2009-2018 trends on PM2.5-SOA in NE Spain as inferred from receptor source apportionment analysis

In 't Veld M., et al 2021 Sci. Total Environ. Submitted

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TFMM-EMEP, May 11-12, 2020

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CSIC

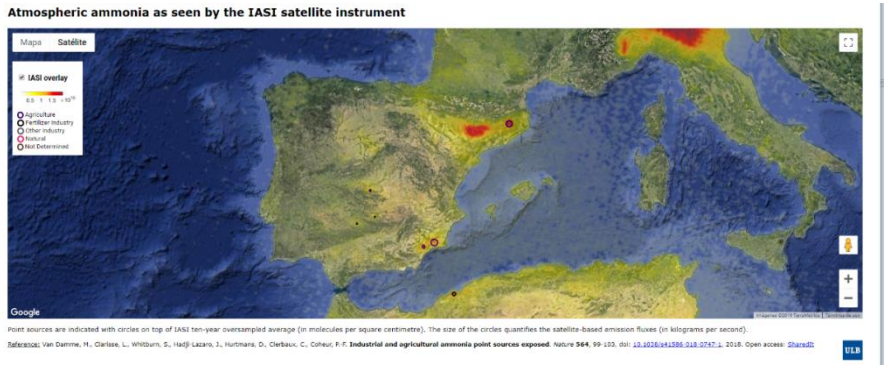
Introduction

- PM levels in NE Spain decreased by 50% since 2005, but since 2010 levels are quite constant
- Countries having adopted already WHOGs for PM2.5 as AQ National Standards seem to continue decreasing PM levels (Kutlar et al., 2017)
- PM speciation at urban (and regional background) available since 1999 (2002)
- Specific EU, national, regional and local policies have been identified as key ones (Querol et al., 2012 and 2014; Pandolfi et al., 2014) and highly affected sulphate, BC, POA, metals
- NE Spain is an O_3 and NH_3 hotspot Querol et al. (2016), Van Damme et al (2018)

Objectives

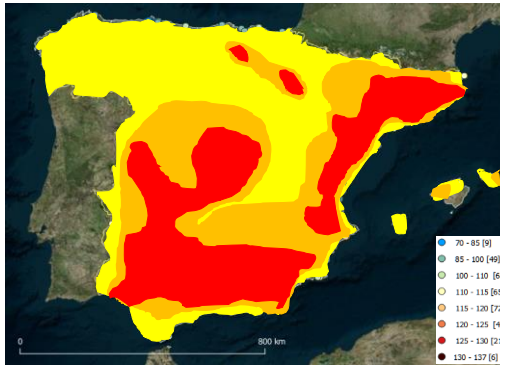
Analysing how SOA has evolved

http://www.ulb.ac.be/cpm/NH3-IASI.html



Exceedance of the WHOAQG

Averaged 93.2 percentile 8h-DM O_3



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MINISTERIO PARA LA TRANSICIÓN ECOLÓGICA Y EL RETO DEMOGRÁFICO

GOBIERNO DE ESPAÑA
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Generalitat de Catalunya

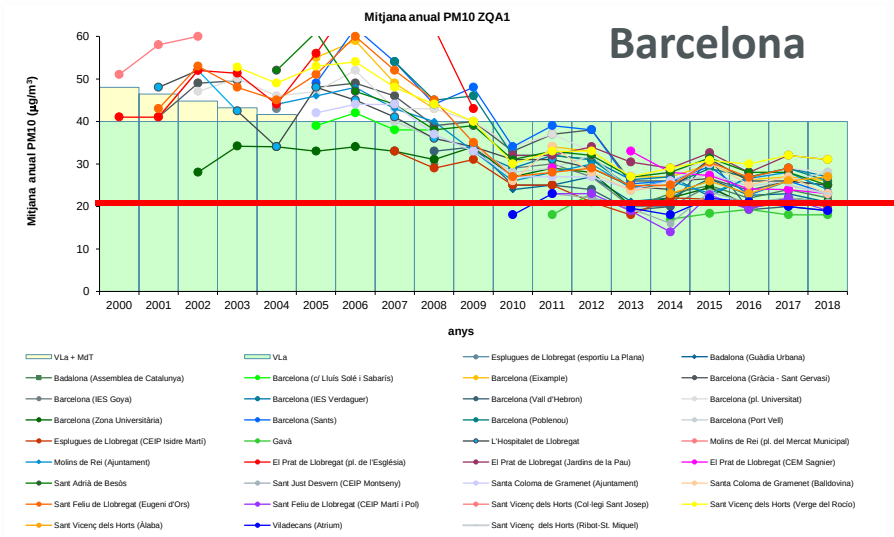
Introduction

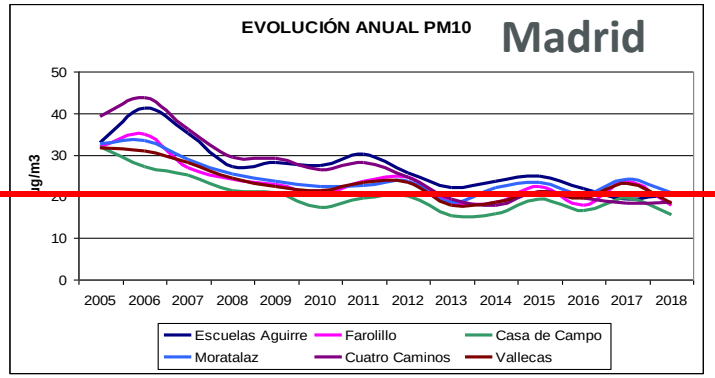
Updated from Kutlar Joss et al., 2017. 20 years of E.U. resistance against science-based «limit values» for PM10 & PM2.5
Int J Pub Health 2017

	Science-based limit values to protect public health				Politically set «limit values»
µg/m³ Annual mean	WHO Air Quality Guideline values	Canada, Iceland, Iran, Australia Switzerland UK	State of California	USA Federal, Mexico Cuba	E.U.
PM_{10}	20	20	20	--	40
$PM_{2.5}$	10	10 (AUST: 8)	12	12	25

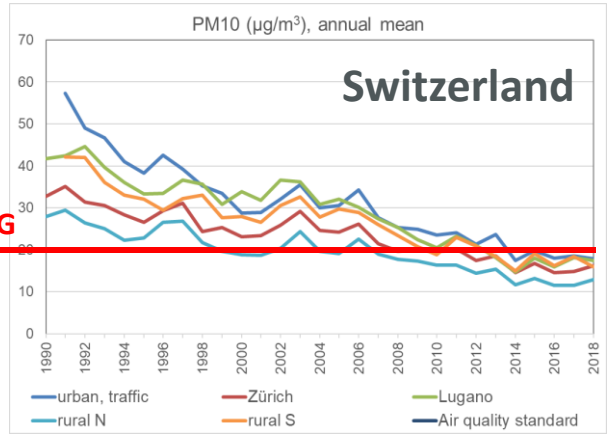
Introduction

PM10

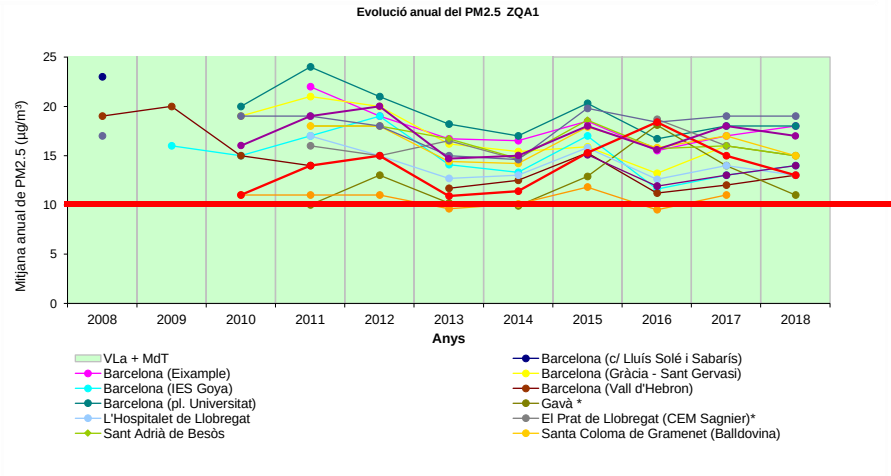


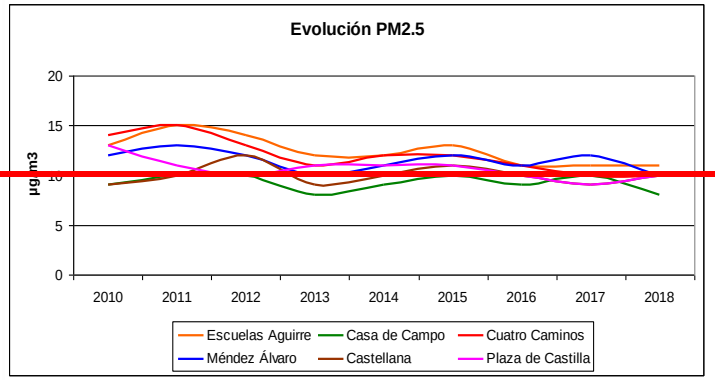


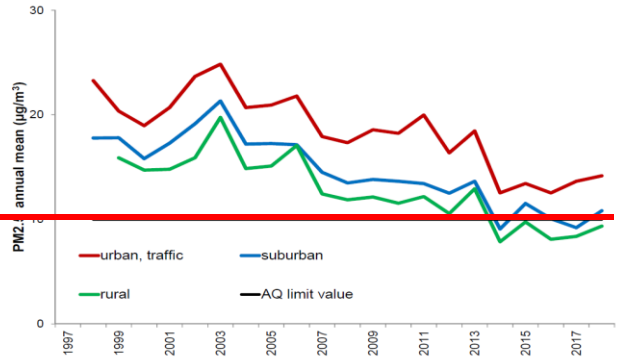
WHO AQG



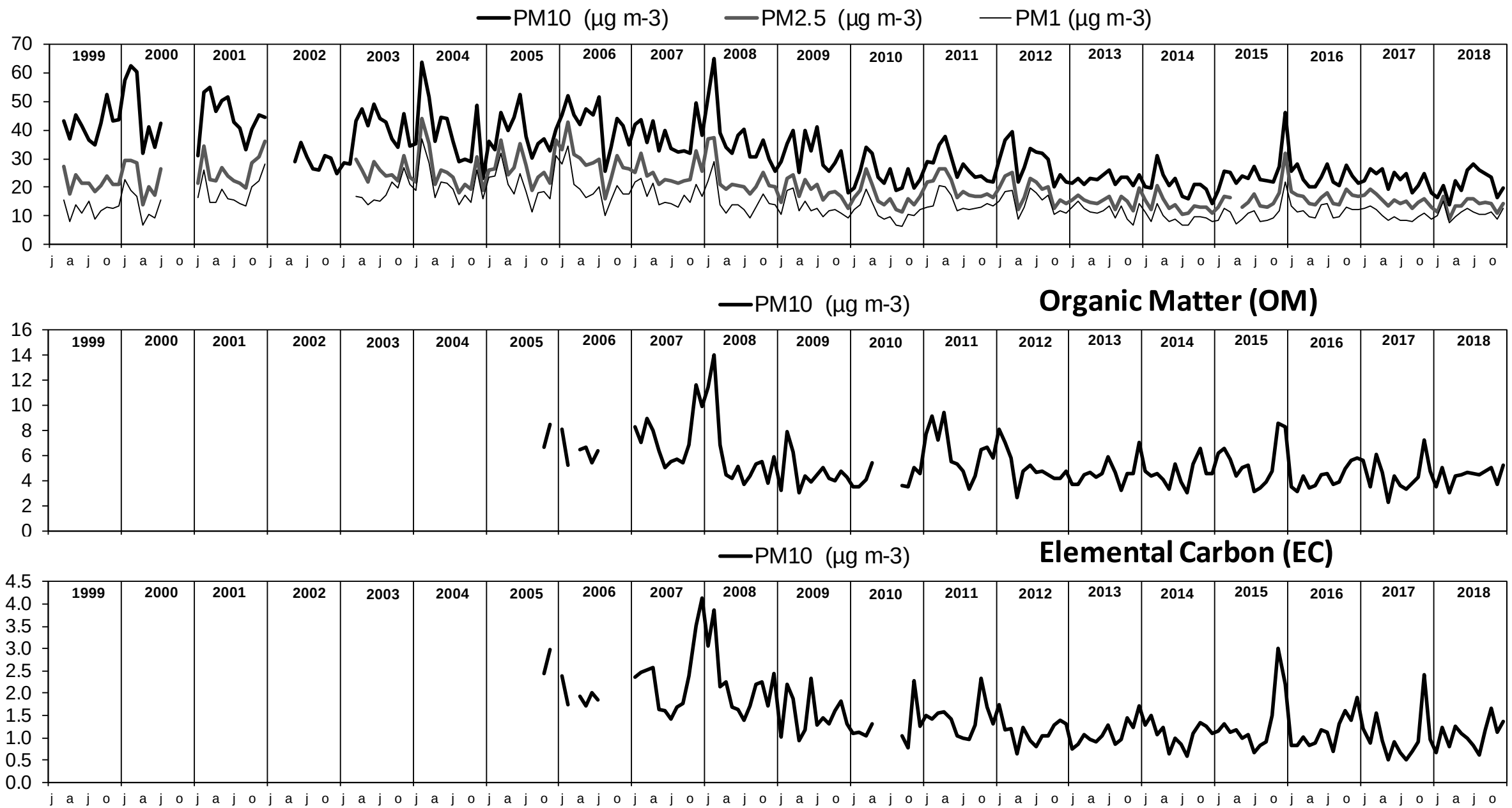
PM2.5





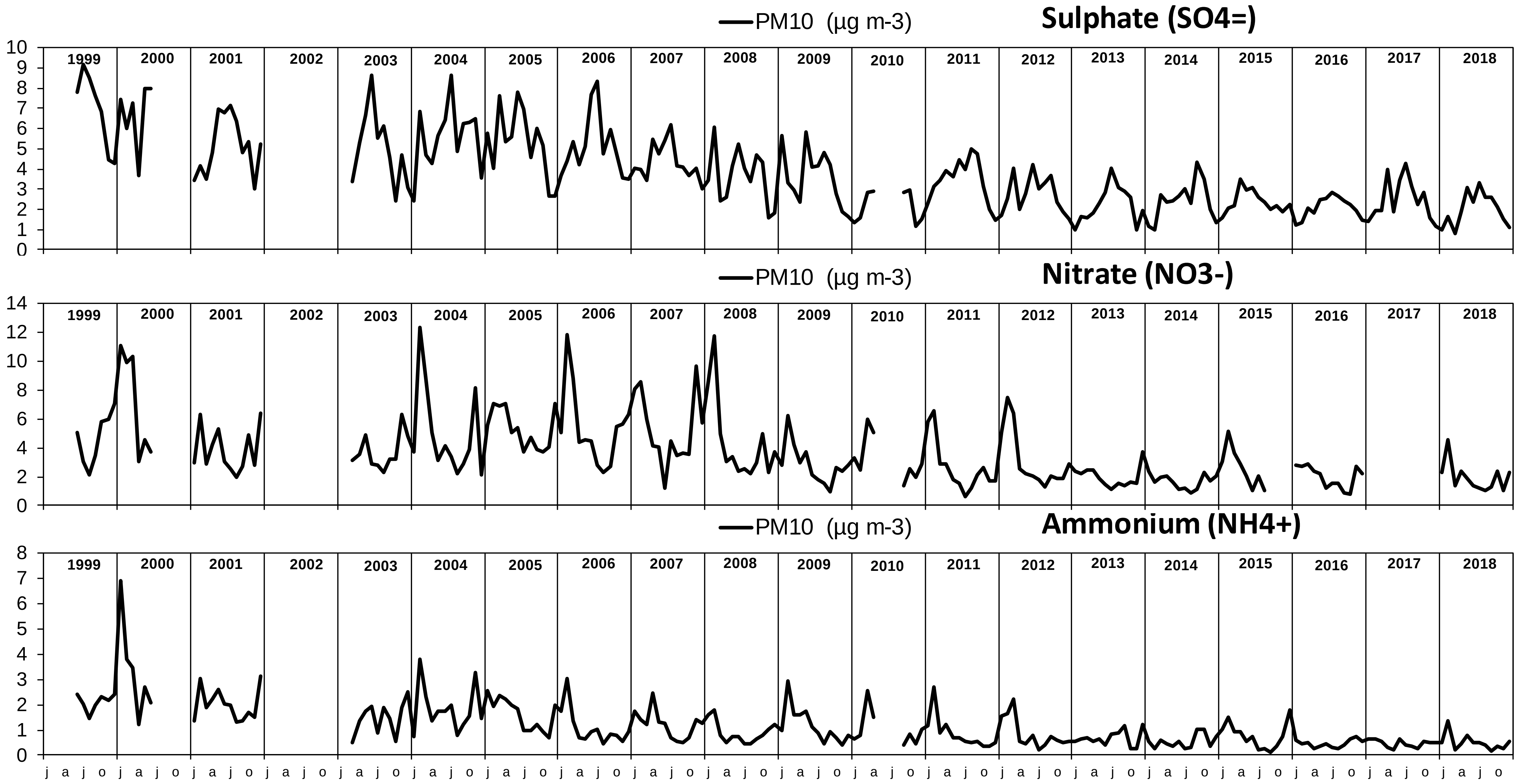


Introduction



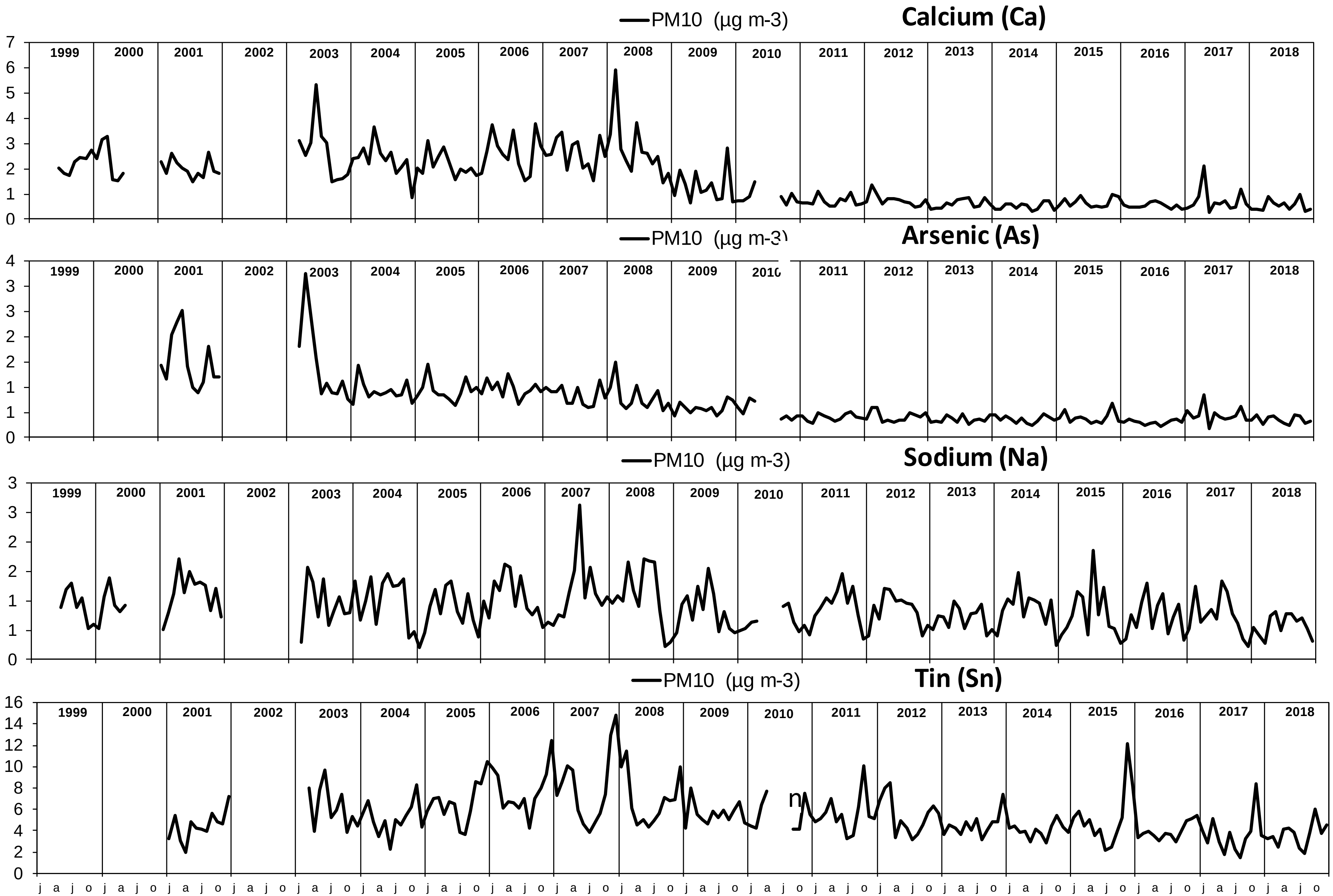
Introduction

BARCELONA 1999-2018



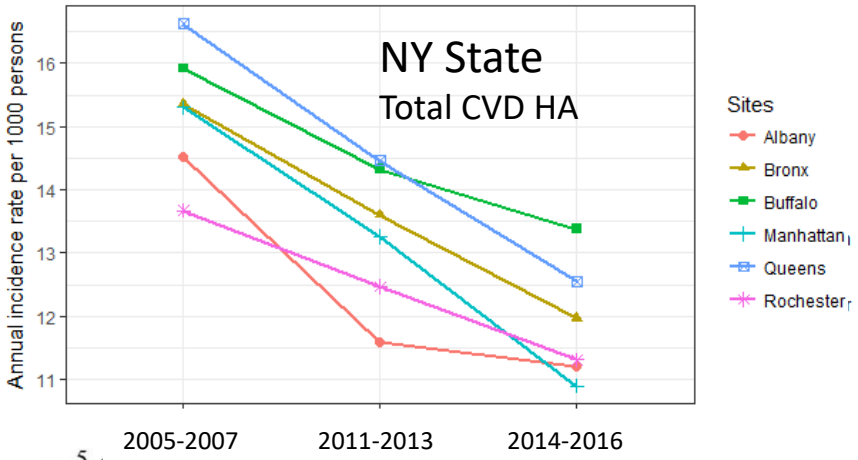
Introduction

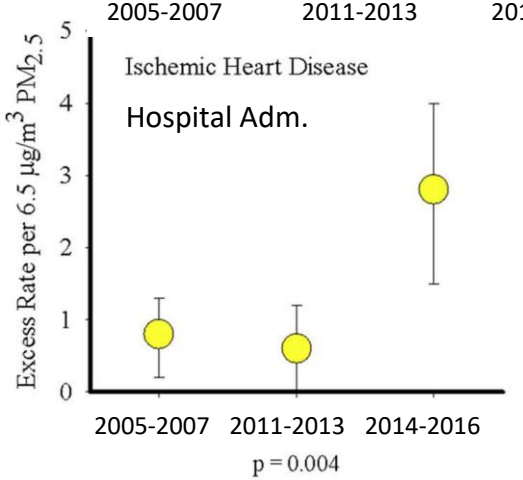
BARCELONA 1999-2018



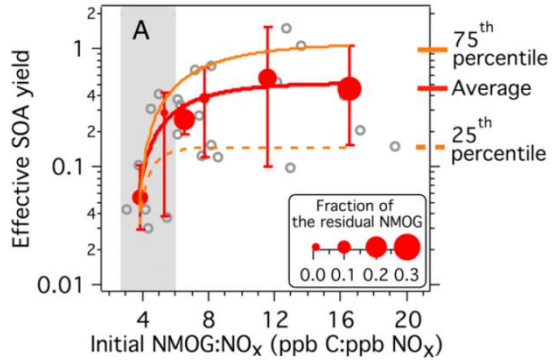
Introduction

Zhang W., Lin S., Hopke P.K. et al., 2018 Env. Poll. 42, 1404-16





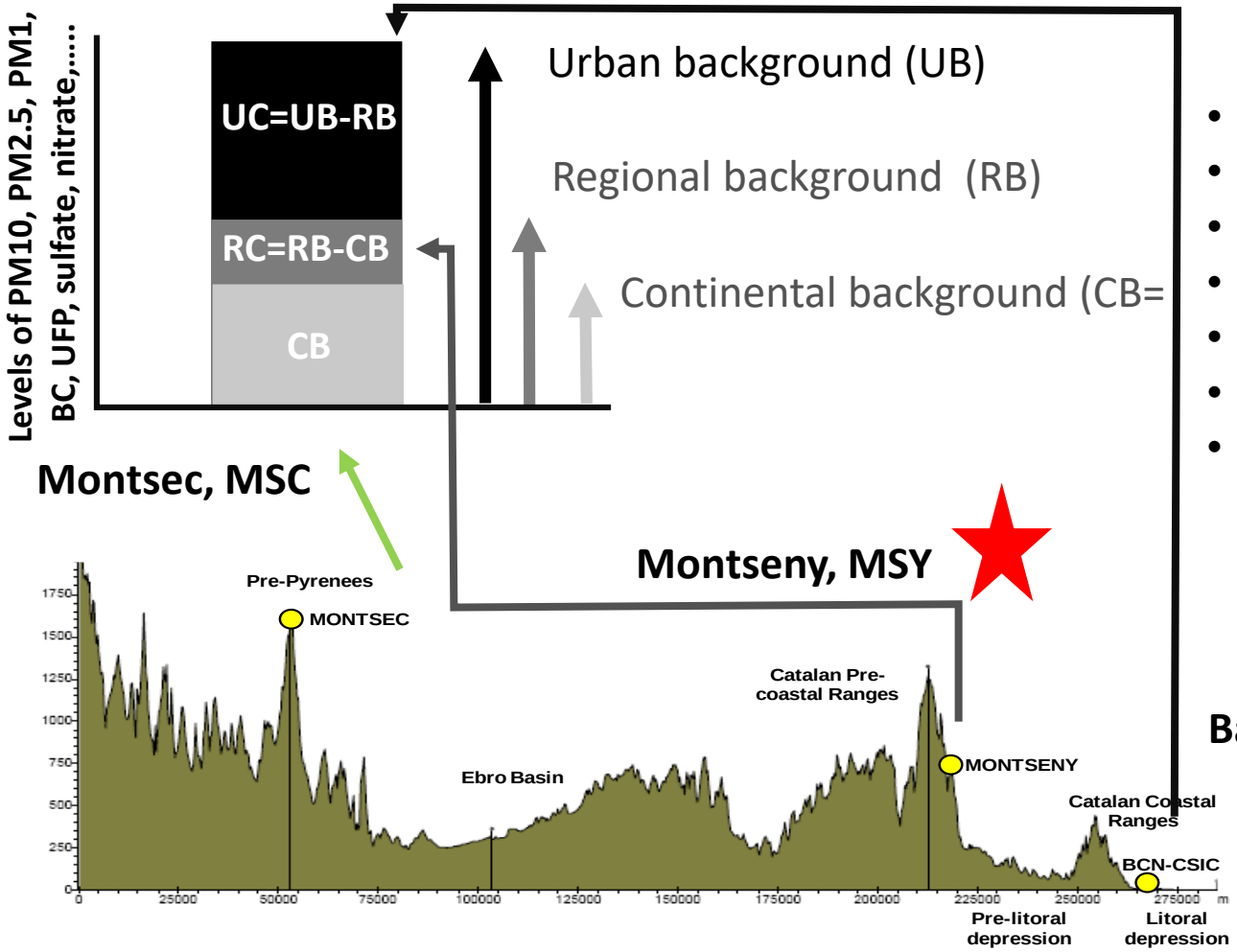
Zhai et Y. al., 2017 PNAS 114, 27, 6987-9
California



Hypothesis: PM2.5 might be more toxic because it has relatively more secondary organic aerosols (SOA)

Scientific and policy challenges to abate secondary organic contributions

Methodology



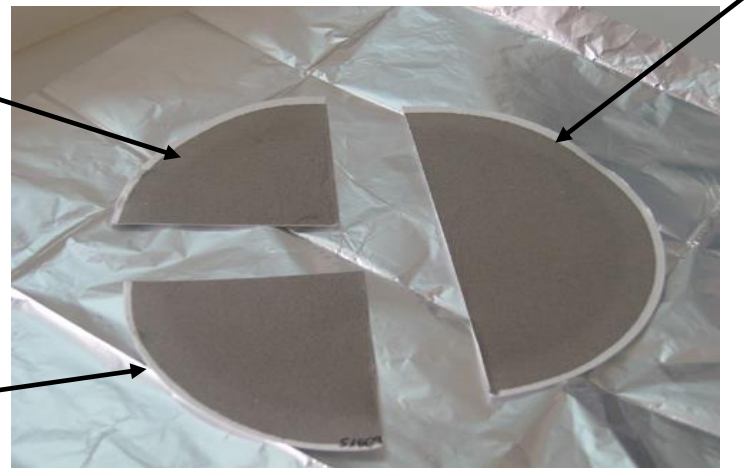
- 2009-2018
- The same OC&EC analysis protocol
- PM2.5
- 24 h
- 1 every 4 days simultaneously at the BCN and MSY
- DIGITEL and MCV high volume samplers (30 m^3h^{-1})
- 15 cm diameter Pall ultrapure quartz microfiber filters

Barcelona, BCN

Methodology

Analyses

OC, EC
Thermo-optical-transmission



Acidic digestion ($HF:HNO_3:HClO_4$)

ICP-AES:

Al, Ca, K, Na, Mg, Fe, Ti, P

ICP-MS:

Li, Ti, V, Cr, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Y, Zr, Cd, Sn, Cs, Ba, La, Ce, Pr, Nd, Hf, Tl, Pb, Bi, Th, U

H_2O leaching

Ion Chromat.:

NO_3^-, Cl^-, $SO_4^=$

Colorimetry FIA and ICP-AES:

NH_4^+, K^+, Ca^{2+}, Mg^{2+},…

Mass determined: 75-85% $PM_{2.5}$

Receptor modeling

- Positive Matrix Factorization (PMF) analysis
- Single site
- Multi-site

Trend analysis

Results

Inter-annual trends PM2.5 and PM2.5 components

PM2.5 µg m^{-3}

-3.4% 2009-2018

Barcelona Urban

-2.3% 2009-2018

BCN	$PM_{2.5}$	OA	SOA	EC	Crustal	Al_2O_3	Na_2O_{dust}	K_2O_{dust}	TiO_2	SIA	$SO_4^{2-}{}_{nss}$	NO_3^-	NH_4^+	Tracers	V	K_{bb}
2009	12.62	3.57	2.21	1.17	1.86	0.25	0.05	0.04	0.01	5.08	2.63	1.36	1.09	0.09	6.61×10^{-3}	0.14
2010	12.30	3.94	2.89	0.90	1.47	0.19	0.04	0.03	0.01	5.35	2.53	1.52	1.30	0.10	4.77×10^{-3}	0.14
2011	13.27	5.02	2.69	1.14	1.25	0.15	0.03	0.02	0.01	5.13	2.80	1.27	1.06	0.08	5.71×10^{-3}	0.22
2012	10.96	4.08	2.99	0.94	0.92	0.13	0.03	0.02	0.01	4.47	2.28	1.25	0.93	0.07	5.11×10^{-3}	0.18
2013	10.18	4.18	3.01	1.01	1.10	0.12	0.03	0.02	0.01	3.35	1.74	0.91	0.71	0.09	3.65×10^{-3}	0.14
2014	9.93	3.71	2.66	0.90	1.19	0.15	0.03	0.02	0.01	3.44	1.90	0.87	0.68	0.07	3.65×10^{-3}	0.15
2015	11.70	4.71	3.47	1.06	1.42	0.17	0.03	0.03	0.01	4.01	1.83	1.25	0.92	0.07	4.08×10^{-3}	0.13
2016	10.42	3.86	2.83	0.89	1.85	0.27	0.06	0.04	0.01	3.44	1.62	1.18	0.65	0.07	3.06×10^{-3}	0.12
2017	10.08	3.91	2.83	0.93	1.51	0.16	0.03	0.03	0.01	3.32	1.67	1.07	0.58	0.06	3.71×10^{-3}	0.09
2018	8.92	3.55	2.63	0.79	1.08	0.12	0.02	0.02	0.01	3.20	1.63	0.94	0.63	0.04	3.22×10^{-3}	0.08
Trend (% yr^{-1})	*-0.3416*	*ns*	*ns*	*ns*	*ns*	*ns*	*ns*	*ns*	*ns*	*-0.2334*	*-0.1224*	*-0.0456*	*-0.0634*	*-0.0050*	*-0.0003*	*-0.0133*
(significance)	*(**)*									*(*)*	*(***)*	*(*)*	*(*)*	*(***)*	*(**)*	*(+)*

-2.4%

Montseny Regional Background

-1.0%

MSY	$PM_{2.5}$	OA	SOA	EC	Crustal	Al_2O_3	Na_2O_{dust}	K_2O_{dust}	TiO_2	SIA	$SO_4^{2-}{}_{nss}$	NO_3^-	NH_4^+	Tracers	V	K_{bb}
2009	7.54	3.06	2.43	0.21	0.58	0.10	0.02	0.02	6.88×10^{-3}	3.02	1.70	0.52	0.80	0.03	1.62×10^{-3}	0.05
2010	6.32	2.79	2.18	0.21	0.40	0.07	0.01	0.01	3.73×10^{-3}	2.59	1.38	0.67	0.54	0.02	1.21×10^{-3}	0.05
2011	7.34	3.51	2.80	0.24	0.44	0.07	0.01	0.01	3.61×10^{-3}	2.93	1.87	0.49	0.57	0.02	1.54×10^{-3}	0.04
2012	7.00	3.57	3.06	0.18	0.52	0.08	0.02	0.01	3.73×10^{-3}	2.53	1.46	0.49	0.58	0.02	1.34×10^{-3}	0.05
2013	5.53	2.73	2.15	0.20	0.43	0.05	0.01	0.01	2.96×10^{-3}	1.97	1.06	0.50	0.41	0.03	1.09×10^{-3}	0.03
2014	5.41	2.49	2.09	0.14	0.32	0.04	0.01	0.01	2.41×10^{-3}	2.18	1.38	0.31	0.49	0.02	1.15×10^{-3}	0.03
2015	6.28	2.79	2.21	0.20	0.59	0.10	0.02	0.02	3.52×10^{-3}	2.46	1.31	0.61	0.53	0.02	1.09×10^{-3}	0.05
2016	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
2017	4.95	2.53	1.96	0.19	0.17	0.01	1.40×10^{-3}	1.10×10^{-3}	1.25×10^{-3}	1.98	1.03	0.47	0.47	0.02	9.09×10^{-4}	0.04
2018	5.54	2.83	2.38	0.15	0.38	0.05	0.01	0.01	1.58×10^{-3}	2.11	1.11	0.50	0.50	0.01	9.82×10^{-4}	0.03
Trend (% yr^{-1})	*-0.2350*	*ns*	*ns*	-0.0047	*ns*	*-0.0070*	*-0.0014*	*-0.0011*	*-0.0004*	*-0.1014*	*-0.0654*	*ns*	*-0.0127*	*-0.0016*	*-0.0001*	*ns*
(significance)	*(**)*			(*)		*(+)*	*(+)*	*(+)*	*(***)*	*(*)*	*(**)*		*(+)*	*(*)*	*(**)*	

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Results

Inter-seasonaltrends PM2.5 and PM2.5 components

Species	Autumn trend Barcelona ($\mu gm^{-3}yr^{-1}$)	α
$PM_{2.5}$	*ns*	
TiO_2	*ns*	
Sea-Spray Na	-0.00940	+
SIA	-0.12277	+
$SO_4^{2-}{}_{nss}$	-0.10106	*
Trace Elements	-0.00620	*
V	-0.00044	**
Ni	-0.00015	*
K_{bb}	-0.01414	*

Species	Winter trend Barcelona ($\mu gm^{-3}yr^{-1}$)	α
$PM_{2.5}$	*ns*	
Na_{ss}	*ns*	
Trace Elements	*ns*	
V	*ns*	

Species	Spring trend Barcelona ($\mu gm^{-3}yr^{-1}$)	α
$PM_{2.5}$	-0.64030	*
Crustal	*ns*	
Mineral	*Ns*	
Al_2O_3	*ns*	
Na_2O_{dust}	*ns*	
K_2O_{dust}	*ns*	
TiO_2	*ns*	
Fe_2O_3	*ns*	
SIA	-0.44996	***
NO_3^-	-0.14758	*
NH_4^+	-0.12767	***
$SO_4^{2-}{}_{nss}$	-0.15924	**
Trace Elements	-0.00620	*
V	-0.00033	**

Species	Summer trend Barcelona ($\mu gm^{-3}yr^{-1}$)	α
$PM_{2.5}$	-0.35454	**
EC	-0.02514	**
Fe_2O_3	+0.00809	**
MnO	-0.00022	**
SIA	-0.18916	**
NH_4^+	-0.05533	*
$SO_4^{2-}{}_{nss}$	-0.15522	*
Trace Elements	-0.00460	*
V	-0.00047	**

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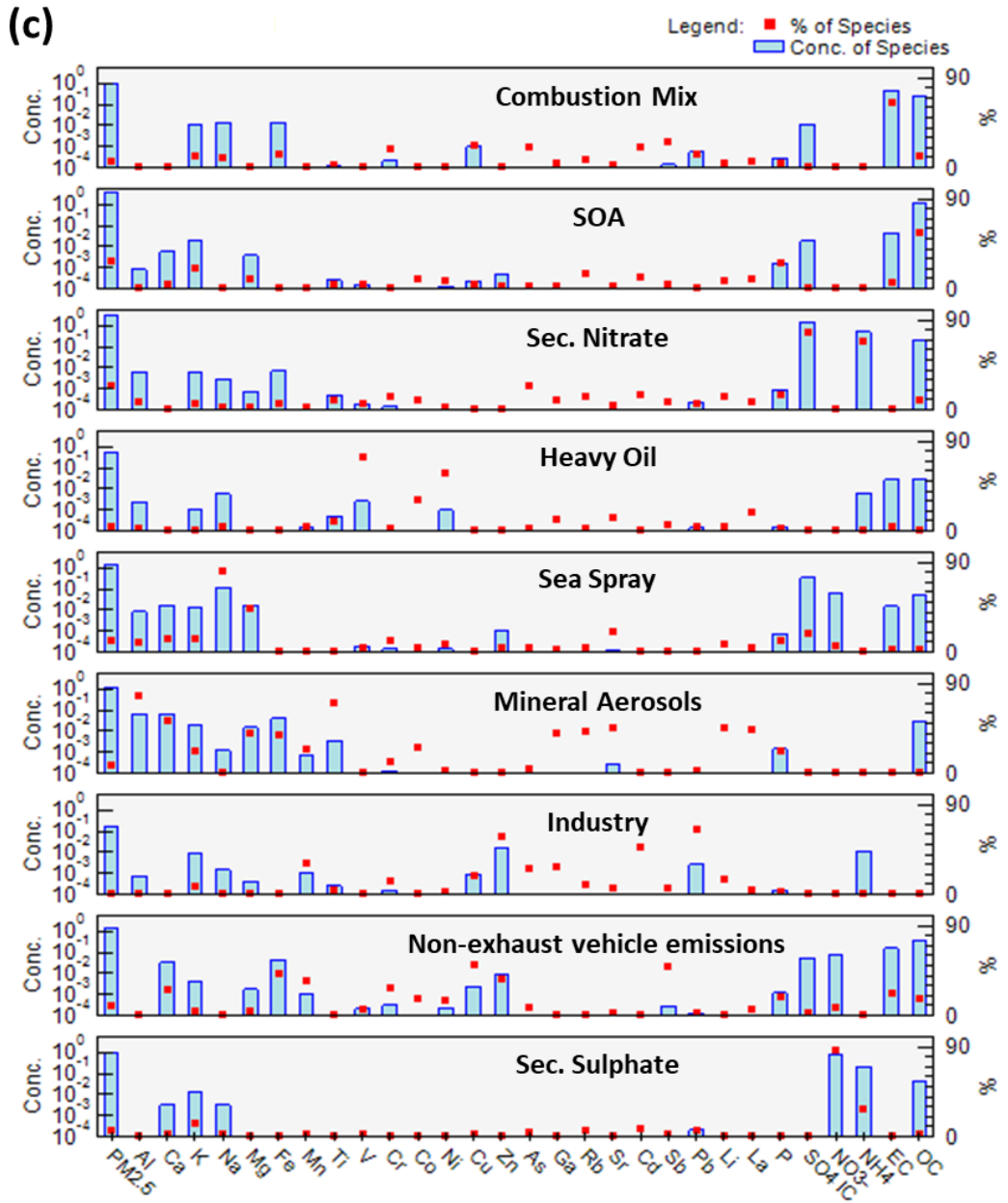
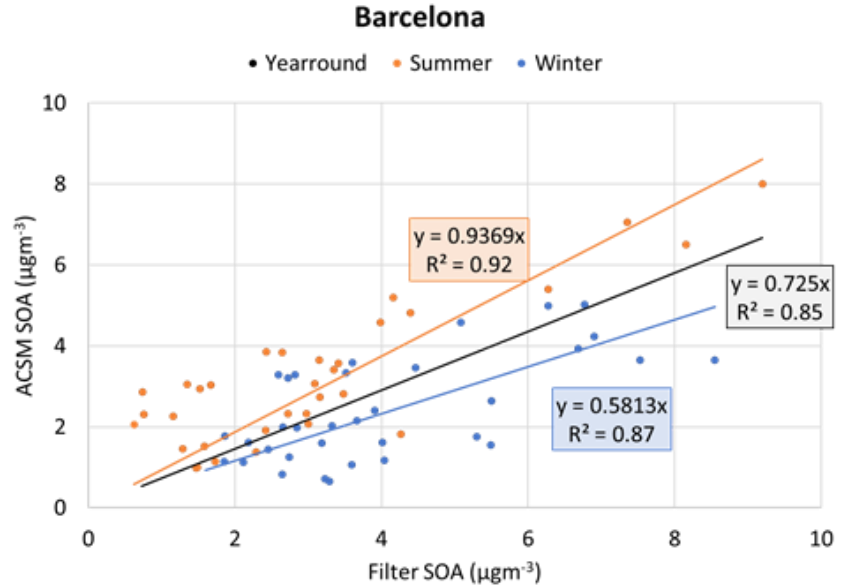
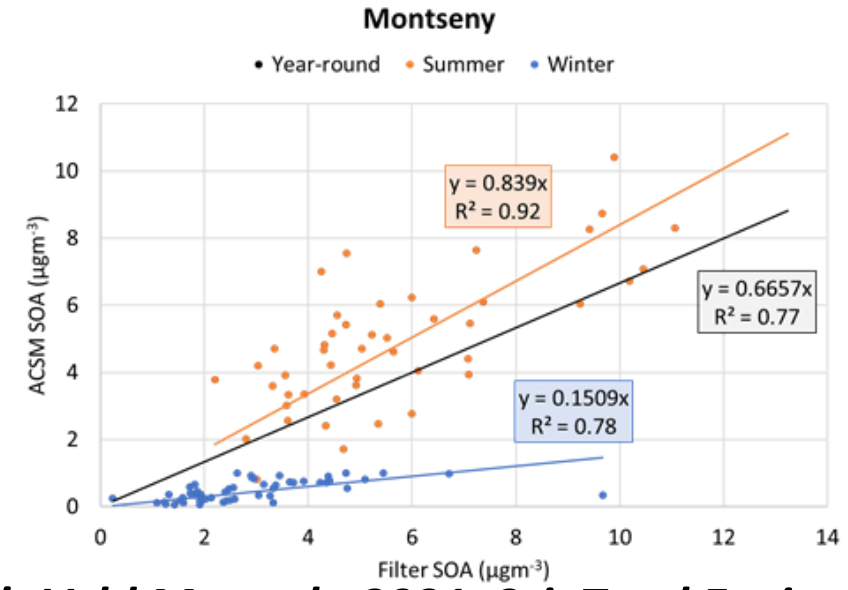
Results

Multi-site-PMF profiles



Receptor modelling (PMF) & validation SOA with ACSM





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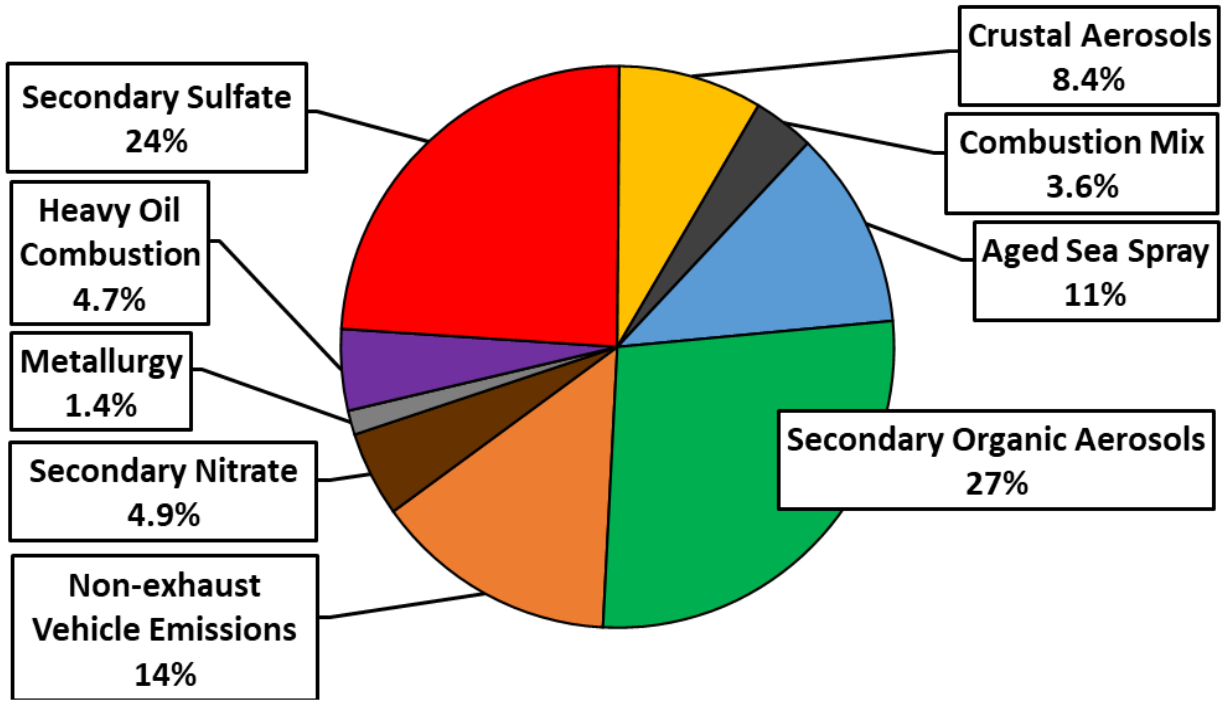
Results

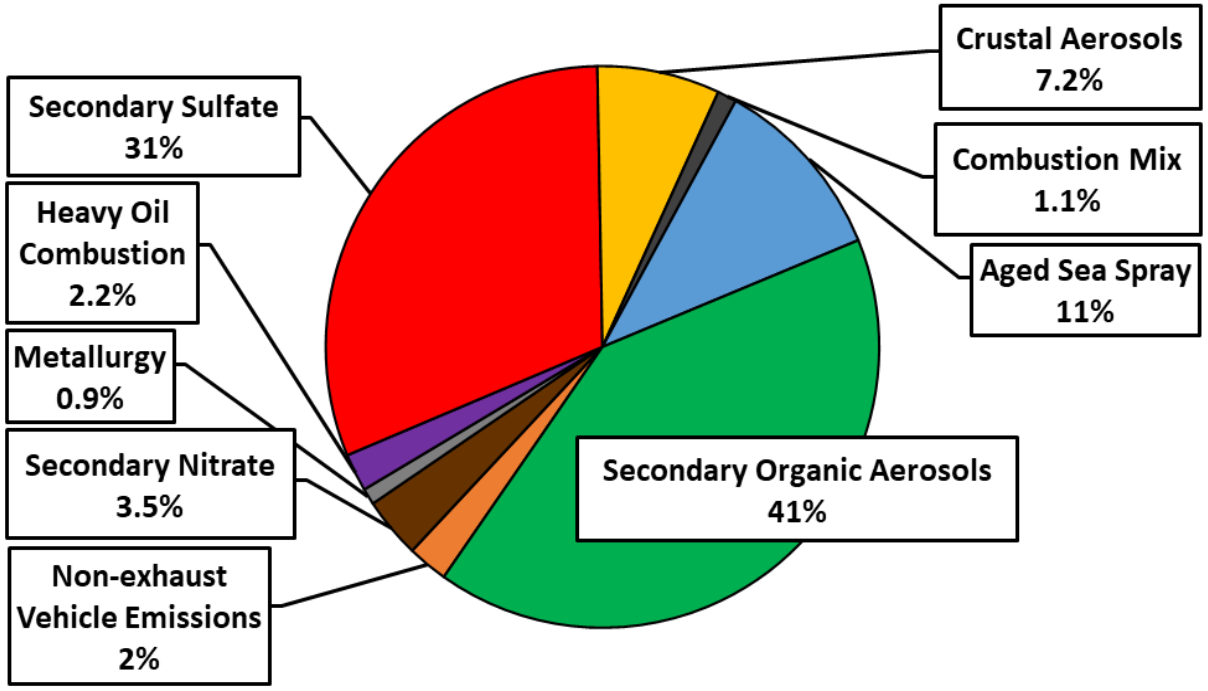
Receptor modelling (PMF)

PM2.5

Multisite Barcelona



Multisite Montseny



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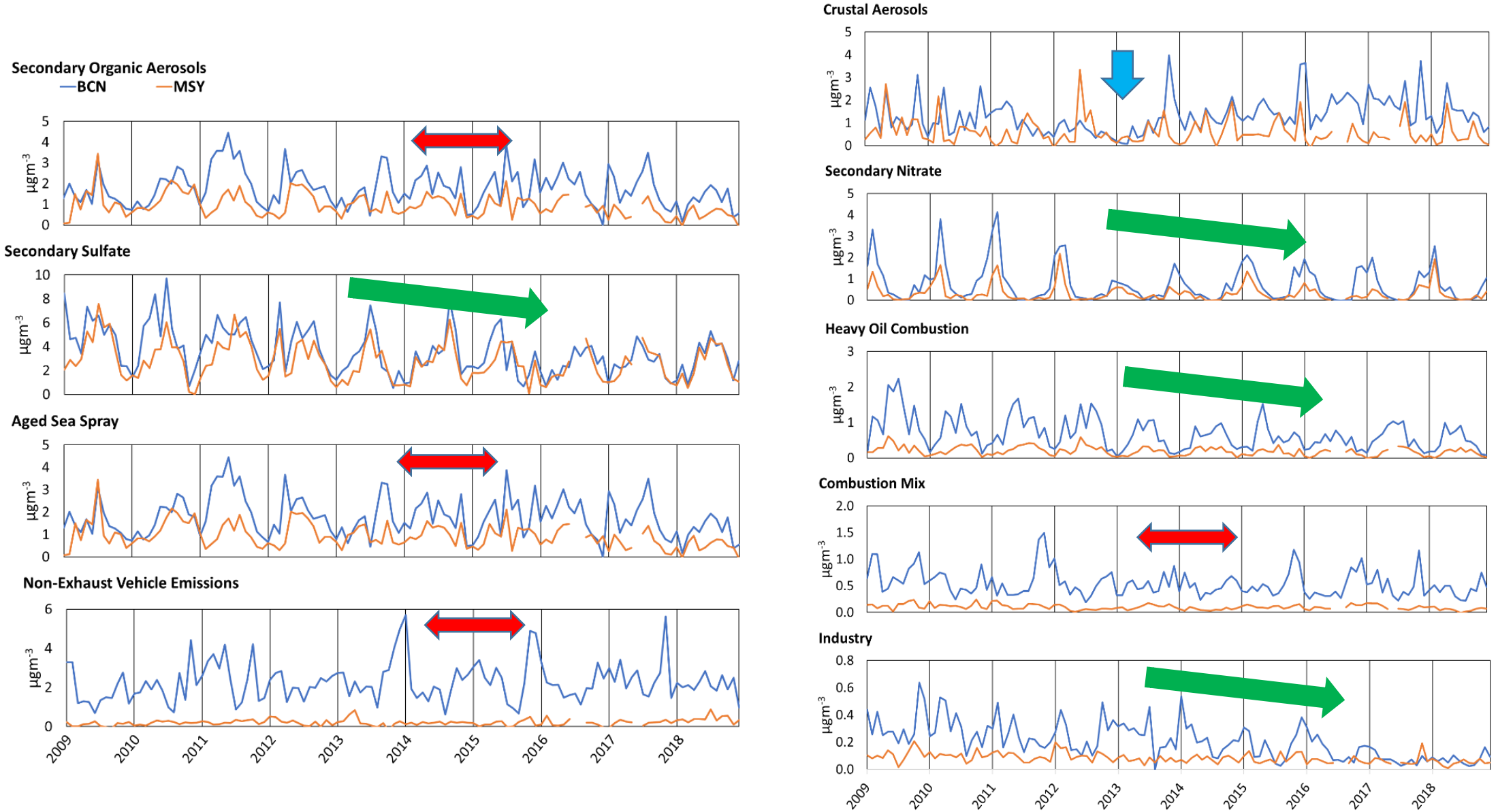
Results

Receptor modelling (PMF)



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Results

PM2.5 compositional trends

SOA from OC/EC method

2009-2018 **+6%** **-10%** **-6** **-3%**

BCN	OA	SOA	EC	SIA	$SO_4^{2-}_{nss}$	NO_3^-	NH_4^+	Crustal	Tracers
2009	30%	18%	10%	39%	22%	9%	8%	13%	1%
2010	33%	24%	8%	41%	20%	11%	10%	12%	1%
2011	39%	28%	9%	36%	22%	7%	7%	9%	1%
2012	39%	29%	9%	37%	21%	9%	7%	9%	1%
2013	42%	30%	10%	32%	17%	8%	7%	10%	1%
2014	38%	28%	9%	33%	19%	8%	6%	12%	1%
2015	41%	30%	9%	32%	16%	9%	7%	12%	1%
2016	38%	28%	9%	31%	17%	9%	6%	18%	1%
2017	40%	29%	9%	32%	17%	9%	5%	14%	1%
2018	41%	30%	9%	34%	18%	10%	7%	12%	1%
Trend (% yr^{-1})		*+0.65*		*-1.03*	*-0.58*		*-0.29*		*-0.04*
(significance)		*(*)*		*(*)*	*(*)*		*(**)*		*(+)*

2009-2018 **+4%**

MSY	OA	SOA	EC	SIA	$SO_4^{2-}_{nss}$	NO_3^-	NH_4^+	Crustal	Tracers
2009	41%	32%	3%	37%	21%	6%	10%	8%	1%
2010	46%	35%	4%	38%	22%	9%	7%	6%	0%
2011	49%	40%	3%	38%	26%	5%	7%	6%	0%
2012	53%	45%	3%	34%	20%	7%	7%	7%	0%
2013	50%	39%	4%	33%	18%	9%	7%	9%	1%
2014	48%	40%	3%	37%	23%	7%	8%	7%	0%
2015	44%	34%	4%	38%	21%	10%	8%	10%	0%
2016									
2017	54%	41%	4%	39%	20%	10%	9%	1%	0%
2018	50%	40%	3%	38%	19%	10%	9%	7%	0%
Trend (% yr^{-1})						+0.41			
(significance)						*(*)*			

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Final considerations

SOA calculated using primary OC/EC ratios

BCN	MSY
2009-2010	2009-2010
SOA 21%	**SOA 33%**
SIA 40%	SIA 38%
2017-2018	2017-2018
SOA 30%	**SOA 41%**
SIA 33%	SIA 38%

SOA with PMF

SOA is the largest source contributing to PM2.5 at both stations,
30% in BCN and 44% in MSY by 2018,
An increase of an additional 9% in BCN and 7% in MSY from 2009

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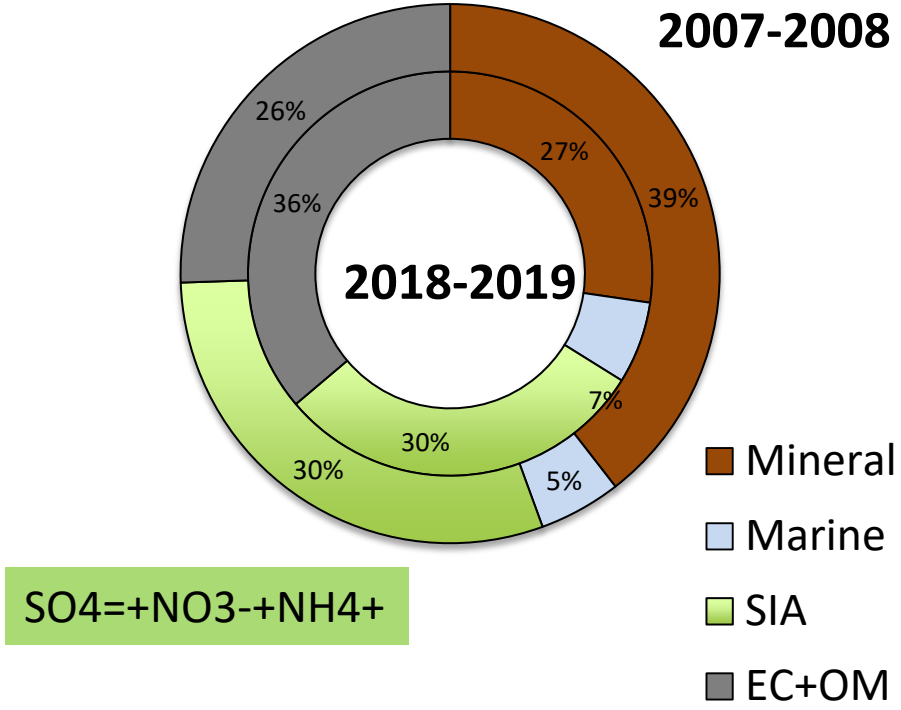
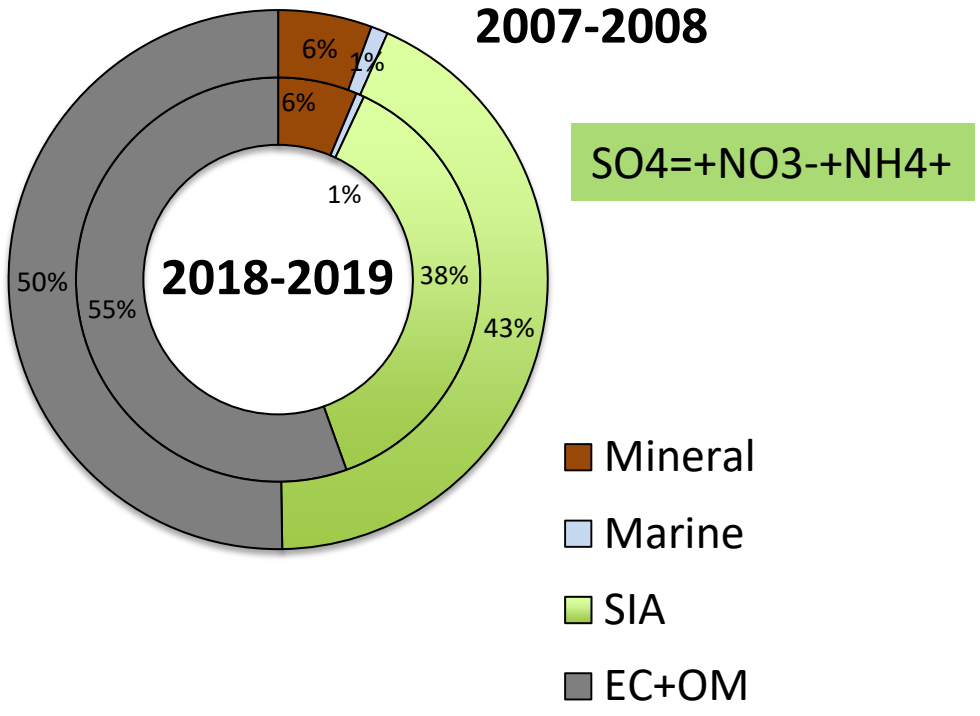
Final considerations

BARCELONA

PM10



PM1



Oxidative potential analysis for twin PM10 and PM1 in Grenoble

G. Uzu & J.L. Jaffrezo

Thank you for your attention!!!!!

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