Point-By-Point Reply to Referee Comment 1 from Anonymous Referee #1

Comment from Referee:

B. Important specific comments: B1) Overall: uncertain and unclear points The referee thinks that the story which authors want to explain may be as follows: (1) validation of the model calculation (WRF/CMAQ) by comparison with observation data (AEROS), (2) explanation and interpretation of the novel cleanness index, CII, estimated for all the municipalities in Japan from the results of WRF/CMAQ, and (3) demonstration of the utility of the index, top 100 clean air cities in Japan. Would you please confirm that the referee's understanding is correct? Such a self-doubt of the referee is due to ambiguity and uncertainness in this study's aim, position, and significance, as follows, especially: (a) What are the differences among the indices? What are advantages to CII? Why CII, not AQI, for example? (b) What is the aim and significance of the top 100 clean air cities in Japan? (c) Descriptions and explanations on the methods and results are unclear. The major data in this study are based on the model calculation, aren't they? However, such critical points are not clearly found in the manuscript.

Author's response:

Yes, your understanding is right. We answered your question (a), (b) and (c) as follows. (a) As you mentioned, the statement about CII was unclear and we added a statement of AQI in Sect. 1. The advantage of CII is to represent the combined effects of multiple pollutants. A comparison study between CII and AQI using WRF-CMAQ and AEROS was added in Sect. 3.3, and please also read the Author's response to C2. We also improved the description of AQI in introduction. (b) We believe the CII has many possibilities to be used in society. We presented "Top 100 clean air cities" as one example of use in society of CII not for scientific meaning. (c) Descriptions of our model simulation in Sect. 3.1 was updated following your comments. Please also read the Author's response to C1.

Author's changes in the manuscript:

Page 2 Line 44 – 50, Page 11 Sect. 3.3

Page 1 Line 23 – 24, Page 17 Line 320 – 321, Page 22 Line 404

Page 5 Sect 3.1

Comment from Referee:

B2) Shallow descriptions without arguments and/or references: Some descriptions are without any arguments and/or references. The referee feels that authors say these descriptions definitively, without explanations.

Eg. Line 65: : : : , because more than 90 - 95 % of Ox is composed of O3.

Eg. Line 136 (similar to Line 65)

Eg. Lines 176-: : : :, because polluted air was transported from East Asia and : : :

Eg. Lines 187-: (similar to Lines 176-)

Eg. Line 188-: typical lifetime of NO2

Eg. Line 251: There are many industrial areas in western Japan, : : :

For example, are these the results of model calculation, or cited from references?

Author’s response:

We added the following references in the manuscript.

- Akimoto, H.: Overview of policy actions and observational data for $PM_{2.5}$ and O_3 in Japan: A study of urban air quality improvement in Asia, (2017).
- Park, M. E. et al.: New approach to monitor transboundary particulate pollution over Northeast Asia. Atmos. Chem. Phys., 14(2), 659-674, (2014).
- Kenagy, H. S. et al.: NOx lifetime and NOy partitioning during WINTER. J. Geophys. Res. Atmos., 123(17), 9813-9827, 2018.
- Li, M. et al.,: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP. Atmos. Chem. Phys., 17(2), 935-963, 2017.

Author’s changes in the manuscript:

Page 4 Line 79, Page 7 Line 188

Page 14 Line 264, 276, 278

Page 21 Line 366

Comment from Referee:

B3) Table 6 and Lines 225-228 Descriptions are only qualitative on the top 100 cities. What is the scientific implication?

Author’s response:

Thank you for pointing it out. We believe the CII has many possibilities to be used in society, and "Top 100 clean air cities" was presented as one example of use in society of CII not for scientific meaning.

Author's changes in the manuscript:

Page 1 Line 23 – 24, Page 17 Line 320 – 321, Page 22 Line 404

Comment from Referee:

B4) Line 239: Would you please indicate authors' opinion why 'except for Osaka'? What is the situations in Osaka?

Author's response:

Discrepancy between WRF-CMAQ and AEROS in Osaka in the previous manuscript was much improved by the following revisions. We changed the x value of O_3 from the daily average to the maximum of hourly value in 24 hours to be consistent between the time-span of x and that of s, following the comment from Anonymous Referee #3 (Major comment 3). Also, following your comment (C7), we investigated correlation between CII and population density (n) for all Japanese municipalities, where the AEROS observation covered more than 80% of the days of the study period, including Osaka. Both WRF-CMAQ and AEROS showed negative correlation between CII and population density, and the linear regression of CII (= Y) with $\log_{10}(n)$ (= X) was consistent within their errors as follows. The slope was -0.034 ± 0.001 and -0.033 ± 0.001 for WRF-CMAQ and AEROS, respectively. The intercept was 0.801 ± 0.002 and 0.796 ± 0.005 for WRF-CMAQ and AEROS, respectively. Therefore, we updated Fig. 7 and the description of CII and the human activity in Sect. 4.3 in the previous manuscript.

Author's changes in the manuscript:

Page 11 Sect. 3.3, Page 17 Sect. 4.3, Page 18 Fig. 9

Comment from Referee:

C. Other comments and Technical corrections: C1) Explanations on model and method are insufficient. Essential parts of the model descriptions are not enough, the referee feels. For example: What is CMAQ? What reactions are considered? How to consider, for example, emission, deposition, secondary formation, transportation and diffusion, for SPM, O3, NO2, and SO2?

Author's response:

CMAQ is a chemical transport model and we used CMAQ to calculate air pollutant amounts for this study. WRF is used for a calculation of meteorological fields (e.g., temperature, wind, and humidity) which is essential for accurate calculation of air pollutant amounts. We added the descriptions of emission, deposition, secondary formation, transportation and diffusion in Sect. 3.1 following your suggestion.

Author's changes in the manuscript:

Page 5 Sect 3.1

Comment from Referee:

C2) The referee thinks that readers want to compare CII with other indices. Thus, please add explanations on other indices. For example, what is AQI? If possible, as a demonstration, please calculate AQI and compare some data and figures based on CII with those based on AQI.

Author's response:

Thank you so much for this comment. We additionally calculated the AQI value based on our WRF-CMAQ model and the AEROS measurements. The correlation coefficients (r) of daily CII value for the study period between WRF-CMAQ and AEROS were compared with those of AQI. Figure 5 in the revised manuscript shows the distribution of r of (a) CII and (b) AQI. The mean of r for all municipalities, where the AEROS observation covered more than 80% of the days of the study period, was 0.664±0.048 (1 σ) and 0.566±0.056 (1 σ) for CII and AQI, respectively. The CII showed better agreement between WRF-CMAQ and AEROS than AQI. This is because of the difference of calculation method. In the calculation of AQI, only the air pollutant that causes the largest health risk is taken into account and the other air pollutants are ignored. In the calculation of CII, all of air pollutants, O_3, SPM, NO_2 and SO_2, are averaged with normalization by their numerical standards. The definition of CII relatively cancels discrepancies in each species in case that the amounts reciprocally vary as O_3 and NO_2 by the chemical reaction; $NO + O_3 \rightarrow NO_2 + O_2$. Therefore, the cancellation of discrepancy in individual species in the definition of CII is a significant advantage for quantifying air cleanness using the proposed model.

Author's changes in the manuscript:

Page 2 Line 44 – 50, Page 11 Sect. 3.3

Comment from Referee:

C3) Line 137: What is 'Fig.3(a)' ?

Author’s response:

Thank you for pointing it out. We corrected this typo.

Author’s changes in the manuscript:

Page 7 Line 190

Comment from Referee:

C4) Line 165 and others: ’1.2 times’, for example, is not proper because CII is defined as ’1 – (ratio of pollution)’ in Eq.(1). In addition, the ’times’ description is also not proper because the CII can be less than zero when the pollution is severe. Meanwhile, for the ’ratio of pollution’, the ’multiple’ description (like ’1.2 times’) is proper, because such ’ratio of pollution’ is proportional to the quantities of pollutants. However, for the difference from unity in Eq.(1), such ’multiple’ description cannot be explained to be proportional to some values.

Author’s response:

We appreciate your comment and agree with your suggestion. The value of 1 – CII is proportional to air pollutants amounts, thus we improved the statement as “The average and standard deviation (1 σ) 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 is proportional to air pollutant amounts, and the air in Tokyo was 1.5 and 2.3 times cleaner, less air pollutant amounts, than those in Seoul and Beijing, respectively.”

Author’s changes in the manuscript:

Page 1 Line 17 – 20, Page 12 Line 252 – 253, Page 22 Line 393 – 396

Comment from Referee:

C5) Line 165: Please indicate the sources of data and information on the pollutions in Seoul and Beijing.

Author’s response:

The data of the pollutants in Seoul and Beijing are derived from our CMAQ model results, as those cities are inside the Domain 1. We added the description in the end of Sect. 3.1.

Author’s changes in the manuscript:

Page 7 Line 167 – 168

Comment from Referee:

C6) Figure 4: Would you please add the histogram of CII determined from AEROS data and compare them with CII from CMAQ? Such additional comparisons can support the reasonability of CII from the model calculation.

Author's response:

Yes, we added the histogram of the CII difference between WRF-CMAQ and AEROS for all Japanese municipalities.

Author's changes in the manuscript:

Page 11 Fig. 5

Comment from Referee:

C7) Overall: Would you please add the figures of CII determined from AEROS data (similar to Figs. 5, 6 and/or 7) and compare them with those from CMAQ? Such comparisons can support the advantages of CII from the model calculation. For example, 'figures acquired from AEROS data are insufficient but those from CMAQ are fine enough to discuss the spatial distributions and temporal variations of CIIs over Japan.'

Author's response:

We performed a comparison study for all AEROS observation sites following your comment, and we discussed the spatial and temporal bias in our model simulation by statistical approach as follows. 498 in 1896 municipalities were covered by the AEROS measurements and the statistical method could be possible by including all AEROS observation sites to cover large number of samples. We deeply appreciate your valuable comment. To investigate the spatial bias between municipalities in our model simulation, we compared the CII mean of all days in the study period for each municipality between WRF-CMAQ and AEROS. The mean and standard deviation (1 sigma) of CII difference (WRF-CMAQ - AEROS) were 0.000 and 0.022, respectively. In the similar way, we investigated the daily temporal bias by comparing the CII mean of all Japanese municipalities for each day between WRF-CMAQ and AEROS. The mean and standard deviation (1 σ) of CII difference were 0.000 and 0.044, respectively. We averaged the CII values for at least 30 days to compare the CII value among municipalities to reduce the temporal bias in CII difference between WRF-CMAQ and AEROS to be less than 0.01. Consequently, we regarded that the CII difference larger than 0.02 is significant.

Author’s changes in the manuscript:

Page 7 Sect. 3.2, Page 8 Fig. 2

Comment from Referee:

C8) Overall: Geographical descriptions of Japan (eg. cities, prefectures, and islands) are insufficient. Readers unfamiliar with Japan cannot understand the information in this paper.

Author’s response:

Thank you so much for pointing it out. We showed the location of the municipalities mentioned in this manuscript.

Author’s changes in the manuscript:

Page 9 Fig. 3, Page 10 Line 206 – 207

Novel index to comprehensively evaluate air cleanness: 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, since clean air deserves a valuable resource as clean water. ~~We~~ Global standard to quantify the level of air cleanness is swiftly required, and we defined a novel concept, namely "Clean aIr Index, CII~~,~~." ~~to quantify the level of air cleanness in terms of a global standard.~~The CII is a simple index defined by the normalization of the amount 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 numerical criteria, namely the Japanese Environmental Quality Standards given by the Ministry of the Environment (MOE). We quantified air cleanness in terms of the CII for the all 1896 municipalities in Japan, and used Seoul and Beijing to evaluate Japanese air cleanness. 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 ~~were validated by comparing~~ calculated by the WRF-CMAQ model ~~and AEROS measurements for selected six cities, and an average~~ showed good agreement with those by the AEROS measurements with a correlation coefficient of ~~> 0.61 was obtained~~0.66±0.05, averaging 498 municipalities where the AEROS measurements have operated. The CII ~~value of Tokyo~~ values averaged for the study period was ~~0.75, which was 1.2 and 1.9 times higher than that of Seoul (0.64) and Beijing(0.39)~~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 extremely clean air, CII ~~> 0.93, occurred~~ ≈ 0.90, occurred in southern remote islands of Tokyo and around west of the Pacific coast, i.e., Kochi, Mie and Wakayama Prefectures ~~, and southern remote islands of Tokyo~~ during summer with transport of clean air from the ocean. The average CII value for the all Japanese municipalities was ~~0.78~~ 0.72 over the study period. We presented "Top 100 clean air cities" in Japan ~~using the CII~~ as one example of CII to be used in society. We confirmed that the CII enabled the quantitative evaluation of air cleanness. The CII can be useful value

~~, for example, for~~ in various scenarios, such as encouraging sightseeing and migration, ~~as "tasty air,"~~ 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). ~~Furthermore~~According to the report from OECD (2016), air pollutant emissions are predicted to increase because of the projected increase in the energy demand, e.g., transportation and power generation, especially in East Asia~~, and~~. 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~~are predicted to increase global GDP from 0.3 % in 2015 to 1.0 % by 2060 (OECD, 2016). Air quality is also an important issue in city planning (e.g., McCarty and Kaza, 2015). Therefore, a~~.

A global standard index to quantify air cleanness 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 index can be a useful communication tool to ~~allow people to make more informed choices~~help decision making. The index should be ~~upgraded with the scientific data, and be~~ understandable/informative not only for scientific experts but also general citizen, and also be upgraded with the scientific data.

Several ~~indexes~~ indices exist for estimating air quality, e.g., Air Quality Index 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 ~~indexes~~ indices is to estimate health risks due to air pollution exposure. These ~~indexes~~ indices were developed based on epidemiological studies and optimized for each country or local area. ~~However, a global standard index for quantifying air cleanness has not been developed~~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 pollution because AQI does not appropriately represent the combined effects of exposure to multiple pollutants. An index to quantify the air quality is still positioned as a developing phase.

In this study, we propose a novel concept of index to quantify air cleanness, "Clean aIr Index (CII)~~.~~" to establish the global standard for quantifying air cleanness. The purpose of CII is to comprehensively evaluate air cleanness by normalizing the amounts of common air pollutants with numerical environmental criteria. In this time, we 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). ~~The CII can be used globally and locally by optimizing the numerical criteria.~~ 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

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. ~~Criterion for photochemical oxidants (Ox) in JEQS was used as the s value for O_3, because more than 90–95 % of Ox is composed of O_3.~~

Air pollutant	Average of model	Numerical criteria (s)	Notes
O_3	~~31.9~~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

Observation System (AEROS) provide highly accurate air pollutant amounts, and ii) the obvious numerical criteria, i.e., the Japanese Environmental Quality Standards given by the Ministry of the Environment (MOE).

In this paper, Sect. 2 ~~introduces~~ defines the CII. Section 3 describes the model for calculating the CII for all Japanese municipalities, and validates the CII ~~by comparing it with that~~ values by comparing with those derived from AEROS measurements. In Sect. 4, air cleanness in each municipality is quantified and the area and season of high air cleanness in Japan is identified using the CII.

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

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

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 ~~this study~~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.

~~The CII can be optimized according to users' requirements by selecting the air pollutants and setting the s values. The air pollutants we selected~~ In this study, the air pollutants used in the CII ~~in this study were~~ are O_3, PM, NO_2 and SO_2 following the WHO AQG (WHO, 2005) as mentioned above, i.e., $\mathrm{N} = 4$. ~~We~~ 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). ~~These~~ The time-span 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 limit 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 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 ~~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. We used the~~ suspended particulate matter (SPM) for PM following the JEQS~~, not $PM_{2.5}$, PM with a diameter of less than 2.5 μm, because the purpose of the CII is to estimate the level of air cleanness that is not a health risk.The amount of SPM was simply assumed as SPM= (PM_{10}+ $PM_{2.5}$)/2 in this study.~~ .NO_2 is a precursor of surface O_3 and is a harmful pollutant. It mostly originates from anthropogenic sources, especially fossil fuel combustion (e.g., power plants and vehicles). ~~Major sources of~~ The environmental SO_2 ~~are emissions from volcanic eruptions (e.g., Read et al., 1993), as~~ level was severe in 1970s in Japan. But the SO_2 ~~emissions from anthropogenic sources have been reduced via regulatory policies (Wakamatsu et al., 2013).~~ 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)~~, including municipalitieswith no stations to monitor air pollutants~~. 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. 3.2) was archived with a unit of FY. The ~~settings of the WRF-CMAQ model used~~ amount of SPM was simply assumed as [SPM] = ([PM_{10}] + [$PM_{2.5}$])/2 in this study~~are described below~~.

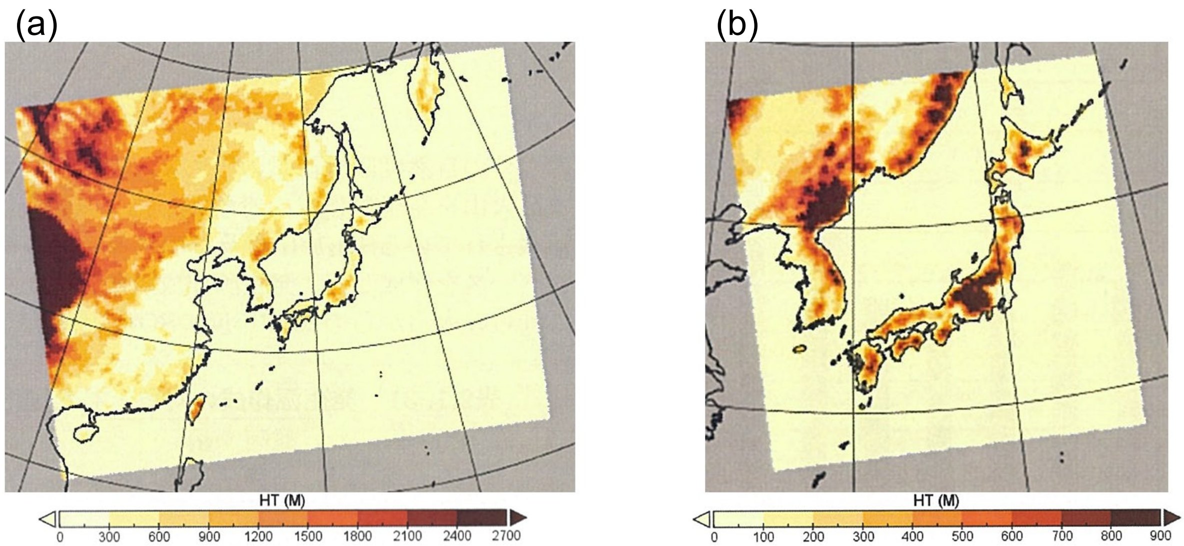

Figure 1. Ranges of (a) Domain 1 and (b) Domain 2 of the WRF-CMAQ models in this study. Color bars denote altitude.

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 Figure 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° × 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). ~~The~~ For the gas-phase chemistry, the 2005 Carbon Bond (CB05) chemical mechanism with toluene update and additional chlorine chemistry ~~(CB05TUCL Whitten et al., 2010; Sarwar et al., 2012) was usedfor the gas-phase chemistry. The used CMAQ model had two model domains, whose regions were the same as those adopted in the WRF model, see Figure 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~~ (CB05TUCL Yarwood et al., 2005; Whitten et al., 2010; Sarwar et 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 ~~model outputs of Domain 1.~~

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. ~~The~~ Those implemented emission inventories did not include interannual changes. Volcanic emissions of SO_2 were ignored ~~, even though there are many active volcanos in Japan, because volcanic activities are irregular and difficult to reproduce~~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-span of the numerical criterion of JEQS. This procedure dilutes an increase of SO_2 due to volcanic eruption.

The used CMAQ model had two model domains, whose regions were the same as those adopted in the WRF model, see Figure 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 Evaluation: Comparison with in situ measurements

~~Location of Japanese municipalities focused on in this study.~~

~~Correlation coefficient (r) of the CII, O_3, SPM, NO_2 and SO_2 between the WRF-CMAQ model simulation and the AEROS measurements in six Japanese cities. Numbers in parenthesis represent mean values of WRF-CMAQ (left) and AEROS (right) for the study period (FY2014–2016).~~

The CII value derived from the amounts of O_3, SPM, NO_2, and SO_2 calculated by the WRF-CMAQ model was compared with that measured by AEROS. 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 (*Kankyosuchi database (in Japanese)*). ~~We selected six cities for the comparison, i.e., Akita, Tokyo, Nagano, Osaka, Fukuoka and Kagoshima. The locations of these cities are shown in Fig. 3. Akita, Tokyo, Osaka, and Fukuoka were selected as representative cities of the four seasonal variation patterns of the surface O_3 in Japan: increase in spring in northeastern area (Akita), increase in spring and summer in Kanto area (Tokyo), increase in spring, summer, and autumn in Kansai area (Osaka), and increase in spring and autumn in western area (Fukuoka) (Akimoto et al., 2015). Nagano was selected as a rural area far from large anthropogenic emission sources. Kagoshima was selected to evaluate the effect of volcanic emission because there is active volcano in Sakurajima island, located approximately 5 km from the Kagoshima municipal office. The WRF-CMAQ results were averaged for all the wards in the comparison of Tokyo, Osaka and Fukuoka cities. The AEROS measurement results were averaged for all observation sites in each city, but in Tokyo, the observation sites in remote islands were omitted. The observation sites in Sakurajima island in Kagoshima were omitted because we ignored SO_2 emission from volcanic eruptions in our model as described in Sect. 3.1.~~ 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).

~~Time~~ Figure 2 shows the time series variations in the daily CII ~~mean derived from WRF-CMAQ and AEROS are compared in Fig. 2~~ value derived from (a) ~~.~~ WRF-CMAQ~~and AEROS showed similar seasonal variations in the CII, i.e., increasing in spring to summer (May–August) and decreasing in autumn to winter (September–April) . This seasonal variation in the CII~~

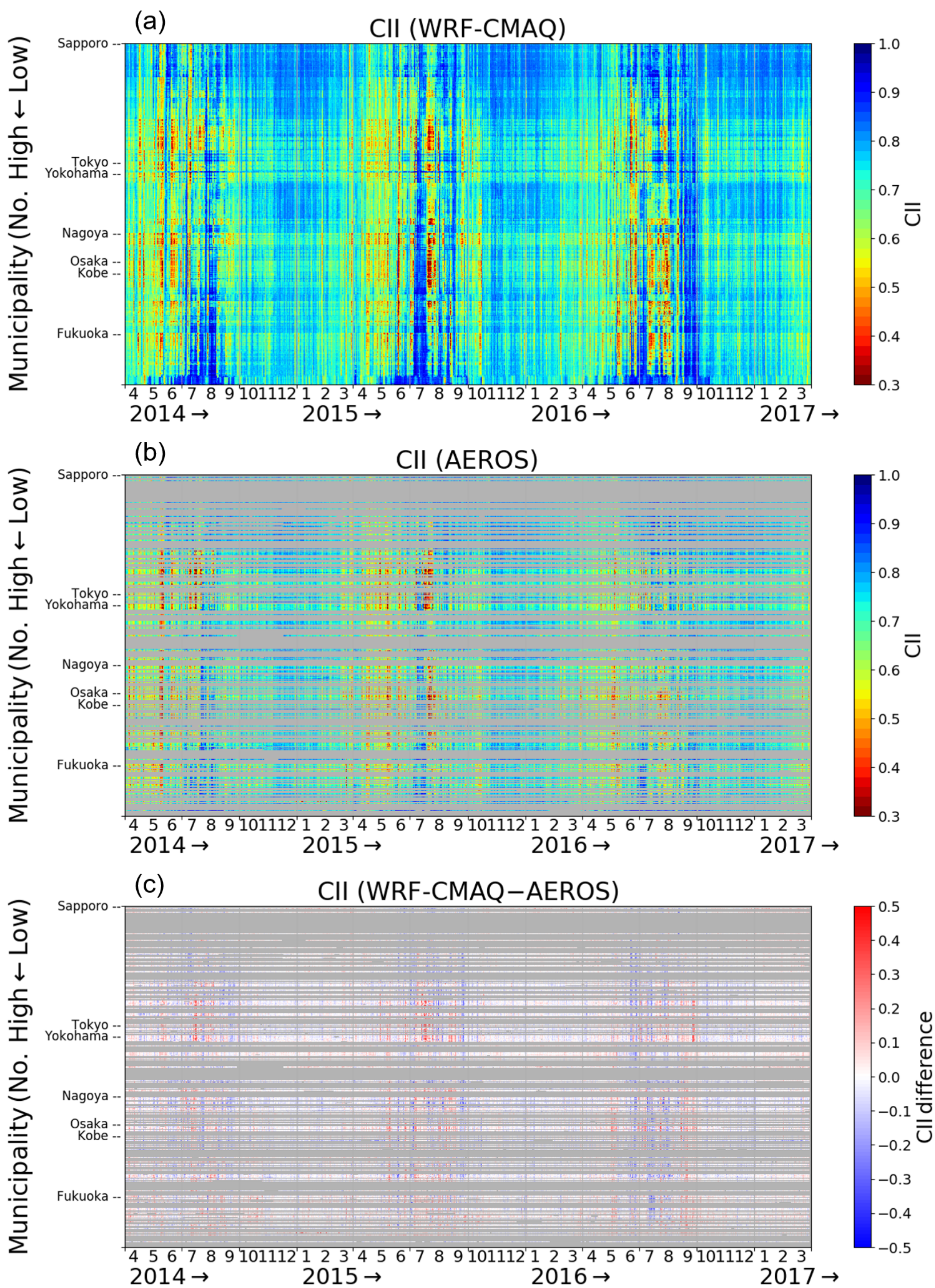

Figure 2. Comparison of CII derived from the WRF-CMAQ model and the AEROS measurements. ~~Left column shows time~~ Spatial-seasonal variation ~~of~~ in CII ~~daily mean. Center column shows scatter plot of them with correlation coefficient~~ values derived from (~~r~~a) ~~. Right column shows histogram of differences in CII~~ the WRF-CMAQ model, (b) the AEROS measurements and (c) their difference (WRF-CMAQ − AEROS). ~~Dashed line is fitting curve with~~ The horizontal and vertical axis corresponds to date of the ~~Johnson SU function~~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.

~~Akita Tokyo Nagano Osaka Fukuoka Kagoshima CII 0.61 0.68 0.61 0.73 0.71 0.65 O_3 ppb0.61 0.67 0.62 0.71 0.69 0.62 SPM μg/$m^3$0.49 0.53 0.45 0.57 0.57 0.31 NO_2 ppb0.25 0.51 0.15 0.56 0.37 0.01 SO_2 ppb0.10 0.36 0.21 0.52 0.19 −0.07~~

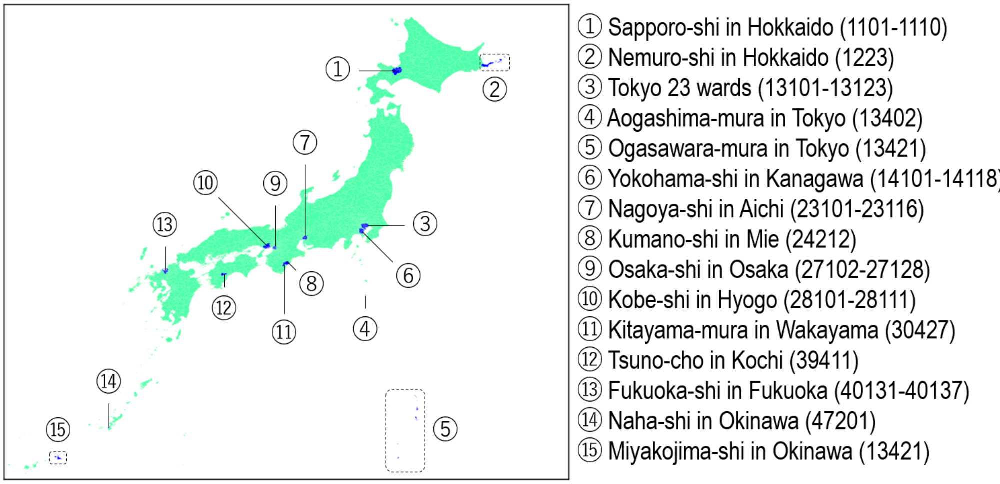

Figure 3. Location of Japanese municipalities focused on in this study. The municipal number is shown in parenthesis.

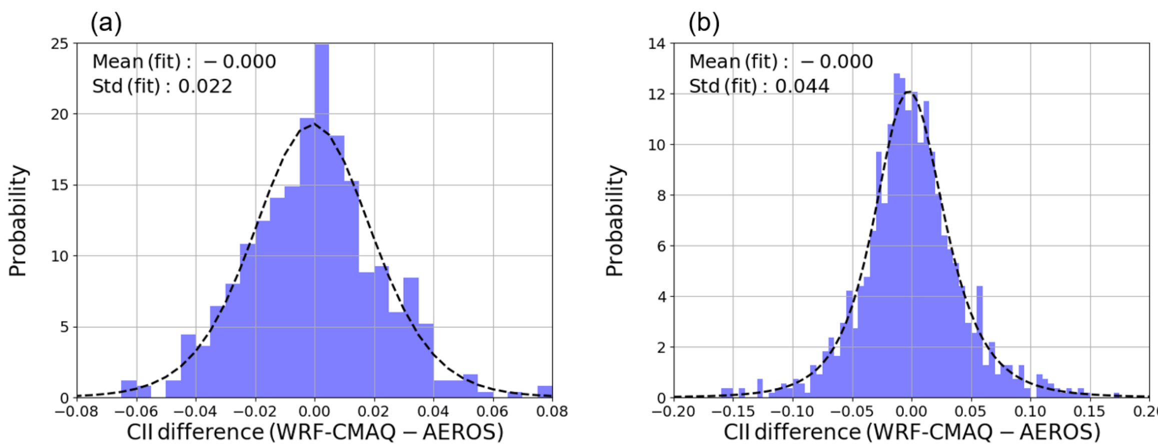

Figure 4. Histogram of CII difference between the WRF-CMAQ model and the AEROS measurements. (a) Their CII mean values of all days in the study period are compared for each municipality. (b) Their CII mean values of all Japanese municipalities are compared for each day. The dashed line represents fitting of the histogram of CII difference by the Johnson SU function. The mean and standard deviation (1 σ) values of the fitting function are shown in the upper left.

~~was observed in the abovementioned six cities. The CII showed good agreement between~~ , (b) AEROS, and (c) their difference (WRF-CMAQ ~~and AEROSwith a correlation coefficient (r)of larger than 0.61. Table ?? shows the r values of the CII, O_3, SPM, NO_2, and SO_2 between WRF-CMAQ and AEROS for the six cities. The r values of the CII were higher than those of O_3, SPM, NO_2 and SO_2 in the 5 cities (not in Nagano), because the amounts of the species were normalized and comprehensively merged in the definition of CII in Eq. (1). This definition of CII relatively cancels discrepancies in each species in case that the amounts reciprocally vary as O_3 and NO_2 as below. NO_2 is a major precursor of O_3, and photolysis of NO_2 provides the oxygen atoms required to generate O_3 in the following reactions:~~

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

$$\mathrm{O + O_2 + M \rightarrow O_3 + M},$$

$$\mathrm{NO + O_3 \rightarrow NO_2 + O_2},$$

~~where M is a third body for the ozone formation reaction. The model underestimates the amount of O_3 and overestimates that of NO_2 in case of large contribution of the reaction (R3), i.e., NO titration effect. This case was observed in~~ – AEROS). 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 ~~comparison in Tokyo, Osaka and Fukuoka, and~~ major cites in Japan are shown in the vertical axis. Figure 3 shows the location of these cities and municipalities discussed in this paper hereafter. We used the AEROS observation sites that cover more than 80 % of days in the ~~r values for CIIwere higher than those for O_3, and NO_2, see Table ??.Therefore, the cancellation of discrepancy in individual species in the definition of CII is a significant advantage for quantifying air cleanness using the proposed model.~~

~~We investigated the precision of the difference in the CII between the WRF-CMAQ model and AEROS measurements~~ 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.

WRF-CMAQ and AEROS showed similar seasonal variations in the CII, i.e., increasing in spring to summer (May–August) and decreasing in autumn to winter (September–April). We discuss the spatial and temporal bias in our calculation to clarify magnitude of significant differences in the CII ~~derived from the WRF-CMAQ model. The histogram of the difference in the CII (~~ value. We compared the CII mean of all days in the study period between WRF-CMAQ ~~– AEROS), the right column of~~ and AEROS for each municipality to investigate the spatial bias in Fig. ~~2, shows~~ 4 (a). The histogram of the CII difference showed an asymmetric distribution~~. We~~ , 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~~ 1 σ) of ~~the fitted Johnson SU distribution was approximately 0.06 (0.054–0.067) . It showed that the WRF-CMAQ model reproduced~~ CII difference were 0.00 and 0.02, respectively. In the similar way, we investigated the daily temporal bias by comparing the CII mean of all Japanese municipalities for each day. The mean and standard deviation (1 σ) 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 ~~within a difference from AEROS of approximately~~ among municipalities to reduce the temporal bias to be less

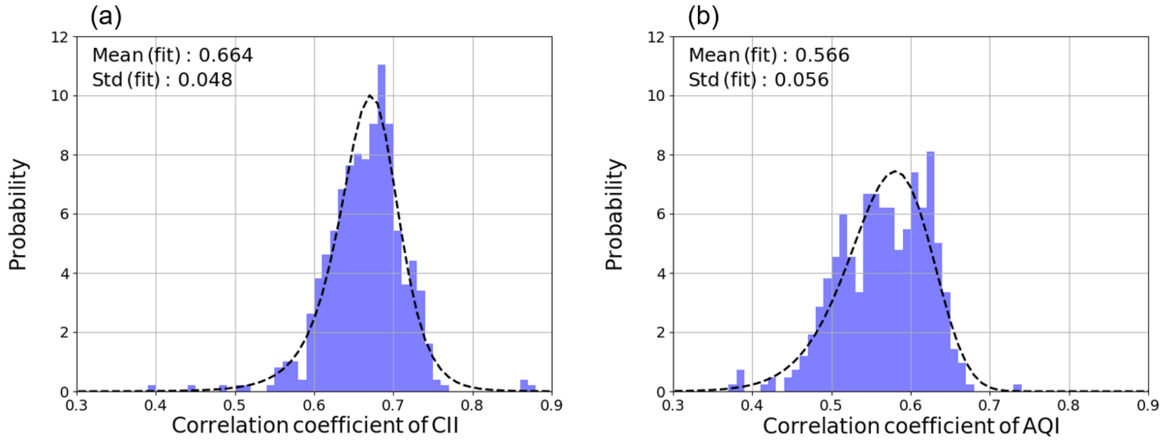

Figure 5. Histogram of correlation coefficient (r) of municipal mean of daily (a) CII and (b) AQI values for the study period between the WRF-CMAQ model and the AEROS measurements. The dashed line represents fitting of the histogram of CII difference by the Johnson SU function. The mean and standard deviation ($1\,\sigma$) values of the fitting function are shown in the upper left.

than 0.01 ~~by averaging 30 values (0.06~~($\approx 0.04/\sqrt{30}$ ~~≈ 0.01~~). Consequently, the difference in CII derived from the WRF-CMAQ larger than ~~0.01~~ 0.02 was significant to be reproduced by AEROS by averaging 30 values.

3.3 Evaluation: Comparison of CII and AQI

In Sect. 3.3, we discuss difference between CII and AQI as a representative of the other indices. We compared these indices calculated from the WRF-CMAQ model and the AEROS measurements. 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 $0.66 \pm 0.05\,(1\,\sigma)$ and $0.57 \pm 0.06\,(1\,\sigma)$, respectively, and the CII showed better agreement between WRF-CMAQ and AEROS then AQI.

This discrepancy between CII and AQI is explained by the difference of the calculation methods. In the calculation of AQI, only the air pollutant that causes the largest health risk is taken into account and the other air pollutants are ignored (US EPA, 2006). In the calculation of CII, all of 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 the CMAQ model

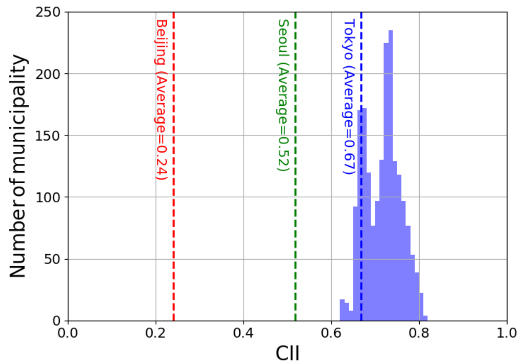

Figure 6. Histogram of average CII over the study period (FY2014–2016) for each municipality in Japan. Red, green, and blue dashed lines represent average CII of Beijing, Seoul, and Tokyo (23 wards), respectively.

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

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

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

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

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 being normalized by the numerical criteria.

4 Visualization of air cleanness in Japan

In Sect. 4, we discuss the area and season of high air cleanness in Japan. Figure 6 shows the average CII over the 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 (~~1~~1 σ) of ~~Tokyo was 0.75~~CII was 0.67±~~0.07, which was 1.2 and 1.9 times higher than those of Seoul (0.64~~0.10, 0.52±~~0.13) and Beijing(0.39~~0.18, and 0.24±~~0.29)~~0.32 in Tokyo, Seoul, and Beijing, respectively. The ~~average CII of 89 % of municipalities were higher than that of Tokyo, and those of all the municipalities were higher than those of~~ value of $1 - \mathrm{CII}$ is proportional to air pollutant amount, and the air in Tokyo was 1.5 and 2.3 times cleaner, i.e., less air pollutant amounts, than those in Seoul and Beijing, respectively.

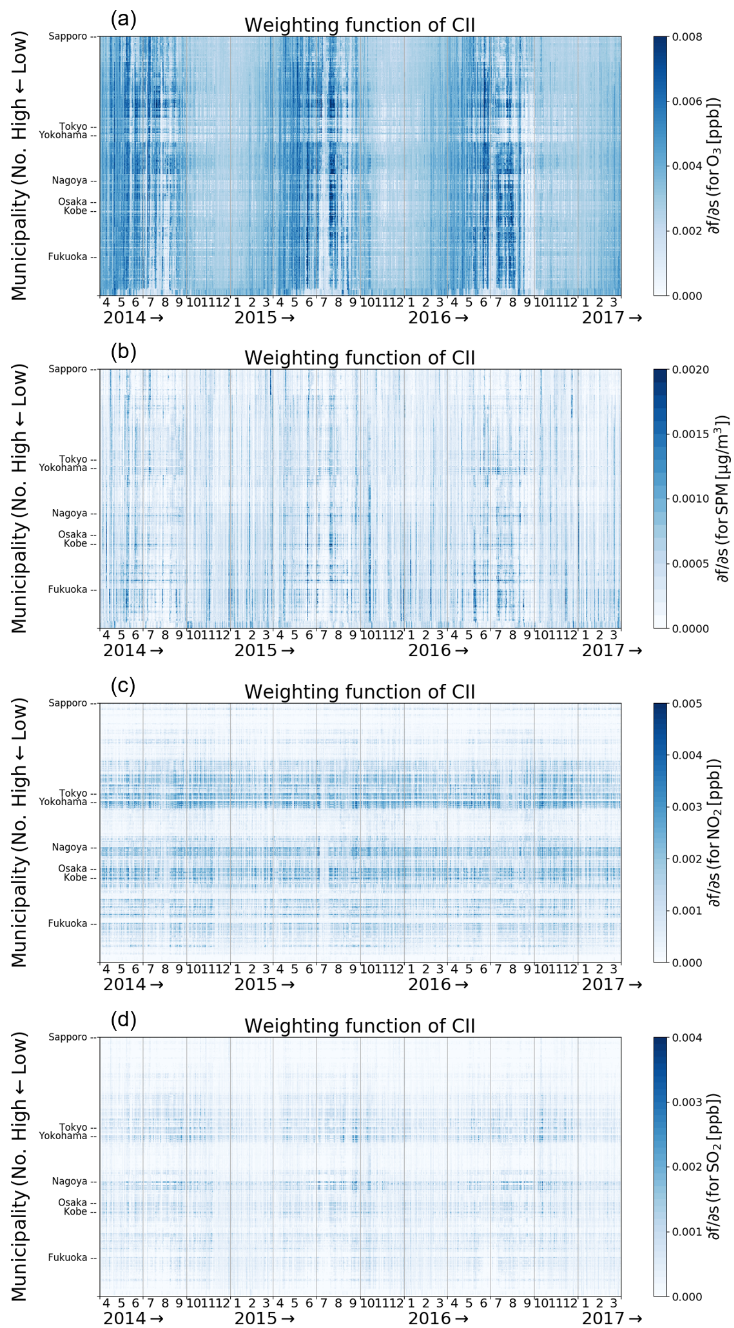

Figure 7. Spatial-seasonal variation ~~in (a) CII and~~ of the weighting function for the numerical criteria, K_s, ~~of~~ for (~~b~~a) O_3, (~~c~~b) SPM, (~~d~~c) NO_2 and (~~e~~d) SO_2 derived from the WRF-CMAQ model. The color scaling of ~~(b–e)~~ is optimized for each panel.

4.1 Spatial-seasonal variation

The spatial-seasonal variations in CII, O_3, SPM, NO_2, and SO_2 in Japan are described in Sect. 4.1. Figure ~~7~~2 (a) shows the daily ~~mean~~ CII value derived from the WRF-CMAQ model for each municipality over the study period. ~~The horizontal and vertical axes correspond to the day and municipal number, respectively, and the lower municipal number corresponds approximately to the municipalities in northeast Japan and vice versa.~~ This figure shows that the CII value 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 ~~0.55~~ 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 (~~0.70~~0.7–0~~.85~~.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. ~~The municipalities in Okinawa Prefecture, southernmost Japan, maintained their higher CII values of > 0.9 during this period.~~ These spatial-seasonal features were reproduced by the AEROS measurements, see Fig. 2 (b).

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 7 ~~(b–e)~~ shows the weighting function for the numerical criteria (K_s) given by

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

~~In the definition of CII,~~ As shown in Eq. (~~1~~3), K_s positively correlates with x, and the CII value monotonically increases with increasing s. The seasonal 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 ~~amount~~ x/s value of O_3 was ~~relatively higher than the value of s compared with~~ 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 NO_2 is approximately a few hours (e.g., Kenagy et al., 2018), and the transport effect was therefore less for ~~this~~ these species. We ignored SO_2 emissions from volcanic eruptions, and the SO_2 distribution consequently corresponded to industrial activities. ~~No significant seasonal variation in K_s was observed for NO_2 and SO_2.~~ The spatial distribution of O_3 was negatively correlated to that of NO_2 primarily because of the ~~NO titration effect, reaction (R.3~~reactions (R1-R3).

~~Consequently, the CII distribution was influenced not only by local emissions but also by transboundary pollution. The variation in O_3 had the most significant effect on seasonal variation in the CII. The spatial distribution of CII corresponded to those of NO_2 and SO_2. The SPM sources constituted both local emissions and transport from outside of Japan, and SPM variation affected both spatial and seasonal variations in CII.~~

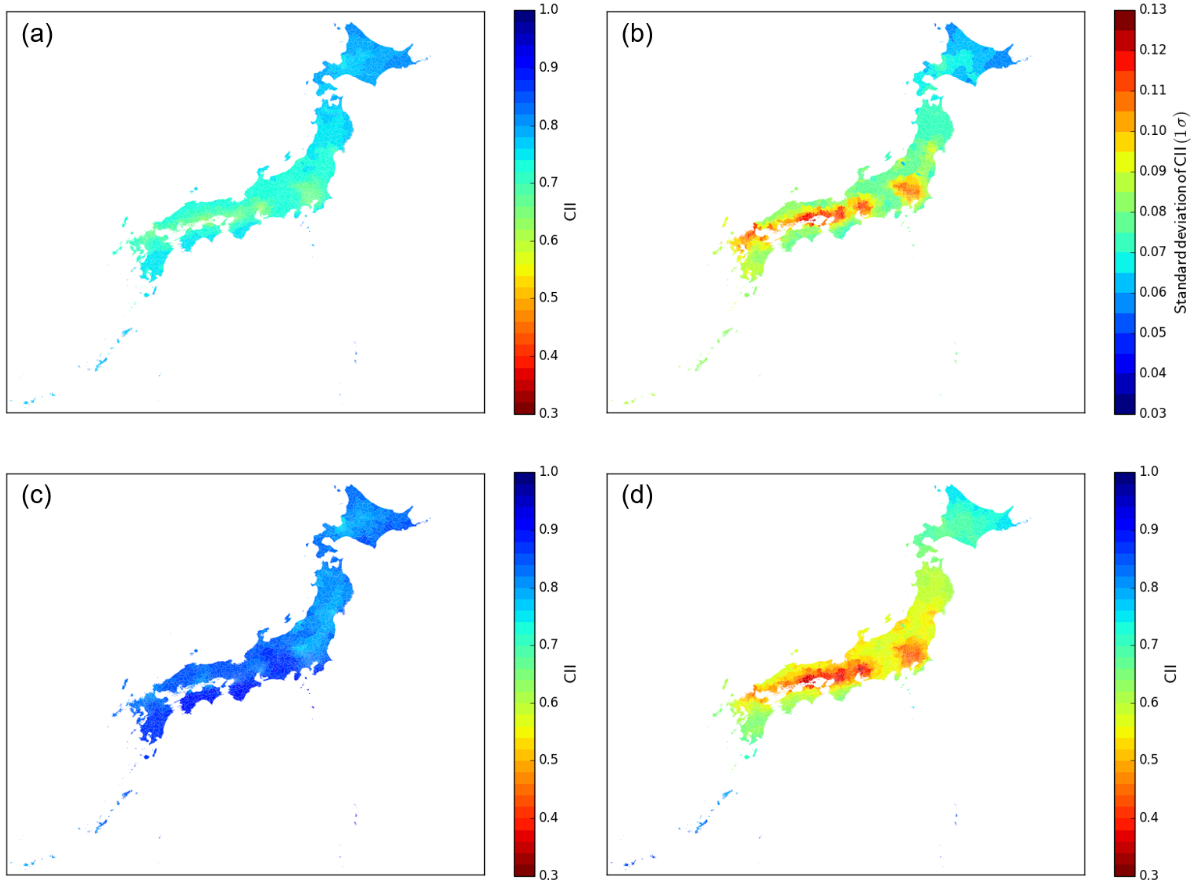

Figure 8. Spatial distributions of CII derived from the WRF-CMAQ model. (a) Mean over the study period (FY2014–2016). (b) Same as (a) but for standard deviation (~~1~~ 1 σ). (c) Mean for 30 days of highest CII average in all Japanese municipalities (10 days from each FY). (d) Same as (c) but for lowest CII average.

4.2 Area and season of high air cleanness

In Sect. 4.2, we discuss the area and season of highest air cleanness over Japan using the CII. First, the CII average over the study period, FY2014–2016, in each municipality was compared ~~,~~ in Fig. 8 (a)~~, and the daily mean value for this period was averaged for each municipality~~. 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, ~~0.78~~0.72±~~0.05 (1~~ 0.04 (1 σ). Table 2 shows the 10 municipalities with the highest average CII values, which ~~all~~ located in eastern Hokkaido and southern remote island in Tokyo. The average CII was approximately ~~0.83–0.84~~ 0.81 in these 10 municipalities ~~,~~ and the standard deviation (~~1~~ 1 σ) over the study period was lower than that in other areas, see Fig. 8 (b)~~. The~~ , which means the CII remained high throughout the year.

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

Municipality	Prefecture	CII
~~Betsukai-cho (1691~~Nemuro-shi (1223)	Hokkaido	~~0.840~~ 0.814
Hamanaka-cho (1663)	Hokkaido	~~0.840~~ 0.813
~~Shibecha-cho (1664~~Akkeshi-cho (1662)	Hokkaido	~~0.839~~ 0.812
~~Akkeshi-cho (1662~~Betsukai-cho (1691)	Hokkaido	~~0.838~~ 0.812
Nakashibetsu-cho (1692)	Hokkaido	~~0.838~~ 0.809
~~Shiranuka-cho (1668~~Kushiro-cho (1661)	Hokkaido	~~0.837~~ 0.809
~~Tsurui-mura (1667~~Rausu-cho (1694)	Hokkaido	~~0.835~~ 0.808
~~Nemuro-shi (1223~~Shibetsu-cho (1693)	Hokkaido	~~0.834~~ 0.808
~~Shibetsu-cho (1693~~Ogasawara-mura (13421)	~~Hokkaido~~ Tokyo	~~0.832~~ 0.808
~~Saroma-cho (1552~~Sarufutsu-mura (1511)	Hokkaido	~~0.832~~ 0.808
Average of all Japanese municipalites		~~0.778~~ 0.717

For example, the CII daily ~~mean in Betsukai-cho~~ value in Nemuro-shi municipality, where the three-year CII average was the highest, was higher than the total municipal average of ~~0.78 in 91~~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 ~~to discuss the CII distribution by same-order precision of 0.01 with the AEROS measurements, see Sect. 3.2~~ (7/9, 7/10, 8/9, 8/10, 8/~~15–8~~15, 8/~~20 in 2014~~16, 8/18, 10/5, 12/6, 1/12 in FY2014; 7/~~13–7~~9, 7/~~19~~13, 7/16–19, 7/22, 8/17, 9/9, 9/10 in ~~2015~~FY2015; and 7/9, ~~8/21,~~ 9/7, 9/12–9/14, 9/19, 9/20, 9/25, 9/27, 9/28 in ~~2016). These~~ 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. 3.2. Almost 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 ~~around~~ in southern remote islands of Tokyo and western Pacific coast area, i.e., Kochi, Mie and Wakayama Prefectures~~, and southern remote islands of Tokyo~~. The average CII of ~~Susaki-shi municipality in Kochi~~ Aogashima-mura municipality in southern remote islands of Tokyo Prefecture was the highest. The average CII of these 10 municipalities was approximately ~~0.93~~0.90, which was ~~1.1 times~~ 0.06, by CII, higher than that of all Japanese municipalities on high-CII days (~~0.88~~0.84). Therefore, the highest CII value occurred on the Pacific coast during summer with the condition of few local pollution.

Similar to the high-CII case, 30 days with the lowest CII average of all Japanese municipalities were selected (4/~~25, 4/~~26, 5/~~13, 5/~~29–6/2, 6/~~16, 7/12 in 2014; 4~~15, 6/16, ~~4~~7/~~24, 4~~12, 7/~~26,~~ 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/~~4, 5/~~27, 5/28, 5/31, 6/10, 6/17, 6/18, 6/~~25~~26, 6/~~26~~27, 8/~~31~~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

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

Municipality	Prefecture	CII
~~Susaki-shi (39206~~Aogashima-mura (13402)	~~Kochi~~ Tokyo	~~0.934~~ 0.902
~~Tsuno-cho (39411~~Hachijo-machi (13401)	~~Kochi~~ Tokyo	~~0.933~~ 0.902
~~Hachijo-machi (13401~~Mikurajima-mura (13382)	Tokyo	~~0.933~~ 0.899
~~Kumano-shi (24212~~Tsuno-cho (39411)	~~Mie~~ Kochi	~~0.933~~ 0.897
~~Kitayama-mura (30427~~Yusuhara-cho (39405)	~~Wakayama~~ Kochi	~~0.933~~ 0.897
~~Mikurajima-mura (13382~~Kumano-shi (24212)	~~Tokyo~~ Mie	~~0.933~~ 0.897
~~Nakatosa-cho (39401~~Kitayama-mura (30427)	~~Kochi~~ Wakayama	~~0.933~~ 0.897
~~Aogashima-mura (13402~~Minabe-cho (30391)	~~Tokyo~~ Wakayama	~~0.932~~ 0.897
Sakawa-cho (39402)	Kochi	~~0.932~~ 0.897
~~Miyake-mura (13381~~Susaki-shi (39206)	~~Tokyo~~ Kochi	~~0.931~~ 0.897
Average of all Japanese municipalites		~~0.880~~ 0.836

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 and Ishigaki-chi in Okinawa~~Tokyo Prefecture. The average CII in these municipalities was ~~0.82~~0.84–0.~~85~~.86, which was approximately ~~1.3 times larger~~ 0.30–0.32, by CII, higher than that of all municipalities on low-CII days (~~0.64~~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 ~~spring~~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.

We selected "Top 100 clean air cities" in Japan ~~using the CII . The~~ as one example of use in society of CII by the following method. The average of 30 highest daily ~~mean~~ CII values in the study period ~~were averaged~~ 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, ~~and~~ around western Japan, especially around the Pacific coast, ~~i.e., Wakayama, Tokushima, Ehime, Kochi, Kumamoto, Oita, Miyazaki, Kagoshima~~ and Okinawa Prefectures, were selected.

4.3 Air cleanness and human activities

Industrial activities, particularly fossil fuel combustion such as vehicles and power plants, are major sources of air pollutants, and air cleanness is strongly related with human activities. ~~It is generally difficult to maintain clean air with large-scale industrial activities, and it is therefore not fair to directly compare air cleanness in municipalities with different human activities.~~ In Sect. 4.3, we discuss the ~~relationship between air cleanness~~ municipalities in Japan with not only air cleanness but also human activity, i.e., ~~CII, and the scale of human activities.~~

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

Municipality	Prefecture	CII
~~Ogasawara-mura (13421~~Miyakojima-shi (47214)	~~Tokyo~~ Okinawa	~~0.852~~ 0.860
~~Ishigaki-shi (47207~~Ogasawara-mura (13421)	~~Okinawa~~ Tokyo	~~0.840~~ 0.857
~~Taketomi-cho (47381~~Tarama-son (47375)	Okinawa	~~0.840~~ 0.857
~~Miyakojima-shi (47214~~Ishigaki-shi (47207)	Okinawa	~~0.835~~ 0.854
~~Tarama-son (47375~~Taketomi-cho (47381)	Okinawa	~~0.835~~ 0.854
Minamidaito-son (47357)	Okinawa	~~0.832~~ 0.848
~~Yonaguni-cho (47382~~Kitadaito-son (47358)	Okinawa	~~0.829~~ 0.845
~~Kitadaito-son (47358~~Yonaguni-cho (47382)	Okinawa	~~0.828~~ 0.841
Kunigami-son (47301)	Okinawa	~~0.824~~ 0.838
Higashi-son (47303)	Okinawa	~~0.824~~ 0.838
Average of all Japanese municipalites		~~0.644~~ 0.544

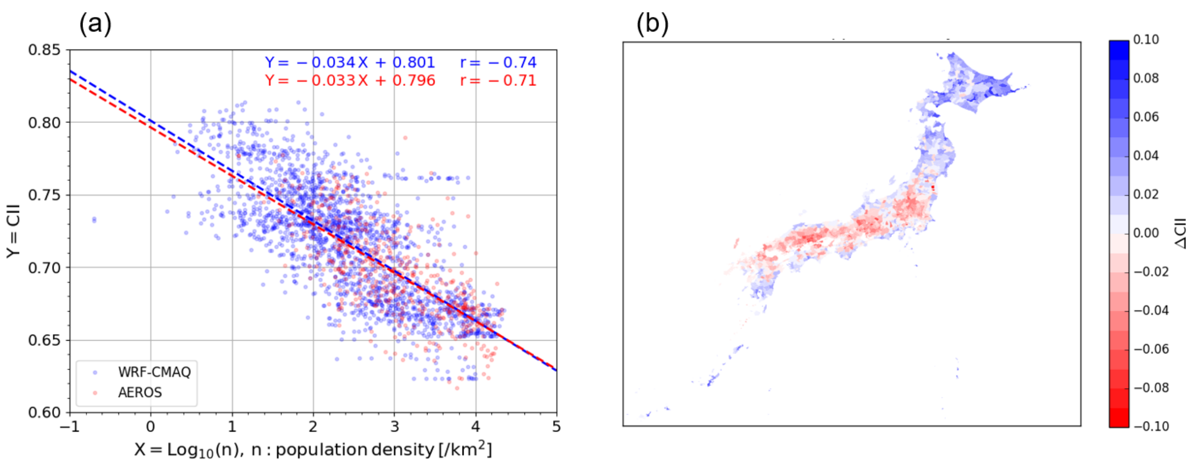

Figure 9. (a) Comparison of CII and population density (n) in each Japanese municipality. ~~Blue dot shows the WRF-CMAQ results. Black dashed~~ Dashed line shows linear regression of CII with $\log_{10}(n)$. Correlation coefficient (r) between CII values and $\log_{10}(n)$ is also shown in the upper right side. ~~Red markers show the CII derived from~~ Blue and red color shows the WRF-CMAQ and AEROS ~~measurements in six cities (Akita, Tokyo, Nagano, Osaka, Fukuoka~~results, ~~and Kagoshima)~~respectively. (b) Distribution of differences in CII from the linear regression (ΔCII) for the WRF-CMAQ model.

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

Municipality
Nemuro-shi (1223), Kushiro-cho (1661), Akkeshi-cho (1662), Hamanaka-cho (1663)
Niijima-mura (13363), Kozushima-mura (13364), Miyake-mura (13381), Mikurajima-mura (13382)
Hachijo-machi (13401), Aogashima-mura (13402), Ogasawara-mura (13421)
~~Tanabe-shi (30206), Minabe-cho (30391), Shirahama-cho (30401), Kamitonda-cho (30404) Wakayama Nachikatsuura-cho (30421), Kozagawa-cho (30~~
Uwajima-shi (38203), Seiyo-shi (38214), Uchiko-cho (38422), Matsuno-cho (38484), Kihoku-cho (38488)
Ainan-cho (38506)
~~Kochi-shi (39201),~~ Aki-shi (39203), ~~Nankoku-shi (39204),~~ Tosa-shi (39205)~~Kochi~~, Susaki-shi (39206), Sukumo-shi (39208), Tosashimizu-shi (39209)
~~Konan-shi (39211), Kami-shi (39212~~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)
Taragi-machi (43505), Yunomae-machi (43506), Mizukami-mura (43507), Asagiri-cho (43514)
Saiki-shi (44205)
~~Miyazaki-shi (45201),~~ Miyakonojyo-shi (45202), Nobeoka-shi (45203), Nichinan-shi (45204)~~Miyazaki~~, Kobayashi-shi (45205) ~~,~~
Hyuga-shi (45206), Kushima-shi (45207), Saito-shi (45208)~~Ebino-shi (45209)~~, Mimata-cho (45341), ~~Takaharu-cho (45361),~~ 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)
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), ~~Yusui-cho (46452),~~ 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)~~Minamidaito-son (47357), Kitadaito-son (47358) Okinawa "Top 100 clean air c~~
~~Tano-cho, Kochi (39303~~Setouchi-cho (46525), Tatsugo-cho (46527), Kikai-cho (46529) ~~/ Otsuki-cho, Kochi (39424)/ Nishinoomote-shi, Kagoshima (4~~
~~Soo-shi, Kagoshima (46217)/ Shibushi-shi, Kagoshima (46221) /~~ Miyakojima-shi (47214), Kunigami-son (47301), Higashi-son (47303), Minamidaito-
Kitadaito-son ~~, Okinawa~~ (47358)~~Tanabe-shi, Wakayama (30206) / Shirahama-cho, Wakayama (30401) / Kamitonda-cho, Wakayama (30404) 0.951 Kai~~

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 ~~(e.g., Kerr and Currie, 1995)~~following Kerr and Currie (e.g., 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, FY2014–2016, 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 had ~~an~~ r ~~value~~ values of $-$~~0.68~~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 ~~black dashed line~~ dashed lines in Fig. 9 (a).

$$\mathrm{CII} = \cancel{-0.016}a \times \log_{10}(n) + \cancel{0.82}b \tag{4}$$

The ~~red markers indicate the CII values derived from the AEROS measurements at the six cities used for comparison study (Fig. 2)~~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 ~~the CII value and~~ $\log_{10}(n)$ and the CII value derived from WRF-CMAQ was reproduced from ~~the AEROSmeasurements and agreed with the linear regression line, except for Osaka~~AEROS, and the parameters of a and b were agreed within their errors.

~~Ten municipalities with highest average ΔCII value over the study period, FY2014–2016. The municipal number is shown in parenthesis. Municipality Prefecture ΔCII CII Obihiro-shi (1207) Hokkaido 0.050 0.827 Sapporo-shi, Atsubetsu-ku (1108) Hokkaido 0.049 0.805 Kushiro-shi (1206) Hokkaido 0.048 0.830 Sapporo-shi, Shiroishi-ku (1104) Hokkaido 0.046 0.802 Nemuro-shi (1223) Hokkaido 0.046 0.834 Nakashibetsu-cho (1692) Hokkaido 0.046 0.838 Kushiro-cho (1661) Hokkaido 0.045 0.831 Sapporo-shi, Chuo-ku (1101) Hokkaido 0.043 0.800 Sapporo-shi, Toyohira-ku (1105) Hokkaido 0.043 0.800 Sapporo-shi, Higashi-ku (1103) Hokkaido 0.043 0.800 — 0.000 0.778~~

~~The~~ 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 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 clean air in spite of small 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), ~~normalized the CIIvalues by the population density n. The difference between the CII and the linear regression line (~~(ΔCII). The positive ΔCII value means that the municipality is categorized in group 1, and the negative ΔCII ~~) can be an additional indicator compensating for the unfair comparison of air cleanness caused by human activities.~~ value does group 4. The distribution of ΔCII in the average for FY2014–2016 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 ~~, similar to the results shown in Table 2, but urban municipalities in Hokkaido were ranked as Sapporo-shi and Obihiro-shi cities. The~~ and Okinawa prefectures. The higher ΔCII values ~~in northeastern Japan, especially Hokkaido, were higher than those in western Japan . The ΔCII value reflects the transport of air pollutants from around the municipality rather than the CII value if the neighboring municipality was a megacity or had industrial factories~~were observed in northeastern

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

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. ~~As discussed above, ΔCII quantified the air cleanness with respect to population density, i.e., human activities. The~~ A combination of CII and ΔCII could be a useful way of evaluating air cleanness ~~.~~ in municipality.

5 Conclusions

We defined a novel concept of index for quantifying air cleanness, namely CII. This index comprehensively evaluates the level of air cleanness 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.

A model simulation was performed to visualize the air cleanness of all 1896 municipalities in Japan using CII. We used O_3, SPM, NO_2, and SO_2 in ~~CII~~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 ~~value~~ values near the surface derived from the model ~~was~~ were evaluated by comparing ~~it with~~ with those of the AEROS in situ observations, operated by the MOE of Japan~~, in Akita, Tokyo, Nagano, Osaka, Fukuoka, and Kagoshima, which cover areas of four patterns of O_3 seasonal variation, a rural area and an area affected by volcanic eruption. The CII correlation coefficient r between the WRF-CMAQ and AEROS exceeded 0.61. The precision of the difference in CII between the~~ . 498 municipalities were covered by the AEROS measurements. The difference of CII between WRF-CMAQ and AEROS was ~~approximated by the Johnson SU function. The CII difference~~ 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 ~~0.01 was significant and could~~ 0.02 was significant to be reproduced by AEROS by averaging 30 values ~~.~~ ~~Over~~ to reduce the temporal bias to be less than 0.01 ($\approx 0.04/\sqrt{30}$). Difference between CII and AQI was also discussed. The correlation coefficient (r) of mean for the study period ~~,~~ between WRF-CMAQ and AEROS was calculated for each municipality. The r of CII and AQI was $0.66 \pm 0.05\,(1\,\sigma)$ and $0.57 \pm 0.06\,(1\,\sigma)$, 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, 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.

Over the study period, FY2014–2016~~, eastern Hokkaido had the highest CII average values of 0.83–0.84, which were 1.1 times higher than the average values of all Japanese municipalities of 0.78.The~~ , the average CII value of Tokyo (23 wards)~~was 0.75, which was 1.2 and 1.9 times higher than those of Seoul (0.64) and Beijing(0.39)~~, 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 ~~around 0.93, occurred~~ approximately 0.90, occurred in southern remote islands of Tokyo and around western the Pacific coast, i.e., Kochi, Mie and Wakayama Prefectures ~~, and southern remote islands of Tokyo~~ during summer with transport of unpolluted air from the ocean. The municipalities in ~~remote islands, such as Ogasawara and Okinawa~~ southern remote islands in Okinawa and Tokyo Prefectures maintained their high CII values of ~~0.82~~0.84–0.~~85~~.86, which was approximately ~~1.3 times higher than the average~~ 0.30–0.32, by CII, higher than that of all municipalities on low-CII days (0.54). Furthermore, "Top 100 clean air cities" in Japan was presented ~~using the CII~~ as one example of CII to be used in society.

~~The relationship between air cleanness and~~ We quantified the air cleanness in municipality with respect to human industrial activities~~could not be fairly compared~~. Population density was used to quantify human activities in this study. A negative correlation between CII and the population density was observed by both the WRF-CMAQ model and the AEROS measurement. The CII (Y) was approximated by a linear function of the common logarithm of population density (X), ~~$Y = -0.016X + 0.82$~~$Y = -0.034X + 0.801$. The differences ~~in~~ of CII from this approximation line (ΔCII) ~~in northeastern Japan, especially Hokkaido, were higher than those in western Japan. The east-west contrast of~~ indicates the CII normalized by human activity. The municipalities with positive ΔCII ~~might be due to large-scale industrial activities in western Japan~~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 in municipality.

The CII can be used in various scenarios, such as encouraging sightseeing and migration, investment and insurance company business, and city planning. ~~For example, Hokkaido is recommended to live because the CII value is constantly high throughout the year. Western the Pacific coast and southern remote islands can be tourist spots for seeking "tasty air " because extremely~~

~~high CII values are temporally given during summer. The CII enabled the quantitative evaluation of air cleanness, and could not only be applied in Japan but also in other countries~~ CII can be used for an advertisement of clean air for promoting sightseeing and migration for local governments. CII is also effective to measure the potential of local brands and tourism resources. Private company is expect 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 quality is related to urban form (e.g., McCarty and Kaza, 2015). As mentioned above, the CII has a potential to be applied to policy as well as company business in cities and countries around the world.

Data availability. The WRF-CMAQ model data in this publication can be accessed by contacting the authors. The AEROS measurement data are available through the following link: https://www.nies.go.jp/igreen. Japanese population density data are available through the following link: https://www.e-stat.go.jp/.

Video supplement. The CII daily mean for all 1896 Japanese municipalities is archived for each month over the study period, FY2014–2016.

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