



# A mixed layer height parameterization in a 3-D chemical transport model: Implications for gas and aerosol simulations

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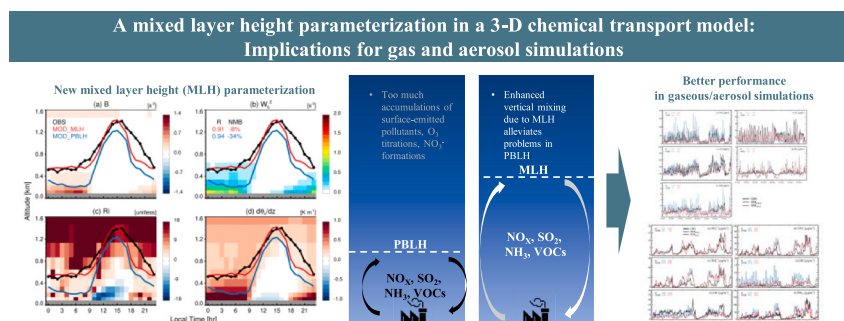
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## HIGHLIGHTS

- We introduce a novel MLH parameterization based on the YSU PBL scheme.
- Simulated MLH shows a better agreement with observed PBLH than the YSU PBL scheme.
- The model with MLH improves NO<sub>x</sub> overestimations and O<sub>3</sub> underestimations.
- Using MLH alters the speed of N<sub>2</sub>O<sub>5</sub> chemistry and vertical distributions of NH<sub>3</sub>.
- These changes lead to better model performance in aerosol simulations.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Vertical mixing within the planetary boundary layer (PBL) is crucial for determining surface-level pollutant concentrations. However, standard PBL schemes in chemical transport models (CTMs) often fail to adequately define the upper bounds of vertical mixing, particularly at night. This limitation frequently results in over-estimated nocturnal concentrations of pollutants near the surface. To address this issue, we propose a parameterization of mixed layer height (MLH) derived from the Yonsei University (YSU) PBL scheme and thoroughly evaluate it by comparing simulations with various observations. We utilized the Weather Research and Forecasting model coupled with GEOS-Chem (WRF-GC) to simulate gas and aerosol distributions over South Korea during the Satellite Integrated Joint Monitoring of Air Quality (SIJAQ) campaign in 2021. The WRF-GC simulations incorporating the MLH parameterization improved the excessive titration of O<sub>3</sub> and the overproduction of HNO<sub>3</sub> and NO<sub>3</sub> in the model. Consequently, the model performances in gaseous and aerosol simulations showed a better agreement with observations, with changes in normalized mean biases (NMBs) of NO<sub>x</sub> (from 50 % to

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–27 %), O<sub>3</sub> (from –49 % to –28 %), NO<sub>3</sub><sup>–</sup> (from 126 % to 91 %), NH<sub>4</sub><sup>+</sup> (from 113 % to 85 %), BC (from 322 % to 135 %), and PM<sub>2.5</sub> (from 58 % to 28 %).

## 1. Introduction

The planetary boundary layer (PBL) is the lowest part of the Earth's atmosphere, where energy, momentum, moisture, gases, and aerosols are exchanged between the Earth's surface and the free atmosphere (Gerbig et al., 2008; Stull, 1988). The PBL height (PBLH) marks the upper limit of the direct influence of the Earth's surface, inhibiting the vertical transport of surface-emitted pollutants to regions above the PBLH (Li et al., 2017). Thus, understanding the structure and diurnal evolution of PBLH is crucial for weather forecasting and pollution dispersion modeling.

Detecting PBLH has been a significant focus in atmospheric sciences for several decades, leading to the development of various estimation methods. A direct measurement of PBLH is impossible; instead, it is estimated indirectly through different observations. Radiosonde measurements are the primary method for detecting PBLH (Li et al., 2021; Liang and Liu, 2010). However, this technique faces challenges in providing comprehensive spatial and temporal coverage due to technical and economic limitations. Remote sensing techniques, such as lidar or satellite observations, can extend the spatiotemporal detection of PBLH, albeit with notable uncertainties (Kim et al., 2021; Park et al., 2022).

Numerical models have been employed to overcome the spatiotemporal limitations of observation-based PBLH detection (Bravo-Aranda et al., 2017; Hong et al., 2006). Various PBL schemes based on thermal and mechanical mixing have been proposed and implemented in weather forecasting and climate models, demonstrating reasonable performances compared to observations (Canuto et al., 2001; Rai and Pattnaik, 2019; Shin and Hong, 2011). The models' PBL schemes are crucial for transferring momentum, heat, and moisture between the surface and the atmosphere (Holtslag and Boville, 1993; Stull, 1988).

The PBL schemes are also essential for simulating the dispersion and fate of surface-emitted pollutants in Chemical Transport Models (CTM) (Jia and Zhang, 2020; Banks and Baldasano, 2016; Onwukwe and Jackson, 2020). However, preceding studies have reported that the conventional PBL schemes may not correctly represent the vertical mixing of pollutants in CTMs because they are highly overweighted on thermally induced convection and underestimate vertical mixing by mechanical forcing (Jordan et al., 2010; Lee et al., 2023; Oak et al., 2019; Travis et al., 2022).

This misrepresentation of PBL schemes in CTMs leads to significant uncertainties in the vertical distributions of pollutants (Shi et al., 2020). Indeed, significant overestimations in primary and secondary aerosols in CTM have been reported due to PBLH underestimation (Lee et al., 2023; Travis et al., 2022). Also, too much accumulation of simulated NO near the surface titrates O<sub>3</sub>, causing overestimations in NO<sub>x</sub> and underestimation in O<sub>3</sub> in the model (Oak et al., 2019; Travis et al., 2022). The issue with PBL representation leads to an imbalance in the exchange of trace gases between the PBL and the free troposphere, causing uncertainty in the model's vertical distribution of OH reactivity (Kim et al., 2022).

Here, we introduce a mixed layer height (MLH) parameterization based on the Yonsei University (YSU) PBL scheme to better represent the vertical mixing of trace gases and aerosols in CTMs. In micrometeorology studies, MLH is conventionally considered as a subset of PBLH. We here do not follow this conventional terminology. Our MLH parameterization aims to reproduce the lidar-detected boundary layer height based on observed vertical distributions of pollutants (Park et al., 2021a; Park et al., 2022). The MLH parameterization and its effect on gaseous and aerosol simulations are validated using observation-based PBLH data from lidar observations and observed gaseous and aerosol

concentrations in South Korea during the Satellite Integrated Joint Monitoring of Air Quality (SIJAQ) campaign in 2021. For convenience hereafter, we refer to “lidar-detected boundary layer height” as “observed PBLH”.

## 2. Observations

South Korea's National Institute of Environmental Research (NIER) conducted the SIJAQ campaign over the Korean peninsula from October to November 2021. The campaign provided an extensive observation dataset of gaseous and aerosol species at Olympic Park (127.13°E, 37.52°N, Fig. 1b), which is located in the middle of a green space surrounded by a high-density residential area and freeways in the Seoul Metropolitan Area (SMA).

Kim et al. (2023) categorized the campaign into five meteorological periods: sustained warm-stagnant (October 10–28, P1), stagnant high (October 29–November 7, P2), migratory low (November 8–14, P3), migratory high (November 15–21, P4), migratory low (November 22–25, P5). The P1 period was influenced by marine stagnation. The P2–P4 periods are a transition to continental influence. The P5 period was under continental influence. An increase in high PM<sub>2.5</sub> and PM<sub>10</sub> concentrations was found in the P2 and P4 periods. Moderate PM concentrations are shown in the P1 period, but relatively clean air quality was found in the P3 and P5 periods.

During the campaign, ion chromatography (Dionex, Sunnyvale, CA, USA) measured NO<sub>3</sub><sup>–</sup>, SO<sub>4</sub><sup>2–</sup>, and NH<sub>4</sub><sup>+</sup> aerosol concentrations every four hours. Organic carbon (OC) and black carbon (BC) aerosol concentrations were analyzed using a thermal, optical transmittance carbon analyzer (Sunset model 5) following the NIOSH method 5040 (Birch and Cary, 1996) with a 4-h temporal resolution. The beta-ray method (Shin et al., 2011; Takahashi et al., 2008) was used to measure hourly particulate matter (PM<sub>2.5</sub>) concentrations.

Gaseous species, including O<sub>3</sub>, SO<sub>2</sub>, and CO, were measured every 1 min with Ozone Analyzer 49iQ, Pulsed Fluorescence SO<sub>2</sub> Analyzer 43i, and Gas Filter Correlation CO Analyzer 48i (Thermo Fisher Scientific, USA), respectively. NO and NO<sub>2</sub> concentrations were observed every 1 min by chemiluminescence with a molybdenum converter (Marley et al., 2004) and Cavity Attenuated Phase Shift (CAPS) Trace-level NO<sub>2</sub> Analyzer.

The AirKorea monitoring network, managed by NIER and affiliated with the Korean Ministry of Environment (<https://www.airkorea.or.kr/eng>, accessed September 2, 2024), provided hourly measurements of volume mixing ratios of SO<sub>2</sub>, CO, O<sub>3</sub>, and NO<sub>2</sub> and mass concentrations of PM<sub>2.5</sub> over the stations at Seoul (Fig. 1b).

An elastic lidar located at Seoul National University (SNU, 126.95°E, 37.45°N, 116 m above sea level, Fig. 1b) measured the attenuated backscatter intensity ( $\beta$ ) and depolarization ratio of the 532 nm laser signal at 15-min intervals, with a vertical resolution of 6 m on average (Pappalardo et al., 2014; Weitkamp, 2006). To ensure data reliability, measurements up to 120 m were excluded due to incomplete overlap issues, which may impact the retrieval of observed PBLH at lower altitudes (Park et al., 2022). Cloud detection was based on total attenuated backscatter values because clouds exhibit stronger signals than aerosols over narrower altitude ranges. We discarded observations with strong backscatter values ( $\beta > 1.5 \times 10^{-5} \text{ m}^{-1} \text{ sr}^{-1}$  below 1 km and  $> 10^{-5} \text{ m}^{-1} \text{ sr}^{-1}$  above 1 km) as cloud presence (Kim et al., 2015).

The observed PBLH was determined as the height at which a drastic decrease in lidar observed  $\beta$  values occurs, assuming a uniform distribution of aerosols within PBL (Kim et al., 2007). The lidar profile was examined in moving bundles of the 3-point (90 m) (the 3-point gradient method) to detect sharp decreases in  $\beta$  (Kim et al., 2015). A possible

deterioration by the nocturnal residual layer is prevented by adopting a threshold value for the vertical gradient of the range-corrected signal, as conducted by [Park et al. \(2021a\)](#). Detailed information about calculating PBL heights using lidar data and its evaluation against other independent observations can be found in [Park et al. \(2021a\)](#). All the observations used in this study and their related information are summarized in [Table 1](#).

### 3. Model description

#### 3.1. The YSU PBL scheme

The YSU PBL scheme is an adapted K-theory model, including countergradient terms to account for the influence of large-scale eddies on the total flux ([Holtslag and Boville, 1993](#); [Hong, 2010](#); [Hong et al., 2006](#)). For  $z \leq \text{PBLH}$ , the turbulence diffusion of prognostic variables ( $C$ ), including zonal wind  $u$ , meridional wind  $v$ , vertical wind  $w$ , potential temperature  $\theta$ , specific humidity  $q$ , and air pollutant concentrations, is computed using Eq. (1) as follows:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial z} \left\{ K_C \left( \frac{\partial C}{\partial z} - \gamma_C \right) - \overline{(wC)}_h \left( \frac{z}{h} \right)^3 \right\} \quad (1)$$

, where  $K_C$  is the eddy diffusivity coefficient,  $\gamma_C$  is a correction to the local gradient to incorporate the contribution of the large-scale eddies to the total flux, and  $\overline{(wC)}_h$  is the flux at the inversion layer.  $z$  is the height of model levels, and  $h$  is PBLH. The detailed equations for calculating each term in the Eq. (1) can be found in [Hong \(2010\)](#).

PBLH,  $h$ , in the Eq. (1) is determined as the first model level from the surface, below which the averaged bulk Richardson number ( $Ri_b$ ) from the surface is equal to or greater than the critical values (0.0 for unstable and 0.25 for stable conditions) ([Hong, 2010](#)). The  $Ri_b$  is calculated using the Eq. (2) below.

$$Ri_b(z) = \frac{g[\theta_v(z) - \theta_s]z}{\theta_{va}U(z)^2} \quad (2)$$

, where  $U(z)$  is the horizontal wind speed at  $z$ , and  $\theta_v(z)$  is the virtual potential temperature at  $z$ .  $g$  is gravitational acceleration.  $\theta_{va}$  is the

**Table 1**

Observations used in this study and related information.

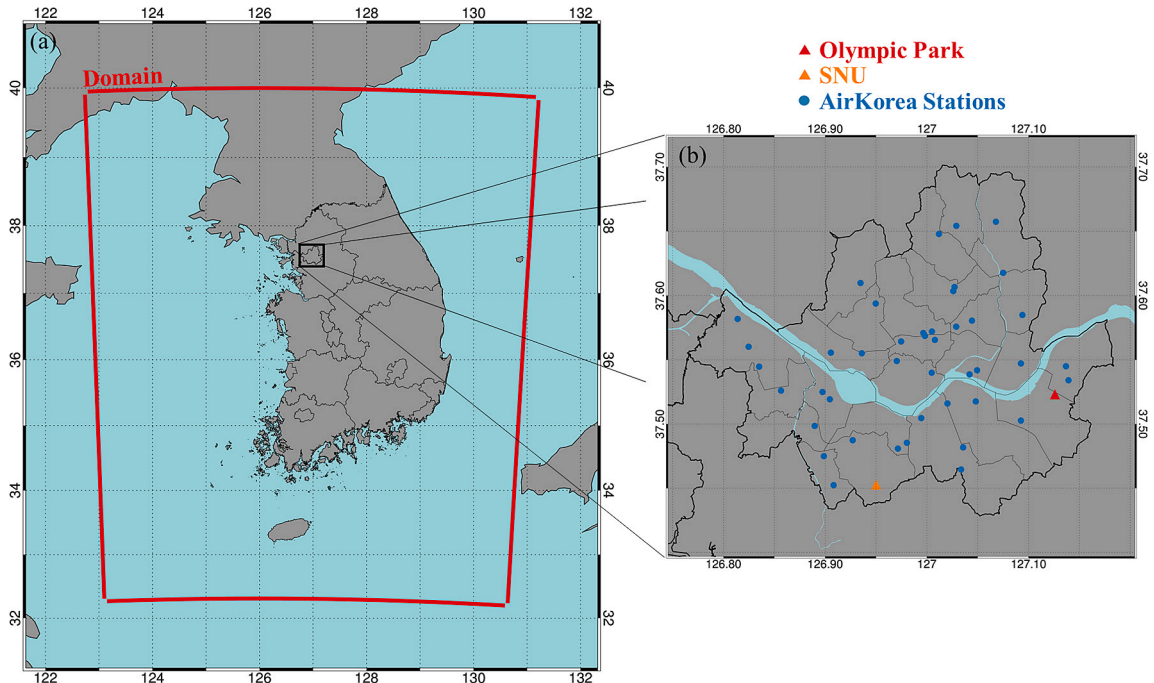
| Species                                                | Instrument                                                                                   | Resolution     | References                                                                     |
|--------------------------------------------------------|----------------------------------------------------------------------------------------------|----------------|--------------------------------------------------------------------------------|
| $\text{NO}_3^-$ , $\text{SO}_4^{2-}$ , $\text{NH}_4^+$ | Ion chromatography                                                                           | 4-h            | (Dionex, Sunnyvale, CA, USA)                                                   |
| BC, OC                                                 | A thermal, optical transmittance carbon analyzer (Sunset model 5) with the NIOSH method 5040 | 4-h            | ( <a href="#">Birch and Cary, 1996</a> )                                       |
| $\text{PM}_{2.5}$                                      | The beta-ray method                                                                          | 1-h            | ( <a href="#">Shin et al., 2011</a> ; <a href="#">Takahashi et al., 2008</a> ) |
| $\text{O}_3$                                           | Ozone Analyzer 49iQ                                                                          | 1-min          | (Thermo Fisher Scientific, USA)                                                |
| $\text{SO}_2$                                          | Pulsed Fluorescence $\text{SO}_2$                                                            | 1-min          | (Thermo Fisher Scientific, USA)                                                |
| CO                                                     | Gas Filter Correlation CO Analyzer 48i                                                       | 1-min          | (Thermo Fisher Scientific, USA)                                                |
| NO                                                     | chemiluminescence with a molybdenum converter,                                               | 1-min          | ( <a href="#">Marley et al., 2004</a> )                                        |
| $\text{NO}_2$                                          | Cavity Attenuated Phase Shift (CAPS)                                                         | 1-min          | ( <a href="#">Marley et al., 2004</a> )                                        |
| $\beta$ , depolarization ratio                         | Trace-level $\text{NO}_2$ Analyzer<br>The elastic lidar                                      | 15-min,<br>6 m | ( <a href="#">Kim et al., 2007</a> ; <a href="#">Kim et al., 2015</a> )        |

virtual potential temperature at the lowest model level.  $\theta_s$  is a temperature calculated as follows:

$$\theta_s = \theta_{va} + \theta_T, \text{ where } \theta_T = a \frac{\sqrt{(w\theta_v')_0}}{w_{s0}} \quad (3)$$

, where the proportionality factor  $a$  is 6.8. The mixed layer velocity scale  $w_{s0}$  is the value at  $z = 0.5 h$ , and the detailed expression for its calculation can be found in [Hong \(2010\)](#).

As will be discussed, however, the PBLH in the YSU PBL scheme is significantly shallower than the observed PBLH ([Oak et al., 2019](#); [Travis et al., 2022](#)), especially during nighttime. We also find that this nighttime underestimation of PBLH is a general problem found in other schemes in WRF, including Mellor–Yamada–Janjic (MYJ) and



**Fig. 1.** Map of the model domain and the locations of the Olympic Park site (127.13°E, 37.52°N), the SNU site (126.95°E, 37.45°N), and the AirKorea sites.

Asymmetrical Convective Model v2 (ACM2) schemes (Lee et al., 2023; Mantovani Júnior et al., 2023). Our parameterization addresses this issue based on the systematic evaluation of simulated PBLH and thermodynamic variables against the available observations during the SIJAQ 2021 campaign over the Korean peninsula.

### 3.2. A new MLH parameterization

Here, we introduce a new parameterization of MLH to detect an upper limit of the thermally/mechanically-induced vertical mixing and replace PBLH in CTMs. We find new criteria to determine MLH based on the detailed analysis of vertical structures and evolutions of related variables such as buoyancy ( $B$ ), wind shear ( $W_s$ ), local Richardson number ( $R_i$ ) (Woods, 1969), and  $\theta_v$  during the SIJAQ 2021 campaign.  $B$  and  $W_s$  are expressed as follows:

$$B(z) = \frac{g}{\theta_v} \frac{\partial \theta_v}{\partial z} \quad (4)$$

$$W_s(z) = \sqrt{\left(\frac{\partial u}{\partial z}\right)^2 + \left(\frac{\partial v}{\partial z}\right)^2} \quad (5)$$

$R_i$  is the ratio of the buoyancy to the flow shear, expressed as:

$$R_i(z) = \frac{B(z)}{W_s(z)^2} \quad (6)$$

For the MLH parameterization, we use  $d\theta_v/dz$  and  $R_i$  to account for mechanical and thermal-induced mixing. At each model level, locally computed  $B$ ,  $W_s$ , and  $R_i$  are examined to determine MLH. We differentiate ways to determine MLH for day and night conditions.

For the night condition, we use  $\theta_v$ , a conservative quantity in the mixed layer, to find the top of the mixed layer. Then, we compare the  $R_i$  value at the top with the lower levels to check whether mechanical mixing can occur based on stability. Based on this concept and through many sensitivity tests, we set criteria to detect the MLH: the altitude below which  $\theta_v$  is uniform ( $|d\theta_v/dz| < 0.0075 \text{ K m}^{-1}$ ) and  $R_i$  begins to increase. For safety purposes, the nighttime upper limit of MLH is set to the nighttime maximum value of observation (800 m) during the campaign. The lower limit of MLH is also set to PBLH from the YSU PBL scheme.

For the daytime condition, we extend MLH to the top of the entrainment zone, where vertical mixing above the PBLH occurs due to countergradients. In the case of the YSU PBL scheme, the top of the entrainment zone is determined based on an arbitrary criterion  $\epsilon$  that is a ratio of the entrainment zone to the depth of the vertical layers of the model. However, this criterion does not work effectively due to the coarse resolution of the vertical grids of the model ( $\sim 120 \text{ m}$  at the first model grid and more coarse at the above grids) unless the vertical resolution gets finer to  $\sim 20 \text{ m}$ . Thus, we increase the criterion value of  $\epsilon$  from 4.6 to 50, considering the interval between the vertical grids of the model, based on a discussion with the developer of the YSU PBL scheme (Hong, 2010; Hong et al., 2006).

Fig. 2 shows simulated vertical profiles of  $\theta_v$ ,  $R_i$ , and their vertical gradients on October 26, 2021, at 22KST during the campaign. The observed and simulated PBLH and MLH are also shown in the figure. The new parameterization appears to capture discontinuity at  $\sim 700 \text{ m}$  well, where both  $\theta_v$  and  $R_i$  start to increase. The determined MLH agrees well with the observed PBLH, whereas the simulated PBLH was significantly low.

Fig. 3 presents campaign mean diurnal variations of observed and simulated PBLH and MLH along with hourly columns of simulated  $B$ ,  $W_s^2$ ,  $R_i$ , and  $d\theta_v/dz$ . The observed PBLH sustains  $\sim 0.5 \text{ km}$  at night and grows up to  $\sim 1.5 \text{ km}$  during the day. The YSU PBL scheme simulates PBLH lower than the observations. The discrepancy in the model is especially significant during the night. This nocturnal underestimation is likely because the YSU PBL scheme diagnoses PBLH based on the averaged bulk Richardson number from the surface to PBLH. This mean quantity likely underestimates local vertical motion caused by mechanical wind shear (S. Hong, personal communication, May 2022).

The MLH parameterization, on the other hand, is based on the local Richardson number, which enables us to consider local stability at each level. The simulated MLH appears to capture the observed PBLH well. The improvement is pronounced at night when non-negligible  $W_s$ , relatively weak  $B$ , and a decrease in  $R_i$  near  $0.5 \text{ km}$  occur, indicating vertical mixing by mechanical forcing.

Although our new parameterization captures the observation well compared to the conventional YSU scheme, we acknowledge that our evaluation is relatively limited to the SIJAQ campaign period in the Korean peninsula. More extensive evaluation of the parameterization by comparing against observations, including radiosonde data, should be

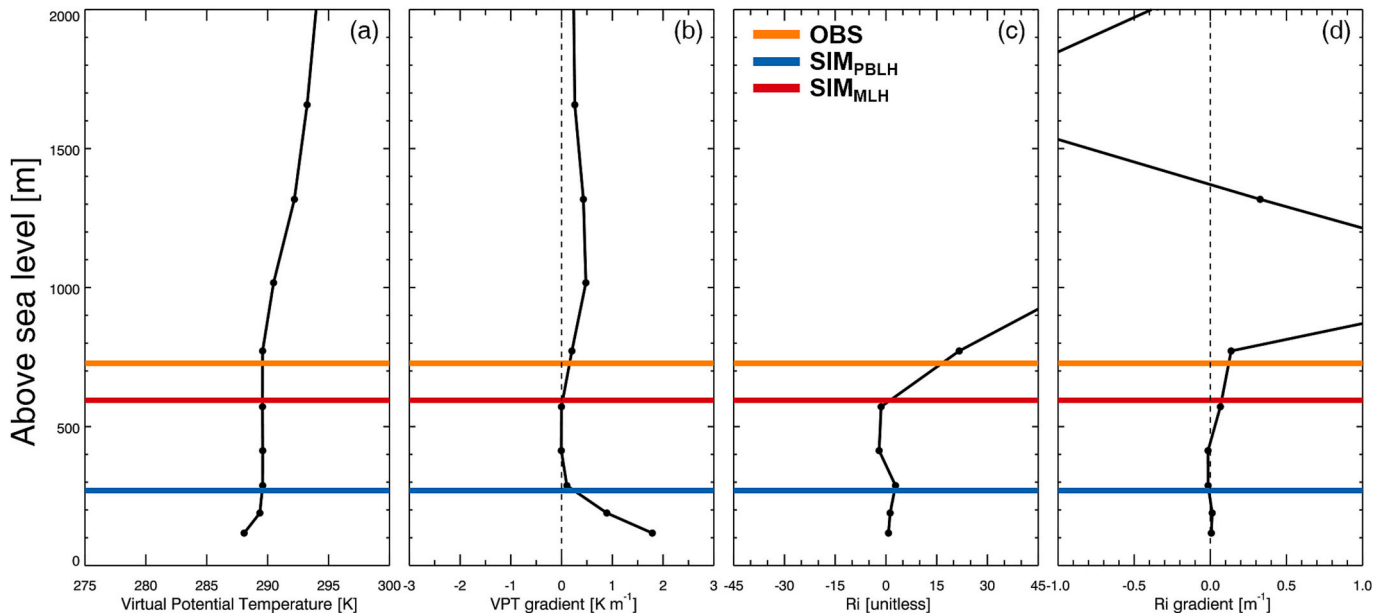
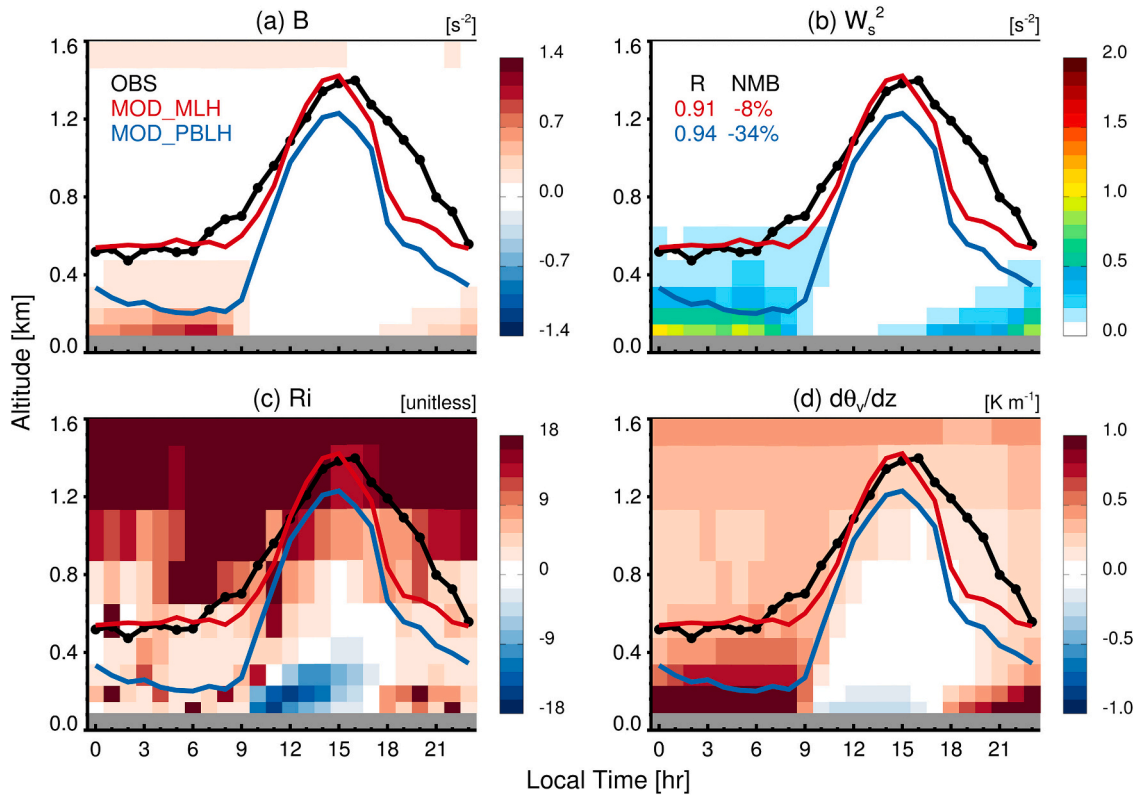


Fig. 2. An example case of MLH determination, presenting simulated vertical profiles of (a)  $\theta_v$ , (b)  $d\theta_v/dz$ , (c)  $R_i$ , and  $dR_i/dz$  on October 26, 2021, at 22KST. Each colored solid line indicates observed PBLH (yellow), simulated PBLH (blue), and simulated MLH (red).





**Fig. 3.** Averaged diurnal variations of observed PBLH (black), simulated PBLH (blue) from the YSU PBL scheme, and simulated MLH (red). Color-shaded areas indicate hourly varying columns of (a)  $B$ , (b)  $W_s^2$ , (c)  $R_i$ , and (d)  $d\theta_v/dz$ , from model simulations during the SIJAQ2021 campaign at the SNU.

necessary for different times and areas.

### 3.3. WRF-GC

We used the WRF-GC version 1.0, an online coupling of the Weather Research and Forecasting (WRF) mesoscale meteorological model (WRF v3.9.1.1) and the GEOS-Chem chemistry model v12.2.1 (Lin et al., 2020). The WRF model provides meteorological variables to drive GEOS-Chem simulations of gas and aerosol species in the atmosphere. More details about the WRF-GC can be found in Lin et al. (2020).

Fig. 1a shows a model domain to cover South Korea (122.7–131.2°E, 32.2–40.0°N) with 9 km × 9 km resolutions and 50 hybrid-sigma vertical layers (up to 10 hPa). We used the final operation global analysis data from the Global Forecasting System of National Centers for Environment Prediction (NCEP-FNL) for meteorological initial and boundary conditions every 6-h.

Table 2 summarizes physical schemes used in WRF simulations. The Rapid Radiative Transfer Model for General Circulation Models (RRTMG) computed short/longwave radiative transfer calculations in the atmosphere (Clough et al., 2005). The Unified Noah land-surface model calculated various processes between the land-surface and the atmosphere in the model (Ek et al., 2003). Grid and subgrid-scale clouds were simulated using the WRF Single Moment 5-class (WSM5) (Hong et al., 2004) and the newer Tiedtke scheme (Hamilton et al., 2011; Tiedtke, 1989). A single-layer urban canopy model (Kusaka and Kimura, 2004) was adopted to simulate urban processes, including building-induced turbulences and the heat island effect. The electronic architectural administration information system (EAIS) of the Ministry of Land, Infrastructure, and Transport of Korea provides statistics for buildings as an input for the urban canopy model (<https://www.eais.go.kr/moct/awp/aec02/AWPAEC02L01>, accessed September 2, 2024). The YSU PBL scheme (Hong et al., 2006) calculated PBLH and variables related to turbulences in PBL. Furthermore, we adopted a new MLH

**Table 2**

Summary of physical schemes used in this study.

| Physical process                           | Option                                                                    | References                             |
|--------------------------------------------|---------------------------------------------------------------------------|----------------------------------------|
| Horizontal grid                            | 78 cells × 93 cells ( $\Delta X = 9$ km)                                  |                                        |
| Vertical Layer                             | 50 layers (up to 10 hPa)                                                  |                                        |
| Short/Long wave radiation                  | The Rapid Radiative Transfer Model for General Circulation Models (RRTMG) | (Clough et al., 2005)                  |
| Land-surface exchange                      | The Unified Noah land-surface model                                       | (Ek et al., 2003)                      |
| Grid-scale cloud                           | The WRF Single Moment 5-class (WSM5)                                      | (Hong et al., 2004)                    |
| Subgrid-scale cloud                        | New Tiedtke scheme                                                        | (Hamilton et al., 2011; Tiedtke, 1989) |
| Urban processes                            | A single-layer urban canopy model                                         | (Kusaka and Kimura, 2004)              |
| Turbulence                                 | The YSU PBL scheme                                                        | (Hong et al., 2006)                    |
| Meteorological initial/boundary conditions | NCEP FNL ( $0.5^\circ \times 0.5^\circ$ )                                 |                                        |

parameterization in the model and conducted several sensitivity studies to understand the effects of mixing on chemistry, especially for nighttime. One thing to note is that MLH was calculated in WRF but only used for the vertical mixing processes of chemistry simulations to evaluate the impacts of MLH on chemistry simulations.

The WRF-GC computes coupled  $O_x$ - $NO_x$ -VOC-halogen-aerosol chemistry in the troposphere (Bey et al., 2001) using unified tropospheric-stratospheric chemistry extension (UCX) (Eastham et al., 2014). The heterogeneous reaction of  $N_2O_5$ , a primary source of nighttime  $HNO_3$  and  $NO_3^-$ , is simulated based on the method by Bertram and Thornton (2009), considering an organic coating effect on the  $N_2O_5$  uptake (McDuffie et al., 2019) in the model. A thermodynamic equilibrium between gaseous and aerosol species is calculated using an aerosol thermodynamic model, ISORROPIA II (Fountoukis and Nenes,

2007). The simple secondary organic aerosol (SOA) scheme is used to simulate a single lumped SOA precursor that is irreversibly converted to a single lumped SOA tracer (SOAS) with a molecular weight of 150 g mol<sup>-1</sup> (Hayes et al., 2015; Hodzic and Jimenez, 2011; Shrivastava et al., 2017). We used MLH and eddy diffusivity from the new MLH parameterization to simulate the PBL mixing of chemical species in the model.

The model of Emission of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012) simulated isoprene, methyl butanol, etc., from biogenic sources based on activity factors, canopy environment, leaf age, and soil moisture. We adopted the SIJAQ v2 emission inventory for anthropogenic emissions over East Asia, developed by Konkuk University. This inventory provides anthropogenic CO, NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, and VOCs emissions for 2019 with the SAPRC-07 chemical mechanism (Carter, 2010). Species in SAPRC-07 were mapped onto the WRF-GC species (Li et al., 2014). The annual total emissions of CO, NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, and VOCs are 0.8, 1.1, 0.3, 0.3, and 1.0 Tg/yr for South Korea, respectively. The chemical boundary conditions were provided every 6-h from the Global/Regional Integrated Model system-Chemistry Climate Model (GRIMs-CCM), developed by coupling the chemistry modules of the GEOS-Chem to the GRIMs general circulation model (Koo et al., 2022; Lee et al., 2022).

We conducted several model simulations with different mixing schemes from 2019.10.17 00UTC to 2019.11.26 00UTC. The simulated PBLH and MLH were validated by comparing them against observed PBLH derived by lidar at SNU. Furthermore, we evaluated the model performances by comparing simulated versus observed gaseous/aerosol concentrations at Olympic Park. For convenience, WRF-GC simulations with PBLH and MLH are hereafter called SIM<sub>PBLH</sub> and SIM<sub>MLH</sub>, respectively.

#### 4. Model evaluation

Fig. 4 shows the hourly mean observed versus simulated PBLH and MLH at SNU during the campaign. The observed PBLH shows typical diurnal variations that increase in the daytime and decrease at night. The daytime peak of observed PBLH reaches up to 1900 m, whereas the nighttime PBLH sustains above 400 m. Simulated values also reproduce observed diurnal variations. As mentioned above, however, simulated

PBLH shows significant underestimation, especially at night, with a normalized mean bias (NMB) of -49 % compared to the observed values. Our comparison is consistent with previous studies reporting nighttime underestimations of simulated PBLH during the KORUS-AQ campaign (Oak et al., 2019; Travis et al., 2022). On the other hand, the simulated MLH presents a better agreement with the observation than the simulated PBLH, showing -10 % of NMB during the campaign. Especially, the new parameterization well captures nighttime observed PBLH of 400–500 m. The simulated MLH also shows better agreement in reproducing daily peaks of observed PBLH than the simulated PBLH.

Here, we also examined the impact of using the simulated MLH instead of the simulated PBLH on model performances in gaseous and aerosol simulations. Figs. 5 and S1 compare gaseous species concentrations from SIM<sub>PBLH</sub> and SIM<sub>MLH</sub> with observations at the Olympic Park site during the campaign. The observed NO and NO<sub>2</sub> concentrations show bimodal increases because of enhancement in mobile emissions in rush hour, reaching up to 200 ppbv and 60 ppbv, respectively. The observed O<sub>3</sub> concentrations typically increase in the daytime and decrease at night. The peak concentration of O<sub>3</sub> reached 75 ppbv during the campaign. The observed CO concentrations also show bimodal increases in rush hour and the ranges of 50–750 ppbv. The observed SO<sub>2</sub> and NH<sub>3</sub> concentrations vary with the ranges of 1.5–4 ppbv and 2–13 ppbv but do not present distinct diurnal variations.

The simulated gaseous species from SIM<sub>PBLH</sub> and SIM<sub>MLH</sub> generally reproduce the observed day-to-day variations of NO<sub>2</sub>, O<sub>3</sub>, and CO, showing moderate correlation coefficients (R) over 0.4. However, for NO, SO<sub>2</sub>, and NH<sub>3</sub>, both SIM<sub>PBLH</sub> and SIM<sub>MLH</sub> show poor correlation coefficients smaller than 0.3. These poor correlations can be attributed to the representation issue of the Olympic Park site in the model. The site is located in the middle of a green space surrounded by a high-density residential area and freeways in the SMA, which cannot be resolved due to the coarse spatial resolution of the model. SIM<sub>MLH</sub> is slightly less correlated with the observations than SIM<sub>PBLH</sub>, except for NO during the campaign, but the difference between the two models is insignificant. However, we find that campaign-averaged diurnal variations were well captured by SIM<sub>MLH</sub> with higher R values than SIM<sub>PBLH</sub>, except for NO<sub>2</sub>, as shown in Fig. S1.

The difference between SIM<sub>PBLH</sub> and SIM<sub>MLH</sub> stands out at night. The

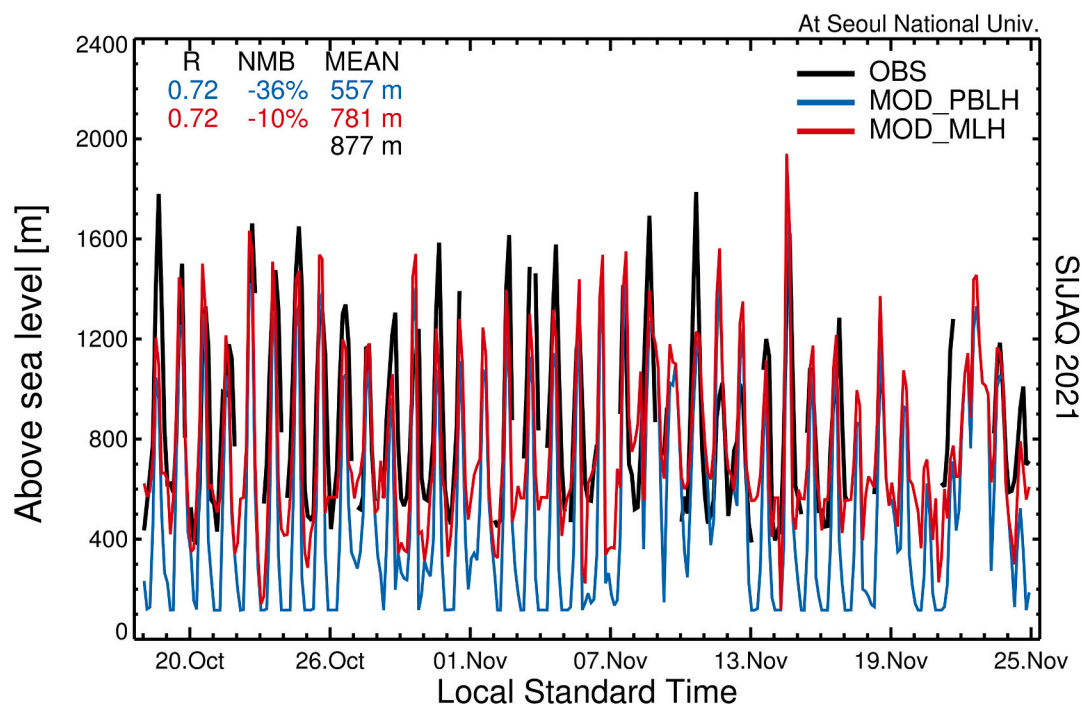
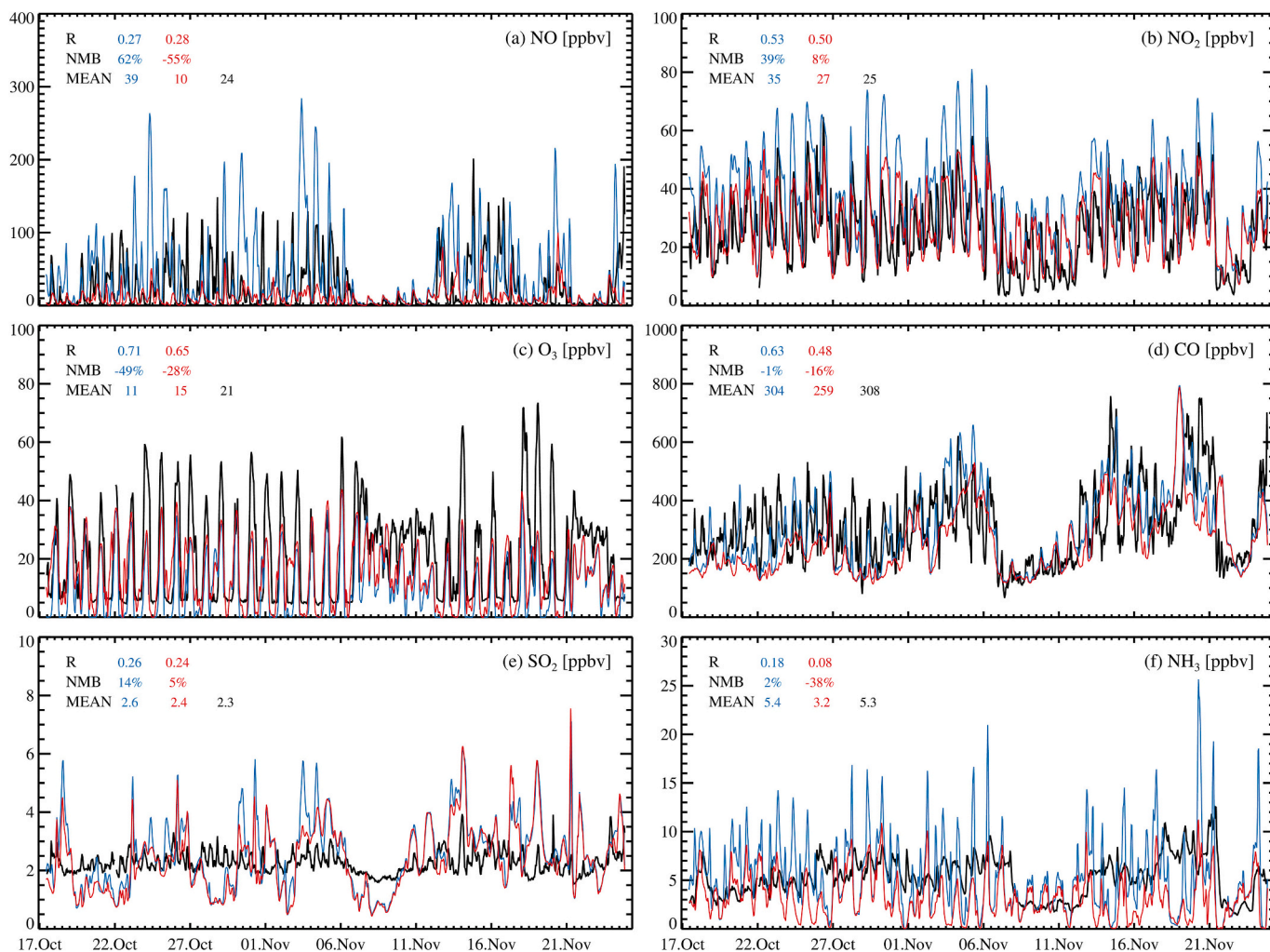


Fig. 4. Hourly time series of the observed PBLH (black) and simulated PBLH (blue) and MLH (red) during the SIJAQ 2021 campaign.



**Fig. 5.** Hourly time series of the observed (black) and simulated gaseous species from SIM<sub>PBLH</sub> (blue) and SIM<sub>MLH</sub> (red), including (a) NO, (b) NO<sub>2</sub>, (c) O<sub>3</sub>, (d) CO, (e) SO<sub>2</sub>, and (f) NH<sub>3</sub> at the Olympic Park site during the SIJAQ 2021 campaign.

nighttime underestimation of PBLH in SIM<sub>PBLH</sub> leads to the accumulation of surface-emitted NO and NO<sub>2</sub> near the surface and its significant overestimation with 62 % and 39 % of NMB, respectively. However, extended mixing volume in SIM<sub>MLH</sub> alleviates this overestimation in NO to -55 % and in NO<sub>2</sub> to 8 % of NMB. The nighttime NO overestimation in SIM<sub>PBLH</sub> also triggers too fast titration of O<sub>3</sub> and significant underestimation in O<sub>3</sub> with -49 % NMB. On the other hand, SIM<sub>MLH</sub> shows some improvement in the nighttime NO-O<sub>3</sub> titration problem of SIM<sub>PBLH</sub>, reducing the NMB of O<sub>3</sub> to -28 %.

Simulated CO and SO<sub>2</sub> in SIM<sub>MLH</sub> are slightly lower than in SIM<sub>PBLH</sub> due to faster ventilation by the extended MLH. Regarding biases in the model, SO<sub>2</sub> gets better, but CO worsens in SIM<sub>MLH</sub>. CO underestimation is attributed to significant uncertainties in the CO emission inventory in East Asia (Park et al., 2021b). Unlike other species, SIM<sub>PBLH</sub> and SIM<sub>MLH</sub> fail to capture observed variation and concentrations of NH<sub>3</sub>. It is likely related to not only uncertainty in the diurnal variation of NH<sub>3</sub> emissions (Jeppsson, 2002; Lonsdale et al., 2017) but also significant uncertainties in measuring NH<sub>3</sub> concentrations due to its high reactivity, water solubility, and low concentrations (Schobesberger et al., 2023; Sutton et al., 2008).

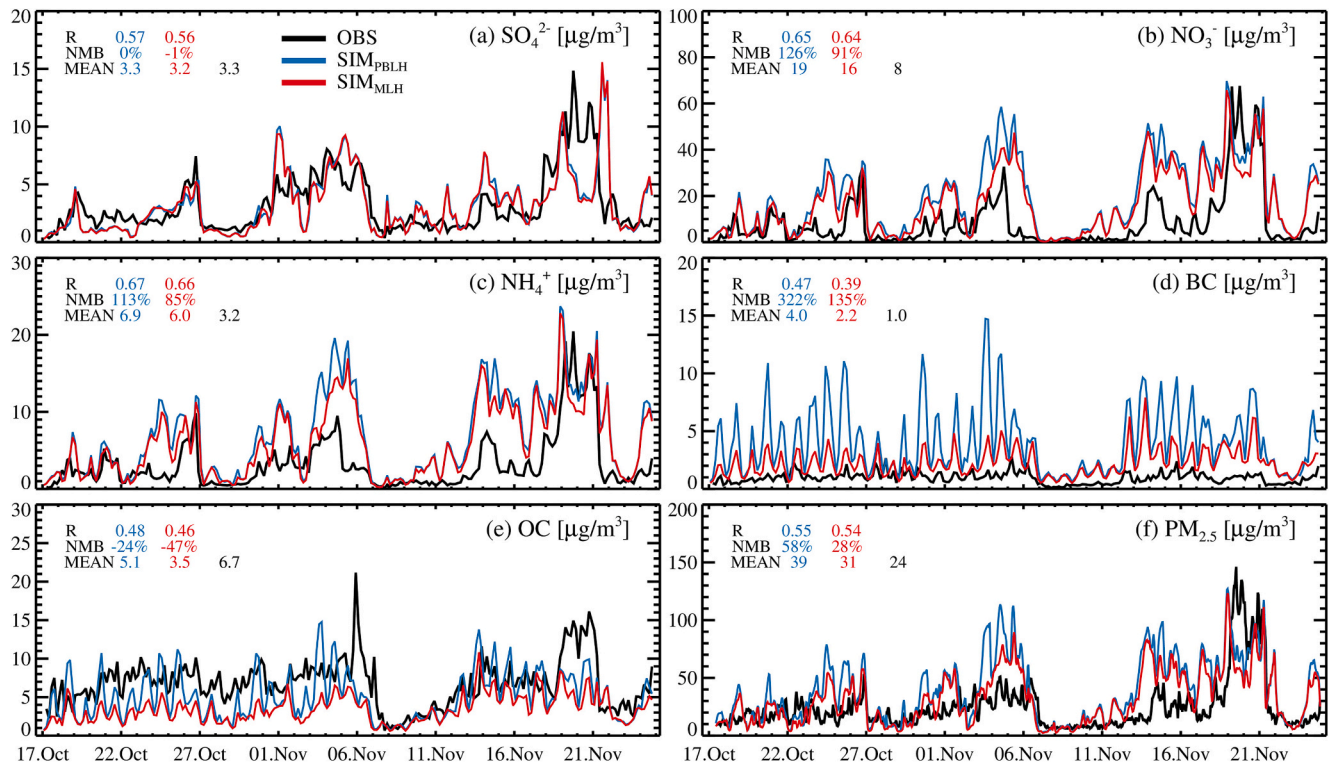
In Figs. 6 and S2, simulated aerosol species from SIM<sub>PBLH</sub> and SIM<sub>MLH</sub> are compared with observations at the Olympic Park site during the campaign. The daily observed SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup> vary consistently, showing almost zero concentrations under conditions favorable to ventilation in P3 but showing peaks of 15 μg m<sup>-3</sup> in SO<sub>4</sub><sup>2-</sup>, 65 μg m<sup>-3</sup> in

NO<sub>3</sub><sup>-</sup>, and 20 μg m<sup>-3</sup> in NH<sub>4</sub><sup>+</sup> in P4. The observed BC and OC also vary similarly to SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and NH<sub>4</sub><sup>+</sup>, with daily ranges of 0–3 μg m<sup>-3</sup> in BC and 1–21 μg m<sup>-3</sup> in OC, respectively. The observed PM<sub>2.5</sub> concentrations vary in the range of 5–150 μg m<sup>-3</sup>, contributed mainly by NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, and OC. The diurnal variations of aerosol species are shown in Fig. S2. Although the 4-hour time resolutions of the aerosol measurements make it difficult to recognize diurnal variations of the species, SIM<sub>MLH</sub> shows better NMBs than SIM<sub>PBLH</sub>, except for OC.

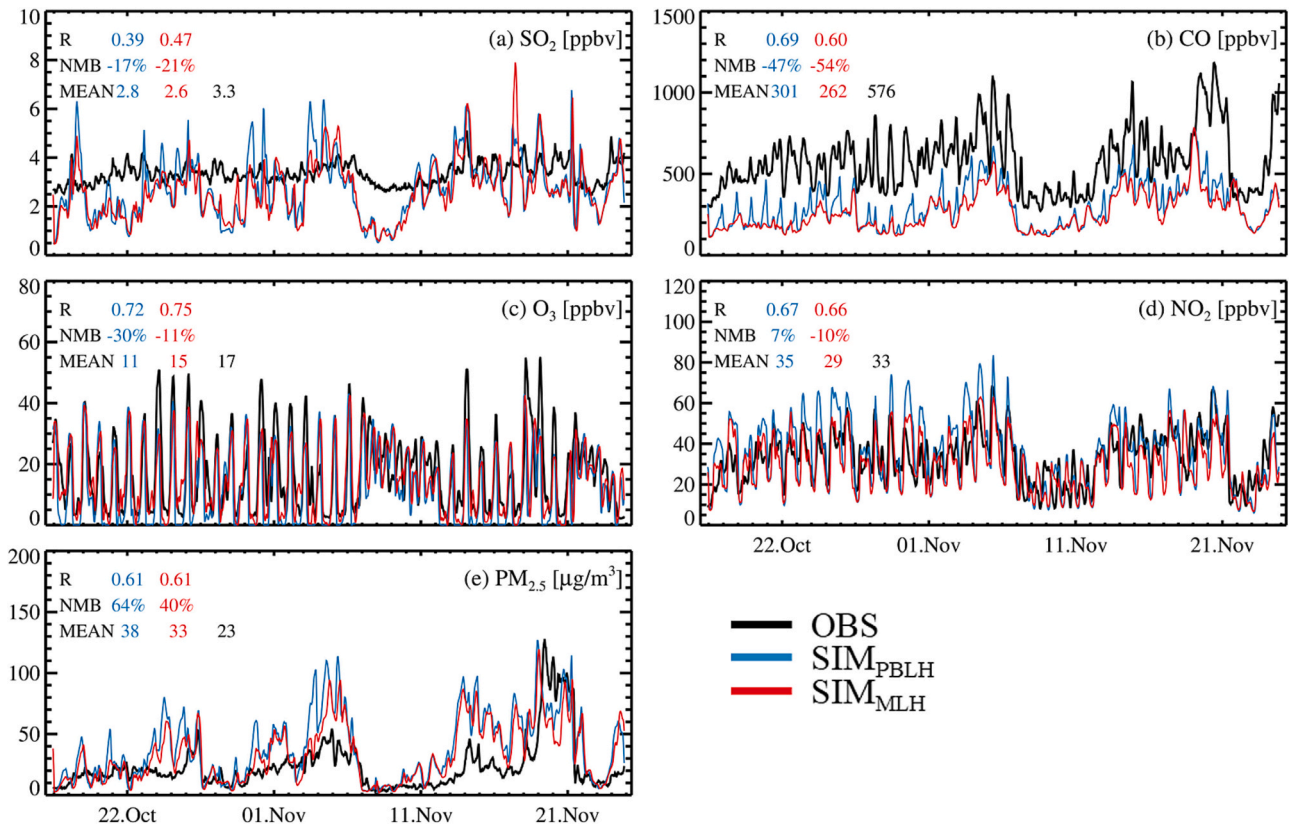
The impacts of using MLH on aerosol species are also evaluated by comparing SIM<sub>PBLH</sub> and SIM<sub>MLH</sub>. Underestimated PBLH causes the accumulation of surface-emitted pollutants near the surface. This aspect is manifested in primary aerosol species, such as BC, which is prone to vertical mixing within the boundary layer. Secondary aerosol concentrations, including NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>, also decrease when using MLH but appear to be buffered by their production processes, such as heterogeneous reactions and thermodynamic partitioning. Interestingly, SO<sub>4</sub><sup>2-</sup> is not sensitive to variation in boundary layer height, showing almost no changes in correlation coefficients and NMBs in SIM<sub>PBLH</sub> and SIM<sub>MLH</sub>. This is likely because SO<sub>4</sub><sup>2-</sup> is mainly produced by the reaction of SO<sub>2</sub> and OH during the daytime and by the aqueous phase oxidation within clouds. Thus, SO<sub>4</sub><sup>2-</sup> is likely well distributed below PBLH/MLH and less sensitive to changes in PBLH and MLH at night.

In the cases of NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>, the extended volume in SIM<sub>MLH</sub> relative to SIM<sub>PBLH</sub> notably decreases their concentrations, resulting in better model performances with decreased NMBs from 126 % to 91 % for





**Fig. 6.** 4-hourly time series of the observed (black) and simulated aerosol species from SIM<sub>PBLH</sub> (blue) and SIM<sub>MLH</sub> (red), including (a)  $\text{SO}_4^{2-}$ , (b)  $\text{NO}_3^-$ , (c)  $\text{NH}_4^+$ , (d) BC, (e) OC, and (f)  $\text{PM}_{2.5}$  at the Olympic Park site during the SIJAQ 2021 campaign.



**Fig. 7.** The hourly time series of the observed (black) and simulated species from SIM<sub>PBLH</sub> (blue) and SIM<sub>MLH</sub> (red), including (a)  $\text{SO}_2$ , (b) CO, (c)  $\text{O}_3$ , (d)  $\text{NO}_2$ , and (e)  $\text{PM}_{2.5}$  averaged over AirKorea sites in Seoul.



$\text{NO}_3^-$  and 113 % to 85 % for  $\text{NH}_4^+$  without noticeable decreases in correlation coefficients. However, overestimations in the model still exist and are likely related to insufficient dry deposition of  $\text{HNO}_3$  (Travis et al., 2022) and the missing uptake of  $\text{HNO}_3$  to coarse particulate matter (Zhai et al., 2023).

We also found that conspicuous overestimation in BC concentrations is alleviated from 322 % to 135 %. However, the underestimation of OC gets worse in  $\text{SIM}_{\text{MLH}}$ , but the secondary organic carbon scheme we used here has significant uncertainties (Brewer et al., 2023; Oak et al., 2022), necessitating further research on organic aerosol chemistry. Finally,  $\text{SIM}_{\text{MLH}}$  shows a better agreement in reproducing observed  $\text{PM}_{2.5}$  concentrations than  $\text{SIM}_{\text{PBLH}}$ , with decreased NMBs from 58 % to 28 %.

We further validated the MLH parameterization by using observations from the AirKorea network. Figs. 7 and S3 compare the observed and simulated species concentrations from  $\text{SIM}_{\text{PBLH}}$  and  $\text{SIM}_{\text{MLH}}$  over Seoul. The observed species show increased concentrations of  $\text{SO}_2$ , CO,  $\text{NO}_2$ , and  $\text{PM}_{2.5}$  in P2 and P4, which is consistent with the observation at the Olympic park (Fig. 5). The peaks in observed CO and  $\text{PM}_{2.5}$  concentrations are up to 1200 ppbv and  $130 \mu\text{g m}^{-3}$ , respectively.

The simulated CO,  $\text{O}_3$ ,  $\text{NO}_2$ , and  $\text{PM}_{2.5}$  concentrations from  $\text{SIM}_{\text{PBLH}}$  and  $\text{SIM}_{\text{MLH}}$  moderately reproduce the observed day-to-day variations with R values over 0.6. We find that the models better capture the observations in a broader region because of the better spatial representation in the models. The impacts of using MLH on the model performance in gaseous and aerosol simulations are consistent with the results at the Olympic park site (Figs. 5 and 6). Underestimation in  $\text{O}_3$  and overestimation in  $\text{PM}_{2.5}$  in the model are improved in  $\text{SIM}_{\text{MLH}}$ . Fig. S3 presents diurnal variations of the observed and simulated species. The tendencies of over/underestimation of species are the same as in Fig. 7.  $\text{SIM}_{\text{MLH}}$  reproduces the diurnal variation better by showing higher R values than  $\text{SIM}_{\text{PBLH}}$ .

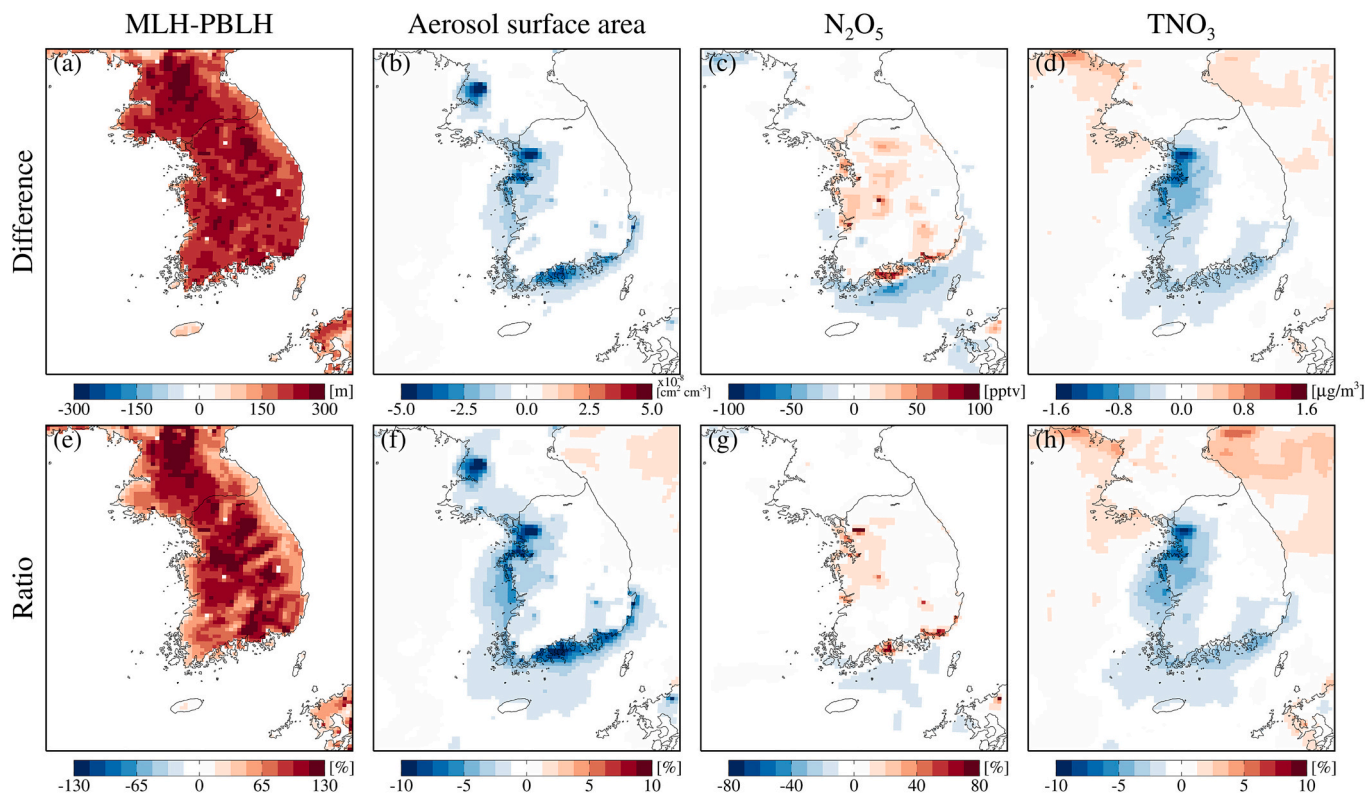
In summary, the new MLH improves the excessive accumulation of surface-emitted species near the surface and subsequent problems. In

particular, the nighttime  $\text{NO}_x\text{-O}_3$  problem is alleviated by suppressing too much titration of  $\text{O}_3$  by overestimated  $\text{NO}_x$ . The NMB of  $\text{PM}_{2.5}$  also decreases due to the improvement in  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ , and BC overestimations. Although aerosol concentrations in  $\text{SIM}_{\text{MLH}}$  are slightly less correlated with the observations compared to  $\text{SIM}_{\text{PBLH}}$ , as shown in Fig. 5, differences are insignificant compared to gaseous species, likely due to the buffered impacts of aerosol production processes. Thus, we conclude that using MLH in the model instead of PBLH improves the model's performance.

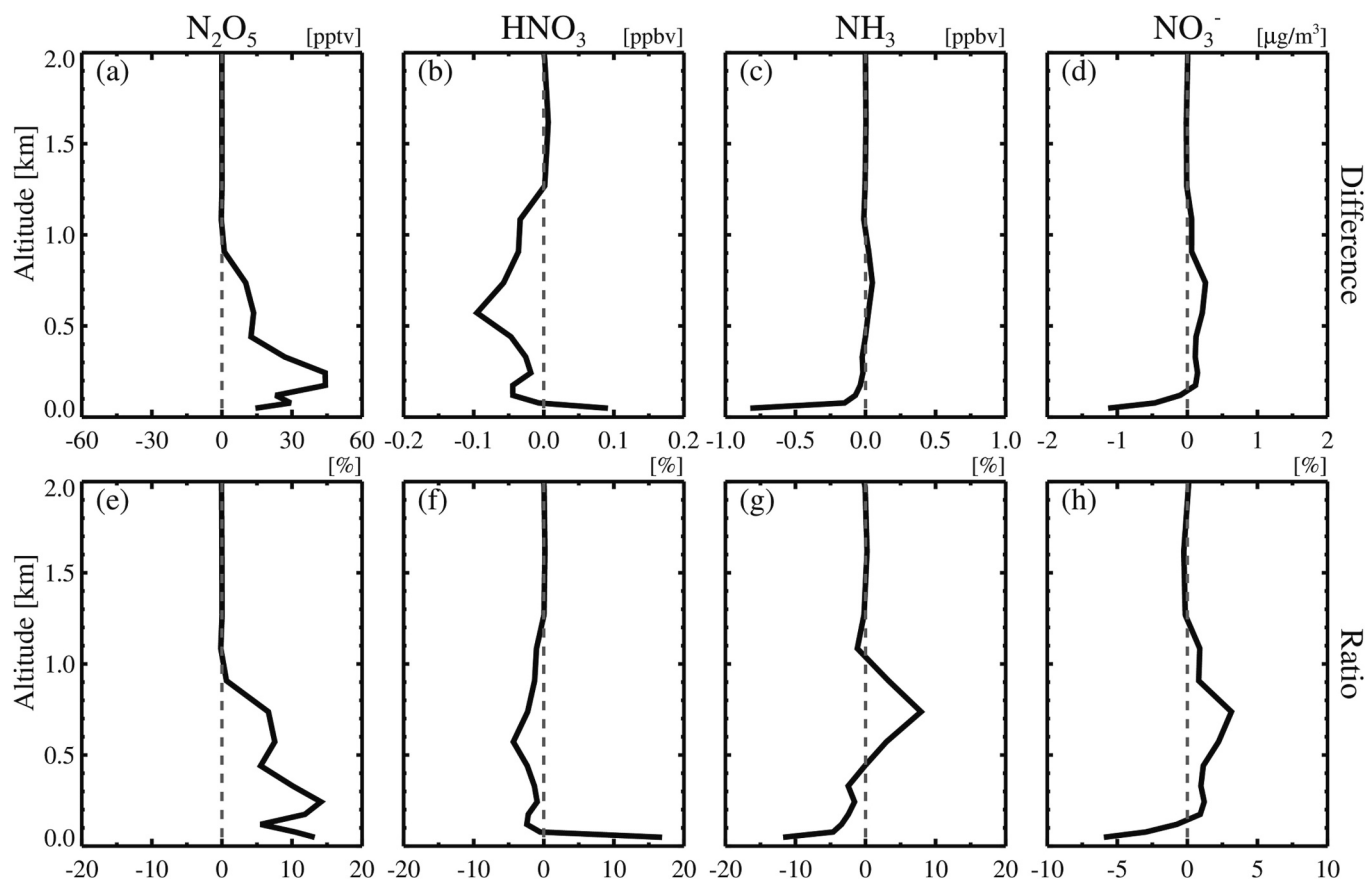
## 5. Impacts of MLH on nighttime chemistry

This section examines how active vertical mixing by PBL correction alters nighttime chemistry simulations. Fig. 8 presents nighttime differences between  $\text{SIM}_{\text{PBLH}}$  and  $\text{SIM}_{\text{MLH}}$ , including aerosol surface areas,  $\text{N}_2\text{O}_5$ , and  $\text{TNO}_3$  ( $\text{HNO}_3 + \text{NO}_3^-$ ) at the surface during the campaign.  $\text{N}_2\text{O}_5$  is one of the nighttime reservoirs of  $\text{NO}_x$  and undergoes heterogeneous reactions on the aerosol surface, producing  $\text{HNO}_3$  (Jacob, 2000). Our PBL correction induces more active vertical mixing of aerosols within the boundary layer and causes a decrease in aerosol surface area in the surface air, consequently slowing down the rate of the  $\text{N}_2\text{O}_5$  heterogeneous reaction. Because the heterogeneous reaction of  $\text{N}_2\text{O}_5$  is the primary process of nighttime  $\text{TNO}_3$  production, the concentrations of  $\text{TNO}_3$  also decrease, along with more dilution of the surface  $\text{TNO}_3$  by the PBL correction. Also, more lofted pollutants are likely transported further by stronger wind above the surface, showing the pronounced impacts of PBL correction in the downwind regions of the significant sources. As a result of PBL correction, aerosol surface area and  $\text{TNO}_3$  concentration are reduced by up to 10 %, but  $\text{N}_2\text{O}_5$  concentrations increase by up to 80 %.

Our PBL correction also alters the thermodynamic equilibrium between  $\text{HNO}_3$  and  $\text{NO}_3^-$ . Fig. 9 shows the impacts of PBL correction on vertical distributions of the species ( $\text{N}_2\text{O}_5$ ,  $\text{HNO}_3$ ,  $\text{NH}_3$ , and  $\text{NO}_3^-$ ) related



**Fig. 8.** Averaged spatial distributions of differences ( $\text{SIM}_{\text{MLH}} - \text{SIM}_{\text{PBLH}}$ ) in (a) boundary layer height, (b) aerosol surface area, (c)  $\text{N}_2\text{O}_5$ , and (d)  $\text{TNO}_3$  ( $\text{HNO}_3 + \text{NO}_3^-$ ) at the surface during the SIJAQ campaign, indicating the impacts of our new parameterization on the model simulations, particularly for nighttime. The ratios of the impacts (e–h) are calculated by  $(\text{SIM}_{\text{MLH}} - \text{SIM}_{\text{PBLH}}) / \text{SIM}_{\text{PBLH}} \times 100$ .



**Fig. 9.** Campaign-mean vertical distributions of the impacts of PBL correction ( $SIM_{MLH} - SIM_{PBLH}$ ) on (a)  $N_2O_5$ , (b)  $HNO_3$ , (c)  $NH_3$ , and (d)  $NO_3^-$  over SMA during the SIJAQ campaign. The ratios of the impacts (e-f) are calculated by  $(SIM_{MLH} - SIM_{PBLH}) / SIM_{PBLH} \times 100$ .

to nighttime  $NO_x$  chemistry. As discussed above, PBL correction slows down the heterogeneous reaction of  $N_2O_5$ , as indicated by  $N_2O_5$  increases and  $TNO_3$  decreases, respectively. Furthermore, PBL correction leads to the dilution of surface-emitted  $NH_3$  and less  $NO_3^-$  production under  $NH_3$ -limited conditions near the surface. This change again affects the reduction in aerosol surface area and  $TNO_3$  production, resulting in a nonlinear feedback between aerosol and  $N_2O_5$  heterogeneous reactions.

## 6. Summary and discussions

We introduce a novel parameterization for the mixed layer height (MLH) based on the Yonsei University (YSU) planetary boundary layer (PBL) scheme. The MLH parameterization is designed to find the top of vertical mixing in the boundary layer using  $R_i$  and  $d\theta_v/dz$ , accounting for thermal and mechanical mixing. The parameterization shows better agreement with the observed PBLH, particularly addressing the underestimation issue of PBLH at night in the model.

The MLH parameterization was further validated by comparing the model performances of  $SIM_{PBLH}$  and  $SIM_{MLH}$  in gaseous and aerosol simulations during the SIJAQ campaign. PBL correction improves the excessive accumulation of surface-emitted pollutants, resulting in improved agreement with the observed concentrations with reduced NMBs of  $NO_x$  (50 % to −27 %) and  $O_3$  (−49 % to −28 %). This approach also improves aerosol simulation, particularly for  $NO_3^-$ ,  $NH_4^+$ , BC, and  $PM_{2.5}$ , compared with the model using the conventional PBL scheme.

The model results reveal that PBL correction significantly impacts nighttime  $NO_x$  chemistry. The enhanced vertical mixing by PBL correction reduces aerosol surface area, decelerating the  $N_2O_5$  heterogeneous reaction while reducing  $TNO_3$  concentrations by up to 10 %. Intriguingly,  $N_2O_5$  concentrations increase by up to 80 %, indicating its sensitivity to PBL correction. Furthermore, PBL correction affects the

thermodynamic equilibrium between  $HNO_3$  and  $NO_3^-$ , which is associated with vertical distributions of  $NH_3$  concentration in the boundary layer. Our findings in this study imply the intricate interplay between aerosols,  $NO_x$  chemistry, and boundary layer mixing, which can play an essential role in determining urban air quality.

## CRediT authorship contribution statement

**Hyeonmin Kim:** Writing – original draft, Visualization, Software, Methodology, Formal analysis, Data curation, Conceptualization. **Rok-jin J. Park:** Writing – review & editing, Supervision, Resources, Methodology, Funding acquisition, Conceptualization. **Song-you Hong:** Writing – review & editing, Methodology, Conceptualization. **Do-Hyeon Park:** Writing – review & editing, Data curation. **Sang-Woo Kim:** Writing – review & editing, Data curation. **Yujin J. Oak:** Writing – review & editing, Conceptualization. **Xu Feng:** Methodology. **Haipeng Lin:** Writing – review & editing, Methodology. **Tzung-May Fu:** Methodology.

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## 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.scitotenv.2024.176838>.

## Data availability

The SIJAQ observational dataset used in this study can be obtained by request through the data archive website (<https://www.sijaq.org/home>, accessed September 2, 2024).

WRF-GC can be downloaded from <https://github.com/jimmielin/wrf-gc-release> (accessed September 2, 2024).

The WRF-GC model with the MLH parameterization in this study can be made available upon request.

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