Impact Assessment of COVID-19 Lockdown on Vertical
Distributions of NO\textsubscript{2} and HCHO From MAX-DOAS
Observations and Machine Learning Models

Sanbao Zhang\textsuperscript{1} \textsuperscript{,} Shanshan Wang\textsuperscript{1,2} \textsuperscript{,} Ruibin Xue\textsuperscript{1} \textsuperscript{,} Jian Zhu\textsuperscript{1} \textsuperscript{,} Aimon Tanvir\textsuperscript{1} \textsuperscript{,} Danran Li\textsuperscript{1} \textsuperscript{,} and
Bin Zhou\textsuperscript{1,2,3} \textsuperscript{β}

\textsuperscript{1}Shanghai Key Laboratory of Atmospheric Particle Pollution and Prevention (LAP\textsuperscript{3}), Department of Environmental Science and Engineering, Fudan University, Shanghai, China, \textsuperscript{2}Institute of Eco-Chongming (IEC), Shanghai, China, \textsuperscript{3}Institute of Atmospheric Sciences, Fudan University, Shanghai, China

Abstract
Responses to the COVID-19 pandemic led to major reductions on air pollutant emissions in modern history. To date, there has been no comprehensive assessment for the impact of lockdowns on the vertical distributions of nitrogen dioxide (NO\textsubscript{2}) and formaldehyde (HCHO). Based on profiles from 0 to 2 km retrieved by Multi-Axis-Differential Optical Absorption Spectroscopy observation and a large volume of real-time data at a suburb site in Shanghai, China, four types of machine learning models were developed and compared, including multiple linear regression, support vector machine, bagged trees (BT), and artificial neural network. Ultimately BT model was employed to reproduce NO\textsubscript{2} and HCHO profiles with the best performance. Predictions with different meteorological and surface pollution scenarios were conducted from 2017 to 2019, for assessing the corresponding impacts on the changes of NO\textsubscript{2} and HCHO profiles during COVID-19 lockdown. The simulations illustrate that the NO\textsubscript{2} decreased in 2020 by 43.8\%, 45.5\%, and 44.6\%, relative to 2017, 2018, and 2019, respectively. For HCHO, the lockdown-induced situation presented the declines of 28.6\%, 32.1\%, and 10.9\%, respectively. In the comparisons of vertical distributions, NO\textsubscript{2} maintained decreasing at all altitudes, while HCHO decreased at low altitudes and increased at high altitudes. During COVID-19 lockdown, the reduction of NO\textsubscript{2} and HCHO from the variation of surface pollutants was dominated below 0.5 km, while the relevant meteorological factors played a more significant role above 0.5 km.

Plain Language Summary
This study evaluated the impact of COVID-19 lockdown on the vertical distributions of NO\textsubscript{2} and HCHO by developing the machine-learning (ML) air pollution prediction models. The ML model allows us to quantify the timing and magnitude influencing real-world air quality responses to surface pollution scenarios and meteorological factors. Compared with the simulations for vertical profiles in the normal scenarios, it can be found that NO\textsubscript{2} maintained decreasing at all altitudes, while HCHO decreased at low altitudes and increased at high altitudes. In addition, the lockdown-induced NO\textsubscript{2} and HCHO were mainly driven by meteorological factors and surface pollutants above and below 0.5 km, respectively.

1. Introduction
The photodissociation of nitrogen dioxide (NO\textsubscript{2}) and formaldehyde (HCHO) in the atmosphere is a process of great importance for atmospheric chemistry. Both take part in the photochemical formation of tropospheric ozone (O\textsubscript{3}) (Brunner et al., 1998), consequently leading to a strong influence on the Earth’s radiation budget. The dominant sources of NO\textsubscript{2} are derived from fossil fuel combustion, biomass burning, lightning, and oxidation of ammonia (Delmas et al., 1997; Logan, 1983). HCHO is known as one of most abundant volatile organic compounds (VOCs), performing a critical role in air quality and atmospheric photochemistry (Sinreich et al., 2007; Volkamer et al., 2005; Wittrock et al., 2006). HCHO can be directly emitted from fossil fuel combustion (Ho et al., 2012; Schauer et al., 2002), biomass burning (Lee et al., 1997), and vegetation (DiGiangi et al., 2011), while secondarily generated by the photooxidation of non-methane VOCs (NMVOCs) (Parrish, Law, et al., 2012).

The COVID-19 pandemic has resulted in unprecedented shutdown on traffic related emissions in megacities in China (Le et al., 2020; Liu et al., 2020). Many recent studies have qualified the deductions on NO\textsubscript{2} and HCHO driven by the COVID-19 lockdowns based on in situ and satellite observations, but they are limited to the results at ground or in the whole column atmosphere. Regarding importance of atmospheric turbulence in mixing layer, high-resolution vertical observations provide a more suitable basis for capturing the vertical transport of air...
pollution. Multi-AXis-Differential Optical Absorption Spectroscopy (MAX-DOAS) equipment is widely utilized for retrieving vertical profiles of aerosol extinction coefficient and trace gases from the spectras of scattered sunlight (Friess et al., 2011; Hendrick et al., 2014; Höninger et al., 2004; Wagner et al., 2004). The conventional observations of atmospheric trace gases, including NO₂, HCHO, nitrous acid (HONO), water vapor (H₂O), sulfur dioxide (SO₂), O₃, glyoxal (CHOCHO), and halogen oxides (e.g., BrO, OClO), have been conducted by MAX-DOAS research teams around the world.

Atmospheric chemical transport models have been widely used to decouple the response of air pollutant concentrations to the changes of emissions and meteorological conditions (Zhao et al., 2020). However, the dynamic analysis of air-quality impacts of sudden changes in emissions has been limited by the challenge in preparing a high-temporal-resolution emission inventory in a timely manner (Zhao et al., 2020). Machine learning (ML) is opening a path toward obtaining fresh insights into the atmospheric pollution research. It has been demonstrated as an alternative and reliable capacity to quantify the timing and magnitude influencing real-world air quality responses to emissions and meteorological factors (Grange et al., 2021; Vu et al., 2019; Y. Wang et al., 2020). Compared with common chemical transport modeling, the advantage of ML model lies in the greater flexibility in utilizing predictive variables and higher computational efficiency. Yang et al. (2021) employed the ML model to assess the impacts of the fluctuations in traffic on NO₂, O₃, and fine particle (PM₂.₅) in the Los Angeles. The quantitative changes from emission reductions were evaluated in ambient air quality in 11 cities globally by (Shi et al., 2021). Notwithstanding, it is still a gray area for evaluating the vertical distribution of fluctuation driven by emissions, chemical reactions, pollutant transport, and meteorology. Thus, the available MAX-DOAS data set provide with a potential to research this further.

The purpose of this study was to assess the impact of COVID-19 lockdown on vertical distributions of NO₂ and HCHO based on MAX-DOAS measurements and ML modeling. This paper is structured as follows: Section 2 gives the details of the MAX-DOAS measurements to retrieve the vertical profiles and the corresponding quality control. In Section 3, an overview of the MAX-DOAS results and the comparisons with independent measurements are displayed. Four types of ML models were compared and evaluated to simulate NO₂ and HCHO profiles. Finally, the evaluation is conducted with the changes in surface pollutant and meteorological factors during COVID-19 lockdown. In Section 4, the summary and conclusions are presented.

2. MAX-DOAS Measurements and Ancillary Datasets

2.1. MAX-DOAS Set Up and Vertical Profile Retrieval

The self-assembled MAX-DOAS instrument was installed on the roof of Dianshan Lake (DSL) Environmental Monitoring Station of Shanghai (31.10°N, 120.98°E), from 17th October 2019 to 31th December 2020. The complex surroundings and circulation characteristics of this site anticipate the frequent influences by pollutants transported from the Northwest (Jiangsu), Southwest (Zhejiang), and East (downtown of Shanghai) (G. Zhang et al., 2021) (Figure 1). The MAX-DOAS system is composed mainly of a scanning telescope, a camera, two stepping motors, a spectrometer (Ocean Optics, QE65 Pro), and a computer operating the MAX-DOAS system (Cheng et al., 2019). The azimuth angle of 5° from the north clockwise was set for avoiding the direct sunlight. The scanning sequence of the telescope is comprised of 10 elevation angles, that is, 3°, 4°, 5°, 7°, 9°, 12°, 15°, 20°, 30°, and 90°. The purpose of the camera is to record the sky condition simultaneously at each elevation angle in real time. Installed a cooled charge-coupled device detector, the spectrometer can measure the spectra covering the UV and visible wavelength ranges (300–480 nm) with a spectral resolution of 0.5 nm full width half maximum.

An essential precondition for deriving the vertical profiles is the inversions of differential slant column densities (DSCDs) of oxygen dimer (O₂), NO₂, and HCHO, which was implemented on the QDOAS software (http://uv-vis.aeronomie.be/software/QDOAS/ last access: 12 January 2021). The spectral analysis was performed with respect to a Fraunhofer Reference Spectrum (FRS) measured at the zenith direction (Hönninger et al., 2004). Here, drawing on the previous research results (Chan et al., 2020; Kumar et al., 2020; Y. Wang et al., 2017), the fitting windows of 351.0–390.0, 415.0–450.0, and 324.5–359.0 nm were suitably used for O₂, NO₂, and HCHO DSCD retrievals, respectively. The detailed properties of the DOAS analyses are provided in Table S1 in Supporting Information S1. Examples of O₂, NO₂, and HCHO DSCDs retrieval from the MAX-DOAS spectrum taken on 30 August 2020 at 09:12 LT are shown in Figure S1 in Supporting Information S1.
Tropospheric vertical profiles of aerosol extinction and volume mixing ratios (VMRs) of NO2 and HCHO can be retrieved for each MAX-DOAS scan by applying an optimal estimation method. In this study, the HEIPRO (Heidelberg Profile) algorithm (Friess et al., 2006), developed by IUP Heidelberg, was performed as the profiling technology. Further details regarding the aerosol and trace gas profile retrievals, including the corresponding aerosol parameters input in SCIATRAN and a priori information settings, are extensively described in S. Zhang et al. (2021). Examples of aerosol extinction coefficient, NO2 and HCHO profile retrievals are exhibited in Figure S2 in Supporting Information S1.

2.2. Quality Control for Retrievals

For ensuring data reliability and high-availability, the threshold of RMS < 1.0 × 10⁻³ and DSCDs detection limits were used to eliminate the unreliable DSCDs and the corresponding qualified results remained 96.42% (O4), 96.15% (NO2), and 94.78% (HCHO), respectively (Text S1 and Table S2 in Supporting Information S1). For preliminary screening of retrieved profiles, the thresholds of 10¹⁶ molecules cm⁻² and 4 × 10¹⁵ molecules cm⁻² were adopted for absolute deviations of measured and modeled NO2 and HCHO DSCDs, respectively. Approximately 5% of both were eliminated.

The atmospheric light path is complex in cloudy sky, especially in the case of rapidly changing clouds (Wagner et al., 1998, 2004, 2011). The profiles contaminated by clouds should create uncertainties to some extent. Therefore, the identification of clouds measured each time can further strengthen quality control of basic data. The feedback difference of color index (CI) and O4 differential air mass factor (DAMF) obtained by MAX-DOAS can reflect different sky conditions (Figure S3 in Supporting Information S1). The details attached to the definitions of CI and O4 DAMF and the complete classification scheme of sky conditions are provided in the Text S2 and Figures S4 and S5 in Supporting Information S1.
The relative fractions of the seven sky conditions for the MAX-DOAS measurement period are presented in Figure 2. The condition of clear sky with low aerosol accounted for the major proportion of 33.27%. Thick clouds and continuous clouds mostly occurred in rainy months. Fog mainly existed in autumn and winter with the lowest frequency of 1.46% all over the year. Y. Wang et al. (2015) assessed the response of different sky conditions to the NO2 and HCHO inversion results, finding the large deviations under fog and thick cloud conditions. If long-term measurements are averaged, the impact of other cloudy conditions mostly cancel out (Kumar et al., 2020; Y. Wang et al., 2017). Therefore, the results judged just as fog and thick clouds (comprising ~22.47%) should be eliminated.

2.3. Ancillary Datasets

Surface PM2.5, NO2, O3, CO, and SO2 concentrations from 2017 to 2020 were provided by the Shanghai Environmental Monitoring Center (Lin et al., 2020). The in-situ instruments are installed at the same measurement station of the MAX-DOAS instrument. Surface HCHO concentrations were retrieved based on a long path differential optical absorption spectroscopy (LP-DOAS) system with a light path of 2.6 km and time resolution of 6 min (Zhu et al., 2020). The LP-DOAS instrument was installed in Fudan University Jiangwan Campus (31.34°N, 121.52°E), around 53 km away from the MAX-DOAS measurement site. Alongside LP-DOAS, a sun photometer was utilized to measure the aerosol optical density.

ERA5 is the new fifth-generation atmospheric reanalysis from the European Center for Medium-Range Weather Forecasts. ERA5 provides hourly around-the-clock meteorological factors from surface up to 0.01 hpa (spanning 137 vertical level) with the spatial resolution of 0.25° × 0.25° (Marshall, 2000). Seven meteorological factors were used in this study, including relative humidity (RH), temperature (T), vertical velocity (W), eastward wind (U), northward wind (V), geopotential height (GH), and boundary layer height (BLH).

The compared HCHO and NO2 VCDs were collected by the TROPOspheric Monitoring Instrument (TROPOMI) on-board of Sentinel-5 Precursor (S5P) satellite (Veefkind et al., 2012). The satellite monitors NO2 and HCHO VCDs at a local Equator overpass time of 13:30 LT with the high spatial resolution of 3.5 × 5.5 km². The HCHO and NO2 VCDs were extracted for 31.10°N, 120.98°E with a radius of 20 km and available after the filtering (filtering conditions: data quality value ≥ 0.75).
3. Results and Discussion

3.1. Overview of MAX-DOAS Retrievals

Vertical profiles retrieved from MAX-DOAS measurements were discussed and verified in this section. The meanly annual surface VMRs and VCDs of NO$_2$ were 8.71 ppbv and $1.46 \times 10^{16}$ molec.cm$^{-2}$, respectively. Monthly mean NO$_2$ is characterized by a V-shaped pattern at different altitudes neglecting the results in February (Figure 3 and S6 in Supporting Information S1). The late fall/winter maximum and summer minimum of NO$_2$ can be associated to the longer photochemical lifetimes caused by the lowest depletion of OH radical in winter (Schaub et al., 2007; Stavrakou et al., 2013). Since the dominant NO$_2$ sources (e.g., industrial fuel combustion and motor vehicle emissions) are at ground level, the major fraction of NO$_2$ column is concentrated below 500 m. Both monthly surface VMRs and VCDs of HCHO performed a coherent variation trend like a “top-hat” with high values between May and August (Figure 3 and S6 in Supporting Information S1). The corresponding means of the entire campaign are 3.89 ppbv and $1.16 \times 10^{16}$ molec.cm$^{-2}$, respectively. The photochemical secondary generation through VOCs oxidation can result in high HCHO during the hot months. Moreover, the strong oxidation with mixed rising VOCs from ground fed back to concentrate HCHO at higher altitudes (∼1.0 km).

Owing to the increasing photochemical processes and the decreasing traffic emission, surface NO$_2$ VMRs can be found the valley between 11:00–15:00 LT and approximately uniformed in different seasons (Figure 4). A weaker role of photochemical reaction advancing NO$_2$ accumulation in afternoon above 1.5 km, resulted in the discrepant diurnal variations even opposite from that at ground. For HCHO, both surface VMRs and VCDs evinced peak appearing around noon (10:00–13:00 LT) in spring and summer, whereas higher concentrations lasting after 13:00 LT in autumn and winter (Figure 4 and S7 in Supporting Information S1). It can be interpreted as the decreasing loss rates for reactions against the OH radical and the increasing production yielded by isoprene oxidation with enhanced temperature in the afternoon (Duncan et al., 2010).
To verify the accuracy of the retrieved NO2 and HCHO from MAX-DOAS measurements, the comparisons with other independent measurements were implemented (Figure S8 in Supporting Information S1). Calculated by the mean around the TROPOMI overpass time 13:00–14:00 LT, MAX-DOAS VCDs agree well with the TROPOMI observations with correlation coefficient ($R$) of 0.89 for NO2 and 0.79 for HCHO. The linear regression slopes between the datasets of MAX-DOAS and TROPOMI were 0.64 for NO2 and 0.71 for HCHO. The underestimation of satellite VCDs is partly related to the a priori profiles employed in the air mass factor calculation (Chan et al., 2020; Kumar et al., 2020). In previous studies, satellite-retrieved NO2 and HCHO VCDs were enhanced significantly by using the MAX-DOAS profile as a priori profiles (Su et al., 2020; Xing et al., 2017). The daily mean of surface NO2 VMRs from MAX-DOAS are consistent with in-situ measurements depicting a high correlation coefficient of 0.80 and slope of 0.88, but systematically lower by ∼20%. The comparison with daily averaged HCHO measured by LP-DOAS also reveals a strong consistency, accompanying $R$ of 0.85 and slope of 0.86. The definite distance between the positions of two instruments partially cause the lower slope of comparison with LP-DOAS. The comparison results presented above demonstrate the highly reliable retrievals from MAX-DOAS measurements.

In order to evaluate the measurement sensitivities of NO2 and HCHO vertical profiles, gain function matrix ($G: \frac{\delta \hat{x}}{\delta y}$) and average kernel matrix (AK: $\frac{\delta \hat{x}}{\delta x}$) are always introduced as the credible indicators. $G$ represents the sensitivity of retrieved vector (retrieved profile: $\hat{x}$) to measured vector (DSCDs: $y$). AK reflects the sensitivity of retrieved vector to true vector (true profile: $x$). As shown in Figure S2 in Supporting Information S1, $G$ and AK tend to change slightly above 3 km for NO2 and above 2 km for HCHO, indicating the low confidence above...
corresponding altitudes. Therefore, the vertical profiles of NO₂ and HCHO are intercepted from 0 to 2 km for the following machine learning simulations.

3.2. Machine Learning Simulation and Evaluation

Based on vertical profiles derived from MAX-DOAS and real-time predictive variables, the machine learning models were developed for predicting NO₂ and HCHO profiles (Figure 5). Predictive variables consisted of ERA5 and in-situ data during the daytime (09:00–15:00). For interpreting the concentrations and the potential transports at different altitudes, seven meteorological factors were adopted, including RH, T, W, U, V, GH, and BLH. The first six factors of different heights are weighted and averaged with height. Surface PM₁₀, CO, NO₂, SO₂, and O₃ concentrations were extracted for reflecting the characteristics of ground pollution. The initial preparation of the raw data was performed, including missing value processing, filtering outliers and normalization. The linear interpolation method was used to fill in when the proportion of missing data was less than 10%. Three-sigma rule of thumb was chosen to eliminate outliers. Predictors were standardized by subtracting the mean and dividing by the standard deviation.

In this study, four ML models were developed and evaluated separately: (a) multiple linear regression (MLR), (b) support vector machine (SVM), (c) bagged trees (BT), and (d) artificial neural network (ANN). According to the training functions, ANN model was divided into two kinds: Levenberg-Marquardt (ANNlm) and Scaled Conjugate Gradient (ANNscg). The detailed configurations of ML models used for this study are elaborated in Text S3 in Supporting Information S1 and the empirical equations of proposed algorithms are presented in Table 1.

Figure 5. The methodology framework of machine learning models.
Here, data for the whole year 2020 was required for training and validation, adopting 10-folds cross validation. After training, each model was applied to predict additional vertical profiles of NO2 and HCHO (17th October–21st November 2019 and 1st January–28th February 2021) to test the corresponding stability. In this study, all of ML models were trained to predict NO2 and HCHO at different altitudes. Within a given ML model, the results of altitudes bins are different. The evaluation indicators of the model performance mainly consider root mean squared error (RMSE), mean absolute error (MAE), Pearson moment correlation coefficient (PMC or R), and index of agreement (IOA). See the definition of this metrics in Text S4 in Supporting Information S1.

Figure 5 presents the methodology framework proposed to develop the statistical predicting models for NO2 and HCHO profiles on hourly scale.

Considering the sensitivity of profile retrievals, these ML models are simply conducted to simulate the NO2 and HCHO profiles below 2 km. Figure 6 depicts the performance statistics obtained on test data set, using the results of altitudes bins are different. The evaluation indicators of the model performance mainly consider root mean squared error (RMSE), mean absolute error (MAE), Pearson moment correlation coefficient (PMC or R), and index of agreement (IOA). See the definition of this metrics in Text S4 in Supporting Information S1.

Table 1

| ML model | Empirical equation |
|----------|--------------------|
| MLR      | \( \hat{f}(x) = a_0 + \sum_{i=1}^{n} a_i x_i \) |
| SWM      | \( \hat{f}(x) = \sum_{i=1}^{n} (a_i - a'_i) K(x, x_i) + b \) |
| BT       | \( \hat{f}(x)_{\text{BT}} = \frac{1}{B} \sum_{b=1}^{B} \hat{f}(x) \) |
| ANN      | \( \hat{f}(x) = s \left( \sum_{j=1}^{m} W_{ij} x_j + b_i \right) \) |

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Considering the sensitivity of profile retrievals, these ML models are simply conducted to simulate the NO2 and HCHO profiles below 2 km. Figure 6 depicts the performance statistics obtained on test data set, using the different fitting models for NO2 and HCHO profiles. It indicates that BT model is the perfect solution owing to its lowest RMSE (MAE) and highest PMC (IOA). BT model exhibit relatively high fidelity in reproducing the observed NO2 and HCHO profiles, with mean PMC (IOA) of 0.86 ± 0.04 (0.83 ± 0.05) and 0.81 ± 0.04 (0.82 ± 0.03) in height, respectively. Lower signal-to-noise ratio of profiles can be the main cause of decreasing in PMC (IOA) at high altitudes. The RMSE from BT model fitting on test data set is averaged out 1.85 ± 1.18 ppbv (0.82 ± 0.03) in height, respectively. Lower signal-to-noise ratio of profiles can be the main cause of decreasing in PMC (IOA) at high altitudes. The RMSE from BT model fitting on test data set is averaged out 1.85 ± 1.18 ppbv (0.82 ± 0.03) in height, respectively. Lower signal-to-noise ratio of profiles can be the main cause of decreasing in PMC (IOA) at high altitudes. The RMSE from BT model fitting on test data set is averaged out 1.85 ± 1.18 ppbv (0.82 ± 0.03) in height, respectively.

MLR model cannot achieve a reasonable agreement with observed profiles, illustrating the nonlinear relation between target gas concentration and predictive variables. Since the inaccurate selection of kernel trick, SVM, an efficiently method performing a non-linear fitting, also cannot improve PMC (IOA) to 0.7 (Richhariya et al., 2021). The kernel trick and its parameters are indeterminate for SVM modeling NO2 and HCHO profiles,
thus they are only determined empirically. ANN model is after all a black-box type of operation with no functional relationships between the target and predictor variables. Besides, it lacks any prior assumption, such as the clear rules or fixed guidelines for optimal ANN architecture, in model building process (Ramalingam et al., 2020). Therefore, four evaluation indicators of the model performance fluctuate violently with height. ANNscg is more suitable than ANNlm in low memory situations, hence slightly better in reproducing NO2 and HCHO profiles. In BT model, resampling is carried out in the original data samples and each sample is fitted with a regression tree model. For reducing variance of the output error and avoid overfitting, these regression trees are grown without pruning and averaged (Breiman, 1996). It can contribute to more flexibility in leveraging predictive variable and higher computational efficiency relative to other models.

An important output of the BT model is the relative importance of all input predictive variables. Figure 7 characterizes the higher importance of main control factors distributed at heights below 0.8 km. Among them, three major governing variables for NO2 profile modeling are, surface NO2, O3, and BLH according to variable importance higher than 0.1. Except for surface NO2, the prominent ranks of O3 and BLH reflect that chemical reactions and pollutant transport act a prevailing role in NO2 distribution. The large regulation on HCHO profile was dominated by CO, O3, and PM2.5. Additionally, HCHO partly follows that emitted from the vehicle and industrial emissions, which is interrelated to CO concentration (Parrish, Ryerson, et al., 2012). PM2.5 affects atmospheric visibility exerting indirectly the impact on photochemical processes. O3 responds to a tracer to identify the secondary sources of HCHO (Lui et al., 2017; Su et al., 2019). Furthermore, higher importance on the factors of PM2.5 and O3 than that of CO, manifests secondary formation dominating HCHO sources at DSL site. This point was documented in accordance with previous studies (Fan et al., 2021; Y. Sun et al., 2021; S. Zhang et al., 2021).

From Figure 7, the predictions are quite sensitive to the in-situ measurements of NOx and O3. Considering the uncertainties from in-situ measurements can propagate through the ML model and affect the final results, it is necessary to quantify these uncertainties. The in-situ surface measurements of ambient air are operated in accordance with China Environmental Protection Standards “HJ 817-2018” for PM2.5 and “HJ 818-2018” for SO2, NO2, O3, and CO. The uncertainties of in-situ measurements for these species were required to be controlled within 5%. For testing the impacts of uncertainties on in-situ measurements, the concentrations of these pollutants adjusted by 5% were input into the BT model to simulate the NO2 and HCHO profiles. The difference of the simulation results before and after adjustment represents the impact of the corresponding final uncertainty on the simulation results. In order to maximize the effect, these differences are taken as absolute values and added. Figure S9 in Supporting Information S1 presents the vertical distribution uncertainties caused by these in-situ measurements and their sum on NO2 and HCHO BT modeling results. The final individual uncertainty is almost less than 2.0%,
and their sum is less than 3.5%. It illustrates that the uncertainties of in-situ measurement have slight influences on the output results of ML model.

With the results discussed above, NO2 and HCHO profiles from actual observation and BT model were compared and differentiated on training (validation) and test data set (Figures S10 and S11 in Supporting Information S1). The biases of NO2 profiles range from −6.7 to 16.7 ppbv on training (validation) data set, mapped to the range of −7.1–21.2 ppbv on test data set. For HCHO, the differences of the two datasets cover −3.1–6.6 and −1.5–5.8 ppbv, respectively. The absolute differences of them appear as a decrease with height, but the deviations of HCHO profiles occurred at higher altitudes (almost below 1.4 km) than NO2 (almost below 1.0 km). It is related to the vertical distribution characteristics of target profiles. Elucidated from Figures S10 and S11 in Supporting Information S1, BT models tending to have larger biases in modeling the extreme values were linked to fewer training data samples than those of middle values (Yang et al., 2021).

### 3.3. Responses of NO2 and HCHO Profiles to COVID-19 Lockdown

With the advantages of high accuracy, fast calculation and easy operation, ML was widely employed in the realm of pollutant prediction. Consequently, the predictions, in previous years at the same period of COVID-19 lockdown, can be available for evaluating the impacts of the lockdowns on NO2 and HCHO profiles. The YRD region implemented strictest lockdown response to public health emergencies from 26th January to 17th February (N. Wang et al., 2021), so it was selected as the period of COVID-19 lockdown in this study. The trained BT model in Section 3.2 were conducted to reproduce NO2 and HCHO profiles in the same period from 2017 to 2019 (Figures S12 and S13 in Supporting Information S1).

The meanly vertical profiles of NO2 presented the deductions of 43.8% (Diff2017,2020), 45.5% (Diff2018,2020), and 44.6% (Diff2019,2020), for 2017, 2018, and 2019, respectively. For HCHO, the lockdown-induced situation resulted in the corresponding mean declines of 28.6%, 32.1%, and 10.9%, respectively (Figure 8). The comparisons of modeling results were implemented with other realistic measurements, holding a relatively good consistency.
The similar variation of NO2 and HCHO VCDs were also reported by previous studies (Huang et al., 2021; Javed et al., 2020; Li et al., 2021; Tanvir et al., 2021). Both NO2 and HCHO generally showed a trend of first decreasing and then increasing with height, appearing the peak around 0.4 km. The reductions on HCHO even reversed as negative at high altitudes, suggesting the reduction in anthropogenic emissions does not necessarily cause HCHO decreasing at high altitudes. It can be attributed to the accelerated HCHO production rate through the oxidation of NMVOCs, driven by higher hydroxyl radical (OH) levels (W. Sun et al., 2021). Besides, multi-scale air quality model has proved that OH enhanced during the COVID-19 lockdown in YRD region (Y. Wang et al., 2021).

Traffic reduction during lockdown led to a lower level in surface PM2.5, NO2, and CO, relative to that in previous years (2017–2019) (Figure S15 in Supporting Information S1). These variation patterns can match with the corresponding variations of NO2 profiles. A decrease in SO2 was remained after 2018, but changed slightly from 2019 to 2020. O3 tended to a decrease from 2017 to 2019, but increased from 2019 to 2020. It has been confirmed that the dropping NOx emissions can alleviate the ozone titration and further enhanced the atmospheric oxidizing capacity (AOC) (Gu et al., 2021). To some extent related to it, the reduction of HCHO by lessening primary emissions can be compensated by the improved secondary formation. In term of the meteorological factors, except for GH and BLH, no intrinsic changes in other meteorological factors between in the COVID-19 lockdown and normal situation (especially in 2019 and 2020). GH alteration can affect the atmospheric oxidation to trigger the ambient air pollution fluctuation (Wu et al., 2019). Higher BLH in COVID-19 lockdown can bring the favorable atmospheric permeability and further diffuse the pollutants (Nguyen et al., 2021).

To further separate the impacts of meteorological factors and surface pollutants on the variations in vertical distribution of NO2 and HCHO, we adopted the method of controlling variables referring to the study by Yang et al. (2021). The predictions were conducted by BT model with two additional scenarios: COVID-19 meteorological condition with normal surface pollution situation (e.g., 2017surf, 2020surf) and COVID-19 surface pollution situation with normal meteorological conditions (e.g., 2017met, 2020surf). The modeling results with 2020surf can represent the influence of surface pollutants in lockdown by maintaining meteorological situation consistency,
and the predictions with $20_{surf}$ correspond to the impact by the special meteorological conditions. A significant discrepancy existed in the reductions driven by two scenarios with the segmentation point of around 0.4 km (Figure 9).

Averaged with height, the NO$_2$ deductions driven by the variation of surface pollutants ($R_{surf}$) were 27.0%, 31.3%, and 21.5%, for 2017–2019, respectively. The corresponding NO$_2$ deductions forced by meteorological changes were 19.5%, 14.3%, and 21.3%. For HCHO, mean $R_{surf}$ were 11.0%, 13.1%, and 4.4% with mean $R_{met}$ of 13.7%, 15.7%, and 6.1%. As for vertical distributions of reductions on NO$_2$ and HCHO, both $R_{met}$ and $R_{surf}$ increased firstly and then decreased, with the peaks at around 0.6 and 0.2 km (Figure 10). The $R_{surf}$ of NO$_2$ and HCHO was higher than $R_{met}$ at the altitudes below 0.5 km, while almost presented a lower value above 0.5 km. It emphasizes the noticeable influence of meteorological factors on the concentration fluctuation, as well as illuminates the deficiencies in the researches limited to surface. As discussed earlier, enhanced AOC driven by reducing NO$_x$ emissions were the main cause of increasing HCHO at high altitudes. Note that all results are based on statistical analysis of data but lacked of exact physical and chemical explanation. This could be revisited by chemical transport mode in the future when accurate emission inventories for the COVID lockdown period are developed.

Despite all this, due to the scarcity of vertical observations, these profiles combined with machine learning simulation are of great value in the field of atmospheric environment research. For instance, these profiles can be employed as reference for validating the modeling results and assimilation data of chemical transport model. In addition, the vertical profiles can be used as the input data in 1D box model for constraining concentrations of corresponding pollutants and studying the relevant chemical mechanisms.

4. Summary and Conclusions

A campaign for monitoring the vertical profiles of NO$_2$ and HCHO was performed by MAX-DOAS in a regionally representative suburban site of Shanghai during 17th October 2019–31th December 2020. For filtering the impacts of sky conditions unfavorable for NO$_2$ and HCHO profile retrieval, an effective classification scheme was proposed based on the distinguishable responses to CI and O$_3$ DAMF. Ultimately, 22.47% of data, judged as fog and thick clouds, was removed for the subsequent discussion. As for retrievals, the monthly variation of NO$_2$ follows generally a V-shaped pattern without considering that in January. For HCHO, seasonal rhythm
was demonstrated a coherent variation trend like a “top-hat” with roughly representing a plateau between May and August. The vertical distribution of NO$_2$ was characterized by major NO$_2$ concentrated below 500 m, while a considerable level of HCHO can be detected until 1 km. The surface VMRs and VCDs of NO$_2$ and HCHO agreed well with in-situ, LP-DOAS and TROPOMI satellite observations, suggesting the highly availability of MAX-DOAS retrievals.

Based on vertical profiles derived from MAX-DOAS and simultaneous predictive variables, ML models were applied to reproduce NO$_2$ and HCHO profiles, avoiding the complex chemical transport model and shortening the simulation duration. Compared and evaluated four types of ML models, BT model was adopted due to its best performance with high fidelity on test data set. BT model exhibits relatively high fidelity with mean PMC (IOA) of 0.86 ± 0.04 (0.84 ± 0.04), and 0.91 ± 0.03 (0.91 ± 0.03) in height on test data set, respectively. The RMSE from BT model fitting on test data set is averaged out 1.82 ± 1.37 ppbv for NO$_2$, and 0.37 ± 0.26 ppbv for HCHO. For MAE by BT model, the mean values are 1.08 ± 0.92 and 0.28 ± 0.18 ppbv, respectively.

The trained BT model was conducted to evaluate the impacts of the lockdowns on NO$_2$ and HCHO by predicting profiles in the same period from 2017 to 2019. Results found that the mean NO$_2$ decreased in 2020 by the changes of 43.8%, 45.5%, and 44.6%, corresponding to 2017, 2018, and 2019, respectively. For HCHO, the lockdown-induced situation presented the declines of 28.6%, 32.1%, and 10.9%, respectively. For diversing the influence of variations in meteorological factors and surface pollutants on NO$_2$ and HCHO distribution, multiple predictions with different scenarios were conducted from 2017 to 2019. The reduction of NO$_2$ and HCHO from the variation of surface pollutants was dominated below 0.5 km, while the relevant meteorological factors played a more significant role above 0.5 km. Moreover, the results indicated that HCHO was offset in reductions at low altitudes even converted into enhancement at high altitudes, owing to enhanced atmospheric oxidation capacity driven by reducing NO$_x$ emissions.

**Data Availability Statement**

The data set of MAX-DOAS, LP-DOAS, and sun photometer can be available at https://data.mendeley.com/data-sets/r4jjhtjyyyp/2 (S. Zhang, 2022) [Data set]. The data set from in situ measurements can be obtained at https://www.aqistudy.cn/ (China MEP, 2013) [Data set]. TROPOMI data are publicly available at https://disc.gsfc.nasa.gov/datasets?keywords=TROPOMI%26page=1 (GES DISC, 2021) [Data set]. The ERA5 data are downloaded...
from European Center for Medium-Range Weather Forecast's MARS archive (CDMS, 2018; https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5) [Data set].

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