Review of gmd-2021-347

Simulation Model of Reactive Nitrogen Species in an Urban Atmosphere using a Deep Neural Network: RND v1.0

by Junsu Gil et al.

General comments:

This manuscript describes a new application of a simple feed forward neural network model to calculate HONO mixing ratios based on a set of other measured variables. While this is an interesting and worthwhile application, the paper lacks the necessary details in the description of the deep learning model and contains no ablation studies which are needed to provide the credibility in the results. I also question the validity of the cross validation and test cases that are discussed, because I doubt that these test cases are truly independent data samples. There is no proof of the generalisation capability of the model, so it may well be that this model fails completely if it were applied to measurement data obtained under different conditions.

In summary, this manuscript falls between "major revisions" and "reject". In computer science conferences it would be ranked "weak reject", which means the paper could be saved if the authors invest substantial work in rerunning their model several times and improving the text.

- We are grateful for your constructive and considerate comments. The point-by-point responses are given below, along with relevant parts of the revised manuscript, where all changes are marked in blue.

Specific comments:

Abstract: Confusing sentence after "In this study,". After reading 3 times I understood that you are resolving the acronym RND here, but this is well hidden. Suggestion: "In this study, a new simulation approach to calculate HONO mixing ratios using a deep learning technique based on measured variables was developed. The 'Reactive Nitrogen species Deep neural network' (RND) has been implemented in Python. It was trained, ..."

- As suggested, the abstract is rewritten for clarification.

- Line 17-22: 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.

Abstract: Why should RND be called a *supplementary* model? What does it supplement?

- As mentioned in your comment L.250/251, HONO mixing ratios estimated from the RNDv1.0 model can be used for various purposes. Finally, you agreed that RND was a ‘supplementary’ tool.

l.35: too vague "observational constraints on individual species". Does this refer to NOy compounds or any species involved in the tropospheric ozone production cycle?

- It refers to NOy species. This sentence is removed in the revised manuscript.

l.40: NOy has been the focus of attention already in the 1990s. See for example papers by Sandy Sillman et al. You may say "renewed attention".

- Yes, you are right. This sentence is meant to emphasize the heterogeneous reaction of nitrogen oxides, and rephrased in the revised manuscript.
- Line 51-54: 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.

l.43: to the uniniated reader it might not be clear what heterogeneous reactions have to do with NOy and ozone chemistry. This would merit one or a few more general sentence(s) to describe NOy chemistry. If this text will get a little longer, please consider sumamrizing the HONO/NOy chemistry in a supplement and refer to it. Nevertheless, one or two sentences will be needed here.

- As suggested, the background information is added to the Introduction in the revised manuscript.
- Line 57-69: In particular, there are growing number of evidence for heterogeneous formation of HONO in relation to high PM2.5 and O3 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 O3 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 NO2 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.

l.44: you could add https://doi.org/10.5194/acp-18-3147-2018 to the list of references here.

- It is cited as follows.
- Line 54-56: 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; Stadtler et al., 2018)

l.52/53: it would be useful to know if there is general agreement among these different measurement methods or if they haven't reached a satisfactory level of consistency yet. In the following sentence, please provide some order of magnitude numbers of observed versus simulated HONO levels (or a value range).

- In several intercomparison studies (Pinto et al.,2014; Yi et al., 2021; Yang et al., 2022), all instruments showed reasonable performance with their inherent weaknesses, depending on conditions such as meteorology,

pollution levels, and so on. In general, however, QC-TILDAS was accepted as a reference method with which all measurements from different techniques were compared.

- The calculated HONO from model explains at most 60~90 % of the observed.
- Line 75-79: 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). 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)

l.57: the recent adaptation of machine learning techniques in atmospheric sciences is more general that "multi layer artificial neural network". In this context, it suffices to say that "machine learning" has been adopted. Then, in a following sentence you can narrow this down to the employment of deep (artificial) neural networks, which have a capability to learn more complex non-linear relations in data, but also require larger amounts of data for training." The selection of references appears a bit arbitrary. For example, there is a whole special issue in Philosophical Transactions A () on machine learning for weather and climate. Indeed, you may want to first provide two or three general references for ML in atmospheric science (with cf.), then write a sentence which refers specifically to atmospheric chemistry/atmospheric composition and provide some more references there.

- Thank you for detailed advice. This part is rewritten as suggested.
- Line 81-95: 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 in 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 of 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).

l.59-62: the description why deep learning might be useful for the analysis of atmospheric chemical measurements remains vague and superficial. You should state explicitly that neural networks learn relations in data (similar to function fitting) and you should state in what way NNs may improve on numerical simulations (I guess you refer to the fact that they are inherently bias-free?).

- The NN architecture has advantage in handling the data which has non-linear relation between dataset. Also it shows good performance when the information of physicochemical process is not clear. And finally, the result from NN architecture can be used to numerical models as input data, and it can contribute to the improvement of prediction performance indirectly. This part is rewritten as follows.
- Line 88-95: 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).

l.62/63: introduction of the model acronym: difficult to disentangle the sentence - see comment on abstract above.

- The full name of the model is separated with quotation marks, and this part is rewritten as follows.
- Line 105-107: 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.

l.67: as this is supposed to be a manuscript for the special issue on "machine learning methods and benchmark datasets", you should add a statement here that the code and training data can be downloaded from ..." (you can of course also refer to the code and data availability section here). Re-usability of your model is a key aspect for this special issue (and for GMD in general).

- As suggested, a statement declaring the reusability of our model, is added to the revised manuscript.
- Line 119-120: The dataset used to train-test-validation can be downloaded from Gil et al., 2021.

l.70: the steps which are described don't guide the development of RND, but describe the typical machine learning workflow.

- Yes, these steps are like a general machine learning model construction workflow which is for users and stated in the text.
- Line 115-118: The development of RNDv1.0 model follows the systematic steps similar to a general machine learning model construction workflow, that including collecting data, preprocessing data, building the DNN, training and validating the model, and testing the performance of the model (Figure 1).

l.77: similar issue - this reads as if every user of RND will first have to perform measurements for her/himself. Please separate the dataset preparation from the model development. The model should be generalizable, i.e. be independent of the specific set of measurements which you describe in the paper.

- Yes, you are right. HONO measurement is not a part of dataset preparation for model run, but for model development. To clarify this point, the section title is modified, and a statement is added in the revised manuscript.
- Line 122: 2.1. Collection of measurement data for model construction
- Line 125-126. It is noteworthy that the HONO measurement data is for model construction and is not required to run the RND model.

l.105: "wind direction should be converted..." - please describe what you did, not what should be done.

- This part is reworded.
- Line 154: Wind direction in degrees were converted to a cosine value for continuity

l.106: "missing values" same as above. Did you filter or interpolate?

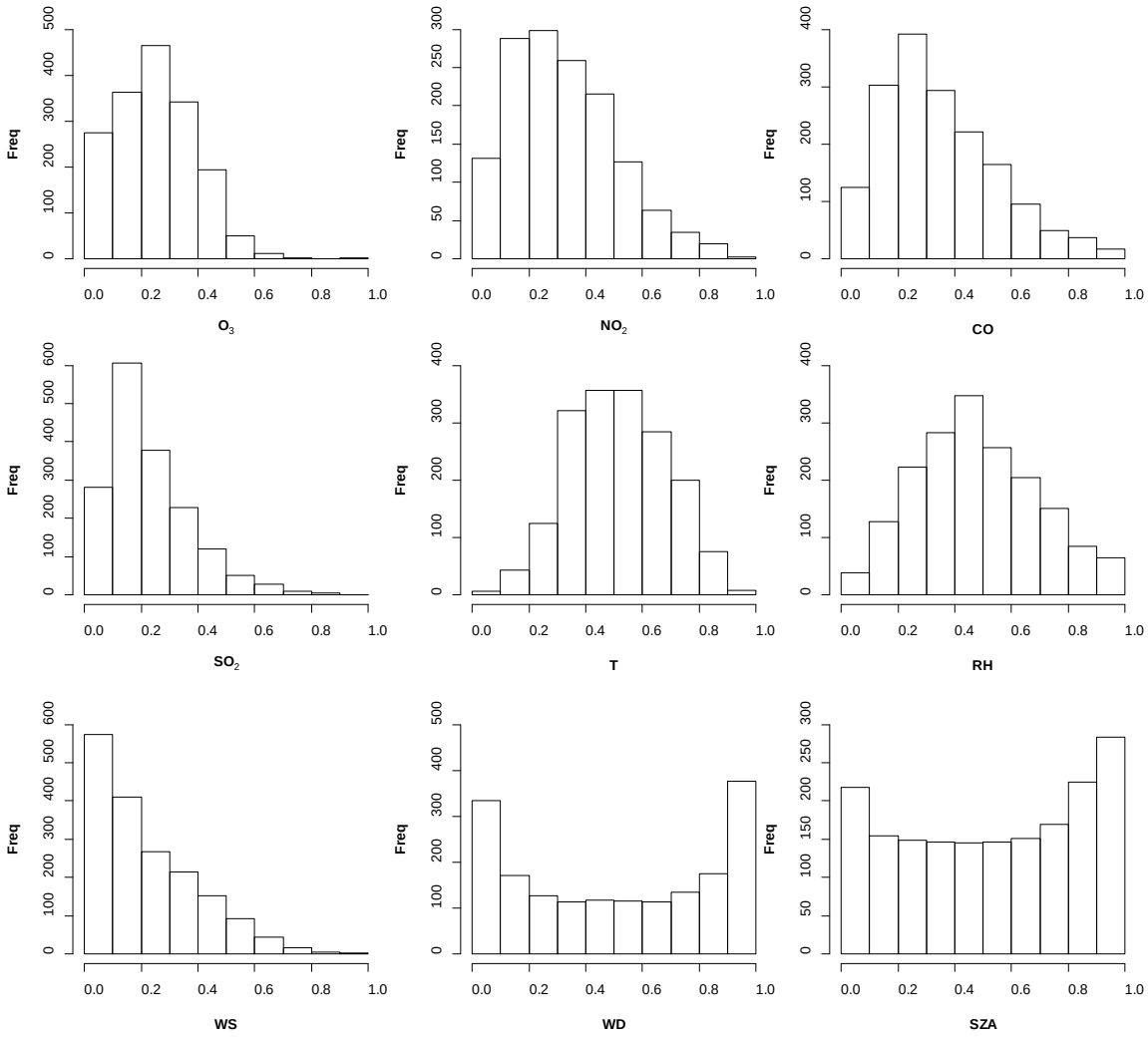
- This part is also reworded. They were filtered.
- Line 153-155: For model operation, data of all variables must exist in each hourly data set. So we conducted data integrity test, and filtered the hour array where missing values are exist.

l.107: what is an "array of measurement data"? Also, what is missing is a description of the time resolution of the measurements and how many independent samples were prepared for the machine learning. How was the train-test-val split done? Have you checked the frequency distributions of the (normalized) variables? Have you considered log transform for non Gaussian variables? How many time steps are included in each sample?

- Actually, 'array' is not necessary in this sentence. To avoid confusion, it is removed in the revised manuscript. For input variable, hourly measurements were used.
- The data were split for train, validation, and test, as shown in Figure 3: Data obtained during May-June in 2016 and in 2019 were used for train, June 2018 were used for validation, and April 2019 were for test. The

number of train, validation and test data are 1122, 381, and 133, respectively, which is stated in chapter 2.4.

- The input data were normalized using min-max scaling method, of which frequency distributions are presented below. Other normalizing method can change the distribution of data set, therefore we used min-max scaling method in this study to preserve its original distribution.
- Line 151: As input variables, hourly measurements of chemical and meteorological parameters are used,
- Line 185-187: 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.



Section 2.3: there is a lot of information missing from the network description: how many nodes per layer? What is the learning rate? How many epochs were trained? Did the learning rate change during training? Did you try out different numbers of layers and nodes per layer to determine the optimum model? Did you perform a hyperparameter search? Also, what exactly is the input data and what exactly is the target output? Loss function... Those things are standard in the machine learning literature and should be adhered to. I see some of this information appears in the figures and the following section (varying the number of nodes), but this belongs in the model description text.

- The detail information of network are written in chapter 2.3 and chapter 2.4 of paper (e.g. nodes per layer, learning rate, epochs, ... and etc.). We performed the hyperparameter test to decide the number of hidden layer and nodes, from 1 to 10, and from 1 to 100, respectively. For a simple

structure, the hyperparameters that may not strongly affect the model performance were not seriously searched. The node number of each hidden layer remained the same with the fixed learning rate. The information on hyperparameters including activation function, loss function, and optimizer, and data are stated in chapter 2.3 and chapter 2.4, as well.

l.136: if June 2018 has been used in the training already, then this month is not an ondependent test dataset any more.

- The data from June 2018 were not used for training. For training, the data from May ~ June in 2016 and 2019 were used.

l.154: does this mean that you always used the same number of modes in each layer? And you did not try to reduce the number of layers? 1600 samples appears rather small for a network with 5 layers.

- In previous study, the HONO simulation using a 1-hidden layer model with ~300 x 8 data resulted in the correlation coefficient of ~ 0.7 (Gil et al., 2021). In addition, the highest IOA and lowest MAE were observed for 5 hidden layers when performance test was conducted for 1, 5, and 10 hidden layers (Gil et al., 2020). Based on this result, the node number was searched with 5 hidden layers through k-fold cross validation. The node number searching test was conducted using the same number of nodes in each layer due to the limitation of computational resources.

l.160: I don't understand this. First you train the network for 2016 to 2019, then you run it again to obtain HONO results? You already have them from the training.(?)

- In this study, the amount of measurement data was not large enough to conduct the full train, validation, and test processes. Therefore, we adopted k-fold cross validation, which was used in other machine learning study for HONO (e.g., Cui et al., 2021). In addition, the traditional validation and test were conducted using the data obtained in June 2018 and April 2019 April. These processes of train, validation, and test are described in section 2.4.

- Line 186-188: 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.

- Line 223-239: 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 NO2 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.

l.167: I doubt that the inability fo the model to capture minima and maxima is due to the limited amount of data. This is a general aspect of regression models and extensively discussed in Kleinert et al (2021): https://doi.org/10.5194/gmd-14-1-2021

- As shown in frequency distribution of input and output variables, the number of high HONO cases are much less than those of low concentrations. Therefore, the ability of model to capture minima and

maxima will be improved with the large amount of data. We hope that recent observations will be incorporated into the model to improve the results.

l.205 and following: this discussion of atmospheric chemsitry doesn't belong into a section describing the application of the model. Is this supposed to be a general discussion section, comparing RND to other (CTM) models?

- As HONO is not properly simulated in general CTM models, their performance could be improved with HONO provided by the RND model. In this section, an example is presented, highlighting the contribution of the RND model rather than an introduction to its practical application.

l.235 Finally, here is a list of the input variables. But is has not been discussed, which variable has which influence on the results. I have a suspicion that the network really makes use only of 3 or 4 of the 9 variables it is given. See Kleinert et al. (2021) for a way how this can be tested with bootstrapping.

- The bootstrap test similar to Kleinert et al. (2021) was conducted by setting each variables to zero with keeping other values and the results were compared with measurements. Among nine input variables, NO2 was found to have the most significant influence on HONO concentration, followed by RH, temperature, and solar zenith angle (Table S1 2). This result is in good agreement with our previous study (Gil et al., 2021) and added to the text and supplementary.
- Line 240-252: 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, NO2 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.

Table S1 The result of bootstrap test using model validation data. The higher errors imply the higher degree of influence.

Variables (X)	MAE	RMSE
-	0.28	0.38
O_3	0.29	0.39
NO_2	0.59	0.85
CO	0.37	0.52
SO_2	0.34	0.46
SZA	0.41	0.60
T	0.52	0.68
RH	0.52	0.72
WS	0.34	0.48
WD	0.29	0.39

l.250/251: the ML model doesn't gain any physical understanding of the HONO chemistry, so it cannot be used to test the existing knowledge. You could use such a tool to forecast HONO levels, for example to determine if it might be worthwhile conducting HONO measurements at a specific location or during a specific time period. You may also be able to use the tool in the context of quality controlling the measurements: any strong disagreement would raise a warning that measurements should be checked with extra care. Also, you can of course use it to estimate HONO concentrations when these were not measured in order to then perform 0D model runs, as you show in Figure 8. And in this light, I would agree with the statement that RND is a "supplementary tool".

- Thank you for sharing ideas. The detail application of the RND is added to the revised manuscript.

- Line 310-313: 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.

l.262: please provide an explicit URL here (you can still add the reference)

- We update the DOI of reference (Gil, J.: RNDv1.0 and example, https://doi.org/10.5281/zenodo.5540180, in, Zenodo, 2021)

Technical corrections:

l.55: related to the comment on l.43: you presume that the reader is familiar with the basics of HONO chemistry, but this cannot be taken for granted.

- More detailed HONO chemistry is added to the introduction (Please seed the response to l.43)

l.30 play instead of plays

- It is corrected (l.46)

l.34 and *it* determines...

- It is corrected (l.50)

Citation: https://doi.org/10.5194/gmd-2021-347-RC1

Reference in answers

Gil, J., Kim, J., Lee, M., Lee, G., Ahn, J., Lee, D. S., Jung, J., Cho, S., Whitehill, A., Szykman, J., and Lee, J.: Characteristics of HONO and its impact on O3 formation in the Seoul Metropolitan Area during the Korea-US Air Quality study, Atmospheric Environment, 2021, https://doi.org/10.1016/j.atmosenv.2020.118182., 2021.

Pinto, J., Dibb, J., Lee, B., Rappenglück, B., Wood, E., Levy, M., Zhang, R. Y., Lefer, B., Ren, X.

R., and Stutz, J.: Intercomparison of field measurements of nitrous acid (HONO) during the SHARP campaign, Journal of Geophysical Research: Atmospheres, 119, 5583-5601, 2014.

Cui, L., and Wang, S.: Mapping the daily nitrous acid (HONO) concentrations across China during 2006-2017 through ensemble machine-learning algorithm, Science of The Total Environment, 147325, 2021.

Kleinert, F., Leufen, L. H., and Schultz, M. G.: IntelliO3-ts v1. 0: a neural network approach to predict near-surface ozone concentrations in Germany, Geoscientific Model Development, 14, 1-25, 2021.

0. In this paper, deep neural network based model is used to calculate nitrous acid (HONO) mixing ratios based on the analysis using HONO measurement data from Seoul between 2016 and 2019. Since I am not an expert in atmospheric sciences, but in data and computer science, I will in my review focus on the computational method used and its validity based on the size and type of the data.

- Thank you for your constructive comments and helpful advice. The point-by-point responses are given below, along with relevant parts of the revised manuscript, where all changes are marked in blue.

1. The paper is generally well written and takes action to document the use of the suggested model. The citation to code availability is missing DOI (and one has to go over to Zenodo to locate the code)

- The DOI of reference is updated (Gil, J.: RNDv1.0 and example, https://doi.org/10.5281/zenodo.5540180, in, Zenodo, 2021)

2. The approach taken is motivated by the success of deep learning based methods in various areas. However, here (as often elsewhere) it is not taken into account, that deep learning is most useful in situations in which there are massive amounts of training data — which is not the case here. There are nine input features and there are 1636 data items (1122 for training and 514 for validation). Hence, the data is not really massive and because the amount of interactions is limited (only nine input variables), its is quite likely that more traditional machine learning methods would work well (e.g., ordinary linear regression could be used to provide a baseline (and could even suffice), then one could see how e.g., support vector machine or random forest would work). In the paper, the use of deep neural networks is argued by them being more useful than traditional models, because they are able to handle large amounts of data. For the data used, there is no reason to assume that it could not be handled using also some of the traditional methods, in particular, when the data is small, more complicated models are quite prone to overfitting.

Suggestion for improvement 1: Test different ML learning models to be able to evaluate properly the usability of the suggested model.

- You are absolutely right. In general, the performance of deep learning (DL) is better than or at least similar to traditional machine learning (ML) such as support vector machine or random forest (Sumathi et al., 2020; Baek et al., 2021). This advantage would be greater with larger data set and even small data set can benefit from it (Dang et al., 2020). DL is also known to be better than general liner regression for data in non-linear relationship.

  The test result of RNDv1.0 demonstrates that it reasonably represents ambient HONO levels and captures well the averaged variation. In comparison, it tends to underestimate high concentrations. This is a weakness of our model but indicates that our model does not overfit the training dataset.

  In the revised manuscript, introduction is fully revised with background information on HONO and the application of DNN to HONO simulation.

  Line 85-104: 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 O3 or PM2.5. 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.

3. My second concern is the feature selection or the lack of it. The model blindly uses the nine input variables from the data. This kind of "taking an ML model off-the-shelf" very rarely produces the best possible results and can seriously affect the performance of the model. In addition to feature selection, it might be also possible to compute some surrogate features, e.g., provide information about dependencies in the modelling domain, reducing the need for the ML models to explicitly model these dependencies.

Suggestion for improvement 2: Use feature selection (for all the models) to search for a best possible set of input features.

- The OH produced from HONO photolysis will fuel the photochemical formation of O3 and PM2.5, which are target species of 0-dimensional photochemical models and chemical transport models (CTM). It is demonstrated in section 3 that the presence of HONO has a significant contribution to the performance of photochemical model.

  In this regard, the purpose of this study is to construct a model for estimating the HONO mixing ratio using atmospheric variables that are continuously and routinely measured, but not to improve the performance of model in which the accuracy matters. We hope that our recent observations will be incorporated into the RND model, and the model will be able to provide robust HONO concentrations for operational forecasting models in the future.

  In a previous study, we built a simple Neural Network model that estimated HONO mixing ratio, and we know that selecting the appropriate variables can increase the accuracy of the model (Gil et al., 2021). In this study, we aim to construct a kind of universal and cheap model to estimate HONO concentration in urban areas using atmospheric variables provided through measurement networks. These input variables that

were used for model construction did not show any meaningful correlations (Figure S2)

In addition, bootstrap test similar to what was done in Kleinert et al. (2021), was conducted by setting each variable to zero with keeping other values and the results were compared with measurements. Among nine input variables, NO2 was found to have the most significant influence on HONO concentration, followed by RH, temperature, and solar zenith angle (Table S2). This result is in good agreement with our previous study (Gil et al., 2021), implying that the input feature used for the model are suitable for estimating HONO concentrations.

In the revised manuscript, the detailed feature selection process is stated in Section1 and Section2.

- Line 105-107: 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.
- Line 151-154: As input variables, hourly measurements of chemical and meteorological parameters are used, including the mixing ratios of O3, NO2, CO, and SO2, 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.
- Line 241-253: 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, NO2 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 of 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.

4. Finally, the testing of the model using data from April 2019, shows some of the limitations of the developed model. It seems that there is an occurrence of concept drift (when the distribution of data changes, the model does not work well anymore). Also, the error might increase due to overfitting of the model. This aspect should be studied further, in particular it would be important to be able to provide the region in which the model's accuracy is on an acceptable level. There is a rich body of literature in detecting concept drift (for a survey, e.g., see Zliobaite I., Pechenizkiy M., Gama J. (2016) An Overview of Concept Drift Applications. In: Japkowicz N., Stefanowski J. (eds) Big Data Analysis: New Algorithms for a New Society. Studies in Big Data, vol 16. Springer, Cham. https://doi.org/10.1007/978-3-319-26989-4_4).

Suggestion for improvement 3: Analyse the region in which the proposed model can be expected to work, at least provide some discussion on the effect of overfitting and concept drift and how theses affect the usability of the model.

- Atmospheric parameters including meteorological factors and chemical constituents show clear diurnal variations, especially in urban areas with high anthropogenic emissions. For example, NO2 reached the maximum during the morning rush hour, decreased down to the minimum in the afternoon, and increased at nighttime. This type of variation remained nearly constant through the year with changes in seasonal amplitude depending on emissions and meteorological factors determining the dilution and transport of air pollutants. The variation in O3 is just opposite to NO2.
- Our model was constructed for urban applications. When the model was tested against data obtained April, model uncertainty was increased. Although our model was trained and validated with data obtained during May-June, the variations in input variables for test period were similar to those of train-validation periods. Considering the result of previous study about HONO formation mechanism, the increased model uncertainty could be due to some factors that were not constrained in the model such as aerosol surface areas.

Therefore, it is quite likely that the increased model uncertainty is not associated with the occurrence of concept drift.

Based on these observations, I would reject the paper in its current form, with the encouragement to resubmit, taking the suggestions for improvement into account.

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