

Applicazione del modello a recettore *dispersion-normalised multi-time resolution PMF* a dati di PM_1 della Pianura Padana



Crova F.^{1,*}, Forello A.C.^{1,2}, Bernardoni V.¹, Calzolari G.^{2,3}, Canepari S.⁴, Argentini S.⁵, Costabile F.⁵, Frezzini M.A.⁴, Giardi F.^{2,3}, Lucarelli F.^{2,3}, Massabò D.^{6,7}, Massimi L.⁴, Nava S.^{2,3}, Paglione M.⁸, Pazzi G.^{2,3}, Prati P.^{6,7}, Rinaldi M.⁸, Russo M.⁸, Valentini S.¹, Valli G.¹, Vernocchi V.⁷, Vecchi R.¹

¹ Dip. di Fisica, Università degli Studi di Milano e INFN-Milano, Milano, 20133

² Dip. di Fisica e Astrofisica, Università degli Studi di Firenze, Sesto Fiorentino, 50019

³ INFN-Firenze, Sesto Fiorentino, 50019

⁴ Dip. di Biologia Ambientale, Sapienza Università di Roma, Roma, 00185

⁵ Ist. di Scienze dell'Atmosfera e del Clima ISAC-CNR, Roma, 00133

⁶ Dip. di Fisica, Università degli Studi di Genova, Genova, 16146

⁷ INFN-Genova, Genova, 16146

⁸ Ist. di Scienze dell'Atmosfera e del Clima ISAC-CNR, Bologna, 40129

* federica.crova@unimi.it



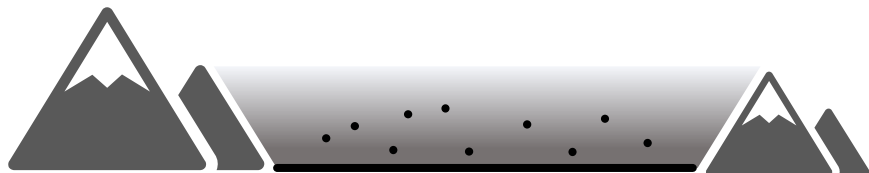
Introduzione

Pianura Padana

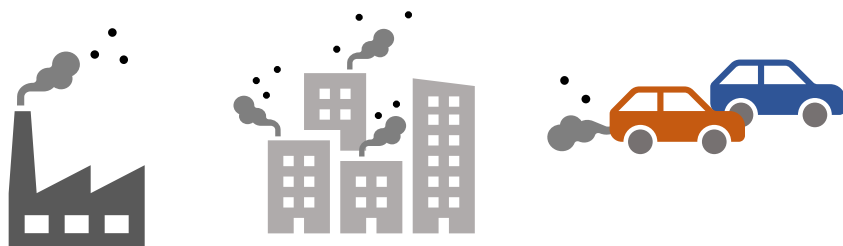


Introduzione

Bacino chiuso
→ Scarsa diluizione atmosferica

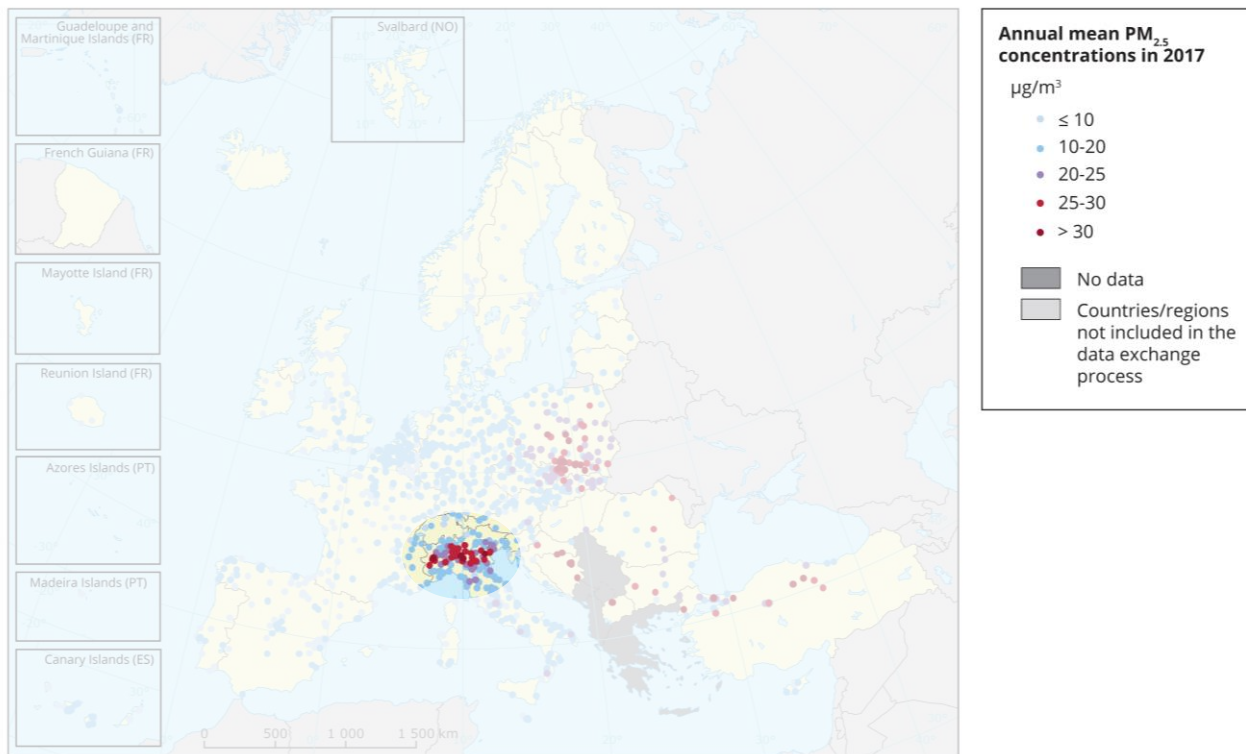


Emissioni intense



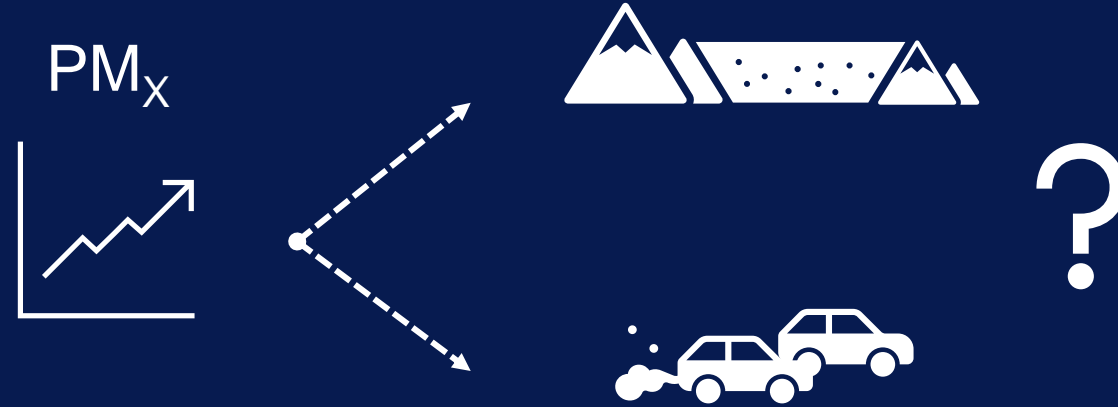
Air quality in Europe — 2019 report
EEA Report | No 10/2019

Map 3.3 Concentrations of PM_{2.5}, 2017 — annual limit value



Note: Observed concentrations of PM_{2.5} in 2017. The possibility of subtracting contributions to the measured concentrations from natural sources and winter road sanding/salting has not been considered. Dots in the last two colour categories indicate stations reporting concentrations above the EU annual limit value (25 µg/m³). Dots in the first colour category indicate stations reporting values below the WHO AQG for PM_{2.5} (10 µg/m³). Only stations with more than 75 % of valid data have been included in the map.

Source: EEA, 2019c.



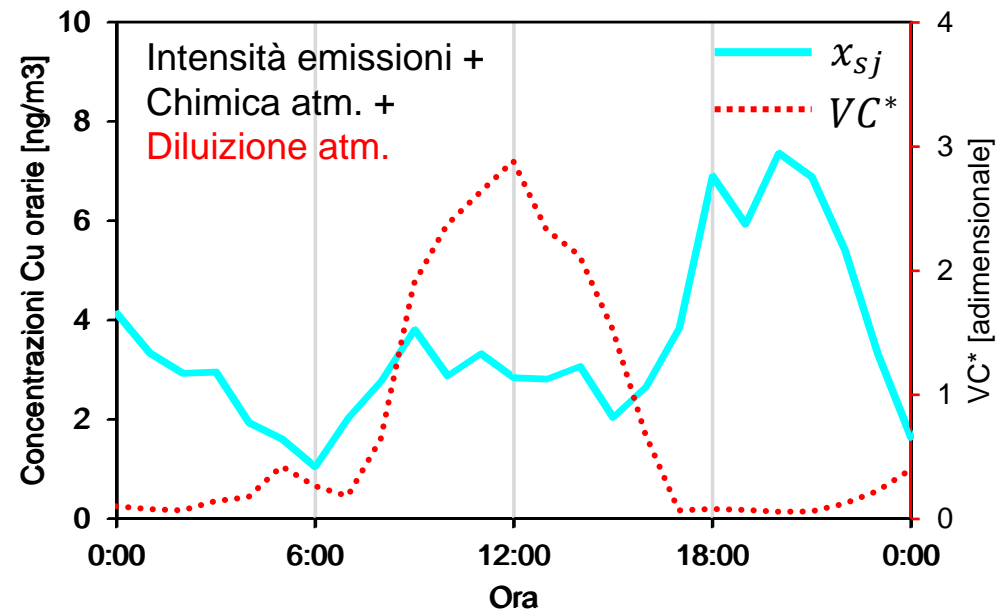
Come possiamo **disaccoppiare** l'effetto della **diluizione atmosferica** dalle **sorgenti di emissione** (e chimica atmosferica) nei modelli a recettore come la PMF?

Dispersion normalised PMF

PMF classica

$$x_{sj} = \sum_{k=1}^P g_{sk} f_{kj} + e_{sj}$$

- $s \rightarrow$ campione
- $j \rightarrow$ specie chimica
- $k \rightarrow$ fattore
- $x_{sj} \rightarrow$ dati di concentrazione in input
- $g_{sk} \rightarrow$ contributo temporale dei fattori
- $f_{kj} \rightarrow$ profilo chimico dei fattori
- $e_{ij} \rightarrow$ residuo di modellizzazione

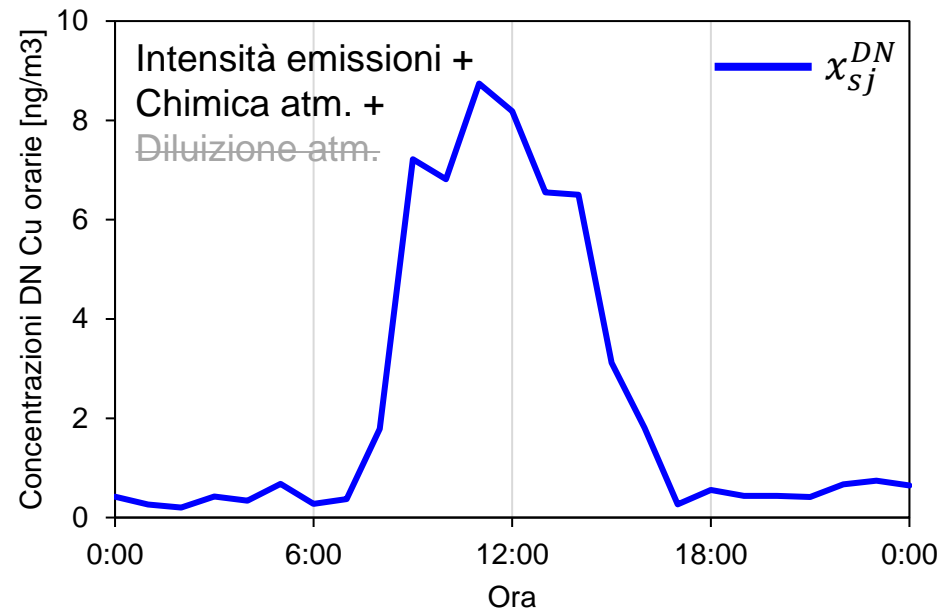


DN-PMF[1]

Ventilation coefficient (proxy per la diluizione):

$$VC_s = MLH_s \cdot v_s \quad x_{sj}^{DN} = x_{sj} \cdot \frac{VC_s}{VC} = x_{sj} \cdot VC_s^*$$

$$x_{sj}^{DN} = \sum_{k=1}^P g_{sk}^{DN} f_{kj}^{DN} + e_{sj}^{DN} \quad g_{sk}^{deDN} = \frac{g_{sk}}{VC_s^*}$$



[1] Dai et al. (2020) Environ. Sci. Technol., 54, 9917

Obiettivi

- Applicazione dell'approccio DN in un sito in **Pianura Padana** (concentrazioni altamente influenzate dalla diluizione atmosferica)

Obiettivi

- Applicazione dell'approccio DN in un sito in **Pianura Padana** (concentrazioni altamente influenzate dalla diluizione atmosferica)
- DN implementato nel modello a recettore avanzato **multi-time resolution PMF^[1]** (DN-MT)

DN-MT

$$x_{sj}^{DN} = \frac{1}{t_{s2} - t_{s1} + 1} \sum_{k=1}^P f_{kj}^{DN} \sum_{i=t_{s1}}^{t_{s2}} g_{ik}^{DN} + e_{sj}$$

t_{s1}, t_{s2} → tempo di inizio e fine del campione s

i → unità temporale nel campione s
più piccolo intervallo di tempo presente nel dataset

$t_{s1} - t_{s2} + 1$ → lunghezza del campione in termini di unità temporali

Uno tra i pochissimi lavori^[2,3] in letteratura

Obiettivi

- Applicazione dell'approccio DN in un sito in **Pianura Padana** (concentrazioni altamente influenzate dalla diluizione atmosferica)
- DN implementato nel modello a recettore avanzato **multi-time resolution PMF^[1]** (DN-MT)
- Approfondimento sulla comprensione dei contributi temporali g_{ik}^{DN}

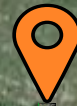
Info sui contributi temporali
consistenti con le concentrazioni originali misurate in atmosfera reale

$$g_{sk}^{deDN} = \frac{g_{sk}^{DN}}{VC^*}$$

Info sulla **intensità effettiva** delle sorgenti di emissione e chimica atmosferica

Campagna di misura

Pianura Padana



Bologna



Campagna di misura

Progetto PRIN-2017 RHAPS [1]

(Redox-activity and Health-effects of Atmospheric Primary and Secondary aerosol)



Sito di fondo urbano

Campagna invernale:

21/01/2021 – 18/03/2021

Campagna estiva:

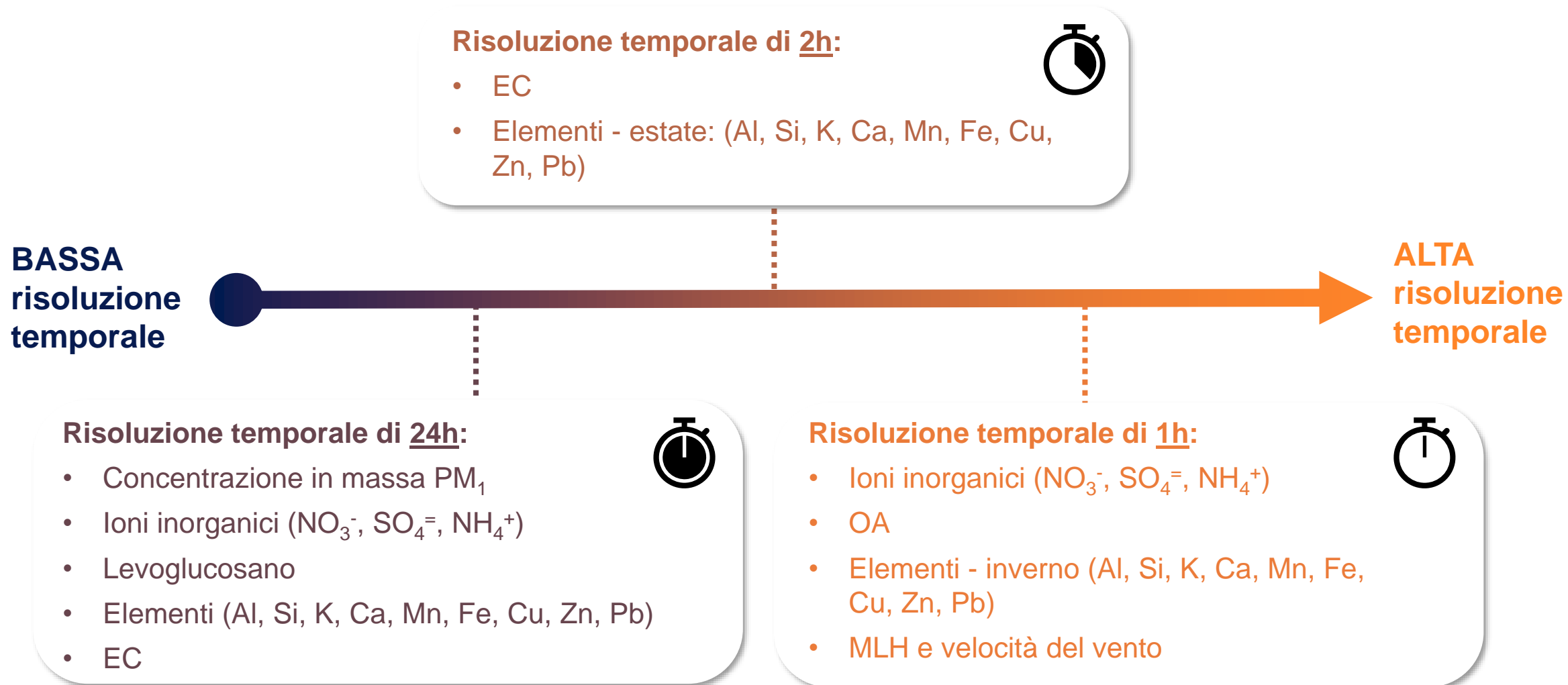
08/06/2021 – 14/07/2021

[1] Costabile et al. (2021) Atmosphere, 13, 5 [Google Earth](#)

Campagna di misura

Caratterizzazione completa e dettagliata del **PM₁**

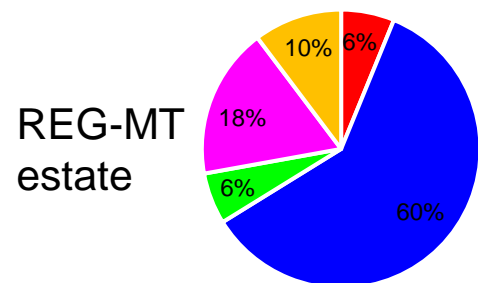
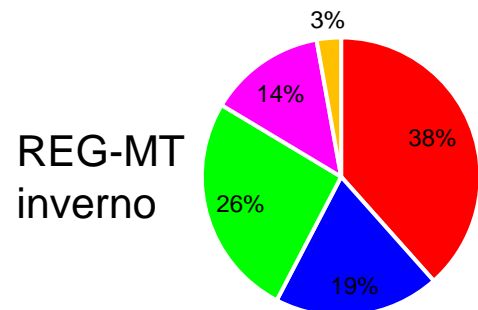
→ **Ampio dataset** (>1000 filtri per analisi offline + strumentazione online)



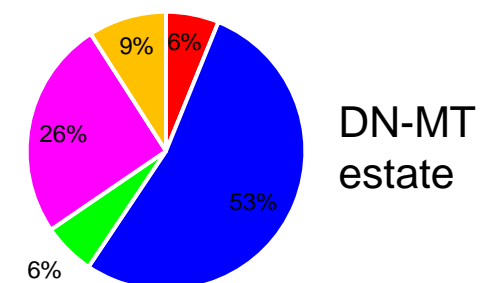
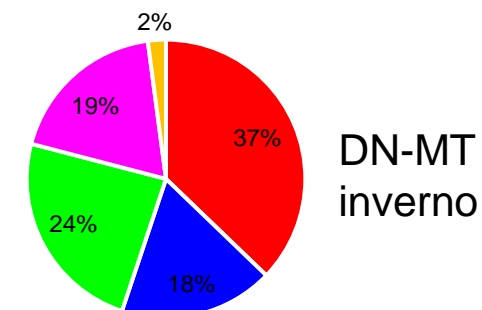
Risultati

Run del modello: applicazione **regolare (REG-MT)** e **dispersion-normalised (DN-MT)**

Soluzione a 5-fattori, identificati come:



- Nitrate-dominated aerosol** (NO_3^- , NH_4^+ , OA)
- Sulphate-dominated aerosol** (SO_4^- , NH_4^+ , OA)
- Biomass burning** (Levo, K, EC, Zn, Pb)
- Urban aerosol** (Mn, Fe, Cu, EC)
- Mineral dust** (Al, Si, Ca, Fe)



REG-MT versus DN-MT

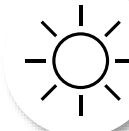
Profili chimici simili

Maggiori differenze nei contributi temporali di fattori specifici

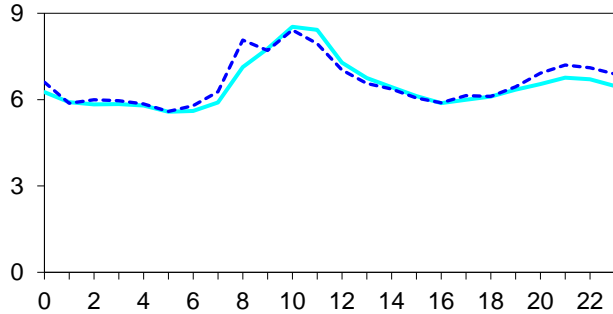
Giorni medi



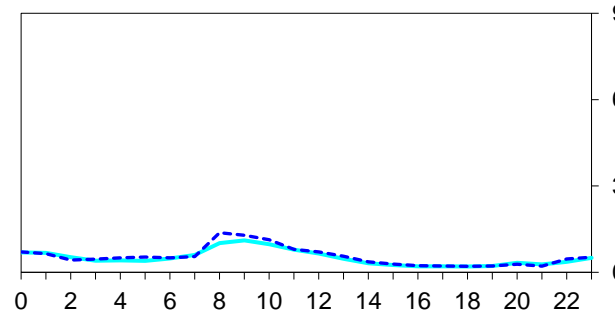
— g_{ik} (REG-MT) - - - g_{ik}^{deDN} (DN-MT)



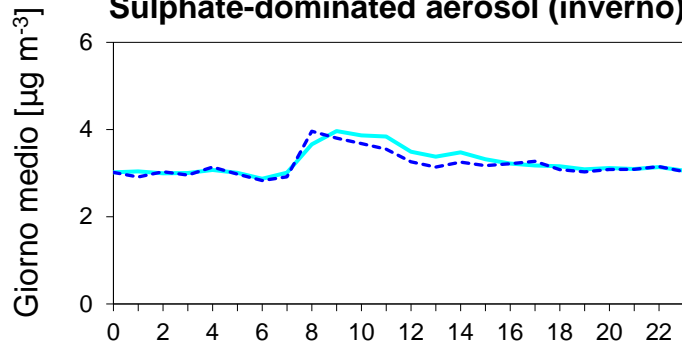
Nitrate-dominated aerosol (inverno)



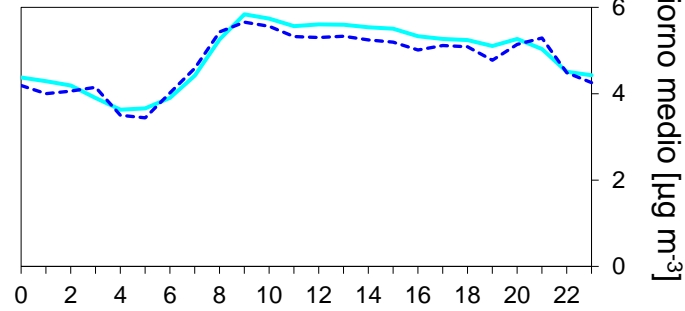
Nitrate-dominated aerosol (estate)



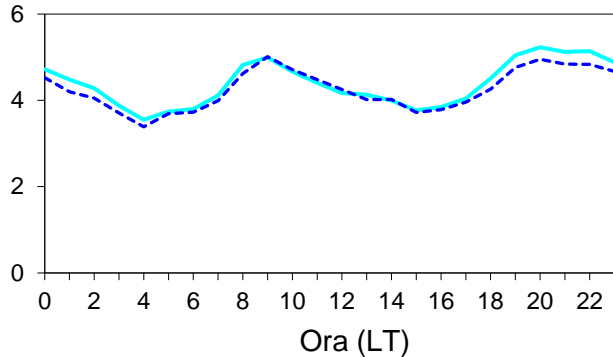
Sulphate-dominated aerosol (inverno)



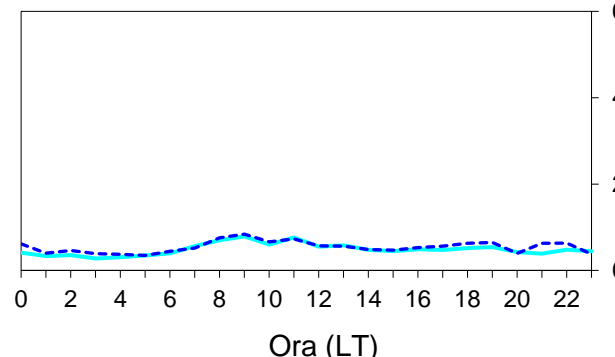
Sulphate-dominated aerosol (estate)



Biomass burning (inverno)



Biomass burning (estate)



REG vs DN:
NON
statisticamente
diversi!
(test di Mann-Whitney)

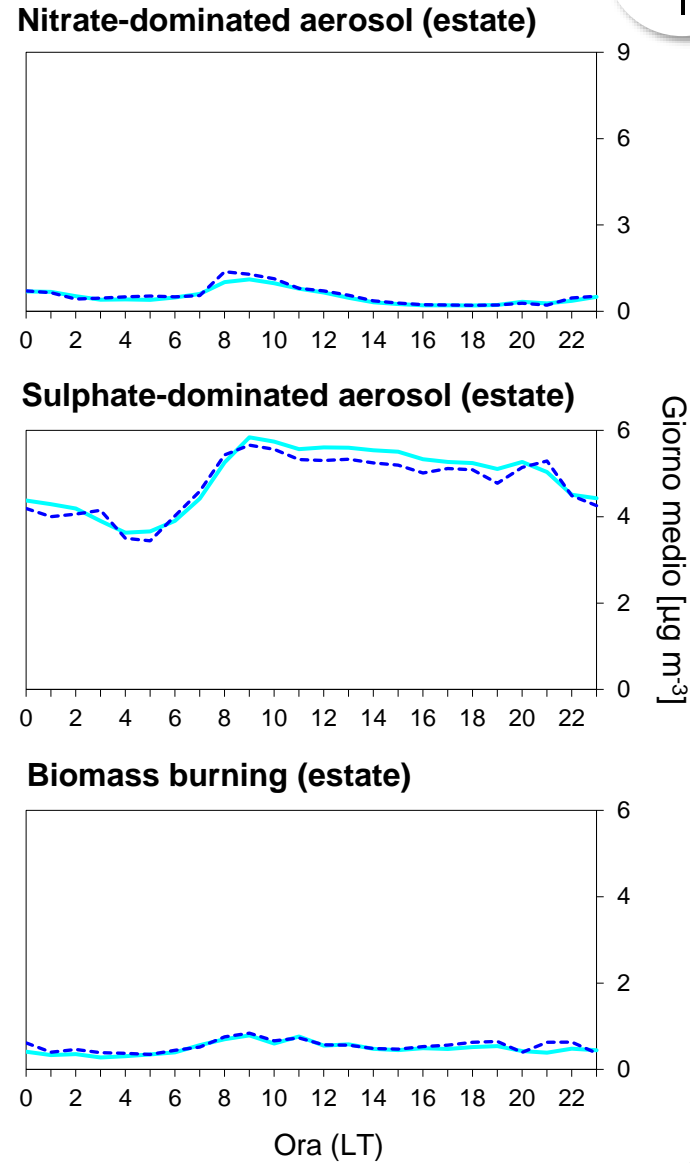
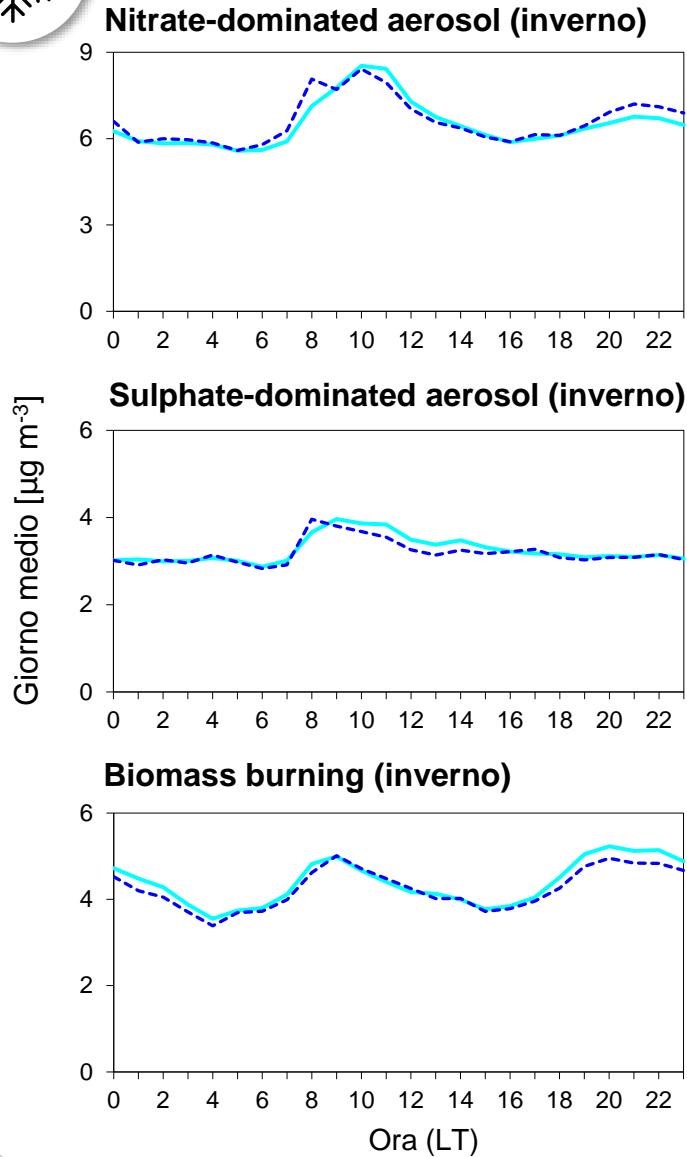
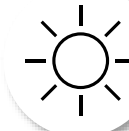


Giorni medi

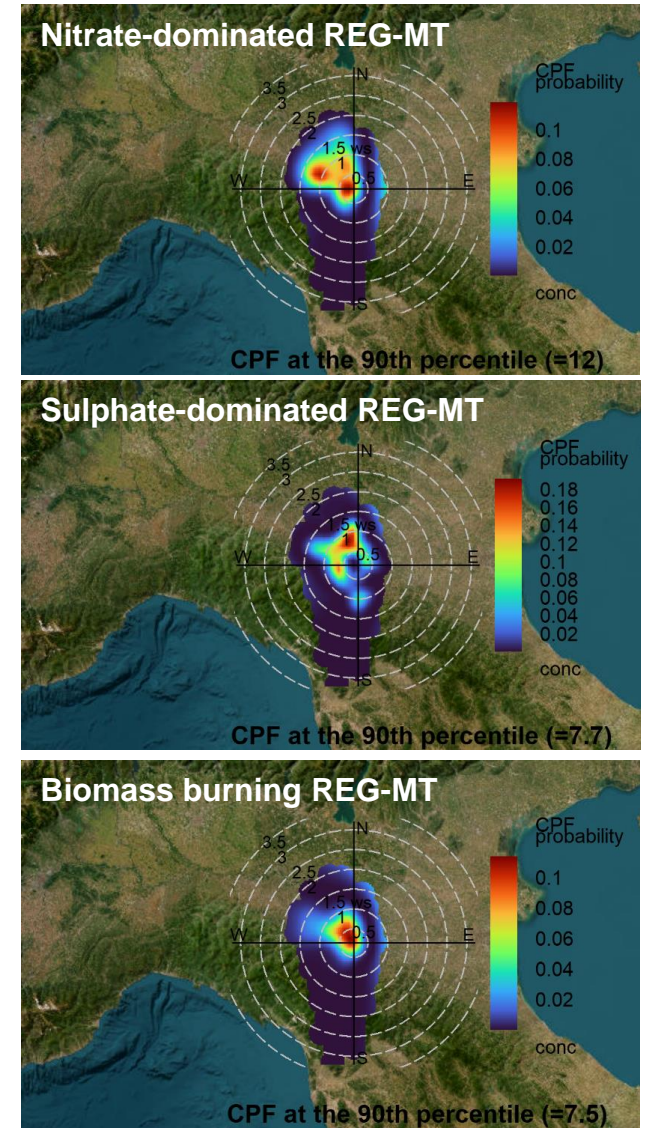


— g_{ik} (REG-MT)

⋯ g_{ik}^{deDN} (DN-MT)



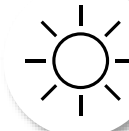
Conditional Probability Function CPF plot (DN-MT ~ equivalente a REG-MT)



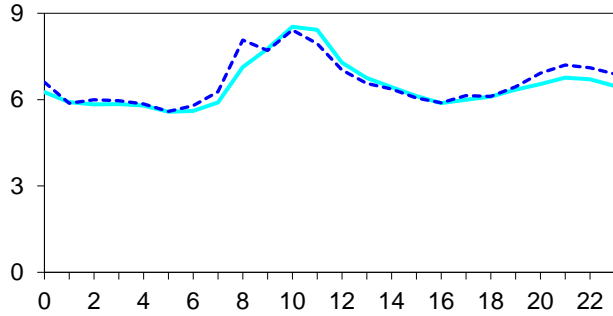
Giorni medi



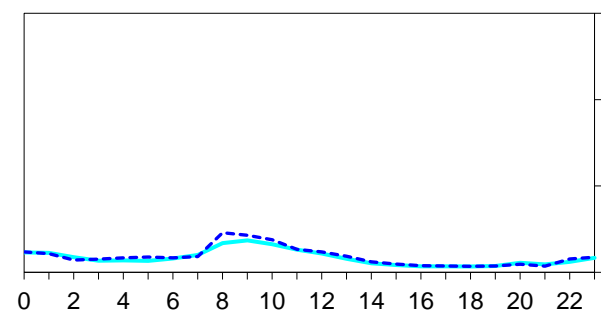
— g_{ik} (REG-MT) - - - g_{ik}^{deDN} (DN-MT)



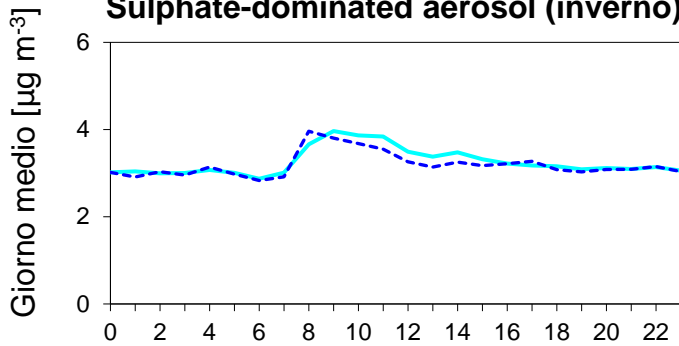
Nitrate-dominated aerosol (inverno)



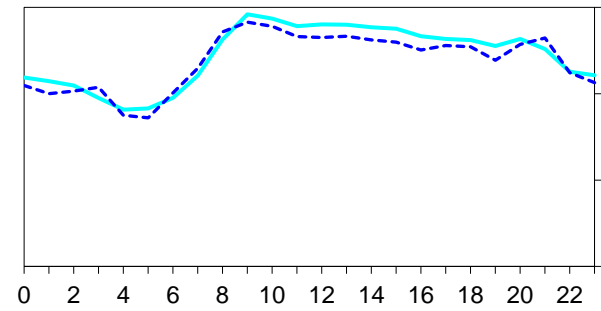
Nitrate-dominated aerosol (estate)



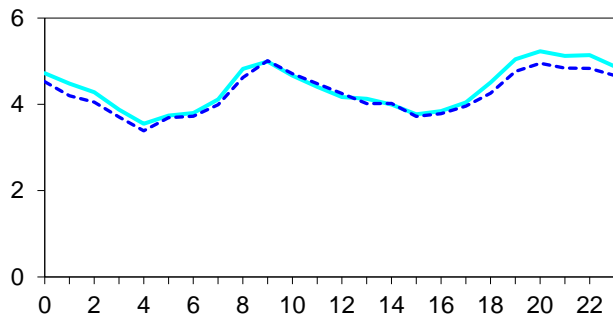
Sulphate-dominated aerosol (inverno)



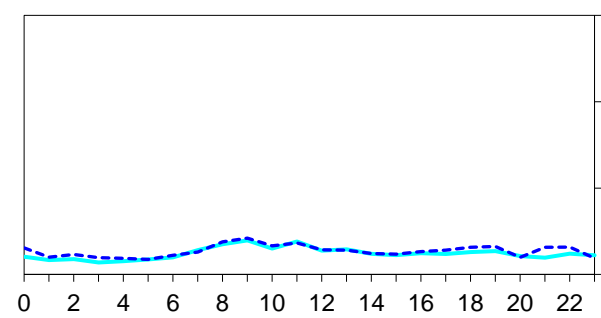
Sulphate-dominated aerosol (estate)



Biomass burning (inverno)



Biomass burning (estate)



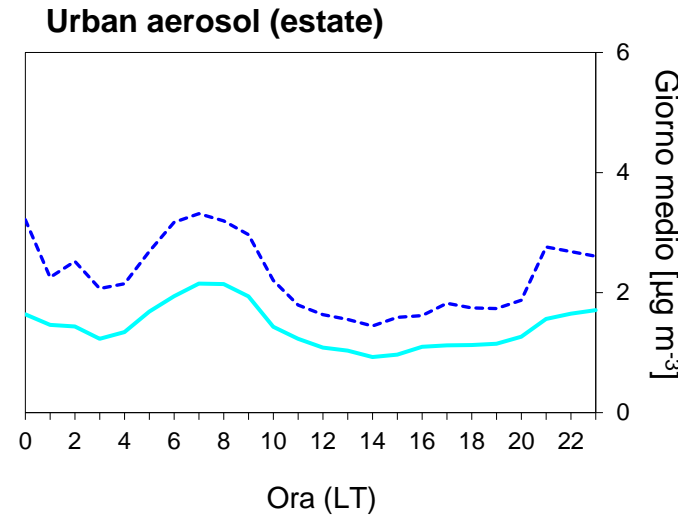
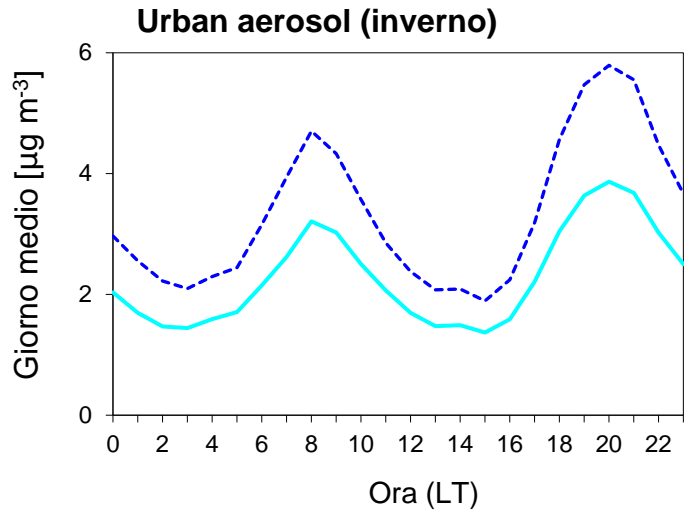
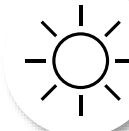
Miscela di aerosol formate/processate da vari processi atmosferici su diverse scale temporali a **scala regionale (di bacino)**



Giorni medi



— g_{ik} (REG-MT) - - - g_{ik}^{deDN} (DN-MT)

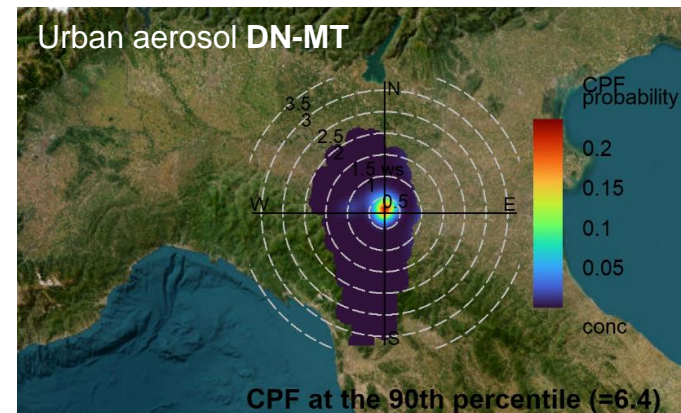
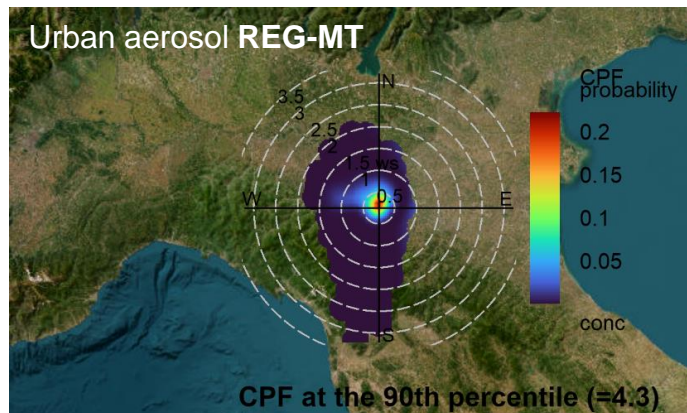


REG vs DN:
Statisticamente diversi!
 (test di Mann-Whitney)

DN-MT **aumenta** il contributo del fattore **urban aerosol**

I pattern del fattore **mineral dust non sono significativi** perchè dominati da episodi di **trasporto di polvere Sahariana** in entrambe le stagioni

Conditional Probability Function CPF plot



- I contributi non provengono mai da una direzione specifica
- Associati a velocità del vento quasi nulle

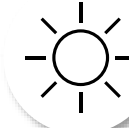
→ **Origine LOCALE**



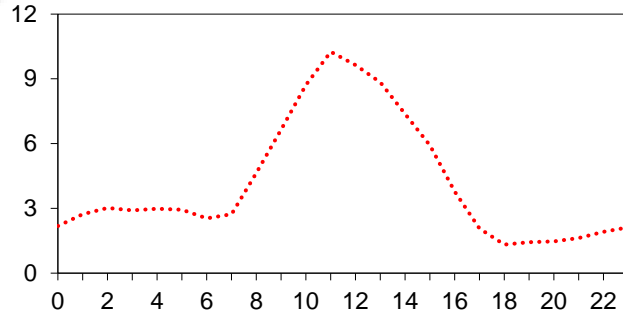
g_{ik}^{DN} per rimuovere la diluizione atmosferica



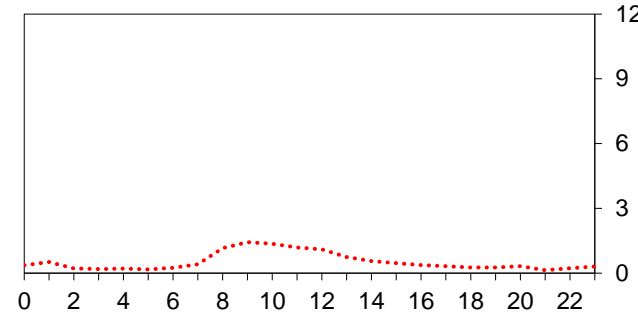
..... g_{ik}^{DN} (DN-MT)



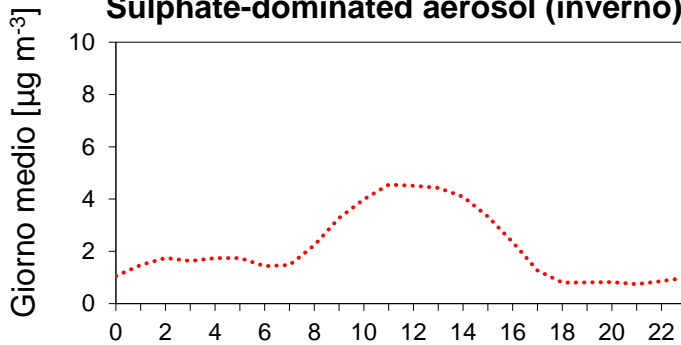
Nitrate-dominated aerosol (inverno)



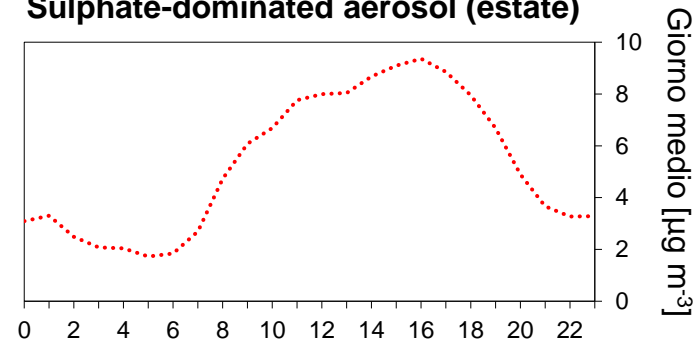
Nitrate-dominated aerosol (estate)



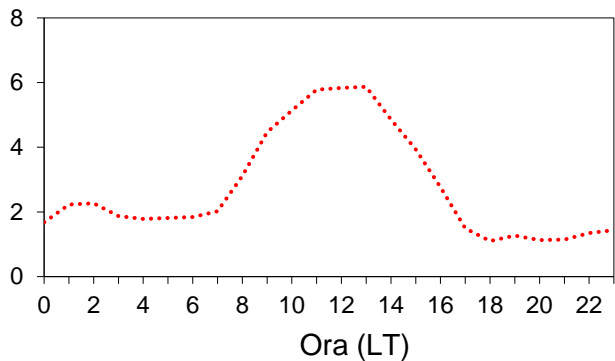
Sulphate-dominated aerosol (inverno)



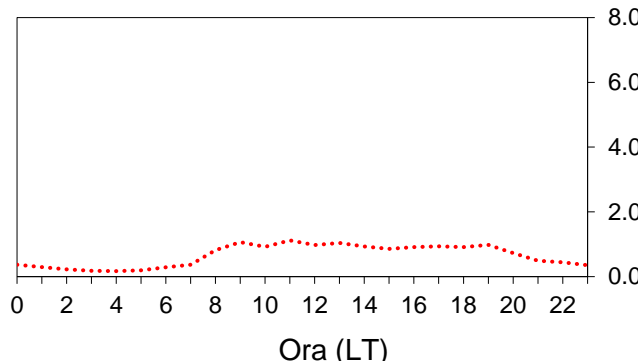
Sulphate-dominated aerosol (estate)



Biomass burning (inverno)



Biomass burning (estate)



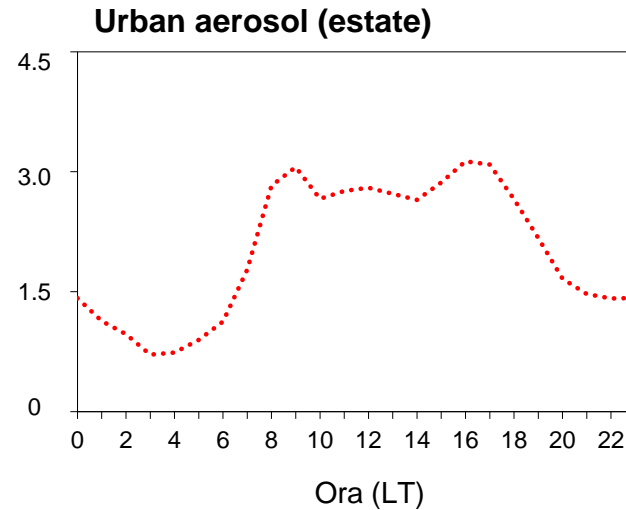
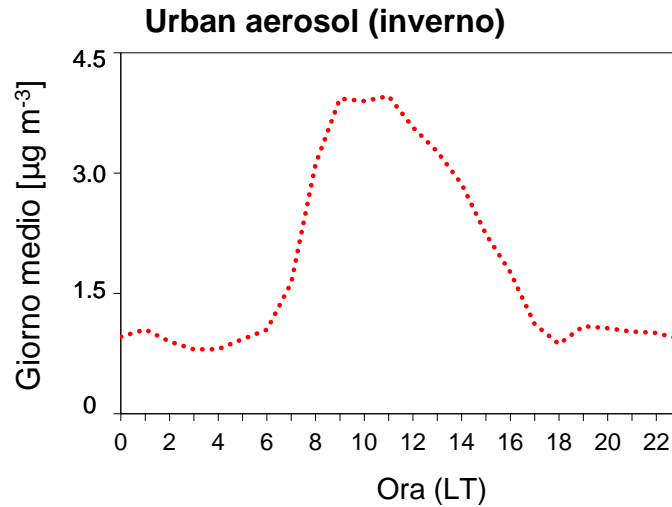
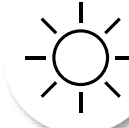
Queste sorgenti sono **attive** nelle **ore centrali** del giorno

Le variazioni giornaliere di emissioni su scala **regionale** sono influenzate dalla **modulazione del MLH**

g_{ik}^{DN} per rimuovere la diluizione atmosferica



..... g_{ik}^{DN} (DN-MT)



Inverno

Sorgente attiva nelle ore centrali del giorno

Massimo del fattore urban aerosol spostato indietro

Estate:

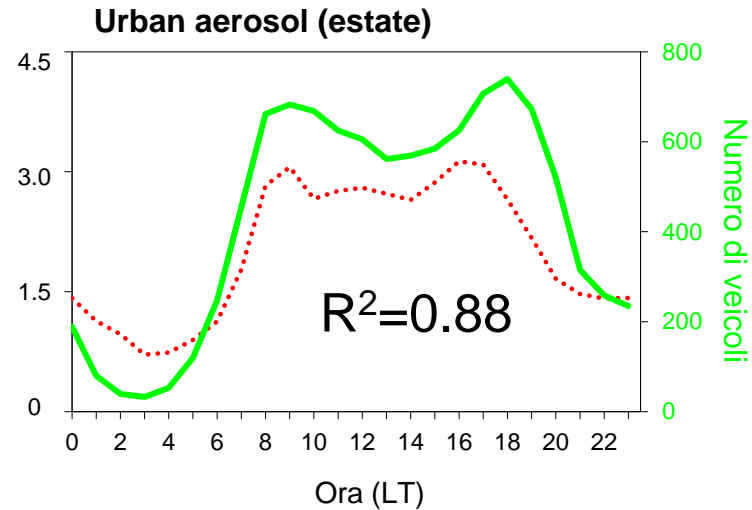
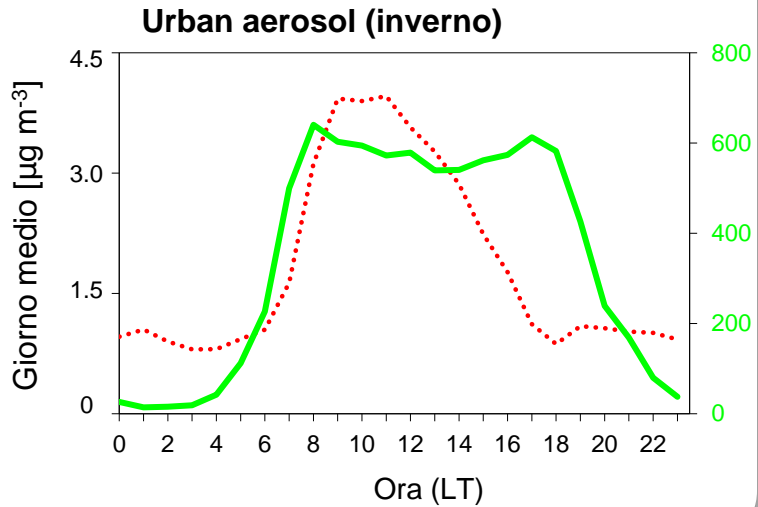
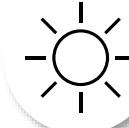
Forma a "M" tipica del traffico

g_{ik}^{DN} per rimuovere la diluizione atmosferica



..... g_{ik}^{DN} (DN-MT)

—— Volume traffico

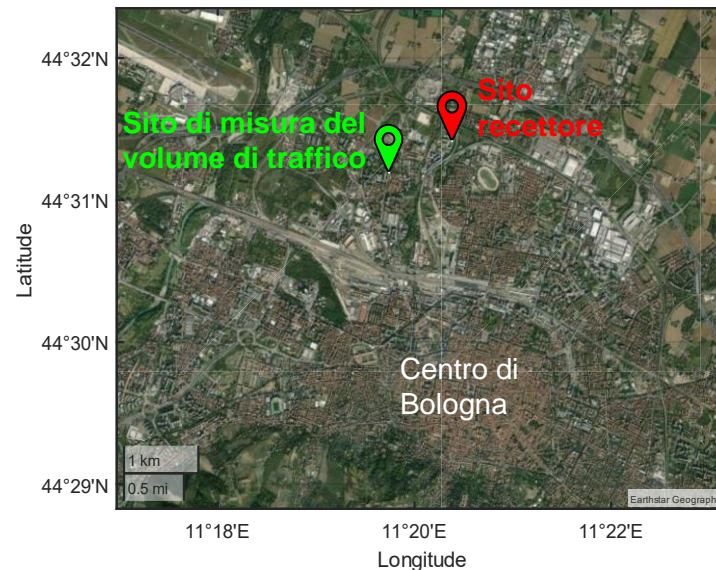


Buon accordo con il volume di traffico in **estate**

→ g_{ik}^{DN} fornisce info sulla **intensità di emissioni effettiva** (il volume di traffico infatti non dipende da dati meteo)

Scarso accordo in **inverno**: il pattern del fattore urban aerosol non presenta il picco serale

→ Trasformazioni che alterano le dimensioni delle particelle PM_1 ?



Conclusioni e prospettive

Prima applicazione del modello DN-MT in un sito Europeo dove la **dispersione atmosferica ha un ruolo fondamentale** sulle concentrazioni di aerosol



I fattori dominati dall'**aerosol secondario** prodotto alla **scala regionale** (di bacino) **non sono influenzati** dall'approccio DN-MT

L'approccio DN-MT ha quasi **raddoppiato** il contributo del fattore **urban aerosol**



Crova et al., *Atmospheric Environment* 316 (2024) 120168



Ulteriori applicazioni dell'approccio DN-MT su dataset diversi

Capire meglio le informazioni sulla **intensità effettiva delle sorgenti** di emissione date dai contribute temporali g_{ik}^{DN}

Grazie a:

Collaboratrici e collaboratori nel progetto RHAPS (*)



UNIVERSITÀ
DEGLI STUDI
DI MILANO



Istituto Nazionale di Fisica Nucleare



UNIVERSITÀ
DEGLI STUDI
FIRENZE



SAPIENZA
UNIVERSITÀ DI ROMA



UNIVERSITÀ
DEGLI STUDI
DI GENOVA

Gruppo di ricerca di Fisica dell'Ambiente, Università degli Studi di Milano

Grazie per la vostra attenzione!

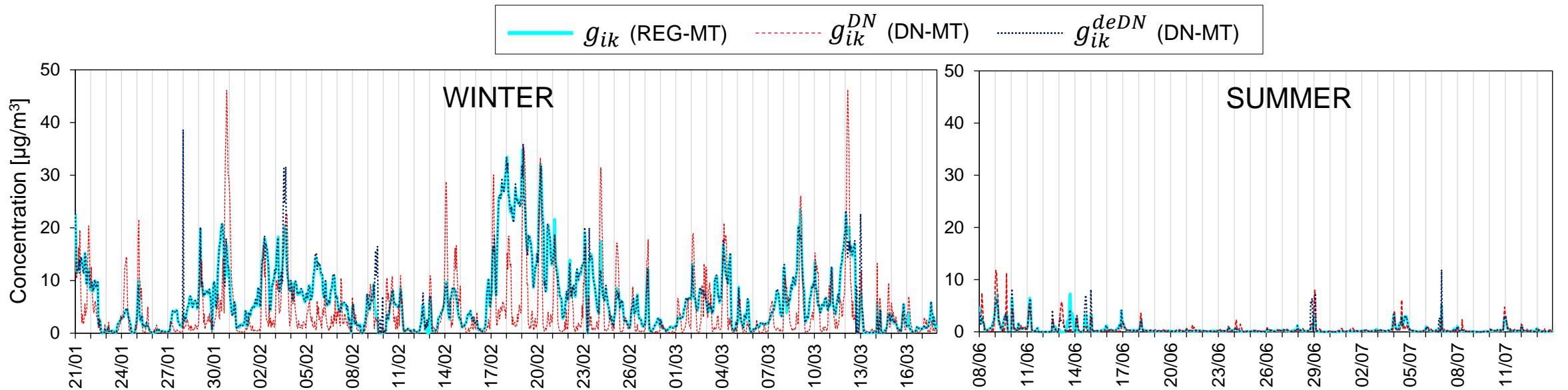
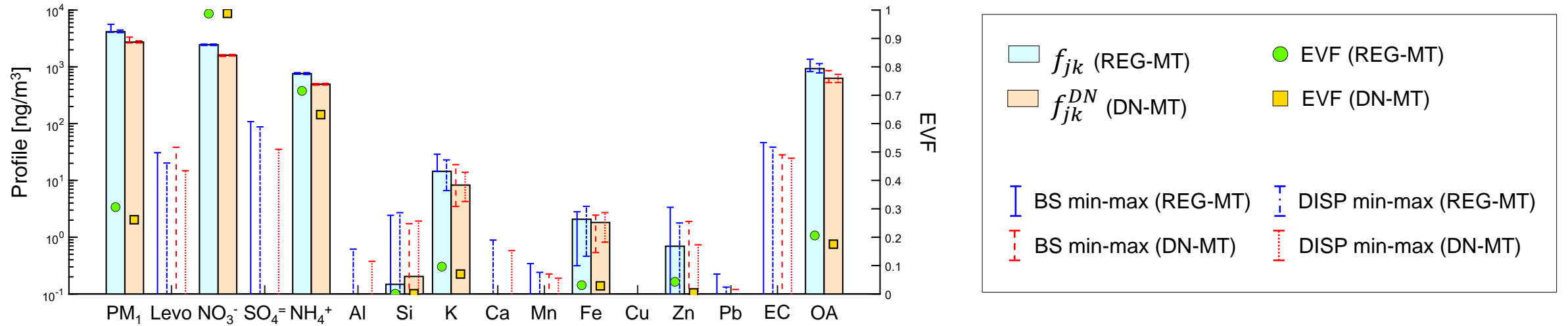
(*) Progetto finanziato dal Ministero dell'Università e della Ricerca (grant number: 2017MSNM8)

federica.crova@unimi.it



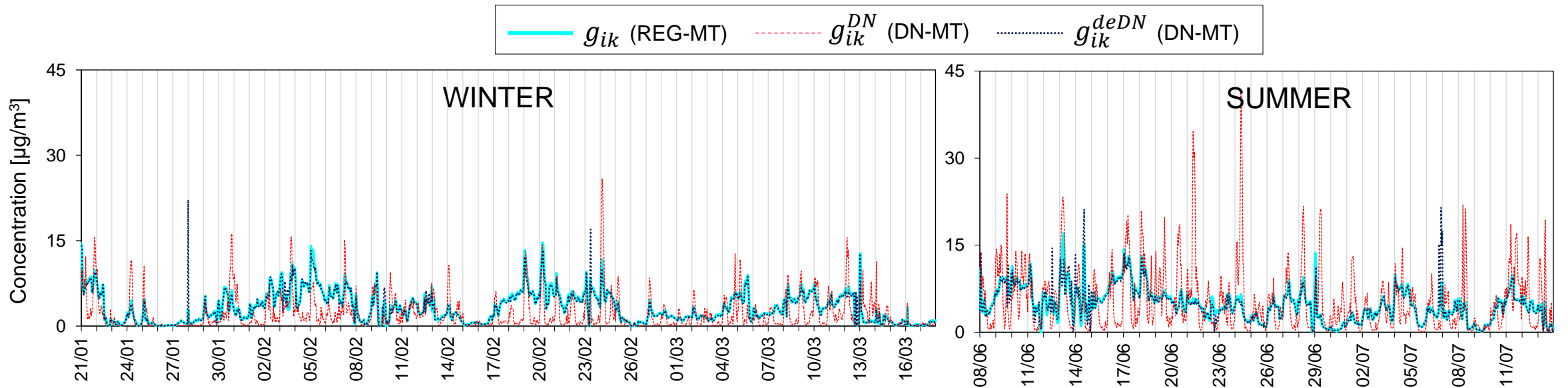
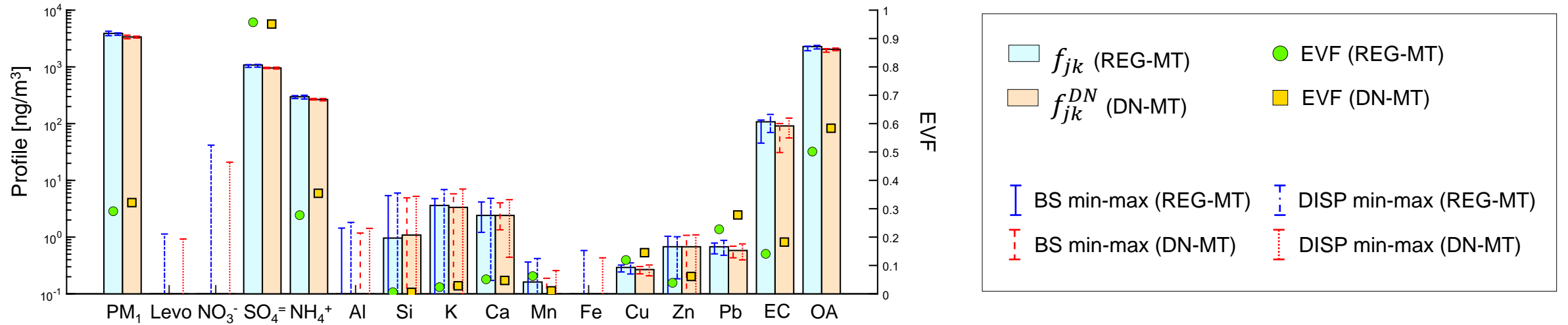
Results: 5-factor solution (REG-MT vs DN-MT)

Nitrate-dominated aerosol



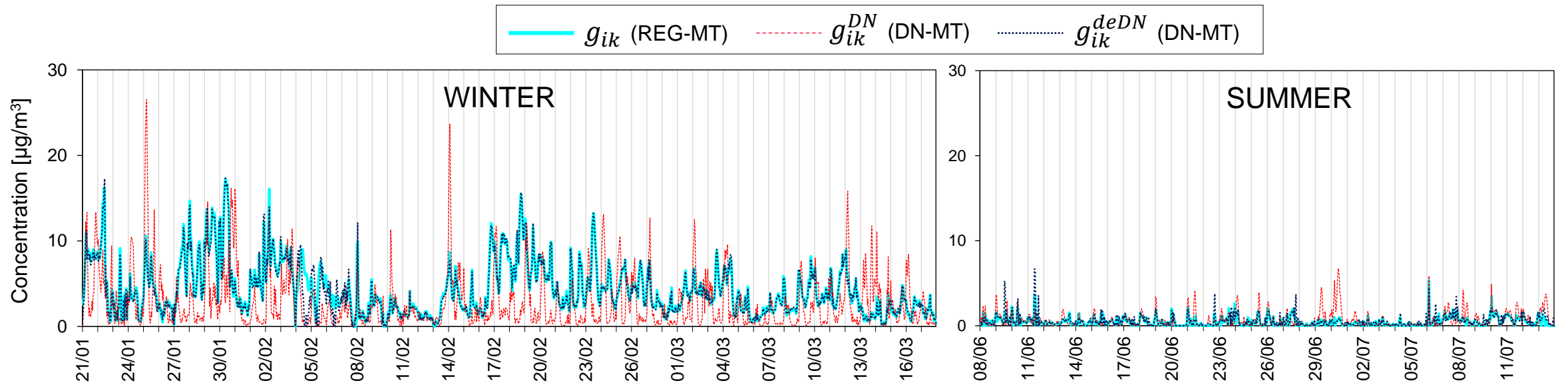
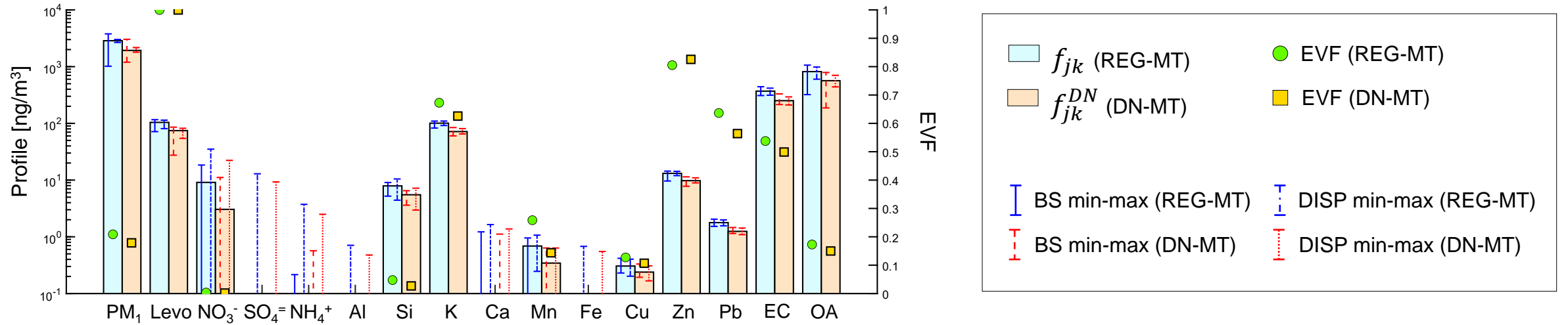
Results: 5-factor solution (REG-MT vs DN-MT)

Sulphate-dominated aerosol



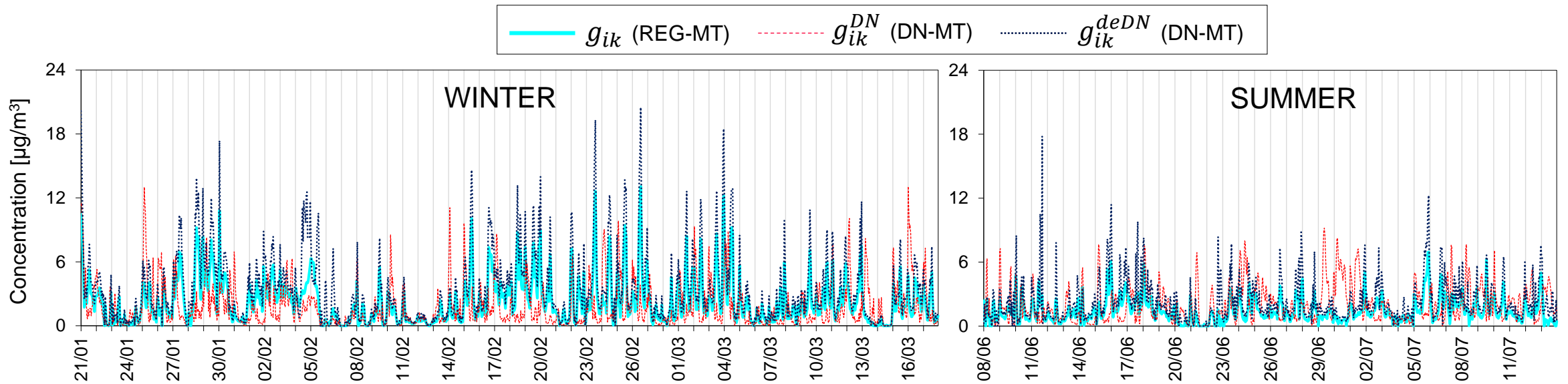
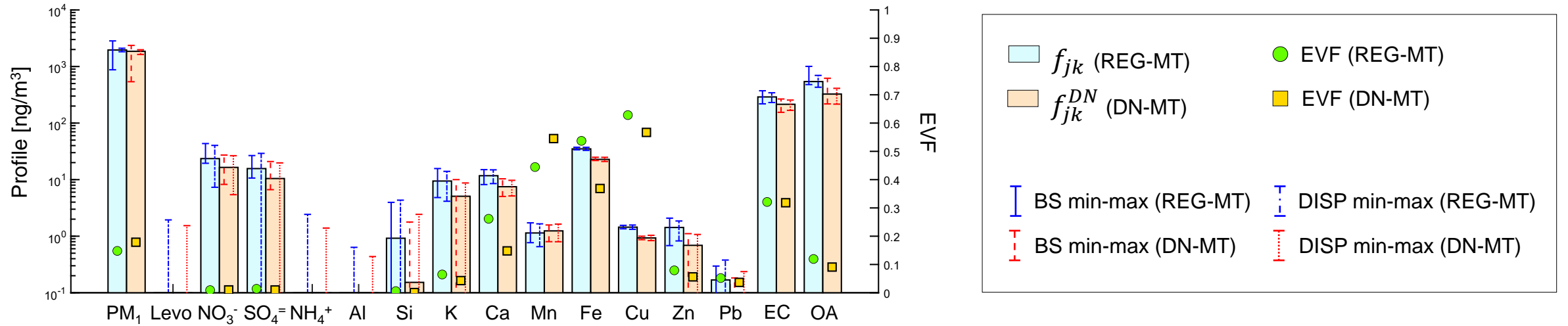
Results: 5-factor solution (REG-MT vs DN-MT)

Biomass burning

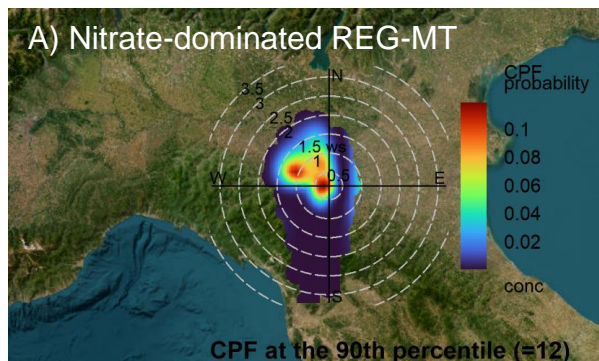


Results: 5-factor solution (REG-MT vs DN-MT)

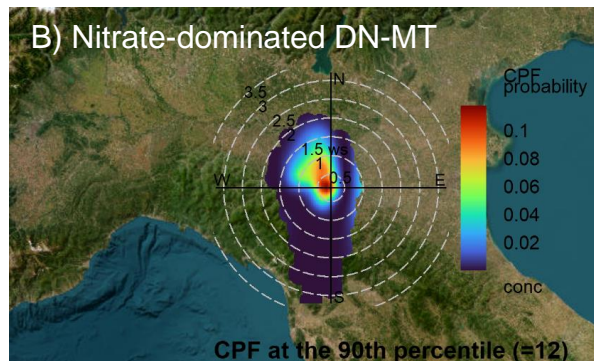
Urban aerosol



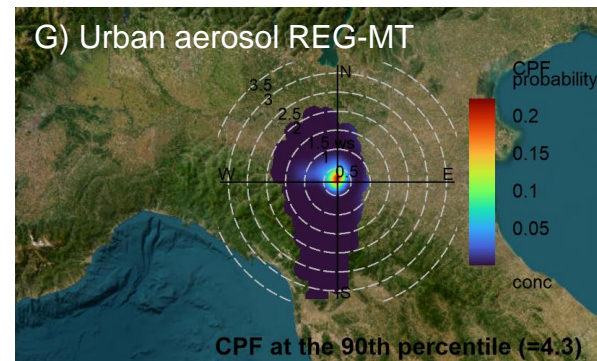
REG-MT



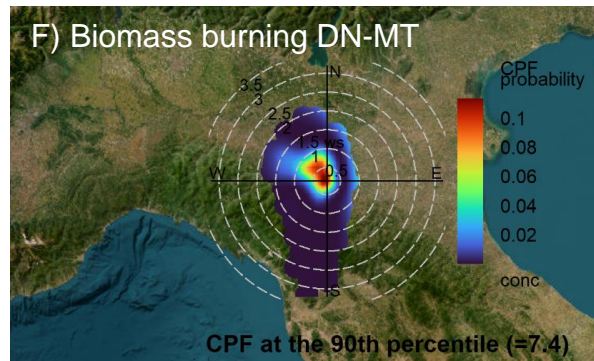
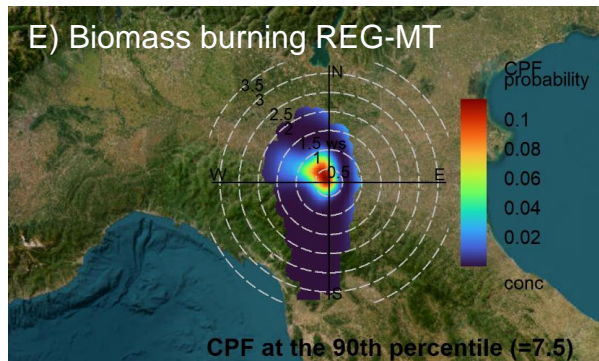
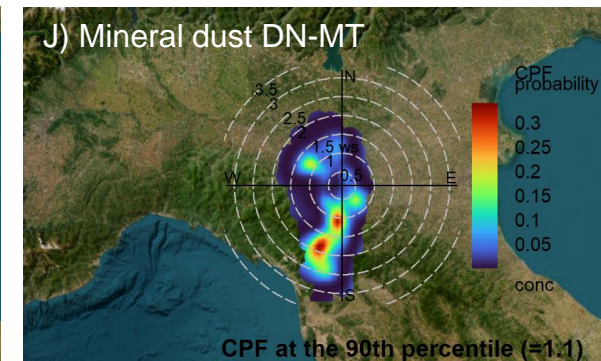
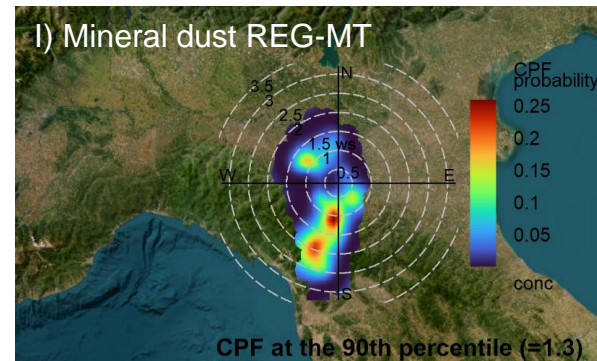
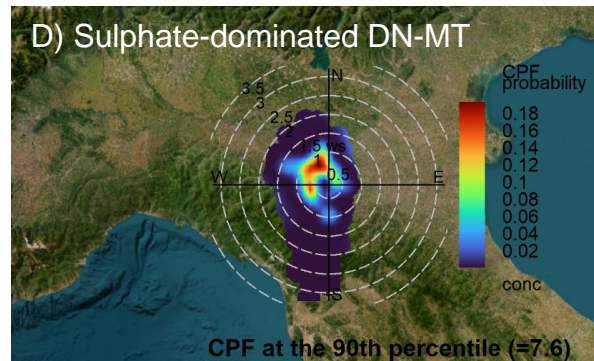
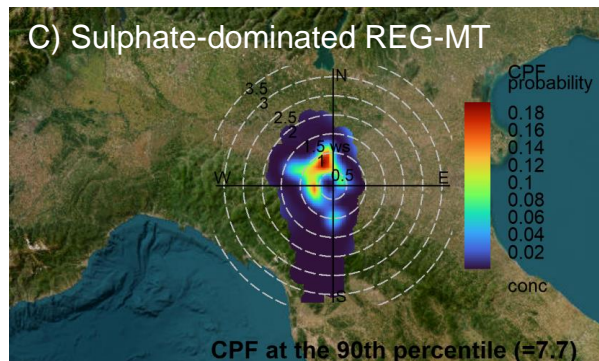
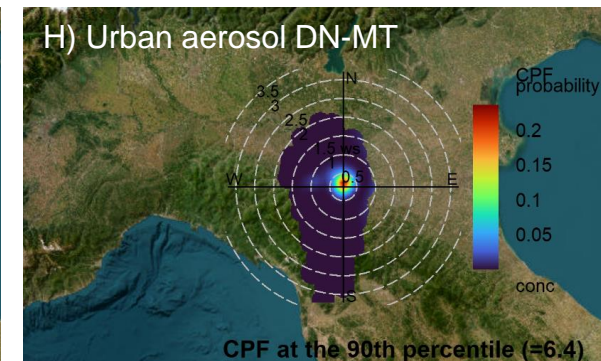
DN-MT



REG-MT

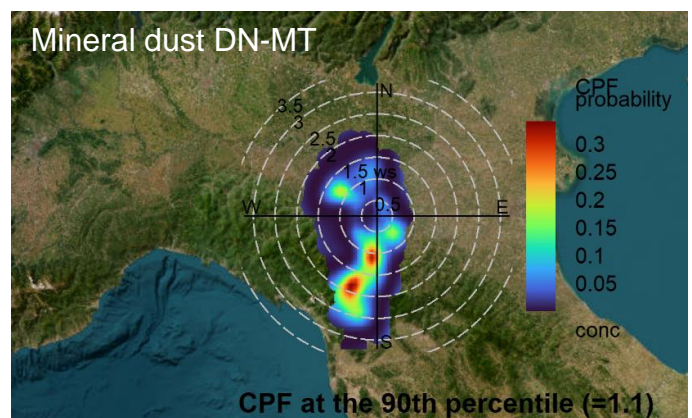
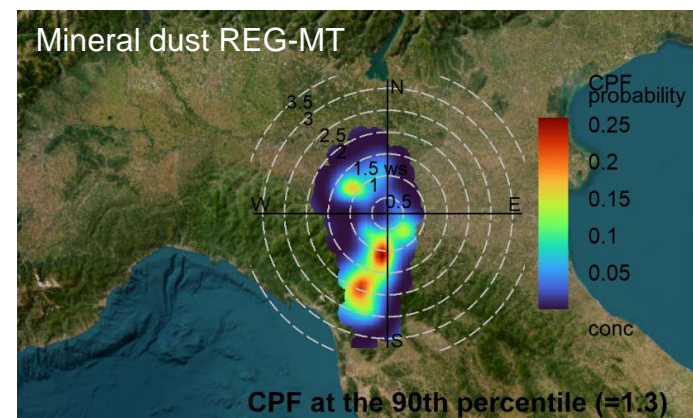
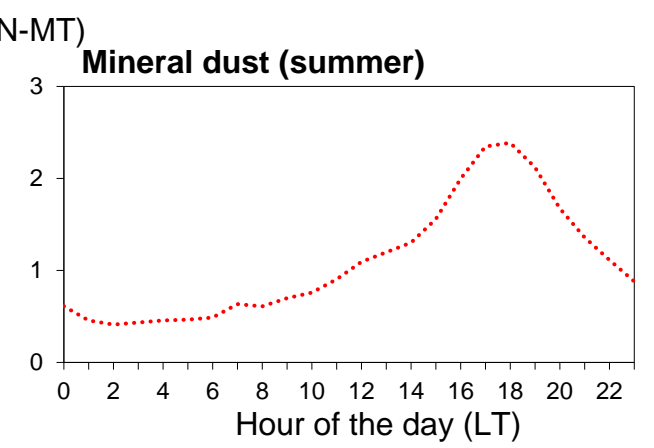
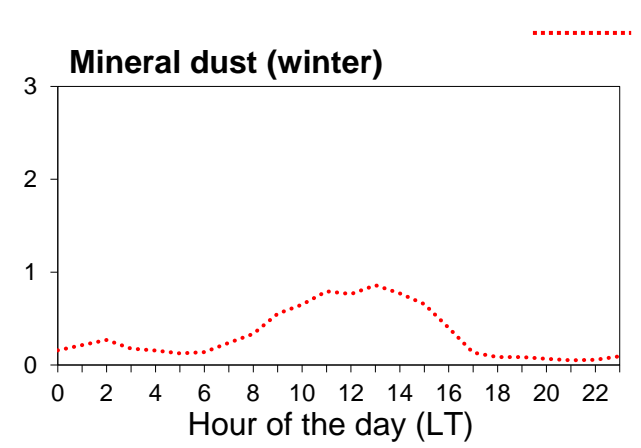
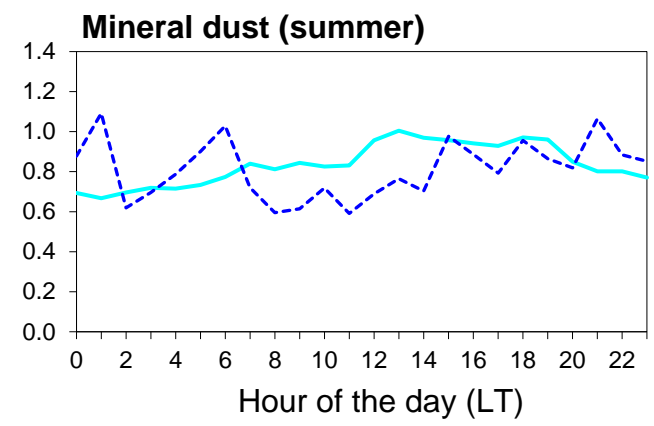
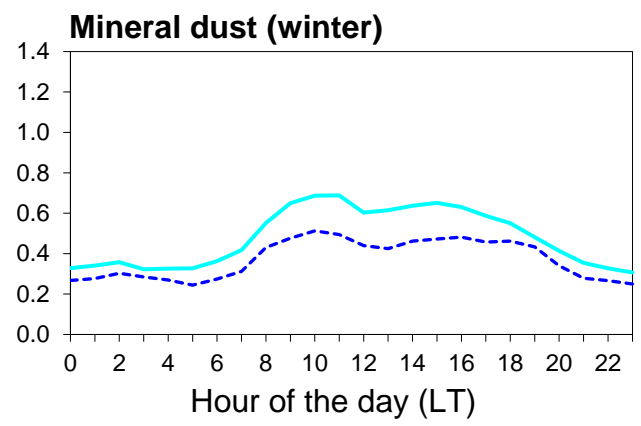


DN-MT



— g_{ik} (REG-MT)

- - - g_{ik}^{deDN} (DN-MT)



Positive Matrix Factorisation (PMF)

PMF

$$x_{sj} = \sum_{k=1}^{n_p} g_{sk} f_{kj} + e_{sj}$$

s → samples

j → species (concentration)

k → factors (sources)

X = input data

G = temporal contributions

F = chemical profiles

E = residuals

X and **G**:

same number of rows

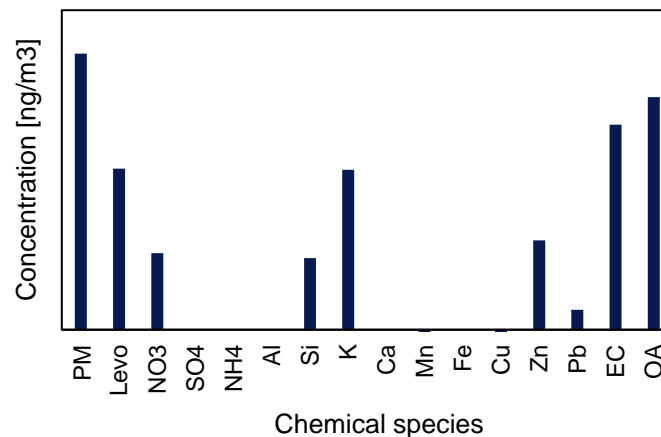
Same time
resolution!

X

Date of sampling	PM mass	Levo	NO3	SO4	NH4	Al	...
21/01/2021	52673.6	356.3	15211.3	1554.0	2447.7	77.6	...
22/01/2021	21067.7	654.4	3758.5	408.7	896.9	22.5	...
...
12/03/2021	9086.5	20.1	281.7	49.9	1421.7	63.9	...
13/03/2021	5997.2	10.3	192.0	40.6	938.3	15.2	...

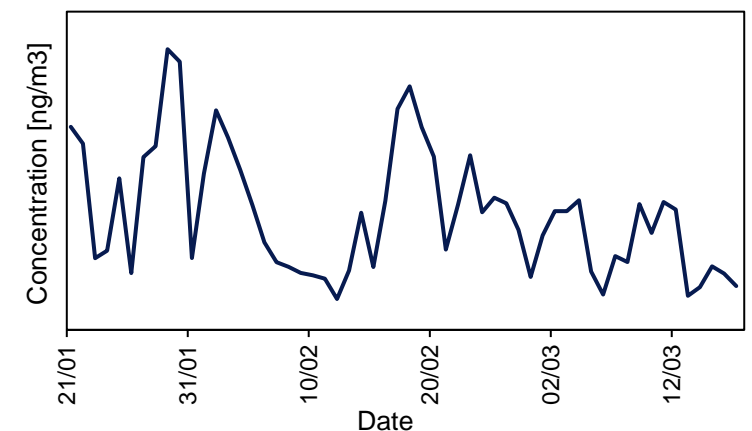
F

Chemical profile of factor k



G

Temporal contribution of factor k



The multi-time resolution receptor model

Regular PMF

$$x_{sj} = \sum_{k=1}^{n_p} g_{sk} f_{kj} + e_{sj}$$

s → samples

j → species (concentration)

k → factors (sources)

X = input data

G = temporal contributions

F = chemical profiles

E = residuals

X and **G**:

same number of rows

Multi-time resolution receptor model ^[1]

$$x_{sj} = \frac{1}{t_{s2} - t_{s1} + 1} \sum_{k=1}^P \left(f_{kj} \sum_{i=t_{s1}}^{t_{s2}} g_{ik} \right) \eta_{jn} + e_{sj}$$

s → samples (different time resolution)

j → chemical species

k → factors

t_{s1}, t_{s2} → start and end time of sample s

i → **time unit** in the sample s

smaller sampling interval in the dataset

$t_{s1} - t_{s2} + 1$ → sample length in terms of time units

η_{jn} → adjustment factors (n =subset of the total number of samples s)

X and **G**:

different number of rows

X number of rows = number of samples

G number of rows = number of time units

The multi-time resolution receptor model

X

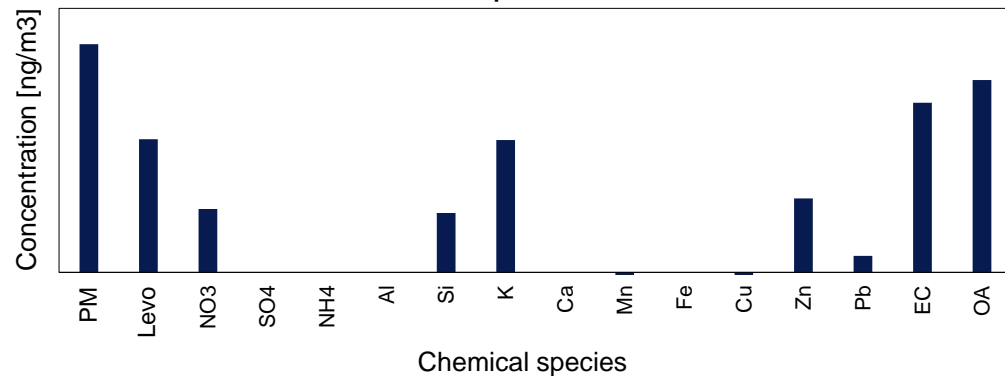
species

	Datestart:	Time	Dateend:	Time	Tzone	Begin	Length	End	PM	PM_std	NO3	NO3_std	SO4	SO4_std	...
	High time resolution samples	21/01/2021	08:00:00	21/01/2021	09:00:00	EST	1	1	1			14212.0	2853.0	5221.4	1054.2
21/01/2021		09:00:00	21/01/2021	10:00:00	EST	2	1	2			11168.8	2244.4	4934.1	996.8	...
21/01/2021		10:00:00	21/01/2021	11:00:00	EST	3	1	3			7166.2	1443.9	2952.7	600.5	...
...	
14/07/2021		06:00:00	14/07/2021	07:00:00	EST	2207	1	2207			67.6	22.9	642.6	137.9	...
14/07/2021		07:00:00	14/07/2021	08:00:00	EST	2208	1	2208			49.6	19.3	431.6	95.6	...
Low time resolution samples	21/01/2021	08:00:00	22/01/2021	08:00:00	EST	1	24	24	52673.6	210694.6	15211.3	1554.0	2447.7	277.6	...
	22/01/2021	08:00:00	23/01/2021	08:00:00	EST	25	24	48	21067.7	84271.0	3758.5	408.7	896.9	122.5	...

	12/07/2021	08:00:00	13/07/2021	08:00:00	EST	2161	24	2184	9086.5	36346.3	281.7	49.9	1421.7	163.9	...
	13/07/2021	08:00:00	14/07/2021	08:00:00	EST	2185	24	2208	5997.2	23988.8	192.0	40.6	938.3	115.2	...

F

Chemical profile of factor k



G

High time resolution temporal contribution of factor k

