Light-absorbing aerosol sources and brown carbon aging in northern France

Presented by:

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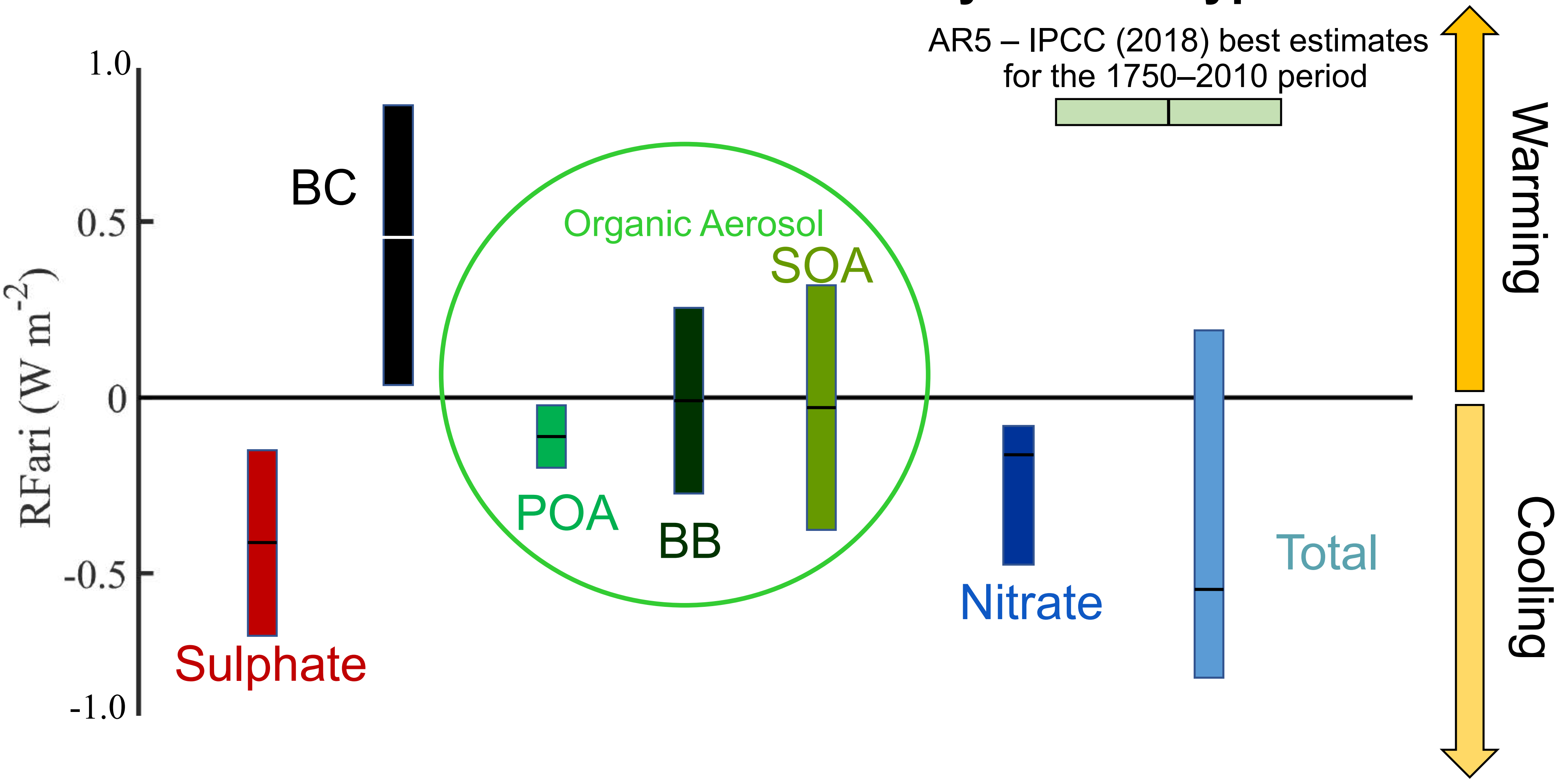
Aerosol radiation interactions by aerosol types



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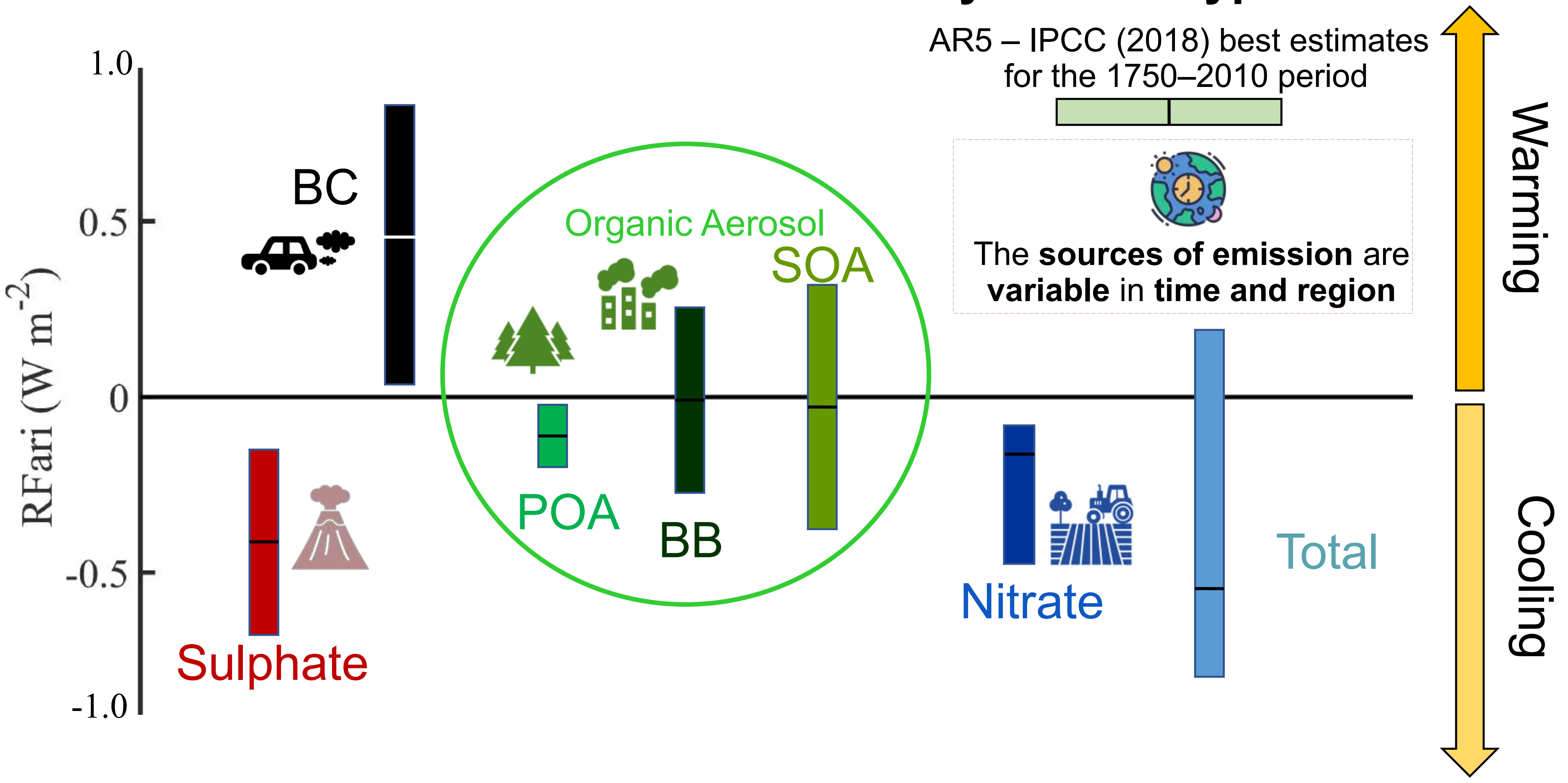

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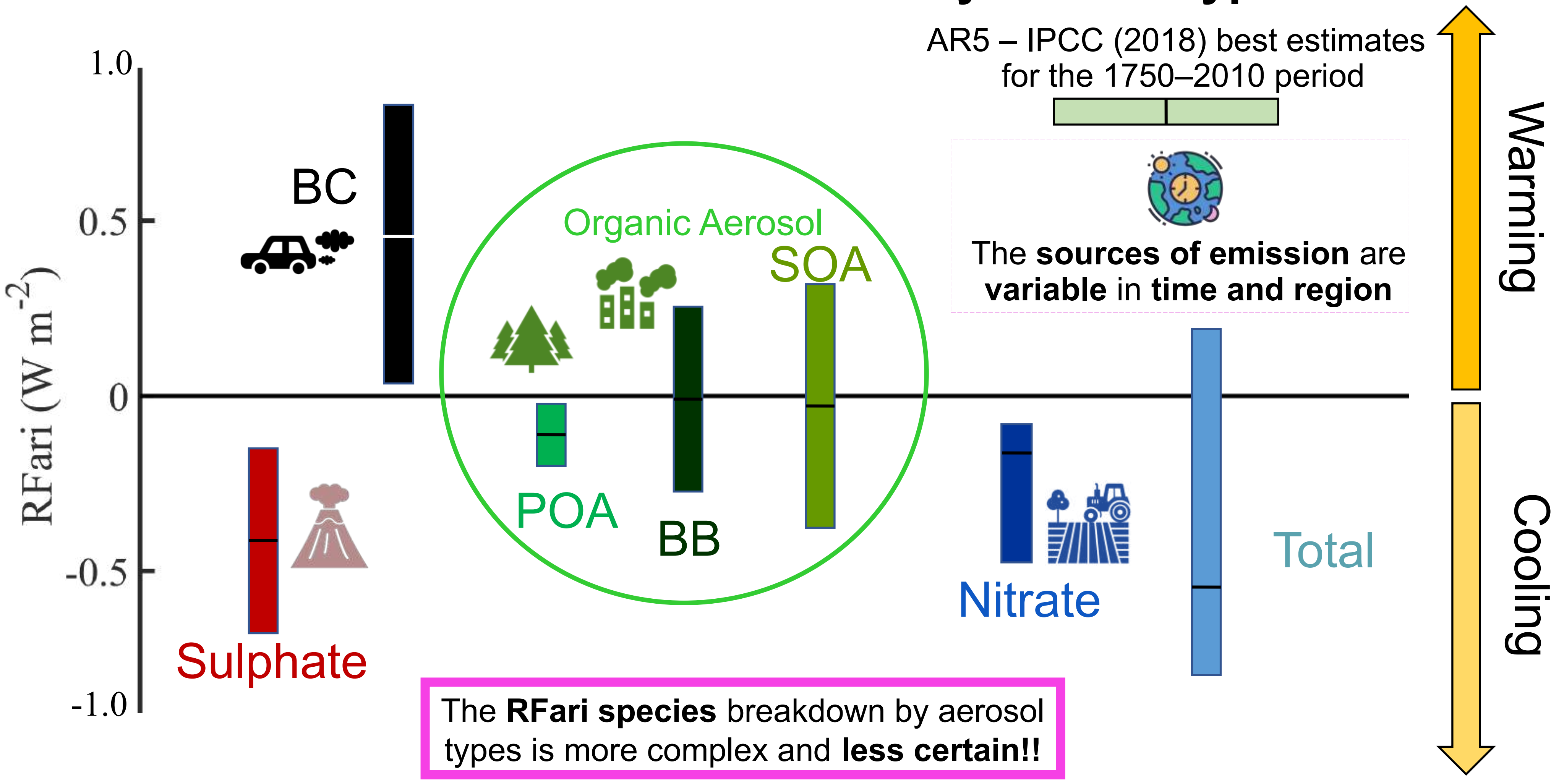


Aerosol radiation interactions by aerosol types

AR5 – IPCC (2018) best estimates

RFari (W m^-2): 1.0, 0.5, 0, -0.5, -1.0

Warming

Cooling

Observations are needed to better understand the RFari by each species

Optical properties ⟷ **Link** ⟷ **Chemical composition**

Need of long-term coincident observations in different regions

The **RFari species** breakdown by aerosol types is more complex and **less certain!!**

ATOLL platform

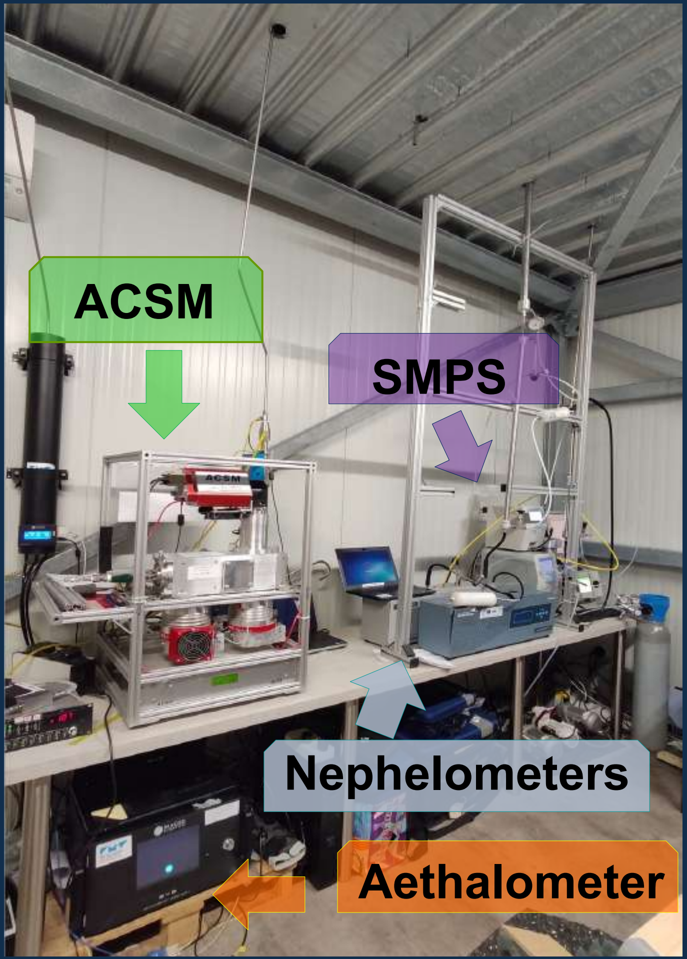
Long-term observations:
chemical & optical properties

ATOLL (ATmospheric Observation in liLLe)
50.6111 °N, 3.1404 °E, 70 m a.s.l.

The **North of France** suffers from significant **particulate pollution events**

- Heavily **populated area**
- High influence of **anthropogenic sources**
- Frequently polluted **continental plumes** arriving to the site

Instrumentation



Instrumentation

Chemical composition

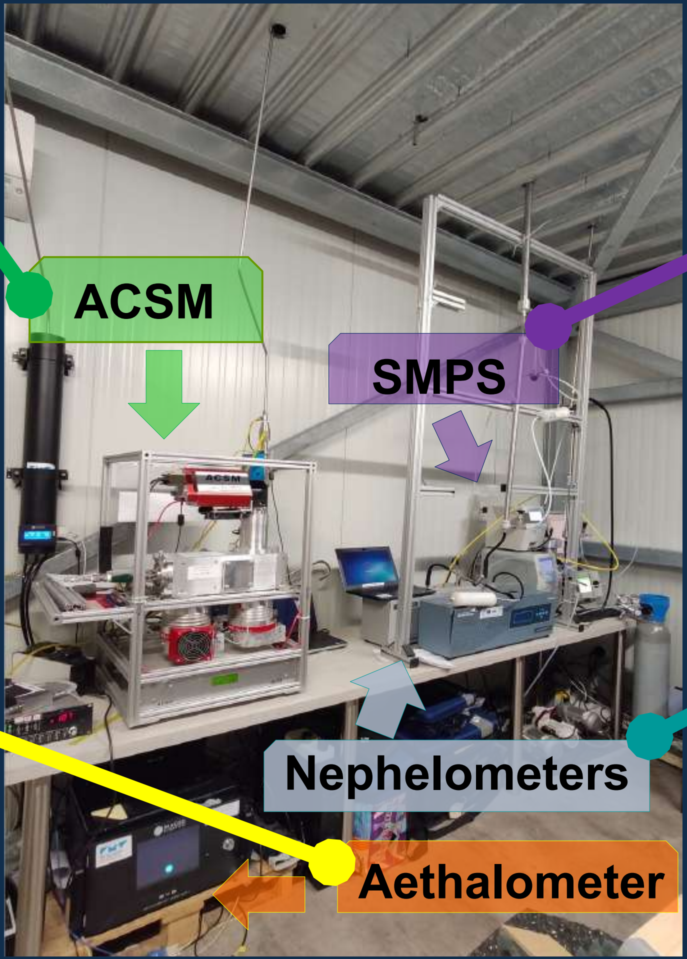
NR-PM_1: Org, NO_3, SO_4, NH_4, Chl

Aerodyne Research Inc.
~30 min

Absorption – PM_1

σ_{abs} & Black Carbon (BC)

AE33, Aerosol d.o.o.
7w (370 – 950 nm)
1 min



Size distribution

15nm – 1µm

SMPS 3936,TSI
5 min

Scattering – PM_1

σ_{scat}

AURORA 3000

450nm, 525 nm, 635nm

1 min

Light-absorbing aerosol sources

Aethalometer

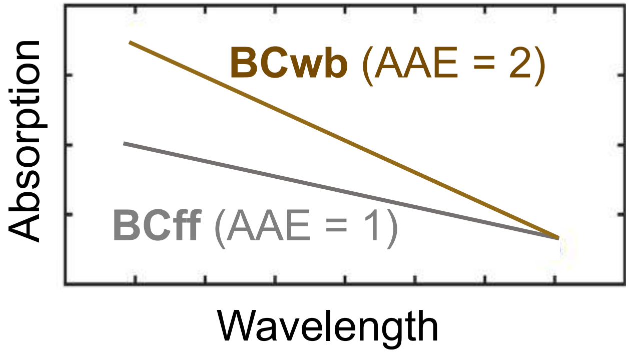
AE33, Aerosol d.o.o.

σ_{abs} (7w) & Black Carbon

Source apportionment

Absorption Ångstrom Exponent (AAE)

BCwb – wood burning & BCff – fossil fuel



ATOLL (2016 – 2020)

Sandradewi et al. 2008

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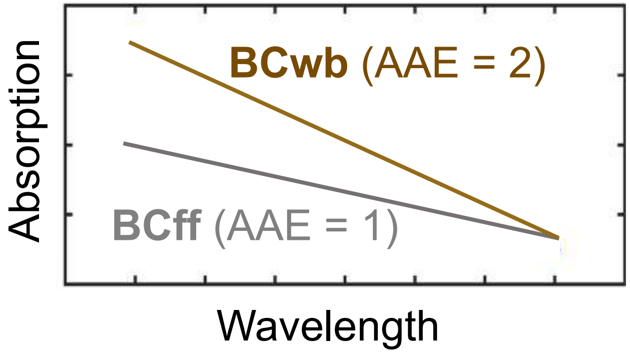

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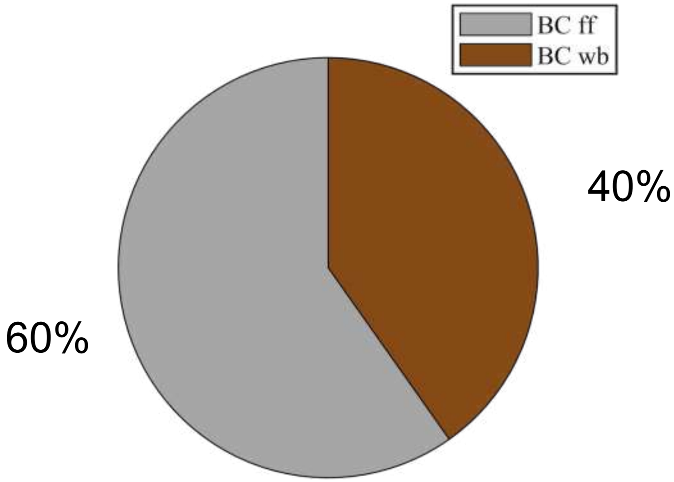

Local vs transported?

➔ % Sectors ?

Sandradewi et al. 2008

INTERPLAY approach

INTERPLAY → combIning iN-siTu obsERvations hysPLit And emissions-inventorY

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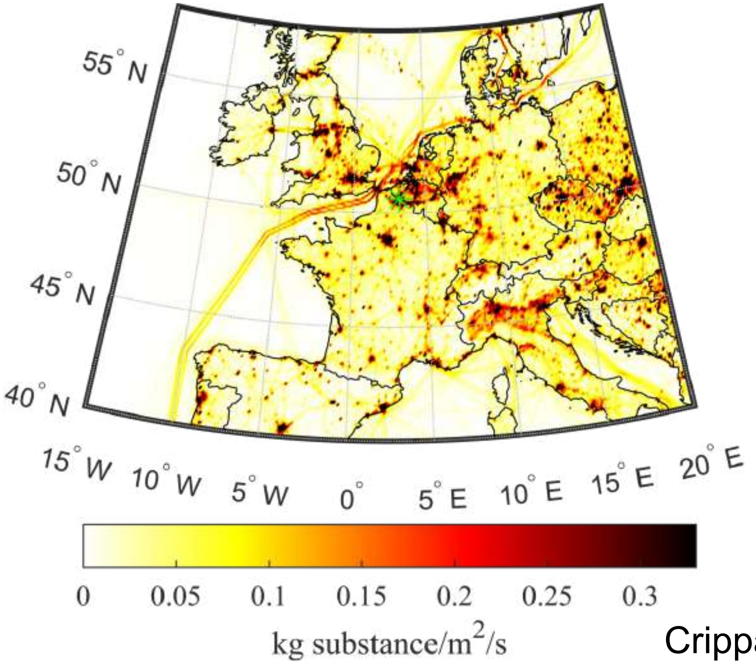
Emission inventory
EDGAR
2018 - Monthly
0.1 degree x 0.1 degree



Crippa et al. 2020, Stein et al. 2015

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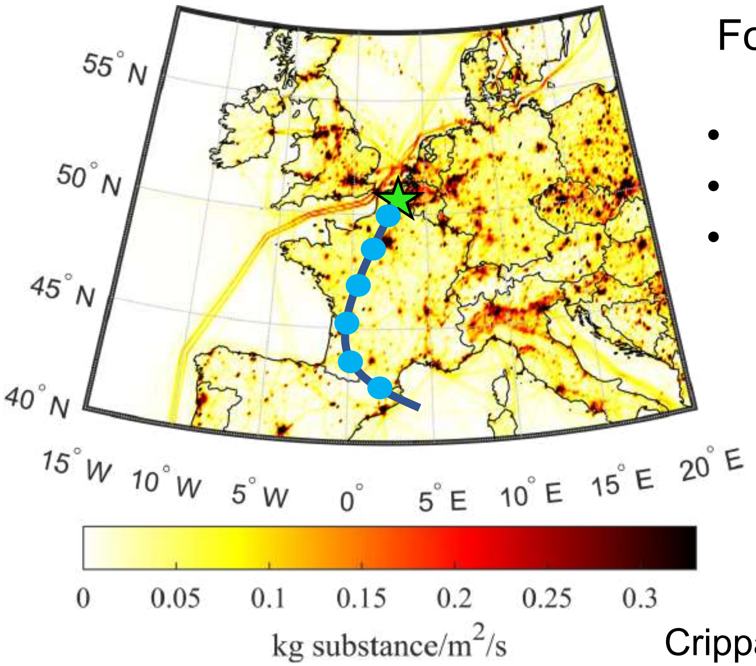
HYSPLIT ATOLL 72 hrs back trajectories
2016 - 2019



For BC observed in ATOLL:

- Source type
- Emission location
- Transport time

Crippa et al. 2020, Stein et al. 2015

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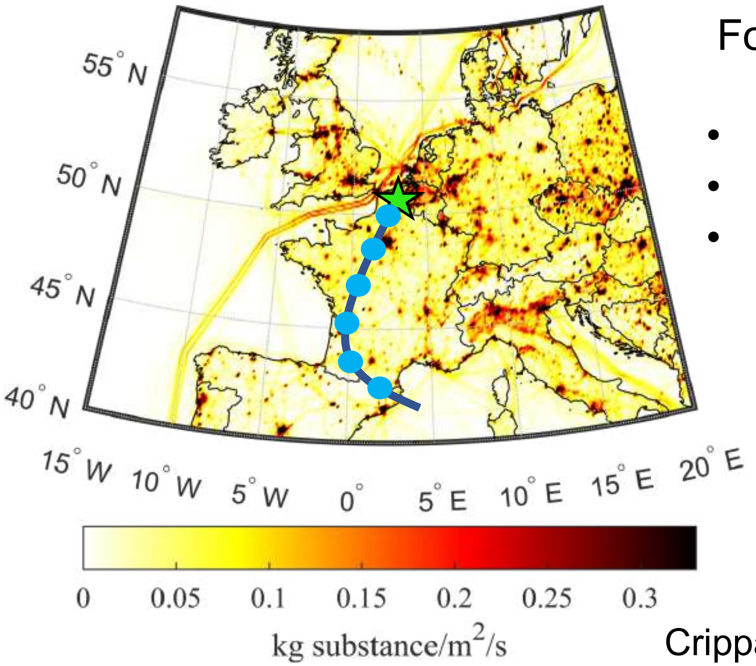

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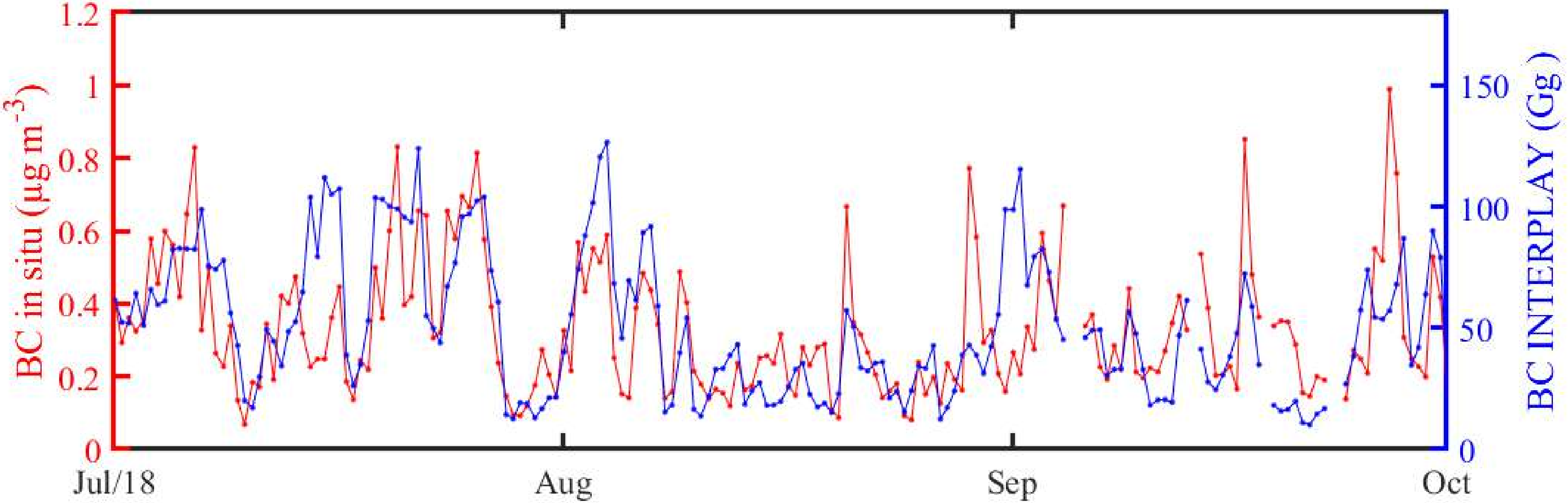
Aethalometer

AE33, Aerosol d.o.o.

σ_{abs} & Black Carbon

2016 - 2019

INTERPLAY vs Aethalometer

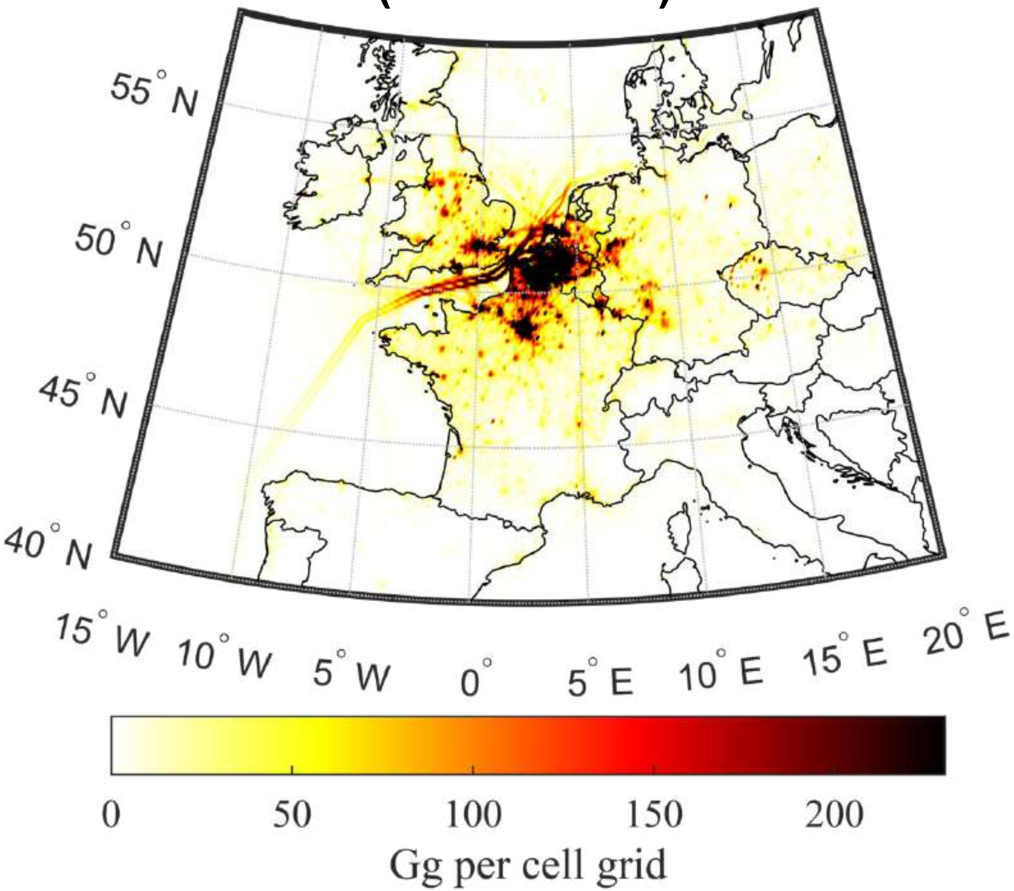


INTERPLAY mimics the BC observed by the aethalometer

→ We can calculate the contribution to BC level at ATOLL by regions & sectors

Spatial contribution to BC level in ATOLL

Multiannual (2016 – 2019)



Spatial contribution to BC level in ATOLL

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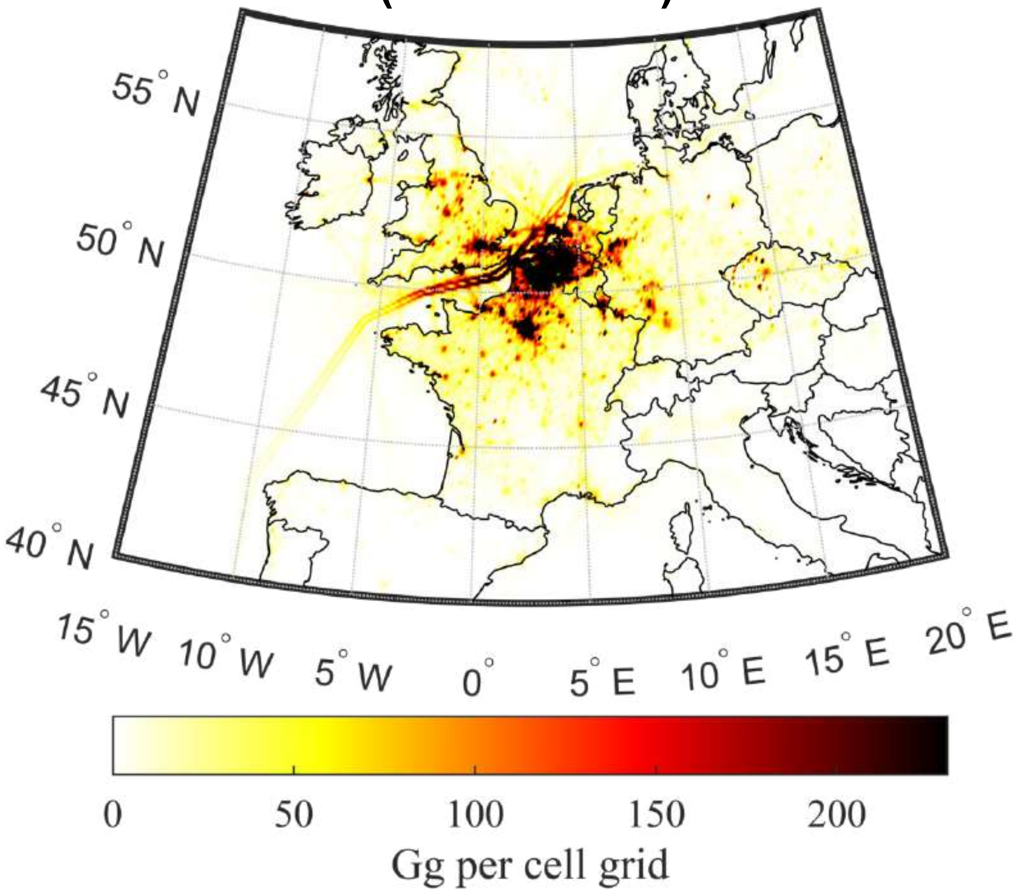
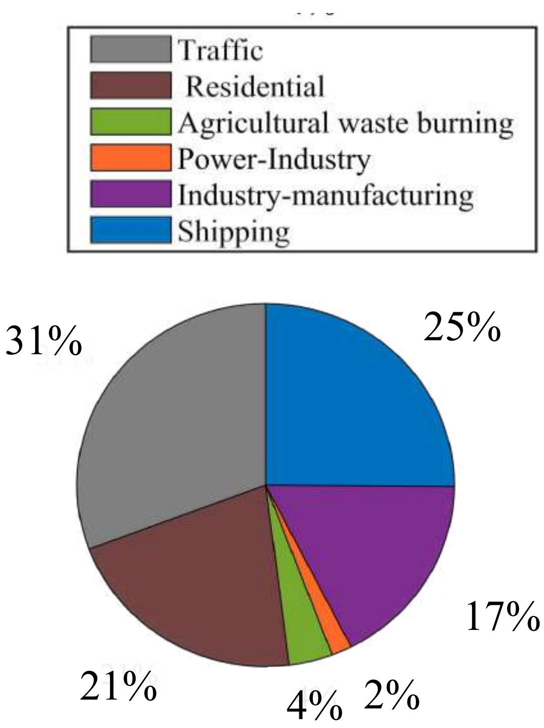




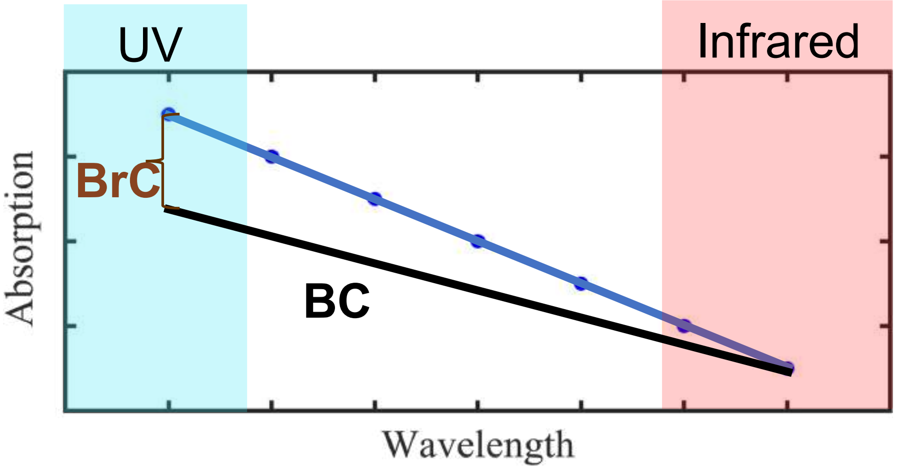
- ATOLL is ~60 km apart from the main port (Calais)

Brown carbon (BrC)

What do we know of absorbing aerosols?

Wavelength dependent:

BC - from UV to infrared
BrC - mostly UV



(Bond & Bergstrom 2006, Laskin et al. 2015, Saleh et al. 2020)

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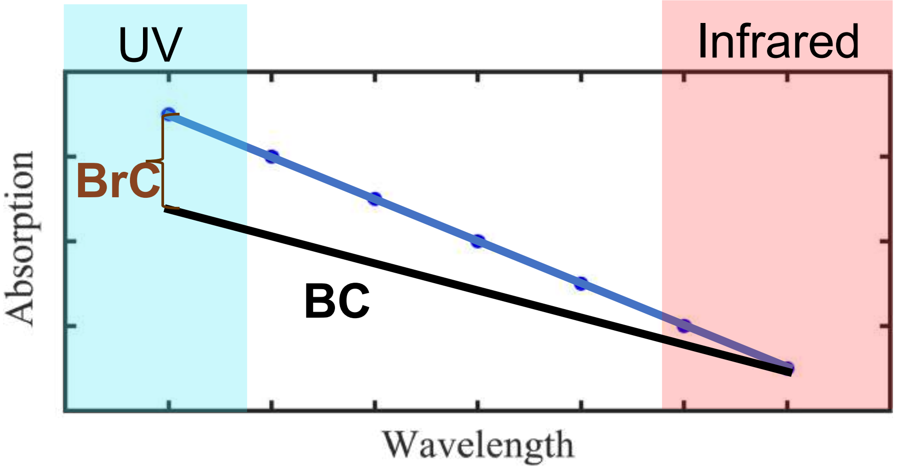
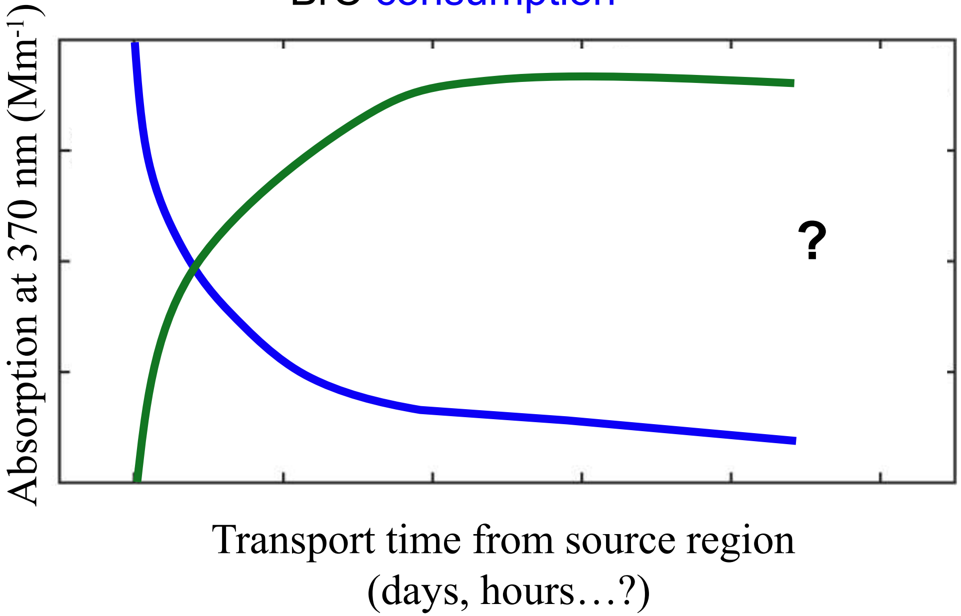


(Bond & Bergstrom 2006, Laskin et al. 2015, Saleh et al. 2020)

Atmospheric aging influences **BrC_{abs} optical properties**

- BrC formation (SOA)
- BrC consumption



BrC formation/consumption time :

Atmospheric observations → ~1day

Laboratory experiments → hours

Wintertime BrC aging

BrC $absorption_{470\ nm}$ lifetime → **1 day**

with

a. **INTERPLAY** (BC footprint)

b. σ_{abs} 7w (370nm – 950nm)

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IMPROVE equation : (Pitchford et al. 2007)

$$\sigma_{abs,\lambda} = \sum_j MAE_{j,\lambda} \times C_j \times f(RH)_j$$

Mass Absorption Efficiency (MAE_j)

Mass concentration of each species (C_j)

Water growth factor $f(RH)_j = 1$ (dry conditions)

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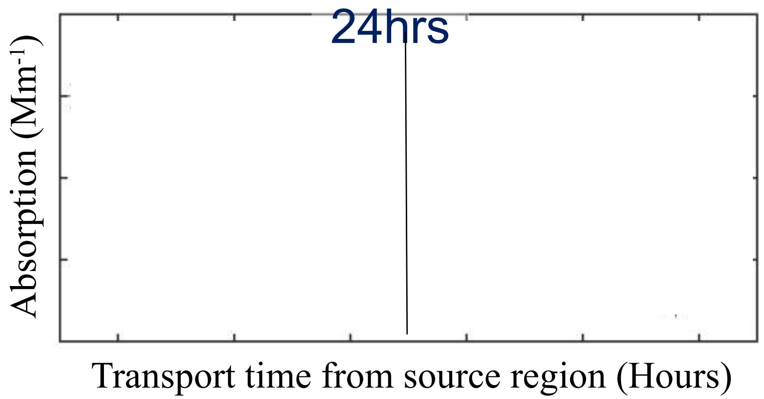
INTERPLAY Two sectors were selected during cold season

BC traffic & BC residential



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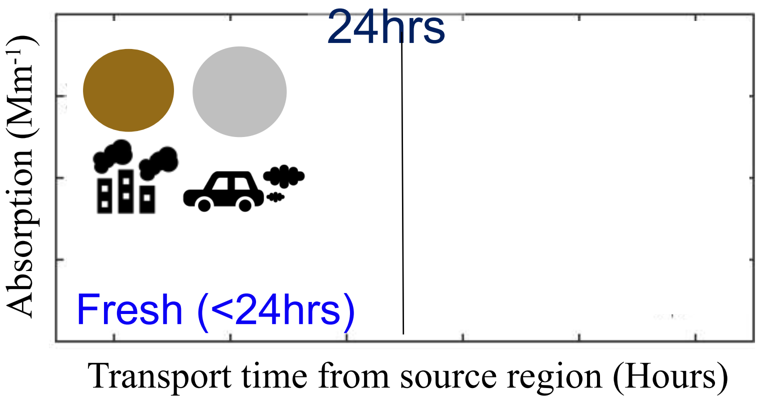

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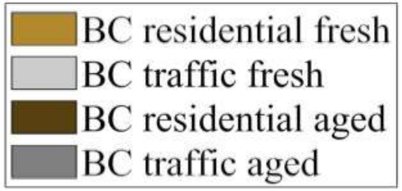

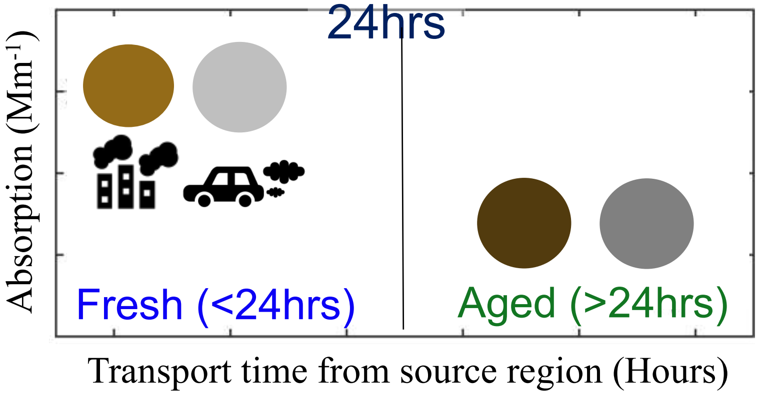

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Multiple Linear Regression (**MLR**)

INTERPLAY

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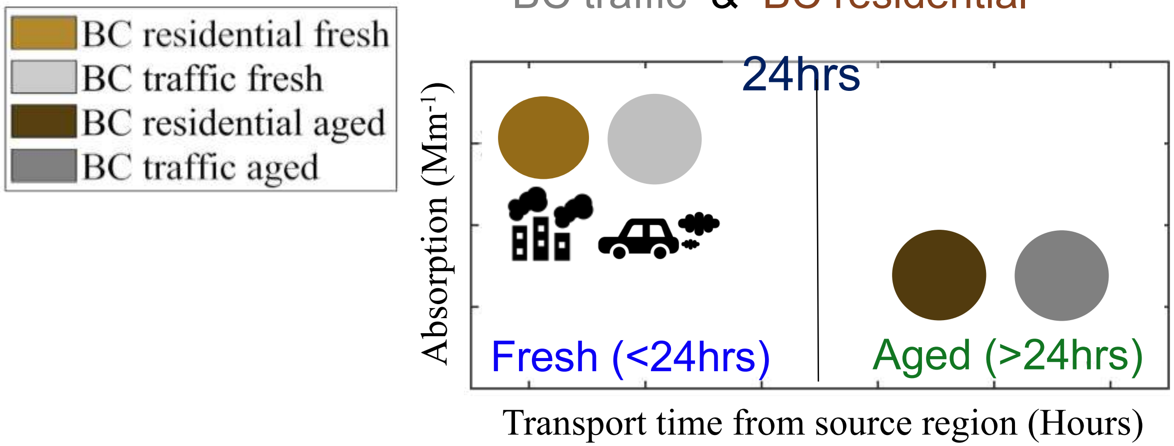

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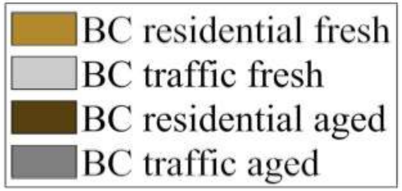
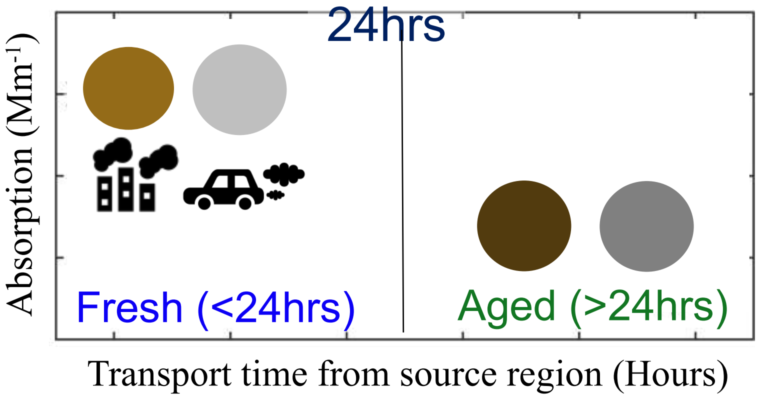


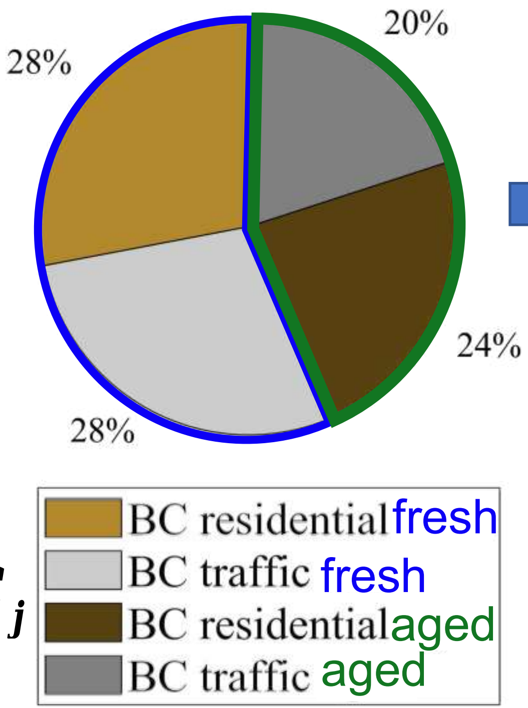


Contribution to BrC$_{470nm}$ by

Fresh (<24hrs) vs Aged (>24hrs)

Wintertime BrC aging

Contribution to **BC mass**
INTERPLAY



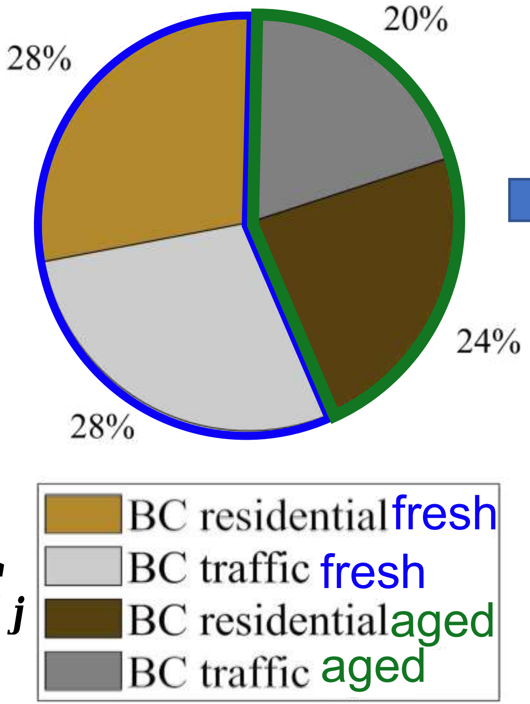
$$C_j$$

$$MAE_{j,\sigma_{BrC,470nm}} \times C_j$$

Velazquez-Garcia et al. to be submitted soon

Wintertime BrC aging

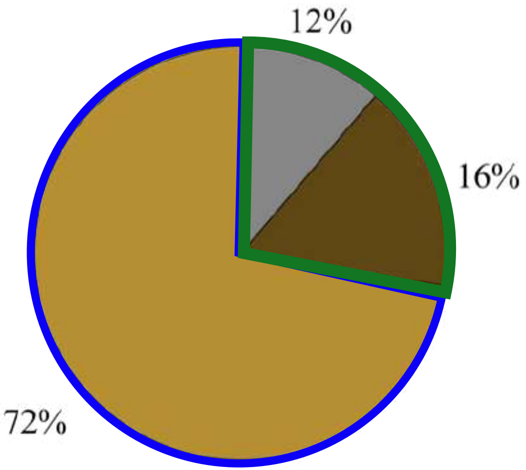
Contribution to **BC mass**
INTERPLAY



C_j

$$MAE_{j,\sigma_{BrC,470nm}} \times C_j$$

Contribution to $\sigma_{BrC,470nm}$



- 28% of residential fresh contributes 72% to BrC_{470nm}
- 28% of taffic fresh does not contribute to BrC_{470nm}

1. **Consumption** (**fresh** particles **contribute more** than **aged**)
2. **After 24hrs of aging** the BrC consumption **is efficient**

Velazquez-Garcia et al. to be submitted soon

Conclusions

- A new tool was developed (**INTERPLAY**) to study the **BC footprint** at the site and **able to** mimic in-situ **observations**

- By applying INTERPLAY **the spatial and sectoral contributions to BC level** at the ATOLL platform **were retrieved**

  - **Origins of BC levels** at ATOLL: **UK, Paris, BENELUX area, western Germany**

  - **Three main sectors** contributing to BC levels: **traffic, shipping & residential (mostly during winter)**

- **Absorption associated to BrC** from residential heating during wintertime **decreases 24 hrs after emission**

Perspectives

- Study **particulate organic nitrates** to investigate the formation of **secondary BrC** using the ACSM and the aethalometer data

- Evaluation of the effect of **aging** on **BrC** to validate **AAE used** for the **source apportionment method** by combining high-time resolution observations with auxiliary data and tools

- Prospects for **widespread use**; applicability to other chemical species and/or in situ measurements.

Thank you

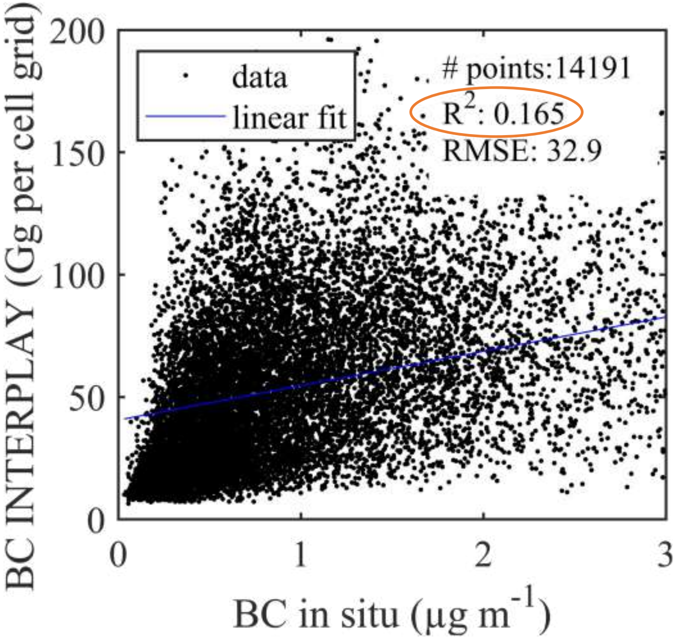
Do you have any questions?

alejandra.velazquez-garcia@meteo.fr

Optimization of INTERPLAY

Integration with in situ observations…?

1 hr → r^2 = 0.165 n=14191

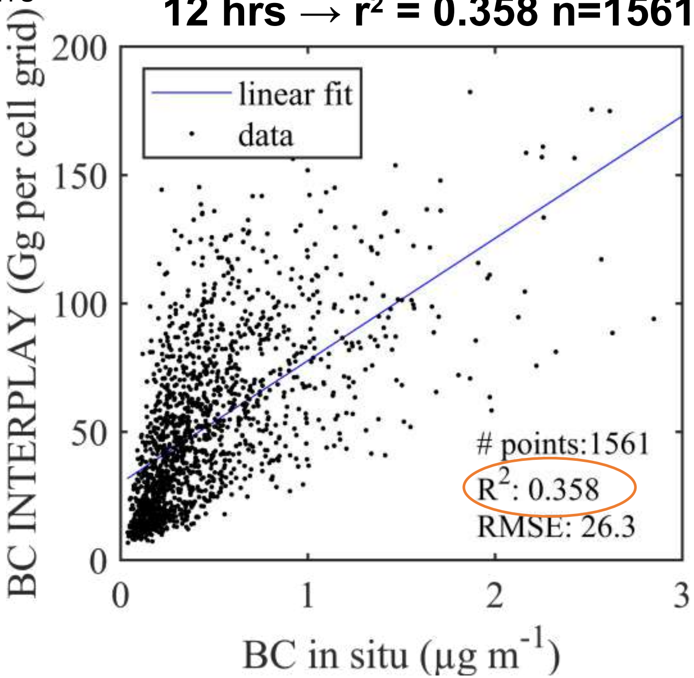


Baseline dataset test

minimize the relevance of sporadic local sources in the site

mean – $BC_{INTERPLAY}$ &
5^{th} percentile – $BC_{IN\ SITU}$

6 hrs → r^2 = 0.288 n=2907
9 hrs → r^2 = 0.335 n=2035
12 hrs → r^2 = 0.358 n=1561



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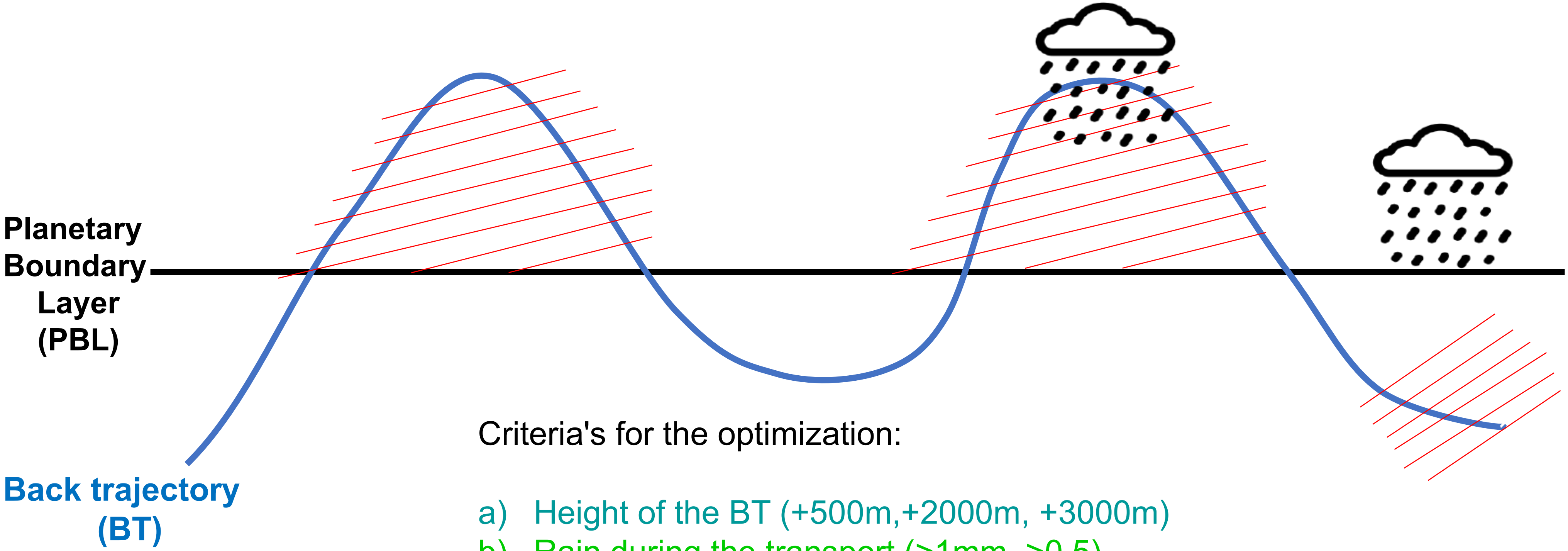


Criteria's for the optimization:

a) Height of the BT (+500m,+2000m, +3000m)
b) Rain during the transport (>1mm, >0.5)
c) Interpolation of BT (10 min)
d) Lille contribution vector (360°, 270°, 180°,…, 30°)
e) Box of BC footprint (1°x1°, 0.8°x0.8°,…, 0.2°x0.2°)
f) Integration with in situ (window average)

Optimization of INTERPLAY

	r² is comparing with in situ observations9	Without PBL criteria	Without rain criteria	Height of BT (PBL+500)	Height of BT (PBL+2000m)	Height of BT (PBL+3000m)	Rain (>1mm)	Rain (>0.5mm)	Interpolation BT (10min)	Without PBL criteria + Rain >1mm + Interpolation of BT of 10 min						Without Interpolation o		
		a	b	a	a	a	b	b	c			d						
										Lille vector (360°)	Lille vector (270°)	Lille vector (180°)	Lille vector (90°)	Lille vector (60°)	Lille vector (30°)	BC footprint (0.2°x0.2°)	BC footprint (0.3°x0.3°)	foo (0.4
a	Without PBL criteria	1	r^2=0.17 n=19932	NA	NA	NA	r^2=0.176 n=14191	r^2=0.180 n=10903	NA	NA	NA	NA	NA	NA	NA	NA	NA	
b	Without rain criteria	r^2=0.17 n=19932	1	r^2=0.151 BT=376000	r^2=0.167 BT=48000	r^2=0.168 BT=12000	NA	NA	r^2=0.169 n=19932	NA	NA	NA	NA	NA	NA	NA	NA	
a	Height of BT (PBL+500m)	NA	r^2=0.151 BT=376000	1	NA	NA	r^2=0.153 BT=376000	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	
a	Height of BT (PBL+2000m)	NA	r^2=0.167 BT=48000	NA	1		r^2=0.171 BT=48000	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	
a	Height of BT (PBL+3000m)	NA	r^2=0.168 BT=12000	NA	NA	1	r^2=0.175 BT=12000	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	
b	Rain (>1mm)	r^2=0.177 n=14191	NA	r^2=0.153 BT=376000	r^2=0.171 BT=48000	r^2=0.175 BT=12000	1	NA	r^2=0.165 n=14191 + without PBL	r^2=0.176 n=14191	r^2=0.171 n=14191	r^2=0.165 n=14191	r^2=0.155 n=14191	r^2=0.147 n=14191	r^2=0.139 n=14191	r^2=0.161 n=14191	r^2=0.164 n=14191	r^2= n=1
b	Rain (>0.5mm)	r^2=0.180 n=10903	NA	NA	NA	NA	NA	1	NA	NA	NA	NA	NA	NA	NA	NA	NA	
c	Interpolation BT (10min)	NA	r^2=0.169 n=19933	NA	NA	NA	r^2=0.165 n=14191	NA	1	r^2=0.176 n=14191	r^2=0.171 n=14191	r^2=0.165 n=14191	r^2=0.155 n=14191	r^2=0.147 n=14191	r^2=0.139 n=14191	r^2=0.161 n=14191	r^2=0.164 n=14191	r^2= n=1

At the end we have…**Without PBL criteria + Rain >1mm + Interpolation of BT of 10 min + Lille vector (180°) + BC footprint square**

Uncertainties associated to INTERPLAY

HYSPLIT

- An error associated with a given trajectory calculation → **15 to 30% of the travel distance** (Stein et al. 2015)

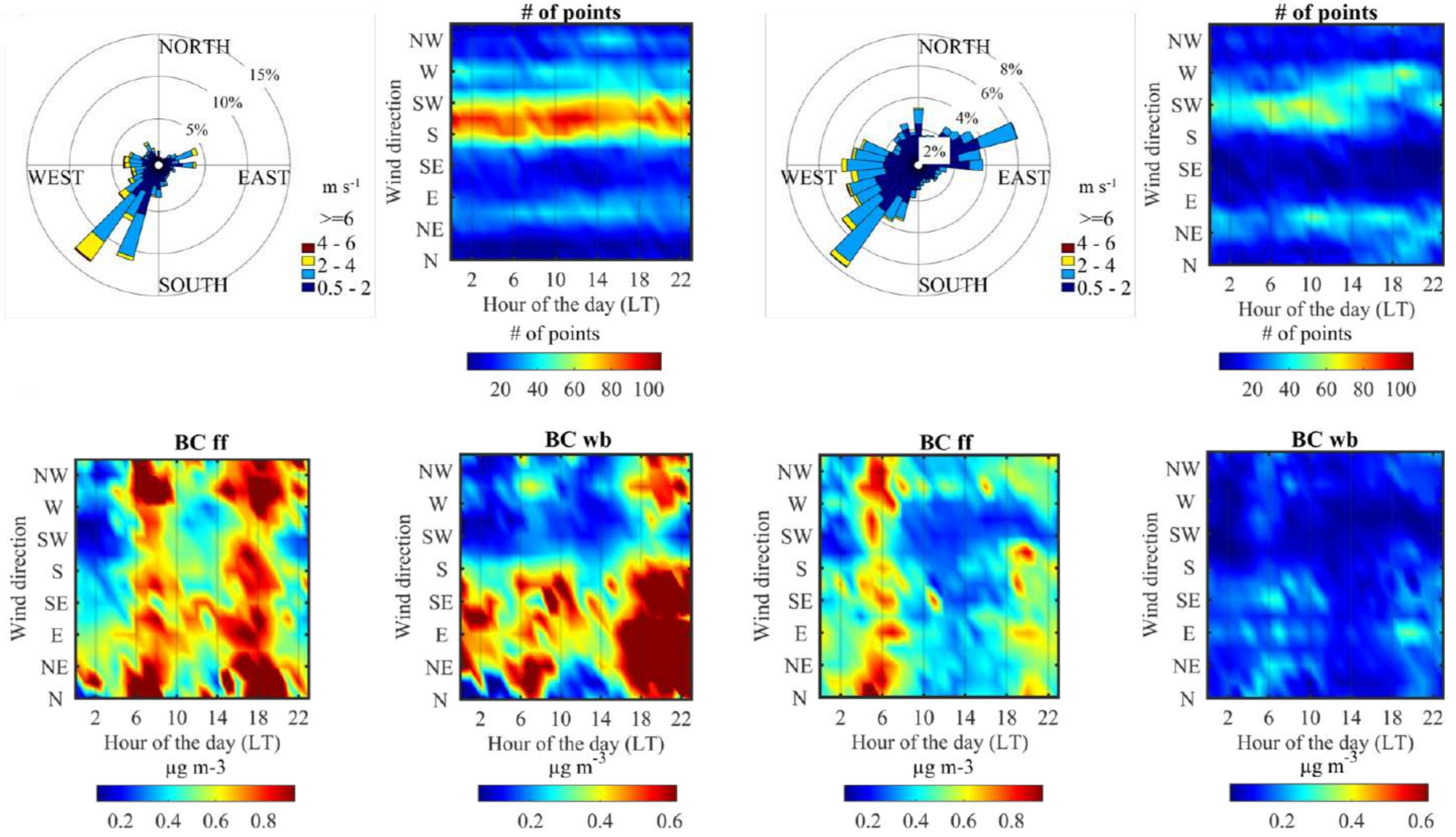
EDGAR v6

Top-down global inventory downscaling national emission data at **fine resolution** (Monthly, 10 km – 10km, sectors)

- **Uncertainties** associated to emission inventories can remain in:
  a) **National total emission estimates:**
     - Caused by model settings, reporting emission sources, gap filling approaches, assumptions or arbitrary choices
  b) **Spatial gridded methodologies:**
     - Spatial discrepancies due to the methodological assumptions, choices of the weighting methodology

  Case of studies:
     - Denier van de Gon et al. 2015 (**EC, OA** – EUCAARI 2005) → **underestimation in wintertime by a factor of 5**
     - Fombetti et al. 2018 (6 EIs) → **a factor of 1.5 – 2 regarding $PM_{2.5}$ EDGARv 2016**
       - ✓ Sectors: **Residential ~ 38%** (100% - all), **Traffic 15%** (25% - all)
       - ✓ Countries: **Paris**, Bucharest, Sofia, Budapest → higher emissions (over allocation of industries)
- Evaluate and improve uncertainties (pollutant and sectorial approach) → Screening method (2 emission inventories, Thunis et. al 2020)

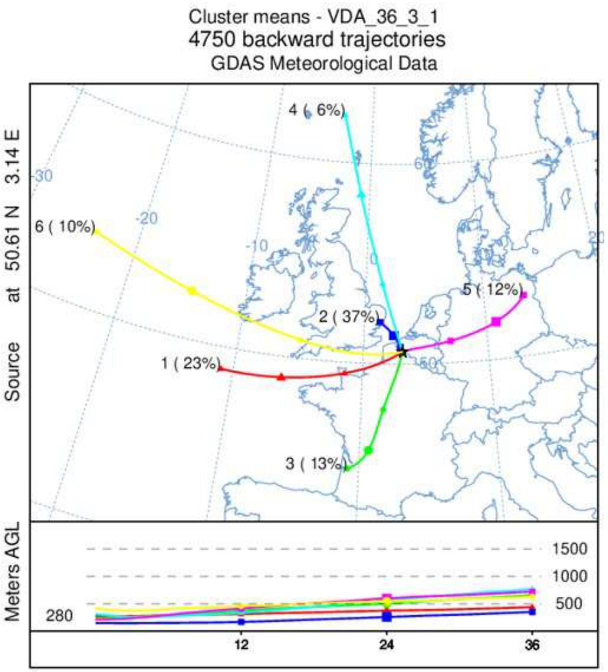
Cold (Oct - Mar) **Warm (Apr - Sep)**

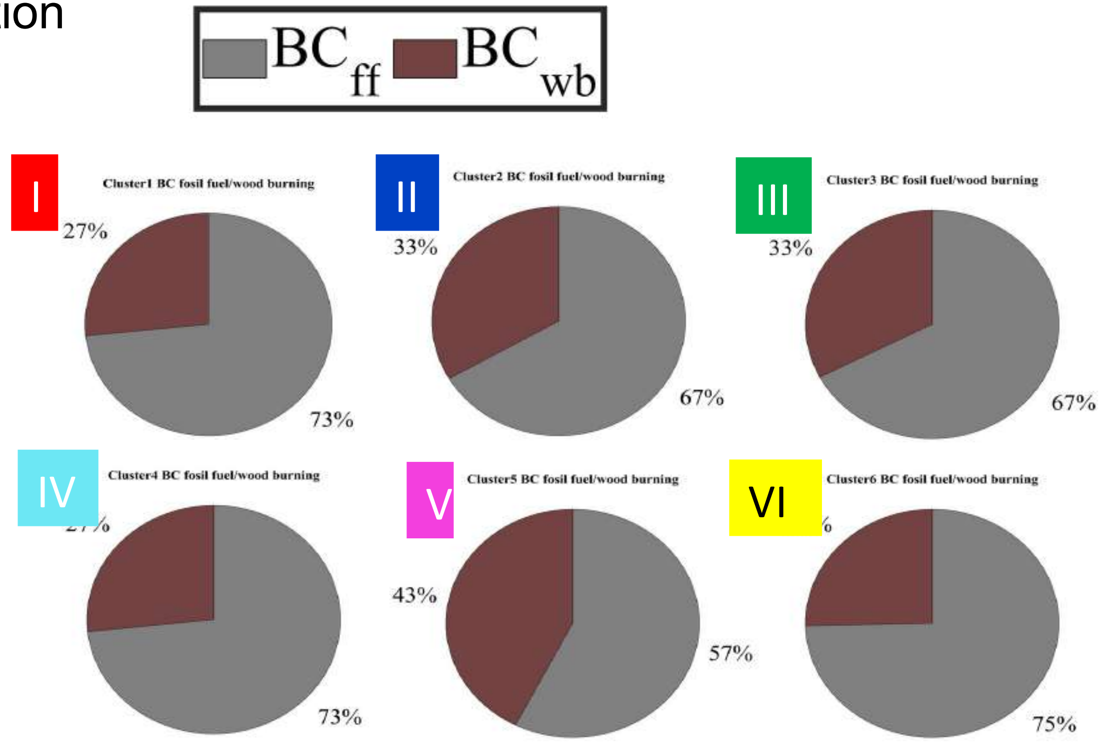


Back trajectories

ATOLL platform
6-cluster solution

I, IV, VI → Marine influence
III, IV, V → Continental influence (BCwb)



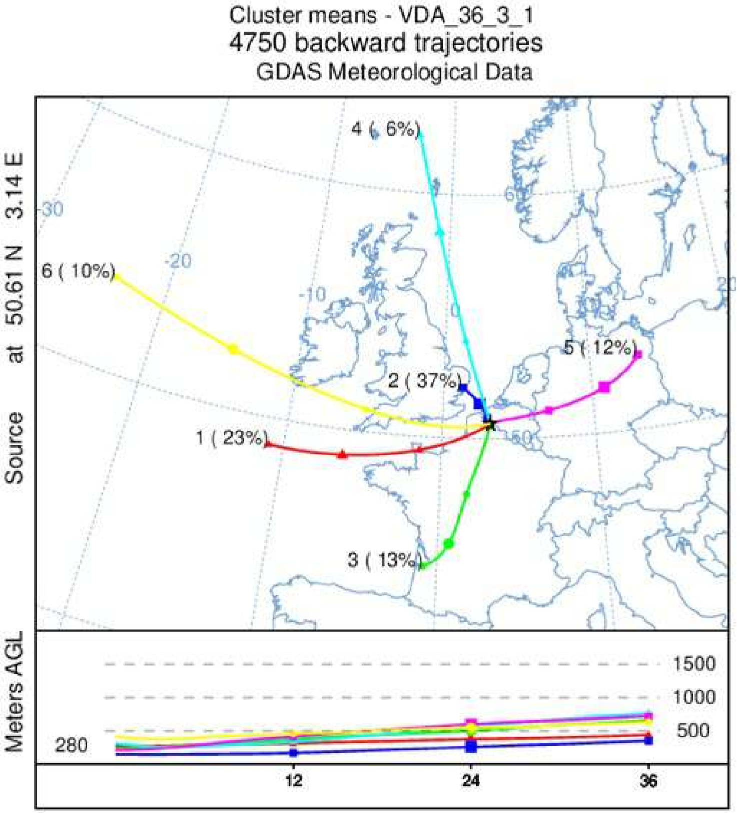


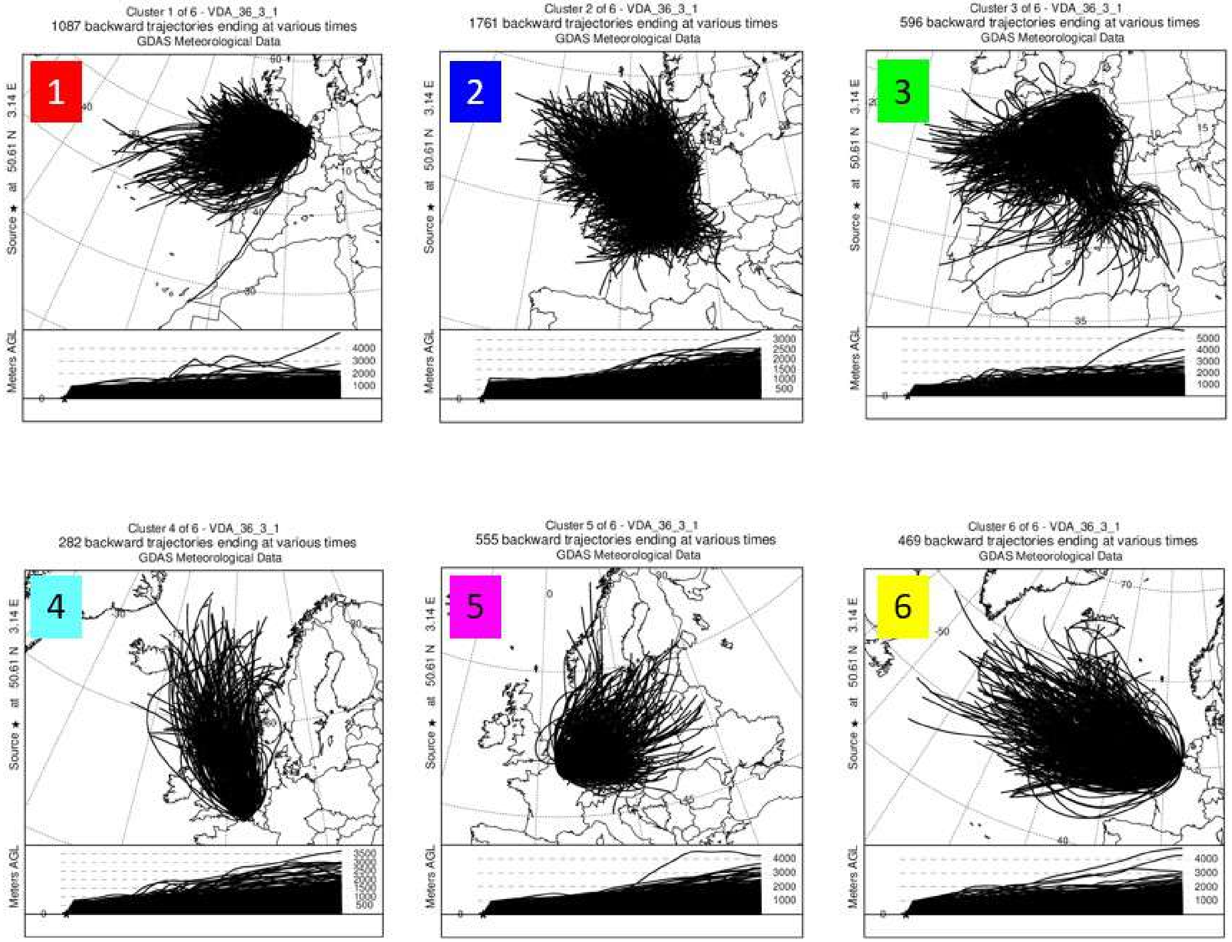
HYSPLIT

Back trajectories

ATOLL platform
6-cluster solution

Limitations!





Objective: **Track the BC – footprint → in situ observations**

HYSPLIT

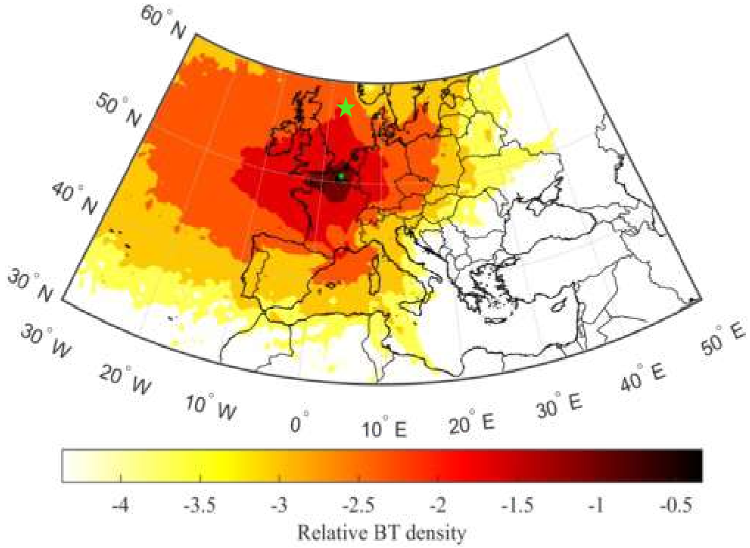
Individual back trajectories (BT)

72 hrs back trajectories – Hourly (2016-2019)

Interpolation (10 min)

Criteria: Rain <1mm (wet deposition)

Half of the PBL & 1 degree GDAS



overlayed onto

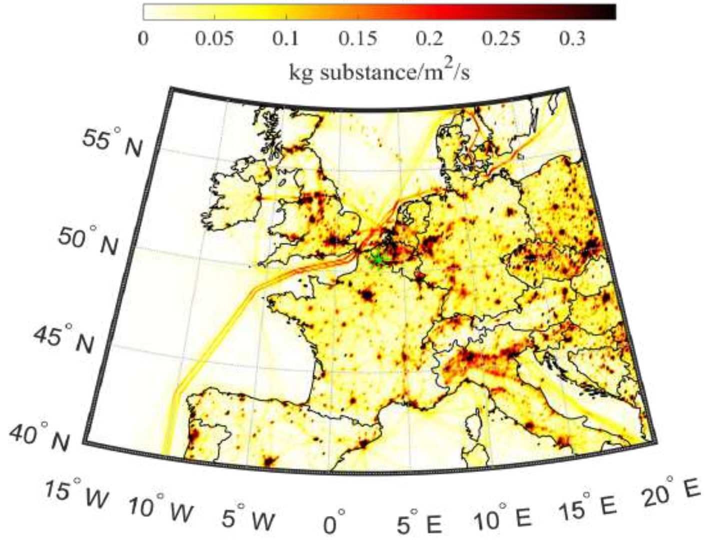
EDGAR v6

Emission inventory - BC

0.1 degree x 0.1 degree

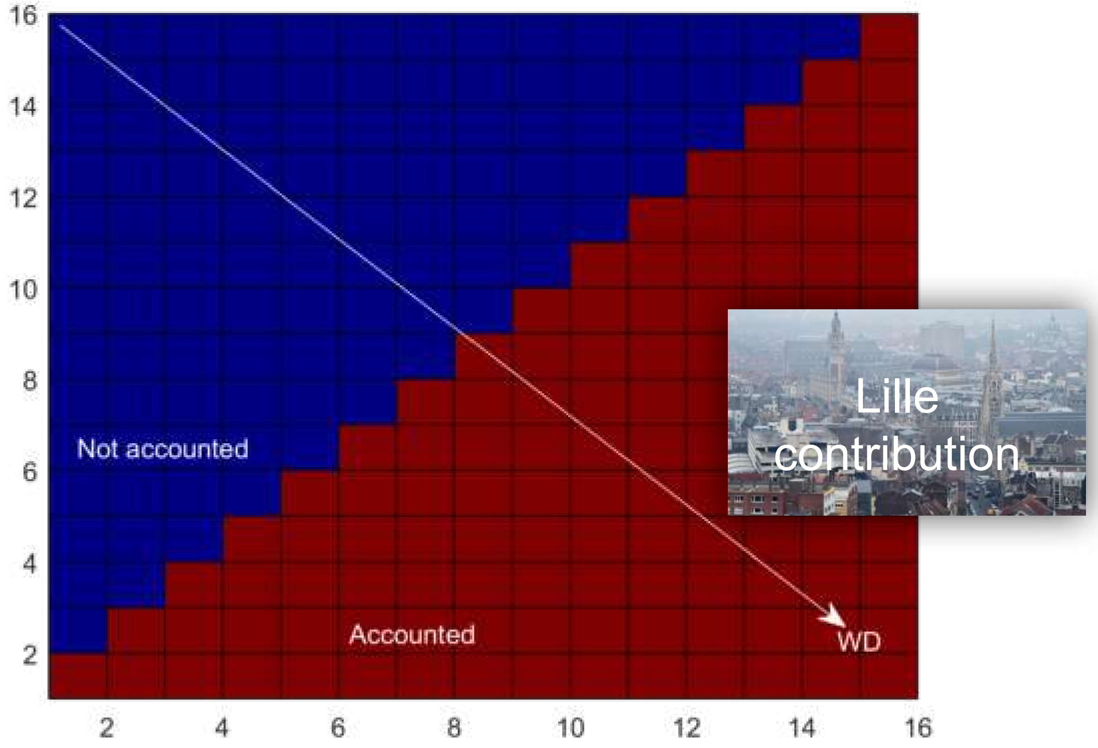
2018 – Monthly - kg m^{-2} s

Main sectors contributing to BC



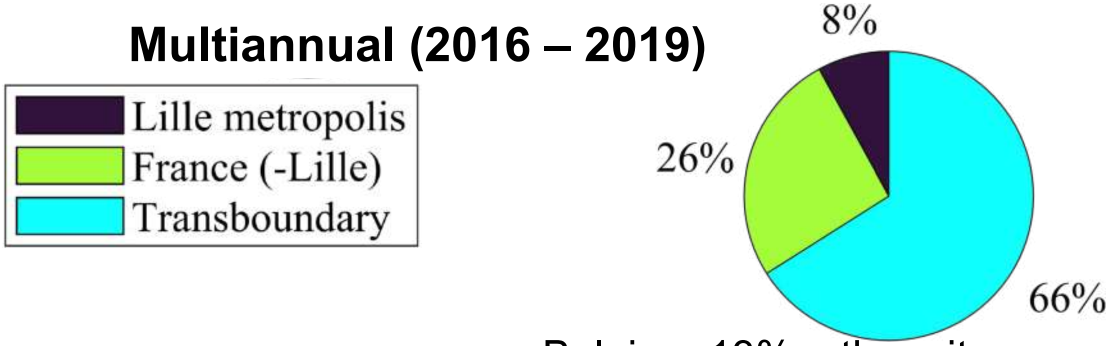
Lille contribution

Local contribution
(arrival sector)
<40km
Semicircle (90°)



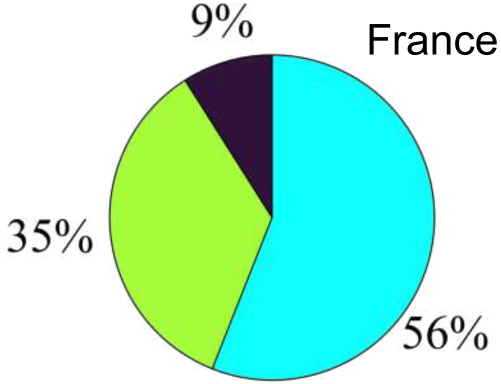
Spatial contribution by <u>country</u> to BC levels in ATOLL

Multiannual (2016 – 2019)



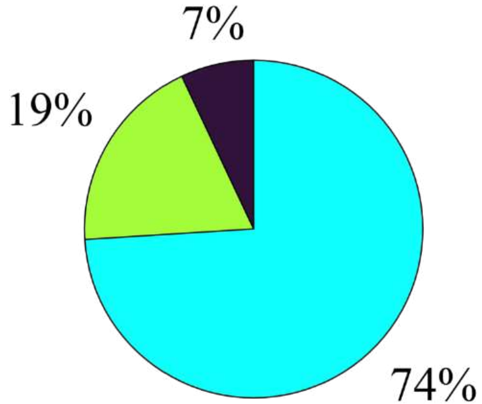
Belgium 13%, other sites

DJF →

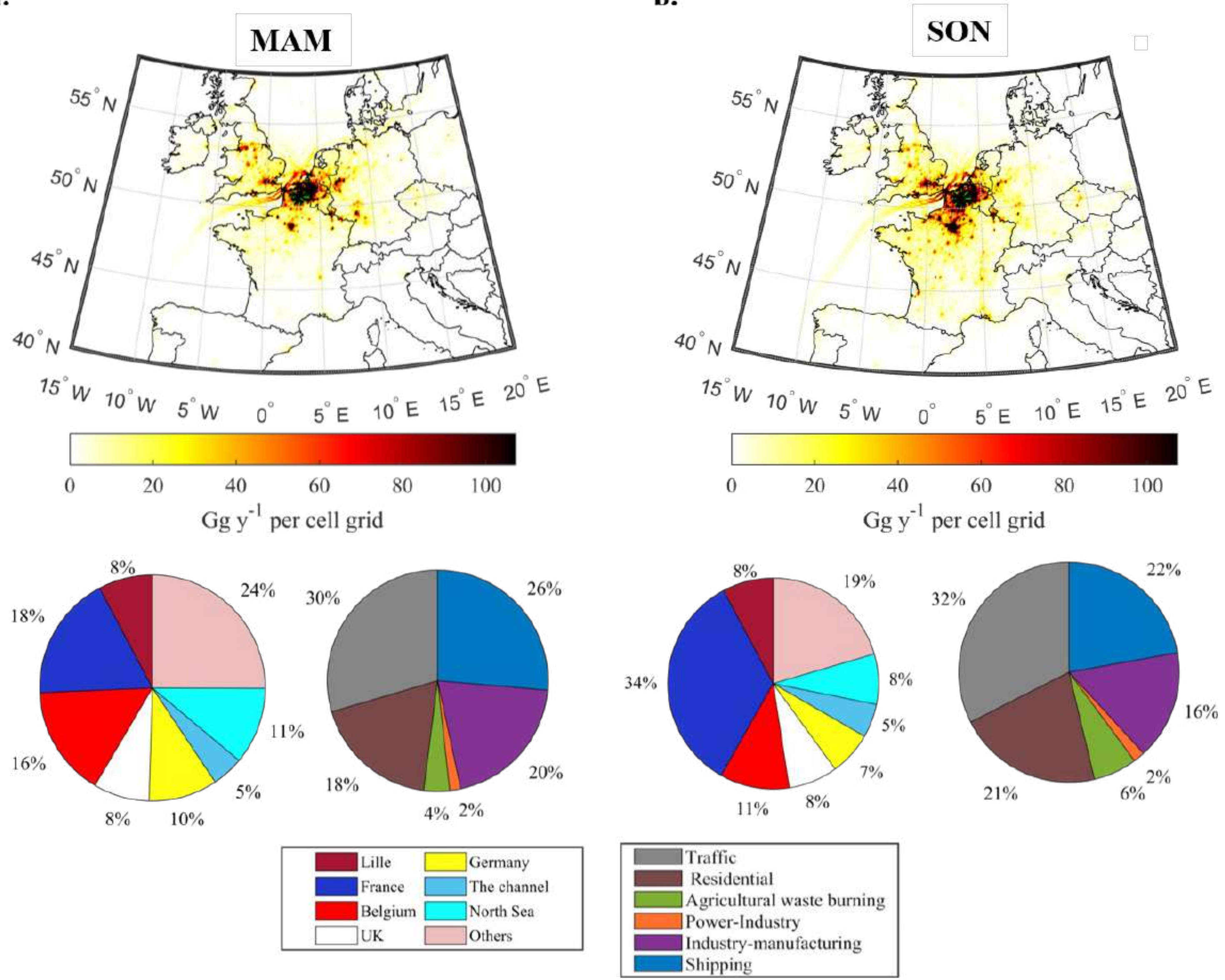


France

JJA →

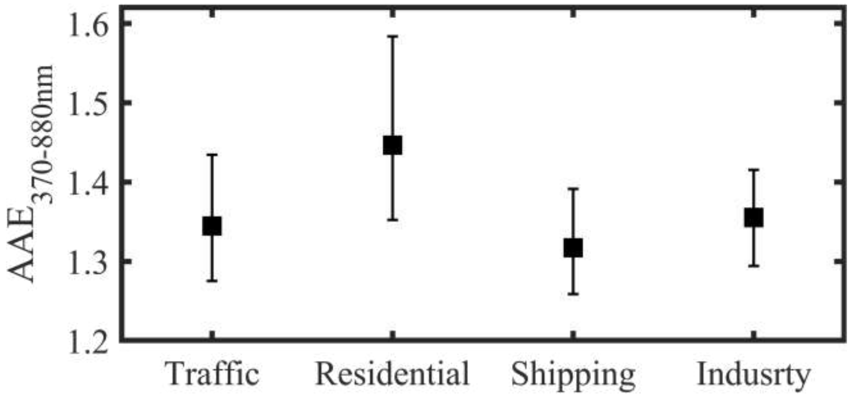


The English Channel 12%
Belgium 18%

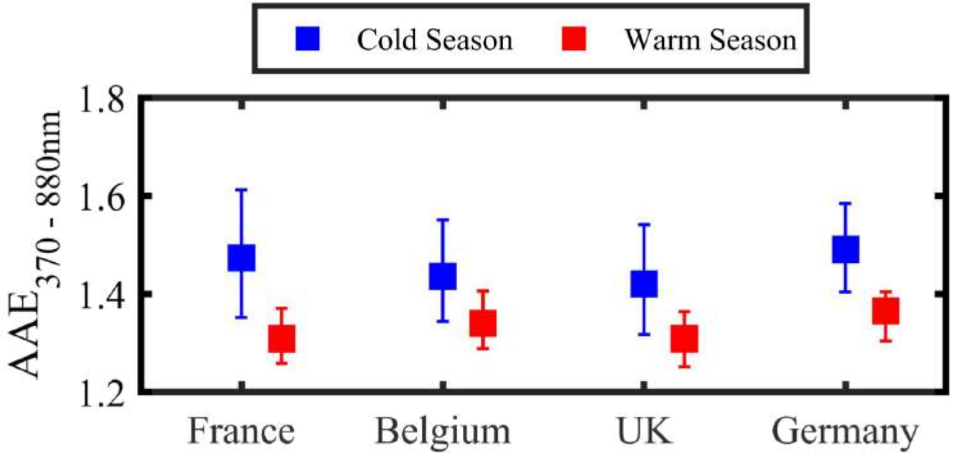

a.
b.
MAM
SON
55° N
50° N
45° N
40° N
15° W
10° W
5° W
0°
5° E
10° E
15° E
20° E
0
20
40
60
80
100
Gg y-1 per cell grid
8%
24%
18%
16%
8%
10%
5%
11%
30%
26%
18%
4%
2%
20%
19%
34%
8%
5%
7%
11%
32%
22%
16%
21%
6%
2%
Lille
France
Belgium
UK
Germany
The channel
North Sea
Others
Traffic
Residential
Agricultural waste burning
Power-Industry
Industry-manufacturing
Shipping

INTERPLAY → in situ
How does the AAE look by region & sectors?

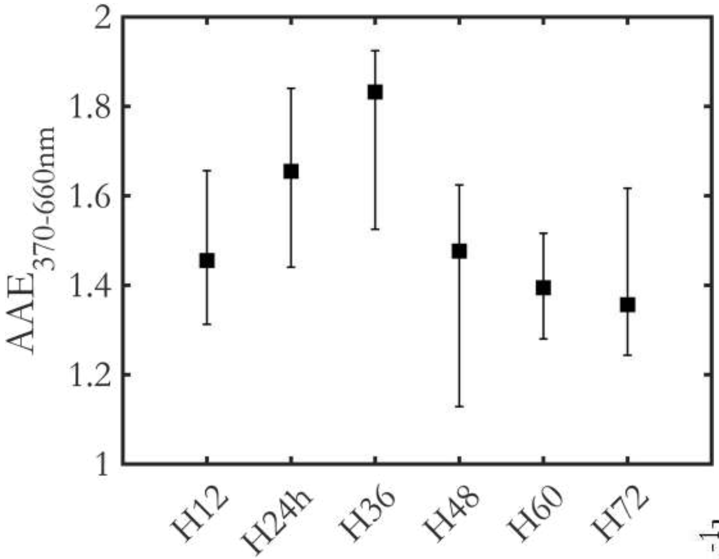
Sectors

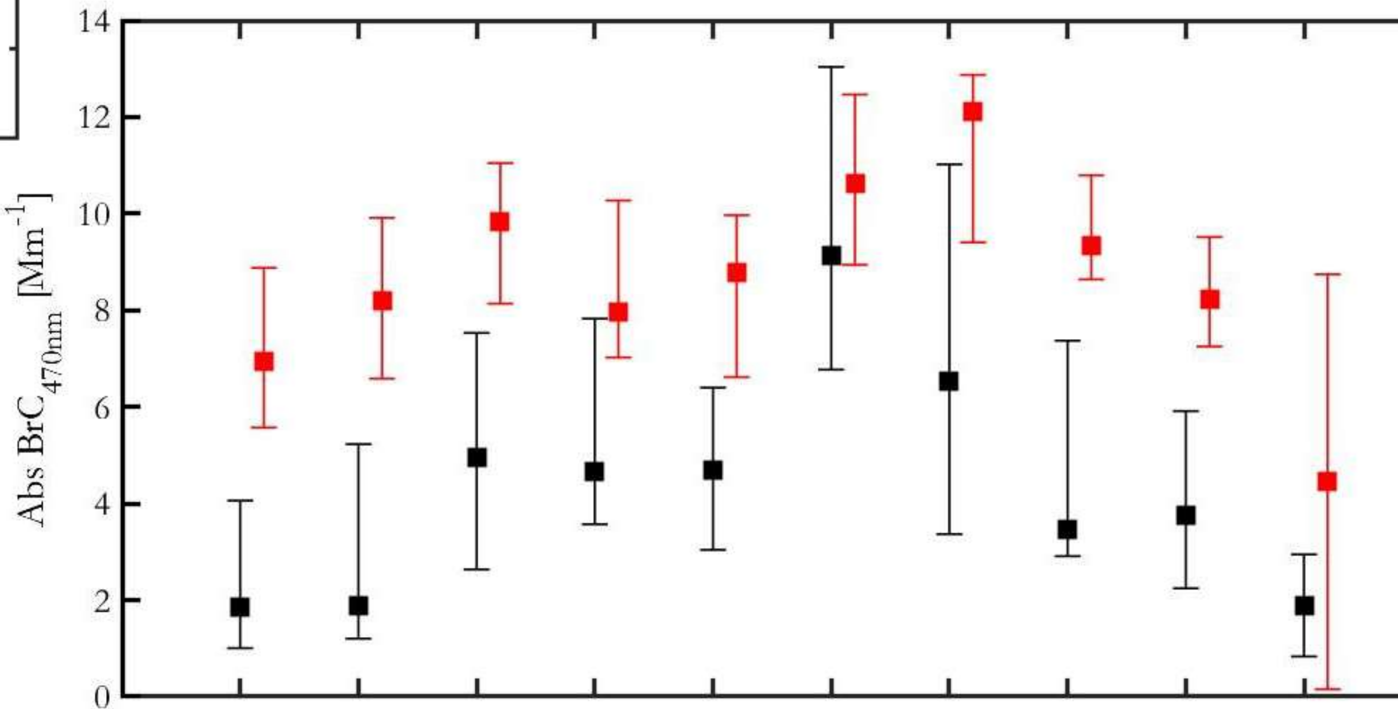


Regions



AAE lifetime



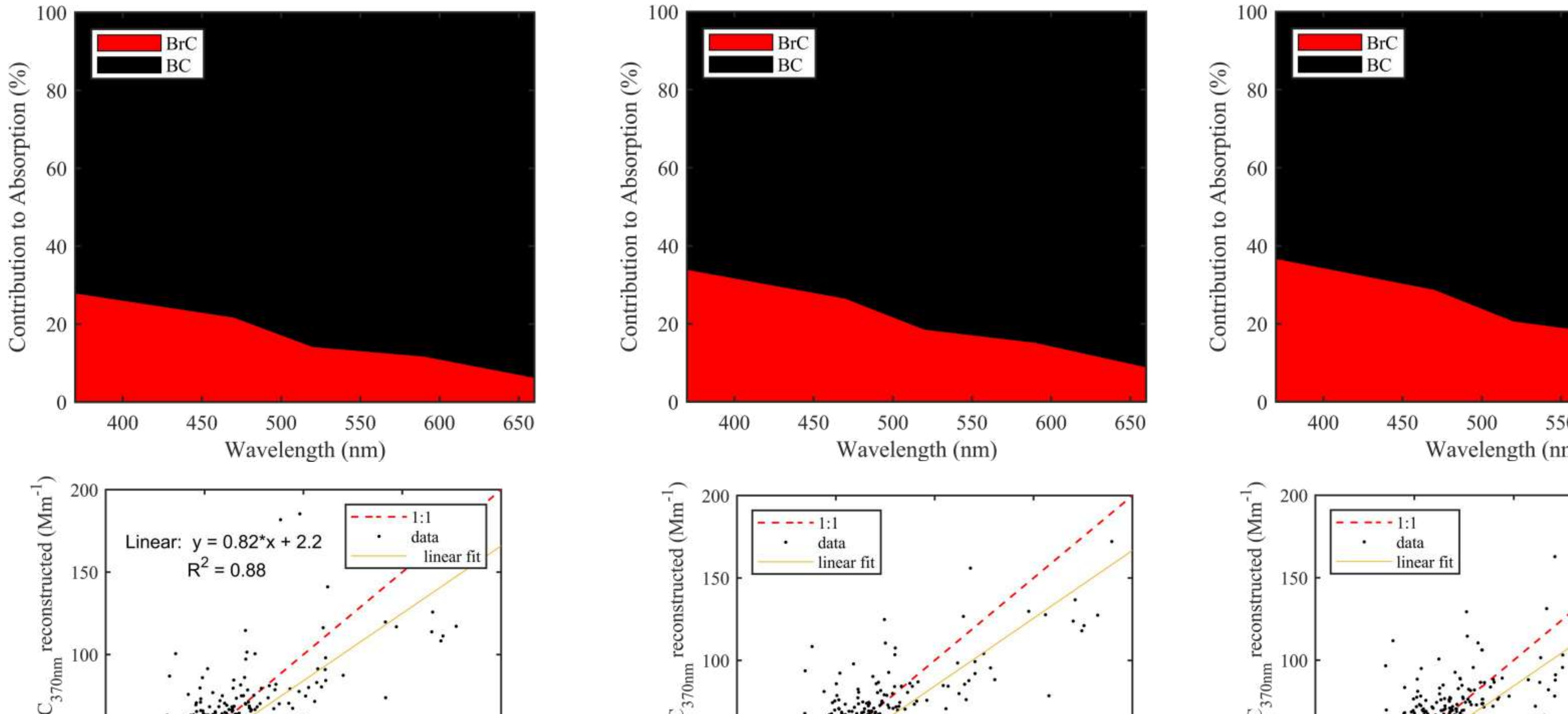


AAE sensitivity test

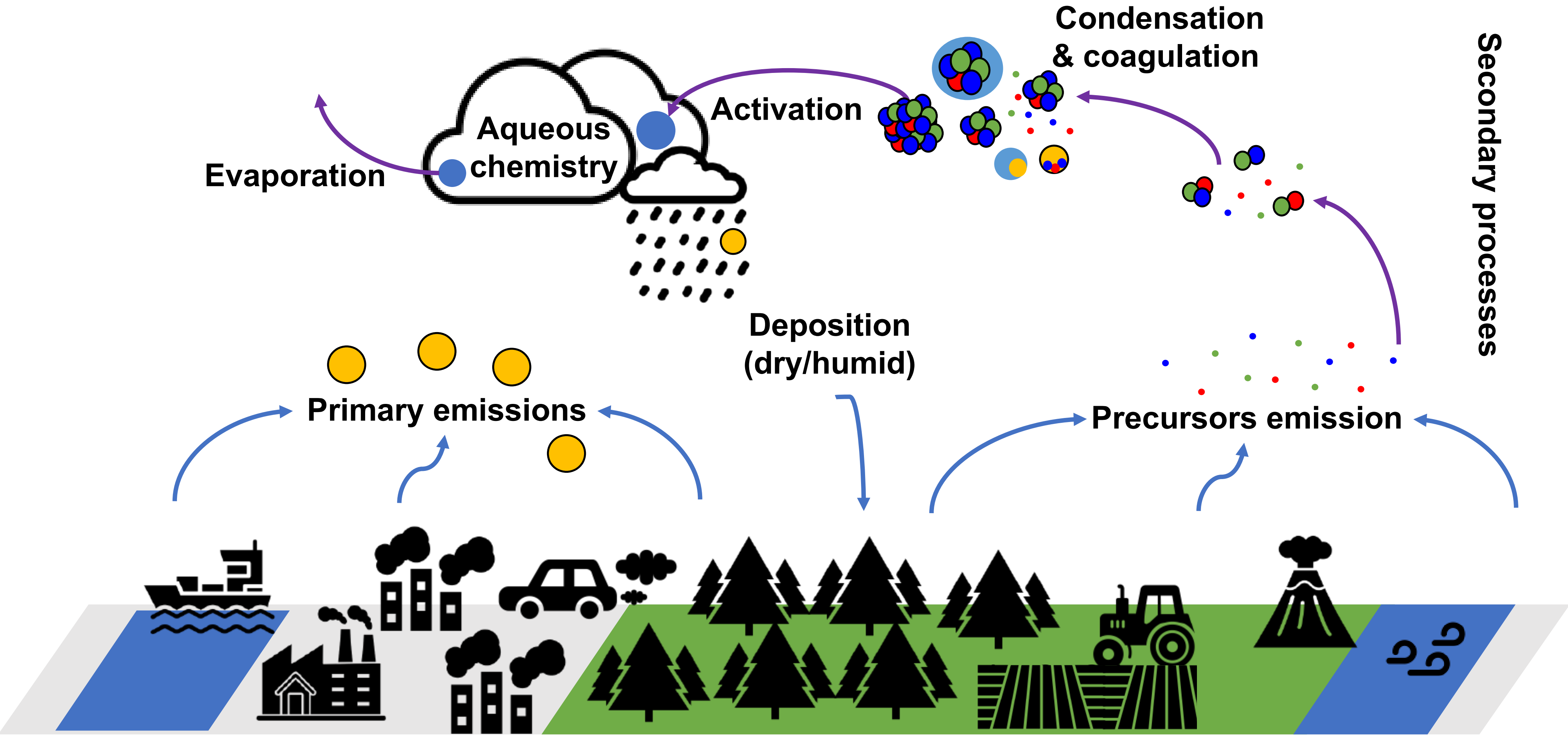
BC fresh
AAE = 1.05

BC coated
AAE = 0.95

BC compacted
AAE = 0.90

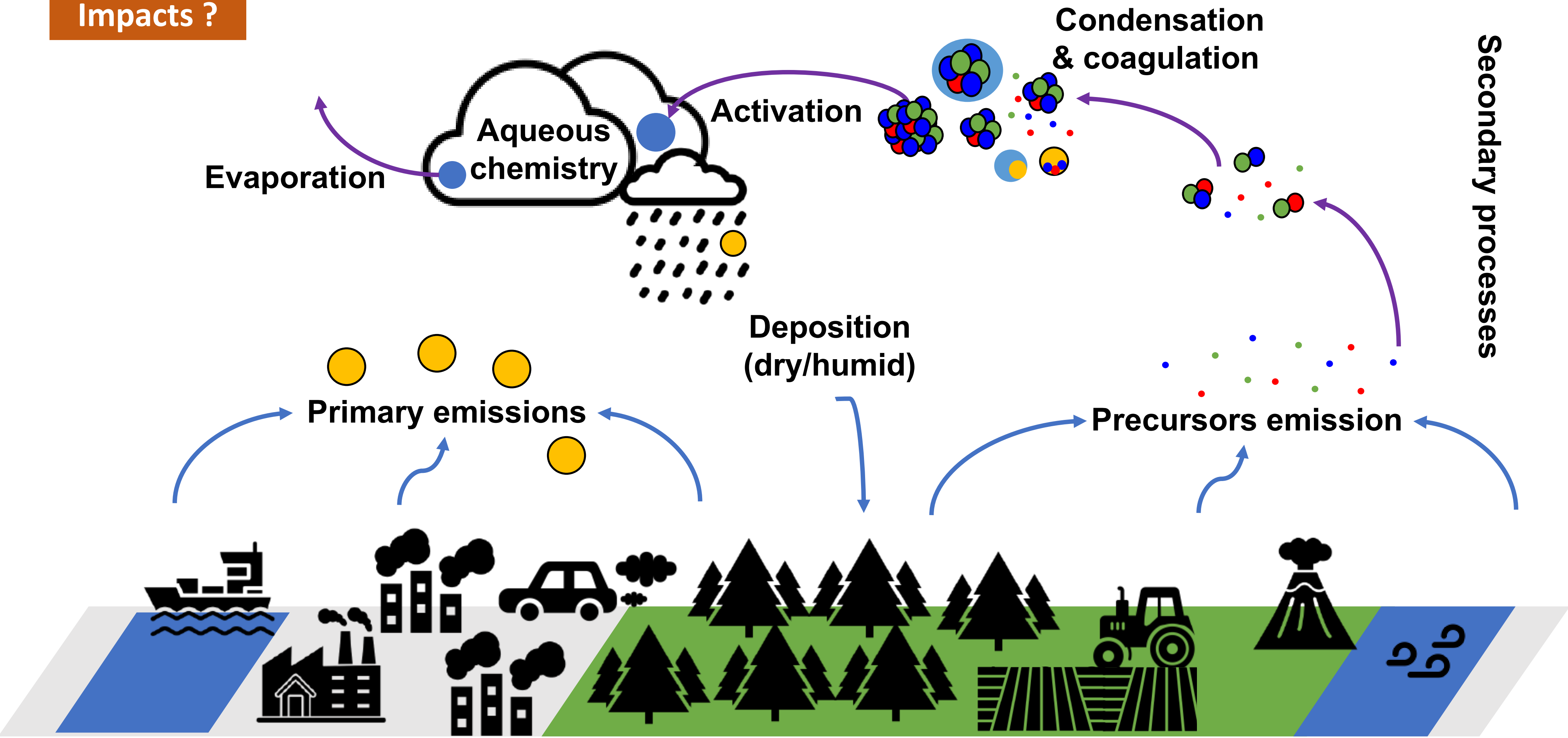


Aerosols and their formation



Aerosols and their formation

Impacts ?



Aerosols and their impacts

Impacts

Air quality

Health **Visibility**

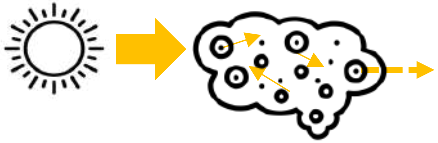
Medina et al. 2021 → air quality models

Air pollution is responsible of
40 000 premature deaths
per year in France

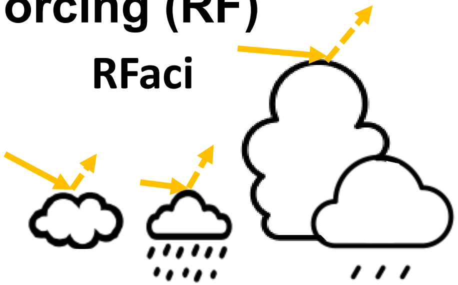
Climate

Radiative Forcing (RF)

RFari



RFaci



Aerosol radiation interaction (ari)

Aerosol cloud interaction (aci)

Aerosols and their impacts

Impacts

Air quality

Climate

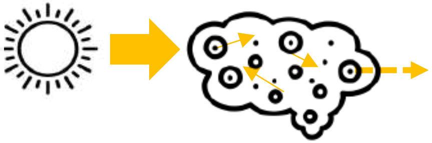
Health

Visibility

Medina et al. 2021 → air quality models

Air pollution is responsible of
40 000 premature deaths
per year in France

RFari

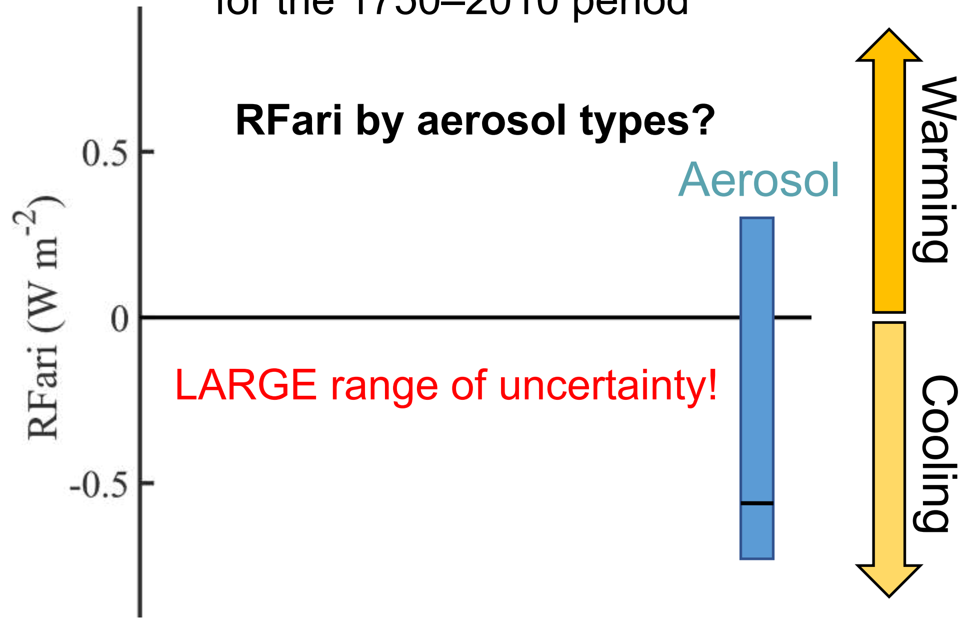


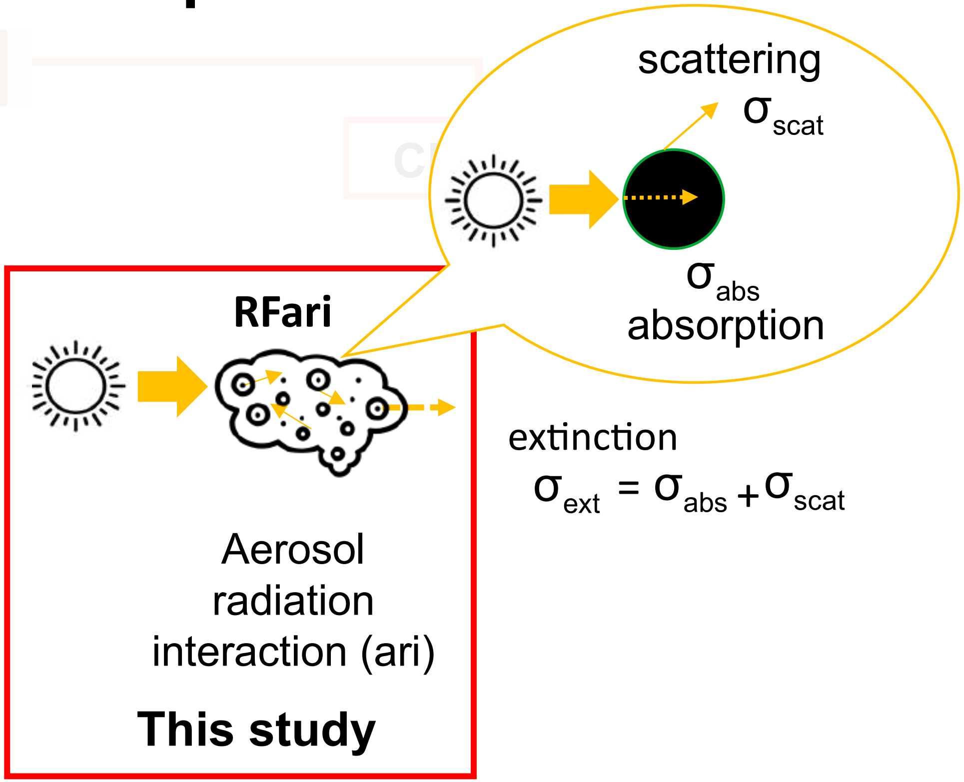
Aerosol
radiation
interaction (ari)

This study

Aerosols and their impacts

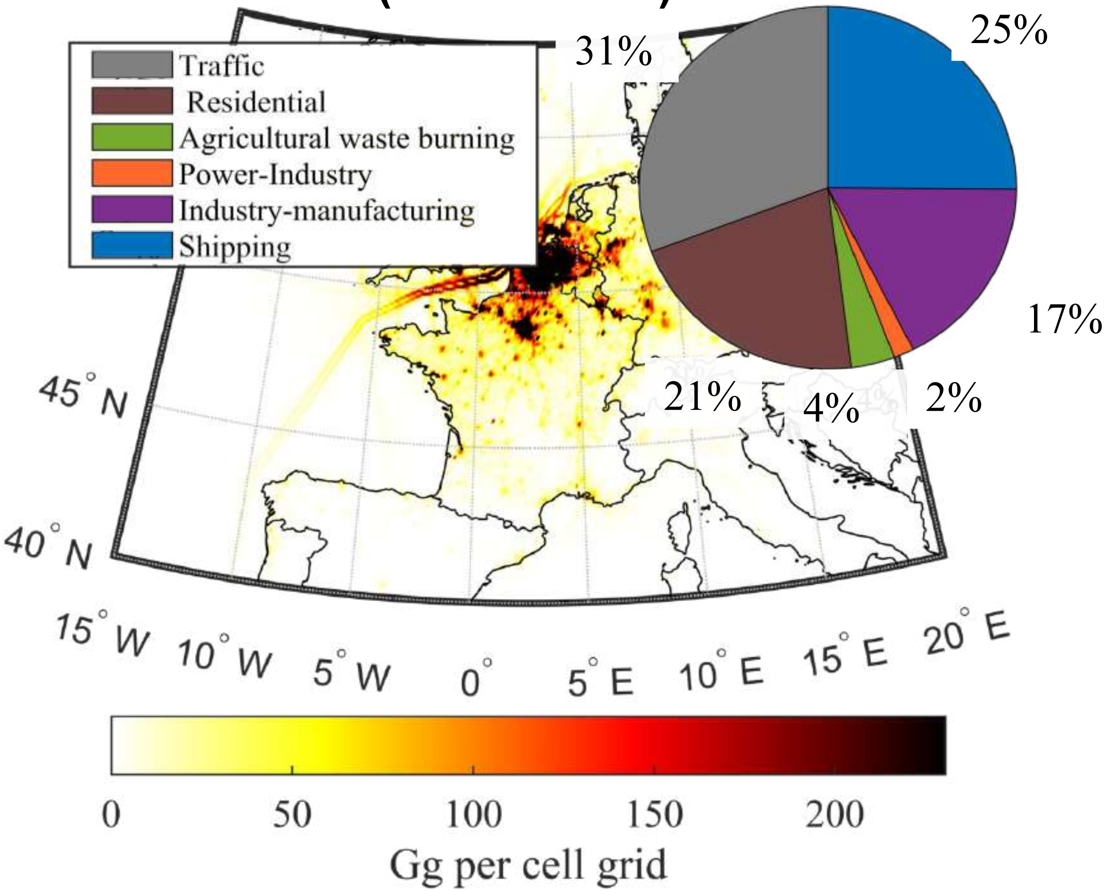
AR5 – IPCC (2018) best estimates
for the 1750–2010 period





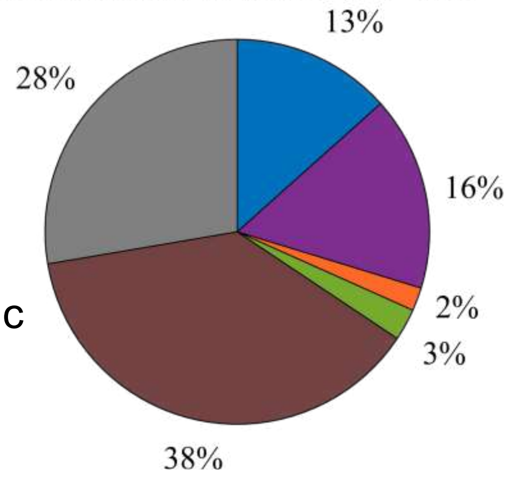
Spatial contribution to BC level in ATOLL

Multiannual (2016 – 2019)



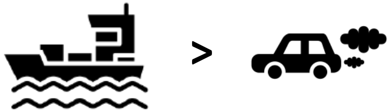
Winter →

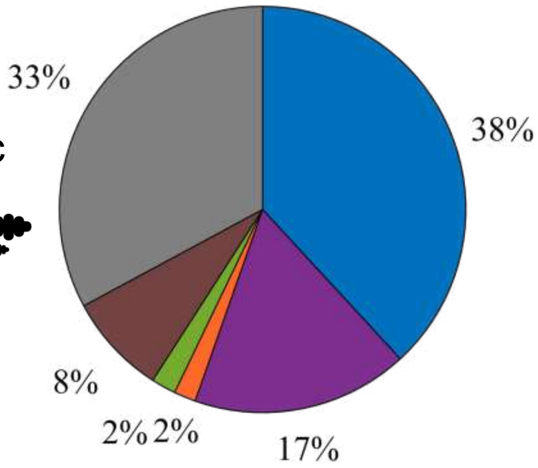
Main contributor:
- Residential > Traffic



Summer →

Main contributor:
- Shipping > Traffic





ATOLL platform

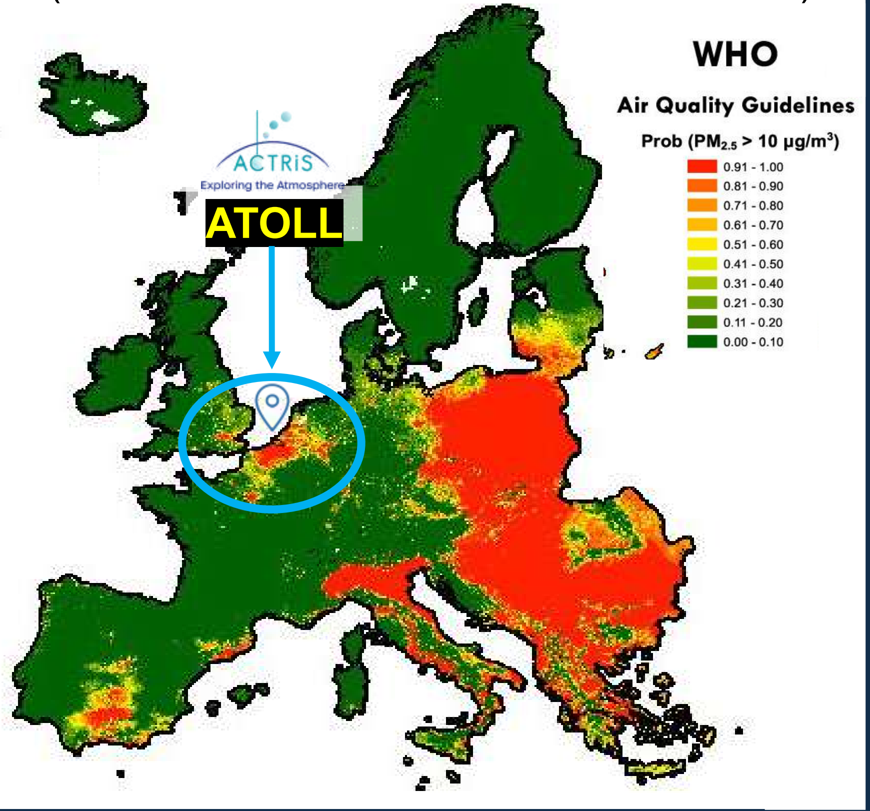
Long-term observations:
chemical & optical properties

ATOLL (ATmospheric Observation in liLLe)
50.6111 °N, 3.1404 °E, 70 m a.s.l.

The **North of France** suffers from significant **particulate pollution events**

- Heavily **populated area**
- Influence of **anthropogenic sources**
- Frequently polluted **continental plumes** arriving to the site

Exceedance of $PM_{2.5}$ concentrations in 2019
(Beloconi et al. 2021 Environ. Sci. Technol)



ATOLL platform

Since 2014

- **Aerosol In situ at the surface**

Since 90's

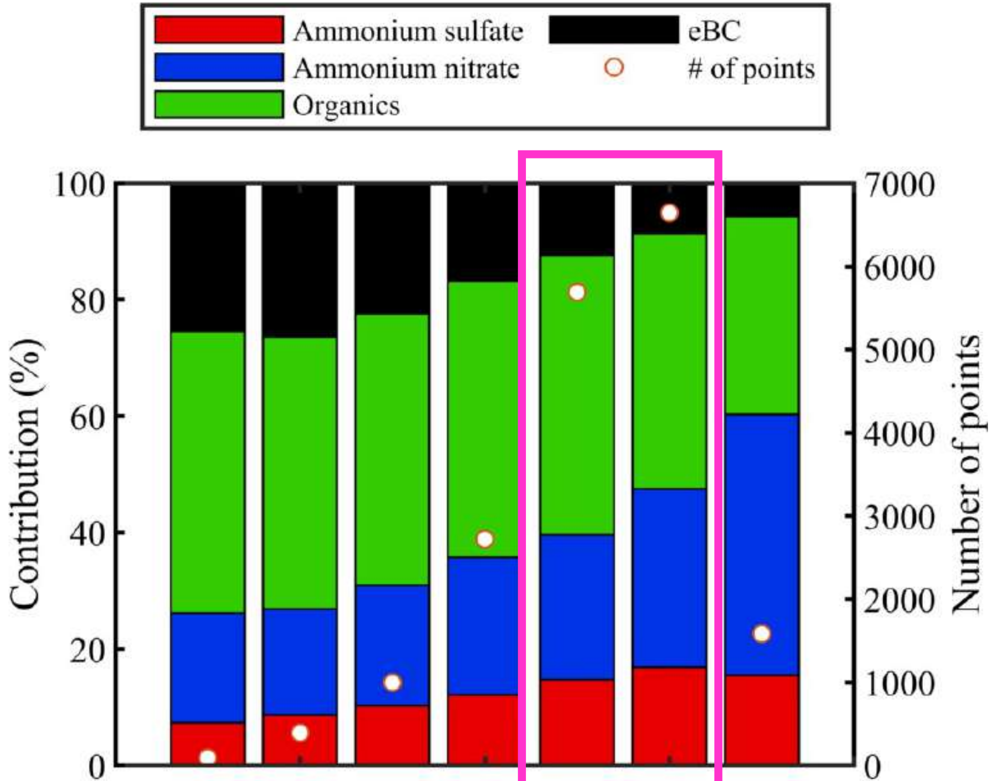
- **Remote sensing** (vertically integrated or resolved)

P5 roof top
Université Lille - Cité Scientifique
Villeneuve d'Ascq Cedex

Chemical contribution to the Aerosol Optical Properties (AOPs)

Ammonium sulfate Ammonium nitrate Organics BC

PM$_1$ fractional composition according to SSA$_{PM1}$ at 525nm:



AAE sensitivity test

Calculation based in observations:

BC ff (1.2)

Conditions:

- Summer (no influence of BB)
- Work days
- Traffic peak (06h00 – 09h00)

BC wb (1.6)

Conditions:

- Winter (influence of BB)
- House heating peak (22h00 – 00h00)