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# Long-Term 30-Year Variation (1990s-2020) of Background Tropospheric Ozone Concentration at Japanese Remote Stations

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**Abstract** Tropospheric ozone (O<sub>3</sub>) is an air pollutant and greenhouse gas and is still a problem in East Asia. O<sub>3</sub> is formed via secondary photochemistry from nitrogen oxide (NO<sub>x</sub>) and volatile organic compound (VOC) precursors. Because of the increase in anthropogenic emissions globally, changes in O<sub>3</sub> concentration over the Northern Hemisphere have been a concern. In this study, using measurements at three remote sites conducted by the Japan Meteorological Agency (JMA), the 30-year (1990s to 2020) long-term trend of background O<sub>3</sub> concentration was analyzed. The results of the fitted curve revealed that background O<sub>3</sub> concentration has changed by within ±0.3 ppbv/year over 30 years. The increasing trend at Ryori (39.03°N, 141.82°E) peaked in 2009 and then decreased, agreeing well with other reports in the mid-latitude zone (40°N–55°N). In contrast, the present study revealed continuous decreasing trends at Minamitorishima (24.28°N, 153.98°E) and Yonagunijima (24.47°N, 123.02°E) at lower latitudes. In addition, the analysis for 2020 clarified that the frequency density shifted to a lower O<sub>3</sub> concentration compared with the dataset for the 1990s to 2019 at all three remote sites in Japan. This suggested that the emission reduction resulting from the COVID-19 pandemic could be a factor causing the decreasing background O<sub>3</sub> concentration.

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#### 1. Introduction

Tropospheric ozone (O<sub>3</sub>) is an air pollutant and greenhouse gas that is produced via photochemical reactions of nitrogen oxide (NO<sub>x</sub>) and volatile organic compound (VOC) precursors (Seinfeld and Pandis 2016). Because of the growing anthropogenic emissions of NO<sub>x</sub> and VOCs since the 1970s (Kurokawa and Ohara 2020; Crippa et al. 2018, 2023), O<sub>3</sub> pollution, particularly over the Northern Hemisphere, has been a major concern. Given its long lifetime, O<sub>3</sub> can be long-range transported away from source regions, for example, from one continent to another (Guerova et al. 2006; Itahashi et al. 2020; Mathur et al. 2022). Thus, O<sub>3</sub> is not only a pollutant on a local scale near intense emission sources, but also a pollutant on hemispheric scales. In this manner, measurements taken at sites least affected by local emission sources can give an indication of background O<sub>3</sub> concentration. The background O<sub>3</sub> levels appeared to have approximately doubled, with the greatest increase since the 1950s, and annual average background O<sub>3</sub> concentration over the mid-latitudes of the Northern Hemisphere has ranged from 20 to 45 ppb (Vingarzan 2004). Long-term measurement data over Europe (beginning in the 1950s and before), North America (beginning in 1984), and Asia (beginning in 1991) show that O<sub>3</sub> has increased at all sites in all seasons at approximately 1%/year relative to the level in 2000 (Parrish et al. 2012). The updated study suggested that the increasing O<sub>3</sub> trend has ended, with a maximum concentration reached in the mid-2000s, followed by slow decrease in Europe and North America (Parrish et al. 2020). The exception to this peak-out trend was found at the Asian site of Mt. Waliguan (36.28°N, 100.90°E, 3.8 km) in Qinghai Province, China.

Because long-range O<sub>3</sub> transport is a concern, the effects of the Asian continent on the downwind region of Japan have been investigated in previous studies (Itahashi et al. 2013, 2015; Chatani et al. 2020; Itahashi 2023). The background O<sub>3</sub> concentration in the early 1990s was estimated as 35–40 ppbv (Akimoto et al. 1996), although the background O<sub>3</sub> concentration in Japan was estimated as higher than the background concentration over the USA and Europe in 1980s (Sunwoo et al. 1994). The 8-year (1989–1997) ozonesonde data at Okinawa, southern Japan, showed an increasing trend of 2.5 ± 0.6%/year (Lee et al. 1998). The O<sub>3</sub> trend from 1998 to 2006 at Mt. Happo (36.70°N, 137.80°E, 1.9 km) during springtime, when dominated by the East Asian continental outflow, showed growth of +1 ppbv/year (Tanimoto 2009). The numerical model simulation also highlighted the impact of O<sub>3</sub> from outside of Japan on these increasing trends (Nagashima et al. 2017). The O<sub>3</sub> trends at Japanese monitoring sites mostly in urban areas have been analyzed from 1980 to 2015, and the analysis suggested that photochemical production and the weakening NO titration effect because of the reduction in NO<sub>x</sub> emissions increased O<sub>3</sub> concentration (Kawano et al. 2022). The development of a historical emission inventory and subsequent model simulation during 2000–2019 indicated that emission regulation

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in Japan partly led to the increase of annual mean O<sub>3</sub> because of weakening NO titration; however, the regulation suppressed high O<sub>3</sub> concentrations (Chatani et al. 2023). Although there are many previous studies discussing O<sub>3</sub> pollution status in Japan, the background O<sub>3</sub> concentration in Japan has not been well examined. In addition, O<sub>3</sub> pollution in 2020, when human activities were severely restricted by COVID-19, should be analyzed for background O<sub>3</sub> concentration because previous research focused mainly on O<sub>3</sub> variation over urban areas. Therefore, in the present work, we analyzed an O<sub>3</sub> concentration measurement dataset at three remote sites in Japan from the 1990s to 2020, and we report the long-term 30-year trend and its change in 2020.

#### 2. Dataset

The Japan Meteorological Agency (JMA) conducted the greenhouse gas measurements at three remote sites, and O<sub>3</sub> has been measured since the 1990s (JMA 2024). Figure 1 shows these sites. Minamitorishima (MNM) is an island in the North Pacific Ocean, in easternmost Japan (24.28°N, 153.98°E, 7 m above sea level (a.s.l.)), at which O<sub>3</sub> measurements started in January 1994. Yonagunijima (YON) is an island in westernmost Japan (24.47°N, 123.02°E, 30 m a.s.l.), at which O<sub>3</sub> measurements started from January 1997. Ryori (RYO) is located on a hilly cape on the Pacific coast of northeast Japan (39.03°N, 141.82°E, 260 m a.s.l.), at which O<sub>3</sub> measurements started in January 1990. All three sites are highly suitable for background O3 measurements because the local emissions are negligible (e.g., Itahashi et al. 2021, 2024). Air samples were introduced into the measuring system from the inlet on the measurement room's roof with a flow rate of 10 L/min. The inlet was 8 m from the ground at MNM and RYO, and 10 m at YON. The measuring system for O<sub>3</sub> consisted of an ultraviolet (UV) absorption monitor that measured O3 concentrations every 10 s. The hourly data were calculated by the arithmetic mean of all measured data over 1 h after removing irregular data and evident errors. The calibration for the UV

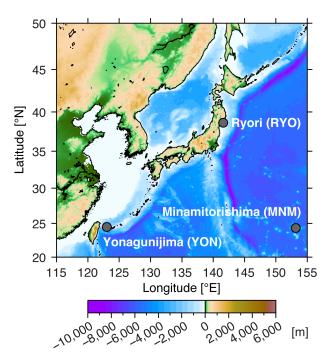


Fig. 1. Geographical map of JMA measurement stations at Minamitorishima (MNM), Yonagunijima (YON), and Ryori (RYO).

O<sub>3</sub> monitor was normally done twice a year based on the calibration system at JMA Headquarters in Tokyo. Monitoring sites with more than 6000 h (corresponding to 250 days) of measurement time are considered effective measuring sites and evaluated as meeting the Japanese environmental quality standard. In this study, we also applied this criterion for the data from MNM, YON, and RYO. The data from RYO in 1990 and 1991 were excluded from the analysis based on this criterion.

#### 3. Results and discussion

All long-term background O<sub>3</sub> concentrations at MNM, YON, and RYO are shown in Fig. 2. At MNM (Fig. 2a), annual mean O<sub>3</sub> concentrations were in the range of 25–35 ppbv, and the median was generally lower than annual mean. The maximum values exceeded 70 ppbv in some years, even at a remote site in the northwest Pacific Ocean. At YON (Fig. 2b), annual mean concentrations were around 40 ppbv, and the median was greater than annual mean. The maximum value exceeded 100 ppbv, which could be affected by the outflow from the Asian continent (i.e., Itahashi et al. 2024). At RYO (Fig. 2c), the annual mean and the median were close and these values were in the range of 35–40 ppbv during the analyzed period.

These datasets were decomposed into the monthly and annual mean data, and then the long-term trends were analyzed following the approach described by Parrish et al. (2019, 2020) for the monthly mean. In this analysis,  $O_3$  concentration ( $[O_3]$ ) is expressed as the least-squares fits of Eq. (1).

$$[O_3] = a + bt + ct^2 + residuals$$
 (1)

Parameters a, b, and c in the power series are the coefficients that quantify the  $O_3$  concentration trend. Time origin t for this quadratic polynomial was chosen as the year 2000 (i.e., t = 2000 in Eq. (1)); therefore, coefficient a was the intercept of the fitted curve at 2000. Coefficient b was the slope of the fitted curve, quantifying the rate of change of  $O_3$  concentration over time at the beginning of the year 2000. Finally, coefficient c was equal to one-half of the time rate of

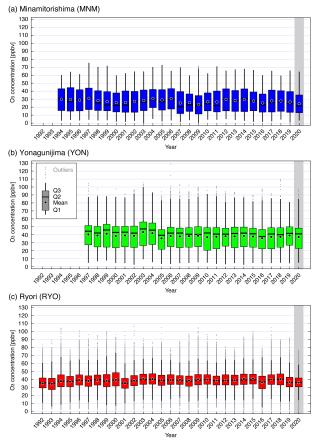


Fig. 2. Timeseries of all hourly measurement data during the analyzed period as box plots at (a) Minamitorishima, (b) Yonagunijima, and (c) Ryori. The lower and upper fences in the box plot are the 25<sup>th</sup> (Q1) and the 75<sup>th</sup> (Q3) percentiles and the thick line in the box plot is the 50<sup>th</sup> percentile (median) (Q2). The whiskers are defined as the minimum (Q1 – 1.5 interquartile range (IQR)) and the maximum (Q3 + 1.5 IQR) values where the IQR is Q3–Q1 and the gray dots are outliers of data beyond the minimum and maximum. The circle is the arithmetic mean. The gray shading indicates the year of the COVID-19 pandemic.

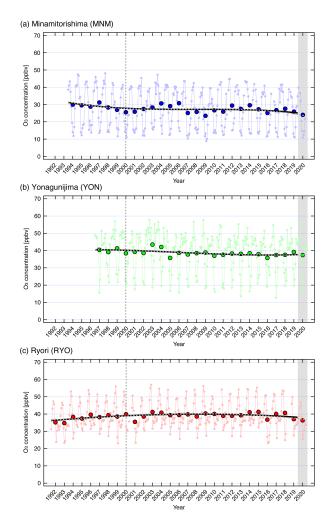


Fig. 3. Timeseries of monthly and annual mean  $O_3$  at (a) Minamitorishima, (b) Yonagunijima, and (c) Ryori. The solid black curve represents the nonlinear, least squares regression fits of a quadratic polynomial to the monthly data from the 1990s to 2019 and the dotted gray curve is the result including the data in the year 2020. The gray shading indicates the year of the COVID-19 pandemic.

change of the fitted curve slope, indicating the acceleration or deceleration of the trend. The monthly and annual mean and the fitted curve for the monthly mean derived based on Eq. (1) are shown in Fig. 3, and the statistical results for the three parameters are listed in Table 1. Because of the larger variation in MNM and YON compared with RYO, the significance level of p < 0.05 was obtained only at RYO in this fitting curve analysis. The a values were 28.0, 40.3, and 38.9 ppbv at MNM, YON, and RYO, respectively (Table 1). The timeseries in Fig. 2 showed that O<sub>3</sub> concentration was lowest at MNM in the Pacific Ocean, whereas those at YON and RYO were higher concentration in 2000. The Pacific marine boundary layer on the US west coast had an a value of  $32.9 \pm 1.1$  ppbv (Table 1 and detailed in Parrish et al. 2020), and the results at MNM in this study were similar. Moreover, the a value at Mace Head, Ireland was around 40 ppbv from the 1980s to the 2010s (Table 1 and detailed in Derwent et al. 2018 and Parrish et al. 2020), mostly consistent with those at YON and RYO in this study. The background O<sub>3</sub> concentration observed around Japan corresponded well to other reported values (e.g., Parrish et al. 2020). Coefficient b was negative at MNM and YON, indicating the decreasing trend for O<sub>3</sub>, whereas b was positive at RYO. Negative slopes are not found at most observation sites in the Northern Hemisphere (Parrish et al. 2020), and this was a unique trend revealed in this study. One possible reason is the difference in latitude; the observation sites reported by Parrish et al. (2020) were at 40°N-55°N, whereas MNM and YON in this study were at 25°N. Although these results at MNM and YON were not significant (p > 0.05), the behavior at different latitudes in the Northern Hemisphere should be studied further. At RYO, which is at the same latitude as the site in the report by Parrish et al. (2020), there was an increasing trend for O<sub>3</sub>. Based on Eq. (1), the year at which the maximum of the fitted curve occurred ( $year_{max}$ ) is obtained as follows (Parrish et al. 2019, 2020).

Site	a (ppbv)	<i>b</i> (ppbv/year)	$(\times 10^{-3} \text{ ppbv/year}^2)$	year <sub>max</sub>	Reference
Minamitorishima (MNM)	$28.0 \pm 1.1$ $(27.8 \pm 1.1)*$	$-0.28 \pm 0.21$ $(-0.28 \pm 0.21)$ *	+35 ± 42 (+44 ± 39)*	_	This study
Yonagunijima (YON)	$40.3 \pm 1.2$ $(40.2 \pm 1.2)$	$-0.15 \pm 0.50$ (-0.17 ± 0.48)	$-12 \pm 76$ $(-8 \pm 69)$	_	This study
Ryori (RYO)	$38.9 \pm 0.8 *$ $(38.8 \pm 0.7) *$	+0.24 ± 0.10* (+0.25 ± 0.10)*	-12 ± 19* (-8 ± 18)*	$2009.5 \pm 7.3$	This study
Pacific marine boundary layer	$32.9 \pm 1.1$	$+0.14 \pm 0.10$	$-25 \pm 11$	$2002.8 \pm 2.2$	Parrish et al. (2020)
Mace Head, Ireland	$40.1 \pm 0.8$ $39.8 \pm 0.6$	$+0.34 \pm 0.07$ $+0.35 \pm 0.06$	$-23 \pm 8$ $-20 \pm 6$	$2007 \pm 6 \\ 2008.8 \pm 3.2$	Derwent et al. (2018) Parrish et al. (2020)
Mt. Waliguan, China	$49.5 \pm 0.4$	$+0.19 \pm 0.05$	+3 ± 7	_	Parrish et al. (2020)

Table 1. Three parameters determined by the fitted curve at three remote sites in Japan and other sites taken from the previous studies.

Note: The parentheses indicate the result of curve fitting including the data from 2020. Significance level of p < 0.05 for the curve fitting is shown by \*.  $year_{max}$  is not derived and shown by – because the value of c is positive at Minamitororishima (MNM) and the values of both b and c are negative at Yonagunijima (YON).

Table 2. Top three lowest values of the observed background O<sub>3</sub> concentration at three remote sites in Japan from the 1990s to 2020.

Site	Statistical metrics	1st	2nd	3rd
Minamitorishima (MNM)	75 <sup>th</sup> percentile (Q3)	34.50 (2009)	35.25 (2020)	36.50 (2016)
	50 <sup>th</sup> percentile (Q2)	20.50 (2007)	21.50 (2020)	21.90 (2011)
	25 <sup>th</sup> percentile (Q1)	11.40 (2009)	11.90 (2007)	13.00 (2020)
	Mean	23.56 (2009)	24.31 (2020)	25.26 (2007)
Yonagunijima (YON)	75 <sup>th</sup> percentile (Q3)	48.30 (2020)	48.50 (2015, 2016)	48.60 (2014)
	50 <sup>th</sup> percentile (Q2)	40.60 (2016)	40.70 (2015)	40.80 (2014)
	25 <sup>th</sup> percentile (Q1)	22.90 (2020)	23.80 (2005, 2010, 2016)	23.90 (2007, 2008, 2009, 2015)
	Mean	percentile (Q2) 20.50 (2007) 21.50 (2020) percentile (Q1) 11.40 (2009) 11.90 (2007) 23.56 (2009) 24.31 (2020) percentile (Q3) 48.30 (2020) 48.50 (2015, 2016) percentile (Q2) 40.60 (2016) 40.70 (2015) percentile (Q1) 22.90 (2020) 23.80 (2005, 2010, 2016) percentile (Q3) 41.60 (2001) 41.70 (1993) percentile (Q2) 35.50 (2001) 35.60 (1993) percentile (Q1) 27.60 (1993) 28.70 (2016)	37.89 (2010)	
Ryori (RYO)	75 <sup>th</sup> percentile (Q3)	41.60 (2001)	41.70 (1993)	<b>42.10</b> (1992, <b>2020</b> )
	50 <sup>th</sup> percentile (Q2)	35.50 (2001)	35.60 (1993)	36.00 (1992)
	25 <sup>th</sup> percentile (Q1)	27.60 (1993)	28.70 (2016)	29.20 (2001)
	Mean		35.30 (1992)	35.56 (2001)

Note: The parentheses indicate the corresponding year and all units for O<sub>3</sub> concentration are ppbv. The year 2020 is shown in bold font.

$$year_{\text{max}} = -b/2c + 2000 \tag{2}$$

At RYO, where there was an increasing trend for  $O_3$ ,  $year_{max}$  estimated from Eq. (2) was  $2009.5 \pm 7.3$ . This peak in the mid-2000s at RYO calculated here agreed well with the results from over the Northern Hemisphere (Table 1 and detailed in Parrish et al. 2020). In Fig. 2, the analysis including 2020 are also overlayed, and the results for the fitted curve are also listed in Table 1. The spread of COVID-19 strongly affected the air quality in 2020 (Liu et al. 2020; Wang et al. 2021; Itahashi et al. 2022). The results for the three parameters were affected by including the year 2020, and the decrease in the values of parameters a and b suggested a further decreasing trend for  $O_3$ .

Finally, we focus on the change in O<sub>3</sub> concentration in 2020 during COVID-19. O<sub>3</sub> concentration at three remote sites showed year-to-year variation (Figs. 2 and 3). Table 2 lists the top three lowest values of the statistical metrics (75<sup>th</sup>, 50<sup>th</sup>, and 25<sup>th</sup> percentiles and arithmetic mean) at each site. Although lower O<sub>3</sub> concentrations occurred in different years at each site, 2020 showed lower concentrations at all three sites (RYO remarked 4<sup>th</sup> or 5<sup>th</sup> lowest concentration, see also Table 3). This result suggested that there was a common factor causing the lower concentration in 2020.

From the emission inventory estimation, anthropogenic NO<sub>x</sub> emissions in 2020 declined 30%–40% (greater than anthropogenic VOC emissions decline) during the lockdown period (Forster et al. 2020; Zheng et al. 2021). Thus, O<sub>3</sub> concentrations in urban areas generally increased because of the weakening of O<sub>3</sub> titration by large NO<sub>x</sub> emission reductions based on a literature review of urban air pollution during the COVID-19 lockdown globally (Gkatzelis et al. 2021). Similarly, increased O<sub>3</sub> concentrations were reported in urban areas (e.g., Tokyo metropolitan area) of Japan (Damiani et al. 2022, 2024). In contrast to these features over urban areas, the background O<sub>3</sub> concentration at Mace Head, Ireland decreased more than concentrations derived considering recent long-term decreasing trends (Parrish et al.

Site	Statistical metrics	O <sub>3</sub> concentration averaged over 1990s to 2019 (ppbv)	O <sub>3</sub> concentration in 2020 (ppbv)
	75 <sup>th</sup> percentile (Q3)	40.80	35.25 (-5.55)
Minimizer of the CARDAR	50 <sup>th</sup> percentile (Q2)	25.90	21.50 (-4.40)
Minamitorishima (MNM)	25 <sup>th</sup> percentile (Q1)	14.80	13.00 (-1.80)
	Mean	27.76	24.31 (-3.45)
	75 <sup>th</sup> percentile (Q3)	50.20	48.30 (-1.90)
V (VON)	50 <sup>th</sup> percentile (Q2)	41.90	41.20 (-0.70)
Yonagunijima (YON)	25 <sup>th</sup> percentile (Q1)	24.40	22.90 (-1.50)
	Mean	38.68	37.29 (-1.39)
	75 <sup>th</sup> percentile (Q3)	45.50	42.10 (-3.40)
Promi (BVO)	50 <sup>th</sup> percentile (Q2)	38.70	36.60 (-2.10)
Ryori (RYO)	25 <sup>th</sup> percentile (Q1)	32.50	31.00 (-1.50)
	Mean	38.90	36.31 (-2.59)

Table 3. Statistical metrics of observed background  $O_3$  concentration at three remote sites in Japan from the 1990s to 2019 and for 2020.

Note: Parentheses indicate the difference between 2020 and from the 1990s to 2019.

2022). This finding was consistent with our analyses using the fitting curve shown in Fig. 3. The global total tropospheric O<sub>3</sub> burden decreased dramatically by about 2% in May and June 2020 (Miyazaki et al. 2021). Based on these results, we further analyzed the frequency density of O<sub>3</sub> concentration before COVID-19 (1990s to 2019) and in 2020 (Fig. 4). The frequency density shifted to a lower concentration in 2020 compared with the 1990s to 2019. The statistical metrics (75th, 50th, and 25<sup>th</sup> percentiles and arithmetic mean) are listed in Table 3. All metrics showed a decrease in 2020 compared with the 1990s to 2019 (Fig. 2), and the changes were greater in the higher concentration range (75th percentile rather than 25th percentile). The analysis of 2020 clarified the decrease in background O3 concentration at remote sites in Japan. The decrease in O<sub>3</sub> concentration could be partly within the year-to-year variation; however, one possible factor causing this decreasing background O<sub>3</sub> concentration is the unprecedented anthropogenic emission reduction resulting from the COVID-19 pandemic in 2020.

#### 4. Conclusion

The long-term near-30-year trend of background  $O_3$  concentration at three remote sites in Japan was analyzed based on a quadratic polynomial fitted curve. The fitted curve results revealed that background  $O_3$  concentration at these remote sites has been changing by  $\pm 0.3$  ppbv/year over the 30 years from the 1990s to 2020. Our study revealed that MNM (24.28°N, 153.98°E) in easternmost Japan and YON (24.47°N, 123.02°E) in westernmost Japan showed a decreasing trend for  $O_3$ . RYO (39.03°N, 141.82°E) in northeast Japan showed an increasing trend for  $O_3$ , with its peak in 2009 and a subsequent decrease. This peak-out  $O_3$  trend was consistent with other analyses of the mid-latitudes in the Northern Hemisphere (40°N–55°N). The unique continuously decreasing background  $O_3$  concentration at a latitude of 20°N should

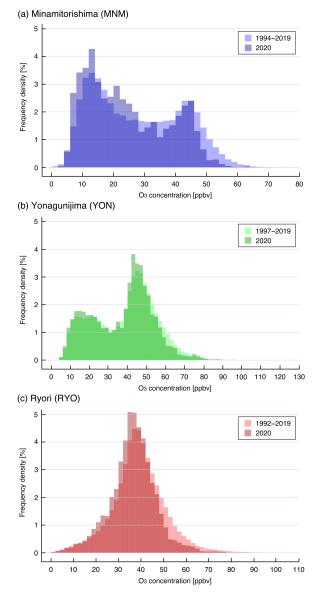


Fig. 4. Frequency density diagram of O<sub>3</sub> concentration at (a) Minamitorishima, (b) Yonagunijima, and (c) Ryori. The light colors indicate the data from the 1990s to 2019 and the dark colors indicate the data for 2020.

be studied further with the available dataset. Detailed analyses for 2020 clarified that background O<sub>3</sub> concentration decreased in all concentration ranges at all three remote sites in Japan, possibly because of the anthropogenic emission reduction owing to the COVID-19 pandemic.

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