Supporting Information for:

# An ensemble learning approach for estimating high spatiotemporal resolution of ground-level ozone in the contiguous United States

**Weeberb J. Requia* [a, b]**

**Qian Di[a, c]**

**Rachel Silvern[d]**

**James T. Kelly[e]**

**Petros Koutrakis[a]**

**Loretta J. Mickley[d]**

**Melissa P. Sulprizio[d]**

**Heresh Amini[a, f]**

**Liuhua Shi[a, g]**

**Joel Schwartz[a]**

* Corresponding Author: SGAN 602, Asa Norte, Brasília, DF, 70830-051, Brazil

weeberb.requia@fgv.br

a - Harvard University, Department of Environmental Health, TH Chan School of Public Health

Boston, Massachusetts, United States

b - School of Public Policy and Government, Fundação Getúlio Vargas

Brasília, Distrito Federal, Brazil

c - Research Center for Public Health, Tsinghua University, Beijing, China

d - Harvard University, John A. Paulson School of Engineering and Applied Sciences

Boston, Massachusetts, United States

e - U.S. Environmental Protection Agency, Office of Air Quality Planning & Standards

Research Triangle Park, NC, United States

f - Department of Public Health, Faculty of Health and Medical Sciences, University of Copenhagen

Copenhagen, Denmark

g - Emory University, Gangarosa Department of Environmental Health, Rollins School of Public Health

Atlanta, Georgia, United States

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# Figure S1

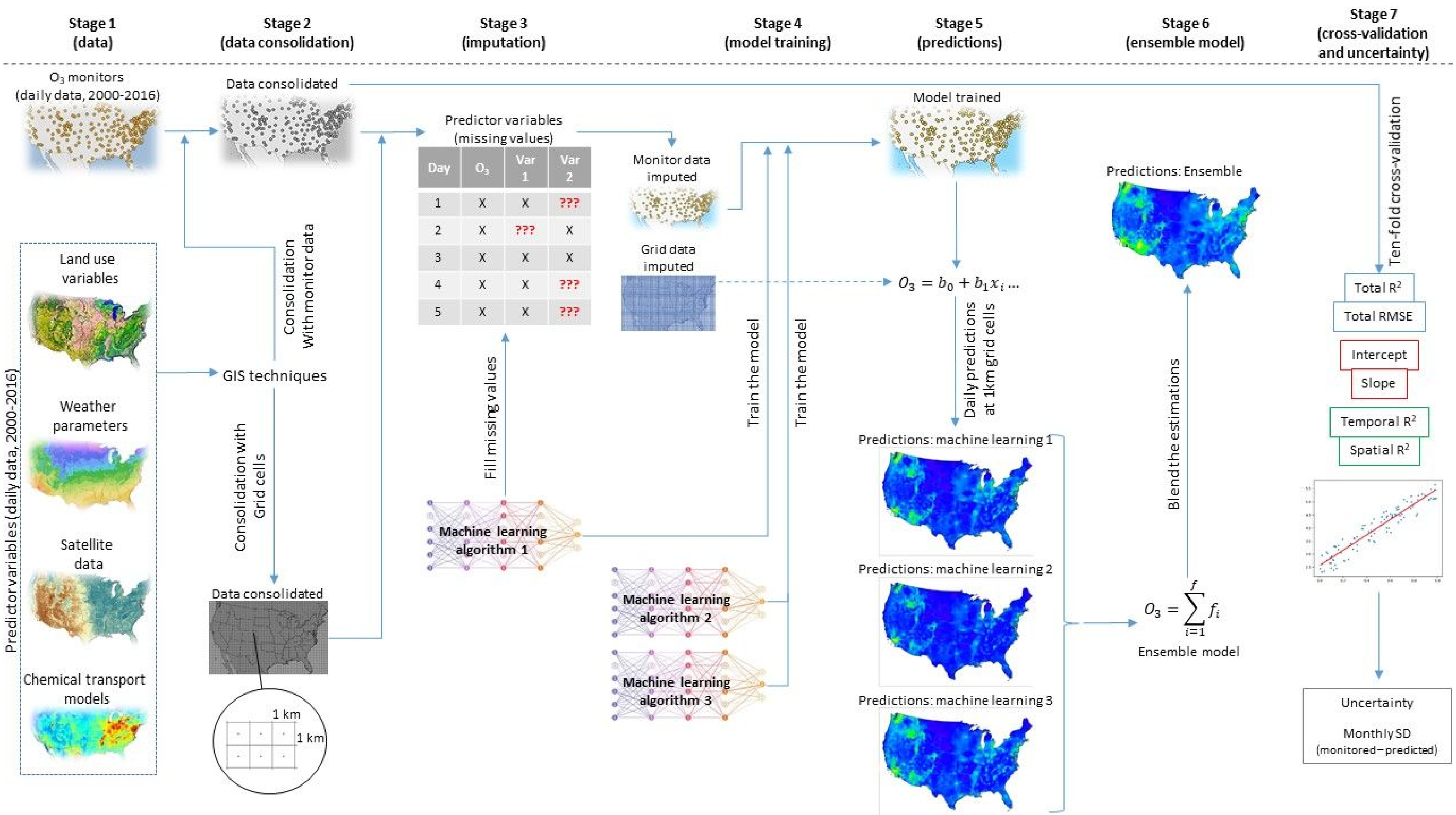


Figure S1 – Study design

## SUPPLEMENTARY MATERIALS 2 (S2)

As we describe in the following sections, we accounted for multi spatial scale 100m, 1km, 10km strategy to capture the predictors. The 1km scale captures predictors on the same scale that we are predicting $O_3$. We include the 10 km scale for some predictors because $O_3$ is a regional pollutant and predictors elsewhere than the grid cell being predicted may be relevant. For example, $NO_2$ and VOC emissions 10km may be relevant, and land use and meteorology predictors are surrogates for such things. Finally, we used a finer scale because some predictors are related to NO emissions which quench $O_3$, and these vary on a fine spatial scale.

### S2. Data source (first stage)

S2.1. $O_3$ ground measurements

We obtained daily maximum 8-hr $O_3$ data from the Environmental Protection Agency (EPA), including the sites from the Air Quality System (AQS) and the sites from the Clean Air Status and Trends Network (CASTNET). In addition to these EPA sites, we also collected data from other regional monitoring datasets. In total, we obtained 2,279 monitoring sites available within the study area during the study period. Note that some monitoring sites did not operate during the entire study period, especially during the winter season. The monitoring sites are not homogenous over the study area. Sites are more densely located in the eastern United States, the industrial Midwest, and the western coast. Figure S2.1 shows the location of the monitoring sites considered in our analysis.

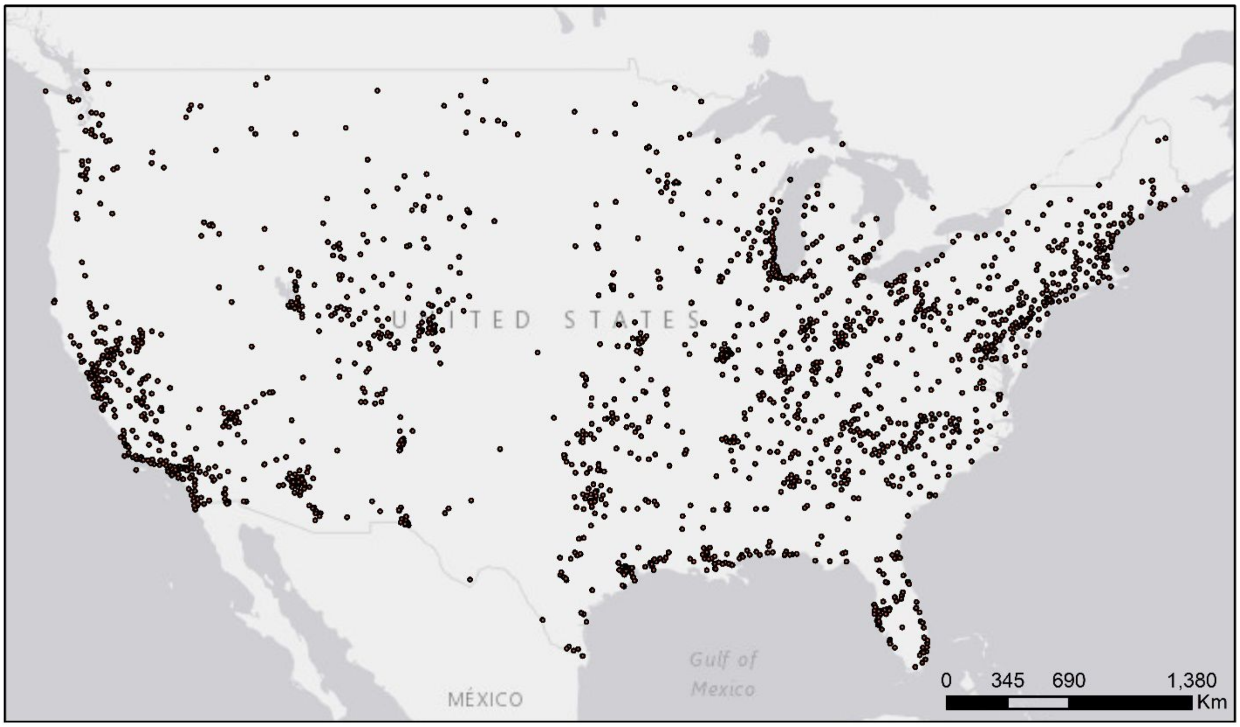


Figure S2.1 – $O_3$ monitoring sites in the United States

S2.2. Meteorological data

Meteorological data were provided by the National Centers for Environmental Prediction (NCEP). The NCEP data is composed by reanalysis datasets from multiple sources, including land-surface monitors, ship, radiosonde, pibal, aircraft, satellite, and other sources. This reanalysis data from NCEP data has high spatio-temporal resolution, which includes daily data with a spatial resolution of 32 km × 32 km over the U.S. The proportion of missing values is relatively low in the dataset [1]. We included 12 groups of meteorological variables, including surface air temperature, accumulated total precipitation, downward shortwave radiation flux at the surface, cloud area fractions, surface albedo, accumulated total evaporation at the surface, planetary boundary layer height, column precipitable water through the troposphere, pressure, specific humidity at 2 m, visibility, and wind speed, which was computed as the vector sum of u-wind (east-west component of the wind) at 10 m and v-wind (north-south component) at 10 m.

S2.3. Chemical transport model and remote sensing data

We used simulation results from two Chemical Transport Models (CTMs) to account for $O_3$ formation, dispersion, and deposition. We also considered CTM predictions for other pollutants in order to represent $O_3$ precursors. Previous studies have used such data to improve the performance of air pollution predictions. Chemical transport simulations represent emissions, transport, chemical reactions, and deposition of pollutants based on state-of-the-science understanding of each of these processes. These mechanistic treatments make CTMs uniquely suited to simulating future and policy scenarios under altered emission conditions in addition to informing statistical modeling studies [2,3]. We also used data based on Remote Sensing (RS) techniques that provide top-down observational constraints for the total column of $O_3$ and its precursors. Note that some chemical transport models also incorporate data from remote sensing to develop model inputs. We describe below the chemical transport and remote sensing data used in our analysis. First, we detail the data used for $O_3$, and then the data used for the $O_3$ precursors.

*S2.3.1. CTM and RS data for $O_3$*

a) GEOS-Chem data:

We used daily simulations of $O_3$ from the GEOS-Chem chemical transport model. This is a global three-dimensional model of tropospheric chemistry based on integrated weather variables from the Goddard Earth Observing System (GEOS) developed by NASA. Full details of the methodology of this model is found in [4]. We performed a nested grid simulation at 0.500° × 0.667°

for North America using boundary conditions from a global model simulation. GEOS-Chem simulates $O_3$ concentrations at different layers through the troposphere. Therefore, to calibrate the tropospheric column of $O_3$ to ground-level $O_3$, we calculated scaling factors as the percentage of surface-level $O_3$ in the total tropospheric column. This approach is similar to that used in modeling $PM_{2.5}$, where aerosol optical depth (AOD) is a column measurement of aerosol and researchers used the vertical profile from a chemical transport model to calibrate AOD to ground-based $PM_{2.5}$ [5,6].

The retrieval algorithm of satellite-based $O_3$ is affected by certain atmospheric factors, including aerosol abundance, surface reflectance, surface albedo, and cloud contamination [7]. To correct possible errors in $O_3$ retrieval, we included in our model variables related to aerosol concentration/aerosol types, cloud coverage, and surface albedo/surface reflectance. We obtained GEOS-Chem variables related to aerosol concentration and aerosol types, which include simulated elemental carbon, organic carbon, sulfate, nitrate, and aerosol mass. The remaining variables used to correct errors in $O_3$ retrieval were obtained from other CTM and RS sources, as described in the next sections. Note that cloud coverage and surface albedo were obtained from the NCEP/NCAR reanalysis dataset, as described above.

b) GEMS data:

We obtained GEMS (Geostationary Environment Monitoring Spectrometer, a satellite-based instrument) $O_3$ data from European Centre for Medium-Range Weather Forecasts (ECMWF). This data is from Copernicus Atmosphere Monitoring Service (CAMS) products that derives GEMS total column $O_3$ at 0.125-degree resolution.

c) OMI data:

We also used tropospheric $O_3$ columns from the Ozone Monitoring Instrument (OMI), an instrument on board the Earth Observing System (EOS)-Aura satellite. The OMI $O_3$ data product is available every day at 13 km × 48 km grid cells. To relate OMI $O_3$ column retrievals to surface-level $O_3$, we used the CTM scaling factors described above.

We also obtained OMI data related to absorbing aerosol index in the ultraviolet and visible ranges (OMAERUVd and OMAEROe). These data are added to the aerosol output from GEOS-Chem to correct possible aerosol-related errors in satellite-based $O_3$ retrieval.

d) CMAQ data:

Daily simulations of $O_3$ were also accessed from the Community Multiscale Air Quality Modeling System (CMAQ). This CTM is a numerical air quality model developed by EPA that simulates the emissions, chemistry and physics of the atmosphere on a 12 km grid. As with GEOS-Chem, we obtained model output from CMAQ related to aerosol concentration and aerosol type, including simulated elemental carbon, organic carbon, sulfate, and nitrate. Note that this was the same set of aerosol data obtained from GEOS-Chem, as described above.

e) MERRA-2 data:

We used total column $O_3$ estimates from Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2). To supplement the aerosol data from GEOS-Chem, OMI, and CMAQ, we also accessed MERRA-2 surface concentrations of sulphate aerosol, hydrophilic black carbon, hydrophobic black carbon, hydrophilic organic carbon, and hydrophobic organic carbon.

f) MODIS data:

We used surface reflectance estimates from MODIS - MOD09A1 [8], which provide estimates of the surface spectral reflectance of TERRA MODIS Bands 1-7 corrected for atmospheric conditions such as gases, aerosols, and Rayleigh scattering. As we described previously, surface reflectance is used to correct possible errors in the $O_3$ retrieval.

*S2.3.2. CTM and RS data for $O_3$ precursors*

As mentioned in the introduction section, $O_3$ formation is based on mechanisms involving photochemical reactions of $O_3$ precursors, including $NO_x$, VOCs, and CO. These precursors are incorporated into the chemical transport simulations through emission inventories. However, the temporal resolution of emissions is limited. To address this limitation, we first used AQS data from U.S. EPA ground monitors (same source used for the $O_3$ data at monitors) to represent daily measurements of $SO_2$, $NO_2$, NOx, and VOCs. Then, we used some chemical and remote sensing data to characterize ozone precursors. These data were accessed from the same sources as described above. We used $NO_2$ concentration from GEOS-Chem model, $NO_2$ column measurement from the OMI satellite instrument (with spatial resolution of 13 km × 24 km), $NO_2$ simulations from CMAQ, and $NO_2$ column concentration simulations from CAMS (Copernicus Atmosphere Monitoring Service), another reanalysis data set.

S2.4. Other predictor variables

The chemical simulation models and data from remote sensing may not capture the very fine spatio-temporal resolution of the atmospheric mechanisms related to $O_3$ formation or removal. Therefore, we considered a set of land use, temporal terms, and some extra variables to represent proxies of the $O_3$ formation or removal. Previous studies have shown that these proxy variables improve the ability to capture the local variation of $O_3$ concentration [2,3,9–11]. The description of these set of variables used in our model is provided below.

*S2.4.1. Land use terms*

We accessed land use variables at 30 m resolution from the National Land Cover Database – NLCD [12]. This database includes water bodies, developed areas, urban areas, barren land, forest, shrub land, herbaceous land, planted/cultivated land, and wetlands. We calculated the proportion of each land-use type in grid cells with 100 m, 1 km, and 10 km horizontal resolution. As a complement for vegetation areas, we also accounted for Normalized Difference Vegetation Index (NDVI). We accessed NDVI data from the MODIS data product MOD13A2 at 1 km × 1 km level (https://cmr.earthdata.nasa.gov/search/concepts/C194001238-LPDAAC_ECS.html). Finally, as an additional proxy variable for local air pollution emissions, we included restaurant density in the model. We obtained the location of restaurants from the U.S. historical business data [13], and then we calculated weighted restaurant density in each 1 km × 1 km grid cell. The weight was based on the amount of emissions, approximated by the number of seats.

*S2.4.2. Elevation*

We accounted for different metrics of elevation, including minimum elevation, maximum elevation, mean elevation, median elevation, standard deviation of elevation, and break line emphasis. We aggregated the data from its original 7.5-arc-second spatial resolution to three different spatial resolution – 100 m × 100 m, 1 km × 1 km, and 10 km × 10 km. These three spatial resolutions were included in the model as separate predictor variables. This data was provided by the Global Multi-Resolution Terrain Elevation Dataset [14].

*S2.4.3. Transportation*

Traffic emissions are important sources of $O_3$ precursors [15,16]. We considered two variables as traffic emission proxies – road density and traffic count. Road density was obtained from the US Census Bureau. The data accessed includes shapefiles representing all roads in the USA. We calculated the spatial density (total length of road in each grid cell) for each 100 m × 100 m, 1 km × 1 km, and 10 km × 10 km grid cell (as for elevation, these three different spatial scales were included in the model as separate predictors). Annual average traffic count data for the contiguous U.S. was provided by ArcGIS Online. We interpolated the original data to 100 m, 1 km, and 10 km spatial resolution.

*S2.4.4. Temporal terms*

To improve the detection of the temporal variation of $O_3$, we accounted for 26 temporal predictor variables. We included in the model 16 dummy variables representing the years (2000-2016), 6 dummy variables for the weekdays, 1 variable representing the Julian days, and 2 variables for the season trends. The variables representing the seasonal patterns were estimated based on sine and cosine functions [17], in which: *sine season = sin(2 x π x doy / 365.24)*; and, *cosine season = cos(2 x π x doy / 365.24)*; where *doy* is the day of year (e.g., 1:365).

*S2.4.5. Spatio-temporal lag of $O_3$ measurements*

We assumed that $O_3$ concentration from nearby monitoring sites are more correlated than from faraway sites, and $O_3$ concentration from neighboring days are more correlated than long ago. These assumptions are based on the spatial and temporal autocorrelation of $O_3$ distributions. Therefore, we included spatially and temporally lagged $O_3$ measurements in the model. We estimated the spatially lagged terms as inverse distance weighted $O_3$ measurements at other locations, as well as their one-day, three-day and five-day lagged moving average values.

*S2.4.6. Temporal lag of several $O_3$ predictors*

We also accounted for temporal lag (1-day lagged moving values) of meteorological variables, including air temperature, total precipitation accumulation, pressure, humidity, and wind speed.

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## SUPPLEMENTARY MATERIALS 3 (S3)

**R script used in the machine learning analyses**

```
########### Ozone model###############
########################################
### Model training - Neural Network ###
########################################
library(h2o)
library(mgcv)
library(parallel)

####################################################################
### Step 1: Set working directory and prepare the dataset ########
####################################################################
##### Set working directory:
setwd("/media/gate/Weeberb/Ozone_model")

##### Open dataset:
InputData <- readRDS("//media/InputData_O3_model.rds")

####################################################################
### Step 2: Grid search - Choose the best model ##################
####################################################################
library(e1071)

##### Define the parameters:
X_Var = c(2:87,94:124,127:length(InputData2))
data1 = InputData2[!is.na(InputData2[,1]),]
set.seed(123)
train_ind <- sample(seq_len(nrow(data1)), size = round(nrow(data1)*0.9))
train_data= data1[train_ind,]

##### Starting H2O:
h2o.init(min_mem_size = "120g",nthreads = 5)

##### Open the data using H2O:
train_h2o<-as.h2o(train_data)
test_data= data1[-train_ind,]
test_h2o<-as.h2o(test_data)

##### Define the parameters
hyper_params=list(epochs=c(40,50,75),hidden=list(c(210,210),c(350,350),c(250,250)),l1=c(1e-
4,1e-5),activation = c("Rectifier"))
```

```
##### Run the model and save the results
modgrid<-h2o.grid("deeplearning",x=X_Var,y=1,epsilon = 1e-08,training_frame =
train_h2o,hyper_params = hyper_params)
saveRDS(modgrid,"grid_search_Neural_Network.rds")

##### Get a list of the highest R2 value:
model_ids <- modgrid@model_ids
models <- lapply(model_ids, function(id) { h2o.getModel(id)})

MaxR2 = 0
MaxID = 0
for(i in 1:18)
{
 TempR = models[[i]]@model$training_metrics@metrics$r2
 if(TempR>MaxR2)
 {
  MaxR2 = TempR
  MaxID = i
 }
 cat(sprintf("%d %f\n",i,TempR))
}



##########################################################################
### Step 3: Run the model - Neural Network ######################
##########################################################################
##### Start H2O:
h2o.init(min_mem_size = "120g",nthreads = 5)
#h2o.init(nthreads=-1,max_mem_size = "400G",port = (54321))    #### To start H2O on
Odyssey

##### Open the data using H2O:
InputData_h2o <- as.h2o(InputData)

### Set "X" variables:
#X_Var_1 = c(2:87,94:124,127:128,130:137,139:length(InputData2))
X_Var_1 = c(2:112, 114:121, 123:length(InputData))

names(InputData)

## Run the model and save the results:
mod_nn_1 <- h2o.deeplearning(x = X_Var_1, # column numbers for predictors
                             y = 1, # column number for label
                             training_frame = InputData_h2o,nfolds=10,standardize = FALSE,
```

```
                    fold_assignment="Modulo",seed=271828,keep_cross_validation_predictions
= TRUE,
                    activation="Rectifier",hidden=c(250,250),epochs=50,
                    epsilon = 1e-08,l1=1e-05,distribution="AUTO")

h2o.saveModel(mod_nn_1,force = TRUE,
"2_Outcome/1_ModelTraining_Neural_Network/2_Model")

summary(mod_nn_1)



########### Ozone model###############
########################################
### Model training - Random Forest ###
########################################
library(h2o)
library(mgcv)
library(parallel)

#####################################################################
### Step 1: Set working directory and prepare the dataset ########
#####################################################################
##### Set working directory:
setwd("/media/gate/Weeberb/Ozone_model")

##### Open dataset:
InputData <- readRDS("//media/InputData_O3_model.rds")

#####################################################################
### Step 2: Grid search - Choose the best model #################
#####################################################################
##### Define the parameters:
X_Var = c(2:87,94:124,127:length(InputData2))
data1 = InputData2[!is.na(InputData2[,1]),]
set.seed(123)
train_ind <- sample(seq_len(nrow(data1)), size = round(nrow(data1)*0.9))
train_data= data1[train_ind,]

##### Starting H2O:
h2o.init(min_mem_size = "120g",nthreads = 5)

##### Open the data using H2O:
train_h2o<-as.h2o(train_data)
test_data= data1[-train_ind,]
test_h2o<-as.h2o(test_data)
```

```
##### Define the parameters
grid_space <- list()
grid_space$ntrees <- c(800,1000,1200)
grid_space$max_depth <- c(7,9,11)
grid_space$nbins <- c(15,20,24)
grid_space$nbins_cats <- c(400,449,500)
grid_space$sample_rate <- c(0.4,0.5)#0.415836

##### Run the model and save the results
modgrid_rf <- h2o.grid("randomForest", grid_id="drf_grid_cars_test", x=X_Var, y=1,
                  training_frame=train_h2o,validation_frame = test_h2o,hyper_params=grid_space)

saveRDS(modgrid_rf,"grid_search_Random_Forest.rds")

##### Get a list of the highest R2 value:
model_ids <- modgrid_rf@model_ids
models <- lapply(model_ids, function(id) { h2o.getModel(id)})

MaxR2 = 0
MaxID = 0
for(i in 1:162)
{
  TempR = models[[i]]@model$training_metrics@metrics$r2
  if(TempR>MaxR2)
  {
    MaxR2 = TempR
    MaxID = i
  }
  cat(sprintf("%d %f ntrees:%d max_depth:%d nbins:%d nbins_cats:%d
sample_rate:%f\n",i,TempR,models[[i]]@parameters$ntrees,models[[i]]@parameters$max_dept
h,models[[i]]@parameters$nbins,models[[i]]@parameters$nbins_cats,models[[i]]@parameters$s
ample_rate))
}


######################################################################
### Step 3: Run the model - Random Forest ########################
######################################################################
##### Start H2O:
h2o.init(min_mem_size = "120g",nthreads = 5)
#h2o.init(nthreads=-1,max_mem_size = "400G",port = (54321))    #### To start H2O on
Odyssey

##### Open the data using H2O:
InputData_h2o <- as.h2o(InputData)
```

```
### Set "X" variables:
#X_Var_1 = c(2:87,94:124,127:128,130:137,139:length(InputData2))
X_Var_1 = c(2:112, 114:121, 123:length(InputData))
names(InputData)

## Run the model and save the results:
mod_rf_1 <-h2o.randomForest(x = X_Var_1,
                     y = 1,
                     training_frame = InputData_h2o,nfolds=10,
                     fold_assignment="Modulo",seed=271828,keep_cross_validation_predictions =
TRUE,
                     ntrees=800,max_depth = 9,nbins = 15,nbins_cats = 449,sample_rate = 0.5)

h2o.saveModel(mod_rf_1,force = TRUE,
"2_Outcome/2_ModelTraining_Random_Forest/2_Model")

summary(mod_rf_1)


########### Ozone model##################
############################################
### Model training - Gradient Boosting ###
############################################
library(h2o)
library(mgcv)
library(parallel)


#################################################################
### Step 1: Set working directory and prepare the dataset ########
#################################################################
##### Set working directory:
setwd("/media/gate/Weeberb/Ozone_model")

##### Open dataset:
InputData <- readRDS("//media/InputData_O3_model.rds")

#################################################################
### Step 2: Grid search - Choose the best model ##################
#################################################################
##### Define the parameters:
X_Var = c(2:87,94:124,127:length(InputData2))
data1 = InputData2[!is.na(InputData2[,1]),]
set.seed(123)
train_ind <- sample(seq_len(nrow(data1)), size = round(nrow(data1)*0.9))
train_data= data1[train_ind,]
```

```
##### Starting H2O:
h2o.init(min_mem_size = "120g",nthreads = 5)

##### Open the data using H2O:
train_h2o<-as.h2o(train_data)
test_data= data1[-train_ind,]
test_h2o<-as.h2o(test_data)

##### Define the parameters
xgb_params1 <- list(learn_rate = c(0.01,0.005,0.007),
                    max_depth = c(6,7,8),
                    sample_rate = c(1.0),
                    col_sample_rate = c(0.4,0.5,0.6),
                    ntrees = c(175,200,250))

##### Run the model and save the results
modgrid_xgb <- h2o.grid("gbm", x = X_Var, y = 1,
                        grid_id = "xgb_params1",
                        training_frame = train_h2o,validation_frame = test_h2o,
                        seed = 1,
                        hyper_params = xgb_params1)

saveRDS(modgrid_xgb,"grid_search_Gradient_boosting.rds")

##### Get a list of the highest R2 value:
model_ids <- modgrid_xgb@model_ids
models <- lapply(model_ids, function(id) { h2o.getModel(id)})
MaxR2 = 0
MaxID = 0
for(i in 1:81)
{
  TempR = models[[i]]@model$training_metrics@metrics$r2
  if(TempR>MaxR2)
  {
    MaxR2 = TempR
    MaxID = i
  }
  cat(sprintf("%d %f\n",i,TempR))
}
```

```
#######################################################################
### Step 3: Run the model - Gradient Boosting#####################
#######################################################################

##### Start H2O:
h2o.init(min_mem_size = "120g",nthreads = 5)
#h2o.init(nthreads=-1,max_mem_size = "400G",port = (54321))    #### To start H2O on
Odyssey

##### Open the data using H2O:
InputData_h2o <- as.h2o(InputData2)

### Set "X" variables:
#X_Var_1 = c(2:87,94:124,127:128,130:137,139:length(InputData2))
X_Var_1 = c(2:112, 114:121, 123:length(InputData))
names(InputData)

## Run the model and save the results:
mod_gbm_1=h2o.gbm(x = X_Var_1,
            y = 1,
            training_frame = InputData_h2o,nfolds=10,
            fold_assignment="Modulo", seed=271828,keep_cross_validation_predictions =
TRUE,
            ntrees=200,learn_rate = 0.007,max_depth = 7,sample_rate = 1,col_sample_rate = 0.5)

h2o.saveModel(mod_gbm_1,force = TRUE,
"2_Outcome/3_ModelTraining_Gradient_Boosting/2_Model")

summary(mod_gbm_1)
```

```
##########################################
### Cross-validation     #####################
##########################################


library(h2o)
library(caret)
library(mgcv)
library(parallel)


for(YEAR in 2015:2016)
{
########### for training #######################
#DirPath = "D:\\Google Drive\\Research\\USTemperature\\"
 DirPath = "/nfs/bigdata_nobackup/a/airpred_d_scratch/"
GCS = "North_America_Equidistant_Conic"
Sep = "/"
VariableID = 99939
NAME = "O3"
EPLISON = 1/1000# change for ozone, NO2.
STARTDATE = as.Date(paste0(YEAR,"-01-01"))
ENDDATE = as.Date(paste0(YEAR,"-12-31"))
N_Core = 61
OPTION = "training"#"prediction"#"training"
SiteName_Train = "AQRVO3"
SiteName_Predict = "AQRVO3"

################ code
source("ModelFunctions.R")

## path
DirPath_Assembled = paste0(DirPath,"assembled_data",Sep)
DirPath_Processed = paste0(DirPath,"processed_data",Sep)
DirPath_Model =
paste0(DirPath,"assembled_data",Sep,"training",Sep,NAME,"_",VariableID,"_CV",Sep)
dir.create(DirPath_Model)
## read location
SiteData_Train<-
ReadLocation(paste0(DirPath_Processed,SiteName_Train,Sep,"Location",Sep,SiteName_Train,"
Site_",GCS))
N_Site_Train <- nrow(SiteData_Train)
SiteData_Predict<-
ReadLocation(paste0(DirPath_Processed,SiteName_Predict,Sep,"Location",Sep,SiteName_Predi
ct,"Site_",GCS))
N_Site_Predict <- nrow(SiteData_Predict)
```

```
## time
N_Day = as.numeric(ENDDATE - STARTDATE + 1)

## read weight
Weight1 =
h5read_robust(paste0(DirPath_Processed,SiteName_Predict,Sep,"Temp",Sep,"SpatialLaggedWe
ightPeak41_",SiteName_Train,"_",SiteName_Predict,".h5"),name = "Weight")
Weight2 =
h5read_robust(paste0(DirPath_Processed,SiteName_Predict,Sep,"Temp",Sep,"SpatialLaggedWe
ightPeak42_",SiteName_Train,"_",SiteName_Predict,".h5"),name = "Weight")
Weight3 =
h5read_robust(paste0(DirPath_Processed,SiteName_Predict,Sep,"Temp",Sep,"SpatialLaggedWe
ightPeak43_",SiteName_Train,"_",SiteName_Predict,".h5"),name = "Weight")

## start h2o vm
h2o.init(min_mem_size = "300g")



## read imputed data
if(file.exists(paste0(DirPath_Model,"InputDataImputed.rds")))
{
  InputData = readRDS(paste0(DirPath_Model,"InputDataImputed.rds"))
  InputData_h2o <- as.h2o(InputData)
}else
{
  ## if not, read non-imputed data
  if(file.exists(paste0(DirPath_Model,"InputData.rds")))
  {
    InputData = readRDS(paste0(DirPath_Model,"InputData.rds"))
  }else
  {
    ## if not again, read data from scratch
    # ##read csv configuration file
    col = c(rep("character",6))
    col[c(2,3)] = "logical"
    col[c(7,8)] = "numeric"
    VariableList =
read.csv(paste0(DirPath,"assembled_data",Sep,"VariableList_",VariableID,".csv"),colClasses =
col)
    VariableList = VariableList[!is.na(VariableList$READ),]
    InputData =
ReadData(DirPath,Sep,VariableID,NAME,SiteName_Train,STARTDATE,ENDDATE,OPTION
,SiteData_Train,VariableList)
    saveRDS(InputData,paste0(DirPath_Model,"InputData_Original.rds"))
```

```
    InputData <- StandardData(DirPath,InputData,VariableID)
    InputData$MonitorData = log(InputData$MonitorData+EPLISON)
    saveRDS(InputData,paste0(DirPath_Model,"InputData.rds"))

  }
  ## imputation
  InputData_h2o <- as.h2o(InputData)
  InputData_h2o =
ImputeData(DirPath,Sep,InputData_h2o,InputData,OPTION,VariableID,SiteData_Train)
  InputData = as.data.frame(InputData_h2o)
  saveRDS(InputData,paste0(DirPath_Model,"InputDataImputed.rds"))
}

## for CV
set.seed(123)
flds <- createFolds(seq(1:nrow(SiteData_Predict)), k = 10, list = TRUE, returnTrain = FALSE)
saveRDS(flds,paste0(DirPath_Model,"CV.rds"))

sink(file=paste0(DirPath_Model,paste0("testH2o_train_output_CV_"),STARTDATE,"_",ENDD
ATE,".txt"),append=T,split=F)

#######################
for(m in 1:10)
{
  if(file.exists(paste0(DirPath_Model,"OutputData","_round",m,".rds")))
  {
    next
  }

  Index_train = which(is.element(data$SiteCode,SiteData[-flds[[m]],"SITECODE"]))
  Index_test = which(is.element(data$SiteCode,SiteData[flds[[m]],"SITECODE"]))

  # variables used in step 1
  X_Var_1 = which(!names(InputData_h2o) %in% c("MonitorData","SiteCode",
"CalendarDay","PM25_Region","NO2_Region","Ozone_Region","Temporal_Lagged_1","Temp
oral_Lagged_2","Temporal_Lagged_3","Spatial_Lagged_1","Spatial_Lagged_2","Spatial_Lagg
ed_3"))

  #step 1: neural network
  TempDir = paste0(paste0(DirPath_Model,"NeuralNetwork_Step1","_round",m,Sep))
  if(length(list.files(TempDir))>0)
  {
    if(length(list.files(TempDir))>1)
    {
      stop("more than one model here exist!",TempDir)
    }
```

```
    Model <- list.files(TempDir)
    mod_nn_1 <- h2o.loadModel(paste0(TempDir,Model[1]))
  }else
  {
    mod_nn_1 <- h2o.deeplearning(x = X_Var_1, # column numbers for predictors
                              y = 1, # column number for label
                              training_frame = InputData_h2o[Index_train,],nfolds=10,standardize =
FALSE,

fold_assignment="Modulo",seed=271828,keep_cross_validation_predictions = TRUE,
                              activation="Rectifier",hidden=c(210,210),epochs=50,
                              epsilon = 1e-08,l1=1e-05,distribution="AUTO")
    h2o.saveModel(mod_nn_1,force = TRUE,TempDir)
  }
  #step 1: random forest
  TempDir = paste0(paste0(DirPath_Model,"RandomForest_Step1","_round",m,Sep))
  if(length(list.files(TempDir))>0)
  {
    if(length(list.files(TempDir))>1)
    {
      stop("more than one model here exist!",TempDir)
    }
    Model <- list.files(TempDir)
    mod_rf_1 <- h2o.loadModel(paste0(TempDir,Model[1]))
  }else
  {
    mod_rf_1=h2o.randomForest(x = X_Var_1,
                           y = 1,
                           training_frame = InputData_h2o[Index_train,],nfolds=10,
                           fold_assignment="Modulo",seed=271828,keep_cross_validation_predictions
= TRUE,
                           ntrees=1000,max_depth = 9,nbins = 20,nbins_cats = 449,sample_rate =
0.41536)
    h2o.saveModel(mod_rf_1,force = TRUE,TempDir)
  }
  ## Step 1: gradient boosting
  TempDir = paste0(paste0(DirPath_Model,"GradientBoosting_Step1","_round",m,Sep))
  if(length(list.files(TempDir))>0)
  {
    if(length(list.files(TempDir))>1)
    {
      stop("more than one model here exist!",TempDir)
    }
    Model <- list.files(TempDir)
    mod_gbm_1 <- h2o.loadModel(paste0(TempDir,Model[1]))
  }else
```

```
 {
  mod_gbm_1=h2o.gbm(x = X_Var_1,
                   y = 1,
                   training_frame = InputData_h2o[Index_train,],nfolds=10,
                   fold_assignment="Modulo", seed=271828,keep_cross_validation_predictions =
TRUE,
                   ntrees=200,learn_rate = 0.007,max_depth = 7,sample_rate = 1,col_sample_rate =
0.5)
  h2o.saveModel(mod_gbm_1,force = TRUE,TempDir)
 }

 ## ensemble
 InputData$pred_nn_1<-as.vector(h2o.predict(mod_nn_1,newdata=InputData_h2o)$predict)
 InputData$pred_gbm_1<-as.vector(h2o.predict(mod_gbm_1,newdata=InputData_h2o)$predict)
 InputData$pred_rf_1<-as.vector(h2o.predict(mod_rf_1,newdata=InputData_h2o)$predict)

 if(file.exists(paste0(DirPath_Model,"Ensemble_Step1","_round",m,".rds")))
 {
  mod_ensemble_1 <- readRDS(paste0(DirPath_Model,"Ensemble_Step1","_round",m,".rds"))
 }else
 {
  cl <- makeCluster(N_Core)
  mod_ensemble_1<-bam(MonitorData ~ s(Other_Lat, Other_Lon,
by=pred_nn_1)+s(Other_Lat, Other_Lon, by=pred_gbm_1)+s(Other_Lat, Other_Lon,
by=pred_rf_1),data=InputData[Index_train,],cluster=cl)
  stopCluster(cl)
  saveRDS(mod_ensemble_1,paste0(DirPath_Model,"Ensemble_Step1","_round",m,".rds"))
 }

 ## show results
 Pred<-predict(mod_ensemble_1,newdata = InputData)
 InputData$pred_ensemble_1 = Pred
 A = summary(lm(MonitorData~pred_ensemble_1,data=InputData[Index_test,]))
 print(sprintf("Step1: Ensemble:%f,%f\n",A$r.squared,sqrt(mean(A$residuals^2))))
 A = summary(lm(MonitorData~pred_rf_1,data=InputData[Index_test,]))
 print(sprintf("Step1: Random Forest:%f,%f\n",A$r.squared,sqrt(mean(A$residuals^2))))
 A = summary(lm(MonitorData~pred_gbm_1,data=InputData[Index_test,]))
 print(sprintf("Step1: Gradient Boosting:%f,%f\n",A$r.squared,sqrt(mean(A$residuals^2))))
 A = summary(lm(MonitorData~pred_nn_1,data=InputData[Index_test,]))
 print(sprintf("Step1: Neural Network:%f,%f\n",A$r.squared,sqrt(mean(A$residuals^2))))

 ########################################
 #### two step modeling...
 dim(Pred)<-c(N_Day,N_Site_Train)
 Pred_1 = apply(Pred,2,function(x) as.numeric(filter(x, rep(1/7,7),sides = 2,circular = TRUE)))
```

```
  Pred_2 = apply(Pred,2,function(x) as.numeric(filter(x,
c(1/16,2/16,3/16,4/16,3/16,2/16,1/16),sides = 2,circular = TRUE)))
  Pred_3 = apply(Pred,2,function(x) as.numeric(filter(x,
c(1/44,4/44,9/44,16/44,9/44,4/44,1/44),sides = 2,circular = TRUE)))
  Pred_4 = Pred%*%Weight1
  Pred_5 = Pred%*%Weight2
  Pred_6 = Pred%*%Weight3
  dim(Pred_1)<-c(N_Day*N_Site_Train)
  dim(Pred_2)<-c(N_Day*N_Site_Train)
  dim(Pred_3)<-c(N_Day*N_Site_Train)
  dim(Pred_4)<-c(N_Day*N_Site_Train)
  dim(Pred_5)<-c(N_Day*N_Site_Train)
  dim(Pred_6)<-c(N_Day*N_Site_Train)

  Temp_h2o = as.h2o(as.data.frame(cbind(Pred_1,Pred_2,Pred_3,Pred_4,Pred_5,Pred_6)))
  colnames(Temp_h2o)<-
c("Temporal_Lagged_1","Temporal_Lagged_2","Temporal_Lagged_3","Spatial_Lagged_1","Sp
atial_Lagged_2","Spatial_Lagged_3")
  InputData_h2o = as.h2o(InputData)
  InputData_h2o = h2o.cbind(InputData_h2o,Temp_h2o)



  ##############################################################
  ### STEP 2
  X_Var_2 = which(!names(InputData_h2o) %in% c("MonitorData","SiteCode",
"CalendarDay","PM25_Region","NO2_Region","Ozone_Region"))

  ## neural network, best choice
  TempDir = paste0(paste0(DirPath_Model,"NeuralNetwork_Step2","_round",m,Sep))
  if(length(list.files(TempDir))>0)
  {
    if(length(list.files(TempDir))>1)
    {
      stop("more than one model here exist!",TempDir)
    }
    Model <- list.files(TempDir)
    mod_nn_2 <- h2o.loadModel(paste0(TempDir,Model[1]))
  }else
  {
    mod_nn_2 <- h2o.deeplearning(x = X_Var_2, # column numbers for predictors
                             y = 1, # column number for label
                             training_frame = InputData_h2o[Index_train,],nfolds=10,standardize =
FALSE,

fold_assignment="Modulo",seed=271828,keep_cross_validation_predictions = TRUE,
```

```
                            activation="Rectifier",hidden=c(210,210),epochs=50,
                            epsilon = 1e-08,l1=1e-05,distribution="AUTO")
   h2o.saveModel(mod_nn_2,force = TRUE,TempDir)
  }

  ## random forest --- the best model
  TempDir = paste0(paste0(DirPath_Model,"RandomForest_Step2","_round",m,Sep))
  if(length(list.files(TempDir))>0)
  {
   if(length(list.files(TempDir))>1)
   {
    stop("more than one model here exist!",TempDir)
   }
   Model <- list.files(TempDir)
   mod_rf_2 <- h2o.loadModel(paste0(TempDir,Model[1]))
  }else
  {
   mod_rf_2 =h2o.randomForest(x = X_Var_2,
                          y = 1,
                          training_frame = InputData_h2o[Index_train,],nfolds=10,
                          fold_assignment="Modulo",seed=271828,keep_cross_validation_predictions
= TRUE,
                          ntrees=1000,max_depth = 9,nbins = 20,nbins_cats = 449,sample_rate =
0.41536)
   h2o.saveModel(mod_rf_2,force = TRUE,TempDir)
  }

  ## Step 2 gradient boosting
  TempDir = paste0(paste0(DirPath_Model,"GradientBoosting_Step2","_round",m,Sep))
  if(length(list.files(TempDir))>0)
  {
   if(length(list.files(TempDir))>1)
   {
    stop("more than one model here exist!",TempDir)
   }
   Model <- list.files(TempDir)
   mod_gbm_2 <- h2o.loadModel(paste0(TempDir,Model[1]))
  }else
  {
   mod_gbm_2 =h2o.gbm(x = X_Var_2,
                  y = 1,
                  training_frame = InputData_h2o[Index_train,],nfolds=10,
                  fold_assignment="Modulo", seed=271828,keep_cross_validation_predictions =
TRUE,
                  ntrees=200,learn_rate = 0.007,max_depth = 7,sample_rate = 1,col_sample_rate =
0.5)
```

```
    h2o.saveModel(mod_gbm_2,force = TRUE,TempDir)
  }

  ## ensemble
  InputData$pred_nn_2<-as.vector(h2o.predict(mod_nn_2,newdata=InputData_h2o)$predict)
  InputData$pred_gbm_2<-as.vector(h2o.predict(mod_gbm_2,newdata=InputData_h2o)$predict)
  InputData$pred_rf_2<-as.vector(h2o.predict(mod_rf_2,newdata=InputData_h2o)$predict)

  if(file.exists(paste0(DirPath_Model,"Ensemble_Step2","_round",m,".rds")))
  {
    mod_ensemble_2 <- readRDS(paste0(DirPath_Model,"Ensemble_Step2","_round",m,".rds"))
  }else
  {
    cl <- makeCluster(N_Core)
    mod_ensemble_2<-bam(MonitorData ~ s(Other_Lat, Other_Lon,
by=pred_nn_2)+s(Other_Lat, Other_Lon, by=pred_gbm_2)+s(Other_Lat, Other_Lon,
by=pred_rf_2),data=InputData[Index_train,],cluster=cl)
    stopCluster(cl)
    saveRDS(mod_ensemble_2,paste0(DirPath_Model,"Ensemble_Step2","_round",m,".rds"))
  }

  ## show results
  Pred<-predict(mod_ensemble_2,newdata = InputData)
  InputData$pred_ensemble_2 = Pred
  A = summary(lm(MonitorData~pred_ensemble_2,data=InputData[Index_test,]))
  print(sprintf("Step2: Ensemble:%f,%f\n",A$r.squared,sqrt(mean(A$residuals^2))))
  A = summary(lm(MonitorData~pred_rf_2,data=InputData[Index_test,]))
  print(sprintf("Step2: Random Forest:%f,%f\n",A$r.squared,sqrt(mean(A$residuals^2))))
  A = summary(lm(MonitorData~pred_gbm_2,data=InputData[Index_test,]))
  print(sprintf("Step2: Gradient Boosting:%f,%f\n",A$r.squared,sqrt(mean(A$residuals^2))))
  A = summary(lm(MonitorData~pred_nn_2,data=InputData[Index_test,]))
  print(sprintf("Step2: Neural Network:%f,%f\n",A$r.squared,sqrt(mean(A$residuals^2))))

  ## save input data and output data
  OutputData =
InputData[,c("CalendarDay","pred_nn_1","pred_gbm_1","pred_rf_1","pred_ensemble_1","pred_
nn_2","pred_gbm_2","pred_rf_2","pred_ensemble_2")]
  saveRDS(OutputData,paste0(DirPath_Model,"OutputData","_round",m,".rds"))
}
saveRDS(OutputData,paste0(DirPath_Model,"OutputData.rds"))
sink()
}
```

**Table S1**

**Table S1 –** List of predictor variables

| |
|---|
| **# Meteorological variables** |
| Accumulated total precipitation |
| Downward Shortwave Radiation on Flux |
| Downward Shortwave Radiation on Flux |
| Accumulated total Evaporation |
| High Cloud Area Fraction |
| Planetary Boundary Layer Height |
| Low Cloud Area Fraction |
| Medium Cloud Area Fraction |
| Precipitable Water for entire atmosphere |
| Visibility |
| Air temperature (surface) |
| Pressure (surface) |
| Specific Humidity at 2m |
| U-wind at 10 m |
| V-wind at 10 m |
| Precipitation rate |
| Latent Heat Flux |
| Sensible Heat Flux |
| Snow Cover |
| Soil Moisture Content |
| Forecast of Total Cloud Cover |
| Upward Longwave Radiation on Flux |
| Omega: A term used to describe vertical motion in the atmosphere |
| Accumulated Snow |
| Cloud coverage |
| Surface albedo |
| |
| **# GEOS-Chem data** |
| Surface-level O3, simulated by GEOS-Chem |
| Surface-level NO2, simulated by GEOS-Chem |
| Elemental carbon - GEOS-Chem variable related to aerosol concentration and aerosol type |
| Organic carbon - GEOS-Chem variable related to aerosol concentration and aerosol type |
| Sulfate - GEOS-Chem variable related to aerosol concentration and aerosol type |
| Nitrate - GEOS-Chem variable related to aerosol concentration and aerosol type |
| Aerosol mass - GEOS-Chem variable related to aerosol concentration and aerosol type |
| |
| **# GEMS data** |
| GEMS total column O3 at 0.125-degree resolution |
| |
| **# OMI satellite data** |
| OMAERUVd_UVA - absorbing aerosol index in the ultraviolet range |
| OMAEROe_UVA - absorbing aerosol index in the ultraviolet range |
| OMAEROe_VISA - absorbing aerosol index in the visible range |
| Satellite-measured column O3 concentration |
| Satellite-measured column SO2 concentration |
| Satellite-measured column NO2 concentration |
| Satellite-measured UV index |
| |
| |

| |
|---|
| **# CMAQ data** |
| Surface-level NO2, simulated by CMAQ |
| Percentage of surface-level NO2 at the total column NO2, simulated by CMAQ |
| Surface-level ozone, simulated by CMAQ |
| Percentage of surface-level ozone at the total column ozone, simulated by CMAQ |
| Surface-level PM2.5 nitrate, simulated by CMAQ |
| Surface-level PM2.5 sulfate, simulated by CMAQ |
| Surface-level PM2.5 elemental carbon, simulated by CMAQ |
| Surface-level PM2.5 organic carbon, simulated by CMAQ |
| |
| **# MERRA** |
| Hydrophilic Black Carbon |
| Hydrophobic Black Carbon |
| Hydrophilic Organic Carbon |
| Hydrophobic Organic Carbon |
| Sulphate aerosol |
| Total column O3 |
| |
| **# MODIS Satellite data** |
| Surface temperature during the day, mean of nearby grid cells |
| Surface temperature at night, mean of nearby grid cells |
| MODIS-measured cloud coverage during the day, mean of nearby grid cells |
| MODIS-measured cloud coverage at night, mean of nearby grid cells |
| Surface temperature during the day, data from the nearest grid cell |
| Surface temperature at night, data from the nearest grid cell |
| MODIS-measured cloud coverage during the day, data from the nearest grid cell |
| MODIS-measured cloud coverage at night, data from the nearest grid cell |
| MAIAC aerosol (aerosol optical depth) data at 470 nm wavelength, from Aqua satellite, retrieved from the nearest grid cell |
| MAIAC aerosol (aerosol optical depth) data at 550 nm wavelength, from Aqua satellite, retrieved from the nearest grid cell |
| MAIAC aerosol (aerosol optical depth) data at 470 nm wavelength, from Terra satellite, retrieved from the nearest grid cell |
| MAIAC aerosol (aerosol optical depth) data at 550 nm wavelength, from Terra satellite, retrieved from the nearest grid cell |
| Viewing angle of the sensor at the Terra satellite |
| Viewing angle of the sensor at the Aqua satellite |
| NDVI value from MODIS MOD13A2, 1 km spatial resolution and 16-day temporal resolution |
| |
| **# CAMS data** |
| NO2 column concentration simulations from CAMS |
| |
| **# Additional air quality data from EPA** |
| Daily measurements of SO2 |
| Daily measurements of NO2 |
| Daily measurements of NOx |
| Daily measurements of VOCs |
| |
| **# NLCD landuse database** |
| Wetland coverage from NLCD data set, the original 30 meter data were aggregated to 10000 meter raster, then we interpolated the 10000-meter raster to locations of interests |
| Water coverage from NLCD data set, the original 30 meter data were aggregated to 10000 meter raster, then we interpolated the 10000-meter raster to locations of interests |

| |
|---|
| Planted coverage from NLCD data set, the original 30 meter data were aggregated to 10000 meter raster, then we interpolated the 10000-meter raster to locations of interests |
| Herbaceous coverage from NLCD data set, the original 30 meter data were aggregated to 10000 meter raster, then we interpolated the 10000-meter raster to locations of interests |
| Shrubland coverage from NLCD data set, the original 30 meter data were aggregated to 10000 meter raster, then we interpolated the 10000-meter raster to locations of interests |
| Barren coverage from NLCD data set, the original 30 meter data were aggregated to 10000 meter raster, then we interpolated the 10000-meter raster to locations of interests |
| Developed area coverage from NLCD data set, the original 30 meter data were aggregated to 10000 meter raster, then we interpolated the 10000-meter raster to locations of interests |
| Wetland coverage from NLCD data set, the original 30 meter data were aggregated to 100 meter raster, then we interpolated the 100-meter raster to locations of interests |
| Water coverage from NLCD data set, the original 30 meter data were aggregated to 100 meter raster, then we interpolated the 100-meter raster to locations of interests |
| Planted coverage from NLCD data set, the original 30 meter data were aggregated to 100 meter raster, then we interpolated the 100-meter raster to locations of interests |
| Herbaceous coverage from NLCD data set, the original 30 meter data were aggregated to 100 meter raster, then we interpolated the 100-meter raster to locations of interests |
| Shrubland coverage from NLCD data set, the original 30 meter data were aggregated to 100 meter raster, then we interpolated the 100-meter raster to locations of interests |
| Barren coverage from NLCD data set, the original 30 meter data were aggregated to 100 meter raster, then we interpolated the 100-meter raster to locations of interests |
| Developed area coverage from NLCD data set, the original 30 meter data were aggregated to 100 meter raster, then we interpolated the 100-meter raster to locations of interests |
| |
| **# Road density obtained from the US Census Bureau** |
| Primary road density; we converted primary road (lines shapefile format) into raster with 100 meter pixel width; then we interpolated them to locations of interests |
| Primary road density; we converted primary road (lines shapefile format) into raster with 1000 meter pixel width; then we interpolated them to locations of interests |
| Primary road density; we converted primary road (lines shapefile format) into raster with 10000 meter pixel width; then we interpolated them to locations of interests |
| Primary and secondary road density; we converted primary and secondary road (lines shapefile format) into raster with 100 meter pixel width; then we interpolated them to locations of interests |
| Primary and secondary road density; we converted primary and secondary road (lines shapefile format) into raster with 1000 meter pixel width; then we interpolated them to locations of interests |
| Primary and secondary road density; we converted primary and secondary road (lines shapefile format) into raster with 10000 meter pixel width; then we interpolated them to locations of interests |
| All road (primary, secondary and terciary road) density; we converted all road (lines shapefile format) into raster with 100 meter pixel width; then we interpolated them to locations of interests |
| All road (primary, secondary and terciary road) density; we converted all road (lines shapefile format) into raster with 1000 meter pixel width; then we interpolated them to locations of interests |
| All road (primary, secondary and terciary road) density; we converted all road (lines shapefile format) into raster with 10000 meter pixel width; then we interpolated them to locations of interests |
| |
| **# Global Multi-Resolution Terrain Elevation Dataset** |
| Maximal elevation, original data was at 7.5 arc-seconds, then aggregated to 100 meter resolution |
| Minimal elevation, original data was at 7.5 arc-seconds, then aggregated to 100 meter resolution |
| Median elevation, original data was at 7.5 arc-seconds, then aggregated to 100 meter resolution |
| Mean elevation, original data was at 7.5 arc-seconds, then aggregated to 100 meter resolution |
| Systematic subsample, original data was at 7.5 arcseconds, then aggregated to 100 meter resolution |
| Breakline emphasis, original data was at 7.5 arc-seconds, then aggregated to 100 meter resolution |
| Standard deviation, original data was at 7.5 arc-seconds, then aggregated to 100 meter resolution |
| Maximal elevation, original data was at 7.5 arc-seconds, then aggregated to 1000 meter resolution |

| |
|---|
| Minimal elevation, original data was at 7.5 arc-seconds, then aggregated to 1000 meter resolution |
| Median elevation, original data was at 7.5 arc-seconds, then aggregated to 1000 meter resolution |
| Mean elevation, original data was at 7.5 arc-seconds, then aggregated to 1000 meter resolution |
| Systematic subsample, original data was at 7.5 arcseconds, then aggregated to 1000 meter resolution |
| Breakline emphasis, original data was at 7.5 arc-seconds, then aggregated to 1000 meter resolution |
| Standard deviation, original data was at 7.5 arc-seconds, then aggregated to 1000 meter resolution |
| Maximal elevation, original data was at 7.5 arc-seconds, then aggregated to 10000 meter resolution |
| Minimal elevation, original data was at 7.5 arc-seconds, then aggregated to 10000 meter resolution |
| Median elevation, original data was at 7.5 arc-seconds, then aggregated to 10000 meter resolution |
| Mean elevation, original data was at 7.5 arc-seconds, then aggregated to 10000 meter resolution |
| Systematic subsample, original data was at 7.5 arcseconds, then aggregated to 10000 meter resolution |
| Breakline emphasis, original data was at 7.5 arc-seconds, then aggregated to 10000 meter resolution |
| Standard deviation, original data was at 7.5 arc-seconds, then aggregated to 10000 meter resolution |
| |
| **# MCD12Q1: a satellite-based landuse types** |
| Water |
| Evergreen Needleleaf forest |
| Evergreen Broadleaf forest |
| Deciduous Needleleaf forest |
| Deciduous Broadleaf forest |
| Mixed forest |
| Closed shrublands |
| Open shrublands |
| Woody savannas |
| Savannas |
| Grasslands |
| Permanent wetlands |
| Croplands |
| Urban and built-up |
| Cropland/Natural Vegetation mosaic |
| Snow and ice |
| Barren or sparsely vegetated |
| Unclassified |
| |
| **# Miscellaneous** |
| Restaurant density |
| Annual average traffic count data for the contiguous U.S. interpolated 100 m |
| Annual average traffic count data for the contiguous U.S. interpolated 1000 m |
| Annual average traffic count data for the contiguous U.S. interpolated 10000 m |
| |
| **# Temporal terms** |
| Dummy variable representing the year 2000 |
| Dummy variable representing the year 2001 |
| Dummy variable representing the year 2002 |
| Dummy variable representing the year 2003 |
| Dummy variable representing the year 2004 |
| Dummy variable representing the year 2005 |
| Dummy variable representing the year 2006 |
| Dummy variable representing the year 2007 |
| Dummy variable representing the year 2008 |
| Dummy variable representing the year 2009 |
| Dummy variable representing the year 2010 |
| Dummy variable representing the year 2011 |

| |
|---|
| Dummy variable representing the year 2012 |
| Dummy variable representing the year 2013 |
| Dummy variable representing the year 2014 |
| Dummy variable representing the year 2015 |
| Dummy variable representing the year 2016 |
| Dummy variable representing the weekday 1 |
| Dummy variable representing the weekday 2 |
| Dummy variable representing the weekday 3 |
| Dummy variable representing the weekday 4 |
| Dummy variable representing the weekday 5 |
| Dummy variable representing the weekday 6 |
| Dummy variable representing the Julian days |
| Variable representing the seasonal pattern – sine season |
| Variable representing the seasonal pattern – cosine season |
| |
| **# Spatio-temporal lag of O3 measurements** |
| Spatially lagged terms as inverse distance weighted O3 measurements at other locations, as well as their 1-day |
| Spatially lagged terms as inverse distance weighted O3 measurements at other locations, as well as their 3-day |
| Spatially lagged terms as inverse distance weighted O3 measurements at other locations, as well as their 5-day |
| |
| **# Temporal lag of O3 predictors** |
| 1-day lagged moving values of air temperature |
| 1-day lagged moving values of total precipitation accumulation |
| 1-day lagged moving values of pressure |
| 1-day lagged moving values of humidity |
| 1-day lagged moving values of wind speed |

**Table S2 - Parameters Tuned for Base Learners**

| Neural Network | | Random Forest | | Gradient Boosting | |
|---|---|---|---|---|---|
| Parameter Name | Final Value after Tuning | Parameter Name | Final Value after Tuning | Parameter Name | Final Value after Tuning |
| Epochs | 50 | Number of trees | 1200 | Learning rate | 0.007 |
| Hidden layer and the number of neurons | 2 hidden layers with 275 neurons in each layer | number of bins for numerical columns | 20 | Number of trees | 200 |
| L1 regularization | $10^{-4}$ | number of bins for categorical columns | 449 | Column sample rate | 0.5 |
| Activation function | Rectifier | Maximum tree depth | 9 | Maximum tree depth | 7 |
| | | Sample rate | 0.42 | Sample rate | 1 |

Note: We used grid search to find optimal value for above parameters and used the final values for model training and model prediction. Take neural network as an example, to do grid search, we tried a series of parameter combinations in a parameter space (i.e., grid), fit neural networks, calculated cross-validated $R^2$, and chose the parameter combination that yielded the best model performance. If the chosen parameter combination was on the edge of parameter space, we slightly expanded the parameter space and repeated the above process.

## Tables S3-S5

**Table S3** – Cross-validation results by region

| Region | Ensemble model | | | | | | Neural Network | Random Forest | Gradient Boosting |
|---|---|---|---|---|---|---|---|---|---|
| | $R^2$ | RMSE (ppb) | Intercept | Slope | Spatial $R^2$ | Temporal $R^2$ | $R^2$ | $R^2$ | $R^2$ |
| East North Central | 0.928 | 4.030 | 0.946 | 0.989 | 0.846 | 0.934 | 0.927 | 0.924 | 0.927 |
| East South Central | 0.912 | 4.161 | 0.383 | 0.986 | 0.779 | 0.924 | 0.909 | 0.909 | 0.911 |
| Middle Atlantic | 0.908 | 4.601 | 3.865 | 0.943 | 0.847 | 0.931 | 0.911 | 0.913 | 0.915 |
| Mountain | 0.862 | 4.642 | 1.594 | 0.969 | 0.789 | 0.891 | 0.855 | 0.855 | 0.857 |
| New England | 0.867 | 4.811 | 1.961 | 0.979 | 0.773 | 0.908 | 0.859 | 0.863 | 0.867 |
| Pacific | 0.891 | 5.467 | 1.295 | 0.970 | 0.879 | 0.896 | 0.881 | 0.884 | 0.886 |
| South Atlantic | 0.912 | 4.283 | 0.667 | 0.991 | 0.881 | 0.915 | 0.910 | 0.906 | 0.908 |
| West North Central | 0.920 | 3.699 | 1.089 | 1.028 | 0.843 | 0.929 | 0.917 | 0.917 | 0.919 |
| West South Central | 0.920 | 4.136 | 0.332 | 1.001 | 0.809 | 0.933 | 0.920 | 0.917 | 0.920 |

Note: Region division was based on U.S. Census Bureau. New England: Connecticut, Maine, Massachusetts, New Hampshire, Rhode Island, Vermont; Middle Atlantic: New Jersey, New York, Pennsylvania; East North Central: Indiana, Illinois, Michigan, Ohio, Wisconsin; West North Central: Iowa, Nebraska, Kansas, North Dakota, Minnesota, South Dakota, Missouri; South Atlantic: Delaware, District of Columbia, Florida, Georgia, Maryland, North Carolina, South Carolina, Virginia, West Virginia; East South Central: Alabama, Kentucky, Mississippi, Tennessee; West South Central: Arkansas, Louisiana, Oklahoma, Texas; Mountain: Arizona, Colorado, Idaho, New Mexico, Montana, Utah, Nevada, Wyoming; Pacific: Alaska, California, Hawaii, Oregon, Washington. Although the Pacific Region includes Alaska and Hawaii, both states were not included in our modeling.

**Table S4** – Cross-validation results by season

| Season | Ensemble model | | | | | | Neural Network | Random Forest | Gradient Boosting |
|---|---|---|---|---|---|---|---|---|---|
| | $R^2$ | RMSE (ppb) | Intercept | Slope | Spatial $R^2$ | Temporal $R^2$ | $R^2$ | $R^2$ | $R^2$ |
| Summer | 0.885 | 5.320 | 0.168 | 1.004 | 0.891 | 0.903 | 0.884 | 0.877 | 0.880 |
| Fall | 0.863 | 4.375 | 0.814 | 0.976 | 0.848 | 0.894 | 0.863 | 0.853 | 0.857 |
| Winter | 0.853 | 4.313 | 0.539 | 0.991 | 0.688 | 0.885 | 0.853 | 0.844 | 0.848 |
| Spring | 0.879 | 4.682 | 0.526 | 0.989 | 0.819 | 0.904 | 0.878 | 0.871 | 0.874 |

Note: The seasons were defined as follows: summer (July – September), fall (October – December), winter (January – March), and spring (April – June).

**Table S5** – Cross-validation results by population density

| Season | Ensemble model | | | | | | Neural Network | Random Forest | Gradient Boosting |
|---|---|---|---|---|---|---|---|---|---|
| | $R^2$ | RMSE (ppb) | Intercept | Slope | Spatial $R^2$ | Temporal $R^2$ | $R^2$ | $R^2$ | $R^2$ |
| Quartile 1 | 0.888 | 4.794 | 0.284 | 0.993 | 0.849 | 0.900 | 0.883 | 0.882 | 0.885 |
| Quartile 2 | 0.911 | 4.388 | 0.018 | 1.002 | 0.875 | 0.924 | 0.908 | 0.903 | 0.907 |
| Quartile 3 | 0.902 | 4.538 | 0.645 | 0.987 | 0.863 | 0.915 | 0.900 | 0.895 | 0.898 |
| Quartile 4 | 0.911 | 4.643 | 0.249 | 1.005 | 0.864 | 0.925 | 0.900 | 0.899 | 0.903 |

**Table S6** – Variables sorted by % of missing values.

| Variables sorted by number of missings | % of missing values |
|---|---|
| MAIACUS_Optical_Depth_047_Terra_Nearest4 | 78.657 |
| MAIACUS_Optical_Depth_055_Terra_Nearest4 | 78.657 |
| MOD04L2_550 | 64.888 |
| MOD11A1_LST_Day_1km_Nearest4 | 63.548 |
| MOD11A1_Clear_day_cov_Nearest4 | 63.548 |
| MOD11A1_LST_Night_1km_Nearest4 | 59.446 |
| MOD11A1_Clear_night_cov_Nearest4 | 59.446 |
| MAIACUS_cosVZA_Terra_Nearest | 27.130 |
| REANALYSIS_gflux_DailyMean | 14.703 |
| REANALYSIS_soilm_DailyMean | 14.703 |
| MOD13A2_Nearest4 | 3.334 |
| CMAQ_NO2 | 3.108 |
| CMAQ_NO2_Vertical | 3.108 |
| CMAQ_Ozone | 3.108 |
| CMAQ_Ozone_Vertical | 3.108 |
| CMAQ_PM25_TOT | 3.108 |
| CMAQ_PM25_Vertical | 3.108 |
| CMAQ_PM25_NO3 | 3.108 |
| CMAQ_PM25_SO4 | 3.108 |
| CMAQ_PM25_EC | 3.108 |
| CMAQ_PM25_OC | 3.108 |
| MERRA2aer_SO4 | 3.061 |
| MERRA2aer_OCPHOBIC | 3.061 |
| MERRA2aer_OCPHILIC | 3.061 |
| MERRA2aer_BCPHOBIC | 3.061 |
| MERRA2aer_BCPHILIC | 3.061 |
| MOD09A1 | 2.696 |
| RoadDensity_prisecroads1000 | 1.345 |
| RoadDensity_prisecroads10000 | 1.345 |
| RoadDensity_roads1000 | 1.252 |

| | |
|---|---|
| USElevation_min100 | 0.232 |
| USElevation_mea100 | 0.186 |
| USElevation_bln100 | 0.186 |
| USElevation_med100 | 0.139 |
| NLCD_Barren100 | 0.139 |
| NLCD_Developed100 | 0.139 |
| NLCD_Herbaceous100 | 0.139 |
| NLCD_Planted100 | 0.139 |
| NLCD_Shrubland100 | 0.139 |
| NLCD_Water100 | 0.139 |
| NLCD_Wetlands100 | 0.139 |
| USElevation_dsc10000 | 0.093 |
| USElevation_max100 | 0.093 |
| USElevation_max10000 | 0.093 |
| USElevation_mea10000 | 0.093 |
| USElevation_med10000 | 0.093 |
| USElevation_min10000 | 0.093 |
| USElevation_std100 | 0.093 |
| USElevation_std10000 | 0.093 |
| USElevation_bln10000 | 0.093 |
| REANALYSIS_hpbl_DailyMax | 0.093 |
| REANALYSIS_shum_2m_DailyMax | 0.093 |
| REANALYSIS_prate_DailyMax | 0.093 |
| REANALYSIS_vis_DailyMax | 0.093 |
| REANALYSIS_apcp_DailyMean | 0.093 |
| REANALYSIS_dlwrf_DailyMean | 0.093 |
| REANALYSIS_dswrf_DailyMean | 0.093 |
| REANALYSIS_evap_DailyMean | 0.093 |
| REANALYSIS_hpbl_DailyMean | 0.093 |
| REANALYSIS_lhtfl_DailyMean | 0.093 |
| REANALYSIS_shtfl_DailyMean | 0.093 |
| REANALYSIS_shum_2m_DailyMean | 0.093 |
| REANALYSIS_snowc_DailyMean | 0.093 |
| REANALYSIS_tcdc_DailyMean | 0.093 |
| REANALYSIS_ulwrf_DailyMean | 0.093 |

| | |
|---|---|
| REANALYSIS_omega_DailyMean | 0.093 |
| REANALYSIS_weasd_DailyMean | 0.093 |
| REANALYSIS_prate_DailyMean | 0.093 |
| REANALYSIS_vis_DailyMean | 0.093 |
| REANALYSIS_hpbl_DailyMin | 0.093 |
| REANALYSIS_shum_2m_DailyMin | 0.093 |
| REANALYSIS_prate_DailyMin | 0.093 |
| REANALYSIS_vis_DailyMin | 0.093 |
| REANALYSIS_hpbl_1Day | 0.093 |
| REANALYSIS_shum_2m_1Day | 0.093 |
| REANALYSIS_prate_1Day | 0.093 |
| REANALYSIS_vis_1Day | 0.093 |
| REANALYSIS_air_sfc_DailyMin | 0.093 |
| REANALYSIS_air_sfc_DailyMean | 0.093 |
| REANALYSIS_air_sfc_DailyMax | 0.093 |
| REANALYSIS_air_sfc_1Day | 0.093 |
| REANALYSIS_windspeed_10m_DailyMax | 0.093 |
| REANALYSIS_windspeed_10m_DailyMean | 0.093 |
| REANALYSIS_windspeed_10m_DailyMin | 0.093 |
| REANALYSIS_windspeed_10m_1Day | 0.093 |
| NLCD_Barren10000 | 0.046 |
| NLCD_Developed10000 | 0.046 |
| NLCD_Herbaceous10000 | 0.046 |
| NLCD_Planted10000 | 0.046 |
| NLCD_Shrubland10000 | 0.046 |
| NLCD_Water10000 | 0.046 |
| NLCD_Wetlands10000 | 0.046 |

# Figures S4-S8

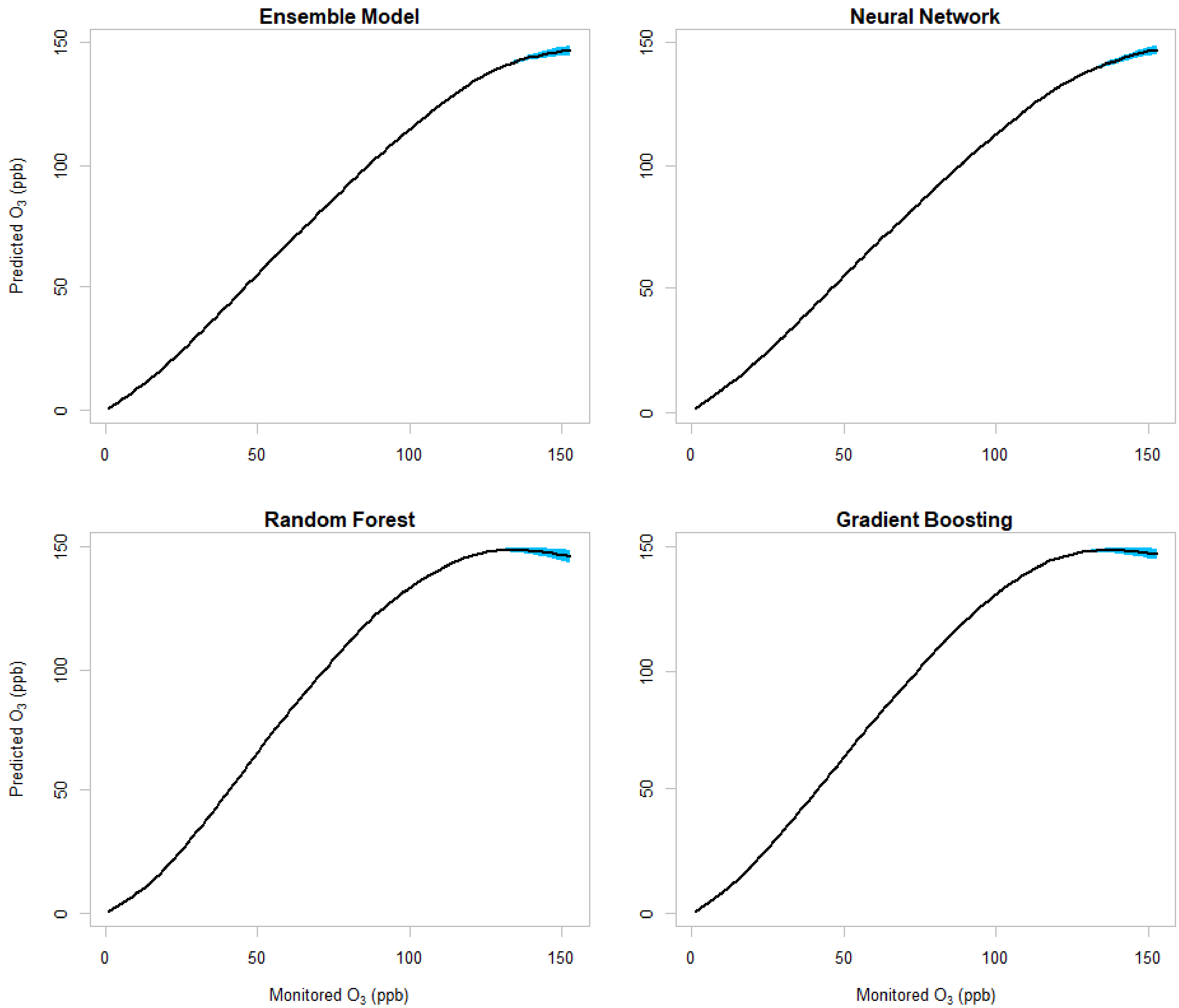
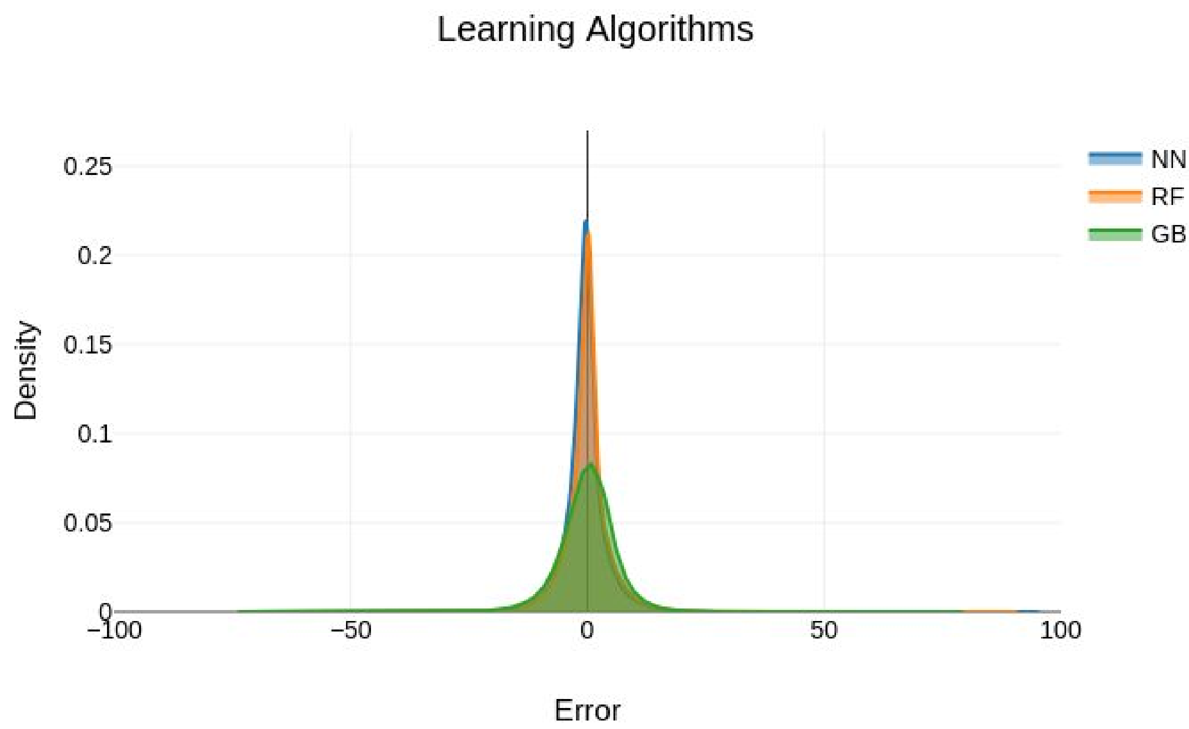



Figure S4 – $O_3$ levels predicted versus measured for the ensemble model and the three machine learning algorithms.

Note: We regressed daily predicted $O_3$ from each model (ensemble, neural network, random forest, and gradient boosting) against monitored $O_3$ using a GAM model with spline on the monitored $O_3$. Blue color represents 95% confidence interval.



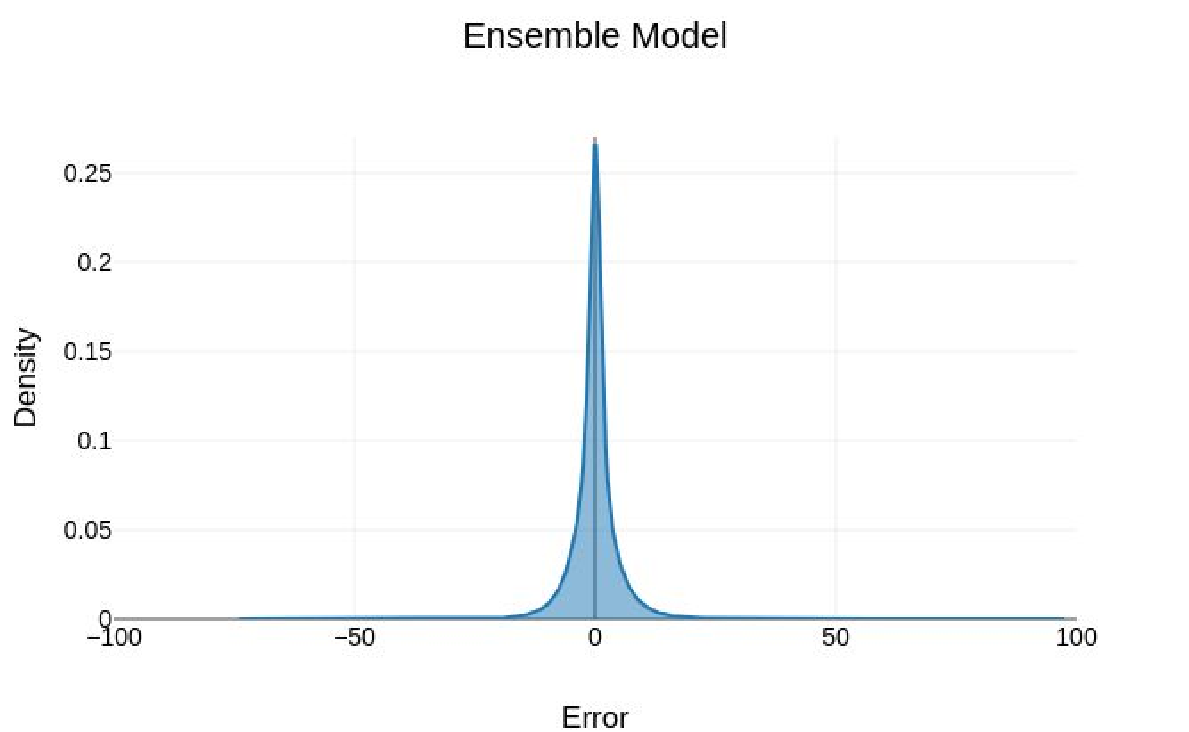
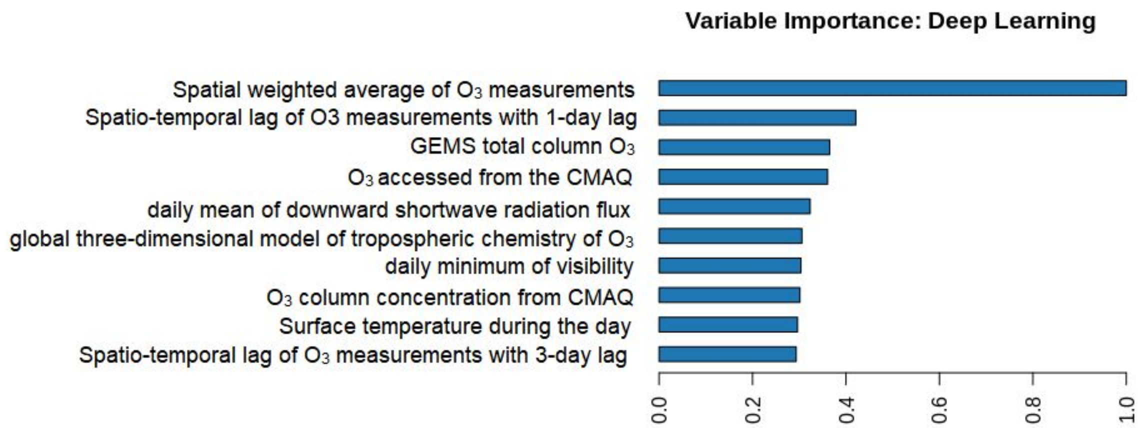
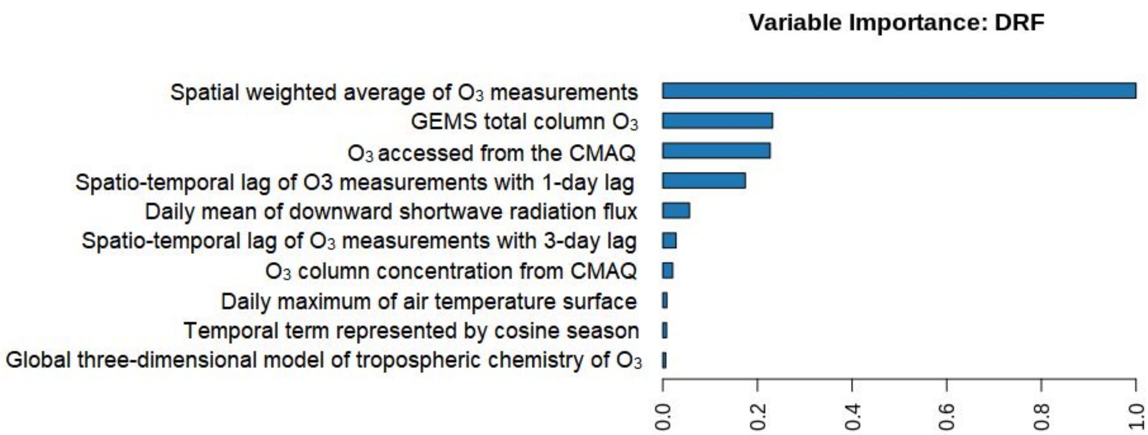




Figure S5 – $O_3$ mapping error estimates (ppb) from cross validation for ensemble model and three machine learning algorithms, where error = predicted – observed values at each site.



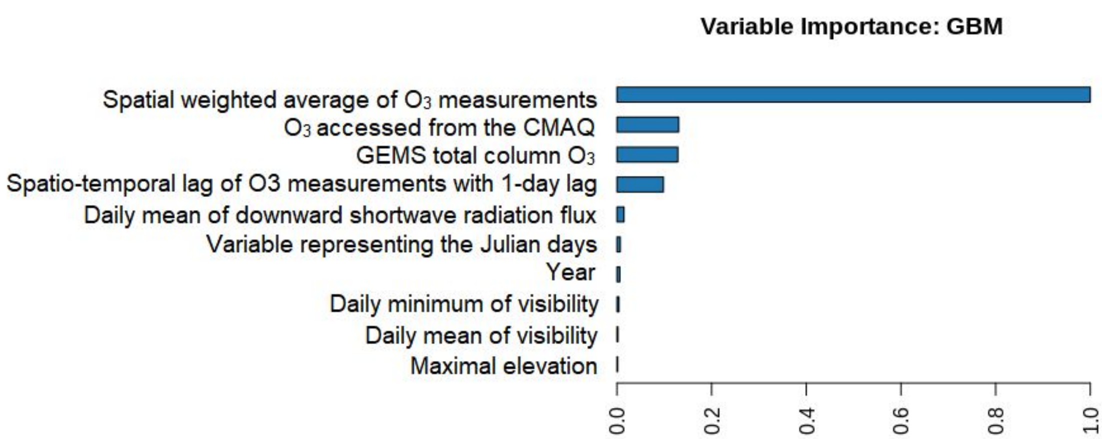




Figure S6 – Relative contribution of predictor variables for the three machine models.

Note: neural network (deep learning), random forest (DRF), and gradient boosting (GBM).

Note 2: We used the H2O package in R to run the three machine learning models. The command "h2o.varimp" extracts the list of variable importance. Some H2O algorithm class has its own methodology for computing variable importance. For random forest and gradient boosting, variable importance is determined by looking at whether a variable was selected to split on during the building process, and how much the squared error (over all trees) improved (decreased) as a result. For neural network, H2O uses the Gedeon method (Gedeon, 1997) - http://users.cecs.anu.edu.au/~Tom.Gedeon/pdfs/ContribDataMinv2.pdf.

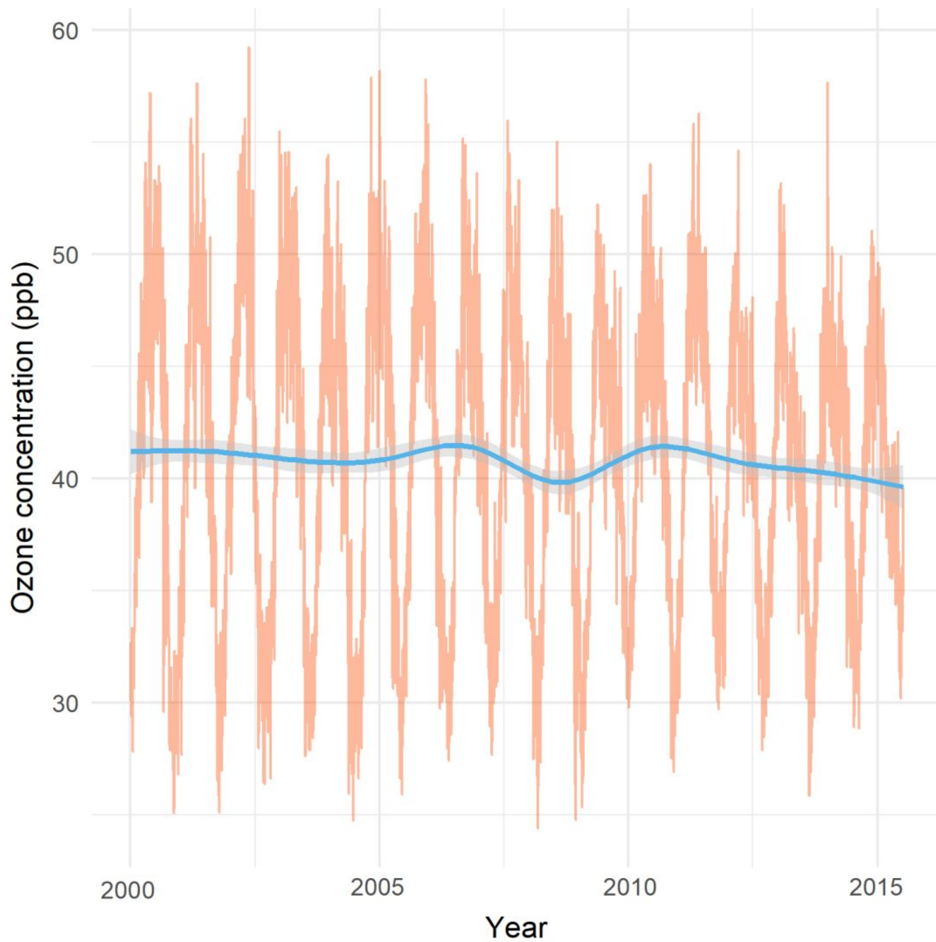



Figure S7 – Temporal trends of $O_3$.

Note: daily nationwide averages (orange line), smoothed conditional means function (blue line).

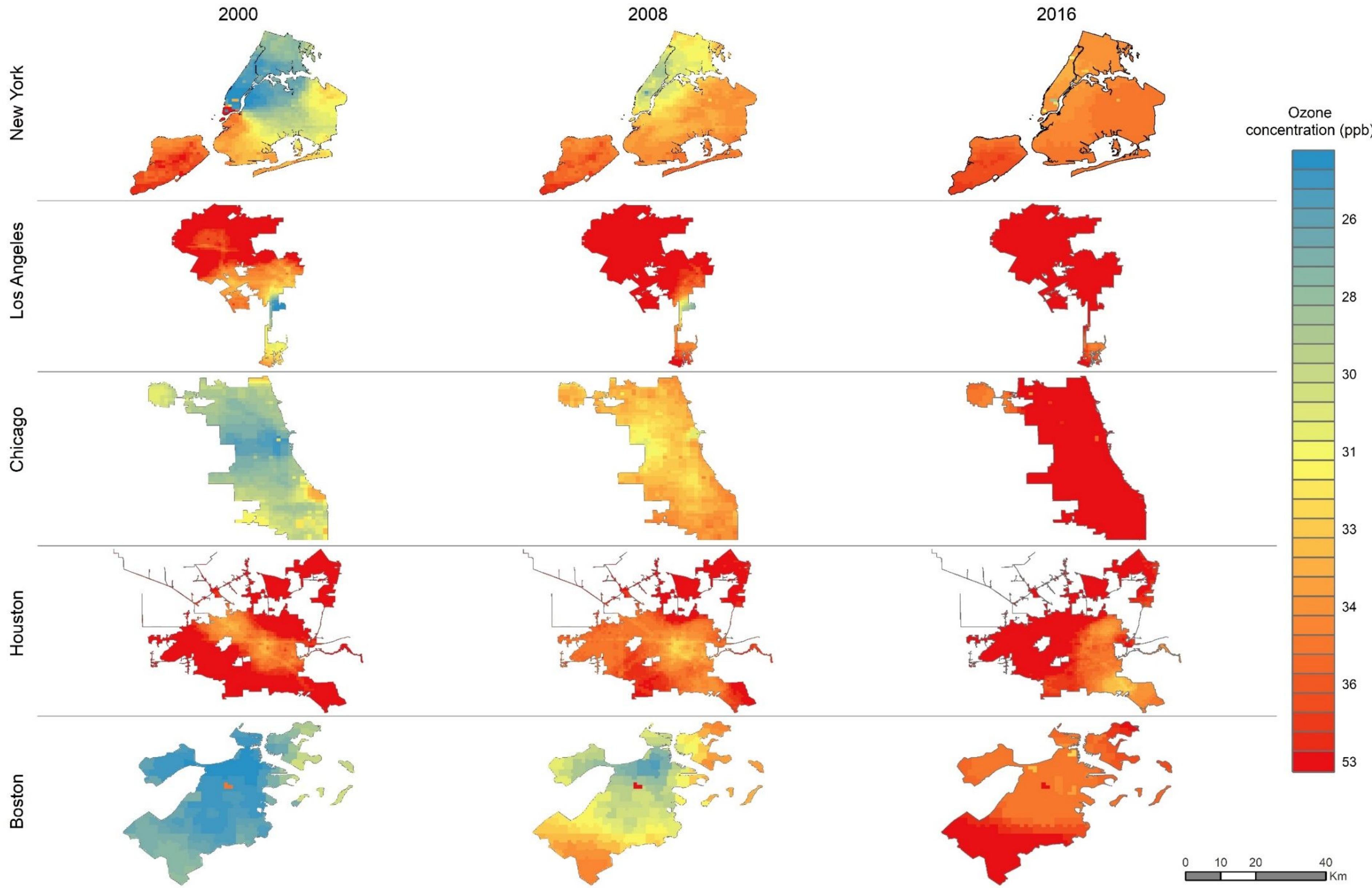



Figure S8 – Spatial distribution of the predicted levels of $O_3$ by the ensemble model for the major cities in the USA.

Note 1: We considered the top 4 cities in the US in terms of population – New York, Los Angeles, Chicago, Houston + Boston.

Note 2: in order to be possible the comparison of different levels of $O_3$ (represented by the legend with a color bar varying from blue [lowest concentration] to red [highest concentration]) over the cities and years, we standardized the symbolization (spatial distribution of the colors representing the ozone variation over space) based on the city and year with the lowest O3 concentration (New York, year 2000).