Boundary-layer aerosols observed in a polluted megacity (Seoul, Korea) from multiple lidar measurements: implications on particulate matter (PM) simulations

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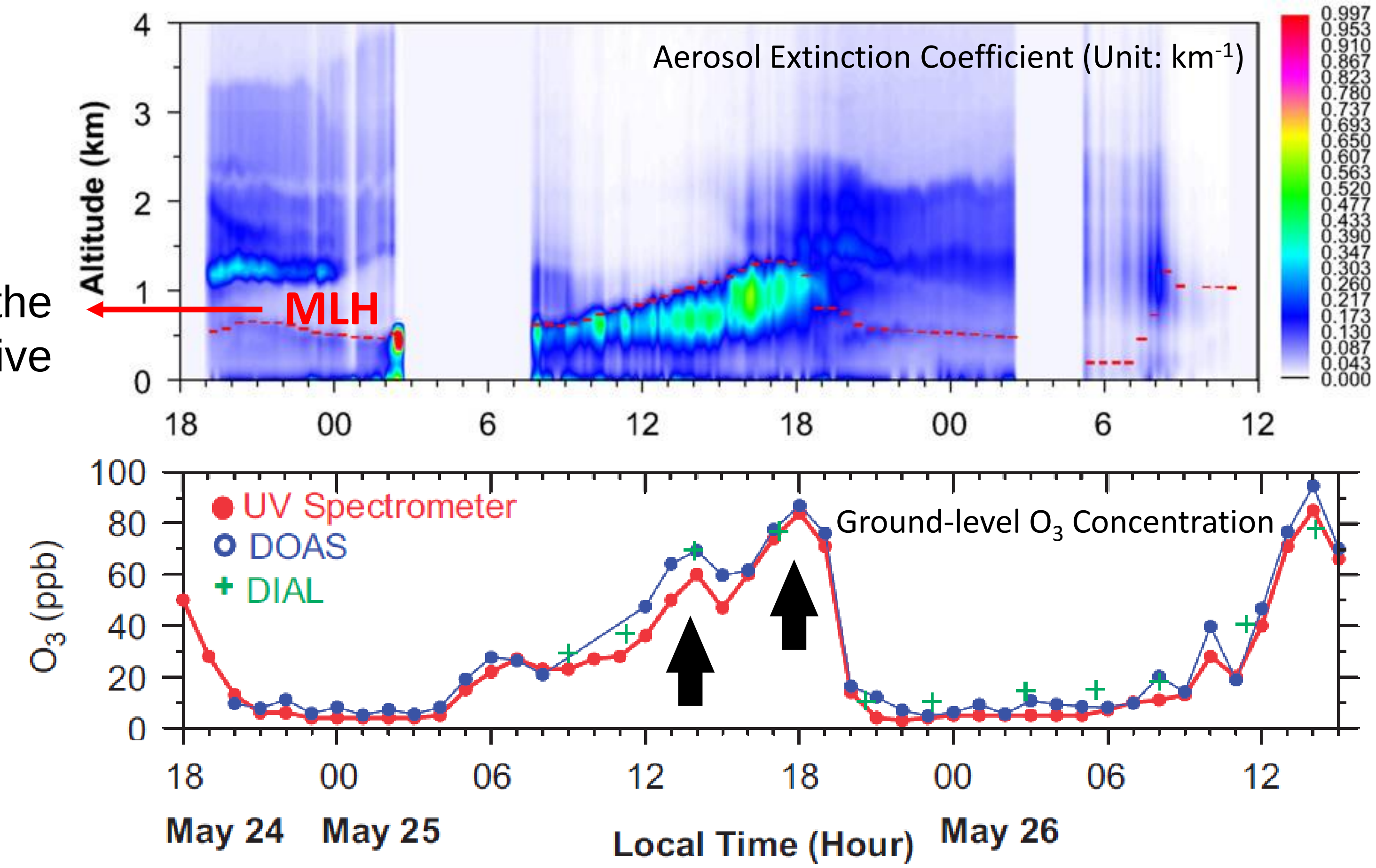
Mixing layer height (MLH)

- The height up to which turbulent mixing creates an environment conducive to the redistribution of temperature, mass, and humidity (Stull 1988; Su et al., 2017).
- Its implications on the vertical distribution of pollutants are widely investigated, especially in regions with high pollution levels.
- Accurate simulation of MLH is especially crucial for models simulating near-surface concentrations of air pollutants (Seo et al., 2015; Compton et al., 2013).

MLH variability and Ground-level Ozone Concentration

(24 -26 May 2000, Olympic Park, Seoul)

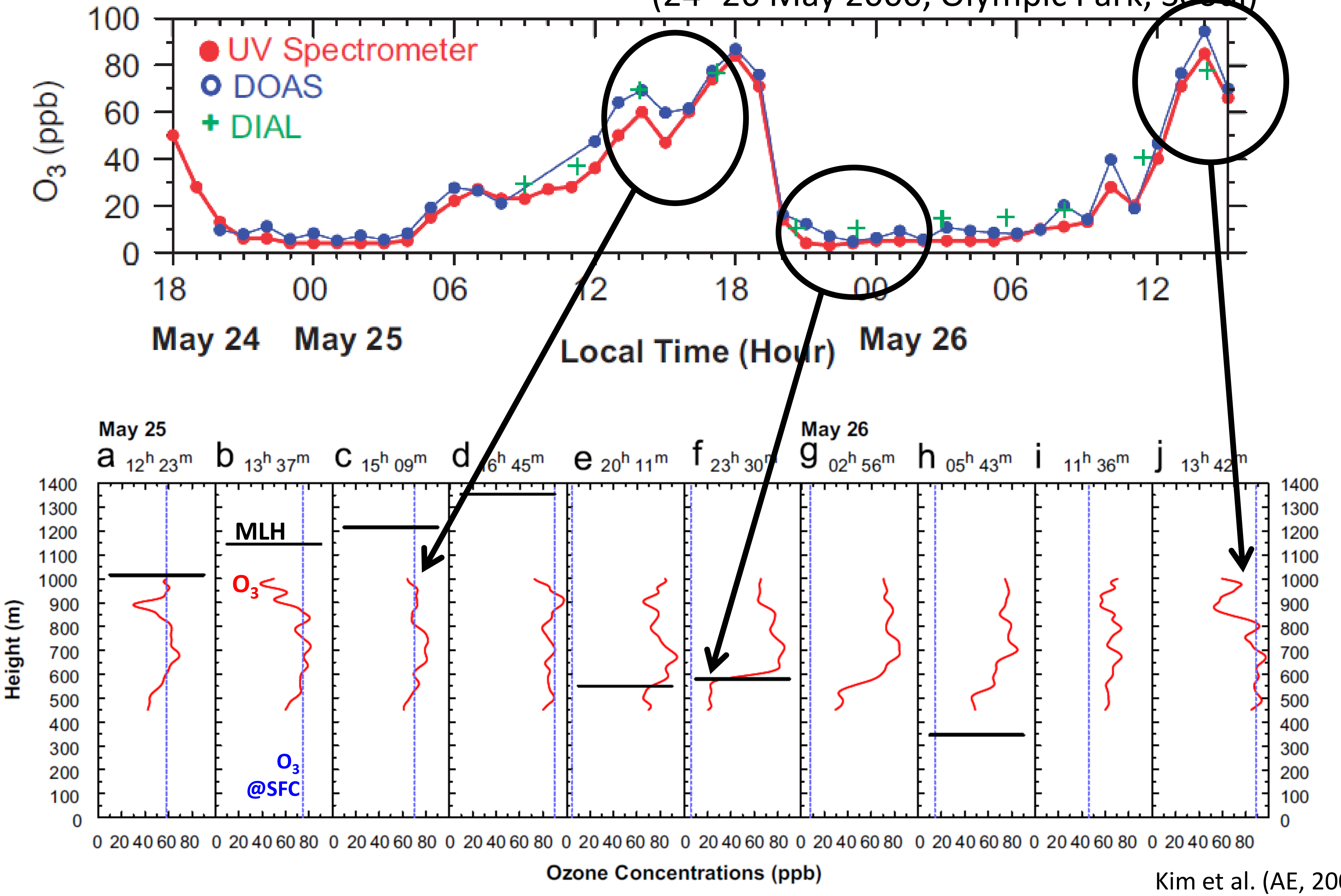
Typical evolution of the MLH under convective situations.



Kim et al. (AE, 2007)

MLH variability and Ground-level Ozone Concentration

(24 -26 May 2000, Olympic Park, Seoul)



Kim et al. (AE, 2007)

Mixing height determination from parameterizations and models

P. Seibert et al. (Atmospheric Environment, 2000)

- Modelling and parameterization of the MH under **stable conditions (SBL)**

e.g.)

$$h = \frac{L_*}{3.8}\left(-1 + \sqrt{1 + 2.28\,\frac{u_*}{fL_*}} \right)$$

Nieuwstadt (1981)

- Modelling and parameterization of the MH under **convective conditions (CBL)**

e.g.)

$$\frac{\mathrm{d}h}{\mathrm{d}t} = A\,\frac{Q_0}{\Delta\Theta} + B\,\frac{u_*^3}{\beta h\,\Delta\Theta} = \frac{Aw_*^3 + Bu_*^3}{\beta h\,\Delta\Theta},$$

Driedonks (1982a)

$$\frac{\mathrm{d}h}{\mathrm{d}t} = (1 + 2A)\,\frac{Q_0}{\gamma_\Theta h} + 2B\,\frac{u_*^3}{\gamma_\Theta \beta h^2} = \frac{(1 + 2A)w_*^3 + 2Bu_*^3}{\gamma_\Theta \beta h^2}.$$

Batchvarova and Gryning (1991)

- Determination of the MH from **NWP model** output

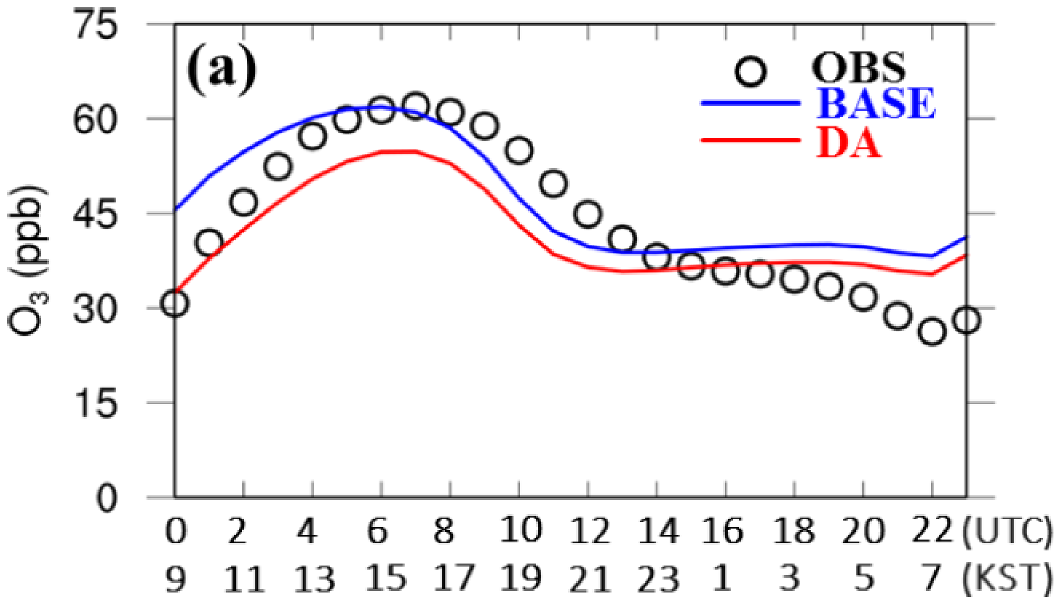
e.g.) The MH is calculated by means of the bulk **Richardson number**:

$$Ri_b = \frac{g}{\theta_0}\,\frac{h(\theta_v(h) - \theta_s)}{(u^2(h) + v^2(h))}$$

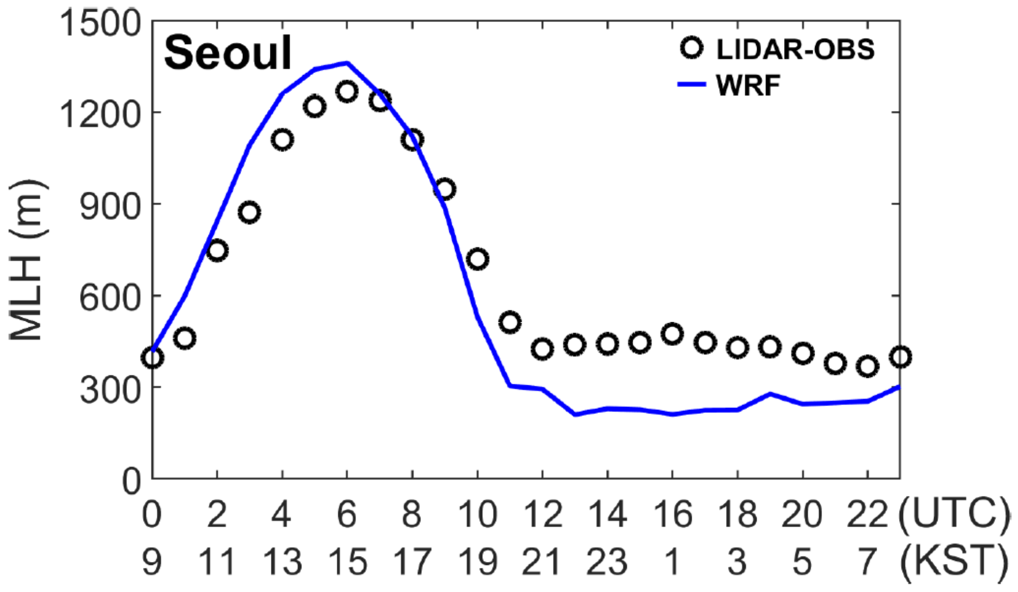
where u(h) and v(h) are the horizontal wind components at height h, $\theta_v(h)$ is the virtual potential temperature at height h, and θs is an appropriate virtual potential temperature near the surface; g/θ0 is the buoyancy parameter, where $\theta_0=0.5(\theta_v(h)+\theta_s)$. The MH is defined as the height at which the bulk Richardson number first equals the critical Richardson number, Ri_c (=0.25)

Hong and Pan (1996)

Model mixing layer height and surface ozone simulations



(KORUS-AQ period, Seoul)



Lee et al., 2020

- Underestimation of the nocturnal mixing layer height (MLH) can be construed as one of the reasons for the overprediction of nighttime ozone mixing ratios.

Mixing layer height in models and vertical profiles of NOx

- Model evaluation showed that aromatic chemistry itself can increase the average net O_3 production in Korea by 37%. The overestimation of the daytime PBL height in the model was found to be responsible for ~10% decrease in both the net O_3 production and NOx loss rates.

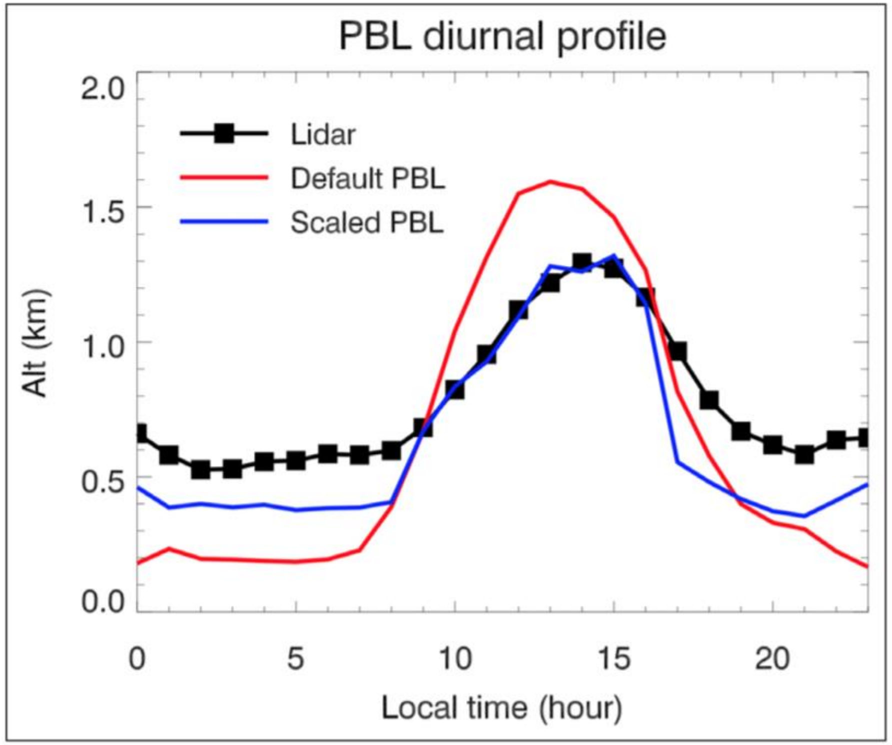


Figure 4: Diurnal profiles of observed (lidar) and simulated PBL heights. Mean diurnal profiles of modeled (colored) and lidar-derived (black) PBL heights at Seoul National University. Red and blue solid lines each indicate the modeled PBL height with no modification and the constrained PBL height using hourly scale factors, respectively. DOI: https://doi.org/10.1525/elementa.394.f4

(KORUS-AQ period, Korea)

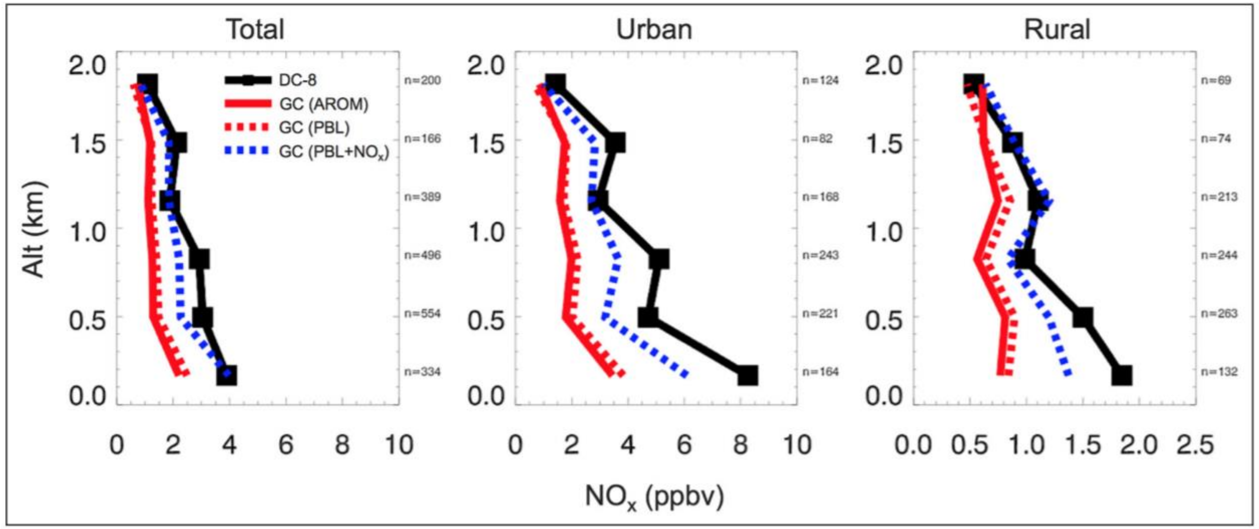


Figure 5: Vertical profiles of observed and simulated NO_x. Mean vertical profiles of observed (black) and simulated NO_x mixings. Colored lines indicate simulated NO_x profiles, and dotted lines indicate results using scaled PBL heights. The number of averaged data is denoted on the left sides of each panel. DOI: https://doi.org/10.1525/elementa.394.f5

Oak et al., 2019

Mixing Height Determination from Profile Measurements

- **Radiosonde**
  - Routine ascents for many years all over the world.
  - Measured data transmitted via international communication networks with very short time delay.
  - Limited height resolution of routine ascents and 2-4 soundings per day.

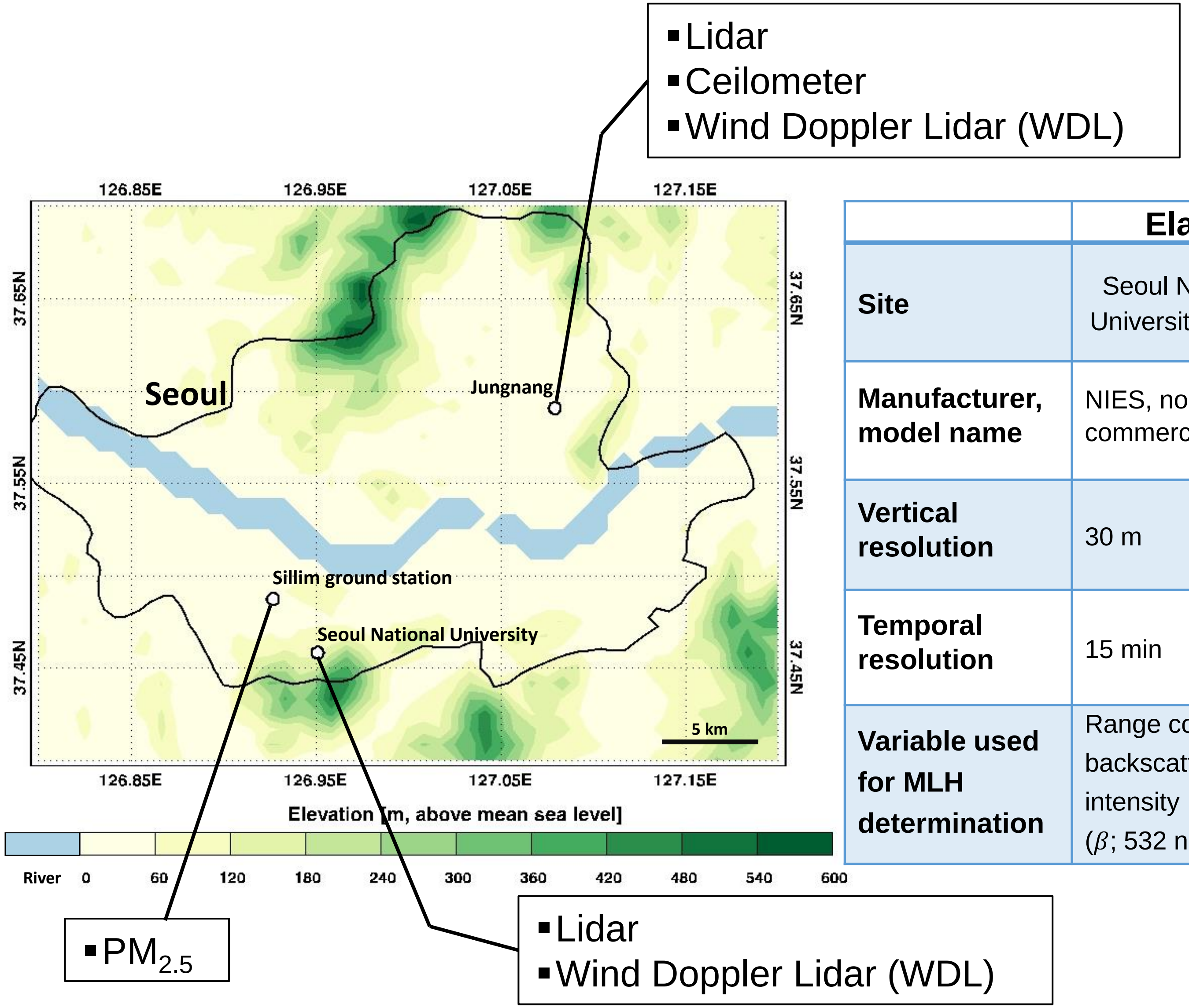
- **Aerosol Lidar** - High sampling rate
  - Return signals originate directly from aerosols ("pollution")
  - Expensive & Tracer necessary & Interpretation sometimes ambiguous

- **Sodar**
  - Relatively simple & not expensive
  - High temporal and vertical resolution
  - Limited sounding range (500 ~ 1000 m) & Sensitive to environmental noise

- **Doppler weather radar/wind profiler**
  - High sampling rate & continuous operation
  - Expensive & Limited vertical resolution

- **Wind Doppler lidar, Ceilometer,** Aircraft, Tethered balloon, Tall tower, etc.

Comparison of MLH determined from profiling instruments



	Elastic aerosol lidar		Ceilometer	WDL	
Site	Seoul National University (SNU)	Jungnang (JNG)	Jungnang (JNG)	Jungnang (JNG)	Seoul National University (SNU)
Manufacturer, model name	NIES, non-commercial	Raymetrics, LB210-D200	Jenoptick, CHM15k	Leosphere, Windcube-200	Mitsubishi, LR-S1D2GA
Vertical resolution	30 m	37.5 m	30 m	50 m	75 m
Temporal resolution	15 min	20 min	20 min	5 min	5 min
Variable used for MLH determination	Range corrected backscattered intensity (β; 532 nm)	Range corrected backscattered intensity (β; 532 nm)	Range corrected backscattered intensity (β; 1064 nm)	Vertical wind speed standard deviation (σ_w)	Vertical wind speed standard deviation (σ_w)

MLH from aerosol lidar, ceilometer, and radiosonde

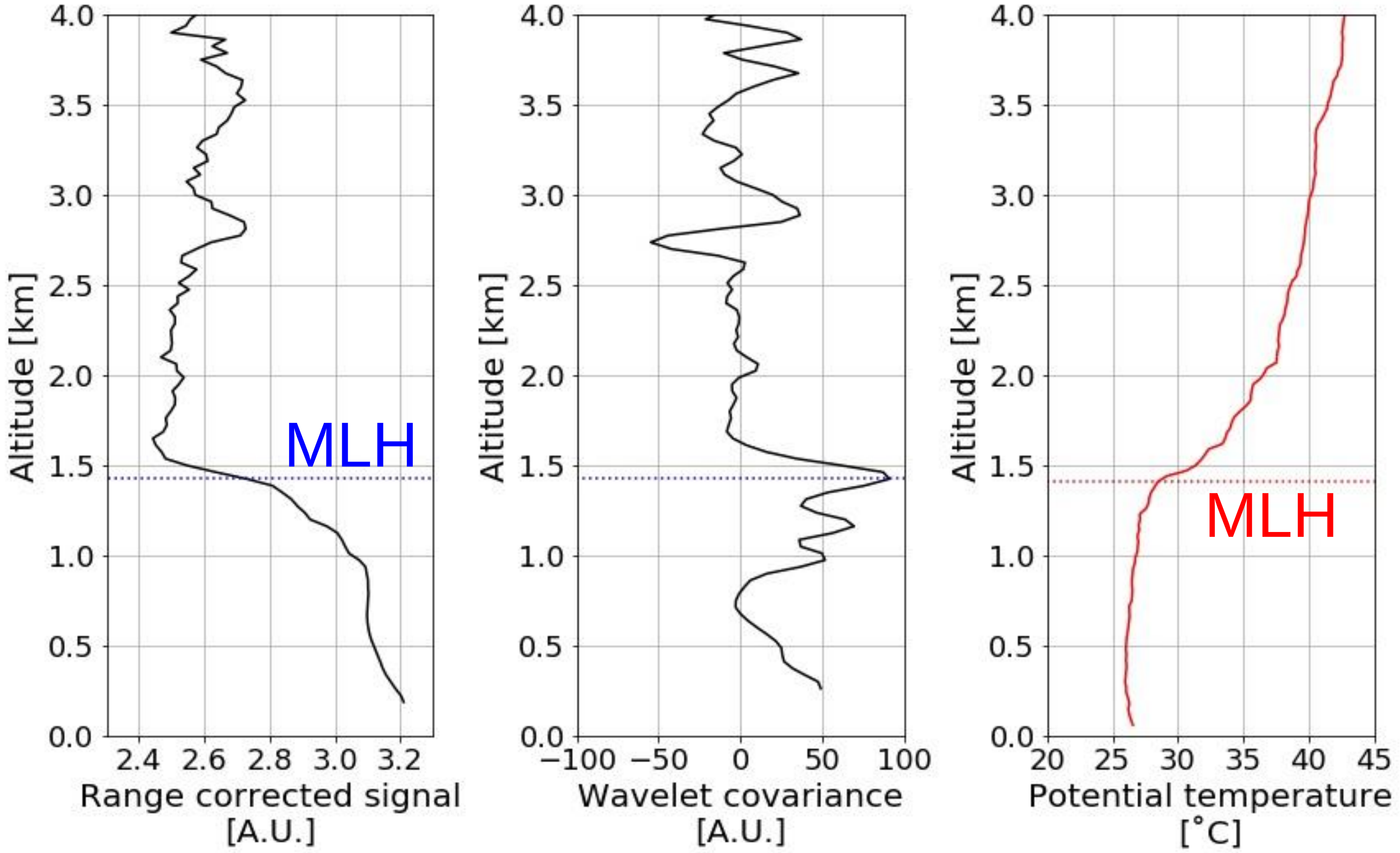
- MLH can be determined as the height of maximum negative gradient of backscattered signal measurements.
- The wavelet covariance transform using the Haar function was used in this study to identify the gradient of profiles.

Haar function

See Figure 1 from Brooks et al (J Atmos OceanicTechnol, 2003)

$$W_f(a,b) = \frac{1}{a}\int_{z_b}^{z_t} f(z)h\left(\frac{z-b}{a}\right)dz$$

1500 LST 4 October 2016, Seoul



MLH from wind doppler lidar

- MLH can be estimated using the **variance of the vertical wind velocity vector (σ_w^2)** measured by WDL (Barlow et al., 2011; Bonin et al., 2018).

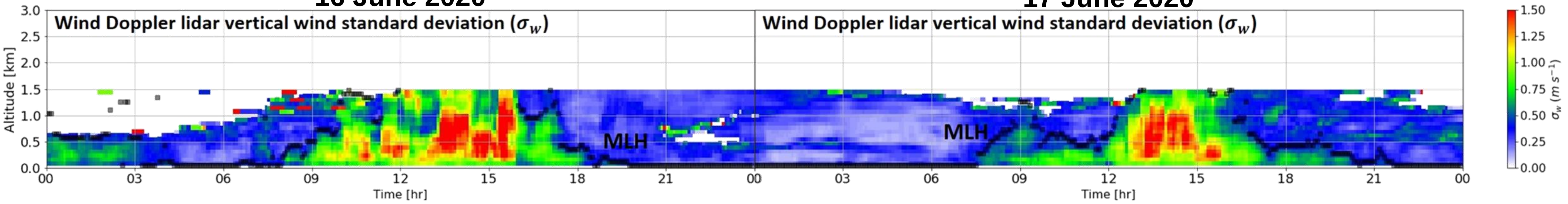
- Threshold values of σ_w^2 are used to determine the height up to where turbulence intensity is sufficient for mixing.

- MLH was defined using a σ_w **threshold value of 0.4 m s^{-1}**.

See vertical velocity variance profile in Figure 2 of Tucker et al (J Atmos OceanicTechnol, 2009)

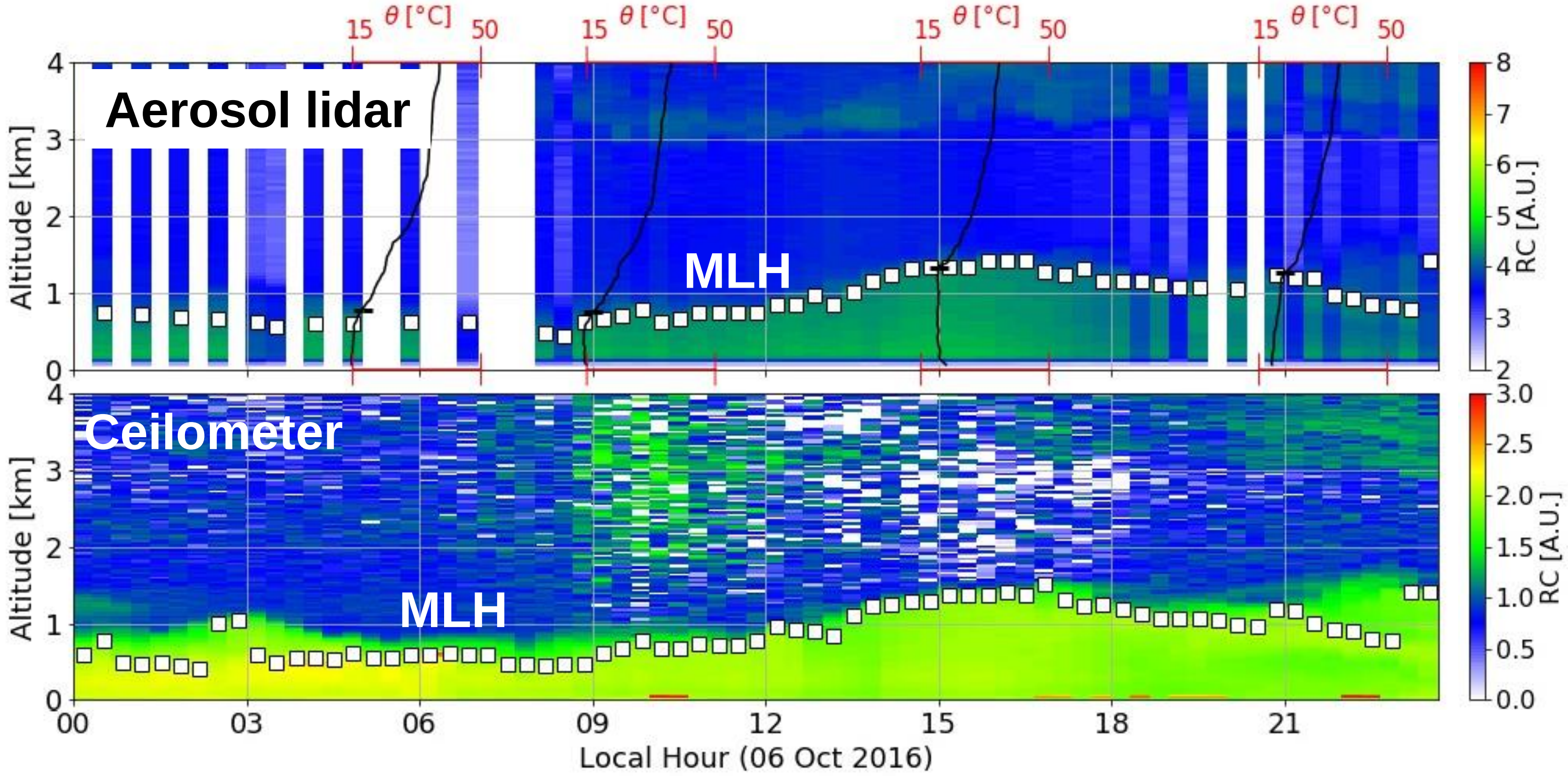
(Tucker et al., 2009)

16 June 2020 **17 June 2020**



Intercomparison of MLH from remote sensing instruments

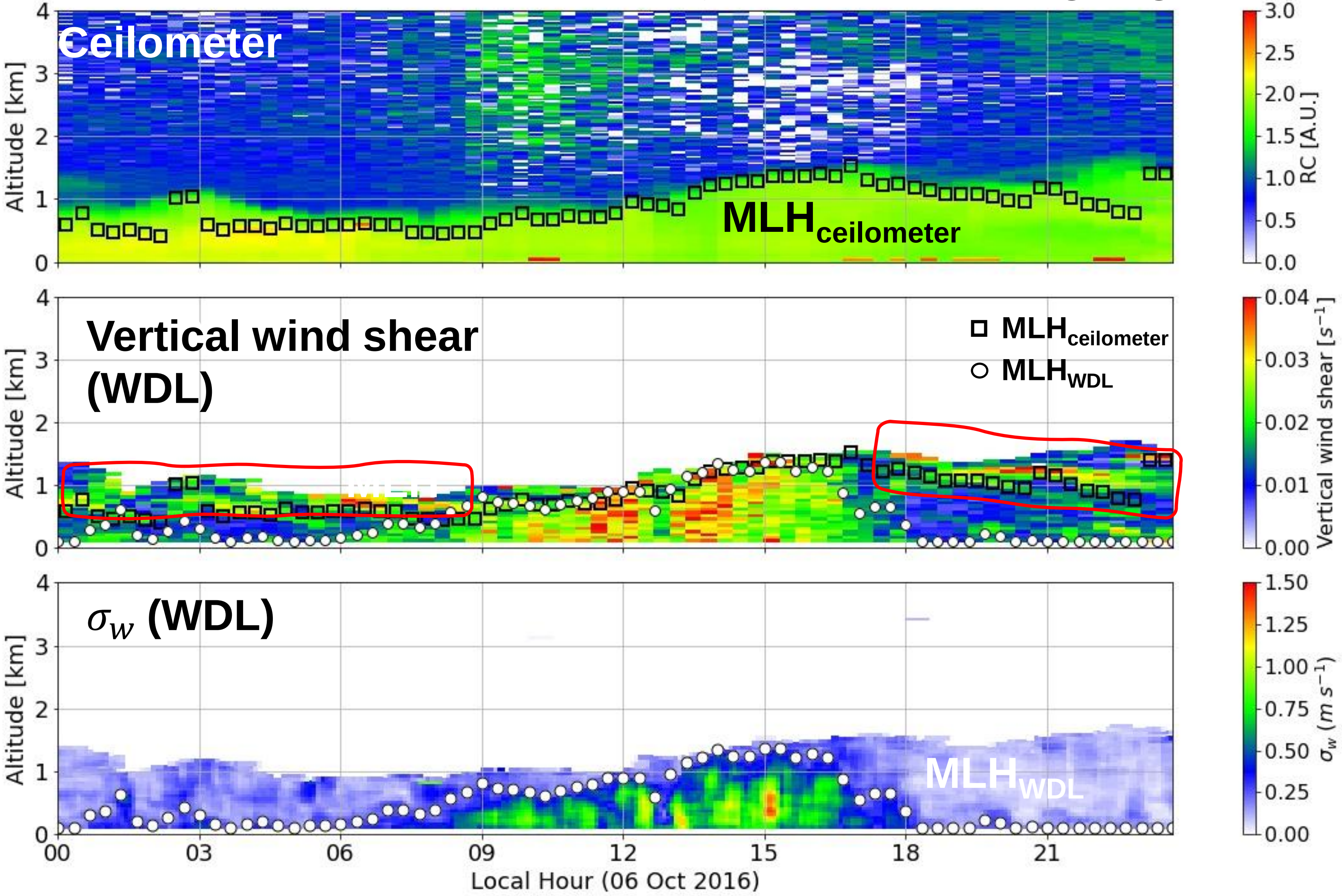
(6 October 2016, Jungnang, Seoul)



- Radiosonde soundings of potential temperature (θ) closely resembled backscattered signal intensity measured by aerosol lidar and ceilometer.

Intercomparison of MLH from remote sensing instruments

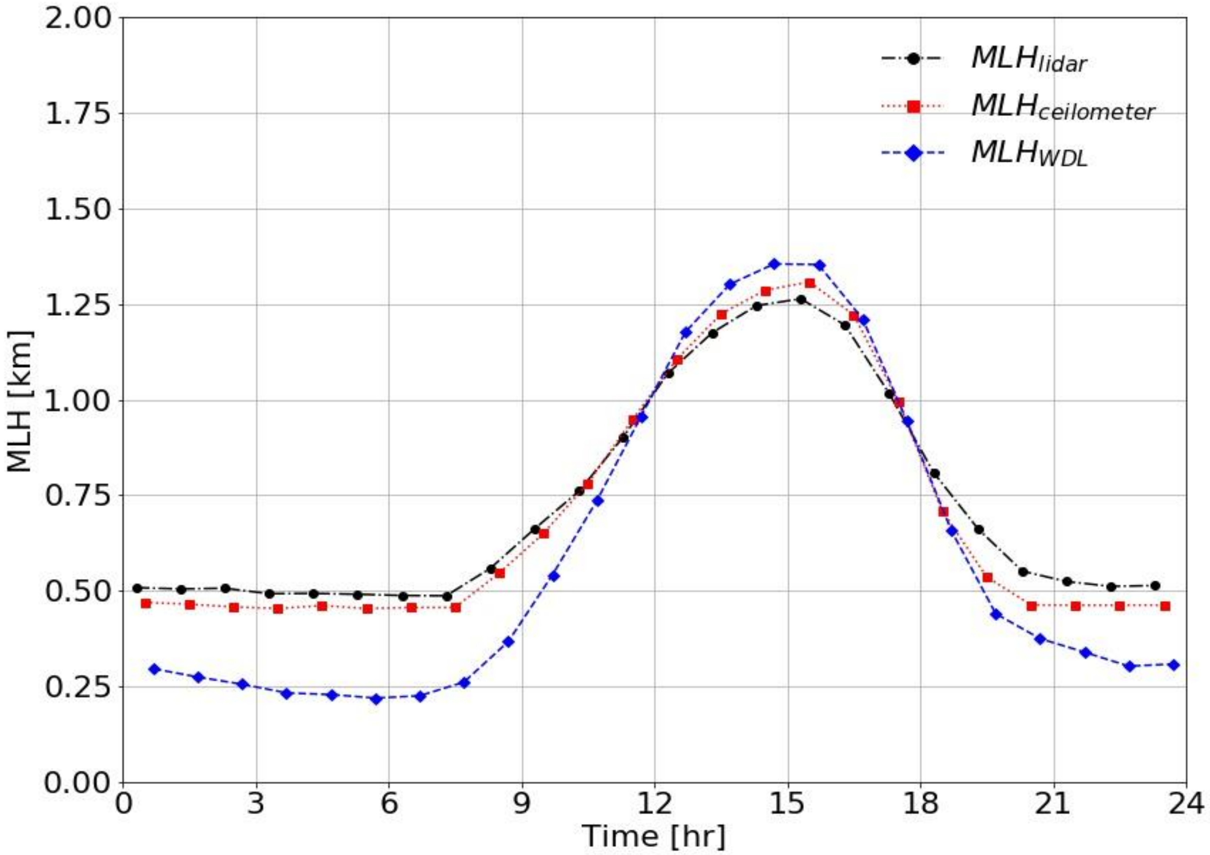
(6 October 2016, Jungnang, Seoul)



- Wind shear may act as a source of mixing during nighttime when other sources of turbulence (e.g., surface heating) are scarce.

Diurnal variation of MLH from lidar, ceilometer, and WDL

MLH diurnal variation (2016 – 2017, Jungnang)

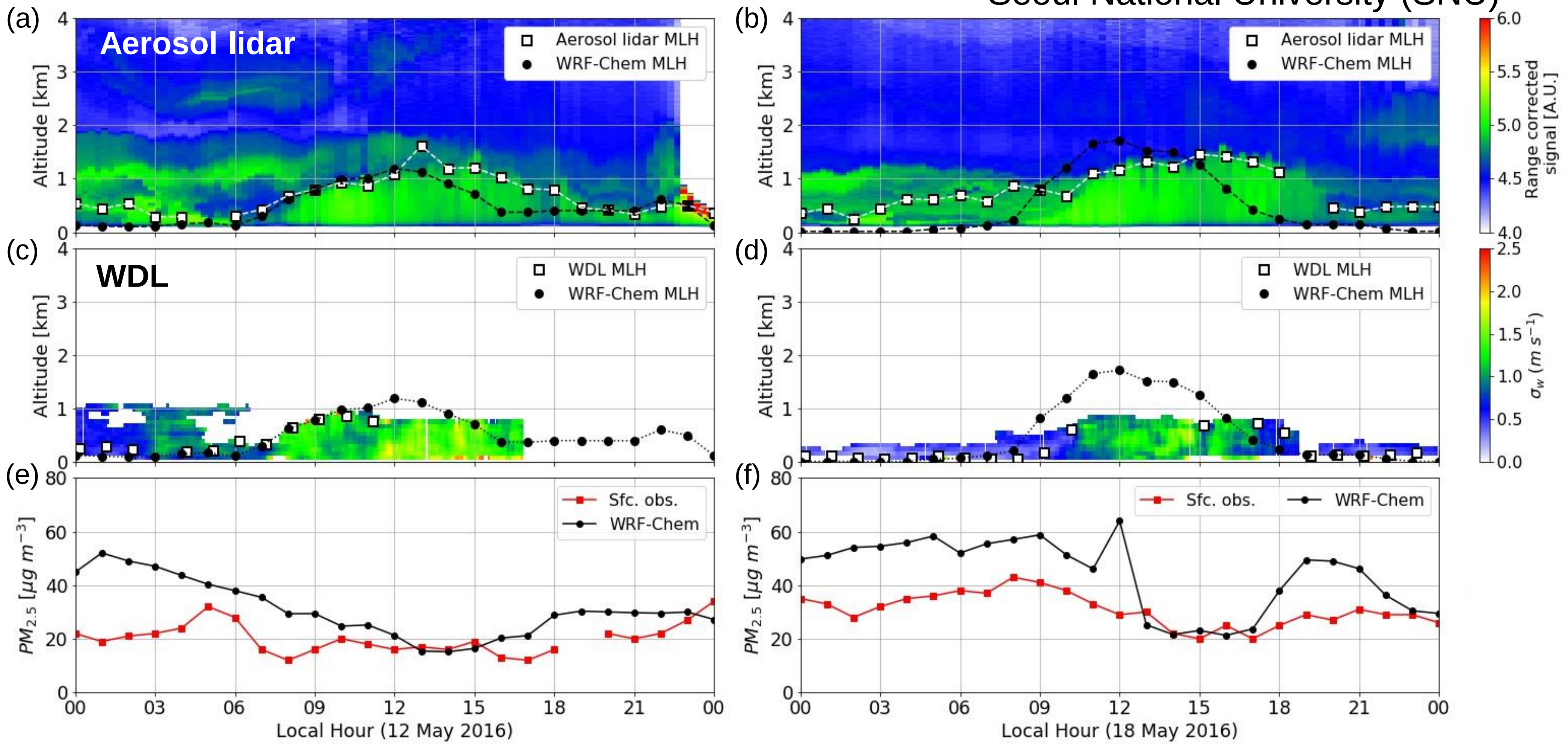


- MLH_{lidar} and $MLH_{ceilometer}$ displayed almost identical diurnal patterns (minimum 0.49 ± 0.13 km and 0.45 ± 0.12 km; maximum 1.26 ± 0.39 km and 1.31 ± 0.43 km, respectively).
- MLH_{WDL} showed the largest diurnal variability (minimum 0.22 ± 0.29 km; maximum 1.35 ± 0.66 km).
- Nocturnal MLH from WDL displayed significantly lower heights than MLH from lidar and ceilometer measurements.

Implications of MLH on surface $PM_{2.5}$ simulations

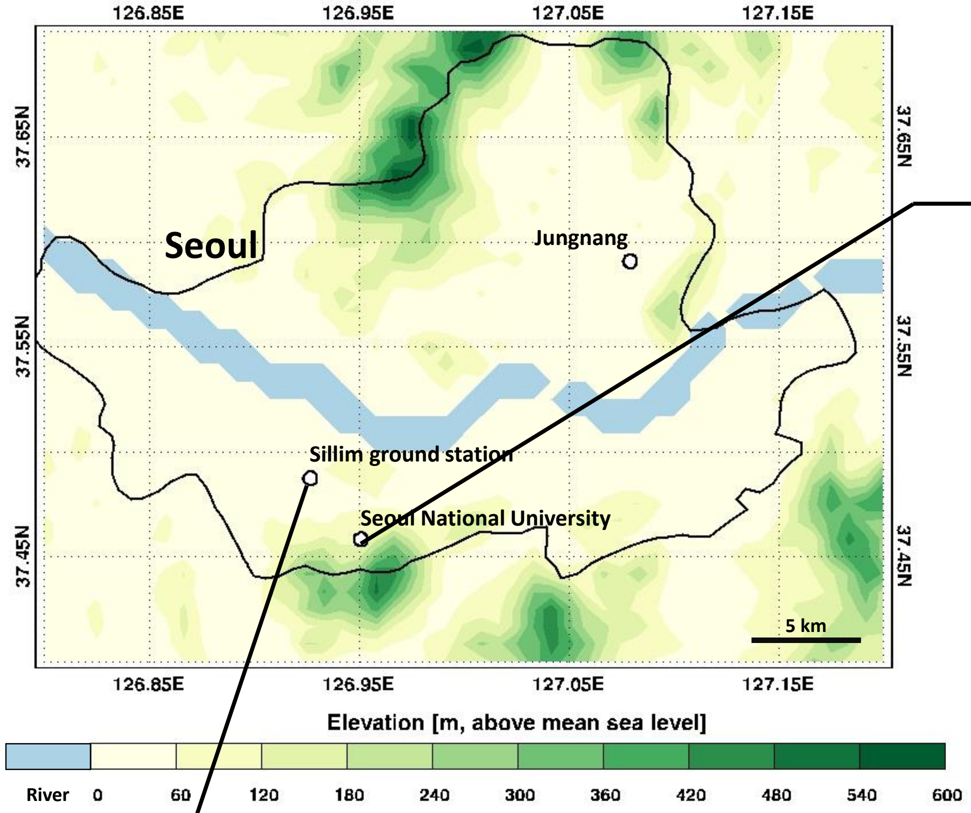
- Comparison of WRF-Chem simulation results of MLH with MLH determined from aerosol lidar and WDL measurements during KORUS-AQ.

Seoul National University (SNU)



(WRF-Chem results courtesy of Hyo-Jung Lee, Pusan National University)

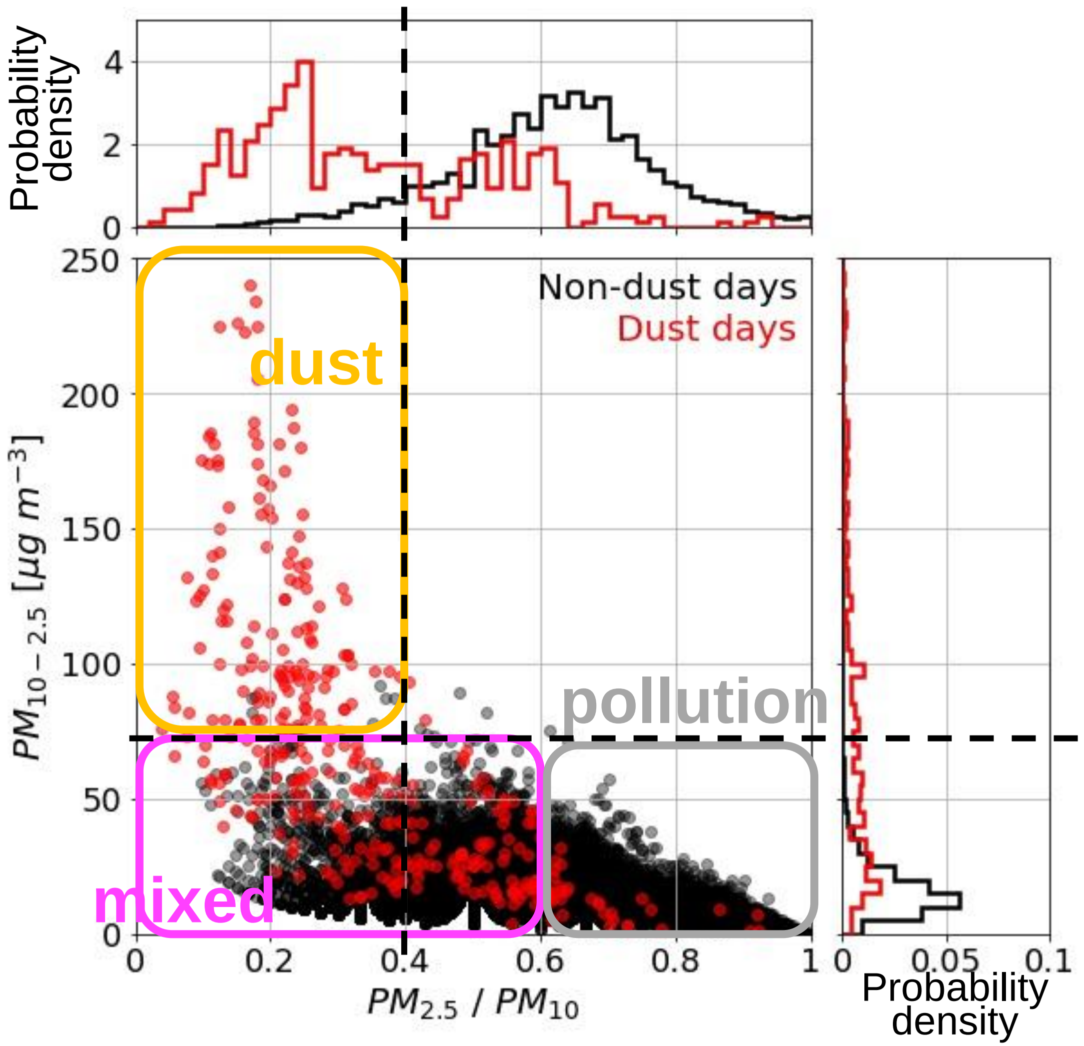
Collocated HSRL and Mie-scattering lidar at SNU



▪ PM_{10} & $PM_{2.5}$

	HSRL	Mie-scattering Lidar
Wavelength	532 nm, 1064 nm	532 nm, 1064 nm
Range resolution	7.5 m	6 m
Temporal resolution	30 s	15 mins
Operation period	**Mar 2016 – Jan 2018**	**June 2006 - present**
	University of Wisconsin-Madison	NIES-Network (AD-NET) & GALION (GAW Aerosol Lidar Observations Network)

Aerosol type classification using surface $PM_{2.5}$ and PM_{10} observations as references

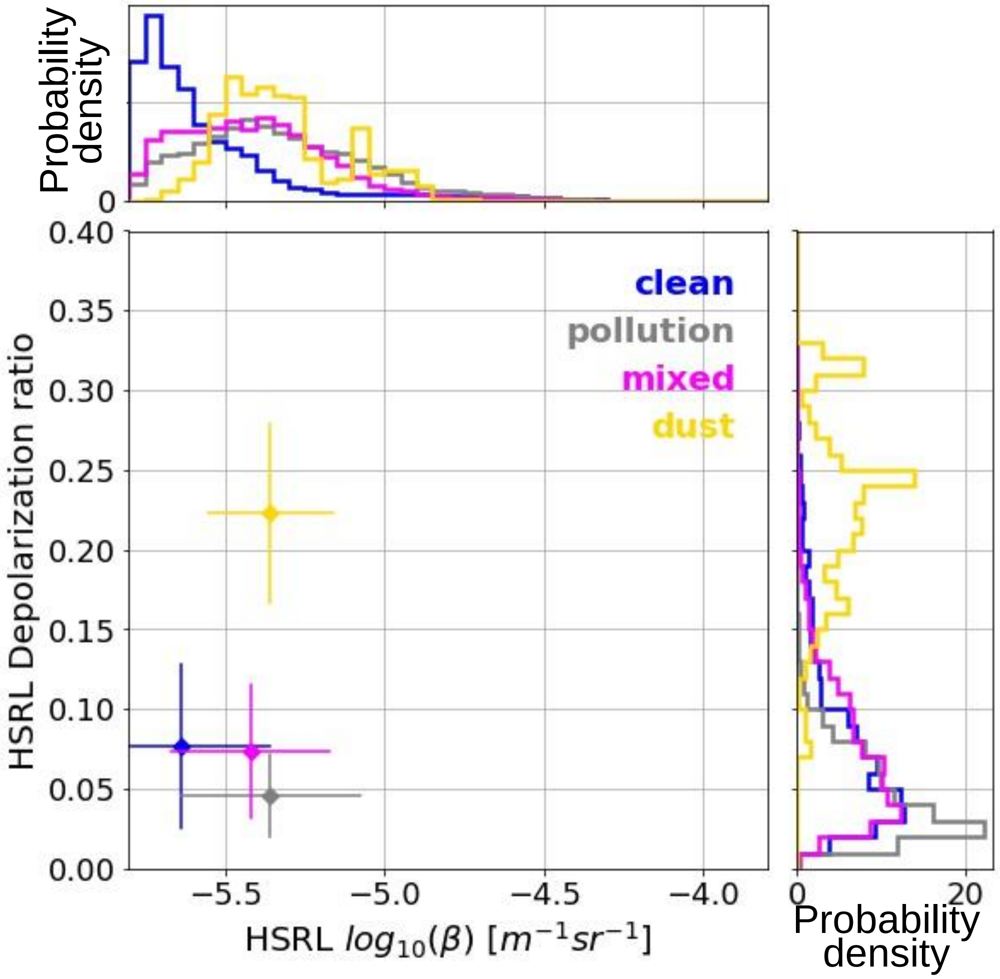


- Surface $PM_{2.5}$ and PM_{10} observations from the Sillim station within the AirKorea network
- Dust days reported by the Korea Meteorological Administration (KMA)

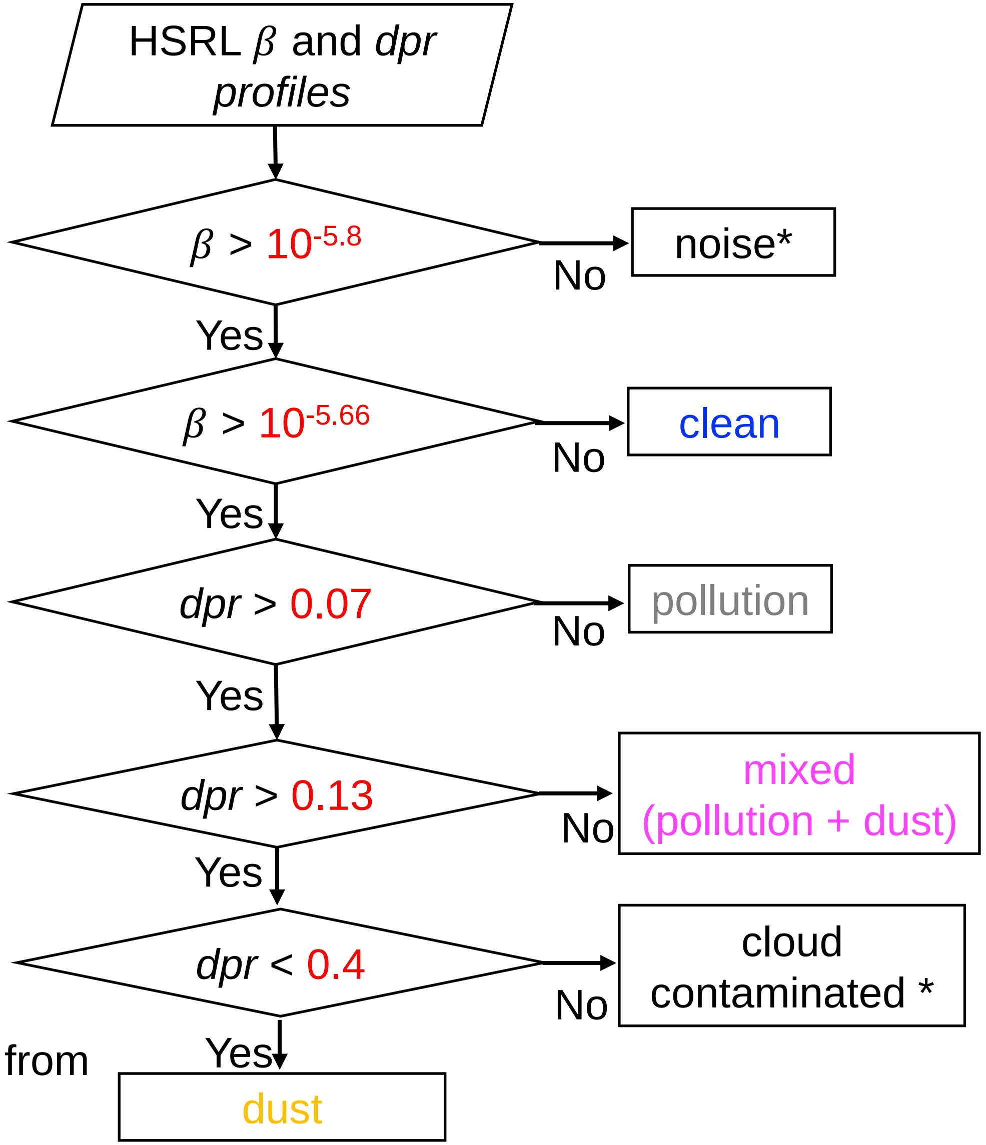
Aerosol type	Classification thresholds	Specifics
clean	• $PM_{2.5}$ < 15 μg m^{-3}	AirKorea standard for "good" air quality
pollution	• $PM_{10-2.5}$ < 75 μg m^{-3} • $PM_{2.5}/PM_{10}$ > 0.6	Determined from measured $PM_{10-2.5}$ and $PM_{2.5}/PM_{10}$ for KMA reported dust days
mixed (pollution +dust)	• $PM_{10-2.5}$ < 75 μg m^{-3} • $PM_{2.5}/PM_{10}$ ≤ 0.6	
dust	• $PM_{10-2.5}$ ≥ 75 μg m^{-3} • $PM_{2.5}/PM_{10}$ ≤ 0.4	

Aerosol type classification using surface PM observations as references

HSRL β and *dpr* within the boundary layer based on PM classification



Aerosol type classification decision tree based on HSRL β and *dpr* measurements

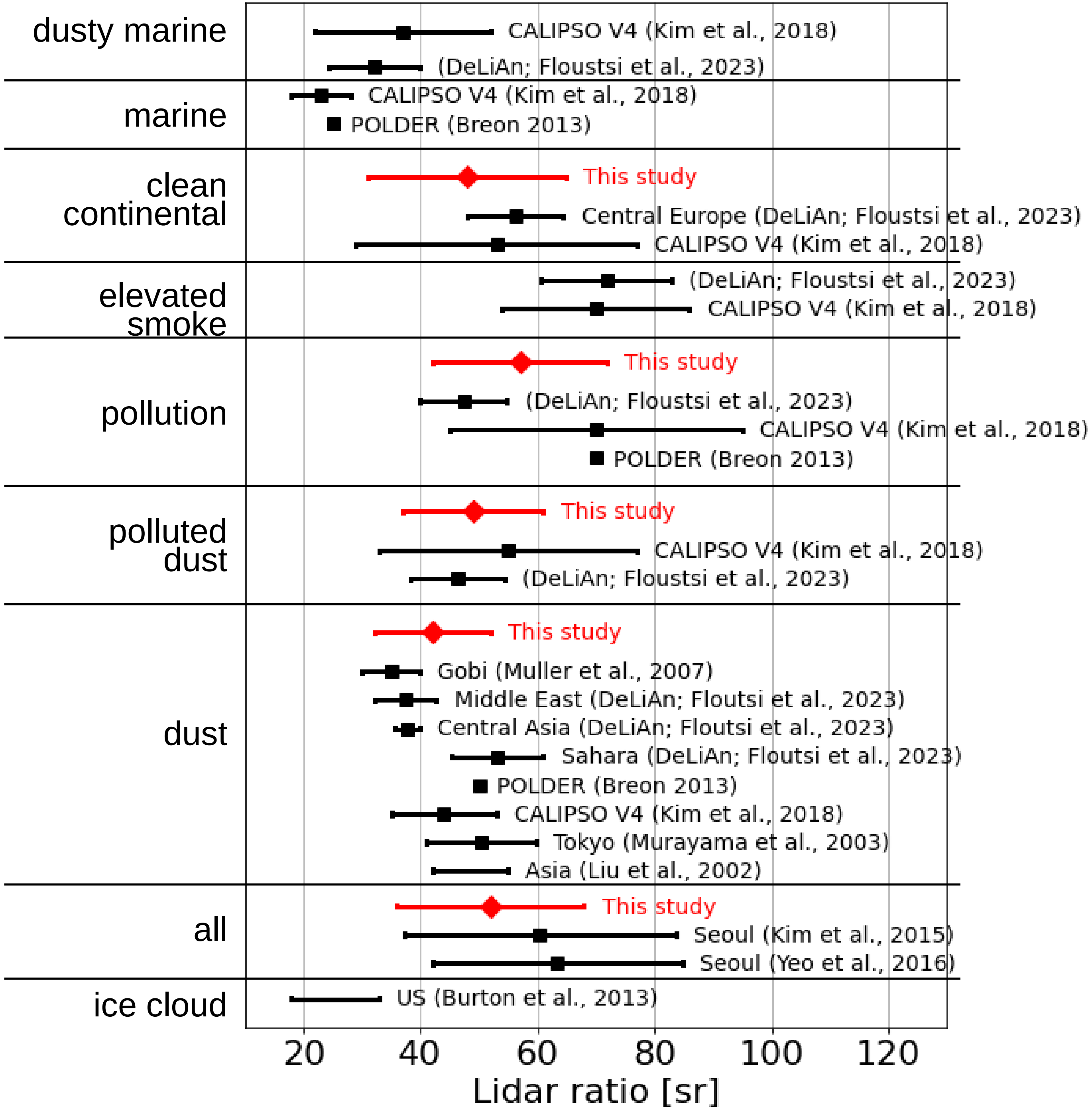


* signals classified as noise and cloud contaminated were excluded from aerosol type classification

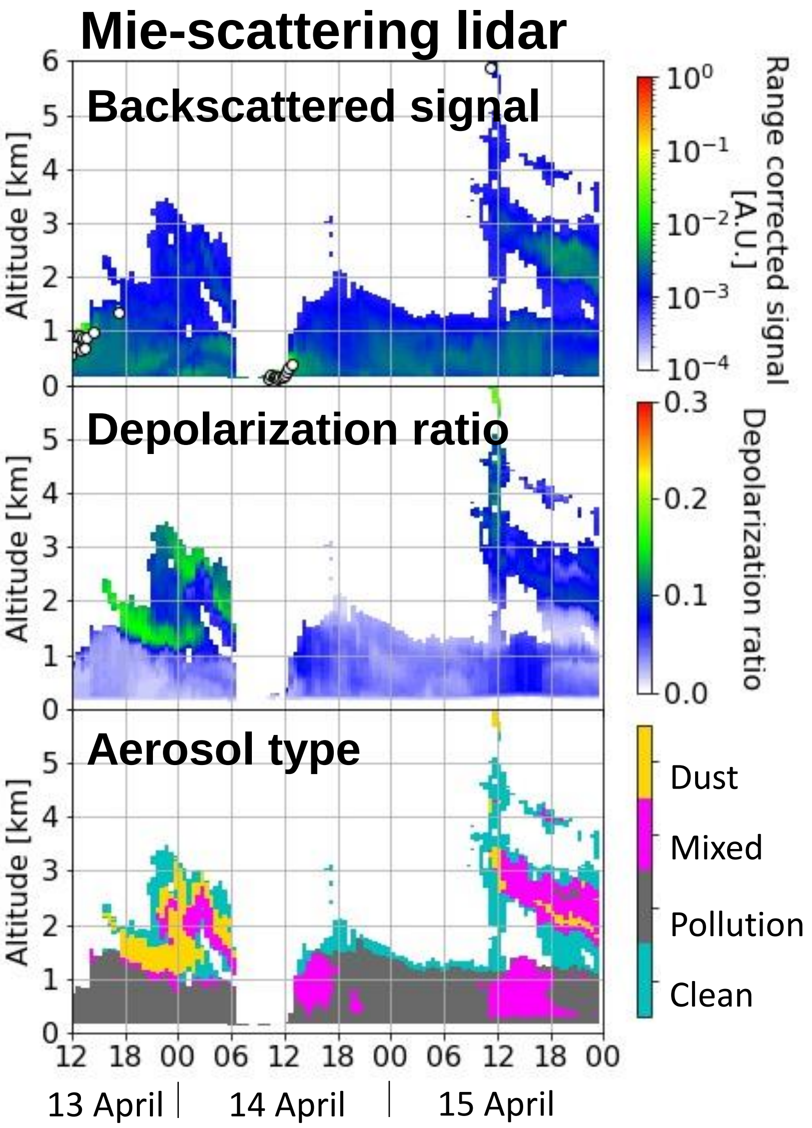
Aerosol specific lidar ratios

Aerosol type-specific lidar ratios from HSRL measurements

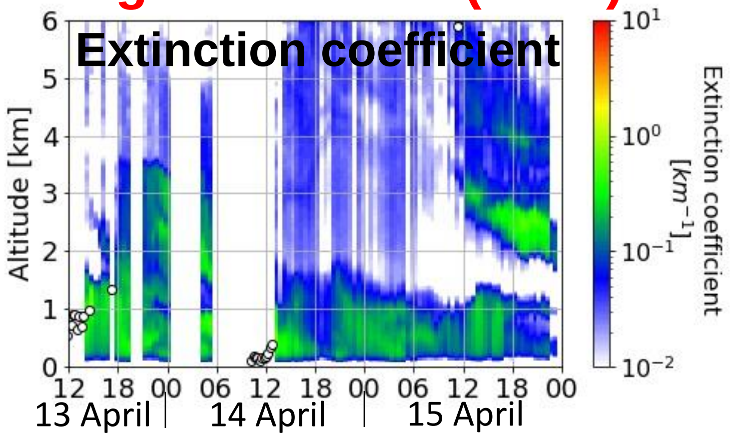
	Mean ± standard deviation [sr]	Median [sr]	Mode [sr]
clean	48 ± 17	46	36
pollution	57 ± 15	57	56
mixed	49 ± 12	49	49
dust	42 ± 10	42	38
total	52 ± 16	53	56



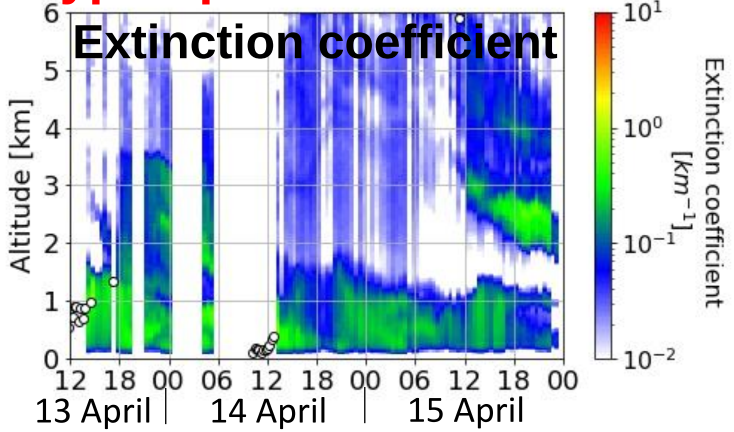
Aerosol type-specific lidar ratio implications on lidar σ_{ext} retreivals



Single lidar ratio (52 sr)

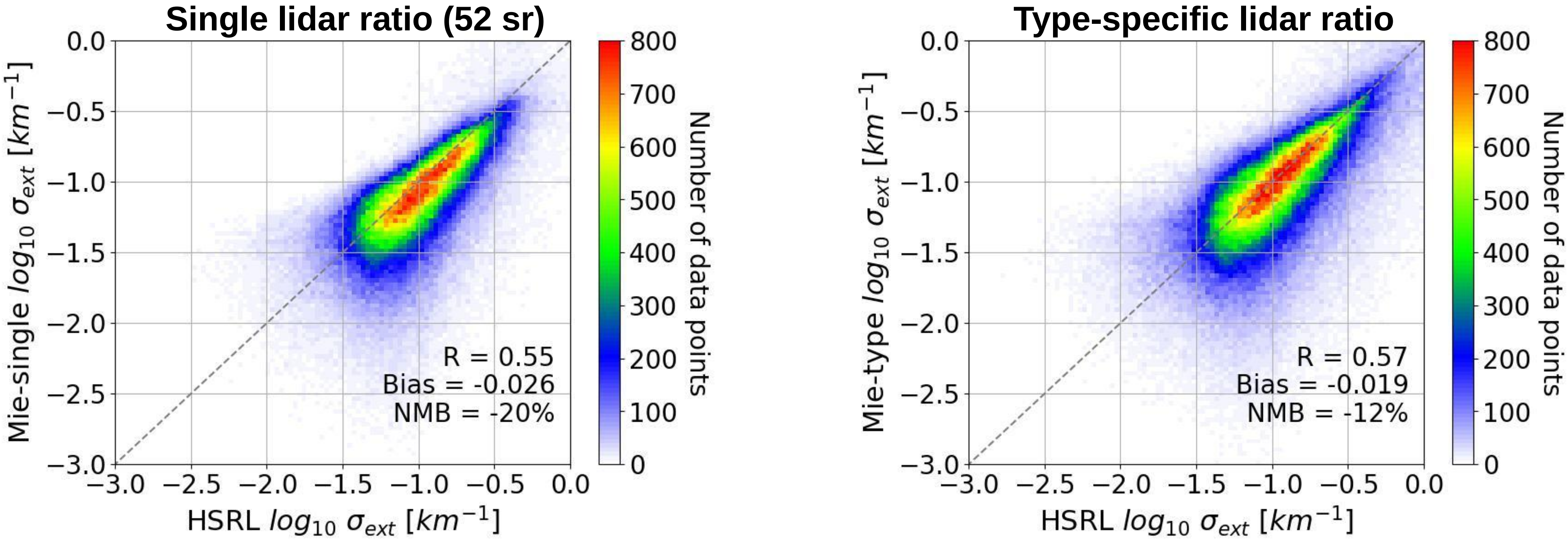


Type-specific lidar ratio



Aerosol type-specific lidar ratio implications on lidar σ_{ext} retreivals

Mar 2016 – Jan 2018
below 6 km

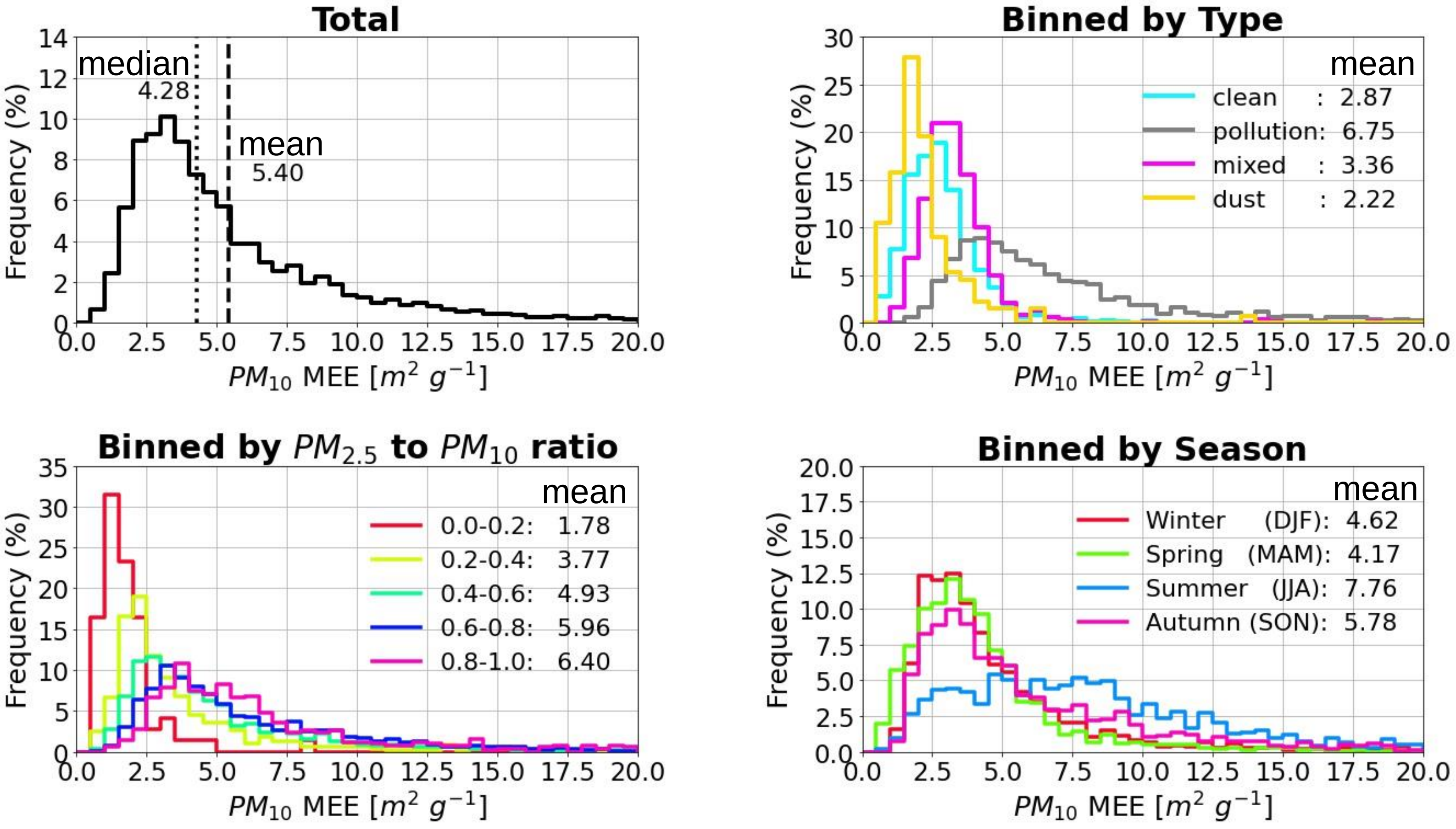


- Applying type-specific lidar ratios to Mie-scattering lidar showed better correlation scores with HSRL $\boldsymbol{\sigma_{ext}}$ measurements compared to Mie-scattering extinction results using a single lidar ratio value ($\boldsymbol{\sigma_{ext}}$ bias decreased by 7 Mm^{-1}).

→ Reduction of bias corresponding to 10% of mean AOD when using type-specific lidar ratios.

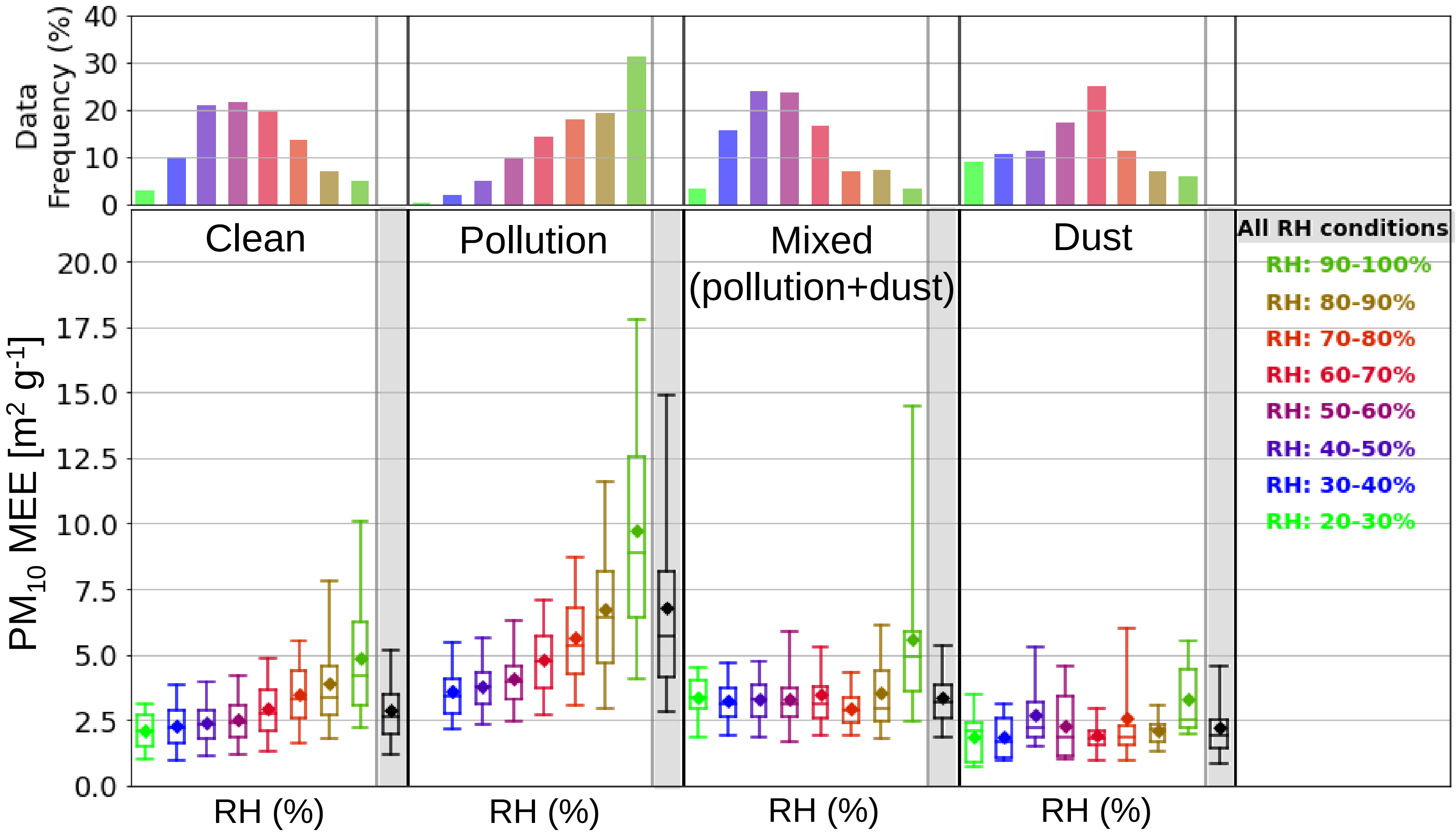
Variability of aerosol mass extinction efficiency (MEE)

$$\text{MEE} = \frac{\sigma_{ext}}{mass\ concentration}$$



- An overall mean (5.4 $m^2\ g^{-1}$) and median (4.28 $m^2\ g^{-1}$) MEE value were observed at Seoul.
- Dust aerosols displayed smallest MEE (2.22 $m^2\ g^{-1}$) and pollution aerosols the largest MEE (6.75 $m^2\ g^{-1}$).
- For low $PM_{2.5}$ to PM_{10} ratios, MEE values decreased, indicating the influence of larger dust particles with low MEE.
- Variability of MEE by season was detected due to the seasonally varying aerosol types observed at Seoul depending on the meteorological condition.

Aerosol type-specific MEE and relative humidity



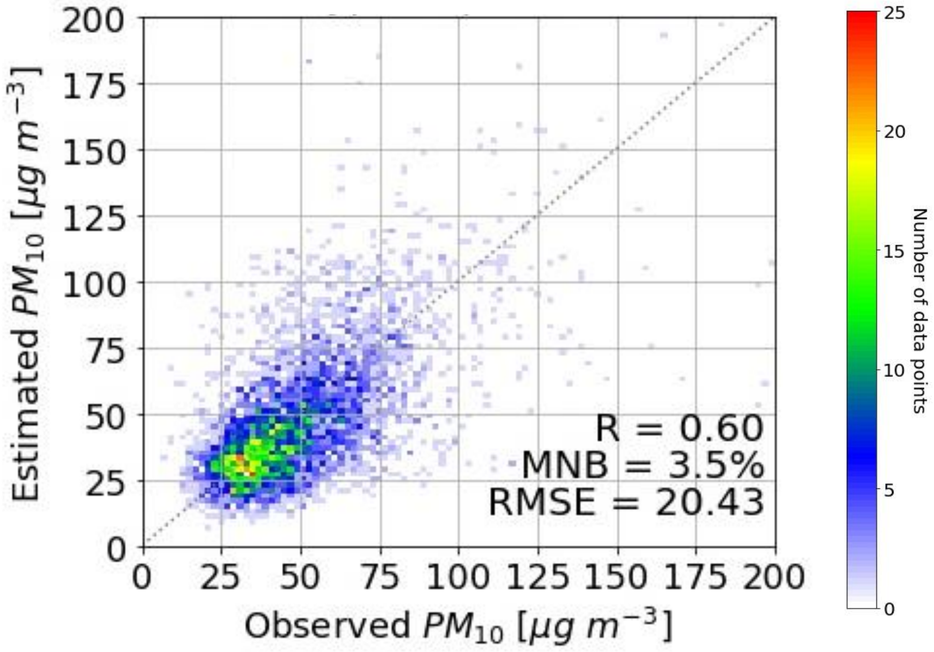
- Different rates of MEE increase with relative humidity was observed due to differences in aerosol hygroscopicity by aerosol type (Li et al., 2021; Pan et al., 2009).
- Variability of the extinction enhancement factors by aerosol type is important in estimating the radiative forcing of aerosols (Pérez-Ramírez et al., 2021; Titos et al., 2021).

The implication of MEE on PM_{10} estimation from lidar measurements

- A look-up table specifying type-specific & RH-dependent MEE values: expected mean normalized bias of 3.5%.

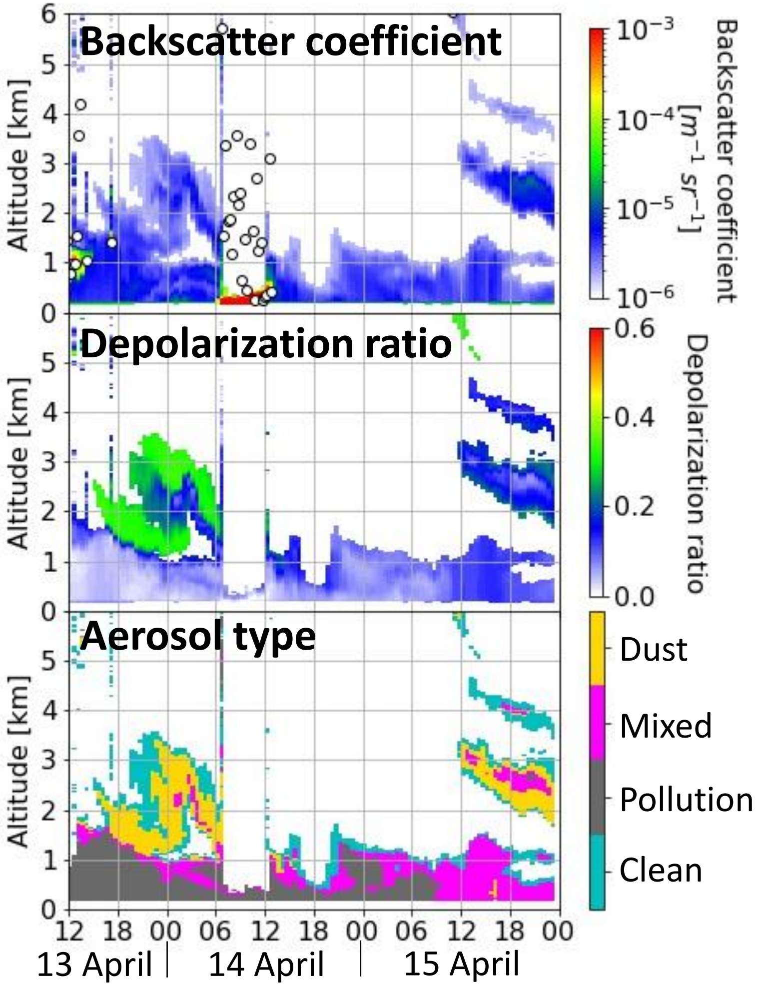
RH-dependent & type-specific MEE

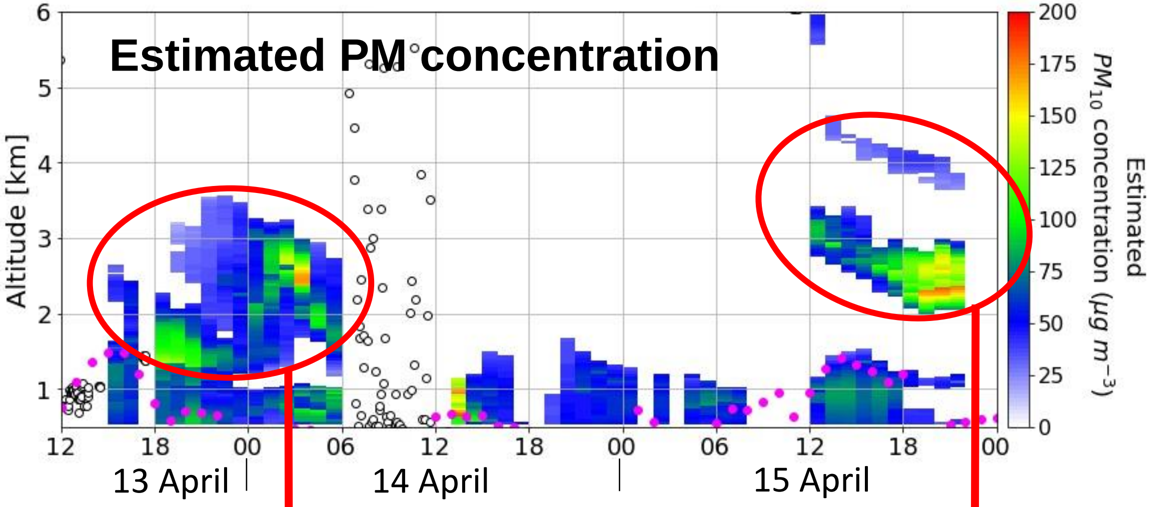
RH range	Clean	Pollution	Mixed	Dust
total	***2.87***	***6.75***	***3.36***	***2.22***
20% ≤ RH < 30%	2.11	2.85	3.36	1.87
30% ≤ RH < 40%	2.26	3.57	3.22	1.83
40% ≤ RH < 50%	2.39	3.79	3.26	2.72
50% ≤ RH < 60%	2.51	4.05	3.27	2.30
60% ≤ RH < 70%	2.90	4.82	3.46	1.90
70% ≤ RH < 80%	3.50	5.61	2.93	2.59
80% ≤ RH < 90%	3.91	6.71	3.55	2.09
90% ≤ RH < 100%	4.88	9.71	5.57	3.28



PM_{10} concentration estimated from lidar measurements

- Extinction coefficient and aerosol type information: HSRL
- Relative humidity information: ERA5 reanalysis data





Elevated dust aerosol layer
Mean PM concentration = **57 μg m^{-3}**

Elevated mixed aerosol layer
Mean PM concentration = **71 μg m^{-3}**

- The elevated dust and mixed aerosol layers observed during 13 – 15 April 2016 had mean PM concentrations of 57 μg m^{-3} and 71 μg m^{-3}, respectively.
- The mean surface PM_{10} concentration during the entire case was 62 μg m^{-3}.

Summary

- MLH from WDL measurements significantly lower nighttime MLH than other measurements (nighttime mean bias between WDL and aerosol lidar MLH = -0.26 km).
- MLH simulations from WRF-Chem PBL-YSU scheme showed close similarity with WDL measurements. However, WDL σ_w and WRF-Chem bulk Richardson number were not good representatives of nocturnal vertical mixing of aerosols (supposedly induced by wind shear).
- WRF-Chem underestimation of nocturnal MLH was speculated to have resulted in overestimation of surface $PM_{2.5}$ due to suppressed vertical mixing of aerosols.
- Applying type-specific lidar ratios to Mie-scattering lidar displayed improved correlations with HSRL σ_{ext} measurements (σ_{ext} bias reduction of 7 Mm^{-1}).
- Reduction of 10% error in AOD was predicted by using type-specific lidar ratios.
- An overall mean (5.4 m^2 g^{-1}) and median (4.28 m^2 g^{-1}) MEE value were observed at Seoul while displaying clear variability by aerosol type and ambient humidity.
- Applying RH and type-dependent MEE values to lidar measurements provided accurate surface PM concentrations (MNB = 3.5%).

Thank you for your attention

Thanks to Sang-woo Kim[1], Man-Hae Kim[1], Jong-Uk Park[1], Robert Holz[2], Ralph Kuehn[2], Edwin Eloranta[2], Ali H. Omar[3], Hyo-Jung Lee[4], Cheol-Hee Kim[4], Atsushi Shimizu[5], Tomoaki Nishizawa[5], Jin-Soo Park[6], and Joonyoung Ahn[6] for their contribution to this work.

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