Reply to Editor

Comment from the editor

Editor Decision: Publish subject to minor revisions (further review by editor) (02 Jul 2020) by Sam Illingworth

Comments to the Author:

Thank you for engaging so throughly with the peer review process, and my apologies for the length of time that this has taken, due to circumstances that were sadly beyond our control. Having now throughly reviewed the manuscript and the referee' comments, we will be delighted to publish this work in Geoscience Communication, once a few small edits have been made, as requested by Referee 2. The referee report from Referee 2 contains several suggestions to improve the readability of the paper, please could address all of these and upload a revised manuscript. Once this has been done I will ensure that I expedite the final editorial review.

Thank you again for your continued patience and professionalism, and I look forward to seeing the updated manuscript.

Many Thanks,

Sam

Author's response

Dear Prof. Illingworth

We greatly appreciate your great effort to improve our manuscript. We answered all comments from the Referee 2 point-by-point as follows. We hope that our manuscript is suitable for publication in GC.

Sincerely yours,

Tomohiro Sato, Yasuko Kasai

National Institute of Information and Communications Technology

Point-By-Point Reply to Report #2 from Anonymous Referee #4

Comment from Referee:

A. General comments from Referee:

This is an interesting study in terms of defining a new straightforward and simple methodology to calculate an index for assessing clean air (CII), which takes account of the four main pollutants. The authors use a model of air pollution over Japan and an alternative index to assess which better matches ground measurements. The CII can also be used in a relative manner by changing the normalisation factor to account for each country's legal framework. The authors apply the CII to the pollution in Japan over a number of years to provide maps and statistics of air quality in the region.

Please see the attached pdf for comments and corrections.

Referee Report: gc-2019-16-referee-report.pdf

Author's response:

Thank you so much for your quite careful reading. We greatly appreciate your efforts to help us improve our manuscript. We answered your valuable comments point by point as follows. We are sure that our manuscript could be more readable and understandable than the previous one. We hope that our manuscript is suitable for publication in GC.

Comment from Referee:

Title: cleanliness is better than cleanness e.g. see

https://www.learnersdictionary.com/qa/cleanliness-and-cleanness

Author's response:

We appreciate your comment. We changed all terms "cleanness" to "cleanliness" following your suggestion.

Author's changes in the manuscript:

Title, Page 1 Line 2, 3, 12, 13, 24, Page 2 Line 33, 47, 48, 49, 56, 57, Page 3 Line 74, Page 11 Line 213, 214, Page 13 Line 222, 223, Page 16 Line 257, 259, 260, Page 18 Line 285, 287, 288, 290, Page 19 Line 304, 315, Page 20 Line 322, 328

Comment from Referee:

P1 Line 15: what do you mean by averaging?

Author's response:

Thank you for pointing it out. It means that the mean of correlation coefficients for 498 municipalities was 0.66±0.05. We improved the sentence as follows.

Author's changes in the manuscript:

Page 1 Lines 15 – 18

"The CII values calculated by the WRF-CMAQ model and the AEROS measurements showed good agreement with a correlation coefficient of 0.66±0.05, averaging 498 municipalities where the AEROS measurements have operated, which was higher than that of Air Quality Index (AQI) of 0.57±0.06."

→

"The CII values calculated by the WRF-CMAQ model and the AEROS measurements showed good agreement. The mean of correlation coefficient for the CII values of 498 municipalities where the AEROS measurements have operated was 0.66±0.05, which was higher than that of Air Quality Index (AQI) of 0.57±0.06."

Comment from Referee:

Page 2 Line 30: assuming that the energy comes from fossil fuels?

Author's response:

Yes, the combustion of fossil fuels is one of the largest sources of air pollutants emission. We improved the sentence as follows.

Author's changes in the manuscript:

Page 2 Line 30

"power generation" → "power generation using fossil fuels"

Comment from Referee:

Page 10 Figure 4: Do you mean frequency?

Page 10 Figure 4: I don't understand the difference between the histograms

Author's response:

Thank you for pointing it out. We showed the histograms by the probability density function, not

by the frequency, in the previous manuscript. But we changed Figures 4 and 5 to the histograms of the frequency following your suggestion because the frequency is more popular than the probability density function as far as we surveyed.

The difference between the panels (a) and (b) in Fig. 4 is that the panel (a) shows the spatial bias of the CII difference between WRF-CMAQ and AEROS, and the panel (b) shows the temporal bias. We improved the statements and Figure 4 as follows.

Author's changes in the manuscript:

Page 9 Lines 184 - 186

"We compared the CII mean of all days in the study period between WRF-CMAQ and AEROS for each municipality to investigate the spatial bias in Fig. 4 (a)."

→

"We compared the CII mean of all days in the study period between WRF-CMAQ and AEROS for each municipality. The CII difference for each municipality is shown in Fig. 4 (a) to investigate the spatial bias."
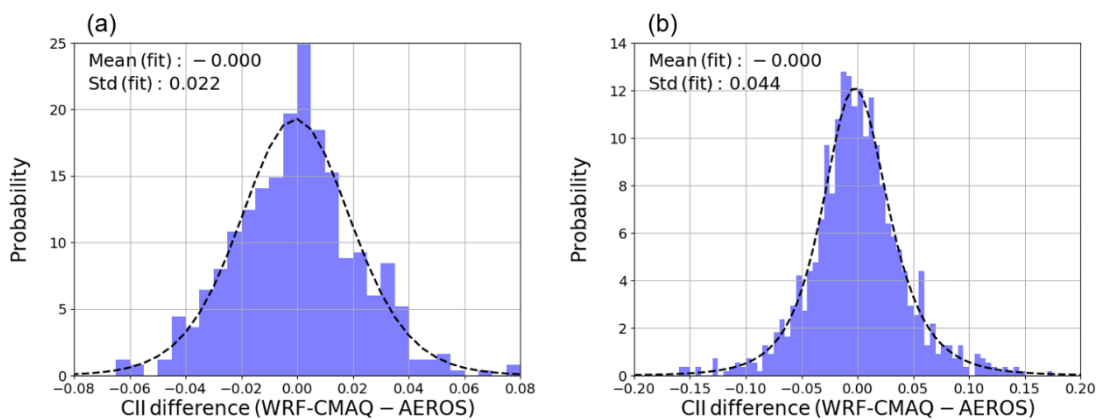
Page 9 Lines 189 – 191

"In the similar way, we investigated the daily temporal bias by comparing the CII mean of all Japanese municipalities for each day."
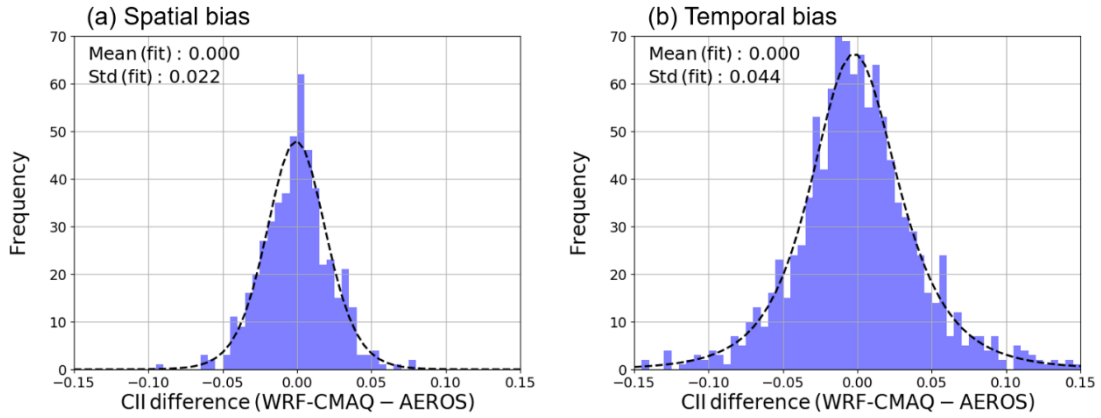
→

"In the similar way, we compared the CII mean of all Japanese municipalities for each day during the study period to investigate the daily temporal bias. Figure 4 (b) shows the histogram of the CII difference for each day."
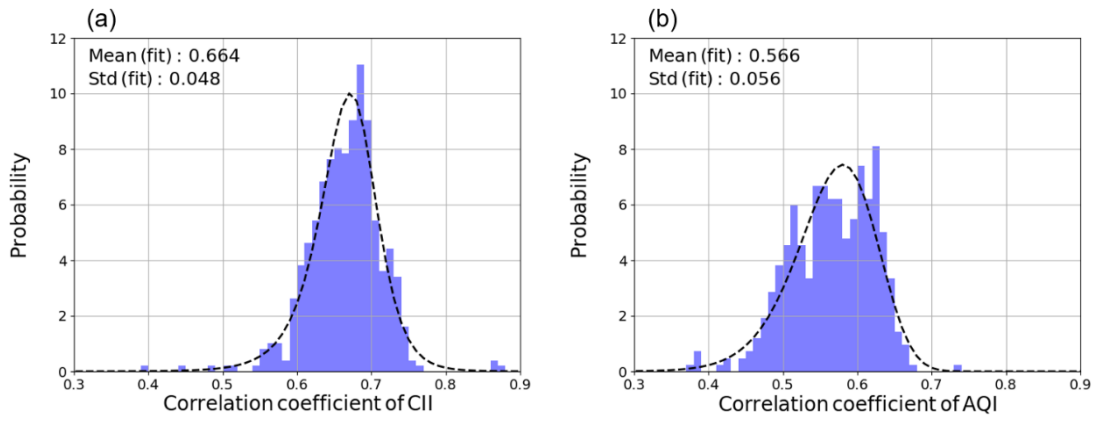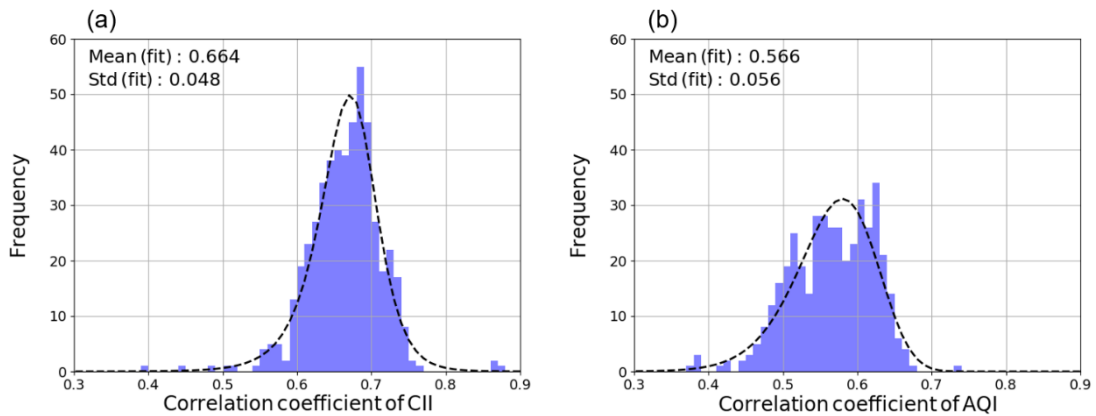
Page 10 Figure 4

→

(a) Spatial bias

Mean (fit) : 0.000
Std (fit) : 0.022

(b) Temporal bias

Mean (fit) : 0.000
Std (fit) : 0.044

Page 11 Figure 5

(a)

Mean (fit) : 0.664
Std (fit) : 0.048

(b)

Mean (fit) : 0.566
Std (fit) : 0.056

→

(a)

Mean (fit) : 0.664
Std (fit) : 0.048

(b)

Mean (fit) : 0.566
Std (fit) : 0.056

Comment from Referee:

Page 10 Lines 197 – 198: what other indices?

Author's response:

We appreciate you comment. We regarded AQI as representative of indices to evaluate the air quality. We improved the statement as follows.

Author's changes in the manuscript:
Page 10 Lines 197 – 198
"the other indices" → "indices to evaluate the air quality"

Comment from Referee:
Page 16 Line 260: We group them into four categories:

Author's response:
Thank you for pointing it out. We followed your suggestion.

Author's changes in the manuscript:
Page 16 Line 260
", i.e.," → ". We group them into four categories:"

Other corrections from Referee:
Referee Report: gc-2019-16-referee-report.pdf

Author's response:
We deeply appreciate your careful reading and many corrections. We improved our manuscript as follows.

Author's changes in the manuscript:
Page 1 Lines 2 – 3: "deserves a valuable" → "is as valuable a"
Page 1 Line 3: "Global" → "A global"
Page 1 Line 5: Add "a set of"
Page 1 Line 10: "obvious" → "fixed"
Page 1 Line 11: Remove "namely", "given" → ", as directed"
Page 1 Line 12: Add "data from"
Page 2 Line 53: "obvious" → "fixed"
Page 6 Line 135: "used CMAQ model had" → "CMAQ model used"
Page 6 Line 152: "is" → "are"

Page 10 Caption of Fig. 4: "Their" → "The"

Page 10 Line 197: Add "the"

Page 10 Line 199: "The correlation coefficient (r) of mean" → "The mean of correlation coefficient (r)"

Page 10 Line 204: "pollutant that causes" → "pollutant that cause", "is" → "are"

Page 10 Line 205: "all of" → "four"

Page 11 Line 212: "with" → "by"

Page 14 Line 235: Add "all", Remove "selected"

Page 14 Line 241: "few" → "little"

Page 18 Line 276: "save" → "have"

Page 19 Table 6: "-0.000" → "0.000"

Page 19 Line 299: "The correlation coefficient (r) of mean" → "The mean correlation coefficient (r)"

Page 19 Lines 302 – 303: "all individuals" → "the individual pollutants"

Page 19 Line 306: Remove "amounts of"

Page 19 Line 313: Add "the"

Page 19 Lines 313 – 314: "was" → "were"

Page 19 Line 314: "CII to" → "how CII could"

Page 19 Lines 315 – 316: ". Population density was used to quantify human activities in this study" → "using population density"

Page 20 Line 319: "normalized" → "weighted"

Page 20 Line 329: "a" → "the"

Novel index to comprehensively evaluate air ~~cleanness~~cleanliness: the "Clean aIr Index"

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Abstract. Air quality on our planet has been changing in particular since the industrial revolution (1750s) because of anthropogenic emissions. It is becoming increasingly important to visualize air ~~cleanness~~cleanliness, since clean air ~~deserves a valuable~~ is as valuable a resource as clean water. ~~Global~~ A global standard to quantify the level of air ~~cleanness~~ cleanliness is swiftly required, and we defined a novel concept, namely "Clean aIr Index, CII." The CII is a simple index defined by the normalization of the amount of a set of individual air pollutants. A CII value of 1 indicates completely clean air (no air pollutants), and 0 indicates the presence of air pollutants up to numerical environmental criteria for the normalization. In this time, the air pollutants used in the CII were taken from the Air Quality Guidelines (AQG) set by the World Health Organization (WHO), namely O_3, particulate matters, NO_2 and SO_2. We chose Japan as a study area to evaluate CII because of the following reasons: i) accurate validation data, as the in situ observation sites of the Atmospheric Environmental Regional Observation System (AEROS) provide highly accurate values of air pollutant amounts, ii) ~~obvious~~ fixed numerical criteria, ~~namely~~ the Japanese Environmental Quality Standards~~given~~, as directed by the Ministry of the Environment (MOE). We quantified air ~~cleanness~~ cleanliness in terms of the CII for the all 1896 municipalities in Japan, and used data from Seoul and Beijing to evaluate Japanese air ~~cleanness~~cleanliness. The amount of each air pollutant was calculated using a model that combined the Weather Research and Forecasting (WRF) and Community Multiscale Air Quality (CMAQ) models for 1 April 2014 to 31 March 2017. The CII values calculated by the WRF-CMAQ model and the AEROS measurements showed good agreement~~with a correlation coefficient of 0.66±0.05, averaging~~. The mean of correlation coefficient for the CII values of 498 municipalities where the AEROS measurements have operated was 0.66±0.05, which was higher than that of Air Quality Index (AQI) of 0.57±0.06. The CII values averaged for the study period was 0.67, 0.52 and 0.24 in Tokyo, Seoul and Beijing, respectively, thus, the air in Tokyo was 1.5 and 2.3 times cleaner, i.e., less amounts of air pollutants, than those in Seoul and Beijing, respectively. The average CII value for the all Japanese municipalities was 0.72 over the study period. The extremely clean air, CII ≈ 0.90, occurred in southern remote islands of Tokyo and around west of the Pacific coast, i.e., Kochi, Mie and Wakayama Prefectures during summer with transport of clean air from the ocean. We presented "Top 100 clean air cities" in Japan as one example of application using CII in society. We confirmed that the CII enabled the quantitative evaluation of air ~~cleanness~~cleanliness. The CII can be useful value in various scenarios, such as encouraging sightseeing and migration,

25 investment and insurance company business, and city planning. The CII is a simple and fair index that can be applied to all nations.

1 Introduction

Air is an essential components for all life on our planet. Air quality has been changing since the industrial revolution (1750s). According to the report from OECD (2016), air pollutant emissions are predicted to increase because of the projected increase
30 in the energy demand, e.g., transportation and power generation using fossil fuels, especially in East Asia. This report also mentions that the global annual market costs are predicted to increase from 0.3 % in 2015 to 1.0 % in 2060 of global GDP because of reduced labor productivity, increased health expenditures, and crop yield losses due to air pollution.

A global standard index to quantify air ~~cleanness~~ cleanliness should be developed as the Global Drinking Water Quality Index (GDWQI), for water quality, defined by UNEP (2007), since clean air is as valuable a resource as clean water is. Such an
35 index can be a useful communication tool to help decision making. The index should be understandable/informative not only for scientific experts but also general citizen, and also be upgraded with the scientific data.

Several indices exist for estimating air quality, e.g., Air Quality Index (AQI) in the United States (US EPA, 2006) and Air Quality Health Index in Canada (Stieb et al., 2008) and Hong Kong (Wong et al., 2013). The purpose of these indices is to estimate health risks due to air pollution exposure. These indices were developed based on epidemiological studies and
40 optimized for each country or local area. The most commonly used index is the US AQI (US EPA, 2006). The AQI ranges from 0 to 500 and is calculated based on the concentrations of the six air pollutants. In the calculation of AQI, an individual AQI for every air pollutants are calculated for a given location on a given day, and the maximum of all individual AQIs is defined as the overall AQI. Hu et al. (2015) performed a comparison study of several indices for air quality using the measurements in China, and showed AQI underestimates the severity of the health risk associated with the exposure to multi-pollutant air
45 pollution because AQI does not appropriately represent the combined effects of exposure to multiple pollutants. An index to quantify the air quality is still under development, and the global standard has not been established yet.

In this study, we propose a novel concept of index to quantify air ~~cleanness~~ cleanliness, "Clean aIr Index (CII)" to establish the global standard for quantifying air ~~cleanness~~ cleanliness. The purpose of CII is to comprehensively evaluate air ~~cleanness~~ cleanliness by normalizing the amounts of common air pollutants with numerical environmental criteria. In this time, we
50 selected surface O_3, particulate matter (PM), NO_2, and SO_2 from the Air Quality Guidelines (AQG) set by the World Health Organization (WHO)(WHO, 2005). As a first approach, we chose Japan for evaluating the CII because of i) the validation data, as the in situ observation sites of the Atmospheric Environmental Regional Observation System (AEROS) provide highly accurate air pollutant amounts, and ii) the ~~obvious~~ fixed numerical criteria, i.e., the Japanese Environmental Quality Standards given by the Ministry of the Environment (MOE).
55 In this paper, Sect. 2 defines the CII. Section 3 describes the model for calculating the CII for all Japanese municipalities, and validates the CII values by comparing with those derived from AEROS measurements. In Sect. 4, air ~~cleanness~~ cleanliness in each municipality is quantified and the area and season of high air ~~cleanness~~ cleanliness in Japan is identified using the CII.

Table 1. Value of numerical criteria (s), O_3, suspended particulate matter (SPM), NO_2, and SO_2 used in this study. We used the criteria of the Japanese Environmental Quality Standards (JEQS) given by the Ministry of the Environment (MOE) of Japan. Average of air pollutant amount calculated by the model for all Japanese municipalities over the study period is shown.

Air pollutant	Average of model	Numerical criteria (s)	Notes
O_3	46.4 ppb	60 ppb	Threshold of the hourly values
SPM	13.5 μg/m^3	100 μg/m^3	Threshold of the daily average for hourly values
NO_2	10.5 ppb	60 ppb	Threshold of the daily average for hourly values
SO_2	1.9 ppb	40 ppb	Threshold of the daily average for hourly values

2 Clean aIr Index (CII)

The CII is a simple index defined by the normalization of each air pollutant amount. The definition of CII is given by

$$\text{CII} = f(x, s) = 1 - \frac{1}{N} \sum_{i}^{N} \frac{x[i]}{s[i]}, \tag{1}$$

where $x[i]$ is the amount of ith air pollutant, $s[i]$ is the numerical criteria for the normalization of $x[i]$, and N is the number of air pollutants considered in the CII. In this equation, a higher CII value indicates cleaner air, with a maximum of 1 indicating the absence of air pollutants. The CII value decreases as the amount of air pollutant increases, with a value of 0 indicating that the amount of air pollutant is equal to the numerical criteria and a negative value indicating that the amount of air pollutant is larger than the numerical criteria.

In this study, the air pollutants used in the CII are O_3, PM, NO_2 and SO_2 following the WHO AQG (WHO, 2005) as mentioned above, i.e., N = 4. The field of this study is Japan, thus, we set the values of s according to the Japanese Environmental Quality Standards (JEQS), which are given by the Ministry of the Environment (MOE) of Japan (Table 1). The time length should be consistent between the x and s values to implement the air pollutant amount in the calculation of CII. In this case, the s value of O_3 is defined as a threshold for 1-hour average, and those of the others are defined as 24-hour average. We employed the maximum of 1-hour average per day for O_3 and daily-mean for the other pollutants. We used the criterion for photochemical oxidants (Ox) in the JEQS as the s value for O_3, because more than 90–95 % of Ox is composed of O_3 (Akimoto, 2017). The CII can be used both globally and locally by defining the setting of s values. In case of applying the CII to compare the air ~~cleanness~~ cleanliness globally, the numerical criteria should be given by the WHO AQG (WHO, 2005).

The selected air pollutants have been of importance for the last 5 decades in Japan, and have been monitored by AEROS from 1970. Surface O_3, which is harmful to human health (e.g., Liu et al., 2013) and crop yields and quality (e.g., Feng et al., 2015; Miao et al., 2017), has been increasing in Japan since the 1980s in spite of the decreasing O_3 precursors, such as NO_X and volatile organic compounds (VOCs) (Akimoto et al., 2015). Nagashima et al. (2017) estimated that the source of surface O_3 is increasing, and approximately 50 % of the total increase was caused by transboundary pollution from China and Korea. We used the suspended particulate matter (SPM) for PM following the JEQS. NO_2 is a precursor of surface O_3 and is a harmful
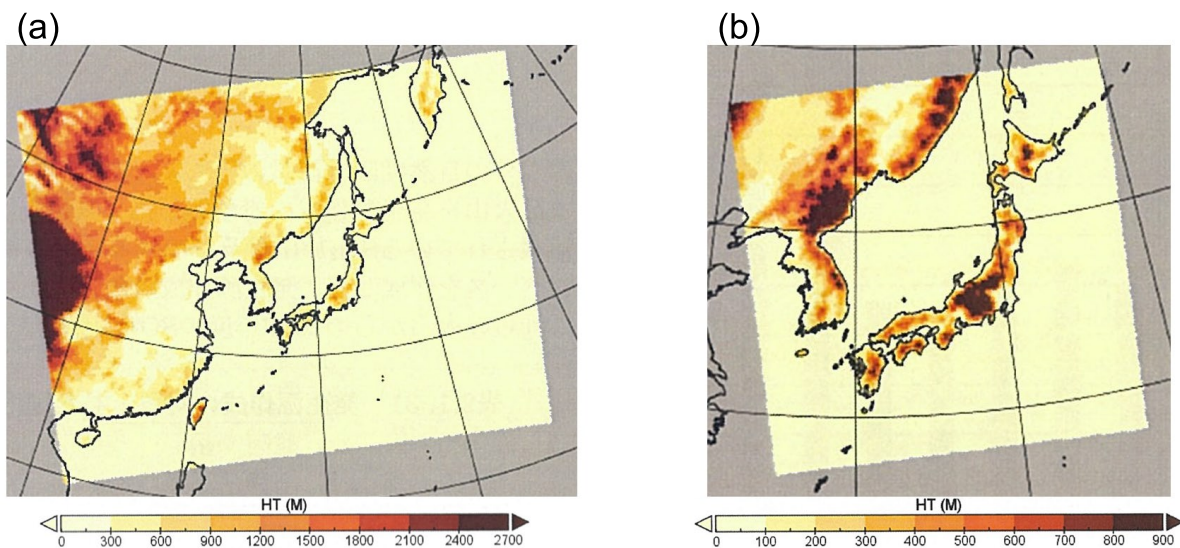
Figure 1. Ranges of (a) Domain 1 and (b) Domain 2 of the WRF-CMAQ models in this study. Color bars denote altitude.

pollutant. It mostly originates from anthropogenic sources, especially fossil fuel combustion (e.g., power plants and vehicles). The environmental SO_2 level was severe in 1970s in Japan. But the SO_2 concentration has been decreasing owing to the use of desulfurization technologies and low-sulfur heavy oil, and JEQS for SO_2 was satisfied at most AEROS sites in 2012 (Wakamatsu et al., 2013).

3   Model simulation

A model simulation was performed to calculate the amounts of O_3, SPM, NO_2, and SO_2 of all Japanese municipalities (1896 in total; note that wards in megacities, such as Tokyo, Osaka, and Fukuoka were counted as independent municipalities). The AEROS measurement network does not cover the all municipalities, thus we employed the model simulation. We combined two regional models; The Weather Research and Forecasting (WRF) model, for calculating meteorological fields (e.g., temperature, wind, and humidity), and the Community Multiscale Air Quality (CMAQ) model, calculating air pollutant amounts using the WRF results as input parameters. Detailed descriptions about the WRF and CMAQ models are written in Sect. 3.1. The calculations were made from 22 March 2014 to 31 March 2017, and the outputs from 1 April 2014 to 31 March 2017 with the interval of every 1 hour were used for analyses. We selected the simulation period with a unit of fiscal year (FY), starting on 1 April and ending on 31 March, because the AEROS measurement dataset that we used to evaluate our simulation (Sect. 3.3) was archived with a unit of FY. The amount of SPM was simply assumed as $[SPM] = ([PM_{10}] + [PM_{2.5}])/2$ in this study.

3.1 WRF-CMAQ settings

We used the WRF model version 3.7 (Skamarock et al., 2008) to calculate the meteorological fields. We set two model domains; which Domain 1 covered East Asia with a horizontal grid resolution of 40 km and 157×123 grid points, and Domain 2 covered main-land Japan with a horizontal grid resolution of 20 km and 123×123 grid points, see Fig. 1. The vertical layers consisted of 29 levels from the surface to 100 hPa. The initial and boundary conditions were obtained from the National Center for Environmental Prediction (NCEP) Final Operational Global Analysis (FNL, ds083.2) data (six-hourly, $1° \times 1°$ resolution) (NCEP FNL, 2000). In the model domain, three-dimensional grid nudging for horizontal wind, temperature, and water vapor mixing ratio as well as two-dimensional grid nudging for sea surface temperature were performed every six hours. Furthermore, we used the following parameterizations: the new Thompson scheme (Thompson et al., 2008) for microphysical parameterization, the Dudhia scheme (Dudhia, 1989) and Rapid Radiative Transfer Model (Mlawer et al., 1997) for short- and longwave radiation processes, the Mellor-Yamada-Janjić scheme (Janjić, 1994) for planetary boundary layer parameterization, and the Betts-Yamada-Janjić scheme (Janjić, 1994) for cumulus parameterization.

The CMAQ model version 5.1 was used as a chemical transport model in this study. Byun and Schere (2006) provided an overview of the CMAQ model, and the updates and scientific evaluations of CMAQ version 5.1 are provided by Appel et al. (2017). For the gas-phase chemistry, the 2005 Carbon Bond (CB05) chemical mechanism with toluene update and additional chlorine chemistry (CB05TUCL Yarwood et al., 2005; Whitten et al., 2010; Sarwar et al., 2012) was used. The core CB05 mechanism (Yarwood et al., 2005) has 51 chemical species and 156 reactions for the compounds and radicals of hydrogen, oxygen, carbon, nitrogen and sulfur. After that, the toluene update (Whitten et al., 2010) has improved the predictions of O_3 and NO_X productions and losses dealing with 59 chemical species and 172 reactions in total. In addition, the implementation of chlorine chemistry (Sarwar et al., 2012) added 7 chemical species and 25 reactions of chlorides, affecting to increase O_3 and reduce nitrates. About the photolysis of molecules, the photolysis rate preprocessor (JPROC) with 21 reactions (Roselle et al., 1999) has been implemented. About the formations of aerosols, the combination of secondary organic aerosol (SOA) formations (Pye and Pouliot, 2012; Pye et al., 2013; Appel et al., 2017), ISORROPIA algorithms (Fountoukis and Nenes, 2007) and binary nucleation (VehkamäKi et al., 2002) has been implemented. 45 kinds of aerosols components, including sulfate, ammonium, black carbon, organic carbon and sea salt, have been considered in this model.

The molecules and aerosols were provided by the emissions (anthropogenic, biogenic and sea salt) from surface or transports from the boundaries of domains, and were transported by the wind fields calculated in the WRF model and the parameterizations of horizontal/vertical diffusions, dry deposition and gravitational settling (see Byun and Schere, 2006; Appel et al., 2017). Anthropogenic emissions were defined using the MIX Asian emission inventory version 1.1 which included emissions by power, industry, residential, transportation and agriculture (Li et al., 2017). This inventory of SO_2, NO_X, PM, VOC, CO and NH_3 for 2015 were estimated by correcting the 2010 data (2008 for NH_3) and implemented into the CMAQ model. The corrections were made using the statistical secular changes in the annual total anthropogenic emissions of pollutants and CO_2 (Crippa et al., 2019), population, amount of used chemical fertilizer and NH_3 emission by farm animals for each country included in the model domains (Japan, China, South Korea, North Korea, Taiwan, Mongolia, Vietnam, and Far East Russia). Biogenic

emissions of VOC were provided by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.10 (Guenther et al., 2012) using the meteorological fields calculated by the WRF model for 2016. Those implemented emission inventories did not include interannual changes. Volcanic emissions of SO_2 were ignored in our model simulation because of the following reason. The SO_2 concentration values were averaged for 24 hours to be consistent with the time length of the numerical criterion of JEQS. This procedure dilutes an increase of SO_2 due to volcanic eruption.

The ~~used CMAQ model had~~ CMAQ model used two model domains, whose regions were the same as those adopted in the WRF model, see Fig. 1, and vertical coordinates of 22 layers; the thickness of the lowest layer was approximately 30 m. The initial and boundary conditions of air pollutants for Domain 1 were obtained from the Model for OZone And Related chemical Tracers (MOZART) version 4 (Emmons et al., 2010), and the boundary conditions for Domain 2 were the model outputs of Domain 1. The MOZART provided the distributions of more than 80 kinds of chemical species and aerosols for the inputs of our model calculations. The amount of pollutants in each Japanese municipality were defined at the longitude/latitude of the municipal office, with the weighted average of the outputs at model grid points near the municipal office using the following equation:

$$\overline{A} = \frac{1}{A_w} \sum_{i=1}^{I} \frac{R^2 - d_i^2}{R^2 + d_i^2} A_i, \quad A_w = \sum_{i=1}^{I} \frac{R^2 - d_i^2}{R^2 + d_i^2}, \tag{2}$$

where $\overline{A}$ is the defined amount of a pollutant at the municipal office, I ($=2$ or 3 mostly) is the number of the model grid points of Domain 2 within $R = \sqrt{0.02}$ degrees of the terrestrial central angle (approximately 16 km) from the office, and A_i and d_i are the simulated amount of a pollutant and distance from the office, respectively, at each model grid point. Note that Okinawa Prefecture and Ogasawara-mura municipality in Tokyo Prefecture were outside Domain 2, and the amount of pollutants at the municipalities in them were thus defined using the model outputs of Domain 1 with $R = \sqrt{0.08}$ degrees (approximately 31 km) in Eq. (2). We also derived the amount of pollutants in Seoul and Beijing for the comparisons with that inside Japan from the model outputs of Domain 1.

3.2 Spatial-temporal variation of CII

The spatial-temporal variations of CII based on the WRF-CMAQ model ~~is~~ are shown in Fig. 2 (a). The horizontal and vertical axes correspond to the date and municipal number, respectively. The lower municipal number corresponds approximately to the municipalities in northeast Japan and vice versa, and the major cites in Japan are shown in the vertical axis. The CII value clearly depended on both area and season. The CII value tended to be higher in summer because of transportation of unpolluted air mass from the Pacific Ocean. In August 2014, July 2015 and September 2016, the CII values of almost all municipalities were higher than 0.9 for a few weeks. However, the local CII values decreased to below 0.5 over a short period from May because of local air pollutant emissions and the enhancement due to photochemical reactions induced by strong UV sunlight. The CII value was moderate (0.7–0.8) and stable from November to February over Japan but gradually decreased from February to May or June because polluted air was transported from East Asia (e.g., Park et al., 2014), and the sunlight strengthened.
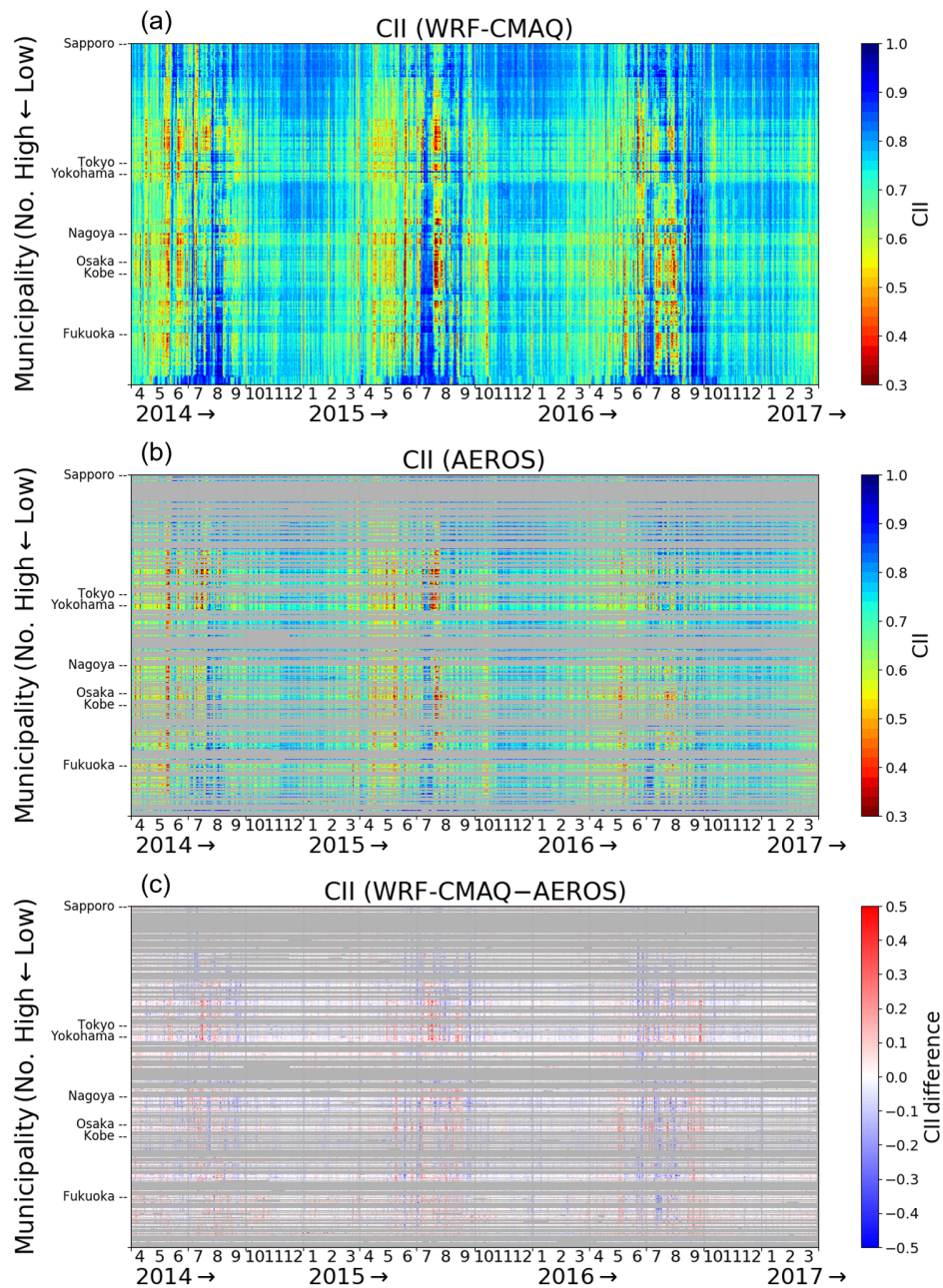
Figure 2. Spatial-temporal variation in CII values derived from (a) the WRF-CMAQ model, (b) the AEROS measurements and (c) their difference (WRF-CMAQ − AEROS). The horizontal and vertical axis corresponds to date of the study period and Japanese municipal number, respectively. The municipalities where the AEROS observation covers less than 20 % of days in the study period are masked by gray color.
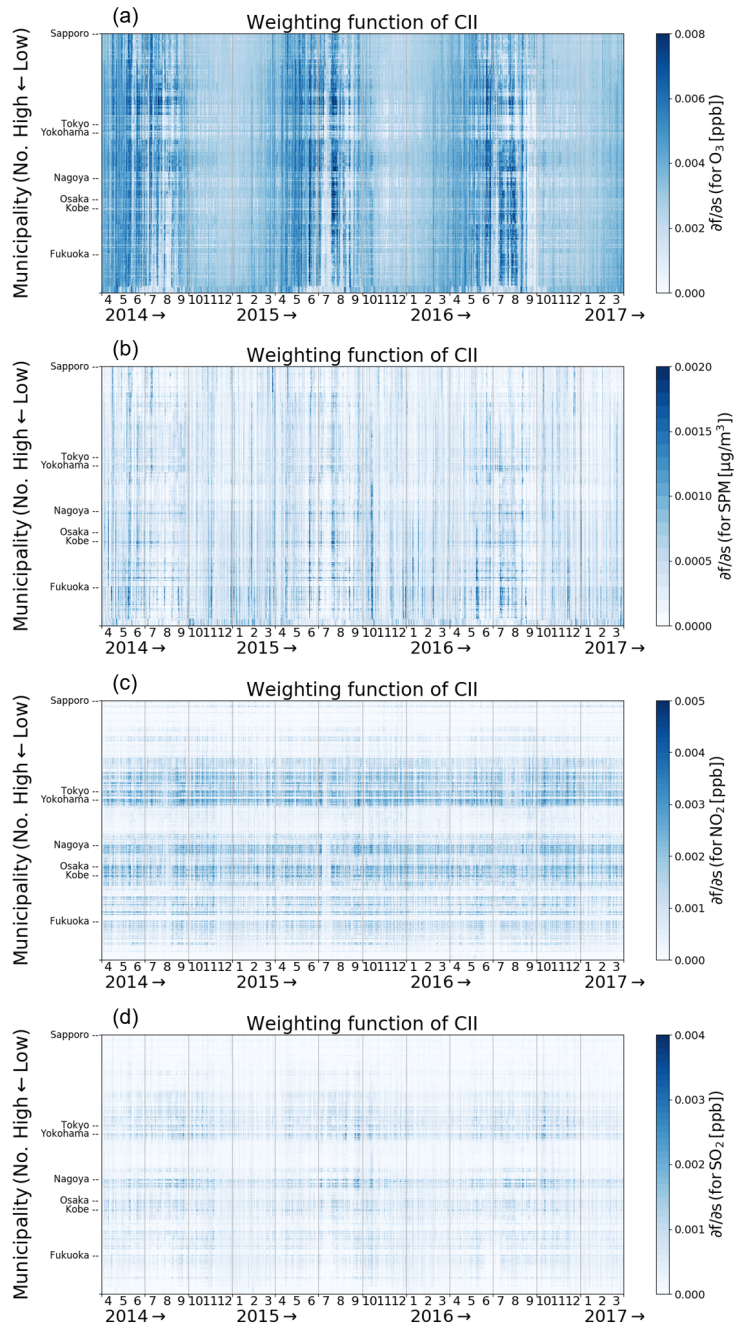
Figure 3. Spatial-temporal variation of the weighting function for the numerical criteria, K_s, for (a) O$_3$, (b) SPM, (c) NO$_2$ and (d) SO$_2$ derived from the WRF-CMAQ model. The color scaling is optimized for each panel.

These spatial-temporal features were reproduced by the AEROS measurements. Figures 2 (b) and (c) show the time series variations in the daily CII value derived from the AEROS measurements and the difference (WRF-CMAQ − AEROS), respectively. AEROS is operated by the MOE of Japan and has 1901 observation sites for monitoring air pollutants in FY2016. The AEROS data were obtained from the atmospheric environment database of the National Institute for Environmental Studies

165 (*Kankyosuchi database (in Japanese)*). We used the AEROS observation sites that cover more than 80 % of days in the study period, and 498 in 1896 municipalities were covered by the AEROS measurements. The AEROS measurement results were averaged for all observation sites in each municipality in case that there were several observation sites in one municipality. In this comparison, the AEROS Ox data were compared to the WRF-CMAQ O_3 data because the composition ratio was larger than 90–95 % O_3 in Ox (Akimoto, 2017).

170 The CII value depends not only on the amount of O_3, SPM, NO_2, and SO_2 (x), but also on their numerical criteria (s), see Eq. (1). A partial differentiation analysis was performed to determine the sensitivities of the s values of O_3, SPM, NO_2, and SO_2 to CII. Figure 3 shows the weighting function for the numerical criteria (K_s) given by

$$K_s[i] = \frac{\partial f(x,s)}{\partial s[i]} = \frac{1}{N} \frac{x[i]}{s[i]^2}. \tag{3}$$

As shown in Eq. (3), K_s positively correlates with x, and the CII value monotonically increases with increasing s. The temporal

175 variation in CII primarily corresponded with the variation in O_3. The average K_s for O_3 was highest among the species used to calculate the CII in this study, because the x/s value of O_3 was higher than those of SPM, NO_2, and SO_2 (Table 1). The value of K_s for SPM in western Japan was higher than that in eastern Japan during winter and spring because of the effect of transboundary pollution from East Asia (e.g., Park et al., 2014). The spatial distribution of CII corresponded to those of K_s for NO_2 and SO_2, which explicitly reflected local emission sources, such as megacities and industrial areas. Typical lifetime of

180 NO_2 is approximately a few hours (e.g., Kenagy et al., 2018), and the transport effect was therefore less for these species. We ignored SO_2 emissions from volcanic eruptions, and the SO_2 distribution consequently corresponded to industrial activities. The spatial distribution of O_3 was negatively correlated to that of NO_2 primarily because of the reactions (R1-R3).

3.3 Evaluation of spatial and temporal bias

We discuss the spatial and temporal bias in our calculation to clarify magnitude of significant differences in the CII value. We

185 compared the CII mean of all days in the study period between WRF-CMAQ and AEROS for each municipality~~to investigate the spatial bias~~. The CII difference for each municipality is shown in Fig. 4 (a) to investigate the spatial bias. The histogram of the CII difference showed an asymmetric distribution, thus we fitted the histogram by using the Johnson SU function, which is a probability distribution transformed from the Normal distribution to cover the asymmetry of the sample distribution (Johnson, 1949). The mean and standard deviation ($1\,\sigma$) of CII difference were 0.00 and 0.02, respectively. In the similar way,

190 we ~~investigated the daily temporal bias by comparing~~ compared the CII mean of all Japanese municipalities for each day during the study period to investigate the daily temporal bias. Figure 4 (b) shows the histogram of the CII difference for each day. The mean and standard deviation ($1\,\sigma$) of CII difference were 0.00 and 0.04, respectively. Hereafter, we average the CII values for at least 30 days to compare the CII value among municipalities to reduce the temporal bias to be less than 0.01 ($\approx 0.04/\sqrt{30}$).
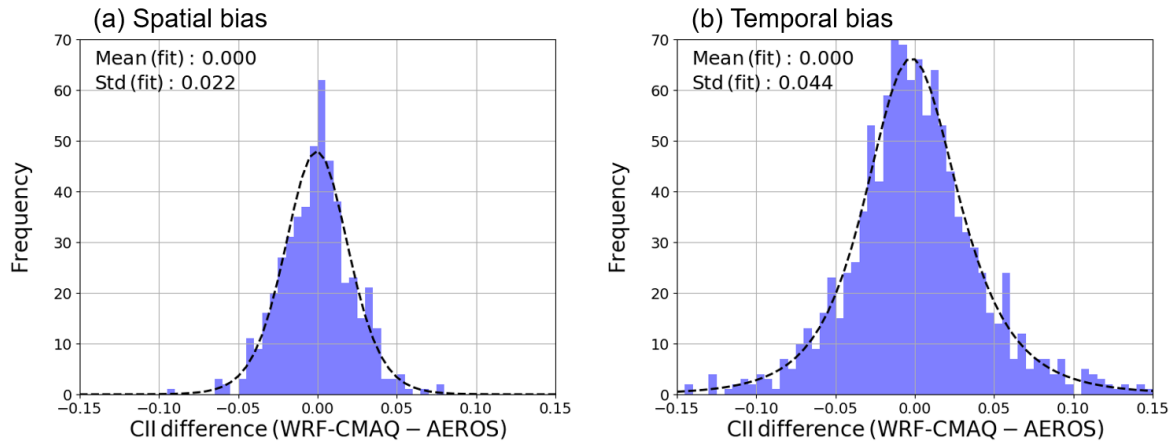
Figure 4. Histogram of CII difference between the WRF-CMAQ model and the AEROS measurements. (a) ~~Their~~ The CII mean values of all days in the study period are compared for each municipality. (b) ~~Their~~ The CII mean values of all Japanese municipalities are compared for each day. The dashed line represents fitting of the histogram of CII difference by the Johnson SU function. The mean and standard deviation ($1\,\sigma$) values of the fitting function are shown in the upper left.

Consequently, the difference in CII derived from the WRF-CMAQ larger than 0.02 was significant to be reproduced by AEROS by averaging 30 values.

3.4 Comparison of CII and AQI

In Sect. 3.4, we discuss the difference between CII and AQI as ~~a representative of the other indices~~ representative of indices to evaluate the air quality. We compared these indices calculated from the WRF-CMAQ model and the AEROS measurements. The mean of the correlation coefficient (r) ~~of mean~~ for the study period between WRF-CMAQ and AEROS was calculated for each municipality. Figure 5 shows the histogram of r for all municipalities for (a) CII and (b) AQI. The histogram was fitted by the Johnson SU function. The r of CII and AQI was $0.66\pm0.05\,(1\,\sigma)$ and $0.57\pm0.06\,(1\,\sigma)$, respectively, and the CII showed better agreement between WRF-CMAQ and AEROS than AQI.

This discrepancy between CII and AQI is explained by the difference of their definitions. In the definition of AQI, only the air ~~pollutant that causes~~ pollutants that cause the largest health risk ~~is~~ are taken into account and the other air pollutants are ignored (US EPA, 2006). In the definition of CII, ~~all of~~ four air pollutants, O_3, SPM, NO_2 and SO_2, are averaged with normalization by their numerical criteria, as Eq. (1). It was reported that the amount of the surface O_3 was overestimated by
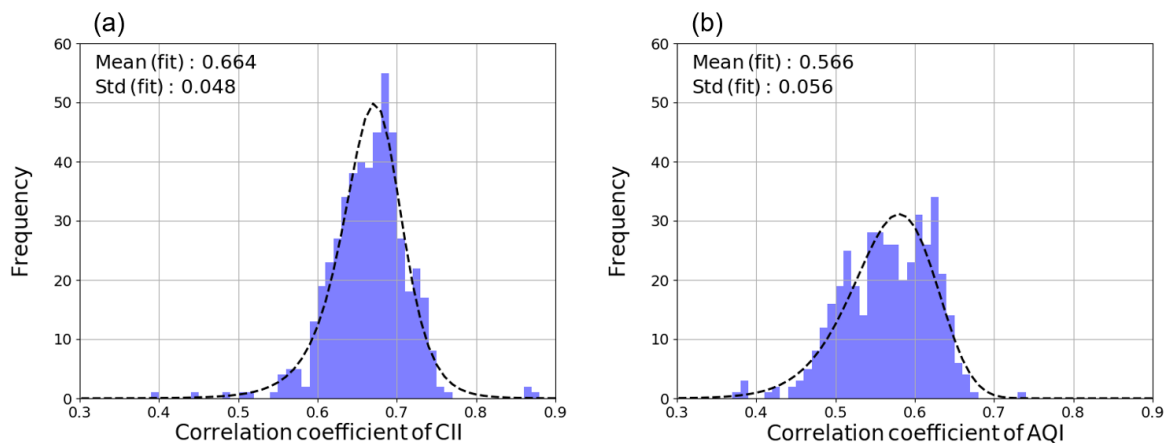
Figure 5. Histogram of correlation coefficient (r) of municipal mean of daily (a) CII and (b) AQI values for the study period between the WRF-CMAQ model and the AEROS measurements. The dashed line represents fitting of the histogram of CII difference by the Johnson SU function. The mean and standard deviation (1σ) values of the fitting function are shown in the upper left.

the CMAQ model (Akimoto et al., 2019). In this case, NO_2 is underestimated because of the following reactions:

$$NO_2 + h\nu \rightarrow NO + O, \tag{R1}$$

$$O + O_2 + M \rightarrow O_3 + M, \tag{R2}$$

210 $$NO + O_3 \rightarrow NO_2 + O_2, \tag{R3}$$

where M is a third body for the ozone formation reaction. This discrepancy is less affected for CII than for AQI because the amounts of air pollutants are averaged ~~with~~ by being normalized by the numerical criteria.

4 Visualization of air ~~cleanness~~ cleanliness in Japan

In Sect. 4, we discuss the area and season of high air ~~cleanness~~ cleanliness in Japan. Figure 6 shows the average CII over the
215 study period (FY2014–2016) for each Japanese municipality. The average CII of 85 % of municipalities were higher than that of Tokyo (23 wards), and those of all the municipalities were higher than those of Seoul and Beijing. Here the JEQS values were employed to the s values to calculate the CII values in Seoul and Beijing to directly compare with those in Japanese municipalities. The average and standard deviation (1σ) of CII was 0.67±0.10, 0.52±0.18, and 0.24±0.32 in Tokyo, Seoul, and Beijing, respectively. The value of $1 -$ CII monotonically increases with air pollutant amounts increase, and the air in
220 Tokyo was 1.5 and 2.3 times cleaner, i.e., less air pollutant amounts, than those in Seoul and Beijing, respectively. The location of the municipalities discussed hereafter is shown in Fig. 7.
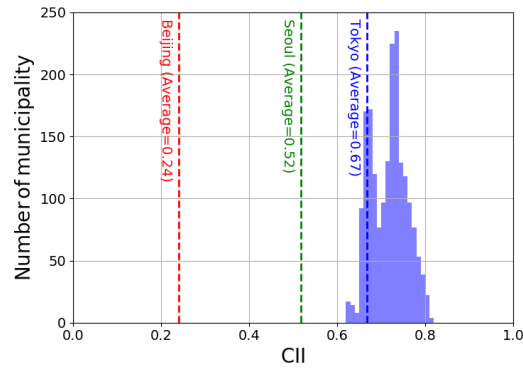
11

Figure 6. Histogram of average CII over the study period (FY2014–2016) for each municipality in Japan. Red, green, and blue dashed lines represent average CII of Beijing, Seoul, and Tokyo (23 wards), respectively.
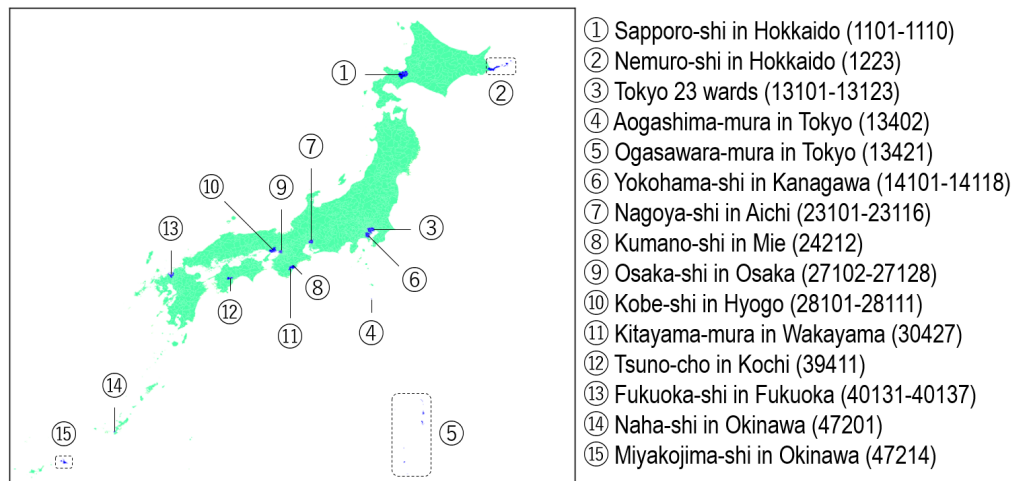

① Sapporo-shi in Hokkaido (1101-1110)
② Nemuro-shi in Hokkaido (1223)
③ Tokyo 23 wards (13101-13123)
④ Aogashima-mura in Tokyo (13402)
⑤ Ogasawara-mura in Tokyo (13421)
⑥ Yokohama-shi in Kanagawa (14101-14118)
⑦ Nagoya-shi in Aichi (23101-23116)
⑧ Kumano-shi in Mie (24212)
⑨ Osaka-shi in Osaka (27102-27128)
⑩ Kobe-shi in Hyogo (28101-28111)
⑪ Kitayama-mura in Wakayama (30427)
⑫ Tsuno-cho in Kochi (39411)
⑬ Fukuoka-shi in Fukuoka (40131-40137)
⑭ Naha-shi in Okinawa (47201)
⑮ Miyakojima-shi in Okinawa (47214)

Figure 7. Location of Japanese municipalities focused on in this study. The municipal number is shown in parenthesis.
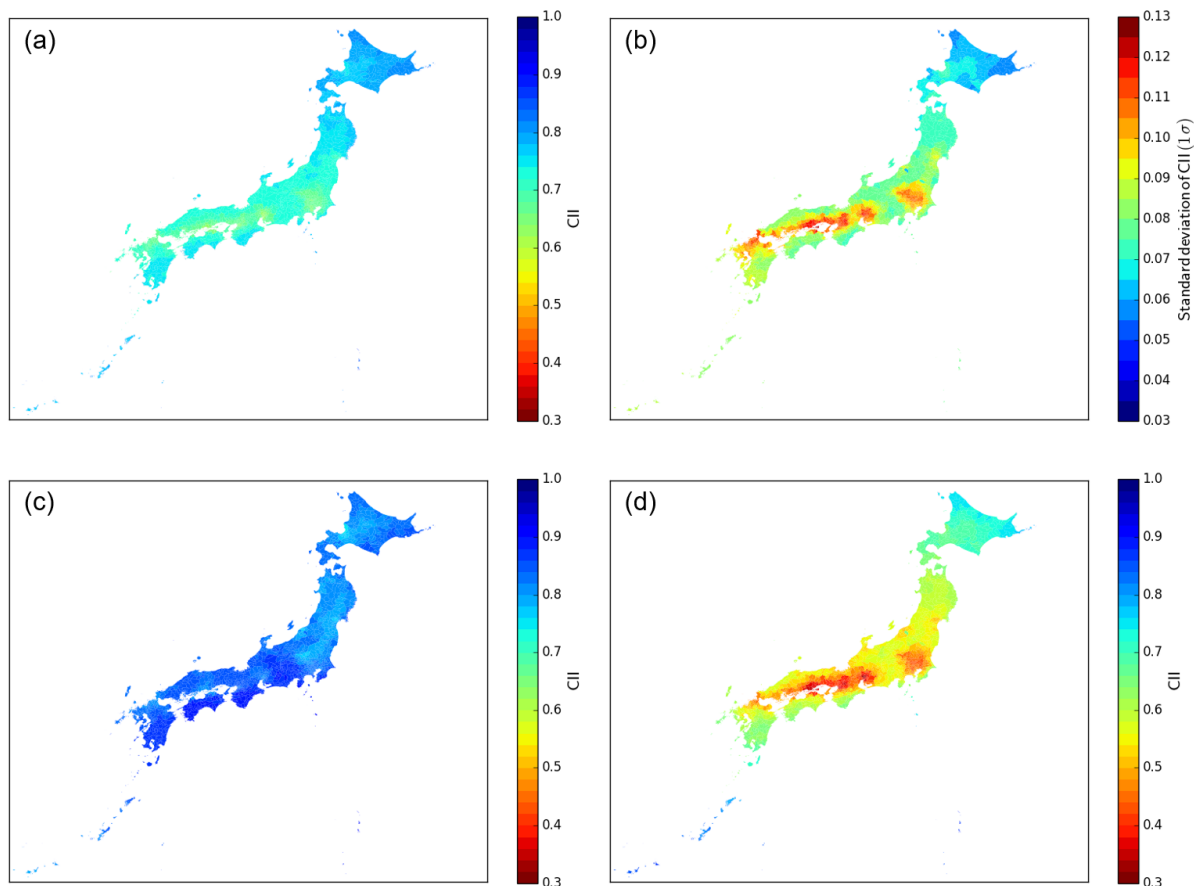
Figure 8. Spatial distributions of CII derived from the WRF-CMAQ model. (a) Mean over the study period (FY2014–2016). (b) Same as (a) but for standard deviation (1σ). (c) Mean for 30 days of highest CII average in all Japanese municipalities (10 days from each FY). (d) Same as (c) but for lowest CII average.

4.1 Area and season of high air ~~cleanness~~cleanliness

We discuss the area and season of highest air ~~cleanness~~ cleanliness over Japan using the CII in Sect. 4.1. First, the CII average over the study period, FY2014–2016, in each municipality was compared in Fig. 8 (a). The CII averages in northern Japan were higher, and those in municipalities around megacities and industrial areas were lower than the average of all municipalities, 0.72 ± 0.04 (1σ). Table 2 shows the 10 municipalities with the highest average CII values, which located in eastern Hokkaido and southern remote island in Tokyo. The average CII was approximately 0.81 in these 10 municipalities and the standard deviation (1σ) over the study period was lower than that in other areas, see Fig. 8 (b), which means the CII remained high

Table 2. Ten municipalities with highest average CII value over the study period, FY2014–2016. The municipal number is shown in paren-thesis.

Municipality	Prefecture	CII
Nemuro-shi (1223)	Hokkaido	0.814
Hamanaka-cho (1663)	Hokkaido	0.813
Akkeshi-cho (1662)	Hokkaido	0.812
Betsukai-cho (1691)	Hokkaido	0.812
Nakashibetsu-cho (1692)	Hokkaido	0.809
Kushiro-cho (1661)	Hokkaido	0.809
Rausu-cho (1694)	Hokkaido	0.808
Shibetsu-cho (1693)	Hokkaido	0.808
Ogasawara-mura (13421)	Tokyo	0.808
Sarufutsu-mura (1511)	Hokkaido	0.808
Average of all Japanese municipalites		0.717

throughout the year. For example, the CII daily value in Nemuro-shi municipality, where the three-year CII average was the highest, was higher than the total municipal average of 0.72 in 95 % of days over the study period.

We discuss the CII distribution in case of high CII average of all Japanese municipalities. We selected 10 days per year, a total of 30 days with the highest average CII values (7/9, 7/10, 8/9, 8/10, 8/15, 8/16, 8/18, 10/5, 12/6, 1/12 in FY2014; 7/9, 7/13, 7/16–19, 7/22, 8/17, 9/9, 9/10 in FY2015; and 7/9, 9/7, 9/12–9/14, 9/19, 9/20, 9/25, 9/27, 9/28 in FY2016). The 30 days CII values were averaged to discuss the CII distribution by same-order precision of 0.02 with the AEROS measurements, see Sect. 3.3. Almost all of these 30 days were ~~selected~~ in summer when unpolluted air was transported from the Pacific Ocean. The average CII values on these 30 days for each municipality are displayed in Fig. 8 (c), and Table 3 shows the 10 municipalities with the highest average CII values on these days. These 10 municipalities located in southern remote islands of Tokyo and western Pacific coast area, i.e., Kochi, Mie and Wakayama Prefectures. The average CII of Aogashima-mura municipality in southern remote islands of Tokyo Prefecture was the highest. The average CII of these 10 municipalities was approximately 0.90, which was 0.06, by CII, higher than that of all Japanese municipalities on high-CII days (0.84). Therefore, the highest CII value occurred on the Pacific coast during summer with the condition of ~~few~~ little local pollution.

Similar to the high-CII case, 30 days with the lowest CII average of all Japanese municipalities were selected (4/26, 5/29–6/2, 6/15, 6/16, 7/12, 7/15 in FY2014; 4/27, 5/13, 5/22, 5/27, 6/12, 6/13, 6/15, 7/31, 8/1, 8/2 in 2015; and 5/27, 5/28, 5/31, 6/10, 6/17, 6/18, 6/26, 6/27, 8/11, 9/1 in 2016). The average of CII values on these 30 low-CII days for each municipality are displayed in Fig. 8 (d), and Table 4 shows the 10 municipalities with the highest average CII values on these days. These 10 municipalities located in southern remote islands, such as Miyakojima-shi in Okinawa Prefecture and Ogasawara-mura in Tokyo Prefecture. The average CII in these municipalities was 0.84–0.86, which was approximately 0.30–0.32, by CII, higher than that of all

Table 3. Same as Table 2 but for the average CII for the 30 high-CII days.

Municipality	Prefecture	CII
Aogashima-mura (13402)	Tokyo	0.902
Hachijo-machi (13401)	Tokyo	0.902
Mikurajima-mura (13382)	Tokyo	0.899
Tsuno-cho (39411)	Kochi	0.897
Yusuhara-cho (39405)	Kochi	0.897
Kumano-shi (24212)	Mie	0.897
Kitayama-mura (30427)	Wakayama	0.897
Minabe-cho (30391)	Wakayama	0.897
Sakawa-cho (39402)	Kochi	0.897
Susaki-shi (39206)	Kochi	0.897
Average of all Japanese municipalites		0.836

Table 4. Same as Table 2 but for the average CII for the 30 low-CII days.

Municipality	Prefecture	CII
Miyakojima-shi (47214)	Okinawa	0.860
Ogasawara-mura (13421)	Tokyo	0.857
Tarama-son (47375)	Okinawa	0.857
Ishigaki-shi (47207)	Okinawa	0.854
Taketomi-cho (47381)	Okinawa	0.854
Minamidaito-son (47357)	Okinawa	0.848
Kitadaito-son (47358)	Okinawa	0.845
Yonaguni-cho (47382)	Okinawa	0.841
Kunigami-son (47301)	Okinawa	0.838
Higashi-son (47303)	Okinawa	0.838
Average of all Japanese municipalites		0.544

municipalities on low-CII days (0.54). The selected 30 days occurred especially at the end of spring and beginning of summer. Generally, the transboundary pollution effect is large in the cold season, and heavy local pollution occurs in summer because of photochemical reactions induced by strong sunlight (e.g., Nagashima et al., 2010). These pollution effects are less pronounced in the remote islands, thus the CII maintained higher values.
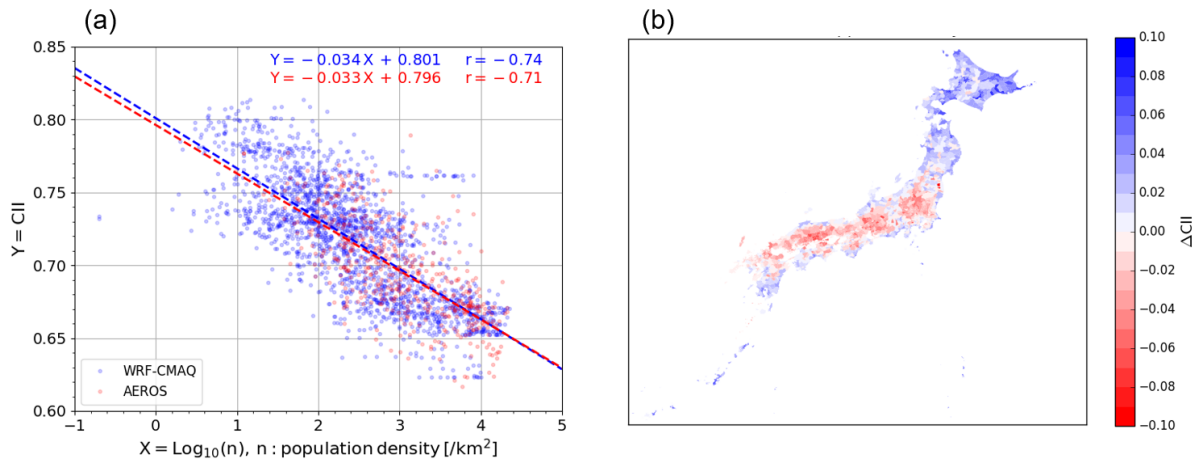
250

Figure 9. (a) Comparison of CII and population density (n) in each Japanese municipality. Dashed line shows linear regression of CII with $\log_{10}(n)$. Correlation coefficient (r) between CII values and $\log_{10}(n)$ is also shown in the upper right side. Blue and red color shows the WRF-CMAQ and AEROS results, respectively. (b) Distribution of differences in CII from the linear regression (ΔCII) for the WRF-CMAQ model.

We selected "Top 100 clean air cities" in Japan as one example of use in society of CII by the following method. The average of 30 highest daily CII values in the study period was calculated for each municipality. The 30 days were selected for each municipality, not as the case of Fig. 8 (c) and (d). Table 5 shows the 100 municipalities with the highest average CII values. The municipalities in remote islands of Tokyo, around western Japan, especially around the Pacific coast, and Okinawa Prefectures, were selected.

4.2 Air ~~cleanness~~ cleanliness and human activities

Industrial activities, particularly fossil fuel combustion such as vehicles and power plants, are major sources of air pollutants, and air ~~cleanness~~ cleanliness is strongly related with human activities. In Sect. 4.2, we discuss the municipalities in Japan with not only air ~~cleanness~~ cleanliness but also human activity~~, i. e. ,~~. We group them into four categories: 1) clean air with high human activity, 2) clean air with low human activity, 3) dirty air with high human activity, and 4) dirty air with low human activity. In this study, the common logarithm of population density (n), $\log_{10}(n)$, was employed to quantify human activities following e.g., Kerr and Currie (1995). The n data were obtained from the 2015 Japanese national census (NSTAC, 2016). Figure 9 (a) shows the scatter plot of $\log_{10}(n)$ and average CII for the study period derived from the WRF-CMAQ model and the AEROS measurements for each municipality. A clear negative correlation between $\log_{10}(n)$ and the CII was observed and the r values were -0.74 and -0.71 for WRF-CMAQ and AEROS, respectively. This negative correlation was formulated by the linear regression with the objective variable of CII and the explanatory variable of $\log_{10}(n)$, as shown by the dashed lines

16

Table 5. "Top 100 clean air cities" in Japan. The municipal number is shown in parenthesis.

Municipality	Prefecture
Nemuro-shi (1223), Kushiro-cho (1661), Akkeshi-cho (1662), Hamanaka-cho (1663)	Hokkaido
Niijima-mura (13363), Kozushima-mura (13364), Miyake-mura (13381), Mikurajima-mura (13382) Hachijo-machi (13401), Aogashima-mura (13402), Ogasawara-mura (13421)	Tokyo
Kaiyo-cho (36388)	Tokushima
Uwajima-shi (38203), Seiyo-shi (38214), Uchiko-cho (38422), Matsuno-cho (38484), Kihoku-cho (38488) Ainan-cho (38506)	Ehime
Aki-shi (39203), Tosa-shi (39205), Susaki-shi (39206), Sukumo-shi (39208), Tosashimizu-shi (39209) Shimanto-shi (39210), Toyo-cho (39301), Nahari-cho (39302), Tano-cho (39303), Yasuda-cho (39304) Kitagawa-mura (39305), Umaji-mura (39306), Geisei-mura (39307), Ino-cho (39386) Niyodogawa-cho (39387), Nakatosa-cho (39401), Sakawa-cho (39402), Ochi-cho (39403) Yusuhara-cho (39405), Hidaka-mura (39410), Tsuno-cho (39411), Shimanto-cho (39412) Otsuki-cho (39424), Mihara-mura (39427), Kuroshio-cho (39428)	Kochi
Taragi-machi (43505), Yunomae-machi (43506), Mizukami-mura (43507), Asagiri-cho (43514)	Kumamoto
Saiki-shi (44205)	Oita
Miyakonojyo-shi (45202), Nobeoka-shi (45203), Nichinan-shi (45204), Kobayashi-shi (45205) Hyuga-shi (45206), Kushima-shi (45207), Saito-shi (45208), Mimata-cho (45341), Kunitomi-cho (45382) Aya-cho (45383), Takanabe-cho (45401), Shintomi-cho (45402), Nishimera-son (45403) Kijyo-cho (45404), Kawaminami-cho (45405), Tsuno-cho (45406), Kadogawa-cho (45421) Morotsuka-son (45429), Shiiba-son (45430), Misato-cho (45431), Takachiho-cho (45441) Hinokage-cho (45442), Gokase-cho (45443)	Miyazaki
Kanoya-shi (46203), Makurazaki-shi (46204), Ibusuki-shi (46210), Nishinoomote-shi (46213) Soo-shi (46217), Kirishima-shi (46218), Shibushi-shi (46221), Amami-shi (46222) Minamikyushu-shi (46223), Osaki-cho (46468), Higashikushira-cho (46482) Kinko-cho (46490), Minamiosumi-cho (46491), Kimotsuki-cho (46492), Nakatane-cho (46501) Minamitane-cho (46502), Yakushima-cho (46505), Yamato-son (46523), Uken-son (46524) Setouchi-cho (46525), Tatsugo-cho (46527), Kikai-cho (46529)	Kagoshima
Miyakojima-shi (47214), Kunigami-son (47301), Higashi-son (47303), Minamidaito-son (47357) Kitadaito-son (47358), Tarama-son (47375), Yonaguni-cho (47382)	Okinawa

in Fig. 9 (a).

$$\text{Approximated CII} = a \times \log_{10}(n) + b \tag{4}$$

270   The parameters of a and b were estimated to be -0.034 ± 0.001 and 0.801 ± 0.002 for WRF-CMAQ, and -0.033 ± 0.001 and 0.796 ± 0.005 for AEROS, respectively. The negative correlation between $\log_{10}(n)$ and the CII value derived from WRF-CMAQ was reproduced from AEROS, and the parameters of a and b were agreed within their errors.

The CII value showed negative correlation with the human activity, thus the municipalities in groups 2 and 3 are in normal situation. The municipalities in group 1 is ideal case because such municipalities are expected to be industrially advanced

275   as well as to succeed to maintain clean air environment. There are some issues in the municipalities in group 4 because such municipalities can not ~~save~~ have clean air in spite of smaller population. It might indicate that there are large air pollution sources, such as large power plant, or air pollutants are transported from the outside. The degree of this categorizing is quantified by difference between the CII and the linear regression line, Eq. (4), (ΔCII).

$$\Delta\text{CII} = \text{CII} - \text{Approximated CII} \tag{5}$$

280   The positive ΔCII value means that the municipality is categorized in group 1, and the negative ΔCII value does group 4. The distribution of ΔCII in the average for the study period is shown in Fig. 9 (b), and Table 6 shows the 10 municipalities with the highest average ΔCII values. All of these municipalities were in Hokkaido and Okinawa prefectures. The higher ΔCII values were observed in northeastern Japan and coastal area. There are many industrial areas in western Japan (Li et al., 2017), which might be one reason for the lower ΔCII values. A combination of CII and ΔCII could be a useful way of evaluating air

285   ~~cleanness~~ cleanliness in municipality.

5   Conclusions

We defined a novel concept of index for quantifying air ~~cleanness~~cleanliness, namely CII. This index comprehensively evaluates the level of air ~~cleanness~~ cleanliness by normalizing the amounts of common air pollutants. A CII value of 1 indicates the absence of air pollutants, and 0 indicates that the amounts of air pollutants are the same as the normalization numerical criteria.

290   A model simulation was performed to visualize the air ~~cleanness~~ cleanliness of all 1896 municipalities in Japan using CII. We used O_3, SPM, NO_2, and SO_2 in this study, and their numerical environmental criteria were taken from the JEQS set by the MOE of Japan. The amounts of these species were calculated via the model combining the WRF model version 3.7 and CMAQ model version 5.1. The time period of the simulation was from 1 April 2014 to 31 March 2017, i.e., FY2014–2016. The CII values near the surface derived from the model were evaluated by comparing with those of the AEROS in situ

295   observations, operated by the MOE of Japan. 498 municipalities were covered by the AEROS measurements. The difference of CII between WRF-CMAQ and AEROS was distributed in 0.00 ± 0.02 and 0.00 ± 0.04 for spatial and temporal bias, respectively. We concluded that the difference in CII derived from the WRF-CMAQ larger than 0.02 was significant to be reproduced by AEROS by averaging 30 values to reduce the temporal bias to be less than 0.01 ($\approx 0.04/\sqrt{30}$). Difference between CII and AQI

Table 6. Ten municipalities with highest average ΔCII value over the study period, FY2014–2016. The municipal number is shown in parenthesis.

Municipality	Prefecture	ΔCII	CII
Naha-shi (47201)	Okinawa	0.095	0.762
Urasoe-shi (47208)	Okinawa	0.091	0.762
Sapporo-shi, Shiroishi-ku (1104)	Hokkaido	0.088	0.759
Sapporo-shi, Chuo-ku (1101)	Hokkaido	0.088	0.761
Ginowann-shi (47205)	Okinawa	0.088	0.762
Sapporo-shi, Toyohira-ku (1105)	Hokkaido	0.087	0.761
Sapporo-shi, Higashi-ku (1103)	Hokkaido	0.086	0.761
Sapporo-shi, Kita-ku (1102)	Hokkaido	0.086	0.761
Tomigusuku-shi (47212)	Okinawa	0.084	0.764
Yonabaru-cho (47348)	Okinawa	0.083	0.762
Average of all Japanese municipalites		—0.000	0.717

was also discussed. The mean correlation coefficient (r) ~~of mean~~ for the study period between WRF-CMAQ and AEROS was
300   calculated for each municipality. The r of CII and AQI was $0.66\pm0.05\,(1\,\sigma)$ and $0.57\pm0.06\,(1\,\sigma)$, respectively. The CII showed better agreement between WRF-CMAQ and AEROS than AQI because of the difference of definition between CII and AQI. The CII averages all normalized air pollutant amounts but the AQI employs only the maximum of ~~all individuals~~the individual pollutants, i.e., any effects from the other air pollutants are ignored. This CII concept to comprehensively evaluate multiple air pollutants could be an advantage to quantify the air ~~cleanness~~cleanliness.
305   Over the study period, FY2014–2016, the average CII value of Tokyo (23 wards), Seoul and Beijing was 0.67, 0.52 and 0.24, respectively. It means that the air in Tokyo was 1.5 and 2.3 times cleaner, i.e., less ~~amounts of~~ air pollutants, than those in Seoul and Beijing, respectively. The CII value varied spatially and temporally, corresponding to variations in O_3, SPM, NO_2, and SO_2. The municipalities in eastern Hokkaido Prefecture had the highest CII average values of approximately 0.81, which was 0.09, by CII, higher than the average values of all Japanese municipalities of 0.72. The extremely clean air with CII values,
310   approximately 0.90, occurred in southern remote islands of Tokyo and around western the Pacific coast, i.e., Kochi, Mie and Wakayama Prefectures during summer with transport of unpolluted air from the ocean. The municipalities in southern remote islands in Okinawa and Tokyo Prefectures maintained their high CII values of 0.84–0.86, which was approximately 0.30–0.32, by CII, higher than that of all municipalities on low-CII days (0.54). Furthermore, the "Top 100 clean air cities" in Japan ~~was~~ were presented as one example of ~~CII to~~ how CII could be used in society.
315   We quantified the air ~~cleanness~~ cleanliness in municipality with respect to human industrial activities ~~. Population density was used to quantify human activities in this study~~using population density. A negative correlation between CII and the population density was observed by both the WRF-CMAQ model and the AEROS measurement. The CII was approximated by a linear

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function of the common logarithm of population density. The differences of CII from this approximation line (ΔCII) indicates the CII ~~normalized~~ weighted by human activity. The municipalities with positive ΔCII values are expected to maintain clean air and to be industrially advanced. Those with negative ΔCII values are expected to have certain issues such as large air pollution source and air pollutants transported from the outside. A combination of CII and ΔCII could be a useful way of evaluating air ~~cleanness~~ cleanliness in municipality.

The CII can be used in various scenarios, such as encouraging sightseeing and migration, investment and insurance company business, and city planning. The CII can be used for an advertisement of clean air for promoting sightseeing and migration for local governments. The CII is also effective to measure the potential of local brands and tourism resources. Private company can be expected to use CII for ESG (Environmental, Social and Governance) investment. If the CII could be associated with life expectancy, the CII can be applied to insurance business especially in Asian region where urban air pollution is a serious problem. City planning is also a possible use of CII because air ~~cleanness~~ cleanliness is related to urban form (e.g., McCarty and Kaza, 2015). As mentioned above, the CII has ~~a~~ the potential to be applied to policy as well as company business in cities and countries around the world.

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