

Synoptic weather and surface ozone concentration in South Korea

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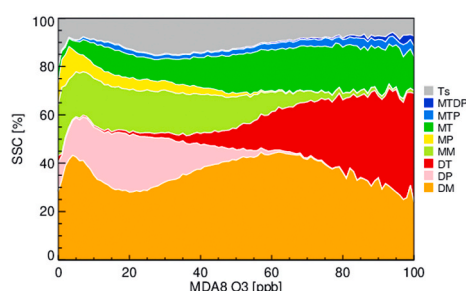
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HIGHLIGHTS

- The relationship between synoptic weather and surface ozone was investigated.
- Dry Tropical (DT) weather type has the highest efficiency in high ozone production.
- Occurrence of DT has increased over fifty years in South Korea.
- It suggests a likely connection to increased ozone in South Korea.

GRAPHICAL ABSTRACT



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ABSTRACT

Seventeen years (2001–2017) of surface observations and spatial synoptic classification (SSC) data are used to analyze the characteristics of surface ozone concentration according to synoptic weather patterns. While weather conditions are known to play an important role in regional air quality, the extent to which synoptic weather patterns affect the production of high ground-level ozone concentrations has not yet been fully quantified. Using thermal characteristics and geographic origins, the SSC method classifies air masses into six types: dry polar (DP), dry moderate (DM), dry tropical (DT), moist polar (MP), moist moderate (MM), and moist tropical (MT). We link daily maximum 8-h ozone concentrations (MDA8 O₃) from 306 monitoring sites to the closest SSC classifications at 17 airport sites and then analyze their association. We find that DM, DT, and MT are commonly associated with high ozone, whereas DT produces ozone with the greatest efficiency, especially high levels of concentration. This finding implies a potentially strong connection between surface ozone and climate change because the occurrence of DT weather has increased by more than three times over the past 50 years in South Korea. Sensitivity tests reveal that mean MDA8 ozone may increase by 3.5% (7.5%) as the DT frequency increases by 200% (300%). The impacts are larger for higher levels of concentration, with 31.7% (63.3%) or more

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prevalence of the >80 ppb range with the same increased DT frequency. We conclude that synoptic weather and its long-term trends play important roles in the increased surface ozone recently seen in South Korea.

1. Introduction

Surface ozone (O_3), one of the important trace gases in the troposphere, is formed from photochemical reactions between two major air pollutants, oxides of nitrogen (NO_x) and volatile organic compounds (VOCs). Heat and solar radiation are also critical in the formation of ozone, resulting in clear seasonal variation: higher ambient ozone concentrations during warmer months and lower concentrations in colder months. Local ozone production is affected by many factors, including anthropogenic sources, such as NO_x and VOCs emissions from mobile, industrial (Bae et al., 2018), and power generation facilities, and natural sources, such as biogenic VOCs emissions (E. Kim et al., 2017). Meteorological conditions are also critical, because ozone photochemistry is basically photon-limited, sensitive to supplied ultraviolet actinic fluxes (Kim et al., 2015). Warm and dry air (i.e., less cloud and more sunlight) favors the production of surface ozone (Bloomer et al., 2009; Camalier et al., 2007; Ryan et al., 1998).

Like other countries in East Asia, South Korea has for several decades suffered a serious degradation of the atmospheric environment, especially from severe cases of haze (H. C. Kim et al., 2017a). While high concentrations of particulate matter have drawn strong public attention (K. Lee et al., 2020), rising concern has been paid to surface ozone concentration (Kim et al., 2019), which has significantly increased in recent years. The puzzle of surface ozone concentration in South Korea lies in its enigmatic long-term increase. Daily maximum 8-h moving average ozone concentrations (hereafter MDA8 ozone), a standard ozone metric of the U.S. EPA's National Ambient Air Quality Standards, increased in Korea from 38.3 ppb in 2001 to 51.9 ppb in 2017, nearly 1 ppb per year, during warm seasons (April to September) (Figure S1). This increase has caused much interest in the research community and among policymakers.

There is no consensus explanation for such an increase in ozone in South Korea. Several speculative reasons include (1) changes in NO_x emissions (Seo et al., 2014); (2) long-range transport, especially from China (Oh et al., 2010); (3) changes in chemical regime (Bae et al., 2020); (4) stratospheric intrusion (Itahashi et al., 2020; Shin et al., 2020); and (5) changes in meteorological condition. Until the early 2000s, increased ozone was usually explained as a result of the rapid economic growth in East Asia and eventual corresponding increase in anthropogenic ozone precursors, especially NO_x emissions. Later, however, ozone continued to increase, perplexingly, even as recently reported surface and space observations suggest a decrease in NO_2 concentrations over East Asian countries, likely due to more strict regulation (Duncan et al., 2016; Krotkov et al., 2016).

Several researchers have suggested that a change of chemical regime over Korea could explain the ozone increase. Surface ozone is determined by the chemical characteristics of the response of tropospheric ozone to its precursors, namely Ozone- NO_x -VOCs chemistry. Under a so-called NO_x -saturated regime, usually in large cities with high NO_x emissions and low VOCs emissions, ozone responds positively to changes in VOCs and negatively to changes in NO_x emissions. On the other hand, ozone increases with increasing NO_x and is insensitive to changes in VOCs under a NO_x -sensitive regime, which occurs mostly in rural areas (Cohan et al., 2005; Sillman, 1999). This chemical regime has been used to explain the increase in surface ozone concentrations in Japan and Taiwan in the early 2000s. Recently, Bae et al. (2020) demonstrated the responses of surface ozone in South Korea using adjusted NO_x emissions based on satellite observations (Bae et al., 2020). Showing negative ozone- NO_x correlation, they suggested that the Seoul Metropolitan Area, South Korea is under a NO_x -saturated chemical regime and that recent efforts to reduce NO_x emissions may have caused surface ozone to

increase. However, this explanation is insufficient to describe the long-term increase, since it remains to be determined whether and when the chemical regime had changed. Hence, explaining the ozone increase due to the NO_x emission increase (in the 1980s and 1990s) as well as the NO_x emission decrease (in the 2000s) could be paradoxical.

Increased surface ozone has been reported in the northern hemisphere (Cooper et al., 2012; Mickley et al., 2004). Allen et al. (2012) stated that atmospheric heating from black carbon and tropospheric ozone has generated a poleward shift of the tropospheric jet, thereby relocating the main division between the tropical and temperature air masses. Parrish et al. (2014) compared models and observations to reproduce the long-term increase in tropospheric ozone. East Asia has one of the strongest increases in tropospheric ozone concentration based on observations from 1970 to 2009 (Allen et al., 2012) or 2000 to 2014 (Chang et al., 2017). Several studies have attributed the increased surface ozone over China to meteorological variations (Lu et al., 2019; Sun et al., 2019). Li et al. (2019), on the other hand, claimed that ozone trends in the North China Plain from 2013 to 2017 were mainly due to decreased $PM_{2.5}$ concentration, which slowed the aerosol sink of hydroperoxy (HO_2) radicals and accelerated ozone production.

Since this increase remains mysterious, we further explore the impact of weather. The influence of synoptic-scale weather on regional air quality has attracted rising interest. Regional and local air quality are affected not only by local precursor emissions but also by meteorological conditions, such as temperature, humidity, wind speed, radiation, and the boundary layer. Changes in meteorological conditions, such as higher temperature, can enhance chemical reactions and increase the release of biogenic emissions. Relative humidity and cloud coverage indicate surface radiances that affect the rate of photolysis. Mixing depth is a key factor in diluting or accumulating pollutants and their precursors. The transport of pollutants and precursors is also controlled by the local and regional wind fields. Synoptic meteorology envelopes all these conditions; a change of synoptic weather pattern is accompanied by dramatic change in these parameters. Therefore, understanding the relationship between synoptic weather and air pollution will help assess the contribution of meteorology and climate to regional air quality (Beaver and Palazoglu, 2009; Chen et al., 2008; Cheng et al., 2007; Liao et al., 2020; Pearce et al., 2011; Triantafyllou, 2001).

Weather types are usually classified based on characteristics of varying types of air mass. Spatial Synoptic Classification (SSC; <http://sh Sheridan.geog.kent.edu/ssc.html>) (Sheridan, 2002) is one of several methods that provide a daily synoptic weather classification product. Since its initial suggestion (Kalkstein et al., 1996), the SSC has a rich legacy in applications for many topics and many regions around the world, including South Korea, and in studies of weather, climate, pollution, and health impacts. Since the SSC mainly uses the thermal properties of an air mass, it has been used to investigate many extreme heat waves and the related epidemiology and the construction of warning systems (Dixon et al., 2016; Hondula et al., 2014).

In South Korea, Kalkstein et al. (2011) developed a national heat-health warning system in collaboration with the Korea Meteorological Administration. Kim (2019) used the SSC in South Korea to demonstrate how synoptic weather impacts topics of epidemiological study. Since many of these studies have focused on the case of extreme heat, they concluded that the occurrence of high temperature air masses (i.e., DT and MT; see Section 2.1) are important to public health. Kim and Yum (2010) used synoptic classification to study the local and synoptic characteristics of the fogs that have formed over Incheon International Airport on the west coast of Korea. Y. Kim et al. (2016) discussed the association of new particle formation according to the displacement of synoptic weather systems. The SSC has also been used in

air-quality studies in South Korea, especially those concerning ozone (Cakmak et al., 2016; Hanna et al., 2011; Kim et al., 2014) and PM_{2.5} (Cakmak et al., 2018, 2016; Liu et al., 2017).

In this study, we aim to investigate the relationship between synoptic weather patterns and surface ozone concentration over South Korea. Following the approaches of Kim et al. (2014), we explore several topics: (1) determining the dominant synoptic weather type for high ozone occurrence, (2) estimating the efficiency of high ozone occurrence, and (3) examining the implications of long-term variations in synoptic weather types for regional air quality. The remainder of this paper is structured as follows. Section 2 explains the data and methodology. Section 3 presents analyses on the characteristics of synoptic weather and surface ozone, as well as their long-term variation. Section 4 concludes.

2. Data and methodology

2.1. SSC

Synoptic weather includes meteorological phenomena that happen on a horizontal length scale of the order of 1000 km or more. The daily weather types on the synoptic scale control many meteorological conditions that are critical in the formation, dispersion, and transport of pollutants. The SSC, a clustering scheme for synoptic weather patterns, has been developed as an analytical tool that can be applied to many natural and socioeconomic issues, including health impacts (Hondula et al., 2014; Lee et al., 2012; Vanos et al., 2014), air pollution (Hebborn and Cakmak, 2015; Hu et al., 2010; Kim et al., 2018; Sheridan et al., 2008; Sullivan et al., 2015), vegetation production, and past and future variations in climate (Bentley et al., 2010; Chow and Svoma, 2011; Hondula and Davis, 2011; Sheridan, 2003). The SSC has been used for global applications, in North America (Delavau et al., 2015; Dyer and Mote, 2007; Liu et al., 2013), Europe (Bower et al., 2007; Makra et al., 2015), and East Asia (Cakmak et al., 2016; Hanna et al., 2011; Kim et al., 2014).

For the present study, we obtained daily SSC classifications at 17 airport meteorological monitoring sites in South Korea directly from the SSC homepage (<http://sheridan.geog.kent.edu/ssc.html>). The SSC utilizes synoptic-scale, daily classifications of different types of air masses based on the thermal and moisture characteristics of each type. Seven meteorological variables classify each day's synoptic pattern: afternoon surface temperature, dew point, dew point depression, wind speed, wind direction, cloud cover, and diurnal temperature range. The SSC algorithm compares the surface observations of each variable with so-called "seed days" that are representative of various air masses for a given day and location. Since seed characteristics vary by time and location, the SSC provides a relative classification system (Sheridan, 2002).

The SSC includes nine synoptic weather types, six main categories and three additional or transitional categories. DP (dry polar) denotes an air mass originating from polar regions and is usually associated with low temperature and low humidity. It is synonymous with the traditional cP (continental polar) air mass classification. DM (dry moderate) air mass has mild and dry characteristics and is often found with zonal flow in the middle latitudes, especially on the leeside of mountains. DT (dry tropical) is the hottest and driest air mass, similar to the traditional cT (continental tropical) air mass. MP (moist polar) air is humid and cool like the traditional mP air mass type, and MM (moist moderate) is assigned in the case of considerably warmer and more humid air. MT (moist tropical) air is warm and very humid (i.e., the traditional mT air mass), often found in the warm section of mid-latitude cyclones. Extreme cases of the MT type are separated into MTP (moist tropical plus) and MTDP (moist tropical double-plus). Most MTP or MTDP cases happen during summer. Days transitioning between one weather type and another, considered to be TR (transitional), are characterized by large shifts in pressure, dew point, and wind pattern. The list and geographical coverage of the meteorological stations used in the study

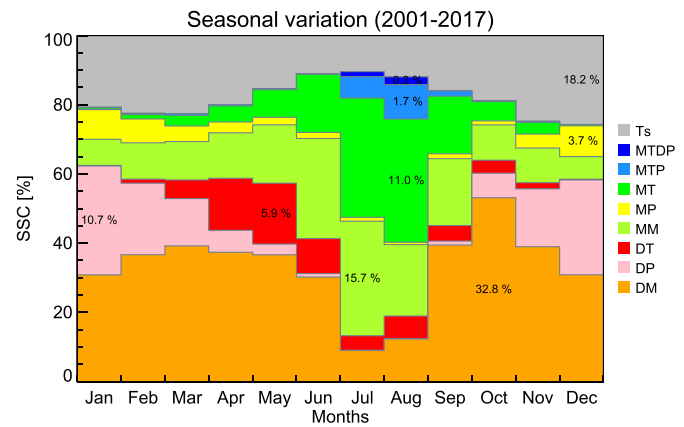


Fig. 1. Seasonal variation of the SSC types in South Korea, 2001–2017.

are shown in Table S1 and Figure S2.

2.2. Observations

Hourly observations of surface ozone concentrations from 2001 to 2017 were collected from AirKorea (<http://www.airkorea.or.kr/>). Data from 306 surface monitoring sites (251 urban, 38 roadside, and 17 background sites) were selected after screening their data quality and availability. Meteorological observations for temperature, dew point, relative humidity, wind speed, cloud fraction, and surface pressure were collected from the surface monitoring sites operated by the Korea Meteorological Administration (KMA; available from the data archive portal at <http://data.kma.go.kr/>). Surface ozone concentrations and meteorological variables were paired with the SSC by assigning the closest SSC site locations to determine daily SSC weather.

3. Results

3.1. Synoptic weather in South Korea

South Korea has distinctive seasonal variations in weather, as the country is affected by both continental and maritime air masses. South Korea is in the Asian monsoon regime, affected by cold air masses from the Asian continent during winter and warm and moist air masses of tropical origin during summer. In traditional air mass classification, several air masses affect South Korea: the Siberian air mass (cP), Okhotsk sea air mass (mP), North Pacific air mass (mT), Equatorial air mass (cT), and Yangtze River air mass. In spring, the Yangtze River air mass is dominant, with migratory cyclones and anticyclones. The routine passage of migratory systems—also known as *three cold days* and *four warm days* in Korean idiom—and the emergence of cases of Asian dust characterize the Korean spring. In early summer, the Okhotsk sea and North Pacific air masses are strong, often causing a rainy season when the strengths of two air masses are balanced. In the summer, the North Pacific air mass is dominant; in the winter, the Siberian air mass is dominant, with prevailing northwesterly wind patterns.

These seasonal variations are reflected in the seasonal SSC patterns. Fig. 1 shows seasonal variation in the SSC as determined from 17 meteorological monitoring sites in South Korea. As expected, the general synoptic weather patterns in South Korea are clearly seasonal. DM is a common weather type in all seasons except July and August when the East Asian summer monsoon is dominant. During the cold season, polar types (DP and MP) prevail as the strong high-pressure system over Siberia (the Siberian High) often extends, and the transboundary movement of the synoptic system affects South Korea. Spring is characterized by consistent passages of transboundary systems, both cyclonic and anticyclonic. Interestingly, from March to May, DT has a remarkable presence during spring. Moist types (MM and MT) are

Table 1

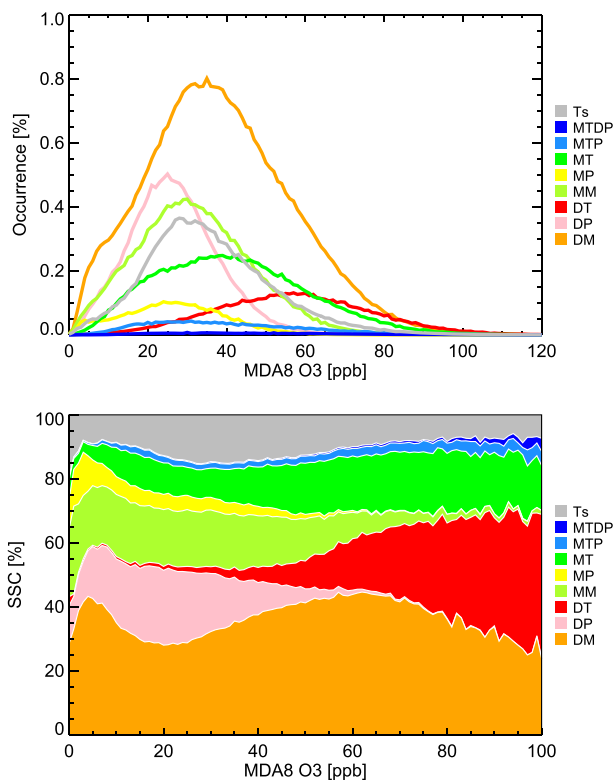
Frequencies of the SSC weather types in South Korea, 2001–2017. Unit: %.

	DM	DP	DT	MM	MP	MT	MTP	MTDP	Ts
January	30.8	31.5	0.2	7.6	8.6	0.5	0.2	0.0	20.7
February	36.7	20.7	1.2	10.5	6.9	1.2	0.4	0.1	22.4
March	39.2	13.7	5.4	11.1	4.5	3.1	0.4	0.0	22.6
April	37.4	6.3	15.1	13.1	3.1	4.7	0.3	0.1	19.9
May	36.6	3.1	17.6	16.9	2.2	8.0	0.2	0.1	15.3
June	30.2	1.0	10.1	28.9	1.7	16.8	0.2	0.0	11.0
July	9.0	0.0	4.2	33.1	1.1	34.5	6.2	1.4	10.4
August	12.3	0.1	6.5	20.6	0.7	35.6	9.9	2.2	11.9
September	39.5	1.1	4.5	19.3	1.4	16.7	1.5	0.1	15.9
October	53.1	7.1	3.8	10.2	1.2	5.6	0.2	0.0	18.8
November	39.0	16.7	1.8	10.0	4.0	3.4	0.3	0.0	24.8
December	30.9	27.4	0.2	6.5	8.9	0.4	0.0	0.0	25.7
Total	32.8%	10.7%	5.9%	15.7%	3.7%	11.0%	1.7%	0.3%	18.2%

Table 2

Statistics of ozone and meteorological variables for each SSC type.

	DM	DP	DT	MM	MP	MT	MTP	MTDP	Ts
T (C)	11.11	1.37	17.96	17.19	5.47	22.91	25.91	27.43	10.63
TD (C)	3.91	−7.26	9.41	12.75	0.08	18.46	21.63	23.56	3.83
RH (%)	64.72	56.35	62.43	77.16	70.67	77.88	79.11	81.29	66.49
WS (m/s)	2.14	2.84	2.01	2.10	2.83	1.99	2.11	2.34	2.10
CLD (1–10)	4.22	3.89	3.76	7.43	6.76	6.30	5.76	5.21	5.13
P (hPa)	1017.98	1022.03	1012.44	1012.75	1020.28	1010.08	1009.59	1008.30	1017.12
MDA8 Ozone (ppb)									
mean	38.8	27.1	57.3	32.3	27.1	41.9	40.4	48.4	36.1
std. dev	18.5	11.1	19.6	14.5	12.8	18.8	19.7	23.3	15.9

**Fig. 2.** (a) Histogram of the MDA8 ozone frequency distribution for each SSC type in South Korea, 2001–2017. Y-axis is normalized across all data. (b) Fractional coverage of each SSC type in 1 ppb ozone bins.

dominant in summer. The start of the summer season happens alongside the summer monsoon season. In this season, the wind flow pattern also changes, and marine flow patterns characterize the humidity of the airshed.

Table 1 summarizes the frequencies of each type of SSC occurring over South Korea from 2001 to 2017. Annually, DM is the dominant type, accounting for 32.8% of synoptic patterns, followed by MM (15.7%) and MT (13%, including MTP and MTDP). DT and MP happen rarely, with frequencies of 5.9% and 3.7%, respectively. SSC types display clear seasonal variation, as the displacement of air masses is strongly affected by the development of monsoons. During the monsoon season, flow patterns change dramatically because of the different heat capacities of ocean and land.

The meteorological characteristics of each type of SSC are summarized in Table 2. As expected from the classification method, each type represents distinctive thermal and moisture properties. Warm air masses (e.g., DT, MT, MTP, and MTDP) have higher temperatures, while dry air masses (e.g., DM, DP, and DT) have drier and less cloudy conditions. Notably, the DT type has the lowest cloud fraction (37.6%) and wind speed (2.01 m/s), providing stagnant conditions with high photolysis potential.

3.2. SSC and high ozone occurrence

Next, we analyze the connection between the occurrence of high ozone and synoptic weather patterns. Fig. 2 demonstrates the extent to which the occurrence of surface ozone differs by SSC weather type. We count the number of occurrences in each 1 ppb ozone bin for each SSC type. A total of 105,553 data points are used for the analyses, and Fig. 2a shows the normalized distribution of MDA8 ozone for each SSC type. Summing the occurrences for all eight SSC types yields 100%. While DM occurs the most frequently throughout the whole concentration range, with a mean value of 38 ppb, the distribution of DT, which occurs less frequently, is skewed toward higher ozone concentrations, with a mean value of 57 ppb. The other types also show distinctive statistical characteristics. Table 2 also presents statistics of MDA8 ozone concentrations for each SSC type.

The link between SSC type and ozone level is clearly presented in Fig. 2b, which shows the occurrence frequency of each SSC type for 1 ppb bins up to 100 ppb. In most cases, DP is associated with lower ozone

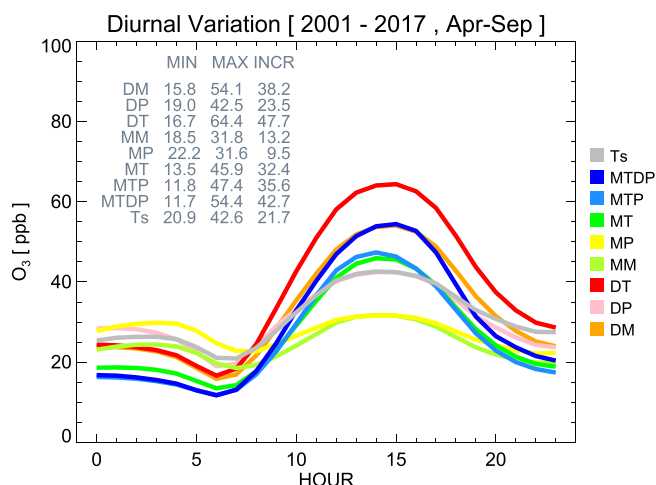


Fig. 3. Diurnal variations in surface ozone concentrations according to the corresponding SSC types during the 2001–2017 warm season (April to September). Minimum, maximum, and increase (i.e., Max-Min) of ozone diurnal variations are also shown for each SSC type (unit: ppb).

concentrations, showing seasonal dependency. MM, on the other hand, happens more during summer but is strongly associated with lower ozone, implying that moist conditions (e.g., more cloud) are not favorable for ozone production. For high ozone concentrations, DT, DM, and MT are the three dominant types, accounting for more than 80% of all events exceeding 80 ppb. DT and MT, both characterized by high temperatures, are associated with high ozone concentrations, but they differ in terms of the efficiency in the high concentration ozone occurrence. A large proportion of high-ozone events happen during the DT synoptic type (dry with high temperatures). This reconfirms that sunlight is critical in ozone formation. Forty-four percent of 100 ppb or higher ozone events happened in the DT type (compared with 25% in DM and 20% in MT), so we conclude that the DT synoptic type has the highest efficiency of high concentration ozone occurrence.

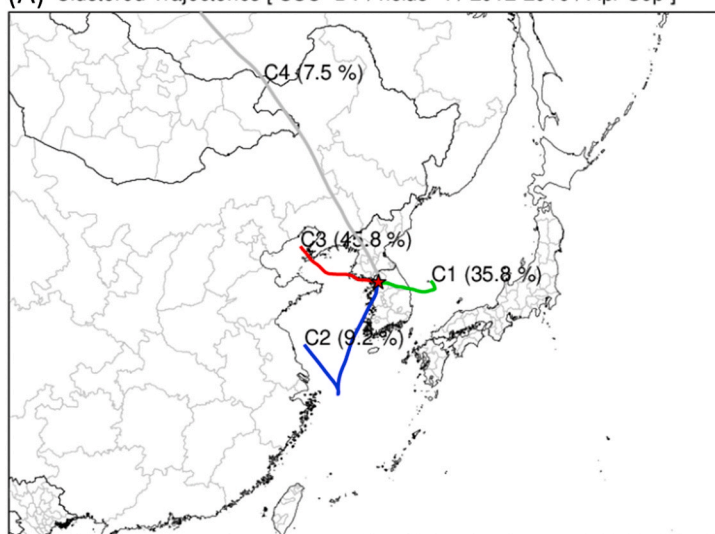
The ozone-production mechanism is usually associated with warmer conditions. High temperature directly enhances chemical processes and biogenic emissions and is also associated with less cloudy conditions, which yield stronger photochemistry. However, with this current method of analysis, we cannot totally exclude the potential contribution

of other factors. For example, DT happens frequently during spring, coincident with the dominant westerly flows over South Korea. Therefore, the efficiency of the DT type for high ozone occurrence may include the transport of ozone and/or its precursors from Chinese emission sources.

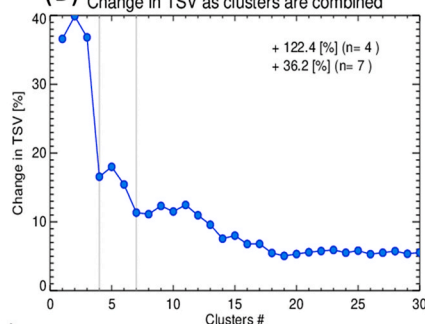
Separating the impacts between local formation and regional transport would be valuable information for regional chemistry and emissions control policy. Although we cannot separate these factors quantitatively at this point, we can explore the characteristics of the high concentration ozone occurring in the region. First, Fig. 3 shows the diurnal variations in hourly ozone concentrations according to each SSC weather type. Consistent with our previous analysis, the diurnal variations in surface ozone concentrations during the warm season show distinctive features for each SSC type. As expected, DT is associated with the highest level of surface ozone during the warm season (i.e., April to September), followed by DM and MT group (MTP and MTDP). Nighttime (including early morning) ozone concentrations in DT are not specifically high compared with those of the other SSC types, suggesting no evidence of elevated background concentrations. During DT synoptic weather, ozone concentration increases by 47.7 ppb from morning (16.7 ppb) to the afternoon peak (64.4 ppb), showing a strong capability of ozone formation. The lowest increase, 9.5 ppb, happens in the MP weather type, which characterizes humid and cold air masses. High daily peak concentrations are mostly associated with a rapid daytime increase, suggesting the role of local formation.

Second, the transport pathways for each SSC type are also analyzed. We conduct five-year (2012–2016) backward trajectory simulations using the NOAA HYSPLIT model (Stein et al., 2015). The daily trajectory simulations reaching the GMP monitor (Gimpo International Airport at a 100-m altitude at midday local time) are classified based on their spatial variances (Stunder, 1996). Fig. 4a show the distributions of the clustered trajectories of the DT synoptic type in the warm season (April–September) during 2012–2016. Four cluster numbers are chosen based on the change in total spatial variance (Fig. 4b). Except a small number of long-range transport cases (7.5%), most trajectories originate from nearby locations, showing a weak wind field during the DT weather type; 55% of trajectories are from Chinese sources (45.8% from northern China and 9.2% from southern China); and 35.8% trajectories from the East Sea (i.e., easterly flow). For these clustered trajectory groups, we calculate the mean MDA8 O₃ concentrations (137 sites in the Seoul metropolitan area). The results show no clear dependency of ozone

(A) Clustered Trajectories [SSC=DT / nclus=4 / 2012-2016 / Apr-Sep]



(B) Change in TSV as clusters are combined



(C)

Cluster	Frequency	MDA8 O ₃ [SMA]
C1	35.8 %	59.7 ppb
C2	9.2 %	63.2 ppb
C3	45.8 %	59.9 ppb
C4	7.5 %	55.0 ppb

Fig. 4. (a) Clustered backward trajectories reaching Seoul, South Korea during the 2012–2016 warm season (April to September) for the DT type, (b) Changes in total spatial variances to decide the number of clusters, and (c) occurrence frequencies and associated MDA8 ozone concentration for each cluster.

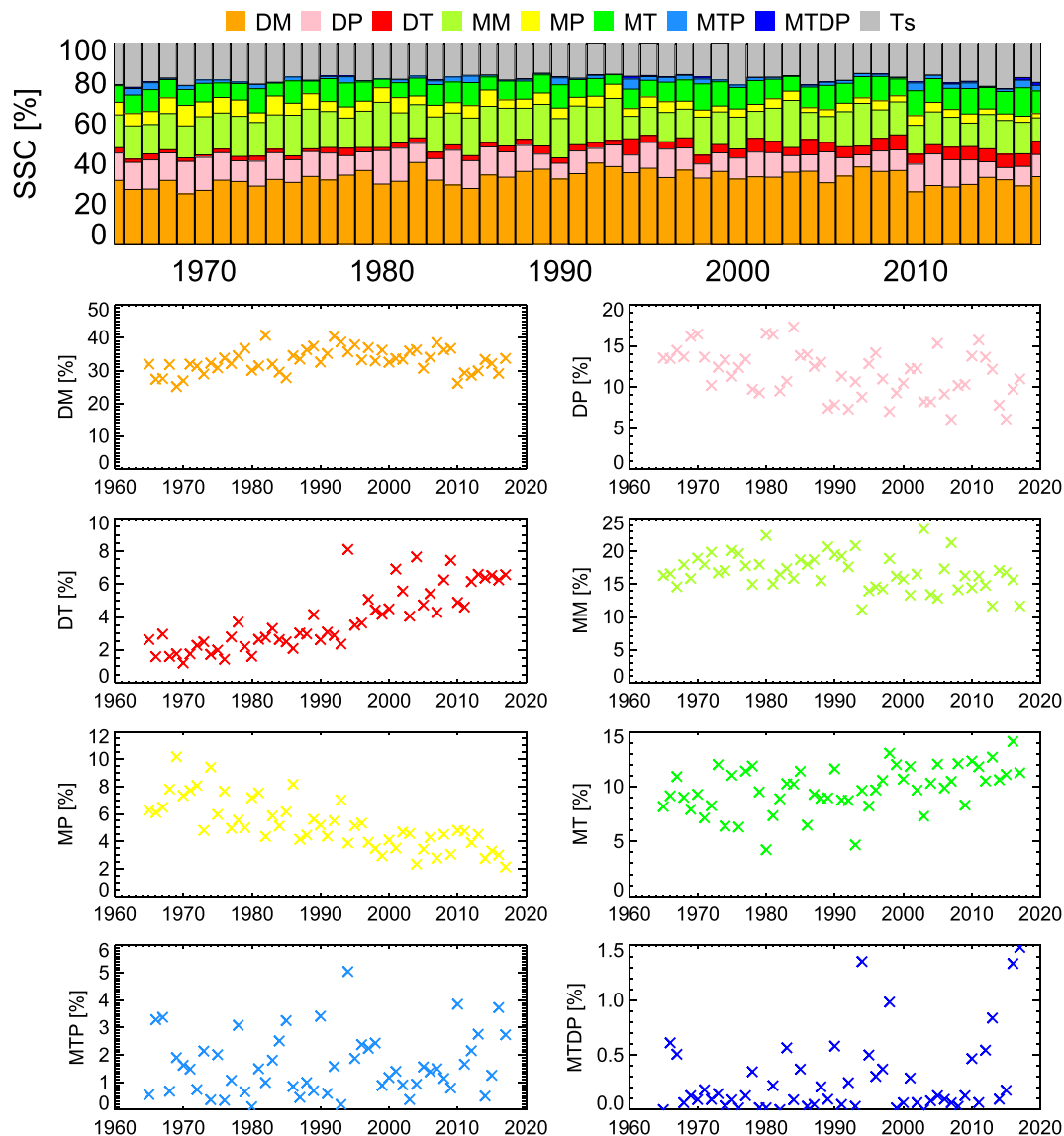


Fig. 5. Interannual variation in SSC types in South Korea, 1965–2017. Normalized to total annual counts in each year. Fractional contributions (upper) and individual SSC (lower) are shown.

concentration on flow pattern direction; the highest concentration is from southern Chinese sources (63.2 ppb) and the lowest is from the long-range case (55.0 ppb) (Fig. 4c). This also implies the likely role of local formation rather than transport from outside.

3.3. Long-term variation in SSC types

Having demonstrated that the production of surface ozone is affected by synoptic weather patterns, we further investigated the long-term variation in SSC types. As the previous analysis showed, certain synoptic weather patterns are tightly associated with the occurrence of high ozone concentration. This clearly implies that changes in synoptic weather can be directly associated with the occurrence of pollution, showing the crucial connections between pollution and weather and between pollution and climate.

To test this, we further investigate the long-term variation in the occurrence frequency of synoptic patterns over the past 50 years. Fig. 5 shows a time series of SSC interannual variation from 1965 to 2017. There are several noticeable changes in the occurrence of SSC types, implying that the South Korean region has been affected by long-term changes in climate. Two noticeable variations are the increasing trend

in DT and MT and the decreasing trend in MP, DP, and MM. DT occurred with an approximately 2% frequency in the 1960s and 1970s but increased up to an 8% frequency in the past two decades. The increase in DT appears to become especially strong after 2000.

Evidently, the long-term variation in SSC types in South Korea is associated with changing climatological characteristics, with an increasing trend in tropical air masses and a declining trend in polar air masses. This is consistent with the findings from the previous literature based on long-term observations (Chung et al., 2004; Chung and Yoon, 2000; H.-S. S. H.-S.S. Kim et al., 2016; Yun et al., 2012) and models (Jeung et al., 2019; Kwon et al., 2007; Yun et al., 2012). Long-term reanalysis datasets also confirm the consistent result in the changes in meteorological variations over South Korea (Fig. 6). We analyze the long-term variations in 2-m temperature and relative humidity over Korea (i.e. 126°E–130°E, 34°N–39°N) using six global reanalysis data sets: JRA-55 (1958–2017), ERA40 (1958–2001), ERA5 (1979–2017), ERA_interim (1979–2017), NCEP/NCAR R1 (1948–2017), and NCEP/NCAR R2 (1979–2017). All the reanalysis data show consistent trends, namely, increasing 2-m temperature and decreasing relative humidity, which characterize the meteorological properties in DT. This change in DT may be related to climate change and could imply

Reanalysis Time-Series

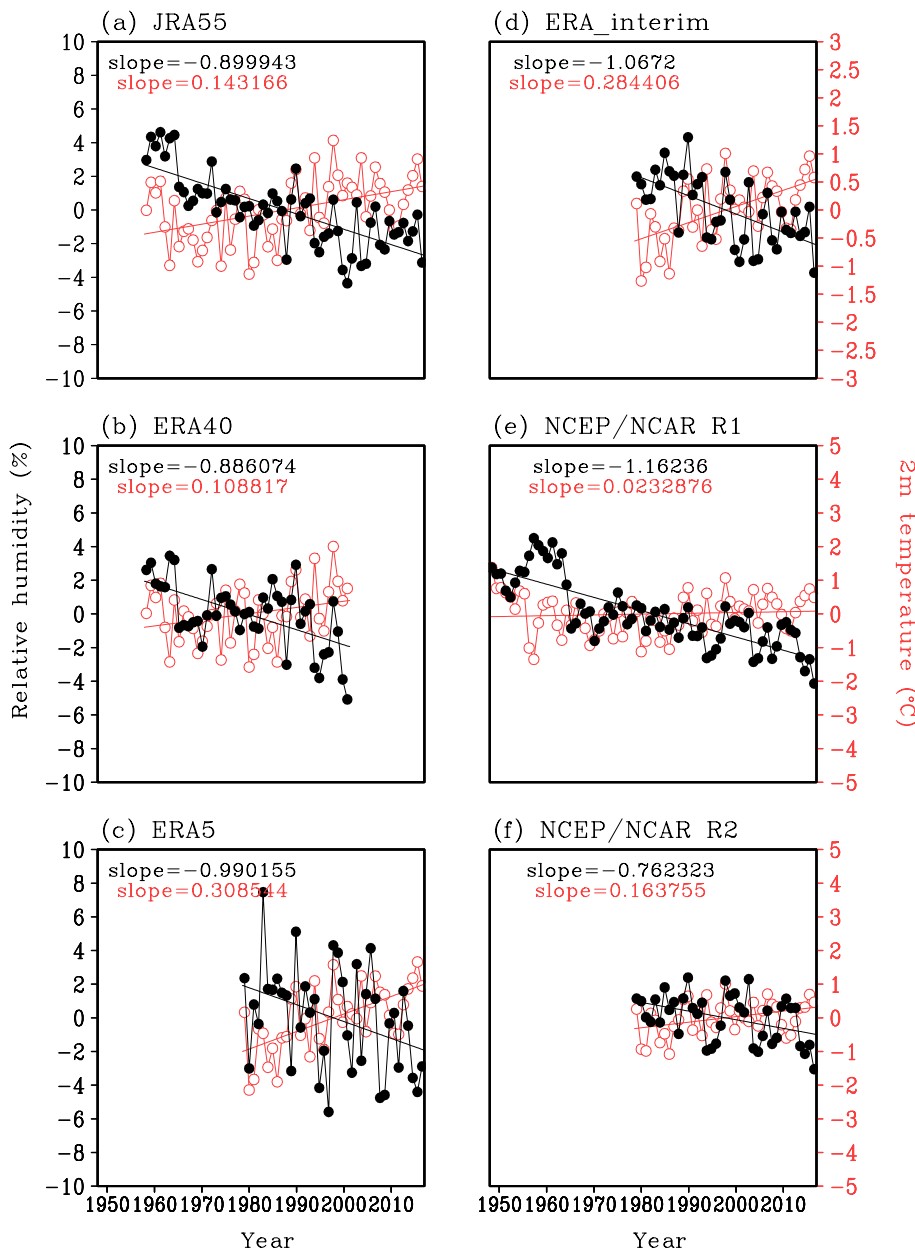


Fig. 6. Interannual variations in 2-m temperature (red line with open circles; °C) and relative humidity (black line with closed circles; %) over South Korea during 1958–2017. Anomalies of annual means and their linear fits are calculated using six global reanalysis datasets: (a) JRA-55 (1958–2017), (b) ERA40 (1958–2001), (c) ERA5 (1979–2017), (d) ERA_interim (1979–2017), (e) NCEP/NCAR R1 (1948–2017), and (f) NCEP/NCAR R2 (1979–2017). Units for slopes are °C 10 years⁻¹ (2-m temperature) and % 10 years⁻¹ (relative humidity). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

long-term changes in air quality. Recent studies have suggested that the increased temperature owing to climate change could weaken the meridional thermal gradient between the equator and poles, causing more stagnant conditions in the region and resulting in deteriorated regional air quality (H. C. Kim et al., 2017b; Mickley, 2004). The recent study by Lee et al. (2020) used CMIP5 single-forcing experiments to demonstrate that climate change can degrade regional air quality, by increasing static stability and reducing wind speed.

3.4. Ozone sensitivity to changes in SSC types

In this section, we estimate how surface-ozone distribution has changed as a result of the increased occurrence of the DT type. Quantifying the changes in surface ozone concentration as a response to incremental changes in the occurrence of the DT type helps explain the relationship between weather and pollution in the region. Although the

DT type occurs less frequently than the other types such as DM or MM, as described in the previous section, it contributes to high ozone production and its frequency has greatly increased in recent decades.

A complete estimate of the effect of changes in synoptic patterns is not easy to obtain, considering the presence of more complicated effects, such as the transport patterns of precursor emissions. However, the sensitivity of ozone production to SSC changes is testable by making two simple assumptions: (1) total occurrence counts are constant and (2) the shape of the ozone distribution for each SSC type is consistent.

Beginning from the probability distribution in Fig. 2a, we change the occurrence frequency of the DT type while preserving the total count of occurrences:

$$n_{total} = n_{DT} + n_{non-DT} = a \cdot n_{DT} + b \cdot n_{non-DT}$$

where n_{DT} and n_{non-DT} indicate the total ozone distribution for the DT type and seven other types, respectively, and a and b are adjustment

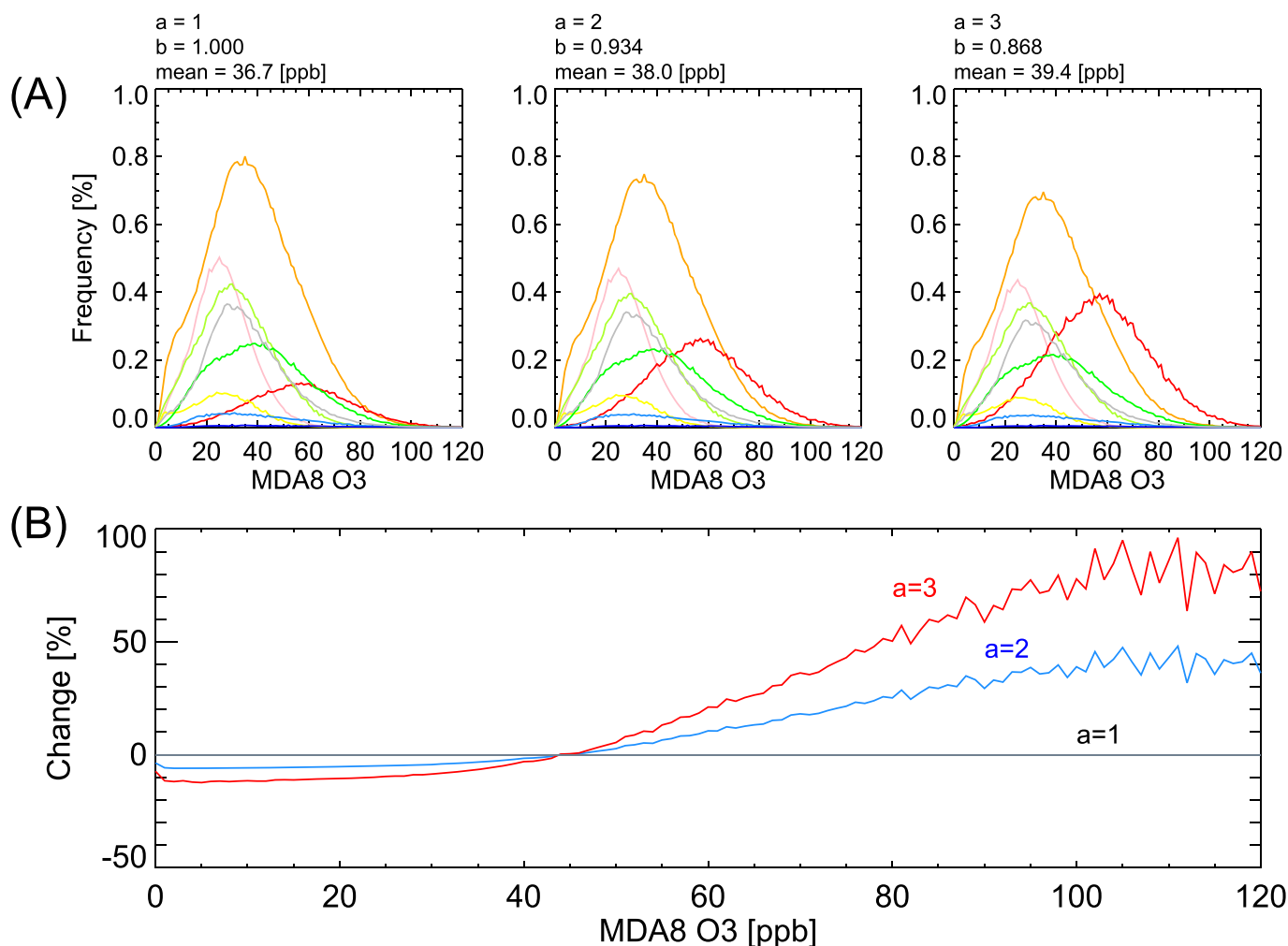


Fig. 7. Sensitivity tests of adjusting the frequency of the DT type. The changed MDA8 ozone distribution assuming original, doubled, and tripled (i.e., $a = 1, 2, 3$) DT occurrences are shown in the upper panel, and the relative changes in the MDA8 ozone distributions after applying the increased DT are shown in the lower panel.

coefficients for the sensitivity test. In this case, increasing the occurrence of DT types by applying $a = 2, 3$, the adjustment coefficient for the other types, b , will decrease, and we can specify its value if the total count, n_{total} , is preserved. It should be noted that we tested the frequency of DT type only. The impact may be larger if the MT type trend is also considered.

Fig. 7 demonstrates how this sensitivity test works. We obtained the non-DT coefficient $b = 0.934, 0.868$ when DT frequency is doubled or tripled (e.g., $a = 2, 3$) with the total count preserved. Since the DT type only account for a small proportion of the total distribution, doubling the DT distribution results in a 6.5% decrease in the other SSC types.

Making these adjustments, the general distributions of MDA8 ozone show noticeable changes in both average and peak values. The averages of MDA8 ozone are increased by 3.5% and 7.5% for $a = 2, 3$, respectively. Moreover, the high ozone-production efficiency of the DT type really alters the higher concentration levels. Fig. 7b presents the change in the MDA8 ozone distribution after doubling or tripling the prevalence of the DT type. At lower concentrations, MDA8 ozone decreases by around 5% or 10% for doubling and tripling, respectively. However, because of the highly skewed distribution of DT ozone, at higher concentration levels, MDA8 ozone increases. A transition happens near 45 ppb in both cases; the occurrence of 80 ppb ozone increases by 25.2% with doubled DT and by over 50.4% with tripled DT frequencies (31.7% and 63.3% for >80 ppb, cumulatively). The efficiency of DT type increases in the higher concentration levels. Higher-level ozone increases quickly—by almost 80%—when $a = 3$ is applied. This is a good example

of the high efficiency of the DT type in producing surface ozone; the DT type thus provides very favorable weather conditions for ozone production.

We do not attribute all the increase in this sensitivity test to the increased prevalence of DT. However, DT clearly provides favorable conditions for high ozone production. In addition, it should be noted that the SSC impact seems to be meteorological but does not include the potential impacts from the changes in transport patterns resulting from each SSC type. As DT represents a typical transport pathway from China, a more detailed investigation of transport impacts is therefore warranted.

4. Conclusion

In this study, we investigated the association between weather and pollution using surface ozone and SSC data over South Korea. We used the synoptic weather classification data to relate meteorological conditions to surface ozone concentrations. The analyses of daily maximum 8-h ozone revealed that DM, MT, and DT are the most common synoptic weather types associated with episodes of high ozone concentration. Among these, the DT type shows the highest correlation with such episodes, implying that air masses' thermal and moisture characteristics are associated with the formation of high ozone concentrations.

We further explored the long-term variation in synoptic weather types, with results demonstrating that the DT type has occurred more frequently in recent years. This increased pattern makes sense given

changes in typical weather in South Korea. Several reports have suggested that the climatological characteristics in South Korea are warming in general.

The findings from this study are summarized as follows:

- 1) Surface ozone distributions show distinctive characteristics for each synoptic weather pattern.
- 2) The occurrence of the DT type is strongly tied to the occurrence of high ozone concentrations. With MDA8 over 100 ppb, the frequency of the DT type is over 44%.
- 3) The long-term variation in the SSC weather types shows clear long-term trends in several of the SSC types. In particular, the DT type has increased in prevalence over the past 50 years. This further implies that any change in the occurrence of DT weather types in the anticipated climate change scenario could affect the development of regional air quality for all regions in the future.
- 4) We estimated the extent to which the change in the prevalence of the DT type affects the total distribution of surface ozone level and occurrence of high ozone levels. Sensitivity tests revealed that when the DT type doubles or triples in occurrence, mean ozone increases by 3% and 7%, respectively. For high ozone, this effect is even more significant.

Based on these findings, we conclude that changes in meteorological condition could have led to the changes in surface ozone concentration trends over South Korea. Notably, we do not exclude the impact of emissions changes on the current ozone trend. The currently observed increase in surface ozone concentrations in South Korea could combine impacts from changes in anthropogenic emission with meteorological conditions. Clearly, the direction of climate change in the region favors conditions for high ozone concentration. In summary, changes in weather type can play an important role in regional air quality. While limited, the findings of this study contribute to our understanding of how long-term and large-scale changes in climate are linked to short-term and local-scale pollution events.

Disclaimer

The scientific results and conclusions, as well as any views or opinions expressed herein, are those of the author(s) and do not necessarily reflect the views of NOAA or the Department of Commerce.

CRediT authorship contribution statement

Hyun Cheol Kim: Conceptualization, Formal analysis, Methodology, Visualization, Writing - original draft. **Dasom Lee:** Conceptualization, Formal analysis, Methodology, Visualization, Writing - original draft. **Fong Ngan:** Writing - review & editing. **Byeong-Uk Kim:** Writing - review & editing. **Soontae Kim:** Funding acquisition, Writing - review & editing. **Changhan Bae:** Writing - review & editing. **Jin-Ho Yoon:** Funding acquisition, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2020.117985>.

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