

Table A1. Geographic information (latitude, longitude, elevation above sea level) for CASTNet sites used in our analysis. Sites determined to be regionally-representative are indicated in **bold**.

Region	Site ID	Station Name	State	Latitude (° N)	Longitude (° W)	Elevation (m ASL)
Northwest	NCS	North Cascades NP	WA	48.5	121.4	109
	OLY	Olympic NP	WA	48.1	123.4	125
	MOR	Mount Rainier NP	WA	46.8	122.1	421
	GLR	Glacier NP	MT	48.5	114.0	976
California	LAV	Lassen Volcanic NP	CA	40.5	121.6	1756
	DEV	Death Valley NM	CA	36.5	116.8	125
	PIN	Pinnacles NM	CA	36.5	121.2	335
	JOT	Joshua Tree NM	CA	34.1	116.4	1244
Mountain West	YOS	Yosemite NP - Turtleback Dome	CA	37.7	119.7	1605
	CNT	Centennial	WY	41.4	106.2	3178
	GTH	Gothic	CO	39.0	107.0	2926
	GRB	Great Basin NP	NV	39.0	114.2	2060
	MEV	Mesa Verde NP	CO	37.2	108.5	2165
	PND	Pinedale	WY	42.9	109.8	2388
	ROM	Rocky Mountain NP	CO	40.3	105.5	2743
	YEL	Yellowstone NP	WY	44.6	110.4	2469
	GRC	Grand Canyon NP	AZ	36.1	112.2	2073
	CAN	Canyonlands NP	UT	38.5	109.8	1814
Plains	CHA	Chiricahua NM	AZ	32.0	109.4	1570
	CAD	Caddo Valley	AR	34.2	93.1	71
	STK	Stockton	IL	42.3	90.0	274
	VOY	Voyageurs NP	MN	48.4	92.8	427
	ALH	Alhambra	IL	38.9	89.6	164
	BVL	Bondville	IL	40.1	88.4	212
	PRK	Perkinsztown	WI	45.2	90.6	472
Great Lakes	VIN	Vincennes	IN	38.7	87.5	134
	ANA	Ann Arbor	MI	42.4	83.9	267
	HOX	Hoxeyville	MI	44.2	85.7	305
	UVL	Unionville	MI	43.6	83.4	201
	SAL	Salamonie Reservoir	IN	40.8	85.7	250
	DCP	Deer Creek	OH	39.6	83.3	267
	LYK	Lykens	OH	40.9	83.0	303
	OXF	Oxford	OH	39.5	84.7	284
	QAK	Quaker City	OH	39.9	81.3	372
	KEF	Kane Exp. Forest	PA	41.6	78.8	622
	LRL	Laurel Hill	PA	40.0	79.3	615
	MKG	M.K. Goddard	PA	41.4	80.1	384
	CDR	Cedar Creek	WV	38.9	80.8	234
	PAR	Parsons	WV	39.1	79.7	510
Northeast	WSP	Washington Crossing	NJ	40.3	74.9	61
	ARE	Arendtsville	PA	39.9	77.3	269
	PSU	Penn State	PA	40.7	77.9	378
	CTH	Connecticut Hill	NY	42.4	76.7	501
	ABT	Abington	CT	41.8	72.0	209
Far Northeast	WST	Woodstock	NH	43.9	71.7	258
	ACA	Acadia NP	ME	44.4	68.3	152
	ASH	Ashland	ME	46.6	68.4	235
	HOW	Howland	ME	45.2	68.7	69
Southeast	BEL	Beltsville	MD	39.0	76.8	46
	BWR	Blackwater NWR	MD	38.4	76.1	4
	VPI	Horton Station	VA	37.3	80.6	920
	PED	Prince Edward	VA	37.2	78.3	150
	SHN	Shenandoah NP - Big Meadows	VA	38.5	78.4	1073
	CDZ	Cadiz	KY	36.8	87.8	189
	CKT	Crockett	KY	37.9	83.1	455
	MCK	Mackville	KY	37.7	85.0	353
	ESP	Edgar Evins	TN	36.0	85.7	302
	GRS	Great Smoky NP - Look Rock	TN	35.6	83.9	793
	SPD	Speedwell	TN	36.5	83.8	361
	COW	Coweeta	NC	35.1	83.4	686
	PNF	Cranberry	NC	36.1	82.0	1219
	BFT	Beaufort	NC	34.9	76.6	2
	CND	Candor	NC	35.3	79.8	198
Florida / Gulf	SND	Sand Mountain	AL	34.3	86.0	352
	CVL	Coffeeville	MS	34.0	89.8	134
	GAS	Georgia Station	GA	33.2	84.4	270
	EVE	Everglades NP	FL	25.4	80.7	2
	SUM	Sumatra	FL	30.1	85.0	14

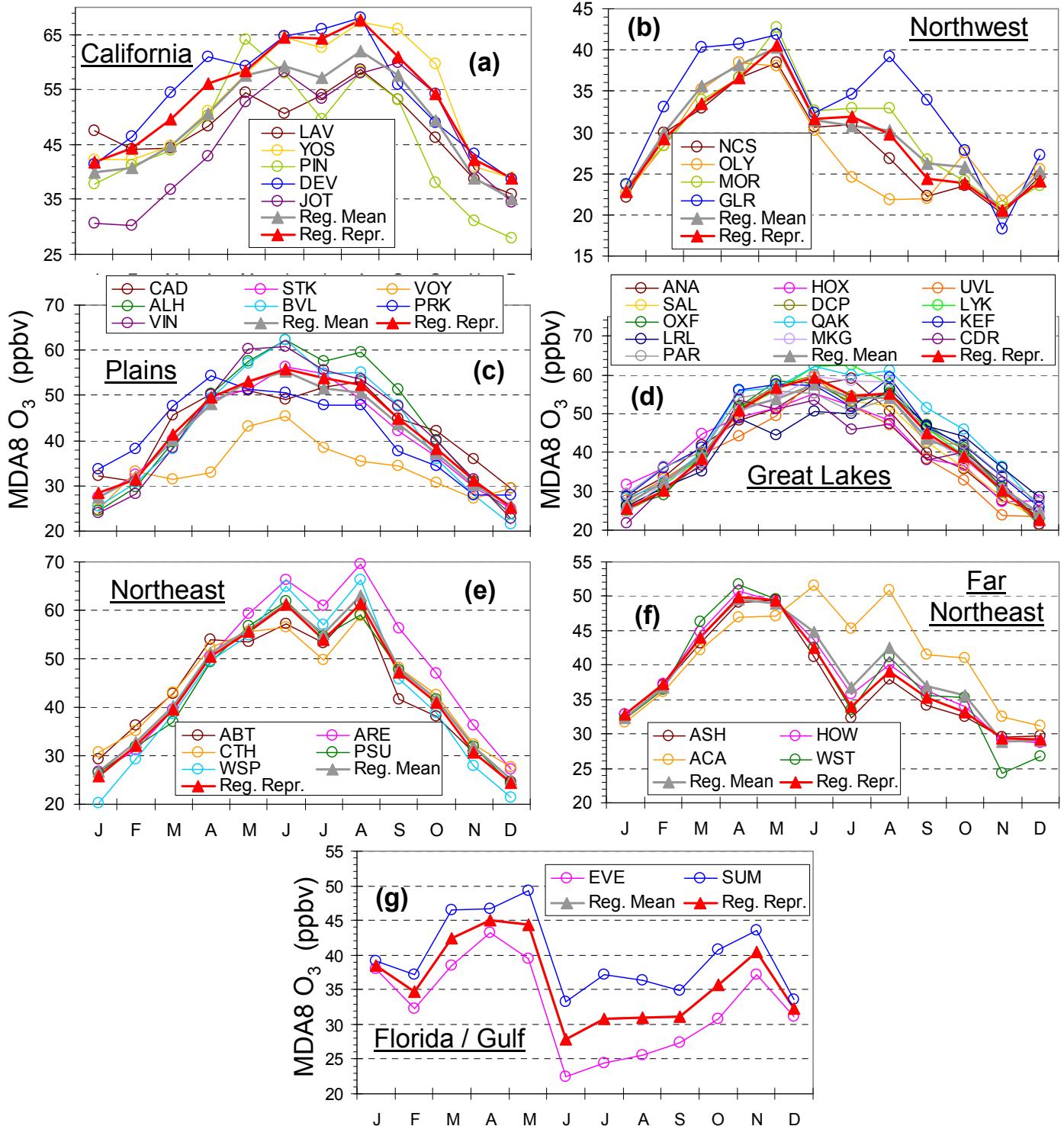


Figure A1. As in Fig. 2, but for the other seven regions. The regionally-representative sites (mean shown as solid red triangles) are: Northwest - MOR, NCS; California - DEV, YOS; Plains - BVL, CAD, STK; Great Lakes - DCP, MKG, OXF, SAL; Northeast - CTH, PSU, WSP; Far Northeast - ASH, HOW; Florida / Gulf - EVE, SUM. Note the range of magnitudes on the y-axes.

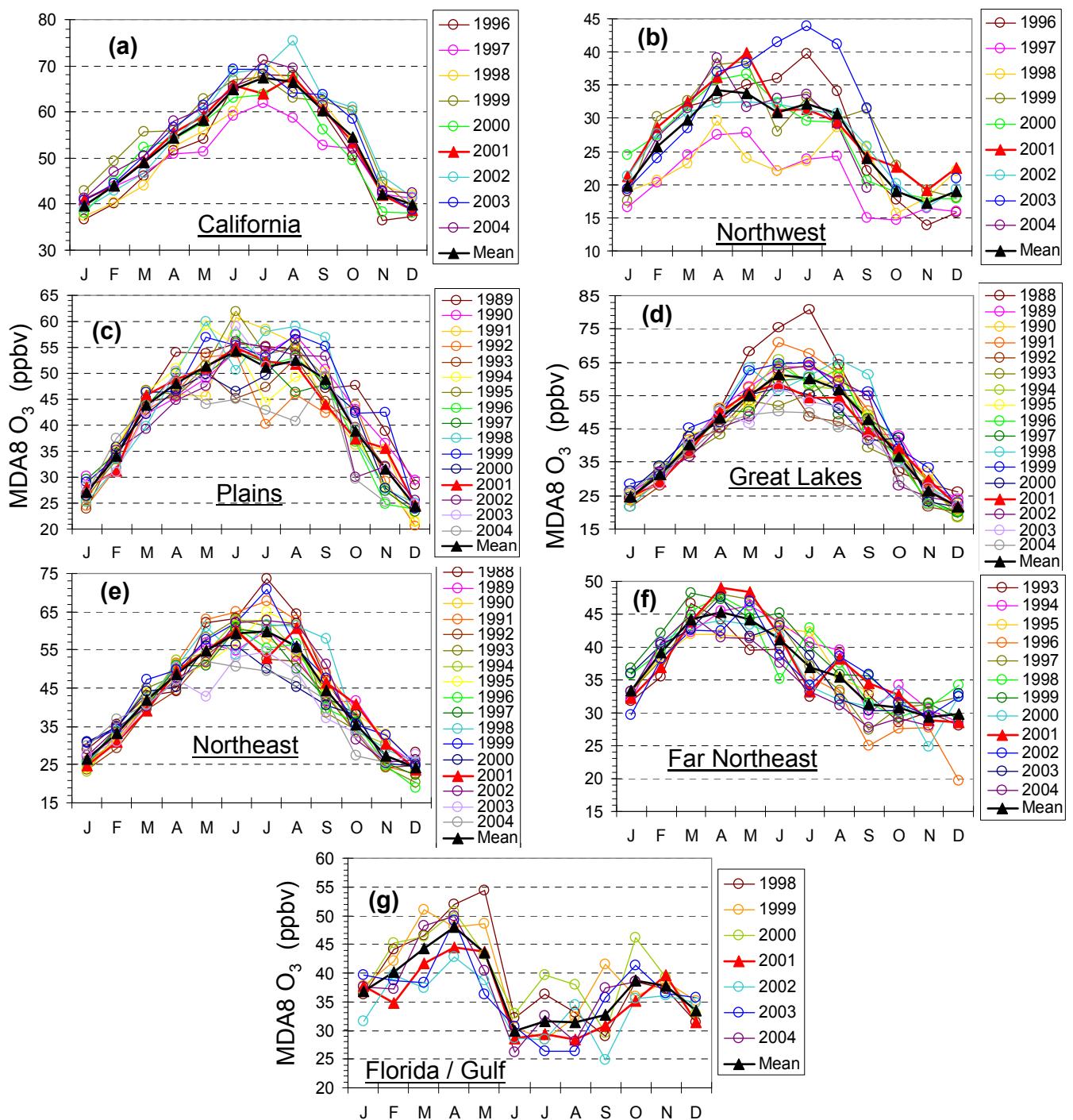


Figure A2. As in Fig. 3, but for the other seven regions.

Table A2. Details of the global models used in our analysis, including emissions inventories and meteorological fields employed for the HTAP simulations. Further description of model characteristics is available at: <http://www.mi.uni-hamburg.de/List-classification-and-detail-view-of-model-entr.567.0.html>. Adapted from Fiore *et al.* [2009].

Model	Resolution (lon x lat x layers)	Institution	Model contact	Anthropogenic emission inventory for O ₃ precursors	Driving meteorology (year 2001)
CAMCHEM-3311m13 [#]	2.5° x 2° x 30	NCAR, USA	Peter Hess	POET for 1997 ^a ; CO fossil fuel and biofuel from a MOPITT inversion ^b	NCEP
ECHAM5-HAMMOZ-v21	2.81° x 2.81° x 21	EPFL, Switzerland	Gerd Folberth	RETRO	ECMWF ERA-40
EMEP-rv26 [#] (NH only)	1° x 1° x 20	EMEP, Norway	Jan Eiof Jonson, Peter Wind	IER / Uni-Stuttgart, based on EDGAR2000; EMEP	ECMWF ERA-40
FRSGC/UCI v01 [#]	2.81° x 2.81° x 37	Lancaster University, UK	Oliver Wild	ACCENT/AR4 ^c	ECMWF IFS ^d
GEMAQ-EC	2° x 2° x 20	Environment Canada	Sunling Gong	AURAMS (regional Canadian, US and Mexico); EDGAR elsewhere	Canadian Meteorological Centre (CMC)
GEMAQ-v1p0 [#]	4° x 4° x 28	York University, Canada	Alexandru Lupu	EDGAR v2	CMC
GEOS-Chem v07 [#]	2.5° x 2° x 30	Harvard University, USA	Rokjin Park	Bey et al., 2001 ^e	NASA GEOS-4
GEOS-Chem v07-res4x5	5° x 4° x 30	CIEMAT, Spain	Marta Garcia Vivanco	EMEP emissions EPA/NEI99 inventory	NASA GEOS-4
GISS-PUCCNI-modeIE [#]	5° x 4° x 23	NASA GISS, USA	Drew Shindell	ACCENT / AR4, with EA emissions of CO and NO _x times 1.66	NCEP, via linear relaxation
GMI-v02f [#]	2.5° x 2° x 42	NASA GSFC, USA	Bryan Duncan	Harvard's merged inventory (NEI99, BRAVO, Streets, EMEP)	NASA GEOS-4
LMDz3-INCA1 [#]	3.75° x 2.5° x 19	LSCE, France	Sophie Szopa	RETRO	ECMWF ERA-40
LLNL-IMPACT-T5a [#]	2.5° x 2° x 48	LLNL, USA	Cynthia Atherton, Daniel Bergmann	POET	NASA GEOS-4- ceres
MOZART-GFDL-v2 [#]	1.88° x 1.88° x 28	GFDL, USA	Arlene Fiore	EDGAR v2	NCEP
MOZECH-v16 [#]	2.81 x 2.81 x 31	FZ Jülich, Germany	Martin Schultz, Sabine Schröder	RETRO	ECMWF ERA-40
Oslo CTM2	2.81° x 2.81° x 40	University of Oslo, Norway	Michael Gauss	EDGAR v3.2	ECMWF - IFS
TM5-JRC-cy2-ipcc-v1 [#]	1° x 1° x 25	JRC, Italy	Frank Dentener, Elina Marmer	ACCENT / AR4	ECMWF

[#] - Model used in NO_x+CO+VOC+aerosol emission perturbation simulations (all 16 used for "base-case" comparisons)

^a - Granier, C. et al. (2004), Present and future surface emissions of atmospheric compounds, European Commission report EVK 2199900011. (Available at <http://www.aero.jussieu.fr/projet/ACCENT/POET.php>)

^b - Pétron, G., C. Granier, B. Khattatov, V. Yudin, J. Lamarque, L. Emmons, J. Gille, and D. P. Edwards (2004), Monthly CO surface sources inventory based on the 2000 - 2001 MOPITT satellite data, *Geophys. Res. Lett.*, 31, L21107, doi:10.1029/2004GL020560.

^c - Bond, T.C. et al. (2004), A technology-based global inventory of black and organic carbon emissions from combustion, *J. Geophys. Res.*, 109, D14203, doi:10.1029/2003JD003697.

^d - van der Werf, G.R., J.T. Randerson, L. Giglio, G.J. Collatz, P.S. Kasibhatla, and A.F. Arellano, Jr. (2006), Interannual variability in global biomass burning emissions from 1997 to 2004, *Atmos. Chem. Physics*, 6, 3423-3441.

^e - based on EDGAR v3.2, see Stevenson *et al.* [2006]

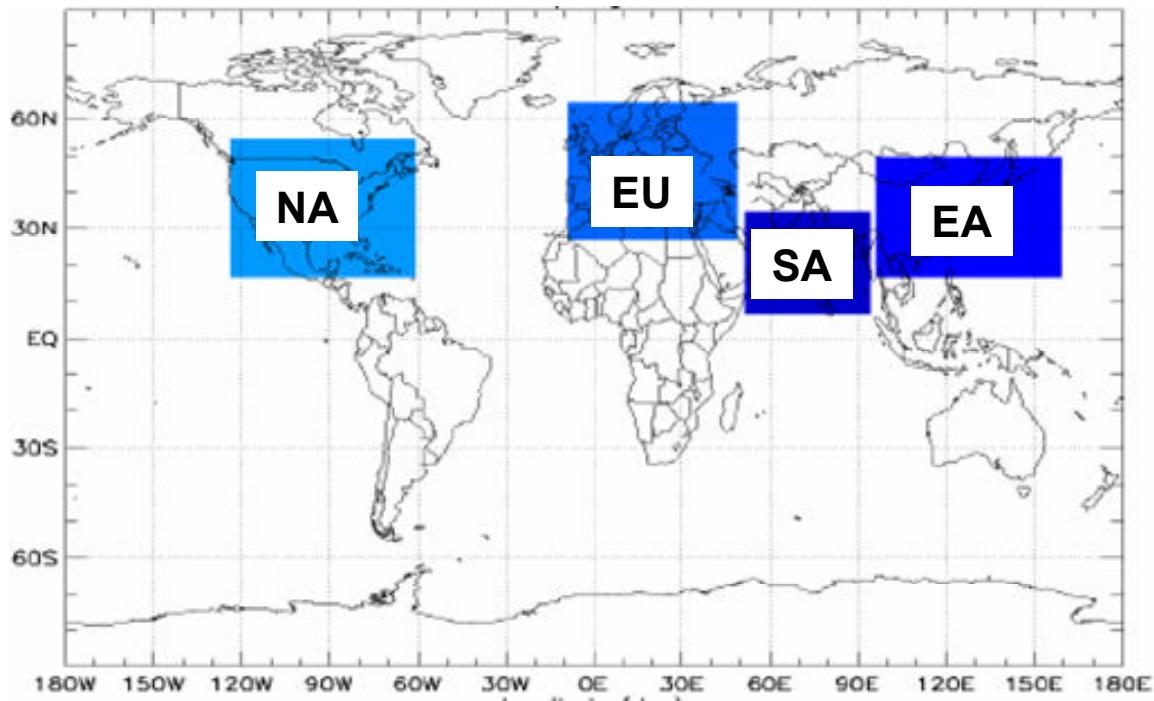


Figure A3. Colored boxes represent the four source regions used in the HTAP emissions reductions simulations: NA - North America, EU - Europe, SA - South Asia, EA - East Asia [adapted from *TF HTAP*, 2007].

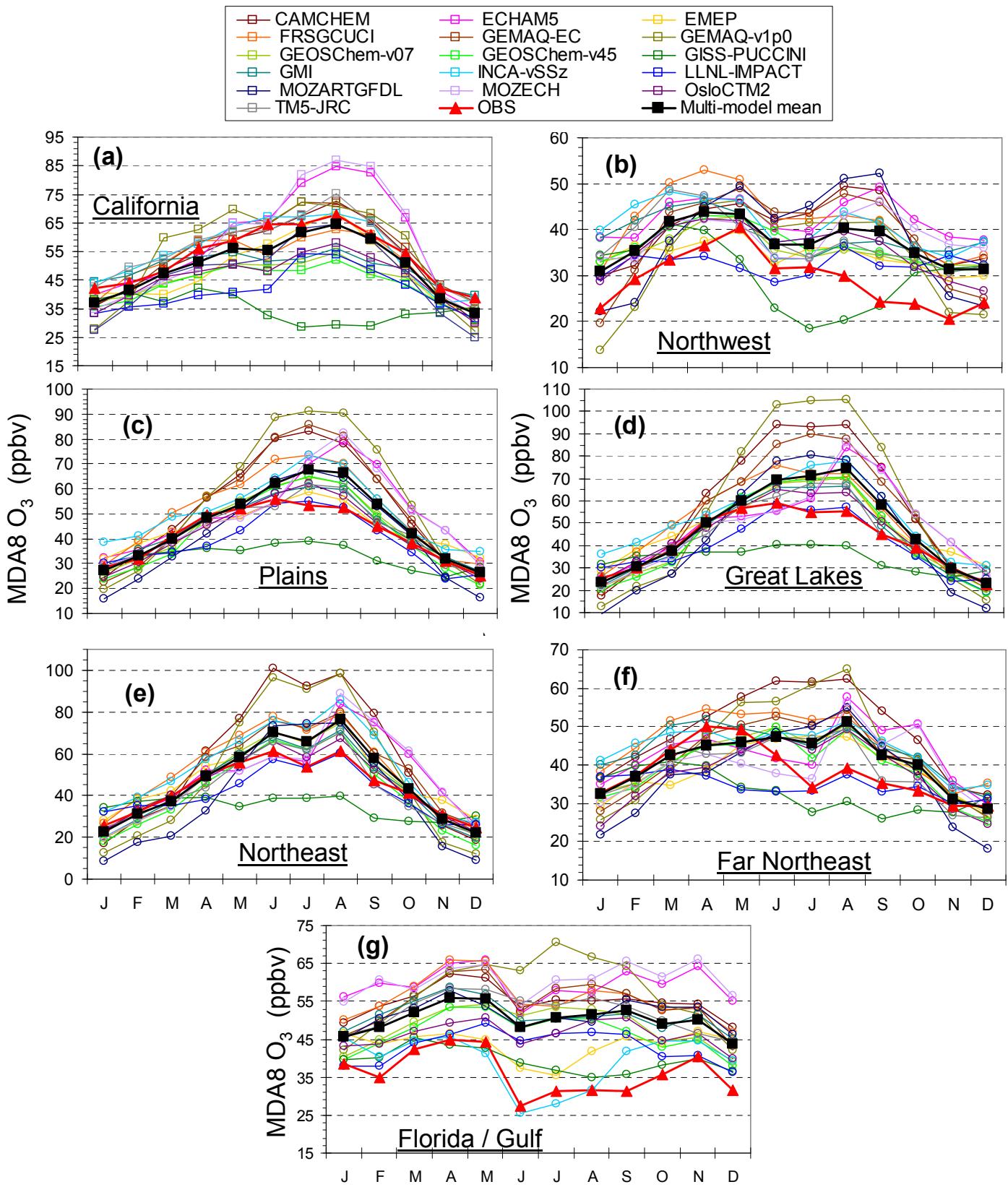


Figure A4. As in Fig. 4, but for the other seven regions.

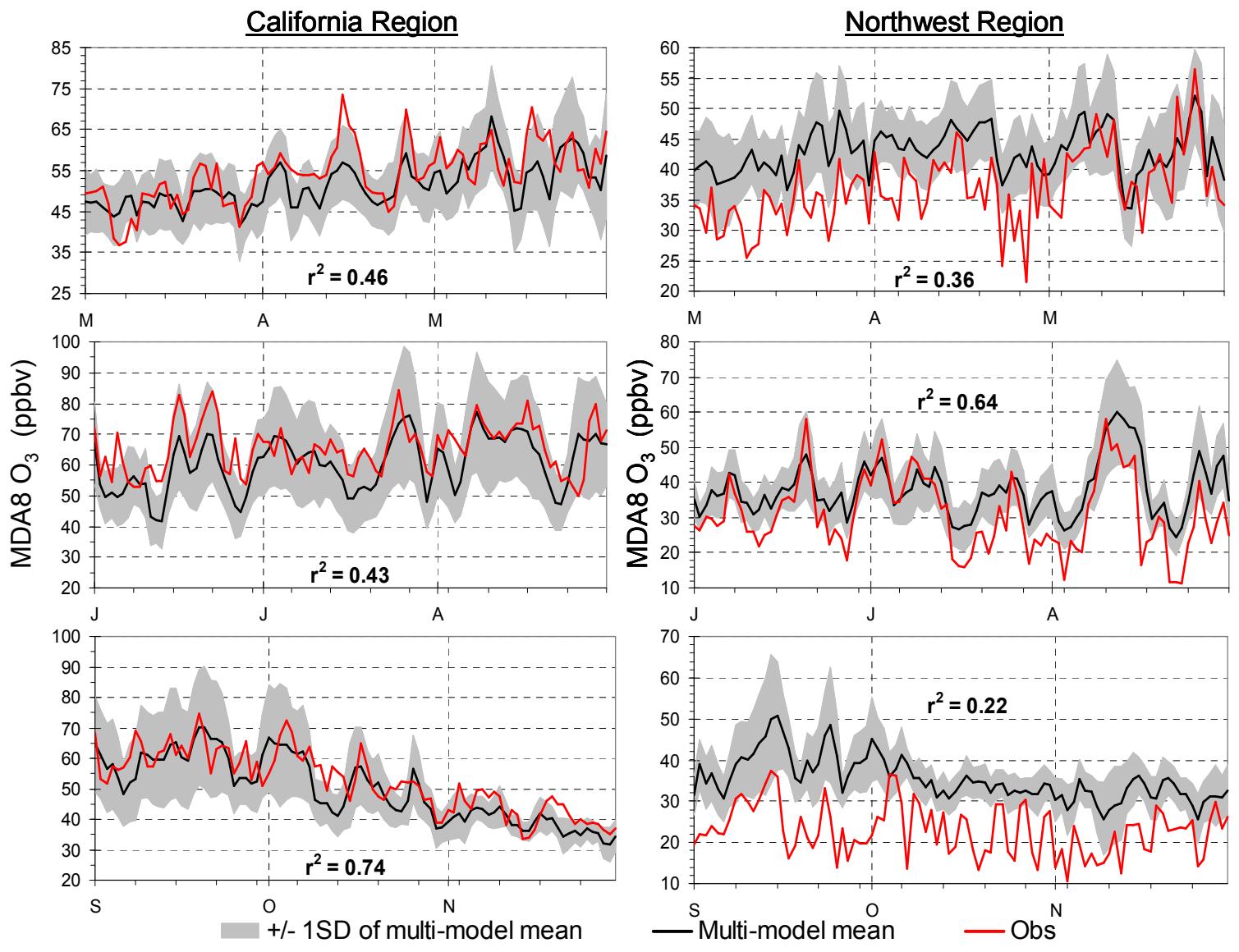


Figure A5. As in Fig. 5, but for the other seven regions.

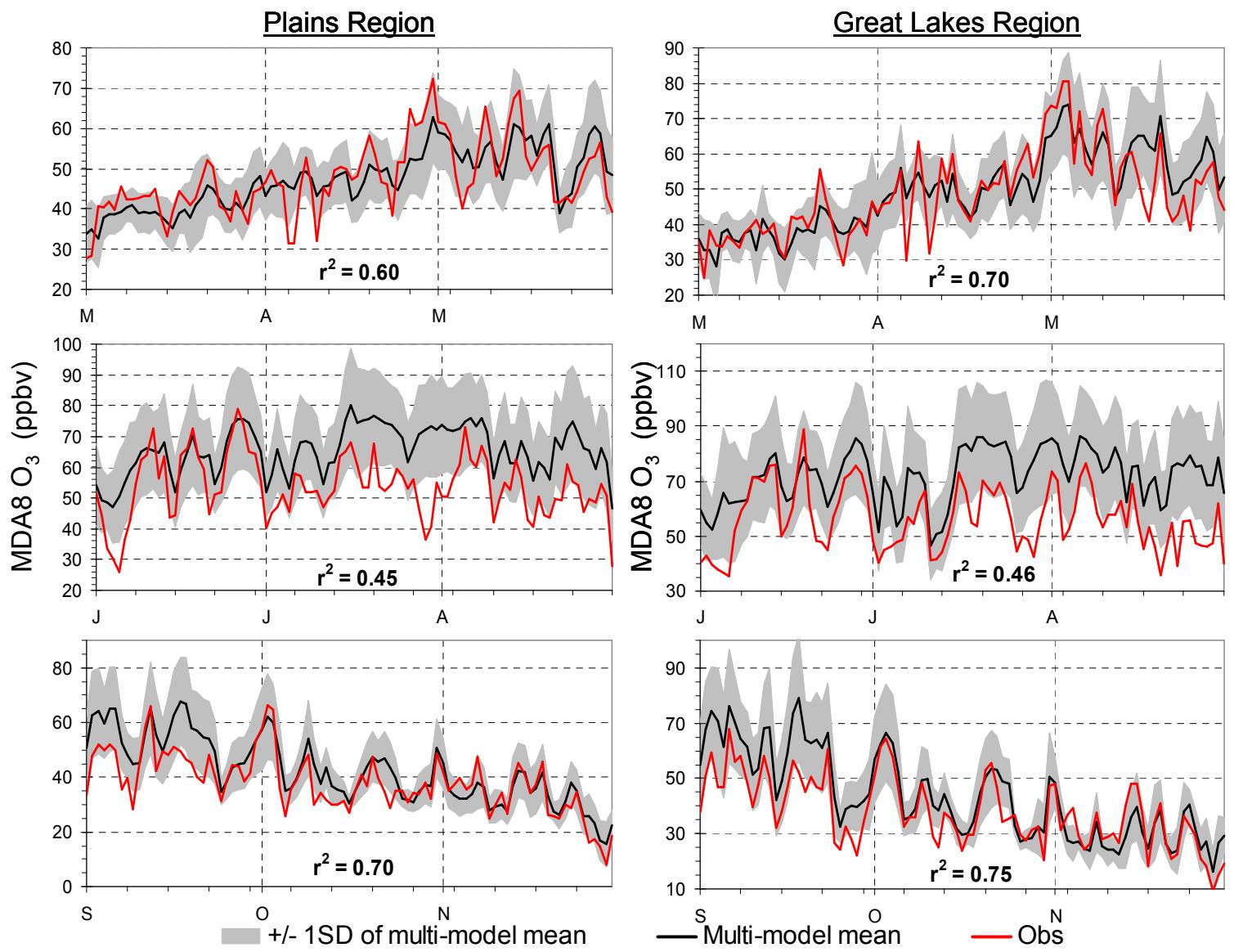


Figure A5. (cont'd)

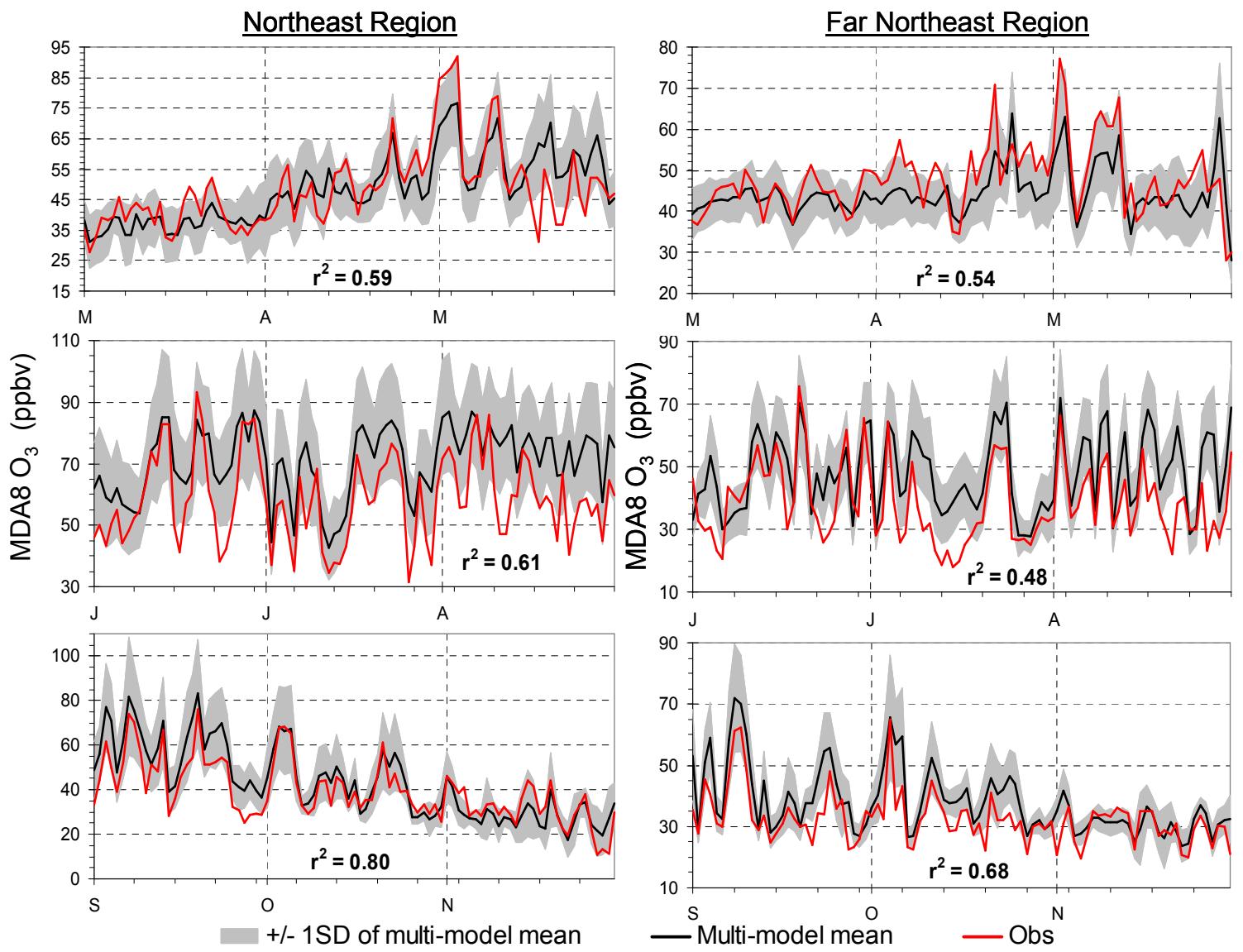


Figure A5. (cont'd)

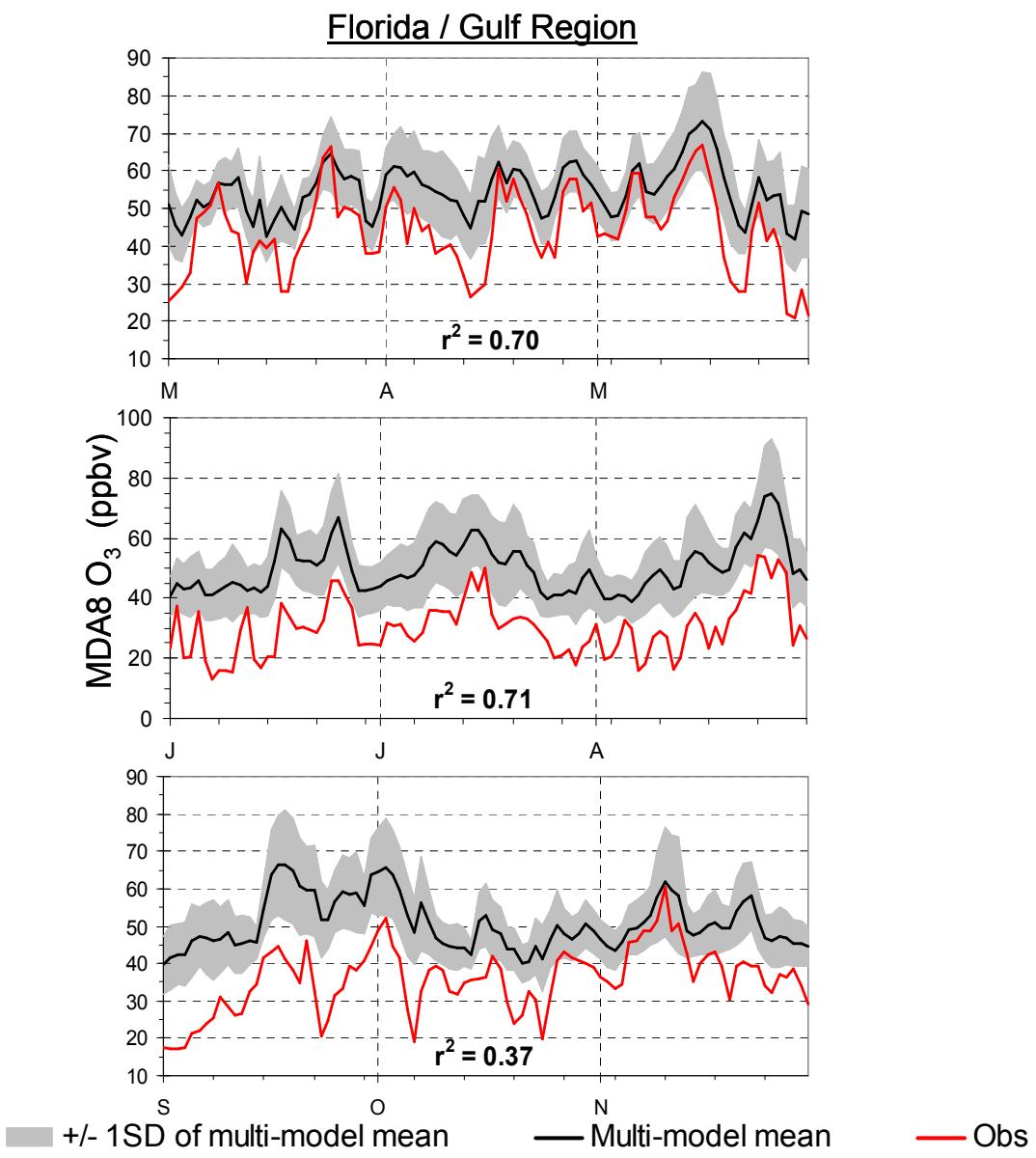


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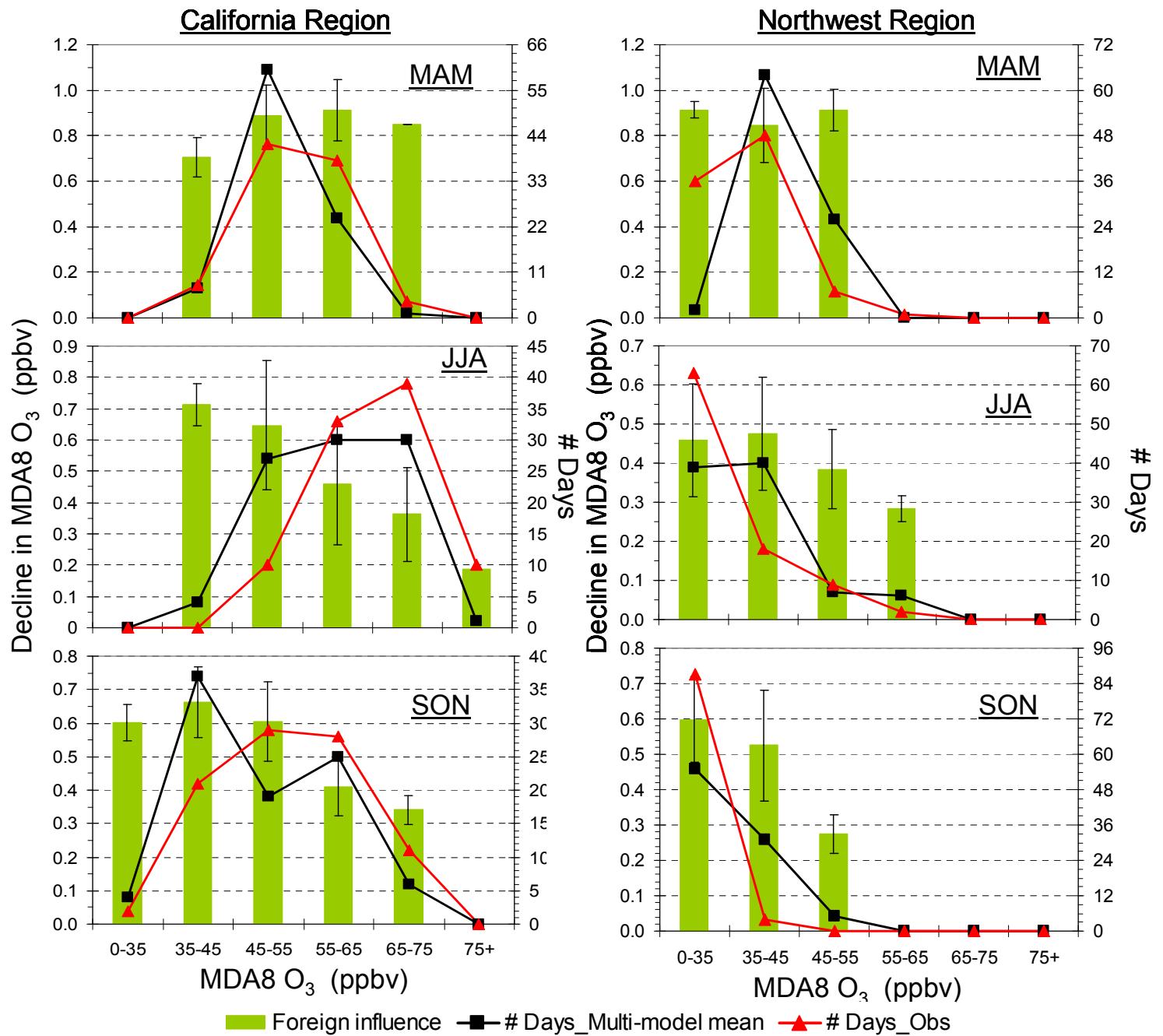


Figure A6. As in Fig. 6, but for the other seven regions.

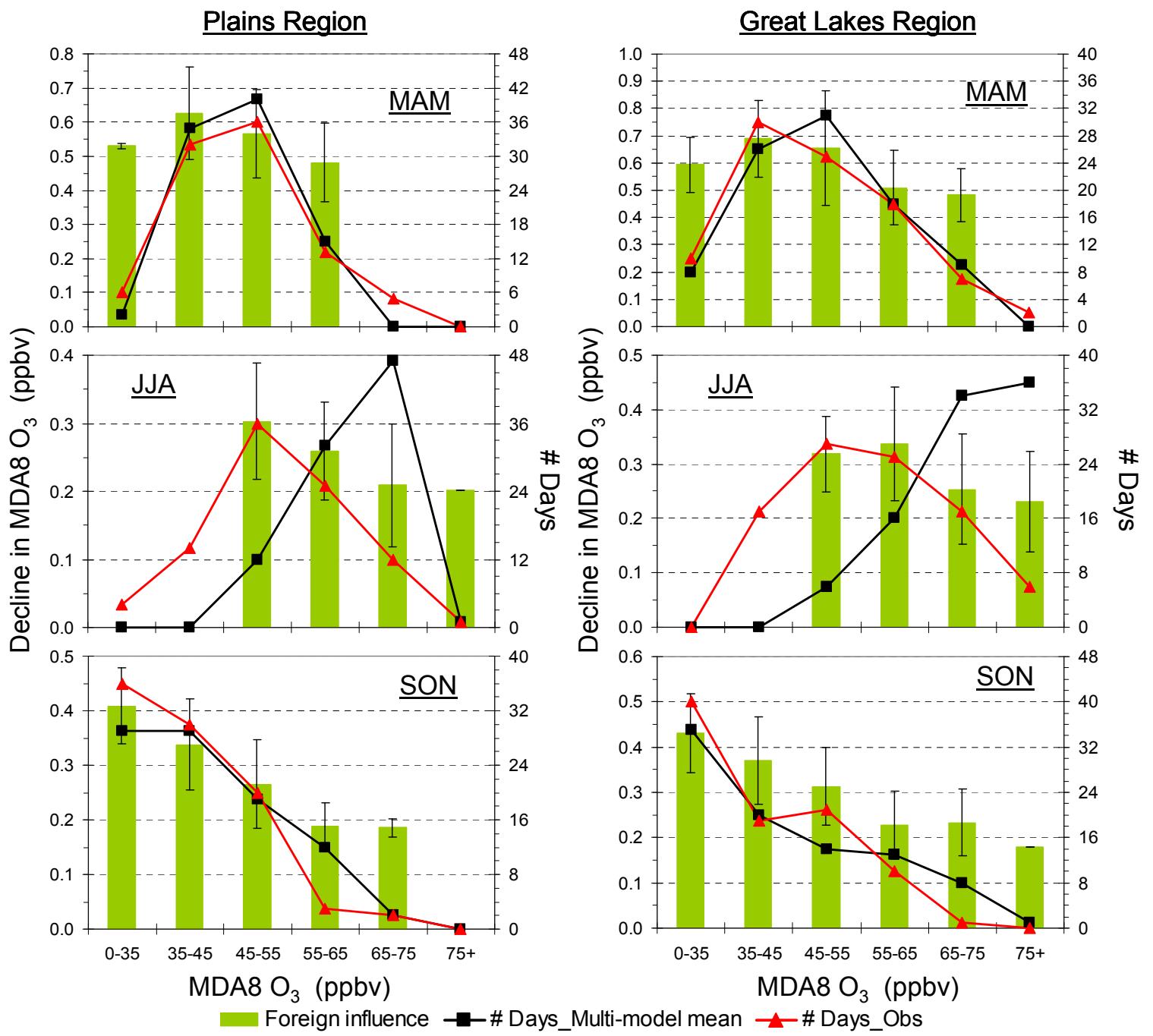


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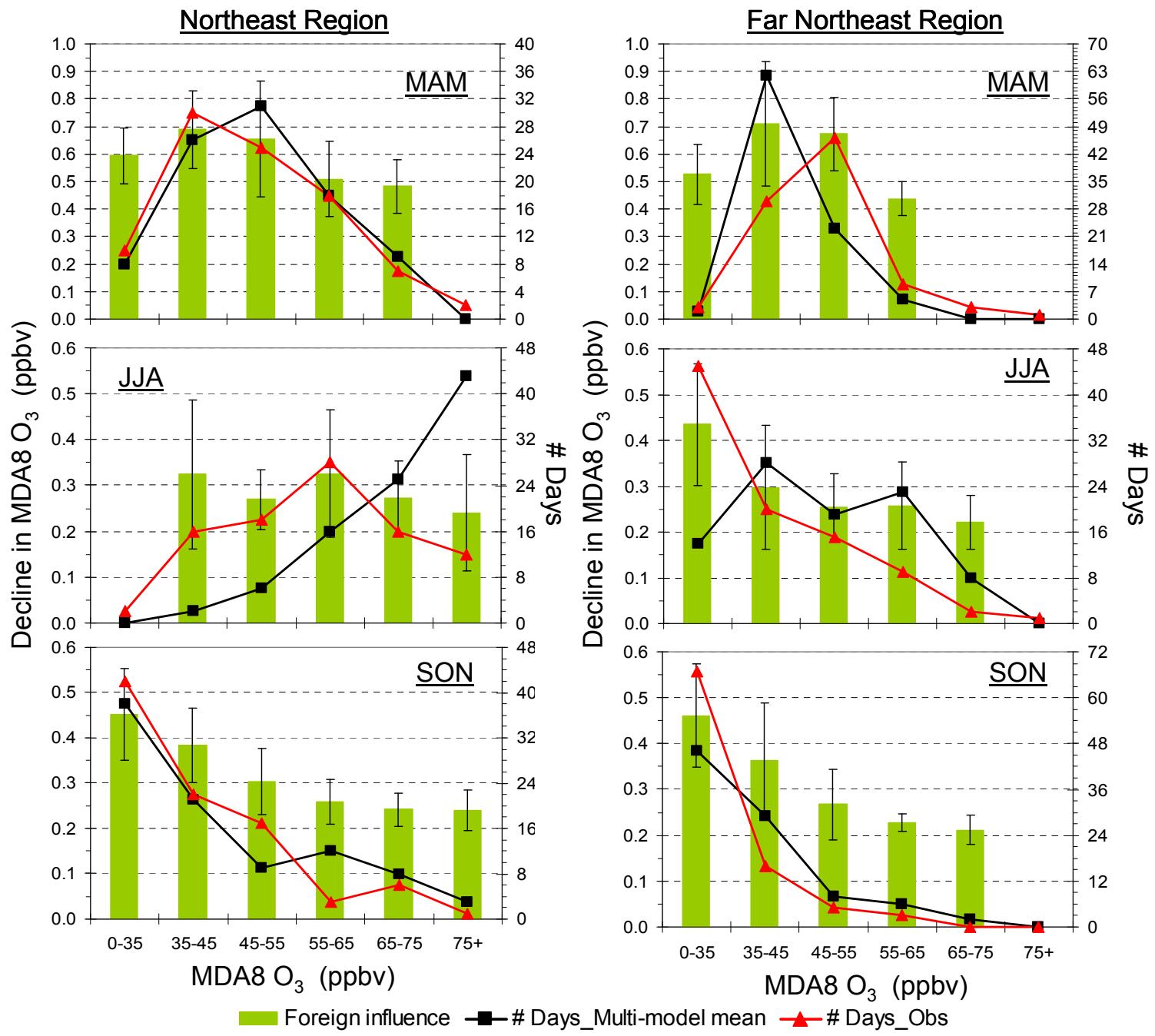


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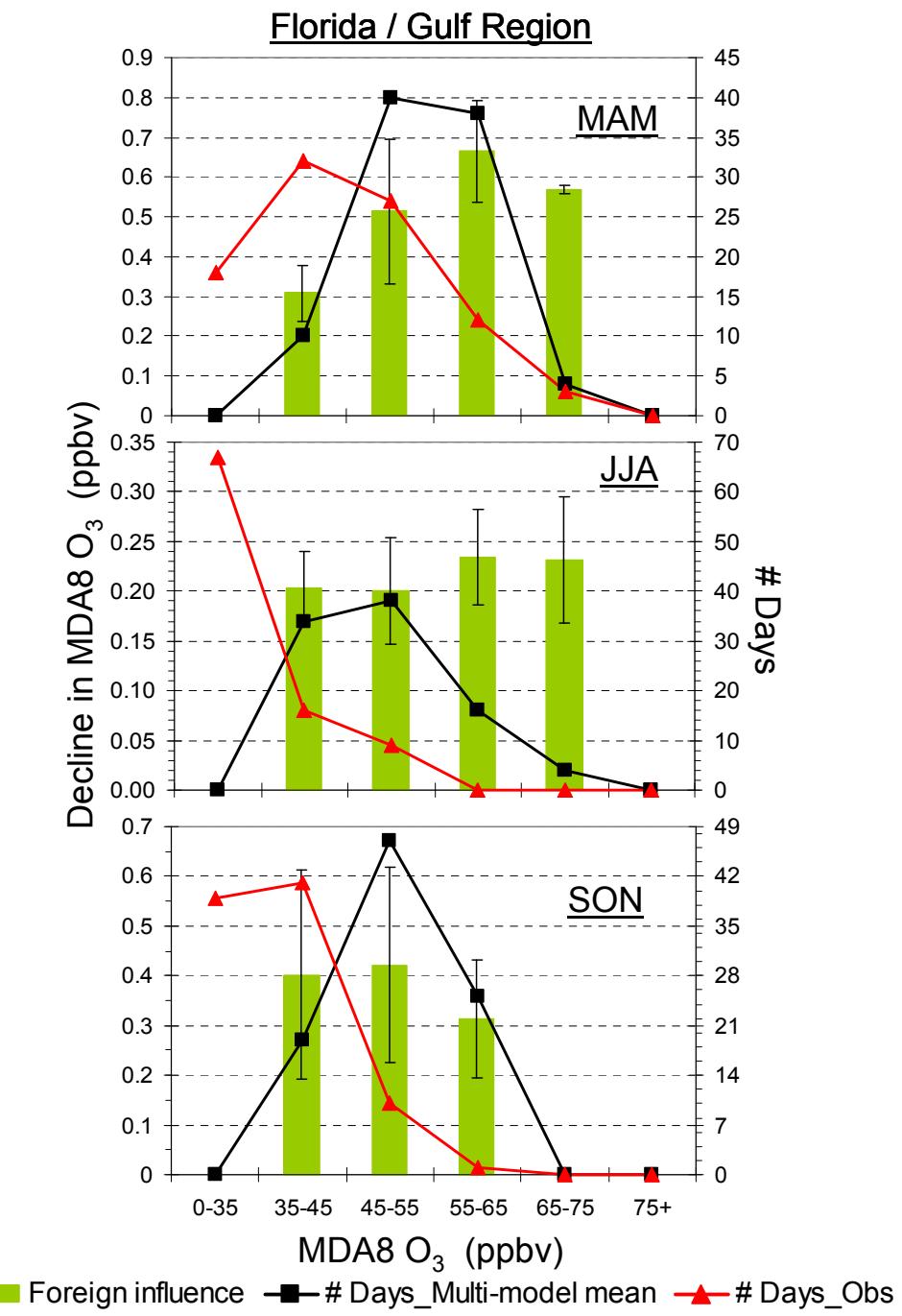


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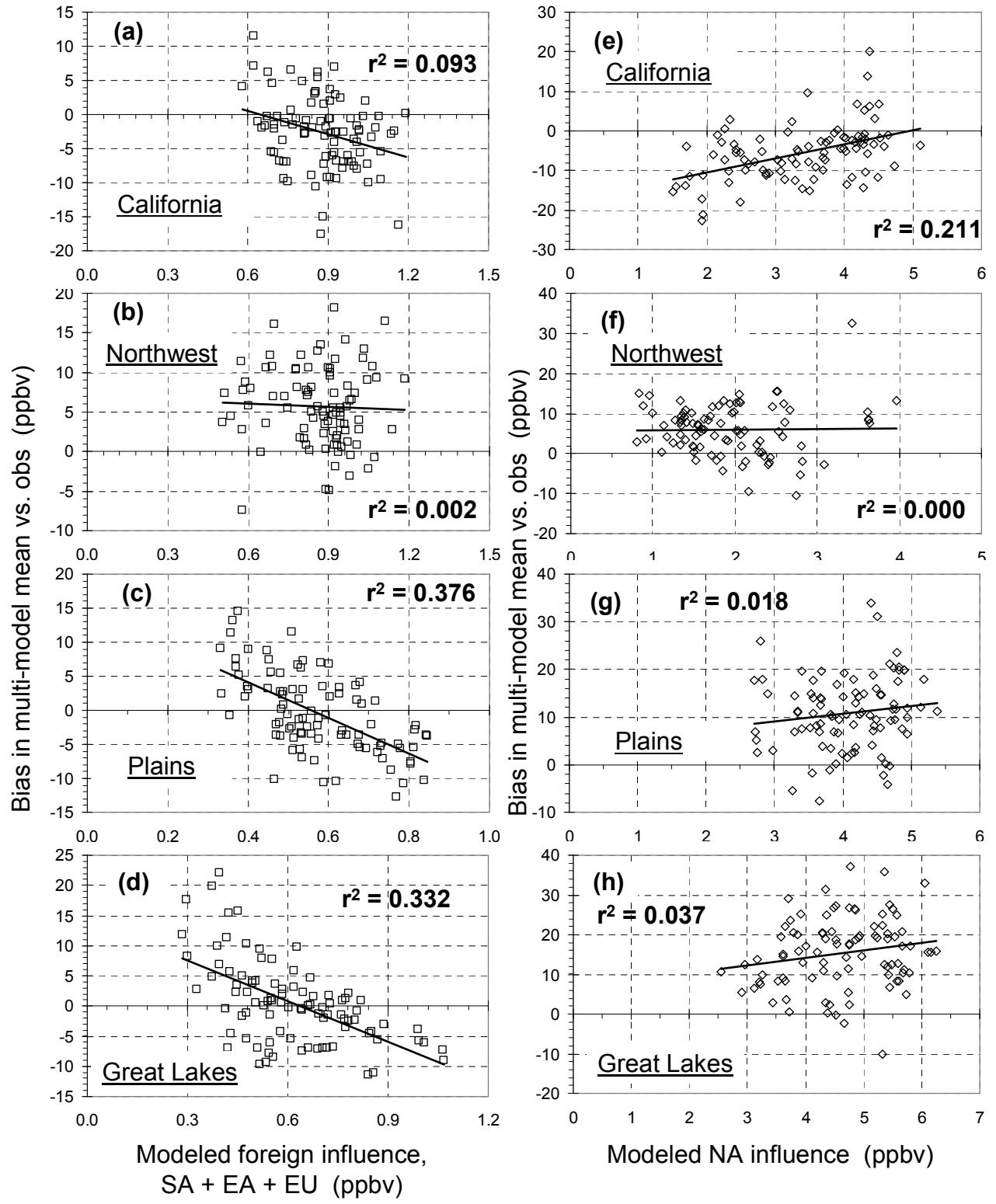


Figure A7. As in Fig. 7, but for the other seven regions.

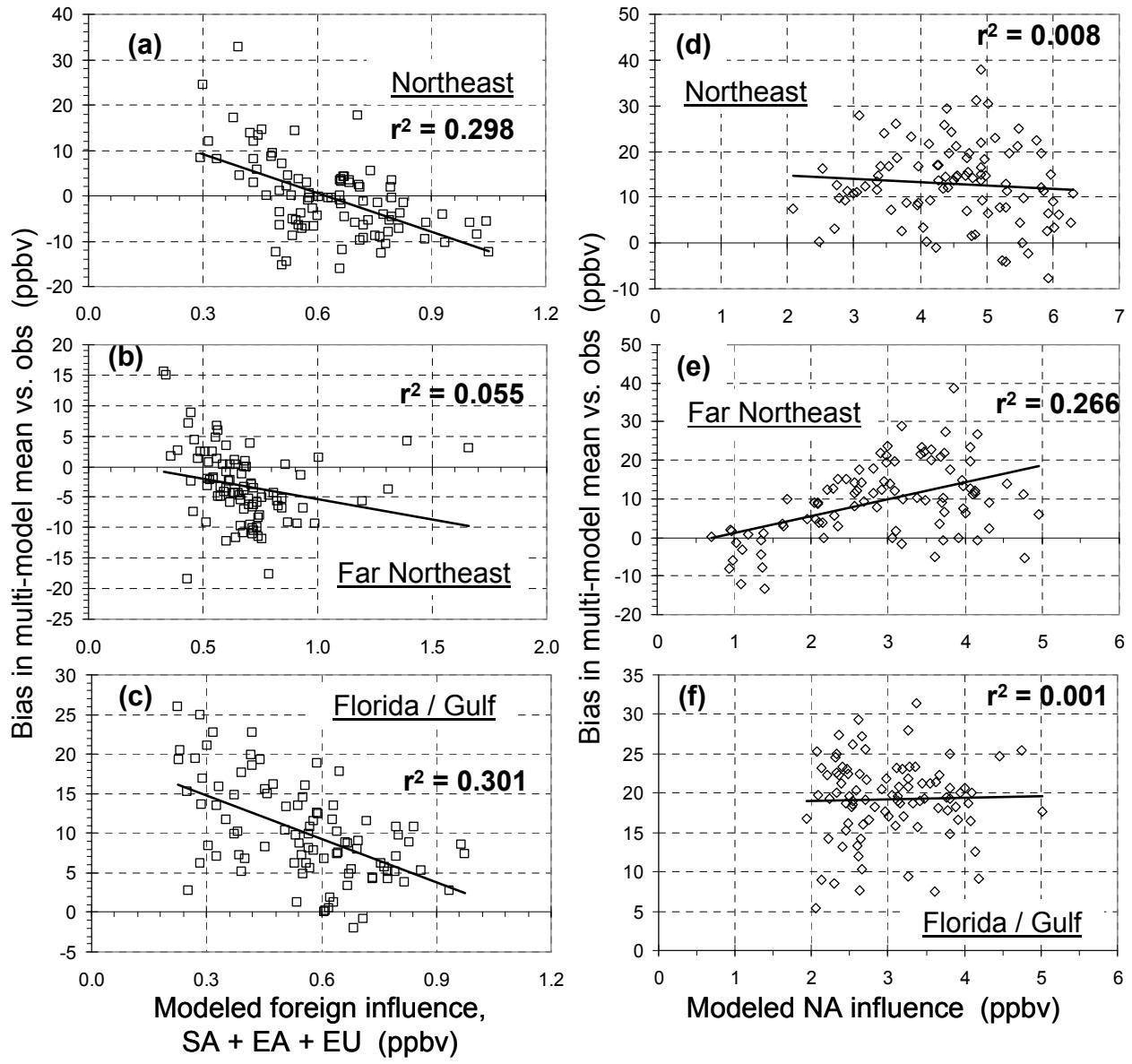


Figure A7. (cont'd)

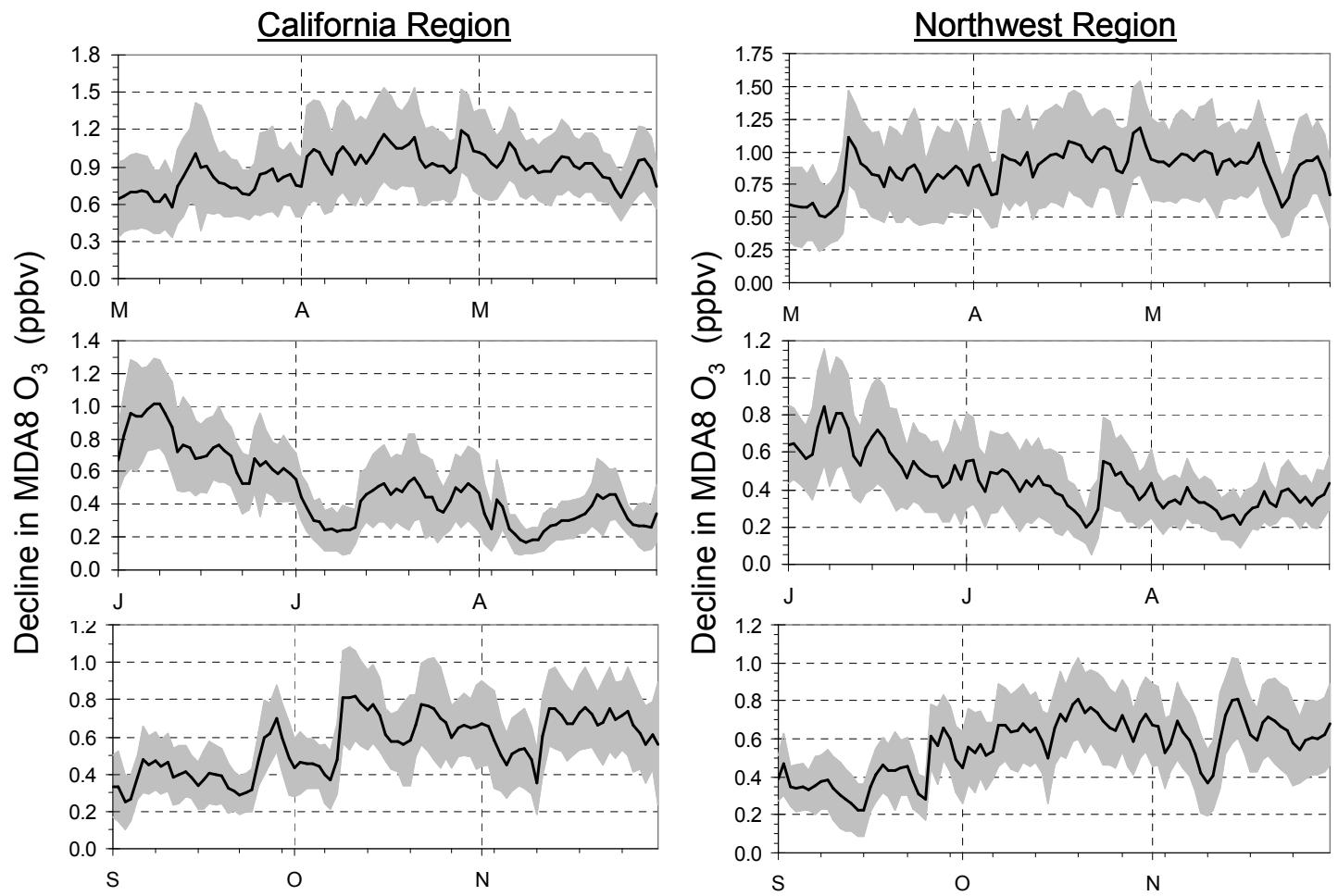


Figure A8. As in Fig. 8, but for the other seven regions.

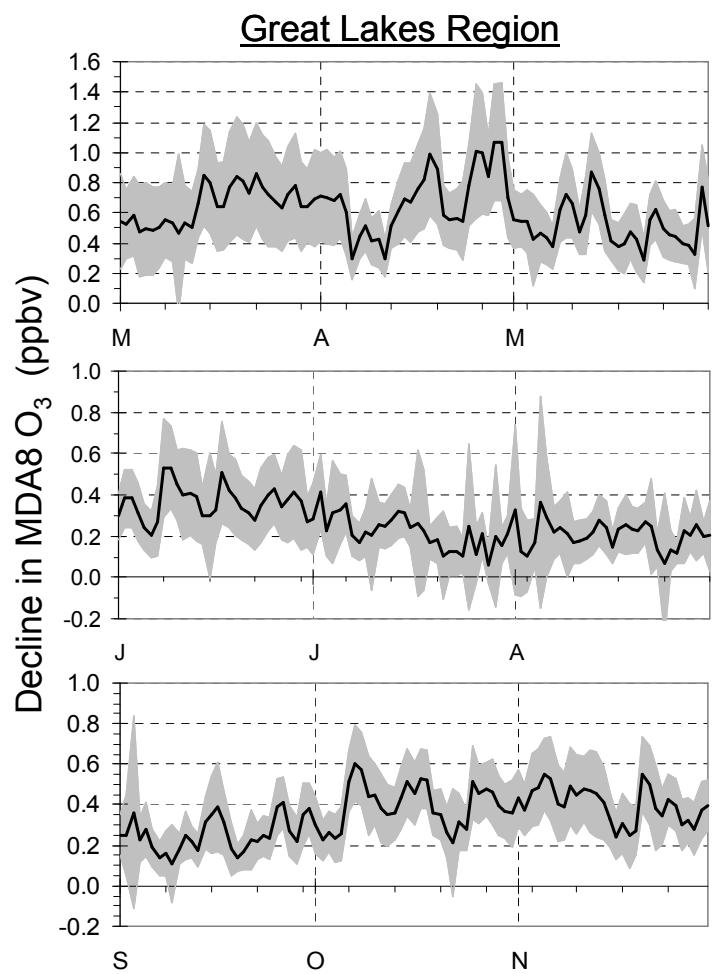
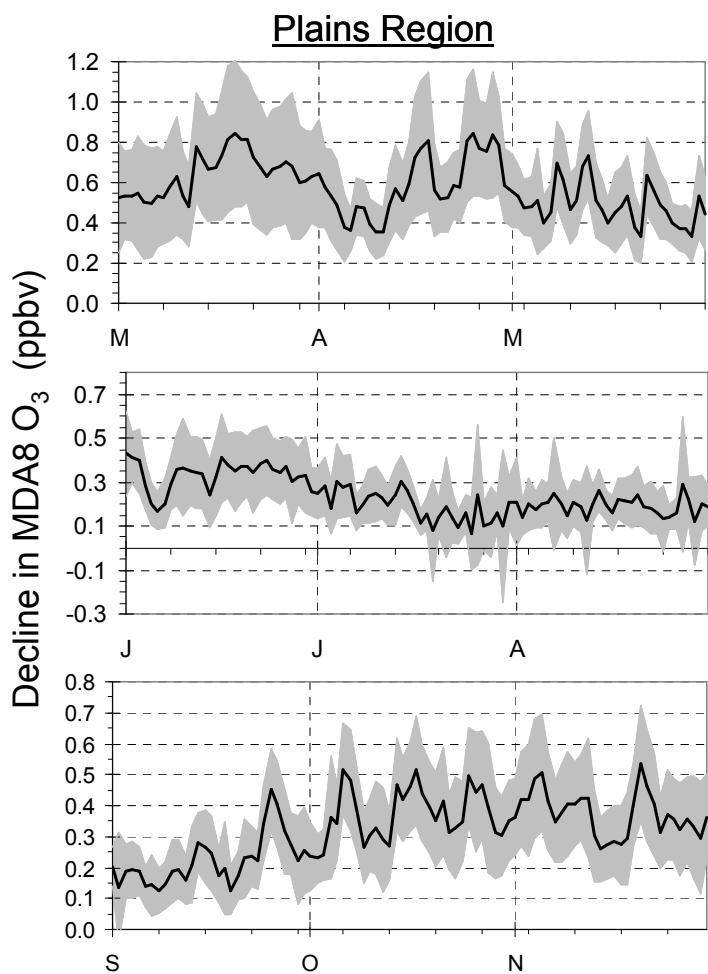


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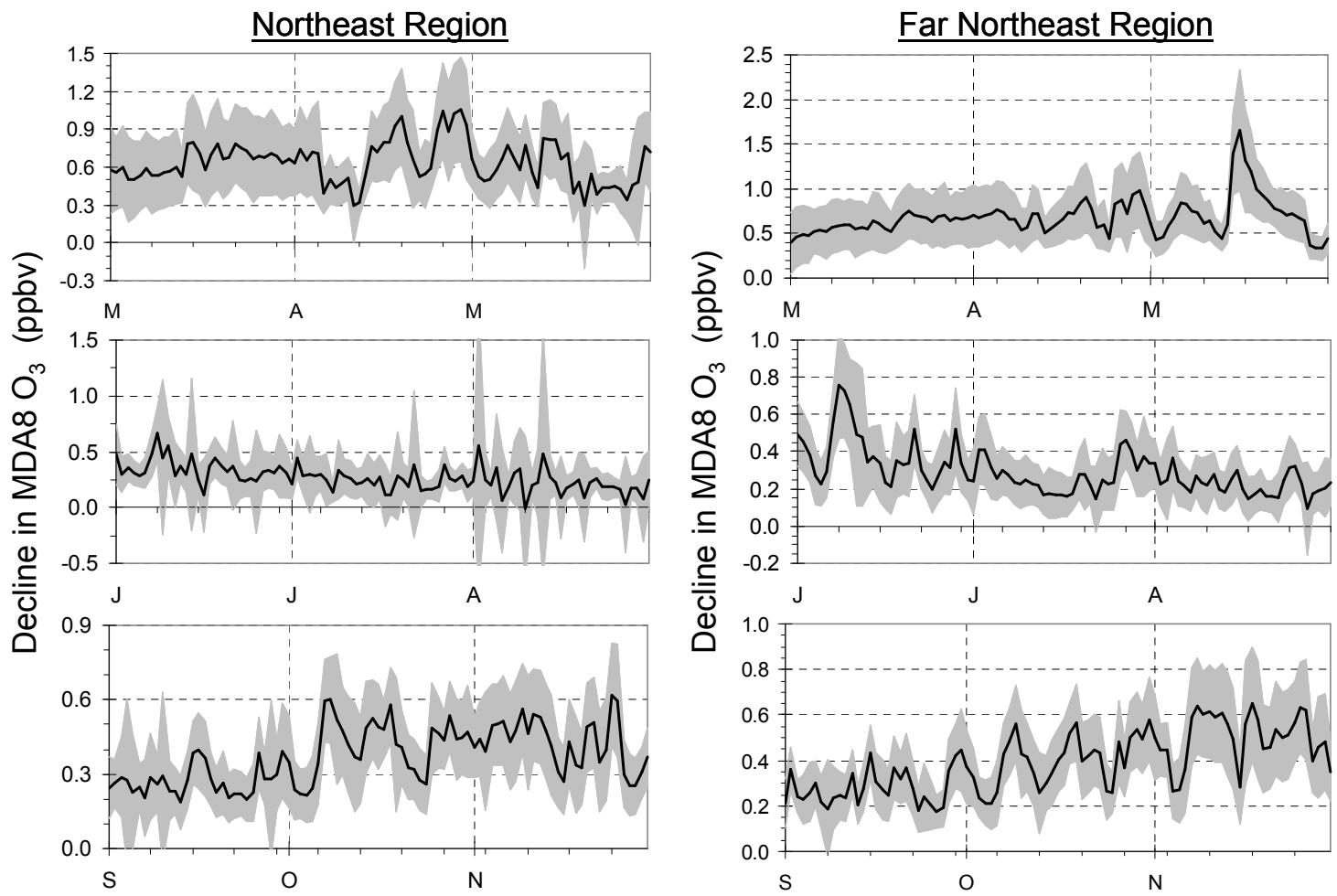


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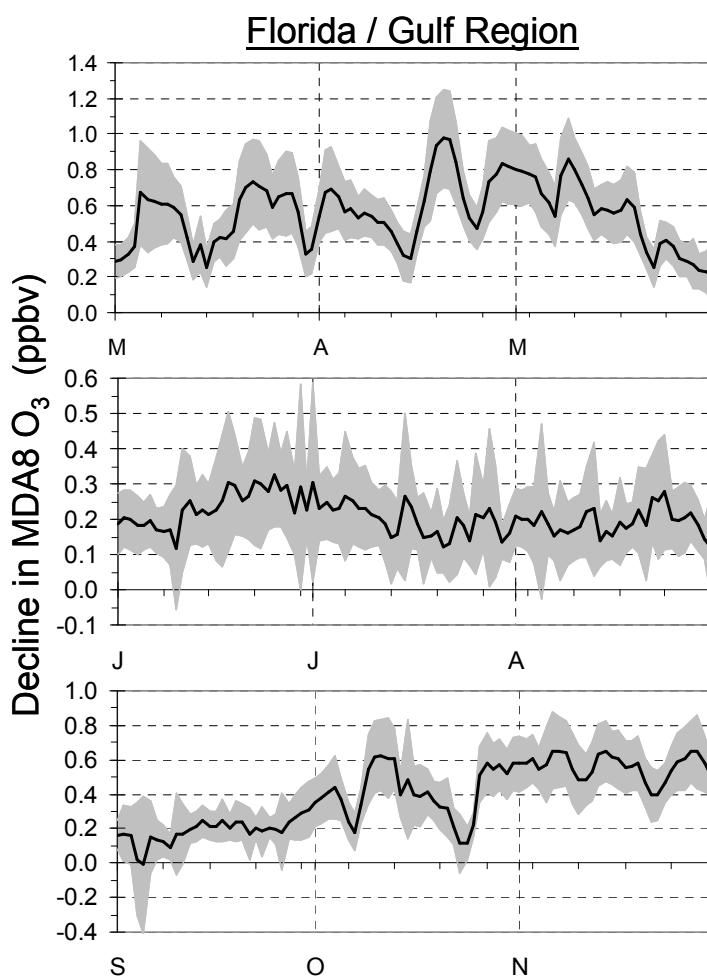


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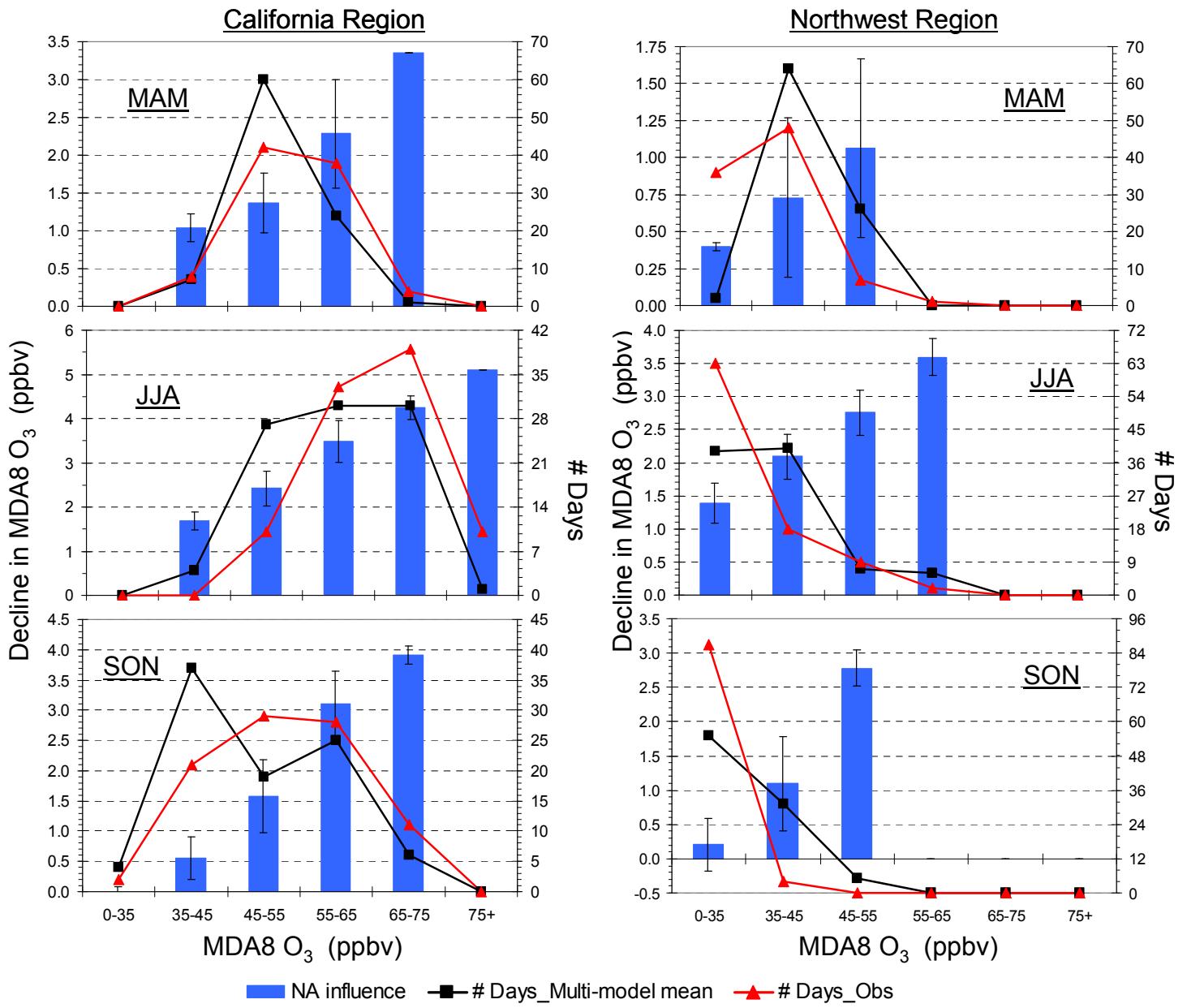


Figure A9. As in Fig. 10, but for the other seven regions.

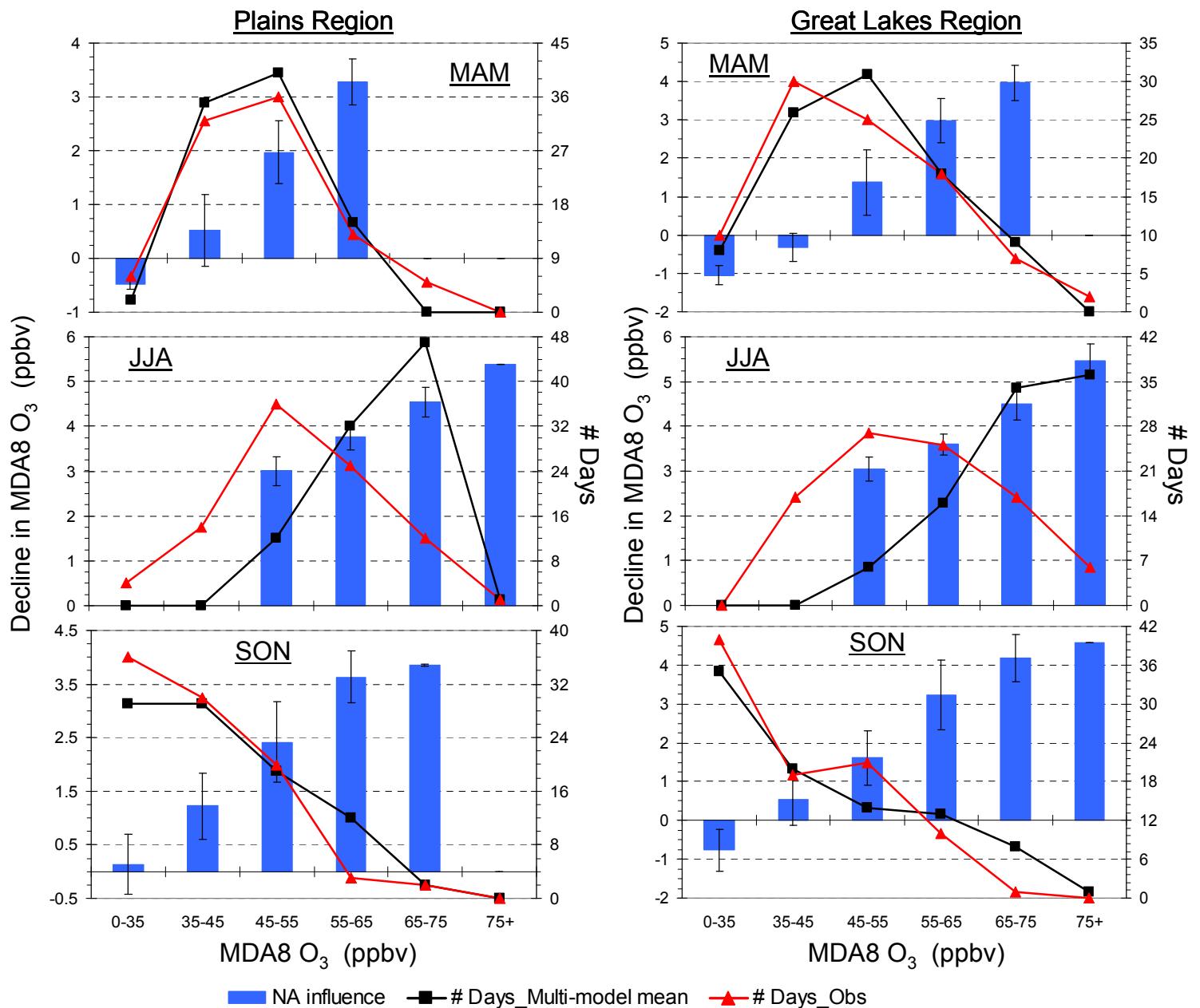


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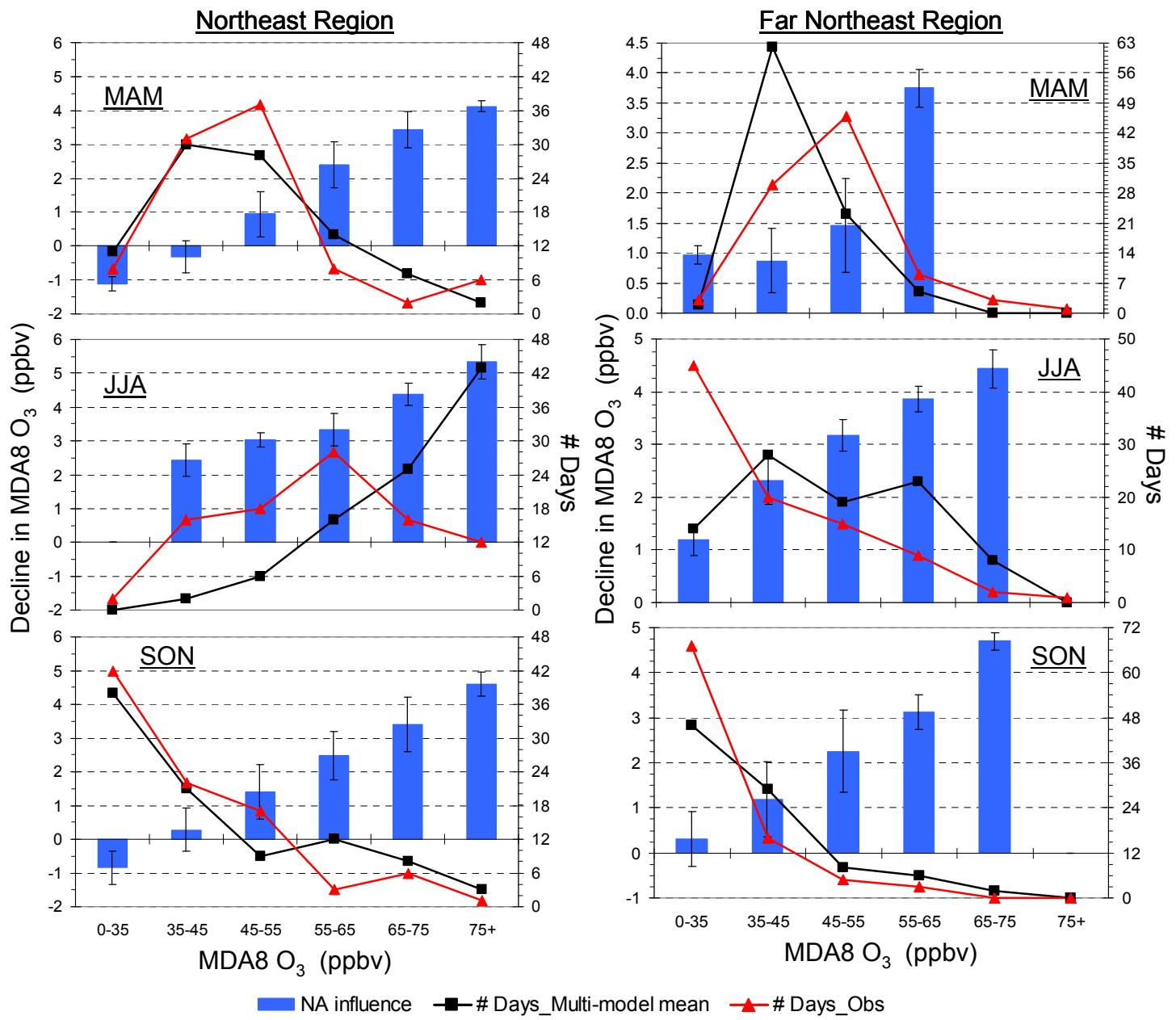


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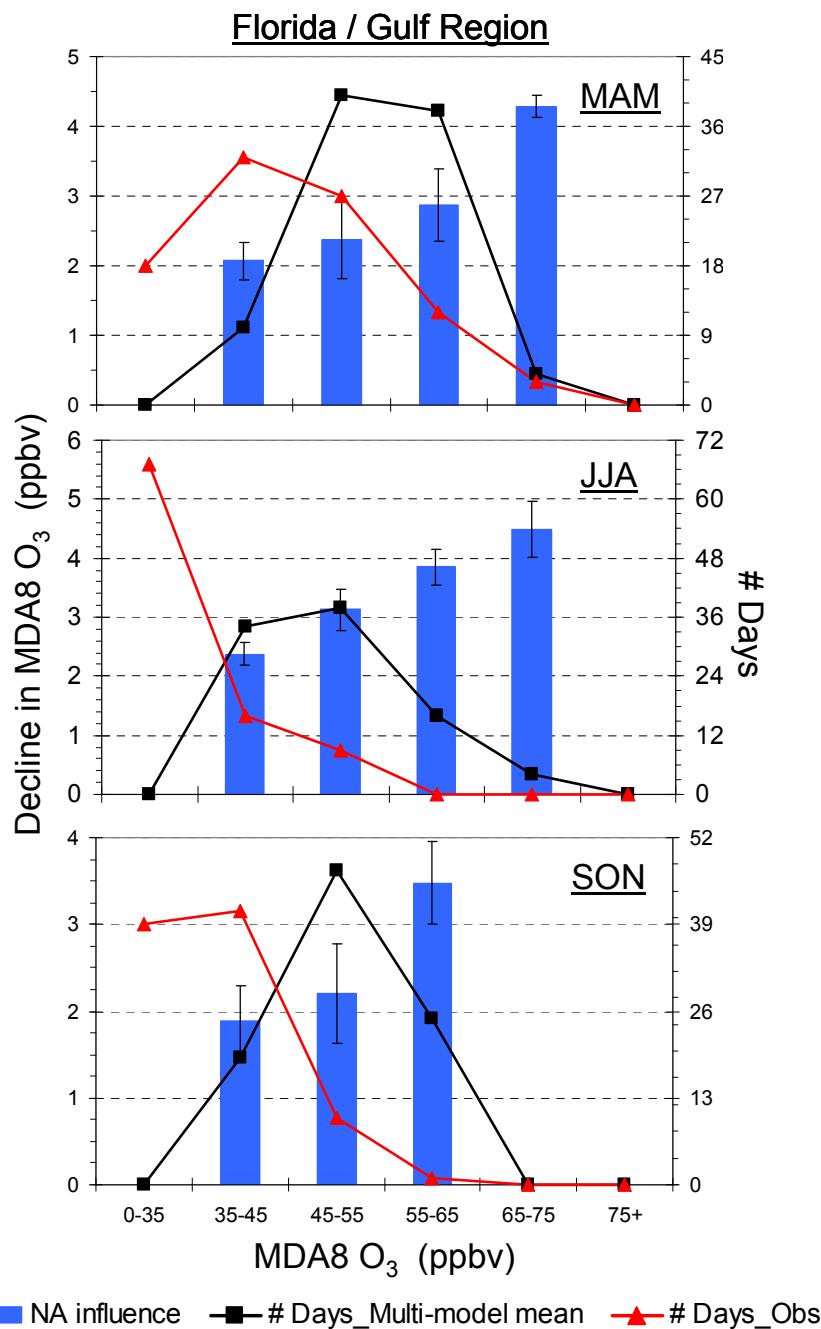


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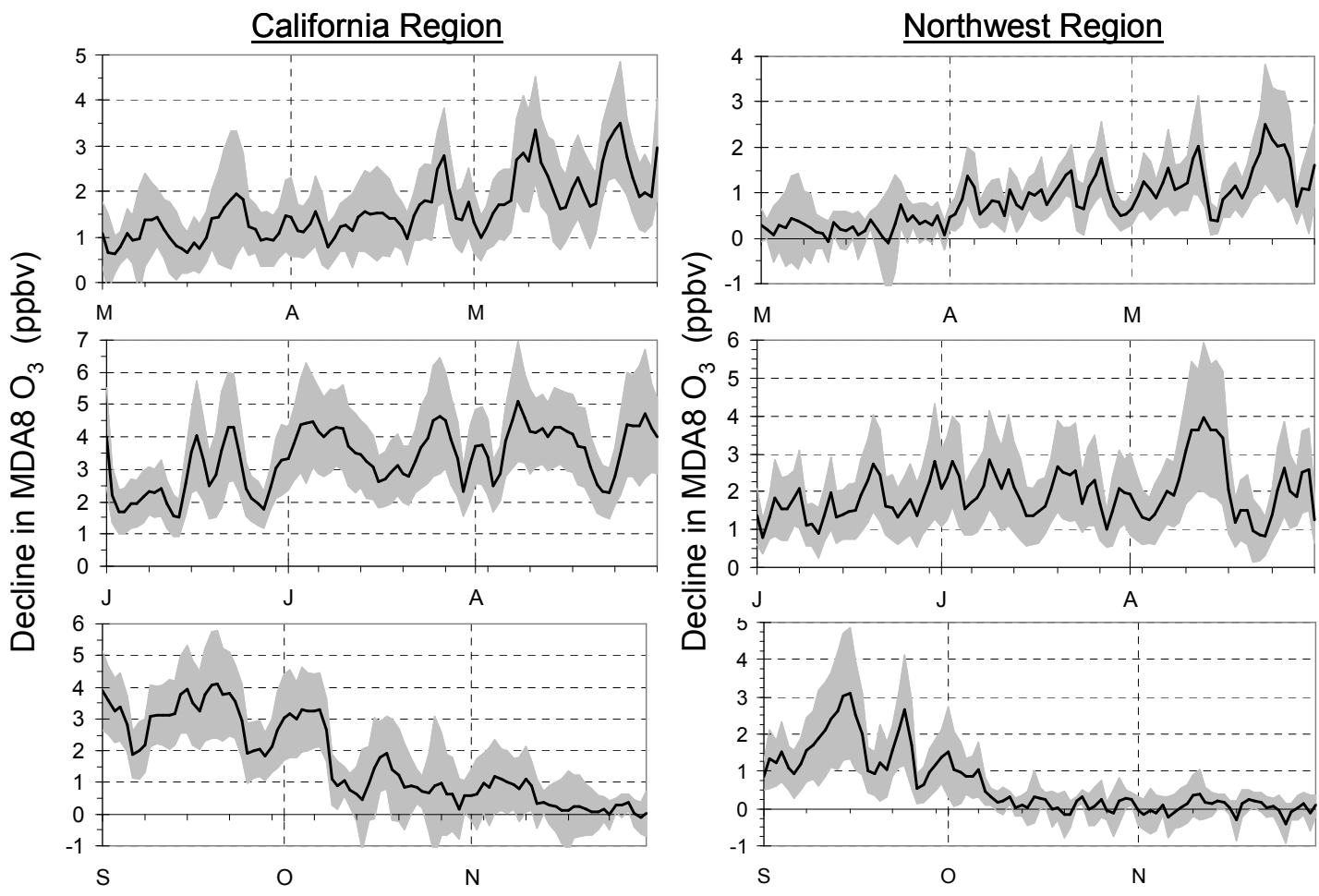


Figure A10. As in Fig. 11, but for the other seven regions.

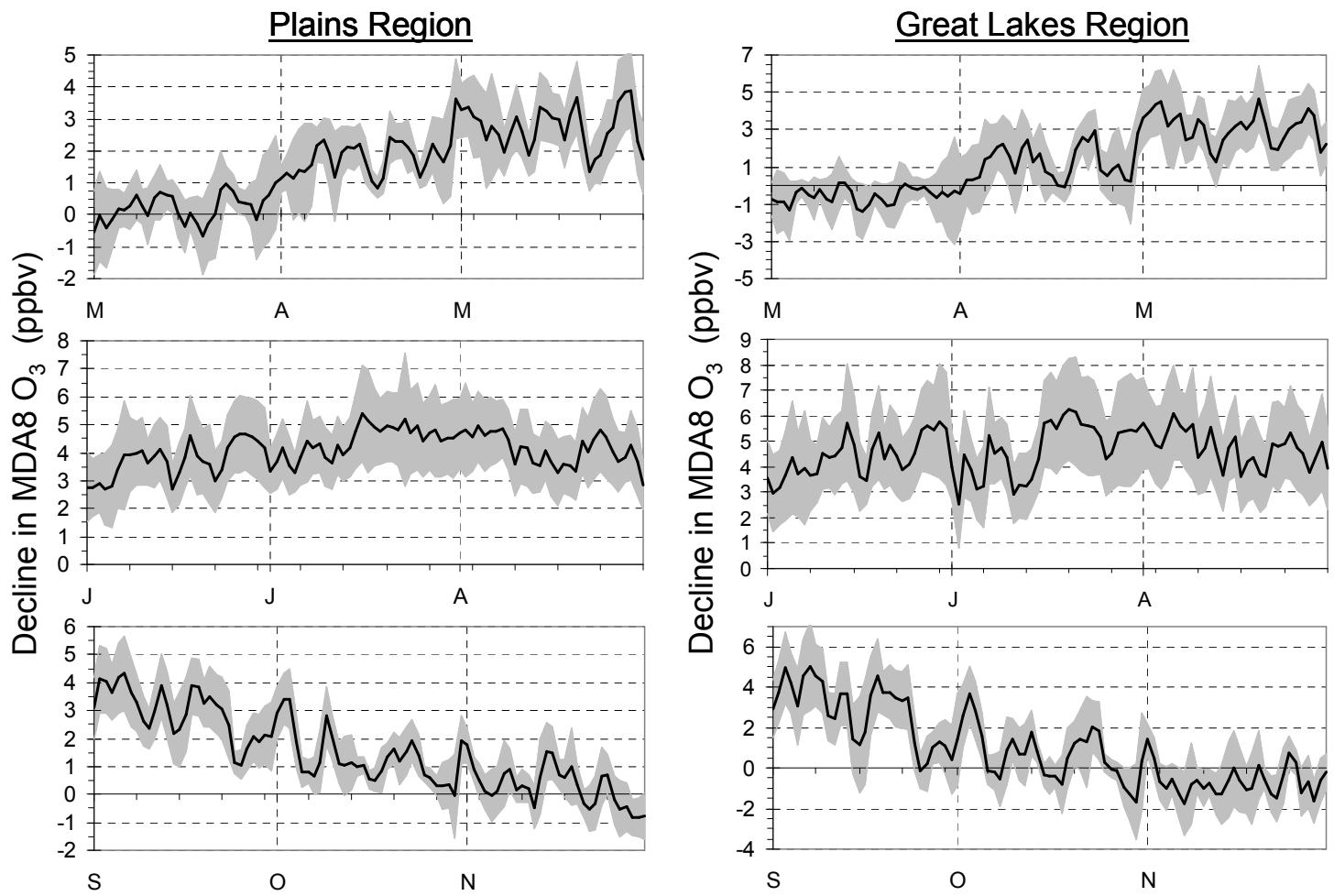


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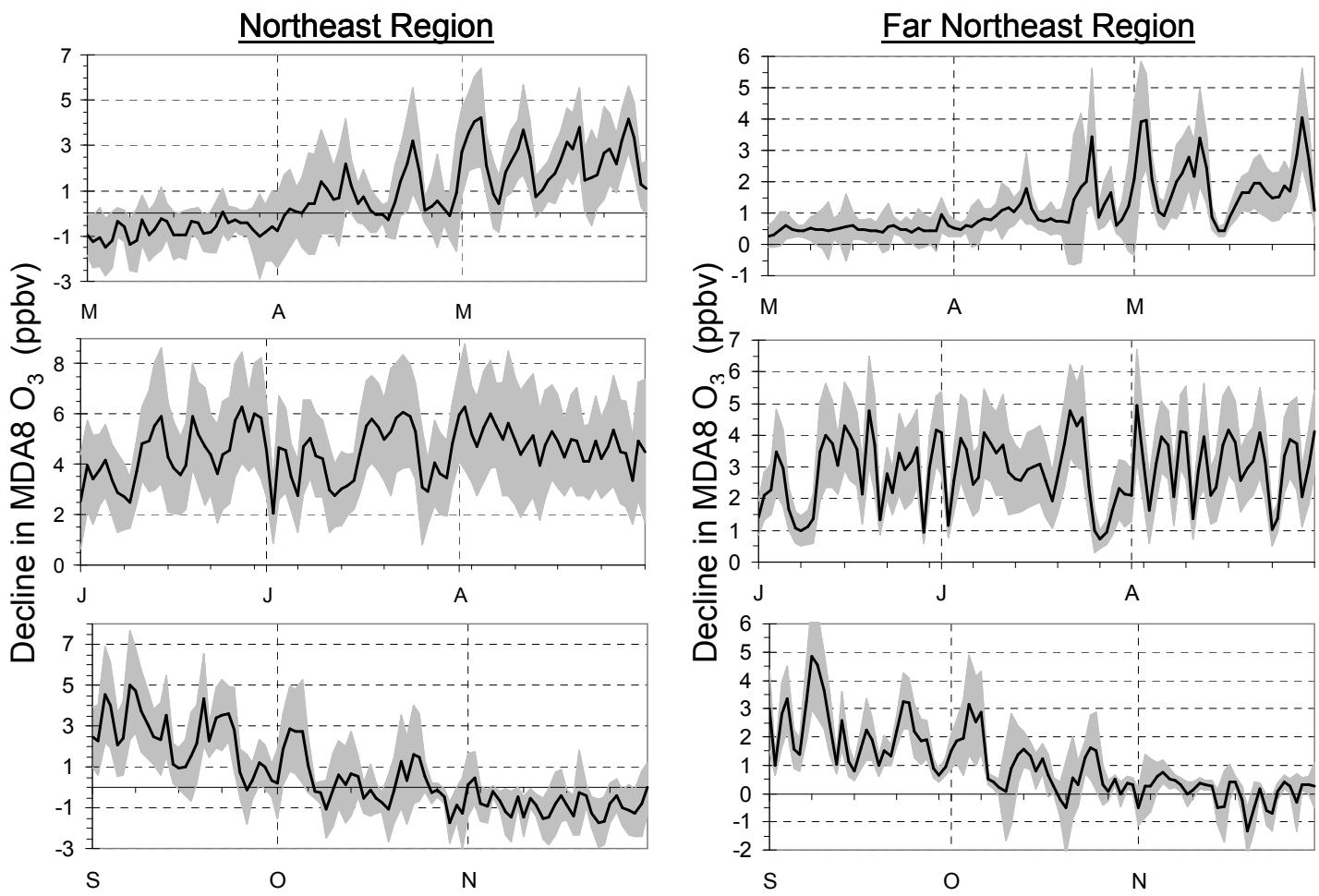


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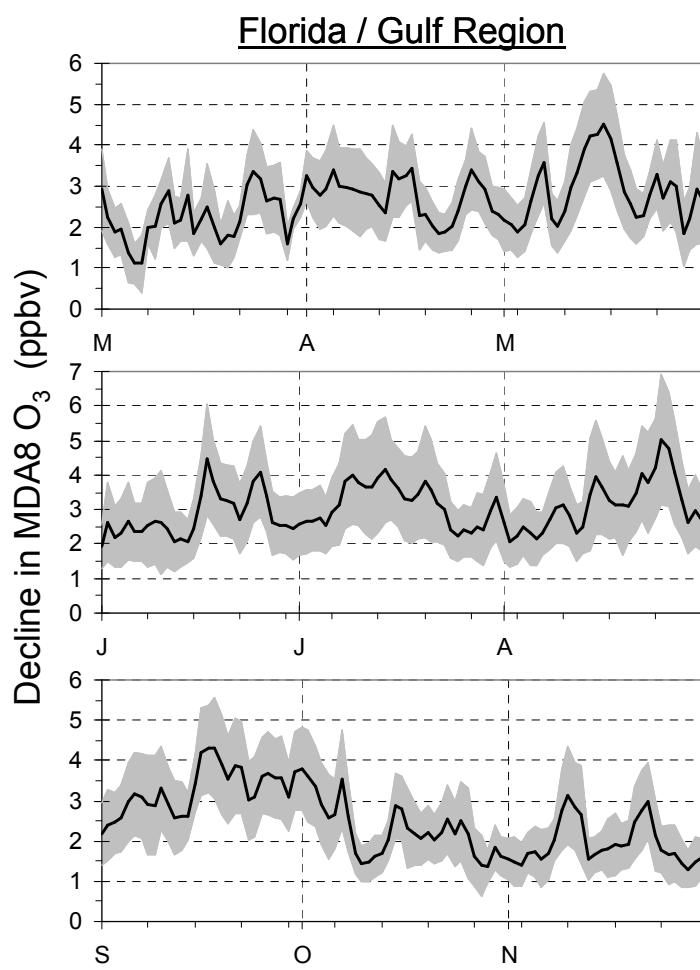


Figure A10. (cont'd)