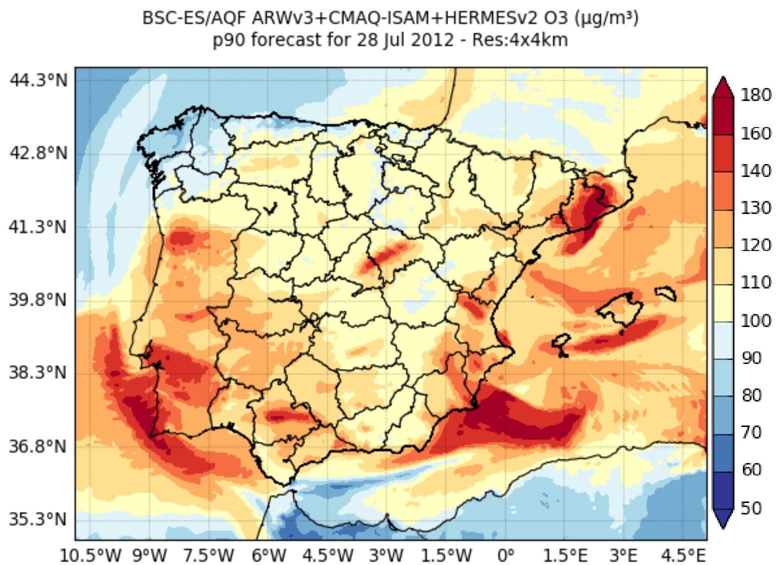
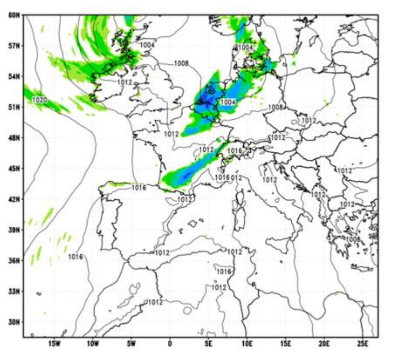
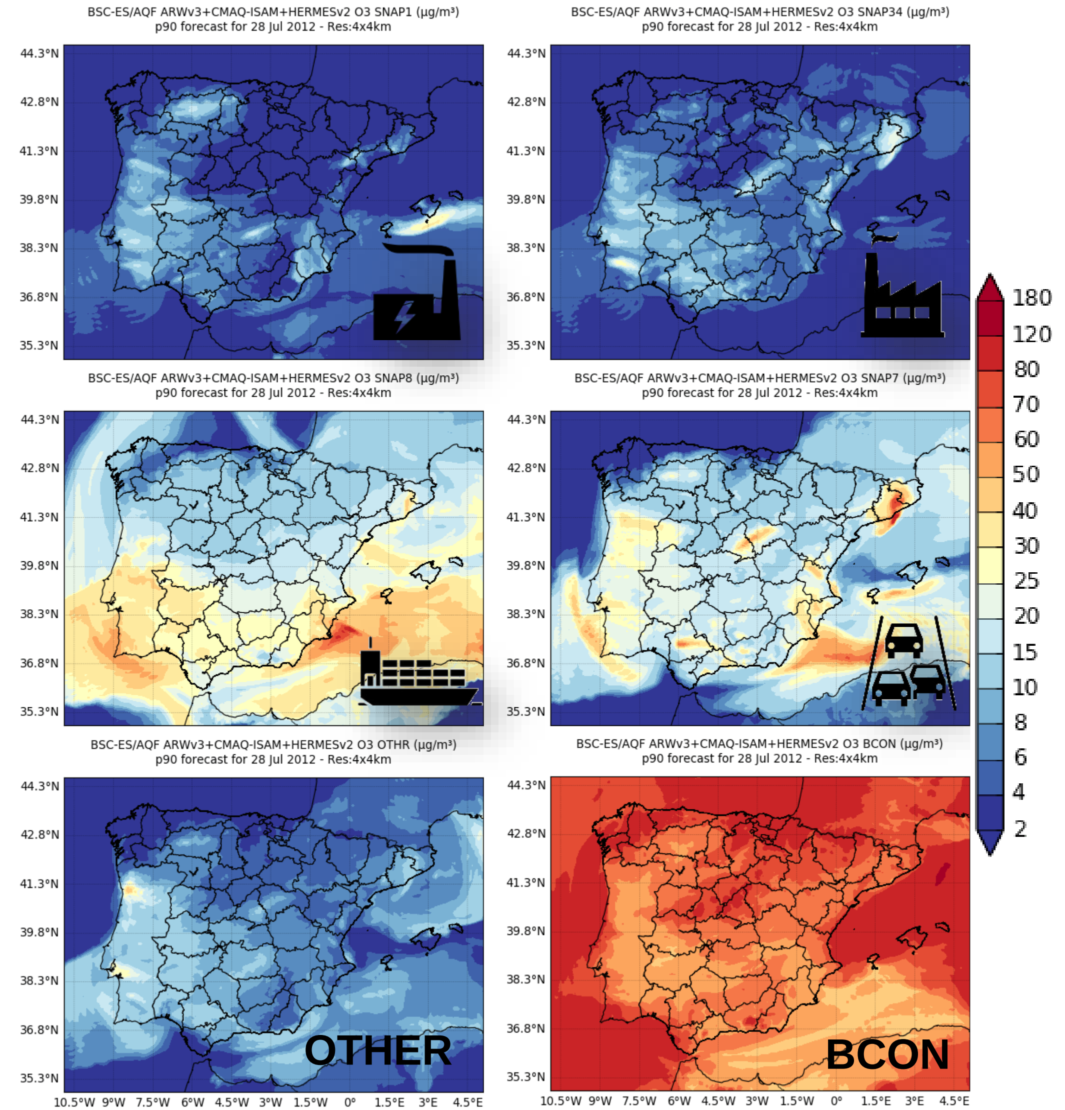
O3 cross sections



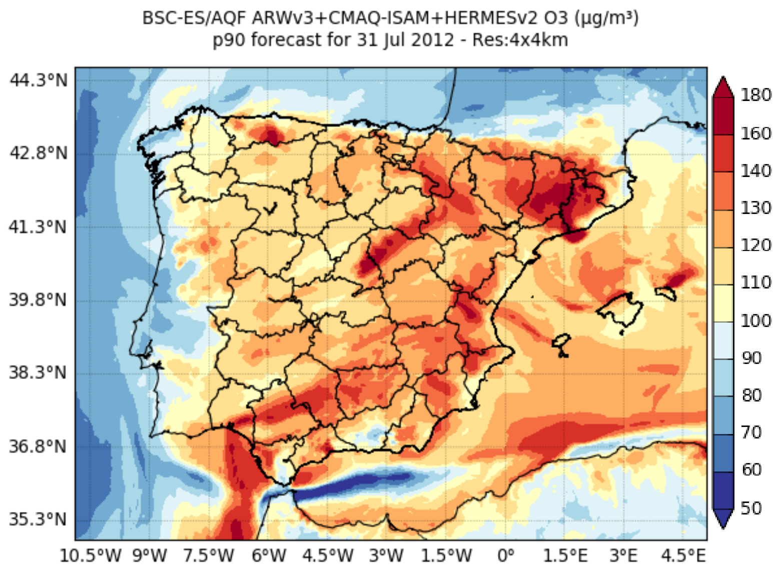
Source-sector contribution during peaks: Nwad – July 28

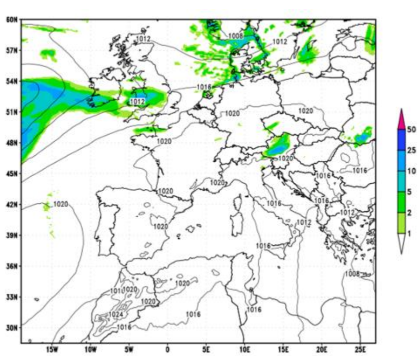


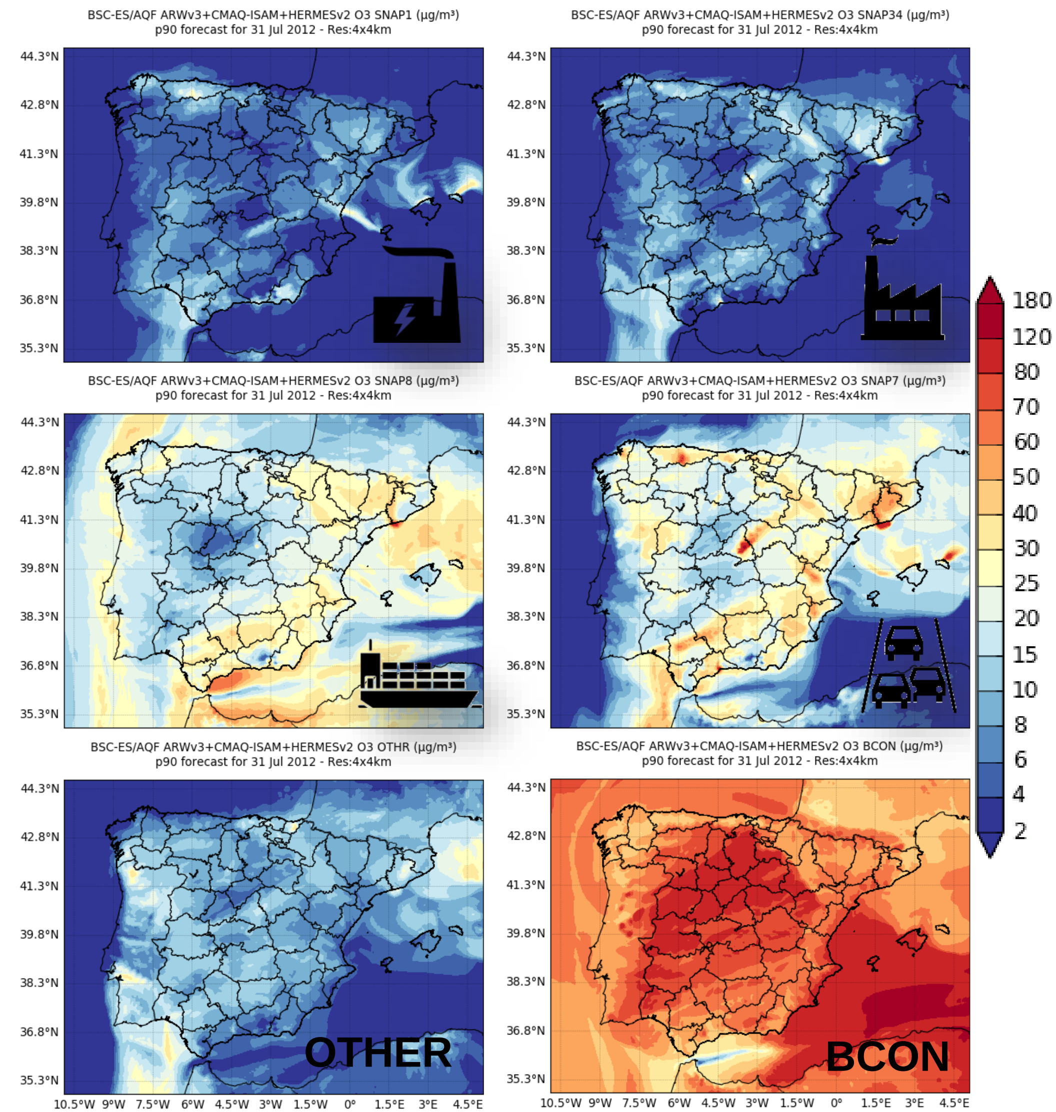




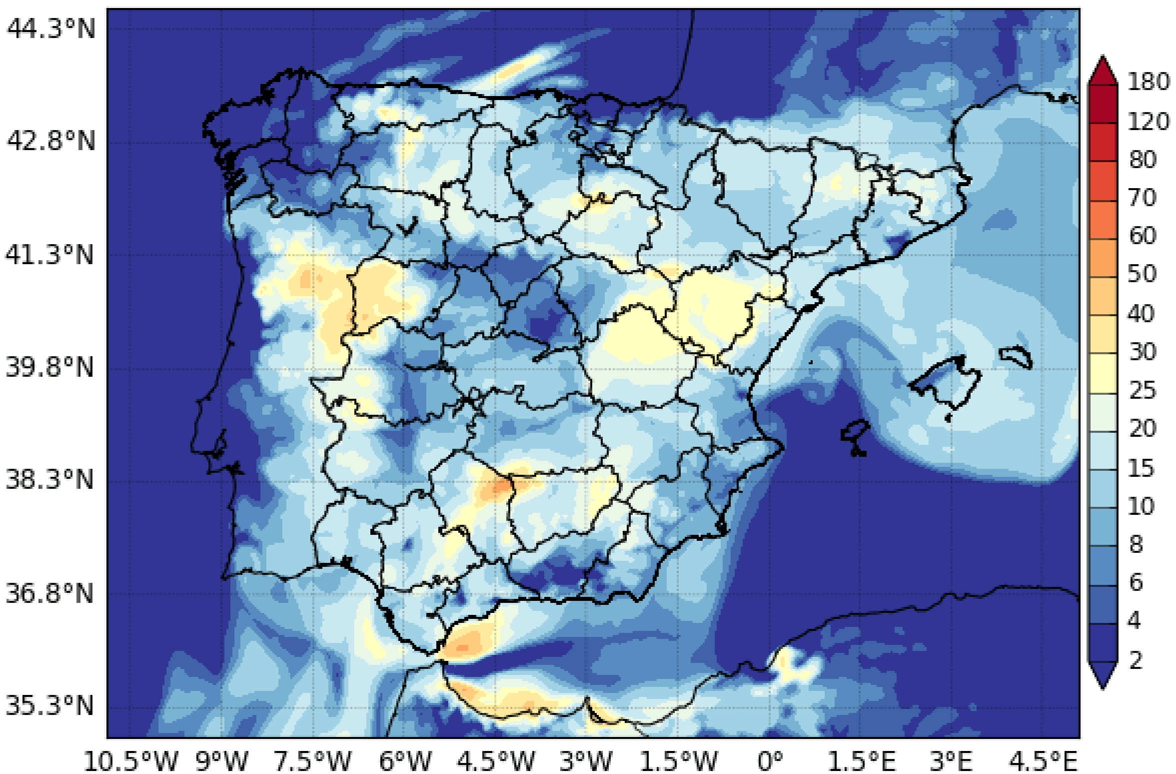
Source-sector contribution during peaks ITL – July







BSC-ES/AQF ARWv3+CMAQ-ISAM+HERMESv2 O3 SNAP7 (µg/m³)
00h forecast for 00UTC 31 Jul 2012 - Res:4x4km



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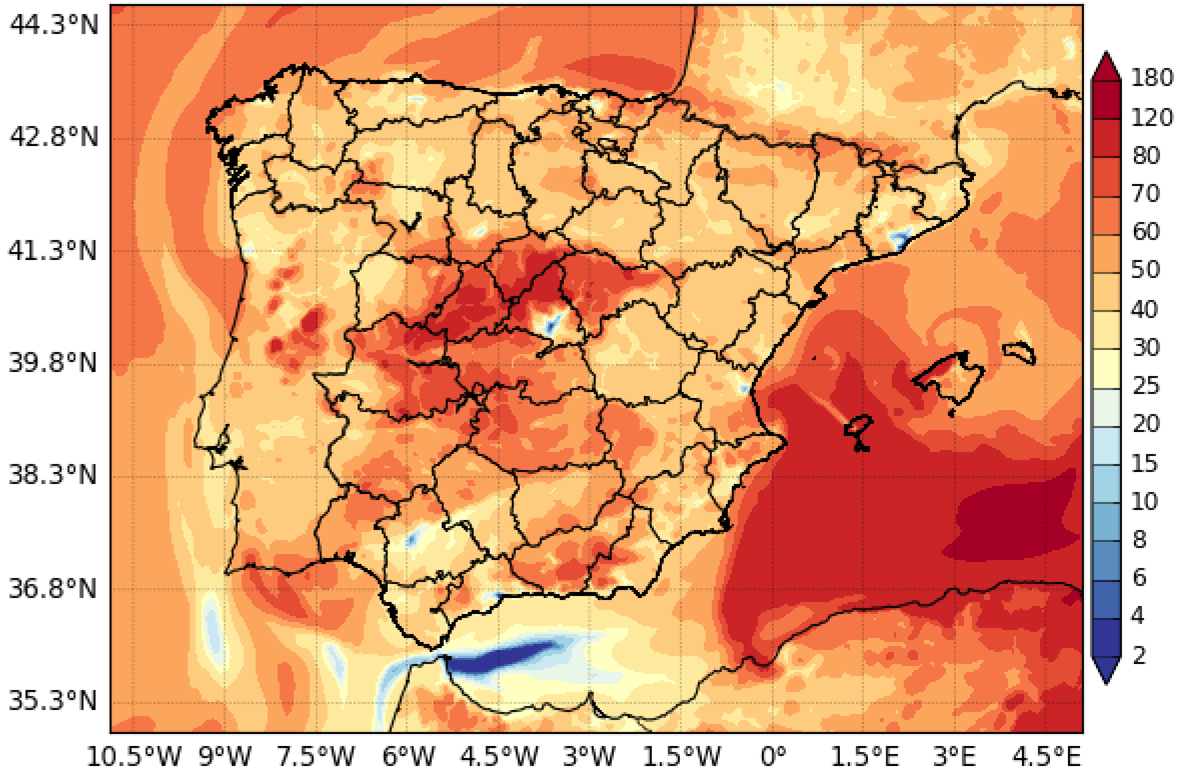
BSC-ES/AQF ARWv3+CMAQ-ISAM+HERMESv2 O3 BCON (µg/m³)
00h forecast for 00UTC 31 Jul 2012 - Res:4x4km



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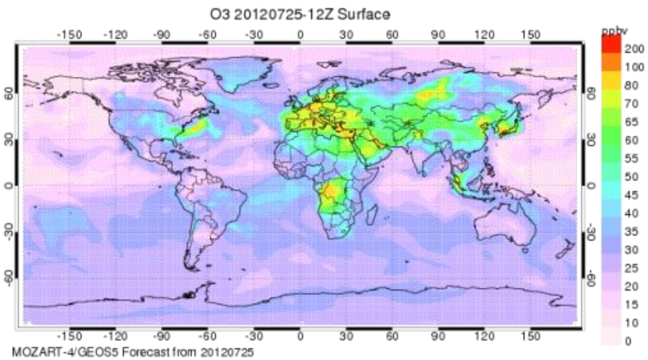

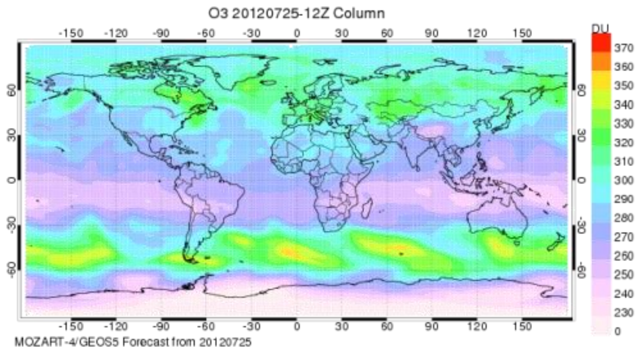

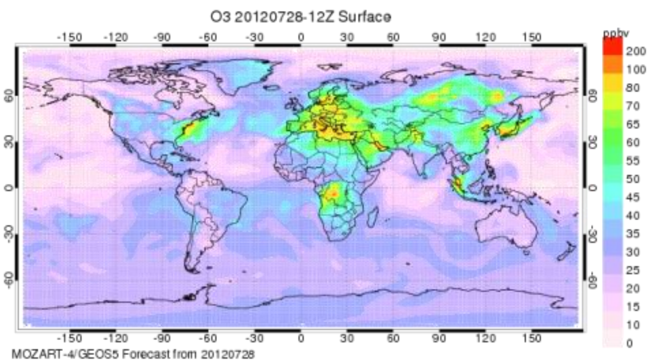

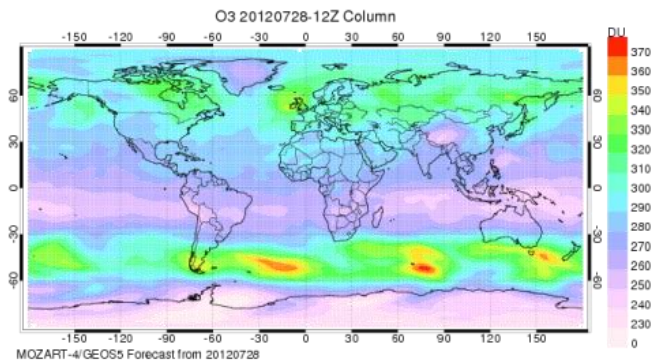

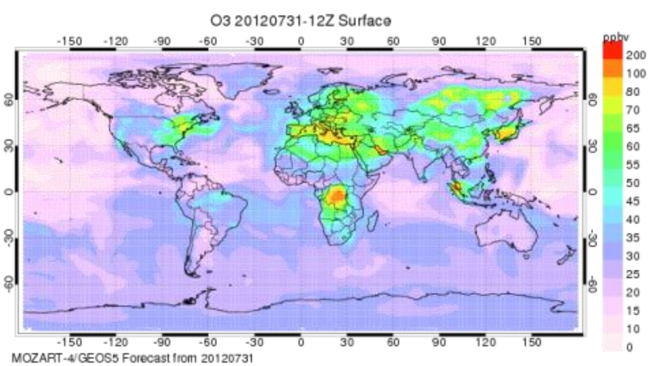

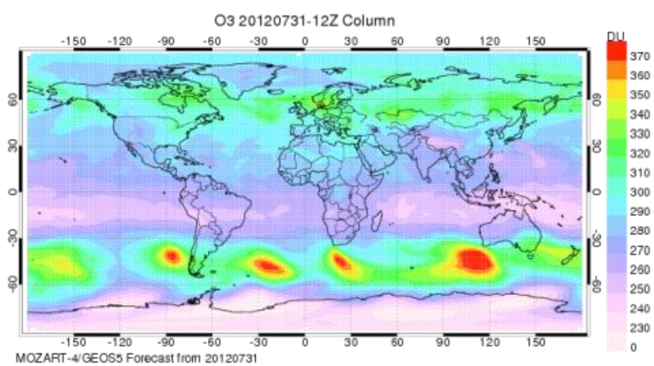

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STATE vs RESPONSE

What are the various contributors to modeled concentrations?

Source contribution approaches: tracks the formation and transport of O_3 and $PM_{2.5}$ from specific sources and allows the calculation of contributions at each hour and grid cell

STATE: relative importance of sources that contribute to high concentrations)

- ✓ Brute force zero out
- ✓ Ozone and PM source apportionment

How will the modeled concentrations change based on changes to emissions?

Source sensitivity approaches: estimates sensitivity coefficients that relate emission changes from specific emission sources to model outcomes at each hour and grid cell

RESPONSE: prediction of how pollutant will respond to reductions in precursor emissions

- ✓ Brute force zero out
- ✓ Decoupled Direct Method (DDM)