

Simulation model of Reactive Nitrogen species in an urban atmosphere using a Deep neural network: RNDv1.0

Junsu Gil¹, Meehye Lee¹, Jeonghwan Kim², Gangwoong Lee², Joonyoung Ahn³, Cheol-Hee Kim⁴

¹ Department of Earth and Environmental Sciences, Korea University, Seoul, South Korea

² Department of Environmental Science, Hankuk University of Foreign Studies, Yongin, South Korea

³ Air Quality Forecasting Center, Climate and Air Quality Research Department, National Institute of Environmental Research (NIER), Incheon, South Korea

⁴ Department of Atmospheric Sciences, Pusan National University, Busan, South Korea

Correspondence to: Meehye Lee (meehye@korea.ac.kr)

Abstract. Nitrous acid (HONO), one of a reactive nitrogen oxide, plays an important role in the formation of ozone and fine aerosols in the urban atmosphere. In this study, a new simulation approach is presented to calculate the HONO mixing ratios using a deep neural technique based on measured variables. The “Reactive Nitrogen species simulation using deep neural network (RND)” is implemented in Python. The first version of RND (RNDv1.0) is trained, validated, and tested with HONO measurement data obtained in Seoul from 2016 to 2021. RNDv1.0 is constructed using k-fold cross validation and evaluated with index of agreement, correlation coefficient, root mean squared error, and mean absolute error. The results show that RNDv1.0 adequately represents the main characteristics of the measured HONO, and it is thus proposed as a supplementary model for calculating the HONO mixing ratio in a polluted urban environment.

1. Introduction

Surface ozone (O₃) pollution has worsened over continental areas (Arnell et al., 2019; Monks et al., 2015; Varotsos et al., 2013; IPCC, 2014). Particularly, a warmer climate is expected to increase the surface O₃ concentrations and peak levels in polluted regions depending on its precursor levels (IPCC, 2021). As a short-lived climate pollutant (SLCP), O₃ interacts with the global temperature via positive feedback (Shindell et al., 2013; Myhre et al., 2017; Stevenson et al., 2013). Therefore, accurate predictions of the mixing ratios and variations

34 of the surface O_3 are essential. While operational models such as the community multiscale air
35 quality (CMAQ) have been widely used for this purpose, uncertainties still arise from poorly
36 understood chemical mechanisms involving reactive nitrogen oxides (NO_y) and volatile organic
37 compounds (VOCs), and the lack of their measurements (Mallet and Sportisse, 2006; Canty et
38 al., 2015; Akimoto et al., 2019; Shareef et al., 2019; Cheng et al., 2022).

39 In the urban atmosphere, NO_y typically includes NO_x ($NO + NO_2$), HONO, HNO_3 ,
40 organic nitrates (e.g., PAN), NO_3 , N_2O_3 , and particulate NO_3^- . These species are produced and
41 recycled through photochemical reactions until they are removed through wet or dry deposition
42 (Liebmann et al., 2018; Brown et al., 2017; Wang et al., 2020; Li et al., 2020). NO_y plays an
43 important role in critical environmental issues concerning the Earth's atmosphere from local air
44 pollution to global climate change (Sun et al., 2011; Ge et al., 2019). The oxidation of NO to
45 NO_2 and finally to HNO_3 is the backbone of the chemical mechanism producing ozone (O_3)
46 and $PM_{2.5}$ (particulate matter with size $\leq 2.5 \mu m$), and determines the oxidization capacity of
47 the atmosphere. Recently, as O_3 has [still](#) increased [even](#) with decreasing NO_x emissions over
48 many regions, including East Asia, interest in the heterogeneous reaction of NO_y , which is yet
49 to be understood, has increased (Brown et al., 2017; Stadtler et al., 2018). Currently, the lack of
50 measurement of individual NO_y species is hindering a comprehensive understanding of the
51 heterogeneous reactions (Anderson et al., 2014; Wang et al., 2017b; Chen et al., 2018b; Akimoto
52 and Tanimoto, 2021; Stadtler et al., 2018).

53 In particular, the evidence for the heterogeneous formation of HONO in relation to high
54 $PM_{2.5}$ and O_3 occurrences in urban areas is increasing (e.g., (Li et al., 2021b)). As an OH
55 reservoir, HONO expedites the photochemical reactions involving VOCs and NO_x in the early
56 morning, leading to O_3 and fine aerosol formation. Nonetheless, its formation mechanism has
57 not been elucidated sufficiently enough to be constrained in conventional photochemical
58 models. In addition to the reaction of NO with OH (Bloss et al., 2021), various pathways of
59 HONO formation have been suggested via laboratory experiments, field measurements, and
60 model simulations: direct emissions from vehicles (Li et al., 2021a) and soil (Bao et al., 2022),
61 photolysis of particulate nitrate (Gen et al., 2022), and heterogeneous conversion of NO_2 on
62 various aerosol surfaces (Jia et al., 2020), ground surface (Meng et al., 2022), and microlayers
63 of the sea surface (Gu et al., 2022). Among these, the heterogeneous reaction mechanism on the
64 surface is of major interest.

65 HONO has been mostly measured during intensive campaigns in urban areas using
66 various techniques, such as a long path absorption photometer (Kleffmann et al., 2006;Xue et
67 al., 2019), chemical ionization mass spectrometry (Levy et al., 2014;Roberts et al., 2010), ion
68 chromatography (VandenBoer et al., 2014;Gil et al., 2020;Ye et al., 2016;Xu et al., 2019),
69 monitor for aerosols and gases in ambient air (MARGA) (Xu et al., 2019), and quantum cascade
70 - tunable infrared laser differential absorption spectrometry (QC-TILDAS) (Lee et al., 2011;Gil
71 et al., 2021). Among these methods, QC-TILDAS has served as a reference for the
72 intercomparison of measurement data obtained using different techniques due to its high time
73 resolution and stability (Pinto et al., 2014). Previous studies have reported that the maximum
74 HONO of several ppb levels has been observed at nighttime. [In comparison, the WRF-Chem
75 and RACM2 model captured approximately 67 %–90 % of the observed HONO in megacities
76 such as Beijing](#) (Tie et al., 2013;Liu et al., 2019).

77 In recent years, machine learning (ML) methods have been [employed](#) in the
78 atmospheric science field for pattern classification (e.g., new particle formation event) and
79 forecasting and spatiotemporal modeling of O₃ and PM_{2.5} (Arcomano et al., 2021;Shahriar et
80 al., 2020;Krishnamurthy et al., 2021;Cui and Wang, 2021;Joutsensaari et al., 2018;Chen et al.,
81 2018a;Kang et al., 2021). Among the ML methods, the neural network (NN) architecture is
82 widely used owing to its powerful ability to process large amounts of data, [realizing
83 performance improvement in comparison to the performance of conventional models through
84 integration with physical equations](#) (Reichstein et al., 2019;Schultz et al., 2021). As an NN
85 architecture, a multilayer artificial NN (ANN) that is denoted as a deep NN (DNN) employs a
86 statistical method that learns nonlinear relations in data and yields the optimum solution for the
87 target species without prior information about the physicochemical processes. DNN is more
88 beneficial than other NN architecture, such as convolution NN or long-short term memory,
89 because it works well for discrete spatiotemporal data. Generally, the performance of DNN is
90 similar to or better than that of other ML methods for small as well as large datasets (Baek and
91 Jung, 2021;Dang et al., 2021;Sumathi and Pugalendhi, 2021).

92 [The DNN method requires lots of data to employ it as atmospheric chemical constituent
93 estimation; therefore, the size of the measurement data is a limiting factor for trace species,
94 such as HONO, that are not routinely measured. In this regard, previous studies had been
95 attempted to estimate the daily average HONO mixing ratio by employing ensemble ML models](#)

with satellite measurements (Cui and Wang, 2021). Furthermore, a simple NN architecture using ground measurement variables that is believed to be deeply involved in HONO formation, was used to calculate the hourly HONO mixing ratio (Gil et al., 2021). The accuracy of the hourly HONO estimated from input variables, such as aerosol surface areas and mixed layer height, is rated better than the daily HONO estimate.

This study aims to develop a user-friendly “reactive nitrogen species simulation using DNN” model (RNDv1.0) that estimates the HONO mixing ratios from the real-time measurements of criteria pollutants and meteorological variables. This study is the first to calculate the HONO mixing ratios using RNDv1.0. The entire construction process is comprehensively described, and the performance is evaluated via comparison with the results of simulations using a commonly used model and observations over several years.

2. Model description

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

2.1. Collection of measurement data for model construction

To construct RNDv1.0, measurement data were obtained, including HONO, reactive gases, and meteorological variables. Note that the HONO measurement data were used for model construction but not required to run the RND model. The HONO mixing ratio was measured in Seoul using a QC-TILDAS system during May–June 2016, June 2018, and April–June 2019 (Lee et al., 2011; Gil et al., 2021), and a MARGA system during May–June 2021 and October–November 2021 (Gil, 2022). When testing and evaluating the atmospheric HONO measurement methods, QC-TILDAS was chosen as the reference method to compare the

125 ambient HONO mixing ratios measured using several different techniques owing to its
126 advantages of low detection limits (~0.1 ppbv) and high temporal resolution (Pinto et al., 2014).
127 More details on measurements can be found elsewhere (Gil et al., 2021; Gil, 2022).

128 HONO was measured at the Olympic Park (37.52° N, 127.12° E) during the Korea–
129 United States Air Quality (KORUS-AQ) study in 2016 (Kim et al., 2020; Gil et al., 2021), at the
130 campus of Korea University (37.59° N, 127.03° E) in 2018 and 2021, and at the site near the
131 Korea University campus (37.59° N, 127.08° E) in 2019 (NIER, 2020) (Figure S1). In addition
132 to HONO, trace gases including O₃, NO₂, CO, and SO₂ as well as meteorological **variables**
133 including temperature (T), relative humidity (RH), wind speed (WS), and wind direction (WD)
134 were measured. Note that HONO was not significantly correlated with any of these variables
135 (Figure S2). The measurement statistics **for the entire experimental periods** are presented in
136 Table 2 and Table S1. In brief, the 10th and 90th percentile mixing ratios of hourly HONO, NO₂,
137 and O₃, were 0.3 and 2.0 ppbv, 10.0 and 47.0 ppbv, and 8.0 and 75.0 ppbv, respectively.

138

139 **2.2. Data preprocessing**

140

141 The observation dataset was prepared for RNDv1.0 model construction. As input
142 variables, hourly measurements of chemical and meteorological **variables** were used, including
143 the mixing ratios of O₃, NO₂, CO, and SO₂, along with T, RH, WS, WD, and solar zenith angle
144 (SZA) to estimate the target species, HONO, as the output. **The WD in degrees was converted**
145 **to a cosine value for continuity. In the last step of data processing, hourly measurement sets**
146 **were removed from the input data set if any of the nine variables were missing.** Finally, 54.2 %
147 of all the available measurement data (2847) were used to construct and evaluate RNDv1.0.

148 Since the measurements of the considered nine variables varied over a wide range in
149 different units, they were normalized to avoid bias during the calculations. Among the widely
150 used normalization methods, *min–max scaling* method was adopted, and **the** input variables
151 were normalized against the minimum and maximum values **herein** (Eq. 1):

152

$$153 \quad x_{\text{sca}} = \frac{x_{\text{raw}} - F_2(X)}{F_1(X)}, \quad (\text{Eq. 1})$$

154

155 where x_{raw} is the raw data, x_{sca} is the scaled value, and the scale factors of F_1 and F_2 correspond
156 to the maximum-minimum and minimum values of the input variable (X), respectively, which
157 are listed in Table 2.

158

159 **2.3. Neural network architecture and hyperparameters**

160

161 The network was built using the above input variables to calculate HONO. RNDv1.0
162 comprises five hidden layers (Figure 2), which employ an exponential linear unit (ELU) as an
163 activation function (Eq. 2).

164

$$165 \quad \text{ELU: } \phi(x) = \begin{cases} e^x - 1 & (x < 0) \\ x & (x \geq 0) \end{cases}. \quad (\text{Eq. 2})$$

166

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

173

174 **2.4. Model training and k-fold cross validation**

175

176 RNDv1.0 was trained, validated, and tested with the HONO measurements obtained
177 during May–June 2016 and June 2018, April–June 2019, and May–June 2021 and October–
178 November 2021, respectively (Figure 3). The number of data used for the training and
179 validation was 1122 and that for testing was 1725.

180 Using the hyperparameters specified in the previous section, the model performance
 181 was first validated using the k-fold cross validation (KFCV) method, which is especially useful
 182 for small datasets (Bengio and Grandvalet, 2003). In the KFCV method (Figure 3), the entire
 183 data are randomly divided into k subsets, of which k – 1 sets are used for training and the
 184 remaining one is used for validation. In this study, k was set to 5. The accuracy was determined
 185 via index of agreement (IOA), which is expressed as follows (Eq. 3):

186

$$187 \quad IOA = 1 - \frac{\sum_{i=1}^n (O_i - P_i)^2}{\sum_{i=1}^n (|P_i - \bar{O}| + |O_i - \bar{O}|)^2}, \quad (\text{Eq. 3})$$

188

189 where O_i , P_i , \bar{O} , and n are the observed value, predicted value, average of the observed values,
 190 and number of nodes, respectively.

191 As IOA varies according to the number of nodes, it was calculated for the measured
 192 ($HONO_{obs}$) and calculated ($HONO_{mod}$) mixing ratios by varying the number of nodes from 0 to
 193 100 in each hidden layer. The best performance was obtained with 41 nodes, for which the
 194 average IOA was 0.89 ± 0.01 (Figure 4). The high IOA value signifies that the performance of
 195 RNDv1.0 is adequate, and it is capable of simulating the ambient HONO mixing ratio using the
 196 routinely measured criteria pollutants and meteorological variables.

197 The performance of RNDv1.0 was compared with that of other models, including
 198 CMAQv5.3.1 (Appel et al., 2021), random forest (RF), and single-layer ANN (Gil et al., 2021),
 199 using the 2016 measurement data. The RF model was constructed using the KFCV method and
 200 the same input variables as RNDv1.0 (Figure S4). Its performance was evaluated based on mean
 201 absolute error (MAE), root mean square deviation (RMSE), and Pearson correlation coefficient
 202 (r):

203

$$204 \quad MAE = \frac{\sum_{i=1}^n |O_i - P_i|}{n}, \quad (\text{Eq. 4})$$

$$205 \quad RMSE = \sqrt{\frac{\sum_{i=1}^n (O_i - P_i)^2}{n}}, \quad (\text{Eq. 5})$$

$$206 \quad r = \frac{cov(O,P)}{\sigma_O \sigma_P}, \quad (\text{Eq. 6})$$

207

208 where σ and cov denote the standard deviation and covariance, respectively.

209 All models except CMAQ simulated the measured HONO mixing ratio fairly well
210 (Figure 5). CMAQ not only underestimated the measured HONO but also failed to represent its
211 diurnal variation (Figure 6). The statistical information about the performance of the four
212 models is presented in Table 3. The mean HONO mixing ratio measured and calculated using
213 CMAQ, RF, ANN, and RNDv1.0 was 0.94, 0.09, 0.95, 0.88, and 0.89 ppbv, respectively. Of the
214 four models, RF exhibited the best performance followed by RND. ANN advantageously
215 calculates HONO more accurately than RND as it uses more input variables, but it has a lower
216 data capture rate (41.5 %) compared to RND (97.7 %) or RF (85.3 %).

217

218 2.5. Model test

219

220 RNDv1.0 and the RF model were tested using data obtained in June 2018, April 2019,
221 and May–June 2021 and October–November in 2021, which were not used for RNDv1.0
222 training (Figure 3). Note that the RF model outperformed the other three models in the training
223 and validation process (Figure 5). Although the performance of RNDv1.0 was slightly lower
224 than that of the RF model, simulated and measured HONO mixing ratios were in good
225 agreement. Interestingly, the performance of the RF model was much worse than RNDv1.0 in
226 the testing process (Figure 7). The IOA and correlation coefficient of the RF model were
227 extremely low (0.29 and -0.02 , respectively).

228 The performance of RNDv1.0 was slightly lower than that of the RF model, but it well
229 traced the HONO mixing ratio. Among the test dataset, the early winter (October–November)
230 data are particularly valuable for demonstrating the applicability of RNDv1.0 because they stem
231 from different weather conditions than the training dataset. For example, HONO mixing ratios
232 reached over 4 ppbv when the daily average $PM_{2.5}$ concentration increased to $120 \mu g m^{-3}$ during
233 severe haze pollution events. Therefore, in the next step, the performance of RNDv1.0 was
234 compared for the two cases by dividing the testing dataset into a group in which all input
235 variables fall within the range of the training dataset and a group which does not meet this

236 criterion. In RNDv1.0, there was no significant difference in performance between the two
237 groups (Figure S5 and Table S2). When the data in which at least one input variable does not
238 fall within the range of the training dataset were excluded from the test dataset, no significant
239 difference was observed in the performance of RNDv1.0 between the two that meet same
240 atmospheric conditions or do not meet the criteria (Figure S5 and Table S2). These extreme
241 atmospheric conditions can make the model performance be worsened. Except for these
242 extremes, RNDv1.0 well traced the variation of the HONO mixing ratio. These results
243 demonstrate the applicability of RNDv1.0, which is not strictly constrained by atmospheric
244 conditions. The influence of input variable are further analyzed in the next section.

245

246 **2.6. Bootstrap test and feature importance**

247

248 A simple bootstrapping test was conducted for both RNDv1.0 and the RF model to
249 evaluate the relative importance of the input variable to the HONO estimates. In this analysis,
250 each variable was set to zero and MAE was calculated as an evaluation metrics (Kleinert et al.,
251 2021). Among the nine input variables of RNDv1.0, NO₂ was found to have the greatest
252 influence on HONO concentration, followed by RH and T (Table 5). The highest MAE of 0.59
253 ppbv could be considered as the maximum uncertainty of RNDv1.0 due to the input variable.
254 The bootstrap test result well agreed with that of our previous study (Gil et al., 2021), where
255 more variables such as aerosol surface area and mixing layer height were incorporated into the
256 model, it highlights the crucial role of precursor gases and heterogeneous conversion in HONO
257 formation.

258 In contrast, in the RF model, O₃ was the most important variable. This is likely due to the
259 distinct inverse relationship between O₃ and HONO in the diurnal patterns, and the O₃
260 variations over a wide range. In conjunction with the evaluation of the test dataset presented in
261 the previous section, the results of the feature importance for the two models demonstrate the
262 ability of RNDv1.0 to simulate the HONO mixing ratio more adequately in urban areas
263 compared to the RF model. Thus, it is reasonable to state that RNDv1.0 constructed using
264 routinely measured criteria pollutants and meteorological variables can sufficiently capture the
265 HONO variability in the urban atmosphere.

266

267 **3. Operation and application of RNDv1.0**

268

269 The RNDv1.0 package is provided as an operational model, and the .h5 files that can
270 be opened in Python. To run RNDv1.0, the measurement data for nine input variables are
271 required and needed to be properly prepared, as described in Section 2.2. Once the input data
272 are ready, open RNDv1.0 with the input data files using the code provided in the example
273 (Figure S3). Then, RNDv1.0 calculates and presents the HONO results as scaled values (x_{sca}),
274 which then can be converted to the HONO mixing ratio (ppbv) via the two scale factors shown
275 in Table 2 (Eq. 5):

276

$$277 \text{HONO (ppbv)} = \text{HONO}_{sca} \times F_1(\text{HONO}) + F_2(\text{HONO}). \quad (5)$$

278

279 The HONO calculated using Eq. 5 can be applied to an urban photochemical cycle
280 simulation. As is already known, the photolysis of HONO is a major source of OH radicals in
281 the early morning when the OH level is low, and this OH affects daytime O₃ formation through
282 photochemical reactions with VOCs and NO_x, which are primarily emitted during the morning
283 rush hour in urban areas. Furthermore, the OH produced from HONO promotes the
284 photochemical oxidation of SO₂ and VOCs, leading to aerosol formation. However, the HONO
285 formation mechanism is still poorly understood, which hinders the accurate simulation of O₃
286 and fine aerosols as well as HONO in conventional photochemical models.

287 The framework for 0-dimension atmospheric modeling (F0AM), which utilizes the
288 MCM v3.3.1 chemical reaction mechanisms (Wolfe et al., 2016), can be used to simulate the
289 diurnal variation of O₃ with the measurements of several reactive gases (NO, NO₂, CO, HCHO,
290 VOCs, and HONO). Detailed information about F0AM can be found in
291 (<https://sites.google.com/site/wolfegm/models>) and in previous studies (Wolfe et al., 2016; Gil
292 et al., 2020). When the F0AM model is run without HONO, it is unable to reproduce the
293 concentration and diurnal cycle of the observed O₃ (Figure 8). In comparison, the model well
294 simulates the O₃ within 2 ppbv when HONO is considered, which is the result of RND v1.0.

295 This is mainly due to the missing OH produced by HONO photolysis in the early morning. Its
296 production rate is estimated to be 0.57 pptv s^{-1} , contributing approximately 2.28 pptv to the OH
297 budget during 06:00–11:00 (Local Sun Time) (Gil et al., 2021). Given that OH is mainly
298 produced from the photolysis of O_3 under high sun, the early morning supply of OH from
299 HONO photolysis will expedite the photochemical cycle involving NO_x and VOCs, promoting
300 O_3 and secondary aerosol formation. The presence of HONO in the photochemical model
301 allows for the accurate estimation of OH radicals; thus, the incorporation of RNDv1.0 into
302 conventional models will improve their overall performance.

303

304 **4. Summary and implications**

305

306 In this study, we developed the RND model to calculate the mixing ratio of NO_y in
307 urban atmosphere using a DNN along with measurement data. The target species of RNDv1.0
308 is HONO, and its mixing ratio is calculated using criteria pollutants, including O_3 , NO_2 , CO,
309 and SO_2 , as well as meteorological variables, including T, RH, WS, WD, and SZA. These
310 variables are routinely measured through monitoring networks. RNDv1.0 was trained and
311 validated using the HONO measurements data obtained in Seoul by adopting a KFCV method
312 and tested with other HONO datasets. The test results demonstrate that RNDv1.0 adequately
313 captures the characteristic variation of HONO.

314 RNDv1.0 was constructed using the measurements made in a high NO_x environment where
315 the maximum NO_2 reached about 80 ppbv. During the measurement period, the HONO mixing
316 ratio was increased up to about 7 ppb under the influence of air masses originating from China.
317 When applying RNDv1.0 to regions or times heavily affected by transport, the model could
318 possibly underestimate the HONO level without more detailed information, such as
319 nanoparticles. Indeed, a previous study showed that HONO formation is closely related to the
320 surface areas of submicron particles (Gil et al., 2021). Nevertheless, RNDv1.0 is
321 advantageously a relatively inexpensive test for measurement quality control and location
322 selection, and it supports the data used for traditional chemistry models based on the current
323 knowledge of the urban photochemical cycle. Therefore, RNDv1.0 can serve as a
324 supplementary tool for conventional forecasting models. Attempts are currently being made to

325 [estimate ground HONO from satellite observations](#) (Clarisse et al., 2011;Theys et al.,
326 2020;Armante et al., 2021), and RNDv1.0 will be useful for validating the satellite-derived
327 HONO.

328

329 **5. Acknowledgements**

330

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332 (2020R1A2C3014592) and Korea Institute of Science and Technology (KIST2E31650-22-
333 P019).

334

335 **6. Code availability**

336

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

339

340 **7. Author contributions**

341

342 JG and ML designed the manuscript and developed the model code. JK, GL, and JA
343 provided [the](#) HONO measurements and CK provided the CMAQ model data. All the authors
344 contributed to the manuscript.

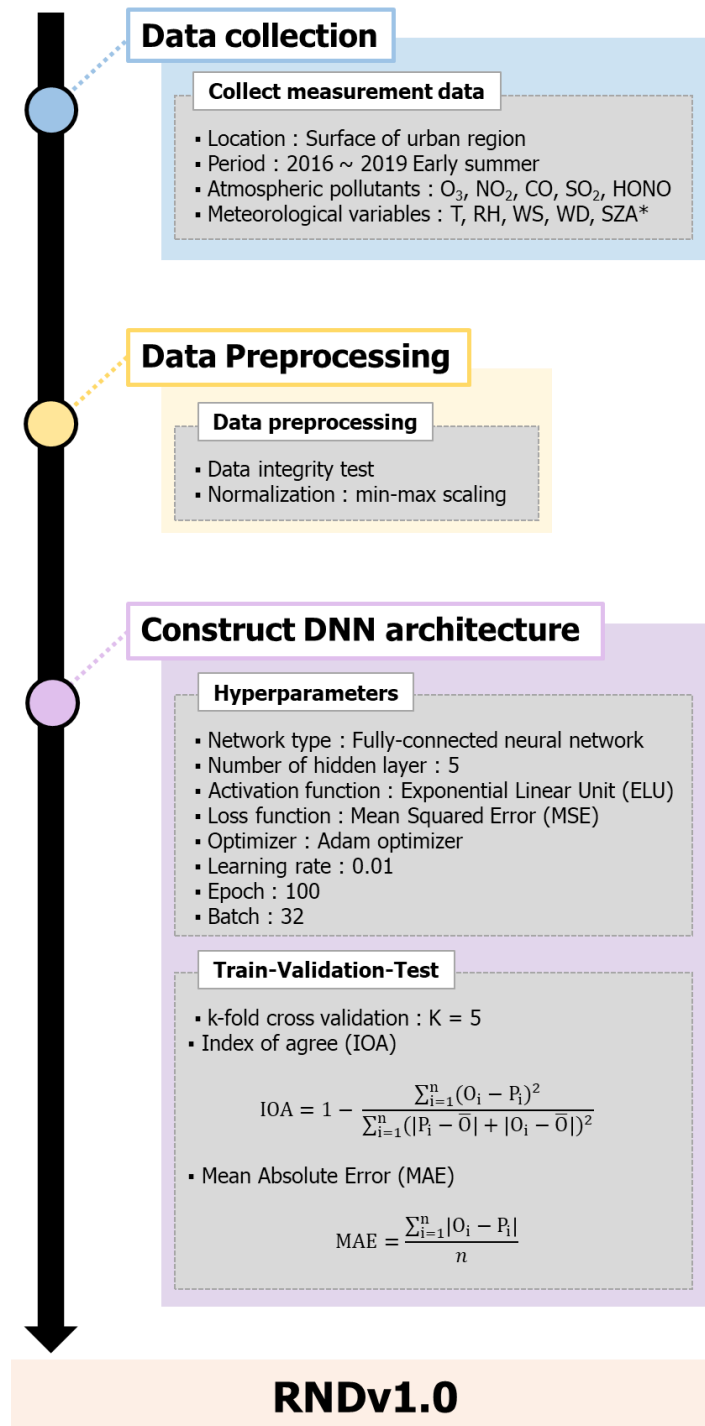
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346 **8. Competing interests**

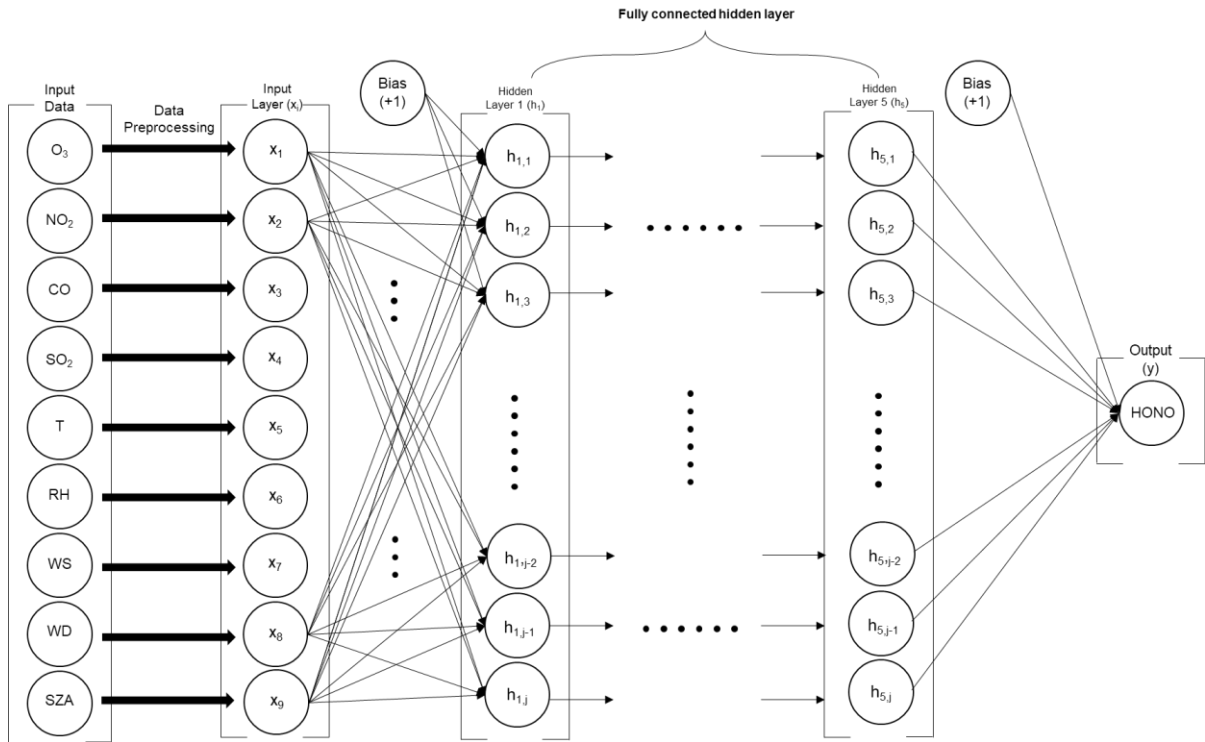
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348 The authors declare that they have no conflict of interest.

349



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

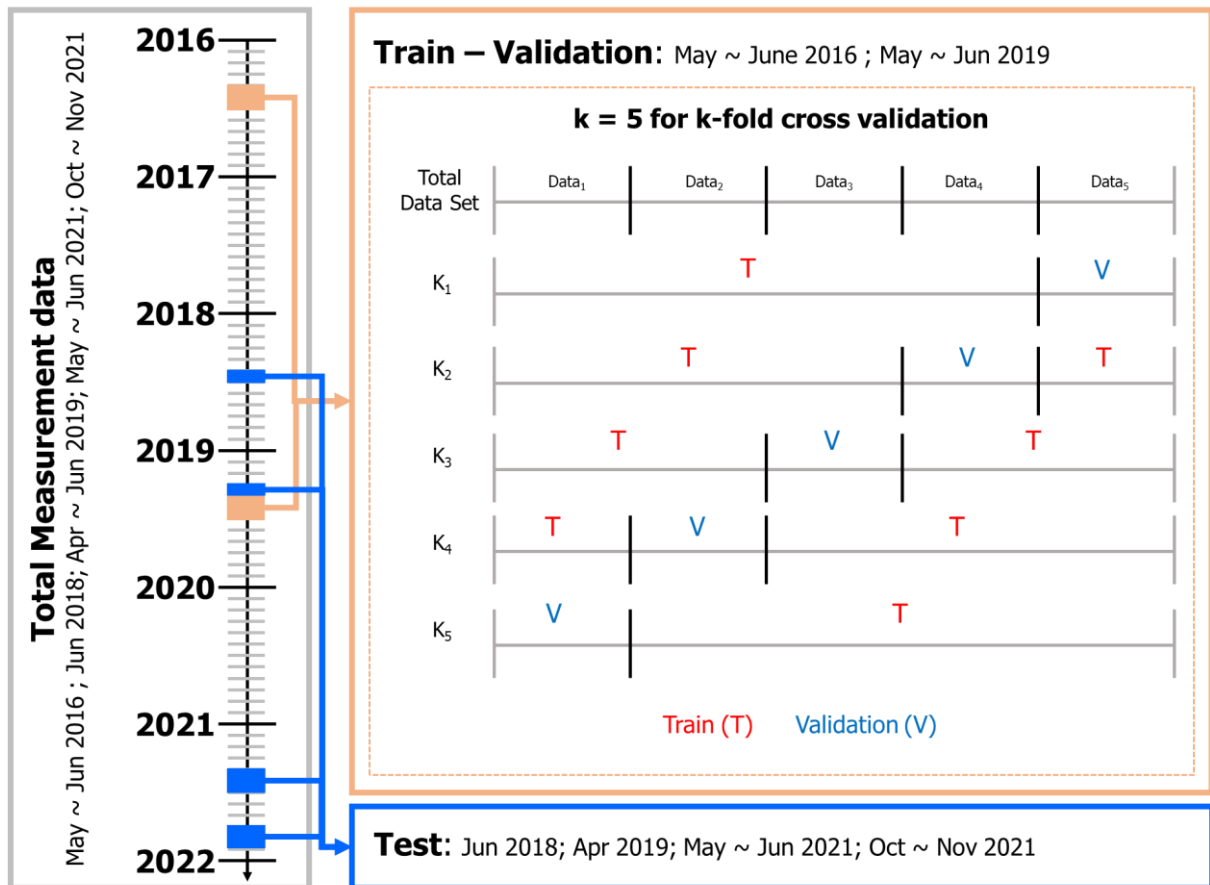


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355 **Figure 2.** Structure of the deep neural network built for RND v1.0.

356

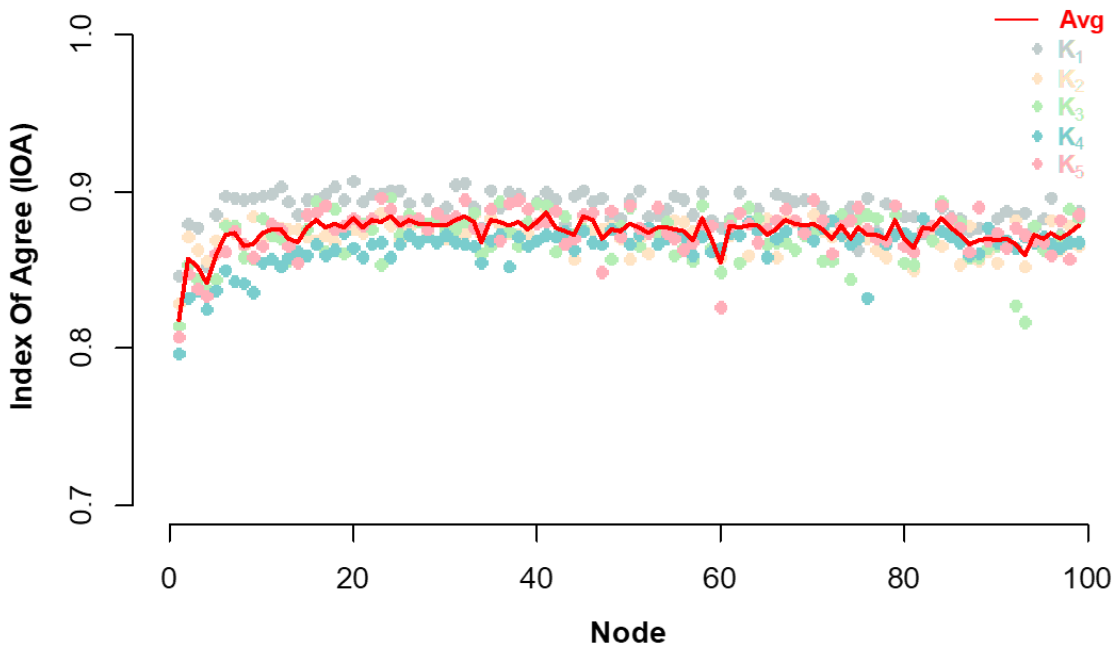
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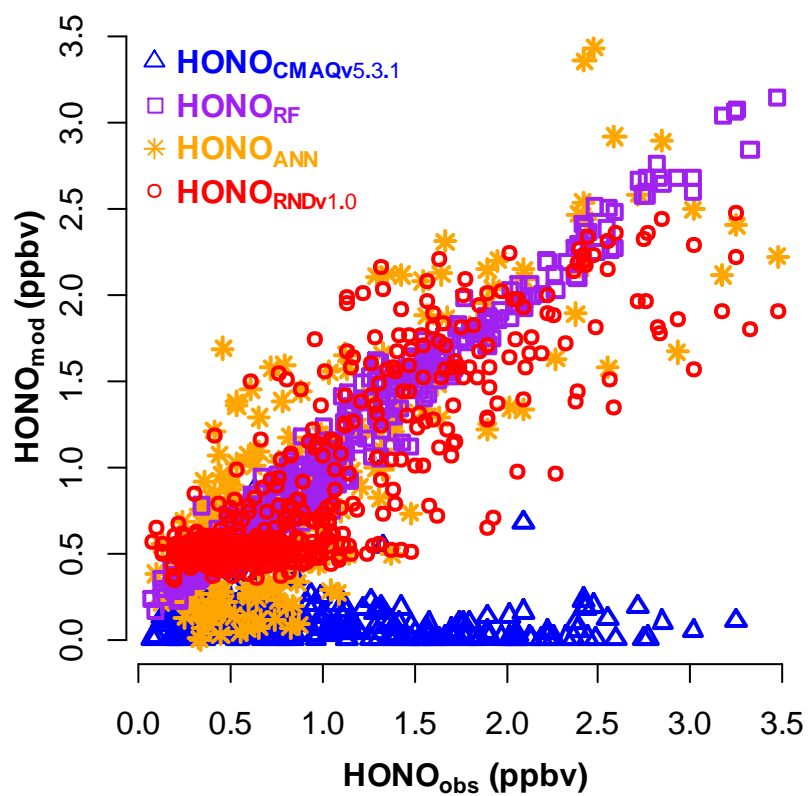
359 **Figure 3.** Training, validation, and test design to build RNDv1.0 using the measurement data.
 360 The k-fold cross validation was performed using randomly divided five subsets of the training
 361 data set.

362



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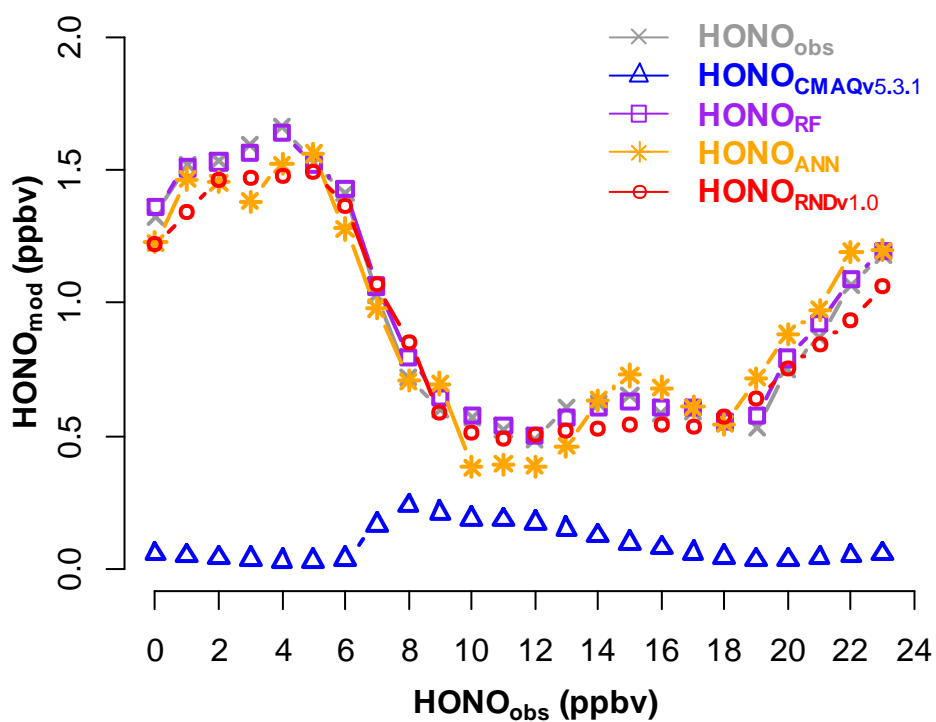
364 **Figure 4.** Index of Agreement (IOA) for k-fold cross validation. Solid circle and red line
 365 represent IOA for each validation (k = 5) and the average of five validation sets at each node
 366 number.



367

368 **Figure 5.** Comparison between the measured HONO (HONO_{obs}) and calculated HONO
 369 (HONO_{mod}) using CMAQv5.3.1 (blue triangle), RF (purple square), ANN (orange star), and
 370 RNDv1.0 (red circle) during the KORUS-AQ campaign (May–June 2016).

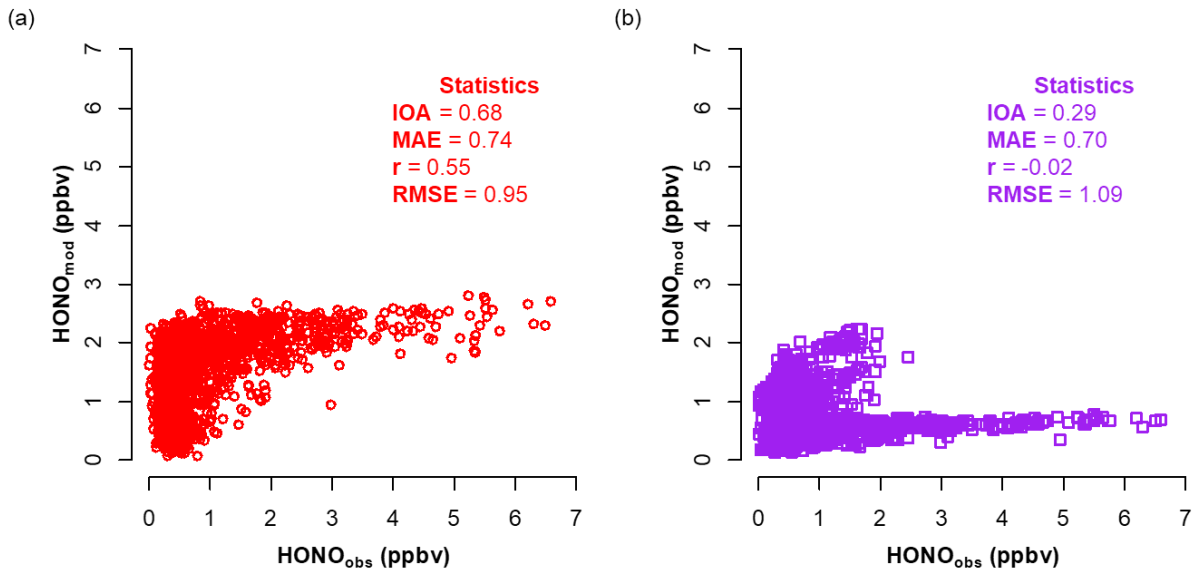
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372

373 **Figure 6.** Average diurnal variation of the measured HONO (HONO_{obs}) and calculated
 374 HONO (HONO_{mod}) using CMAQv5.3.1 (blue triangle), RF (purple square), ANN (orange
 375 star), and RNDv1.0 (red circle) during the KORUS-AQ campaign (May–June 2016).

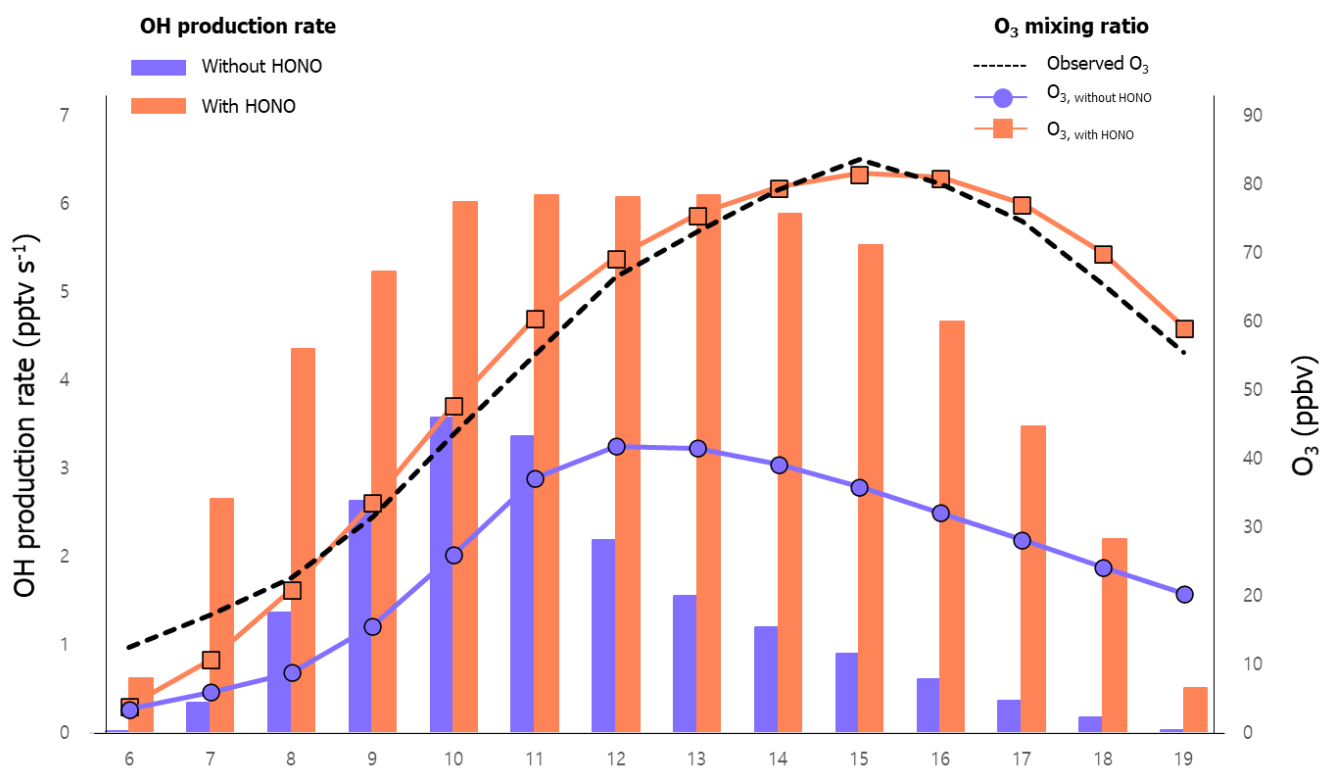
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378 **Figure 7.** Relationship between measured HONO (HONO_{obs}) and modeled HONO (HONO_{mod})
 379 using (a) RNDv1.0 and (b) a Random Forest model for the test dataset.

380



382

383 **Figure 8.** For June 2016, the diurnal variations of O₃ (line) and OH production rate (bar)
 384 calculated using the FOAM photochemical model with (orange) and without (blue) HONO
 385 estimated from the RNDv1.0 model. The measured and calculated O₃ values are compared.

386

387 **Table 1.** Resources for constructing the 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	<i>Python library</i>
Keras	v2.4.3	<i>Python library</i>
Pandas	v1.0.5	<i>Python library</i>
Numpy	v1.18.5	<i>Python library</i>

388 *GPU denotes graphic processing unit

389 **Table 2.** Input variables and their concentrations (10th–90th percentile of the hourly
 390 **measurements**), coverage, and scale factors for the RNDv1.0 model. Measurements were
 391 conducted 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 (°)	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 (°)	99.4	359.565	0.235
Output Variables				
HONO	0.3–2.0 (ppbv)	81.1 %	3.447	0.013

392 * Maximum–Minimum

393 ** Minimum value

394

395 **Table 3.** Performance of the chemical transport model (CMAQv5.3.1) and machine learning
396 (ML) models, including Random Forest (RF), Artificial Neural Network (ANN), and RNDv1.0,
397 on the measurement data from 2016 KORUS-AQ campaign, which were used for training.

	CMAQv5.3.1	RF	ANN	RNDv1.0
IOA	0.44	0.99	0.86	0.9
r	-0.07	0.99	0.81	0.84
MAE	0.82	0.1	0.38	0.27
RMSE	1.06	0.12	0.41	0.37

398

399

400 **Table 4.** Results of the bootstrap test of measurement data used to train the RF and RNDv1.0
 401 models. The greater the MAE, the greater the influence of the variable.

Variable	RF		RNDv1.0	
	MAE	Feature Importance	MAE	Feature Importance
-	0.10	-	0.28	-
O ₃	0.57	1	0.29	8
NO ₂	0.24	4	0.59	1
CO	0.19	7	0.37	5
SO ₂	0.17	8	0.34	6
Solar zenith Angle (SZA)	0.25	2	0.41	4
Temperature (T)	0.21	5	0.52	2
Relative humidity (RH)	0.25	3	0.52	2
Wind speed (WS)	0.20	6	0.34	6
Wind direction (WD)	0.13	9	0.29	8

402

403

404 **Reference**

405

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