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A High-Performance Convolutional Neural Network for Ground-Level Ozone Estimation in Eastern China

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Abstract: Having a high-quality historical air pollutant dataset is critical for environmental and epidemiological research. In this study, a novel deep learning model based on convolutional neural network architecture was developed to estimate ground-level ozone concentrations across eastern China. A high-resolution maximum daily average 8-h (MDA8) surface ground ozone concentration dataset was generated with the support of the total ozone column from the satellite Tropospheric Monitoring Instrument, meteorological data from the China Meteorological Administration Land Data Assimilation System, and simulations of the WRF-Chem model. The modeled results were compared with in situ measurements in five cities that were not involved in model training, and the mean $R^2$ of predicted ozone with observed values was 0.9, indicating the good robustness of our model. In addition, we compared the model results with some widely used machine learning techniques (e.g., random forest) and recently published ozone datasets, showing that the accuracy of our model is higher and that the spatial distributions of predicted ozone are more coherent. This study provides an efficient and exact method to estimate ground-level ozone and offers a new perspective for modeling spatiotemporal air pollutants.

Keywords: ground-level ozone; deep learning; convolutional neural network

1. Introduction

Ground-level ozone and short-lifetime greenhouse gases are harmful to human health at high concentrations, with a significant effect on radiative forcing [1]. Although China has implemented strict atmospheric protection measures in recent years, ozone pollution is still worsening [2]. An annual mean trend of $3.3 \pm 4.7 \text{ ug/m}^3$ of maximum daily average 8-h (MDA8) ozone in China between 2015 and 2019 was reported [3]. In 2015 and 2020, the ozone-related all-cause and respiratory health impact in China increased by 94.61 and 96.54%, respectively [4]. Ozone pollution is particularly severe in eastern China. In 2015, eastern China had the highest annual mean MDA8 ozone concentration in the country [5], which increased by ~13% from 2015 to 2017 [6]. This is also a region with high population density. There is an urgent need to obtain accurate information about the spatiotemporal distribution of ozone in this region.
China has established more than 1600 air-quality monitoring sites since 2013. Observations from the state-managed monitoring network are important for understanding the atmospheric environment. However, the construction and maintenance of these sites have a high resource cost, and most of the sites have limited representative areas [7]. Hence, several methods have been developed to compensate for the inadequate spatiotemporal coverage of monitoring sites, which can generally be divided into numerically driven and data-driven models. Numerically driven models, such as WRF-Chem and GEOS-Chem, simulate the three-dimensional distribution of atmospheric components considering the pollutant emissions, meteorological data and chemical reactions [8–10]. There is strong interpretability of these models due to the clear calculation of causality between pollutants and predictive variables [11]. The advantage of numerically driven models is that they can generate ground-level ozone data with high spatiotemporal resolution, but they require a huge amount of calculation. In addition, the accuracy is not very high because of our imperfect understanding of ozone-related chemical mechanisms and much uncertainty in emission source data [12]. For example, a recent study used WRF-Chem to map the ground-level ozone distribution over Asia at 8 km spatial resolution, and the Pearson’s correlation coefficient (R) of simulations with in situ observations was 0.51 [13].

Data-driven models establish regression equations between ozone and its predictors (e.g., satellite retrievals and meteorological data). These models have been widely adopted because of their characteristics of high accuracy and fast calculation [14]. Data-driven models use linear fitting methods (e.g., land-use regression (LUR) and geographically weighted regression) [15–17], so their performance is easily affected by collinear variables. More importantly, linear functions are insufficient to describe the multiple complex interactions between ozone and its predictors. With the development of computer performance and machine learning (ML) theory, ML models are becoming more popular. A comparison between various LUR and ML models in ozone estimation across the United States indicated that ML models had significant superiority [18]. Currently, many typical ML models, such as random forest [19,20], gradient-boosting decision tree [21], and deep neural network [22], have been successfully applied in mapping the spatial distribution of air pollution. Although most studies have reported acceptable accuracy and reasonable robustness, these models cannot include spatiotemporal information about air pollution, which does not conform with the prior knowledge that air pollution has a significant spatiotemporal correlation. As for spatiotemporal models, the widely used model is geographically and temporal weighted regression (GTWR), which uses a local multiple linear regression to consider the spatiotemporal heterogeneity of the predictors and air pollution [23]. Although the GTWR model usually performs better than some global linear models, its performance is still limited by linear fitting. Some scholars have improved ML algorithms to reinforce the processing of spatiotemporal information. For example, spatiotemporal kriging was embedded in a random forest model for NO\textsubscript{2} estimation; the out-of-bag errors in this model take into account the spatiotemporal interpolations of kriging [24]. An exponential spatiotemporal weighting function was introduced in a generalized regression neural network for estimating PM\textsubscript{2.5}, the spatiotemporal similarity between different samples is expressed [25]. A geospatially local CatBoost model was developed for ozone estimation; an adaptive threshold function was used to determine the modeling scope [26]. Although these studies reported higher accuracy, the expression of spatiotemporal information is experiential and relatively simple, and the modeling process is complicated due to too many hyperparameters being present, which are usually determined by a computationally intensive nested-loop process.

Deep learning, a subfield of machine learning, can extract high-level features from large data to achieve accurate parameter estimation by using several hidden computational layers [27]. The convolutional neural network (CNN) is a commonly used deep learning architecture, which can process spatiotemporal information simultaneously using multilayer convolution filters. Compared to the multilayer perceptron, the advantages of CNN are that the spatiotemporal coherence of inputs can be preserved by applying convolution to adja-
cent inputs using different filters, and the number of trainable parameters can be reduced using the local receptive field [28]. The characteristics of CNN match the requirements of atmospheric modeling well, since many pollution and predictors exhibit spatiotemporal coherence. For example, CNN can learn the temporal pattern of input variables for predicting site observations in advance and conducting outlier detection [29,30]. Furthermore, CNN can extract useful spatiotemporal features for estimating surface air pollution spatial distribution [31,32], which is also this paper’s objective.

In this study, a novel CNN-based architecture was developed for estimating ground-level ozone concentrations. We took advantage of the flexible structure of CNN and used different filters to process the multi-scale spatiotemporal information. The inputs of our model mainly include Tropospheric Monitoring Instrument (TROPOMI) ozone retrievals, China Meteorological Administration Land Data Assimilation System (CLDAS) meteorological data, and WRF-Chem model simulations of three types of air pollution (ozone, NO\(_2\), and PM\(_{2.5}\)). To the best of our knowledge, this study is the first to adopt CNN architecture for ground-level ozone estimation. After validating the overall fitting ability and spatiotemporal extrapolation of our model, full-coverage daily MDA8 ozone at 0.05° spatial resolution over eastern China in 2020 was generated.

2. Materials and Methods

2.1. Data Description

2.1.1. Study Area and Ground-Level Ozone Concentration Data

Figure 1 shows a schematic diagram of our study area, which ranges from 26.98° to 38.45°N and 113.96° to 124.04°E. The study area includes 4 provinces (Shandong, Anhui, Jiangsu, and Zhejiang) and one province-level municipality (Shanghai). It is one of the most economically developed and densely populated areas in China. In addition, Shandong has the largest chemical industrial base nationwide, and is a region with one of the highest levels of pollutant emissions in China [33]. This area can be used as a representative area for ground-level ozone modeling.

![Figure 1. Schematic diagram of study area.](image-url)

We collected hourly ground-level ozone concentration data from the China National Environmental Monitoring Center (CNEMC) [34]. Ozone concentrations were measured using UV-spectrophotometry, and the data quality at all monitoring sites was validated using HJ 818-2018 specifications [35]. The arrangement of sampling sites followed HJ 664-2013 specifications, which mainly ensure stable environmental conditions around sites, the proper distance between sites and pollution sources, and the horizontal distance between the sampling port and the highest obstacle [36].
2.1.2. Satellite-Based Total Ozone Column (TOC) Data

Although satellite-retrieved TOC cannot directly correspond quantitatively to the ground-level ozone concentration, many studies have reported the acceptable accuracy for monitoring long-term ozone variation using a ozone monitoring instrument (OMI) TOC [37]. In particular, Liu et al. evaluated the OMI TOC in east China over ten years and indicated that there is a reasonable consistency between OMI TOC and ground-level ozone [38]. Furthermore, OMI TOC has been widely used as a useful proxy variable for ground-level ozone in both numerical models and machine learning models [14,19,39]. However, there are too many missing values in OMI data due to the sensor’s physical obstruction (the so-called row anomaly). As a successor to OMI, TROPOMI is carried on the Sentinel 5 Precursor (S5P) satellite, which launched in 2017. Compared to OMI, TROPOMI is equipped with a more sophisticated hyperspectral imager for the measurements of seven bands, the spatial resolution is significantly improved, and the signal-to-noise ratio is increased by 1–5 times. TROPOMI TOC is retrieved from Huggins spectral bands (325–335 nm) using the GODFIT direct fitting algorithm, which is a least-squares cost function minimization method based on the differences between measured and simulated radiances. The comparison between TROPOMI TOC and ground-based TOC measurements shows that the quality of the TROPOMI TOC is excellent with a bias of max. 3.5–5% [40]. We downloaded the TROPOMI level 2 total ozone column (5.5 km x 3.5 km) from the Copernicus Open Access Hub [41]. The S5P satellite passes over our study area at about 13:30 local time. Most of the S5P passing times are within the period of the highest daily concentration of ozone over eastern China. Statistical results show that the coefficient of determination ($R^2$) and mean absolute error (MAE) between the ozone concentration at the S5P passing time and MDA8 simulation were 0.86 and 11.63 µg/m$^3$, respectively. Therefore, the ozone concentration at the time of S5P passing largely reflects the daily MDA8 ozone level.

2.1.3. Meteorological Data

Precipitation (PRE), surface pressure (SP), specific humidity (SH), downward short-wave radiation flux (DSR), 2 m temperature (TMP), and 10 m wind speed (WIN) were obtained at 0.0625° spatial resolution by CLDAS version 2.0 [42]. Compared with measurements of ground sites, the correlation and root mean square error (RMSE) for PRE, SP, SH, DSR, TMP, and WIN were 0.72 and 0.94 mm/H, 0.96 and 3.74 hPa, 0.93 and 4.76%, 0.9 and 31.9 W/m$^2$, 0.97 and 0.88 K, and 0.82 and 0.83 m/s, respectively [43,44]. In addition, the boundary layer height (BLH) and 2 m dew point temperature (DPT) at 0.25° spatial resolution were derived from the ERA5 dataset [45].

These meteorological data are important in ground-level ozone formation. Ozone is the result of the photochemical reaction of precursor pollutants, and DSR can reflect the solar radiation intensity, which has a direct impact on photochemical reaction rate [46]. TMP reflects the intensity of radiation to a certain extent and also affects the photochemical reaction rate [47]. PRE means there is little radiation near the surface, which can slow the photochemical process. In addition, depositing on water droplets could promote the depletion of ozone [48]. DPT and SH carry the information of water vapor, which can convert NOx into water-soluble HNO$_3$, slowing down the production of ozone [49]. BLH is a function of many meteorological factors, which has a variety of complex nonlinear effects on ozone formation; for example, lower BLH transports less moisture to higher layers to form clouds, so the clouds become more transparent and the photolysis rates for NO$_2$ could be increased [50]. Wind can disperse concentration from the emission sources, so there is always a negative correlation between WIN and ground-level ozone concentration [51].

2.1.4. WRF-Chem Simulations and Other Data

To increase the ozone sources information in our model, we obtained WRF-Chem simulations from the Regional Atmospheric Environmental Modeling System (RAEMS). RAEMS, which aims to provide information on major atmospheric pollutants, such as PM$_{2.5}$ and ozone, and key meteorological factors for eastern China, was built based on
WRF-Chem version 3.2 [52] and the Multi-Resolution Emission Inventory for China [53]. The R value between simulations and in situ observations of PM$_{2.5}$ and ozone is ~0.7 and that of NO$_2$ is ~0.5 [54].

The normalized difference vegetation index (NDVI) was obtained from the MODIS 16-day product [55]. Surface elevation (digital elevation model (DEM)) was derived from the Shuttle Radar Topography Mission [56]. They are important variables that reflect ground information. Day of year (DOY) is a commonly used representation of temporal information, which is important for air pollutant modeling.

Specific information of the data used in this paper can be found in Table S1.

2.2. Data Preprocessing

The Technical Specification for the Environmental Air Quality Assessment for China addresses the maximum daily 8 h moving average from 8:00 to 24:00 local time as the daily MDA8 ozone level [57]. As for data quality control, MDA8 ozone data will be discarded if there not 14 items of hourly ozone measurements for that time period, unless the MDA8 ozone concentration exceeds the limits of the air quality standard. We processed hourly ozone measurements of the CNEMC and RAMES simulations into MDA8 concentrations based on the specifications. Hourly meteorological data of CLDAS and ERA5 were also averaged over that time period. Hourly simulations of PM$_{2.5}$ and NO$_2$ were processed into daily averages.

It is worth noting that the space coverage of satellite is not uniform due to the uncertainty of observation conditions and sensor’s operation status. A quality assurance value, which ranges from 0 (error) to 1 (all is well), of each pixel is provided with the TROPOMI data. The readme of TROPOMI Level-2 product recommends users to use pixels associated with quality values above 0.5 to exclude outliers caused by cloud cover, the presence of snow and ice, etc. As a result, the deficiency rate of TROPOMI data was ~2.8%. In this paper, the ordinary Kriging (OK) method was used for supplementing missing data. To evaluate the effectiveness of this method, a randomized sensitivity experiment was performed according to a real case (Oct. 18), which is the worst day in our data, with a missing rate of 8.9%. Then, we imputed missing data using the OK method from TROPOMI data and the bilinear interpolation method from ERA5 TOC data, respectively. The comparison results show that OK method has better performance with a higher $R^2$ (seen in Figure S1), and a more similar spatial distribution (seen in Figure S2). As for other data, there are few missing values, and we believed that the OK method can guarantee the data quality.

After supplementing missing data, all data except for ground-level ozone concentration data were resampled into 0.05° spatial resolution by performing cubic convolution interpolation, in which the values of new pixels can be determined by fitting curves through the values of the 16 nearest input pixels. The grid that the site fell into was used as a sampling center grid. We used two spatiotemporal data-matching methods. The first method was used to extract the long-term time series of the center grid, with 10 days of single-point data of each variable fused into a 2-D matrix with a size of 10 × 15 (10 days and 15 variables). The second method was used to extract the short-term spatiotemporal data of the center grid, with 5 days of spatial data of each variable merged into a 4-D matrix with a size of 5 × 7 × 7 × 15 (5 days, 7 × 7 in the spatial dimension, and 15 variables). Figure S3 shows a schematic diagram of our matching methods.

2.3. Model Development

Our model consists of two sub-networks, and was developed based on CNN and the Convolutional Block Attention Module (CBAM). The CBAM is formed by the channel attention module (CAM) and the spatial attention module, which is an efficient feature-refinement module with a somewhat increased number of parameters [58]. The CNN can be divided into 1-D and 2-D CNN according to the number of dimensions of the sliding convolution kernels. The convolution kernels of the 2-D CNN slide along two dimensions of the inputs, usually corresponding to spatial scope, so this CNN is widely
used in image processing. The 1-D CNN generally corresponds to temporal dimension, so it can be applied in time-series processing. Because the applications of these algorithms are extensive and the principles are complex, the schematic formulas will not be repeated in this study.

A schematic diagram and the specific parameter settings of our model are shown in Figure 2. For sub-network 1, the input is a 4-D matrix, as mentioned above. We first applied a combination of CAM and 2-D CNN to the inputs, and 2-D CNN with $1 \times 1$ kernel size was applied to each variable, calculating the temporal dimension. The $1 \times 1$ convolution kernel, also known as network-in-network, is used for cross-dimension information interaction. Here, this algorithm was used for the aggregated temporal information. The temporal feature map of each variable was obtained through two combination layers, and the calculated feature size was $7 \times 7 \times 15$. Then, a CBAM and 2-D CNN with $7 \times 7$ convolution kernel size were applied to this feature, aiming to extract the spatial and channel (the information between different variables) features, and finally a feature map was obtained with a size of $3 \times 3 \times 64$. This sub-network represents a low-cost and crude way to process temporal information while focusing on extracting spatial and channel information. For sub-network 2, three layers of 1-D CNN were applied to 10-day time-series inputs, obtaining a $6 \times 16$ feature map. This sub-network worked to extract long-term temporal features. In the end, the features extracted from the two sub-networks were flattened into a 1-D vector and connected, then passed into two fully connected layers for regression. In brief, the model can be summed up as follows:

\[
\text{surface ozone} = F (\text{TROPOMI TOC} + \text{PRE} + \text{SP} + \text{SH} + \text{DSR} + \text{TMP} + \text{WIN} + \text{BLH} + \text{DPT} + \text{WRF-Chem simulations} + \text{NDVI} + \text{DEM} + \text{DOY})
\]

Figure 2. Model architecture.

In addition, the model’s optimizer is Adam, the loss function is the mean square error, the learning rate is reduced by a factor of 0.2 with 5 epochs based on validation loss, and the model stops training when validation loss has stopped improving in 10 epochs. The model was developed using the TensorFlow2.4 machine learning platform.

2.4. Model Validation
2.4.1. Cross-Validation

We adopted a 10-fold cross-validation (CV) strategy to validate model performance. To test the overall fit, the widely adopted sample-based CV was selected [14,20,39]. In sample-
Based CV, all matched samples are randomly divided 10-fold, of which 9 folds are used for model training, and the rest are used for validation. This process is repeated 10 times to ensure that all samples are tested. To test spatiotemporal predictive ability, we adopted space-based and time-based CV, whose operations are similar to sample-based CV, but the sample division criteria are spatial grid (0.05°) and DOY (day of year), respectively [59]. A schematic diagram of the 3 CV methods is shown in Figure 3.

![Figure 3. Cross-validation methods.](image)

### 2.4.2. Predictive Performance for Major Cities

To further investigate the spatial predictive ability of our model, we evaluated the model performance for 5 major cities, 4 provincial capital cities (Jinan, Nanjing, Hangzhou, and Hefei), and Shanghai. The sites in these cities did not participate in the model training process, and the mean values of all site observations/model predictions in each city were used as city-level ozone concentrations. The geographical locations of the 5 cities are shown in Figure S4.

### 2.4.3. Statistical Metrics

Statistical metrics included coefficient of determination ($R^2$), RMSE, mean absolute percentage error (MAPE), and mean absolute error (MAE), as well as whether the model can provide accurate peak predictions, which is more important for environmental research. We preformed peak validation for samples with MDA8 ozone concentration $>160$ ug/m$^3$, which is the interim target 1 (IT-1) formulated by the World Health Organization. These peaks occupy approximately 9% of all samples; the probability histogram of site observations can be found in Figure S5. The statistical metrics of peak validation include the hit rate ($H_R$), false alarm ratio ($F_{AR}$), missing rate ($M_R$), and threat score ($T_S$).

### 3. Results and Discussions

#### 3.1. Statistical Description

During 2020 in eastern China, the annual MDA8 ozone measurement was $101 \pm 44$ ug/m$^3$ (mean ± standard deviation), with a median of $97$ ug/m$^3$ and a 90th percentile value of $160$ ug/m$^3$. The monthly ozone measurement ranged from $52 \pm 20$ to $136 \pm 40$ ug/m$^3$, and was the highest in May and the lowest in December (Figure S6a). Spatially, the annual measurements of Shandong, Jiangsu, Anhui, Shanghai, and Zhejiang were $107 \pm 48$, $103 \pm 44$, $97 \pm 40$, $99 \pm 39$, and $94 \pm 40$, respectively. The overall spatial pattern was high in the north and low in the south (Figure S6b).

We matched 89,331 pairs of variables by monitoring sites for model building, and collected 13,104,264 pairs of variables for ozone data grid generation. Statistical descriptions of these variables can be found in Table S2.
3.2. Overall Fitting Results

Figure 4 displays the results of the three CV methods. The CV-R^2 (CV-RMSE) values of sample-based (Figure 4a), space-based (Figure 4b), and time-based (Figure 4c) CV were 0.94 (10.26 ug/m^3), 0.91 (12.79 ug/m^3), and 0.83 (17.74 ug/m^3), respectively. The results indicate that the ozone estimations of our model are highly consistent with ground-level measurements. Note that compared with sample-based CV, the performance of space-based and time-based CV had a certain decline. This problem is widespread in machine learning based air pollution modeling, mainly because of the unsatisfactory generalization performance of data-driven models. However, our model’s spatiotemporal prediction ability reached an acceptable level, and statistically surpassed some recent studies. The Ts (HR) values of sample-based, space-based, and time-based CV were 0.76 (0.85), 0.69 (0.81), and 0.59 (0.71), respectively, indicating the model has good ability to predict peaks, which is important for air pollution studies, especially when assessing human exposure levels.

![Figure 4. Density scatter plot of model predictions and in situ observations: (a) sample-based CV; (b) space-based CV; and (c) time-based CV.](image-url)

3.3. Spatiotemporal Variations of Model Performance

The spatial distributions of R^2 values of the three CV methods are shown in Figure 5. In sample-based (Figure 5a) and space-based CV (Figure 5b), sites with R^2 values greater than 0.9 accounted for ~86 and ~80%, respectively. In time-based CV (Figure 5c), R^2 values exceeded 0.8 at ~80% of sites. The performance of our model was relatively poor in the southwest mountainous and northeast coastal areas, mainly because of the sparse distribution of monitoring sites in these regions. On the individual month level (Figure 6), the model performed best in summer and worst in winter. The R^2 values ranged from 0.91 to 0.97 in sample-based CV and from 0.86 to 0.95 in space-based CV. In time-based CV, the model performance fluctuated more, and the R^2 value was highest in June (0.90) and lowest in December (0.72). In general, there was no obvious spatiotemporal nonstationarity in our model’s performance; no exceptionally low R^2 values were shown in the three CV methods for each site and month. In addition, the mountainous and northeastern coastal areas are relatively sparsely populated, and summer is often the season with the highest ozone concentration, which further reduces the disadvantage of our model for environmental research.

3.4. Prediction for Major Cities

The time-series diagram (Figure 7) shows that the model’s predictions were in good agreement with observations in all cities. For Jinan, Nanjing, Hangzhou, Hefei, and Shanghai, the R^2 values were 0.89, 0.87, 0.84, 0.83, and 0.83, and the RMSE values predicted by our model were 18.25, 16.62, 13.84, 19.09, and 16.07 ug/m^3, respectively. The Jinan area had the lowest MAPE of 17.12% and the Hefei area had the highest MAPE of 23.18%, but
the performance was still very good. The results indicate that our model can provide robust spatial interpolation predictions.

3.3. Spatiotemporal Variations of Model Performance

The spatial distributions of R² values of the three CV methods are shown in Figure 5. In sample-based (Figure 5a) and space-based CV (Figure 5b), sites with R² values greater than 0.9 accounted for ~86 and ~80%, respectively. In time-based CV (Figure 5c), R² values exceeded 0.8 at ~80% of sites. The performance of our model was relatively poor in the southwest mountainous and northeast coastal areas, mainly because of the sparse distribution of monitoring sites in these regions. On the individual month level (Figure 6), the model performed best in summer and worst in winter. The R² values ranged from 0.91 to 0.97 in sample-based CV and from 0.86 to 0.95 in space-based CV. In time-based CV, the model performance fluctuated more, and the R² value was highest in June (0.90) and lowest in December (0.72). In general, there was no obvious spatiotemporal nonstationarity in our model’s performance; no exceptionally low R² values were shown in the three CV methods for each site and month. In addition, the mountainous and northeastern coastal areas are relatively sparsely populated, and summer is often the season with the highest ozone concentration, which further reduces the disadvantage of our model for environmental research.

Figure 5. R² value of sites in (a) sample-based CV, (b) space-based CV, and (c) time-based CV.

Figure 6. Individual month CV results.
by our model were 18.25, 16.62, 13.84, 19.09, and 16.07 ug/m³, respectively. The Jinan area had the lowest MAPE of 17.12% and the Hefei area had the highest MAPE of 23.18%, but the performance was still very good. The results indicate that our model can provide robust spatial interpolation predictions.

Figure 7. Time series plots of predictions and observations by our model in Jinan, Nanjing, Hangzhou, Hefei, and Shanghai.

4. Discussions

4.1. Comparisons

We generated the daily ozone data grid for 2020 by using the optimal model in sample-based CV, and the fitting results are shown in Figure S7. Two published datasets, China High Air Pollutants (CHAP) and Tracking Air Pollution in China (TAP), were selected to contrast the consistency. CHAP ozone was estimated based on the spatiotemporal extremely randomized trees model, the performance of which in eastern China (sample-based $R^2 = 0.93$) is similar to our results (sample-based $R^2 = 0.94$) [60]. TAP ozone was
generated using a chemical transport model and the data fusion method with $R^2$ equal to 0.7 nationwide [61]. First, the quarterly averages (Figure 8) showed a high degree of consistency between our data, CHAP, TAP, and site observations (the errors of daily averages can be seen in Figure 9), and the predicted ozone showed coherent spatial distribution without visible boundaries caused by model overfitting, which confirms the reasonability of our results. Second, daily comparisons (Figure 10) showed that our data have mainly two advantages: (i) there were no obvious interpolation centers, and the predictions in locations of monitoring sites were more consistent with the surrounding data; and (ii) more detail on ozone distributions was provided due to the higher spatial resolution.

Figure 8. Quarterly averages of MDA8 ozone of CHAP, TAP, our predictions, and site observations.
Figure 9. Errors between different ozone datasets and ground measurements.

Figure 10. Daily distribution of MDA8 ozone of CHAP, TAP, our predictions, and site observations.

With regard to the statistical metrics of the models, we compared our model (CNN) with the two sub-networks and three machine learning models, namely the deep neural network (DNN), random forest (RF), and light gradient-boosting machine (LGBM) models. DNN, RF, and LGBM are widely used and easy to deploy, and we built them using TensorFlow 2.4, SciKit-learn 0.24.2, and LightGBM 3.2.1, respectively. The CV results and main parameter settings of these models are summarized in Table 1, and the density scatter plots can be found in Figure S8. As shown in Table 1, the performance of CNN was superior in comparison to the other models, with the $R^2$ being increased by 3–8%, 1–5%, and 2–4% for sample-based, space-based, and time-based CV, respectively.
Table 1. Statistical metrics of models’ CV results.

| Model   | Parameters | Sample-Based CV |        |        |        | Space-Based CV |        |        |        | Time-Based CV |        |        |
|---------|------------|----------------|--------|--------|--------|----------------|--------|--------|--------|----------------|--------|--------|
|         |            | R²   | RMSE | Tₕ    |        | R²   | RMSE | Tₕ    | R² | RMSE | Tₕ |
| RF      | 350/31/1/2 | 0.87 | 15.56 | 0.71  | 0.87  | 15.58 | 0.71 | 0.81  | 18.68 | 0.67 |
| LGBM    | gbdt/800/26/0.04/21 | 0.91 | 12.78 | 0.76  | 0.90  | 13.99 | 0.73 | 0.80  | 19.58 | 0.65 |
| DNN     | 512/128/80/32/1 | 0.87 | 15.57 | 0.72  | 0.86  | 16.08 | 0.71 | 0.81  | 19.02 | 0.68 |
| Sub-network 1 | - | 0.92 | 12.29 | 0.81  | 0.88  | 15.07 | 0.77 | 0.81  | 18.88 | 0.71 |
| Sub-network 2 | - | 0.91 | 12.72 | 0.80  | 0.90  | 13.40 | 0.73 | 0.80  | 19.33 | 0.67 |

1 RF parameters correspond to number of weak estimators, maximum depth, minimum samples required to split, and minimum samples required to be at a leaf node; LGBM parameters correspond to boosting type, number of weak estimators, maximum depth, number of leaves, learning rate, and minimum number of samples in leaf node; DNN parameters correspond to number of units/neurons in each layer.

4.2. Uncertainty Analyses

4.2.1. Training Sample Size

The number of training samples is among the determining factors for the performance of a data-driven model. In the above validation, we adopted a 10-fold CV method, where training data accounted for 90% of the total samples. Here, we conducted 2-fold to 9-fold CV, exploring the sensitivity of our model to the volume of training data (Figure S9). With an increase in training samples, the results of the three CV strategies slightly improved, with R² value increments from 0.932 to 0.943, 0.892 to 0.911, and 0.804 to 0.824 for sample-based, space-based, and time-based CV, respectively. The performance of our model remained high even if the training dataset was the same size as the testing dataset, suggesting that the influence of the amount of training data on the model performance is limited.

4.2.2. Site Density

A sampling radius of 0.25° was used to calculate the spatial density of sites in the study area. The calculated spatial density ranged from 0.9 to 21.8 (Figure S10a), and we divided the sites into 22 groups with equal spacing of 1. As shown in Figure S10b, model accuracy improved rapidly in the first three groups, and then began to oscillate slightly, suggesting that when the spatial density of the site is greater than 3, the performance of the model does not heavily depend on that.

4.2.3. Spatiotemporal Sampling Size

Spatiotemporal sampling size determines the amount of information contained in the inputs. For sub-network 1, we tested a spatial sampling size range from 3 × 3 to 13 × 13 (Figure S11a). Our experimental results show that the model performance remained stable when the spatial sampling size was 7 × 7, which corresponds to 0.35° × 0.35° geographic space. For sub-network 2, we tested time steps ranging from 2 to 13. As shown in Figure S11b, when the time steps were greater than 7, the R² value of the three kinds of CV methods no longer increased significantly, and 10 time steps was finally selected in this paper.

4.2.4. Configurations of the Sub-Networks

For sub-network 1, we used the CBAM for feature refinement. Although the algorithm is widely used in computer vision, it has not been reported to be applied in atmospheric pollutant estimation. We compared 2-D CNN without CBAM with sub-network 2; the number of parameters in CBAM (1350) accounted for 2.2% of the total parameters (59,958), but the R² was increased by 3.0, 3.4, and 3.3% in sample-based, space-based, and time-based CV, respectively (Table S3). We think that the spatiotemporal relationship between different variables and ozone is theoretically different, and CBAM can be used for slight correction of the relationship.
For sub-network 2, RNN-based models are generally regarded as more accurate for time-series processing. We compared two widely used RNN-based models, the gated recurrent unit (GRU) [62] and long short-term memory (LSTM) [63] models, with 1-D CNN. Three groups of experiments were carried out to observe the performance of different models with approximately the same number of parameters; we set the number for each group at ~8200, ~20,000, and ~32,000. The structures of the models were roughly the same (i.e., multiple hidden layers for feature extraction and fully connected layers for regression), and there were 10 time steps. As summarized in Table S4, there were no significant differences in the performance of the three models using the same number of parameters. The reason may be that RNN-based models focus on memorizing information of long time series, which is not required for this task (e.g., 1-D CNN can achieve convergence with >7 time steps). Meanwhile, the training speed of RNN-based models is much greater than that of CNN-based models even if the number of parameters is the same, because the recursive algorithm cannot run in parallel. From this, 1-D CNN was determined to be the preferred model for our task.

5. Conclusions

In summary, a regional CNN-based model was developed and a high-resolution maximum daily average 8-h surface ground ozone concentration dataset during 2020 was generated. The model was formed by two sub-networks, which were used to extract the 10-day time series features and 5-day spatiotemporal features. The proposed model achieved high performance, with sample-based, space-based, and time-based CV $R^2$ ($T_S$) values of 0.94 (0.85), 0.91 (0.81), and 0.83 (0.71), respectively. To further evaluate the spatial generalization ability of our model, we tested five cities that were not part of the model training, and the $R^2$ values were 0.89, 0.87, 0.84, 0.83, and 0.83 for Jinan, Nanjing, Hangzhou, Hefei, and Shanghai, respectively. The superiority of the model in terms of accuracy was confirmed by comparing it with commonly used machine learning methods, and the generated ozone data grid was superior to existing datasets in terms of spatial distribution coherence and resolution. This paper presents a novel deep learning method for ozone estimation that can provide more accurate data for atmospheric environment and epidemic research.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/rs14071640/s1, Figure S1: Scatter plot of impute total ozone column: (a) Kriging; (b) ERA5. Figure S2: Imputed samples. Figure S3: Spatiotemporal match methods: (a) 10-day single-point data; (b) 5-day spatiotemporal data. Figure S4: Geographical location of provincial capital cities (Jinan, Nanjing, Hangzhou, and Hefei) and Shanghai. Figure S5: Probability histogram of ozone observations. Figure S6: (a) Monthly and (b) spatial variations of ozone measurements. Figure S7: Model fitting results. Figure S8: Density scatter plots of models' CV results. Figure S9: Validation results of different CV fold values. Figure S10: (a) Spatial density of ground-level monitoring sites and (b) CV results of sites with different densities. Figure S11: Variations of CV $R^2$ values: (a) time steps; (b) spatial sampling sizes. Table S1: Data descriptions. Table S2: Statistical descriptions of input variables. Table S3. Comparison between 2-D CNN and sub-network 2. Table S4. Comparison between 1-D CNN, GRU and LSTM.

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