S1. Methodology

S1.1 Comparison of long-term trends in O_3 annual averages with 3-yr averaged data

Linear trends were tested both for O_3 annual averages and 3-yr average O_3 data with the non-parametric Theil-Sen approach. Although, slightly larger O_3 growth rates are determined for the smoothed data than for the annual averages as shown in Fig. S2, non-significant differences (p>0.05) were observed between both Sen slopes. Considering this, and that the smoothing of O_3 annual averages could lead to miss significant features in the current trends (Carslaw et al., 2007; Carslaw, 2015), in the current study, O_3 annual averages with no smoothing were used to determine the long-term trends reported at the 3 metropolitan areas.

S1.2 Monitoring of O_3 in the Mexico City Metropolitan Area (MCMA)

The MCMA is located in Central Mexico (19°30'N, 99°02'W) and covers an area of around 9,500 km^2 (Fig. 1). It lies at an average altitude of 2,500 m asl and is surrounded completely by mountains. The MCMA has a population of 20.1 million inhabitants, which makes it the most populous city in Mexico and third in the world (INEGI, 2010). The climate is mild with an annual average temperature of 12-16°C and annual average rainfall of 820 mm. For decades, air pollution emissions in the MCMA, primarily from the industrial, transport and services sectors, resulted in air quality that was and is considered to be very poor by comparison with other large cities in Mexico and with other megacities worldwide (INE, 2011).

Within the MCMA, air quality monitoring is carried out by the Atmospheric Monitoring System of Mexico City (SIMAT), which measures 7 criteria air pollutants (O_3, SO_2, NO, NO_2, CO, PM_{10} and $PM_{2.5}$), and 5 meteorological parameters (WS, WD, Temp, RH and UV-SR). SIMAT currently operates a network of 34 stations to monitor air quality, and 15 stations to record meteorological parameters (SEDEMA, 2016a). Data are stored on a central server as hourly averages pending validation by SIMAT. The validated SIMAT data archive comprises hourly measurements of the above 7 criteria air pollutants and 5 meteorological parameters from January 1986 to December 2014. It is managed by the Government of Mexico City, and data were downloaded from their web site (http://www.aire.df.gob.mx/default.php).

S1.3 Monitoring of O_3 in the Guadalajara Metropolitan Area (GMA)

The GMA is located NW of Mexico City (20°39'N, 103°21'W; Fig. 2a), covers an area of 3,450 km^2, and with a population of 4.5 million inhabitants, is the second most populous urban area in Mexico (INEGI, 2010). It is located at an average altitude of 1,550 m asl. The climate is mild with dry springs and wet summers. The annual average temperature is 20.9°C and annual average rainfall is 1,030 mm. The GMA experiences occasional pollution events due to local emissions from automobiles, industry, and commercial and services activities (INE, 2011; SEMADET, 2014). Air quality is monitored within the GMA by the Atmospheric Monitoring System of the Jalisco State (SIMAJ). SIMAJ currently operates 10 stations, which report hourly data for 6 criteria pollutants (CO, NO, NO_2, O_3, PM_{10}, and SO_2) and four meteorological parameters (WS, WD, Temp and RH) (SEMADET, 2016). The SIMAJ data archive

contains hourly data recorded from January 1996 to December 2014. It is managed by the Environment and Territorial Development Secretariat of the Jalisco State government, with data downloaded from their web site (http://siga.jalisco.gob.mx/aire/Datos.html).

S1.4 comparison of estimated trends from *openair* and MAKESENS 1.0 macro

To rule methodology influences on the reported trends, Sen slopes for O_3 de-seasonalised annual averages obtained with the MAKESENS 1.0 macro were compared with those obtained with the Theil-Sen function included in the *openair* package for R software (Salmi et al., 2002; Carslaw, 2015). No significant differences ($p<0.05$) were found between the slopes determined from both approaches as shown in Fig. S4. Hence, all trends reported here were obtained with *openair*.

REFERENCES:

Carslaw, D. C., and Carslaw, N.: Detecting and characterising small changes in urban nitrogen dioxide concentrations, Atmos. Environ., 41, 4723-4733, doi:10.1016/j.atmosenv.2007.03.034, 2007.

Carslaw, D. C.: The openair manual - open-source tools for analysing air pollution data, Manual for version 1.1-4, King's College London, 2015.

INE (Instituto Nacional de Ecologia): Cuarto almanaque de datos y tendencias de la calidad del aire en 20 ciudades mexicanas 2000-2009, INE-SEMARNAT, México, D.F., 405 pp., 2011.

INEGI (National Institute of Statistics and Geography): XIII Censo General de Población y Vivienda 2010, México, available at: http://www.censo2010.org.mx/, last Access: 22 May 2016, 2010.

SEDEMA (Secretaria del Medio Ambiente de la Ciudad de Mexico): Sistema de Monitoreo Atmosférico, available at: http://www.aire.df.gob.mx/default.php, last access: 21 May 2016, 2016a.

SEMADET, (Secretaria de Medio Ambiente y Desarrollo Territorial): Inventario de Emisiones de Contaminantes Criterio del Estado de Jalisco 2008, SEMARNAT, Gobierno del Estado de Jalisco, available at: http://semadet.jalisco.gob.mx/sites/semadet.jalisco.gob.mx/files/ inventario _de_ emisiones_ cc_jalisco_2008.pdf, last Access: 23 May 2016, 2014.

SEMADET, (Secretaria de Medio Ambiente y Desarrollo Territorial): Sistema de Monitoreo Atmosférico de Jalisco, available at: http://siga.jalisco.gob.mx/aire/ last access: 21 May 2016.
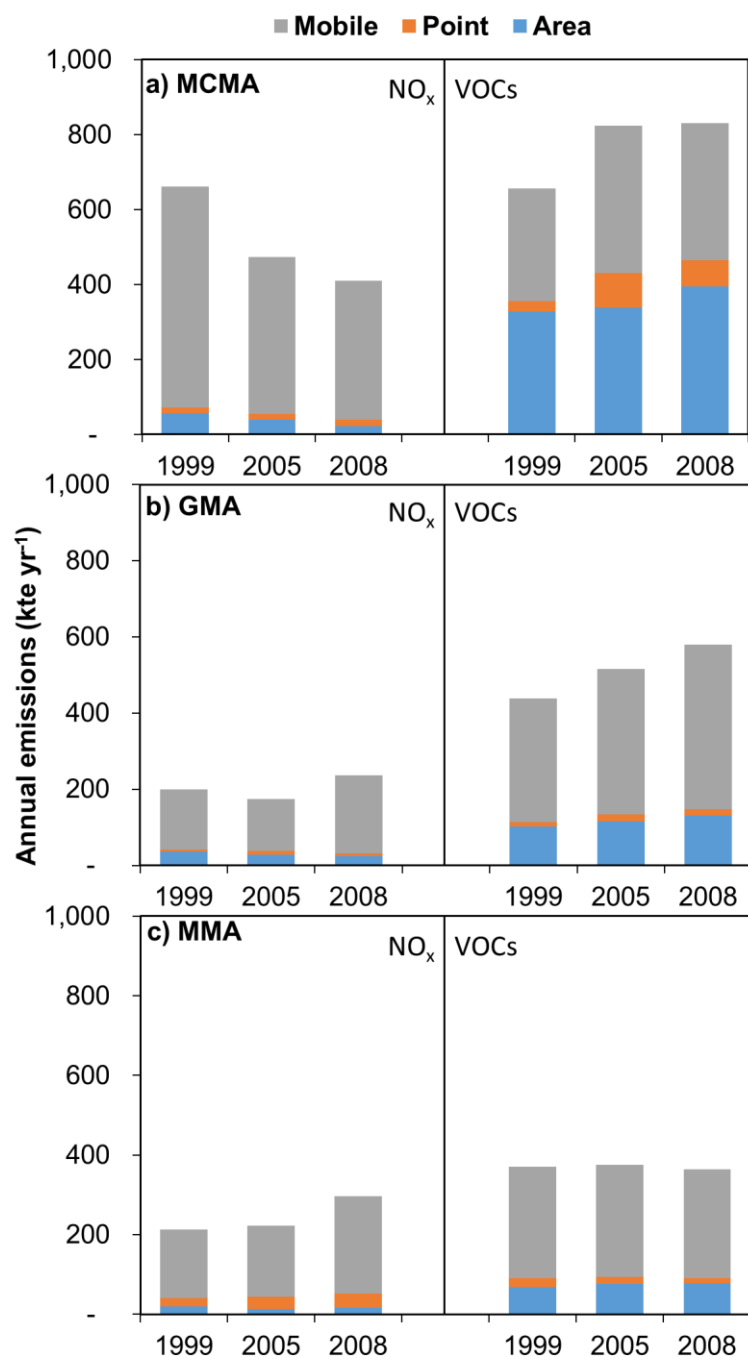
Fig. S1. NO$_x$ and VOCs anthropogenic emission estimates reported in the NEI for the 1999-, 2005- and 2008-base years for the MCMA, GMA and MMA. Adapted from: SEMARNAT, 2006, 2014, 2015.
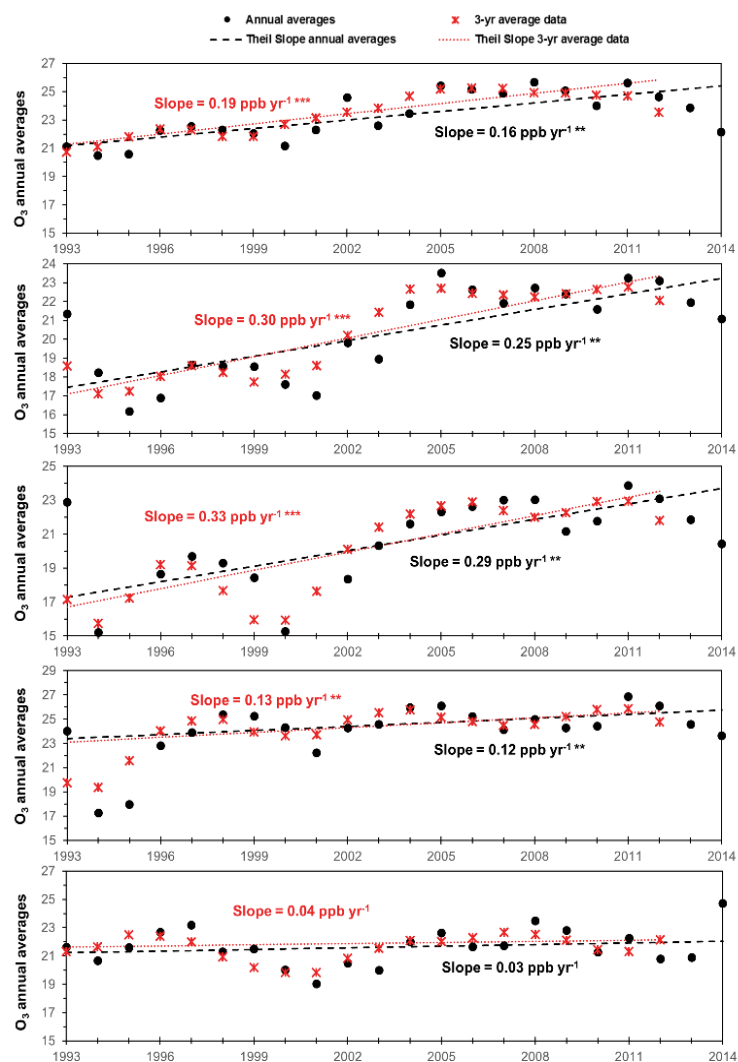
Fig. S2. Comparison of long-term trends in for O$_3$ annual averages (1993-2014), and 3-yr average O$_3$ data (1993-2012). The dashed lines represent the Theil slopes. Statistical significance is expressed as $p < 0.1 =^+$, $p < 0.05 = *$, $p < 0.01 = **$ and $p < 0.001 = ***$.
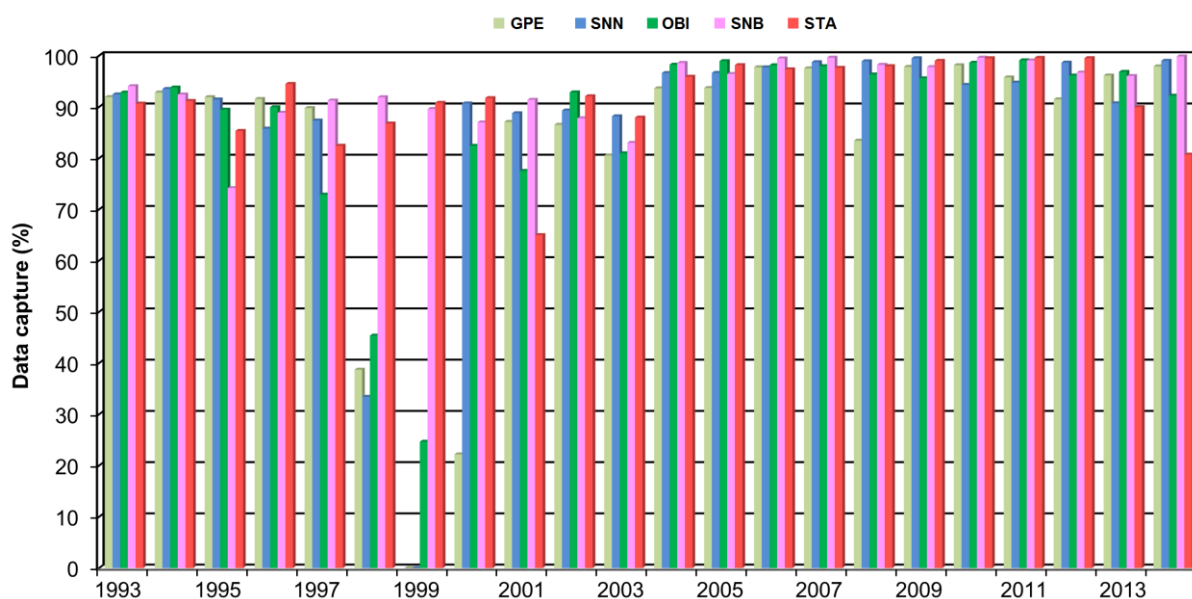
Fig. S3. Data capture of 1-h averages recorded for O$_3$ at the 5 monitoring sites within the MMA during 1993-2014.
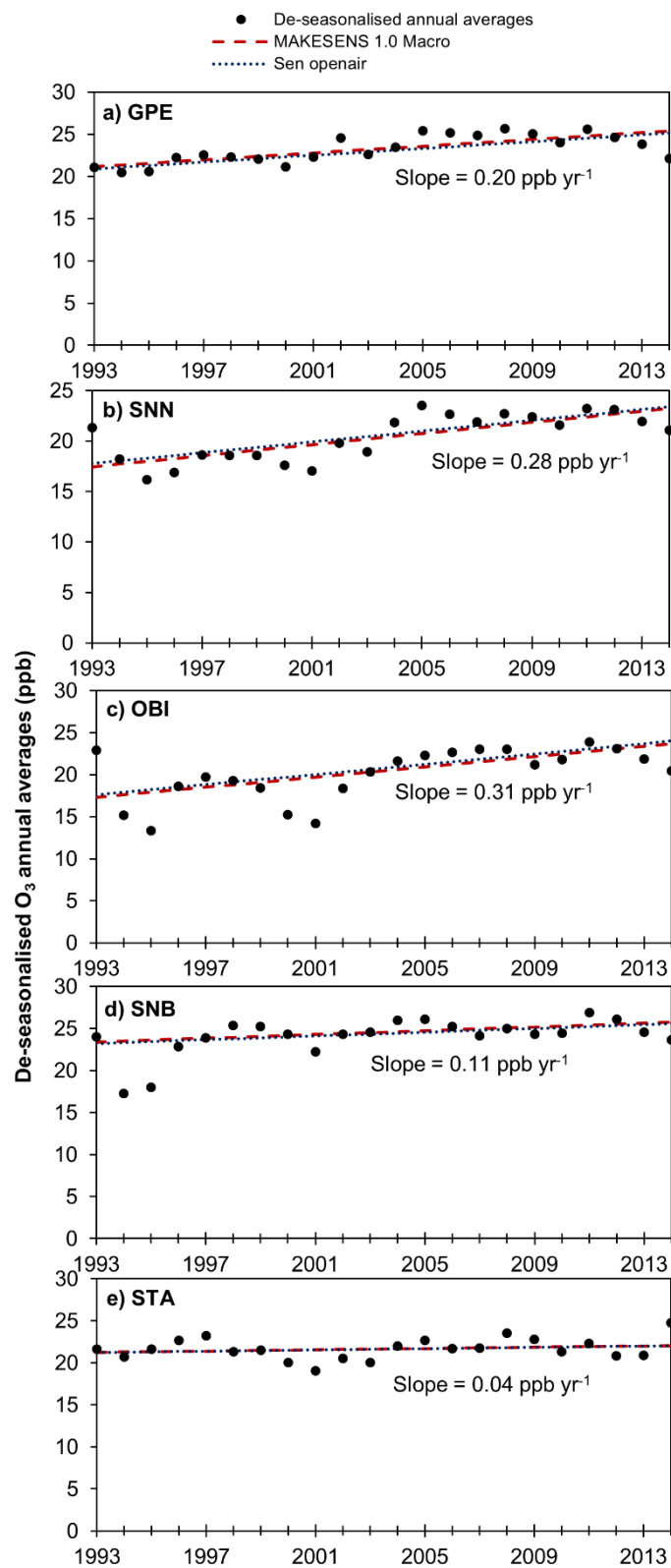
Fig. S4. Comparison of Sen slopes determined with the Theil-Sen function included in the *openair* package for R(Carslaw, 2015), and those obtained with the MAKESENS 1.0 macro (Salmi et al., 2002).
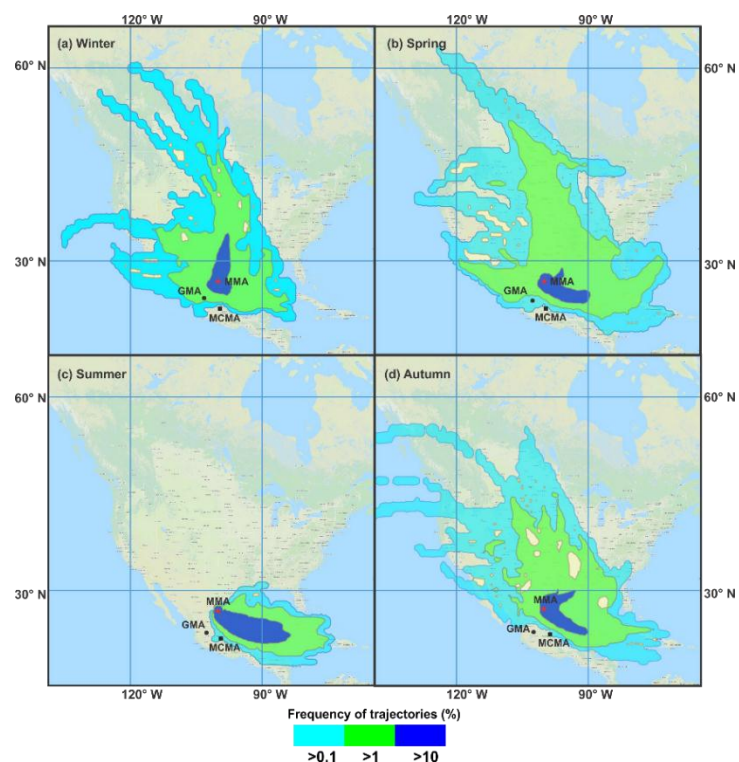
Fig. S5. HYSPLIT frequency plots of 96-h back trajectories from the MMA by season during 2014. Each panel represents 3-months of data, with a new trajectory represented every 6 hours.
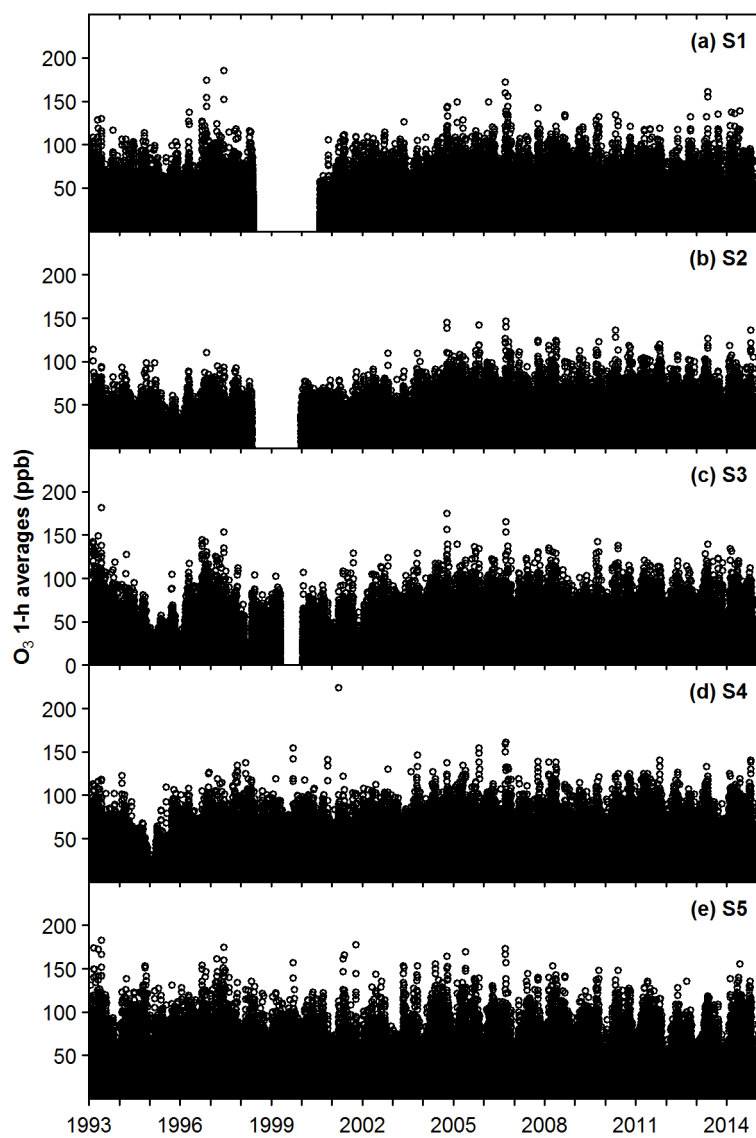
Fig. S6. 1-h averages of O_3 calculated at the 5 sites within the MMA during 1993-2014.
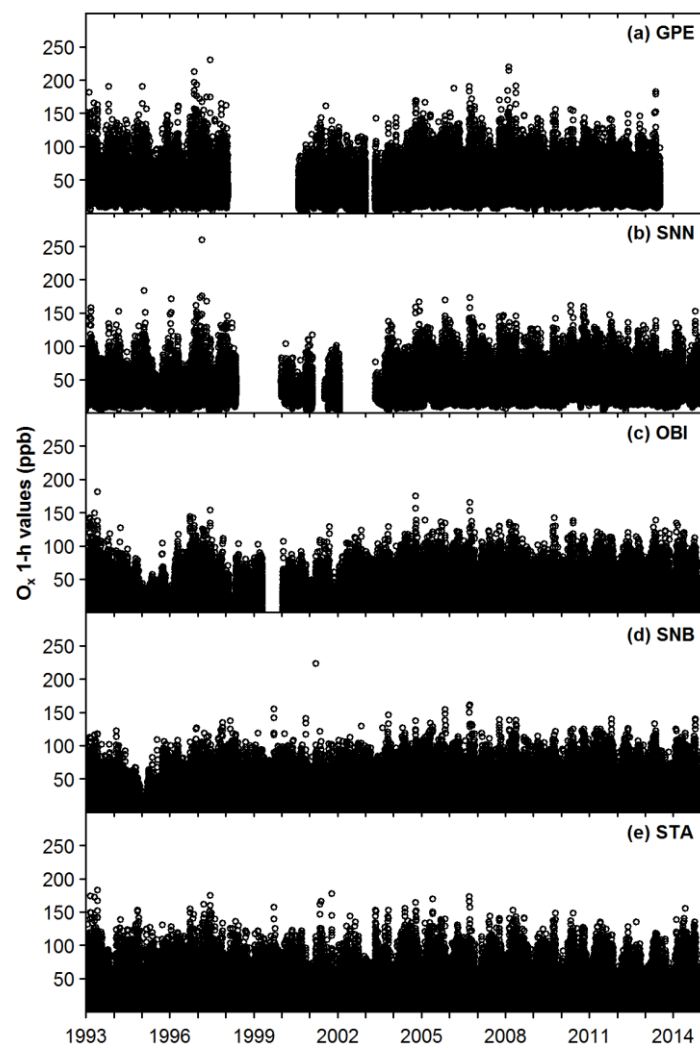
Fig. S7. 1-h values of O_X calculated at the 5 sites within the MMA during 1993-2014.
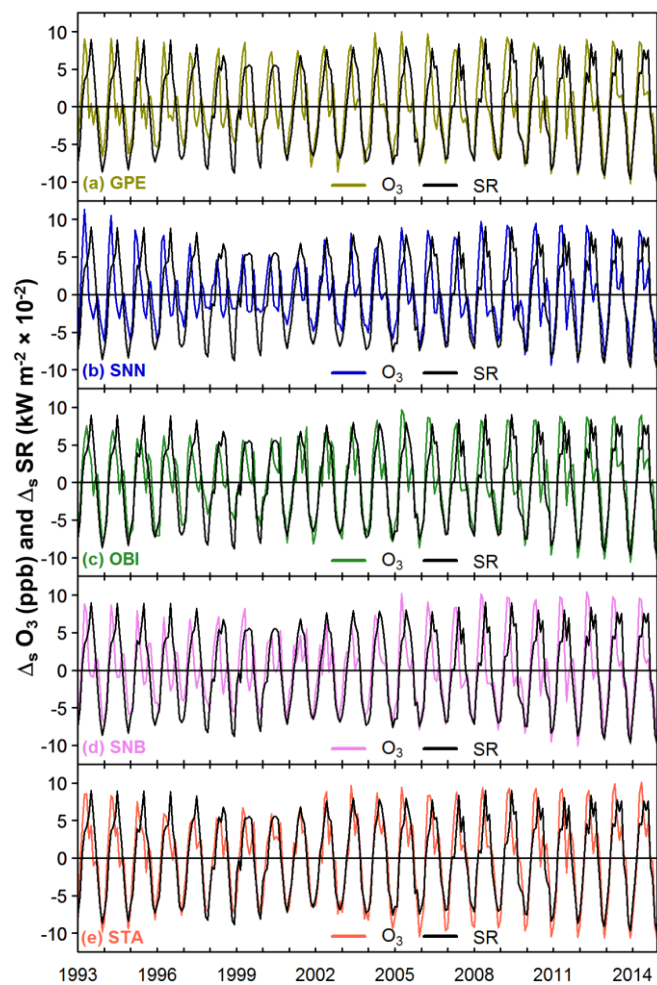
Fig. S8. Seasonal variations in O_3 mixing ratios and SR at the 5 sites within the MMA, constructed from filtered data using the STL technique developed by Cleveland et al. (1990).
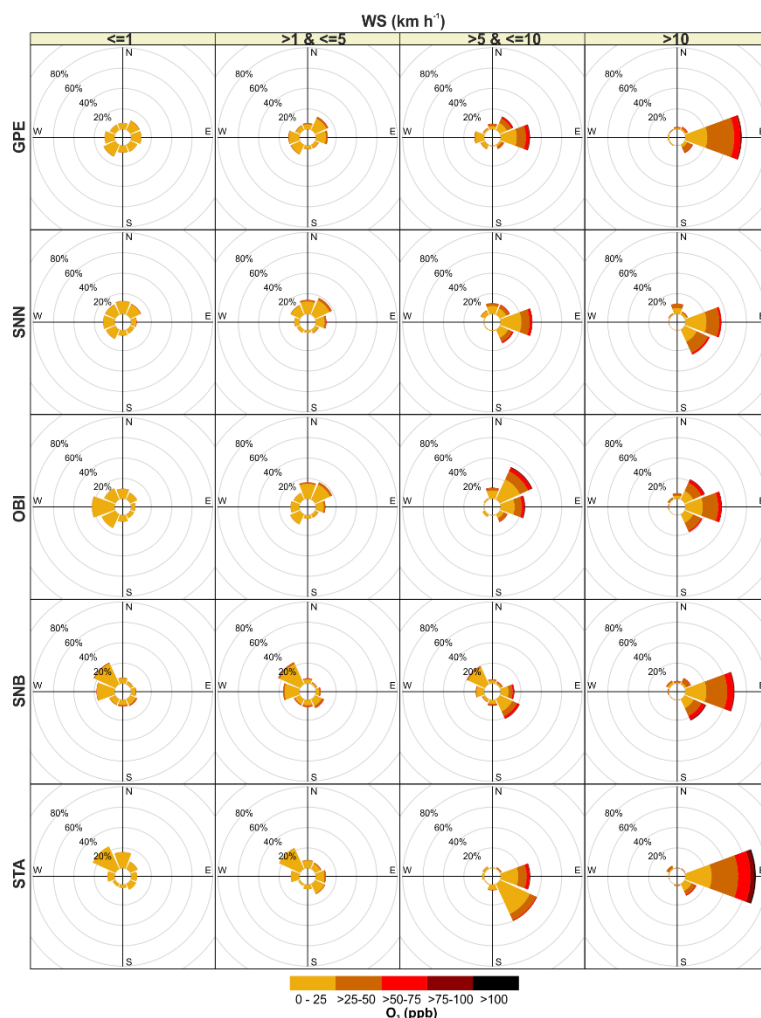
Fig. S9. Pollution roses for O$_3$ 1-h averages by wind speeds recorded at the 5 monitoring sites within the MMA during 1993 to 2014.
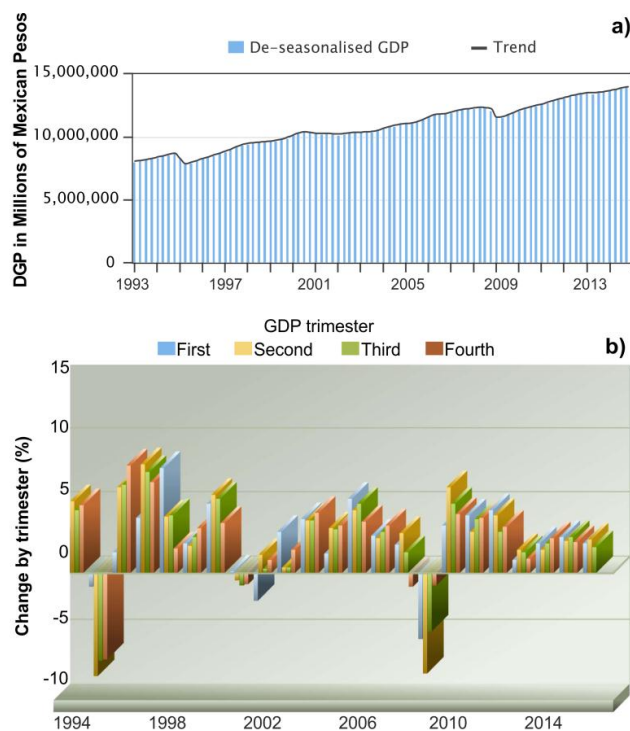
Fig. S10. a). Domestic gross product (DGP) by trimester and its trend during 1993-2014 in Mexico. **b).** Change in percentage in the DGP. The dip observed in 1995 is due to the economic crisis experienced in Mexico between 1994-1996, the one observed in 2001-2002 is due to the economic recession in the

Mexican economy during the period, and the one observed in 2009 derives from the global recession period experienced between 2008-2009.
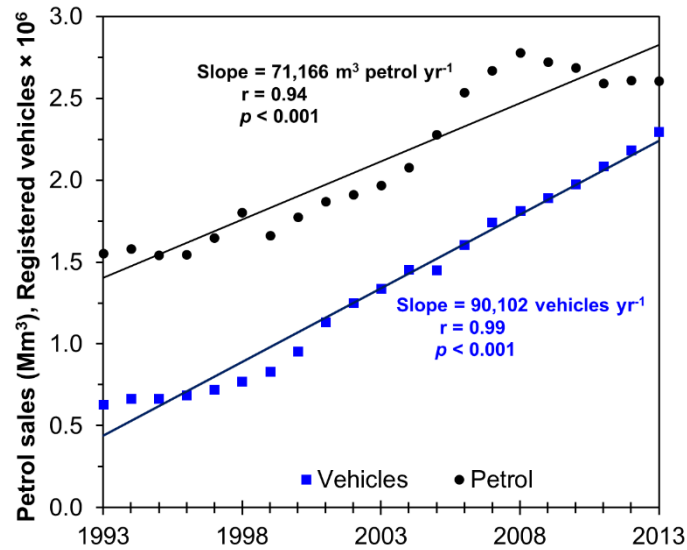
Fig. S11. Petrol sales in the MMA and registered vehicles in the Nuevo Leon state during 1993-2013.

Table S1. Statistics of O$_3$ hourly averages in units of ppb at the 5 monitoring sites in the MMA during 1993-2014.

Year	GPE					SNN					OBI					SNB					STA				
	Min	AVG	SD	Median	Max	Min	AVG	SD	Median	Max	Min	AVG	SD	Median	Max	Min	AVG	SD	Median	Max	Min	AVG	SD	Median	Max
1993	0	23	17	20	130	2	31	16	28	113.5	0	31	21	26	181.5	0	32	19	27	118	0	30	23	23	182.5
1994	0	21	16	19	113.5	0	19	12	17	98.5	0	15	15	11	127	0	18	14	15	122.5	0	21	22	15	153
1995	0	24	16	22	103	0	19	12	17	98	0	14	11	13	104.5	0	20	15	17	109	0	24	21	19	130.5
1996	0	23	17	21	174	0	18	13	15	110	0	20	19	15	144	0	24	18	21	126.5	0	23	22	17	154
1997	0	22	18	19	185.5	0	18	14	16	94.5	0	19	18	15	153.5	0	23	18	20	134.5	0	23	24	17	174.5
1998	0	27	18	27	115.5	0	20	13	19	76.5	0	18	16	14	103.5	0	26	20	23	137	0	21	20	16	135.5
1999	NR	NR	NR	NR	NR	0	25	22	20	52	0	21	17	17	102.5	0	25	19	21	154.5	0	22	21	17	157
2000	0	15	13	13	105.5	0	18	13	15	78	0	16	14	12	107	0	25	19	20	141	0	20	21	13	122.5
2001	0	22	17	19	111	0	16	12	15	88	0	14	14	10	129	0	21	17	17	223.5	0	19	20	12	177.5
2002	0	26	19	22	114	0	21	15	19	109	0	21	18	16	123.5	0	25	19	22	129.5	0	21	21	15	143.5
2003	1	22	18	17	126	1	18	14	15	109	1	20	18	14	129	1	25	19	20	146	1	20	21	14	153
2004	0	25	19	22	144	0	23	17	19	145	0	23	21	17	175	0	28	21	24	137	0	23	24	16	164
2005	0	27	21	24	149	0	25	19	21	142	0	23	21	17	139	0	28	22	23	154	0	24	23	17	169
2006	0	27	21	24	172	0	25	19	21	146	0	25	21	19	165	0	28	22	23	161	0	23	22	16	173
2007	1	27	19	23	142	0	23	17	19	124	0	23	20	17	130	1	25	20	20	139	1	22	21	15	144
2008	0	28	21	23	134	1	25	18	20	124	0	24	21	19	135	1	27	21	22	138	0	25	22	18	153
2009	0	28	20	25	132	0	25	17	21	123	0	22	18	18	142	1	26	20	22	121	0	25	21	19	148
2010	1	26	19	23	134	1	23	17	19	136	1	22	19	18	138	1	26	20	21	125	1	24	21	18	148
2011	0	29	20	25	119	0	26	18	22	120	0	26	21	21	121	1	30	22	25	140	1	26	23	20	135
2012	0	26	18	23	132	1	25	16	22	107	0	23	19	20	120	0	27	20	24	126	0	23	19	19	135
2013	1	26	19	23	161	2	24	17	21	126	1	23	19	19	139	1	26	19	22	133	1	23	19	18	118
2014	1	24	19	20	139	2	23	17	19	136	1	21	19	17	134	2	26	21	21	140	1	27	23	21	155

NR: No records
AVG: Annual average.
SD: Standard deviation of the average.

Table S2. Statistics of O_X hourly data in units of ppb at the 5 monitoring sites within the MMA during 1993-2014.

Year	GPE					SNN					OBI					SNB					STA				
	Min	AVG	SD	Median	Max	Min	AVG	SD	Median	Max	Min	AVG	SD	Median	Max	Min	AVG	SD	Median	Max	Min	AVG	SD	Median	Max
1993	5	37	21	33	190	4	35	17	32	158	6	51	27	45	330	7	42	21	37	250	10	46	28	39	286
1994	3	35	17	31	148	6	32	14	30	153	2	41	21	36	233	7	32	16	30	191	8	46	27	39	251
1995	2	35	17	32	190	3	30	15	28	184	4	39	18	35	160	4	32	18	29	193	11	44	23	39	211
1996	2	36	20	33	213	3	32	16	30	171	6	46	25	41	220	7	37	20	34	219	10	45	26	38	196
1997	5	36	19	32	230	5	33	17	31	260	5	44	25	39	200	6	37	19	33	204	6	46	27	38	213
1998	8	40	19	36	162	6	38	14	36	146	2	40	23	37	216	2	40	22	37	209	2	37	23	33	181
1999	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	7	41	18	38	142	2	30	19	26	171	5	39	24	34	208
2000	2	34	18	32	102	2	27	14	25	110	2	46	31	40	290	2	31	20	26	163	2	39	26	32	179
2001	2	34	18	31	161	4	31	14	29	117	2	51	27	46	251	2	26	17	21	228	2	34	25	27	207
2002	2	37	20	34	130	2	23	17	21	99	6	48	23	43	233	2	29	21	23	133	2	32	21	26	146
2003	2	29	18	25	143	5	30	17	27	138	2	30	21	25	160	3	29	14	26	108	2	31	21	26	139
2004	4	37	20	33	169	6	33	17	30	167	3	37	22	33	225	3	39	19	35	158	2	38	25	31	182
2005	8	42	20	39	166	8	40	19	38	170	8	42	22	38	179	10	41	21	37	196	5	46	26	40	192
2006	7	41	20	37	190	9	39	18	36	173	11	47	22	42	185	8	42	21	38	183	2	51	26	46	239
2007	7	38	19	35	170	8	39	17	36	146	13	45	22	41	189	9	40	19	37	162	2	47	26	42	185
2008	7	41	21	38	220	6	40	18	37	145	9	45	22	41	171	10	44	21	40	172	2	42	25	36	188
2009	4	40	18	37	155	7	37	16	34	142	10	41	18	38	168	10	43	19	40	166	5	43	22	37	176
2010	7	39	19	35	155	8	46	18	43	162	10	43	22	39	168	6	43	19	40	145	2	34	23	30	181
2011	7	41	19	38	142	3	41	19	38	147	3	40	23	36	147	2	43	22	39	165	2	45	26	39	172
2012	8	37	17	34	149	2	37	16	34	136	13	42	19	38	153	12	43	19	40	163	6	46	22	41	219
2013	8	39	18	36	182	7	34	16	31	138	3	40	20	35	162	6	42	20	39	158	3	43	21	40	222
2014	NR	NR	NR	NR	NR	8	37	17	33	153	6	36	21	30	175	3	34	22	30	164	3	37	22	31	173

NR: No records.
AVG: Annual average.
SD: Standard deviation of the averages.