# Origine des particules fines en région parisienne: Résultats des campagnes de terrain et modélisation numérique


**M. Beekmann[1],**
**H. Petetin[1,8], Q.J. Zhang[1,9], A. S. H. Prevot[2], J. Sciare[3], V. Gros[3], V. Ghersi[4], A.Rosso[4], M. Crippa[2], F. Freutel[5], L. Poulain[6] , F. Drewnick[5], A. Borbon[1], A. Fortems-Cheiney[1], G. Dufour[1], A. Wiedensohler[6] , S. N Pandis[7] , and U. Baltensperger[2]**

*and the MEGAPOLI Paris campaign teams*

*[1]LISA/IPSL, France, [2]PSI, Switzerland. [3]LSCE/IPSL, France, [4]AIRPARIF, France, [5]MPI-Chemistry, Germany. [6]IfT, Germany [7]FORTH, Greece, [8] now Laboratoire d'Aérologie, France, [9]now at Aria Technologies, France.*



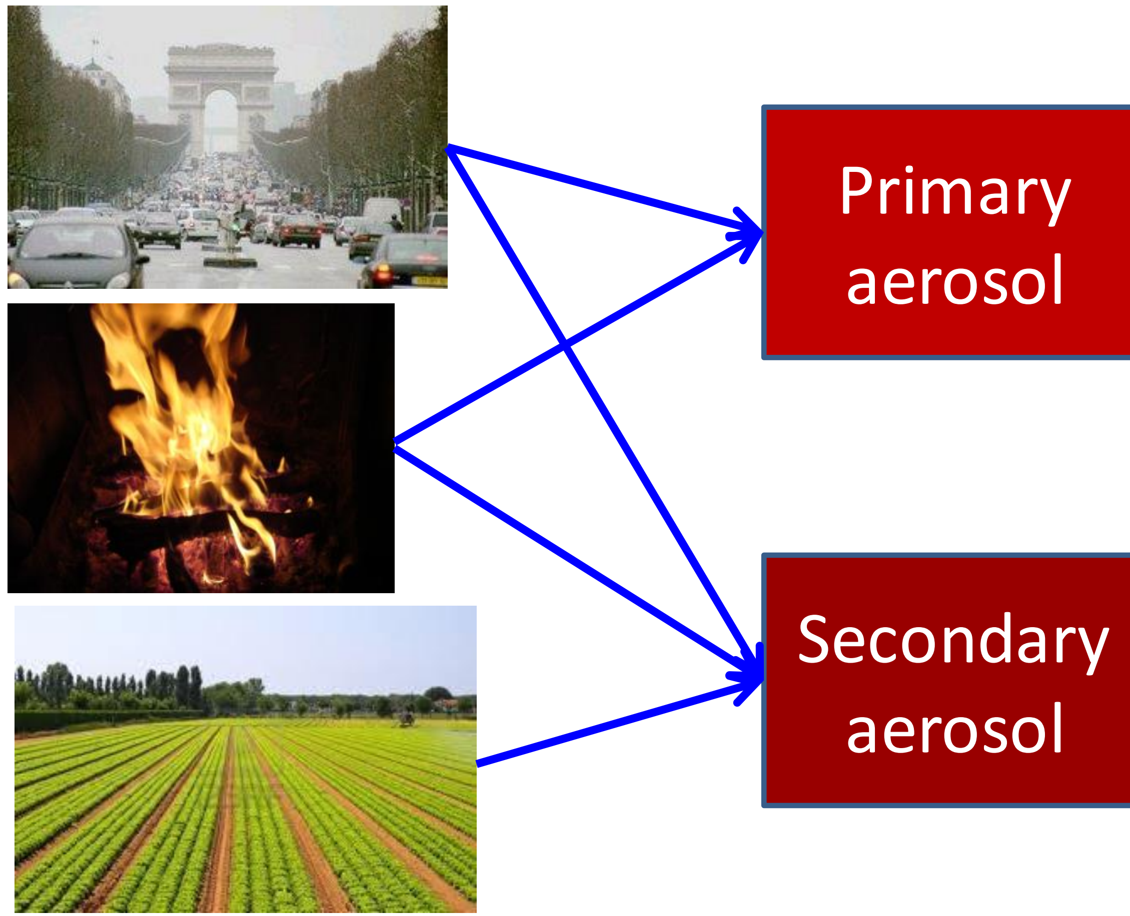
*Journée OCAPI, Pôle Observations de l'IPSL, 10 Décembre 2015*

# *Context:*

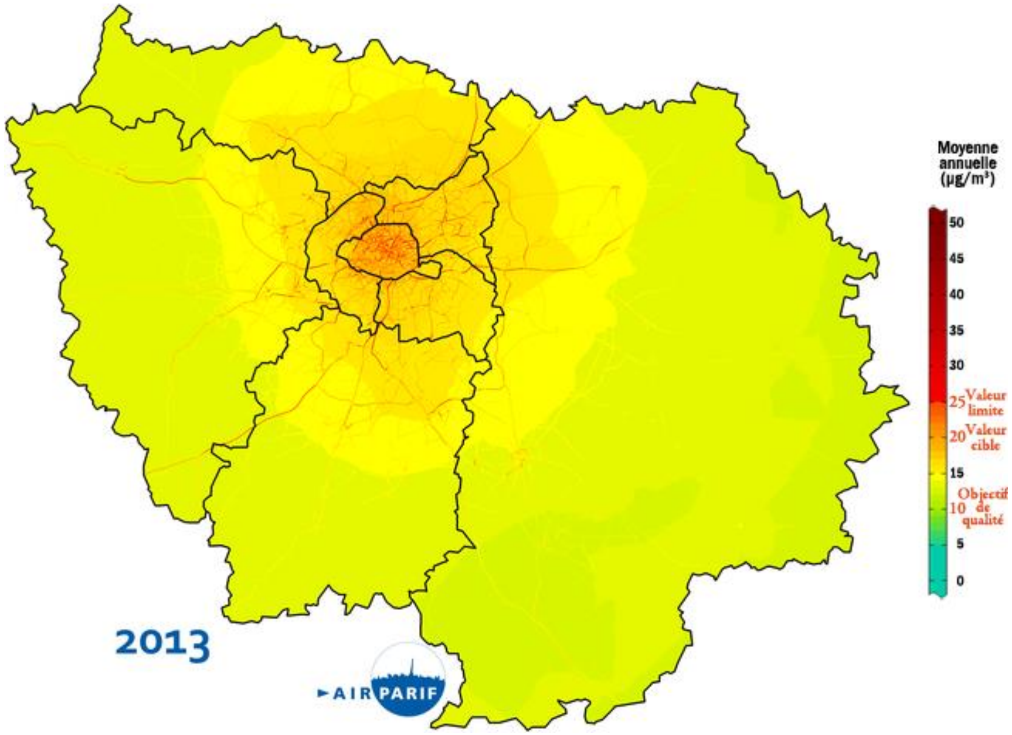
## *Still large uncertainties on origin of primary and secondary particulate matter*

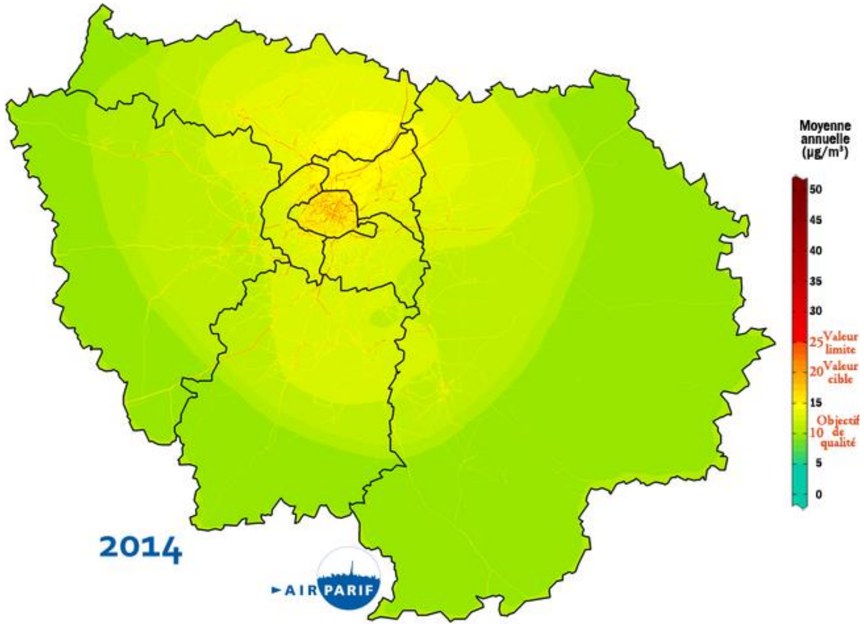
*Source sectors*

- *Traffic, industry*
- *Wood burning, fires*
- *Agriculture*
- *Forests*



# Qualité de l’air en Ile de France:
## champs annuels des particules fines PM2.5



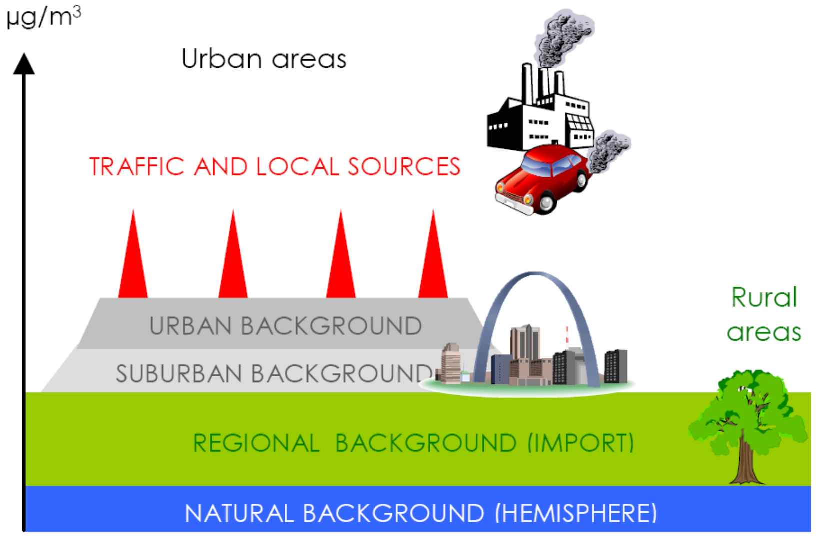
- Data sets :

- Megapoli intensive campaign: Paris region, 7/2009, 1,2/2010, ground based + mobile + aircraft facilities, aerosol + gaz characterisation
- Particules, Paris region: 1 year 2009-2010, PM2.5 composition on several sites
- Francipol: Paris region, spring, summer 2010, 1 urban site, VOC's , $NH_3$, $HNO_3$

- Model simulations:

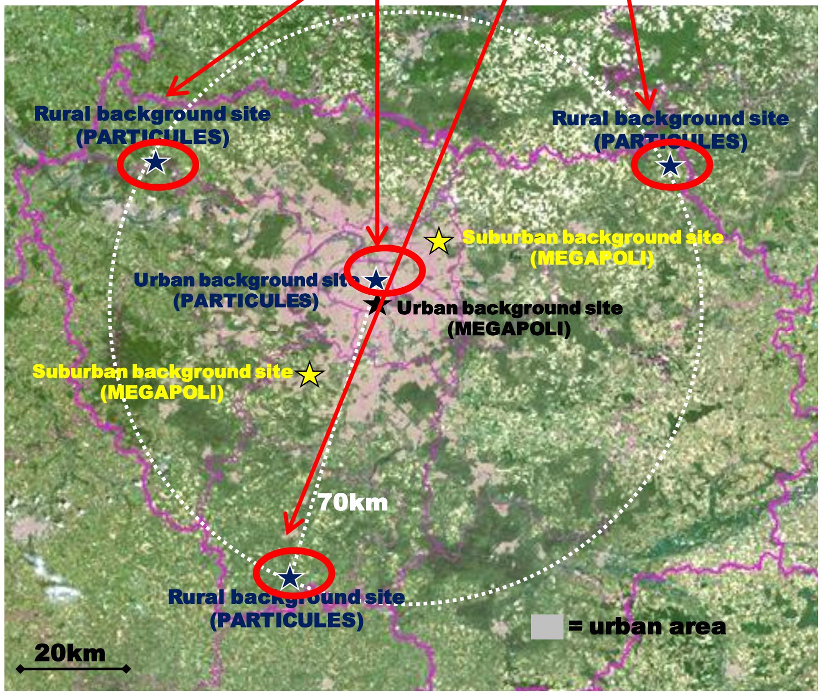
- CHIMERE regional chemistry transport model

# Local versus Regional contribution of $PM_{2.5}$




Subtractive approach

$$[PM_{2.5}]_{PARIS} = [PM_{2.5}]_{urban} - [PM_{2.5}]_{rural}$$

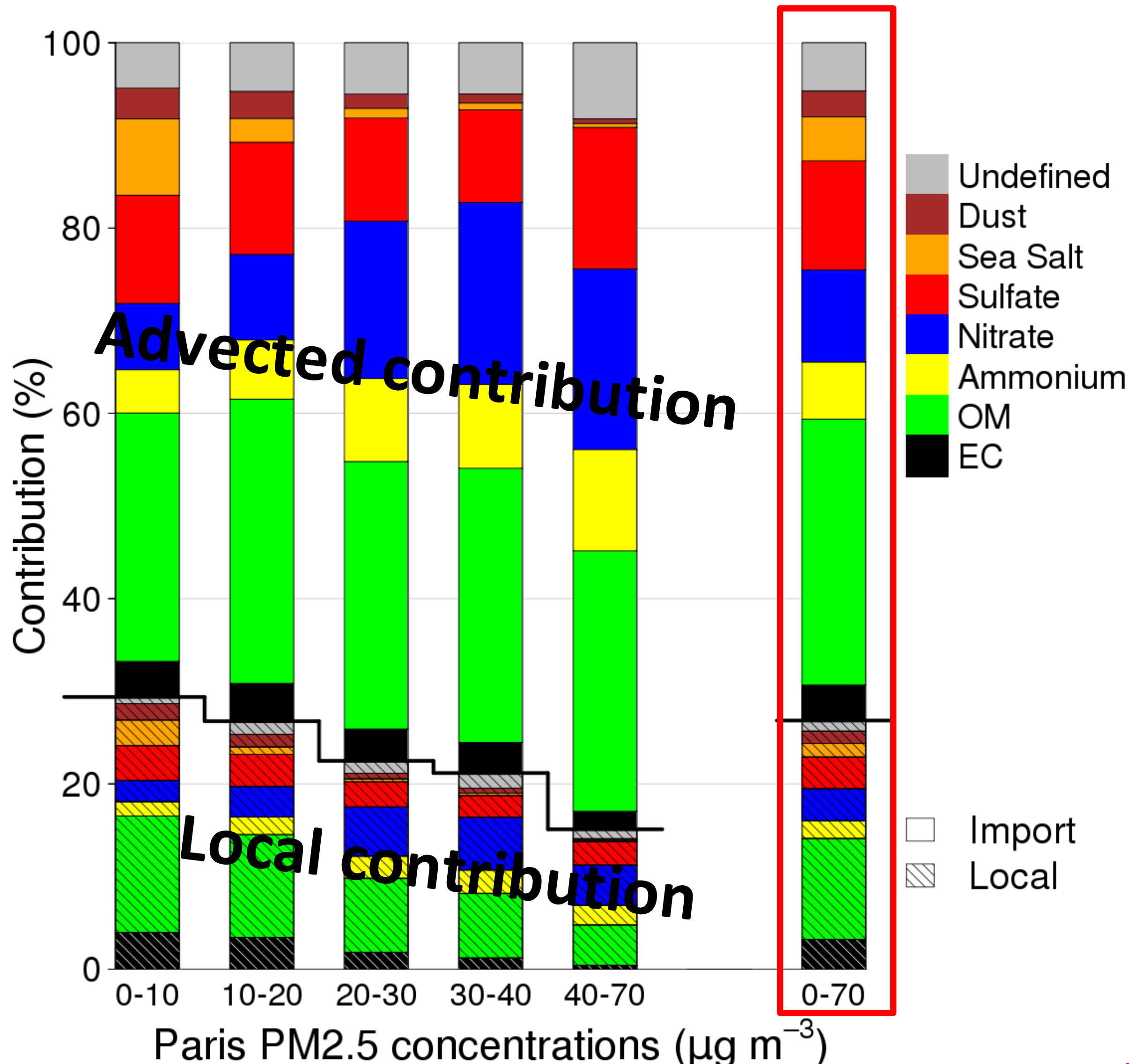



Results from the Particules campaign
September 2009 – September 2010
Daily PM2.5 major components
from filter samples + PILS system + Sunset

This approach has been applied earlier
for Berlin (Lenschow et al., 2001)

Bressi et al., ACP 2013

# Local versus Imported Contribution to PM2.5



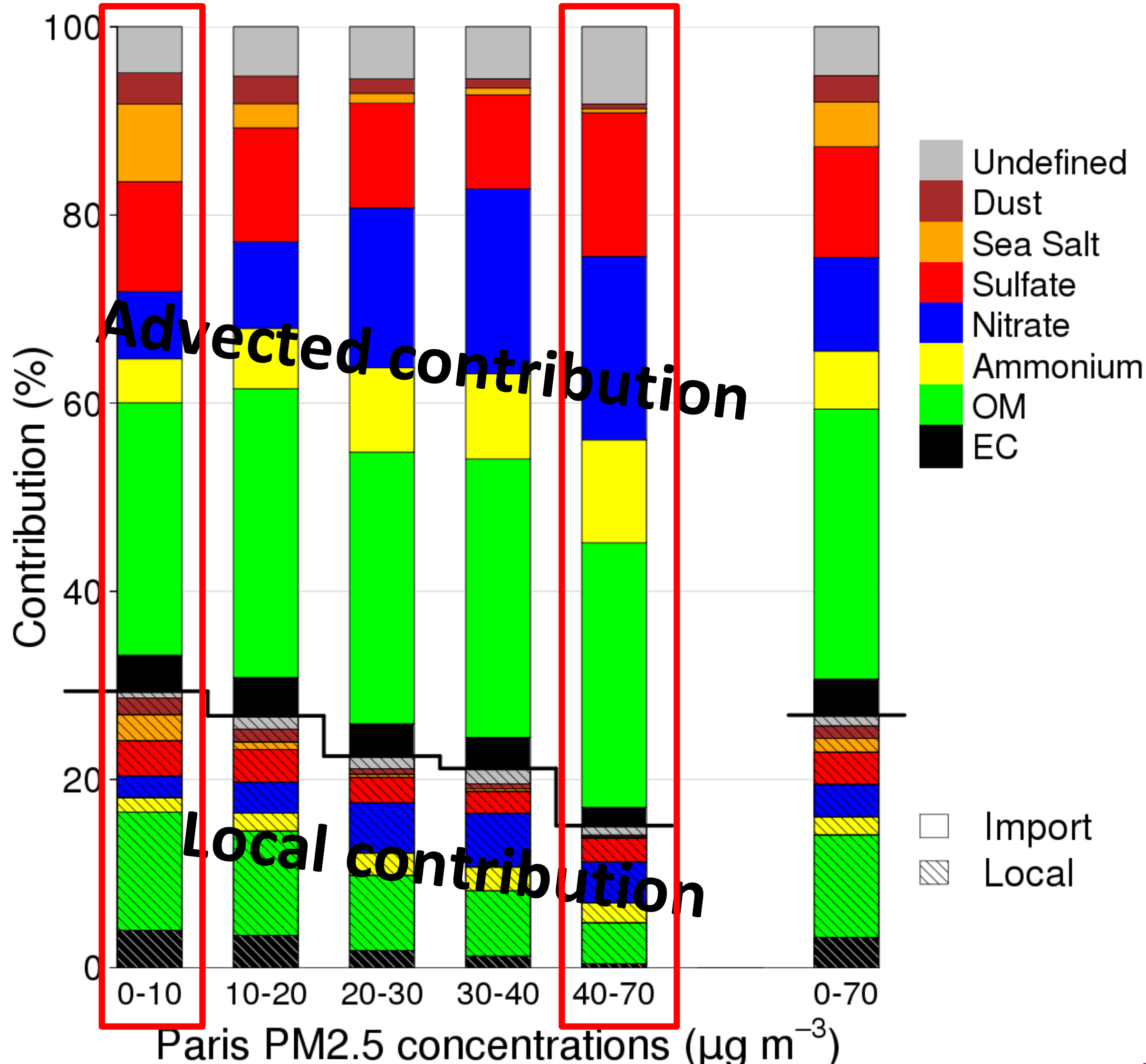

At Paris urban downtown site, September 2009 – August 2010 average

**⇒ ~70 % of PM2.5 is imported from outside the agglomeration**

**⇒ this was an expected result for sulfate and nitrate, but it is new for organic aerosol**

**Beekmann et al., ACP, 2015, Petetin et al., GMD, 2014**

# Local versus Imported Contribution to PM2.5




At Paris urban downtown site, September 2009 – August 2010 average

**⇒ ~70 % of PM2.5 is imported from outside the agglomeration**

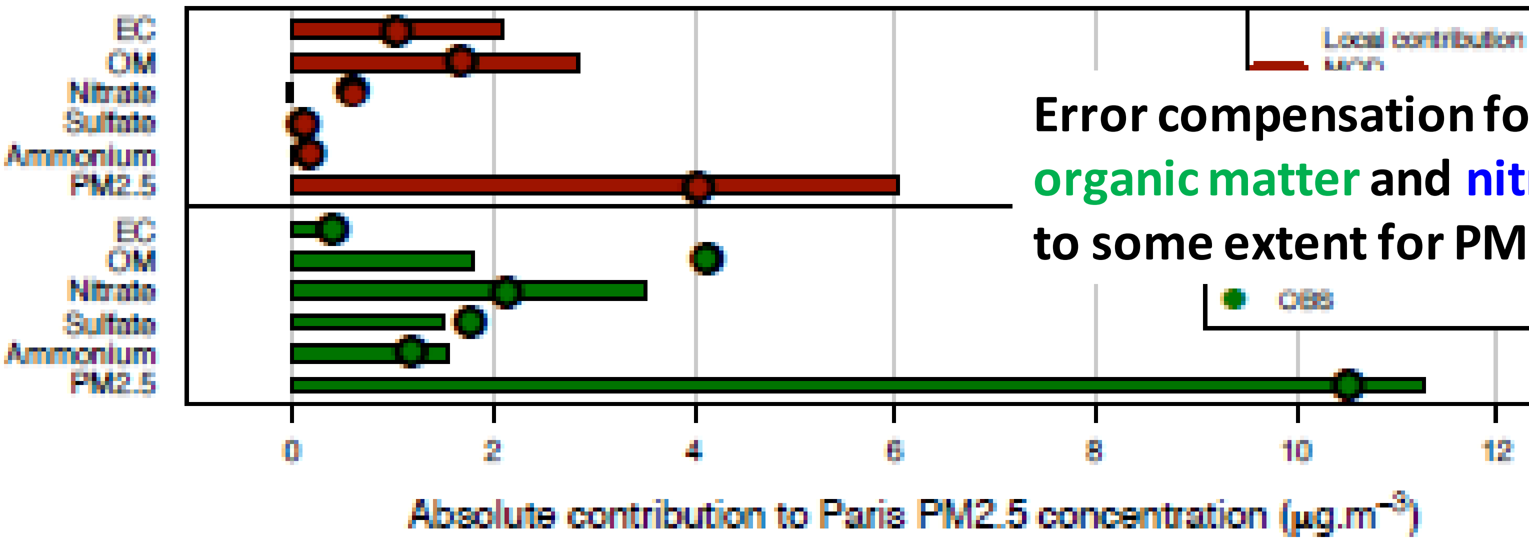
**⇒even more for large PM2.5 concentrations**

**⇒PM2.5 variability is mainly advection controlled**

**Beekmann et al., ACP, 2015, Petetin et al., GMD, 2014**

# A novel approach for air quality model evaluation

⇒ **Use derived observations of urban local and advected PM concentrations for model evaluation**




**Error compensation for organic matter and nitrate, to some extent for PM2.5**

**Mean (Sep 2009 – Sep 2010) local (top, red) and advected (bottom, green) contributions to greater Paris PM2.5 urban background, for the CHIMERE model (bars) and observations (filled circles)**

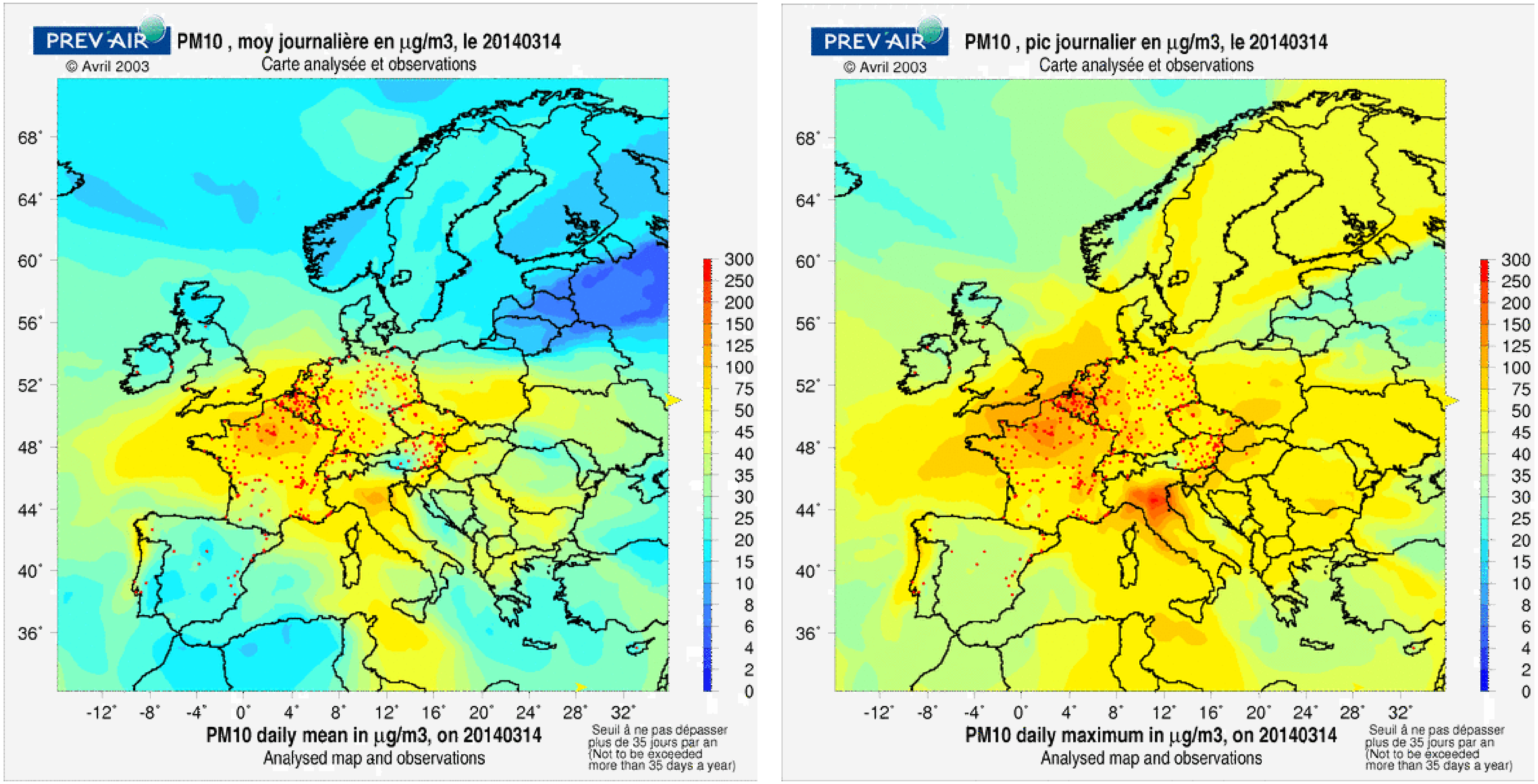
**Petetin et al., GMD, 2014**

Chimere LMD INERIS lisa [www.lmd.polytechnique.fr/chimere/](www.lmd.polytechnique.fr/chimere/)

**Menut et al. 2014 (GMD 6)**

# **Origin of ammonium nitrate aerosol**
## example of spring 2014 pollution period

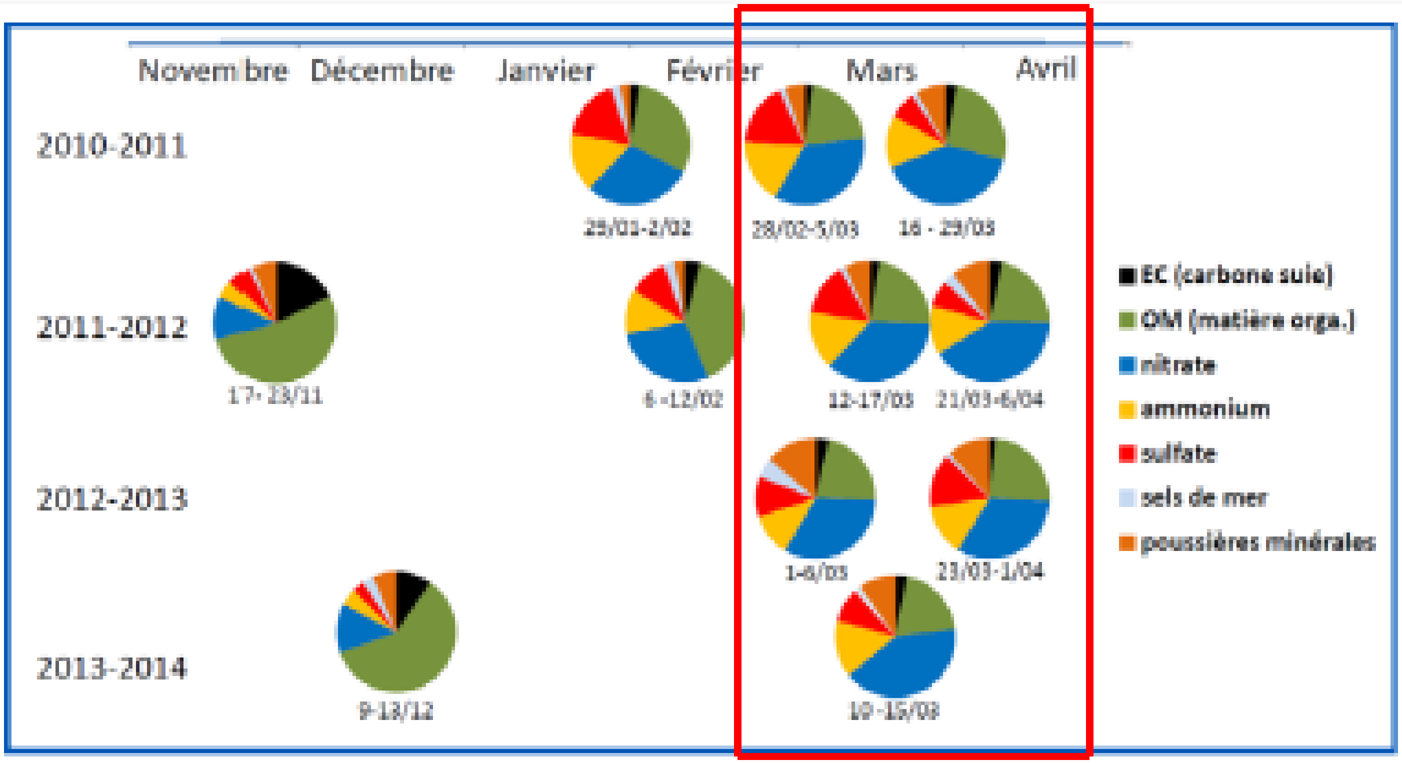
**Analysed daily average $PM_{10}$** **Analysed daily peak $PM_{10}$**




**PREVAIR PM10 *analysis*, based on CHIMERE air quality model simulations and assimilation of Airbase surface observations**

http://www.prevair.org/en

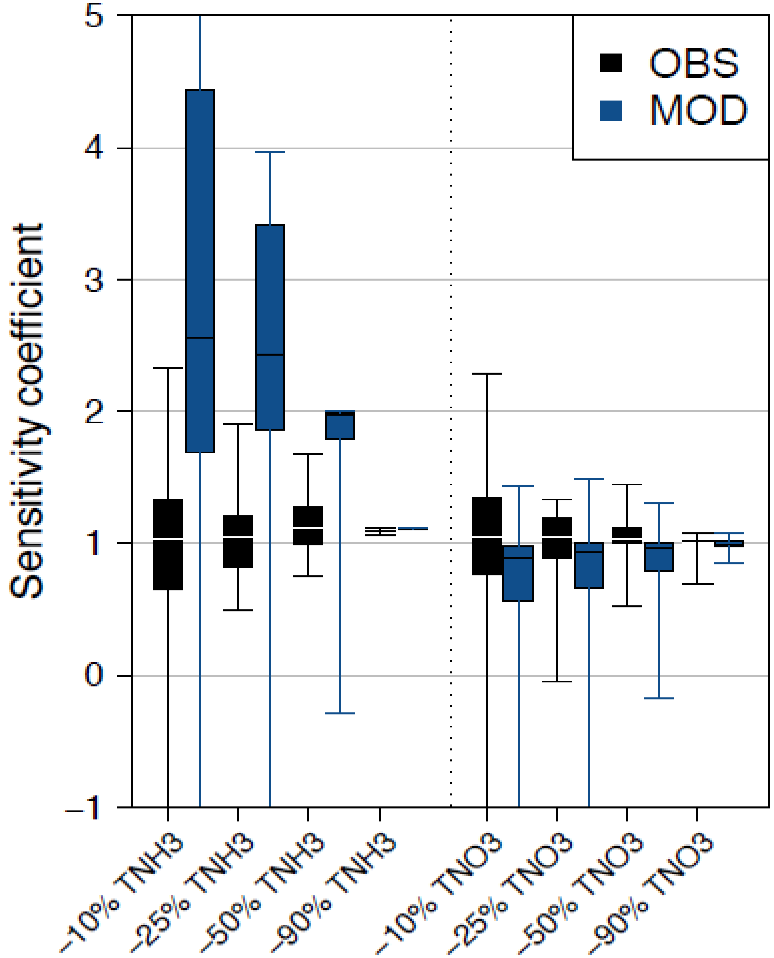
# PM2.5 composition during pollution episodes




***=> March , April : ~50% NH4 NO3***

**PM2.5 aerosol composition at a rural background site (Petit-Quevilly )during the 10 largest PM pollution episodes between 2010 and 2014**
**(Source Air Normand)**

# Simulated and observed local sensitivity of nitrate formation to precursors in Paris



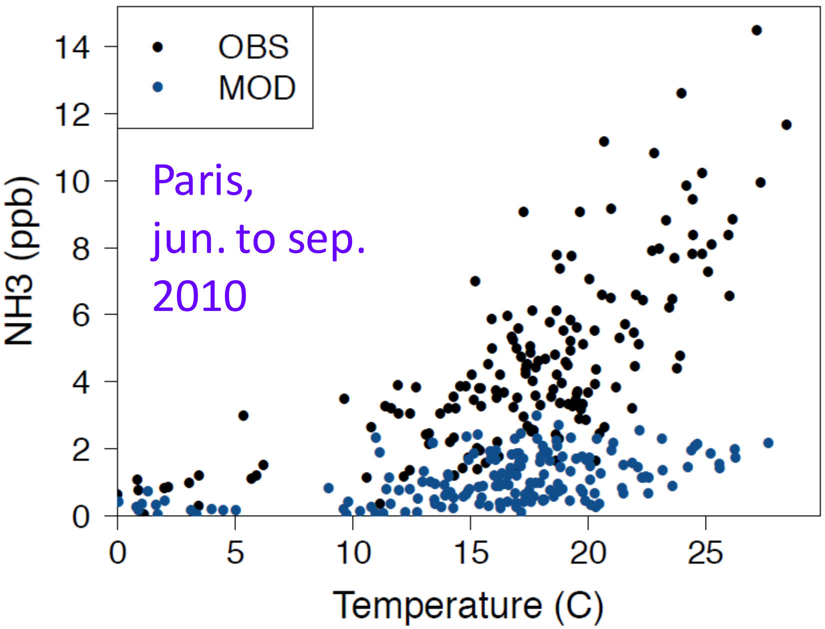

Sensitivity coefficient $S_x$:

$$S_x = \frac{\Delta NO_3}{NO_3} \frac{x}{\Delta x}$$

calculated with the ISOROPIA model from observed and simulated

$TNH_3 = NH_3 (g) + NH_4^+ (p)$

$THNO_3 = HNO_3 (g) + NO_3^- (p)$

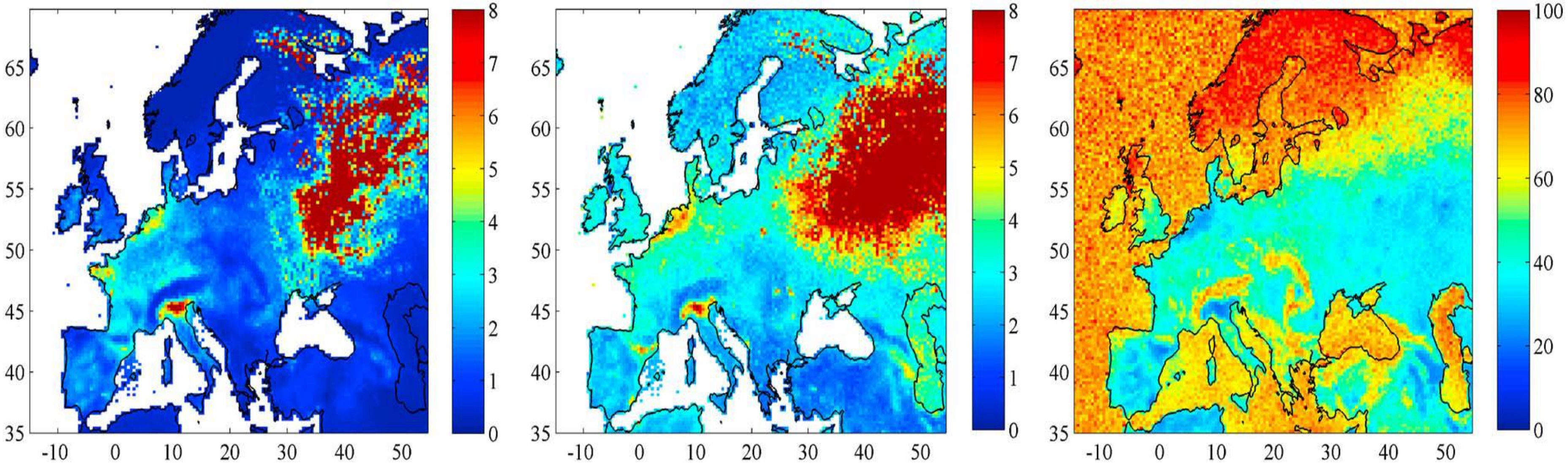



Petetin et al., ACPD, 2015

*How to improve from this?*

*=> include temperature dependence in emission estimates by using explicit NH3 evaporation model => link VOLT'AIR + CHIMERE* **Hamaoui et al.2014**

*=> use NH3 satellite observations to infer NH3 emissions (Cheiney et al., in progress)*




**LOTUS-EUROS Simulation**

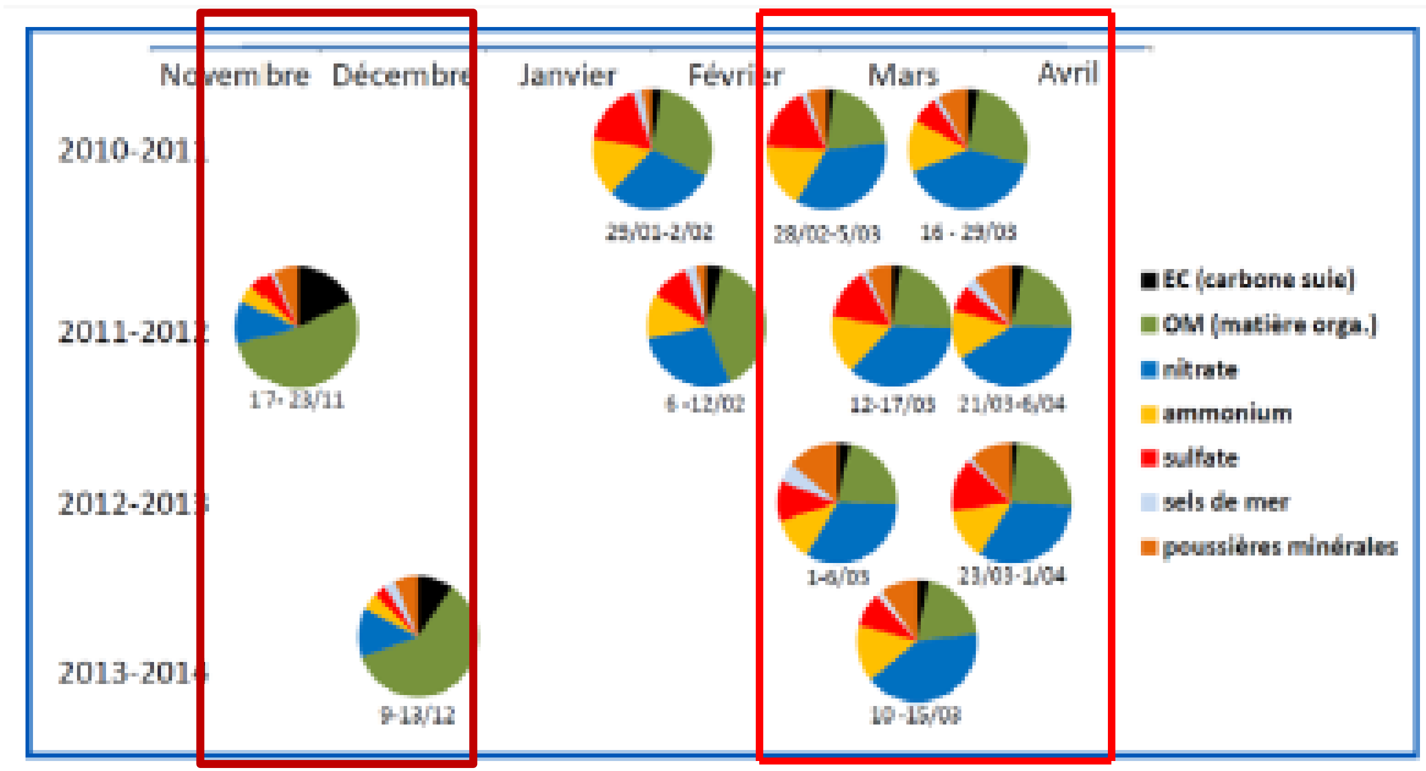
**IASI observation**

**IASI error**

**Atmospheric NH3 columns (mg/m2) between 2008 et 2011**

Van Damme et al., 2014

# PM2.5 composition during pollution episodes



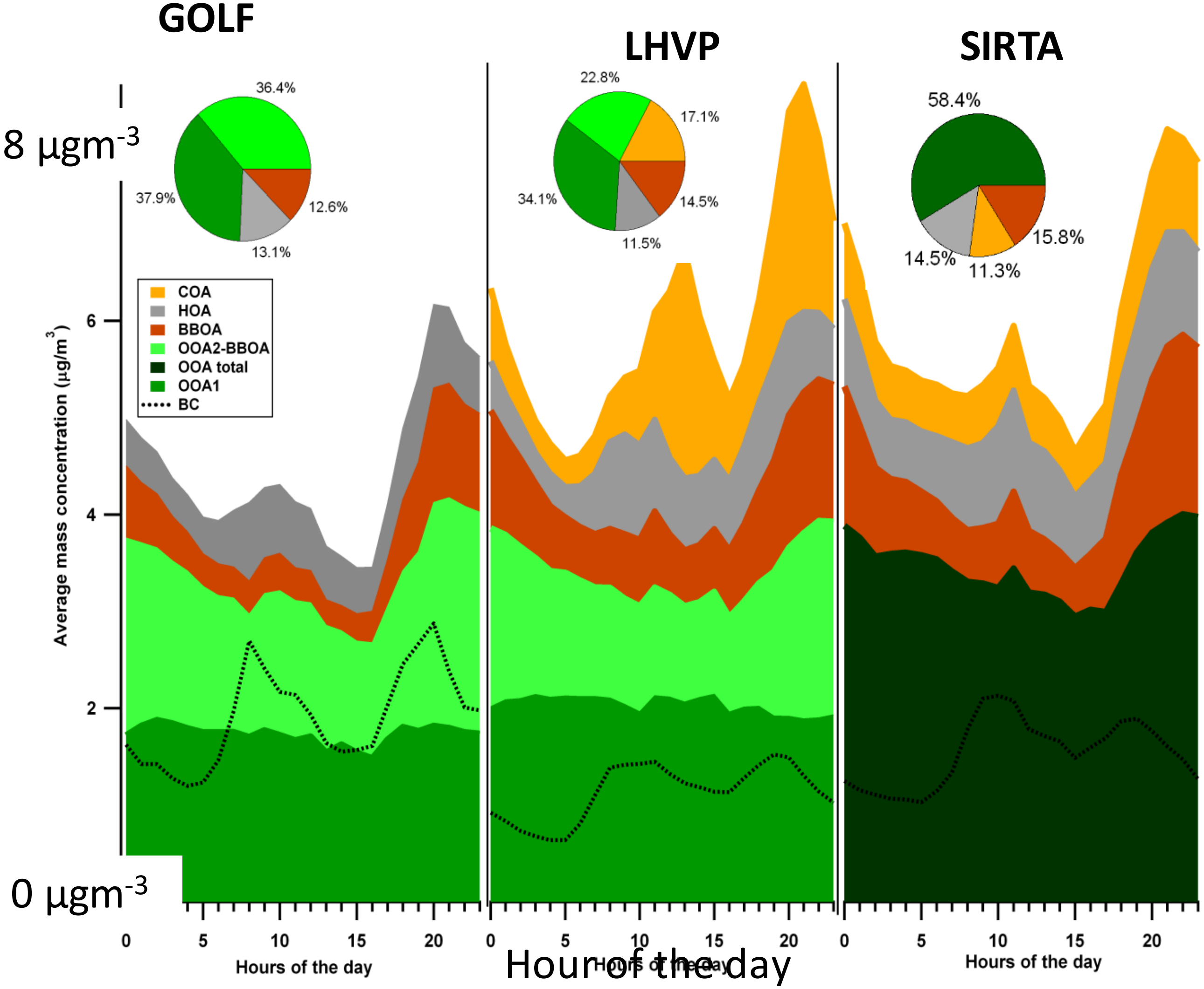

***=> Winter : major organic matter origin***

**PM2.5 aerosol composition at a rural background site (Petit-Quevilly )during the 10 largest PM pollution episodes between 2010 and 2014**

**(Source Air Normand)**

# Diurnal cycles of organic aerosols sources MEGAPOLI winter campaign (PM1)

**From Positive Matrix Factorization of Aerosol Mass Spectrometer measurements**


**Primary :**

**cooking =11-17%**

**(35% for meal hours)**

**traffic=11-13%;**

**woodburning=13-16%;**

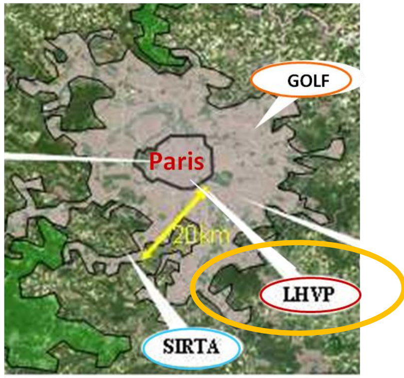
**Secondary :**

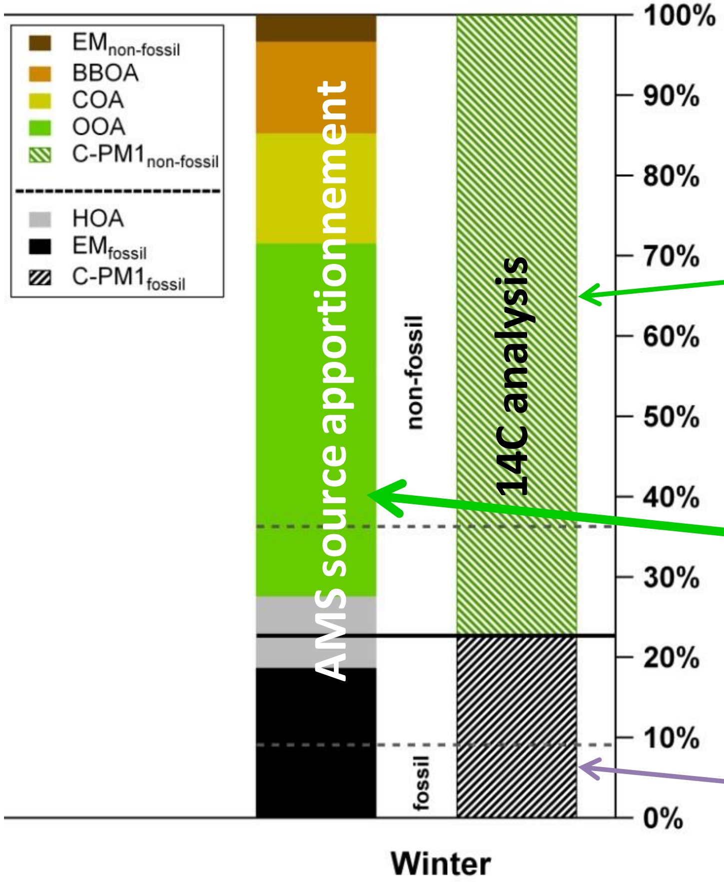
**22- 36% woodburning**

**unidentified non-fossil 34-38%**

Crippa et al., 2013a,b, c

# Fossil fuel vs. non fossil carbonaceous aerosol (PM1) – LHVP winter



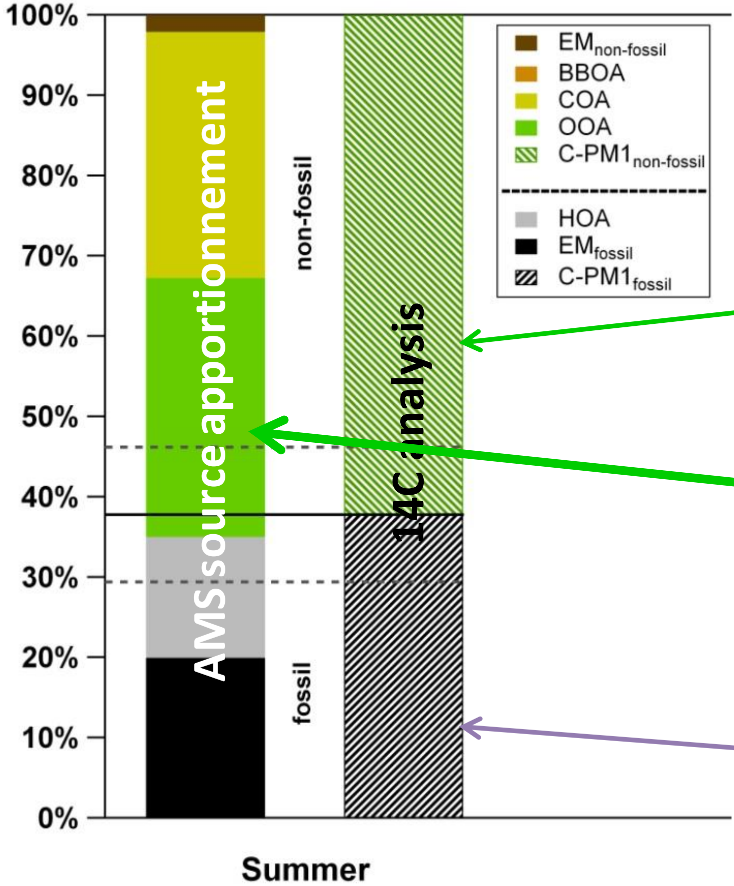

Non-fossil fuel C
*=> major fraction*

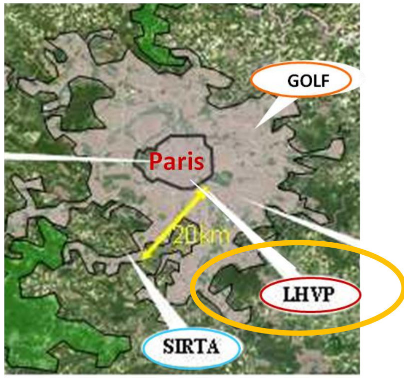
=> SOA is essentially of non-fossil origin from wood burning and /or biogenic emissions

Fossil fuel C
*=> minor fraction*

Beekmann et al., ACP,2015 P. Zotter, PSI, $^{14}$C measurements S. Szidat, Univ. Bern

# Fossil fuel vs. non fossil carbonaceous aerosol (PM1) – LHVP summer



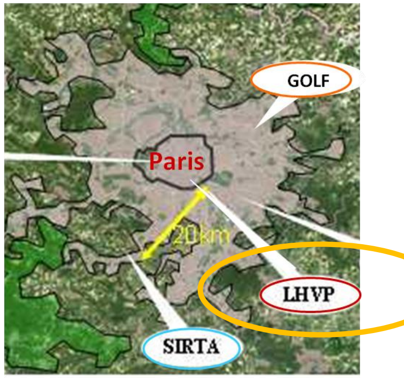

**Non-fossil fuel C**
***=> major fraction***

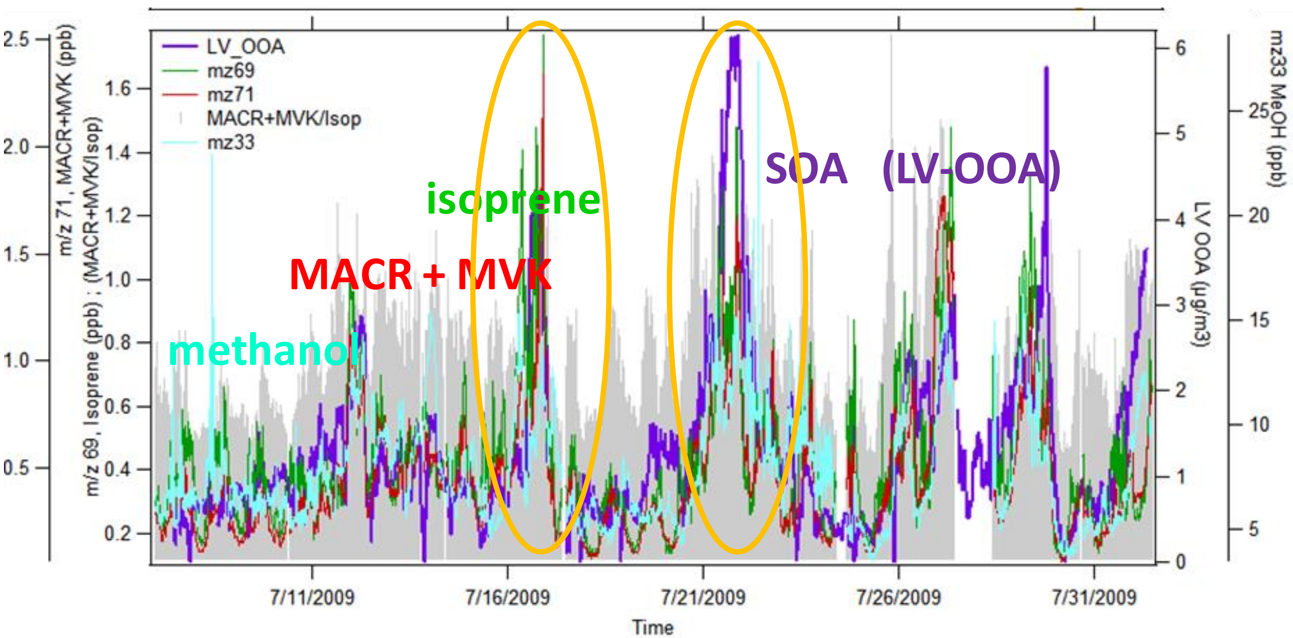
**=> SOA is of major non-fossil origin mostly from biogenic emissions**

**Fossil fuel C**
***=> minor fraction***

**Beekmann et al., ACP, 2015 P. Zotter, PSI, $^{14}C$ measurements S. Szidat, Univ. Bern**

# Secondary aerosol variability and biogenic emission tracers (PTRMS measurements) during the summer campaign

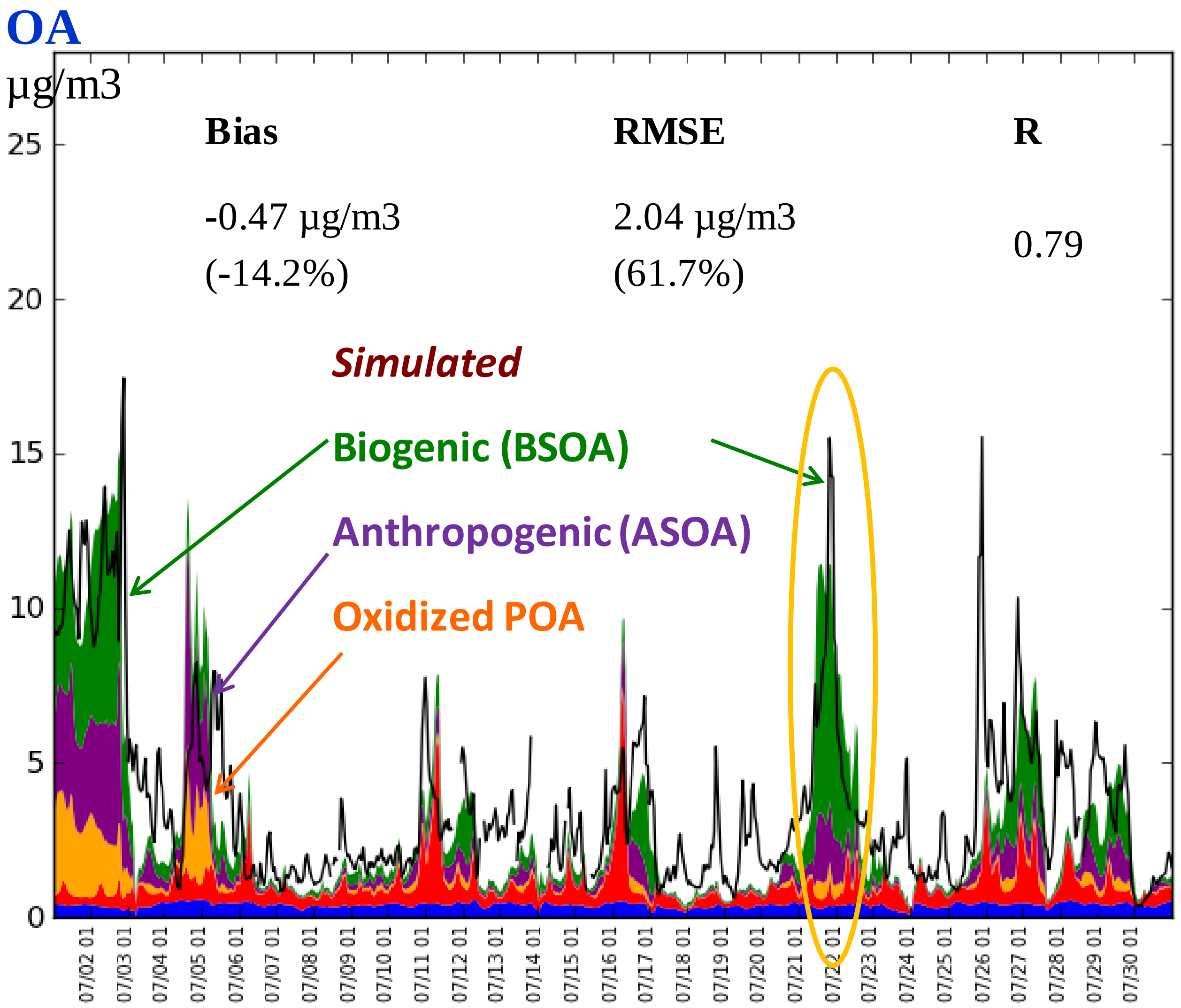


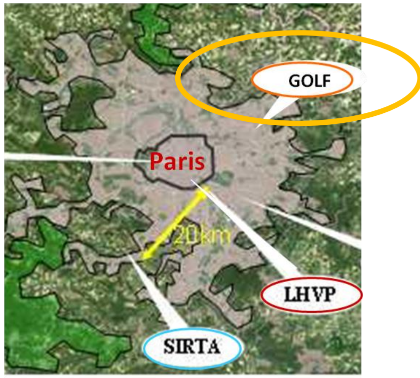




⇒ ***SOA peaks related to biogenic origin***

**Courtesy**
**N. Marchand, LCE,**
**L. Poulain, IfT,**
**M. Crippa, PSI**

# Organic aerosol modelling




**CHIMERE CTM**

**2) Volatility basis set**

POA volatile

POA + ASOA + BSOA

chemical aging

***Better agreement***

***Less POA***

***More SOA***

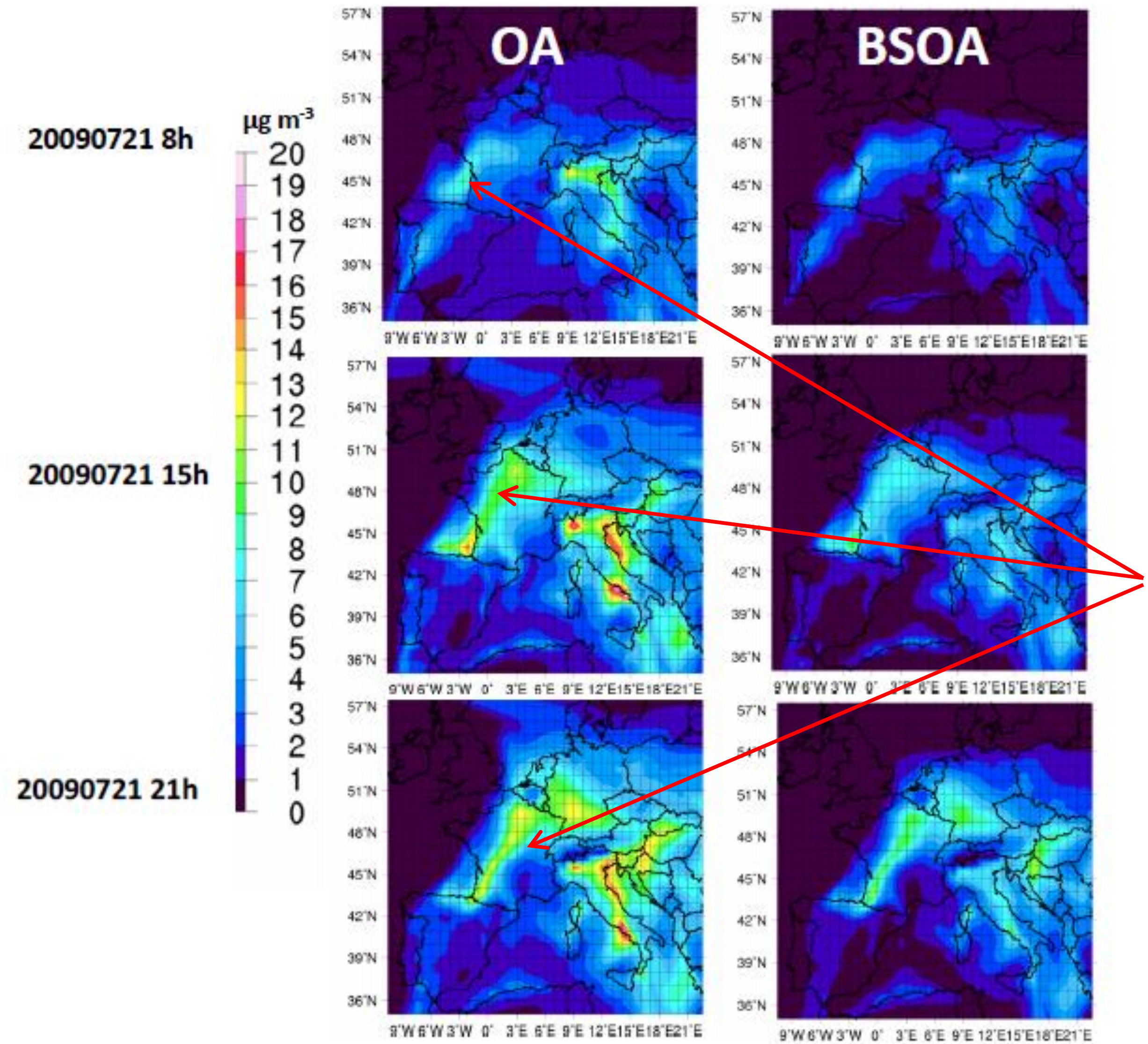
**Biogenic (BSOA)**

**Anthropogenic (ASOA)**

**Oxidized POA**

Time series July 2009

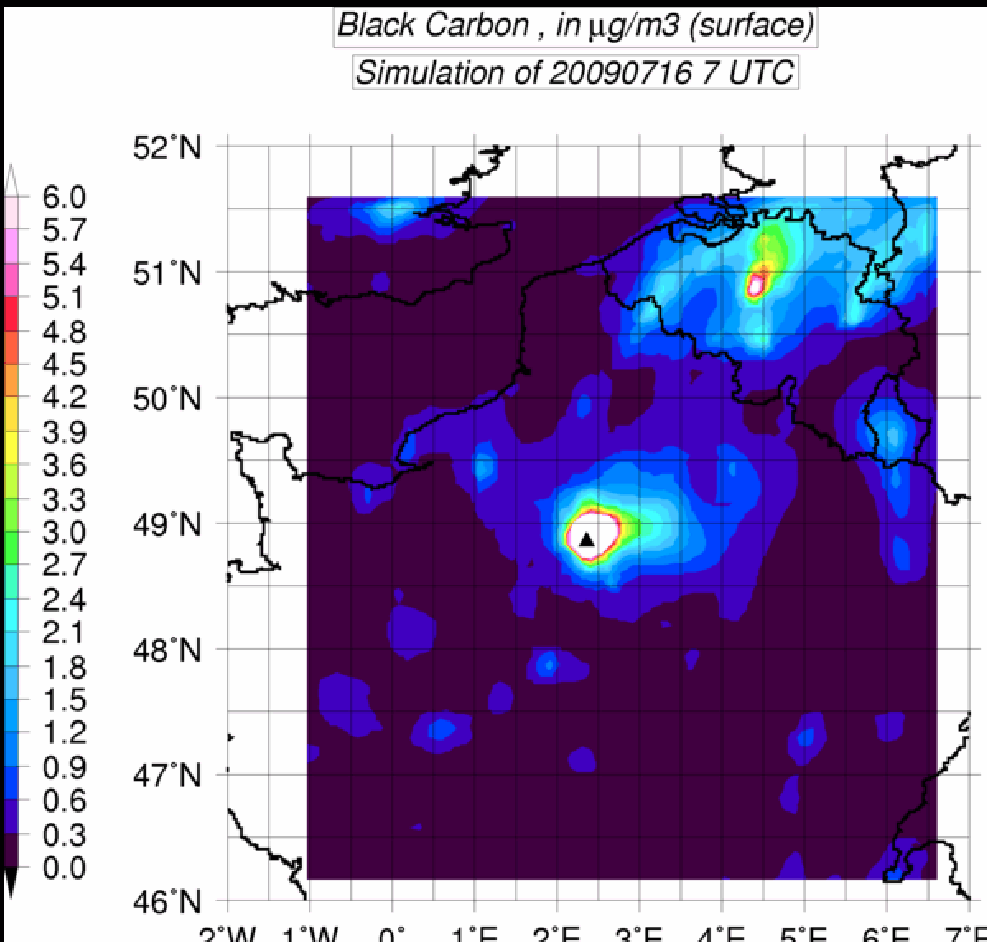
**Zhang et al., ACP, 2013**



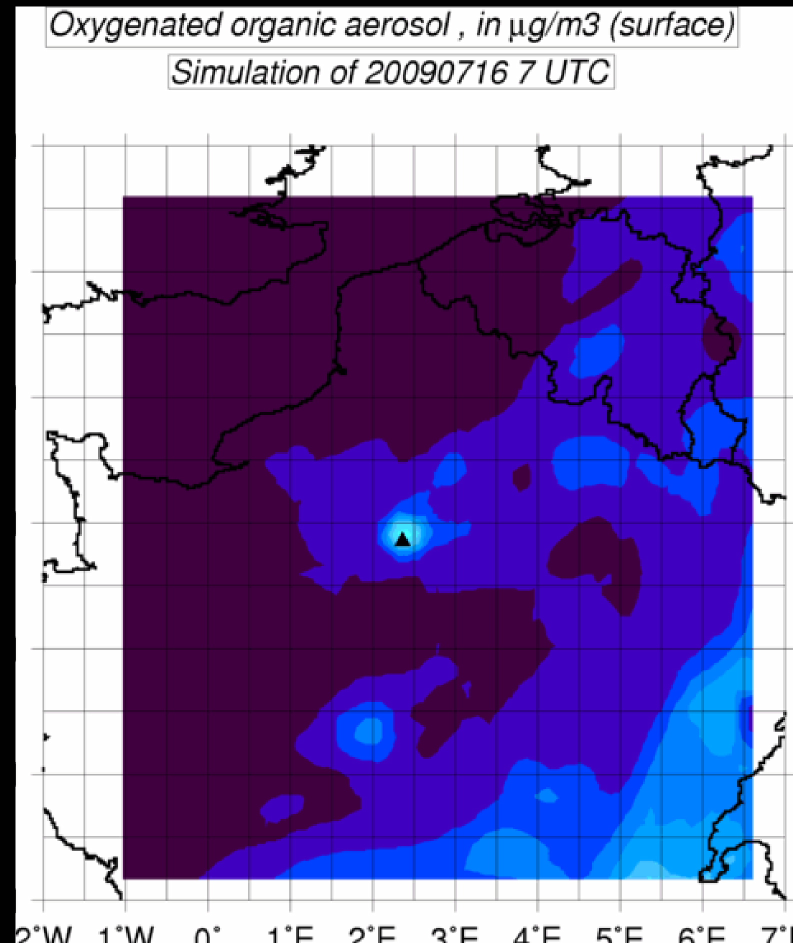

**Advection of BSOA formed from biogenic emissions in Northern Spain and South-western France to Paris**

# Simulation of Paris polluant plume - CHIMERE

## Black carbon



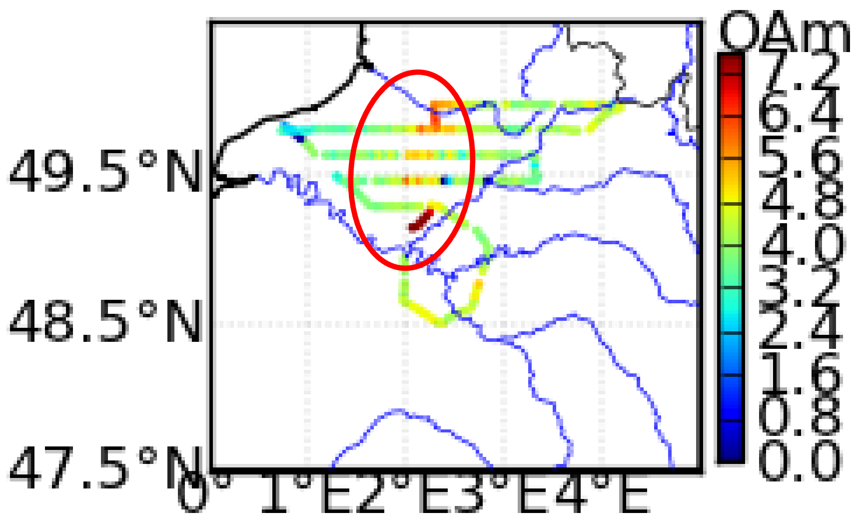

## Secondary organic aerosol



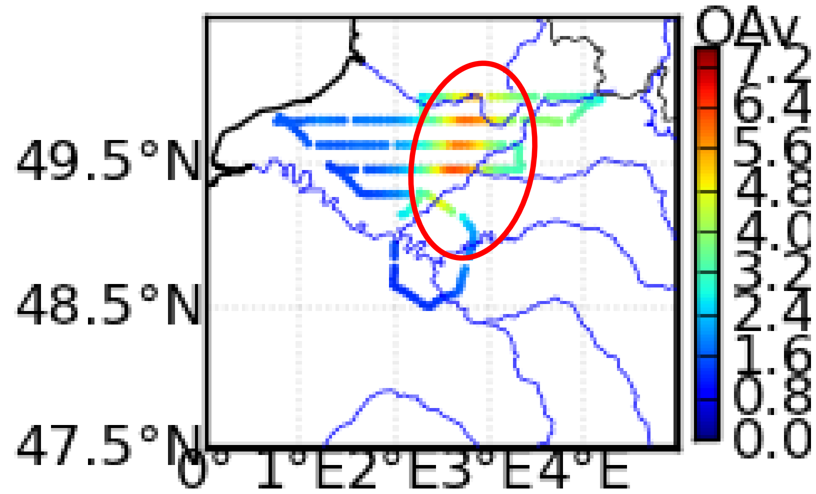

Simulations CHIMERE-VBS Q.J. Zhang, LISA/CNRS

# Paris plume developement (July 16)

**ATR-42 Airborne observed OA µg/m3**




**CHIMERE simulated OA µg/m**




**Freney et al., ACP, 2013**

***=> Simulations ~ right for the right reasons ?***

# SOA vs. Ox plots

SOA versus Ox ($O_3$+$NO_2$) plots can be used to normalise SOA formation with respect to precursor concentration and photochemical activity (Herndon et al. 2008)

COV + OH -> -> -> $\alpha_1$ Ox + ..... (for high pour NOx)

COV + OH -> -> -> $\alpha_2$ AOS + .....

Slope SOA vs. Ox correspond to $\alpha_1$ / $\alpha_2$

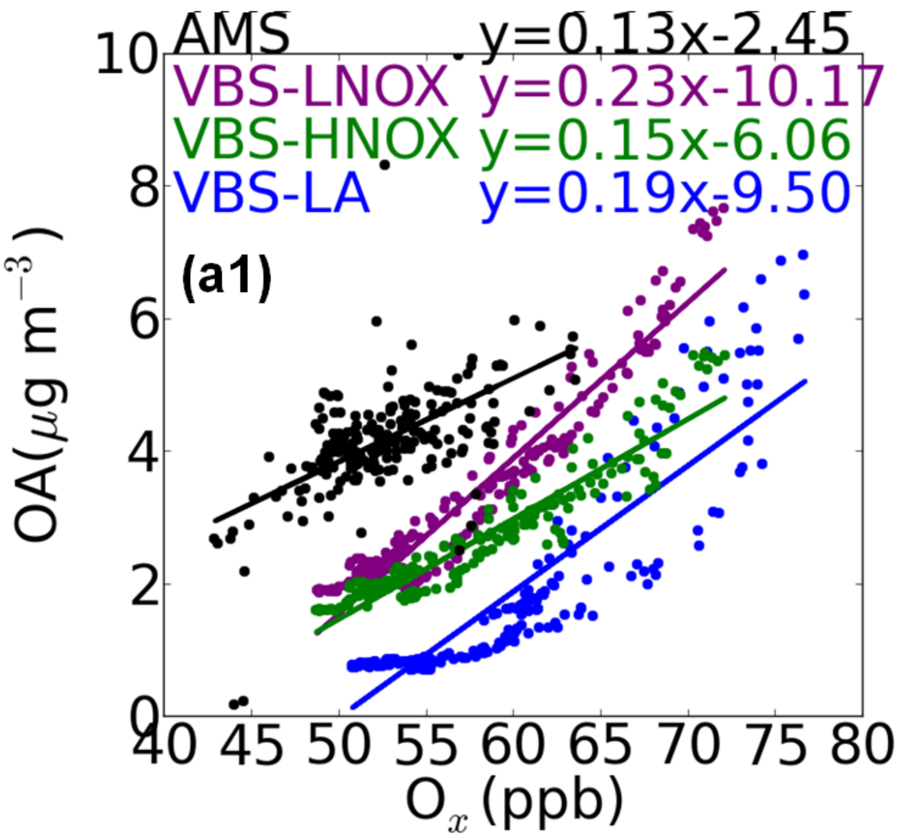
# SOA vs. Ox plots

SOA versus Ox ($O_3$+$NO_2$) plots can be used to normalise SOA formation with respect to precursor concentration and photochemical activity (Herndon et al. 2008)

COV + OH -> -> -> $\alpha_1$ Ox + ..... (for high pour NOx)

COV + OH -> -> -> $\alpha_2$ AOS + .....

Slope SOAvs. Ox correspond to $\alpha_1 / \alpha_2$




**AMS OA and Ox (O3+NO2) observations July 16**

**VBS-LNOX: Volatility basis set with high SVOC yield (low NOx)**

**VBS-HNOX: Volatility basis set with low SVOC yield (high NOx)**

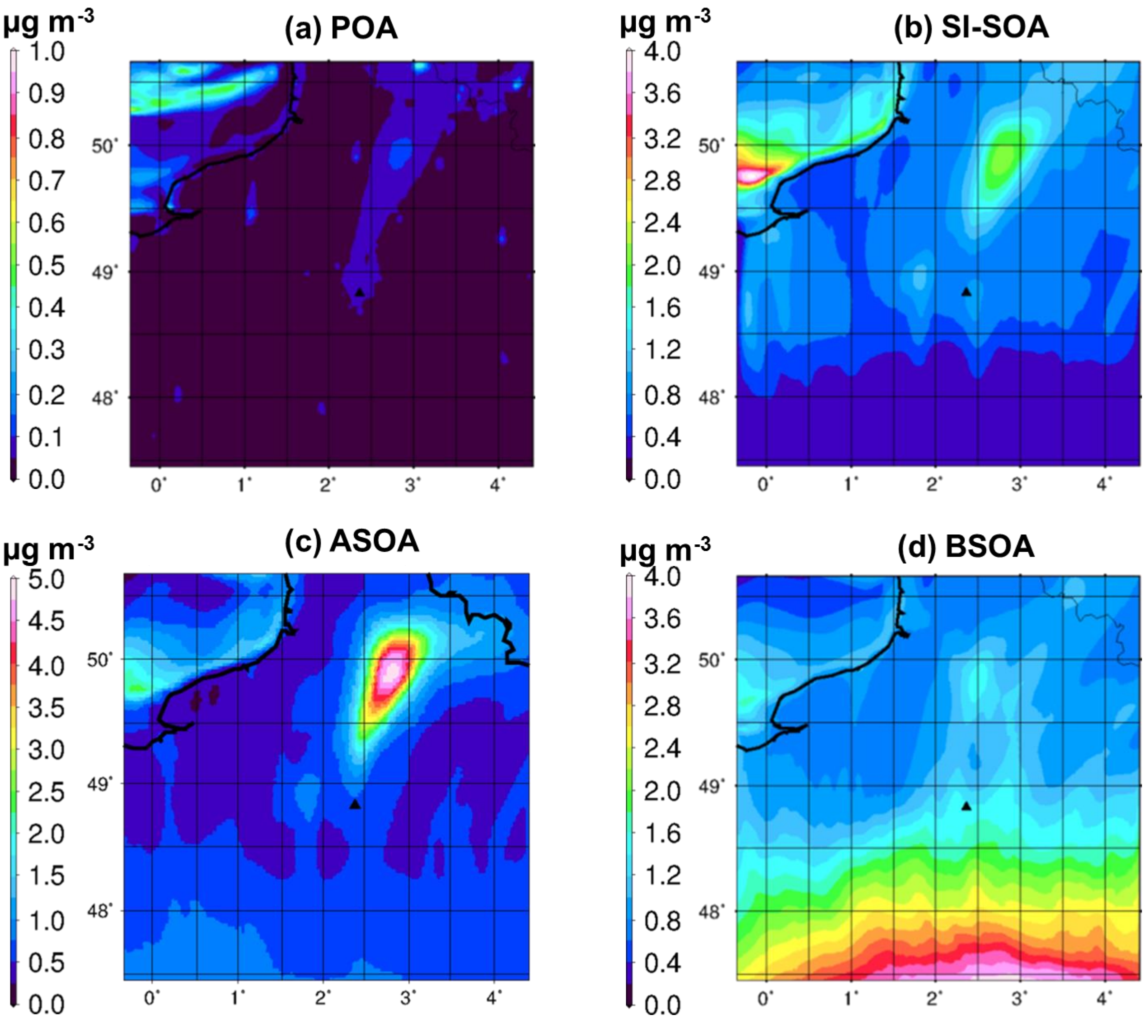
**VBS-LA: Volatility basis set with SI-SOA formation only, but with 3 times higher POA emissions**

***=> both VBS-LNOX and VBS-LA agree rather well with observed slope***

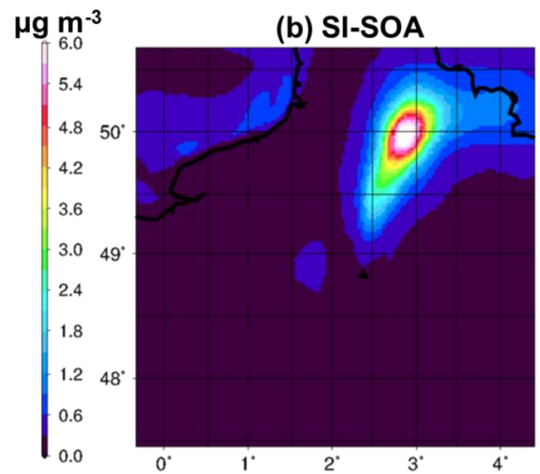
**Zhang et al., ACP, 2015**

# ….. but give a different share of ASOA and SI-SOA

## VBS-HNOX




## VBS-LA




***ASOA:***
**secondary organic aerosol from aromatic VOC**

SI-SOA:
**secondary organic aerosol from primary organic aerosol**

# Conclusions et Perspectives

- 70 % du PM2.5 est transporté vers la région parisienne depuis l'extérieur
- Pics de printemps de nitrate d'ammonium: meilleure connaissance des sources de $NH_3$ et de $HNO_3$ nécessaires pour bien évaluer les sensibilités par rapport aux émissions de trafic et d'agriculture

*=> modélisation des processus d'émissions, modélisation inverse*

- Combinaison des mesures AMS-PMF and $^{14}C$ puissante pour l'attribution des sources :
  => fraction moderne dominante pour le fond urbain parisien (>60% en été, <80% en hiver): activités de cuisine (été, hiver), combustion de bois (hiver), aérosol organique secondaire (été, hiver ?)

*⇒ meilleures cadastres d'émissions pour le chauffage par le bois, activité des cuisine incluant les émissions semivolatiles*

- Contribution d'aérosol organique d'origine fossile (ASOA, SI-SOA) dans le panache parisien

*=> mesure de traceurs pour sources spécifiques (ASOA vs. SI-SOA BBOA vs BSOA*