## **1** Supplementary Tables

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Table S1. Column integrated AOD and SSA values used for the LUT construction for each research flight, observed
 from the nearest AERONET site.

Variables	17 OCT 2020	3 NOV 2020	5 NOV 2020	24 NOV 2022	25 NOV 2022
(Acronym)	(Chungnam)	(Jaechon)	(Pohang)	(Chungnam)	(Chungnam)
AOD (440 nm)	0.18	0.09	0.21	0.35	0.28
SSA (440 nm)	0.925	0.96	0.96	0.937	0.944
AERONET site	Anmyon	Gangneung_WNU	Gangneung_WNU	Anmyon	Anmyon
(Lat, Lon)	(36.539N, 126.330E)	(37.771N, 128.867E)	(37.771N, 128.867E)	(36.539N, 126.330E)	(36.539N, 126.330E)

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**Table S2**. Reference conditions for the sensitivity test per research flights

Variables (Acronym)	17 OCT 2020 (Chungnam)	3 NOV 2020 (Jaechon)	5 NOV 2020 (Pohang)	24 NOV 2022 (Chungnam)	25 NOV 2022 (Chungnam)
NO <sub>2</sub> VCD	1.00 DU	1.00 DU	1.00 DU	1.00 DU	1.00 DU
PBLH	1.0 km	1.0 km	1.0 km	1.0 km	1.0 km
ALB	0.15	0.15	0.15	0.15	0.15
SZA	48 °	54 °	54 °	57 °	57 °
ALT	1,600 m	1,600 m	2,900 m	1,500 m	1,700 m
VZA	0 °	0 °	0 °	0 °	0 °
RAA	0 °	0 °	0 °	0 °	0 °

## **Table S3**. Details of research flight for the airborne HIS observation. (See Fig. 4 for the acronyms of industrial NO<sub>2</sub> point sources)

Date	Target domain	Potential emission point sources	Airplane type
17 October 2020	Chungnam	PP, PC, SY	Cessna 208
3 November 2020	Jaechon	СК	Cessna 208
5 November 2020	Pohang	SY	Cessna 208
24 November 2022	Chungnam	PP, PC, SY	Beechcraft 1900D
25 November 2022	Chungnam	PP, PC, SY	Beechcraft 1900D

## 14 Supplementary Figures



Figure S1. Relationship between R values of the wavelength pairs and the NO<sub>2</sub> VCDs based on forward RTM
 simulations.

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Figure S2. CMAQ-driven vertical profiles of NO<sub>2</sub> volume mixing ratio (left) and number densities (right) at (a) Chungnam, (b) Jaechon, and (c) Pohang domain according to the PBLH. The shaded area shows the  $10^{th}$ and  $90^{th}$  percentiles of NO<sub>2</sub> concentrations at each altitude per PBLH conditions, and the numbers in parentheses are the corresponding vertical profiles accounted for the analysis.





**Figure S3**. Random noise level per pre-binned spatial pixels (binning 50 raw spatial CCD columns; shown in

28 different colors) under integrated exposure time of 1 second.



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**Figure S4**. Sensitivity of Q-value calculated from simulated radiances at wavelength-pair 2 (i.e., Type\_A: 435.689, 437.015 nm; Type\_B: 433.037, 434.363 nm) depending on (a) NO<sub>2</sub> VCD, (b) PBLH, (c) albedo (ALB; reflectivity), (d) solar zenith angle (SZA), (e) observation altitude (ALT), (f) viewing zenith angle (VZA), and (g) relative azimuth angle (RAA) considering atmospheric condition at Pohang and its corresponding reference conditions (shown in supplementary Table S2).



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**Figure S5**. Sensitivity of Q-value calculated from simulated radiances at wavelength-pair 3 (i.e., Type\_A: 439.932, 441.258 nm; Type\_B: 442.849, 444.175 nm) depending on (a) NO<sub>2</sub> VCD, (b) PBLH, (c) albedo (ALB; reflectivity), (d) solar zenith angle (SZA), (e) observation altitude (ALT), (f) viewing zenith angle (VZA), and (g) relative azimuth angle (RAA) considering atmospheric condition at Pohang and its corresponding reference conditions (shown in supplementary Table S2).



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Figure S6. (a) The hyperspectral imaging sensor (HIS), manufactured by Headwall Photonics, Inc., used in
this study. (b) Cessna 208 Caravan aircraft (left) and the HIS mounted configure on the camera hole of Cessna
plane (right), and (c) Beechcraft 1900D (right) and the HIS mounted on an external canister on a pylon beneath
an aircraft (left).







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Figure S8. Effective Albedo calculated from VIIRS BRDF kernel and the HIS observation geometry along the research flight track conducted at Chungnam (a-c), Jaechon (d), and Pohang (e) area. Flight dates are

shown at the top of each figure.



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Figure S9. Time series of NO<sub>2</sub> VCD measured from HIS (red line) and TROPOMI (blue line) with TROPOMI
overpass time shown in vertical dashed line (left) and the scatter plot comparing the HIS and TROPOMI NO<sub>2</sub>
VCDs with the corresponding HIS NO<sub>2</sub> VCD quantiles (10, 25, 75, 90<sup>th</sup>) shown over the TROPOMI NO<sub>2</sub>
VCD range (right) for the research flight at (a) 17 October 2020, (b) 24 November 2022, and (c) 25 November
2022 over the Chungnam domain.



Figure S10. Time series of NO<sub>2</sub> VCD measured from HIS (red line) and TROPOMI (blue line) with TROPOMI overpass time denoted (left) and the scatter plot comparing the HIS and TROPOMI NO<sub>2</sub> VCDs with the corresponding HIS NO<sub>2</sub> VCD quantiles (10, 25, 75, 90<sup>th</sup>) shown over the TROPOMI NO<sub>2</sub> VCD range (right) for the research flight on 3 November 2020 over the Jaechon domain.



Figure S11. Time series of NO<sub>2</sub> VCD measured from HIS (red line) and TROPOMI (blue line) with
TROPOMI overpass time denoted (left) and the scatter plot comparing the HIS and TROPOMI NO<sub>2</sub> VCDs
with the corresponding HIS NO<sub>2</sub> VCD quantiles (10, 25, 75, 90<sup>th</sup>) shown over the TROPOMI NO<sub>2</sub> VCD range
(right) for the research flight on 5 November 2020 over the Pohang domain.