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Source apportionment studies review

Vanessa N. dos Santos María Teresa Pay

Barcelona

Outile

- Why source apportionment (SA)?
- SA methods
- Review of receptor SA applications (very short!)
- Review of modelling SA applications (science & service)
- Brainstorming

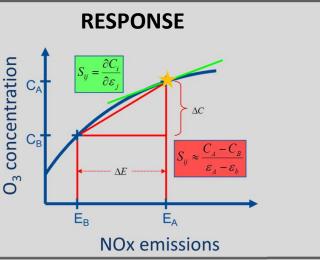


SA and SS: different purposes

Source Apportionment (SA) SA quantifies the contribution of a source to the pollutant concentration. Which is the origin of high O₃ concentration in Vic? Is it coming from the traffic in Barcelona?

Source Sensitivity (SS)

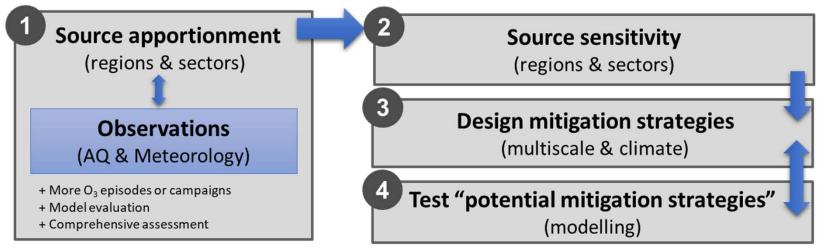
- SS estimates the impact of changing emission sources on pollutant concentration.
- What happened with the O3 in Vic if emission from traffic in Barcelona are reduced?



Why source apportionment (SA)?

• Policy point of view:

- Annex XV of the AQD: information about sources has to be provided to establish the causes that determined an exceedance (PM10, PM2.5 and NO2)
- SA outputs has to be reported regions (regional, urban, local) and sectors (traffic, industry, residential, etc.).
- SA point relevant sources to be reduced \rightarrow previous step to SS studies.
- Research point of view
 - Unravelling the origin of air pollution in combination with observations.
 - Understanding the modelling uncertainties related with emissions.
 - Understanding the relative importance of imported vs. regional contribution.



SA modelling

Source oriented model **Receptor oriented model** (SM) (RM) **Emissions Inventory** Source-compositions Meteorology Chemistry -X Source Impacts Receptor (monitor) Air Quality Emissions-based Model (3D Chemical Transport Model (CTM): CMAQ, CAMx, STEM, GEOS-**Receptor Model:** Chem...) CMB, PMF, UNMIX...

- Too much temporal variability
- Applicable only at the receptor
- Too little temporal variability
- Uncertain inputs (meteo., emissions)
- Secondary pollutants can be apportioned.

None can be evaluated by direct observation

SA in Source Modelling (mainly CTM)				
	Source Sensitivity (SS)			
1) Brute Force (BF) or Emission Re				
1.1) Zero-out	Advantages:			
• ΔE _i /E _i = 100%	 Straightforward to any CTM 			
• $C_j = C - C_{all sectors-j}$	Disadvantages:			
1.2) Small ERP	 Not addressing the non-linearity in the attribution 			
 ΔE_j/E_j ~15-20% to find ΔC_j/ΔE_j 	(total mass not preserved)			
 C_j = ΔC_j/ΔE_j x E_j 	 Not real atmospheric conditions. 			
	 Number of runs proportional to sources (j). 			
1.3) Source allocation	Advantages:			
 ΔE/E_j from 0 to ~50% to find 	 Straightforward to any CTM 			
$\Delta C_{j} / \Delta E_{j}$ till linearly	 Valid for annual and seasonal studies (non-linearity 			
 Combination of ERP with SSR 	become less important)			
model	 Mass preserved by normalization. 			
 C_j = ΔC_j/ΔE_j x E_j 	Disadvantages:			
	 Not addressing the non-linearity in the attribution. 			
	 Not real atmospheric conditions. 			
	 Number of runs proportional to sources (j) and the 			
	number of levels of emission reduction ($\Delta E/E_{ini}$).			
	 Optimum ΔC_j/ΔE_j are model- and year-dependent 			
2) Decoupled direct method (DDM)			
2.1) DDM and HDDM	Advantages:			
 Instantaneous ΔC_{j,t}/ΔE_{j,t} 	• Source sensitivities ($\Delta C_{j,t} / \Delta E_{j,t}$) calculated on-line (1 run)			
 C_j = ΔC_j/ΔE_j x E_j 	 HDDM allows for second order derivatives 			
 DDM first-order sensitivity 	Disadvantages:			
 HDDM high-order sensitiviy 	 Not addressing the non-linearity in the attribution. 	\prec		
	Tagging method (TG)			
1) PM	Advantages:			
 Primary aerosol 	 Real atmospheric conditions (reactive tracers). 			
 Secondary inorganic aerosol 	 Source attribution (C_j) calculated on-line (1 run) 			
 Secondary organic 	 Addressing the non-linearity in the attribution. 			
2) O₃	 Computationally efficient, works in parallel with the 			
 O3_NOx_VOC_limiting 	CTM			
 O3_only_NOx_precursors 	 Mass preserved (renormalize if need). 			
 O3_only_VOC_precursors 	Disadvantages:			
• O3_equal_NOx_VOC	High memory and storage if the tagged source increase.			

- SS (BF or DDM) can be use for SA
 - $C_j = \Delta C_j / \Delta E_j \times E_j$
 - Assume linearity

- TG only provides SA
- Reactive tracers
 - Take into account nonlinearity in the attribution.

SSR model: algebraic relationships between gridded precursor emissions and concentrations linked by a series of unknown coefficients that are identified based on a limited series of full CTM simulations

O₃ tagging methods

Model	Algorithm	Tag pollutant	Ref	Obs
MOZART-4, CAM-chem	-	O3_only_nox_pr ecursors	Emmons et al. (2012)	Global . Tagging O3 produced from NO sources through updates to an existing chemical mechanism
Box model	-	O3_only_voc_pr ecursors	Butler et al. (2011)	Tagging O3 produced from VOC sources through updates to an existing chemical mechanism
ECHAM/MESS y 2.52	TAGGING 1.0	O3_equal_NOx_ VOC	Grewe et al. (2017)	Global . Competition between O3 precursors allowed. Large number of additional tracers
CAM-chem	TOAST 1.0	O3_equal_NOx_ VOC	Butlet et al. (2018)	Global. Competition between O3 precursors allowed. Large number of additional tracers
CMAQ	-	O3_equal_NOx_ VOC	Zhang et al., (2011)	Regional . Expand the SAPRC mechanism (significant increase of reactions e.g. from 224 to 1680)
CAMx	OSAT	O3_limiting	Dunker et al., 2008	Regional . Computationally efficient, works in parallel with the CTM
CMAQ > 4.7.1	ISAM	O3_limiting	Kwok et al., (2015)	Regional. Computationally efficient, works in parallel with the CTM

PM tagging methods

Model	Algorithm	Tag pollutant	Ref	Obs
CACM	SOEM	Secondary PM	Ying and Kleeman (2006)	Regional. Accurate but computationally prohibitive
CAMx	PSAT	SIA, EC, POM, IONS	Yarwood et al. (2004)	Regional . Computationally efficient, works in parallel with the CTM
CAMx-2008	PSAT	PM (SOA with VBS)	Skyllakou et al. (2014)	Regional.
LOTOS-EUROS	PSAT based	SIA, EC, POM, IONS	Kranenburg et al. (2013)	Regional. Lower troposphere (3.5 km asl)
CMAQ	TSSA	SIA, EC, POM, IONS	Wang et al. (2009)	Regional.
CMAQ > 4.7.1	ISAM	SIA, EC, POM, IONS	Kwok et al. (2013)	Regional.

What we can do with ISAM?

TAG CLASS	Species in EMISfile	Species in IC/BC, CGRID, DRYDEP, WETDEP
EC	PEC	AECI, AECJ
OC	POC, PNCOM	APOCI, APOCJ, APNCOMI, APNCOMJ
SULFATE	SO2, SULF, PSO4	S02, SULF, AS04I, AS04J, SULRXN
NITRATE	PNO3, NO2, NO, HONO	ANO3I, ANO3J, HNO3, NTR, NO2, NO, NO3, HONO, N2O5, PNA, PAN, PANX
AMMONIUM	NH3, PNH4	NH3, ANH4I, ANH4J
OZONE	PNO3, NO2, NO, HONO ALD2, ALDX, ETH, EHTA, ETOH FORM, IOLE, ISOP, MEOH, OLE PAR, TERP, TOL, XYL	O3N, O3V ANO3I, ANO3J,HNO3, NTR, NO2, NO, NO3, HONO, N2O5, PNA, PAN, PANX VOC
PM25_IONS	PCL, PNA, PMG, PK, PCA PFE, PAL, PSI, PTI, PMN PMOTHR	ACLI, ACLJ, ANAI, ANAJ, AMGJ, AKJ, ACAJ AFEJ, AALJ, ASIJ, ATIJ, AMNJ AOTHRI, AOTHRJ
СО	C0	со

Inputs: Tagged region* (mask) + tagged sector* (emission by sector) + total emissions **Configuration options:** (1) only regions (2) only sector (3) regions + sector

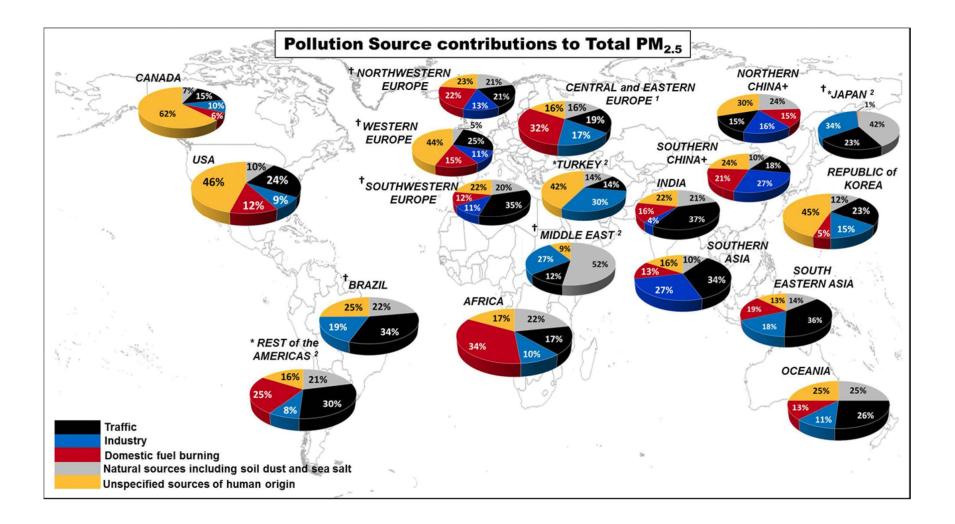
*optional

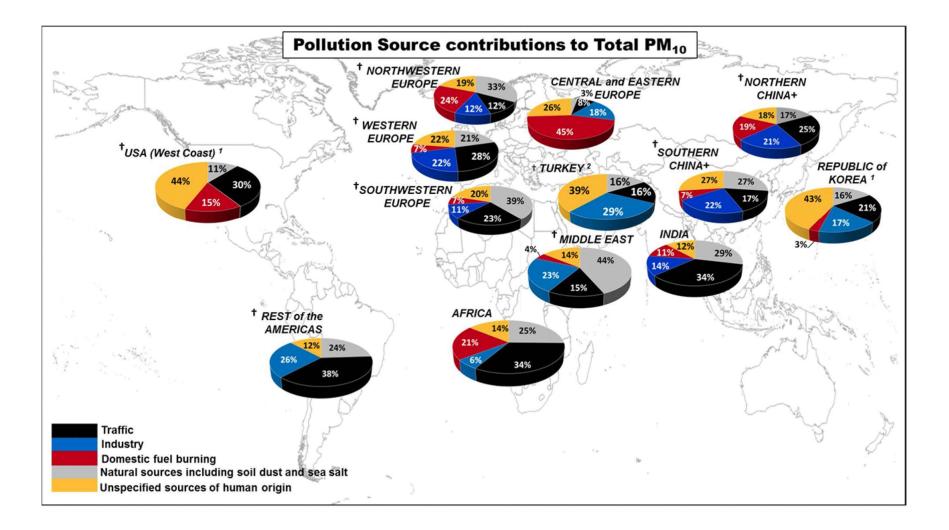
Tagged pollutants:	NO-Tagged pollutants:
• Fine aero.: EC, SO4, NO3, NH4, POA,	 Fine aero: SOA, dust*
IONS, CO	 Coarse aero: SO4, NO3, NH4, dust*,
• Ozone: O3 (O3N + O3V)	unspec**
	*dust = desert dust (transboundary) **unspec: asoil, acors, aseacat (¿urban+national?)

Review of receptor SA applications



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PM SA receptors: AIRUSE project

- Period: full year 2013
- **Sites**: 3 UB, 1 SB, 1 T.
- Measurements: OC, EC, anions, cations, major and trace elements and levoglucosan
- Model: USEPA PMF5

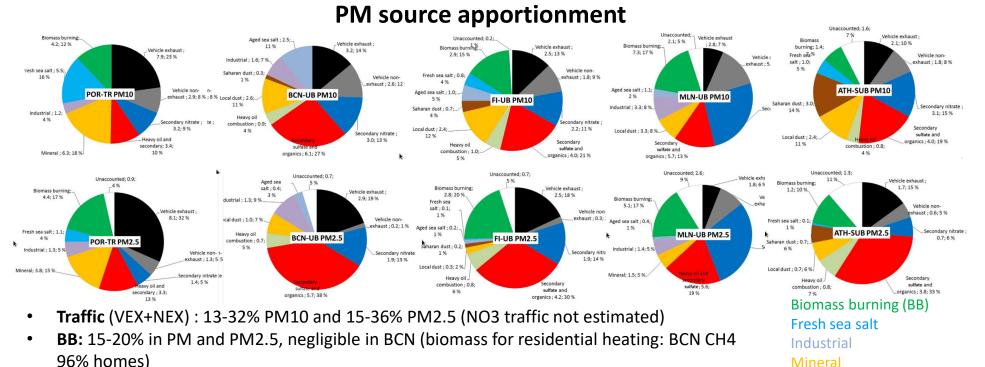


Heavy oil and secondary

Vehicle non-exhaust (VEX)

Vehicle exhaust. (NEX)

Secondary nitrate



- Industry (metallurgy) 4-11% PM10, 5-12% PM2.5 only in POR, BCN and MLN (no clear impact in the other cities)
- Natural contribution: SS 13%PM10 POR, DD 14% PM10 ATH.

Review of modelling SA applications



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Review (1/3): Regional sources

Global EU - Operational

			Regional sources		
N.	Study	Model/SAM/Domain of simulation/Resolution	Attributed sources/Receptor	Period	Attributed pollutants
1	Ikeda et al.(2017)	GEOS-Chem/TG/Global/2°x2.5°	Anthropogenic and BB in world regions/Arctic region	5 years (2007-2011)	BC
2	Zhang et al. (2017)	GEOS-Chem/BF/Global/100 km2	Consumption-production /World regions	1 year (2007)	PM2.5
3	Zhang et al. (2015)	CAM5/TG/Global/1.0°X1.0°	Anthropogenic and BB in world regions/Arctic region	1 year (2001)	BC
4	Fargeli et al., 2018 (EMEP SR matrices)	EMEP MSC-W/BF ⁶ /Europe (EU) and part of Asia and Africa/0.3°x 0.2°	EU countries /EU countries	1 year (2016)	PM2.5, EC Fine and coarse; O3
5	Lu et al. (2017)	NAQPMS/TG/Eastern and central China /27 km ² - 3 km2 ¹	Administrative regions/Wuhan region	1 day (12/10/2014) and 5 days (15 – 20/10/2014)	PM2.5
6	Skyllakou et al. (2014)	CAMx/TG/Europe/36 km2	Paris metropolitan regions ² (local, mid- range, long-range)/Paris Metropolitan, downwind areas	2 months (7/2009 and1-2/2010)	PM2.5_[EC, SO4, SOA, POA]
7	Local-external attribution CAMS	EMEP/BF ⁴ /EU/ 0.25°x0.125°	3 regions (local, rest EU, others / EU cities (38)	Operational ⁵	PM10, PM2.5, O3 ⁶
8	Country attribution CAMS	EMEP/BF ⁴ /EU/0.25°x0.125°	Country / EU cities (38)	Operational ⁵	PM10, PM2.5, O3 ⁶ , PM10_ [Dust, SS, SO4, NH4, NO3, POM, EC, SOA, PMw, FFire, Rest PPM]
9	Country attribution CAMS	LOTOS- EUROS/TG ⁷ /EU/0.25°x0.125	Country / EU cities (38)	Episodes ⁸	PM10
10	TOPAS	LOTOS-EUROS/TG/EU, part of Asia and North Africa/0.5°x0.25°	(1) Countries, (2) SNAP, (3) SNAP local and non-local ⁹ /EU cities (201) and EIONET stations	Operational ¹⁰	PM2.5, PM10

Review (2/3): Sectors

US EU EU regions

	Sectors						
N.	Study	Model/SAM/Domain of simulation/Resolution	Attributed sources/Receptor	Period	Attributed pollutant		
11	Paulot et al. (2014)	GEOS-Chem/BF/US/2°x2.5°	Agriculture for food exports/US	1 year (2015)	PM2.5		
12	Napelenok et al. (2014)	CMAQ/TG/US/36km-12km2	10 BB sources ¹³ /SE. US	1 year (2007)	Levoglucosan, PM2.5_[EC, OC]		
13	Karamchandani et al. (2017)	CAMx/TG/Europe and part of Africa/23 km2	SNAP/16 EU cities	2 months (8/2010 and 2/2010)	PM2.5, O3		
14	Thunis et al. (2018)	CHIMERE/Source Allocation ¹⁴ /EU and part of Asia/7km2	(1)SNAP ¹⁵ (2) cities/EU cities (150)	1 year (2010)	PM2.5		
15	Hendriks et al. (2013)	LOTOS-EUROS/TG/EU/28 km2 LOTOS-EUROS/TG/The Netherlands/7 km2	SNAP ¹¹ National and foreign/The Netherlands	3 years (2007- 2009)	PM2.5, PM2.5_[OC, EC, SS, SO4, NO3, MD], PM10, PM10_[same]		
16	Hendriks et al., (2016)	LOTOS-EUROS/TG/EU/0.5°x0.25°	Agriculture /N. western EU (focus Flanders)	5 years (2007 – 2011)	PM2.5, PM10, NH3, PM2.5_NH4,		
17	Pirovano et al. (2015)	CAMx/TG/Po Valley/5 km2	Agriculture, domestic heating, road transport ¹² /Po Valley region	1 year (2005)	PM2.5, PM2.5_[EC, OC]		
18	Bove et al. (2014)	CAMx/BF/Western and central EU/10km2 CAMx/BF/urban Genova/1km2	5 SNAP sectors ¹⁶ / urban Genova	4 months (06- 08/2011 and 15/11-15/12/2011)	PM2.5		

Review (3/3): Transportation

Aviation Shipping Traffic

		Transpor	tation (Shipping, aviation, on-road)		24
N.	Study	Model/SAM/Domain of simulation/Resolution	Attributed sources/Receptor	Period	Attributed pollutant
19	Yim et al. (2015)	GEOS-Chem/BF/global/4°x5° CMAQ/BF/NA/36 km2 CMAQ/BF/EU/40.5 km2 CMAQ/BF/Asia/50 km2	Aviation (full-flight (FF) and Landing+takeoff (LTO)/global, EU, NA, Asia, others	1 year (2006)	PM2.5 and O3 ¹⁸
20	Camaro et al. (2017)	GATOR-GCMOM/BF/Global/4°x5° GEOS-5/BF/Global/2.5°x2° NASA GISS/BF/Global/2.5°x2° CAM5/BF/Global/2.5°x2° GEOS-Chem/BF/Global/4°x5°	MOM/BF/Global/4°x5° BF/Global/2.5°x2° S/BF/Global/2.5°x2° Aviation (all altitudes + LTO)/ global surface air quality		PM2.5, O3
21	Vennam et al. (2017)	1) CMAQ/BF/N. Hemisphere (1) Aviation (FF)/HE Vennam et al. (HEMI)/108 km2 (1) Aviation (FF)/HE		1 year (2005)	O3, PM2.5, PM2.5_[SO4, NO3, NH4, EC, POA, SOA, crustal]
22	Bo et al. (2019)	CAMx/BF/East Asia/36km2	Aviation (LTO) ¹⁹ +airports/China	1 year (2016)	NOX, SO2, PM2.5
23	Monteiro et al. (2018)	CHIMERE/BF/EU/27 km2 CHIMERE/BF/Portugal/3 km2	Shipping/Portugal	1 year (2006)	PM ₁₀ , NO ₂ , O ₃
24	Aksoyoglu et al. (2016)	CAMx/BF/EU/19x13km2	Shipping, biogenic emissions/EU	1 year (2006)	O3, PM2.5, PM2.5_[EC, POA, secondary NO3, SO4, NH4, SOA], Deposition_[N, S, dry SO2, wet SO4]
25	Lv et al. (2018)	CMAQ/BF & TG/All China and parts of East Asia/36 km2	Shipping distances from coast/ E. China	1 year (2015)	PM2.5, PM2.5_[EC, POA, PSO4, Secondary SO4, NO3, NH4, SOA ¹⁷]
26	Valverde et al. (2016)	CALIOPE/TG/Iberian Peninsula/4 km2	Traffic /Madrid, Barcelona and areas downwind these cities	6 days (circulation types) in 2012	O3, NOX, VOC
27	Días-Robles et al. (2008)	CMAQ/BF/Southeastern US/36km2	Diesel fuel sources /SE. US cities ²¹	Episode: 13 days year (27/08- 09/08/1999)	PM2.5, PM2.5_[SO4,NO3,NH4,POA, SOA, EC, crustal and others]

Summary of SA review

- Type of **SA information**:
 - 2 operational services for O3 and PM (CAMS y TOPAS)
 - 23 research studies (12 BF, 11 TG)
- What is the **most popular SA method** for studying <u>EU international shipping</u> contribution?
 - BF is the most popular (both O3 and PM).
- What is the **most popular SA method** in <u>global/hemispheric studies</u>?
 - Mainly BF for PM, addressing BC and PM2.5 (Global TG studies on O3 not included yet).
 - Mainly addressing just one sector (BB, aviation, consumption/production in world regions)
- Which **period/year** is used for SA studies in EU?
 - Most popular year is 2010, no EU studies latter than 2011.
 - Only LOTOS-EUROS cover more than 1 year. Mainly 1 month summer/winter.
 - tagging & BF, mainly on PM, only 1 on O3 using tagging.
- Which type of **region allocation studies** have been done?
 - Country-to-country (EMEP, 2018), annual and based on BF-SR matrix.
 - Transboundary / country / city (core vs greater) or local.
 - World regions (aggregation of countries)

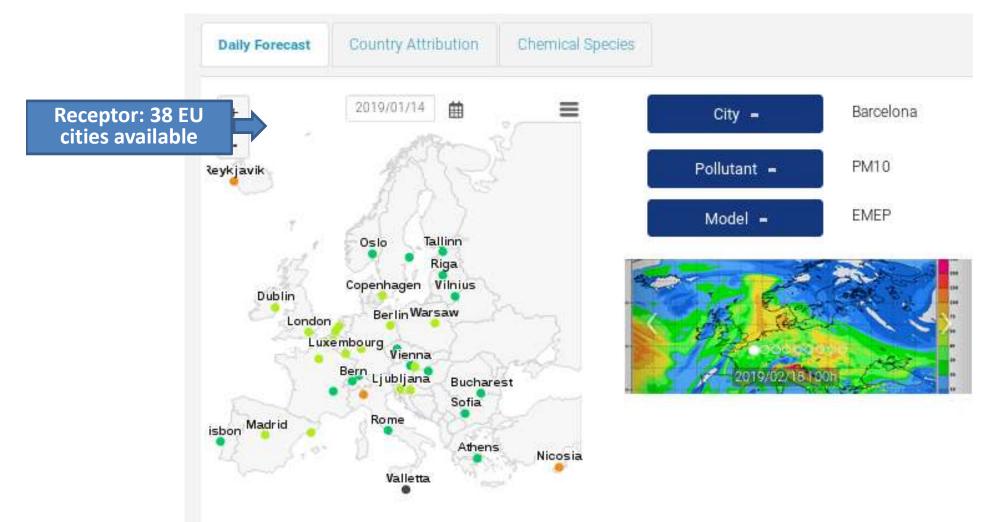
NOTE: SA studies on O_3 should be complemented with more studies.

SA services



		Regional sources		
Service	Model/SAM/Domain of simulation/Resolution	Attributed sources/Receptor	Period	Attributed pollutant
Local-external attribution CAMS	EMEP/BF ⁴ /EU/ 0.25°x0.125°	3 regions (local, rest EU, others / Main EU cities (38 total)	Operational⁵	PM10, PM2.5, O3 [€]
Country attribution CAMS	EMEP/BF ⁴ /EU/0.25°x0.125°	Country emissions, natural and others/Main EU cities (38)	Operational⁵	PM10, PM2.5, O3 ⁶ , PM10_ [Dust, SS, SO4, NH4, NO3, POM, EC, SOA, PMw, FFire, Rest PPM]
Country attribution CAMS	LOTOS- EUROS/TG ⁷ /EU/0.25°x0.125	Country emissions/ Main EU cities (38)	Episodes ⁸	PM10
TOPAS	LOTOS-EUROS/TG/EU, part of Asia and North Africa/0.5°x0.25°	 (1) Countries, (2) SNAP, (3) SNAP local and non- local emissions, natural emissions, others, rest⁹/Main EU cities (201) and EIONET stations 	Operational ¹⁰	PM2.5, PM10

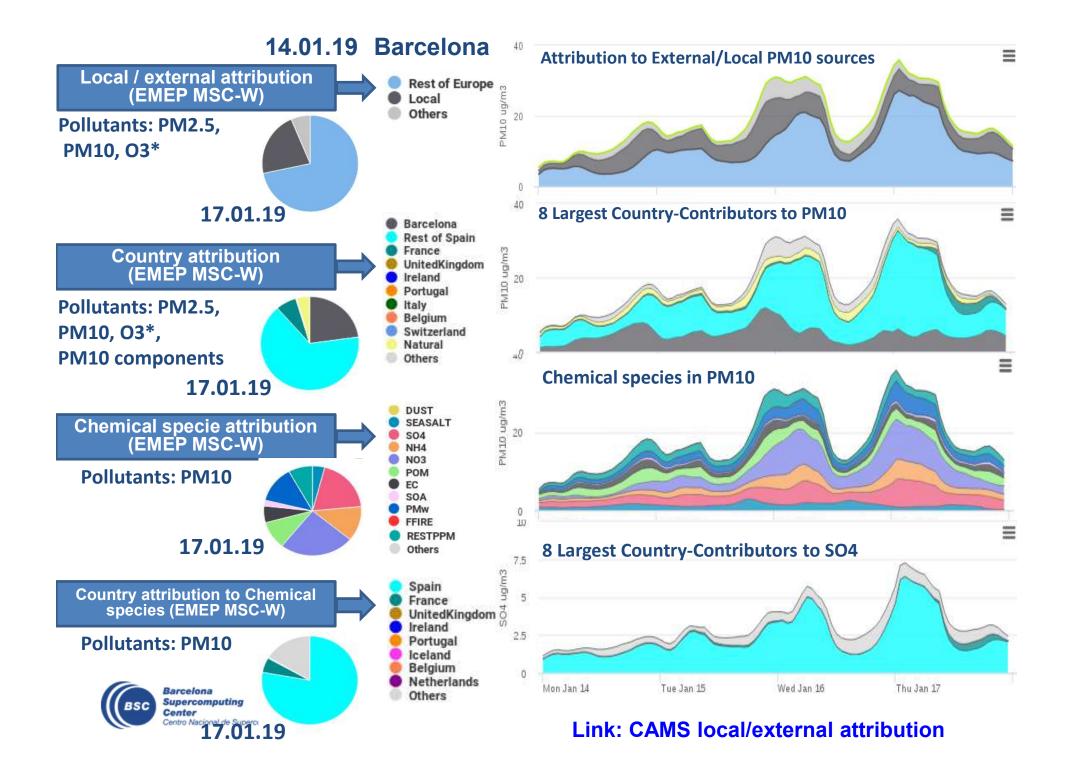
CAMS SA Services

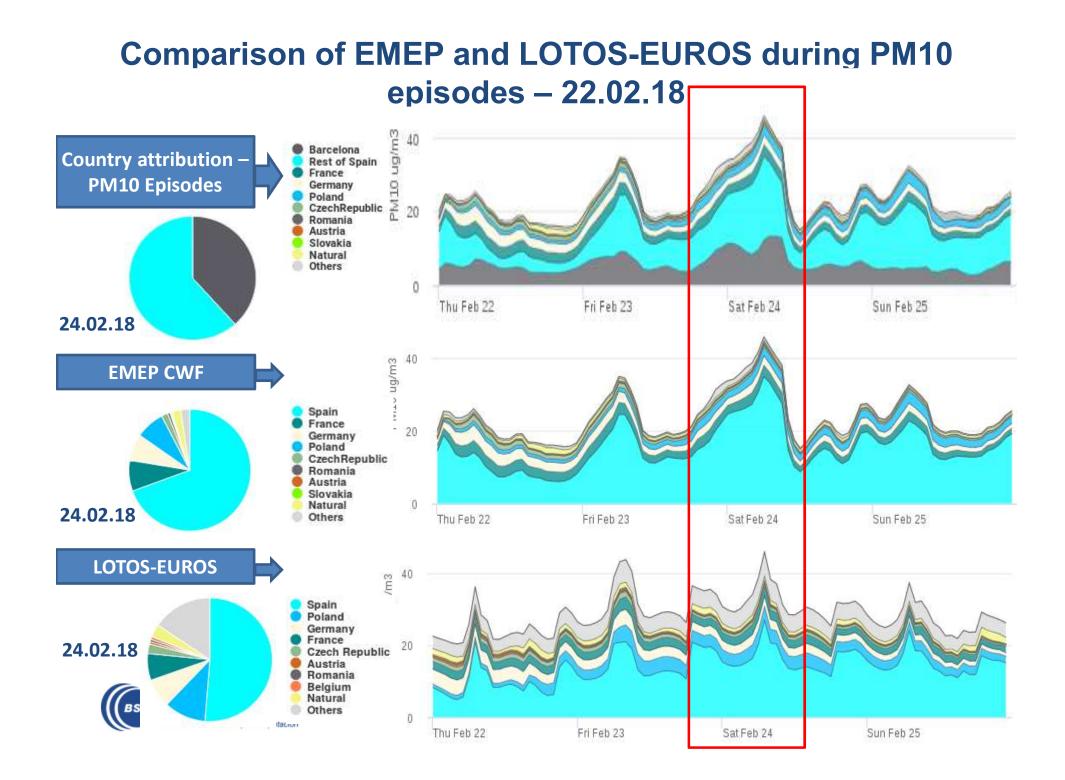


Link: CAMS local/external attribution

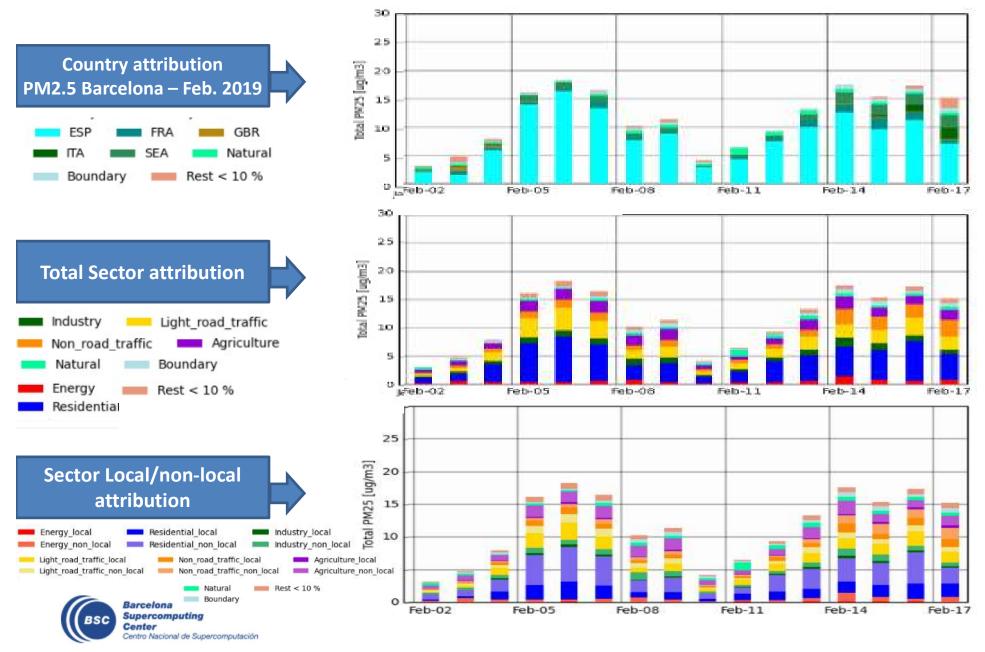


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TOPAS SA Services (LOTOS EUROS)



Limitation of SA services

Overall

- No research publication on the SA products.
- Emission database: TNO-MACC 2011
- Coarse horizontal resolution: 0.25° x 0.125° (CAMS) and 0.5° x 0.25° (TOPAS).
- Comprehensive O3 tagging not available.
- High uncertainty in PM modelling.

CAMS (EMEP)

- Calculates sensitivity of 15% perturbations extrapolated to 100% perturbation.
- Several precursors are perturbed simultaneously.
- Brute force is inaccurate for source apportionment of secondary pollutants (nonlinear) (E.g. negative contributions for O3 in some events).
- Use 9-grids as "optimum size" for EU capitals, due to the resolution (0.25° x 0.125°).

TOPAS (LOTOS-EUROS)

- Vertical resolution < 5 km
- Does not include secondary organic aerosols (SOA)
- Cannot tag O₃ nor SOA

Sector contribution to PM and O3



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Region

		Regio	ns	
Chudu	Model/SAM/Domain	Attributed	Period	Sector Contributions
Study	/Resolution	sources/Receptor	Period	PM2.5/PM10/O3
Thunis et al. (2018)	CHIMERE/Source Allocation ¹⁴ /EU and part of Asia/7km2	(1)SNAP ¹⁵ (2) regions (city core, greater city and country)/ EU cities (150)	1 year (2010)	PM2.5: City core: ~26% (up to 50% in Milan, Paris, Madrid) Greater city: ~31% (up to 60 in Paris, Madrid Athens, Turin, and Milan) National: ~56% (up to 80% in Milan, Warsaw, Brescia, Paris and Turin)
Skyllako u et al. (2014)	CAMx/TG/Europe/36 km²	Metropolitan Paris regions ⁵ (local, mid-range <mark>, long-</mark> range)/Metropolitan Paris, downwind areas	2 months (7/2009 and1- 2/2010)	PM2.5_EC: 64% local, 30% mid range, 6% long range PM2.5_SO4: 6% local, 23% mid range, 71% long range PM2.5_SOA: 7% local, 45% mid range, 48% long range
Kiesewe tter and Amman (2014)	GAINS and CHIMERE/BF/EU/7 km2	(1) SNAP (2) regions (transboundary, national, urban)/ main EU cities	1 year (2009)	PM2.5: Urban: up to 40% National: 20-40% N EU, 40-60% C EU, 60- 80% N Italy and N Spain. Transboundary: 0-20% S EU, 20-60% C and N EU

Selected Nomenclature for Air pollution (SNAP): agriculture, road and non-road transport, industries, energy production, Centro Nacional de Supercomputación waste treatments, residential emissions.



Agriculture

	<i>1</i>		Agriculture	
Study	Model/SAM/Domain/R	Attributed	Period	Sector Contributions
100000	esolution	sources/Receptor		PM2.5/PM10/03
Hendriks et al. (2013)	LOTOS- EUROS/TG/EU/28 km ² LOTOS-EUROS/TG/The Netherlands/7 km2	SNAP ¹¹ National and foreign /The Netherlands	3 years (2007- 2009)	PM2.5: Average: National: 7.5%, Foreign: 4.5% PM10: Average: National: 6%, Foreign: 4.5%
Hendriks et al., (2016)	LOTOS- EUROS/TG/EU/0.5°x0.2 5°	Agriculture /Flanders	5 years <mark>(</mark> 2007 – 2011)	РМ2.5_NH4: 47% (1.6 µg/m3) РМ10: 8.8%
Karamchandani et al. (2017)	CAMx/TG/EU and part of Africa/23 km2	SNAP/EU cities (16)	2 months (8/2010 and 2/2010)	PM2.5 (mean 2 months): Med: ≤9%; East: 12 – 16%; Central: ≤ 15%; Nordic: ≤ 12% O3 (mean 1 month): < 5% in the MDA8 all 16 cities
Pirovano et al. (2015)	CAMx/TG/Po Valley/5 km2	Agricultural, domestic heating, road transport ¹² /Po Valley region	1 year (2005)	PM2.5: 0.2 – 1.6 μg/m3
Thunis et al. (2018)	CHIMERE/Source Allocation ¹⁴ /EU and part of Asia/7km2	(1)SNAP ¹⁵ (2) cities/150 EU cities	1 year (2010)	PM2.5: Annual: ≤40% (Dresden 40%). Mean of 150 EU cities: 23%



Aviation

		Aviation		
Study	Mandal/CANA/Damain (Davalution	Attributed	Period	Sector Contributions
Study	Model/SAM/Domain /Resolution	sources/Receptor	Period	PM2.5/PM10/O3
Yim et al. (2015)	GEOS-Chem/BF/global/4°x5° CMAQ/BF/NA/36 km2 CMAQ/BF/EU/40.5 km2 CMAQ/BF/Asia/50 km2	Aviation (full-flight (FF) and Landing+takeoff (LTO)/global, EU, NA, Asia, others	1 year (2006)	PM2.5 (FF/LTO, ng/m3): Global: 6.2/0.5; EU: 9.0/1.2; NA: 18.2/4.8; Asia: 15.1/0.7; Others: 3.8/0.3 O3 (FF/LTO, ppb/ppt): Global: 0.6/10.7; EU: 1.1/25.4; NA: 1.0/29.8; Asia 0.9/12.5; Others: 0.5/8.8
Camaro et al. (2017)	GATOR-GCMOM/BF/Global/4°x5° GEOS-5/BF/Global/2.5°x2° NASA GISS/BF/Global/2.5°x2° CAM5/BF/Global/2.5°x2° GEOS-Chem/BF/Global/4°x5°	Global commercial aviation (all altitudes + LTO)/ global surface air quality	1 year (2006) ²⁰	PM2.5: Annual: -1.9% to 1.2% (CTMs: 0.14 to 0.40%, CRMs: -1.9 to 1.2%) O3: Annual: 0.3% to 1.9%
Vennam et al. (2017)	1) CMAQ/BF/N. Hemisphere (HEMI)/108 km2 2) CMAQ/BF/N. America (NA)/36 km2	(1Aviation (FF)/HEMI-NA, HEMI- EU, HEMI-EA (2) Aviation (FF)/NA	1 year (2005)	PM2.5: N.HEMI: 0.2% (0.013 μg/m3) ²⁴ , NA: 0.1% (0.002 μg/m3) O3: N. HEMI: 1.3% (0.46 ppbv), NA: 0.1% (0.03)
Bo et al. (2019)	CAMx/BF/East Asia/36km2	Aviation (LTO) ¹⁹ , airports/China	SA-1 year (2016) SS-1 year (2020)	PM2.5: Annual: 0.098% (< 0.033 μg/m3)



Shipping

Shipping				
Study	Model/SAM/Domain /Resolution	Attributed sources/Receptor	Period	Sector Contributions
				PM2.5/PM10/03
Monteiro et al. (2018)	CHIMERE/BF/EU/27 km2 CHIMERE/BF/Portugal/3 km2	Shipping/Portugal	1 year (2006)	PM10: Annual: EU: < 7 μg/m3), PT: 2 to 7 μg/m3 O3: EU: < 14 μg/m3, PT: < 9 μg/m3
Aksoyo <mark>glu</mark> et al. (2016)	CAMx/BF/EU/19x13km2	Shipping /EU	1 year (2006)	PM2.5: Annual: < 45% (< 3.5 μg/m3) O3: Annual: < 10% (-7 ²² to 6 ppb)
Lv et al. (2018)	CMAQ/BF & TG/All China and parts of East Asia/36 km2	Shipping at distances from the coast/ Eastern China	<mark>1</mark> year (2015)	PM2.5: Annual: <15% ²³ (< 5.2 μg/m3)
Bove et al. (2014)	CAMx/BF/Western and central EU/10km2 CAMx/BF/urban area Genova/1km2	5 SNAP sectors ¹⁶ / Urban Genova	3 months (06- 08/2011) (15/11- 15/12/2011)	PM2.5: Annual: 10%–15%



On-road transport

On-road transport					
Study	Model/SAM/Domain /Resolution	Attributed sources/Receptor	Period	Sector Contributions	
				PM2.5/PM10/O3	
Valverde et al. (2016)	CALIOPE/TG/Iberian Peninsula/4 km2	Madrid and Barcelona traffic/areas downwind (200km from cities)	6 days (circulation types) in 2012	O3 (daily maximum): Traffic Madrid up to 24%, Traffic Barcelona up to 8%	
Bove et al. (2014)	CAMx/BF/Western and central EU/10km2 CAMx/BF/urban area Genova/1km2	5 SNAP sectors ¹⁶ / urban Genova	3 months (06- 08/2011) (15/11- 15/12/2011)	PM2.5: 3 months average: 40–50%v	
Pirovano et al. (2015)	CAMx/TG/Po Valley/5 km2	Agricultural, domestic heating, road transport ¹² /Po Valley region	1 year (2005)	PM2.5: 3.9 to 10.7 μg/m328	
Días-Robles et al. (2008)	CMAQ/BF/SE. US/36km2	Diesel fuel sources/SE. US cities ²¹	Episode: 13 days (27/08- 09/08/1999)	PM2.5: Episode: 8% ²⁶ , EC: 78%, OA: 17% , NO3: 14%, SO4: 8%	
Hendriks et al. (2013)	LOTOS-EUROS/TG/EU/28 km ² LOTOS-EUROS/TG/The Netherlands/7 km2	SNAP ¹¹ National and foreign /The Netherlands	3 years (2007- 2009)	PM2.5: Annual: Dutch: 4.5%, Foreign: 5% PM10: Annual: Dutch: 3.5%, Foreign: 6%	
Karamchanda ni et al. (2017)	CAMx/TG/EU and part of Africa/23 km2	SNAP/EU cities (16)	2 months (8/2010 and 2/2010)	PM2.5 Med: 7–18%; East.: ≤18%; Central: 12–19% Nordic: 9–22% O3 Med: 15–24%; East.: 18–35%; Central: 11–19%; Nordic: 12–24%	
Thunis et al. (2018)	CHIMERE/Source Allocation ¹⁴ /EU and part of Asia/7km2	(1)SNAP ¹⁵ (2) cities/EU cities (150)	1 year (2010)	PM2.5: Annual: 33-48% in E EU, 20-25% N Italy, <10% rest EU (mean 13%)	



Biomass Burning (BB)

Biomass Burning					
Study	Model/SAM/Domain /Resolution	Attributed sources/Receptor	Period	Sector Contributions	
				PM2.5/PM10/03	
Hendriks et al. (2013)	LOTOS- EUROS/TG/EU/28 km ² LOTOS-EUROS/TG/The Netherlands/7 km2	SNAP ¹¹ National and foreign /The Netherlands	3 years (2007- 2009)	PM2.5: Annual: Dutch: < 1%, Foreign: 4% PM10: Annual: Dutch: <1%, Foreign: 3.5%	
Karamchandani et al. (2017)	CAMx/TG/EU and part of Africa/23 km2	SNAP/EU cities (16)	2 months (8/2010 and 2/2010)	PM2.5: Summer: <5% (Oslo 11%), Winter: 15-47% main cities in EU, outlier BCN 11% O3: Summer: < 5% in the H1MDA8	
Pirovano et al. (2015)	CAMx/TG/Po Valley/5 km2	Agricultural, domestic heating, road transport ¹² /Po Valley region	1 year (2005)	PM2.5: Summer (PM2.5/EC/OC): 0.8/0.2/0.3 μg/m3 Winter(PM2.5/EC/OC): 10.1/1.2/1.4 μg/m3	
Thunis et al. (2018)	CHIMERE/Source Allocation ¹⁴ /EU and part of Asia/7km2	(1)SNAP ¹⁵ (2) cities/EU cities (150)	1 year (2010)	PM2.5: Annual: 33-48% in E EU, 20-25% N Italy, <10 % rest EU (mean contrib. 13%)	
Bove et al. (2014)	CAMx/BF/Western and central EU/10km ² CAMx/BF/urban area Genova/1km ²	SNAP, others ¹⁶ / urban Genova	3 months (06- 08/2011) (15/11-15/12/2011)	PM2.5: Summer: 0.1-0.2 μg/m3 Winter: 1.5-1.6 μg/m3	



Combustion in energy production and industry

		Combustion in energy pro	oduction and industr	У	
Study	Model/SAM/Domain /Resolution	Attributed sources/Receptor	Period	Sector Contributions	
				PM2.5/PM10/O3	
Ka <mark>ramchandani</mark> et al. (2017)	CAM <mark>x/</mark> TG/EU and part of Africa/23 km2	SNAP/EU cities (16)	2 months (8/2010 and 2/2010)	PM2.5 (Energy) Med: ≤15%; East: 9–24%; Central: 6–15%; Nordio 6–12% O3 (Energy): Med: 5–8%; East: 9-17%; Central: ≤ 13%; Nordic: 13%	
Karamchandani et al. (2017)	CAMx/TG/EU and part of Africa/23 km2	SNAP/EU cities (16)	2 months (8/2010 and 2/2010)	PM2.5 (Industry) Med: ≤11%; East: 6–9%; Central: ≤10%; Nordic: ≤7% O3 (Industry): Med: 6–9%; East: ≤7%; Central: <5%; Nordic: ≤5%	
Hendriks et al. (2013)	LOTOS-EUROS/TG/EU/28 km ² LOTOS-EUROS/TG/The Netherlands/7 km2	SNAP ¹¹ National and foreign regions/The Netherlands	3 years (2007- 2009)	PM2.5 (Energy): Annual: Dutch: < 1%, Foreign: 5% PM10 (Energy): Annual: Dutch: <1 %, Foreign: 7%	
Hendriks et al. (2013)	LOTOS-EUROS/TG/EU/28 km ² LOTOS-EUROS/TG/The Netherlands/7 km2	SNAP ¹¹ National and foreign /The Netherlands	3 years (2007- 2009)	PM2.5 (Industry): Annual: Dutch: < 2.5%, Foreign: 5% PM10 (Industry): Annual: Dutch: <2 %, Foreign: 6.5%	
Thunis et al. (2018)	CHIMERE/Source Allocation ¹⁴ /EU and part of Asia/7km2	(1)SNAP ¹⁵ (2) cities/150 EU cities	1 year (2010)	PM2.5 (Energy + industry): Annual: ≤47% (M. Ludwigshafen 47%, Bilbao 46%). Mean of 150 EU cities: 20%	



Brainstorming



Barcelona Supercomputing Center Centro Nacional de Supercomputación

Country-to-country

- Rationale:
 - Long-life pollutants: O₃, SIA, NO2 and also for deposition.
 - Only EMEP model has done that for annual contribution using BF (SR matrices) for PM2.5, EC, O3.
- Research questions:
 - What is the <u>contribution each EU country to air pollution in other countries</u>? In what extend the air pollution problem in one EU country is due to emissions in other EU countries?
 - The same question but a combination of country + sector
 - Which are the <u>top country contributors</u> to air pollution problems in other countries
 - Which is the contribution from <u>non-EU countries</u>? ("the neighbours", e.g. N Africa)
 - Combination (region+sector)
 - NOTE: international <u>shipping and BCON</u> will be other sources.





Agriculture

- Rationale:
 - Significant source of NH₃ and precursor of PM.
 - <u>NH₃ emissions have increased</u> 3% from 2013 to 2016 in Europe and Spain (EEA, 2018; EMEP, 2018).
 - <u>40% of the EU land</u> area is destined to agriculture (EEA, 2017).
 - <u>Contribution</u>. PM2.5: 23% on annual average, main contribution in C EU. Among <u>main contributor to PM2.5 episode</u>s (winter and fall) in EU in 2015 and 2016 (EEA, 2017, 2018). O3: <5%.

Research questions:

- Country-to-country:
 - What is the contribution of agriculture in each EU country to PM in other countries?
 - Which are the top country contributors to air pollution problems in other countries?
- **Consumption/production:** e.g: Spain exports agricultural products to the rest of EU. What is the contribution of agricultural consumption of EU to PM2.5 emissions in Spain?





Aviation

• Rationale:



- Aviation is a growing sector (5% y⁻¹) and its contribution to AQ is not yet fully understood.
- AQ impacts (<u>takeoff and landing</u>, surface): increase in premature mortality (Yim et al., 2015; Keuken et al., 2015). Low contribution of <u>cruise level (8 12 km asl</u>)
- Contribution to PM2.5/O3: < 2%
- Limitations: domain hemispheric (more frequent flights)
- Research questions:
 - **Technique:** What is the contribution of aircrafts to air quality in EU, at surface and at cruise level using <u>tagging method</u>?
 - Activity: Which is the contribution of <u>commercial planes</u> (tourism) compared to <u>cargo planes</u> (international trade)?
 - **Destination:** What is the contribution of <u>international flights</u> in comparison to <u>EU originated flights</u>?



Shipping

- Rationale:
 - Significant source for NO₂, SO₂, O₃ and PM (SIA, EC).
 - Contribution: PM2.5 < 45% annual, O3 < 10% annual. Higher in summer than in winter, especially high coastal areas.
 - No many studies based on tagging in EU (only Karamchandani et al. (2017) but for the full SNAP8)
- Research questions:
 - Technique: What is the contribution of shipping to AQ in EU using <u>tagging</u> <u>method</u>?
 - Tourist effect:
 - What is the contribution from <u>cruise-ship</u> and <u>cargo-ship emissions</u>?
 - Is shipping contribution increasing in warm <u>months by tourism or higher</u> <u>photochemistry</u>?
 - **Consumption/Production**: where are the ships coming from? Which EU country is the largest polluted in terms of shipping demand?





On-road traffic

- Rationale:
 - Significant source of NO2, SO2, PPM.
 - Contribution: PM2.5 up to 50% (annual), O3 8-24% (episode) in the main Spanish cities.
 - No CTM studies on fuel type or exhaust/non-exhaust
 - No studies on the contribution of traffic in commuting zones.
- Research questions:
 - Fuel type: What is the contribution of diesel vehicles vs. gasoline vehicles? What about the modern diesel vehicles? (Platt et al., 2017)
 - Exhaust/non-exhaust: What is the contribution each process?
 - Vehicle type: motos vs. cars vs. high duty



OPEN Gasoline cars produce more carbonaceous particulate matter than modern filter-equipped diesel cars

5 October 2016 10 May 2017 online: 13 July 2017

S. M. Platt^{1,3}, I. El Haddad¹, S. M. Pieber³, A. A. Zardini³, R. Suarez-Bertoa³, M. Clairotte³, K. R. Daellenbach³, R. J. Huang^{1,5,6}, J. G. Slowik³, S. Hellebust^{1,13}, B. Temime-Roussel⁴, N. Marchand⁴, J. de Gouw^{5,6}, J. L. Jianez^{6,7}, P. L. Hayes¹, A. L. Robinson⁹, U. Baltensperger¹, C. Astorga³ & A. S. H. Prévůt¹

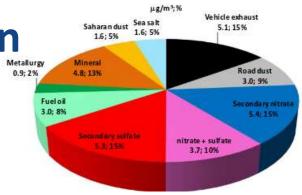




PAISA: PM contribution in Spain

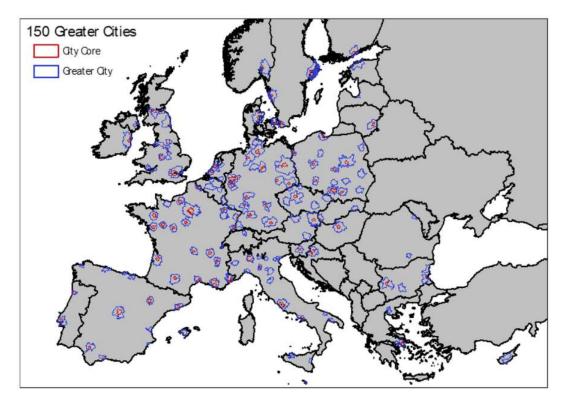
- Rationale:
 - High PM in episodes, different origin winter and summer.
 - SA RM (AIRUSE) in BCN (24% traffic, 40% SIA, 13% Mineral, SIA from traffic not accounted).
 - Modelling SA studies covering main Spain on annual basis and using brute force and top-down emissions (Kiesewetter and Amman, 2014; Thunis et al., 2018).
- Research questions:
 - **Regions:** what is the contribution of agriculture in each EU country to PM in other countries?
 - Imported: Sea + EU + BCON + Dust
 - Country: ES cities
 - Local: each city administrative area (6) \rightarrow NEW: use Thunis et al. (2018)
 - Sector:
 - Sectors: agriculture, energy+industry, residential, traffic,
 - Natural sources: SS + dust
 - Period: Which is the contribution of regions and sectors in episodes?





PM10 (2003-2014)

Definition of a city Thunis et al. (2018)

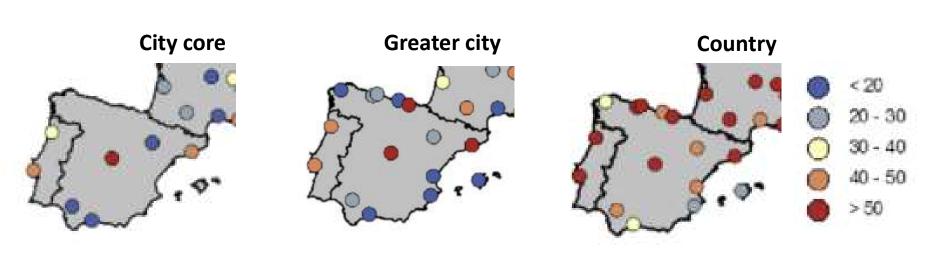


Core city: local administrative units, with population density > 1500/km2 and population > 50.000, where the majority of the population lives in an urban center (OECD, 2012)

Greater city: functional urban area (OECD, 2012). It consists of the core city + the wider communting zone, defined as the surrounding travel-to-work areas where at least 15& of the employed residents work in the city.

City contribution to PM2.5 in 2009

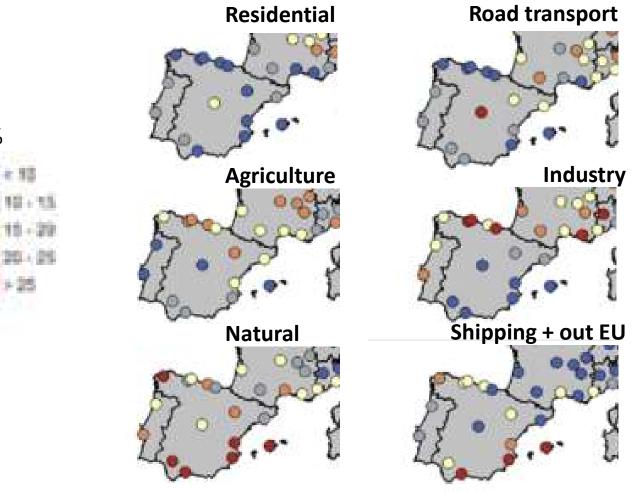
Thunis et al. (2018)



The commuting area important contribution in BCN

Source contribution to PM2.5

Thunis et al. (2018)





Region and sector contribution to PM2.5 in 2009

Kiesewetter and Amman (2014)

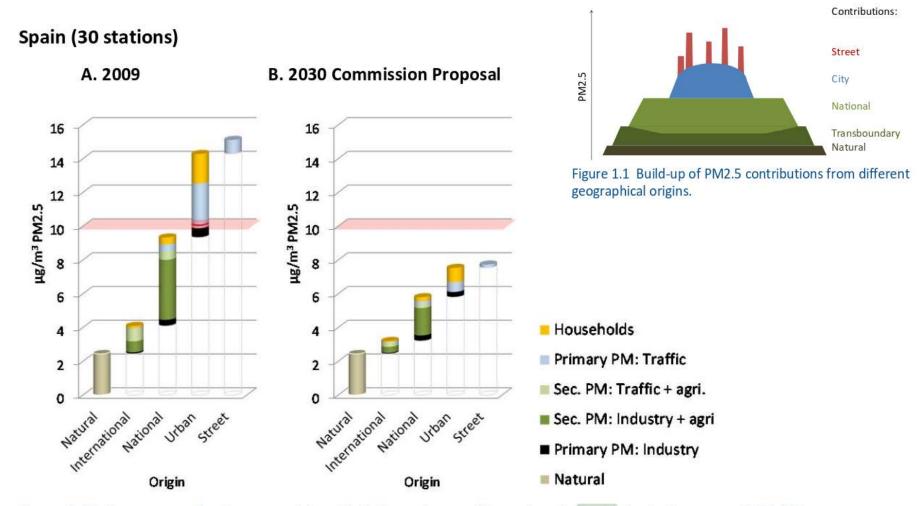


Figure 2.20. Source contributions to ambient PM2.5 at urban traffic stations in Spain, in the base year 2009 (A) and for 2030 assuming adoption of the Clean Air Policy Package proposed by the Commission (B). Source: IIASA GAINS.

Period of study

Pollutant	Period
03	 Summer (July) 2015 (most warmest Jul in records) Summer (July) from 2000-2015
PM2.5	 Winter (Feb) and Summer (Jul) 2015 Winter (Feb) and Summer (Jul) from 2000-2015 Last PM2.5 episodes in EU

- PAISA:
 - 1) Winter (Feb) and Summer (Jul) 2015





Barcelona Supercomputing Center Centro Nacional de Supercomputación



Thank you

YourEmail@bsc.es

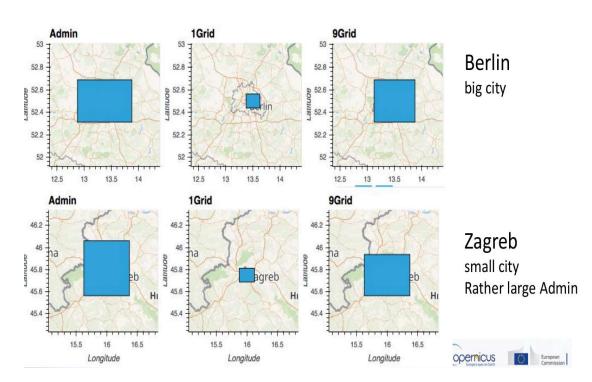
Supplementary material



Country to country: personalized grid size for city boundaries ?

CAMS/TNO uses 9-grid cells to estimate the contribution of local/external emissions to EU cities. While the approach may be suitable for some cities, it may be inaccurate for very small or very large ones.

Missing: To evaluate country attribution ideas using tailored grid-sizes for each main EU capital to estimate local/external contributions.





1) Country to country attribution – CAMS and TNO

Summary of CAMS / TNO Services

Source apportionment products / Projects	Model/ Approach	Source/ Receptor	Time res./ Frequency/ Period/ Mode	Pollutant				
Local - external source attribution/ CAMS	EMEP / Brute force ¹	Local, Rest EU, Others /Main EU cities (38 total)	Hourly/ Daily/ Nov. 2016 until today +3days/ Forecast	${\rm PM}_{10},{\rm PM}_{2.5},\\{\rm O}_3*$				
Country attribution / CAMS	EMEP / Brute force ¹	5 - 10 largest country contrib- utors /Main EU cities (38 total)	Hourly/ Daily/ Nov. 2016 until today +3days/ Forecast	$\begin{array}{c} \mathrm{PM}_{10},\\ \mathrm{PM}_{2.5}, \mathrm{O}_3^*, \mathrm{PM}_{10}\\ \mathrm{chemical \ species}^2 \end{array}$				
Country attribution/ CAMS	LOTOS-EUROS / Tags ³	5 - 10 largest country contrib- utors/Main EU cities (38 total)	Hourly/ A few times per year/ Episodes ⁴ / Analysis	PM_{10}				
Country attribution/ TOPAS	LOTOS-EUROS / Tags ⁵	5 - 10 largest country contrib- utors/Major cities (201 total) or sta- tions	Daily/ Daily/ Nov. 2016 until 43 days from yesterday/ Analysis	$PM_{10}, PM_{2.5}$				

EMEP report – Blame matrices

Model: EMEP MSC-W;

Method: Brute force (15% proportionally extrapolated to 100%).

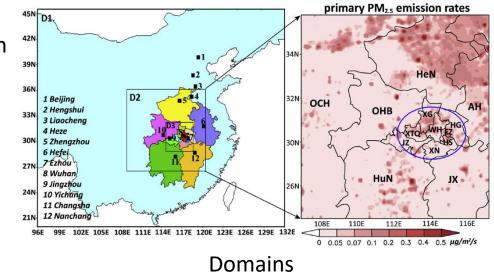
Pollutants: PM components: NH3, VOC, EC; OC; SOx, NOx, fine EC, Coarse EC; O3.

Table C.11: 2016 country-to-country blame matrices for PM2.5. Units: ng/m³ per 15% emis. red. of NH₃. Emitters →, Receptors ↓.

	AL	АМ	AT	AZ	ΒА	BE	BG	BY	СН	CY	cz	DE	DK	EE	E5	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KG	кz	LT	LU	LV	MD	
AL	69	-0	1	0	0	0	0	0	0	0	1	2	0	0	0	0	0	0	0	3	1	3	0	-0	4	0	-0	0	0	0	0	AL
AM	0	61	0	12	-0	-0	0	0	0	0	-0	0	-0	-0	0	-0	0	-0	2	-0	-0	0	-0	-0	0	0	-0	0	0	0	0	AM
AT	0		103	0	0	1	0	1	4	0	15	47	0	0	0	0	4	1	0	0	3	8	0	0	13	0	0	0	0	0	0	AT
AZ BA	-0 1	4	0	53 -0	-0 94	-0	-0 1	0	0	-0	-0 5	-0 7	-0	-0 0	-0 0	-0	-0	-0 0	4	-0 0	-0 18	-0 12	-	-0	0	0	-0	0	-0 0	-0 0	0	AZ
BE	0	-0	1	-0	0	185	-0	0	2	-0	2	92	2	0	1	0	66	36	-0	-0	10	0	3	0	2	-0	-0	0	7	0	0	BE
BG	1	0	3	0	1	0	97	1	0	0	2	4	0	0	0	0	0	0	0	6	2	7	0	-0	2	-0	-0	0	0	0	1	BG
BY	0	0	0	0	0	0	0	75	0	-0	2	11	1	0	0	0	2	1	0	0	0	2	0	0	0	0	0	4	0	1	1	BY
CH	-0	0	2	0	-0	1	-0	0	90	-0	1	27	0	0	1	0	15	1	0	-0	0	0	0	0	17	-0	-0	0	0	0	0	CH
CY	0	0	0	0	-0	-0	-0	-0	0	45	-0	0	-0	-0	0	-0	0	-0	0	0	0	0	-0	-0	0	-0	-0	-0	-0	-0	0	CY
CZ DE	0	0	20	-0	1	2	0	1	2	0	188	93 270	2	0	1	0	10 25	3	0	0	3	17	0	0	3	0	-0	1	0	0	0	CZ
DK	0	-0	1	-0	0	14	0	1	4	-0	2	73	92	0	1	0	25	12	-0	-0	0	1	3	0	0	-0	-0	1	0	0	0	DK
EE	0	0	0	0	0	0	0	5	0	-0	1	10	3	34	0	4	1	2	0	0	0	1	0	0	0	-0	0	4	0	5	0	EE
ES	0	0	0	-0	-0	0	-0	0	0	-0	0	1	0	0	79	0	5	1	-0	-0	0	0	0	-0	1	-0	-0	0	0	0	0	ES
FI	0	0	0	0	0	0	0	2	0	-0	0	4	1	1	0	15	0	0	0	0	0	0	0	-0	0	-0	0	1	0	0	0	FI
FR	0	0	1	0	-0	9	0	0	5	0	1	26	1	0	4	0	122	13	0	0	0	0	1	0	6	-0	-0	0	1	0	0	FR
GB GE	-0 -0	-0 2	0	-0	-0	-0	0	0	0	-0	-0	17	-0	0-0	2	0	19 -0	172	-0 26	-0	0	0	7	0	1	-0 -0	-0	0	0	0	0	GB GE
GL	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	0	-0	-0	0	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	-0	GL
GR	2	0	1	0	0	0	5	0	0	0	1	1	0	-0	0	0	0	0	0	54	0	2	0	-0	1	-0	-0	0	0	0	0	GR
HR	0	0	11	0	16	0	1	1	1	0	9	11	0	0	1	0	1	0	0	0	71	16	0	-0	26	-0	0	0	0	0	0	HR
HU	0	0	16	0	2	0	1	1	1	-0	17	22	1	0	1	0	2	1	0	0	10	117	0	0	8	0	0	1	0	0	0	HU
IE	-0	0	0	0	-0	3	-0	0	0	-0	0	8	1	0	0	0	8	43	0	-0	0		52	0	0	0	-0	0	0	0	0	IE
IS IT	-0 0	-0 0	0	-0	-0 0	0	-0 0	0	0	-0	0	1	0	0	0	-0 0	0	1	-0	-0 0	-0	-0	0	3	0 193	-0	-0	0	0	0	-0 0	IS IT
KG	-0	0	0	0	-0	-0	-0	-0	0	-0	-0	-0	-0	-0	-0	-0	-0	-0	0	-0	-0	-0		-0	0	18	2	-0	-0	-0	-0	KG
ΚZ	-0	0	0	0	-0	0	-0	0	0	-0	0	0	0	-0	0	0	0	0	0	-0	-0	0	0	-0	0	1	34	0	0	0	0	κz
LT	0	0	1	0	0	1	0	18	0	-0	3	19	4	1	0	1	2	2	0	0	0	2	0	0	1	-0	0	53	0	5	0	LT
LU	0	0	3	-0	0	47	0	0	2	0	- T.	137	1	0	1	0	58	17	0	0	0	0	2	0	2	0	-0	0	74	0	0	LU
LV MD	0	0	1	0	0	1	0	13	0	-0 0	2	14	3	4	0	1	1	2	0	0	0	1	0	0	0	-0	0	17	0	33 0	0 62	MD
ME	10	-0	2	-0	6	0	0	1	0	0	1	4	0	0	0	0	0	0	-0	0	3	4	0	-0	5	0	-0	0	0	0	02	ME
MK	10	0	2	0	1	0	3	1	0	0	2	4	0	0	0	0	0	0	0	14	2	6	0	-0	2	-0	-0	0	0	0	0	MK
MT	0	0	0	0	0	0	0	0	0	0	0	1	0	0	4	0	3	0	0	1	0	1	0	0	11	0	-0	0	0	0	0	MT
NL	-0	-0	1	-0	0	45	-0	0	1	-0	-	107	3	0	1	0	35	51	-0	-0	0	0	4	0	1	-0	-0	0	1	0	0	NL
NO	-0	-0	0	-0	-0	0	-0	0	0	-0	0	5	2	0	0	0	1	1	-0	-0	0	0	0	-0	0	-0	-0	0	0	0	0	NO
PL PT	0	0	4	0	-0	2	0	5	1	0	22	61 1	0	0	1 25	0	6	3	-0	0	1	10	0	0	1	0	-0	2	0	1	1	PL PT
RO	0	0	3	0	1	0	5	1	0	0	3	6	0	0	0	0	1	0	0	0	2	12	0	-0	2	0	0	0	0	0	2	RO
RS	2	0	6	0	5	0	5	1	1	0	6	10	0	0	0	0	1	0	0	1	7	19	0	-0	3	-0	-0	0	0	0	0	R5
RU	0	0	0	0	0	0	0	2	0	-0	0	1	0	0	0	0	0	0	0	0	0	0	0	-0	0	0	4	0	0	0	0	RU
SE	0	-0	0	-0	0	1	0	1	0	-0	0	13	6	0	0	1	1	2	-0	0	0	0		-0	0	-0	-0	1	0	0	0	SE
SI	0	0	28 12	0	1	0	0	1	1	-0	8 29	15 29	0	0	1	0	1	0	0	0	23	9 45	0	-0 0	64 4	0	0	0	0	0	0	SI SK
TJ	-0	0	-0	0	-0	-0	.0	-0	0	-0	-0	-0	-0	-0	-0	-0	-0	-0	0	-0	-0	-0		-0	-0	1	-0	-0	-0	-0	-0	TJ
TM	-0	0	-0	1	-0	-0	-0	0	0	-0	-0	-0	0	-0	-0	-0	-0	-0	0	-0	-0	-0		-0	-0	-0	0	0	-0	-0	0	TM
TR	0	0	0	0	0	0	1	0	0	0	0	0	0	-0	0	-0	0	0	0	0	0	0	0	-0	0	-0	-0	0	0	0	0	TR
UA	0	0	1	0	0	0	1	8	0	0	2	6	1	0	0	0	1	0	0	0	0	4	0	-0	1	0	0	1	0	0	3	UA
UZ	-0	0	-0	0	-0	-0	-0	0	0	-0	-0	-0	0	-0	-0	-0	-0	0	0	-0	-0	-0		-0	-0	2	2	0	-0	0	0	UZ
ATL BAS	-0 0	-0 0	0	-0	-0 0	0	-0 0	0	0	-0	0	1 49	0	0	2	-0	3	4	-0	-0 0	-0	-0	1	-0	0	-0 -0	-0	0	0	0		BAS
BAS	0	0	1	0	0	0	5	1	0	-0	0	49	1/	0	0	0	0	0	2	2	0	1		-0	1	-0	-0	4	0	0		BAS
MED	1	0	0	0	-0	0	0	0	0	-0	0	1	0	-0	4	-0	2	0	0	-0	0	0	0	-0	7	-0	-0	0	0	-0		MED
NOS	0	-0	0	-0	0	12	0	0	0	-0	1	42	10	0	1	0	26	65	-0	0	0	0	5	-0	0	-0	-0	0	0	0		NOS
AST	-0	0	0	1	-0	-0	-0	0	0	0	0	0	0	-0	-0	-0	-0	-0	0	-0	0	0	0	-0	0	0	0	0	-0	0		AST
NOA	0	0	0	0	-0	0	0	0	0	-0	0	0	-0	-0	2	-0	0	0	0	-0	-0	-0	0	-0	-1	-0	-0	-0	0	-0		NOA
EXC EU	0	0	1	0	0	1 5	1	2	1	0	2	10 40	1	0	3 11	0	5 21	4 15	0	0	1 2	2	2	0	4	-0	6 -0	1	0	0	0	EXC
20		AM	AT						CH		cz	DE										HU	IE			KG					MD	20

2) Lu et al., 2017 (Atm. Env.)

- Objectives: To identify major sources in central China and quantify their contribution to PM2.5 during haze episodes.
- Model: NAQPMS / Method: Tagging
- Domain/Receptor: Eastern and central China (27 x 27 km); Hubei province and nearby areas (9 x 9 km); Wuhan and nearby cities (3 x 3 km) / Wuhan.
- Time frame: two PM2.5 episodes (ep1: 12.10.2014; ep2: 15 20. 10.2014).



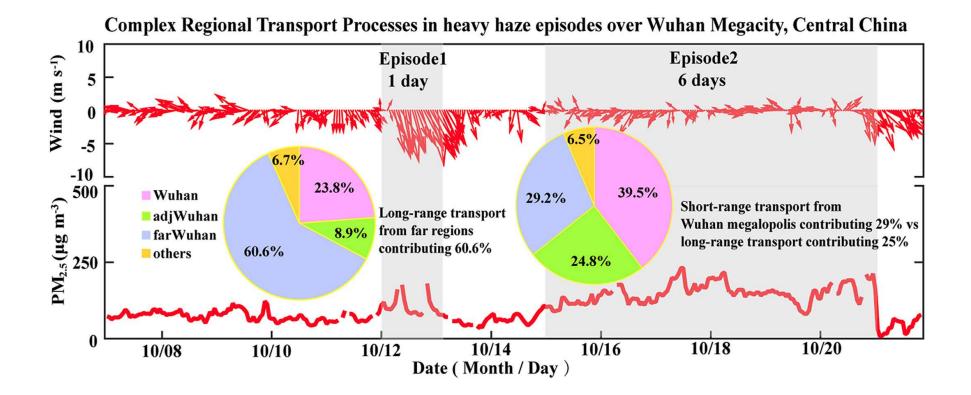
- Tagged species: PM2.5: Primary and secondary aerosols
- Tagged emissions: area sectors, lateral and top boundaries, ICON.

- Local (Wuhan); Adjwuhan (Ezhou (EZ); Huangshi (HS); Huanghang (HG), Xiaogan (XG), Jingzhou (JZ), Xianning (XN), Xiantao Tianmen -Qianjiang (XTQ); FarWuhan (western Hubei province (OHB), Henan (HeN), Anhui (AH), Jiangxi (JX) and Hunan (HuN)); Other areas of China (OCH); Other regions that are not China (OTH)).

2) Lu et al., 2017 (Atm. Env.)

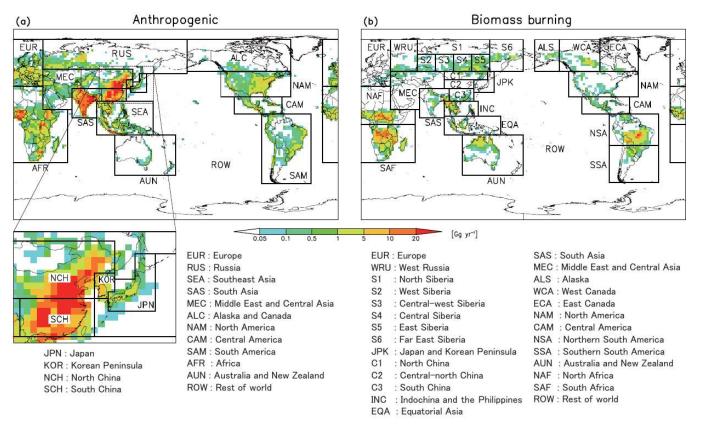
Main results and Conclusions:

- Meteorology played a large role in haze episodes (PM2.5) in Wuhan.
- Non-episode days: local contribution about 60%.
- Implications: AQ in Wuhan depends on cooperation with other cities.



3) Ikeda et al. 2017 (ACP)

- Objectives: Main source regions of the world and their contribution to BC concentrations in the Arctic region.
- Model: GEOS-Chem v. 9-02 / Method: tagging
- Domain/Receptor: global (2° x 2.5 °)/ Arctic region
- Time frame: 5 years (2007 2011)
- Tagged specie: Black carbon (BC)
- **Emission Tags:** Regions of the world and two activity sectors (anthropogenic and biomass burning including forest fires):



3) Ikeda et al. 2017 (ACP)

Main results and Conclusions:

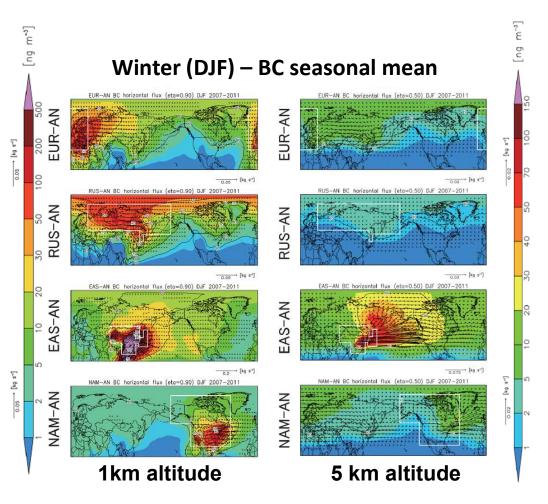
-Main contributors to BC conc. varied with altitude, most likely due to meteorological conditions.

-RUS (AN): 62% of annual mean at surface level (1 km) and 9.8% (5 km).

-Most BC emitted by EU and Russia reached the Arctic in winter and spring through the lower troposphere.

-BC from East Asian-travelled to the Arctic via middle troposphere in the same seasons.

-Up-lifting of air masses in Russia and EU was limited by cold winter temperatures (more stable conditions).

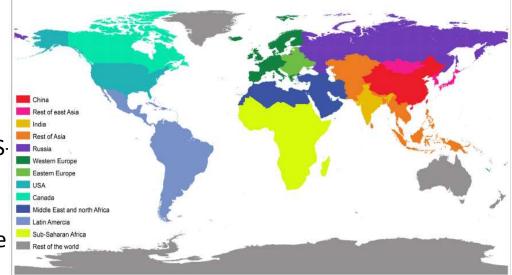


- East Asia was the most important contributor in terms of BC in the middle troposphere (41%), most likely due to uplifting of air masses, especially in early spring.

4) Zhang et al., 2017 (Nature)

 Objectives: Impacts that emissions from production and emissions embodied in consumption have on premature mortality regionally and worldwide.

- Models: Emission inventory; MRIO; GEOS-Chem; health impacts model / Method: Brute force
- Domain/receptor: global / regions of the world
- **Time frame:** 2007
- Pollutant: PM2.5
- Targeted Emissions: 13 Regions + production / consumption.



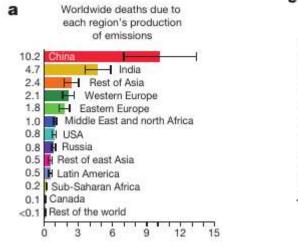
4) Zhang et al., 2017 (Nature)

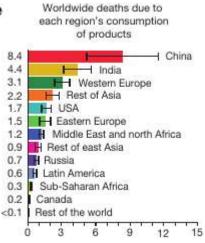
Main results and conclusion:

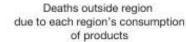
- Globally, the impact of PM2.5 on premature mortality was greater for international trade (22%, 762 400 deaths) than for long-range transported pollution (12%, about 411 100 deaths).

 The consumption in USA and Western Europe were linked to over 108.600 premature deaths in China as consequence of international trade.

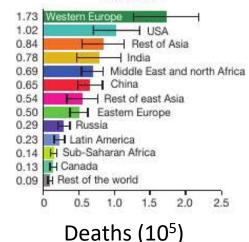
- The use of cleaner technologies by developing countries could potentially help improving air quality without disrupting the international trade system.







g

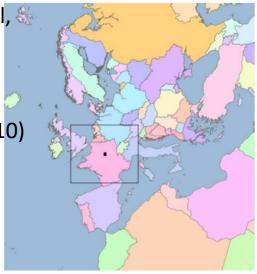


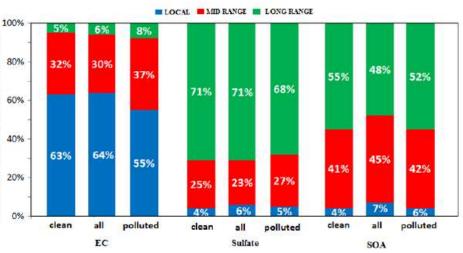
Deaths (10⁵)

6) Skyllakou et al., 2014 (ACP)

- Objectives: To identify main PM sources in Paris and estimate local, medium-range and long-range transport contributions.
- Model: PMCAMx-2008
- Method: tagging (PSAT)
- Time frame: summer (1-30.07.2009) and winter (10.01 09.02.2010)
- Domain: EU (36 x 36 km²)
- Receptor: Paris metropolitan area and downwind areas
- Tagged species: PM2.5 (EC, SO4, SOA)
- Tagged emissions: ICON; source areas at 50 km (local), 500 km (mid-range), > 500 km (long-range including BCON) from center of Paris metropolitan area.

 Main results and conclusions: Local contribution to PM2.5 during both winter and summer: 13%; mid-range contribution (<500 km): 36% and long-range contribution (> 500 km + BCON): 51%.





6) Skyllakou et al., 2014 (ACP)

Percentage

20%

10%

0%

Zero-out

PSAT

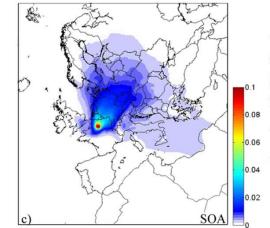
EC

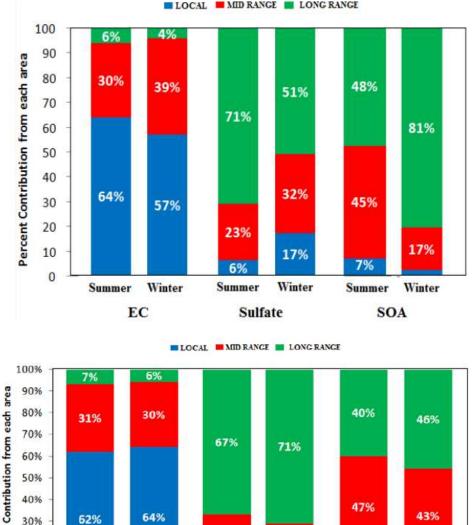
Local sources contributed to at least 50% of Primary PM components (EC, POA), while secondary PM (SO4, SOA) were mostly attributed to mid and long range transport.

- SOA plume originated by Paris reached as far as 800 km, however absolute contribution was negligible (< 0.1 μg/m3).

 Areas outside Paris (100 – 500 km) contributed to more than 45% of SOA mostly due to VOC emissions (followed by oxidation) in these

areas.





25%

8%

Zero-out

23%

6%

PSAT

Sulfate

13%

Zero-out

11%

PSAT

OA

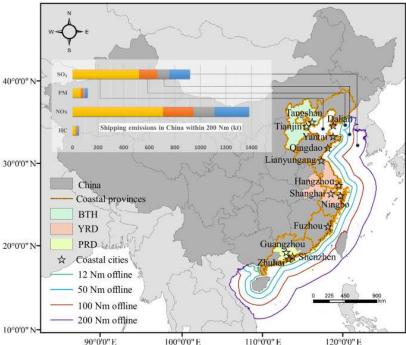
SA by activity sector: Shipping



Barcelona Supercomputing Center Centro Nacional de Supercomputación

7) Lv et al., 2018 (ACP)

- Objectives: To estimate the contribution of shipping emissions to PM2.5 concentrations on the eastern coast of China.
- Models: CMAQ v.5.0.1 and v.5.2 / Method: brute force + tagging
- Domain / receptor: All China and parts of Asia (36 x 36 km) / Eastern China.
- Time frame: 1 year (2015)
- Tagged species: PM2.5 (EC, POA, PSO4 + secondary SO4, NO3, NH4).
- Tagged emissions: shipping emissions within 0-12; 12-50; 50-100; 100-200 nautical miles from the shore.



Study area and contribution of different maritime areas for the total shipping emissions. The yellow, red, gray and blue columns represent the amount of shipping emissions in the areas within 12, 12–50, 50–100 and 100–200 Nm of the Chinese coastline, respectively.

7) Lv et al. 2018 (ACP)

Main results and conclusions:

-Contribution of 5.2 μ g/m3 to annual mean PM2.5 in eastern China, especially for the Yangtze River delta area.

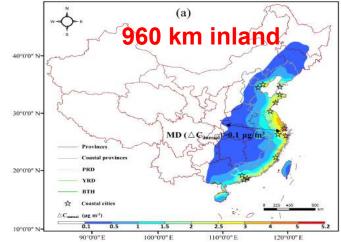
-Plume reached up to 960 km inland China (negligible increase, 0.1 μ g/m3).

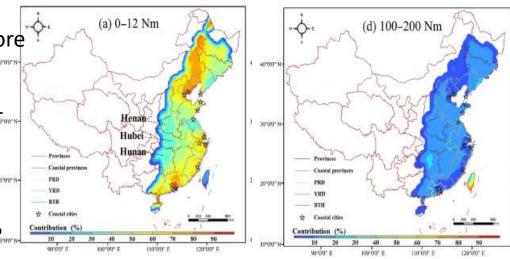
-Largest contribution for secondary pollutants: SO4, NO3 and NH4.

-Impacts higher in the summer for most cities of the central coast, when sea-to-shore winds prevailed.

-Sea-to-shore winds increased PM2.5 1.8 – 2.7 times in comparison to days of other wind directions.

- Ships within 12 Nm contributed 30 – 90% of PM2.5 in summer, especially in coastal areas.





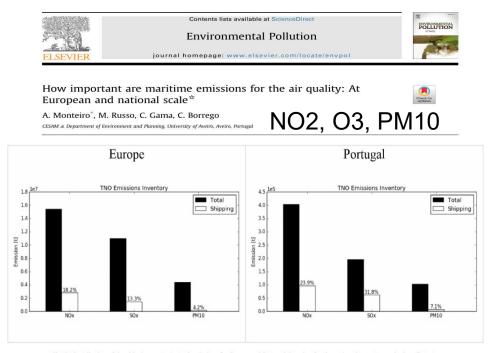
8) Monteiro et al. 2018 (Environ. Pollut.)

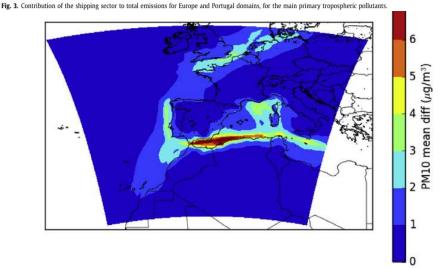
 Objectives: To estimate the contribution of shipping emissions to pollutant concentrations on regional (EU) and national levels (Portugal).

- Model: CHIMERE / Method: Brute force
- Domain: EU (27 km²); Iberian Peninsula (9 km²); Portugal (3 km²) / Receptor: EU and Portugal
- Time frame: 1 year (2006)
- Pollutant: PM10; NOx, O3, SOx
- Main conclusions:

-Contribution of shipping to total emissions in EU: 18,2 % for NOx, 13% for SOx and **4.1%** PM10; in Portugal: 24% NO2, 32% SOx and 7% PM10. **PM2.5 not included.**

-Mediterranean and North seas were the most affected by shipping contribution.

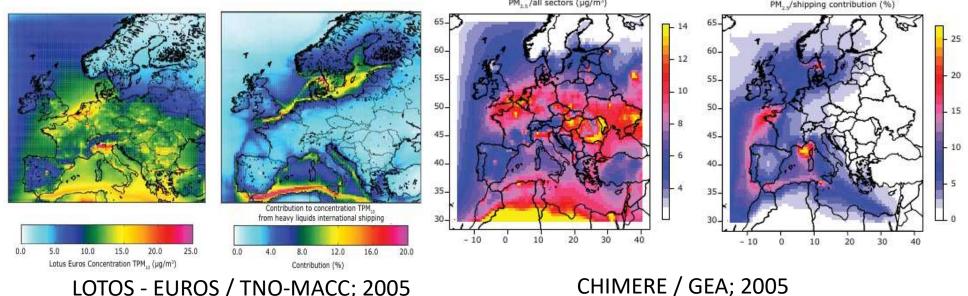




8) EEA technical report, 2013

- **EEA technical report (2013)**: The impact of international shipping on EU air quality and climate forcing.

- Literature review on mostly measurement-based studies but includes **LOTOS-EUROS** estimations for Heavy oil combustion from international shipping (4 – 8% contribution in EU for PM10), and CHIMERE simulation based on brute force. CHIMERE estimates annual contribution of PM2.5 of 5 - 10%. Monteiro et al. (2018) reported 4.1% contribution to PM10.



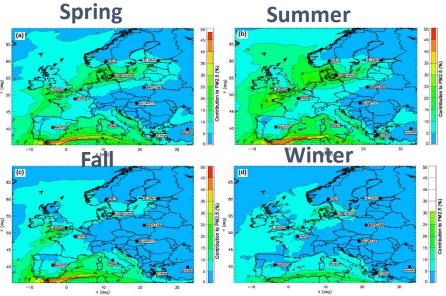
9) Aksoyoglu et al., 2016 (ACP)

- Objectives: To estimate shipping contribution to air quality in EU.
- Model: CAMx / Method: Brute force
- Domain: EU (19 x 13 km²) / Receptor: EU
- Time frame: 1 year (2006)
- Pollutants: O3, PM2.5 (EC, POA, NO3, NH4, SO4, SOA)
- Main results and conclusions:
- Highest contribution in summer.

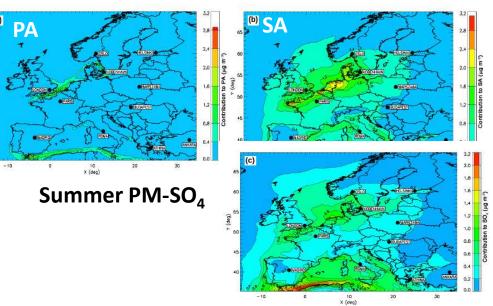
 Contribution in summer, up to 50% of annual PM2.5 in Mediterranean and 20 – 25% near English channel and North Sea.

- Winter contribution was lower: 5 - 10% in near PA English Channel and 15-20% in Mediterranean: -Highest contribution was for secondary aerosols. PM-SO₄ up to 60% increase in Mediterranean - summer.

-Ship NOx emissions combined with NH3 from inland activities in Benelux region, increased concentrations of NO3 and NH4 in the area due to NH4NO3 formation.



Contrib. ship emissions to PM2.5 (%)



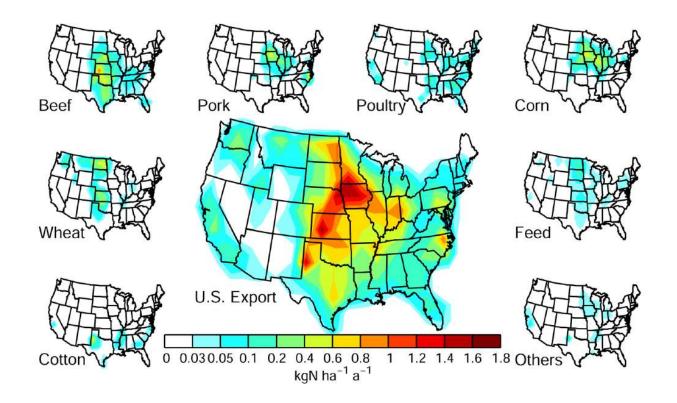
SA by sector activity: Agriculture



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10) Paulot et al., 2014 (Environ. Sci. Technol.)

- Objectives: To estimate NH3-emissions from food exports, and evaluate costs related to PM2.5 premature mortality.
- Models: GEOS-Chem, MASAGE / Method: brute force
- Domain / receptor: USA (2 ° x 2.5°) / USA
- Time frame: 1 year (2015)
- Pollutants: PM2.5 (NH4NO3, NH4SO4)

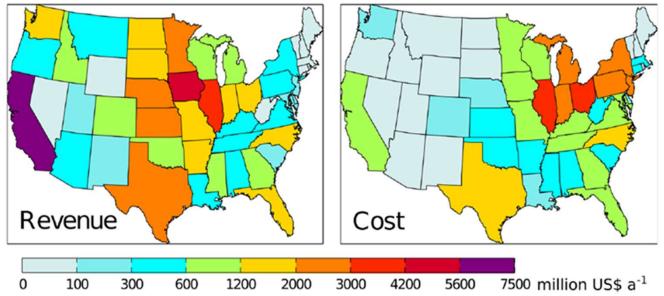


10) Paulot et al., 2014 (Environ. Sci. Technol.)

Main results and conclusions:

- 11% NH3 emissions in US is from food exports.

- PM2.5 related premature mortality caused by food exports is equivalent to 50% of gross food export value.



Comparison between annual gross revenue and health cost of agricultural solely driven by increased exposure to PM2.5 due to NH3 emissions from export for individual states. The health cost as computed here is agricultural export.

SA by sector activity: Aviation



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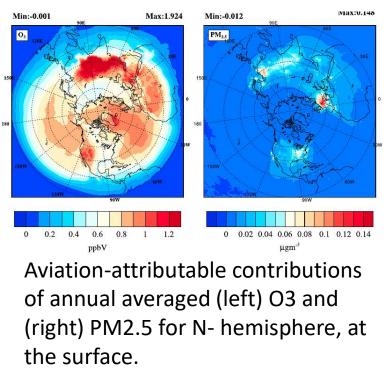
11) Vennam et al. 2017 (JGR)

 Objectives: To estimate contribution of aviation (full flight) to PM2.5 at surface and upper layers of the atmosphere (up to 12 km) on North-hemisphere and regional scale (USA, Asia and EU). To evaluate effects of increasing horizontal resolution.

- Model: CMAQ / Method: Brute force
- Domain: Northern Hemisphere (108 x 108 km²) and USA (36 x 36 km²) / Receptor: Asia, USA and EU
- Time frame: 1 year (2005)
- Pollutants: PM2.5 (SO4, NO3, NH4, EC, POA, SOA, crustal); O3
- Main results and conclusions:

- Contribution to PM2.5 annual mean highest at cruise level (3.6% in EU at 8 – 12 km altitude) and small at surface (max in EU: 0.5%).

- Coarse resolution caused an increase of 13% for PM2.5 and 70% for O3 in North America (108 x 108 km vs 36 x 36 km over USA).



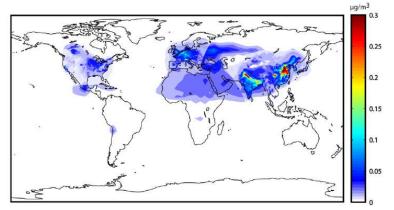
11) Yim et al., 2015 (Environ. Res. Lett.)

- Objectives: Contribution of aviation to surface
 PM2.5 and O3 conc. at global, regional and local.
 Implication of this contribution to premature
 mortality and health costs.
- Model: GEOS-Chem; CMAQ / Method: Brute force
- Domain: Global (GEOS-Chem: 4° x 5°), regional (CMAQ: EU (40.5 km), North America (36 km), Asia
 (50 km) / Receptor: global, EU, USA, Asia
- **Pollutant:** O3; PM2.5
- Time frame: 1 year (2006)
- Main results and conclusions:

-Full flight emissions increased PM2.5 at surface by 0.0062 μ g/m3 globally, 0.009 μ g/m3 in North America, 0.018 μ g/m3 in EU and 0.015 μ g/m3 in Asia.

-Globally, aviation was linked to an excess of 13 920 PM2.5-related deaths, 25% caused by take off and landing.

- Aviation emissions caused 5000 annual premature deaths globally (people living within 20 km), 38% in EU.



PM2.5 annual mean increase due to aviation emissions in 2006 (μg/m3). CMAQ + GEOS-Chems

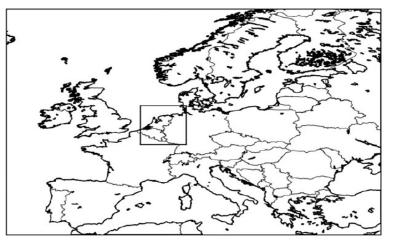
SA by sector activity: SNAP sectors or onroad transport



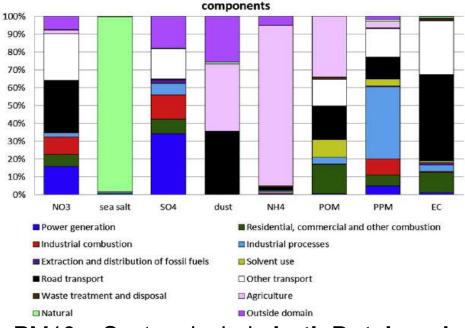
Barcelona Supercomputing Center Centro Nacional de Supercomputación

12) Hendriks et al., 2013 (Atmos. Environ.)

- **Objectives:** Main sources of PM in Netherlands, including episodes.
- Model: LOTOS-EUROS v1.8 / Method: Tagging
- Domain / receptor: EU (28 x 28 km), Netherlands (7 x 7 km) / Netherlands
- Time frame: 2007-2009
- Tagged species: Primary and secondary PM, except SOA; sea salt; mineral dust.
- Tagged emissions: Dutch / foreign emissions (rest of EU) + activity sectors (SNAP level 1) + natural emissions + boundary conditions (outside mother domain) + aloft conditions.



Contribution of emission sources to PM



PM10 – Sectors include **both Dutch and Foreign** contribution.

12) Hendriks et al., 2013 (Atmos. Environ.)

Dutch

0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9

Main Results and Conclusions:

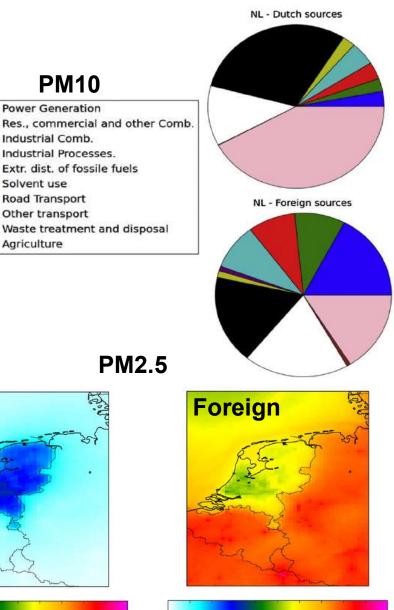
- 70 - 95 % of all PM observed in Netherlands were from anthropogenic origins.

-1/3 of PM were from domestic emissions, mainly agriculture and road transport. Sectors large contributors including in episodes.

-2/3 of the PM in Netherlands originated from foreign emissions (other EU countries).

-Foreign contrib. was larger for PM2.5 than for PM10 due to lifetime differences.

 Note on model uncertainty: Model underestimated both PM2.5 and PM10 by a 40%, possibly due to underestimations of N and OM, and unaccounted SOA.

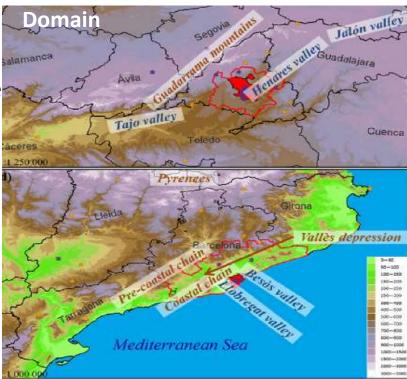


0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0

13) Valverde et al. 2016 (Sci. Total Environ.)

- Objectives: To determine contribution of on-road transport to O3 conc. in Barcelona and Madrid;
 To evaluate role of typical synoptic conditions on the transportation of on-road O3 plume in both cities.
- Model: CALIOPE / Method: Tagging (ISAM)
- Domain/Receptor: EU (12 x 12 km) and Iberian
 Peninsula (4 x 4 km) / Madrid and Barcelona metropolitan areas.
- **Time frame:** 1 day for each most common CTs in Iberian Peninsula (2012) (NW advection; Iberian thermal low; E/NE adv.; Atlantic high; W/SW adv.; Zonal Western adv.)
- Tagged species: O3, NOX, NMVOC
- Tagged emissions: on-road transport sector;

metropolitan areas of Barcelona and Madrid (O3T-MAD and O3 T-BCN); BCON to Iberian Peninsula (O3-BCON); other sectors inside and outside metropolitan areas + on-road outside metropolitan areas (O3-OTHER); ICON (O3-ICON).



13) Valverde et al. 2016 (Sci. Total Environ.)

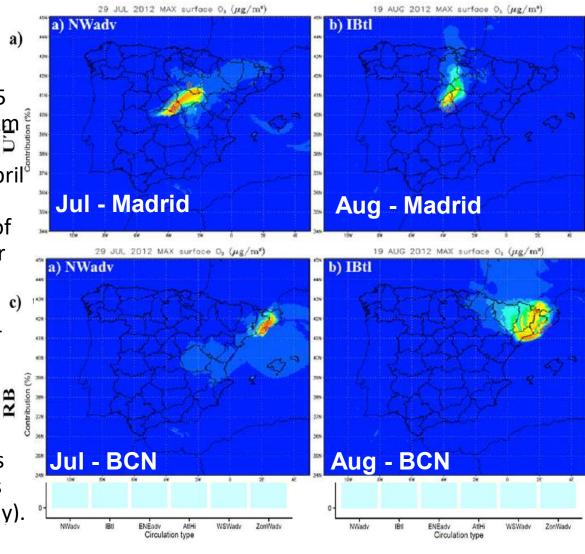
Main results and conclusions:

 On-road transport emissions was minor contributor to the total O3 conc. in both cities (MAD: 0.2 – 23.5 %; BCN: 0.5 -7.7 %), within an 200 km area. On road contribution was highest around midday and from April – Sept.

-MAD plume influenced large area of IP, while BCN plume influenced over Mediterranean Sea.

Main contributor to O3 conc. (34 – 96 %) was boundaries of Iberian Peninsula (O3-BCON).

- Contribution of imported O3 was higher during cold circulation types than during warm circulation types (70 - 96% and 35 - 70 % respectively).



O3T-BCN O3T-MAD O3-OTHER O3-ICON O3-BCON

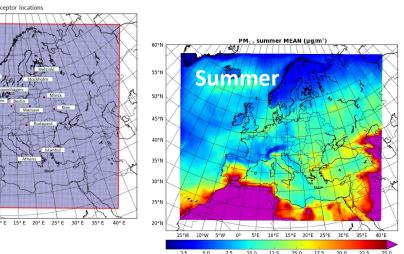
14) Karamchandani et al., 2017 (ACP)

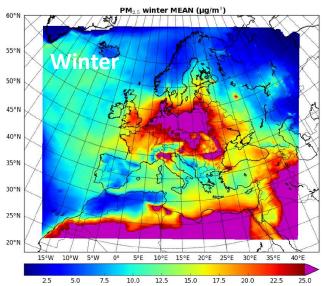
- Objectives: To estimatimate sector contributions to PM2.5 and O3 conc. in tell cities.
- Model: CAMx
- Method: tagging (PSAT/OSAT)
- Time frame: 1 year (2010). Tagging: August 2010; February 2010
- Domain: EU (23 km²)
- Receptor: 16 EU cities
- Tagged species: PM2.5 + O3
- Tagged emissions: source sectors (SNAP), biogenic emissions, sea salt, BCON + ICON

Main resuls and conclusions:

-PM2.5 conc. And source contributions varied greatly among
 EU cities between winter and summer months. Warsaw:
 summer: 13 μg/m3, winter: 38 μg/m3
 -BCON was the main contributor for
 PM2.5 in Mediterranean cities in summer

(Barcelona, Lisbon, Athens and Istanbul, 38 – 49%). Likely Saharan dust. The second highest contrib., for Barcelona and Lisbon was non-on road emissions (e.g. shipping)



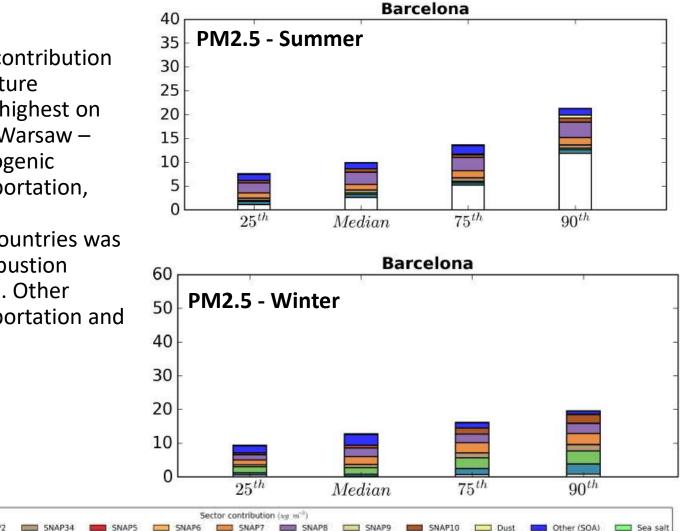


14) Karamchandani et al., 2017 (ACP)

-Summer: on-road vehicle contribution less than 5% – 13%. Agriculture contributed to < 5% - 14% (highest on Minsk, Budapest, Kiev and Warsaw – Eastern EU). Main anthropogenic contributors: energy, transportation, industry and agriculture.

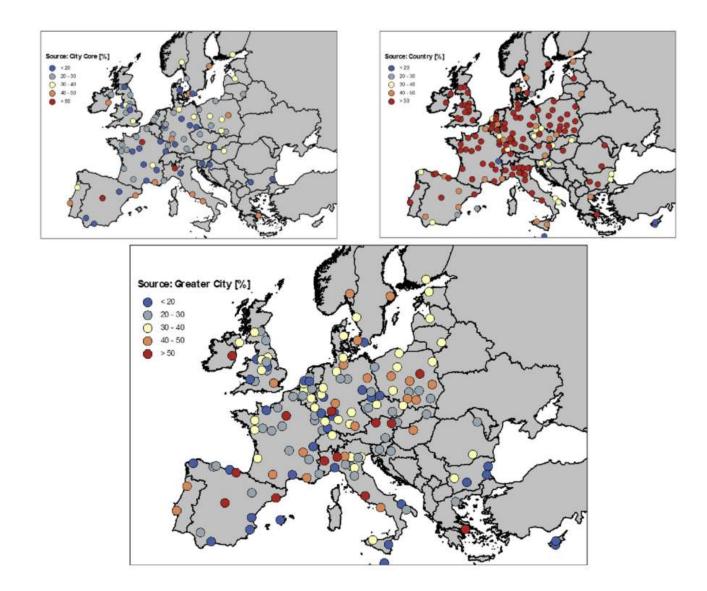
-Winter: PM.5 in most EU countries was dominated residential combustion (SNAP2) sources (11 – 47%). Other contributors: energy, transportation and agriculture.

IC/BC

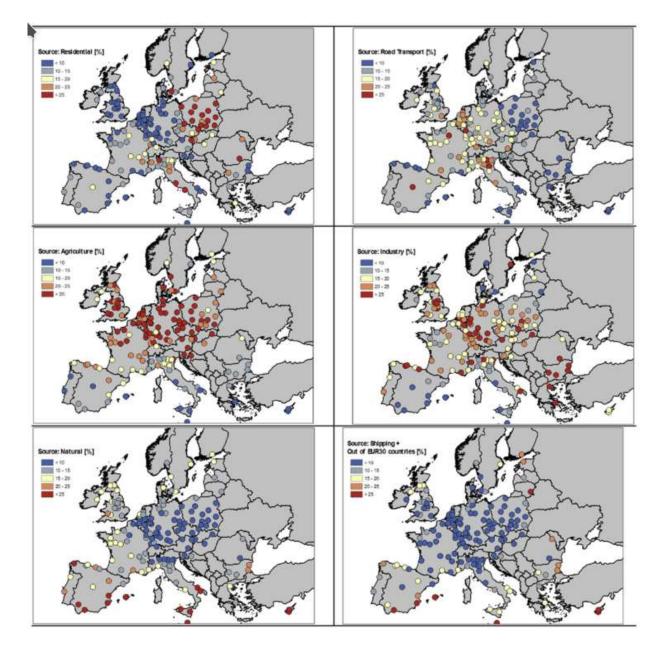


1-Energy industries; 2-Non-industrial (residential) combustion 34-Industry*; 5-Extraction and distribution of fossil fuels; 6-Solvent and other product use; 7-Road transport (includes exhaust, evaporative, tire–brake–road wear); 8-Non-road transport; 9-Waste treatment; 10-Agriculture

Thunis et al. (2018)



Thunis et al. (2018)



Kiesewetter and Amman (2014)

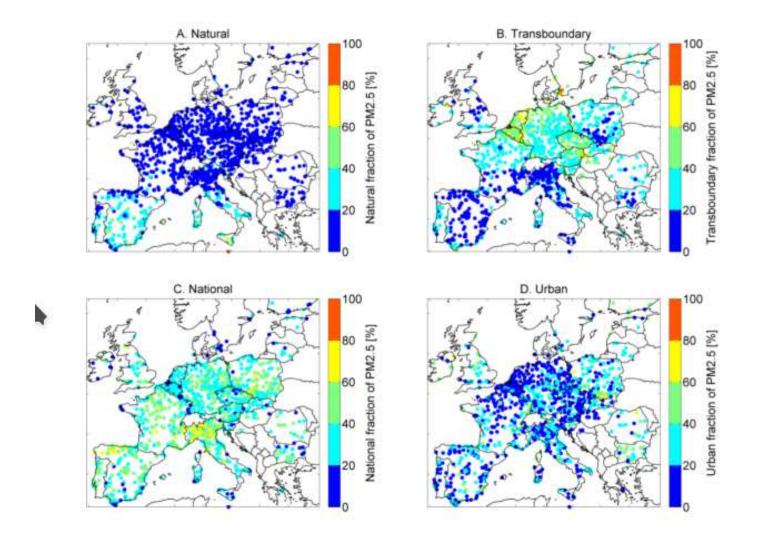


Figure 2.1. Spatial origins of PM2.5 at background monitoring stations covered by GAINS, shown as relative fractions of total modelled PM2.5 at each station in 2009.