Ozone formation sensitivity study using machine learning coupled with the reactivity of VOC species

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Abstract

The formation of ground-level ozone (O_3) is dependent on both atmospheric chemical processes and meteorological factors. In this study, a random forest (RF) model coupled with the reactivity of volatile organic compound (VOC) species was used to investigate the O_3 formation sensitivity in Beijing, China, from 2014 to 2016, and evaluate the relative importance (RI) of chemical and meteorological factors to O_3 formation. The results showed that the O_3 prediction performance using concentrations of measured/initial VOC species (R^2 = 0.82/0.81) was better than that using total VOCs (TVOCs) concentrations (R^2 = 0.77). Meanwhile, the RIs of initial VOC species correlated well with their O_3 formation potentials (OFPs), which indicate that the model results can be partially explained by the maximum incremental reactivity (MIR) method. O_3 formation presented a negative response to nitrogen oxides (NO_x) and relative humidity (RH), and a positive response to temperature (T), solar radiation (SR) and VOCs. The O_3 isopleth calculated by the RF model were generally comparable with those calculated by the box model. O_3 formation shifted from a VOC-limited regime to a transition regime from 2014 to 2016. This study demonstrates that the RF model coupled with the initial concentrations of VOC species could provide an accurate, flexible, and computationally efficient approach for O_3 sensitivity analysis.

1. Introduction

Ground-level ozone (O_3) pollution, which can cause adverse human health effects such as cardiovascular and respiratory diseases, has received increasing attention in recent decades (Cohen et al., 2017). Oxidation of volatile organic compounds (VOCs) will produce peroxyl radicals (RO_2) and hydroperoxyl radicals (HO_2). The RO_2/HO_2 can accelerate the conversion from NO to NO_2, subsequently, formation of O_3 by photolysis of NO_2 in the presence of O_2 (Wang et al., 2017a). The production and loss of RO_2 and HO_2 are highly dependent on the concentration ratio of VOCs and NO_x in the atmosphere. Hence, atmospheric O_3 concentrations or production rates show a nonlinear relationship with VOCs and NO_x. Moreover, the O_3-VOC-NO_x sensitivity is readily influenced by VOC species (Tan et al., 2018), meteorological parameters (Liu et al., 2020a; Liu et al., 2020), and even atmospheric particulate matter (Li et al., 2019), thus, exhibiting high temporal and spatial variability. Therefore, it is urgent to develop an accurate and highly efficient method for timely assessing the sensitivity regime of O_3 production and evaluating the effectiveness of a potential measure on O_3 pollution control. The sensitivity of O_3 formation can usually be analysed using observed indicators, such as ozone production efficiency (OPE, $\Delta O_3/\Delta NOz$) (Wang et al., 2010; Lin et al., 2011), HCHO/NO_y (Martin et al., 2004), and H_2O_2/NOz (or H_2O_2/HNO_3) (Sillman 1995; Hammer et al., 2002; Wang et al., 2017a), observation-based model (OBM) (Vélez-Pereira et al., 2021) and chemical transport models including community multiscale air quality (CMAQ) (Djalalova et al., 2015) and Weather

Research and Forecasting with Chemistry (WRF-Chem) model (Wang et al., 2020a).

The observed indicators can be utilized to quickly diagnose the sensitivity regime of O_3 production. However, the accuracy is sensitive to the precision of tracer measurements. OBMs combine *in-situ* field observations, remote sensing measurements and chemical box models, which are built on widely-used chemistry mechanisms (e.g., MCM, Carbon Bond, RACM or SAPRC), and applied to the observed atmospheric conditions to simulate the *in-situ* O_3 production rate (Mo et al., 2018). The sensitivity of O_3 production to various O_3 precursors, including NO_x and VOCs can be diagnosed based on the empirical kinetic modeling approach (EKMA) or quantitatively assessed with the relative incremental reactivity (RIR). Chemical transport models, which are driven by meteorological dynamics and incorporated with the emissions of pollutants and the complex atmospheric chemical mechanism, provide a powerful tool for simulating various atmospheric processes, including spatial distribution, regional transport *vs.* local formation, source apportionment and production rates of pollutants and so on (Sayeed et al., 2021). At present, OBMs are widely used to investigate O_3 formation sensitivity in China. Previous studies indicated that O_3 formation in urban areas of China is located in a VOC-limited or a transition regime and varies with time and location (Ou et al., 2016; Wang et al., 2017a; Zhan et al., 2021). Although both OBMs and chemical transport models can assess the sensitivity of O_3 production and predict the O_3 pollution level in a scenario of control measures, the calculation accuracy is affected by the uncertainty of input parameters

(Tang et al., 2011; Yang et al., 2021b). Thus, they are mostly applied to sampling cases with a short time span (days or weeks) (Xue et al., 2014; Ou et al., 2016).

Compared to traditional methods, machine learning (ML) is able to capture the main factors affecting atmospheric O_3 formation in a timely manner with great flexibility (without the constraints of time and space) and high computational efficiency (Wang et al., 2020c; Grange et al., 2021; Yang et al., 2021a). Although attentions should be paid to the robustness of machine learning because it depends on the input dataset (observations or outputs of chemical transport models), previous studies have demonstrated that cross-validation and data-normalization can well reduce the dependence of the model on input data and improve the robustness of the model (Wang et al., 2016; Wang et al., 2017b; Liu et al., 2021; Ma et al., 2021a). Thus, it is a promising alternative to account for the effects of meteorology on air pollutants and has been intensively used in atmospheric studies (Liu et al., 2020a; Hou et al., 2022).

Recently, ML based on convolutional neural network (CNN), random forest (RF) and artificial neural network (ANN) models have been applied in simulating atmospheric O_3 and shown good performance in O_3 prediction (Ma et al., 2020; Xing et al., 2020). For example, Ma et al. (2021a) simulated O_3 concentrations in the Beijing-Tianjin-Hebei (BTH) region from 2010-2017 using an RF model that considered meteorological variables and output variables from chemical transport models, and the correlation coefficient (R^2) between the observed and modelled O_3 concentrations was greater than 0.8. Liu et al. (2021) also reported a high accuracy (80.4%) for classifying

pollution levels of O_3 and fine particulate matter with aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) at 1464 monitoring sites in China using an RF model. Thus, the RF model has shown good performance in terms of prediction accuracy and computational efficiency (Wang et al., 2016; Wang et al., 2017b).

Although ML is widely used to understand air pollution, many ML studies have used total VOCs (TVOCs) to simulate O_3 formation and rarely considered the effect of VOC species on O_3 formation sensitivity (Feng et al., 2019; Liu et al., 2021; Ma et al., 2021a). Thus, they were unable to identify the chemical reactivity of a single species to O_3 formation, which may lead to underestimations or even misunderstandings of the role of VOCs in O_3 formation because the same concentration of TVOCs with different compositions may lead to different OPEs. In addition, VOCs react with OH radicals during atmospheric transport, which is the most important sink of VOCs (Carlo et al., 2004; Liu et al., 2020b). Makar et al. (1999) reported that the isoprene emissions were underestimated by up to 40% if the OH oxidation is not considered. Other studies indicated that the initial concentrations of VOCs, which account for the photochemical loss of VOCs during transport, were more representative of pollution levels in the sampling area than the observed VOCs (Yuan et al., 2013; Zhan et al., 2021). However, whether the ML model can identify the connection between the reactivity of VOC species and O_3 formation sensitivity has not been clarified.

It should be noted that physical interpretability of the results is an important question when ML models are applied in atmospheric studies (Hou et al., 2022).

However, explanations of ML results (e.g., RI) are somewhat vague because ML is a "black-box" model from the point view of chemical mechanism (Hou et al., 2022; Taoufik et al., 2022). In this study, we used the RF model to evaluate the prediction performance of atmospheric O_3 using the TVOCs, measured VOC species and photochemical initial concentration (PIC) of VOC species, which is calculated based on the photochemical-age approach (Shao et al., 2011). We compared the relative importance (RI) of the precursors (VOC species, NO_x, $PM_{2.5}$, CO) and the meteorological parameters (temperature, solar radiation, relative humidity, wind speed and direction) on O_3 formation in the summer of Beijing from 2014 to 2016. We also discussed the possibility of connecting the RIs of VOCs with their OFPs and the changes in O_3-VOC-NO_x sensitivity based on the RF model from 2014 to 2016. Our study indicates that the RF model combined with initial concentrations of VOC species can simulate O_3 concentrations well and provides a flexible and efficient tool for O_3 modelling in a near real-time way.

2. Methods

2.1 Sampling site and data

The sampling site (40.04°N, 116.42°E) is located at the campus of Chinese Research Academy of Environmental Sciences and was described in our previous work (Zhang et al., 2021). Briefly, the station is located two kilometers from the north 4^{th} ring road and surrounded by a mixed residential and commercial area. The concentrations of VOCs, NO_x, CO, O_3 and $PM_{2.5}$ were measured at 8 m above ground level at this location. Meteorological parameters, including temperature (T), relative humidity (RH),

wind speed and direction (WS&WD), solar radiation (SR), were monitored at 15 m above ground level. VOCs were measured by an online commercial instrument (GC-866, Chromatotec, France), which consisted of two independent analysers for detecting C_2-C_6 and C_6-C_{12} hydrocarbon components. More details about the observations can be found in the Supplemental Materials (S1). The calculation of initial VOCs and sensitivity tests can be found in the Supplemental Materials (S2).

2.2 Random forest model

The random forest (RF) is a type of ensemble decision tree that can be used for classification and regression (Breiman 2001). During the training process, the model creates a large number of different decision trees with different sample sets at each node, and then averages the results of all decision trees as its final results (Breiman 2001). To avoid over-fitting, we trained the random forest model using cross-validation for the normalized data, which can improve the robustness of the model. Briefly, we randomly divided the normalized data into 12 subsets, then alternately took one subset as testing data along with the rest as training data. By doing this, every data point has an equal chance being trained and tested. The length of the input data from 2014 to 2016 were 1190, 1062 and 872 rows, respectively, in which different types of VOCs, NOx, CO, $PM_{2.5}$ and meteorological parameters (including temperature, relative humidity, solar radiation, wind speed and direction) were used as input variables and O_3 as output variables. The mean values (±standard deviation) of input/output parameters are shown in Table S1. Approximately one-third of the samples are excluded from the sample,

158 when the decision tree is built and used to calculate the out-of-bag data error. Hence,
159 RF can evaluate the RI of variables via the changes in out-of-bag (OOB) data error
160 (Svetnik et al., 2003),

161 $$RI_i = \sum (errOOB2_i - errOOB1_i) / N \quad (1)$$

162 where N represents the number of decision trees, and errOOB1 and errOOB2 represent
163 the out-of-bag data error of feature *i* before and after randomly permuting the
164 observation, respectively. The RI_i used to evaluate the importance and sensitivity of
165 feature *i* to O_3 formation in this study. More details about workflow of RF model and
166 the hyperparameter tuning can be found in the Text S3. The optimized parameters are
167 shown in Table S2. To verify the stability of the model, we performed a significance
168 test on the model results. The results showed that there was no significant difference
169 among the different tests (P>0.05, R^2>0.98).

170 When plotting the O_3 formation sensitivity curves, we made a virtual matrix of
171 inputs by varying the concentrations of NOx and VOCs from 0.9 to 1.1 times (with a
172 step of 0.01) of their mean values while keeping all other inputs unchanged (i.e., the
173 mean values). Then, the new matrix was used as testing data, while all the measured
174 data were taken as training data. Thus, the testing data should represent the mean
175 sensitivity regime of O_3 in Beijing, while the training data actually covered all the
176 sensitivity regimes of O_3 formation to guarantee a sufficient coverage in the NOx-
177 limited regime for the RF model simulations. The EKMA curves were plotted using the
178 daily maximum 8-h (MDA8) O_3. More details can be found in the SI.

3. Results and discussion

3.1 Overview of air pollutants and meteorological conditions

Figure 1 shows the time series of air pollutants and meteorological parameters during the observations from 2014 to 2016. In 2014, 2015 and 2016, the wind direction was dominated by northwest winds (Figure S1), with mean wind speeds of 3.1 ± 2.7 m s^{-1}, 2.3 ± 2.2 m s^{-1}, and 1.3 ± 1.2 m s^{-1}, respectively, and the mean daytime temperature were 22.3 ± 5.8, 23.9 ± 5.0 and 24.0 ± 4.4 °C, respectively. The average value of SR decreased from 162.9 to 150.8 W m^{-2} during the observation period. As shown in Figure 1F-G, in 2014, 2015 and 2016, the mean VOC concentrations were 20.3 ± 10.9, 15.8 ± 8.3 and 12.1 ± 7.7 ppbv, respectively, while the mean initial VOC concentrations were 28.1 ± 25.7, 27.2 ± 32.6 and 16.4 ± 16.1 ppbv, respectively. The calculation of initial VOCs and sensitivity tests can be found in the Supplemental Materials (S2). Both the measured VOCs and initial VOCs showed a decline along with a decrease in $PM_{2.5}$ concentration from 67.2 ± 53.5 to 61.1 ± 48.6 μg m^{-3} due to the Air Pollution Prevention and Control Action Plan in China (Zhao et al., 2021). However, O_3 concentrations showed a slight downward trend from 44.3 ± 32.4 to 42.7 ± 27.9 ppbv from 2014 to 2015 and then reach to 44.0 ± 29.6 ppbv in 2016. A slight upward trend was observed for NO_x concentrations (Figure S2). As shown in Figure 1F-G, the concentrations of four types (alkanes, alkenes, alkynes, and aromatics) of VOCs showed significant differences from 2014 to 2016 due to the variations in emission sources (Zhang et al., 2021). In addition to VOC species, the variations in other parameters, such as

meteorological conditions and $PM_{2.5}$, should have a complex influence on O_3-VOC-NO_x sensitivity (Li et al., 2019; Ma et al., 2021b).

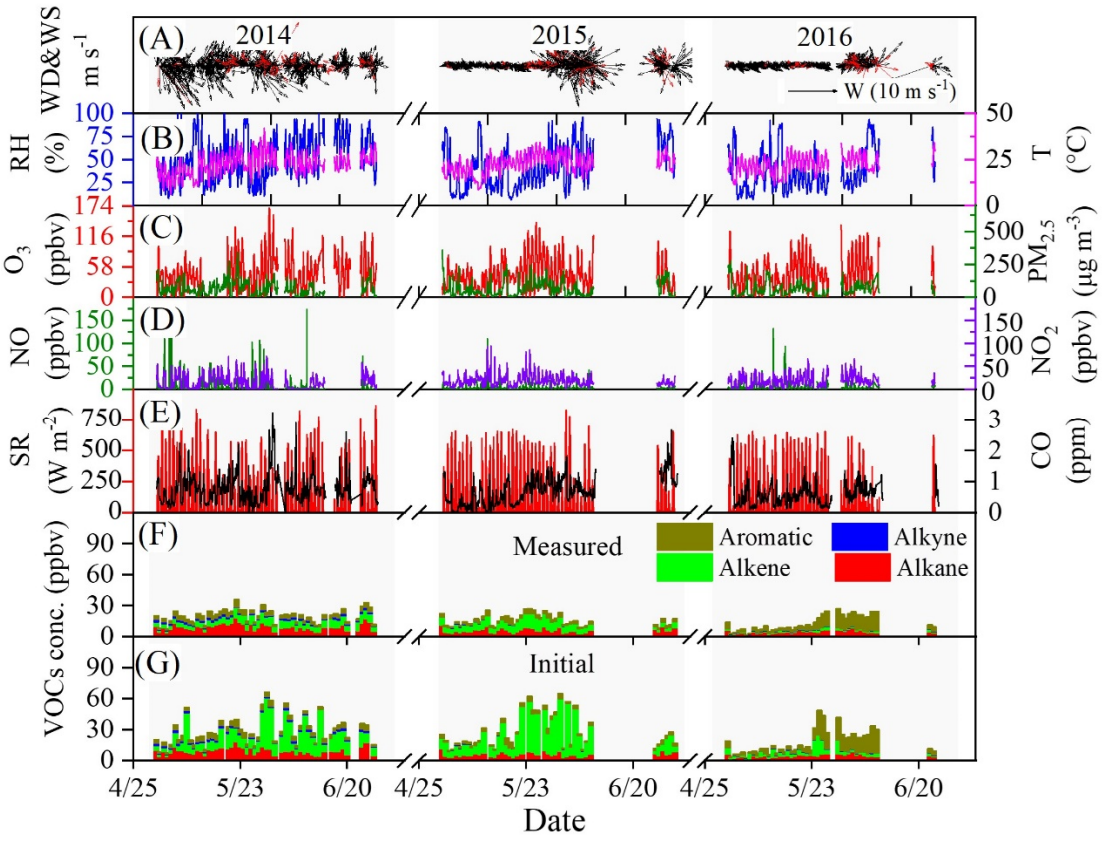
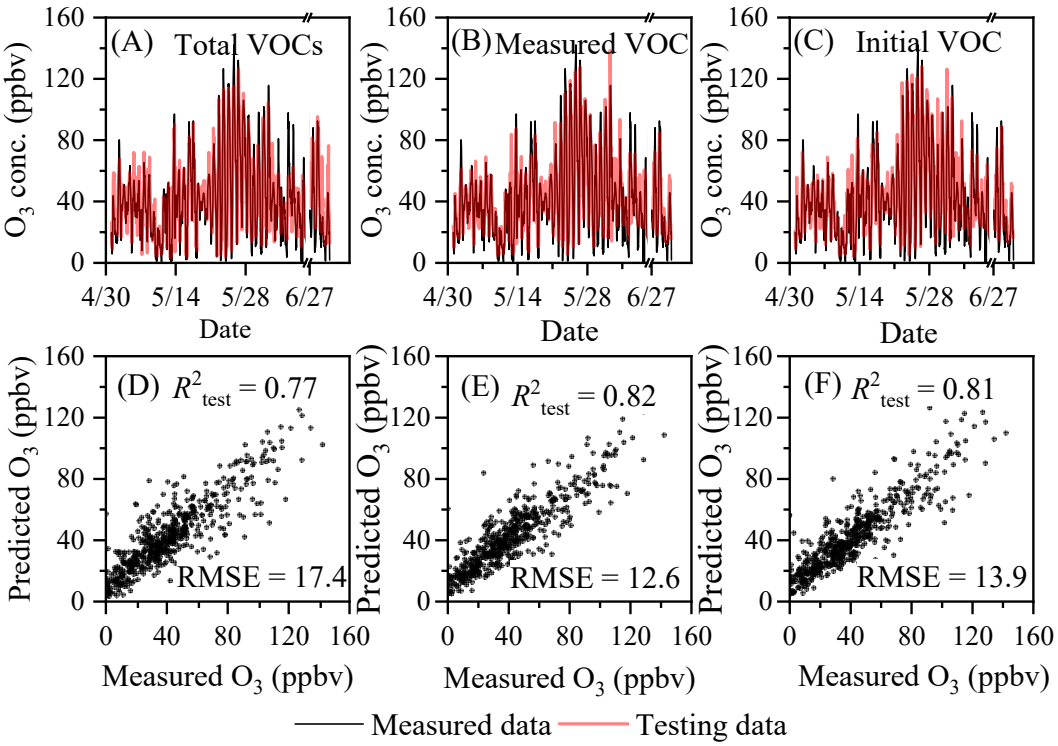


Figure 1. Time series of air pollutants and meteorological parameters during observations in Beijing. (In A, the red arrows represent the O_3 concentration exceed 74.6 ppbv according to the national ambient air quality standard.)

3.2 Prediction performance of the model.

To build a robust model, we evaluated the prediction performance of the RF model for the ambient O_3 simulation. Figure 2 shows the O_3 prediction performance in 2015 when chemical species (including VOCs, NO_x, $PM_{2.5}$, CO) and meteorological factors (i.e., WS, WD, SR, T and RH) were used as inputs in the RF model. The prediction performance of RF model for 2014 and 2016 is shown in Figures S3 and S4 respectively.

The details of the modelling and input parameters are shown in Table S2. Figure 2A-C shows the time series of the measured and modelled O_3 concentrations, which were simulated using the TVOCs, measured VOC species and initial VOC species as part input variables along with the same set of other parameters. The correlation coefficients (R^2) of the training data were 0.77, 0.82 and 0.81 for the TVOCs, measured VOC species and initial VOC species, respectively. The corresponding root mean squared errors (RMSEs) for the predicted O_3 concentrations were 17.4, 12.6 and 13.9. Figure 2D-F shows the prediction performance of the testing dataset under these three circumstances. When the TVOCs were split into measured or initial VOC species, the R^2 increased obviously as the number of data features increased. Therefore, the VOC composition has a significant influence on O_3 prediction using the RF model. In previous studies using TVOCs, the influence of VOC composition was neglected (Liu et al., 2021; Ma et al., 2021a). Our results indicate that the RF model can accurately predict O_3 concentrations when the concentrations of measured/initial VOC species are considered.



227

228 **Figure 2.** Comparison of the predicted and measured O_3 concentrations in Beijing in
229 the summer of 2015. (A and D: TVOC concentrations; B and E: measured
230 concentrations of VOC species; C and F: initial concentrations of VOC species)

231 It should be pointed out that if the training dataset does not have sufficient
232 coverage in the NOx-limited regime, then the trained algorithm essentially attempts to
233 extrapolate in that regime, which is prone to overtraining. To avoid such overtraining,
234 a 12-fold cross-validation by randomly dividing the observation data in each day into
235 12 subsets and alternately taking one subset as testing data and the rest as training data
236 ensures that each data point has an equal chance of being trained and tested. The curves
237 of the predicted O_3 concentrations in Figure 2 were spliced using the testing datasets in
238 all runs. Thus, our results actually covered all the sensitivity regimes of O_3 formation.
239 This means that the model is robust

240 **3.3 Relative importance of major factors**

Figure 3A shows the RIs of different ambient factors, including chemical and meteorological variables on O_3 formation. The difference in the RIs is also compared using the TVOCs and the VOC species as inputs. Chemical factors (including VOC species, NO_x, $PM_{2.5}$ and CO) accounted for 79.1% of the contribution to O_3 production in the summer of 2016. Meanwhile, VOC species accounted for approximately 63.4% of O_3 production while the RIs using TVOC concentrations accounted for only 2.1%. Ma et al. (2021b) analysed the contribution of meteorological conditions and chemical factors to O_3 formation on the North China Plain (NCP) using the CMAQ model in combination with process analysis and found that chemical factors dominate O_3 formation in summer. Using probability theory, Ueno et al. (2019) also found that VOCs/NO_x dominate O_3 production compared to meteorological variables. Thus, our results are similar to those of previous studies based on chemical models (Ueno et al., 2019), which demonstrates that the RF model can reflect the contribution of VOC species to O_3 production even if the observed VOC species are used.

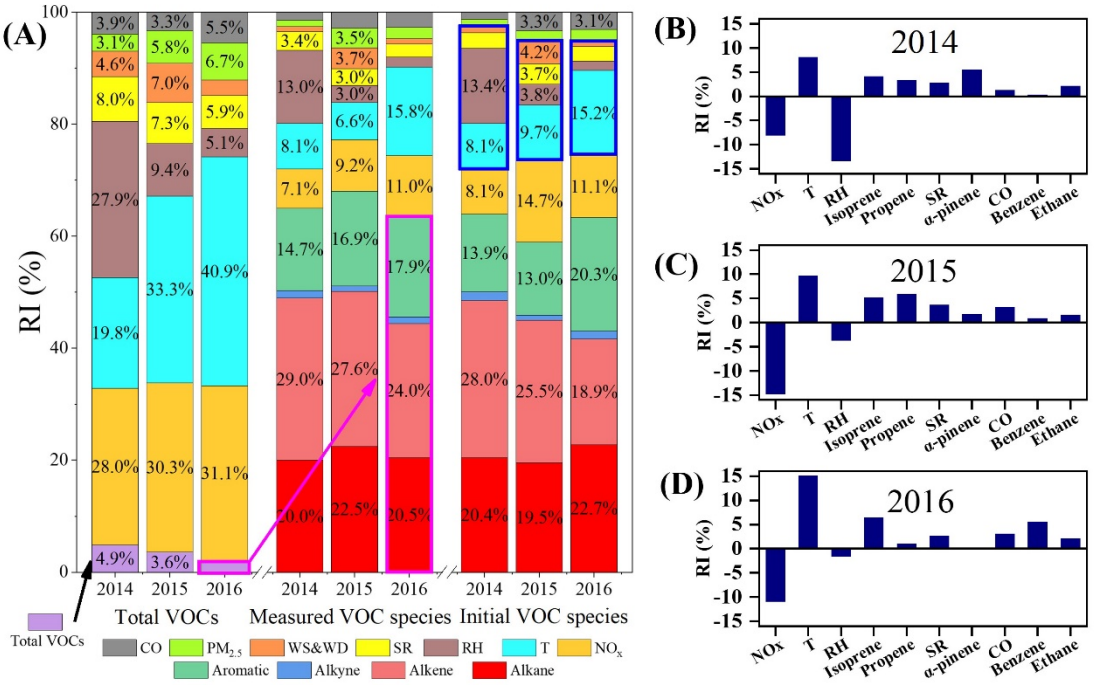


Figure 3. Percentage of RI for O_3 precursors and meteorological parameters (A) and the top 10 factors with high values of RI in 2014-2016 (B-D: using initial concentrations of VOC species).

Here, we compared the RIs of VOCs calculated using the initial VOC species and the observed VOC species with the O_3 formation potentials (OFPs). The OFPs were calculated by the maximum incremental reactivity (MIR) method (Carter 2010). As shown in Figure S5, the RIs showed good correlations with the OFP. Interestingly, the initial concentrations of VOC species improved the correlation coefficients between the RIs and OFPs. Furthermore, we calculated the RIs and OFPs of different species using the observed data during the campaign study in Daxing District in the summer of 2019 (Zhan et al., 2021), and a stronger correlation was observed between the RIs of the initial VOC species and the OFPs (Figure S6). These results indicate that the RIs of the initial VOCs species in the ML model should partially reflect the chemical reactivity of

VOCs to produce O_3 in the atmosphere.

Although the RIs calculated using the initial VOC species slightly changed compared to those calculated using the observed VOCs (Table S3), VOCs still dominated O_3 formation (Figure 3A). For example, the initial VOCs dominated O_3 production in 2014, 2015, and 2016, with RI values of 64.0, 59.0 and 63.3% respectively. Li et al. (2020a) used a multiple linear regression (MLR) model to study the contribution of anthropogenic and meteorological factors to O_3 formation in China from 2013-2019 and found that meteorological factors accounted for 36.8% and anthropogenic factors accounted for 63.2%, which is similar to our results. Figure 3B-D shows the top 10 factors having a strongly influence on O_3 production. Interestingly, NO_x and RH showed negative responses to O_3 formation, while other variables, including T, SR, CO and all of the VOCs, showed positive responses. Thus, a decrease in NO_x or RH will lead to an increase in O_3 concentration while a decrease in T, SR, CO and VOCs will lead to a decrease in O_3 concentration. Although O_3 formation is highly related to the photolysis of NO_2, a previous study demonstrated that it is VOC-limited in summer in Beijing (Zhan et al., 2021). This finding is consistent with the observed negative response of O_3 to NO_x in this work. High RH usually coincides with low surface O_3 concentrations in field observations, which can be ascribed to the inhibition of O_3 formation by the transfer of NO_2/ONO_2-containing products into the particle phase and the promotion of dry deposition of O_3 on the surface (Kavassalis et al., 2017; Yu 2019). In addition, it has been shown that RH is negatively related to the

rate constant of HONO formation (Hu et al., 2011). Thus, RH might also affect the O_3 formation by influencing atmospheric OH radicals from photolysis of HONO. It should be noted that the negative response of ozone to RH might also be resulted from the dependence of RH on other parameters/conditions, such as SR. However, RH and SR showed a bad correlation ($r < 0.1$). We further tested the dependence of the RI on RH and SR with or without the counterpart as input. The stable RI values (Table S4) mean that RH and SR are independent from each other. These previous works can well explain the observed negative response of O_3 to RH in Figure 3B-D. Previous studies have observed a positive correlation between the O_3 concentration and T or SR (Steiner et al., 2010; Paraschiv et al., 2020; Li et al., 2021). Temperature can directly affect the chemical reaction rate of O_3 formation (Fu et al., 2015), and SR can promote the photolysis of NO_2 (Hu et al., 2017; Wang et al., 2020b), thus accelerating O_3 formation. As mentioned above, O_3 formation is VOC-limited in Beijing; thus, a positive response of O_3 concentration to VOCs is observed in Figure 3B. Interestingly, the RIs of isoprene showed an increasing trend from 2014 to 2016 because of the obvious reduction in anthropogenic VOCs (Figure S7) (Zhang et al., 2021). In the context of global warming, studies should focus on the factors that affect O_3 formation, including biogenic emissions, T and SR. Thus, additional efforts will be required to reduce anthropogenic pollutants in the future.

3.4 Ozone formation sensitivity

To further analyse the sensitivity of O_3 to VOCs and NO_x from 2014 to 2016, we

311 plotted sensitivity curves for O_3 generation using the RF model, and the results are
312 shown in Figure 4A-C. Moreover, EKMA curves in 2015 were also obtained using the
313 OBM (Figure 4D). As shown in Figure 4A-C, O_3 formation was sensitive to VOCs in
314 the summer of Beijing during our observations, which is consistent with previous
315 studies that used box models (Li et al., 2020b) and chemical transport models (Shao et
316 al., 2021). This result is also consistent with the RIs of VOCs or NO_x to O_3 formation
317 (Figure 3B-D). Interestingly, the O_3 formation sensitivity to VOCs decreases or
318 gradually shifts from the observed point to the transition regime from 2014 to 2016
319 (Figure 4A-C), which is similar to that reported by Zhang et al. (2021). These
320 phenomena can be ascribed to the increased relative importance of meteorological
321 factors, such as T, SR, and RH, for O_3 formation and the variation in anthropogenic
322 VOC emissions (Steiner et al., 2010; Ma et al., 2021b).

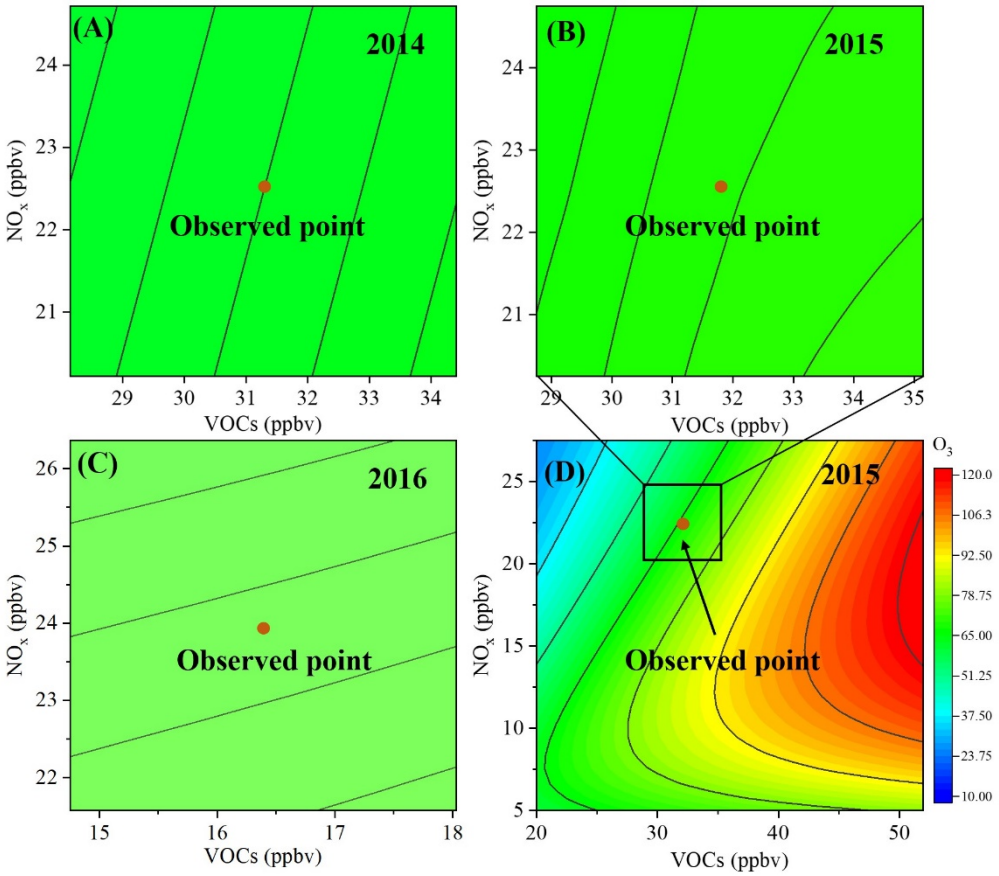


Figure 4. Ozone formation sensitivity curves from 2014-2016. (A, B, C: calculated by the RF model for 2014, 2015, and 2016, respectively. D: calculated by the OBM for 2015.)

We compared the relative error of simulated MDA8 O_3 calculated using the RF and OBM model in 2015, as shown in Figure S8. The mean relative error of simulated MDA8 O_3 between RF model and Box model was 15.6%. Hence, a combination of the RF model and initial VOCs species can accurately depict the sensitivity regime of O_3 formation, while the calculated RIs correlate well with the OFPs.

4. Conclusions

In summary, this work investigated O_3 formation sensitivity in the summer from 2014-2016 in Beijing using the RF model coupled with the reactivity of VOC species.

335 The results show that the prediction performance of O_3 by the RF model was
336 significantly improved when measured/initial VOC species were considered compared
337 to TVOCs. Furthermore, after the photochemical loss of VOC species during transport
338 was corrected, the RIs of the VOC species were well correlated with the OFPs of VOC
339 species calculated using the MIR method, thus indicating that the RIs in the ML model
340 reflect the chemical reactivity of VOCs. Meanwhile, both NO_x and highly reactive
341 species (such as isoprene, propene, benzene) played an important role in O_3 formation.
342 An increased contribution of temperature to O_3 production was observed, which
343 implied the importance of temperature to O_3 pollution in the context of global warming
344 conditions. Both the RF model and the box model results showed that O_3 formation was
345 sensitive to VOCs in Beijing, although the sensitivity regime shifted from VOC-limited
346 regime to a transition regime from 2014 to 2016. Due to the high computational
347 efficiency of ML, the O_3 formation sensitivity plotted by the RF model coupled with
348 the reactivity of VOC species can provide an accurate, flexible and efficient approach
349 for analysing O_3 sensitivity in a near real-time way.
350
351 **Code and data availability**
352 The datasets of VOCs and meteorology are available and will be provided by the
353 corresponding authors Yongchun Liu (liuyc@buct.edu.cn) and Hong Li
354 (lihong@craes.org.cn) upon request. The code can be seen in GitHub
355 (https://github.com/z-12/amt-2021-367.git). The solar radiation data are publicly

356 available via www.copernicus.eu/en.

357 **Supplement**

358 Supplementary information is available for this paper.

359 **Author contributions**

360 Junlei Zhan designed the idea and wrote this manuscript; Yongchun Liu and Hong Li
361 provided useful advice and revised the manuscript; Wei Ma performed box model
362 simulations; and Xin Zhang, Xuezhong Wang, Fang Bi, Yujie Zhang and Zhenhai Wu
363 conducted the campaign and compiled the data. All authors contributed to the
364 discussion of the results and writing of the manuscript.

365 **Competing interest**

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

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373

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