Comparison of source apportionment approaches in a real-case model application

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Overview

In this study we

- compare PM_{10} tagged species (TS) contributions with brute force impacts (BF, or emission reduction impacts) at different emission reduction levels (ERLs)
- analyse the non-linear response of the studied sources
- examine the behaviour of PM_{10} and its components in selected areas (urban, rural, industrial) where different chemical regimes prevail

The focus is on the SIA formation, with particular reference to ammonium nitrate (NH_4NO_3) and ammonium sulfate ($(NH_4)_2SO_4$).

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Novelties of the study

- Comparison of BF and TS in a real-world situation
- Reduction of sources one by one (by contrast to reduction by precursor)
- Analysis of PM_{10} and its main inorganic components
- Explore the links between the chemical regime and non-linear responses
- Theoretical framework about the factors that influence the linear response of sources

Limitations of the study

- Does not consider the organic secondary PM_{10} components
- Only for year averages

Simulation with tagged species and brute force approaches

- Models: CAMx and FARM
- Tagged species module: PSAT
- Pollutant: PM_{10}
- Area: Po Valley
- Reference year: 2010
- Time window: full year
- Domain: 580 x 400 km^2
- Grid step: 5 km x 5 km approx.
- Meteorology: WRF 14 layers
- Emissions: EMEP (Europe), ISPRA (Italy), INEMAR (regional) processed with SMOKE
- Thermodynamic model: ISORROPIA (Nenes et al., 1998)

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Non linearity

Types of linear/non-linear responses analysed in this study:

a) match sum of the single sources and the total PM_{10} mass (= additivity)

indicator: similarity between TS and BF (because TS is mass conservative)

b) match single source emission reduction and multiple source emission reduction (= additivity)

indicator: interaction terms ($\hat{c}$) of the Stein and Alpert (1993) decomposition

c) match change in precursor emissions and change in PM_{10} concentrations (= consistency)

indicator: comparison between different emission reduction levels (ERLs)

In addition, we have studied the link between non-linearity and chemical regime by combining:

- Interaction terms
- The “free ammonia” / “total nitrate” ratio (GR), Ansari and Pandis (1998)

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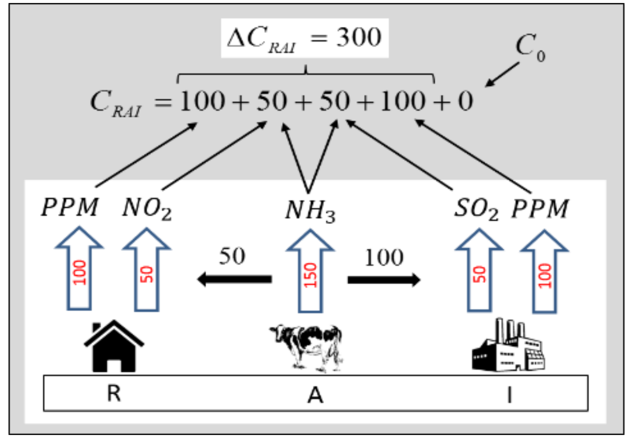
Stein and Alpert decomposition (theoretical example)

Stein and Alpert decomposition
2 sources

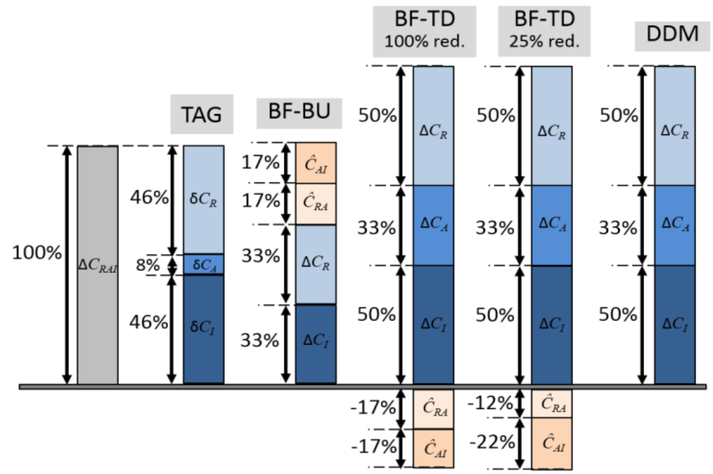
$$\Delta C_{AB} = \Delta C_A + \Delta C_B + \hat{C}_{AB}$$

Stein and Alpert, 1993



Stein and Alpert decomposition
3 sources

$$\Delta C_{ABC} = \Delta C_A + \Delta C_B + \Delta C_C + \hat{C}_{AB} + \hat{C}_{AC} + \hat{C}_{BC} + \hat{C}_{ABC}$$



Clappier et al., 2017

The interaction terms ($\hat{C}$) of the factor decomposition measure the consistency between the impacts obtained with single source reductions compared to those of multiple source reductions and therefore can be used as indicators of non-linearity.

Simulations with Brute Force approach

Simulation set / Reduced sources	CAMx 100%	CAMx 50%	CAMx 20%	FARM 50%	FARM20%
No reduction	Base case CAMx			Base case FARM	
AGRICULTURE (AGR)	x	x	x	x	x
INDUSTRY (IND)	x	x	x	x	x
ROAD TRANSPORT (TRA)	x	x	x	x	x
RESIDENTIAL (RES)				x	x
AGR-IND	x	x	x	x	x
AGR-TRA	x	x	x		x
IND-TRA	x	x	x	x	x
RES-IND				x	x
RES-TRA				x	
RES-AGR				x	
AGR-IND-TRA	x	x	x		x
RES-IND-TRA				x	x

Simulations in blue are not discussed in this presentation

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“free ammonia” / “total nitrate” Gas Ratio

The gas ratio (GR, Ansari and Pandis,1998) was used to define the chemical regime in each of the simulations :

$$GR = ([NH_3] + [NH_4^+] - 2[SO_4^{2-}]) / ([HNO_3] + [NO_3^-])$$

where concentrations are $nmol.m^{-3}$ or in $nmol.mol^{-1}$ of air (ppb).

The GR value defines three different chemical regimes:

(a) $GR>1$, in which NH_4NO_3 formation is limited by the availability of HNO_3,

(b) $0<GR<1$, in which NH_4NO_3 formation is limited by the availability of NH_3, and

(c) $GR<0$, in which NH_4NO_3 formation is inhibited by H_2SO_4

Situations giving rise to non-linearity

- **Double counting**

when the concentration of two PM_{10} precursors is near to stoichiometric ratios, the sum of single source reduction is double (or near) than the double source reduction at once.

- **Precursor limited chemical regime**

the reduction in the emissions of the non-limiting precursor leads to a response which is less than double counting

- **Competition**

the interaction between two sources A and B can be affected by a third one C when the precursors emitted by sources B and C compete to react with the one emitted by source A

- **Thermodynamic equilibrium**

when the reaction products (NO_3^-, NH_4^+) are in thermodynamic equilibrium with the reagents (NH_3, HNO_3)

- **Compensation**

processes which alter the ΔC expected to result from a given ΔE in a theoretical exercise (e.g. advection, influence of sources not considered in the studied system, etc.)

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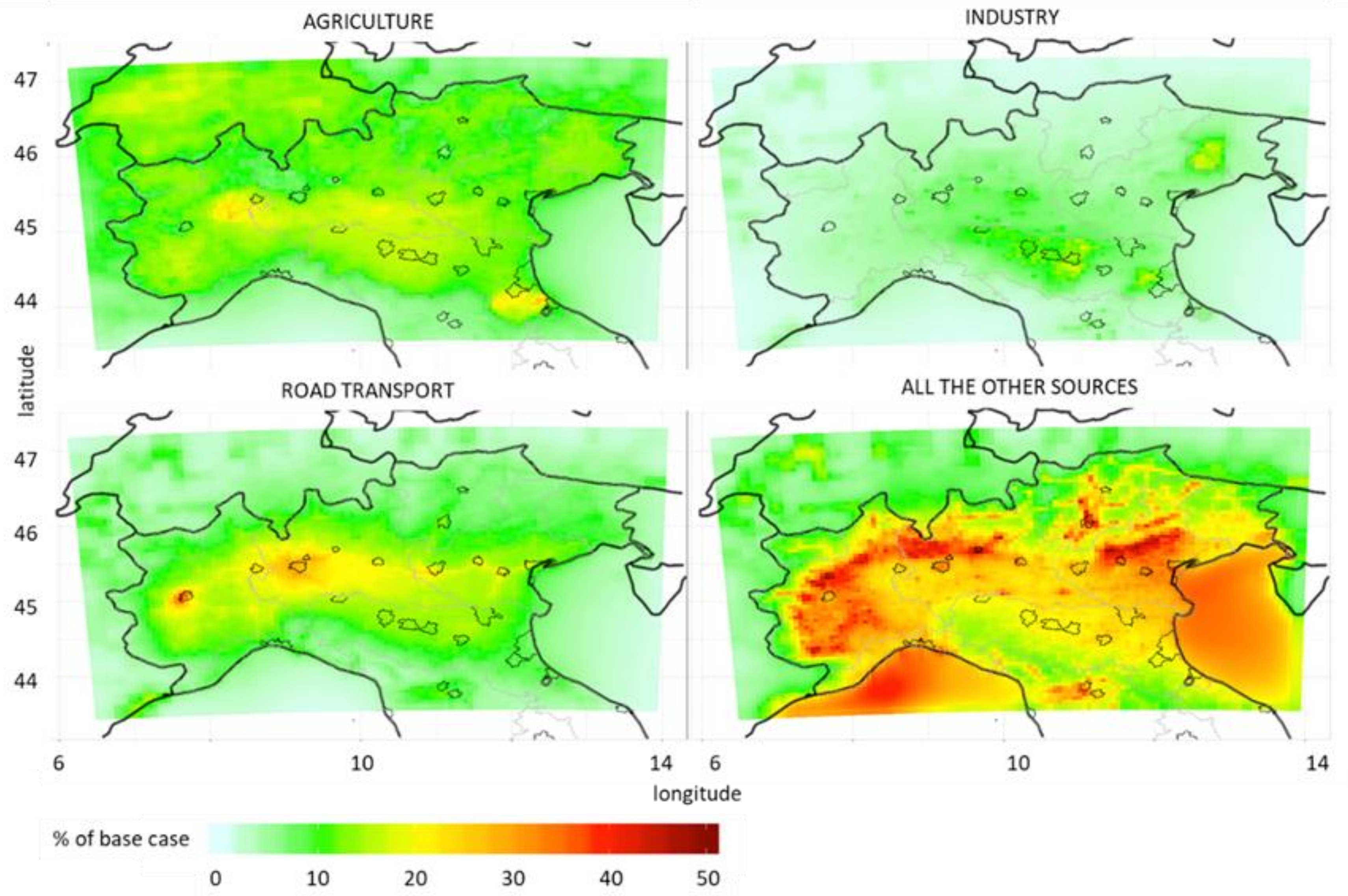
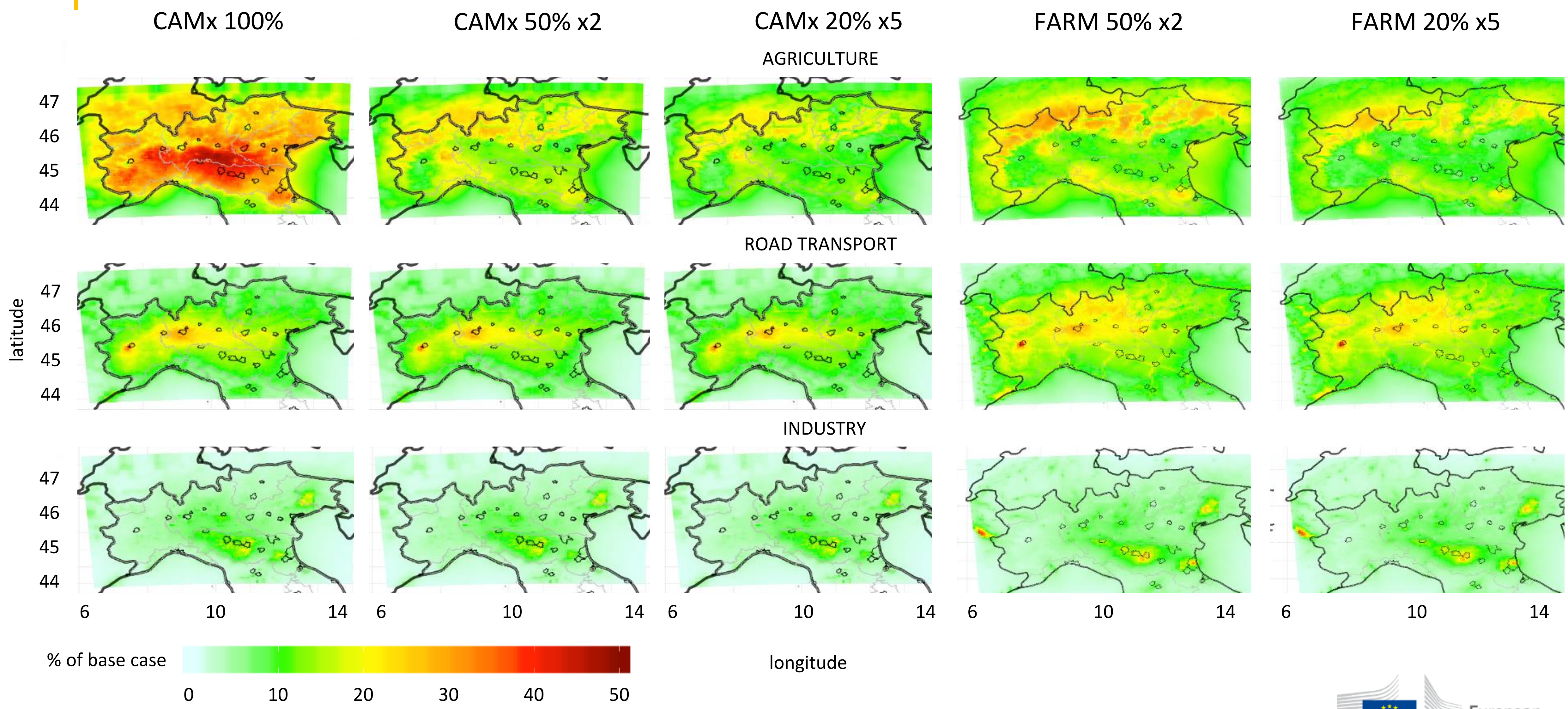
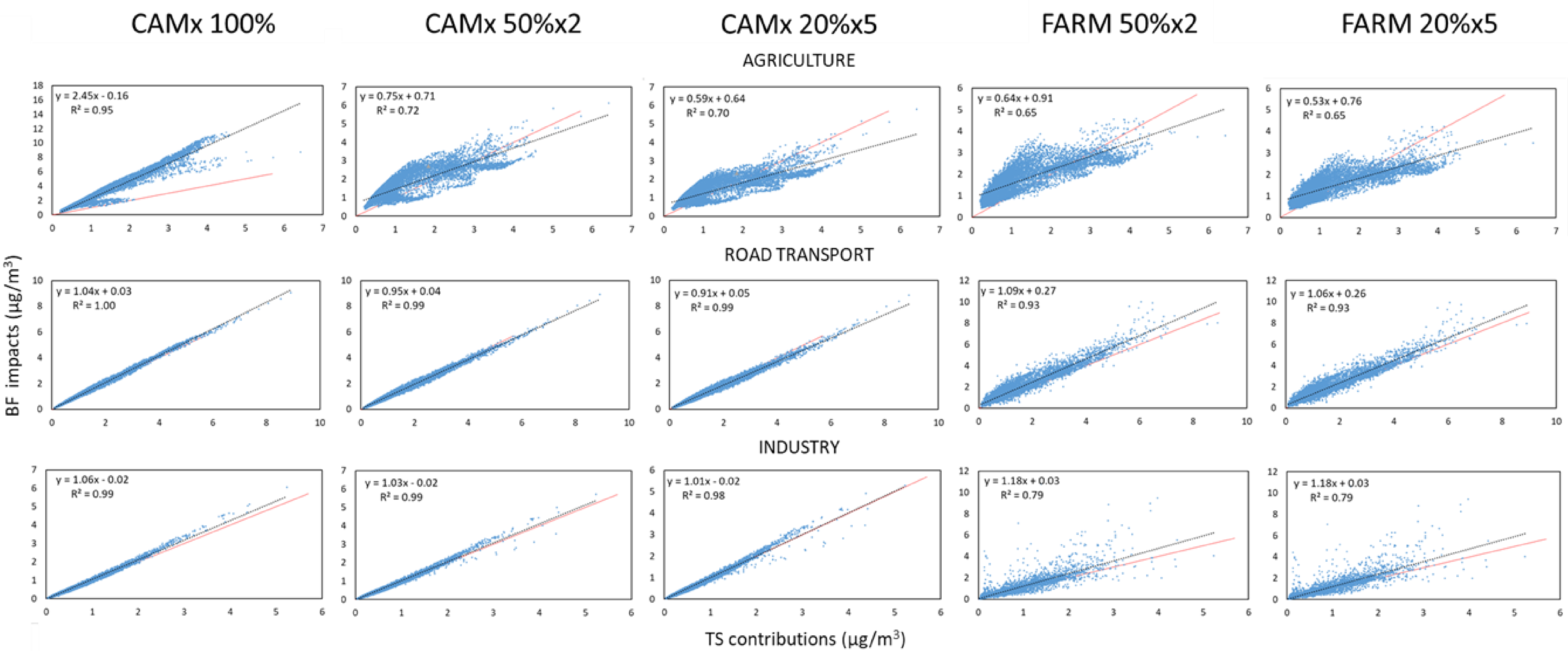
Annual contributions of PM_{10} sources, TS (CAMx PSAT)



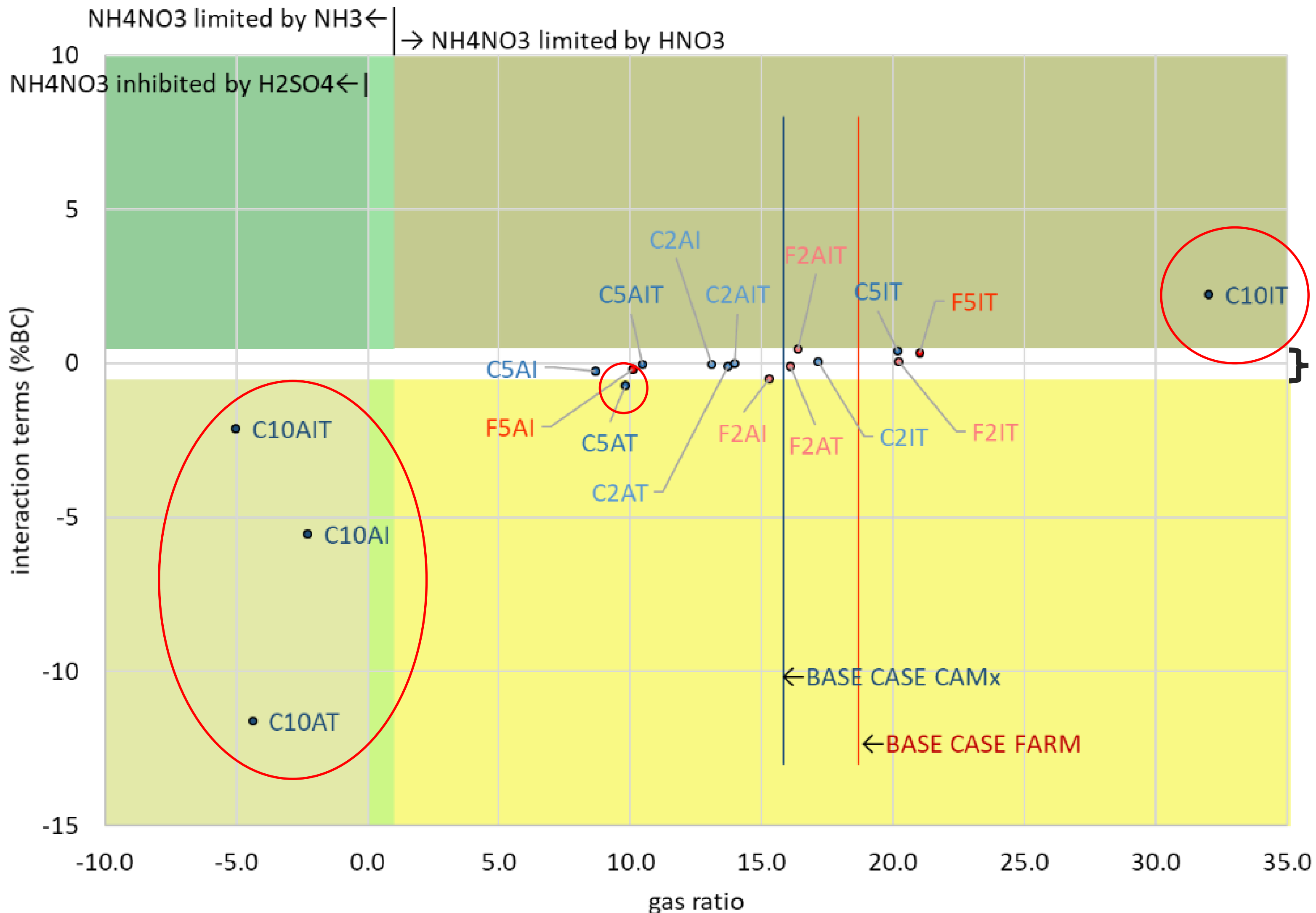
Annual impacts of sources on PM_{10}, BF



PM_{10} annual contributions (TS) vs impacts (BF)



GR vs interaction terms in a rural site (Cremona province)



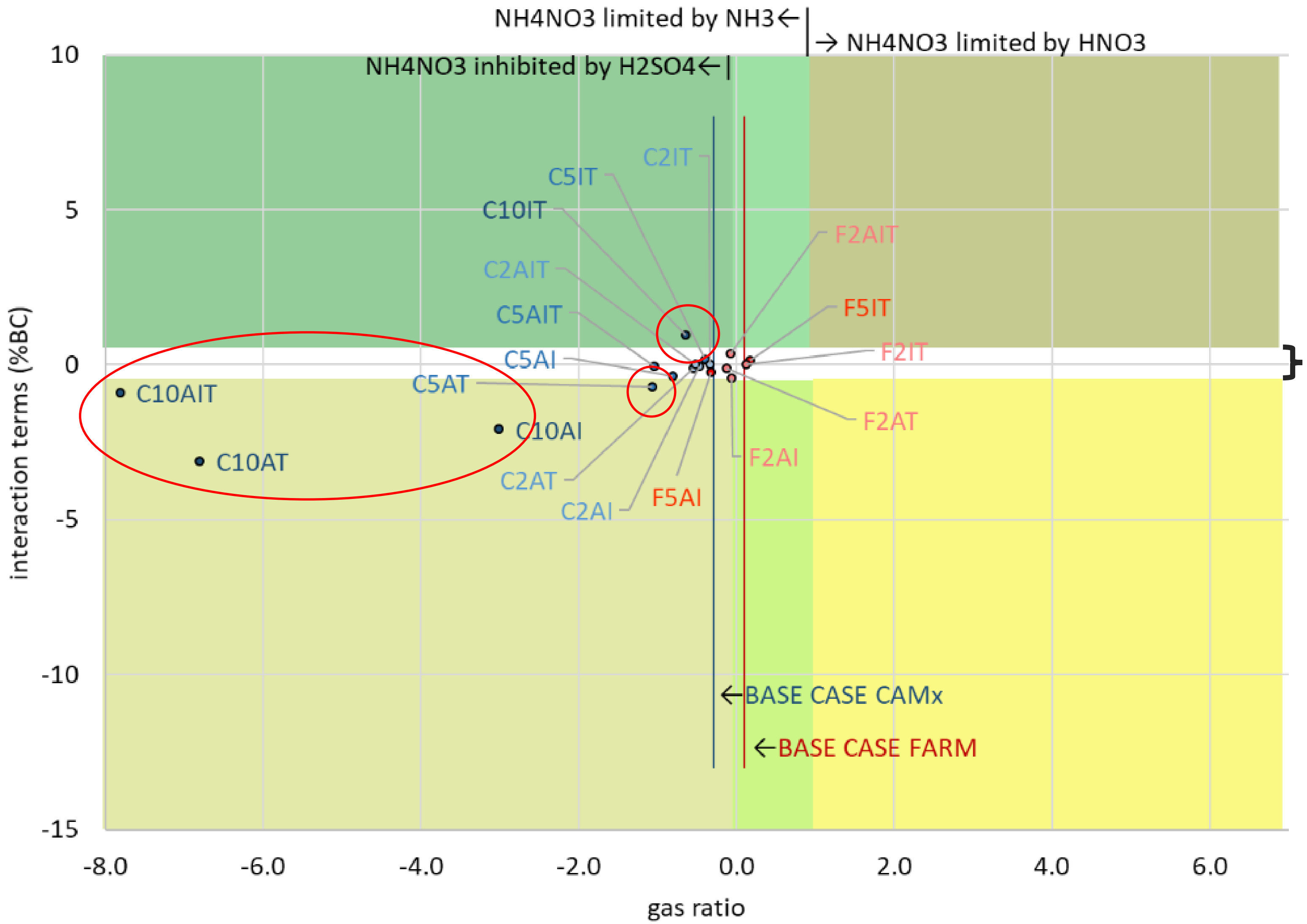
interaction terms > 0

negligible interaction terms

interaction terms < 0

Legend
- C: CAMx and F: FARM.
- 10, 5 and 2 indicate the 100%, 50% and 20% ERLs, respectively.
- A: agriculture, I: industry and T: transport

GR vs interaction terms in an urban site (Milan)



interaction terms > 0

negligible interaction terms

interaction terms < 0

Legend
- C: CAMx and F: FARM.
- 10, 5 and 2 indicate the 100%, 50% and 20% ERLs, respectively.
- A: agriculture, I: industry and T: transport

Conclusions

- The differences in PM_{10} attributed to AGR between TS and BF at 100% ERL reach a factor 2.
- There are observable inconsistencies between different ERLs, in particular between 100% and 50% and 20%.
- The joint analysis of the interaction terms and the gas ratio (GR) showed how changes in the chemical regime leads the non-linear response of PM_{10} concentrations to emissions reductions.
- The association between AGR with non-linear responses is due to the key role of NH_3.
- TS and BF lead to comparable PM_{10} apportionments for the other studied sources IND, TRA (and RES, not shown).

Conclusions 2

- When there is a clear non-linear response (interaction terms), precaution is needed in the interpretation of the results from both BF and TS:

  - in BF it is not appropriate to sum of the impact of the sources obtained by single source reduction because they may not match the total PM (additivity),

  - TS leads to distortion in the allocation of secondary aerosol because it does not account for indirect effects

- Moreover, in case of non-linear responses, also extending the results of BF for a specific ERL to another (e.g. 20 to 50 or 100%) could be misleading (consistency).

- More work is needed to check the linearity for shorter averaging intervals (daily, hourly) and for secondary organic aerosol.

Thank you

Belis, C. A., Pirovano, G., Villani, M. G., Calori, G., Pepe, N., and Putaud, J. P.: Comparison of source apportionment approaches and analysis of non-linearity in a real case model application, Geosci. Model Dev., 14, 4731–4750, https://doi.org/10.5194/gmd-14-4731-2021, 2021.

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