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

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S1. Data processing of surface and satellite observations

Spatial comparisons of surface (AirKorea) observations are all based on 2023 annual mean values using all hours of the day at each monitoring site except for May−June MDA8 O_3. We use level 2 satellite products for TROPOMI and GEMS, which we apply quality filters listed in Table S1 and area-weight averaging to map on a 0.1° × 0.1° grid. We use gridded level 3 products for MOPITT (1° × 1°), OMI (0.25° × 0.25°), and IASI (0.125° × 0.125°) and regrid them to 0.1° × 0.1° for consistency.

For annual trend analyses we only use AirKorea sites that contain continuous records from 2015 to 2023. We construct surface annual mean timeseries by averaging the hourly timeseries across these sites. We construct satellite annual mean timeseries using all pixels over South Korea and sample GEMS observations at the overpass time of OMI and TROPOMI. We use GOCI and GOCI-II AOD observed during the overpass time of GOCI and correct for GOCI-II's low bias by applying a scale factor of 1.047 (= 0.875/0.836). These are regression slopes of GOCI and GOCI-II from the validation against AERONET by Lee et al. (2023).

Table S1. Quality flags used for trace gas satellite observations.

Species	Instrument	Quality flag
CO	MOPITT	N/A
	TROPOMI	qa_value > 0.5
SO_2	OMI	QualityFlags_SO2 = 0
	GEMS	AlgorithmQualityFlag = 0
NO_2	OMI	N/A
	TROPOMI	qa_value > 0.5
	GEMS	AlgorithmQualityFlags ≤ 112, AMFQualityFlags ≤ 64, FinalAlgorithmFlags ≤ 1
NH_3	IASI	N/A
HCHO	OMI	data_quality_flag = 0, -0.5×10^{16} ≤ column_amount ≤ 10×10^{16}
	TROPOMI	qa_value > 0.5
	GEMS	FinalAlgorithmFlags = 0, 1, 3
CHOCHO	OMI	N/A

	GEMS	FinalAlgorithmFlags = 0, 1, 3

S2. HCHO and CHOCHO columns and loss frequencies obtained by aircraft observations

We use the DC-8 aircraft vertical profiles from near the surface to 8 km altitude conducted over the SMA during KORUS-AQ to obtain aircraft-observed column measurements and loss frequencies. HCHO and CHOCHO mixing ratios were measured using the University of Colorado CAMS instrument (Richter et al., 2015) and Gwangju Institute of Science and Technology CAESAR instrument (Min et al., 2016), respectively. OH was measured using the Penn State ATHOS instrument (Faloona et al., 2004) and photolysis frequencies were measured by the NCAR CAFS instrument (Shetter and Müller, 1999).

We first compute mean vertical profiles of HCHO and CHOCHO mixing ratios within 0–8 km at 1 km (Δz) vertical resolution. We convert them into number densities (n_i in molecules cm^{-3}) at each altitude (i) using observed pressure and temperature, then integrate within the column to obtain $\mathrm{VCD} = \sum_i n_i \Delta z_i$ in molecules cm^{-2}. Loss frequencies of HCHO and CHOCHO against OH oxidation and photolysis (two channels for HCHO, three channels for CHOCHO) are defined as follows:

$$L_{HCHO} = k_{HCHO+OH}[OH] + j_{HCHO(1)} + j_{HCHO(2)}\,, \quad \text{(Eq. S1)}$$

$$L_{CHOCHO} = k_{CHOCHO+OH}[OH] + j_{CHOCHO(1)} + j_{CHOCHO(2)} + j_{CHOCHO(3)}\,, \quad \text{(Eq. S2)}$$

where $k_{HCHO+OH} = 5.5 \times 10^{-12} e^{125/T}$, $k_{CHOCHO+OH} = 3.1 \times 10^{-12} e^{340/T}$, T is temperature, and j indicates photolysis frequencies. We compute mean vertical profiles of L_{HCHO} and L_{CHOCHO} within 0–8 km at 1 km vertical resolution and integrate within the column to obtain $L = \frac{\sum_i L_i n_i}{\sum_i n_i}$, where L_i is the mean loss frequency at altitude i. Values at each time of day and the number of aircraft observations used are presented in Table S2.

Table S2. HCHO and CHOCHO columns and loss frequencies from aircraft observations.

Local time (LT)	7–9	9–11	11–13	13–15	15–17
VCD_{HCHO}[a]	1.32 (695)[c]	1.60 (89)	1.59 (472)	1.86 (366)	2.33 (223)
VCD_{CHOCHO}[b]	0.53 (469)	0.88 (75)	0.94 (344)	0.92 (272)	0.95 (346)
L_{HCHO} (hr^{-1})	0.29 (755)	0.41 (92)	0.44 (519)	0.40 (383)	0.35 (485)
L_{CHOCHO} (hr^{-1})	0.48 (755)	0.65 (92)	0.65 (519)	0.57 (383)	0.52 (485)

[a] Units are in 10^{16} molecules cm^{-2}.

[b] Units are in 10^{15} molecules cm^{-2}.
[c] Number of aircraft observations used are indicated in parentheses.

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