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Article

Supplement to "30 Years of Air Quality Trends in Japan"

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S1. Air quality standard in Japan, US and Europe and WHO guidelines

Table S1-1 shows the limit values and the date of notification of Japan's air quality standards and also shows the number of stations complying with the air quality standards as of 2018 [S1]. According to this table, the compliance rate of SPM, NO_2, SO_2, and CO standards have achieved almost 100% for both AAQMS and RsAQMS. On the other hand, for $PM_{2.5}$, 93.5% of AAQMS (765 out of 818 stations) and 93.1% of RsAQMS (216 out of 232 stations) have not reached 100%. For O_3, the compliance rate is almost 0% for both AAQMS (1 out of 1,155 stations) and RsAQMS (0 out of 28 stations).

Table S1-1. Limit values and notification dates of air quality standards in Japan and the number of stations complying with the standards in 2018

Species	Unit	Air Quality Standard			Notification date	# of attainment stations (all stations) (1)	
		Annual	Daily	Hourly		AAQMS	RsAQMS
$PM_{2.5}$	$\mu g/m^3$	15	35		September 9, 2009	765 (818)	216 (232)
SPM	mg/m^3		0.10	0.20	May 8, 1973	1292 (1294)	384 (384)
Ox	ppm			0.06	May 8, 1973	1 (1155)	0 (28)
NO2	ppm		0.06		July 11, 1978	1233 (1233)	390 (391)
SO_2	ppm		0.04	0.1	May 16, 1973	947 (948)	49 (49)
CO	ppm		10	20	May 8, 1973	56 (56)	226 (226)

Note:
(1) as of 2018

Table S1-2 compares the air quality standards of Japan, the U.S. and Europe, and the WHO guidelines for particulate matter ($PM_{2.5}$, PM_{10}, SPM) and gaseous components (O_3, NO_2) [S2][S3][S4]. The unit of particulate matter is unified as $\mu g/m^3$, and the concentration unit for gaseous components is unified as ppb. Among gaseous substances, the following conversions are used for substances for which the ambient air quality standard is expressed in $\mu g/m^3$ (assuming 20°C and 1 atm).

O_3: 1 ppb = 1.997 $\mu g/m^3$, NO_2: 1 ppb = 1.913 $\mu g/m^3$

The percentile values in the table represent the percentile of the annual measurement results in ascending order. For example, in the case of the 98th percentile value of the daily concentration, the eighth highest value among the 365 daily concentration data per year must not exceed the air quality standard. The notation "n.a." indicates that there is no such tolerance setting.

Table S1-2 Comparison of air quality standards (particulate matter and gaseous component) among Japan, the United States, Europe, and WHO guidelines

Species		Japan		US		Europe		WHO	
$PM_{2.5}$	annual	15 μg/m^3	mean	12 μg/m^3	mean	25 μg/m^3	mean	10 μg/m^3	mean
	daily	35 μg/m^3	98%	35 μg/m^3	98%	−−	−−	25 μg/m^3	99%
PM_{10}, SPM	annual	−−	−−	−−	−−	40 μg/m^3	mean	20 μg/m^3	mean
	daily	100 μg/m^3	98%	150 μg/m^3	99.7%	50 μg/m^3	90%	50 μg/m^3	99%
O_3	hourly	60 ppb	n.a.	−−	−−	−−	−−	−−	−−
	8-hour	−−	−−	70 ppb	99%	60 ppb (120 μg/m^3)	93%	50 ppb (100 μg/m^3)	n.a.
NO_2	annual	−−	−−	53 ppb	mean	21 ppb (40 μg/m^3)	mean	21 ppb (40 μg/m^3)	mean
	daily	60 ppb	98%	−−	−−	−−	−−	−−	−−

From this table, the air quality standards for particulate matter ($PM_{2.5}$, PM_{10}, and SPM) can be compared with each other because the definition of averaged time for all of them is almost the same (e.g. annual average), although the definitions for PM_{10} and SPM are not completely identical. However, it is difficult to directly compare the standard values of O_3 and NO_2 in each region because the averaged time of the standard values is different. For example, O_3 is defined by 1-hour and 8-hour averaged values, and NO_2 is defined by annual and daily averaged values. Therefore, in order to be able to compare the air quality standards of O_3 and NO_2 in each region, we used the air pollutant concentration data in Japan and calculated different indices from the same air pollutant concentration database to investigate the relationship between them.

The left panel of Figure S1 plots the annual maximum hourly concentration and the annual 4th highest daily maximum of the 8-hour concentration (A4MDA8) of O_3 from the data of air pollutant concentrations at the AAQMS and RsAQMS in Japan from 1990 to 2017 (n=26,882). The right figure of Figure S2 also shows the relationship between the annual 98% value of the daily mean concentration of NO_2 and the annual mean value of NO_2 (n=51,398). The red rectangles in the figures indicate values that fall between 55 ppb and 65 ppb on the horizontal axis, a range roughly equivalent to Japan's air quality standards for O_3 and NO_2.

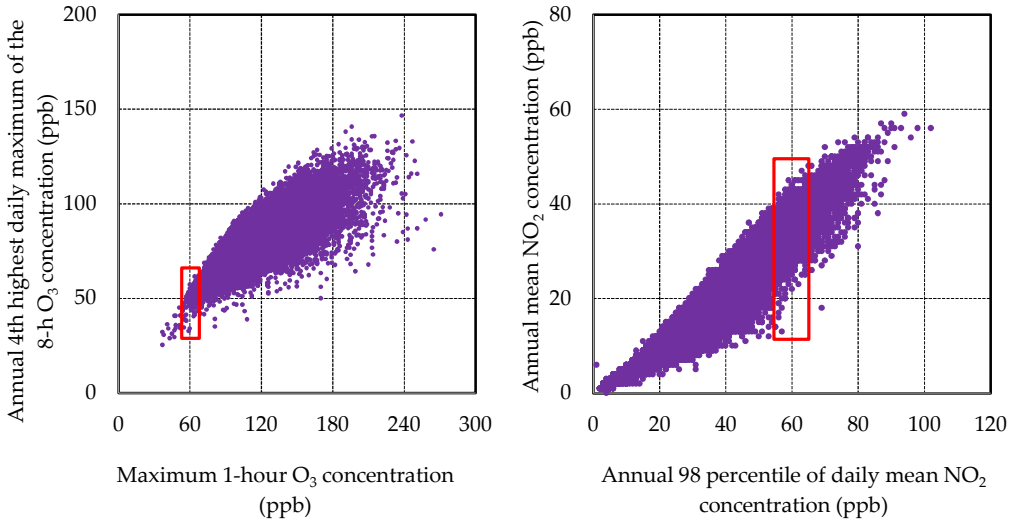

Figure S1. Conversion of air quality standards for O_3 (left figure) and NO_2 (right figure) (horizontal axis: Japanese air quality standards, vertical axis: European and U. S. air quality standards and WHO air quality guidelines)

From these figures, it can be seen that there is a relationship between the respective indices of O_3 and NO_2, although the variation is large. Averaging the data included in the red rectangle, mean concentration of A4MDA8 was 50.3 ± 5.3 (ppb). This value is lower

than the air quality standards of the United States and Europe, and is almost equivalent to the WHO guidelines. These results indicate that the air quality standards for O_3 in Japan are more stringent than those in the United States and Europe. This is thought to be one of the reasons why there are almost no stations in Japan that comply with the air quality standards for O_3. However, the percentage of stations in Japan where the A4MDA8 concentration is less than 70 ppb, which is equivalent to the U.S. air quality standard, is only about 10% [S5], so a further reduction of O_3 concentration is necessary. Similarly, averaging the NO_2 concentrations enclosed in the red squares, the mean annual NO_2 concentration was 33.7±4.4 ppb. This value was found to be lower than the U.S. air quality standards, but higher than the European air quality standards and WHO guidelines.

S2 Supplement to figure 1 in main text

Figure 1 in main text shows a map of the distributions of ambient air quality monitoring stations (AAQMS) and roadside air quality monitoring stations (RsAQMS) and Figure S2 shows example photos of an AAQMS and a RsAQMS [S6].

Figure S2. Photographs of an ambient air quality monitoring station (AAQMS) and a roadside air quality monitoring station (RsAQMS) [6]

Figure 1 in main text also shows the positional relationship from Region 1 to Region 8 considering the geographical commonality of Japan. The characteristics of each area are as follows:

Region 1 (Tohoku/Hokkaido) A region with a high latitude.

Region 2 (Kanto) An area that includes Tokyo, the capital, and surrounding prefectures and which has a concentration of economic activities.

Region 3 (Chubu) A mountainous area in the center of Japan and the area to the north of the mountains.

Region 4 (Tokai) Including Nagoya City, which is one of Japan's three major metropolitan areas, a mountainous area in the center of Japan and the plains on the south side.

Region 5 (Kinki) Japan's second largest commercial area, including the metropolis of Osaka City.

Region 6 (Chugoku) An area that is easily affected by trans-boundary air pollutants from the Asian continent because it is located close to the continent.

Region 7 (Shikoku) Surrounded by high mountains to the north and south, it is a climatically mild region, but it is susceptible to the emissions of vessels in the region.

Region 8 (Kyushu/Okinawa). An area which is affected by trans-boundary transportation from the Asian continent.

S3 $PM_{2.5}$ concentrations (annual average concentrations) in South Korea and China

Figure S3 shows the $PM_{2.5}$ concentrations (annual average concentrations) at three sites in South Korea and five sites in China from 2010 to 2019 [S7].

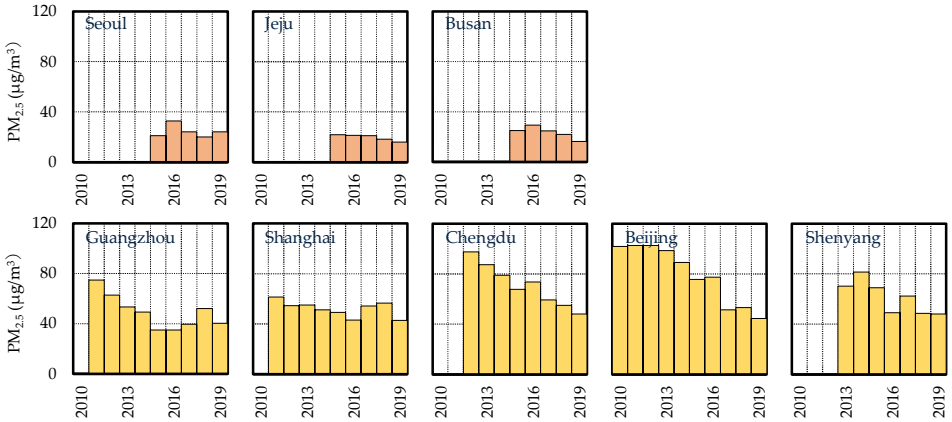

Figure S3. Annual average $PM_{2.5}$ concentration in cities in South Korea and China

S4 Ozone and related substances (nitrogen oxides, non-methane hydrocarbons)

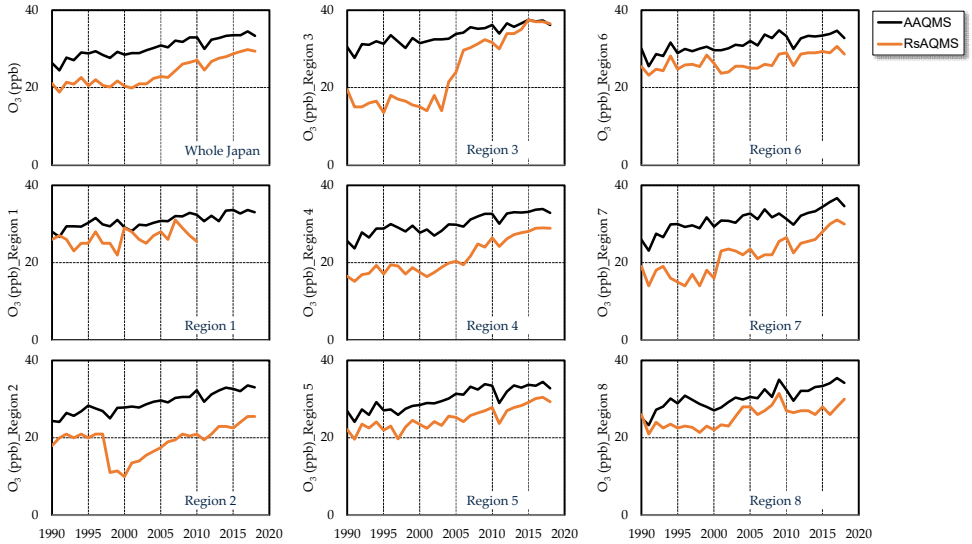

Figure S4-1. Annual average of 1-hour daytime ozone concentrations at the AAQMS and RsAQMS of the nation and each region from 1990 to 2018

Table S4-1. Annual average of 1-hour daytime ozone concentrations at the AAQMS for 1990-1992 and 2016-2018, averaged by region, and their ratios

Ozone	Concentration (ppb)		
AAQMS	1990~1992	2016~2018	$C_{2016\sim2018}$ / $C_{1990\sim1992}$
Whole Japan	26.2	33.8	1.29
Region 1	28.0	33.1	1.18
Region 2	25.0	32.9	1.32
Region 3	29.8	36.9	1.24
Region 4	25.7	33.5	1.30
Region 5	26.1	33.6	1.29
Region 6	28.1	33.8	1.20
Region 7	25.5	35.6	1.40
Region 8	25.2	34.6	1.37

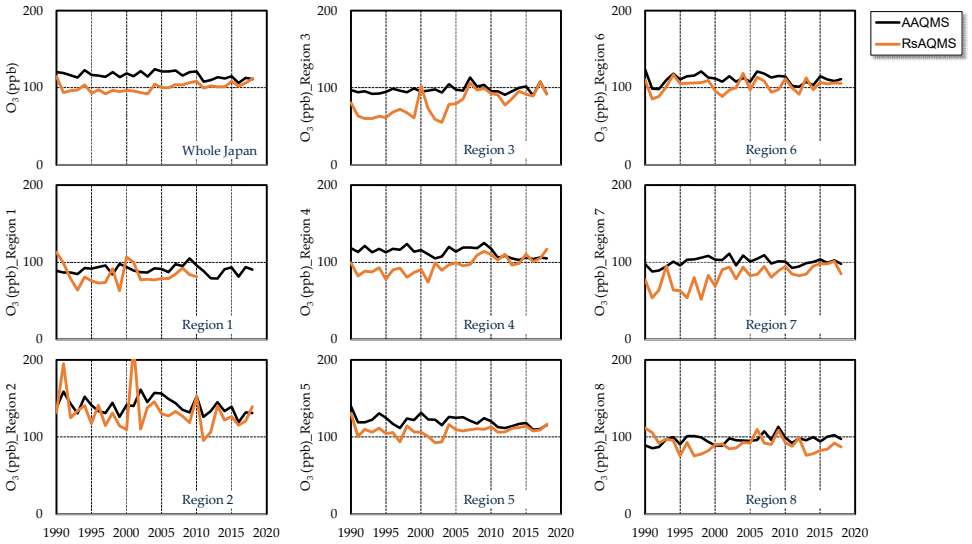

Figure S4-2. Annual maximum 1-hour daytime ozone concentrations at the AAQMS and RsAQMS averaged by region from 1990 to 2018

Table S4-2. Annual maximum daytime 1-hour ozone concentrations at the AAQMS for 1990-1992 and 2016-2018, averaged by region, and their ratios

Ozone	Concentration (ppb)		
AAQMS	1990~1992	2016~2018	$C_{2016\sim2018} / C_{1990\sim1992}$
Whole Japan	118.5	109.9	0.93
Region 1	87.5	88.5	1.01
Region 2	146.5	127.3	0.87
Region 3	95.6	96.6	1.01
Region 4	117.7	105.3	0.90
Region 5	125.8	111.8	0.89
Region 6	106.8	110.1	1.03
Region 7	91.6	99.9	1.09
Region 8	87.3	100.0	1.15

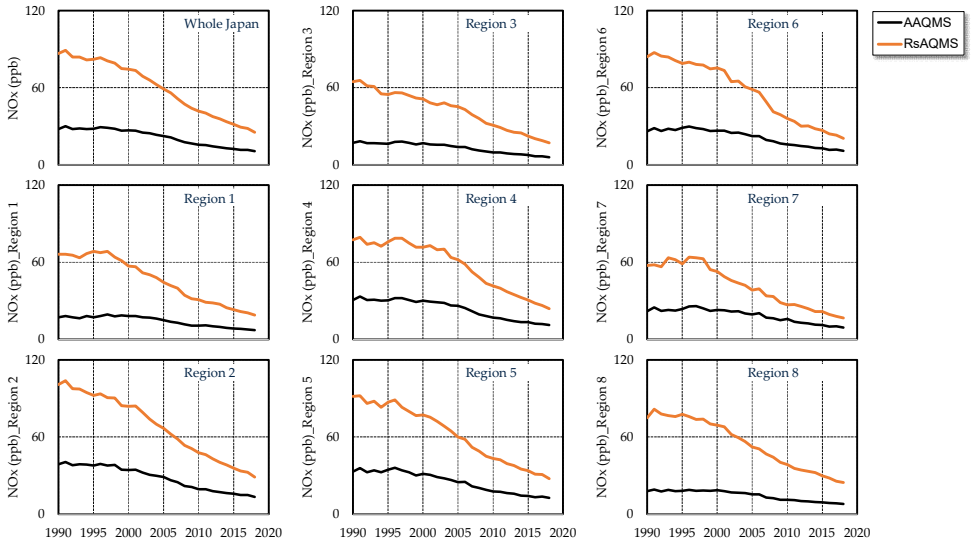

Figure S4-3 Annual average NOx concentrations at the AAQMS and RsAQMS of the nation and each region from 1990 to 2018

Table S4-3 Annual average NOx concentrations at the AAQMS and RsAQMS of the nation and each region in 1990, 2000, 2010 and 2018, and the ratio of concentrations in 2018 to 1990

NOx		Concentration (ppb)				
		1990	2000	2010	2018	C_{2018} / C_{1990}
Whole Japan	AAQMS	28	27	16	11	0.38
	RsAQMS	87	74	42	25	0.29
Region 1	AAQMS	17	18	11	7	0.42
	RsAQMS	66	57	31	19	0.29
Region 2	AAQMS	39	34	19	14	0.35
	RsAQMS	101	84	48	29	0.29
Region 3	AAQMS	17	17	10	6	0.35
	RsAQMS	65	51	31	17	0.26
Region 4	AAQMS	31	30	17	11	0.37
	RsAQMS	77	72	42	24	0.31
Region 5	AAQMS	33	31	18	13	0.38
	RsAQMS	92	77	43	27	0.30
Region 6	AAQMS	26	26	16	11	0.41
	RsAQMS	84	75	36	21	0.24
Region 7	AAQMS	22	23	16	9	0.42
	RsAQMS	58	53	27	17	0.29
Region 8	AAQMS	18	19	11	8	0.45
	RsAQMS	75	69	39	25	0.33

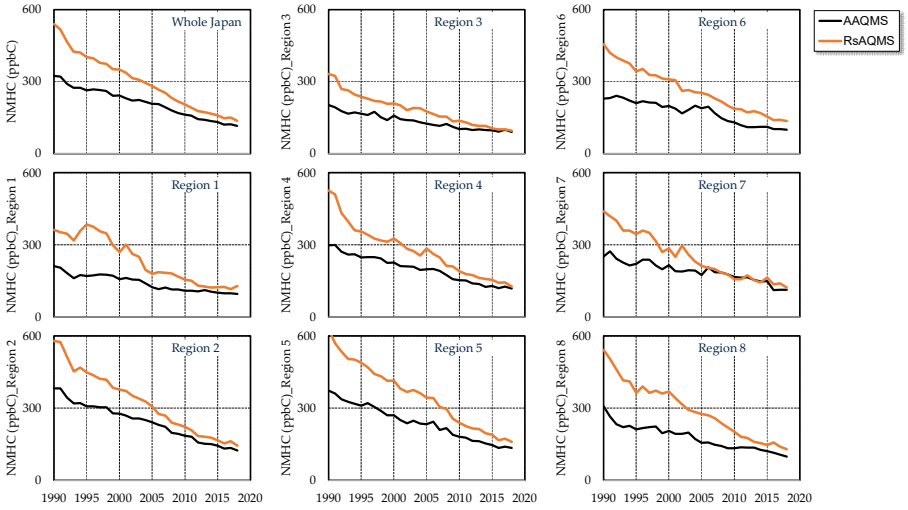

Figure S4-41 Annual average NMHC concentrations at the AAQMS and RsAQMS of the nation and each region from 1990 to 2018

Table S4-4 Annual average concentration of NMHC at the AAQMS and RsAQMS of the nation and each region in 1990, 2000, 2010 and 2018, and the ratio of concentrations in 2018 to 1990 and 2010 to 2000

NMHC		Concentration (ppbC)					
		1990	2000	2010	2018	C_{2018} / C_{1990}	C_{2018} / C_{2000}
Whole Japan	AAQMS	324	242	163	116	0.36	0.67
	RsAQMS	539	350	205	138	0.26	0.59
Region 1	AAQMS	211	158	110	96	0.45	0.69
	RsAQMS	363	270	156	129	0.36	0.58
Region 2	AAQMS	383	277	186	124	0.32	0.67
	RsAQMS	580	377	222	145	0.25	0.59
Region 3	AAQMS	203	159	103	91	0.45	0.65
	RsAQMS	333	210	138	96	0.29	0.65
Region 4	AAQMS	299	227	153	119	0.40	0.67
	RsAQMS	526	326	191	126	0.24	0.58
Region 5	AAQMS	373	271	181	135	0.36	0.67
	RsAQMS	630	414	239	159	0.25	0.58
Region 6	AAQMS	229	199	131	100	0.44	0.66
	RsAQMS	456	309	187	136	0.30	0.61
Region 7	AAQMS	252	215	166	113	0.45	0.77
	RsAQMS	440	285	157	123	0.28	0.55
Region 8	AAQMS	307	205	134	98	0.32	0.65
	RsAQMS	542	369	202	130	0.24	0.55

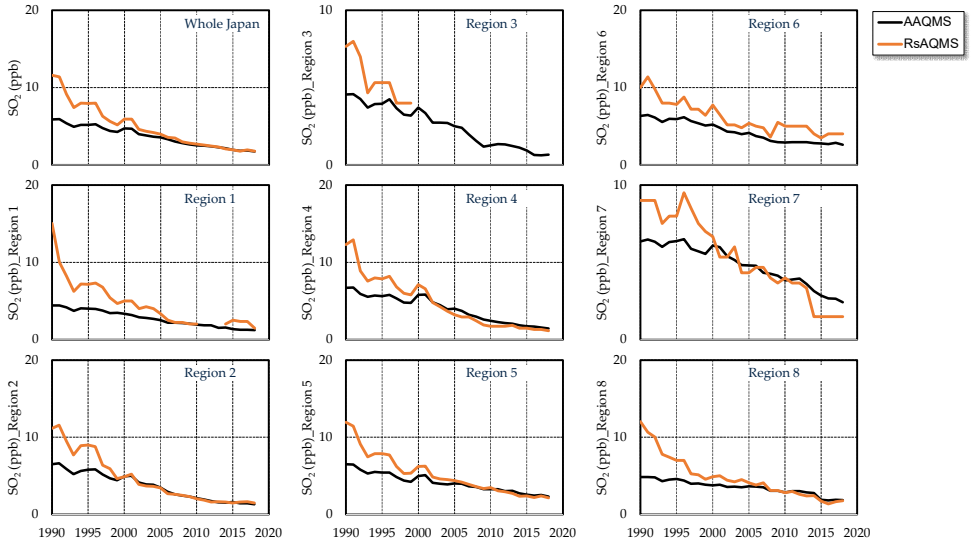

Figure 2 Annual average SO_2 concentrations at the AAQMS and RsAQMS of the nation and each region from 1990 to 2018

Table S4-5 Annual average concentration of SO_2 at the AAQMS and RsAQMS of the nation and each region in 1990, 2000, 2010 and 2018, and the ratio of concentrations in 2018 to 1990

SO_2		Concentration (ppb)				
		1990	2000	2010	2018	C_{2018} / C_{1990}
Whole Japan	AAQMS	5.9	4.7	2.5	1.7	0.29
	RsAQMS	11.6	5.9	2.7	1.8	0.15
Region 1	AAQMS	4.4	3.3	2.0	1.2	0.28
	RsAQMS	15.0	5.0	2.0	1.5	0.10
Region 2	AAQMS	6.5	4.9	2.1	1.3	0.20
	RsAQMS	11.2	4.9	2.0	1.5	0.13
Region 3	AAQMS	4.5	3.7	1.3	0.7	0.15
	RsAQMS	7.7	--	--	--	--
Region 4	AAQMS	6.7	5.8	2.4	1.4	0.22
	RsAQMS	12.3	7.1	1.7	1.2	0.09
Region 5	AAQMS	6.5	5.0	3.3	2.3	0.36
	RsAQMS	11.9	6.2	3.5	2.1	0.18
Region 6	AAQMS	6.3	5.2	2.9	2.6	0.41
	RsAQMS	10.0	7.8	5.0	4.0	0.40
Region 7	AAQMS	6.4	6.1	3.8	2.4	0.38
	RsAQMS	9.0	6.7	4.0	1.5	0.17
Region 8	AAQMS	4.8	3.8	2.8	1.9	0.38
	RsAQMS	12.0	4.9	2.8	1.8	0.15

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