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Evaluation of the MONARCH-simulated NO_2 and HCHO tropospheric columns against Sentinel-5P TROPOMI observations over the Iberian Peninsula

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Context

MITIGATE project that aims at getting insights on the sensitivity of O_3 to NOx and VOC precursors over Spain using TROPOMI tropospheric columns (TrC) observations, combined with surface in-situ observations and air quality modeling.

Data

- TROPOMI TrC-NO_2 and TrC-HCHO (PAL and OFFL products), preprocessed with the CAMS Satellite Operator (CSO) tool
- Surface NO_2 and O_3 observations from EEA AQ eReporting database, with quality assurance filtering (using GHOST)
- MONARCH air quality simulation (0.1°x0.1°) over year 2019, with HERMESv3 bottom-up anthropogenic emissions

Overall statistics

	Pollutant	Timescale	Unit	MB	nMB	RMSE	nRMSE	slope	PCC	N	Mo	Mm
Tab1a	O_3	h	ppbv	2.3	7	12.2	38	0.61	0.66	1505025	32.0	34.2
	O_3	d	ppbv	2.2	7	9.3	29	0.59	0.63	62624	32.0	34.3
	O_3	d1max	ppbv	0.5	1	9.3	20	0.66	0.69	62624	45.7	46.3
	O_3	d8max	ppbv	1.5	3	9.0	21	0.69	0.70	61251	42.1	43.6
	NO_2	h	ppbv	-1.8	-24	7.5	99	0.66	0.64	1129145	7.6	5.8
	NO_2	d	ppbv	-1.9	-24	5.3	66	0.76	0.73	43643	8.0	6.1
	NO_2	d1max	ppbv	-2.9	-16	13.1	70	0.76	0.65	43643	18.7	15.8
	NO_2	d8max	ppbv	-2.6	-19	8.9	65	0.79	0.71	39114	13.7	11.1
	TrC-NO_2	d	Pmolec/cm^2	-0.3	-25	0.9	62	0.44	0.67	1435065	1.4	1.1
	TrC-NO_2	md	Pmolec/cm^2	-0.3	-24	0.5	36	0.65	0.87	65819	1.4	1.0
	TrC-HCHO	d	Pmolec/cm^2	-1.4	-26	5.1	96	0.26	0.47	1626707	5.3	3.9
	TrC-HCHO	md	Pmolec/cm^2	-1.3	-26	1.9	37	0.85	0.88	65817	5.1	3.7
Tab1b	NO_2(RAW)	13UTC	ppbv	-2.1	-43	4.6	95	0.52	0.62	33361	4.8	2.7
	(corr:HNO3/PAN)	13UTC	ppbv	-1.5	-39	3.9	100	0.59	0.69	33361	3.9	2.4
	(corr:HNO3/PAN/NTR)	13UTC	ppbv	-1.2	-36	3.5	106	0.62	0.72	33361	3.3	2.1
	TrC-NO_2	d	Pmolec/cm^2	-0.8	-28	2.2	79	0.42	0.76	33361	2.8	2.0

Tab1a: Overall evaluation of MONARCH over Spain in 2019 at hourly (h), daily mean (d), daily 1-hour maximum (d1max), daily 8-hour maximum (d8max), and monthly (md) time scales.

Tab1b: Specific evaluation of surface NO_2 in early afternoon with raw observations, and using the corrections proposed by Lamsal et al. (2008) :
"RAW" : no correction
*"HNO3/PAN/NTR" : (NO_2/(NO_2 + 0.95*PAN + 0.35*HNO_3 + NTR)*
*"HNO_3/PAN" : (NO_2/(NO_2 + 0.95*PAN + 0.35*HNO_3)*

Discussion of NO_2 results

Negative bias on both surface NO_2 and TrC-NO_2 (*Tab1a, Fig1*)

Possible issues in the observations :

- **Part of the negative bias on surface NO_2 possibly due to positive artefacts in the chemiluminscence measurements**, but cannot explain the strong bias in early afternoon (*Tab1b, Fig2*)
- **Expected negative artifact on the TROPOMI TrC-NO_2 measurements** (from -23 to -37% in clean/slightly polluted, and -51% in highly polluted areas)

Fig1

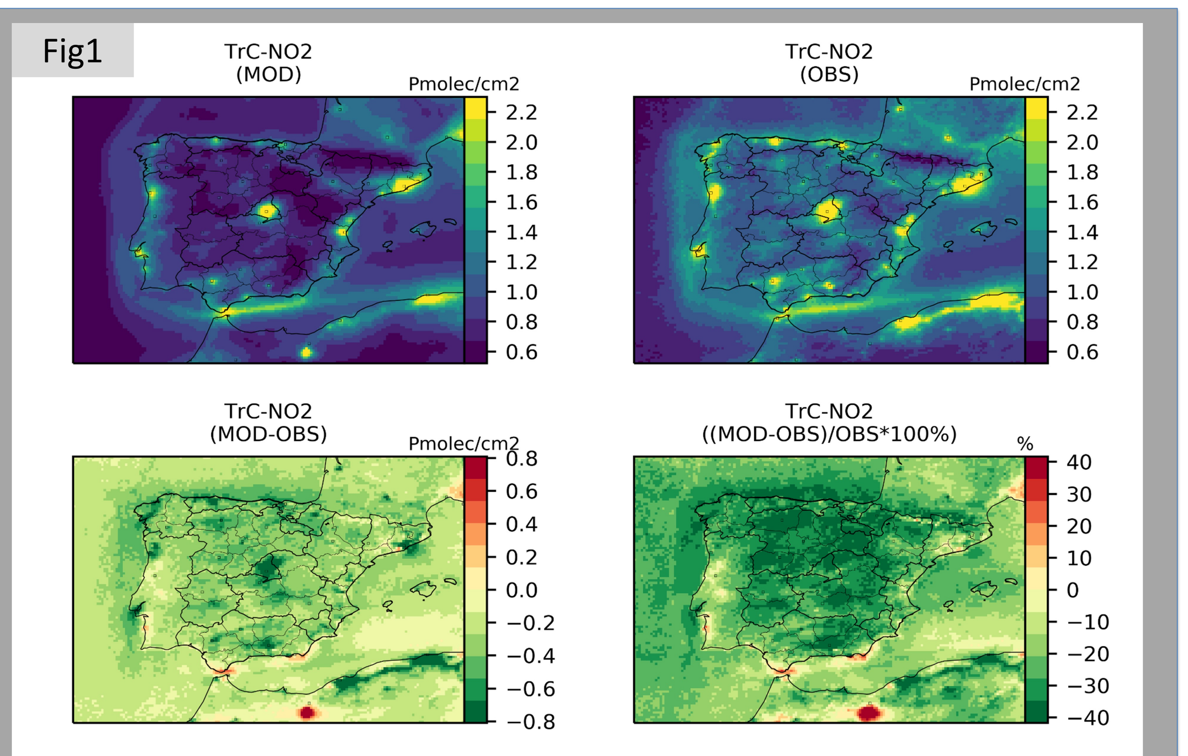

Fig1: Annual map of TrC-NO_2 in MONARCH, TROPOMI, and absolute and relative differences.

Fig2

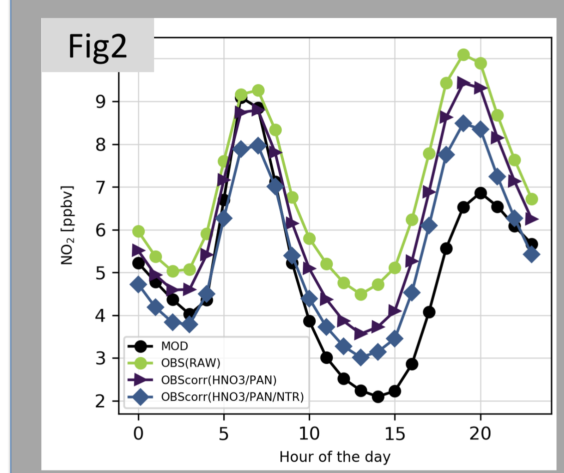

Fig2: Mean diurnal profile of surface NO_2 in MONARCH and in observations, without and with correction from Lamsal et al. (2008) (see caption of Tab1).

Fig3

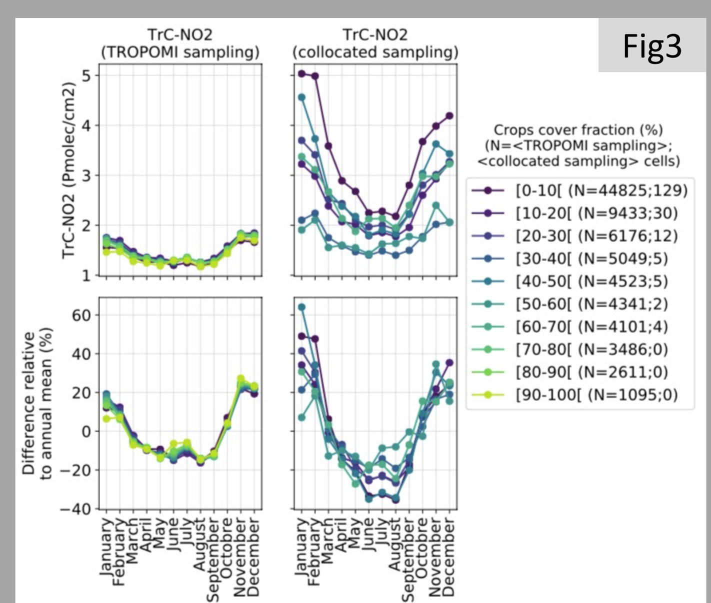

Fig3: Monthly profiles of TROPOMI TrC-NO_2 with full sampling (left panels) or collocated with surface monitoring stations (right panels), as a function of the crops cover fraction over Spain in 2018-2021 (from Petetin et al., 2023)

Possible issues in the model, explored with sensitivity tests :

- Vertical diffusion coefficient (Kz) divided by 2 : bias on surface NO_2 improved from -20 to -10% but no change on TrC-NO_2
- Soil NO emissions multiplied by 10 : bias on surface NO_2 improved from -20 to 0%, bias on TrC-NO2 improved from -29 to -12%, but PCC slightly reduced from 0.87 to 0.82 in TrC-NO2. TROPOMI observations show a slight increase of TrC-NO2 in June-July in areas dominated by crops (*Fig3*)

Discussion of HCHO results

Reasonably good spatial distribution of TrC-HCHO but generalized negative bias over the Iberian Peninsula (*Tab1a, Fig4*)

Possible issues in the observations :

- TROPOMI TrC-HCHO measurements may be positively biased in low-HCHO areas, and negative biased in high-HCHO areas (Vigouroux et al., 2020)

Sensitivity test :

- Biogenic emissions multiplied by 2 : bias on TrC-HCHO changed from -14 to +10%, and slope improved from 0.27 to 0.39, thus **uncertainties on biogenic VOC emissions could explain part of the negative bias on TrC-HCHO but no improvement of correlation here**

Fig4

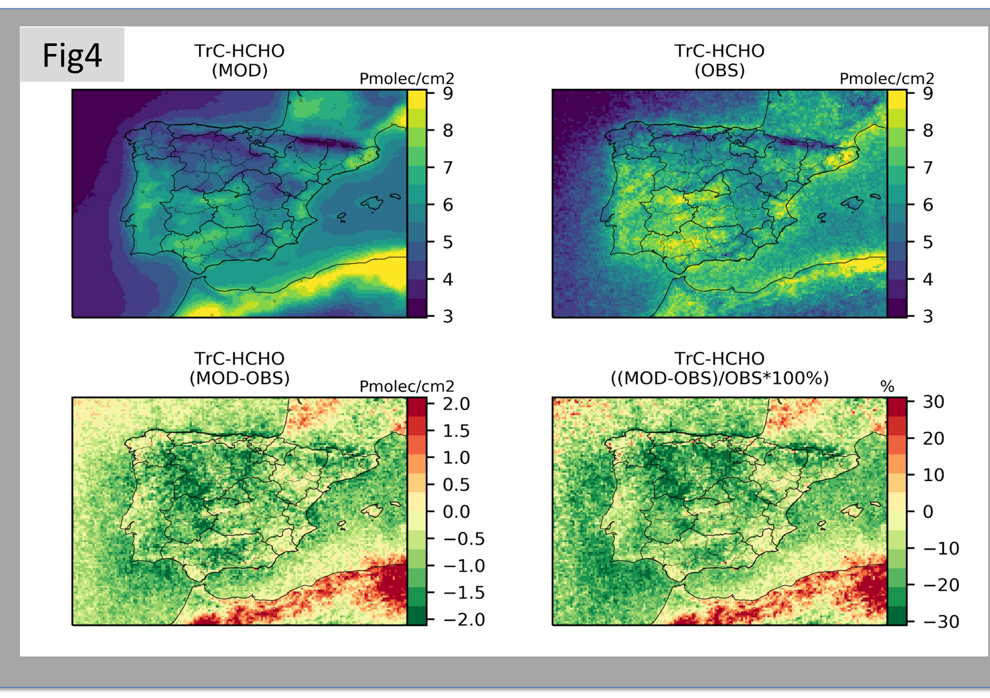

Fig4: Annual map of TrC-HCHO in MONARCH, TROPOMI, and absolute and relative differences.

Exploring the sensitivity of O_3 to its NOx and VOC anthropogenic precursors

Different emission scenarios simulated with MONARCH:

- REF : base case emissions
- ANOx25 : -25% of NOx anthropogenic emissions over Spain
- AVOC25 : -25% of VOC anthropogenic emissions over Spain

Tab2

Variable	Unit	Timescale	Mean	Response to NOx	Response to VOC
O_3	ppbv	d8max	54.5	-1.4 (-2.6%)	-0.0 (-0.0%)
O_3	ppbv	d	43.0	-0.8 (-1.8%)	-0.0 (-0.0%)
NO_2	ppbv	d	1.5	-0.3 (-17.3%)	0.0 (+0.2%)
HCHO	ppbv	d	2.0	-0.0 (-2.2%)	-0.0 (-0.9%)
VOC	ppbv	d	40.2	0.3 (+0.6%)	-0.9 (-2.7%)
E[NOx]	kg/m2/s	d	8.7e-10	-2.1e-10 (-24.5%)	2.4e-20 (+0.0%)
E[VOC]	kg/m2/s	d	1.3e-09	7.7e-22 (-0.0%)	-3.1e-10 (-24.8%)
TrC-NO_2	Pmolec/cm2	d	0.9	-0.1 (-6.0%)	0.0 (+0.2%)
TrC-HCHO	Pmolec/cm2	d	9.1	-0.1 (-1.1%)	-0.0 (-0.2%)

Tab2: Impact of -25% reduction of NOx and VOC anthropogenic emissions.

On average over Spain, ANOx25 leads to -17 and -6% of surface NO_2 concentrations and TrC-NO_2, respectively; minor impact of AVOC25 on HCHO (±0.2%). When averaged over the entire country, **$O_3^{(d8max)}$ decreases by -2.6% in ANOx25, but almost no response to AVOC25.**

When analysing the $O_3^{(d8max)}$ response against TrC-($HCHO/NO_2$) (*Fig5*), results appear consistent with past studies (e.g., Martin et al., 2004; Jin et al., 2017): **the $O_3^{(d8max)}$ response to ANOx25 shifts from negative to positive values when TrC($HCHO/NO_2$) increases but relatively large ambiguous/transition zone where TrC-($HCHO/NO_2$) values alone are insufficient for predicting the sign of the O_3 response.**

Fig5

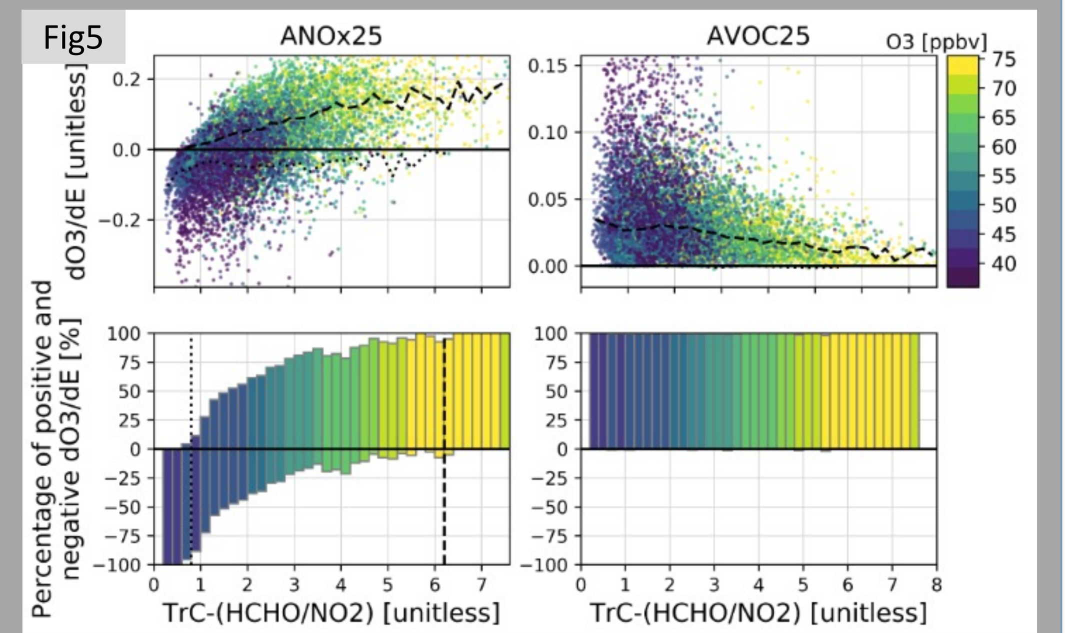

Fig5: Surface $O_3^{(d8max)}$ response to ANOx25 and AVOC25 emission scenarios over Spain, against modelled TrC-($HCHO/NO_2$) ratios (as seen by TROPOMI, i.e. applying averaging kernels). Only polluted cells with TrC-NO_2 above 2.5 Pmolec/cm² are considered here, period April-September 2019.

When analysing the probability of high surface $O_3^{(d8max)}$ against TrC-NO_2 and TrC-HCHO (*Fig6*), we see that these space-based information can provide some insights on these $O_3^{(d8max)}$ episodes. **TROPOMI observations may be used in combination with model-based relationships between O_3 and TrC-NO_2 and TrC-HCHO in order to get some probabilistic information on the category of O_3 response** (*Fig7*), but more in-depth investigations are required to get more robust and quantitative information on the O_3 sensitivity to NOx and VOC precursors

Fig6a

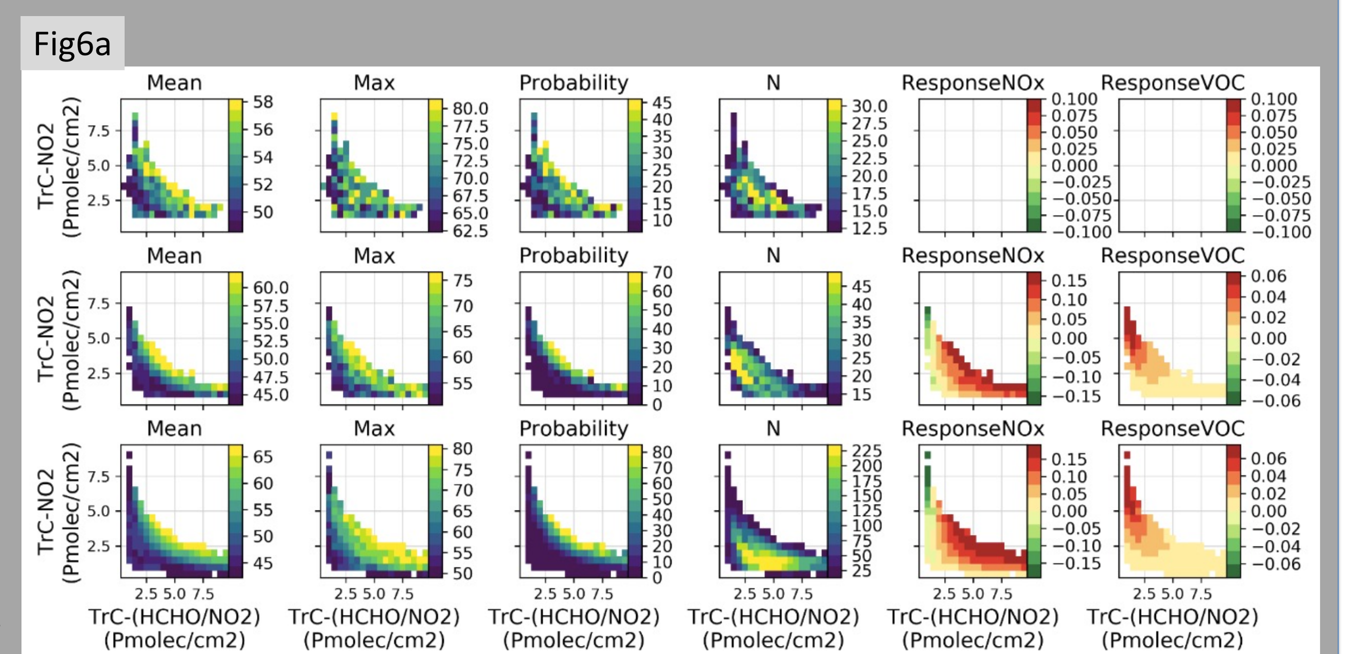

Fig6b

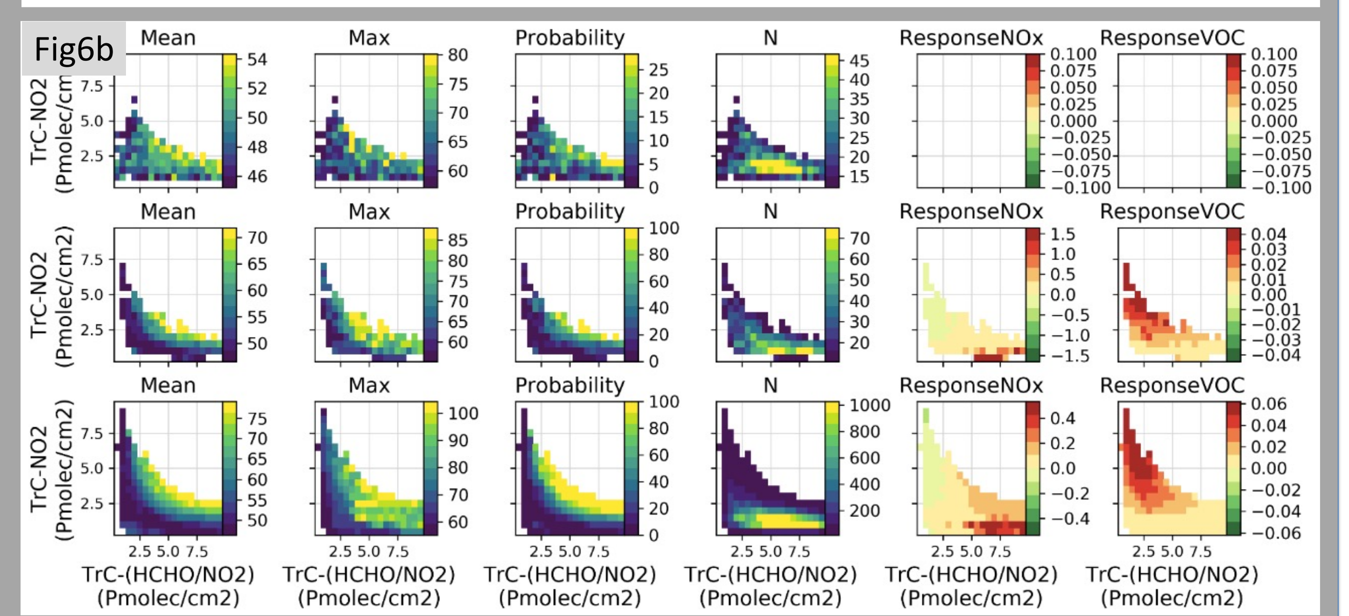

Fig6: Mean $O_3^{(d8max)}$, maximum $O_3^{(d8max)}$, probability of $O_3^{(d8max)}$ above 60 ppbv, number of points, and $O_3^{(d8max)}$ response to ANOx25 and AVOC25 emission scenarios, in observations (top panels), MONARCH collocated with observations (middle panels) and all MONARCH (bottom panels), in Madrid region (Fig6a) and Catalonia (Fig6b).

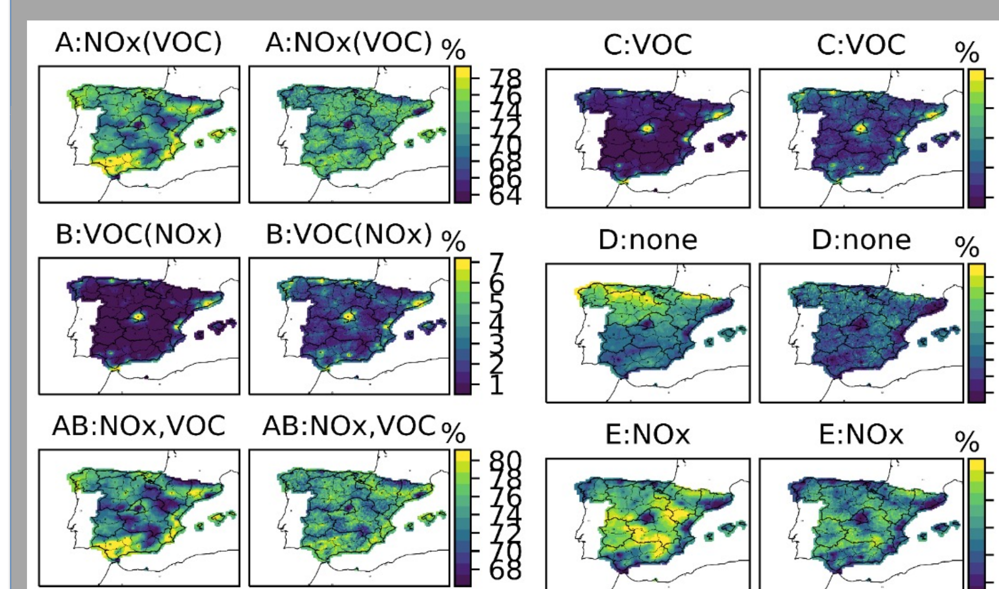

Fig7

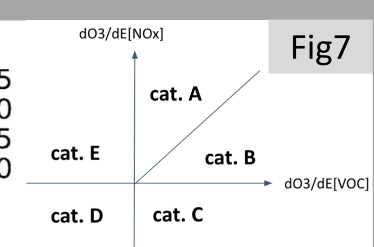

Fig7: Probability of occurence of different categories (from A to E) of O_3 response to NOx and VOC emission reductions as obtained in MONARCH (panel columns 1 and 3), and scaled using the observed TrC-($HCHO/NO_2$) observations (panel columns 2 and 4).

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