Simulation Model of Reactive Nitrogen Species in an Urban Atmosphere using a Deep Neural Network: RNDv1.0

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Abstract

Nitrous acid (HONO), one of the reactive nitrogen oxides (NO_y), plays an important role in the formation of ozone (O_3) and fine aerosols ($PM_{2.5}$) in the urban atmosphere. In this study, a new simulation approach to calculate HONO mixing ratios using a deep neural technique based on measured variables wad developed. The 'Reactive Nitrogen species simulation using Deep neural network' (RND) has been implemented in Python. It was trained, validated, and tested with HONO measurement data obtained in Seoul during the warm months from 2016 to 2019.

A k-fold cross validation and test results confirmed the performance of RND v1.0 with an Index Of Agreement (IOA) of 0.79 ~ 0.89 and a Mean Absolute Error (MAE) of 0.21 ~ 0.31 ppbv. The RNDV1.0 adequately represents the main characteristics of HONO and thus, RND v1.0 is proposed as a supplementary model for calculating the HONO mixing ratio in a high-NO_x environment.

1. Introduction

Surface ozone (O_3) pollution has been reported to be worsen over continental areas (Arnell et al., 2019;Monks et al., 2015;Varotsos et al., 2013;IPCC, 2014). In particular, a warmer climate is expected to increase surface O_3 and intensity of surface O_3 peaks in polluted regions, depending on its precursor levels (IPCC 2021). As one of the short-lived climate pollutants (SLCPs), O_3 also interacts with the global temperature via positive feedback (Shindell et al., 2013;Myhre et al., 2017;Stevenson et al., 2013). Therefore, it is imperative to accurately predict the mixing ratios and variations of surface O_3. While operational models such as community multiscale air quality (CMAQ) have been used widely for this purpose, uncertainties still arise from poorly understood chemical mechanisms involving reactive nitrogen oxides (NOy) and volatile organic compounds (VOCs), and lack of their measurements (Mallet and Sportisse, 2006;Canty et al., 2015;Akimoto et al., 2019;Shareef et al., 2019;Cheng et al., 2022).

In the urban atmosphere, NO_y typically includes NO_x ($NO + NO_2$), HONO, HNO_3, organic nitrates (e.g., PAN), NO_3, N_2O_3, and particulate NO_3^-. These species are produced and recycled through photochemical reactions until they are removed through wet or dry deposition (Liebmann et al., 2018;Brown et al., 2017;Wang et al., 2020;Li et al., 2020). NO_y play an important role in critical environmental issues concerning the Earth's atmosphere, spanning from local air pollution to global climate change (Sun et al., 2011;Ge et al., 2019). The oxidation of NO to NO_2, and finally to HNO_3, is the backbone of the chemical mechanism producing ozone (O_3) and $PM_{2.5}$ (particulate matter of size $\leqslant$ 2.5 μm), and it determines the oxidization capacity of the atmosphere. Recently, as O_3 has increased along with a decrease in NO_x emission over many regions including East Asia, interest in the heterogeneous reaction of reactive nitrogen oxides, which is yet to be understood, has been newly raised (Brown et al., 2017;Stadtler et al., 2018). Currently, the lack of measurement of individual NO_y species hindered a comprehensive understanding of the heterogeneous reactions (Anderson et al., 2014;Wang et al., 2017b;Chen et al., 2018b;Akimoto and Tanimoto, 2021;Stadtler et al., 2018).

In particular, there are growing number of evidence for heterogeneous formation of HONO in relation to high $PM_{2.5}$ and O_3 occurrence in urban areas (e.g., (Li et al., 2021b)). As an OH reservoir, HONO will expedite the photochemical reactions involving VOCs and NOx in the early morning, leading to O_3 and fine aerosol formation. Nonetheless, its formation

mechanism has not been elucidated clearly enough to be constrained in conventional photochemical models. In addition to the reaction of NO with OH (Bloss et al., 2021), various pathways of HONO formation have been suggested from laboratory experiments, field measurements and model simulations: direct emissions from vehicles (e.g., (Li et al., 2021a)) and soil (e.g.,(Bao et al., 2022)), photolysis of particulate nitrate (e.g., (Gen et al., 2022)), and heterogeneous conversion of NO_2 on various aerosol surfaces (e.g., (Jia et al., 2020)), ground surface (e.g.,(Meng et al., 2022)), and microlayers of sea surface (e.g., (Gu et al., 2022)). Among these, heterogeneous reaction mechanism at surface is major concern in recently HONO study.

HONO has been measured mostly during intensive campaigns in urban areas using various techniques such as a long path absorption photometer (LOPAP) (Kleffmann et al., 2006;Xue et al., 2019), chemical ionization mass spectrometry (CIMS) (Levy et al., 2014;Roberts et al., 2010), ion chromatography (IC) (VandenBoer et al., 2014;Gil et al., 2020;Ye et al., 2016;Xu et al., 2019), and quantum cascade tunable infrared laser differential absorption spectrometry (QC-TILDAS) (Lee et al., 2011;Gil et al., 2021). Of these methods, QC-TILDAS has served as a reference for intercomparison of measurement data from different techniques due to high time resolution and stability (Pinto et al., 2014). These studies reported the maximum HONO of several ppb levels at nighttime. In comparison, the model captured at most 67~90 % of the observed HONO in megacities such as Beijing (Tie et al., 2013;Liu et al., 2019).

In recent years, Machine Learning (ML) method has been adopted in the atmospheric science for pattern classification (e.g. New Particle Formation event) and forecasting and spatiotemporal modelling of O_3 and $PM_{2.5}$ (Arcomano et al., 2021;Shahriar et al., 2020;Krishnamurthy et al., 2021;Cui and Wang, 2021;Joutsensaari et al., 2018;Chen et al., 2018a;Kang et al., 2021). Among ML methods, the Neural Network (NN) architecture is widely used owing to its powerful ability to process large amounts of data, allowing improvement in the performance of conventional models through being integrated with physical equations (Reichstein et al., 2019;Schultz et al., 2021). As a NN architecture, a multi-layer artificial neural network, referred to as a Deep Neural Network (DNN), employs a statistical method that learn non-linear relations in data and obtain the optimum solution for the target species without prior information on the physicochemical processes. DNN has advantages over other NN architecture

such as Convolution NN (CNN) or Long-Short Term Memory (LSTM) because it works well for discrete spatiotemporal data. In general, the performance of DNN is similar to or better than other ML methods for small number of data as well as large data set (Baek and Jung, 2021;Dang et al., 2021;Sumathi and Pugalendhi, 2021).

When the DNN method is applied to atmospheric chemical constituents, it requires large amount of data for training and thus, the size of measurement data becomes a limiting factor for trace species such as HONO, which are not routinely measured such as O_3 or $PM_{2.5}$. In this regard, the daily average HONO mixing ratio was attempted to be estimated using ensemble ML models with satellite measurements (Cui and Wang, 2021). In comparison, the hourly HONO mixing ratio was calculated using a simple NN architecture with measured variables, which were thought to be closely linked with HONO formation (Gil et al., 2021). The accuracy of the hourly HONO estimated from input variables such as aerosol surface areas and mixed layer height was better than the daily HONO estimate.

In this study, we aimed to construct a user-friendly 'Reactive Nitrogen species simulation using DNN' (RND) model and estimate HONO mixing ratio using routinely measured atmospheric variables in a highly polluted urban area. Finally, the model results will be incorporated into operational photochemical models for air quality forecasting and improve their performance. Since this is the first attempt to calculate HONO mixing ratios using a first version of RND model (RNDv1.0), we describe the entire modeling process and evaluate the model results by comparing them with the measurements.

2. Model description

The development of RNDv1.0 model follows the systematic steps similar to a general machine learning model construction workflow, including collecting data, preprocessing data, building the DNN, training and validating the model, and testing the performance of the model (Figure 1). The RNDv1.0 was written in Python and necessary libraries to build and operate RNDv1.0 are listed in Table 1. The dataset used to train-test-validation can be downloaded from Gil et al., 2021.

2.1. Collection of measurement data for model construction

As the first step constructing the RNDv1.0, measurement data were obtained including HONO, reactive gases, and meteorological parameters. It is noteworthy that the HONO measurement data is for model construction and is not required to run the RND model. The HONO mixing ratio was measured using a Quantum Cascade – Tunable Infrared Laser Differential Absorption Spectrometer (QC-TILDAS) system in Seoul during May–June 2016, June 2018, and April-June 2019 (Lee et al., 2011;Gil et al., 2021). When testing and evaluating atmospheric HONO measurement methods, QC-TILDAS has been chosen as the reference method for comparing ambient HONO mixing ratios measured using several different techniques owing to its advantages of low detection limits (~ 0.1 ppbv) and high temporal resolution (Pinto et al., 2014). More details on measurements can be found elsewhere (Gil et al., 2021). HONO was measured at Olympic Park (37.52°N, 127.12°E) during the Korea-United States Air Quality (KORUS-AQ) study in 2016 (Kim et al., 2020;Gil et al., 2021), at the campus of Korea University in 2018 (37.59°N, 127.03°E), and at the site near the campus in 2019 (37.59°N, 127.08°E) (NIER, 2020) (Figure S1). Of the three sites, the Korea University campus and Olympic Park have served as measurement sites representing the air quality of Seoul. In fact, it has been known that O_3 and $PM_{2.5}$ levels are strongly influenced by the synoptic circulation throughout the Korean peninsula (Peterson et al., 2019;Jordan et al., 2020). In addition, trace gases including O_3, NO_2, CO, and SO_2 and meteorological parameters including temperature (T), relative humidity (RH), wind speed (WS) and direction (WD) were measured. Note that HONO was not significantly correlated with any of these variables (Figure S2). The measurement statistics are presented in Table 2 and Table S1. Briefly summarizing, the 10^{th} and 90^{th} percentile mixing ratios of HONO, NO_2, and O_3 are 0.3 ppbv and 1.9 ppbv, 10.7 ppbv and 48.2 ppbv, and 12.0 ppbv and 80.9 ppbv, respectively for the entire experiment periods.

2.2. Data preprocessing

In the next step, the observation data set was prepared for RNDv1.0 model construction. As input variables, hourly measurements of chemical and meteorological parameters are used, including the mixing ratios of O_3, NO_2, CO, and SO_2, along with temperature (T), relative humidity (RH), wind speed (WS), wind direction (WD), and solar zenith angle (SZA) to estimate the target species, HONO, as the output. Wind direction in degrees were converted to a cosine value for continuity. As a last step in data processing, missing values were filtered out from the input dataset. Finally, 50.7 % of all available measurement data (1636) were used to construct the RNDv1.0 in this study.

Since the measurements of these nine variables vary over a wide range in different units, they were normalized to avoid bias during the calculations. Among the widely used normalization methods, '*min-max scaling*' method was adopted and input variables were normalized against the minimum and maximum values in this study (Eq. 1):

$$x_{sca} = \frac{x_{raw} - F_2(X)}{F_1(X)}, \quad (1)$$

where x_{raw} is raw data of input variable (X), x_{sca} is scaled data of X, F_1 and F_2 are scale factors of X, and are given for each input variable used in Table 2.

2.3. Neural network architecture and hyperparameters

At this stage, the network is built to calculate HONO using those input variables. The RNDv1.0 is composed of five hidden layers (Figure 2), which employed an exponential linear unit (ELU) as an activation function (Eq. 2).

$$\text{ELU}: \phi(x) = \begin{cases} e^x - 1 & (x < 0) \\ x & (x \geq 0) \end{cases}. \quad (2)$$

In a DNN, an activation function creates a nonlinear relationship between an input variable and an output variable. When constructing a DNN model, an ELU has the advantage of a fast-training process and better performance in handling negative values than other activation functions (Wang et al., 2017a;Ding et al., 2018). In addition, the mean squared error and Adam optimizer were applied as loss function and optimize function, respectively. The learning rate, epoch, and batch were set to 0.01, 100, and 32, respectively.

2.4. Train, validation, and test

The RNDv1.0 model was trained, validated, and tested with HONO measurements obtained during May ~ June in 2016 and 2019, in June 2018, and in April 2019, respectively (Figure 3). The number of data used for train, validation, and test were 1122, 381, and 133, respectively.

With the hyperparameters specified in previous section, the performance of the model was firstly validated using the k-fold cross-validation method, which is especially useful when the size of dataset is small (Bengio and Grandvalet, 2003). In the k-fold cross-validation method (Figure 3), the entire data is randomly divided into k subsets, of which k-1 sets were used for training and the rest one was used for validation. k was set to 5 in this study. The accuracy was determined by Index Of Agreement (IOA) and Mean Absolute Error (MAE) expressed by the following equation (Eq. 3, Eq. 4):

$$\mathrm{IOA} = 1 - \frac{\sum_{i=1}^{n}(O_i - P_i)^2}{\sum_{i=1}^{n}(|P_i - \bar{O}| + |O_i - \bar{O}|)^2}, \quad (3)$$

$$\mathrm{MAE} = \frac{\sum_{i=1}^{n}|O_i - P_i|}{n}, \quad (4)$$

where O_i, P_i, $\bar{O}$, and n are the observed value, predicted value, average of the observed values, and number of nodes, respectively. The overall accuracy of

As IOA and MAE vary according to the number of nodes, they were calculated for the measured ($HONO_{obs}$) and calculated ($HONO_{mod}$) mixing ratios by varying the number of nodes from 0 to 100 in each hidden layer. The best performance was found with 41 nodes, with which

the averaged IOA and MAE were 0.89 ± 0.01 (mean ± standard deviation) and 0.31 ± 0.02 ppbv, respectively (Figure 4). The high level of IOA and low MAE demonstrates that the performance of RNDv1.0 model is adequate, and it is capable of simulating the ambient HONO mixing ratio using the routinely measured chemical and meteorological parameters. In particular, MAE was commensurate with the detection limit of HONO measurement.

After the network validation, HONO mixing ratio was calculated for May ~ June in 2016 and 2019, and the model results were compared with the measured values (Figure 5). The average mixing ratios of measured and calculated HONO was 0.94 ppbv and 0.89 ppbv in 2016, and 1.02 ppbv and 0.96 ppbv in 2019, respectively. The MAE and IOA of the measurement and calculation were 0.27 ppbv and 0.90 in 2016, and 0.29 ppbv and 0.91 in 2019, respectively, demonstrating the ability of the RNDv1.0 to simulate ambient HONO levels. In both cases, however, the model slightly underestimated the highest and lowest HONO mixing ratios, which is mainly due to the limited number of data used for training, but also related to the intrinsic nature of DNN. The model calculation well captured the diurnal variation of ambient HONO with a slight underestimation (Figure 6). In addition, the correlation between $HONO_{mod}$ and $HONO_{obs}$ was better in 2019 (MAE = 0.06 ppbv) than in 2016 (MAE = 0.08 ppbv). Since the MAE of the two cases was far below the detection limit of HONO measurements (~ 0.1 ppbv), the RNDv1.0 is considered adequate to simulate HONO in urban areas.

Finally, the RND model was validated and tested against the measurement data obtained in June 2018 and April 2019. The calculated HONO mixing ratios are compared with those measured in Figure 7, and their MAE and IOA are listed in Table 3. The two sets of model performance test showed that the model reasonably traced what was observed. As the validation result of RND, the MAE and IOA of the calculated and measured in June 2018 are comparable to those of 2016~2019 result. However, the MAE and IOA of the April 2019 measurements were relatively poor compared to the validation results. Especially, the MAE of the April 2019 is about twice as high as those of validation.

In these two test periods, HONO levels were lower than those observed on validation days (Figure 5), and the model tended to overestimate high HONO concentrations. The large discrepancy in April 2019 is probably due to seasonality: the difference in meteorological and chemical regime of the atmosphere. For example, the monthly average temperature, relative humidity, and NO_2 mixing ratio of Seoul in 2019 were 12.1 ℃, 50.9 %, and 29 ppbv in April

2019 and 22.5 ℃, 60.6 %, and 21 ppbv in June 2019 (https://cleanair.seoul.go.kr; https://weather.go.kr). Note that the RNDv1.0 model was trained with the 9 variables measured in early summer (Table 2). Therefore, the more measurement data spanning a full year for training, the more accurate the model estimates will be.

2.5. Influence of input variables to HONO concentration

Additionally, a simple bootstrapping test was conducted by setting each variable to zero with keeping other variables (Kleinert et al., 2021). Then, the importance of each input variable to HONO concentration was evaluated using MAE and root mean square error (RMSE). Of nine input variables, NO_2 was found to have the most significant influence on HONO concentration, followed by RH, temperature, and solar zenith angle (Table S2). The result of bootstrap test is in good agreement with those from our previous study (Gil et al., 2021), where more detailed information such as aerosol surface area and mixing layer height were incorporated into the model and highlighted the role of precursor gases and heterogeneous conversion in HONO formation. Therefore, these results demonstrate that the RND model constructed using routinely observed variables, reasonably traced the level of HONO in urban atmosphere.

3. Operation and application of RNDv1.0

The RNDv1.0 package is provided as an operational model, .h5 files that can be opened in Python. To run the RNDv1.0, the measurement data for nine input variables are required and need to be properly prepared as described in Section 2.2. A sample of preprocessed input dataset is provided as a .csv file (Dataset_for_model.csv). Once the input data is ready, open the RND model with input data files using the code provided in the example (Figure S3). Then, RND v1.0 calculates and presents the HONO results as scaled values (x_{sca}), which will be finally converted to HONO mixing ratio (ppbv) by the two scale factors in Table 2 (Eq. 5):

$$\mathrm{HONO\ (ppbv)} = \mathrm{HONO_{sca}} \times \mathrm{F_1(HONO)} + \mathrm{F_2(HONO)}. \quad (5)$$

The result of the RNDv1.0, HONO, can be applied to an urban photochemical cycle simulation. It is already known that the photolysis of HONO is a major source of OH radicals in the early morning when the OH level is low, and this OH affects daytime O_3 formation through photochemical reactions with VOCs and NO_x, which are primarily emitted during morning rush hour in urban areas. Therefore, the OH produced from HONO expedites photochemical reactions, promoting O_3 formation. However, the HONO formation mechanism is still poorly understood, and concentrations are not correctly simulated in conventional photochemical models; therefore, the absence of HONO causes great uncertainty in O_3 prediction (Figure 8).

The 0-Dimension Atmospheric Modelling (F0AM) utilizing the MCM v3.3.1 chemical reaction mechanisms (Wolfe et al., 2016), can be used to simulate the diurnal variation of O_3 with the measurements of several reactive gases (NO, NO_2, CO, HCHO, VOCs, and HONO). Detailed information about F0AM can be found in (https://sites.google.com/site/wolfegm/models) and in previous works published elsewhere (Wolfe et al., 2016; Gil et al., 2020). When the F0AM model is run without HONO, it is not able to reproduce the concentration and diel cycle of the observed O_3 (Figure 8). In comparison, the model simulates the O_3 well within 2 ppbv when adding HONO, which is the product of RND v1.0. This is mainly due to the missing OH produced by HONO photolysis in the early morning. Its production rate is estimated to be 0.57 pptv s^{-1}, contributing approximately 2.28 pptv to OH budget during 06:00 ~ 11:00 (LST) (Gil et al., 2021). Given that OH is mainly produced from the photolysis of O_3 under high sun, the early morning source of OH will expedite the photochemical cycle involving NO_x and VOCs, promoting O_3 and secondary aerosol formation. Since the presence of HONO in the photochemical model allows for accurate estimation of OH radicals, the incorporation of RND into conventional models will improve their overall performance.

4. Summary and implications

In this study, we developed the RND model to calculate the mixing ratio of NO_y in an urban atmosphere using a DNN along with measurement data. The target species of RNDv1.0 is HONO, and its mixing ratio is calculated using trace gases including O_3, NO_2, CO, and SO_2, and meteorological variables including T, RH, WS, and WD, along with the SZA. These variables are routinely measured through monitoring networks. The RNDv1.0 was trained and validated using the HONO measurements obtained in Seoul by adopting a k-fold cross validation method and tested with other HONO datasets measured using the same instrument. The validation and test results demonstrate that RND adequately captures the characteristic variation of HONO and confirms the efficacy of RND v1.0.

RNDv1.0 was constructed using measurements made in a high NO_x environment during early summer (May–June). It is noteworthy that in this period, the HONO mixing ratio was raised above 3 ppbv with the highest O_3 levels under stagnant conditions. If RND is applied to areas under significant influence of outflows, the model possibly overestimates or underestimate the level of HONO without detailed information such as nanoparticles. In the previous study, the formation of HONO was shown to be intimately related with surface areas of submicron particles (Gil et al., 2021). Nevertheless, the HONO concentration produced from RNDv1.0 with routine measurements provides the benefit of relatively inexpensive test for measurement quality control, location selection, and supports the data used for traditional chemistry model based on the current knowledge of the urban photochemical cycle. Therefore, it is reasonable to argue that RND can serve as a supplementary tool for conventional photochemical models.

5. Acknowledgements

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6. Code availability

The RND model codes (.h5 files) with preprocessed sample data can be downloaded from (Gil, 2021).

7. Author contributions

JG and ML designed the manuscript and developed the model code. JK, GL, and JA provided the measurement data and validated the model. All the authors contributed to the manuscript.

8. Competing interests

The authors declare that they have no conflict of interest.

Figures and Tables

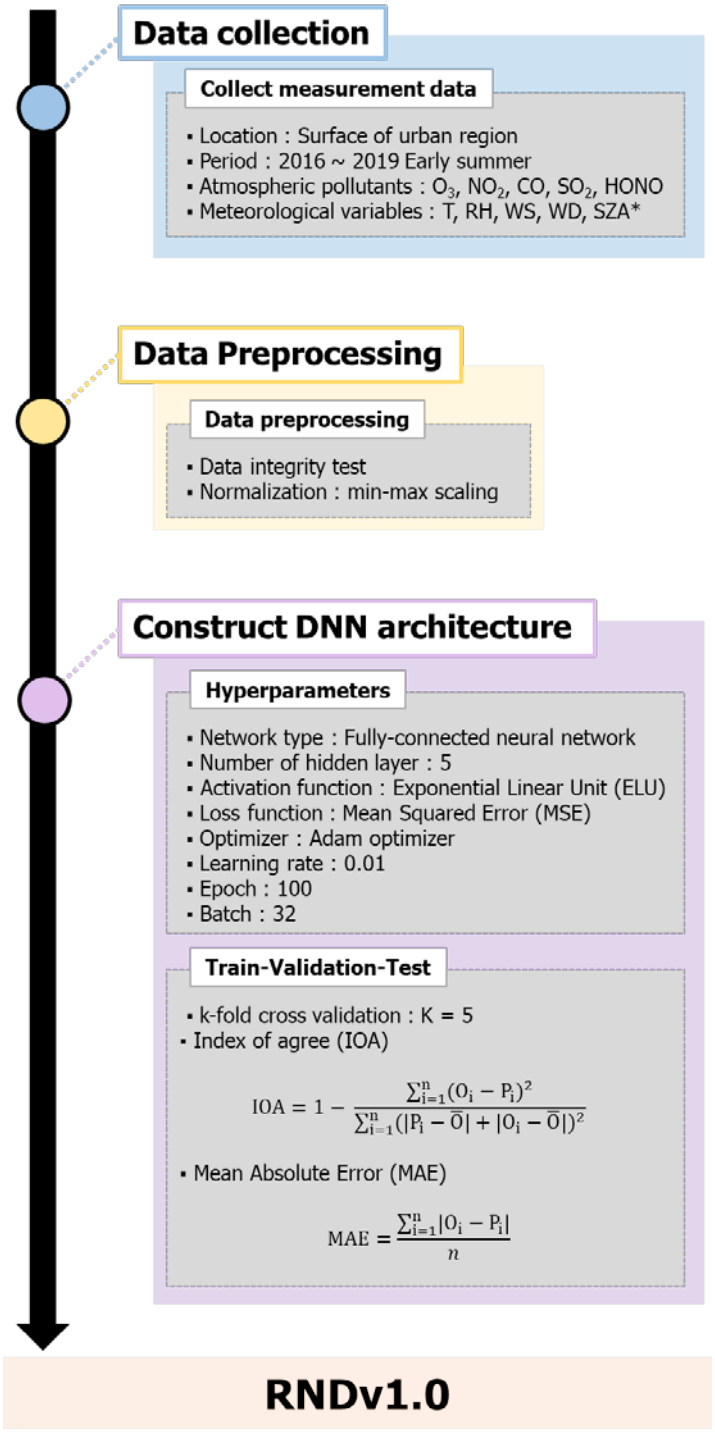


Figure 1. The main processes for configuring the RNDv1.0 (*: calculated values)

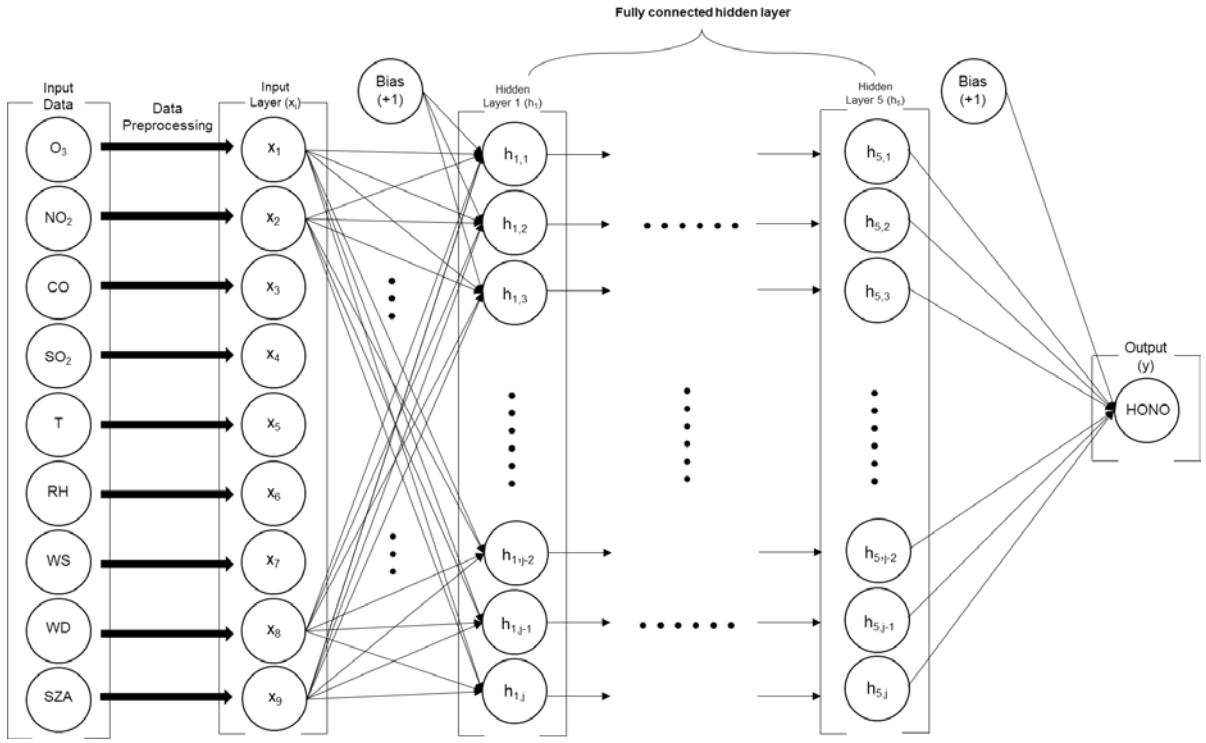


Figure 2. The structure of deep neural network built for RND v1.0.

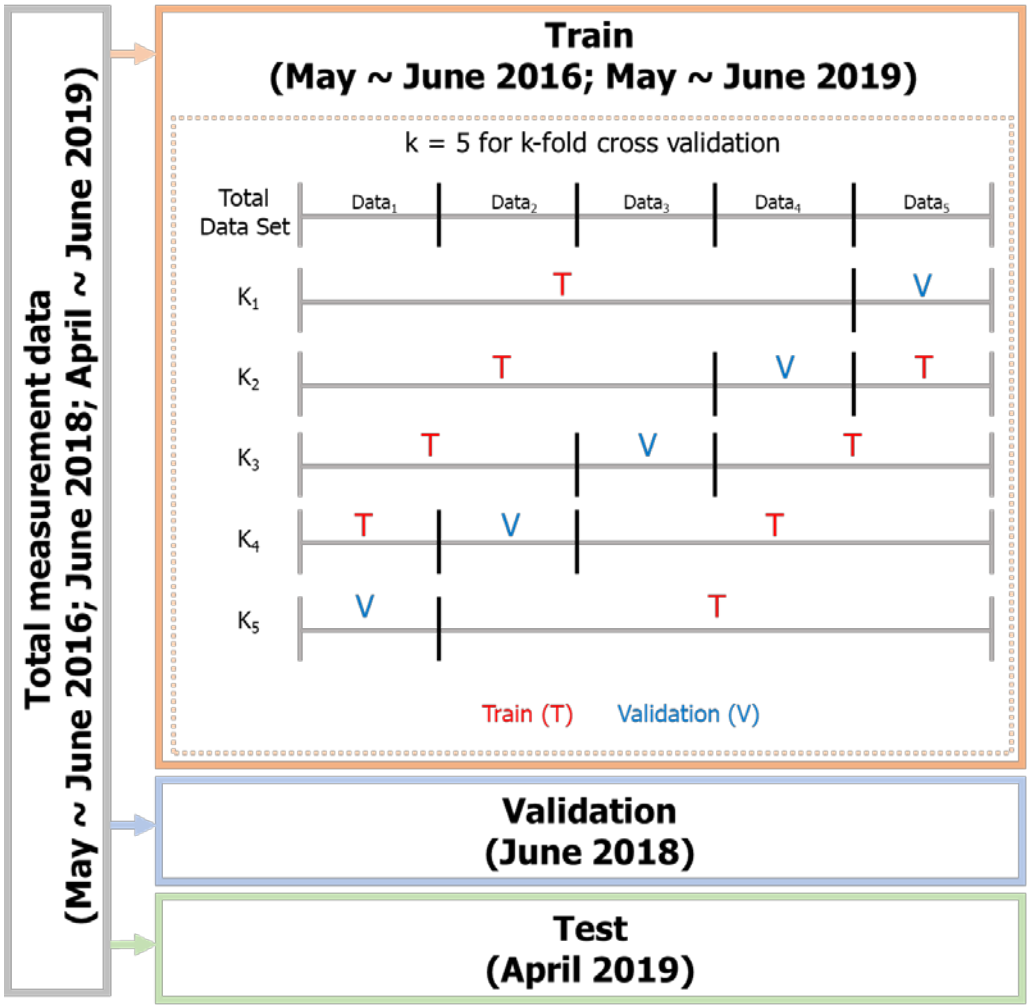


Figure 3. Design of training, validation, and test to build RNDv1.0 using measurement data. **The k-fold cross validation were performed using randomly divided five subsets of training data set.**

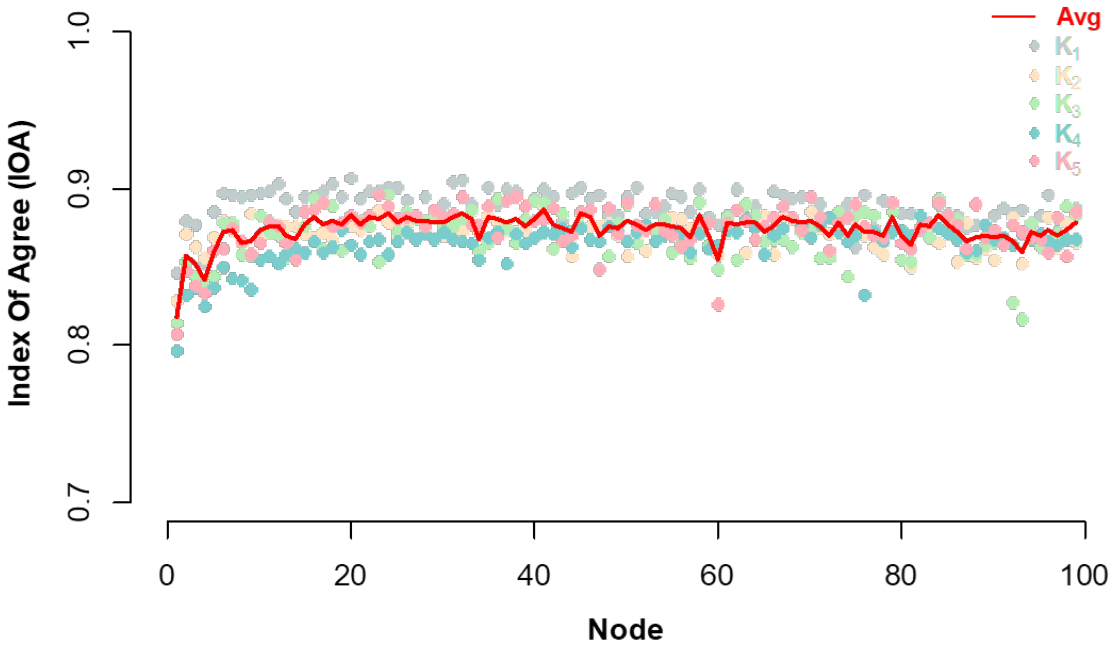
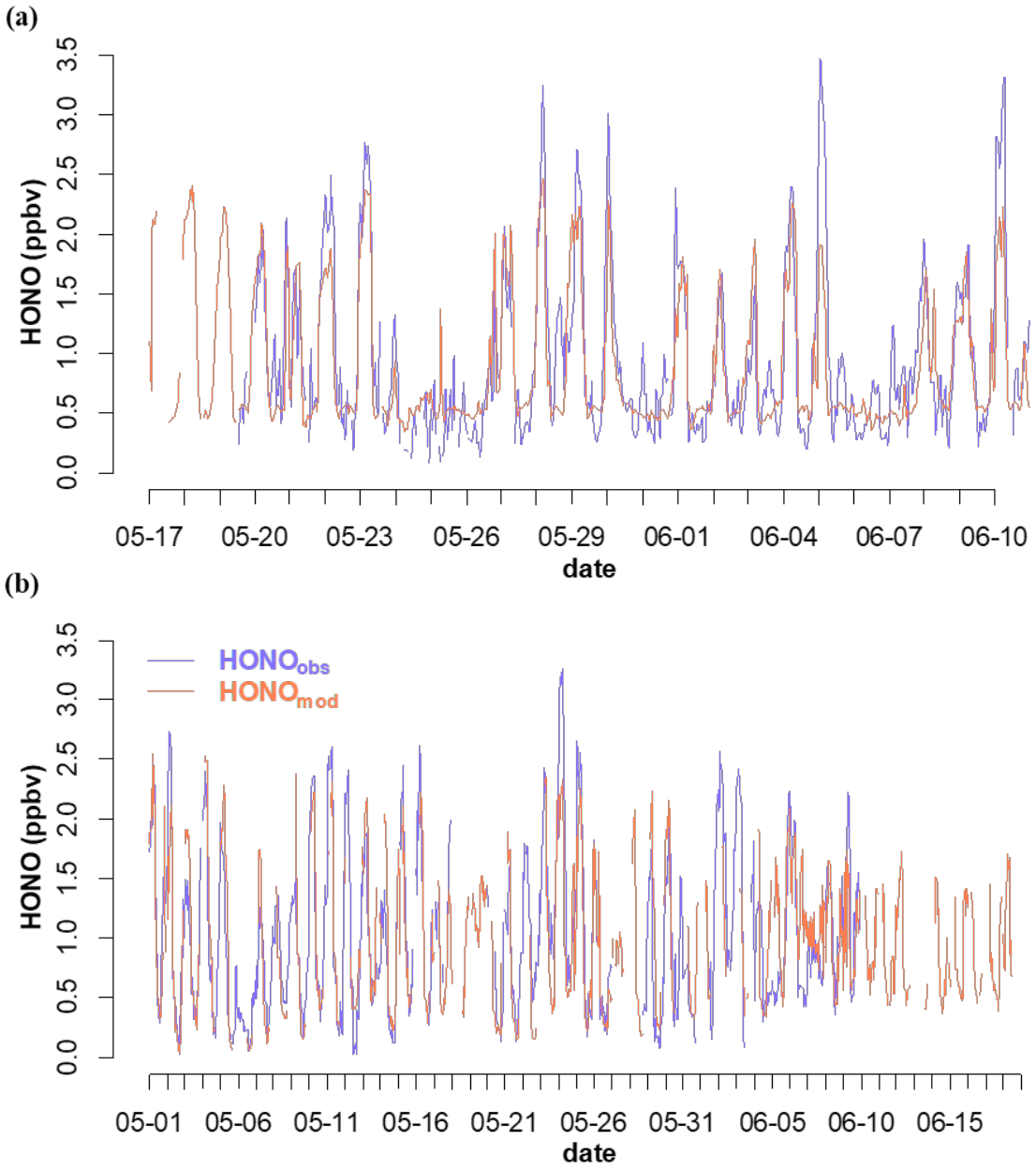


Figure 4. Index Of Agreement (IOA) for k-fold cross validation. Solid circle and red line represent IOA for each validation (k=5) and the average of 5 validation sets at each node number.



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355 **Figure 5.** Comparison between the measured ($HONO_{obs}$) and calculated ($HONO_{mod}$) HONO
356 mixing ratios in Seoul during May~June in (a) 2016 and (b) 2019. The blue and red lines
357 indicate the measured and calculated HONO mixing ratio, respectively.

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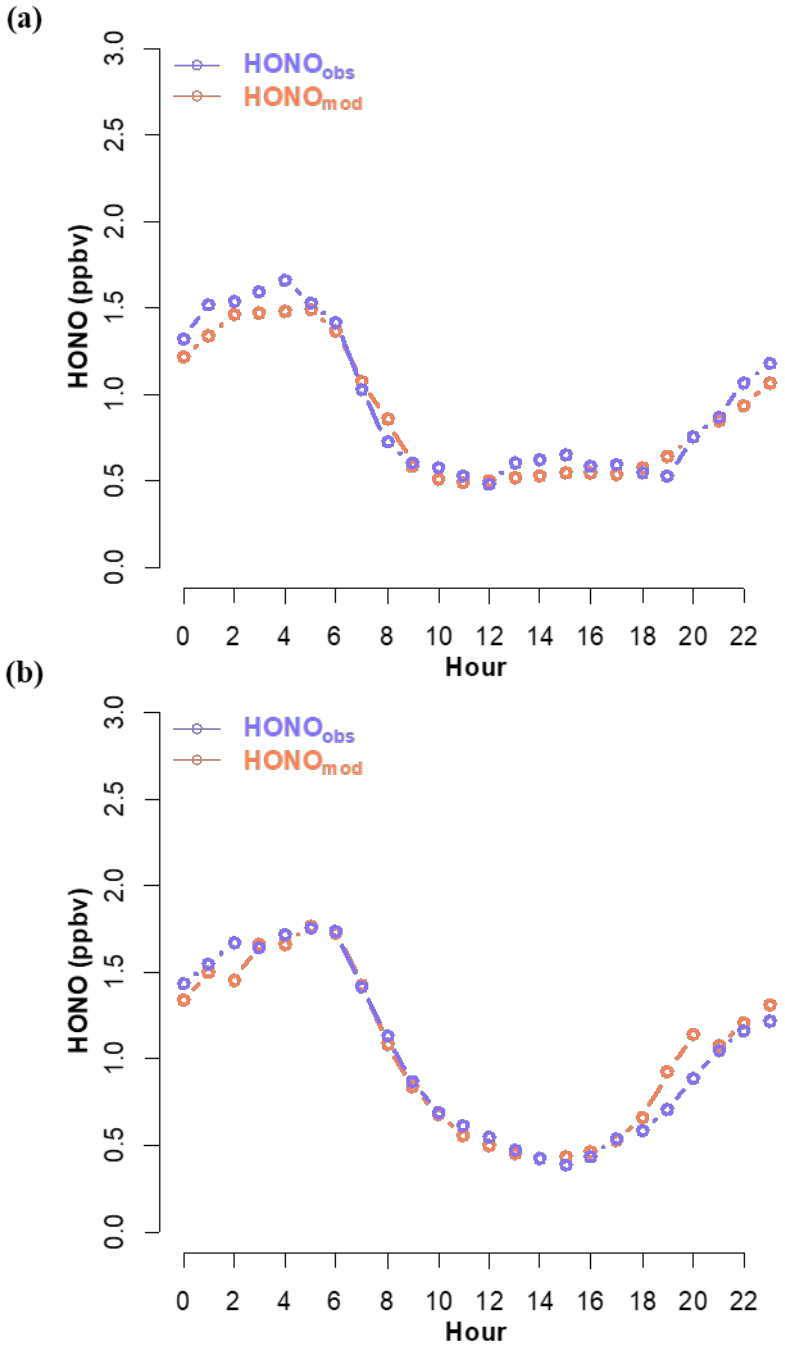

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360 **Figure 6**. Average diurnal variations of the measured ($HONO_{obs}$) and the calculated ($HONO_{mod}$)
361 HONO mixing ratios in Seoul during May ~ June in (a) 2016 and (b) 2019.

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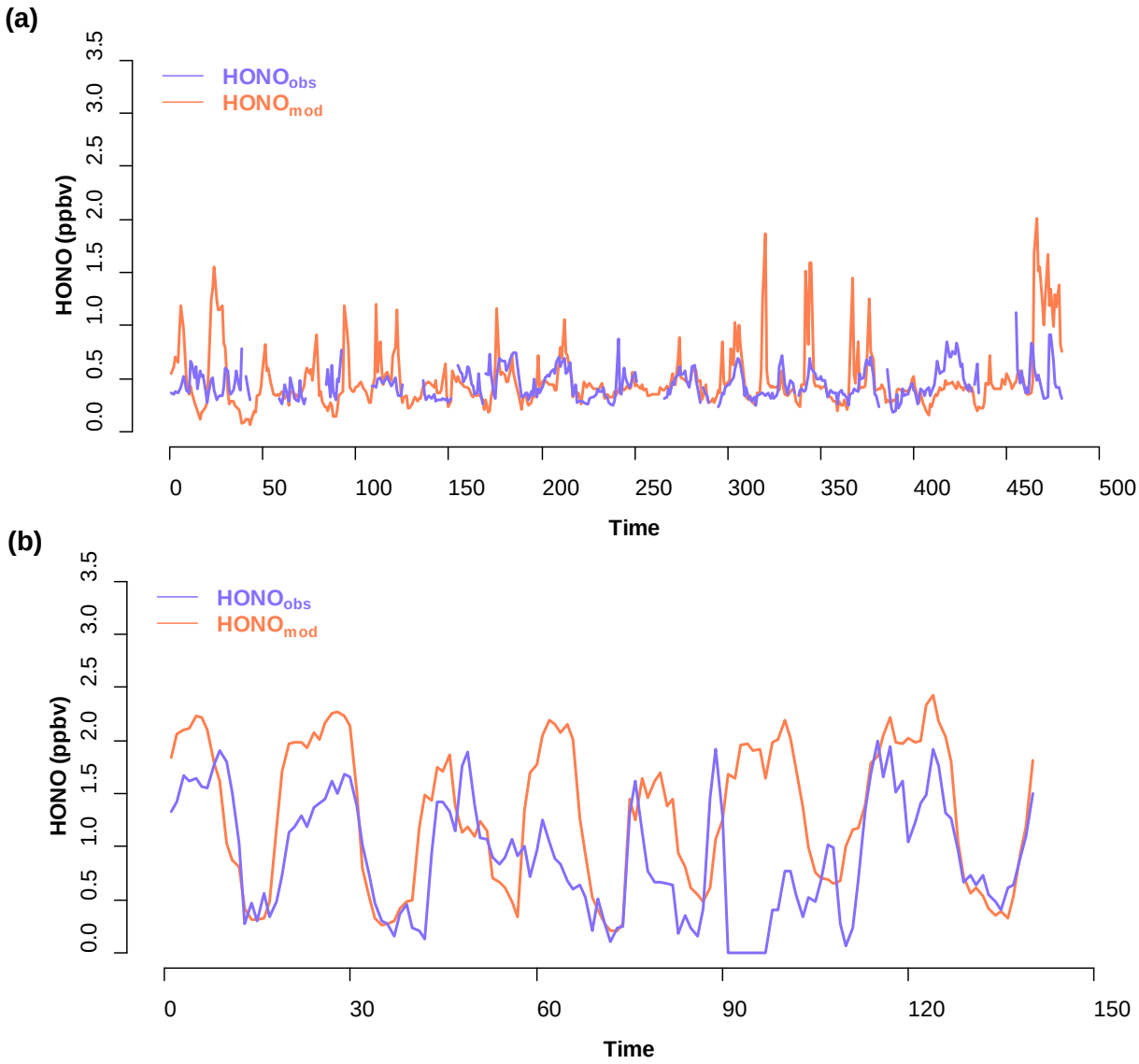
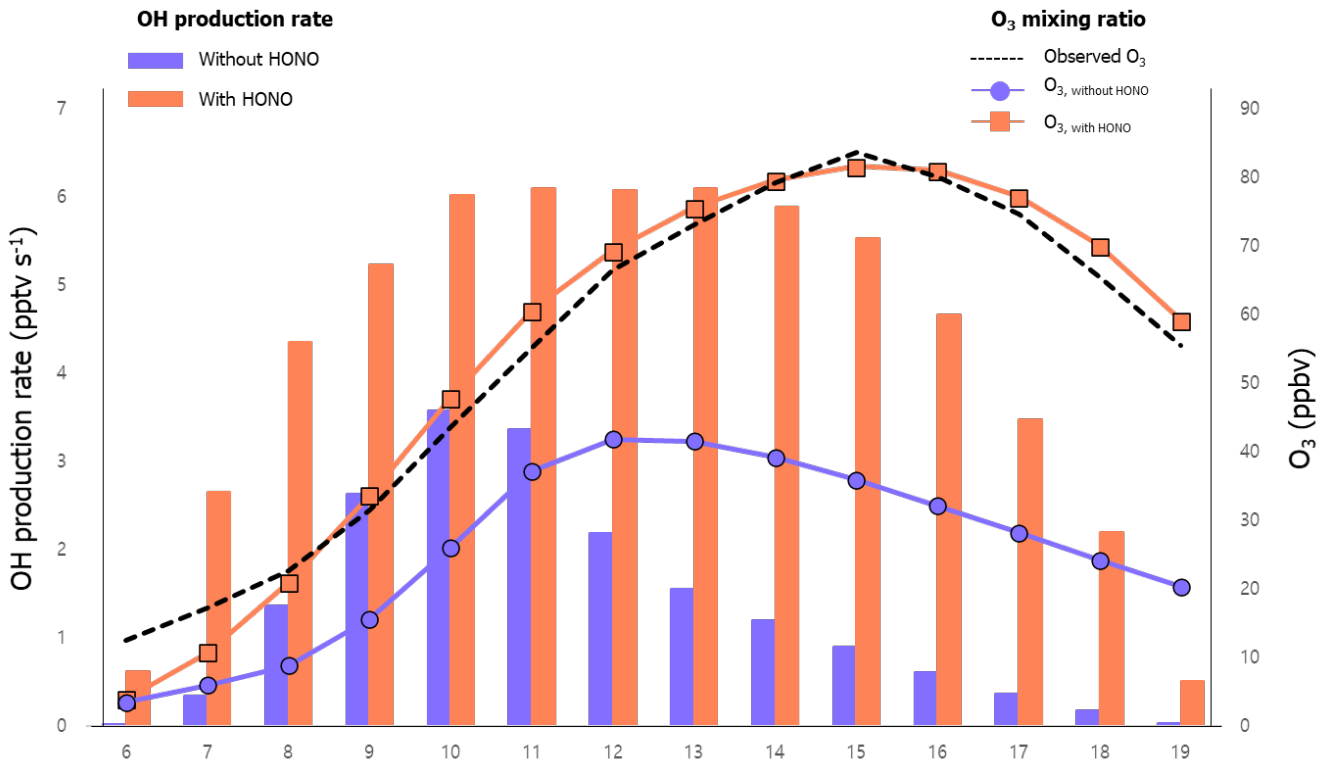


Figure 7. Comparison between the measured ($HONO_{obs}$) and calculated ($HONO_{mod}$) HONO mixing ratios in Seoul during (a) June 2018 and (b) April 2019. The blue and red lines indicate the measured and calculated HONO mixing ratio, respectively. The x axis indicates the hour from the beginning of the experiment, which is (a) 00:00 on 1st June 2018 and (b) 00:00 on 12th April 2019.



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372 **Figure 8**. For June 2016, diurnal variations of O_3 (line) and OH production rate (bar) calculated
373 from the F0AM photochemical model with (orange) and without (blue) HONO estimated from
374 the RNDv1.0 model. The measured O_3 is compared with the calculated.

375

Table 1. Resources for constructing RND model.

	Version	Remark
Python	v3.8.3	
CUDA	v10.1	*If using GPU
CuDNN	v7.6.5	*If using GPU
Tensorflow	v2.3.0	*Python library*
Keras	v2.4.3	*Python library*
Pandas	v1.0.5	*Python library*
Numpy	v1.18.5	*Python library*

*GPU denotes graphic processing unit

Table 2. Input variables of the RNDv1.0 model and their ranges (10^{th} and 90^{th} percentile) observed in Seoul during May ~ June in 2016 and 2019.

	10^{th}~90^{th} percentile (unit)	Coverage (%)	Scale Factor1 (F_1)*	Scale Factor 2 (F_2)**
Input Variables				
O_3	12.1 ~ 90.4 (ppbv)	95.5	204.738	0.842
NO_2	11.0 ~ 48.6 (ppbv)	80.6	79.925	2.375
CO	252 ~ 743 (ppbv)	95.1	975.248	137.253
SO_2	1.9 ~ 6.4 (ppbv)	95.6	12.479	0.958
Solar Zenith Angle	22.7 ~ 118.4 (º)	100.0	112.317	14.195
Temperature	15.9 ~ 26.7 (°C)	99.4	24.240	8.610
Relative Humidity	29.2 ~ 79.1 (%)	99.4	88.545	10.555
Wind Speed	0.2 ~ 3.7 (m/s)	99.4	7.581	0.005
Wind Direction	45.4 ~ 287.5 (º)	99.4	359.565	0.235
Output Variables				
HONO	0.3 ~ 2.0 (ppbv)	81.1%	3.447	0.013

* Maximum – Minimum

** Minimum value

Table 3. The result of validation and test of RNDv1.0 model using measurement data.

Measurement data	Validation		Test	
	MAE (ppbv)	IOA	MAE (ppbv)	IOA
May 2016*	0.26	0.93		
June 2016*	0.29	0.86		
June 2018	0.21	0.79		
April 2019			0.56	0.65
May 2019*	0.26	0.93		
June 2019*	0.36	0.76		

*Re-using the data already used for training

Reference

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