

Air quality trends and regimes in South Korea inferred from 2015–2023 surface and satellite observations

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Abstract.

Air pollutant emissions in South Korea have been changing rapidly over the past decade. Here we analyze the resulting 2015–2023 trends in air quality and regimes using surface (AirKorea), aircraft (KORUS-AQ), and satellite (low Earth orbit, geostationary) measurements. Surface concentrations of primary pollutants have decreased at rates (CO : $-2.6 \pm 0.7\% \text{ a}^{-1}$, SO_2 : $-6.4 \pm 0.8\% \text{ a}^{-1}$, NO_2 : $-4.4 \pm 0.8\% \text{ a}^{-1}$) consistent with the national CAPSS emissions inventory and satellite observations. CAPSS indicates no trend in volatile organic compound (VOC) emissions, consistent with satellite observations of formaldehyde (HCHO) and glyoxal (CHOCHO), but surface aromatic concentrations show a $5.0 \pm 3.9\% \text{ a}^{-1}$ decrease. Peak season (May–June) maximum 8-hour daily average (MDA8) surface ozone (O_3) exceed the 60 ppbv standard everywhere at AirKorea sites, with an increase of $0.8 \pm 0.9 \text{ ppbv a}^{-1}$ in the 90th percentile averaged across all sites indicating VOC-limited conditions for O_3 production. However, satellite HCHO/ NO_2 ratios indicate a shift from VOC- to NO_x -limited as NO_x emissions decrease. Most AirKorea sites is in the Seoul Metropolitan Area (SMA) where vestiges of VOC-limited conditions persist; we find no O_3 increases over the rest of Korea. Fine particulate matter ($\text{PM}_{2.5}$) has been decreasing at $5.0 \pm 1.6\% \text{ a}^{-1}$ but the nitrate (NO_3^-) component has not. Satellite NH_3/NO_2 ratios show that $\text{PM}_{2.5} \text{ NO}_3^-$ formation was NH_3 -sensitive before 2019 but is now becoming NO_x -sensitive as NO_x emissions decrease. Our results indicate that further NO_x emission decreases will reap benefits for both O_3 and $\text{PM}_{2.5} \text{ NO}_3^-$ as their production is now dominantly NO_x -sensitive.

1. Introduction

South Korea experienced rapid development over the past 30 years with an annual average GDP growth rate of 5% (S. Song and G. Lee, 2020). This has resulted in high emissions of carbon monoxide (CO), sulfur dioxide (SO₂), nitrogen oxides (NO_x ≡ NO + NO₂), nonmethane volatile organic compounds (NMVOCs), and particulate matter (PM) (Y. Kim and G. Lee, 2018). Subsequent atmospheric chemistry of these precursors produces surface ozone (O₃) and additional fine PM_{2.5} (less than 2.5 μm diameter), which are the main pollutants of concern for air quality. 30,000 premature deaths per year are presently attributed to air pollution in South Korea (hereafter referred to as Korea) (Oak et al., 2023; J. Choi et al., 2024). National air quality standards were tightened in 2018 for O₃ (60 ppbv maximum 8-hour daily average or MDA8) and for PM_{2.5} (15 μg m⁻³ annual, 35 μg m⁻³ 24-hour). None of the sites in the nationwide AirKorea governmental surface network meet the O₃ standard as of 2022, and only 4% meet the 24-hour PM_{2.5} standard, despite governmental efforts to decrease emissions.

The need to decrease emissions responsible for air pollution has been recognized since the 1980s, prompting early control policies to regulate solid fuel use and outdoor combustion, and promote clean fuels. This effectively reduced SO₂, CO, and directly emitted (primary) PM (Y. Kim and G. Lee, 2018). More recent efforts by the Korean Ministry of Environment (MOE) have targeted NO_x emissions. However, O₃ pollution has been getting worse at a rate of 1.0–1.5 ppbv a⁻¹ over 2000–2021 (S. W. Kim et al., 2023). PM_{2.5} has decreased though unevenly (J. Jeong et al., 2022; H. M. Lee et al., 2024; Pendergrass et al., 2022, 2024), with an increasing contribution from secondary components produced chemically in the atmosphere including secondary organic aerosol (SOA) and particulate nitrate (NO₃⁻) (H. M. Lee et al., 2024).

Synoptic meteorology and transport from China also contribute to seasonal and long-term variations of pollutants over Korea (D. Park et al., 2021; H. M. Lee and R. Park, 2022; J. Jeong et al., 2024). Photochemical O₃ production is largest during the summer, but the summer monsoon brings clean marine air masses into the Korean peninsula resulting in lower O₃ levels in July–August compared to May–June (Wie et al., 2018; H. M. Lee and R. Park, 2022). May–June has additional contributions to O₃ from wildfires, stratospheric intrusions, and transport from China (H. M. Lee and R. Park, 2022). PM_{2.5} is highest during the colder months (October–

March), due to shallow mixing depths and stagnant conditions over the Korean peninsula (J. Jeong et al., 2024), but here again transport from China makes an important contribution (D. Park et al., 2021). PM_{2.5} pollution in China has decreased considerably over the past decade in response to emission controls (Zhai et al., 2019) and this has decreased its influence on Korea (Bae et al., 2021). By contrast, O₃ pollution in China has gotten worse (K. Li et al., 2021).

Photochemical O₃ production takes place by oxidation of VOCs and CO in the presence of NO_x, and can be either NO_x- or VOC-limited depending on the concentrations of these precursors. Formation of PM_{2.5} NO₃⁻, which is a major component of wintertime secondary PM_{2.5} in Korea and is mainly present as ammonium nitrate, can be either NO_x- or ammonia (NH₃)-sensitive again depending on the concentrations of these precursors. These dependences define chemical regimes that are important to identify for emission control strategies. O₃ sensitivity to NO_x versus VOCs can be diagnosed using formaldehyde (HCHO) to NO₂ ratios measured from satellites, where HCHO and NO₂ are proxies for VOCs and NO_x emissions (Martin et al., 2004; Duncan et al., 2010). Similarly, PM_{2.5} NO₃⁻ sensitivity to NO_x versus NH₃ can be diagnosed using NH₃ to NO₂ ratios measured from satellites (Dang et al., 2023, 2024).

Satellites indeed offer a growing resource for monitoring air pollutants, trends, and regimes over Korea. These are mostly low-Earth orbit (LEO) instruments that observe at specific times of day including the Measurements Of Pollution In The Troposphere (MOPITT) (Edwards et al., 2004) and TROPOspheric Monitoring Instrument (TROPOMI) (Veefkind et al., 2012) for CO, the Ozone Monitoring Instrument (OMI) (Levelt et al., 2006) and TROPOMI for SO₂, NO₂, HCHO, and glyoxal (CHOCHO), and the Infrared Atmospheric Sounding Interferometer (IASI) (Van Damme et al., 2014) for NH₃. Additional geostationary instruments over East Asia with continuous hourly observations include the Geostationary Ocean Color Imager (GOCI) and GOCI-II for aerosol optical depth (AOD) (M. Choi et al., 2018; S. Lee et al., 2023). The Geostationary Environment Monitoring Spectrometer (GEMS), launched in February 2020, provides the first hourly observations of gases by solar backscatter including SO₂, NO₂, HCHO, and CHOCHO (J. Kim et al., 2020).

Here we analyze recent 2015–2023 trends in air quality in Korea by exploiting surface, airborne, and satellite observations to provide insights for the effectiveness of past regulation policies and future management. We interpret the trends in terms of the major drivers and evaluate consistency with annual bottom-up emission estimates from the Clean Air Policy

Support System (CAPSS) of the MOE (S. Choi et al., 2022). We start from 2015 when PM_{2.5} observations from the AirKorea network became available, with subsequent milestones including the May–June 2016 Korea-United States Air Quality (KORUS-AQ) field campaign (Crawford et al., 2021) and satellite observations from TROPOMI (starting in May 2018) and GEMS (starting in November 2020). We use HCHO/NO₂ and NH₃/NO₂ indicators from the satellite data to diagnose O₃ and PM_{2.5} chemical regimes and their trends.

2. Air quality observing system for South Korea

We make use of air quality observations in Korea from surface sites, aircraft, and satellites to analyze annual, diurnal, and spatial variations of pollutants. The National Institute of Environmental Research (NIER) operates the AirKorea surface network of 650 monitoring sites as of 2023 (<https://www.airkorea.or.kr/eng>, last access: 8 January 2025), providing hourly data on CO, SO₂, NO₂, O₃, PM₁₀ (smaller than 10 µm diameter), and PM_{2.5} concentrations. Monthly VOCs data (56 species) are available at a few urban sites. For annual trend analyses we use observations from AirKorea sites that have continuous records from 2015 to 2023. We use least-squares linear regression to calculate annual trends and their error standard deviations at 95% confidence level. The KORUS-AQ field campaign in May–June 2016 included a detailed chemical payload onboard the DC-8 aircraft with extensive vertical profiling at different times of day (Crawford et al., 2021). This was used by Yang et al. (2023) to infer diurnal profiles of NO₂ vertical column densities (VCDs) and we will do the same here for HCHO and CHOCHO. Details on data processing and trend analysis are described in the Supplement.

Satellite observations for air quality over Korea used in this work are compiled in Table 1. For all instruments we filter out cloudy scenes using a cloud fraction threshold of 0.3. We further filter out geostationary observations with a solar zenith angle larger than 70°. Additional quality filtering is summarized in Table S1. CO is retrieved in both the shortwave and thermal infrared (SWIR and TIR). NH₃ is retrieved in the TIR. All other gases are retrieved in the ultraviolet-visible (UV-VIS). Tropospheric O₃ can also be retrieved in the UV but the measurements are difficult because of air scattering and the stratospheric column overhead, and different products are inconsistent over Korea (Gaudel et al., 2018), therefore we do not use them here. GOCI and GOCI-II AOD retrievals are for 550 nm wavelength.

OMI and TROPOMI make afternoon overpasses at 13:30 local time (LT). We make use of morning overpasses for MOPITT (10:30 LT) and IASI (9:30 LT). We use hourly daytime observations from GEMS (7:45–16:45 LT), GOCI (9:30–16:30 LT), and GOCI-II (8:15–17:15 LT). For annual trend analyses we use GEMS observations made between 12–14 LT for consistency with the overpass time of OMI and TROPOMI measuring the same gases. We find no significant differences in observed trends when using surface observations sampled at satellite overpass times and therefore use all hours of the day.

Table 1. Satellite observations used in this work.

Instrument	Launch	Species ^a	Spatial resolution ^b	Version	Reference
<i>Low Earth orbit</i>					
MOPITT	1999	CO	$22 \times 22 \text{ km}^2$	V9	Deeter et al. (2022)
OMI	2004	SO ₂ , NO ₂ , HCHO, CHOCHO ^c	$13 \times 24 \text{ km}^2$	V3	González Abad et al. (2015); Krotkov et al. (2017); C. Li et al. (2020); Kwon et al. (2024)
TROPOMI	2017	CO, NO ₂ , HCHO	$5.5 \times 3.5 \text{ km}^2$	V2.4.0	Landgraf et al. (2016); De Smedt et al. (2018); van Geffen et al. (2022)
IASI	2006	NH ₃	$12 \times 12 \text{ km}^2$	V4	Clarisse et al. (2023)
<i>Geostationary orbit</i>					
GEMS	2020	SO ₂ , NO ₂ ^d , HCHO, CHOCHO	$3.5 \times 7.7 \text{ km}^2$ at 37.5° latitude	V2.0.0	NIER (2020); Ha et al. (2024); G. T. Lee et al. (2024); Oak et al. (2024)
GOCI	2011	AOD	$2 \times 2 \text{ km}^2$ ^f	GOCI-II	M. Choi et al.
GOCI-II	2020	AOD ^e	$2.5 \times 2.5 \text{ km}^2$	YAER ^g	(2018); S. Lee et al. (2023)

^a Total atmospheric columns except for NO₂ (tropospheric column).

^b Native pixel resolution of retrieval.

^c Provided at $1^\circ \times 1^\circ$ by Kwon et al. (2024).

^d Bias-corrected by Oak et al. (2024).

^e Bias-corrected by applying a scale factor of 1.047 to account for low bias in GOCI-II. See Supplement for details.

^f Resolution of aggregated pixels for final aerosol product.

^g Yonsei Aerosol Retrieval.

3. Air quality distributions and trends in South Korea

Here we analyze spatial distributions and temporal trends of individual air pollutants using surface and satellite observations, and compare the trends to the annual bottom-up estimates of anthropogenic emissions from CAPSS, reported with a two-year lag (<https://www.air.go.kr/eng/main.do>, last access: 8 January 2025). CAPSS includes city/county/district (Korean; si/gun/gu) level emissions for source categories including fuel combustion, manufacturing, solvent use, mobile sources, agriculture, and anthropogenic biomass burning (biofuel, agriculture).

Figure 1 shows major anthropogenic source regions in Korea. There are seven major cities with populations larger than one million. The Seoul Metropolitan Area (SMA; $37\text{--}37.8^\circ\text{N}$, $126.4\text{--}127.5^\circ\text{E}$) is the largest urban area which includes Seoul, Incheon, and surrounding suburbs, with concentrated electronics and chemical industry. The southeast region including Busan and Ulsan is the second largest urban area and has petrochemical facilities, oil refineries, and steel/ship/automobile manufacturing industries.

3.1. Carbon monoxide (CO)

CO levels in Korea have consistently remained below the national air quality standards (9 ppmv 8-hour, 25 ppmv 1-hour) since the late 1990s (NIER, 2023). CO is nevertheless a useful tracer of pollution and plays an important role in driving ozone formation in Korea (Gaubert et al., 2020; H. Kim et al., 2022). Anthropogenic CO emissions in CAPSS are 45% from transportation (passenger vehicles, heavy-duty vehicles, ships) and 32% from biomass burning (agricultural waste incineration, biofuels). Figure 2a–c compares 2021 CAPSS CO emissions with 2023 average surface CO and TROPOMI VCDs. Concentrations are highest in urban and industrial areas, but there is also a relatively high background reflecting the long atmospheric lifetime of CO (2 months on average). Low VCDs over mountainous areas are due to surface

elevation reducing the background column. This effect of surface elevation on VCDs is less apparent for shorter-lived species with weaker background contributions.

Figure 2d shows 2015–2023 CO trends, demonstrating consistency between CAPSS and atmospheric observations. CAPSS emissions and AirKorea surface concentrations decrease at similar rates of -2.3 ± 1.7 and $-2.6 \pm 0.7\% \text{ a}^{-1}$. MOPITT decreases at a rate of $-0.9 \pm 0.5\% \text{ a}^{-1}$, slower than surface concentrations because of the background contribution to the VCDs ($\sim 2 \times 10^{18} \text{ molecules cm}^{-2}$). Chong et al. (2023) previously found a MOPITT CO decrease of $-0.6 \pm 0.1\% \text{ a}^{-1}$ during 2005–2018. It is estimated that Chinese emissions contributed 21–25% to the downward trend between 2016 and 2022 (J. Park et al., 2024; E. Kim et al., 2024).

3.2. Sulfur dioxide (SO₂)

SO₂ levels in Korea have consistently remained below the national air quality standards (20 ppbv annual, 50 ppbv 24-hour) over the past two decades due to large reductions of emissions from power plants and the petrochemical industry (NIER, 2023). However, there is continuing motivation for emission controls because SO₂ is a precursor to PM_{2.5} sulfate (SO₄²⁻). Figure 3a–c compares 2021 CAPSS SO₂ emissions with 2023 average surface SO₂ and GEMS VCDs for all available observations. GEMS displays enhancements in the SMA, mid-south coast (power plants, petrochemical/steel industry) and northeastern regions (cement/concrete/pulp industry), consistent with OMI SO₂ hotspots previously identified for 2011–2016 (Chong et al., 2020).

Figure 3d shows good agreement between the CAPSS-reported emission trends and atmospheric observations. CAPSS-reported emissions have decreased at a rate of $-9.9 \pm 3.3\% \text{ a}^{-1}$, while surface SO₂ concentrations and OMI VCDs have decreased at similar rates of $-6\% \text{ a}^{-1}$ since 2015. However, there is large uncertainty in the satellite observations that likely contributes noise to the trend (C. Li et al., 2020). J. Park et al. (2024) found that recent trends (2016–2022) in national mean surface SO₂ were driven by reductions in both domestic (25%) and Chinese (16%) emissions, explaining the 41% decrease shown in Figure 3d.

3.3. Nitrogen dioxide (NO₂)

NO₂ levels exceeded the national standards (30 ppbv annual, 60 ppbv 24-hour) at 28% of the AirKorea sites in 2015 but fewer than 1% in 2022 (NIER, 2023). NO_x emissions in Korea are dominated by the transportation sector, which account for 64% of the CAPSS inventory. Control

of NO_x emissions is more recent than for CO and SO₂ and has been motivated not only by the NO₂ standards but also by the potential to reduce PM_{2.5} NO₃⁻. CAPSS NO_x emissions declined by 23% from 2015 to 2021 in response to policies including stronger regulation on heavy-duty diesel engines (S. Song and G. Lee, 2020) and governmental PM management plans implemented in 2019 (Bae et al., 2022; J. Jeong et al., 2024).

Figure 4a–c compares 2021 CAPSS NO_x emissions with 2023 average surface NO₂ and GEMS tropospheric VCDs. Here we use the Oak et al. (2024) GEMS product calibrated to TROPOMI to remove artifacts. Surface concentrations and VCDs display similar spatial distributions, with highest values in the SMA and other urban areas in the southeast. Figure 4d shows that surface NO₂ and OMI tropospheric VCDs have decreased over the 2015–2023 period by 32% and 36%, respectively. The trend in CAPSS-reported emissions ($-4.8 \pm 2.7\% \text{ a}^{-1}$) is consistent with surface observations ($-4.4 \pm 0.8\% \text{ a}^{-1}$) and OMI VCDs ($-4.6 \pm 0.8\% \text{ a}^{-1}$) during 2015–2023. Meteorology-corrected trends in tropospheric VCDs observed by ground-based remote sensing instruments at urban sites decreased at similar rates (-5.0 to $-5.4\% \text{ a}^{-1}$) during 2015–2020 (Y. Choi et al., 2023). Long-term (2005–2019) records show that significant decreases in surface and OMI NO₂ began in 2015 (S. Seo et al., 2021). CAPSS shows an increase from 2015 to 2016, which is due to updates in emission factors (S. Choi et al., 2020). E. Kim et al. (2024) found that only 2% of the observed 23% decrease in surface NO₂ during 2016–2021 over Korea was attributable to the Chinese contribution.

Figure 4e shows the 2021–2023 seasonal mean hourly variations of surface NO₂ and GEMS VCDs over the SMA. Both surface and column NO₂ are higher by a factor of two during the cold season, which can be explained by the longer NO_x lifetime (Shah et al., 2020). NO_x emissions in the SMA have small seasonal variations as they are dominated by mobile sources (Pandey et al., 2008; H. M. Lee and R. Park, 2022; Yang et al., 2024). The emissions are higher in the daytime (7–18 LT) than at night but do not show significant rush hour enhancements because traffic load is sustained with little variability throughout the daytime (Yang et al., 2024). Therefore, the peak in surface NO₂ concentrations at 8–9 LT is not due to the rush hour but to accumulation of daytime emissions in a shallow mixed layer (Moutinho et al., 2020). NO₂ then decreases in the morning by dilution as the mixed layer grows from solar heating, and increases again in the evening when the mixed layer collapses (J. Li et al., 2021). Increasing NO₂ photolysis as the morning progresses would also be expected to lower NO₂ concentrations but this is offset by

entrainment of O₃ from aloft as the mixed layer grows, such that the NO₂/NO_x ratio increases during the morning hours (Yang et al., 2024). Geostationary satellite observations provide unique information on the diurnal variation of NO₂ VCDs (Tian et al., 2018; Cheng et al., 2019; Edwards et al., 2024; Xu et al., 2024). This is illustrated in Figure 4e for the SMA. A NO_x budget analysis by Yang et al. (2024) showed that NO₂ VCDs in Seoul increase steadily in the morning from accumulation of emissions as they are not affected by mixed layer growth, reaching a steady state in the afternoon due mostly to loss from ventilation.

3.4. Nonmethane volatile organic compounds (NMVOCs)

NMVOC emissions include important contributions from both anthropogenic and biogenic sources. More than half of anthropogenic VOC (AVOC) emissions according to CAPSS are from solvent use while transportation is responsible for less than 10%, although the latter may be underestimated by a factor of 2–3 according to source apportionment studies (Y. Kim and G. Lee, 2018; S. Song et al., 2019; Kwon et al., 2021). CAPSS also does not account for residential emissions of volatile chemical products (VCPs), which could be large in Korea as indicated by observations of elevated ethanol during KORUS-AQ (Beaudry et al., 2024; Travis et al., 2024). Annual total AVOC emissions are estimated to be a factor of two larger than biogenic VOCs (BVOCs) on a national level (Jang et al., 2020). However BVOCs play an important role in O₃ and SOA formation during summer (H. K. Kim et al., 2018; Oak et al., 2022; H. M. Lee and R. Park, 2022), when their emissions are comparable to those of AVOCs (J. Choi et al., 2022).

Figure 5a–b compares 2021 total AVOC emissions from CAPSS and BVOC emissions calculated from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2012). The two have contrasting distributions, with AVOCs mostly urban and industrial. Shown in Figure 5c is the distribution of BTEX (≡ benzene + toluene + ethylbenzene + xylenes) concentrations observed at AirKorea sites, with high values over urban areas consistent with CAPSS. Benzene is elevated on the west and southern coasts where it originates from the steel industry, oil refineries, and petrochemical facilities (Y. Seo et al., 2014; Fried et al., 2020; C. Cho et al., 2021). Toluene, xylenes, and ethylbenzene are abundant in the SMA (S. Song et al., 2019; S.-J. Kim et al., 2021; Y. Lee et al., 2023) due to emissions from traffic and solvent use (Simpson et al., 2020).

Figure 5d–e shows spatial distributions of CHOCHO and HCHO VCDs from GEMS. These are common intermediates in the oxidation of NMVOCs, but CHOCHO is preferentially produced from aromatics (Kaiser et al., 2015; J. Li et al., 2016). Satellite observations are most sensitive to precursor NMVOCs with short lifetimes and prompt HCHO or CHOCHO yields including isoprene, alkenes, toluene, and xylenes (Palmer et al., 2003; Chan Miller et al., 2017; Bates et al., 2021). The GEMS CHOCHO and HCHO VCDs are elevated in major industrial regions, but CHOCHO shows hotspots for manufacturing industries (Incheon, Changwon) while HCHO shows hotspots for petrochemical facilities (Yeosu, Ulsan). HCHO observations are also more distributed, reflecting the larger BVOCs contribution from isoprene.

Figure 5f shows the CHOCHO to HCHO ratio $R_{GF} = \text{VCD}_{\text{CHOCHO}}/\text{VCD}_{\text{HCHO}}$, illustrating the contrast in their sources. R_{GF} is generally higher under anthropogenic dominance (Chen et al., 2023). Values range from 0.02 in rural regions to more than 0.05 in the SMA and Busan. In the US, R_{GF} values are below 0.03 even under polluted conditions (Chan Miller et al., 2017) and are down to 0.01 in rural regions with dominant biogenic sources (Kaiser et al., 2015). GEMS R_{GF} values in Korea are higher than 0.01 everywhere, indicating a more important role for AVOC emissions than in the US where these emissions have been strongly regulated for decades (Parrish et al., 2009; Warneke et al., 2012). Unlike for other pollutants and in contrast to the US, regulation of AVOCs in Korea has been limited (S. Song and G. Lee, 2020; J. Kim et al., 2023). Figure 5g shows no significant trends in AVOC emissions and satellite observations of CHOCHO and HCHO, although surface BTEX decreased at $-5.0 \pm 3.9\% \text{ a}^{-1}$ during 2015–2023.

Figure 6 compares diurnal variations of HCHO and CHOCHO VCDs in the SMA observed from GEMS (2021–2023) and DC-8 aircraft vertical profiles from near the surface to 8 km altitude during KORUS-AQ (May–June 2016). Computation of VCDs from the aircraft data is described in the Supplement. Mean loss frequencies of HCHO and CHOCHO against oxidation by OH and photolysis in the aircraft data average 0.42 h^{-1} and 0.61 h^{-1} , respectively at 11–15 LT. We find that the GEMS columns are lower than the aircraft columns, consistent with previously reported low biases in satellite retrievals of CHOCHO (–50%) and HCHO (–40% to –20%) (Zhu et al., 2016; Chan Miller et al., 2017; Zhu et al., 2020). HCHO VCDs are more than twice higher during the warm season (April–September) than the cold season (October–March), consistent with a biogenic contribution to HCHO, while CHOCHO VCDs show no seasonal difference. Both GEMS and aircraft show slight HCHO and CHOCHO increases in the

morning from photochemical production (G. T. Lee et al., 2024), flattening by midday. The aircraft data show a late afternoon rise in HCHO for which we have no explanation and might reflect sparse sampling.

3.5. Ozone (O₃)

None of the AirKorea monitoring sites met the MDA8 standard of 60 ppbv for O₃ as of 2022 (NIER, 2023). O₃ peaks in May–June in Korea (Figure 7a) with contributions from domestic emissions, wildfires, stratospheric intrusions, and transport from China (H. M. Lee and R. Park, 2022). Several studies have reported on the O₃ increase in Korea over the past two decades, using different O₃ concentration metrics and time periods (J. Seo et al., 2018; Yeo and Kim, 2022; S. W. Kim et al., 2023). We find that May–June 90th percentile MDA8 O₃ calculated for individual AirKorea sites and then averaged across all sites shows an increase of 0.8 ± 0.9 ppbv a⁻¹ during 2015–2023 (Figure 7b).

Previous studies found that O₃ formation in major cities in Korea is in the VOC-limited regime, where decreasing NO_x emissions causes O₃ to increase (S. Kim et al., 2018; Oak et al., 2019; Souri et al., 2020; H. J. Lee et al., 2021; S. W. Kim et al., 2023). However, as NO_x emissions have decreased (Figure 4) whereas VOC emissions have not (Figure 5), O₃ formation may shift to a NO_x-limited regime. The HCHO to NO₂ column ratio ($R_{FN} = \text{VCD}_{\text{HCHO}}/\text{VCD}_{\text{NO}_2}$), an indicator for O₃ sensitivity to NO_x versus VOCs (Martin et al., 2004; Duncan et al., 2010), increased steadily from 2015 to 2023 as seen from OMI, TROPOMI, and GEMS (Figure 7c). Recent studies over Northeast Asia suggest that NO_x-limited regimes are found where $R_{FN} > 2$ –3 (Ren et al., 2022; Jin et al., 2020, 2024). Here we use 2.5 as a threshold and find that Korea is now mostly in the NO_x-limited regime. Figure 7d–e shows May–June 2023 90th percentile MDA8 O₃ and its sensitivity regimes inferred from GEMS R_{FN} . Most of the country is in NO_x-limited and transition regimes while VOC-limited conditions are largely limited to the central SMA and Busan. The broader SMA and urban southeastern Korea are in a transition regime where O₃ is sensitive to both NO_x and VOC emissions. These latter regions experience the most severe O₃ pollution as both NO_x and VOCs contribute to O₃ formation.

Also shown in Figure 7b are May–June 90th percentile MDA8 O₃ trends for AirKorea sites in different sensitivity regimes based on the 2023 GEMS R_{FN} . The O₃ increase during 2015–2023 is only found in the VOC-limited areas (1.6 ± 0.8 ppbv a⁻¹). O₃ in NO_x-limited areas

does not show any noticeable increase. Reports of O₃ increases in Korea based on data from the AirKorea sites may be biased by the AirKorea sites being concentrated in the SMA, which has been mostly VOC-limited, but this is now changing as NO_x emissions decrease. Our analysis suggests that O₃ pollution in Korea is now poised to decrease everywhere in response to continued NO_x emission controls.

An additional challenge for Korea to meet its air quality standard is the high background originating from East Asia, estimated to be 55 ppbv (Colombi et al., 2023). During the COVID-19 lockdown in 2020 precursor emissions significantly dropped in China but not in Korea (Koo et al., 2020), which led to reduced long-range transport of O₃ and hence lower background levels over Korea (S. W. Kim et al., 2023). This could explain the large decrease in O₃ found between 2019 and 2020, especially in NO_x-limited areas which are more sensitive to background contributions than local emissions.

3.6. Particulate matter (PM)

PM levels have steadily decreased in Korea over the 2015–2023 period with more than 95% of the AirKorea sites meeting the annual PM₁₀ standard (50 µg m⁻³) since 2018. However, only 27% of sites met the PM_{2.5} annual standard (15 µg m⁻³) in 2022, and only 4% met the 24-hour standard (35 µg m⁻³) (NIER, 2023). Figure 8a–c compares 2023 average PM₁₀, PM_{2.5}, and GOCI-II AOD which share similar spatial distributions. GOCI-II (2021–2023) is biased low relative to GOCI (2015–2020), and therefore we apply a scale factor of 1.047 to the GOCI-II data (Lee et al., 2023). Annual trends in PM₁₀ ($-4.0 \pm 1.7\% \text{ a}^{-1}$), PM_{2.5} ($-5.0 \pm 1.6\% \text{ a}^{-1}$), and AOD ($-4.4 \pm 2.4\% \text{ a}^{-1}$) over Korea during 2015–2023 are consistent (Figure 8d). J. Park et al. (2024) found that 14% of the observed 33% decrease in PM_{2.5} during 2016–2022 over Korea was attributable to the Chinese contribution.

Figure 8e shows seasonal mean hourly variations of surface PM_{2.5} and GOCI AOD. Surface PM_{2.5} peaks in winter to early spring, mostly attributable to sulfate-nitrate-ammonium aerosols (Zhai et al., 2021) and is at its minimum in summer during the monsoon period (H. M. Lee et al., 2024). Conversely, AOD peaks in spring and summer (March–August) due to dust events, chemical production of secondary aerosols, and hygroscopic growth at high relative humidity (Zhai et al., 2021). PM_{2.5} peaks at 9–11 LT local time and then decreases until late afternoon as the mixed layer grows and dilutes surface concentrations (Jordan et al., 2020). AOD rises in the

morning and peaks in midday reflecting photochemical production (P. Kim et al., 2015; Lennartson et al., 2018).

2015–2021 PM_{2.5} observations in Seoul shows that all major PM_{2.5} components decreased except for NO₃⁻, which accounts for 25% of total PM_{2.5} during winter to early spring (H. M. Lee et al., 2024). Winter NO₃⁻ formation depends nonlinearly on NO_x and NH₃ emissions, with dominant sensitivity to either precursor that can be diagnosed from the NH₃/NO₂ VCD ratio and the NO₂ VCD in satellite observations (Dang et al., 2023, 2024). Figure 9a–b compares 2021 CAPSS NH₃ emissions and 2023 average NH₃ VCDs observed by IASI. 76% of anthropogenic NH₃ emissions in Korea originate from livestock manure management according to CAPSS. Transportation is also a significant source in urban areas (T. Park et al., 2023). Highest VCDs are found in the southern SMA, where livestock farming is concentrated, and corresponding to a PM_{2.5} hotspot (Figure 8b). Despite high NH₃ emissions in the southeast coast, VCD enhancements are not observed there due to high SO₂ emissions (Figure 3a) and expected high SO₄²⁻ production converting gas-phase NH₃ to particle-phase ammonium (NH₄⁺). Figure 9d indicates that annual total NH₃ emissions have shown little change while NH₃ VCDs have significantly increased since 2015. Decreases in SO₂ emissions and the resulting SO₄²⁻ in both Korea and China have left more NH₃ available for NO₃⁻ formation (J. Jeong et al., 2022).

Figure 9c shows NO₃⁻ sensitivity regimes inferred from GEMS NO₂ and IASI NH₃ VCDs during the cold season (October–March) in 2023. Figure 9e shows the evolution of the sensitivity regimes inferred from OMI NO₂ and IASI NH₃ from 2015 to 2023. As NO_x emissions have decreased, we find that NO₃⁻ formation over Korea has transited from an NH₃-sensitive to a NO_x-sensitive regime. NH₃-sensitive conditions are now mostly limited to parts of the SMA, and as NO_x emissions continue to decrease we can expect NO₃⁻ formation to be controlled by NO_x emissions everywhere. Our analysis indicates that Korea will increasingly benefit from controlling NO_x emissions to improve both O₃ and PM_{2.5} air quality in the future.

4. Conclusions

We analyzed the distributions and 2015–2023 trends of major air pollutants in South Korea using the AirKorea surface network, aircraft, and satellite observations. Air quality in Korea has

improved for primary pollutants over the past two decades, but surface O₃ and PM_{2.5} still widely exceed national standards despite emission controls.

Surface CO and SO₂ levels have stayed below air quality standards since the late 1990s, while NO₂ is now below the air quality standard at almost all AirKorea sites. Anthropogenic CO and SO₂ show steady and consistent declines from 2015 to 2023 in both surface concentrations and satellite vertical column densities (VCDs), consistent with the trends from the CAPSS national emissions inventory. NO₂ surface concentrations decreased by 32% from 2015 to 2023 while tropospheric NO₂ VCDs decreased by 36%, consistent with the 23% decrease of NO_x emissions in CAPSS.

Anthropogenic VOC emissions show no significant trend from 2015 to 2023 in the CAPSS inventory. This is consistent with HCHO and CHOCHO VCDs from satellites, although surface observations show a decrease in BTEX aromatic compounds. Satellite HCHO observations show contributions from both anthropogenic and biogenic VOCs, while CHOCHO is more specifically associated with BTEX. Diurnal variations of HCHO and CHOCHO over the Seoul Metropolitan Area (SMA) observed from the GEMS geostationary satellite instrument show a morning increase and a leveling off by midday. Aircraft vertical columns over the SMA during the KORUS-AQ campaign show similar diurnal variations but a late afternoon HCHO increase.

Surface O₃ levels in Korea peak in May–June, and observations at AirKorea sites show an average increase of 0.8 ppbv a⁻¹ in 90th percentile MDA8 O₃ from 2015 to 2023. Such an O₃ increase has been attributed to the effect of NO_x emission reductions under VOC-limited conditions for O₃ production. However, we find from the evolution of the satellite HCHO/NO₂ ratio from 2015 to 2023 that the O₃ formation regime in Korea has been shifting from VOC- to NO_x-limited. GEMS satellite observations for 2023 indicate that most regions in Korea are now NO_x-limited or in a transition regime, and that VOC-limited conditions are confined to the central SMA and Busan. We find that the O₃ increase at AirKorea sites is limited to sites still in the VOC-limited regime, whereas there is no increase for sites in the transition or NO_x-limited regimes. Our results suggest that O₃ across Korea is poised to decrease in response to continued NO_x emission controls.

Annual trends during 2015–2023 in PM₁₀, PM_{2.5}, and AOD show consistent decreases of 4–5% a⁻¹. Diurnal variations in AODs seen from the GOCI satellite instrument show the importance of photochemical production as a source of PM. The only PM_{2.5} component not

found to show a significant decrease over the 2015–2023 period is nitrate (NO_3^-). From the NH_3/NO_2 ratio observed by satellites and its trend over the 2015–2023 period, we find that $\text{PM}_{2.5}$ NO_3^- formation in Korea was mostly NH_3 -sensitive but has become increasingly NO_x -sensitive as NO_x emissions have decreased. As of 2023, NO_3^- formation across Korea is dominantly NO_x -sensitive except in parts of the SMA.

The vigorous NO_x emission controls in Korea have not yet yielded results in terms of decreasing O_3 and $\text{PM}_{2.5}$ NO_3^- . However, our results show that they have effectively shifted O_3 production from a VOC-limited to a NO_x -limited regime and NO_3^- formation from an NH_3 -sensitive to a NO_x -sensitive regime. As NO_x emissions continue to decrease, the benefits for decreasing O_3 and $\text{PM}_{2.5}$ should become apparent.

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Data availability

AirKorea surface network data are available at <https://www.airkorea.or.kr/eng>. CAPSS annual emissions are available at <https://www.air.go.kr/eng/main.do>. MEGAN BVOC emissions are available at https://geos-chem.s3.amazonaws.com/index.html#HEMCO/OFFLINE_BIOVOC/v2021-12/0.25x0.3125/. KORUS-AQ aircraft data are available at <https://www-air.larc.nasa.gov/cgi-bin/ArcView/korusaq>. Satellite products are available at MOPITT CO <https://15ftl01.larc.nasa.gov:22000/misrl2l3/MOPITT/MOP03J.009/>; OMI SO_2 <https://dx.doi.org/10.5067/Aura/OMI/DATA3008>, NO_2 <https://dx.doi.org/10.5067/Aura/OMI/DATA3007>, HCHO <https://dx.doi.org/10.5067/Aura/OMI/DATA3010>, CHOCHO <https://doi.org/10.7910/DVN/Q1O2UE>; TROPOMI CO <https://dx.doi.org/10.5270/S5P-bj3nry0>, NO_2 <https://dx.doi.org/10.5270/S5P-9bnp8q8>, HCHO <https://dx.doi.org/10.5270/S5P-vg1i7t0>; IASI NH_3 <https://iasi.aeris-data.fr/nh3/>; GEMS SO_2 , HCHO, CHOCHO <https://nesc.nier.go.kr/en/html/index.do>, NO_2 <https://doi.org/10.7910/DVN/ZQQJRO>; GOCI AOD available upon request.

Author contributions

Original draft preparation, data processing, analysis, investigation, and visualization were done by YJO. DJJ contributed to project conceptualization. Review and editing were done by DJJ,

DCP, RD, HC, SL, and JK. DCP, NKC, and SK provided additional resources and support in analysis.

Competing interests

The contact author has declared that none of the authors has any competing interests.

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Figures

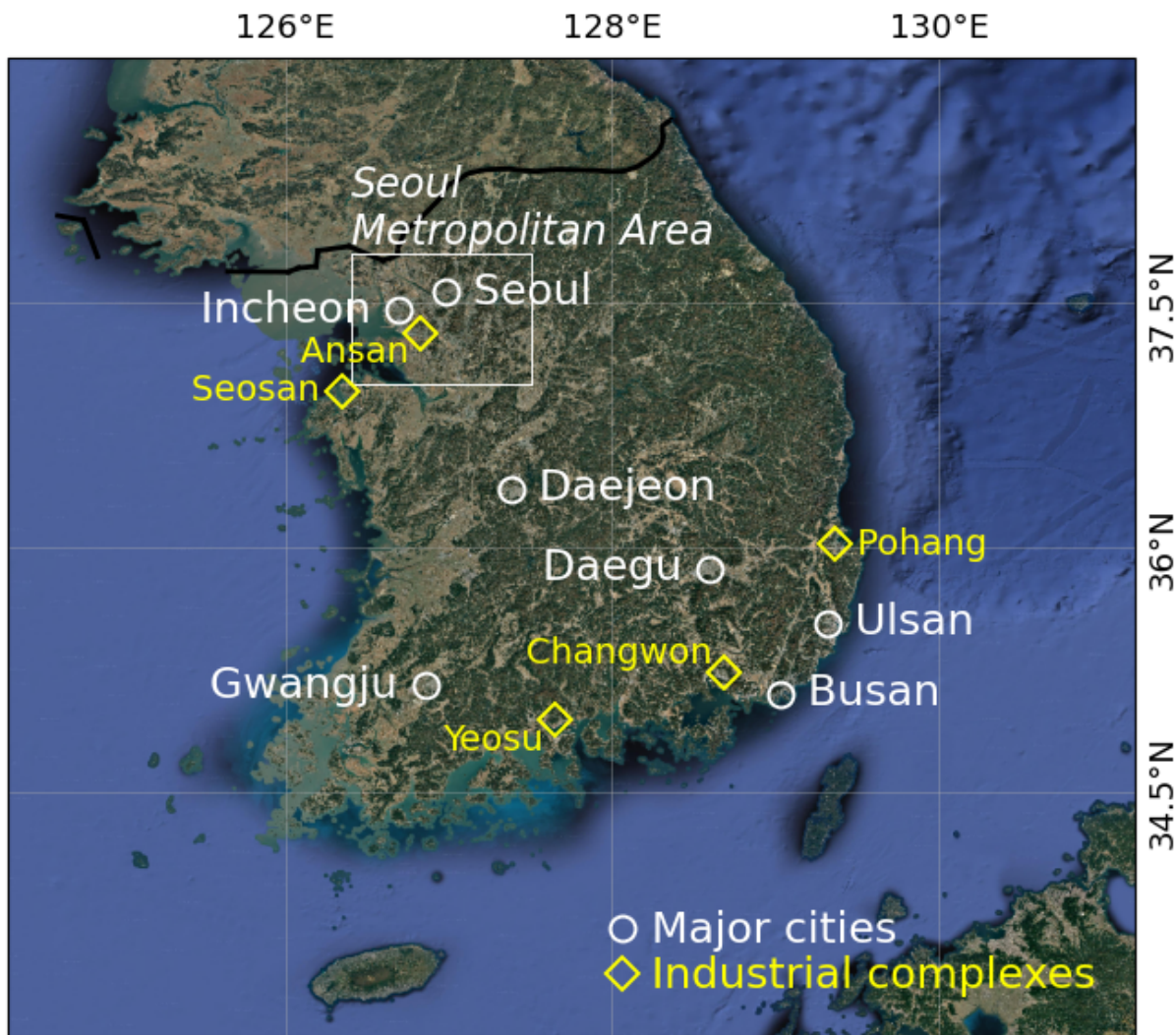


Figure 1. Major source regions in South Korea. Large cities (>1 million) and industrial complexes are indicated in white and yellow colors. The Seoul Metropolitan Area (SMA) is defined as the rectangular domain covering 37–37.8° N and 126.4–127.5° E. Background surface imagery is from © Google Earth.

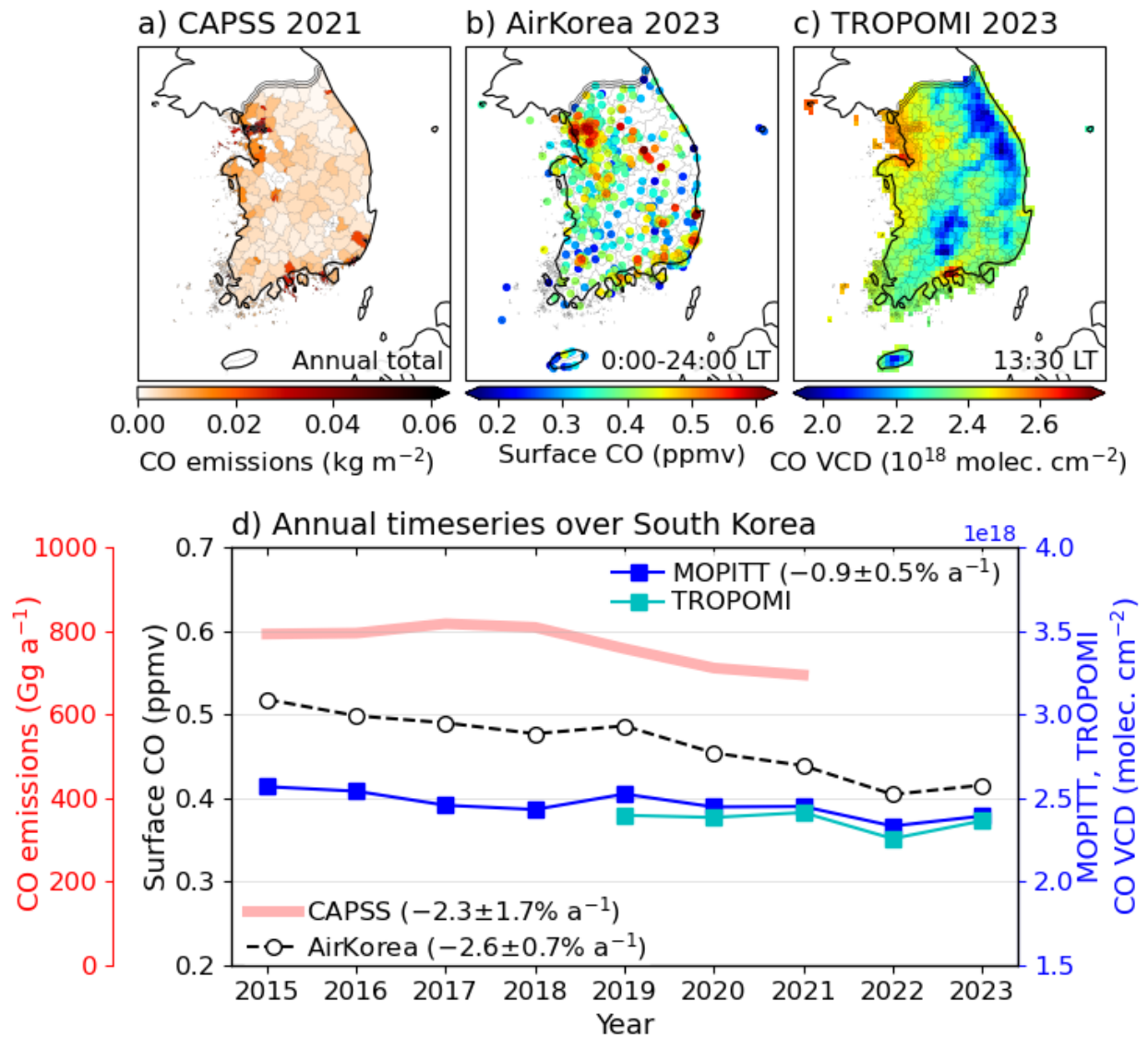


Figure 2. Annual mean CO distributions and trends in South Korea. Top panels show spatial distributions of (a) 2021 anthropogenic CO emissions from CAPSS, (b) 2023 average AirKorea surface CO concentrations for 0–24 local time (LT), and (c) 2023 average TROPOMI CO vertical column densities (VCDs) at 13:30 LT mapped on a $0.1^\circ \times 0.1^\circ$ grid. Lower panel (d) shows 2015–2023 trends in CAPSS CO emissions, surface CO averaged over all AirKorea sites with continuous records (0–24 LT), and CO VCDs from MOPITT (10:30 LT) and TROPOMI (13:30 LT) averaged over South Korea. Statistically significant trends (p -value < 0.05) are given inset with error standard deviations at 95% confidence level.

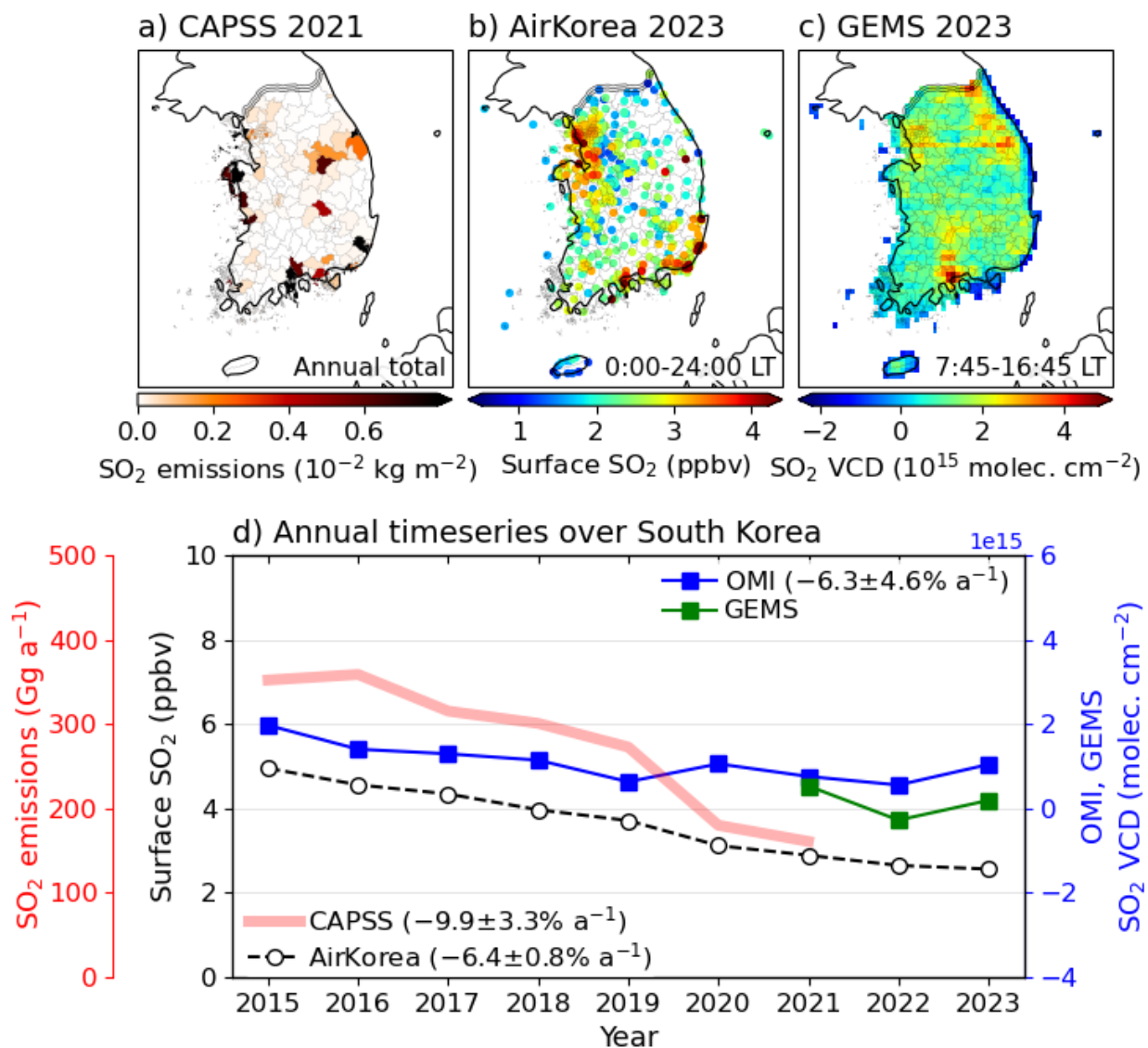


Figure 3. Annual mean SO₂ distributions and trends in South Korea. Top panels show spatial distributions of (a) 2021 anthropogenic SO₂ emissions from CAPSS, (b) 2023 average AirKorea surface SO₂ concentrations for 0–24 LT, and (c) 2023 average GEMS SO₂ VCDs for the 7:45–16:45 LT observation period mapped on a $0.1^\circ \times 0.1^\circ$ grid. Lower panel (d) shows 2015–2023 trends in CAPSS SO₂ emissions, surface SO₂ averaged over all AirKorea sites with continuous records (0–24 LT), and SO₂ VCDs from OMI (13:30 LT) and GEMS (sampled at OMI overpass time) averaged over South Korea. Statistically significant trends (p -value < 0.05) are given inset with error standard deviations at 95% confidence level.

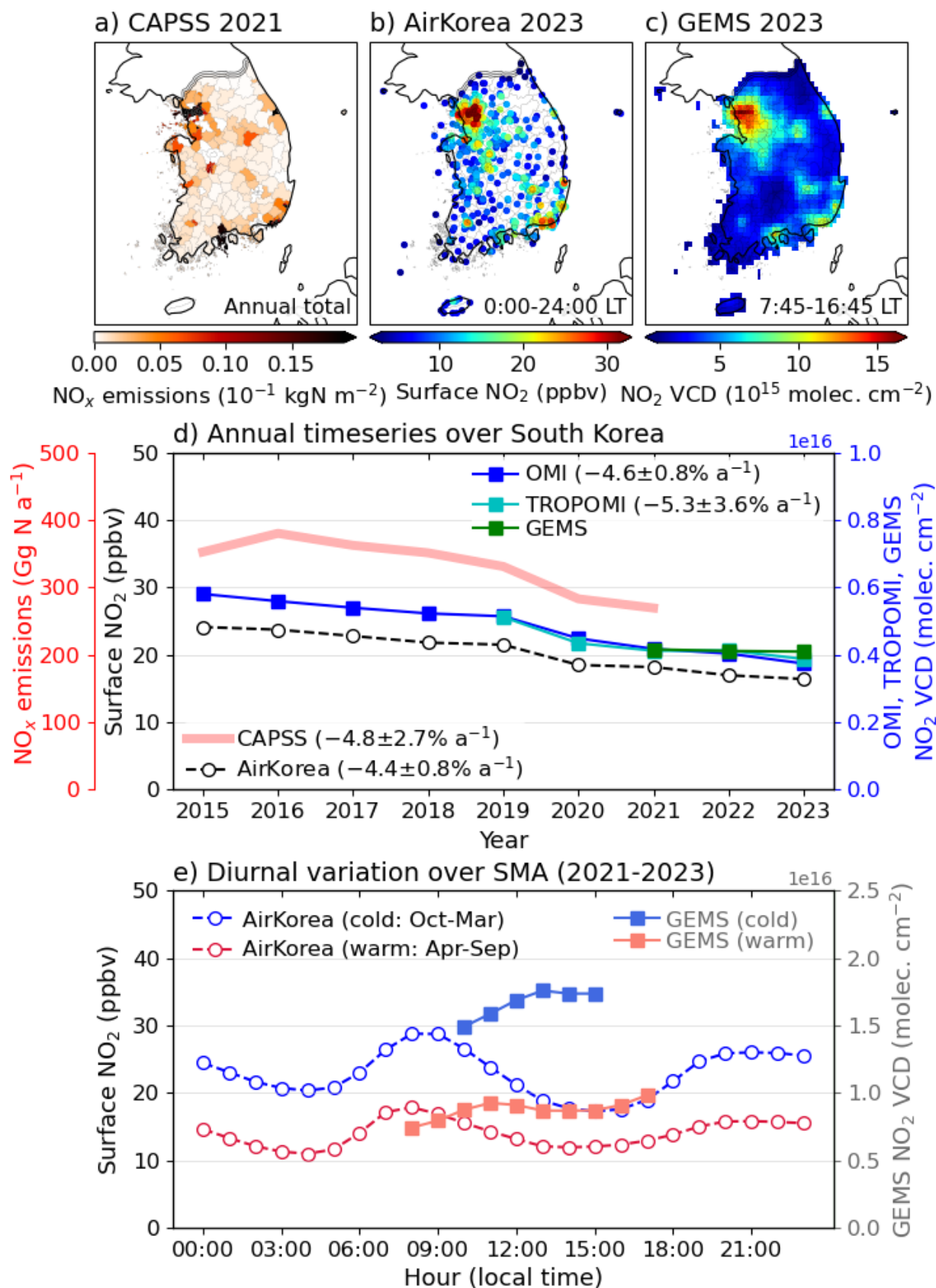


Figure 4. Annual mean NO₂ distributions and trends in South Korea. Top panels show spatial distributions of (a) 2021 anthropogenic NO_x emissions from CAPSS, (b) 2023 average AirKorea surface NO₂ concentrations for 0–24 LT, and (c) 2023 average GEMS tropospheric NO₂ VCDs for the 7:45–16:45 LT observation period mapped on a 0.1° × 0.1° grid. Middle panel (d) shows 2015–2023 trends in CAPSS NO_x emissions, surface NO₂ averaged over all AirKorea sites with continuous records (0–24 LT), and tropospheric NO₂ VCDs from OMI (13:30 LT), TROPOMI (13:30 LT), and GEMS (sampled at OMI overpass time) averaged over South Korea. Statistically significant trends (*p*-value < 0.05) are given inset with error standard deviations at 95% confidence level. Lower panel (e) shows 2021–2023 seasonal mean (cold: October–March, warm: April–September) diurnal variations of AirKorea surface NO₂ concentrations and GEMS VCDs over the SMA.

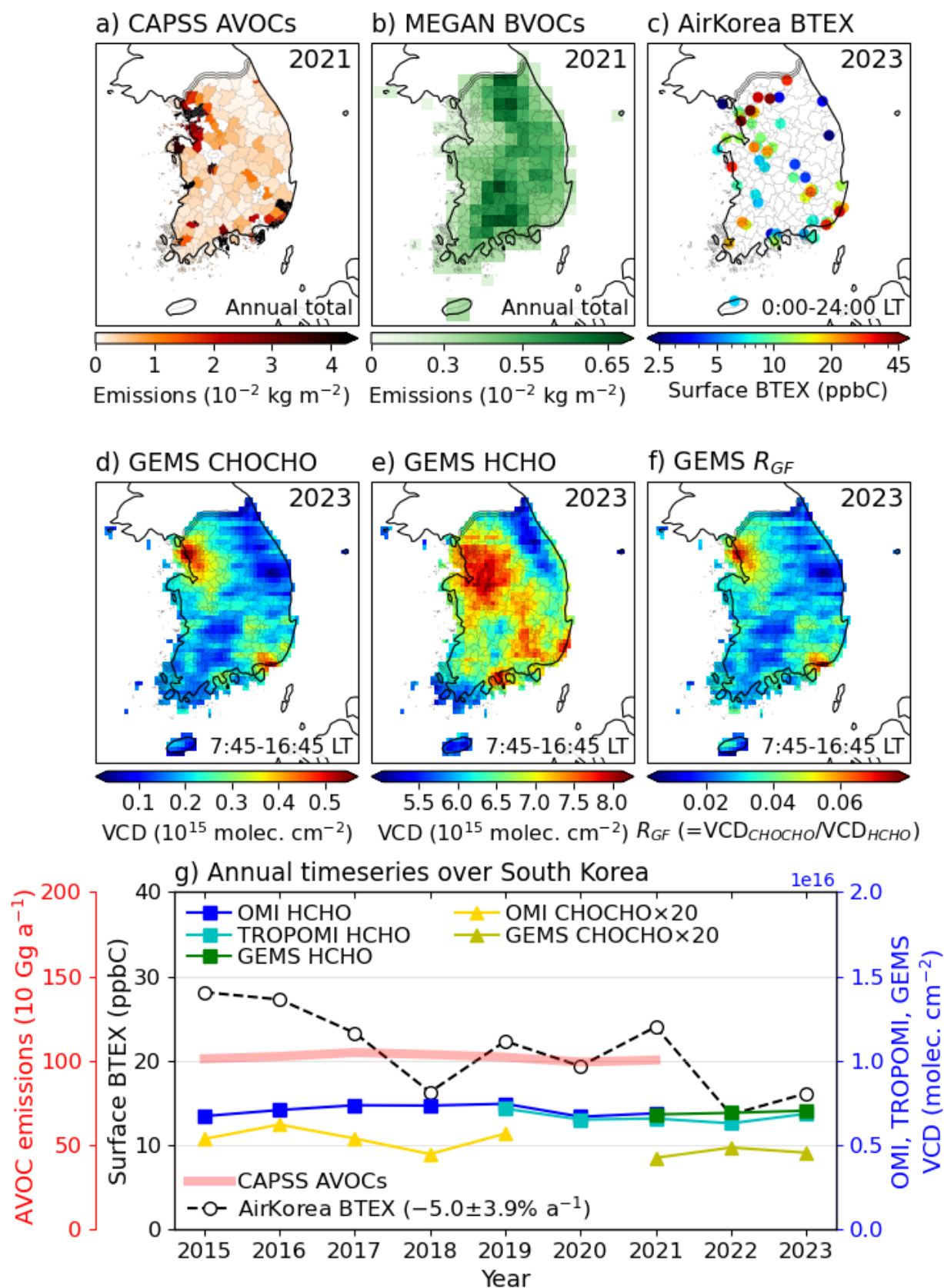


Figure 5. Annual mean NMVOC distributions and trends in South Korea. Top panels (a–b) show 2021 anthropogenic VOC (AVOC) emissions from CAPSS and biogenic VOC (BVOC: sum of isoprene, monoterpenes, sesquiterpenes, acetaldehyde, acetone, methanol, ethanol) emissions from MEGAN, and (c) 2023 average AirKorea surface BTEX ($\equiv$ benzene + toluene + ethylbenzene + xylenes) concentrations for 0–24 LT. Middle panels (d–f) show spatial distributions of 2023 average GEMS glyoxal (CHOCHO) VCDs, formaldehyde (HCHO) VCDs, and glyoxal to formaldehyde ratio R_{GF} ($= \text{VCD}_{\text{CHOCHO}}/\text{VCD}_{\text{HCHO}}$) for the 7:45–16:45 LT observation period mapped on $0.1^\circ \times 0.1^\circ$ grids. Lower panel (g) shows 2015–2023 trends in CAPSS AVOC emissions, surface BTEX averaged over AirKorea sites with continuous records (0–24 LT), and CHOCHO (20 times magnified) and HCHO VCDs averaged over South Korea from OMI (13:30 LT), TROPOMI (13:30 LT), and GEMS (sampled at OMI overpass time). Statistically significant trends (p -value < 0.05) are given inset with error standard deviations at 95% confidence level. None of the satellite data show significant trends over the 2015–2023 period.

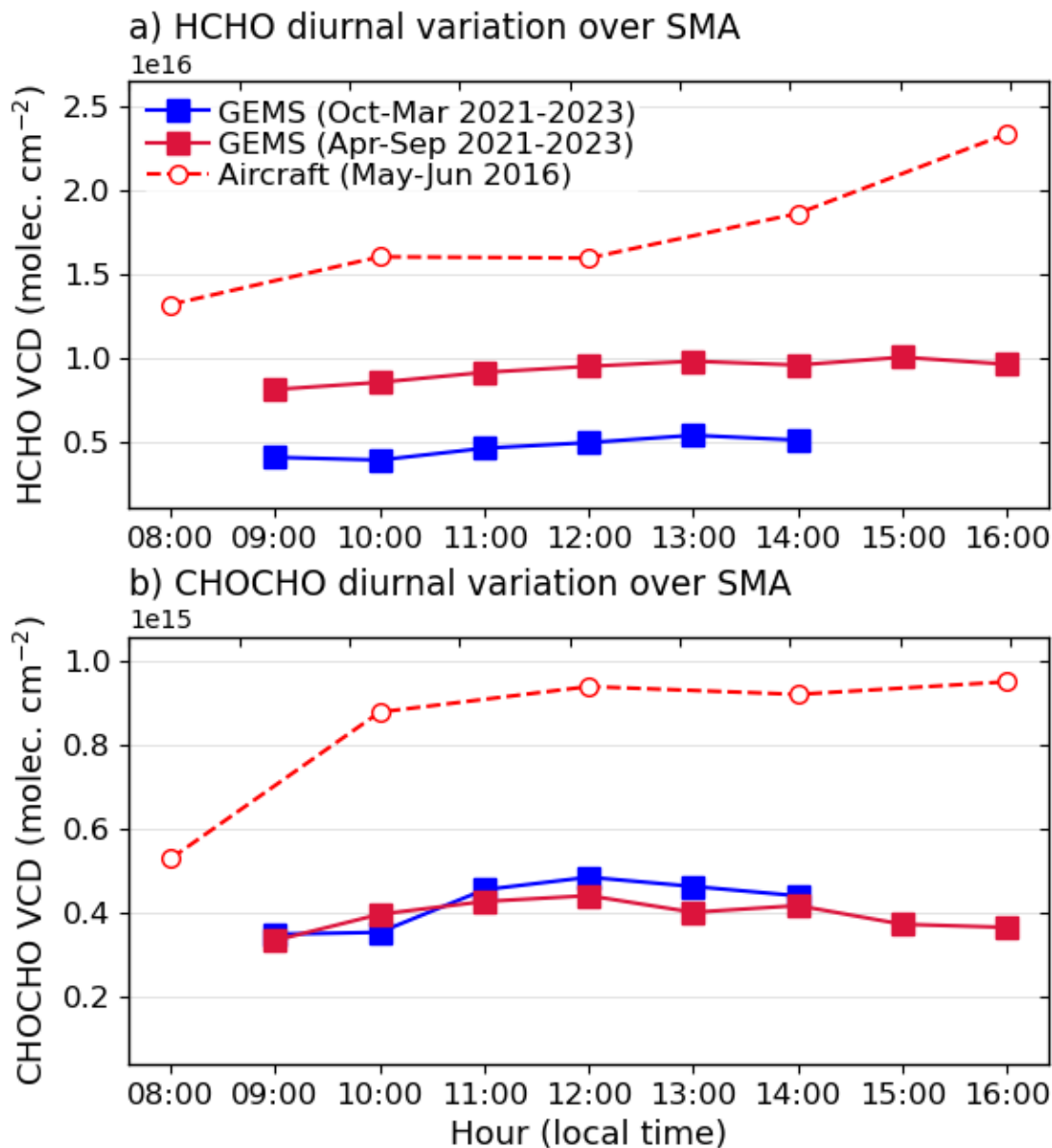


Figure 6. Diurnal variations of HCHO and CHOCHO VCDs in the SMA. Upper panel (a) shows seasonal mean (blue: October–March, red: April–September) diurnal variations of HCHO VCDs from GEMS 2021–2023 observations and from KORUS-AQ (May–June 2016) DC-8 aircraft vertical profiles from near the surface to 8 km altitude over the SMA. Lower panel (b) shows the same for CHOCHO VCDs.

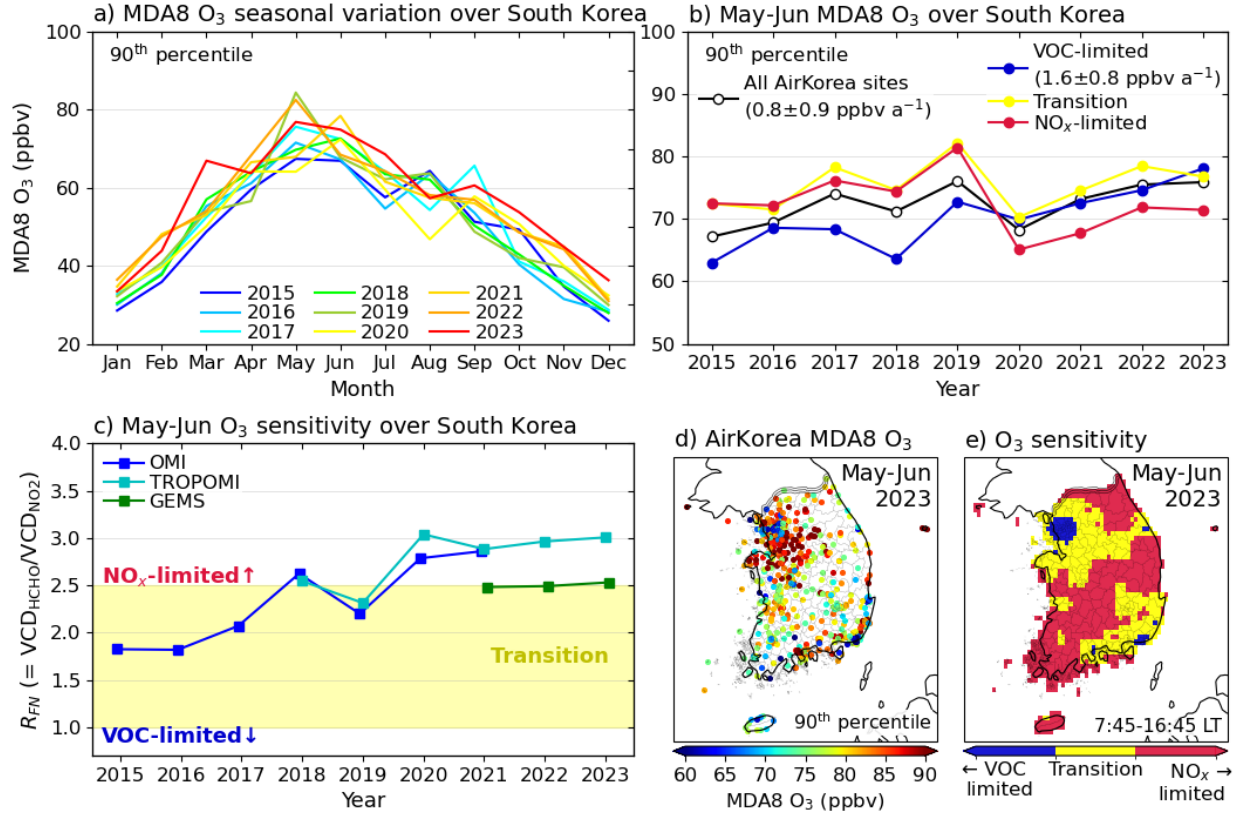


Figure 7. O₃ distribution, trend, and sensitivity to precursors in South Korea. Values are shown for the 90th percentile maximum 8-hour daily average (MDA8) at individual AirKorea sites with continuous records. Top panels show averages of 90th percentile MDA8 O₃ for 2015–2023 as (a) monthly variations in individual years and (b) long-term trends in May–June (when concentrations are highest) for sites in different sensitivity regimes inferred from 2023 GEMS observations. Statistically significant trends (p -value < 0.05) are given inset with error standard deviations at 95% confidence level. Lower left panel (c) shows May–June average timeseries of formaldehyde to NO₂ ratios R_{FN} ($= \text{VCD}_{\text{HCHO}}/\text{VCD}_{\text{NO}_2}$) averaged over South Korea from OMI (13:30 LT), TROPOMI (13:30 LT), and GEMS (sampled at OMI overpass time). Lower right panels show spatial distributions of May–June 2023 average (d) AirKorea 90th percentile MDA8 O₃ and (e) O₃ sensitivity regimes inferred from GEMS R_{FN} (7:45–16:45 LT) mapped on a $0.1^\circ \times 0.1^\circ$ grid. O₃ sensitivity regimes are based on a R_{FN} threshold of 2.5 from Jin et al. (2024).

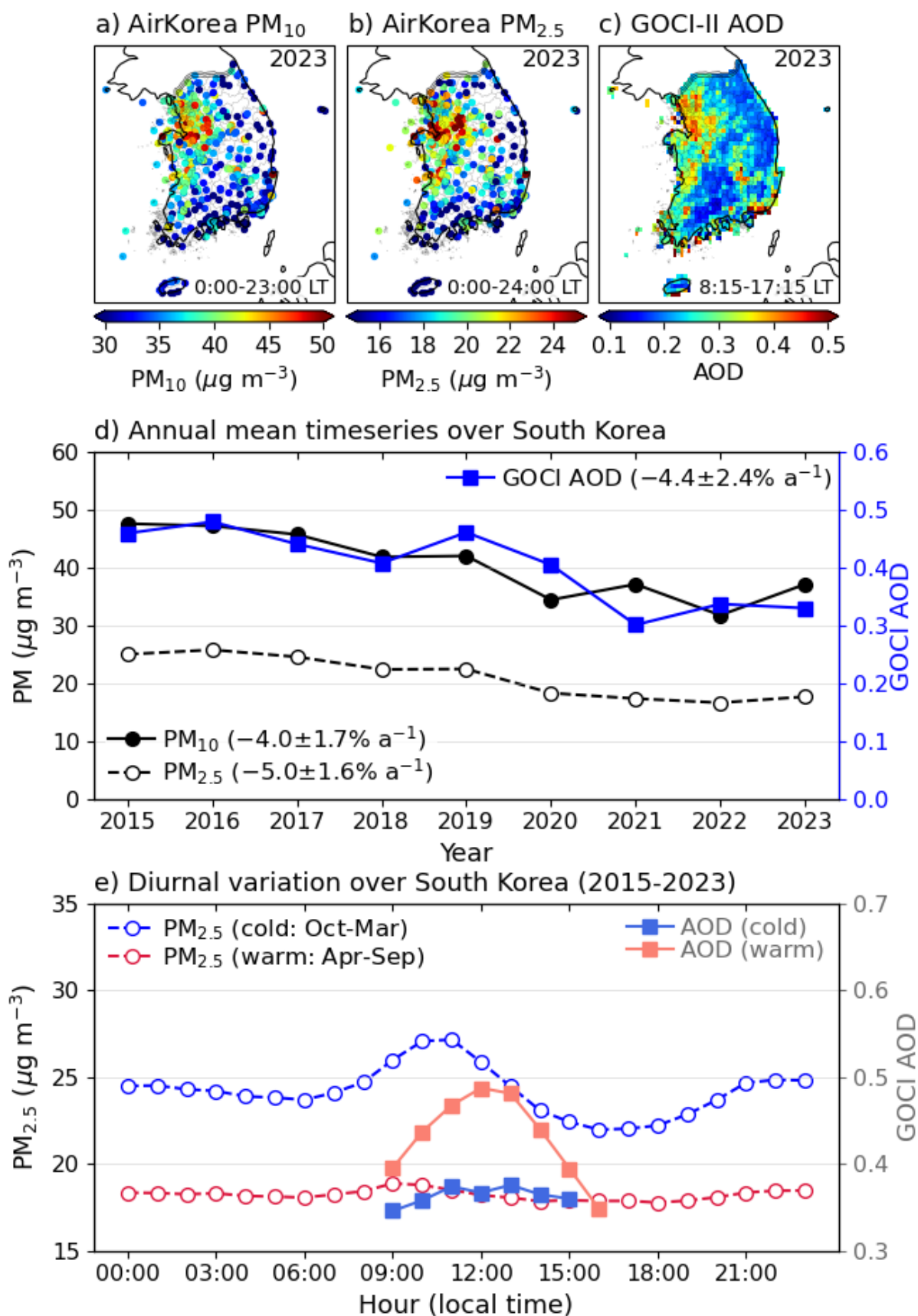


Figure 8. Annual mean PM and aerosol optical depth (AOD) distributions and trends in South Korea. Top panels (a–b) show spatial distributions of 2023 average AirKorea PM₁₀ and PM_{2.5} for 0–24 LT, and (c) 2023 average GOCI-II AOD for the 8:15–17:15 LT observation period mapped on a 0.1° × 0.1° grid. Middle panel (d) shows 2015–2023 trends in PM₁₀ and PM_{2.5} averaged over all AirKorea sites with continuous records (0–24 LT), and GOCI (GOCI; 2015–2020, GOCI-II; 2021–2023) AOD (9:30–16:30 LT) averaged over South Korea. Statistically significant trends (p -value < 0.05) are given inset with error standard deviations at 95% confidence level. Lower panel (e) shows 2015–2023 seasonal mean (cold: October–March, warm: April–September) diurnal variations of AirKorea PM_{2.5} concentrations and GOCI AOD over South Korea. GOCI-II data have been corrected for a low bias relative to GOCI (Table 1).

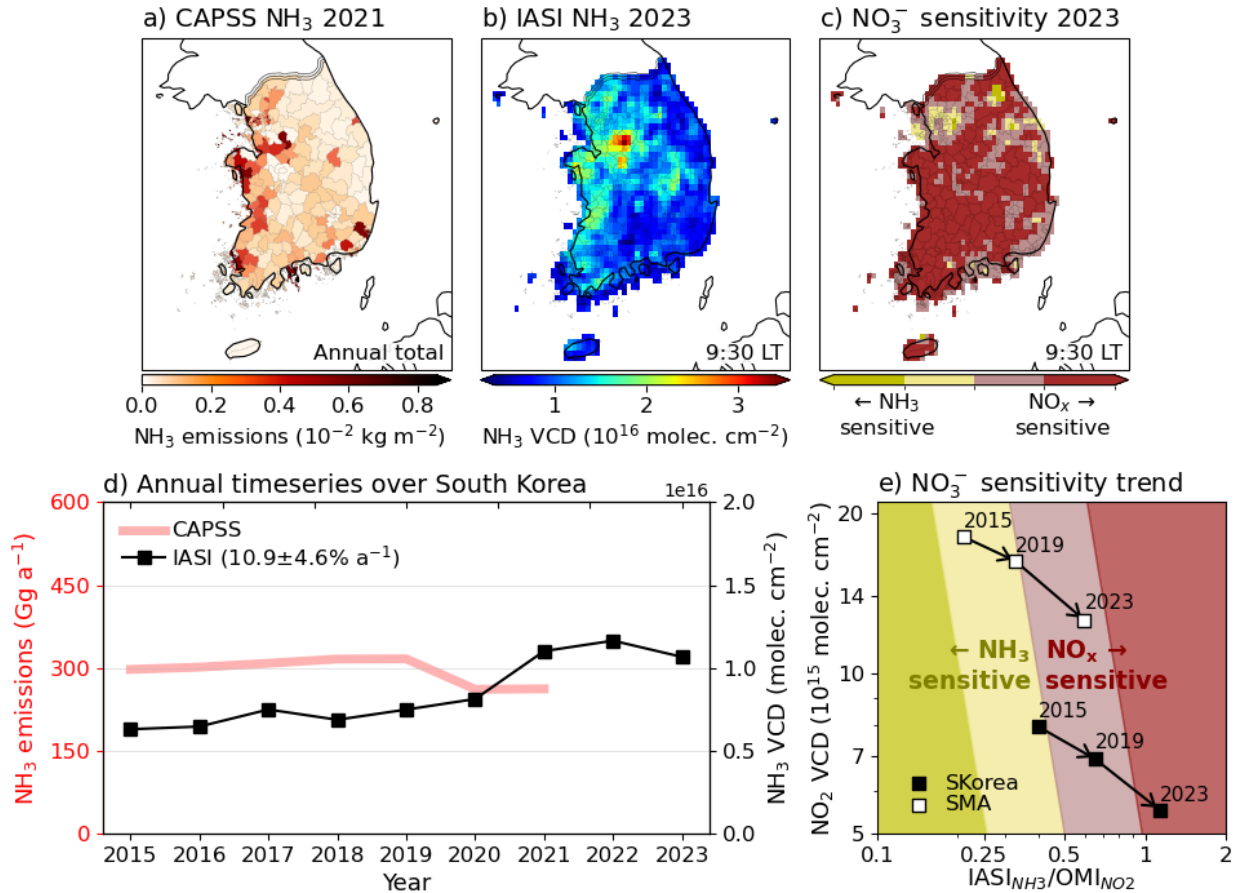


Figure 9. Annual mean NH₃ distributions, trends, and PM_{2.5} nitrate (NO₃⁻) sensitivity in South Korea. Top panels show spatial distributions of (a) 2021 anthropogenic NH₃ emissions from CAPSS, (b) 2023 average IASI NH₃ VCDs at 9:30 LT overpass, and (c) 2023 cold season (October–March) NO₃⁻ sensitivity regimes inferred from IASI NH₃ and GEMS NO₂ (sampled at IASI overpass time). VCDs are mapped on a 0.1° × 0.1° grid. Lower panel (d) shows 2015–2023 trends in CAPSS NH₃ emissions and IASI NH₃ VCDs (9:30 LT) averaged over South Korea. Statistically significant trends (p -value < 0.05) are given inset with error standard deviations at 95% confidence level. Lower right panel (e) shows the cold season NO₃⁻ sensitivity trends averaged over South Korea and over the SMA. NO₃⁻ sensitivity regimes are based on winter thresholds from Dang et al. (2024).