The sensitivity of surface ozone concentration to geographically-distributed VOC and NOx emissions over Kao-Ping air basin in Taiwan

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1. INTRODUCTION

Kao-Ping air basin (KPAB) located in the southwestern region of Taiwan has been recognized for O_3 non-attainment area in years mainly due to its high dense population, intensive industrial outputs and heavy traffic volume (Taiwan Environmental Protection Administration, 2015). Furthermore, the topography and weather condition in southern Taiwan also favor O_3 formation in this region. The Central Mountains Range perform a cumulative effects of air pollutant in downwind of Kao-Ping air basin due to unfavorable dispersion effect, which leads to more elevated air pollution in KPAB than in other upwind regions. Notably, the highest O_3 concentration occurred particularly in Autumn and the monthly averaged O_3 concentrations increase from northern to southern Taiwan, indicating that transport of emission plays important role in O_3 pollution formation in Southern Taiwan.

2. OBJECTIVES

We use the community multiscale air quality (CMAQv5.0.2) coupled with high decoupled direct method (HDDM) to quantify O_3 sensitivity and contributions of NOx and VOC emissions from local and upwind regions in KPAB under the typical stagnation conditions that existed on 1–5 October, 2010, with an emphasis on the nonlinearity of the response. In particular, we model the interactions between KPAB and other air basins emissions sources and quantify the impacts of emissions inventory perturbations on the calculated responses of ozone to emission controls. In addition, a backward trajectory analysis was employed to locate the source of the accumulated O_3. The result is expected to determine which sources are responsible for O_3 formation in KPAB quantitatively and to characterize O_3 responses for hypothetical emission reductions management strategies.

3. METHODS

Model configuration

- CMAQ DDM v5.0.2 with CB05 (gas chemistry) and AERO6 (aerosol chemistry) is driven by meteorological fields generated by the Weather Research and Forecasting (WRF) version 3.7
- The one-way nested approach with four-dimensional data assimilation (FDDA) in WRF3.7 was performed to construct model domain with 80×70 horizontal grids at a resolution of 81 km × 81 km (Domain 1) down to 80×70 horizontal grids at a resolution of 27 km × 27 km (Domain 2), 80×70 horizontal grids at a resolution of 9 km × 9 km (Domain 3) and 135×90 horizontal grids at a resolution of 3 km × 3 km (Domain 4), with all domains centered at 25^0N, 125^0E nested on a Lambert conformal projection over East Asia shown in Figure. 1.
- CMAQ modeling performance validation was set up to simulate the period of September 28 to October 31 in 2010 (3 days spin-up run).
- Initial and boundary (IC/BC) conditions of CMAQ were derived from GEOS-Chem which is incorporated various emission inventories for different sectors, including MIX Asia Emission Inventory with a data resolution of 0.25^0x0.25^0 for anthropogenic emission over Asia was used. Biogenic emission is generated by the Model of Emissions of Gases and Aerosols from Nature (MEGANv2.10). Biomass burning emission is developed from the Global Fire Emissions Database (GFED).
- The emission inventory used in 3-km domain was developed from the Taiwan Emission Data System (TEDS 8.1) (Table 1 and Figure 2) and Taiwan Biogenic Emissions Inventory System (TBEIS-2) released by the Taiwan EPA with a data resolution of 1 km ×1 km.

WPS Domain Configuration

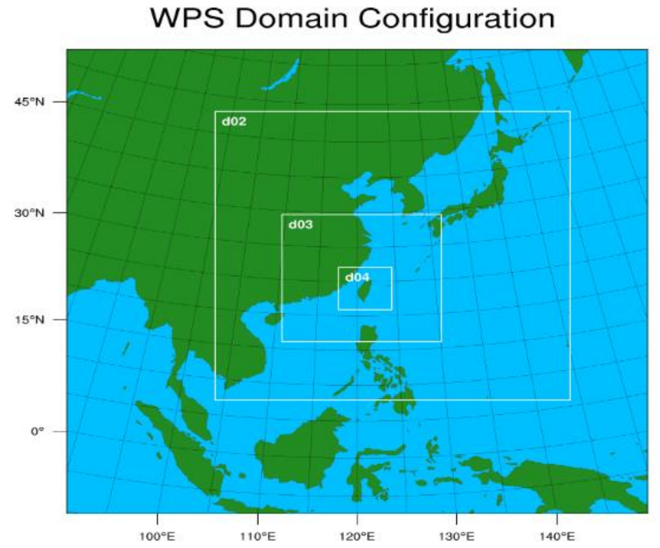


Figure. 1 CMAQ modeling domain

Table 1. Episode average anthropogenic NOx and VOC Emissions rate (ton/day) by defined sensitivity regions in October 1-31, 2010&

Regions	ANOx	AVOC
NCMAB	301 (100%)	830 (97%)
CTAB	122 (100%)	226 (74%)
YCNAB	211 (100%)	183 (64%)
KPAB	222 (100%)	264 (81%)
YLHTAB	152 (100%)	90 (23%)

&The percentage of total emission was shown in parentheses.

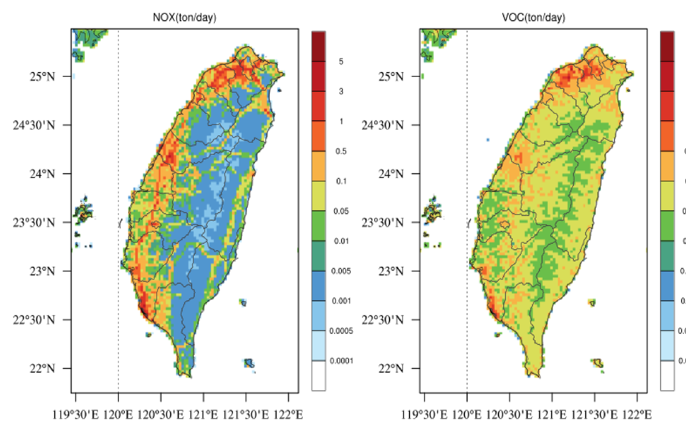


Figure 2. Episode average anthropogenic NOx and VOC emissions (ton/day) in October 1-31, 2010.

Sensitivity analysis

- The period modeled for sensitivity analysis during 1–5 October 2010 is chosen, a high-pressure system was located over northern China, showing the subsidence accompanied by the strong high-pressure system led to a clear, sunny sky over the KPAB.
- CMAQ-HDDM simulations were performed to investigate the response of O_3 formation in the KPAB to geographically-distributed reductions of anthropogenic emissions of NOx (ANOx) and VOC (AVOC) emission precursors from local (i.e. Kao-Ping Air Basin (KPAB) and other 4 upwind regions i.e. North and Chu-Miao Air Basin (NCMAB), Central Air Basin (CTAB), Yun-Chia-Nan Air Basin (YCNAB), and Yi-Lan and Hua-Dong Air Basin (YLHDAB) as shown in Table 2 and Fig. 3.

Table 2. CMAQ-HDDM sensitivity experiments

Order	Regions	Sensitivity name
First	NCMAB(North+ChuMiao) CTAB(Central) YCNAB(YuChiNa) KPAB (KaoPing) YLHDAB (Ilan+HuaTung)	NCMNOX, IHTNOX, CTNOX, YCNNOX, KPNOX (same as VOC)
Self-Second		2NCMNOX, 2IHTNOX, 2CTNOX, 2YCNNOX, 2KPNOX (same as VOC)
Cross-second		2NCRO,2CCRO,2YCRO,2KCRO, 2ICRO
	Domain-wide	TWNBC, TWNIC

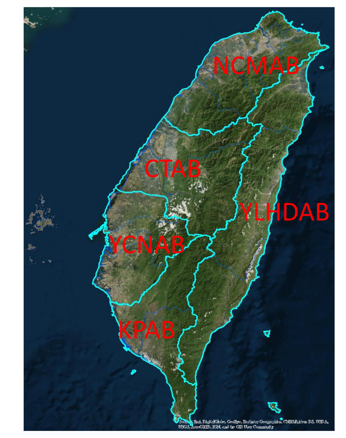


Figure 3. Defined regions for sensitivity analysis

4. RESULS

- Simulated surface meteorological fields were examined against surface hourly observations from 20 ground observation sites of Taiwan Central Weather Bureau (CWB) across whole nation.

Table 3. Bias and error in WRF-generated meteorological parameter fields during October 1-31, 2010 with respect to the Taiwan Central Weather Bureau (CWB) surface observations

	T(^{0}K)		WS(m/s)		WD(%)	
	MBE	MAGE	MBE	RMSE	WNMB	WNME
NCMAB	-0.8	1.3	1.9	2.7	-0.3%	11%
CTAB	-0.6	1.3	2.1	2.8	2%	9%
YCNAB	-1.1	1.5	0.2	1.5	-1%	9%
KPAB	-1.3	1.6	0.6	1.8	-1%	10%
YLHDAB	-0.9	1.5	1.9	2.8	5%	11%
Average	-0.8	1.3	1.9	2.7	-0.3%	11%

- Simulated surface air pollutant concentration were from 15 air quality monitoring stations of Taiwan Air Monitoring Station Network (TAQMN) over KPAB

Table 4. Key model performance metrics for the O_3 during the October of 2010

	O_3	O_3(>60ppb)	O_3(1hr-max)	O_3(8hr-max)	NO_2	VOC
Num. of obs.	10,973	2,221	11,160	11,160	10,937	11,076
Mean obs.	37.8	80.4	83.0	65.6	17.8	244.8
Mean mod.	35.6	76.2	74.4	64.8	30.6	307.7
Mean bias (MB), ppb	3.3	2.9	4.8	4.7	2.7	-22.6
Mean absolute gross error (MAGE), %	16.1	15.1	18.9	14.1	11.3	125.6
Mean normalized mean bias (MNB), %	28.5	-5.8	10.6	8.7	19.5	14.4
Mean normalized error (MNE), %	43.1	22.9	22.5	21.5	63.6	53.7
Root mean Square error (RMSE), ppb	20.5	20.5	21.3	21.4	15.6	186.2
Correlation coefficient (R)	0.7	0.7	0.5	0.6	0.4	0.3

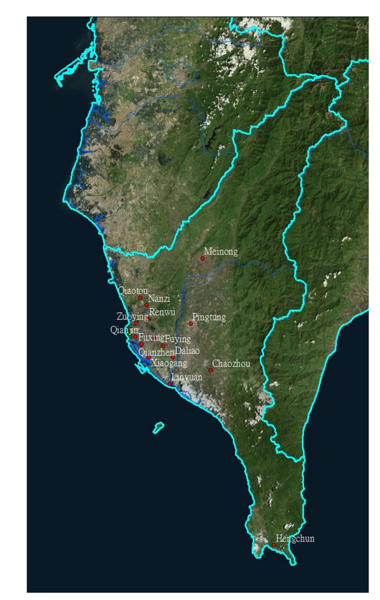


Figure 4. Domain-wide for modeling evaluation and sensitivity analysis

- The comparison of first- and second order sensitivity coefficients between CMAQ-HDDM and those approximated by the brute force method for the response of ozone concentrations to perturbations in domain-wide (3-km domain) anthropogenic emissions ANOx and AVOC

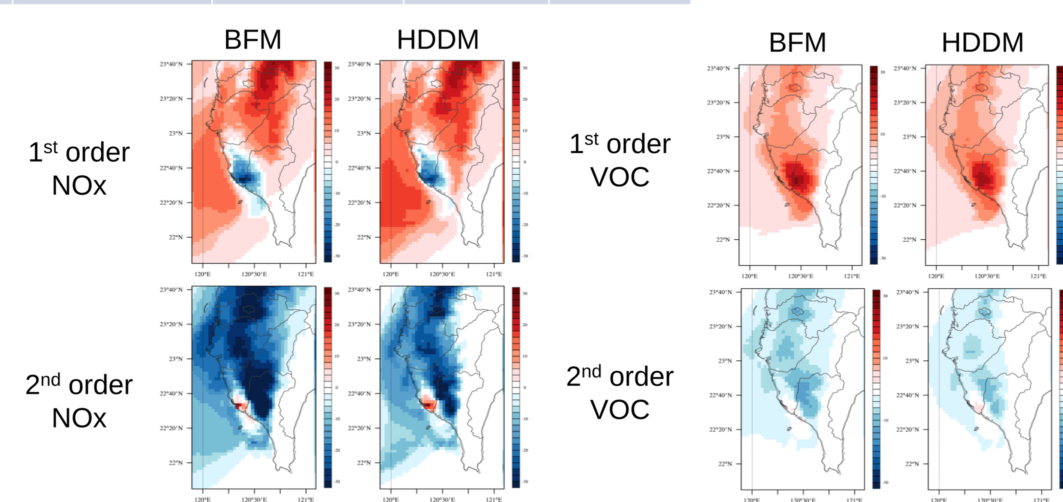


Figure 5- Spatial comparison between HDDM and BFM for the 8-hour daily average ozone sensitivity (ppb) to domain-wide ANOx and AVOC for October 1-5, 20103

Table 5. Statistical comparison of daily maximum 8h O_3 concentration of brute force method and HDDM for October 1-5, 2010

Emission	[a]BFM(μg/m³)	[b]DDM(μg/m³)	[c]NMB(%)	[d]NME(%)	R^2
1st order domain-wide ANOx	6.56	7.69	17.32	18.44	0.97
1st order domain-wide AVOC	4.26	4.7	10.16	11.87	0.99
2nd order domain-wide ANOx	-11.95	-7.63	-36.17	36.63	0.93
2nd order domain-wide AVOC	-3.05	-1.91	-37.21	37.73	0.95

[a]Calculated by eqs. 1 using 10% perturbations. [b]Averaged over domain. [c]Normalized mean bias Σ(HDDM -BF)/ Σ(BFM), comparing coefficients for each day and grid cell. [d]Normalized mean error, Σ|(HDDM - BFM)|/ Σ|(brute force)|.

Fig 6. Spatial distribution of daily maximum 8h ozone sensitivity (ppb) in KPAB on 1-5 October 2010 to ANOx and AVOC from different source regions in KPAB

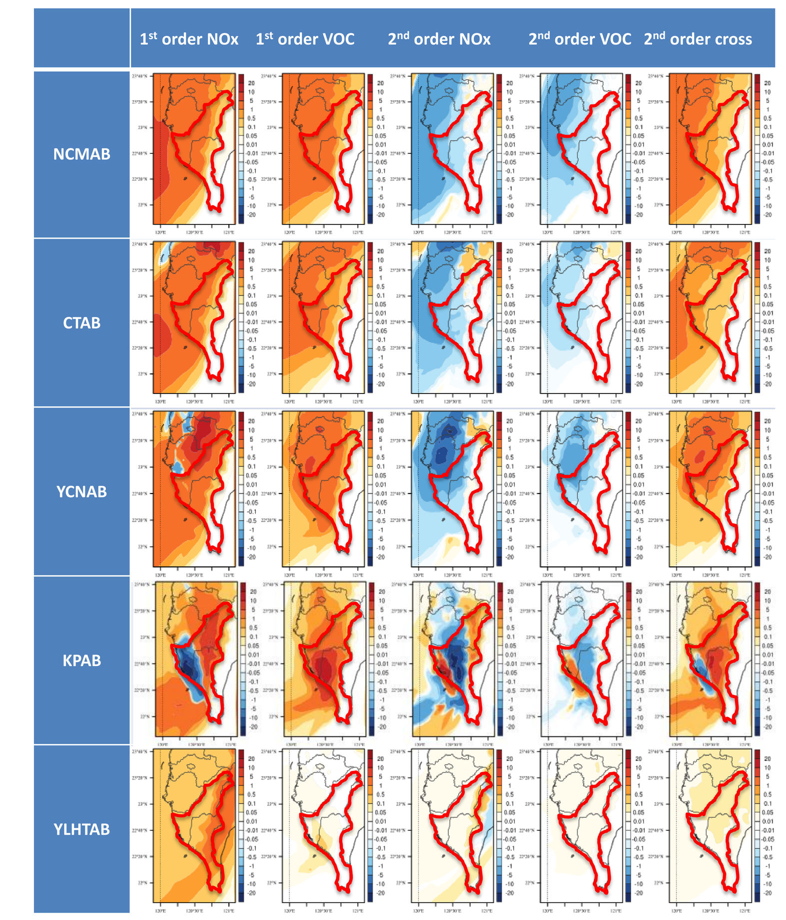


Fig 6. Source contributions of local and upwind ANOx and AVOC emissions for daily maximum 8h ozone concentration in KPAB on 1-5 October 2010.

1st order	NCMAB_NOx	1.1	9%
	NCMAB_VOC	1.2	10%
	CTAB_NOx	1.2	9%
	CTAB_VOC	0.6	5%
	YCNAB_NOx	2.3	18%
	YCNAB_VOC	1.5	12%
	KPAB_NOx	-0.3	-3%
	KPAB_VOC	4.4	35%
	YLHDAB_Nox	0.6	5%
	YLHDAB_VOC	0.0	0%
2nd order (Self)	NCMAB_NOx	0.1	1%
	NCMAB_VOC	0.1	1%
	CTAB_NOx	0.1	1%
	CTAB_VOC	0.0	0%
	YCNAB_NOx	0.5	4%
	YCNAB_VOC	0.1	1%
	KPAB_NOx	1.8	14%
	KPAB_VOC	0.3	2%
	YLHDAB_Nox	0.0	0%
	YLHDAB_VOC	0.0	0%
2nd order (Cross)	NCMABNOxVOC	-0.4	-4%
	CTABNOxVOC	-0.3	-2%
	YCNABNOxVOC	-0.8	-6%
	YLHDABNOxVOC	-0.1	0%
	KPNOxVOC	-2.3	-19%
	IC	0.0	0%
	BC	1.3	11%
	Contribution	12.5	100%
	Simulated ozone	62.4	

Fig 7. Episode average decomposition of the zero-out source contributions from local and upwind ANOx and AVOC emissions to ozone over KPAB

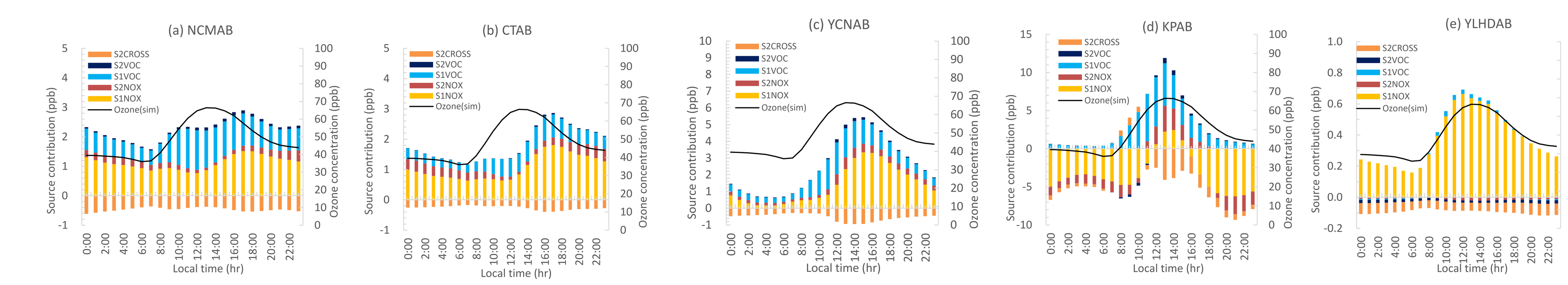
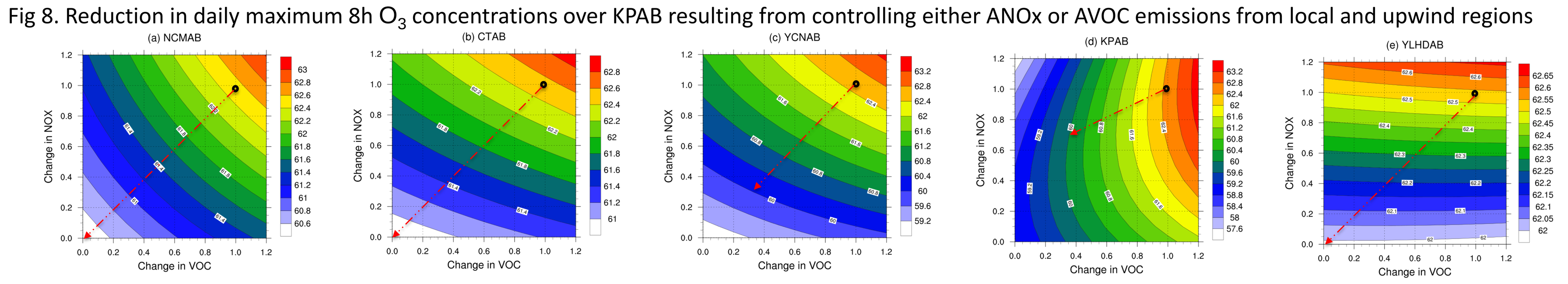


Fig 8. Reduction in daily maximum 8h O_3 concentrations over KPAB resulting from controlling either ANOx or AVOC emissions from local and upwind regions



5. CONCLUSIONS

Upwind anthropogenic emission shows NOx-limited regime over KPAB while local anthropogenic emission forms VOC-limited regime in the core area of KPAB and NOx-limited regime in rural area of KPAB

- First-order response of ozone to upwind ANOx and AVOC emissions is entirely positive (NOx-limited) over KPAB while that to local ANOx emission is typically positive in rural area where ozone concentrations are highest but is negative (VOC-limited) in the core area of KPAB.
- Self-sensitivity of ozone typically exhibits strongly concave response to local ANOx and AVOC emissions, indicating far more nonlinear ozone formation from KPAB than from other upwind regions.
- The cross-sensitivity of upwind and local ANOx and AVOC emission exhibits overall negative in the core area of KPAB, reflecting that as ANOx (AVOC) emissions are reduced, ozone becomes less sensitive to AVOC (ANOx).

Whereas local anthropogenic emission dominate ozone contribution, ozone contribution from upwind anthropogenic emissions combined is significant.

- Anthropogenic emissions from NCMAB, CTAB, YCNAB, KPAB, YLHDAB, IC and BC contribute 16%, 13%, 28%, 29%, 4%, 0% and 10% of daily maximum 8h ozone concentration, respectively.
- Daytime ozone response to local emissions evolves diurnal variations, mostly positive sensitivities (NOx-limited) but negative sensitivities (VOC-limited) during the night while positive first-order response of ozone to upwind ANOx shows entirely NOx-limited ozone formation.
- Four sensitivity coefficients stand out for their contributions of significant magnitude, that is, $S^{(1)}_{KPAB}$ VOC, $S^{(1)}_{YCNAB}$VOC, $S^{(1)}_{YCNAB}$NOx, $S^{(2)}_{KPAB}$NOx, $S^{(2)}_{KPAB}$NOxVOC. $S^{(1)}_{KPAB}$ VOC was the largest contributor to ozone over KPAB.
- Considering that the KPAB region requires roughly 2.4 ppb of additional ozone reduction to attain the daily maximum 8h ozone standard, this ozone reduction could be achieved for these episodes either by a 55% reduction from local AVOC emissions, >60% reduction from YCNAB emissions or >100% reduction from NCMAB or CTAB emissions. The combination of mitigation policies is also possible, where the cross-sensitivity coefficients would play a role in interactions between the impacts.

ACKNOWLEDGEMENTS: The research has been funded by a scholarship from the Ministry of Education, Taiwan.