

ISTANBUL TECHNICAL UNIVERSITY ★ GRADUATE SCHOOL

**PARTICULATE MATTER 2.5 – PM2.5 MAPPING USING MODIS SATELLITE
DATA AND MULTIVARIATE NON-LINEAR REGRESSION ANALYSIS
OVER MARMARA REGION – TURKEY**



Ph.D. THESIS

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Department of Geomatics Engineering

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JUNE 2021

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JUNE 2021

İSTANBUL TEKNİK ÜNİVERSİTESİ ★ LİSANSÜSTÜ EĞİTİM ENSTİTÜSÜ

**MODIS UYDU VERİLERİ VE ÇOK DEĞİŞKENLİ DOĞRUSAL OLMAYAN
REGRESYON ANALİZİ İLE PARTİKÜLER MADDE 2.5-PM2.5
HARİTASININ MARMARA BÖLGESİ-TÜRKİYE İÇİN ÜRETİLMESİ**

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To my family and friends,



FOREWORD

A journey of a thousand miles begins with a single step, and so did this dissertation. It started with an idea. I would not have been able to achieve this without the endless and unconditional support I have received from wonderful colleagues, family members, and friends.

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ABBREVIATIONS

AE	: Angstrom Exponent
AERONET	: Aerosol Robotic Network
AHI	: Advanced Himawari Imager
AOD	: Aerosol Optical Depth
ARIMA	: Autoregressive Integrated Moving Average
NOA	: National Observatory of Athens
AVHRR	: Advanced Very High-Resolution Radiometer
BC	: Black Carbon
BLH	: Boundary Layer Height
C6.1	: Collection 6.1
CTM	: Chemical Transport Model
CUT	: Cyprus Technical University
CV	: Cross-Validation
D.B.	: Deep Blue
D.T.	: Dark Target
DBDT	: Deep Blue Dark Target
ECMWF	: European Centre for Medium-Range Forecast
EEA	: European Environment Agency
ENVI	: Environment for Visualizing Images
EOS	: Earth Observation Satellite
EPA	: Environmental Protection Agency
E.U.	: European Union
GAM	: Generalized Additive Model
GBM	: Gradient Boosting Machine
GeoTIFF	: Geospatial Tagged Image File Format
GMT	: Greenwich Mean Time
GOCI	: Geostationary Ocean Colour Imager
HDF	: Hierarchical Data Format
HYSPLT	: Hybrid Single-Particle Lagrangian Integrated Trajectory model
IDL	: Interactive Data Language
KNN	: K-Nearest Neighbour
L2	: Level 2
LME	: Linear Mixed Effect
L.T.	: Local Time
MAE	: Mean Absolute Error
MAIAC	: Multi-Angle Implementation of Atmospheric Correction
MERIS	: Medium Resolution Imaging Spectrometer
MERRA-2	: Modern-Era Retrospective Analysis for Research and Application
METU	: Middle East Technical University
MISR	: Multi-angle Imaging Spectroradiometer
MODIS	: Moderate Resolution Imaging Spectroradiometer
NAQMS	: National Air Quality Monitoring System
NASA	: National Aeronautics and Space Administration
NAPS	: National Air Pollution Surveillance

NCP	: North China Plains
NDVI	: Normalized Difference Vegetation Index
netCDF	: network Common Data Format
N.N.	: Neural Network
O.C.	: Organic Carbon
OECD	: Organization for Economic Co-operation and Development
PBL	: Planetary Boundary Layer
PM	: Particulate Matter
PMSD	: Particle Number Size Distribution
R.F.	: Random Forest
RH	: Realtive Humidity
RMSE	: Root Mean Squared Error
SLP	: Sea Level Pressure
STD	: Standard Deviation
T	: Temperature
UHKİA	: Ulusal Hava Kalite İzleme Ağı
U.K.	: United Kingdom
USA	: United Stated of America
USGS	: United States Geological Survey
WHO	: World Health Organiztion
WS	: Wind Speed

SYMBOLS

%	: Percentage
°C	: Centigrade
µg/m³	: Microgram per cubic meter
ÅE	: Angstrom Exponent
AOD₄₄₀	: Aerosol Optical Depth at 440 nm wavelength
AOD₅₅₀	: Aerosol Optical Depth at 550 nm wavelength
AOD_{ambient}	: Aerosol Optical Depth at an ambient humidity
AOD_{ho}	: Aerosol Optical Depth at the surface level
Exp(PM_{2.5}, R.H.)	: Exponential of the PM _{2.5} with respect to relative humidity
Km²	: Squared kilometer
µm	: Micrometer
NaCl	: Sodium Chloride
NH₃	: Ammonia
NO₃	: Nitrate
P_(i,j)	: Temporal predictor of the month i and season j
PM_{2.5}	: Particulate Matter with an aerodynamic diameter of less than 2.5 µm
PM_{2.5(m)}	: Measured Value of PM _{2.5}
PM_{2.5(p)}	: The predicted value of PM _{2.5}
PM_{2.5(ambient)}	: PM _{2.5} at an ambient relative humidity
Q1	: First quartile
Q3	: Third quartile
R²	: Coefficient of determination
RMSE^{rel}	: Relative Root Mean Square Error
SO₄	: Sulphate
β	: Fixed coefficient of the variable
σ_{sp(R.H.)}	: Particle diameter at an ambient relative humidity
σ_{sp(R.H.)dry}	: Particle diameter at a dry relative humidity
f(RH)	: Particle extinction coefficient



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PARTICULATE MATTER 2.5 – PM2.5 MAPPING USING MODIS SATELLITE DATA AND MULTIVARIATE NON-LINEAR REGRESSION ANALYSIS OVER MARMARA REGION – TURKEY

SUMMARY

This study estimated and mapped the dry-mass concentrations of PM_{2.5} (Particulate Matter 2.5) on the ground-level for the Marmara Region, Turkey, during 2013-2017, using multivariate nonlinear regression analysis. The study was conducted using AOD₅₅₀ (Aerosol Optical Depth at 550 nm) derived from the collection C6.1 of MODIS (Moderate Resolution Imaging Spectroradiometer) Aqua and Terra of the NASA (National Aeronautics and Space Administration), meteorological variables of the ERA-5 assimilation of the ECMWF (European Centre for Medium-Range Weather Forecasts), and PM_{2.5} ground measurements of the UHKİA/NAQMS (National Air Quality Monitoring System) ground-stations network. MODIS AOD data were validated against AERONET (Aerosol RObotic NETwork) ground-based sunphotometer AOD obtained from three sites around Turkey. In addition, MODIS AOD was compared to MERRA-2 (Modern-ERA Retrospective Analysis for Research and Application) Version 2 of AOD 550nm.

The study aims mainly to map the PM_{2.5} dry-mass concentrations using MODIS C6.1 AOD₅₅₀ gridded data over the Marmara Regions, Turkey, for the period between 2013 and 2017. Since a wide variety of health issues are attributed to PM_{2.5}, we aim to assess human exposure to high PM_{2.5} concentrations in the region due to analyzing the spatiotemporal variability of PM_{2.5}. Besides, an objective is to evaluate the efficiency of the MODIS sensor to retrieve AOD at 550 over the study area using both sunphotometer and gridded data. The fourth objective is to examine the associations between meteorological conditions and PM_{2.5}. Finally, Turkey is a candidate country for a European Union membership. Therefore, we aim to assess the vulnerability of humans in the Marmara region to excessive PM_{2.5} concentrations based on the E.U.s' air quality regulations.

Datasets of AOD obtained from three sites (METU-ERDEMLI, ATHENS-NOA, and Cyprus CUT-TEPAK) of AERONET ground-based sunphotometer were used to validate MODIS AOD. Collocated AOD pixels from the Terra and Aqua daily rasters were averaged and combined to improve the availability AODs. However, MODIS Terra, Aqua, and combined Terra/Aqua AOD datasets were validated independently against AERONET. Also, the three MODIS datasets were compared to the MERRA-2 AOD datasets of Morning, Noon, combined Morning/Noon, respectively. Furthermore, the statistics of both dataset validation versus AERONET were investigated under different R.H. conditions.

According to the statistics of seasonal and daily regression analysis, MODIS AOD is better than MERRA-2 by the mean of R^2 , MAE, and $RMSE^{rel}$. Combined AODs from MODIS Terra/Aqua and MERRA-2 Morning/Noon exhibited better validation results than the individual datasets against AERONET AOD. Apparent annual cycling was demonstrated by the three datasets of AOD over Turkey. Though, MODIS

underestimated while MERRA-2 overestimated AERONET. In addition, MODIS was found more efficient than MERRA-2 in detecting extreme episodes of AOD over Turkey. Particles' hygroscopic growth by humidity changes aerosols' microphysical characteristics. It was found that the R^2 of the regression analysis increases in low RH conditions. However, no significant changes were found in the MAE/RMSE^{rel}. Therefore, MODIS combined Terra/Aqua AOD dataset has been chosen to estimate PM_{2.5} concentrations over the Marmara Region.

The linear correlation between ambient MODIS AOD and PM_{2.5} has been found weak ($R^2=0.152$). The hygroscopic growth effects of R.H. (Relative Humidity) on the particles were corrected by the nonlinear extinction coefficient $f(RH)$. Also, BLH (Boundary Layer Height) has been utilized to estimate the AOD at the ground level (AOD_{h0}). The MODIS combined Terra/Aqua AOD outcomes were divided by the BLH values obtained from ERA-5 meteorological gridded datasets. Daily mean pixel values of relative humidity, boundary layer height, temperature, wind speed, precipitation, and sea level pressure, were calculated from averaging every days' provided ERA-5 rasters. However, daily pixel values of resultant wind speed have been calculated from the vertical and horizontal wind speed rasters. The meteorological variables were added to the MODIS AOD in a multivariate additive model to improve the PM_{2.5}–AOD relationship. Meteorological variables along with the $f(RH)$ and BLH corrections have been found increasing the additive models' R^2 into 0.492.

Collection window of 3x3 pixels (30x30 km²) of the MODIS AOD was used to calculate the daily values of AOD over every PM_{2.5} measuring site. Thus, to increase the daily AOD values' representativity, only the collected number of daily matchups resulted from averaging more than 5 pixels were considered in the model. Since R.H. changes the aerosols' microphysical properties, daily matchups with extreme humidity events (R.H.>90%) were excluded from the model. Besides, minimal values of PM_{2.5} (PM_{2.5}<5 µg/m³) are hard to be detected by the statistical models; thus, they were excluded to reduce bias and improve R^2 . Although 7% of the data were excluded, the models' R^2 increased to 0.602. At the last stage of the model, monthly and seasonal predictors were added to optimize the models' efficiency and statistics. The last stage of the model caused a 14% inclination for the models' R^2 .

Clear annual cycling of PM_{2.5} was detected by both the model and datasets of ground stations. The monthly average PM_{2.5} was found to decrease in summer and reach its maximum in the winter season. The concentrations reached their maximum in January (39 µg/m³) and reached their minimum in July (13 µg/m³). This could be explained by burning low-quality coal and fossil fuels for heating in the cold months. Also, the model tends to underestimate and overestimate PM_{2.5} in the months of winter and summer, respectively. Daily maps of PM_{2.5} were averaged for every month to reduce the missing pixels and produce monthly- seasonal- and annual-average maps of PM_{2.5}. Availability of pixels in every raster ranged between 33% and 66% for winter and summer, respectively. Compared to other cities of the region, the monthly average map of PM_{2.5} has shown high concentrations over Edirne, Bursa, and Kocaeli. This is due to the intensive burning of coal and fossil fuels for heating.

According to the E.U.s' (European Union) limit of PM_{2.5} (25 µg/m³), the seasonal and annual maps were classified to produce the E.U.s' exceedance maps. The seasonal exceedance maps showed that the PM_{2.5} exceeds the limit in most of the Marmara Region in winter. However, more than 90% of the region is lower than the E.U.s' limit

for the other three seasons. The annual exceedance maps clearly showed that the area of exceedance declined from 11.8% to 2.1% between 2013 and 2017, respectively.

Finally, the model outcomes of PM_{2.5} were validated against ground measured PM_{2.5}. Since PM_{2.5} seasonal and annual variability were found in the Marmara Region, leave-one-out CV Cross-Validation (CV) was conducted on a seasonal and annual basis. The number of available matchups of the leave-season-out for winter was found the least among the four seasons. However, a good agreement is found between the model-estimated and ground-measured PM_{2.5}. The R^2 ranged between 0.51 and 0.65 for spring and summer, respectively. The final R^2 and $RMSE^{rel}$ were 0.687 and 27%, respectively. However, the model underestimated the extreme events of PM_{2.5}, especially in winter.





MODIS UYDU VERİLERİ VE ÇOK DEĞİŞKENLİ DOĞRUSAL OLMAYAN REGRESYON ANALİZİ İLE PARTİKÜLER MADDE 2.5-PM2.5 HARİTASININ MARMARA BÖLGESİ-TÜRKİYE İÇİN ÜRETİLMESİ

ÖZET

Bu çalışmada, çok değişkenli doğrusal olmayan regresyon analizini kullanarak Marmara Bölgesi, Türkiye için 2013-2017 yılları arasında yer seviyesinde PM2.5 (Partikül Madde 2.5) kuru kütle konsantrasyon dağılımı tahmin edilmiş ve dağılım haritası üretilmiştir. Çalışma, NASA'ya ait TERRA ve AQUA uydularında bulunan MODIS C6.1 (The Moderate Resolution Imaging Spectroradiometer) algılayıcısı ile elde edilen AOD550 nm (Aeorosol Optik Derinlik-550 nm) verileri, ECMWF tarafından üretilen ERA-5 meteorolojik parametreleri, Ulusal Hava Kalitesi İzleme Sistemi'ne ait yer istasyonlarından elde edilen yersel PM2.5 verileri kullanılarak gerçekleştirilmiştir. Uydu görüntülerinden elde edilen MODIS AOD verilerinin doğruluk değerlendirmesi için Türkiye ve yakın çevresinde bulunan üç AERONET istasyonundan elde edilen AOD değerleri kullanılmıştır. Ayrıca, MERRA-2 AOD verilerinin doğruluk performansı ile MODIS AOD verilerinin doğruluk performansı bu çalışma kapsamında AEORONET AOD verileri kullanılarak karşılaştırılmıştır.

Çalışmada, MODIS C6.1 AOD550 verileri kullanarak 2013 ve 2017 yılları arasında Marmara Bölgesi, Türkiye için aylık PM2.5 kütle konsantrasyon haritalarının üretilmesi amaçlanmıştır. Ayrıca, Marmara Bölgesi'nde yaşayan nüfusun PM2.5 kirleticilerine maruz kalma durumunun mekânsal-zamansal PM2.5 analizleri ile değerlendirilmesi hedeflenmiştir. Avrupa Birliği (AB) üyeliğine aday bir ülke olan Türkiye'nin Marmara Bölgesi'ndeki PM2.5 konsantrasyonlarının AB'nin hava kalitesi yönetmeliklerine uygun olarak değerlendirilmesi önerilmiştir. Bunlara ek olarak, çalışma alanı için seçilen meteorolojik parametreler ile PM2.5 arasındaki istatistiksel ilişkinin araştırılması amaçlanmıştır.

Türkiye ve yakın çevresinde yer alan üç farklı AERONET istasyonundan (ODTÜ-ERDEMLİ, Atina-NOA ve Kıbrıs CUT-TEPAK) elde edilen AOD veri setleri, MODIS algılayıcısı ile elde edilen AOD değerlerinin doğruluk değerlendirmesi için kullanılmıştır. AERONET güneş fotometreleri, AOD değerlerini 550 nm' de elde etmemektedir. Doğruluk değerlendirmesinde kullanmak için AERONET istasyonlarında ölçülen AOD 440 nm ve AE₄₄₀₋₅₅₀'den (Angstrom Exponent) kullanılarak AERONET AOD 550 nm hesaplanmıştır. Ayrıca, günlük yarım saatte bir ölçülen AERONET AOD değerlerinin ortalaması günlük sabah, öğlen ve sabah/öğlen (gün ortası) saatleri için hesaplanmıştır. Terra ve Aqua uydularına ait MODIS C6.1 AOD görüntüleri coğrafi konum dosyaları ve ENVI IDL 5.1 de üretilen bir algoritma kullanılarak geometrik düzeltilmesi gerçekleştirilmiştir. MODIS Terra ve Aqua birleşik AOD görüntülerini üretmek için ortak pikseller ArcGIS 10.4 bulunan otomatik ve iteratif bir model kullanılarak birleştirilmiştir. Günlük Terra, Aqua ve Terra/Aqua AOD değerlerini hesaplamak için boyutu 30x30 km (dokuz MODIS piksel) olan pencere kullanılmıştır. Terra ve Aqua sonuçlarının birleştirilmesinin MODIS AOD'nin kullanılabilir veri kapasitesini yaklaşık %24,5 artırdığı görülmüştür.

Bunların yanı sıra, MERRA-2 AOD verileri (.netCDF) GeoTiff raster verilere dönüştürülmüştür. Sabah ve Öğlen MERRA-2 raster verilerinin, Sabah/Öğlen (birleştirilmiş Sabah/Öğlen) AOD veri setini oluşturmak için ortalaması alınmıştır. Ancak, MERRA-2 sürekli bir veri seti olduğu için, MODIS Terra ve Aqua'dan farklı olarak, MERRA-2'nin sabah ve öğlen veri setlerinin birleştirilmesi AOD'nin kullanılabilirliğini etkilememiştir. Terra ve Sabah, Aqua ve Öğlen ve Terra/Aqua ve Sabah/Öğlen alt veri setleri sırasıyla AERONET Sabah, Öğlen ve Sabah/Öğlen için doğrulanmıştır. AOD'nin Türkiye için zaman serileri oluşturulmuş ve üç veri seti için de analiz edilmiştir. Ayrıca, bağıl nem aerosollerin mikrofiziksel özelliklerini etkilediğinden, MODIS ve MERRA-2'nin AOD alt veri setleri mevsimlere göre farklı RH koşulları için değerlendirilmiştir.

Mevsimsel ve günlük regresyon analizi istatistiklerine göre, R^2 , MAE ve $RMSE^{rel}$ ortalamaları göz önüne alındığında, MODIS AOD algılayıcısının performansı MERRA-2'den daha iyi olduğu belirlenmiştir. Doğruluk değerlendirmesi sonuçlarına göre MODIS Terra/Aqua ve MERRA-2 Sabah/Öğlen'den birleştirilmiş AOD'ler, AERONET AOD'ye karşı diğer veri setlerinden daha yüksek doğruluk değerlerine sahip olduğu tespit edilmiştir. Buna rağmen, MERRA-2 AERONET değerlerini değerinden fazla tahmin ederken MODIS ise AERONET değerlerini değerinden daha az olarak tahmin etmiştir. Ayrıca MODIS'in, Türkiye için ekstrem olayların yaşandığı dönemleri tespit etmede MERRA-2'den daha etkin olduğu görülmüştür. Partiküllerin nemden kaynaklı higroskopik büyümesi, aerosollerin mikrofiziksel özelliklerini değiştirmektedir. Regresyon analizinin sonucunda elde edilen R^2 değerinin düşük RH koşullarında arttığı belirlenmiştir. Bununla birlikte, MAE/ $RMSE^{rel}$ değerlerinde istatistiksel anlamda önemli bir değişiklik tespit edilmemiştir. Bu nedenle, Marmara Bölgesi üzerindeki PM2.5 konsantrasyonlarını dağılımını belirlemek için birleştirilmiş MODIS Terra/Aqua AOD veri seti kullanılmıştır.

UHKİA/NAQMS hava kalitesi yer ölçüm istasyonları her 30 dakikada bir PM2.5 verisi sağlamaktadır. Günlük PM2.5 veri seti elde etmek amacıyla, her istasyon için PM2.5 ölçümlerinin günlük ortalaması hesaplanmıştır. Bir ArcGIS 10.4 otomatik modeli kullanılarak her yersel PM2.5 ölçen istasyon sahası için birleştirilmiş MODIS Terra/Aqua AOD'nin günlük ortalama değerleri $30 \times 30 \text{ km}^2$ (dokuz MODIS piksel penceresi) alan için hesaplanmıştır. Kullanılan dokuz piksel, UHKİA/NAQMS yersel hava kalitesi istasyonu konumu piksel penceresinin ortasına gelecek şekilde seçilmiştir. Yersel istasyon PM2.5 ölçümlerinin ve MODIS AOD'nin veri kullanılabilirliği sırasıyla %94,2 ve %49,5 olarak tespit edilmiştir. Her iki veri setinde de eksik veriler olduğundan, kullanılabilir veri kümelerinin eş zamanlı olanlarını eşleştirme için bir Python 3.8 betiğinden yararlanılmıştır. PM2.5 ile uydu algılayıcısından elde edilen AOD arasındaki istatistiksel ilişkiyi kontrol edebilmek için öncelikle basit bir doğrusal korelasyon hesaplanmıştır.

MODIS AOD ve PM2.5 arasındaki doğrusal korelasyon değerinin düşük olduğu belirlenmiştir ($R^2=0.152$). Bu, atmosferik nemin neden olduğu su alımının higroskopik büyüme etkileri ile açıklanabilir ve doğrusal olmayan sönüm katsayısı $f(RH)$ ile ölçülebilir. Bu nedenle, sönüm katsayısı, RH'nin [Relative Humidity (Bağıl Nem)] PM2.5 parçacıkları üzerindeki higroskopik etkilerini düzeltmek için kullanılmıştır. Ayrıca uydular, BLH [boundary layer height (sınır katmanı yüksekliği)] boyunca tüm AOD değerlerini (sütun biçimli AOD) birlikte kaydetmektedir, fakat yer istasyonları PM2.5'i yer seviyesinde ölçmektedir. Bu sebepten dolayı, yer seviyesinde (AOD_{h0}) AOD'yi tahmin etmek için BLH kullanılmıştır.

ECMWF ERA-5'in gridli verilerinden türetilen, (i) sınır katmanı yüksekliği (BLH), (ii) Bağıl Nem (RH), (iii) Sıcaklık [Temperature (T)], (iv) dikey rüzgar hızı [vertical wind speed (VWS)], (v) yatay rüzgar hızı [horizontal wind speed (HWS)], (vi) toplam yağış [total precipitation (TP)] ve (vii) deniz seviyesi basıncı [sea level pressure (SLP)] için günlük ortalama değerleri hesaplanmıştır. VWS ve HWS'den rüzgar hızını (RWS) hesaplamak için hız denklemi, kullanılmıştır. ECMWF ERA-5'in meteorolojik veri setleri, netCDF veri formatı olarak sağlanır. NetCDF gridli veri setleri, Python 3.8'deki netCDF4 kitaplığı kullanılarak GeoTIFF veri formatına dönüştürülmüştür. Otomatik bir ArcGIS 10.4 modelinin yardımıyla, her ERA-5 meteorolojik değişkeninin günlük ortalama değerlerini hesaplamak için aynı piksel pencereleri (30x30 km²) kullanılmıştır. Sütun biçimli AOD'lerin MODIS AOD verileri üzerindeki etkileri, BLH değişkeni kullanılarak giderilmiştir.

Birleştirilmiş MODIS Terra/Aqua AOD sonuçları, yer seviyesinde ($h_0 = 0$) MODIS AOD'yi hesaplamak için ERA-5 meteorolojisinden elde edilen BLH değerlerine bölünmüştür. Nem ve sütun düzeltmeleri, PM2.5 ile MODIS AOD arasındaki ilişkiyi iyileştirmiştir. PM2.5 ile MODIS AOD arasındaki doğrusal olmayan korelasyon için R^2 0,152'den 0,393'e yükselmiştir. Daha sonra, PM2.5-AOD ilişkisini iyileştirebilmek için çok değişkenli bir modelden yararlanılmıştır. Model, PM2.5'i bağımlı değişken olarak değerlendirirken, diğer yedisi (MODIS AOD ve altı meteorolojik değişken) bağımsız değişkenler olarak modele dahil edilmiştir. Meteorolojik değişkenlerin MODIS AOD ile birlikte çok değişkenli modele eklenmesi, R^2 'nin 0,492'ye çıkmasıyla sonuçlanmıştır.

Günlük MODIS AOD değerleri, 3x3 piksel penceresinin sonuçlarının ortalaması alınarak hesaplanmıştır. MODIS AOD'ların temsil edilebilirliğini artırmak için, ortalama beş pikselden daha az olan AOD'lar modelin dışında bırakılmıştır. MODIS AOD kullanılabilirliğinde %7'lik bir azalmaya rağmen, genel R^2 0,528'e yükselmiştir. Ayrıca, minimum PM2.5 değerlerinin modeller tarafından tahmin edilmesi zor olduğu için, beş $\mu\text{g}/\text{m}^3$ ($\text{PM}_{2.5} < 5 \mu\text{g}/\text{m}^3$) altındaki PM2.5 yer ölçüm değerleri modelden çıkarılmıştır. Bu işlem, R^2 'yi 0,554'e ulaşacak şekilde arttırmıştır. Ancak, kullanılabilir AOD veri miktarı %41.02'den %38.82'ye düşmüştür.

Daha önce değinildiği gibi, atmosferik nem, su alımı ile higroskopik büyümeye bağlı olarak aerosollerin mikrofiziksel özelliklerini değiştirmektedir. Aşırı bağıl nem olayları ($\text{RH} > \%90$) ile günlük eşleşmelerin %4.4'ünün modelden çıkarılması R^2 'yi 0.602'ye çıkarmıştır. Marmara Bölgesi üzerindeki PM2.5'te belirgin bir zamansal değişiklik tespit edilmiştir. Modellerin istatistiksel sonuçlarını iyileştirmek için aylık ve mevsimsel tahmin ediciler modellere eklenmiştir. Böylece %4'lük bir R^2 artışı ile 0,681 değerine ulaşılmıştır.

Günlük zaman serileri, 12 istasyonun PM2.5 ölçümlerinin ve Marmara Bölgesi için üretilen günlük PM2.5 değerlerinin ortalaması alınarak oluşturulmuştur. Bölgedeki MODIS görüntülerinde AOD piksellerinin ortalama kullanılabilirliği 59,4 olarak bulunmuştur. PM2.5'in net yıllık döngüsü, hem model sonuçları hem de yer istasyonlarının PM2.5 veri setleri ile gösterilmiştir. Günlük ortalama PM2.5 değerlerinin yaz aylarında (Haziran ve Temmuz) azaldığı, Ocak ayında ise maksimum seviyesine ($98,24 \mu\text{g}/\text{m}^3$) ulaştığı görülmüştür. Ayrıca, günlük zaman serileri, PM2.5 tahmin modelinin görece yüksek ortalama günlük PM2.5 değerlerini ($70 \mu\text{g}/\text{m}^3$ 'ün üzerindeki PM2.5 değerleri) olduğundan daha az tahmin ettiğini göstermiştir.

Aylık zaman serilerini analiz edebilmek amacıyla, her ay için PM2.5'in günlük haritaları ve yersel PM2.5 istasyon ölçümlerinin ortalaması alınmıştır. Günlük zaman

serilerinde olduğu gibi, kış aylarında (Aralık, Ocak ve Şubat) PM2.5'te belirgin bir artış yaşandığı ve 38.84 $\mu\text{g}/\text{m}^3$ seviyesine ulaşıldığı tespit edilmiştir. Buna karşılık olarak, PM2.5 konsantrasyonlarının bölgede yaz aylarında (Haziran, Temmuz ve Ağustos) minimum seviyeye indiği görülmektedir. Günlük zaman serisi verilerini aylık zaman serileri için birleştirmek, modellerin sonuçları ve yersel ölçümleri arasındaki istatistiksel değerleri iyileştirmiştir. Hesaplanan R^2 değeri 0,568'den 0,721'e yükselirken, RMSE değeri 9,32 $\mu\text{g}/\text{m}^3$ 'ten 4,48 $\mu\text{g}/\text{m}^3$ 'e düşmüştür.

Her iki zaman serisi için, modelin yersel ölçümleri olduğundan daha az hesapladığı görülmüştür. Hem günlük hem de aylık zaman serileri için değeri birden düşük RMB değerleri (sırasıyla 0.863 ve 0.927) elde edilmiştir. Kışın görülen yüksek PM2.5 konsantrasyonları, antropojenik faaliyetler ve meteorolojik koşullarla ilişkilidir. Fosil yakıtların ve düşük kaliteli kömürün (özellikle linyit kömürü) kışın kullanılması, insan kaynaklı PM2.5 emisyonlarını artırmaktadır. Ayrıca, kışın düşük sıcaklıklar, partiküllerin düşey karışmasını ve mekanik konveksiyonu zayıflatarak PM2.5 konsantrasyonlarını yükseltmektedir.

PM2.5 dağılım haritalarının üretilmesi için Python 3.8'in netCDF4 kitaplığı ve ArcGIS 10.4 otomatikleştirilmiş model kullanılmıştır. Meteorolojik değişkenler Python'daki netCDF4 betiğinden yararlanılarak ayrı ayrı hesaplanmış ve sonrasında GeoTIFF dosyalarına dönüştürülmüştür. Elde edilen meteorolojik değerler daha sonra MODIS Terra/Aqua'nın AOD görüntülerine entegre edilmiştir. Eksik pikselleri azaltmak ve aylık, mevsimsel ve yıllık ortalama PM2.5 dağılım haritalarını oluşturmak amacıyla, her bir ay için günlük PM2.5 haritalarının ortalaması alınmıştır. Kullanılan her görüntüde kullanılabilir piksellerin miktarı, kış ve yaz için sırasıyla %33 ile %66 arasında değişmektedir. Aylık ortalama PM2.5 dağılım haritaları, Edirne, Bursa ve Kocaeli şehirleri için yüksek PM2.5 konsantrasyonları göstermiştir. Aşırı ısınma, endüstriyel faaliyetler ve doğal gaz yerine fosil yakıtların kullanılması Marmara Bölgesi'ndeki bu üç şehirde belirgin bir PM2.5 artışına neden olmuştur.

AB'nin (Avrupa Birliği) PM2.5 (25 $\mu\text{g}/\text{m}^3$) limiti esas alınarak, mevsimsel ve yıllık PM2.5 limit aşım haritaları oluşturulmuştur. Mevsimsel limit aşım haritaları, kış mevsiminde PM2.5'in Marmara Bölgesi'nin büyük bir bölümünde sınırı aştığını göstermiştir. Bununla birlikte, bölgenin %90'ından fazlası diğer üç sezon için AB'nin sınır değerinin altında olduğu belirlenmiştir. Yıllık aşım haritaları, limit değerinin aşıldığı alanların Marmara Bölgesinde 2013 ve 2017 yılları arasında %11,8'den %2,1'e düştüğünü açıkça göstermiştir. Bu durumun, bölgedeki hava kalitesine ilişkin kısıtlayıcı politikalarla ilişkili olabileceği düşünülmektedir.

Son olarak, PM2.5'in model sonuçlarının, hava kalitesi istasyonlarının ölçtüğü yersel PM2.5 değerleri kullanılarak doğruluk değerlendirmesi yapılmıştır. Marmara Bölgesi'nde mevsimsel ve yıllık PM2.5 doğruluk değerlendirmesi için, "leave-one-out CV Cross-Validation (CV)" (birini dışarıda bırakma çapraz doğrulaması) yöntemi kullanılmıştır. Kış mevsiminin dışarda bırakıldığı doğruluk değerlendirmesi uygulamasında mevsimsel değerlendirmede en düşük sonuç elde edilmiştir. Bununla beraber, model kullanılarak tahmin edilen PM2.5 değerleri ile istasyonlarda ölçülen yersel PM2.5 değerleri arasında iyi bir uyum tespit edilmiştir. R^2 değerleri, ilkbahar ve yaz için sırasıyla 0,51 ile 0,65 arasında değişmiştir. En son R^2 ve RMSE^{rel} değerleri ise sırasıyla 0.687 ve %27'dir. Ancak model, özellikle kış mevsiminde yaşanan ekstrem olaylara ait PM2.5 değerlerini daha düşük olarak tahmin etmiştir.

1. INTRODUCTION

Air quality measures the degree of pollution in the air we breathe. Air pollution is considered a severe environment-originated risk to human health and the ecosystem. The pollutants of the atmosphere significantly impact human health and Earth's energy system balance. Breathing pollutants, especially atmospheric aerosols, both in the short- and long-term, cause serious respiratory diseases. According to the WHO, “breathing low-quality air causes one out of nine death cases in the world” (WHO, 2016). Besides, aerosols pollutants affect Earth's radiation budget by changing cloud properties and scattering solar light (Che et al, 2019).

The atmospheric aerosols are suspended liquid and solid particulates in the air. They originate from natural and anthropogenic sources and are formed from secondary atmospheric chemical reactions (Anderson et al, 2012; McMurry, 2002). Natural sources of atmospheric aerosols include arid and semiarid dust, sea spray, wildfires, and volcanic debris (Viana et al, 2014). Industry, agriculture, and combustion, on the other hand, present the anthropogenic sources (Bellouin et al, 2005). Anthropogenic emissions of aerosols are significantly impacting climate and human health (Zhao et al, 2019). They influence Earth's climate and radiation budget directly by solar light scattering in the cloud-free atmosphere and indirectly by changing the cloud properties (IPCC, 2013). Atmospheric aerosols, especially their fine fraction, are associated with plenty of respiratory and cardiovascular diseases (Hoek et al, 2013).

Particulate Matter 2.5 (PM_{2.5}) is the suspended-fine portion of atmospheric aerosols with an aerodynamic diameter equals or less than 2.5 μm (Spurny, 1996; Zhang et al, 2018). Chemical compounds like sulfates SO_4 , nitrates NO_3 , ammonia NH_3 , sodium chloride NaCl , organic carbon OC, black carbon BC, inorganic dust, and water are the major constituents of PM_{2.5} (An et al, 2015; Gong et al, 2015; Ram et al, 2012). PM_{2.5} is directly emitted to (Primary) or formed in (Secondary) the atmosphere (U.S.EPA, 2019). Primary particles, such as dust, organic carbon (OC), and black carbon (BC), are released from combustion processes. On the other hand, Chemical Reactions form secondary particles of PM_{2.5}, such as sulfate ions (SO_4) and nitrate (NO_3)

(Kleindienst et al, 2012; Wang et al, 2014). PM_{2.5} mass contains various primary and secondary organic and inorganic chemical constituents.

Ambient-outdoor PM_{2.5} is a significant inhalable air pollutant associated with various pulmonary and cardiovascular diseases. PM_{2.5}-induced health effects are categorized broadly into acute effects from short-term exposures (e.g., hourly or daily) and chronic conditions from long-term exposures (e.g., monthly or annual) (Hart et al, 2011; Kloog et al, 2013). Long-term exposures to PM_{2.5} cause chronic bronchitis, premature deaths, and decreasing lung function. Increasing respiratory symptoms, cardiac arrhythmias, and heart attacks, on the other hand, are associated with short-term exposures to PM_{2.5} (US EPA, 2004; WHO, 2013). Besides, mortality and morbidity are robustly related to PM_{2.5} mass concentrations (Dockery et al, 1993; HEI, 2000). In 2014, 428,000 premature deaths were attributed to the ambient PM_{2.5} in the European continent (EEA, 2017), while 2.9 million globally died in 2016 (WHO, 2016).

PM_{2.5} contributes to the transmission of contagious diseases, such as SARS-CoV-2. The small size (less than 2.5 μm) and long residence time of PM_{2.5} are suitable for penetrating lungs and reaching blood circulation. Coronavirus germs are carried over the PM_{2.5} particles deep into the lungs, increasing the chance of having infected (Nor et al., 2021). In addition, PM_{2.5} is responsible for developing severe symptoms in cases that already have SARS-CoV-2. Chronic exposure to high concentrations of PM_{2.5} can progressively and slowly inflame respiratory tracts (Ali et al, 2021). It produces excessive amounts of mucus and lessens ciliary movements. Therefore, PM_{2.5} increases patients' mortality from severe symptoms of SARS-CoV-2 (X. Wu et al, 2020).

Besides the adverse effects on human health, several environmental drawbacks are linked to PM_{2.5}. Ambient PM_{2.5} significantly influences the function of the aquatic ecosystem Zhao et al (2014), climate forcing Karumuri et al (2019), and atmospheric visibility Wang et al (2006). Since aquatic ecosystems are an ultimate sink for atmospheric aerosols Verma et al (2013), PM_{2.5} is accumulated in water bodies through wet and dry depositions and intensifies its toxicity (Hartono et al, 2017). Fine particulate matter is affecting the Earth's radiation budget and energy balance. PM_{2.5} constituents, like BC and mineral dust, absorb solar radiation (Aldabash et al, 2020) and disturb atmospheric long-wave radiation progress (Hu et al, 2017). Ambient

PM_{2.5} is also considered the leading air pollutant deteriorating atmospheric horizontal visibility (Sun et al, 2020). It forms hazes, impairs vision, and increases traffic accidents. Health and environmental consequences of exposure to PM_{2.5} still a complicated challenge for the world and governments.

Environment and public health organizations worldwide seek to regulate ambient PM_{2.5} limit values to safeguard public health and ecosystems. For instance, the World Health Organization (WHO) defined PM_{2.5} daily and annual mean limits in the air as 25 µg/m³ and 10 µg/m³, respectively (WHO, 2005). On the other hand, the United States Environmental Protection Agency (USEPA) has been more environmentally cautious and imposed strict measures to reduce environmental harm. It has set daily mean primary, daily mean secondary, and annual mean (primary and secondary) limits of PM_{2.5} as 12 µg/m³, 15 µg/m³, and 35 µg/m³, respectively (U.S.EPA, 2014). Since diesel motor vehicles are a significant source of fine particulate matter (Hatzopoulou et al, 2013), USEPA is also committed to reducing the PM_{2.5} emissions of vehicles from 31 µg/km to 22 µg/km in the United States (Kuklinska et al, 2015). European Union addressed only the annual PM_{2.5} maximum concentration as 25 µg/m³ (IIASA, 2014). Levels of safe fine particulate matter concentrations continue to be a severe threat to the public health and ecosystems and an environmental concern for international organizations and local authorities.

Air quality in the Marmara Region

Multiple human activities have contributed to deteriorating air quality in the Marmara region. The region is located in a very vital position with respect to Turkey. It is aligned between Turkey's European and Asian sides, connecting the European Union and the Black Sea countries. The Marmara Region represents a very critical commercial pathway between Turkey and Europe. Individuals and goods movements between Turkey and Europe are organized through five land cross points located in the region; Pazarkule and Ipsala are at Greece border, Kapıkule, Hamzabeyli, and Dereköy are at the Bulgarian border. The region involves a large-scale local and international transportation and highways networks.

Human activities such as urbanization movements and transportation are known to deteriorate air quality. The region is experiencing speedy urbanization over the last decade. At the expense of vegetated areas, urbanized areas have increased by 6.8%

between 2012 and 2018 in the region (Genel & Guan, 2021). Massive constructions and mega infrastructures are being built in the area, such as the İstanbul Airport (2018), Yavuz Sultan Selim Bridge (2016), Osman Gazi Bridge (2016), Marmaray Tunnel (2013), the Gebze–İzmir Highway (2019), and Northern Marmara Motorway (2018). In addition, 4.1 million of them are in Istanbul, seven million operated vehicles from different types are registered in the region, (Akgül et al, 2016).

Industrialization and urbanization have increased population density in the Marmara Region. As the most extensive contributor to the Turkish economy among other Turkey regions, the Marmara region contributes by 48% of Turkey's total industrial production (Demiraslan & Akıncı, 2018). Therefore, both local and regional migrants tend to settle in the region, increasing population density. Having a capacity for a large migrant influx has been identified as a primary issue for the Marmara Region, especially in Istanbul (OECD, 2008). These factors made the region the most populated area of Turkey, with 30% of the total Turkey's population (Kiliç et al, 2014).

Furthermore, anthropogenic activities in the Marmara Region have put the air quality and pollution levels on the stack. Some parts of the Marmara Region, mainly rural areas, still use low-quality lignite coal and fossil fuels for heating, especially in the winter (Karaca, 2012). Besides, anthropogenic emissions originated from industrial facilities, and traffic loads increase the concentrations of carbonaceous and particulate pollutants in the Marmara Region's air (Nur Kara, 2018). Besides, the area is extensively affected by the long-range transported mineral dust episodes from North Africa and Arabic Sahara (Koçak et al, 2007). Those mentioned above anthropogenic, human, and atmospheric factors have impacted the air quality and air pollution in the Marmara Region. Thus, the region was chosen as a study area to investigate PM_{2.5} comprehensively.

Meteorology and PM_{2.5}

Different meteorological parameters influence PM_{2.5} concentration levels in the atmosphere. PM_{2.5} mass concentration is affected by Total Precipitation TP, Wind Speed WS Toro et al. (2014), Height of the Planetary Boundary Layer BLH Wang et al. (2019), Temperature Kim et al. (2016), Sea Level Pressure SLP Aromolo et al. (2015), and Relative Humidity RH Lou et al. (2017) by facilitating or barring particle's dispersion.

Precipitation's falling speed is higher than the particles dry deposition velocity (Saha and Moorthy, 2003). Thus, precipitation washes settled particles on surfaces and deposits down suspended particles from the atmosphere. It has been proved that precipitation is negatively correlated with PM_{2.5} and lowers its concentrations (Chithra and Shiva Nagendra, 2014; Tiwari et al, 2013). Zhou et al. (2020) studied the effect of precipitation on PM_{2.5} over the Yangtze River in China. The authors found that high precipitation intensities are efficient in the PM_{2.5} wet deposition, particularly in high atmospheric loads of PM_{2.5}. The same relationship with the precipitation, however with PM_{2.5}/PM₁₀ ratio was investigated in Bogota, Columbia, during 2007-2011 by (Blanco-Becerra et al, 2015). The results showed that the precipitation reduces PM_{2.5} in the atmosphere; though, not as significant as it reduces PM₁₀. Precipitation substantially influences declining levels of aerosols pollution in the atmosphere.

WS plays a vital role in PM_{2.5} concentration levels. It influences pollutants' dispersion and dilution, where high wind speed distributes the particle load and lowers PM_{2.5} concentration (Cheng and Li, 2010). Previous studies widely investigated the effects of wind speed on the PM_{2.5} in the atmosphere. Pu et al. (2011) analyzed the relationship between the PM_{2.5} concentration and four categories of winds in China. Their results showed wind speed is positively correlated with the degree of dilution and dispersion in PM_{2.5}. However, the potential of PM_{2.5} local and regional emission sources can suppress wind speed effects and increases PM_{2.5} concentration. High enough WS can transport PM_{2.5} particles on regional and global spatial scales, thus increasing PM_{2.5} concentrations. Similar results were found in (Wang & Ogawa, 2015). They demonstrated at high WS; a negative WS-PM_{2.5} relationship turned into a positive. The authors associated this relation with the long-range transport of particles originated from geographical areas far from Nagasaki by the effect of high WS.

The third meteorological variable affecting PM_{2.5} is the Planetary Boundary Layer PBL. PBL is a strongly turbulent layer located at the lowest part of the atmosphere near the Earth's surface. Meteorologic conditions within the PBL affect clean and polluted air exchanging and particle transport and dispersion (Petäjä et al, 2016). The PBL interferes with aerosols' vertical structure as the bulk of the locally generated fine aerosols tends to be concentrated within the PBL (Su et al, 2018).

On the one hand, aerosols' cooling effects and negative radiative forcing can suppress the PBL development (Qu et al, 2017). Therefore, the PBL has an inversely linear relationship with the PM_{2.5} mass concentration (Luan et al, 2017). Sekiguchi et al. (2018) found a 9% decrease of the BLH over East Asia in a study to evaluate PM_{2.5} direct effects on the PBL in 2014. However, PBL can also be studied using passive remote sensing datasets. It was analyzed using micro-pulse LiDAR MLP continuous measurements (Pan et al, 2019). The study was conducted in Shanghai, China, during 2010-2015 and exhibited a 30-50 $\mu\text{g}/\text{m}^3$ increase of PM_{2.5} concentration causes a 100 m decrease of the BLH. Particulate matter accumulation within the PBL reduces BLH and negatively impacts the evolution of PBL.

On the contrary, stable stratification between different layers of the atmosphere makes PBL less turbulent (Vickers and Mahrt, 2004). It forms a barrier at the top of the PBL, preventing particles from transporting to the free troposphere (Han et al, 2009). Thus, it weakens the particle exchange and increases PM_{2.5} concentrations (Ding et al, 2017; Zilitinkevich et al, 2012). Even though it is uncommon, this positive relationship was found in previous studies in (Du et al, 2013; Q. Zhang et al, 2009). A total of 17 in-situ craft measurements between 2005 and 2006 over Beijing, China were used to analyze the effects of PBL on aerosols vertical distribution by (Q. Zhang et al, 2009). An apparent inversely relationship between the turbulence in the PBL (vertical mixing and horizontal transport) and the aerosol concentrations was presented. Besides, Du et al. (2013) also found an incline of particulate matter concentration associated with a decline of the BLH over Xi'an city, China, during 2007-2009. PBL is a substantial atmospheric constituent that influences the level of particulate pollution near the Earth's surface.

How temperature impacting PM_{2.5} dry mass concentrations was also investigated by previous studies. The relationship between PM_{2.5} and Temperature varies according to the meteorological and topographic characteristics. Some studies demonstrated that Temperature and PM_{2.5} are negatively correlated Chen et al. (2016); however, other studies showed a positive relationship between them (Li et al, 2015). On the one hand, high temperatures can increase PM_{2.5} through secondary-induced aerosols formation Jun Li et al. (2013) and soil dust resuspension (Vecchi et al, 2004). The temperature was found to increase PM_{2.5} concentrations due to soil and dust resuspension by Kozáková et al. (2017) and Kegler et al. (2001) in Central Europe and Arizona, USA,

respectively. Furthermore, Chen et al. (2017) found that temperature is the most positive influential meteorological parameter on different components of PM_{2.5} in the Jing-Jin-Ji region, China, during 2014-2015.

On the other hand, the temperature has an inverse relationship with PM_{2.5} due to the particles' convection and anthropogenic emissions. Temperature causes air convection and enhances the meteorological conditions for diffusion of particulate matter and reducing PM_{2.5} concentrations (Lu et al, 2017). This mechanism of PM_{2.5} dilution was proved by (C. Zhang et al, 2015). The authors analyzed the cross-correlation between PM_{2.5} and different meteorologic parameters for 2014 in Beijing and Hong Kong, China. The cross-correlation results showed that air convection becomes frequent when the temperature rises. Thus decreases PM_{2.5} mass concentrations over the study area.

Besides, coal consumption for heating in low-temperature seasons, especially in developing regions, contributes to increasing particulate matter concentrations (Xu et al, 2018). Demiraslan & Akıncı (2018) have demonstrated the effect of burning coal in winter on increasing PM₁₀ and SO₂ components in the Marmara Region. The study was conducted by analyzing measurements obtained from 35 ground stations. Increasing particulate matter PM pollution by coal consumption in Turkey, especially in the Marmara Region, was also stated in (Karaca, 2012). The study was conducted using GIS and statistical approaches to evaluate Turkey's air quality profile and classify its climatic zones. Q. Ma et al. (2017) also investigated contributions of coal burning to PM_{2.5} concentrations. They utilized GEOS Chemical Transport Model (GEOS-Chem) simulations. The study results showed a 35% contribution of burning coal to PM_{2.5} over China in 2013. To conclude, both meteorological impacts of temperature and anthropogenic activities can invert the relationship between temperature and PM_{2.5} into negative.

Due to impacting other meteorological variables, meteorology and spatial characteristics were also found to affect the relationship between atmospheric pressure and PM_{2.5}. On the one side, SLP may indirectly influence PM_{2.5} by affecting other factors, such as PBL and WS. High-pressure systems generally generate stagnant environments, thus accompany high concentrations of PM_{2.5} (Hsu and Cheng, 2016). Besides, High SLP is always accompanied by low PBL; therefore, it hinders the particles vertical movement and accumulates pollutants (Wu et al, 2017). The

spatiotemporal relationship between PM_{2.5} and different meteorological parameters was investigated by X. Li et al. (2017) in 173 cities in China during 2014-2015. The spatial results presented an increase in PM_{2.5} concentrations where SLP increases and a 20% contribution of SLP to the PM_{2.5} spatial distribution. Similar effects of SLP on the PM_{2.5} were found for China in 2017 by (Yang et al, 2020). When they quantified the unidirectional casualty effects of meteorology on PM_{2.5}, they discovered that PM_{2.5} increases due to particle's upward movement in low atmospheric pressures.

Conversely, some other studies found that the atmospheric pressure and PM_{2.5} can be negatively correlated. Low SLP causes the lower-level wind to converge and rise into a vertical direction, thus accelerating the dispersion of PM_{2.5} (You et al, 2017). Meanwhile, high SLP can indirectly reduce PM_{2.5} by affecting other factors, such as accelerating WS (Jian et al, 2012). Although the negative correlation between PM_{2.5} and SLP is possible in literature, it is found in a small number of studies compared to the positive association. To address this negative PM_{2.5}-SLP relationship, Jian et al. (2012) have utilized an Autoregressive Integrated Moving Average (ARIMA) statistical model to investigate the effect of meteorological factors on submicron particles in China in 2009. The negative correlation between PM_{2.5} and SLP was shown apparently in the results. Similar results were also demonstrated by (C. Zhang et al, 2015).

Relative Humidity RH is a critical meteorological parameter that affects PM_{2.5} microphysical properties and scattering of solar light due to various processes, such as hygroscopic growth by water uptake. Aerosol's hygroscopic properties affect the single scattering albedo, forward-scattering fraction, and asymmetry factor of the ambient aerosols (Gassó et al, 2016). Thus, it is essential to estimate water's contribution to aerosol's total columnar mass when retrieving PM_{2.5} concentrations from sensor data. Aerosol hygroscopicity is often described by aerosol's scattering enhancement factor (denoted by $f(RH)$). It is usually defined as a ratio between aerosols scattering coefficients at ambient RH ($\sigma_{sp}(RH)$) and dry reference RH <40% ($\sigma_{sp}(RH)_{dry}$), $f(RH) = \sigma_{sp}(RH) / \sigma_{sp}(RH)_{dry}$ (Zieger et al, 2011). This relationship was investigated in detail in previous studies (Yan et al, 2009; Zieger et al, 2011).

Significant gaps have existed in quantifying the relationships between RH and PM_{2.5}. This lack is due to their nonlinear relationships and uncertainties about the sensitivity of PM_{2.5} to RH (P. Wang et al, 2015). However, many previous studies concluded

that RH influences PM_{2.5} mass concentration significantly. This relationship was examined over Yangtze River, China, during 2014-2015 by (Lou et al, 2017). The results presented a positive PM_{2.5}-RH association in RH<70%; However, the PM_{2.5}-RH relationship turns into negative if RH>70%. Besides, J. Chen et al. (2014) proposed a retrieval method to estimate aerosol hygroscopicity parameter based on $f(RH)$ and Particle Number Size Distribution (PNSD) in-situ measurements in China, 2010. Results showed a sharp increase in $f(RH)$ related to RH increasing and that the high RH is associated with a high time variance of $f(RH)$.

Meteorological conditions have the most significant effects on dilution, diffusion, and accumulation of PM_{2.5} in the atmosphere (Pohjola et al, 2002; Tai et al, 2010). Meteorological variables datasets, such as ERA5, were used widely to retrieve meteorological variables for the purpose of PM_{2.5} estimation (J. P. Guo et al, 2009; Z. Li et al, 2017; Q. Zhang et al, 2009). The meteorological datasets of the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA5 contain various advantages by means of resolution, coverage, and operability. The temporal resolution of the ERA5 datasets is in near-real-time (1-hour); however, rasters are delivered as 3-hourly for the ensemble data assimilation. This temporal resolution is significantly higher than the ERA-Interim (the previous ECMWF assimilation dataset) and MERRA-2, 79-km and 65-km, respectively.

Meteorological datasets of ERA5 are continual over space and time. ECMWF provides historical meteorological gridded datasets of covering the entire Earth's surface starting from 1950. ERA5 ingests satellite sensors data, air crafts measurements, and ground measurements compared to other assimilated datasets that ensemble only ground measurements data. Besides, manipulation of ERA5 gridded datasets is feasible with respect to cost, processing time, and types of equipment. Products of ERA5 are downloaded automatically using Python scripts for free of charge with a .netCDF format. The downloaded rasters do not involve missing pixels and on the same size and extension. Therefore, processing meteorological rasters are conducted quickly using Python scripting and requires ordinary hardware. Furthermore, a wide variety of studies found the meteorological variables of the ERA5 datasets effective in retrieving meteorological conditions (J. P. Guo et al, 2009; Z. Li et al, 2017; Q. Zhang et al, 2009). For the advantages mentioned above of ERA5, it was chosen in the present study to retrieve meteorological conditions for the Marmara Region.

In-situ measurements vs. satellite

Retrieving atmospheric PM_{2.5} mass concentration data is conducted using either ground-level real-time measuring stations and/or satellite remotely sensed Aerosol Optical Depth AOD-dependent estimation models. However, each method contains advantages and disadvantages.

On the one hand, in-situ measurements of PM_{2.5} concentrations are conducted using globally distributed ground-level measuring stations. Ground stations provide high-quality PM_{2.5} measurements with the minimum error rate, irrespective of cloud cover and surface conditions (Gupta and Christopher, 2008). High temporal resolution characterizes ground-station measurements (e.g., 30 minutes and one hour) (Ramos et al, 2016). However, this traditional approach has disadvantages. The number of air quality monitoring sites is limited for the associated high costs, both fixed and operational costs, leading to sparseness in air quality data (Stevens et al, 2014). The low density of measuring stations makes the spatial coverage incomprehensive, provides unreliable results, especially in rural areas (Lee et al, 2015). Also, since monitoring station networks are point-source of data, they miss the spatial continuity (Shi et al, 2018).

PM_{2.5} in situ measuring ground stations provide high quality and high temporal resolution measurements; however, they have considerable shortcomings related to low spatial coverage and high cost. In the Marmara Region, the network of National Air Quality Monitoring System (TR: Ulusal Hava Kalite İzleme Ağı UHKİA) is composed of 12 PM_{2.5} ground-measuring stations (URL-5). The station's datasets are used by multiple researchers related to study PM_{2.5} in the region. They were utilized in Kahya et al. (2017) to investigate the spatiotemporal variation through the EU, USEPA, and WHO PM_{2.5} exposure regulations, the Marmara region, Turkey, during 2013-2014. Finally, Zeydan and Wang (2019) mapped PM_{2.5} mass concentrations in Turkey during 2011-2015. MODIS AOD data and in-situ PM_{2.5} measurements of 20 NAMQS ground stations in Ankara and the Marmara region were used. The PM_{2.5}-AOD relationship was established using linear multivariate regression analysis and HYSPLT back trajectories model.

On the other hand, recent advances in Earth Observation Satellites (EOS's) spatial coverage, spectral resolution, and historical datasets availability have motivated

remote sensing to be a widely used approach to estimate PM_{2.5} concentration (Donkelaar et al, 2012; Zheng et al, 2017). Polar satellites' sensors provide large AOD scenes at multiple spectral wavelengths covering a large extent of the Earth (Aldabash et al, 2020). Therefore, satellite sensors grant the advantages to estimate PM_{2.5} concentrations on comprehensive territorial and broad areas coverage (Hua et al, 2019). Since they are spatially continuous, another potential of the remote sensing-based PM_{2.5} estimation approach is filling the spatiotemporal gaps left by ground stations (Ma et al, 2016). Remote sensing sensors also provide historical gridded meteorological datasets with low costs (Levizzani and Cattani, 2019). However, drawbacks of satellite-derived aerosol and meteorological products include relatively low spatial resolution and low temporal resolution Mei et al. (2019), and the clouds in the atmosphere bring down their availability (Gueymard and George, 2005). Remote sensing-based PM_{2.5} retrieval models are spatially comprehensive, though, low spatial and temporal resolution-related drawbacks.

Satellite AOD datasets involve uncertainties due to aerosol columnar distribution and surface interaction (J. Li et al, 2015). Besides, AOD data collected by satellite sensors contain a degree of uncertainty. Therefore, validation of the sensor datasets of AOD is a required step before PM_{2.5} estimation. AOD datasets obtained from Aerosol Robotic NETwork AERONET ground-based sunphotometer observations are routinely utilized to validate satellite AOD datasets (Virtanen et al, 2018). Filonchyk and Hurynovich (2020) demonstrated the surpassing of MODIS C6.1 on C6 when the performance of DT, DB, and DTDB of MODIS collections 6 and 6.1 were assessed using 10 AERONET sites over China and Eastern Europe during 2001–2018. Similarly, Bhaskaran et al. (2011) compared AODs retrieved from MODIS bi-product with near-simultaneous AERONET AOD data over three sites in Australia in 2011. The results showed MODIS overestimating AERONET on low AODs; conversely, AERONET overestimated MODIS on high AOD values. Over Turkey, the efficiency of MODIS AOD was comprehensively evaluated by (Aldabash et al, 2020). The MODIS C6.1 Terra and Aqua AOD were validated and compared against AOD datasets of AERONET and MERRA-2. The authors found a high degree of compatibility of MODIS and AERONET AOD over Turkey. However, MODIS overestimated, and MERRA-2 underestimated AERONET AOD. Incommensurate results, Eck et al. (2019) found that MODIS C6.1 DT, DB, and MAIAC AODs

underestimate the AERONET AOD dataset and don't retrieve AOD efficiently on high burning days in Indonesia.

1.1 Literature Review

With the evolvments of satellite sensors and meteorological parameters measuring methods, knowledge of PM_{2.5} using remote sensing has increased immensely into the last two decades. Besides, the development of physical and chemical aerosols characterizing methods facilitated the global efforts to quantify concentrations and ambient aerosol sources (Hidy, 2019). Studies of retrieving near-Earth PM_{2.5} mass concentration using remote sensing are divided into at least three groups of algorithms (Hoff and Christopher, 2009): (1) empirical algorithms that regress PM_{2.5} directly with the gridded AOD. (2) algorithms that use, in addition to the PM_{2.5} and gridded AOD, multiple meteorological and land use parameters in a Multivariate regression, (3) Machine Learning and Mode-Based algorithm

AOD-PM_{2.5} empirical relationships

Aerosols affect the Earth's energy budget by absorbing and scattering solar light. Therefore, many previous studies established an empirical relationship between PM_{2.5} and different products of satellite-derived AOD. Glantz et al. (2009) have presented an empirical correlation between MODIS and MERIS satellite AOD datasets and ground-measured PM_{2.5} over Southern Sweden. The authors analyzed MODIS and MERIS AOD products according to Bremen aerosol satellite retrieval BEAR approach. The results have shown an AOD-PM_{2.5} linear relationship with R^2 of 0.59 and 0.58 for MODIS and MERIS, respectively, and a low slope affected by particle's hygroscopic growth. However, the AOD derived from the Multiangle Imaging Spectro Radiometer (MISR) sensor was found more reliable in correlating PM_{2.5} over the contiguous US by (Liu et al, 2004). The authors linked the MISR AODs with EPA's PM_{2.5} ground-measured data on an average seasonal basis. A strong correlation was found between AOD and PM_{2.5} with $R^2 = 0.66$ and Slope=1.00. The excellent AOD-PM_{2.5} agreement was also found for Jefferson County, the USA, in 2002 in a previous study by (Wang and Christopher, 2003). The correlation coefficient R for the MODIS-derived AOD and PM_{2.5} correlation was found 0.7 and 0.9 daily and monthly, respectively. Some case studies required pre-processing corrections to enhance the AOD-PM_{2.5} empirical relationship. RIVM LiDAR-based cloud screening method

was used to detect cloud-contaminated AOD values and quantify the vertical aerosol profile MODIS L1.5 AOD and PM_{2.5} ground-level data over Cabauw for the Netherlands in 2007 (Schaap et al, 2008). Without LiDAR cloud screening, results presented a good PM_{2.5}-AOD correlation with $R^2=0.56$ in September and spring months, while variation was very high in the overall regression. However, LiDAR cloud screening enhanced the overall linear regression; R^2 increased from 0.33 to 0.48. Besides, Zeng et al. (2018) applied a vertical-humidity correction method on the Advanced Himawari Imager AHI AOD dataset to estimate hourly PM_{2.5} mass concentrations over Hebei, China, 2017. Vertical humidity correction significantly improved the direct linear relation between AOD and PM_{2.5} (R increased from 0.19 to 0.47 and from 0.61 to 0.76), and model-retrieved PM_{2.5} was found consistent with the ground-measured PM_{2.5}.

Machine learning and Mode-Based approaches

Other authors supported machine learning models by meteorological, land use, AOD, and time predictors to estimate the PM_{2.5} mass concentrations. Random Forest (RF) model is the most frequently applied model within this approach (G. Chen et al, 2018; Stafoggia et al, 2020; Yazdi et al, 2020). The authors have implemented multi-stage RF models that digest AOD and other climatological, spatial, and temporal predictors to estimate PM_{2.5} concentrations. Although R^2 values of the CV were found acceptable in the three studies, the models still have very high statistical values of bias. The RF model was found able to detect most of the spatial variability of PM_{2.5}; however, it was weak in detecting temporal PM_{2.5} variation. The model was utilized to estimate PM_{2.5} in the summer season only by (Stafoggia et al, 2020). Besides, Z. Chen et al. (2018) and Yazdi et al. (2020) have applied the model to estimate PM_{2.5} for short periods (one year and two years) for London and China, respectively. Meteorological datasets of ERA5 were also involved in the machine learning studies. A Four-stages RF model was fed using ERA5-induced atmospheric predictors by Schneider et al. (2020) to estimate PM_{2.5} concentrations over Great Britain during 2008–2018.

Mode-based approaches depend on globally designed models to retrieve PM_{2.5}. GEOS-CHEM is widely used in this scope. Van Donkelaar et al. (2006) simulated the factors contributing to PM_{2.5}-MODIS AOD relationships using a global chemical transport model (GEOS-CHEM) over the USA during 2000-2001. They found that

Including the aerosol extinction's relative vertical profile in the simulation increased R-value from 0.36 to 0.71. However, visibility data integration into the GEOS-CHEM were found significant to estimating PM_{2.5} over the North China Plain (NCP) for 2014 ($R = 0.79$). Conversely, in an inverse process, Mölders et al. (2012) assessed the Weather Research and Forecast model and a chemistry package PM_{2.5} using mobile ground-measured PM_{2.5} and meteorological data. It was found that humidity and PBL are associated with discrepancies between modeled and measured PM_{2.5}.

Meteorological variables-based empirical relationships

The spatial and temporal variability of PM_{2.5} is affected by meteorological conditions. Meteorological parameters (SLP, WS, BLH, T, SLP, and RH) are added to the empirical relationship between MODIS AOD and ground measured PM_{2.5} in many previous studies. Tian and Chen (2010) established an empirical relation between ground-level PM_{2.5} and MODIS C005 AOD data over Ontario, Canada, for 2004. The authors aided the developed model by five meteorological parameters RH, SLP, T, BLH, and WS, obtained from the NASA Global Modelling Assimilation Office GMAO and assimilated based on Goddard Earth Observing System (GEOS). They modified ambient AOD into dry AOD based on an empirical equation to enhance the PM_{2.5}-AOD relation. The results showed a significant influence of the meteorological parameters and AOD correction on the empirical regression, $P < 0.001$ and $R^2 = 0.64$. Meteorology parameters were also found effective when utilized with AOD derived by the MISR sensor. Depending on an empirical regression model to estimate PM_{2.5} using MISR AOD, GEOS-assimilated RH, and BLH, and other geographic and temporal factors, Liu et al. (2005) estimated PM_{2.5} over the Eastern United States during 2001. Models' meteorological parameters were significant ($p < 0.0001$) in predicting PM_{2.5}, and the results showed a considerable influence of RH and BLH on correlating PM_{2.5} and MISR AOD. However, R^2 values were found to increase clearly when meteorology parameters correlated using the generalized additive model in the previous study by (Huang et al, 2020). The authors estimated PM_{2.5} concentrations by employing a general additive model containing MODIS C6 AOD and meteorology factors (BLH, RH, and WS) of the European Centre for Medium-Range Weather Forecasts (ECMWF ERA5) over Hubei, Central China, between 2016 and 2018. The results showed the model's satisfactory performance in estimating ground-level PM_{2.5} with low root mean squared error RMSE and high correlation coefficient R^2

(RMSE=13.1 $\mu\text{g}/\text{m}^3$; $R^2 = 0.89$). Besides, the trend analysis results detected a decreased PM2.5 trend over Hubei between 2016 and 2018, and the ERA5 meteorology datasets were found reliable in retrieving meteorological conditions. Using the same meteorological dataset (ERA5), Nabavi et al. (2019) found a strong correlation between PM2.5 in-situ measurements and ERA5-induced RH and BLH over Tehran, Iran, during 2011-2016. Also, Ding et al. (2019) have used the precipitation data of the ERA5 to investigate year-to-year meteorological differences that may impact the PM2.5 concentrations in Nanjing, eastern China, during 2011-2019. With the recent advances in telecommunication technology, phones were also used to monitor outdoor PM2.5. Meteorology variables of ERA5 were utilized along with phone-measured PM2.5 ground measurements in a study conducted by (H. Guo et al, 2020). The authors applied a two-level hierarchical linear model that depends on mobile phone-based measurements of PM2.5 and six ERA5-induced meteorological variables to estimate PM2.5 in Shenzhen, China, 2012.

Meteorological variables-based methods have many advantages over machine learning and linear approaches. Significant hygroscopic effects of the relative humidity and transported dust on the satellite AOD have weakened the linear empirical relationship between MODIS AOD and ground-measured PM2.5. Therefore, the PM2.5-AOD linear regression method was excluded from estimating PM2.5. Machine learning and mode-based methods benefit from the anthropogenic and land use data. Although anthropogenic emissions impact the atmospheric particulate matter, PM2.5 is significantly diluted, diffused, and accumulated by the atmosphere's meteorological conditions (Pohjola et al, 2002; Tai et al, 2010).

In addition, compared to machine learning models, meteorology-based methods do not require a high number of ground-measuring stations. In the Random Forest approach, the model robustness depends on training density based on ground station measurements (Stafoggia et al, 2019). However, Meteorology-based methods can be applied with any number of ground measuring stations. A limited network of PM2.5 ground stations is found in the Marmara Region, with only 12 stations. Besides, inter-period PM2.5 missing data does not affect the time coverage of models' outputs in the meteorology-based methods. Simultaneously, machine learning can estimate PM2.5 concentrations for the time-period covering of the ground measurements only.

Missing data is a significant concern when choosing an appropriate method for the PM_{2.5} estimation. All of the datasets of the variables ingested in the machine learning models, especially AOD, should be continual over space and time. Unfortunately, a significant amount of missing pixels due to clouds existed in the atmosphere are involved in the MODIS datasets of AOD, which hinders applying a machine learning approach. Therefore, all of the AOD datasets processed by machine learning models are assimilated datasets, not remotely sensed, especially the MAIAC AOD dataset. Contrarily, methodology-based datasets can be applied with the minimum percentage of available pixels. Furthermore, the implemented method corrects the atmospheric humidity's hygroscopic effects and treats the AOD and meteorology factors in the same ambient conditions. Besides, it clarifies how every meteorological condition impacts PM_{2.5}. It quantifies the relationship between every meteorological variable and PM_{2.5} separately. For the abovementioned reasons, a nonlinear meteorology-based approach was utilized to estimate PM_{2.5} in the present study.

1.2 Objectives

Particulate pollution, especially dry mass concentrations of PM_{2.5}, is a serious challenge for local governments, human health as well as environmental ecosystems. Our study aims to achieve the following objectives:

1. The Marmara region in Turkey is experiencing massive urbanization and industrialization movements for 30 years. Therefore, this study aims to map the PM_{2.5} mass concentrations over the region between 2013 and 2017, using MODIS C6.1 AOD550 gridded data.
 - Establish a nonlinear statistical relationship between ground-measured PM_{2.5}, MODIS AOD, and meteorological variables.
 - Apply the statistical model equations on the MODIS AOD scenes and ERA5 meteorology data in different months and seasons.
 - Averaging daily PM_{2.5} dry mass concentrations maps into monthly, seasonal, and annual maps for the five-year study period.
2. Concentrations of PM_{2.5} are varying per space and time. The study aims to Spatiotemporally analyze PM_{2.5} concentrations over the region between 2013-2017.
 - Comparative analysis of the PM_{2.5} concentrations maps in different months.

- Specifying seasons of high and low concentrations of PM2.5
 - Investigate the annual trend of PM2.5 for five years.
 - Analyzing monthly and daily time series of the PM2.5.
3. Humans' health is the most affected by the vulnerability to PM2.5. We also aim to assess human exposure to PM2.5 pollutants by specifying the cities of high concentrations.
 4. As a recognized candidate for membership in the European Union, the study's objective is to assess PM2.5 concentrations in Turkey based on the EUs' PM2.5 environmental regulations.
 - Classifying seasonal and annual maps of PM2.5 concentrations based on the EUs' annual threshold of PM2.5.
 - Assessing and comparing the exceedance coverage for the EUs' limit, both on a seasonal and annual basis.
 - Comparing exceedance coverage occurring in different areas of the region.
 5. The efficiency of space instruments is different in acquiring AOD data. Comprehensive validation of the MODIS instrument to measure AOD550 using both point source (AERONET) and gridded (MERRA-2) AOD data.
 - Validation of the used AOD dataset of AOD (MODIS C6.1 AOD550) using ground-based sunphotometer AOD datasets.
 - Comparing the AOD product of MODIS with another product, MERRA-2.
 - Study time-series and annual cycling of AOD over Turkey.
 - Assessing the detectability of the MODIS sensor to the extreme episodes of AOD.

Contrary to EU member states, the first PM2.5 measuring stations were launched in Turkey in 2013. Among Turkey, only one similar study by Zeydan and Wang (2019) was conducted during 2013-2015 to map PM2.5 over the Marmara Region. To the best of our knowledge, this comprehensive long-term study will be the first to map the PM2.5 over the Marmara Region. Besides, it investigates (examines) the relationship between every meteorological parameter and PM2.5. Furthermore, the present study is considered a nucleus for a real-time PM2.5 monitoring model that combines PM2.5 in-situ measurements, remote sensing data, and meteorological data. Finally, the

largest and the most comprehensive remote sensing AOD data validation process over Turkey is included in our study.



2. METHODOLOGY AND DATA

2.1 Study Area

Turkey is located in the north-east part of the Earth (33° N–42° N, 21° E–44° E). Turkey's area is 783,000 km², for which only 3% falls in Europe, while 97% lies in the Asian continent. Turkey's population increased by 1.4% in 2019 to reach 84.34 million inhabitants by 2020 (European Commission, 2020). Green areas cover 59% of Turkey's total area, of which 75% are agricultural areas, while only 25% are covered by forests (OECD, 2017). Because of the irregular topography, the climate in Turkey is varying considerably. The steppe climate is typical in the Anatolian plateau; the Mediterranean climate characterizes the coastal areas of the Mediterranean and Ege seas, while the climate of continents typifies the area of the Black Sea. Air quality in Turkey has been dramatically influenced by excessive fuel consumption, industrialization, urbanization, and construction (Kahya et al, 2017). Turkey is contributing to more than 50% of the anthropogenic PM₁₀ emissions in Europe EEA (2019), and more than 97.2% of the population in Turkey is breathing harmful air (URL-6).

The present study area is the Marmara Region (TR: Marmara Bölgesi), which is located in the north-west part of Turkey (39° N–42° N, 26° E–31° E). The Marmara region comprises 11 cities surrounding the Marmara sea and bridges the European and the Asian sides of Turkey (Table 2.1 and parts a and b of Figure 2.1, respectively). In Table 2.1, AMSL is above mean sea level (URL-8). Area values in km² were obtained from the Google Earth Engine where populations of the cities for 2020 are available online at (URL-2). In Figure 2.1, LULC data were obtained from MODIS 13A1 sensor on a 14-days and 500 temporal and spatial resolutions (URL-7). In addition, the figure shows ; a) Land cover map of Turkey, b) Map of the Marmara Region and the locations of the ground stations.

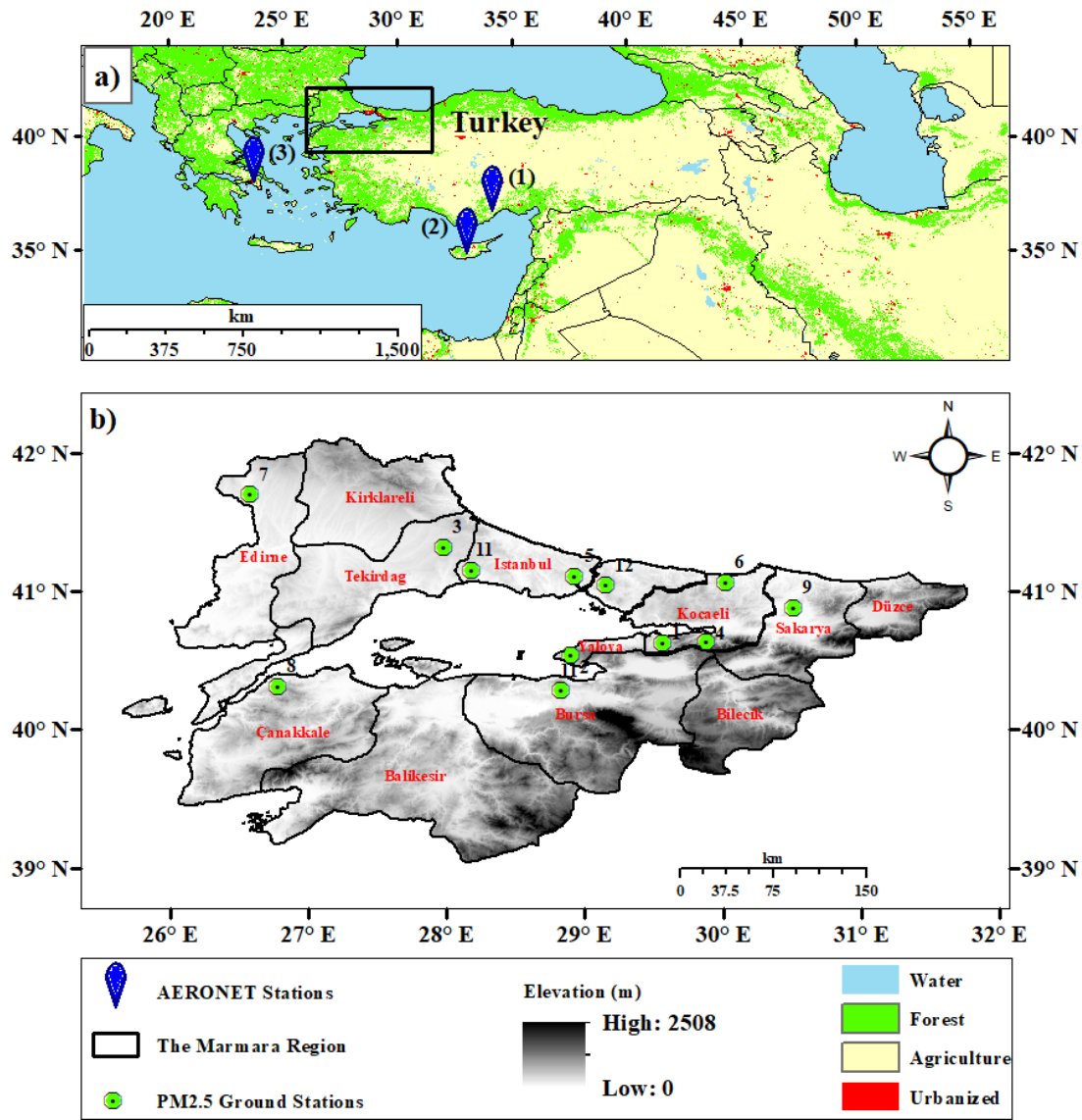


Figure 2.1: The Landuse Landcover (LULC) maps of the study area.

The Marmara Region area is 67,000 km², presenting around 9% of Turkey's total area. Approximately 33% of Turkey's population lives in the Marmara Region, with 27.5 million inhabitants (population density is 379 inhabitants/km²). It is characterized by a moderate and warm climate (Sertel and Ömerci, 2011). The peak and the lowest mean monthly temperatures (3 C°–25 C°) are in summer and winter, respectively. Contrarily, the highest mean monthly relative humidity and precipitation (63% and 1 mm) values are in the winter season (Figure 2.2).

Various extensive anthropogenic activities have contributed to the air pollution in the Marmara Region. Urbanization in the Marmara Region changes land cover from the crop into urban and deteriorates air pollution (Sertel et al., 2011). Besides, urbanization rates in the Marmara region, particularly in Istanbul, are higher than in all other Turkey

regions (Arat & Pamuk, 2019). Industry-induced air pollutants are a major constituent of the air pollution in the Region.

Table 2.1: Spatial information about the cities of the Marmara Region.

City	AMSL (m)	Area	Population	Continent
Balıkesir	310	12719.86	1,189,057	Asia
Bilecik	627	4384.15	209,925	Asia
Bursa	1791	11240.83	2,787,539	Asia
Çanakkale	192	9911.43	511,790	Eur./Asia
Edirne	453	6391.48	400,280	Europe
Istanbul	55	5558.83	14,377,018	Eur./Asia
Kırklareli	203	6667.33	343,723	Europe
Kocaeli	465	3690.95	1,722,795	Asia
Sakarya	1204	4965.38	932,706	Asia
Tekirdağ	110	6452.55	906,732	Europe
Yalova	735	862.17	226,514	Asia

More than 50% of Turkey's whole industrial activities have existed in the region (Yücekaya, 2018). According to the Netherlands Enterprise Agency, “diesel engines and trucks are a significant source of particulate matter pollution in Turkey as a whole” (Nur Kara, 2018). Since the region operates as the main transportation passage, both locally between cities and internationally between Turkey and Europe, the Marmara Region contributes highly to transportation-induced particulate pollution.

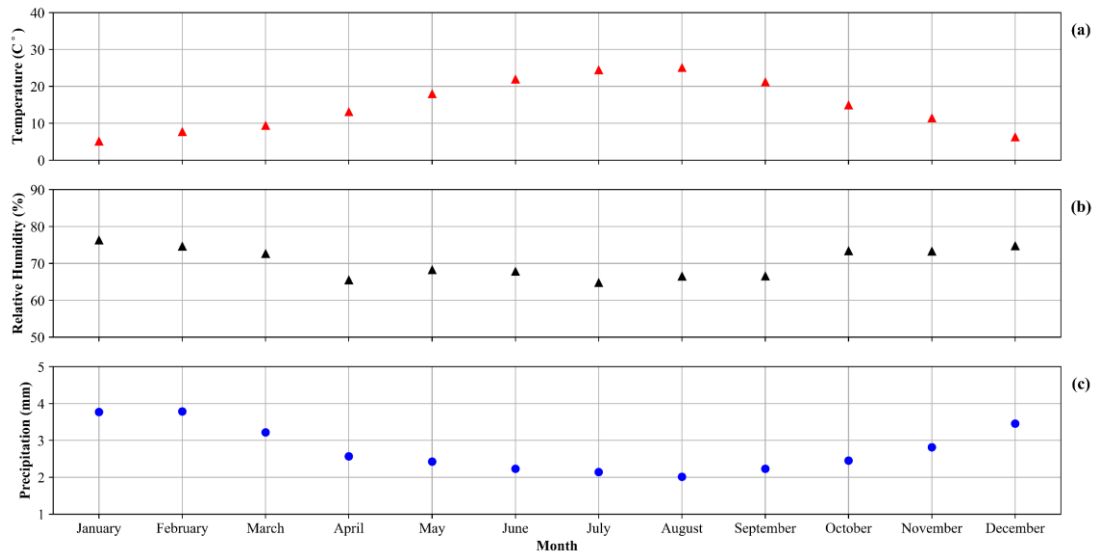


Figure 2.2: Map of Monthly mean (a) Temperature, (b) Relative humidity, and (c) Total precipitation of the European Center for Medium-Range Forecasting (ECMWF) ERA5 in the Marmara Region between 2013 and 2017.

2.2 Aerosol and Meteorology Products

Spatial and temporal datasets about the Earth and atmosphere improved our understanding of processes and dynamics occurring in the lower atmosphere and land. Concerning spatial coverage, datasets about atmospheric parameters, including AOD, are available as gridded or point-source time series. However, according to the data collection instrument, datasets are divided into remotely-sensed and direct-measured data. Gridded datasets including 1) AOD retrieved from the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor; 2) the Version-2 of the Modern-Era Retrospective analysis for Research and Applications (MERRA-2) AOD dataset; 3) The 5th generation of the European Center for Medium-Range Weather Forecasts (ECMWF ERA5) reanalysis of the global climate atmospheric parameters. On the other hand, the AErosol RObotic NETwork (AERONET) and the Turkish Ministry of Environment and Urbanization (TCCŞB) ground stations provide point-source ground-based data about AOD and PM_{2.5}, respectively.

2.2.1 Aerosol products

MODIS-derived AOD

MODIS is a key instrument aboard Terra and Aqua Earth Observation Satellites (EOS). Terra is a descending satellite that passes from the north to the south across the equator in the morning, it is known as EOS AM-1. On the contrary, Aqua is an ascending satellite that passes the equator from the south to the north in the afternoon and is known as EOS PM-1 (Z. Zhang and C. Moore, 2015). US National Aeronautics and Space Administration (NASA) launched Terra and Aqua MODIS as a sun-synchronous satellite in December 1999 and May 2002, respectively (Xiong et al, 2009). Usually, both instruments would view the whole surface of the Earth every 1 to 2 days. MODIS sensor measures signals in 36 spectral bands between 405 and 1438.5 nm, and it has a 2330 km viewing swath width (Srivastava et al, 2016). NASA provides MODIS aerosol products free of charge via Level-1 and Atmosphere Archive and Distribution System (LAADS) of the Distributed Active Archive Center (DAAC) Earth Data search platform. Its granules are downloaded in a Hierarchical Data Format HDF; each is containing 36 spectral bands.

Since the atmosphere and the surface properties contribute to the signal received by the MODIS sensor, several algorithms have been developed to retrieve aerosol

properties and produce MODIS aerosol products (M. Zhang et al, 2017). MODIS generates AOD products over land using Deep Blue (DB) and Dark Target (DT) algorithms (Wei et al, 2019). Pixel-based choosing between DB and DT depends on selection criteria based on the Normalized Difference Vegetation Index (NDVI) value as follows: 1) If $NDVI > 0.3$, DT AOD data is used; 2) If $NDVI < 0.2$, DB AOD data is used; and 3) If $0.2 \leq NDVI \leq 0.3$, mean value of both DT and DB AOD is used (Bilal et al, 2018). The latest collection of MODIS AOD datasets (C6.X) provides the DT and DB AOD products separately at 550nm wavelength (AOD550) on 3x3 km spatial resolution. It contains a combined DTDB AOD550 product; however, its spatial resolution is 10x10 km, achieved along the sensor's track at nadir (Gupta et al, 2020).

The second level of the latest MODIS collection (level 2 collection 6.1 (L2/C6.1)) at a 10x10 km² nominal spatial resolution are available in one NASAs' online database. In the present study, we have used the MODIS C6.1/L2 combined DTDB in MODIS Terra and Aqua AOD at 550 nm wavelength "Dark_Target_Deep_Blue_AOD_550_Combined" of the period 2013-2017. The spatial and temporal resolutions of the dataset included are 10x10 km² and 1-day, respectively. However, the scenes are provided as the fourth version of Hierarchical Data Format (.HDF4) on 2800x3140 km² longitudinal and latitudinal dimensions. In addition, a 1-km geolocation field supplements every scene of the MODIS AOD products for georeferencing. The geolocation field files including surface height above the geoid, longitude, latitude, azimuth angle, solar zenith angle, satellite zenith angle, and a land/sea mask. The dataset was entirely downloaded through the NASA Earth Data Search platform (URL-4).

MERRA-2 AOD reanalysis

MERRA-2 upgrades NASA's former satellite-era (1980 onward) reanalysis system to incorporate further observations and marked improvements to the Goddard Earth Observing System, version 5 (GEOS-5), Earth system model (Gelaro et al, 2017). Many advances have been made to the MERRA-2 GEOS-5, including advances in the hydrologic cycle representation, Ozone, stratosphere (Bosilovich et al, 2015; Molod et al, 2012), and analyzed fields of aerosol that have been radiatively coupled to the atmosphere (Randles et al, 2017). MERRA-2 is assimilating bias-corrected AOD outcomes from MODIS Collection 5 (C5), Advanced Very High-Resolution

Radiometer (AVHRR), MISR instruments, and Level 2 ground-sensed AERONET AOD (Heidinger et al, 2014; Holben et al, 1998; Kahn et al, 2005).

MERRA-2 collections are comprehensive through space, near-real-time, and compensate for sensors missing data due to clouds present in the atmosphere. Data collections in MERRA-2, including AOD, are provided entirely on the same spatial grid with 576x361 points on the latitudinal and longitudinal directions, respectively (Pinheiro et al, 2020). The temporal resolution of MERRA-2 collections is semi-instantaneous. GEOS-5 provides AOD collections every six hours at the synoptic times (00 GMT, 06 GMT, 12 GMT, and 18 GMT) (M. G. Bosilovich et al, 2016). Furthermore, MERRA-2 facilitated reading the files of the collections. Since the variables in MERRA-2 are stored as netCDF-4, which is similar to HDF-5, both the netCDF-4 and HDF-5 are read by the same tools.

MERRA-2 assimilates MODIS C5.1, MIRS, AVHRR satellite-derived AOD, and level 2 AERONET ground-sensed AOD in Earth system modeling. All pixels of MERRA-2 are computed on a 50x50 km grid; though, the AOD outputs of MERRA-2 are provided on a spatial resolution of $0.625^{\circ} \times 0.5^{\circ}$ ($\approx 62.5 \times 50$ km) (Manney et al, 2017). MERRA-2 AOD dataset has a semi-real time (1-hour) temporal resolution provided at the middle of every hour. In our study, the "Goddard Earth Sciences Data and Information Services Centre "GIOVANNI" platform GIOVANNI (2020), was utilized to download MERRA-2 AOD at 550 nm hourly, with a 62.5×50 km pixel resolution for five years, between 2013 and 2017. However, every day, the data's timescale is constrained within five hours (7:30 to 12:30 LT) to be compatible with the MODIS Terra and Aqua satellite overpassing times. The files of the datasets were downloaded on (.netCDF4) file format by using the WGET software.

AERONET ground-based AOD

The AERONET is a global federation of ground-based sunphotometers established by NASA and a group of local and international agencies and corporations (Holben et al, 1998). Since these measurements are point-source of columnar AOD data, AERONET was distributed over diverse and expansive areas at high temporal resolution (Giles et al, 2019). Sunphotometers provide ground-based remote sensing ambient observations about aerosol microphysical properties in a global context every 15 minutes (Xie et al, 2015). Under cloud-free conditions, AERONET reports observations of AODs,

inversion products, and precipitable water on multiple wavelengths (Giles et al, 2019; Tariq and Ul-Haq, 2020). The network imposes instruments calibration and data processing (Goloub et al, 2014). The second level (Level2) of the latest/third version (Version3) of the AERONET aerosol data are fully automated, cloud-screened, and pre-and post-field calibrated datasets (Aldabash et al, 2020; Aldabash and Balcik, 2019).

The certainties in the space-borne AOD products are primarily based on the validation between simultaneous sensor scenes and ground-measured AOD datasets, generally represented by AERONET AOD (Chu et al, 2002; Tong et al, 2020). AERONET ground-measuring stations are providing local measurements of AOD and aerosol microphysical properties on different wavelengths. In the present study, the quality-assured and cloud-screened (level 2 version 3 (L2/V3)) hourly measurements of the AERONET AOD and Ångström exponent (AE) were downloaded for the period during 2013-2017 from the AERONET official webpage (URL-1). The hourly dataset was downloaded for three AERONET stations: CUT-TEPAK in Cyprus, Athens-NOA in Athens, Greece, and METU-ERDEMLI in Mersin-Turkey (Table 2.2). In the Table 2.2, the AOD is the mean value of hourly AOD while the AMSL is the measured above mean sea level (Raptis et al, 2020; Tutsak & Koçak, 2019). Among the three selected AERONET stations, METU-ERDEMLI is located within Turkey's border; however, the rest two stations are located near to Turkey (Figure 2.1.a). The hourly measurements of (AE_{440/675}) and AOD at 440 nm were utilized to calculate AOD at the wavelength of MODIS and MERRA-2 of 550 nm.

Table 2.2: Spatial information and mean AOD during 2013-2017 for the Aerosol Robotic Network (AERONET) ground-measuring stations.

No.	Station	Country/City	Land-use	AOD	AMSL	Coordinates
(1)	METU Erdemli	Turkey/Mersin	Vegetation	0.213	71	36.34° N / 34.14° E
(2)	CUT-TEPAK	Cyprus/Limassol	Urban	0.168	11	34.68° N / 33.04° E
(3)	Athens-NOA	Greece/Athens	Discretely Urban	0.157	105	37.97° N / 23.72° E

2.2.2 Meteorology products

ECMWF ERA5

ERA5 is the latest version of the ECMWF's ERA series, 1 through 5, follows the ERA-Interim Analysis (ERA4). The ECMWF is operating and organizing the ERA5 datasets through the European Union (EU) flagship program for monitoring the Earth's

environment using satellite and in-situ observations (Copernicus) (Albergel et al, 2018). ERA5 ingested reprocessed datasets obtained from various satellite instruments, such as Terra- and Aqua-MODIS (Hersbach et al. (2020), Ocean and Sea Ice Satellite Application Facilities (OSI-SAF), SPOT VGT1, SPOT VG2 (Tall et al, 2019). A large number of hourly estimates of atmospheric, land, and oceanic variables are assimilated in the ERA5, including T, RH, SLP, WS, and BLH. The whole dataset of ERA5 covers the period between 1950-ongoing, which is divided into two clusters: one makes up climate data store entries for 1950 - 1978 (preliminary back extension) and the other covers from 1979 onwards (final release plus timely updates) (Hersbach et al, 2020).

The ECMWF's improved consistency of assimilation, which minimizes data reanalysis increments, is exhibited by the ERA5 (Hoffmann et al, 2018). Datasets of the ERA5 are provided on a high spatial and temporal resolution of $0.25^{\circ} \times 0.25^{\circ}$ ($\approx 30 \times 30$ km) and 1-hour, respectively. In the present study, we have obtained the hourly datasets of seven meteorological parameters (BLH, RH, SLP, T, TP, VWS, and HWS) for the Marmara Region during 2013-2017. Every day in the study period, the hourly gridded datasets were averaged for hours between (7:00 and 13:00 LT) to be compatible with MODIS Terra and Aqua overpasses.

NAQMS (UHKİA) PM_{2.5} measurements

National Air Quality Monitoring Stations (NAQMS) (TR: Ulusal Hava Kalitesi İzleme Ağı – UHKİA) is a local federation of distributed air quality measuring ground stations over Turkey. Turkish Ministry of Environment and Urbanization started to establish NAQMS in the last two decades to harmonize Turkey's air quality standards with the EU (Zeydan and Wang, 2019). NAQMS ground stations measure chemical and particulate air pollutants, including SO₂ (Sulfur dioxide), NO₂ (Nitrogen dioxide), Ozone (O₃), PM₁₀, and PM_{2.5}. In the Marmara region, 13 of these stations are measuring hourly PM_{2.5} mass concentrations. However, the stations measuring PM_{2.5} are located in heavily urbanized areas, lacking in covering rural areas. Moreover, data about PM_{2.5} are available only as short-term data, starting in March 2013.

The total number of UHKİA/NAQMS PM_{2.5} ground-measuring stations in the Marmara Region is 12; distributed among seven cities (Table 2.3). Table 2.3 provides

AMSL values (in Meter unit) that were obtained from (Zeydan & Wang, 2019). The unit of the minimum value (Min), the maximum value (Max), and the standard deviation (σ) of the dataset is $\mu\text{g}/\text{m}^3$, while Data is the the percentage of available days for every statiuon during 2013-2017. They are providing PM2.5 ground-measured data in the region only since 2013. NAQMS is providing surface-level PM2.5 measurements on a high temporal resolution of 1-hour. In the present study, we have downloaded hourly PM2.5 datasets of the 12 stations in the Marmara Region for the period between 2013 and 2017 from the Air Monitoring Network of the Turkish Ministry of Environment and Urbanization (UHKİA, 2020). The mean value of seven hourly measurements in each day (7:00 – 13:00 LT) was calculated to fulfill the MODIS and ERA5 dataset's time consistency.

Table 2.3: Spatial and statistical data for the PM2.5 ground measuring NAQMS stations in the Marmara Region.

No.	Station	City	Coordinates	AMSL	Min.	Max.	Mean	σ	Data
1	Altınova	Yalova	40.70° N / 29.51° E	16	0.49	76.52	21.99	10.56	92%
2	Armutlu	Yalova	40.53° N / 28.78° E	21	0.44	87.67	18.03	9.00	94%
3	Çerkezköy	Tekirdağ	41.32° N / 27.98° E	146	0.18	113.10	23.89	15.35	93%
4	Gölcük	Kocaeli	40.72° N / 29.79° E	29	0.73	86.59	21.33	13.78	95%
5	Kağıthane	İstanbul	41.09° N / 28.97° E	43	0.24	156.89	29.23	19.37	94%
6	Kandıra	Kocaeli	41.13° N / 30.00° E	48	0.25	79.69	15.73	9.75	92%
7	Karaağaç	Edirne	40.85° N / 26.64° E	36	0.21	140.35	20.75	13.91	95%
8	Lapseki	Çanakkale	40.40° N / 26.77° E	12	0.28	90.56	16.42	8.57	93%
9	Ozanlar	Sakarya	40.79° N / 30.40° E	26	1.73	196.23	34.56	25.06	97%
10	Silivri	İstanbul	41.07° N / 28.26° E	8	1.91	78.71	19.81	11.28	96%
11	Uludağ Üni.	Bursa	40.22° N / 28.87° E	289	1.59	102.97	26.72	14.11	97%
12	Ümraniye	İstanbul	41.02° N / 29.10° E	149	1.27	136.32	26.67	14.18	96%

2.3 Methodology

In the present study, MODIS AOD was validated against AERONET AOD and compared to MERRA-2 AOD with respect to the period 2013-2017. MODIS AOD, accompanied by ERA5 meteorological variables, was utilized to map monthly PM2.5 mass concentrations over the Marmara Region during 2013-2017. The approach used in the validation of MODIS AOD and mapping PM2.5 is explained in detail in the following three sections and in the flow chart of (Figure 2.3).

2.3.1 MODIS AOD validation

Space-based satellite sensors are extensively used to obtain global scale data about aerosol. However, several assumptions and different conditions are involved in retrieving satellite-based aerosol data (Virtanen et al, 2018). Thus, ground-based

aerosol data should be used to validate the retrieved outcomes of satellite aerosol datasets (Witthuhn et al, 2020). In this stage, MODIS Terra, Aqua, and combined Terra/Aqua AOD datasets were validated against AERONET stations AOD as well as compared with morning, noon, and combined morning/noon (named with a semi-day) MERRA-2 AOD products.

In this study, MODIS and MERRA-2 AOD at 550 nm were validated against and seasonally compared to AERONET AOD with respect to the period 2013 -2017. The following nine steps explain the approach used in the comparison between MODIS/MERRA-2 and AERONET: (i) AOD DB/DT retrievals were obtained from the MODIS Terra and Aqua scientific datasets of “Dark_Target_Deep_Blue_AOD_550_Combined” by extraction and georeferencing against corresponding geolocation files using ENVI IDL iterative script. (ii) Mosaicking was conducted to merge the adjacent layers in each dataset over the study area to produce daily layers using ArcGIS automated model built in the model builder tool. (iii) MODIS Aqua and Terra AOD collocated pixels values were averaged and combined. (iv) MERRA-2 AOD hourly values were averaged over morning and noon hours (7:00 – 11:00 and 11:00 – 14:00, respectively) as well as over these two periods (named with semi-day period) and extracted over the three AERONET sites. (v) Both MODIS and MERRA-2 daily values were extracted using 30x30 km² (3x3 MODIS pixels) window and operated based in an automated ArcGIS extraction model. The spatial window size was chosen after considering the average surface wind speed (0.447 m s⁻¹) in the study area (URL-9). (vi) Since AERONET stations are not providing AOD at 550 nm wavelength, the AE_{440/675} and AOD at 440 nm were used to calculate AOD at the MODIS wavelength 550 nm using equation 2.1.

$$AOD_{550} = AOD_{440} \left(\frac{440}{550} \right)^{AE_{440/675}} \quad (2.1)$$

AOD₅₅₀ and AOD₄₄₀ are the Aerosol Optical Depth at wavelengths 550 and 440, respectively; AE_{440/675} is the Angstrom Exponent for the wavelengths 440 and 675 nm. The AERONET sunphotometer provides each parameter on hourly basis.

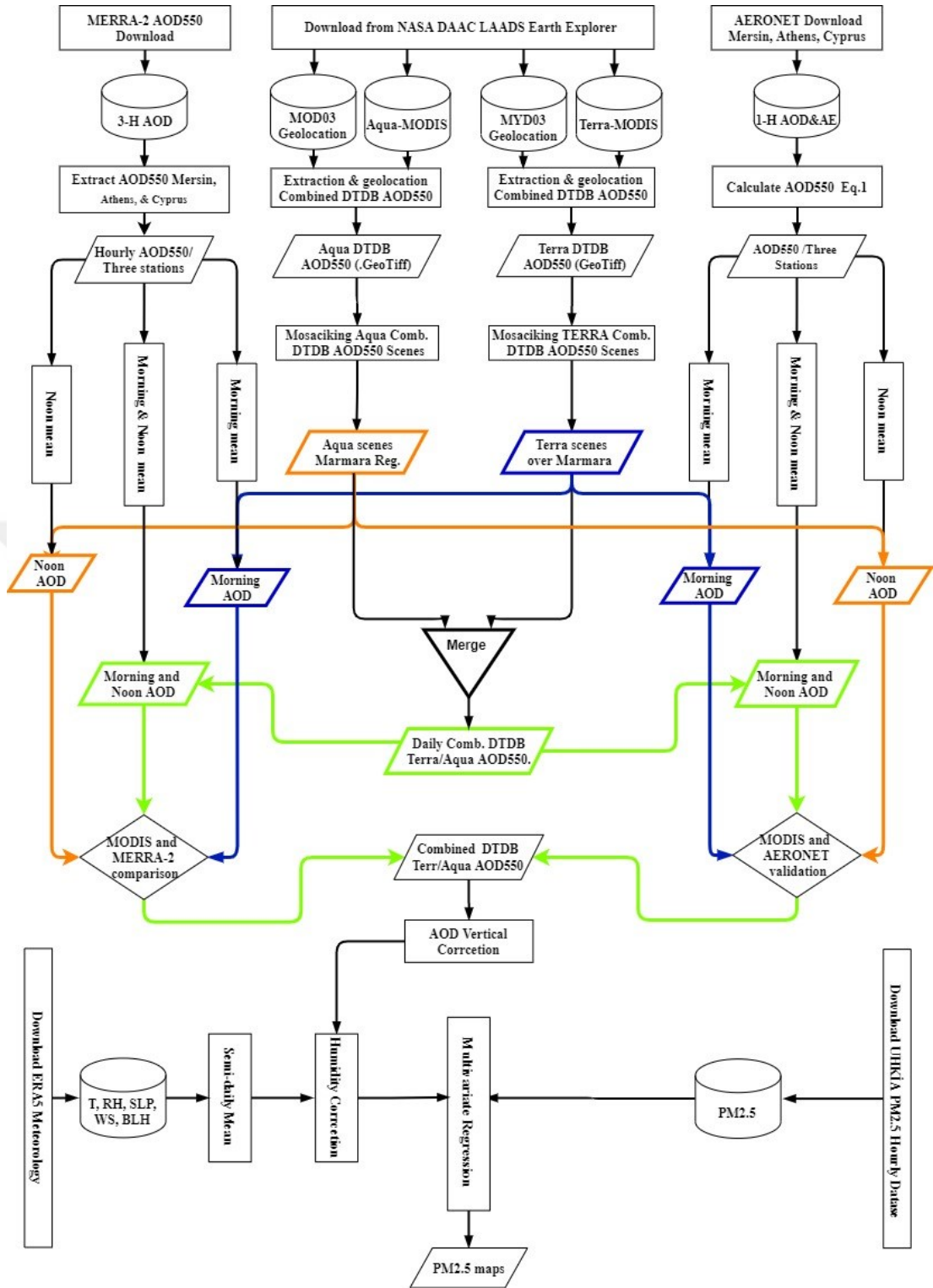


Figure 2.3: Flow chart explaining the data used and the methodology applied in the present study. The MODIS AOD validation processes over the morning, noon, and semi-day were colored by the blue, orange, and green lines. The cylindrical, rectangular, triangular, and diamond-shaped elements indicate a dataset, a process, a merge, and an output, respectively.

(vii) AERONET AOD hourly observations were separated and averaged according to the three periods described in (iv). (viii) ECMWF daily RH was extracted over the

three stations and averaged for each season. (IX) The statistical metrics of the coefficient of determination (R^2), root mean square error (RMSE), mean absolute error (MAE), relative mean bias (RMB), and relative root mean square deviation ($RMSE^{rel}$) were calculated (using equations 2.2 – 2.6) and investigated for the satellite and ground-based AOD validation. N is the number of collocated points, and RMB indicates overestimation and underestimation if the value is higher or lower than one, correspondingly.

$$R^2 = 1 - \frac{\sum_{i=1}^N (AOD_{SD(i)} - AOD_{AER(i)})^2}{\sum_{i=1}^N (AOD_{SD(i)} - \mu AOD_{SD})^2} \quad (2.2)$$

$$MSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (AOD_{SD(i)} - AOD_{AER(i)})^2} \quad (2.3)$$

$$MAE = \sum_{i=1}^N |AOD_{SD(i)} - AOD_{AER(i)}| \quad (2.4)$$

$$RMB = \frac{\mu AOD_{SD}}{\mu AOD_{AER}} \quad (2.5)$$

$$RMSE^{rel} = \frac{\sqrt{\frac{1}{N} \sum_{i=1}^N (AOD_{SD(i)} - AOD_{AER(i)})^2}}{\mu AOD_{AER}} \quad (2.6)$$

AOD_{SD} and AOD_{AER} are the AOD derived by MODIS satellite and AERONET, whereas μAOD_{SD} and μAOD_{AER} are the same parameters' mean values, respectively.

2.3.2 Point data and gridded data processing

In the current stage — for every day between 2013 and 2017—hourly gridded datasets of seven ERA5 meteorological parameters (RH, BLH, T, SLP, TP, VWS, and HWS) for the Marmara Region were averaged over seven hours (7:00 and 13:00 LT). Vertical (VWS) and horizontal wind speeds (HWS) were utilized to calculate the wind speed resultant (RWS) using equation 2.7.

$$RWS = \sqrt{(VWS)^2 + (HWS)^2} \quad (2.7)$$

An ArcGIS10.4 extraction automated model, designed by the model builder tool, for extraction was used to operate a 30x30 km² (3x3 MODIS pixels) extraction window to obtain the daily values from the ERA5 parameters as well as the combined MODIS

Terra and Aqua AOD layers over the 12 PM2.5 ground measuring stations. However, the PM2.5 ground measuring stations were centered in the middle of the extraction window, and, on the contrary of the validation process, the extraction outcomes that included at least five MODIS pixels were chosen as daily collocated points. For the compatibility of MODIS AOD, ERA5 meteorology, and PM2.5 data over the time, PM2.5 ground-measured data were also averaged over the same period ERA5 was averaged (7:00-13:00 LT).

2.3.3 Model design and validation

Humidity correction

Satellite sensors retrieve AOD outcomes in the atmosphere under ambient conditions. However, aerosol optical properties – particularly the aerosol light scattering – are changed due to the experienced particle's hygroscopic growth in the ambient conditions (Zieger et al, 2010). Contrarily, PM2.5 ground-measuring stations are recording the dry mass concentrations of PM2.5 at the ground-surface level. Since aerosol particles' size is changed due to water uptake, aerosol light scattering is significantly dependent on the relative humidity (RH) in the ambient atmospheric conditions. Thus, the relative humidity correction aims to quantify the hygroscopicity effects of the dry-measured ground-level PM2.5 mass concentrations (Koelemeijer et al, 2006; Wu et al, 2019). The particles extinction coefficient $f(RH)$ equation 2.8 is the ratio between the particles at ambient and at dry ($RH < 40\%$) relative humidity (Nabavi et al, 2019; Wang et al, 2010; Zeng et al, 2018).

$$f(RH) = \frac{PM2.5_{(ambient)}}{PM2.5_{(dry)}} \quad (2.8)$$

thus, the PM2.5 at ambient condition is calculated using equation 2.9

$$PM2.5_{(ambient)} = PM2.5_{(dry)} * f(RH) \quad (2.9)$$

Where $PM2.5_{(dry)}$, $PM2.5_{(ambient)}$, and $f(RH)$ are the PM2.5 values at dry conditions, PM2.5 at ambient conditions, and particles extinction coefficient, respectively.

The $f(RH)$ is an exponential formula equation 2.10 as a function of RH, in which the value of the extinction coefficient is non-linearly increasing as the relative humidity increases (Hoff and Christopher, 2009; Tsai et al, 2011; Veefkind et al, 1996).

$$Exp_{(PM2.5, RH)} = f(RH) = \frac{1}{\left(1 - \frac{RH}{100}\right)^g} \quad (2.10)$$

Where RH refers to the ambient relative humidity, and g is an empirical constant experimentally assigned to one (Bhattarai et al, 2019; Chen et al, 2017; Wu et al, 2019; Zheng et al, 2015).

Vertical correction

AOD is a columnar measurement presenting the aerosol optical and microphysical properties by integrating all of the aerosol extinction light along the atmospheric column (Boiyo et al., 2018). However, PM2.5 ground stations measure dry mass concentrations of PM2.5 at the surface level (Height ≈ 0). Thus, it is in consequence, to conduct a direct association between ground-measured PM2.5 and sensor-derived AOD directly (Gong et al, 2017). Therefore, we have applied a vertical correction on the MODIS AOD to obtain AOD at the surface level. It is assumed that the aerosol particles are uniformly distributed through the boundary layer height (BLH) since it is an actively turbulent atmospheric layer (Gong et al, 2017; Koelemeijer et al, 2006; Wu et al, 2019). Thus, the AOD at the level of the Earth's surface (AOD_{h0}) is obtained from equation 2.11 (Koelemeijer et al, 2006; Li et al, 2018).

$$AOD_{h0} = \frac{AOD_{ambient}}{BLH} \quad (2.11)$$

Where AOD_{h0} is the Aerosol Optical Depth at ground level ($h=0$) and BLH is the boundary layer height.

Excluding outliers

The window size was chosen based on the mean wind speed in the Marmara Region (3.3 m s^{-1}) according to, which means that particulate aerosol can be carried for 16 km in one hour. The increasing number of averaged pixels for AOD was found to improve the representativity of AODs and reduces bias (Glantz et al, 2009; Mei et al, 2019). The PM2.5 station is centered at the middle of the nine pixels, and we selected the extraction outcomes that contained at least five MODIS pixels.

Since low PM2.5 values are difficult to be detected by the statistical models, PM2.5 values less than five $\mu\text{g}/\text{m}^3$ were excluded from the model. This step was found effective by (Nabavi et al, 2019). The authors removed very low values of PM2.5,

which falls lower than the first quartile (Q1) they found an increase in the R^2 from 0.61 to 0.72. In addition, the threshold of $5 \mu\text{g}/\text{m}^3$ is very likely to be recorded by interactive activities around the ground stations, such as vehicle passing or domestic emissions.

Furthermore, water uptake by relative humidity changes the microphysical characteristics of aerosols (Burgos et al, 2019; Zieger et al, 2010). Solar light scattering of aerosols particles in ambient conditions where RH is more than 90%, was considered dominated by water uptake (Shinozuka et al, 2007). The authors have found a strong correlation ($R^2=0.77$) between satellite AOD and PM2.5. For the present study, the daily matchups with RH values more than 90% were removed to improve the regression and reduce bias.

Model design

By conducting the humidity and vertical corrections on the PM2.5 ground measurements and the MODIS AOD, both the variables are changed into surface-ambient condition; thus, the regression can be operated at this stage. The seven-parameter additive model of the exponential of the simulated MODIS AOD, RH, BLH, SLP, T, TP, and RWS were designed using Minitab 16 software. Besides, since the anthropogenic and meteorological conditions are varying over time; and aerosol is a time-dependent variable, monthly and seasonal experimental predictors was added to optimize the accuracy of the model and reduce bias (Mao et al, 2012; Stafoggia et al, 2020; Yazdi et al, 2020), see equation 2.12.

$$PM2.5 = \frac{P(i,j)}{\text{Exp}(PM2.5, RH)} * [\beta_1 AOD_{h0} + \beta_2 RH + \beta_3 BLH + \beta_4 SLP + \beta_5 T + \beta_6 TP + \beta_7 RWS + \varepsilon] \quad (2.12)$$

where PM2.5 is the daily ground-level predicted PM2.5 dry mass concentration ($\mu\text{g}/\text{m}^3$); $P_{(i,j)}$ is the predictor of the month i in the season j; AOD is the ambient daily MODIS AOD (unitless); RH is the relative humidity (%); BLH is the boundary layer height (meter); SLP is the sea level pressure (hecto Pascals); T is the temperature in (Calvin); TP is the total precipitation depth (meter); and RWS is the resultant wind speed in (meter per second). Before the mapping process, the spatial resolution of both the MODIS AOD and ERA5 gridded layers was rescaled into ($3 \times 3 \text{ km}^2$) using an ArcGIS bilinear convolution resampling tool. Pixel sizes were made equal to facilitate the mathematical operations (sum, divide, multiplication, and subtraction) on the

pixels using ArcGIS. Also, the area of the Marmara Region as well as distances between the PM2.5 ground stations are small to be analyzed on the MODIS and ERA-5 spatial resolutions (10 km and 30 km, respectively). Therefore, decreasing pixel size into 3x3 km² was chosen based on previous studies conducted the analysis of PM2.5 (Glantz et al, 2009; Nabavi et al, 2019; Z. Wang et al, 2010; Zeydan & Wang, 2019). In addition, daily maps of the PM2.5 ground concentrations were averaged into monthly maps to enlarge the cloud-free spatial coverage of MODIS AOD data and enable data presentation. The maps of the average-monthly and -annual PM2.5 concentrations are presented in the next chapter, Chapter 3. Concentrations of the PM2.5 were classified and presented in the maps using discrete-period legends to reach a maximum level of data visualization.

Model validation

According to previous studies by Joharestani et al. (2019); Ma et al. (2014); Rodríguez et al. (2010), we utilized a Leave-One-Out cross-validation method to check the robustness of the designed model. We have cross-validated the PM2.5 predictions using two validation processes; both Leave-Year-Out and Leave-Season-Out cross-validations. The first process was repeated 12 times by excluding one ground station from modeling to be used for the validation process. At the same time, the second process (Leave-Season-Out) was done by excluding one season out of the total years of the study period, 2013-2017. Validation results were evaluated using the coefficient of determination (R^2), root mean square error (RMSE), relative mean bias (RMB), and the relative root squared error (RMSE^{rel}) using the equations 2.13 – 2.16, respectively.

$$R^2 = 1 - \frac{\sum_{i=1}^N (PM2.5_{(p)i} - PM2.5_{(m)i})^2}{\sum_{i=1}^N (PM2.5_{(p)i} - \mu PM2.5_{(m)i})^2} \quad (2.13)$$

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (PM2.5_{(p)i} - PM2.5_{(m)i})^2} \quad (2.14)$$

$$RMB = \frac{\mu PM2.5_{(p)}}{\mu PM2.5_{(m)}} \quad (2.15)$$

$$RMSE^{rel} = \frac{\sqrt{\frac{1}{N} \sum_{i=1}^N (AOD_{SD(i)} - AOD_{AER(i)})^2}}{\mu AOD_{AER}} \quad (2.16)$$

Where $PM_{2.5(p)}$ is the predicted $PM_{2.5}$ value; $PM_{2.5(m)}$ is the ground measured $PM_{2.5}$ value; $\mu PM_{2.5(p)}$ and $\mu PM_{2.5(m)}$ are the mean values of the whole predicted and measured $PM_{2.5}$ values, respectively.





3. RESULTS

3.1 Results

3.1.1 Validation of MODIS AOD

MODIS and MERRA-2 daily AOD are compared to AERONET measurements in Figure 3.1, subdivided with respect to the time of the day. MERRA-2 reanalysis contain the highest number of collocated values (N), as it being a continual time series. The figures show that MODIS agree according to the 1:1 line and squared correlation coefficient somewhat better than MERRA-2. However, slightly lower $RMSD^{rel}$ and MAE values are found for MERRA-2. For AOD corresponding to MODIS Aqua and Terra combined, and MERRA-2 Semi-Day, higher R^2 values and $RMSD^{rel}$ are found compared to separated platforms and for morning and noon hours, respectively.

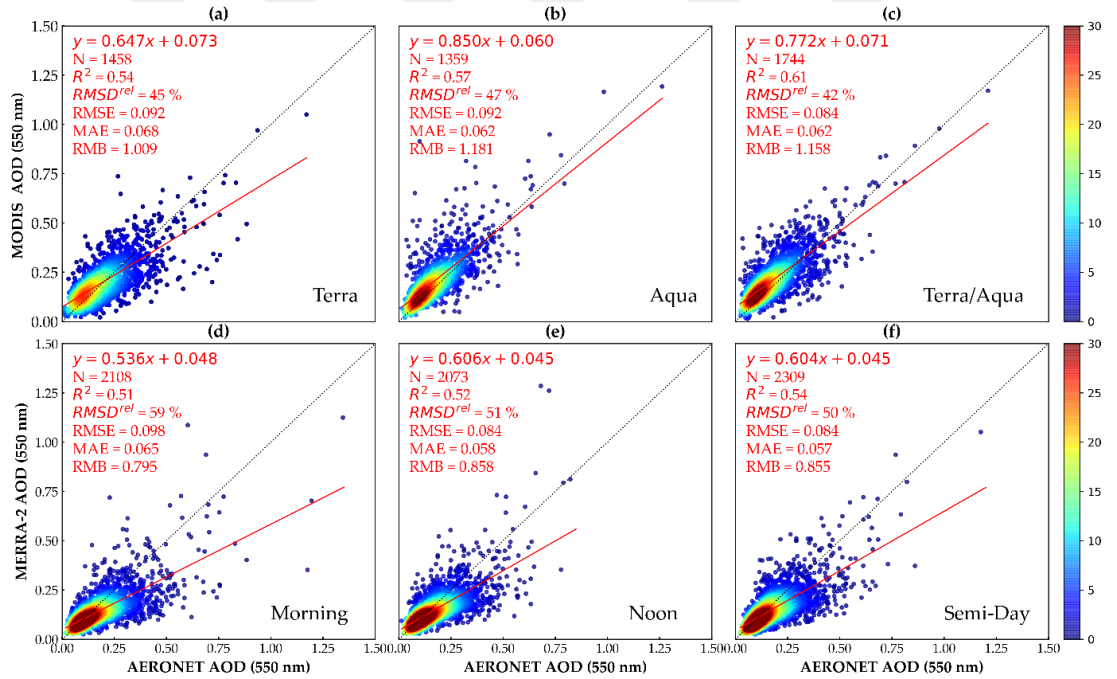


Figure 3.1: MODIS Aqua and Terra, and MERRA-2 AOD against AERONET. (a) MODIS Terra, (b) MODIS Aqua, (c) MODIS Aqua and Terra combined, (d) MERRA-2 morning , (e) MERRA-2, (f) MERRA-2 combined (semi-day) datasets vs. AERONET. The match points values are indicated by the colour bars.

The largest deviation to the 1:1 line is found for MODIS Terra and MERRA-2 AOD obtained for morning hours. Concerning the R^2 and the 1:1 line in the comparison with AERONET, Shaheen et al. (2020) also found an agreement for MODIS better than MERRA-2; However, MERRA-2 was found associated with less bias and slightly closer to the 1:1 line. The study mentioned earlier proposed merging MERRA-2 and MODIS AOD to minimize bias and obtain AOD closed to AERONET. The obtained results with MERRA-2 and MODIS over inlands are in line with the previous study by (Sogacheva et al, 2020). They found that the R^2 values for the validation against AERONET was 0.72 and 0.66 for the MODIS and MERRA-2, respectively. AOD Morning hours of MERRA-2 and MODIS Terra contain the largest deviation to the 1:1 line. The latter results with MODIS Terra are in line with Robert C. Levy et al. (2018), who found a significantly larger bias for the AOD derived by this satellite sensor than the MODIS Aqua. The authors found that the bias values associated with MODIS Aqua and Terra sensors are 0.041 and 0.014, respectively. Accordingly, Schutgens et al. (2020) generalized the MODIS Aqua dataset to represent the MODIS AOD. However, MODIS Terra presented the best consistency with AERONET over the East Mediterranean (Georgoulas et al, 2016).

Time series of MODIS, MERRA-2, and AERONET AOD

MODIS and MERRA-2 monthly averaged AOD were compared to AERONET AOD over the study period of 5 years. MODIS and AERONET observations are not continuous time series, while combining monthly values resulted in only one missing month out of 60 for both platforms. For MODIS, available days included in the monthly averaging ranged between 44% in January and 92% in August. Figure 2 shows that the MODIS AOD values are higher compared to both AERONET and MERRA-2 with respect to the period 2013 - 2015, while more comparable with AERONET for 2016 and 2017. Figure 3.2 shows also that MODIS and MERRA-2 somewhat overestimate and underestimate AOD, respectively, with respect to AERONET AOD when the full period is taken in consideration. Larger deviation in RMB is found when comparing MODIS and MERRA-2 AOD, where substantially lower values are found for the latter platform both with respect to monthly and daily averaging in Figure 3.1. Georgoulas et al. (2016) explained the latter results with the long-range transport of mineral dust aerosol over the Anatolian Plateau in Turkey from

the Middle East and North Africa in summer. This result was also supported by (Perez et al., 2006).

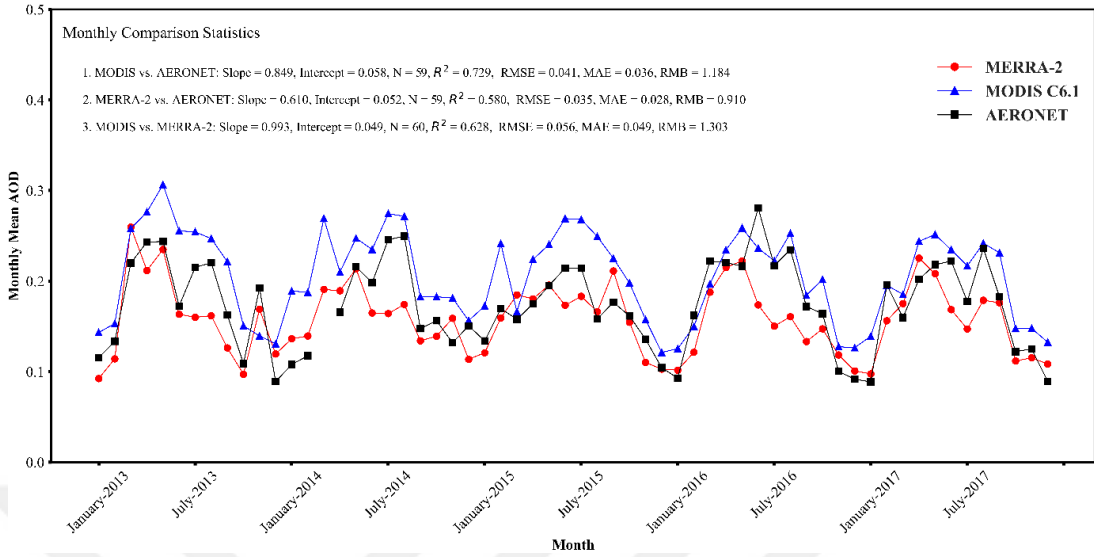


Figure 3.2: Monthly mean AOD and the corresponding statistics for MERRA-2, MODIS, and AERONET.

Table 3.1 presents statistics between MODIS and MERRA-2 against AERONET with respect to season and the three present AERONET sites. For the purpose of investigating seasonal variation, four seasons were classified: (i) winter (December, January and February – DJF), (ii) spring (March, April and May – MAM), (iii) summer (June, July and August – JJA), and (iv) autumn (September, October and November – SON). For MODIS, available days are 63, 85, 71 and 51% for summer, autumn and winter respectively. The availability for AERONET AOD is largest in August (94%) and reached minimum in January (44%), while the seasonal availability is 57, 75, 57 and 44% for spring, summer, autumn and winter, respectively. For the ATHENS-NOA station, Table 3.1 shows that the lowest RH and highest R^2 were recorded in summer for both MODIS and MERRA-2. Similar findings are found for the stations METU-ERDEMLI and CUT-TEPAK according to spring/summer and winter, respectively. Considering the other statistical parameters in Table 3.1 no clear relation with RH is found.

Table 3.1: Seasonal statistical metrics of MODIS and MERRA-2 AOD against AERONET for the (I) Athens-NOA, (II) METU-ERDEMLI, and (III) CUT-TEPAK.

Station (I)		ATHENS-NOA Athens							
Dataset		MODIS C6.1				MERRA-2			
Season	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	
RH (%)	73	67	64	72	73	67	64	72	
N	51	95	241	148	148	157	322	235	
Slope	0.604	0.831	0.882	0.673	0.564	0.662	0.691	0.540	
Intercept	0.061	0.063	0.072	0.094	0.031	0.054	0.033	0.062	
R ²	0.545	0.590	0.666	0.527	0.533	0.576	0.652	0.511	
RMSE	0.051	0.079	0.083	0.125	0.059	0.074	0.062	0.073	
MAE	0.036	0.061	0.063	0.079	0.039	0.053	0.044	0.047	
Station (II)		METU-ERDEMLI Mersin							
RH (%)	64	56	56	63	64	56	56	63	
N	101	176	277	183	120	235	323	215	
Slope	0.771	0.819	0.746	0.804	0.653	0.596	0.559	0.464	
Intercept	0.069	0.079	0.053	0.067	0.016	0.050	0.015	0.042	
R ²	0.563	0.641	0.614	0.560	0.503	0.567	0.539	0.520	
RMSE	0.071	0.086	0.079	0.078	0.069	0.084	0.137	0.082	
MAE	0.054	0.067	0.058	0.056	0.052	0.060	0.114	0.058	
Station (III)		CUT-TEPAK Cyprus							
RH (%)	56	64	65	63	56	64	65	63	
N	105	170	100	97	140	201	103	110	
Slope	0.780	0.794	0.723	0.921	0.567	0.646	0.668	0.435	
Intercept	0.054	0.102	0.111	0.050	0.030	0.053	0.062	0.063	
R ²	0.798	0.645	0.639	0.699	0.634	0.589	0.542	0.573	
RMSE	0.047	0.104	0.085	0.066	0.062	0.087	0.062	0.093	
MAE	0.036	0.075	0.060	0.048	0.040	0.048	0.049	0.053	

Figure 3.3 shows annual mean scenes of MODIS Aqua and Terra combined and MERRA-2 AOD (left and middle column, respectively) and corresponding differences in AOD between the two platforms (right column) with respect to the present investigation period. For the years 2013, 2015 and 2016 the figures show that MODIS AOD is higher than MERRA-2 for nearly the full study area. MODIS $10 \times 10 \text{ km}^2$ and MERRA-2 $50 \times 62.5 \text{ km}^2$ pixel sizes were converted to a grid of $50 \times 50 \text{ km}^2$ to obtain the same spatial resolution when comparing the two datasets.

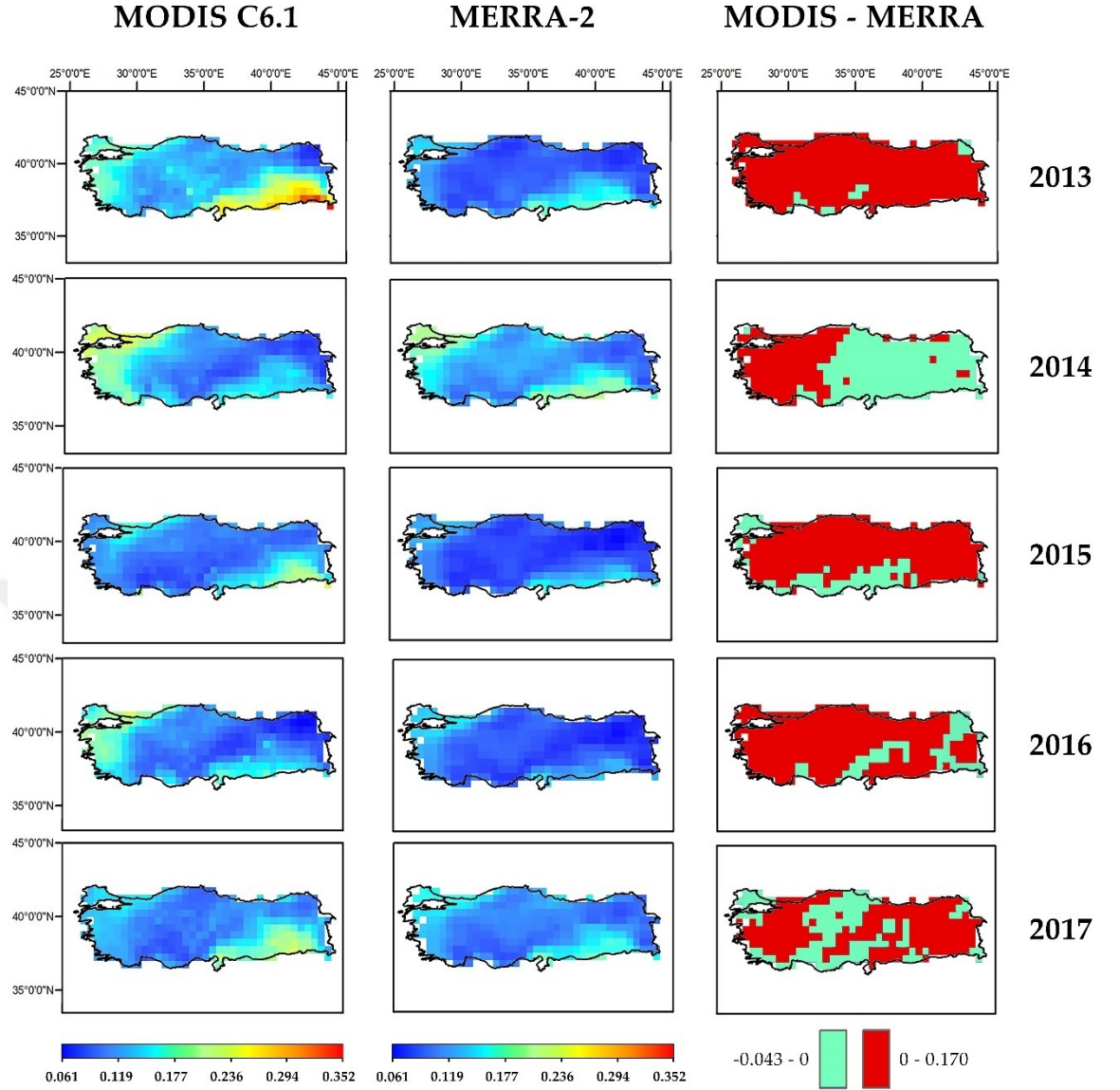


Figure 3.3: Average annual scenes of MODIS Aqua and Terra combined and MERRA-2 are presented in the first and second column, respectively, while the third column shows the difference in AOD between MODIS and MERRA-2 over Turkey.

Extreme events

Aerosol extreme events were specified for MODIS/MERRA-2 and AERONET as daily mean AOD greater than or equal to the threshold value 0.5 (Sehatakhani et al, 2016; Van Donkelaar et al, 2011). Extreme events were investigated separately for the three AERONET stations and included if daily mean values are available for all three platforms. According to AERONET-derived AOD, Athens, METU, and CUT-TEPAK have recorded 8, 20, and 7 extreme events, respectively, during the five years included in the present investigation. Table A.1 in Appendix A shows that MODIS detected a total of 18 (54%) extreme events out of the 33 by AERONET, while MERRA-2

detected only 8 (24%). Previous studies such as that of H. Shi et al. (2019) showed that AOD events of dust and haze were not detected very well by MERRA-2. Li et al. (2009) demonstrated the poor performance of MODIS in detecting extreme loadings of mineral dust aerosol over 13 AERONET stations in China. Jin et al. (2019) showed that although MODIS somewhat underestimated extreme aerosol events, the satellite sensor was actually effective in detecting extreme events recorded by four AERONET stations around the Yangtze River in China.

3.1.2 Data description

Tables 3.2 presents statistics for daily datasets of seven parameters included in the model of estimating PM_{2.5}. Measurements of PM_{2.5} obtained by 12 ground-stations were daily averaged with respect to 56 months (Between March/2013 and October/2017) for the Marmara Region. The results of the table show relatively high bias and variation in the dataset of PM_{2.5}. The RMSE^{rel} is 48.68%, and the values resulted from the daily averaging ranged between 6.07 µg/m³ (Minimum) and 70.03 µg/m³ (Maximum).

Table 3.2: Descriptive statistics of the used parameters with respect to the whole study period (2013–2017). Statistics of PM_{2.5} were calculated from averaging daily values obtained from the 12 ground stations, while statistics of AOD and meteorological parameters by averaging the raster datasets (MODIS and ERA-5, respectively) over the Marmara Region.

Parameter	Mean	Median	Min.	Max.	STD	RMSE ^{rel}	Confidence Interval	
							5%	95%
PM _{2.5} (µg/m ³)	21.17	18.224	6.069	70.027	10.310	48.68	0.489	0.016
AOD550 nm	0.220	0.194	0.013	1.014	0.133	60.45	0.007	0.000218
BLH (m)	510.1 1	506.43	20.68	1530.00	289.74	56.80	13.749	0.440
RH (%)	70.07 8	69.983	43.020	95.731	9.886	14.11	0.469	0.015
SLP (Bar)	1.016	1.015	0.995	1.039	0.007	0.69	0.0003 2	1.0079E-05
T (C°)	15.29 6	15.508	-4.421	28.571	7.403	48.40	0.351	0.011
TP(mm)	0.356	0.067	0.000	5.942	0.683	191.85	0.032	0.001
WS (ms ⁻¹)	3.222	3.027	0.276	12.534	1.559	48.39	0.074	0.002

The latter results roughly indicate that the annual average EUs' concentration limit of PM_{2.5} (25 µg/m³) was not surpassed in the present study area. Concerning the aforementioned study period, the mean and median values of the PM_{2.5} were 21.18 µg/m³ and 18.22 µg/m³, respectively.

Datasets of meteorological parameters are spatially and temporally continual. However, AOD datasets of MODIS involve a high percentage of missing pixels because of clouds. Available pixels in AOD scenes of MODIS included in the daily averaging ranged between 38% in January and 88% in August over the Marmara Region. The daily mean of AOD for the present study period is 0.22. The table shows that the AOD dataset range is somewhat short; the maximum and minimum values are 0.013 and 1.014, respectively. However, comparing to other datasets of variables, large $RMSE^{rel}$ (60.45%) was found in the AOD.

Opposing to AOD and PM2.5, Table 3.2 presents that the lowest variation is found in the dataset of the sea level pressure (SLP) parameter. The STD of the SLP dataset was 0.007 Bar, while the highest and lowest daily average SLP values were 0.995 Bar and 1.039 Bar, respectively. The SLP dataset contained the least $RMSE^{rel}$ (0.69%) compared to datasets of other variables. On the contrary, the highest $RMSE^{rel}$ (191.85%) was found for the total precipitation dataset (TP). The latter finding means that the sea level pressure is not varying significantly in the study area with respect to time. However, significant variation in the TP dataset is caused by days with no rainfall (TP = 0 mm) in the Marmara Region.

The daily air temperature is moderate for the present study area. The mean and median of the Temperature (T) dataset were 15.3 C° and 15.5 C°, respectively. Over the course of the study period, the temperature ranged between -4.4 C° (Minimum) and 28.6 C° (Maximum). However, relative humidity (RH) is somewhat high and not varying significantly for the Marmara Region. RHs' daily mean and median are almost identical (70%), while its minimum and maximum values are 43% and 95.7%, respectively. Table 3.2 showed a low $RMSE^{rel}$ value of RHs' dataset (14.11%) compared to the temperature dataset. Contrarily, the datasets of wind speed (WS) and boundary layer height (BLH) contained high $RMSE^{rel}$, 48.3% and 56.80%, respectively.

Table A.2 presents statistics of the datasets of the seven parameters with respect to seasons. Years were divided into seasons (as in section 3.1.2) to investigate seasonal variations in the datasets. The table shows similar values of seasonal average concentrations of PM2.5 for spring and autumn ($\approx 21 \mu g/m^3$), while the highest and lowest values were in winter ($28.76 \mu g/m^3$) and summer ($15.54 \mu g/m^3$), respectively. The bias of daily mean PM2.5 values with respect to the seasonal mean is the largest

in summer ($STD = 13.2 \mu\text{g}/\text{m}^3$), while autumn is the season where $RMSE^{rel}$ is the highest (51.10%). Contrarily to the whole of Turkey as a study area (section 3.1.1), the seasonal mean of MODIS AOD was equal for every two successive seasons $AOD = 0.25$ (for spring and summer) and $AOD = 0.16$ (for winter and autumn). However, seasonal mean datasets of AOD contained a relatively large bias comparing to other datasets of the parameters; $RMSE^{rel}$ is 72.5%, 64.5%, 45.2%, and 56.7% for winter, spring, summer, and autumn, respectively.

Winter months are humid in the Marmara Region. The seasonal mean relative humidity in winter reached up to 93%, with mean and median of 73.3% and 75%, respectively. Opposing to winter, the latter parameter of RH declines for spring and summer ($RH \approx 67.5\%$). With respect to bias from the seasonal mean, $RMSE^{rel}$ ranged between 15.4% and 11.5% for spring and summer, respectively. On the other hand, the highest $RMSE^{rel}$ values among seasonal datasets of BLH were found for spring (65.10%) and winter (67.47%). Table A.2 shows that the highest seasonal mean for the datasets of BLH is for autumn ($BLH = 647 \text{ m}$), while it is comparable for other seasons ($BLH \approx 460 \text{ m}$).

Precipitation is not continuous over the course of the years in the Marmara Region. Four of seasons, including winter, contain days with no rainfall ($TP = 0$). Availability of rainy days is different among seasons. For the present study period, the seasonal availability of days with rainfall is 76.6%, 71.7%, 70.7%, and 52.5% for winter, spring, summer, and autumn, respectively. In addition, daily mean precipitation is maximum for seasons of low temperature, 0.476 mm and 0.445 for winter and spring, correspondingly. The seasonal mean temperature declines in winter ($T = 5.9 \text{ C}^\circ$) and reaches its maximum in summer ($T = 23.72 \text{ C}^\circ$). Contrarily, the largest and smallest seasonal $RMSE^{rel}$ values of temperature datasets were found for summer and winter, 9.84% and 67.78%, respectively. Characteristics of all the utilized datasets, both for aerosol and meteorology, are presented in Table A.3.

Figure 3.4 illustrates Violin and Box-Whisker combined plots for daily mean $PM_{2.5}$ with respect to 12 ground-measuring stations in the Marmara Regions. Concerning five years study period, the $PM_{2.5}$ annual mean limit was exceeded in only three datasets of the stations; Ozanlar, Kağıthane, and Ümraniye. A one-year analysis for the same datasets conducted by Kahya et al. (2017) presented $PM_{2.5}$ values measured by the latter three stations higher than other stations and illustrated right-skewed

distribution. The datasets of these stations contained the most extended ranges of data, between $0.8 \mu\text{g}/\text{m}^3$ and $117.8 \mu\text{g}/\text{m}^3$. Datasets involved missing values because of instruments malfunction. The percentage of missing measurements among stations ranged between 3% and 8% (Table 2.3). The figure shows that the lowest and highest mean values of PM_{2.5} with respect to the whole dataset are for Kandıra and Ozanlar, respectively. However, the same values were assigned to Lapseki and Ozanlar in (Kahya et al, 2017). Zeydan & Wang (2019) also supported these findings.

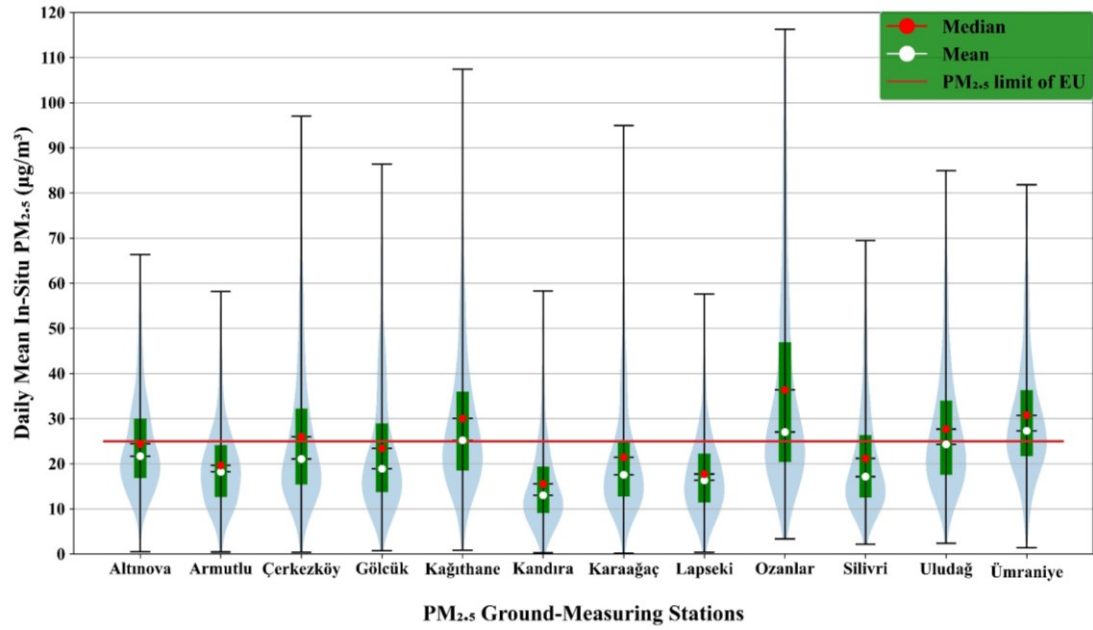


Figure 3.4: Violin and Box-Whisker combined plot for average daily PM_{2.5} with respect to 12 ground-based sites in the Marmara Region during 2013–2017. Values of the Median, Mean, and EU limit of PM_{2.5} are indicated by the red points, white points, and the red horizontal line, respectively. However, Green boxes present PM_{2.5} values between the 1st quartile (Q1-lower limit) and 3rd quartile (Q3-upper limit).

3.1.3 Estimation model of PM_{2.5}

The model of estimation dry mass concentrations of PM_{2.5} included eight stages. Table 3.3 presents every steps' contribution to models' accuracy with respect to the coefficient of determination (R^2). The models' development steps were applied for every station separately. Linear regression between MODIS AOD and ground measured PM_{2.5} was found weak respecting R^2 ; overall R^2 is 0.15. Engel-Cox et al. (2004); X. Xu & Zhang (2020) also demonstrated a low R^2 for the linear regression between ground-measured PM_{2.5} and MODIS AOD. However, applying the non-linear (exponential extinction coefficient) $f(RH)$ correction to PM_{2.5} increased the

models' accuracy by 29.87%. In addition, converting AOD obtained by MODIS (columnar AOD) into ground-level AOD using the BLH (vertical correction) contributed to 15.12% of the overall models' accuracy. Correcting ground-measured PM2.5 and MAIAC AOD using $f(RH)$ and ERA-Interim BLH, respectively, in Nabavi et al. (2019); increased the R^2 of the PM2.5-AOD correlation from 0.17 to 0.67. In addition, X. Xu & Zhang (2020) demonstrated the high effectiveness of BLH and $f(RH)$ corrections on increasing R^2 for PM2.5-AOD relationships. The corrections in the latter study results in increasing the R^2 from 0.52 to 0.78.

Table 3.3 shows that adding meteorological variables to the model contributed by 19.28% of the overall model accuracy; R^2 increased from 0.393 to 0.492. Multiple studies have quantified the effects of meteorological variables on PM2.5 estimations models. Yazdi et al. (2020) have utilized five meteorological variables (BLH, WS, SLP, T, and RH) to estimate PM2.5 for London, UK. The results showed that they contributed to 20.24% of the models' overall R^2 . In addition, Lianfa Li et al. (2017) have found that the meteorological variables contributed to 9.2% of the R^2 . In the same study, among meteorological variables, the highest and lowest contributions to R^2 are associated with adding RH (0.03) and TP (0.008), respectively. These findings inline with (Y. Chen et al., 2020; X. Wang et al., 2019).

Table 3.3: Contribution of the individual models' stages to the overall R^2 .

Stage	Overall R^2	Contribution to R^2	Data Availability
PM2.5-AOD Linear Regression	0.152	N/A	48.02%
Non-linear Correction	0.314	29.87%	48.02%
Vertical Correction	0.393	15.12%	48.02%
Climatology Parameters	0.492	19.28%	48.02%
Five-Pixels Filter	0.528	6.80%	41.02%
5 $\mu\text{g}/\text{m}^3$ PM2.5 Filter	0.554	4.91%	38.82%
RH >90% Filter	0.602	9.07%	34.42%
Temporal Predictors	0.681	14.96%	34.42%

The results in Table 3.2 show that excluding daily matchups of AOD resulted from averaging less than five pixels decreased data availability from 48.02% to 41.02%. Yet, the present step increased the overall R^2 to 0.528. The latter raised to 0.554 after removing collocated values with average PM2.5 daily ground measurements less than five $\mu\text{g}/\text{m}^3$; however, it decreased data availability to 38.82%. Nabavi et al. (2019) found that excluding PM2.5 values below the first quartile (Q1) enhanced their

proposed model accuracy. Besides, PM_{2.5} values of less than three $\mu\text{g}/\text{m}^3$ were invalidated by (Sofowote et al, 2018)8.

In addition, days contains RH values of more than 90% were also removed from the model, and the R^2 inclined to 0.602. Stages 6 and 7 were applied to estimate PM_{2.5} over North America by (Shinozuka et al, 2007). Although a large aerosol vertical distribution, removing 4% of the dataset with $\text{RH}>90\%$ resulted in correlating ambient AOD with PM_{2.5} ($R^2 = 0.77$). Finally, overall R^2 reached its maximum (0.681) by adding the temporal (monthly and seasonal) predictors to the model. K. Huang et al. (2018); L. Mao et al. (2012) found temporal predictors are effective in enhancing the predictability of PM_{2.5} estimation models. Multiplying PM_{2.5} ground measurements by the particles extinction coefficient $f(\text{RH})$ increases values of PM_{2.5}. Accordingly, daily mean values of PM_{2.5} were divided by the $f(\text{RH})$ to obtain the dry mass concentrations of PM_{2.5} (section 2.3.3). However, this step decreased the R^2 to 0.67. This result is supported by (Nabavi et al., 2019). The latter stated that the R^2 decreased slightly after dividing RH-corrected PM_{2.5} values by $f(\text{RH})$.

The study period covers 1706 days (56 months). Pixels covered by clouds as well as missing ground-stations measurements affected the availability of matched points for every station (N). The latter ranged between 38.2% and 55.8% (Overall availability is 48.02%). However, filtering the pixels covered by clouds, days of high relative humidity ($\text{RH}>90\%$), and low PM_{2.5} ($\text{PM}_{2.5} < 5 \mu\text{g}/\text{m}^3$) eliminated the number of daily matchups. Table 3.3 presents the availability of daily matchups associated with every step. In contrast, Table 3.4 presents the fixed estimators (β_i) and associated significance (P) for the meteorological variables with respect to every measuring station. Results show that the AOD and RH have overall positive fixed estimators. However, overall negative estimators are presented with respect to BLH, T, TP, WS, and SLP. Tai et al. (2010) analysed correlations between meteorological variables and PM_{2.5} between 1998 and 2008 over the US. They found that temperature, relative humidity, and sea level pressure are positively correlated, while total precipitation is negatively correlated with PM_{2.5}. Jianhua Wang & Ogawa (2015) also found a negative correlation for total precipitation. However, the temperature was found positively correlated with PM_{2.5}.

The table shows that the significance of SLP and AOD is larger than 0.01 for six and eight stations out of 12, respectively. Yet, other variables were found significant for

all datasets of the measuring sites. The highest and lowest R^2 are presented for Karaağaç (0.729) and Ümraniye (0.575) PM2.5 measuring sites. Nonetheless, datasets of Kağıthane and Silivri contained the highest and lowest $RMSE^{rel}$, 44.48% and 10.73%, respectively. After dividing the model-obtained PM2.5 by $f(RH)$, predicted PM2.5 is compared to ground-measured PM2.5 in Figure 3.5, which is subdivided with respect to the ground-measuring sites. The subfigures show that the predicted PM2.5 for Lapseki and Armutlu agree according to the 1:1 line somewhat better than other stations. Stations associated with relatively high PM2.5, i.e., Karaağaç, Kağıthane, and Ozanlar, are also related to high bias values, $RMSE^{rel}$ and MAE. The largest deviation to the 1:1 line was found for Ozanlar and Kağıthane. The smallest $RMSE^{rel}$ (27%) was found for Armutlu, Lapseki, silivri, and Uludağ, while Çerkezköy contained the largest $RMSE^{rel}$ (38%). The RMSE ranged between 9.81 $\mu g/m^3$ for Ozanlar and 4.02 $\mu g/m^3$ for Lapseki. These findings inline with the spatiotemporal analysis of PM2.5 for the Marmara region conducted by (Kahya et al, 2017).

Table 3.4: Contribution of the models' stages to overall R².

Station	Intercept		AOD _{h0}		RH (%)		BLH (m)		SLP (Bar)		T (C°)		TP (mm)		WS (ms-1)		Statistics	
	β1	P	β1	P	β2	P	β3	P	β4	P	β5	P	β6	P	β7	P	R ²	RMSE ^{rel} (%)
Altinova	129.44	P = 0.295	2628.06	P < 0.001	1.893	P < 0.001	-5.037	P = 0.108	112.37	P = 0.351	-1.671	P < 0.001	2.107	P = 0.028	-6.004	P < 0.001	0.609	33.16
Armutlu	173.96	P = 0.026	5165.37	P < 0.001	1.564	P < 0.001	-4.893	P < 0.01	-223.01	P < 0.01	-0.164	P = 0.017	2.635	P < 0.001	-3.353	P < 0.001	0.672	32.24
Cerkezkoy	164.77	P = 0.108	10585.26	P < 0.001	0.933	P < 0.001	-2.952	P = 0.238	-143.99	P = 0.151	-1.537	P < 0.001	1.076	P = 0.269	-1.479	P < 0.001	0.603	32.06
Golcuk	39.13	P = 0.804	5911.62	P < 0.001	1.570	P < 0.001	-14.297	P < 0.001	61.92	P = 0.688	-3.453	P < 0.001	-2.944	P < 0.01	-3.710	P < 0.001	0.625	37.42
Kagithane	626.13	P < 0.01	418.37	P = 0.285	2.342	P < 0.001	-0.135	P = 0.831	-564.00	P < 0.01	-4.865	P < 0.001	4.288	P = 0.015	-6.412	P < 0.001	0.604	44.48
Kandira	284.12	P < 0.001	2862.78	P < 0.001	1.239	P < 0.001	0.007	P < 0.001	-280.07	P < 0.001	-1.705	P < 0.001	-1.640	P = 0.010	-2.613	P < 0.001	0.674	32.18
Karaagac	578.20	P < 0.001	382.66	P = 0.507	3.789	P < 0.001	-1.089	P = 0.744	-1077.15	P < 0.001	-2.599	P < 0.001	-1.622	P = 0.086	-5.610	P < 0.001	0.629	31.28
Lapseki	55.36	P < 0.01	190.64	P = 0.690	1.446	P < 0.001	0.423	P = 0.808	164.70	P = 0.041	0.504	P < 0.001	-0.073	P = 0.879	-1.772	P < 0.001	0.561	28.12
Ozanlar	777.60	P < 0.001	3403.12	P < 0.001	2.385	P < 0.001	-15.516	P < 0.001	-820.75	P < 0.001	-6.131	P < 0.001	-4.034	P = 0.011	-2.322	P = 0.055	0.681	39.20
Silivri	38.47	P = 0.888	1006.53	P < 0.01	-0.771	P = 0.772	-1.732	P < 0.001	19.89	P = 0.853	-2.366	P < 0.001	0.032	P < 0.01	-3.862	P < 0.001	0.668	10.73
Uludag	420.85	P < 0.001	1264.14	P < 0.01	2.173	P < 0.001	-3.737	P = 0.106	-424.09	P < 0.001	-2.056	P < 0.001	0.550	P < 0.01	-7.492	P < 0.001	0.672	23.61
Umraniye	199.82	P = 0.171	909.66	P = 0.122	1.886	P < 0.001	-11.080	P < 0.01	-195.88	P = 0.168	-2.536	P < 0.001	-0.292	P = 0.082	-2.519	P < 0.001	0.575	26.19
Combined	257.14	P < 0.001	1796.52	P < 0.001	1.946	P < 0.001	-1.504	P < 0.01	-241.74	P < 0.001	-2.775	P < 0.001	-0.073	P < 0.01	-3.880	P < 0.001	0.681	29.48

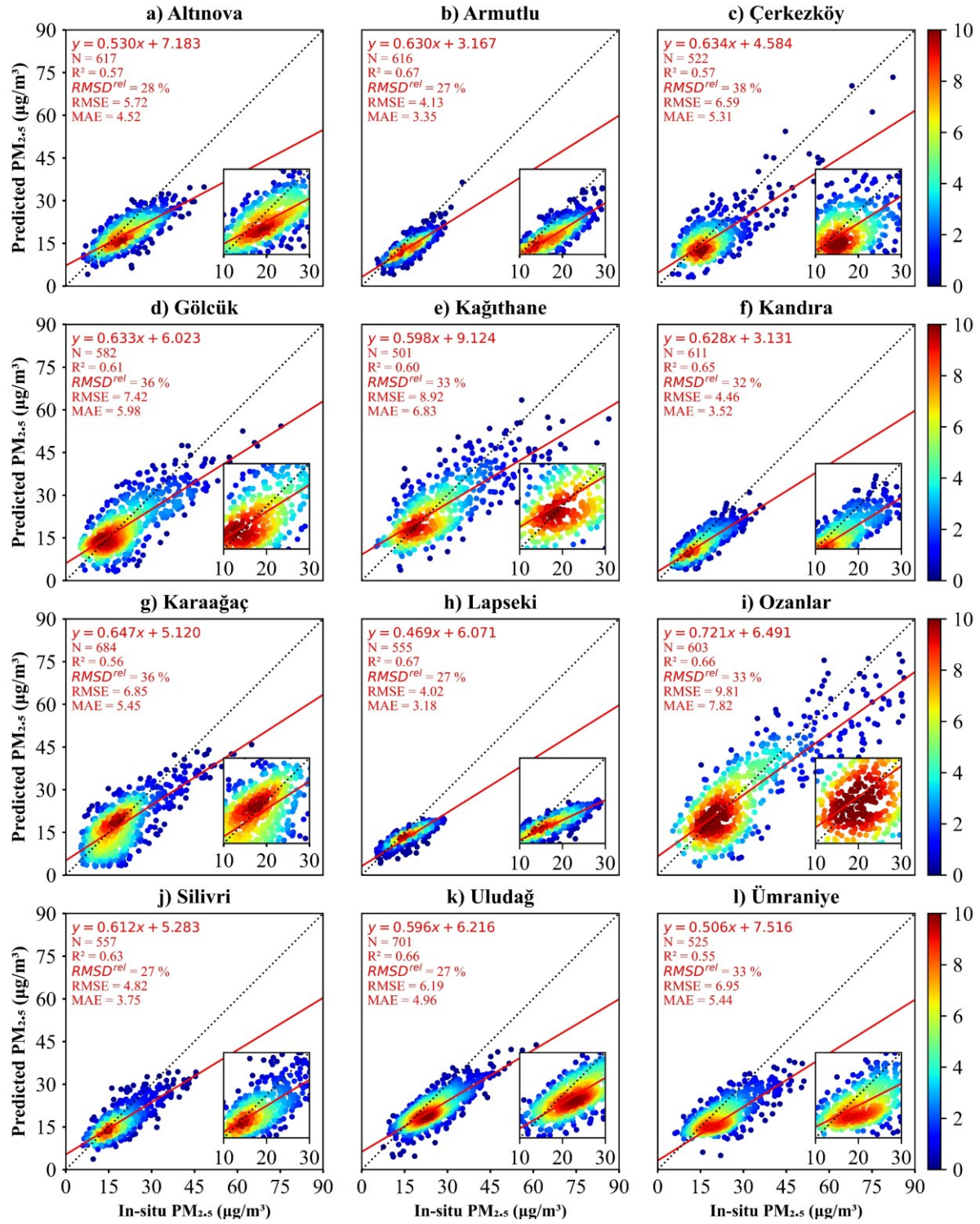


Figure 3.5: Comparison between the model-predicted and the ground-measured PM_{2.5} values over the 12 stations individually. The frequency of match points values is indicated by the color bars.

Figure 3.6 combines the comparisons between predicted and measured PM_{2.5} for the 12 measuring sites. The overall R² is 0.67, while the total number of matched daily points (N) is 7074, comprising 34.44% of the study periods' whole daily points.

The comparisons' bias declined to 28% and 5.94% with respect to $RMSE^{rel}$ and RMSE, respectively; however, the overall R^2 is 0.67.

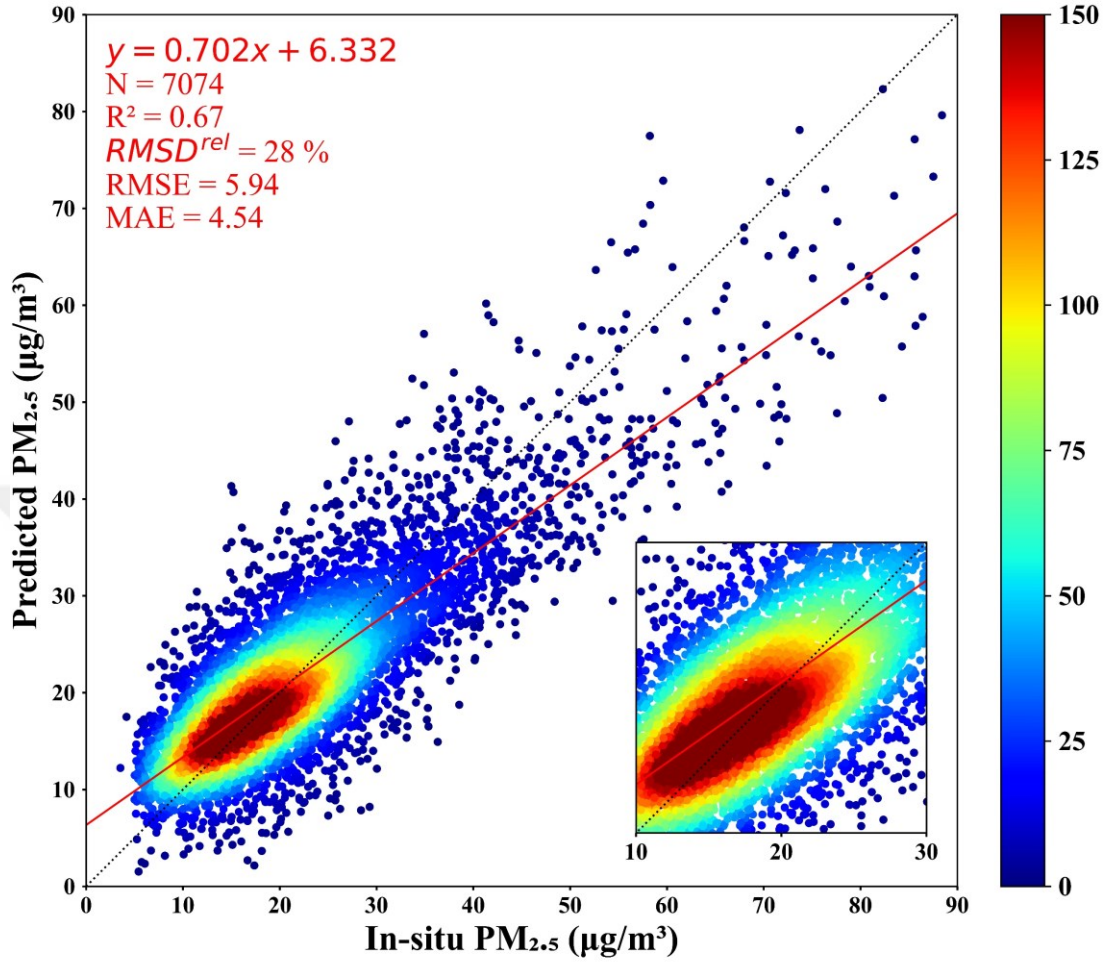


Figure 3.6: Comparison between the model-predicted and the ground-measured PM_{2.5} values over the 12 stations combined. The frequency of match points values is indicated by the color bars.

3.1.4 Time series of measured and predicted PM_{2.5}

Model-predicted PM_{2.5} was compared to ground-measured PM_{2.5} over the study period of 2013-2017. Ground-measured PM_{2.5} was averaged according to 12 ground-based sites in the Marmara Region, while model-predicted PM_{2.5} resulted from averaging daily scenes (rasters) of estimated PM_{2.5}. For ground-measured PM_{2.5}, available measurements included in the daily averaging ranged between 10 and 12 values per day, while the availability of rasters included in averaging the model-predicted time-series was 78.53%. Despite the four months missing from the time-series of the ground-measurements dataset (Jan/2013, Feb/2013, Nov/2017, and Dec/2017), we have estimated PM_{2.5} concentrations for these months. The model equations were applied to the meteorological parameters and MODIS AOD for the

missing period to predict PM_{2.5}. Figure 3.7 presents a comparison between daily time-series for ground-measured and model-predicted PM_{2.5} during the present study period. The figure plots show that, for both time-series, PM_{2.5} concentrations decrease in July and reach their maximum in December/January. With respect to monthly variability of PM_{2.5}, Flores et al. (2020) also found maximizing and minimizing of PM_{2.5} in July and December, respectively, for four sites in Istanbul. Besides, (Kahya et al., 2017) presented that the minimum and maximum of PM_{2.5} occurs in the Marmara Region in July and January, respectively. However, PM_{2.5} concentrations are seen similar in spring and autumn. Statistics in Figure 3.7 shows that the predicted PM_{2.5} somewhat underestimated ground measured PM_{2.5}; the relative mean bias (RMB) is 0.863. Accordingly, extreme events in the ground-measured time-series are underestimated by the model-derived PM_{2.5}. Also, the comparison presented a relatively high RMSE (9.32 $\mu\text{g}/\text{m}^3$) and MAE (6.70 $\mu\text{g}/\text{m}^3$).

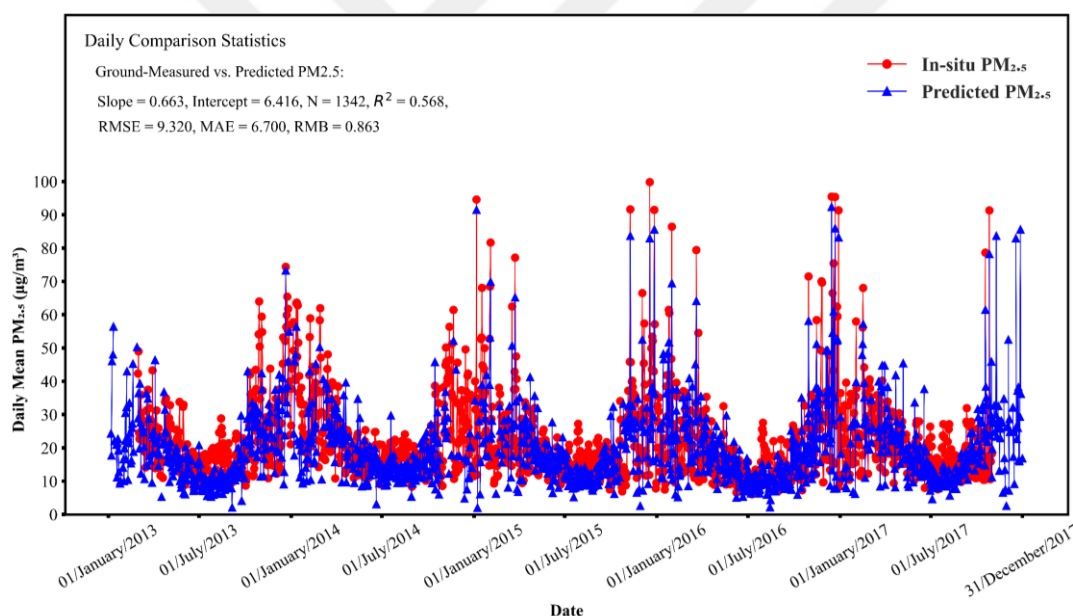


Figure 3.7: Daily time series of the measured and predicted average daily PM_{2.5} over the Marmara Region during 2013-2017.

Plots and statistics comparing monthly time-series of ground-measured and model-derived PM_{2.5} are illustrated in Figure 3.8. Despite discontinuity of daily-average time-series (Figure 3.7), combining monthly values resulted in a continual monthly time-series (Figure 3.8). For the predicted PM_{2.5}, the availability of days included in the monthly averaging ranged between 44.3% in January and 93.4% in July. The figure shows that, for both time-series, the lowest and highest concentrations of PM_{2.5} are found in summer and winter, respectively. These findings are supported by (Flores et

al. 2020; Kahya et al. 2017; Tepe & Doğan, 2021). Statistics of the comparison show that the monthly time-series agree to the 1:1 line better than the daily time-series with respect to R^2 ; in addition, it is associated with less bias (RMSE, MAE). Similar to the daily time-series, model-derived underestimated the ground-measured monthly PM2.5, yet RMB was found closer to one (RMB=0.927) than in daily time-series.

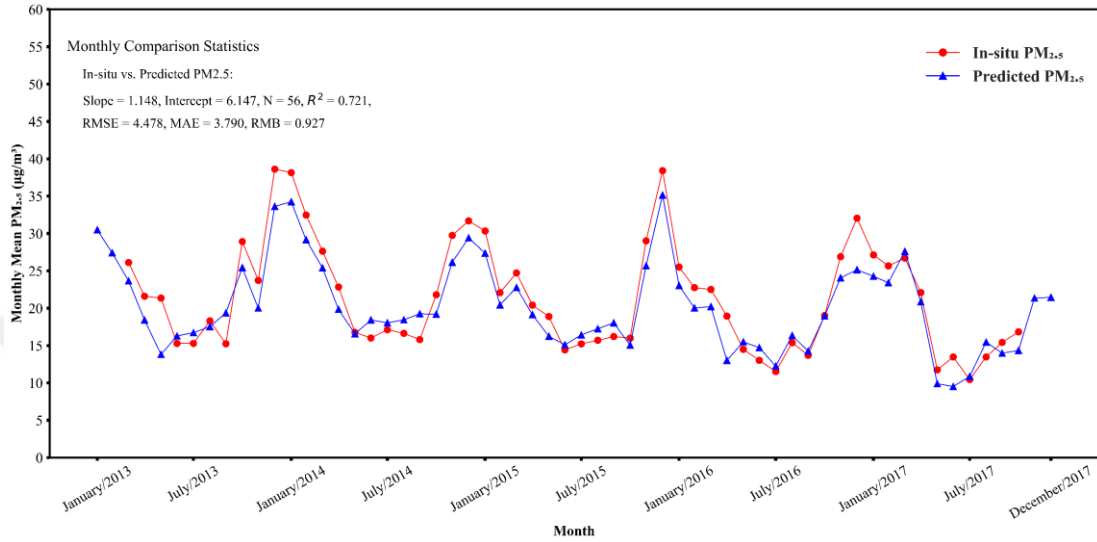


Figure 3.8: Monthly time series of the measured and predicted average daily PM2.5 over the Marmara Region during 2013-2017.

Figure 3.9 shows the month-average PM2.5 obtained for ground measurements and model-derived PM2.5 between 2013 and 2017. Since four months are missing from the dataset of the ground-measurements, month-average values of January, February, November, and December were resulted from averaging four average monthly values of PM2.5. However, five monthly values were included in averaging of the other eight months. The figure shows that the model overestimated ground measurements in the heating months (July, August, and September). However, for the other months of the year, the ground measurements overestimated the model derived PM2.5. In addition, the highest and lowest month-average values of PM2.5 are presented for December and July, respectively.

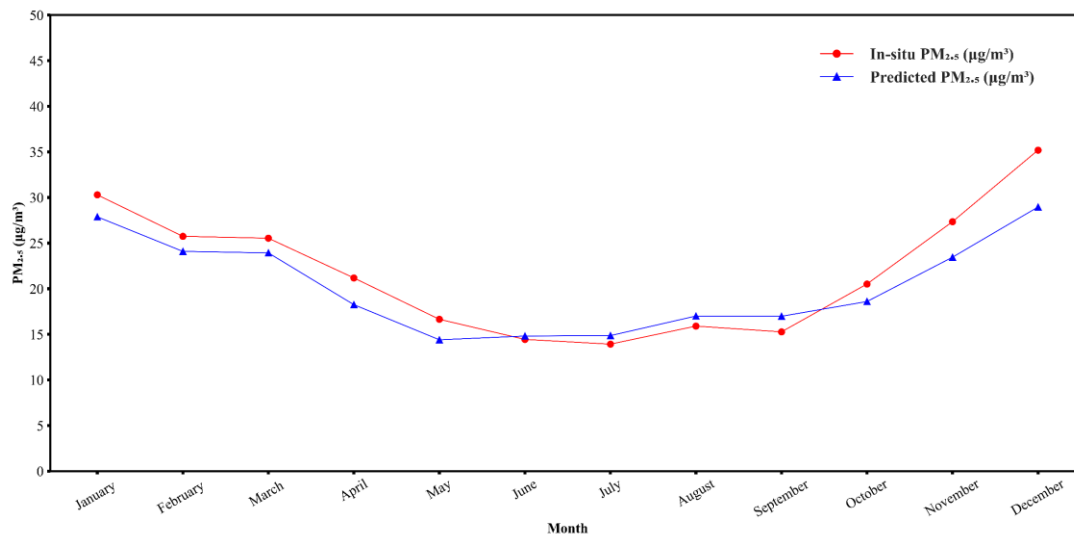


Figure 3.9: Monthly-averaged of the measured and predicted PM_{2.5} over the Marmara Region during (2013-2017).

3.1.5 Spatial distribution of PM_{2.5}

Monthly Spatial Distribution

Maps of average-monthly dry mass concentration of PM_{2.5} are presented in Figure 3.10 for the Marmara region during 2013-2017, while Figure B.1–Figure B.5 present them for every year separately. According to Figure 3.10, the highest monthly concentrations of PM_{2.5} occurred in December (29.26 µg/m³) and January (28.60 µg/m³), while the lowest occurred in June and July, 14.86 µg/m³ and 14.46 µg/m³, respectively. The subfigures (a), (b), (c), (k), and (l) of Figure 3.10 and Figures B.1–B.5 show that high PM_{2.5} concentrations are associated with Edirne, Sakarya, and Bursa cities (see Figure 2.1); yet comparable concentrations are found for the other cities. Low average monthly concentrations of PM_{2.5} for Çanakkale city are presented in all of the figures and subfigures. The maps of the months between April and September (Figure 3.10 (d), (e), (f), (g), (h), and (i)) show relatively low concentrations of monthly mean PM_{2.5}, less than 22 µg/m³. In contrast, especially in the Black Sea, Figures B.1–B.5 displayed high concentrations along the study areas' coastal lines. In addition, the scenes of January and December in the same figures included significantly missing pixels.

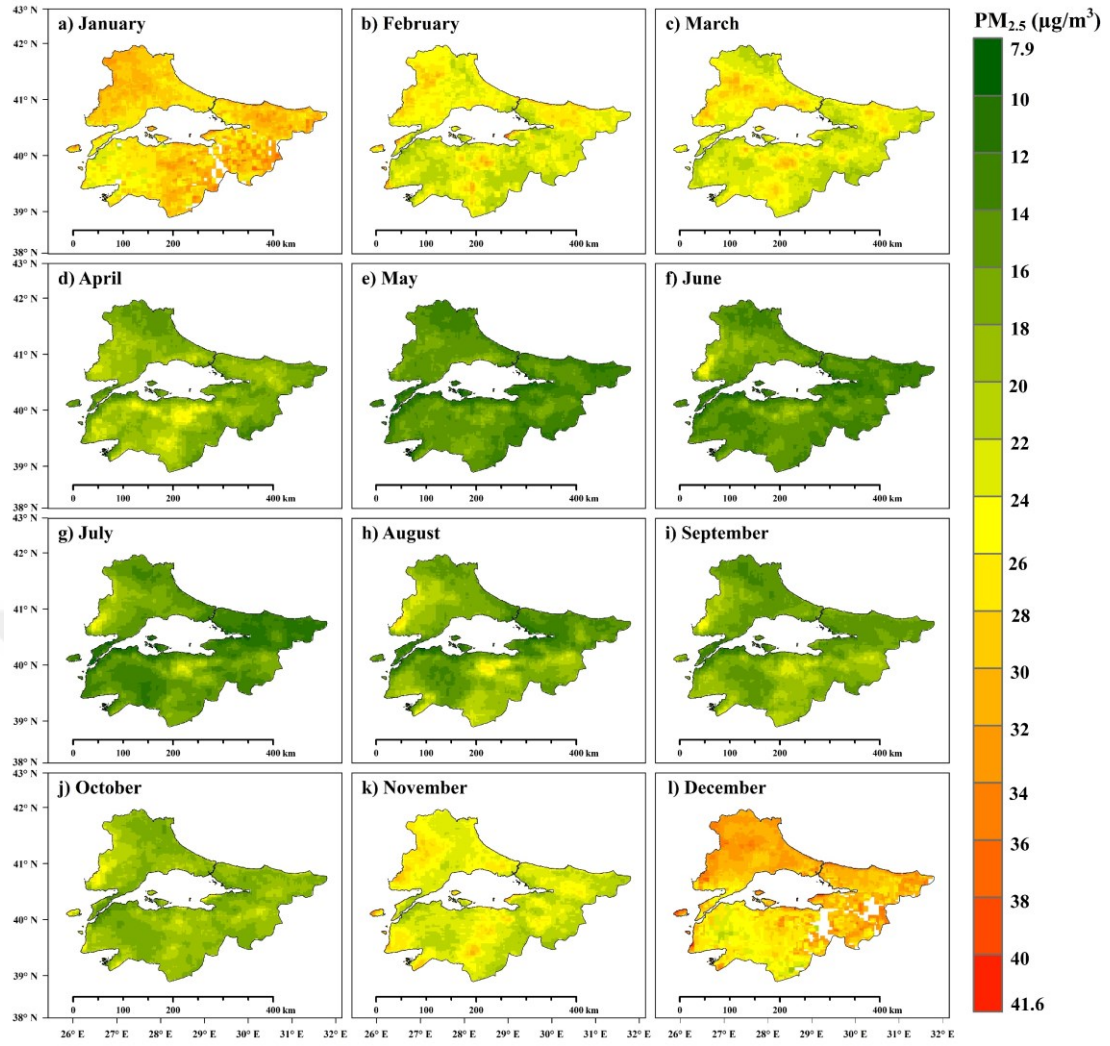


Figure 3.10: Maps of monthly average PM_{2.5} concentrations estimated for the Marmara region during 2013-2017.

Seasonal Spatial Distribution

Seasonal average PM_{2.5} concentrations maps (Figure 3.11) were produced by averaging the monthly maps with respect to the season. Figure 3.11 shows that Edirne, Bursa, and Sakarya cities are associated with high concentrations of PM_{2.5} compared to other cities of the Marmara Region; however, the concentrations were found low for Lapseki city. High PM_{2.5} concentrations of Edirne and Sakarya were presented by Zeydan & Wang (2019), while (Kahya et al., 2017) indicated the low concentrations of Lapseki in Çanakkale. Regarding other cities, Figure 3.10 and Figure 3.11 show moderate PM_{2.5} concentrations on a monthly and seasonal basis. Among seasons, (Figure 3.11 (a)) presented that winter is associated with high PM_{2.5} concentrations compared to other seasons for the entire study area. However, the summer map (Figure 3.11 (c)) presents the lowest concentrations of PM_{2.5} with respect to the season.

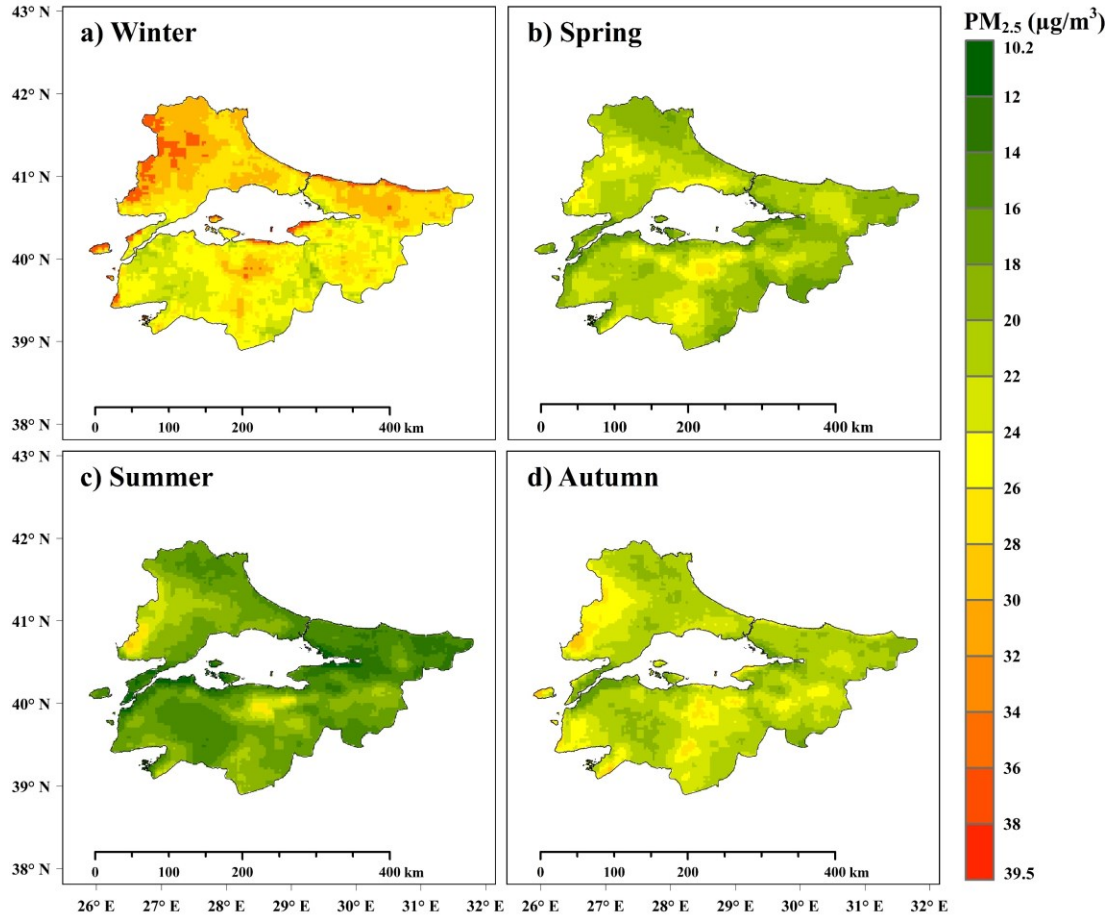


Figure 3.11: Maps of seasonal average PM_{2.5} concentrations estimated for the Marmara region during 2013-2017.

Annual Spatial Distribution

Annual average maps of PM_{2.5} for the years between 2013 and 2017 were presented in Figure 3.12. The maps show an apparent decrease in PM_{2.5} concentrations between 2013 and 2017. The annual average PM_{2.5} concentrations in 2013 and 2017 were 21.11 µg/m³ and 18.21 µg/m³, respectively; while they were 22.6 µg/m³, 20.34 µg/m³, and 18.56 µg/m³ for 2014, 2015, and 2016, respectively. Despite the decrease in PM_{2.5} concentrations shown by the annual average maps, they are consistent with the seasonal and monthly maps with respect to the cities of low and high PM_{2.5} concentrations.

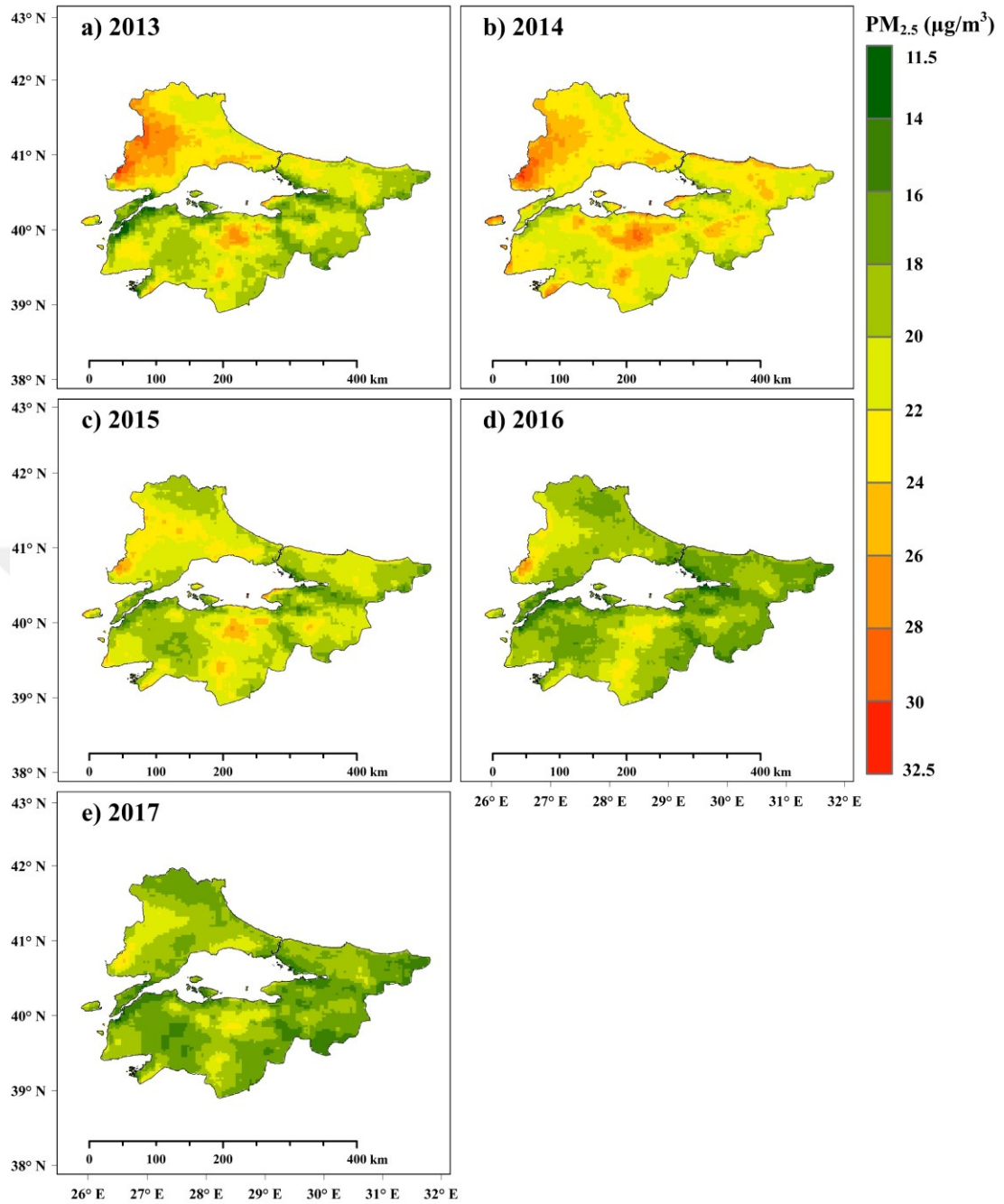


Figure 3.12: Maps of seasonal average PM_{2.5} concentrations estimated for the Marmara region during 2013-2017.

Annual average maps of PM_{2.5} for the years between 2013 and 2017 were presented in Figure 3.12. The maps show an apparent decrease in PM_{2.5} concentrations between 2013 and 2017. The annual average PM_{2.5} concentrations in 2013 and 2017 were 21.11 µg/m³ and 18.21 µg/m³, respectively; while they were 22.6 µg/m³, 20.34 µg/m³, and 18.56 µg/m³ for 2014, 2015, and 2016, respectively. Despite the decrease in PM_{2.5} concentrations shown by the annual average maps, they are consistent with the

seasonal and monthly maps with respect to the cities of low and high PM_{2.5} concentrations.

3.1.6 EU standards of PM_{2.5}

European Environment Agency (EEA) of the European Union (EU) has regulated the maximum allowed annual average of PM_{2.5} dry mass concentration as 25 µg/m³ (EU legislation; Directive 2008/50/EC). Although the EEA did not specify the average seasonal limits of PM_{2.5}, we used this limit (25 µg/m³) to study PM_{2.5} exceedance with respect to seasonal and annual averages. Exceedance maps of annual and seasonal average PM_{2.5} were presented in Figure 3.13 and Figure 3.14, respectively. Maps illustrated in Figure 3.13 show that the annual average PM_{2.5} exceeded EUs' limit for the years between 2013 and 2016. The spatial analysis for the annual exceedance maps demonstrated that the PM_{2.5} concentrations exceeded the EU's limit over 9.88% and 12.15% (6,620 km² and 8,140 km²) of the Marmara Region's area for 2013 and 2014, respectively. Most of the exceedance occurred over the cities of Edirne and Bursa. In other words, according to Table 2.1, around 8.2% of the total population (2.25 million inhabitants) are exposed to unhealthy PM_{2.5} concentrations during 2013-2014. In addition, while the exceedance occurred in Edirne and Bursa cities on a large scale, a negligible exceedance on a small-scale was shown for parts of Istanbul and Sakarya. However, the area of exceedance was found to decline for the next three years, during 2015-2017. According to Figure 3.13 (e), almost no PM_{2.5} values more than 25 µg/m³ were detected in 2017 over the study area.

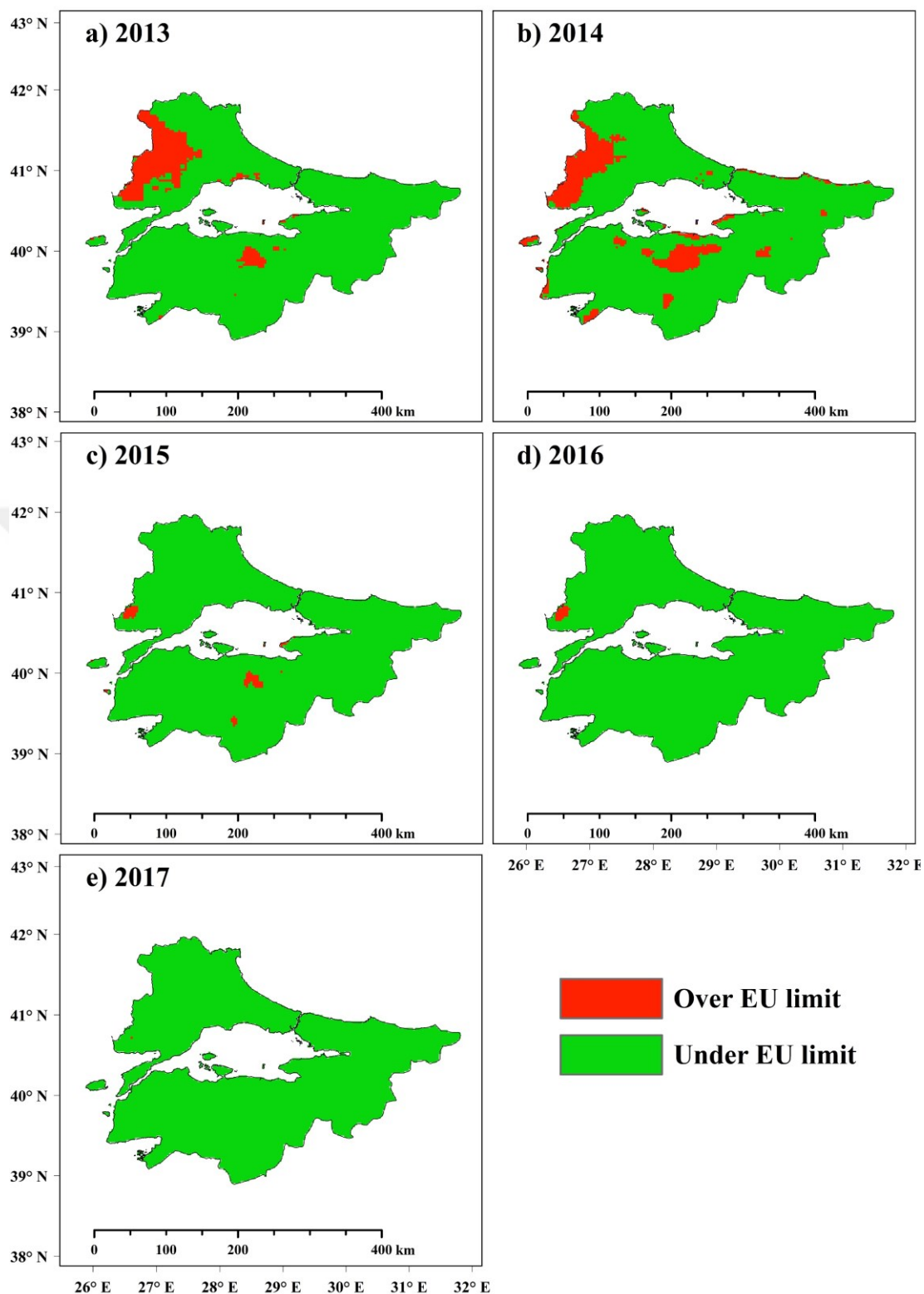


Figure 3.13: Exceedance maps of annual average PM_{2.5} concentrations to the EUs' limit of PM_{2.5}.

Figure 3.14(a) shows that the seasonal average concentrations of PM_{2.5} exceeded the EUs' standards in winter for most of the Marmara Region. The seasonal-average PM_{2.5} concentrations were found exceeding 25 µg/m³ in 73.4% (49,205 km²) of the study area in winter during 2013-2017. According to the figure mentioned above, the exceedance in winter occurred almost in all cities except Çanakkale and parts of Bursa and Yalova. Looking at the population distribution in Table 2.1, taking into account the EU's limit of PM_{2.5}, it is estimated that around 81.4% of the total population (22.38 million inhabitants) living in the Marmara region are exposed to risky PM_{2.5} concentrations in the winter season.

On the other hand, the areas exceeding the EU's limit for the spring, autumn, and summer are very small compared to the winter season. Figure 3.14(b) and Figure 3.14(d) show that the exceedance areas were found almost identical for spring and autumn. Concentrations of PM_{2.5} went beyond 25 µg/m³ over only 4.1% (2,754 km²) of the study area. In addition, the region was found to share similar coverage for each season, where more than 91% of the exceedance occurred over Bursa and Edirne cities. Based on Table 2.1, it can be estimated that around 1.05 million inhabitants (3.8% of the total population) used to breathe air with PM_{2.5} concentrations of more than 25 µg/m³ in the spring and autumn seasons during 2013-2017. For summer, the exceedance happened over 1.17% of the Marmara region over Bursa and Edirne only, while most of the study area did not witness any exceedance for the EU's limit. Finally, it can be seen from Figure 3.14 that, for Edirne and Bursa cities, the PM_{2.5} exceeded the limit of the EU in the four seasons. Information about Land Use and Land Cover LULC data for every station can be found in detail in Figure B.6.

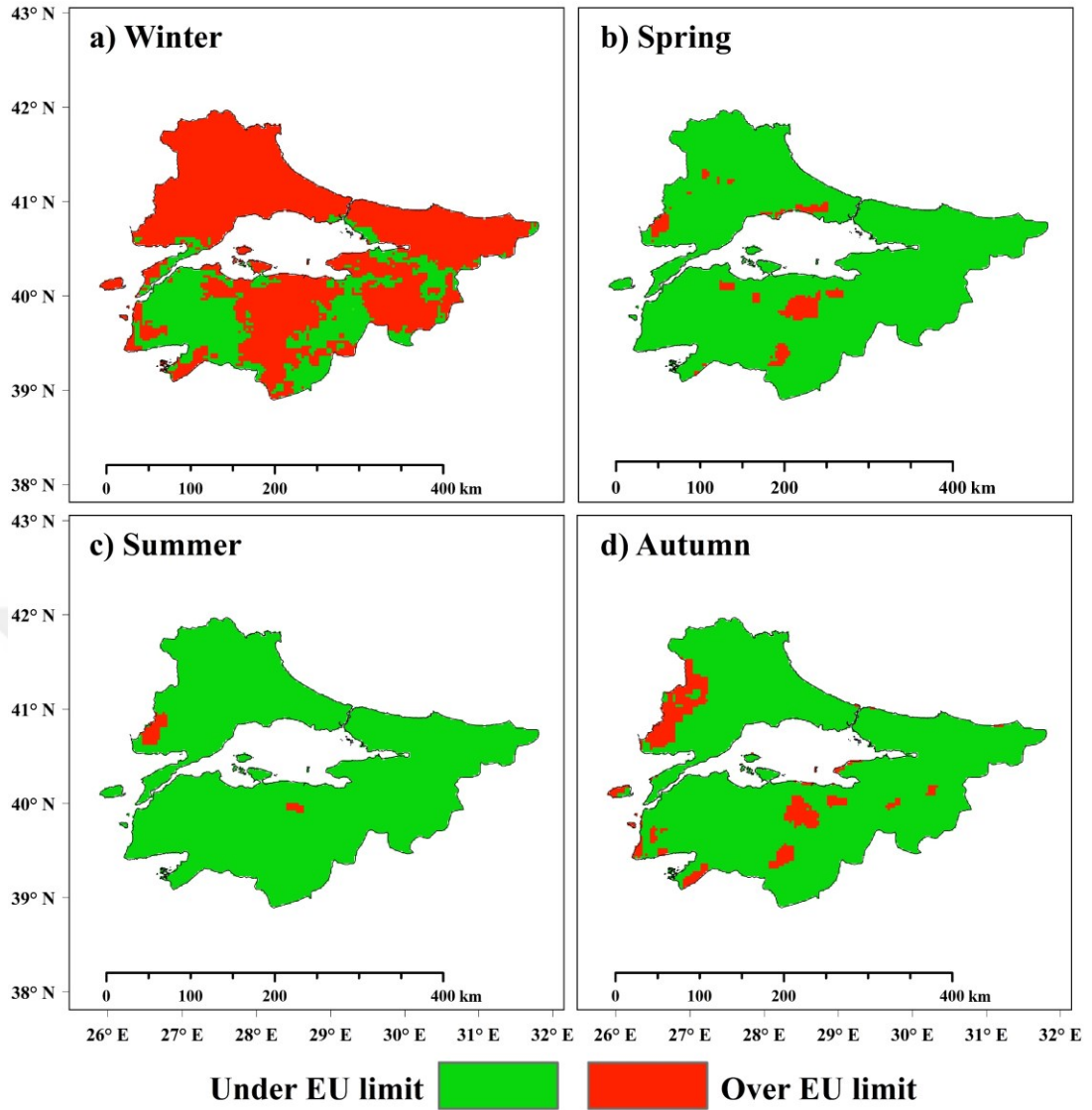


Figure 3.14: Exceedance maps of annual average PM_{2.5} concentrations to the EU's limit of PM_{2.5}.

3.1.7 Validation of PM_{2.5} estimation model

Leave-One-Out cross-validation method was conducted concerning season and year. Figure 3.15 and Figure 3.16 show results of Leave-Season-Out and Leave-Year-Out cross-validations, respectively. Figure 3.15 shows that the summer season contains the highest number of collocated daily matchups ($N = 3417$) while the lowest was for winter ($N = 965$). In contrast, bias for winter was found higher than for the summer with respect to $RMSE^{rel}$ and MAE. Both seasons were found similarly agreeing with the 1:1 line with respect to R^2 . Similar N , $RMSE^{rel}$, and MAE cross-validation results were found for both autumn and spring. However, R^2 for autumn ($R^2 = 0.58$) is found higher than for spring ($R^2 = 0.51$).

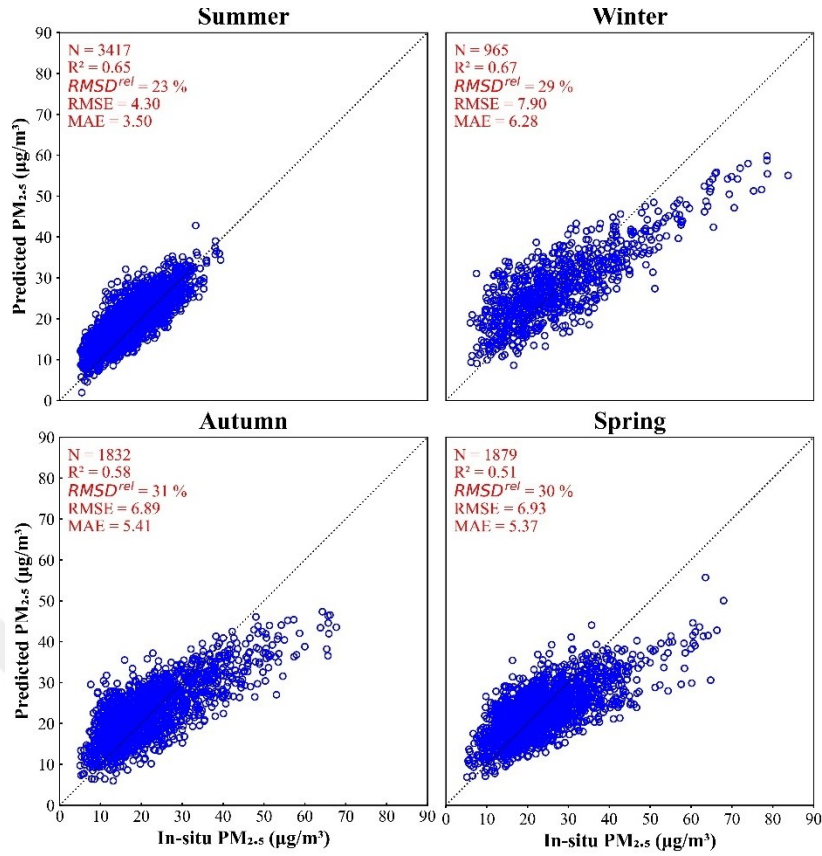


Figure 3.15: Seasonal cross-validation of the whole 12 stations over the five-year study period (2013-2017) between the models' outcomes and ground-measured PM_{2.5} values.

Statistics of Leave-Year-Out cross-validation are presented in Figure 3.16. The number of matchups for 2013 (N = 1482) and 2017 (N = 1516) are less than of the other three years. The R² ranged between 0.52 and 0.65 for 2017 and 2013, respectively. The average of RMSE^{rel} for the annual cross-validation (38.6 %) was found larger than of seasonal cross-validation (28.25%).

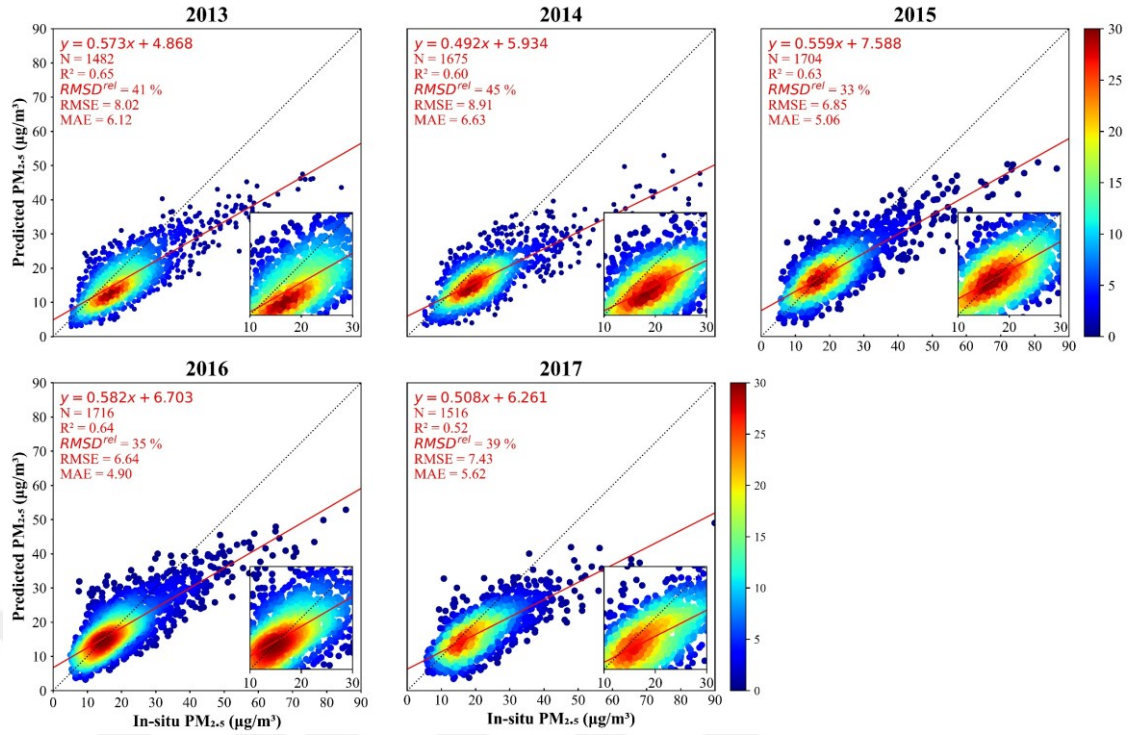


Figure 3.16: Annual cross-validation of the whole 12 stations over the five-year study period (2013-2017) between the model's outcomes and ground-measured PM2.5 values. The frequency of match points values is indicated by the color bars.

The whole matchups included in the cross-validation were combined into one plot, and the statistics were calculated in Figure 3.17. According to the latter, agreeing to the 1:1 line and bias were enhanced by combining all the daily matchups in one scatter plot. The overall R^2 increased to 0.687 while $RMSE^{rel}$ decreased to 27%. J. W. Xu et al. (2015) estimated the PM2.5 over eastern China using AOD determined from the GOCI satellite instrument. Their results presented that combining seasonal validation scatters increased R^2 from 0.49 to 0.66. The scatter plot of Figure 3.17 shows that the model underestimates the high values of PM2.5, i.e., more than 60 $\mu\text{g}/\text{m}^3$, yet overestimated low values of PM2.5 daily matchups (less than 22 $\mu\text{g}/\text{m}^3$).

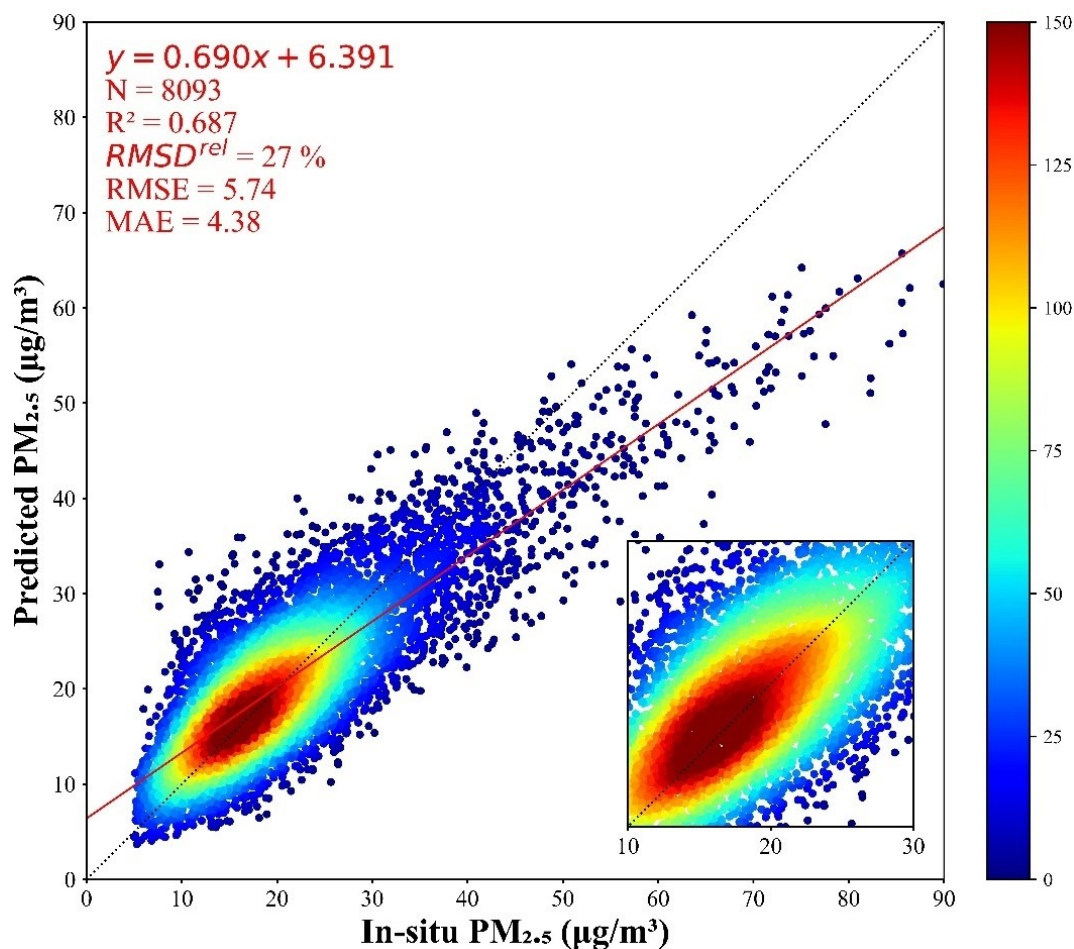


Figure 3.17: Total cross-validation of the whole 12 stations over the five-year study period (2013-2017) between the model's outcomes and ground-measured PM_{2.5} values. The frequency of match points values is indicated by the color bar.

3.2 Discussion

3.2.1 Validation of MODIS AOD

The present study evaluated AOD (550 nm)-derived from MODIS observations and MERRA-reanalysis against three AERONET ground-based stations: one is located in Turkey, while the others are in Athens, Greece, and on Cyprus, with respect to the period 2013–2017. The availability in both MODIS Aqua and Terra AOD is found to be low due to cloudy conditions and hazy days (Che et al, 2019; F. Mao et al, 2015). Even so, the statistics are higher when combining AOD from the two platforms. By considering MODIS Aqua and Terra combined, the availability in AOD is 37%, 32%, and 34% for the AERONET Athens-NOA, Mersin ERDEMLI, and Cyprus CUT-TEPAK stations, respectively. In the validation against AERONET, AOD derived by MODIS Aqua and Terra combined is associated with higher R^2 and lower $RMSD^{rel}$

compared to AODs derived from separated platforms. These findings are in line with retrievals of AOD over land and ocean by (McPhetres & Aggarwal, 2018). The authors in the latter study found that the R^2 was 0.95, 0.93, and 0.92 for the combined Terra/Aqua, Terra, and Aqua, respectively; However, the opposite occurs for retrievals of AOD over land.

In the comparison with AERONET, it was found that MODIS overestimates AOD by about 20%, both considering daily and monthly averaging. This is in line with previous studies where the overestimation of AERONET by MODIS was estimated by 10% (Sayer and Knobelspiesse, 2019; Wei et al, 2019). On the other hand, MERRA-2 underestimates AOD with 9–20% with respect to daily and monthly averaging. (H. Shi et al., 2019) found also an underestimation of MERRA-2 AOD. This might be due to nitrate pollutants that are not included in the AOD model (Randles et al, 2017). The spatial resolution of MERRA-2 AOD is substantially lower than MODIS ($62.5 \times 50 \text{ km}^2$ and $10 \times 10 \text{ km}^2$, respectively), which may contribute to the underestimation of the former platform. In addition, AODs derived from MISR (Microwave Integrated Retrieval System) are included in the MERRA-2 reanalysis, which are associated with higher uncertainties than the MODIS AODs. The Error Envelope (EE) is $\pm (0.2 \text{ AOD})$ and $\pm (0.05 + 0.15 \text{ AOD})$ for MISR and MODIS, respectively (Kahn et al, 2005).

The discrepancies that are found in the present study between satellite-retrieved and ground-based measured AOD may be due to a variety of factors such as the selection of satellite pixels and the approach to reducing surface reflectance. Increasing the number of available AODs with respect to the box with the number of satellite pixels used and collocation window size during sampling increases correlation and declines bias (Georgoulas et al, 2016). Increasing the numbers of available MODIS and AERONET AOD values for each MODIS–AERONET pair reduces bias and improves the results of the validation (Levy et al, 2010). Globally, satellite-derived AOD is usually associated with smaller bias compared to retrievals of AOD at a particular site, and the high spatial resolution of aerosol gridded data reveals aerosols features efficiently (Levy et al, 2018). Inhomogeneous land cover and surface properties cause large uncertainties in the estimations of surface reflection and thus satellite AOD (Jin et al, 2019).

AOD derived from both MODIS observations and MERRA-2 reanalysis is the highest in summer and lowest in winter over Turkey. One reason for this finding might be due

to soil moisture declining in summer associated with higher temperatures, which resuspends surface dusts (Prasad et al, 2005). The long-range transport of dust aerosols from the Middle East and North Africa probably also increases aerosol loadings over Turkey in the summer (Ali et al, 2017; F. Li & Ramanathan, 2002). For example, the same results have been found for AOD over Saudi Arabia India, and Nigeria (Ayanlade et al, 2019; Buchard et al, 2017; Sivaprasad & Babu, 2014). Table 3.1 shows that the R^2 obtained in the comparison between MODIS/MERRA-2 and AERONET AOD is correlated to atmospheric RH. These results are compatible with previous studies (Hobbs, 2002; Kent et al, 1998; Schutgens et al, 2013). However, when investigating other statistical parameters in the present study, such as slope, offset, RMSE, and RMB, a relation with RH does not occur when all three AEROENET sites are taken into account. Besides, extreme aerosol conditions such as dust events were detected more effectively by MODIS than MERRA-2. Furthermore, the accuracy of MERRA-2 AOD with respect to AERONET measurements was found to be lower than that of MODIS AOD. The latter was also found by (Jin et al., 2019). These findings may be due to the spatial resolution, which is substantially higher for MODIS compared to MERRA-2.

The scarcity of sunphotometer sites in the study area is considered a challenge when validating satellite AOD. Utilizing AOD datasets of AERONET stations located outside of the study area affects the statistical results of the MODIS AOD validation. For example, there are no ground-based AERONET sunphotometers found in the Marmara Region. Thus, AOD datasets of the nearest AERONET sites (MTEU-ERDEMLI, Athens-NOA, and CUT-TEPAK), alongside the MERRA-2 assimilated dataset, were used to validate the AOD dataset of MODIS C6.1 for the Marmara Region. Furthermore, cloud cover in the atmosphere reduces the availability of AOD pixels. For the MODIS sensor, AOD pixels are missed if clouds are in the atmospheric column. Therefore, the availability of AOD daily matchups included in the validation will witness a significant decrease. For cloudy months, especially in winter, the low availability of AOD daily matchups reduces the representativity of AOD datasets.

3.2.2 Data description

Datasets of three different sources were used to estimate PM_{2.5} over the Marmara Region concerning the period 2013–2017; (i) PM_{2.5} ground measurement obtained

from 12 ground-stations of the NAQMS/UHİKA local network, (ii) AOD 550nm obtained from combining MODIS C6.1 Terra and Aqua datasets, and (iii) ECMWF ERA-5 datasets of six meteorological variables (BLH, RH, T, WS, TP, and SLP). Although temporal resolution for the PM_{2.5} ground measurements is very high (30 minutes), their spatial coverage is discontinuous. Contrarily, meteorology datasets of ERA-5 are spatially and temporally continual as well as satisfy a global coverage. However, the spatial resolution of ERA-5 is relatively low (30 x 30 km²) compared to the MODIS C6.1 AOD (10 x 10 km²). The latter provides two scenes of AOD 550 per day, which is considered high temporal resolution comparing to other sensors.

Raster scenes of MODIS AOD are significantly affected by clouds and hazes. Pixel values of MODIS AOD are missed if clouds are found in the atmosphere (Aldabash et al, 2020). Availability of AOD pixels in every scene for the Marmara Region ranged between 38% and 88% in January and August, respectively. Depending on the cloud cover in the atmosphere, MODIS AOD pixels availability is different among geographical zones. (Chi et al, 2020) presented that the availability of AOD obtained from MODIS Aqua was 12% over China during 2018-2019. Contrarily, over the Asian continent, the availability of inland AOD obtained from DB/DT of the MODIS C6.1 was found 27% and 52% for January and August, respectively (Gupta et al, 2020). Therefore, the availability of MODIS AOD for the present study area is overall acceptable to retrieve the concentrations of PM_{2.5}. Compared to other regions such as China (Zhang et al., 2019; Zhong et al., 2017), a low daily average of AOD (AOD = 0.22) was found for 2013-2017. AOD is increased significantly by dust episodes which are infrequent in Turkey. Small desert landcover and large green areas in Turkey mitigate soil resuspension and dust production. In Turkey, the vast majority of dust is originated from surrounding desert areas, especially Egypt, Saud Arabia, and Libya (Shaheen et al., 2020).

Ground-stations of measuring PM_{2.5} are distributed evenly in seven cities over 72,000 km² in the Marmara region. Every station's measurements are affected by the surrounding anthropogenic activities and meteorological characteristics (Cheng & Li, 2010; Jianhua Wang & Ogawa, 2015). Along the five years study period, the highest daily mean of PM_{2.5} was presented for the datasets of Ozanlar (28.34 µg/m³), Kağıthane (25.31 µg/m³), and Ümraniye (27.84 µg/m³). Ozanlar and Ümraniye are located on the eastern side of the Marmara Region. Both stations might be affected by

the emissions of the surrounding industrial zones and vehicular emissions out of the highway. Besides, the Ozanlar station is located in Sakarya city, which is affected by the transported dust from the middle east and north African desert (TMMOB, 2019).

Concerning the Kağıthane, it is one of the most crowded counties of the Marmara Region, where 448,025 inhabitants live on 14.52 km² (URL-3). This is increasing the likelihood of high rates of anthropogenic emissions. In addition, the station is located in a relatively low-elevation area (32 m AMSL). Low elevations could preserve low wind speeds and prevent pollutants' dispersion (Cheng & Li, 2010). This was supported by a previous study (Pu et al, 2011). Besides, the range of datasets (Max-Min) is long for the three of the stations. This might be due to extreme events of anthropogenic aerosols in the study area. These results are consistent with (Zeydan & Wang, 2019). Contrarily, stations of Lapseki and Karaağaç are located in open coastal regions and demographically non-crowded areas. This resulted in low mean values of PM2.5 as well as short ranges of their data.

Trends of the meteorological variables were found different along the course of the year in the Marmara Region. The temperature reaches its maximum in summer, while winter was found cold and humid. Therefore, a large bias ($RMSE^{rel} = 48.40\%$) was found in the temperature dataset. Contrarily, $RMSE^{rel}$ was found low (14.11%) for the dataset of RH. Humidity is somewhat high (mean RH $\approx 70\%$) and does not vary significantly among the seasons. However, the lowest variation among the datasets of meteorological variables was found in the SLP. The latter is affected by elevation differences as well as humidity (Turgut & Usanmaz, 2016). Except for the Uludağ PM2.5 measuring station, the elevation is not varying significantly in the study area; for nine of the stations, it ranged between 8 and 48 AMSL only (Table 2.3).

3.2.3 PM2.5 estimation model

The present study estimated PM2.5 dry mass concentrations for the Marmara Region with respect to the period 2013-2017, using MODIS-derived AOD (550 nm), meteorological variables obtained from the ECMWF ERA-5, and PM2.5 measurements of the UHKİA/NAQMS ground stations. Linear correlation between AOD and PM2.5 is found to be weak due to multiple reasons. Satellite AOD is a columnar measurement that presents the integration of all aerosol's light extinction, while PM2.5 is measured on ground-level elevation (Ying Li et al, 2018). Besides,

satellites' sensors acquire AOD in ambient conditions ($RH > 40\%$). Microphysical properties of aerosol and solar light scattering are affected by RH due to hygroscopic growth by water uptake (Burgos et al, 2019; Zieger et al, 2011). However, ground measurements of PM_{2.5} are obtained in dry conditions where RH is less than 40% (X. Wang et al, 2019). These findings are in line with the estimation of PM_{2.5} by (Kumar et al, 2007; Shao et al, 2017). Therefore, it is crucial to convert dry aerosol properties into equivalent amounts at ambient RH (Yu et al, 2018; Zang et al, 2019).

PM_{2.5}-AOD correlation is improved by converting satellite-derived AOD and ground-measured PM_{2.5}. By applying the non-linear $f(RH)$ and the BLH corrections on the PM_{2.5}-AOD correlation, it was found that the R^2 increased from 0.152 to 0.393. The non-linear correction, based on extinction coefficient $f(RH)$, is applied to convert PM_{2.5} dry-mass measurements into the ambient state (Lianfa Li et al, 2017; Nabavi et al, 2019). In addition, the relationship between AOD and PM_{2.5} is potentially improved by the appropriate conversion of the column-integrated MODIS AOD to the ground-level aerosol extinction coefficient using BLH (J. Li et al, 2015; Lianfa Li et al, 2017). (Zang et al, 2019) stated that multiplying dry mass PM_{2.5} by $f(RH)$ ($AOD \sim PM_{2.5dry} \times f(RH)$) improved R^2 from 0.56 to 0.65. Besides, these results are consistent also with retrieving PM_{2.5} over the inlands by (Chew et al, 2016; Hoff & Christopher, 2009; Nabavi et al, 2019). In the latter study over Tehran, R^2 of the PM_{2.5}-AOD relationship reached 0.72 (Nabavi et al, 2019).

The relationship between satellite AOD and ground-measured PM_{2.5} is affected by the meteorological conditions. Adding the meteorological variables (RH, BLH, SLP, T, TP, and WS) somewhat improved the model of PM_{2.5} estimation; the overall R^2 increased from 0.393 to 0.492. The relative humidity was found positively correlated, and the strongest meteorological predictor increased R^2 of the correlation between PM_{2.5} and AOD. This might be due to the increasing hygroscopic volume of aerosols by water uptake of the atmospheric RH (J. Chen et al, 2014; Zieger et al, 2010). In addition, the refractive index and particle size of aerosols are changed via hygroscopic growth caused by RH (J. Chen et al, 2014; Covert et al, 1972). Therefore, excluding matchups of extremely humid days ($RH > 90\%$) contributed by 9.07% to the overall R^2 (Table 3.3). However, this stage caused a 4.4% reduction in data availability. These results are in line with the PM_{2.5} prediction over North America by (Shinozuka et al,

2007). They found that the PM_{2.5} is robustly correlated with AOD ($R^2 = 0.77$) after removing 4% of the data with RH>90%.

In the comparison between meteorological variables, BLH was found strongly negatively correlated ($\beta < 0$) with PM_{2.5}. As the bulk of aerosols stay volatile within the BLH, it represents the area available for dispersion and dilution of PM_{2.5} in the atmosphere (Su et al, 2018; C. Wang et al, 2019). Vertical convection and turbulence force within the BLH satisfy particulate pollutants' transport and mixing (Rost et al, 2009). Therefore, high turbulence and convection in high BLH dilute and reduce concentrations of PM_{2.5} (Petäjä et al, 2016). In addition, radiative forcing and cooling effects of particulate pollutants, including PM_{2.5}, can stifle the development of BLH (Qu et al, 2017). This negative relationship between BLH and PM_{2.5} is consistent with previous studies by (Luan et al, 2018; Miao et al, 2019; Su et al, 2018).

In the same context, the meteorological variable of temperature was found significant to the model and correlated negatively with the PM_{2.5}. This might be due to vertical convection caused by temperature (Lu et al, 2017). The air is elevated vertically by the effects of heating, diffusing PM_{2.5} particles, and reducing their concentrations. Also, burning fossil fuels for heating in low-temperature seasons, especially in the rural areas, increases emissions of PM_{2.5} in the atmosphere. These findings are in line with investigating the effects of temperature on PM_{2.5} concentrations by (Q. Ma et al, 2017; C. Zhang et al, 2015). However, some previous studies presented a positive impact of high temperature on increasing PM_{2.5} concentrations due to secondary-induced aerosols formation and soil dust resuspension (Jun Li et al, 2013b; Kegler et al, 2001; Vecchi et al, 2004).

The variable of resultant wind speed is found inversely strongly significant in the model of PM_{2.5}. The significance P of the WS was less than 0.001 for 11 out of 12 stations, and the β was -3.88. Wind disperses atmospheric particulate matter and dilutes PM_{2.5} concentrations in areas where wind speeds are low Pu et al. (2011); Jianhua Wang & Ogawa (2015), e.g., the Marmara Region (mean WS = 3.3 ms⁻¹). Contrarily, high wind speeds transport particulate pollutants on a regional and continental basis, thus increases PM_{2.5} concentrations. Aldabash et al. (2020) ; Shaheen et al. (2020) stated somewhat that Turkey is affected by the long-range transport of mineral dust from the Arabic and African Sahara. However, mineral dust contributes to increasing the PM₁₀ concentrations rather than PM_{2.5} (Al-Taani et al,

2019; Vasilatou et al, 2017). These results are consistent with analyzing PM_{2.5} and PM₁₀ concentrations over Istanbul by (Çapraz & Deniz, 2021).

Contrarily to the temperature, wind speed, and BLH, the total precipitation and sea level pressure variables were found weakly significant to the model. The P-value was found significant for only six and four stations for the SLP and TP, respectively. Precipitation is not continuous in the present study area. The dataset of the TP contains more than 40% of the no-rainfall days. Besides, elevation AMSL is not varying significantly in the Marmara Region. Therefore, the SLP dataset did not include an enormous variation to be detected by the model.

Filtering the number of available AODs collected by the pixel window(at least five pixels) increased the R^2 from 0.493 to 0.528 in the fifth stage of the model. Correlation of the PM_{2.5}–AOD relationship is improved by increasing the number of available satellite AODs included in the collection window (30x30 km²) (Georgoulas et al, 2016). This enhances the representativity of the averaged daily MODIS AODs to the atmospheric AOD. However, this stage caused a 2.2% decrease in the available collocated PM_{2.5}–AOD daily matchups. Increasing the available area of AODs from 25x25 km² to 50x50 km² increased the correlation coefficient R from 0.78 to 0.83 (Georgoulas et al, 2016). These findings are in line with previous studies by (Chi et al, 2020; Tong et al, 2020). The availability of AOD pixels in latter two study decreased by 7% (R^2 increased from 0.61 to 0.72) and 9.4% (R^2 increased from 0.58 to 0.96) in the studies conducted by Chi et al. (2020) and Tong et al. (2020), respectively.

Removing outliers in the sixth and seventh stages has contributed to 14.98% o the overall R^2 of the model. Average daily values of PM_{2.5} less than five µg/m³ were considered outliers. Since small PM_{2.5} concentrations are interactive values, it is hard to be estimated by the model. Nabavi et al. (2019) found an increase in the model accuracy when 3% of the daily matchups were excluded; PM_{2.5} values lower than the first quartile (Q1)). Since aerosol microphysical properties are affected by the RH, RH's extreme daily values (RH>90%) were also excluded from the model. Although this stage reduced 4.4% of the available PM_{2.5}–AOD collocated points, the R^2 increased to 0.602. Shinozuka et al. (2007) found a high PM_{2.5}–AOD correlation after removing 4% of the data, including RH>90%. This result is compatible with (Glantz et al, 2009; Nabavi et al, 2019).

Adding temporal predictors significantly increased the model's accuracy. The concentrations of PM_{2.5} variability are time-dependent. Therefore, factorize the predicted values based on the month and season helped the model to detect temporal variations of PM_{2.5} better. The concentrations of PM_{2.5} in the winter season are driven significantly by anthropogenic emissions; however, climatology affects the PM_{2.5} concentrations in summer. This leads to over-and underestimating PM_{2.5} in winter and summer, respectively (Figure 3.8 and 3.9). Temporal predictors helped effectively in constraining the estimations and reducing the bias of the model. Temporal predictors were widely used to estimate PM_{2.5} using satellite AOD (Hu et al, 2017; Schneider et al, 2020; Stafoggia et al, 2020).

3.2.4 Spatiotemporal variability of PM_{2.5}

In the time-series analysis of PM_{2.5}, it was found that model-predicted PM_{2.5} underestimates ground-measured PM_{2.5} in the winter times. Both considering daily and monthly averaging, RMB is 0.863 and 0.927, respectively. PM_{2.5} particles are induced by anthropogenic activities, particularly heating, in the Marmara Regions (HEAL, 2021). This underestimation is due to anthropogenic emissions, e.g., BC, OC, SO₄, and NO₂, which were not included in the PM_{2.5} estimation model. Moreover, the modeled PM_{2.5} concentrations cover the whole study area, while the ground measuring stations cover only 12 sites. The spatial resolution of the ECMWF ERA-5 is substantially lower than MODIS AOD C6.1 (30x30 km² and 10x10 km², respectively), which may contribute to the underestimation of the predicted PM_{2.5}. This is in line with previous studies by (Maciejewska, 2020; Parde et al, 2020). The authors found that the PM_{2.5} is underestimated when increasing the pixel size of MODIS AOD data.

Maps of monthly- and seasonal-average and time-series analysis agreed that the PM_{2.5} derived from both the estimation model and ground measuring stations is the highest in winter and the lowest in summer over the Marmara Region. This finding may be due to a variety of factors related to anthropogenic activities and meteorological conditions. A reason for this is due to burning coal and fossil fuels for heating in the winter season, especially in the rural areas of the region (HEAL, 2020; TMMOB, 2019). Moreover, low temperature in the winter season reduces mechanical vertical convection and contribute to pollutants accumulation near to ground surface (Q. Ma et al, 2017; C. Zhang et al, 2015). Boundary layer height is also lowered in winter, which

reduces vertical mixing of particles and increases the concentrations of PM_{2.5}. This finding is consistent with the Spatiotemporal analysis of PM_{2.5} over the Marmara Region conducted by (Kahya et al, 2017; Zeydan & Wang, 2019). Kahya et al. (2017) only analysed the PM_{2.5} ground measurements over the Marmara Region. However, their study did not include a PM_{2.5} maps for the region. In addition, Zeydan & Wang, (2019) mapped the PM_{2.5} over the study area in the heating seasons. Whilst the other three seasons, especially winter where the area witnesses high PM_{2.5} episodes, were not included in their study.

High concentrations of PM_{2.5} were found along the Marmara Sea and Black Sea's coastal lines in the winter season. This is due to the high AODs along the coast caused by sea salt scattering (SSA) aerosols. Radiative forcing of SSS aerosols is larger than anthropogenic aerosols radiative forcing (Vinoj & Satheesh, 2004). Therefore, high values of AODs are detected by the MODIS sensor along the shorelines of the present study area. High breaking waves activity in winter stimulates higher SSS aerosol production (Clarke et al., 2003). Thus, the model predicted high concentrations of PM_{2.5} along shorelines. These findings are supported by (Tao et al, 2014).

Furthermore, for the cities of Edirne, Bursa, and Kocaeli, concentrations of PM_{2.5} were found higher than in other cities of the Marmara Region. Concerning Edirne, especially in winter, particulate pollutants are sourced from domestic heating, mining works, and highway traffic (Gül & Yorulmaz, 2019). Networks of natural gas are not widely established in many of Edirne's areas, especially the rural zone such as Keşan and Çerkezköy. Particularly in winter, people in Keşan and Çerkezköy burn low-quality coal and fossil fuels intensively for domestic heating purposes. The annual consumption of locally-produced and imported coal in Edirne is more than 35,000 tons/year (THEP, 2020). Lignite coal which includes a high percentage of sulfur is burned intensively in Keşan and Çerkezköy (Özdemir & Boyraz, 2002). Besides, the international and inter-cities D550 highway is passing from Edirne, increasing diesel emission of PM_{2.5}. Because of the anthropogenic emissions, Keşan is the most air polluted area of Turkey (Özşahin et al, 2016).

Despite being the most elevated Marmara Regions' area (155 m AMSL), PM_{2.5} concentrations were found seriously high in Bursa city. In the comparison of monthly and seasonal maps, it was found that PM_{2.5} concentration is high along the course of the year in a part of Bursa city, particularly over the county of İnegöl. According to the

2020 air quality report by Right of Clean Air Platform, “Bursa is the third air polluted city in Turkey with respect to particulate pollutants (PM₁₀ and PM_{2.5}), and 68% of its population breathe harmful air” (THHP, 2020). This is due to the anthropogenic emissions from the industrial facilities located in Bursa and burning fossil fuels and low-quality lignite coal, especially in İnegöl county (PALA, 2019). The industrial zone emissions in İnegöl, especially the automobile industrial zone, and burning plastic wastes contributed immensely to the PM_{2.5} pollution in Bursa (BBB, 2017).

Estimated concentrations of PM_{2.5} were found high in the Kocaeli city in the north-east of the Marmara Region. Anthropogenic emissions of PM_{2.5} in Kocaeli are divided into static and anthropogenic sources. The static sources are factories emissions, residential areas, and mining activities. However, traffic load and highways related emissions are considered dynamic sources (KÇŞİM, 2016). Vehicular emissions of PM_{2.5} are significant in Kocaeli. The city contains the largest automobile industrial zone in Turkey. In addition, with only 1.7 million inhabitants, more than one million vehicles are registered in Kocaeli (Yılmaz et al, 2020).

Annual-mean maps of PM_{2.5} concentrations presented an apparent negative trend of PM_{2.5} between 2013 and 2017. This might be due to increasing population consciousness and establishing governmental policies for air quality control. Natural gas subscriptions in rural areas of Bursa and Edirne, i.e., Keşan, Karaağaç, and İnegöl, were facilitated, and governmental loans were provided. Besides, fines were assigned toward illegal commerce of low-quality coal, especially in Edirne. Since 2013, more than 60 air quality monitoring stations were fixed in the Marmara Region; According to Edirne city's Environment and Urbanization Administration, Edirne's air quality became harmonious with the EU legalizations of air quality since 01.01.2019 (THEP, 2020).

3.2.5 EU standards of PM_{2.5}

As a country seeking to join the European Union (EU), Turkey is trying to fulfill air quality standards specified by the EU. European Environment Agency (EEA), the EUs' organization concerned by the air quality standards, has established a maximum annual-average limit of PM_{2.5} in the “Directive 2005/50/EC” as 25 µg/m³ (European Commission, 2005). We have generalized the latter value on both annual- and seasonal-average of PM_{2.5}. It was found that the exceedance for the EUs' limit

occurred apparently in the Edirne and Bursa with respect to the period 2013-2016. This is due to the excessive emitting of heating- and industrial-induced anthropogenic aerosols in these cities, especially in winter (section 3.2.4). However, the exceedance area declined by approaching 2017. In 2017, the annual-average PM_{2.5} over the whole Marmara Region was almost lower than the EUs' limit (25 $\mu\text{g}/\text{m}^3$). This might indicate the effectiveness of the established regulations aimed to control air quality in the Marmara Region.

Despite the small annual-average exceedance found, an extensive exceedance coverage area was found in the seasonal-average exceedance maps. In winter, more than 70% of the PM_{2.5} concentrations were found higher than the EUs' limit of PM_{2.5}. However, all the PM_{2.5} concentrations of the area are almost less than the EUs' limit in summer. These findings indicate how burning coal and fossil fuel in winter increases the PM_{2.5} concentrations in the region. According to the average-seasonal results, the emissions of PM_{2.5} in the Marmara Region are concentrated in the winter season. However, emissions in the rest three seasons are very low. Besides, the PM_{2.5} concentrations are lower than the EUs' limit for more than 90% of the Marmara Region. Furthermore, this may be due to the weak vertical convection related to low temperatures in winter that contribute immensely to accumulate PM_{2.5} over the region (section 3.2.3).

3.2.6 Validation of PM_{2.5} estimation model

The present model estimated the concentrations of PM_{2.5} for the Marmara Region during 2013-2017 using MODIS-derived AOD and ground-measured PM_{2.5}. Seasonal and annual variations were found in ground-level PM_{2.5}. Therefore, estimated concentrations of PM_{2.5} were validated against ground-measured PM_{2.5} using Leave-One-Out Cross-Validation (CV) with respect to the seasons and years included in the study period (J. W. Xu et al, 2015). The availability in daily matchups for the CV in winter was found lower than in other seasons. This is due to the cloud conditions and hazy days that reduce MODIS AOD pixels' availability in winter (Che et al, 2019; F. Mao et al, 2015). Also, the bias for the CV in winter was found more considerable than any other season. The humidity-induced hygroscopic growth in winter is substantially higher than in other seasons, which changes the aerosol microphysical properties (AOD) by water uptake (Burgos et al, 2019; Zieger et al,

2011). These results are compatible with the previous studies by (Chang, 2019; Kuang et al, 2016).

On the other hand, the bias values of $RMSE^{rel}$, MAE, and RMSE of the Leave-Year-Out CV were found apparently larger than the ones in the Leave-Season-Out CV. One reason for this finding might be due to similarity in the daily matchups' microphysical properties in the one season. However, different meteorological and microphysical conditions affect aerosols over one year (Molero et al, 2020; Sreekanth et al, 2019). Since the months of (Jan/Feb 2013) and (Nov/Dec 2017) are missing from the PM_{2.5} ground measurements dataset, their CVs' numbers of collocated points (N) are less than of the other three years CV. However, it was found that the agreement to the 1:1 line is similar for the period 2013–2016, while it is lower for 2017. Combining all the daily matchups into one scatterplot improved the agreement to the 1:1 line and decreased bias. However, high values of PM_{2.5} were found underestimated in the overall CV. Finally, the values of R^2 and $RMSE^{rel}$ indicated the model robustness to estimate the ground-level PM_{2.5} over the Marmara Region.

4. CONCLUSIONS

4.1 Conclusions

The number of available daily AODs obtained from a single platform increased from 30% to 34%, over the three AERONET sites, by combining MODIS Aqua and Terra outcomes. For MODIS, the availability of days included in the seasonal averaging ranged between 51% and 58% for winter and summer, respectively. In contrast, with respect to month, the values ranged between 92% for August and 44% for January. It was found in the comparison between MODIS and AERONET that the highest R^2 and lowest bias ($\text{RMSD}^{\text{rel}}/\text{MAE}$) are in the MODIS/MERRA-2 combined datasets. MODIS AOD is more consistent with AERONET than MERRA-2, which can be interpreted from the higher R^2 and lower bias values. The three of the platforms detected an apparent annual variation in AOD. In comparison with AERONET, MODIS AOD overestimates, and MERRA-2 underestimates AERONET. In addition, fewer days with extreme events (24%) were detected by MERRA-2 than by MODIS (55%). Furthermore, based on the statistical results, there is an apparent relationship between relative humidity and the accuracy of MODIS and MERRA-2 AOD.

Although the Marmara Region is a medium-range study area with nonsignificant variation in elevation, there is high variability in measured PM_{2.5} among the ground-stations. Ground-stations measurements of PM_{2.5} were found high and low in winter and summer, respectively. The five-years (2013-2017) average of the ground-measured PM_{2.5} is 21.18 $\mu\text{g}/\text{m}^3$, below the EUs' annual limit of PM_{2.5} (25 $\mu\text{g}/\text{m}^3$). Besides, the meteorological variables of SLP and TP tend to be weakly variable in the study area. However, apparent seasonal variations were found in the meteorology of T, RH, and BLH. The latter meteorological variables are more significant (less P-value) to the PM_{2.5} estimation model than the SLP and TP.

Despite strong linear relationships between PM_{2.5} and satellite AOD were found for other study areas, extremely low R^2 was found in the PM_{2.5}-AOD linear correlation for the Marmara Region. Hygroscopic growth by RH and vertical distribution of

aerosols lower the R^2 of the PM_{2.5}–AOD linear correlation. R^2 of the PM_{2.5}–AOD relationship increases significantly after the corrections of PM_{2.5} and MODIS AOD using the nonlinear extinction coefficient $f(RH)$ and BLH, respectively. Adding the meteorological conditions as a mixed additive model improves the consistency between the corrected PM_{2.5} and MODIS AOD, which means that the meteorology has a significant effect on the PM_{2.5} variability in the present study area. Excluding outliers represented by the extreme episodes of RH and small values of PM_{2.5} enhanced the model predictability, which means higher R^2 and lower RMSE^{rel}/MAE. Furthermore, the increasing number of collected AOD pixels in the spatial window enhances the model statistics (R^2 and RMSE) due to the increasing representativity of collected daily MODIS AODs.

Meteorological variables datasets of the ECMWF ERA-5 are temporally and spatially continual. However, comparing to MODIS C6.1 AOD scenes, low spatial resolution (30x30 km²) of the ERA-5 meteorological products contributed to models' underestimation of high PM_{2.5} values. MODIS AOD dataset involved more than 55% of missing pixels over the present study area. Availability of AOD pixels ranged between 33% and 66% for winter and summer, respectively. Low MODIS daily AODs in winter reduced representativity of AOD; thus, contributed to an underestimation of PM_{2.5}. Locally emitted anthropogenic emissions increase the concentrations of PM_{2.5} in the Marmara Region. There is a clear seasonal and annual variation in PM_{2.5} over the study area. Seasonal model validation in winter is associated with the highest bias (RMSE^{rel}/MAE) because of the RH hygroscopic effects. However, the bias of the annual validation was found higher than in the seasonal validation.

The absence of anthropogenic variables from the model as well as the relatively low spatial resolution of ERA-5, made the model underestimate ground measured PM_{2.5}, especially in winter. Burning coal and fossil fuels for heating in the Marmara Region elevates PM_{2.5} concentrations in the aforementioned season. Intensive burning of low-quality fuels and industrial activities increases the concentrations of PM_{2.5} apparently in the cities of Edirne, Bursa, and Kocaeli. In most of the present study area, the concentrations of PM_{2.5} are seriously exceeding the EUs' limit in winter. However, for the other three seasons, seasonal-average PM_{2.5} is not exceeding EUs' limit for more than 90% of the Marmara Region. Although the study period covers only five years, average-annual concentrations of PM_{2.5} exhibited a negative trend,

both in the model-estimated and ground-measured PM_{2.5}. Finally, MODIS C6.1 AOD and ERA-5 meteorology were effectively utilized to estimate PM_{2.5} dry mass concentrations over the Marmara Region.

4.2 Recommendations

The research estimated dry-mass concentrations of PM_{2.5} on ground-level for the Marmara Region using MODIS C6.1 AOD and meteorological datasets of ECMWF ERA-5. During conducting the research stages, the researcher identified a variety of areas for interests, and recommendations are summarized as follows:

1. Datasets of MODIS AOD include a high percentage of missing pixels due to cloud cover, especially in winter. More research is needed to fill the gaps of AOD pixels to increase AODs' availability.
2. Particulate pollution, especially PM_{2.5}, derives severe health disorders for humans as well as environmental problems. Therefore, more researches are needed to quantify and monitor PM_{2.5} pollutants for Turkey.
3. Concentrations of PM_{2.5} are in a complicated nonlinear relationship with meteorological conditions and anthropogenic activities. Machine learning approaches are needed to be utilized to clarify how PM_{2.5} is affected by every predictor.
4. Variation of land use and land cover of every geographic zone changes the spatiotemporal variability of PM_{2.5}. Thus, enlarging the networks of ground-measuring stations can provide accurate quantitative data about PM_{2.5} pollution.
5. The UHKİA/NAQMS ground measuring stations of PM_{2.5} was established for the Marmara Region in 2013. However, no available ground measurements of PM_{2.5} before 2013. Based on historical land use and meteorological data, methods to assimilate historical datasets of PM_{2.5} concentrations for Turkey are needed to be conducted.
6. Aerosols' microphysical properties, i.e., hygroscopic growth, size distribution, and extinction coefficient, are different among climatic zones. We do recommend studying the physical and chemical characteristics of atmospheric aerosols in multiple climatic zones in Turkey.

7. The field of atmospheric aerosols in Turkey is in deficit need of more research to improve our understanding of the atmosphere. Therefore, it is recommended to establish multi-national research teams concerned with the studies of atmospheric aerosols.
8. Mapping and analyzing atmospheric aerosols involves a wide range of uncertainties. Uncertainty in the present study was found in the stages of MODIS AOD validation, modeling PM_{2.5}, and mapping the dry mass concentrations. Uncertainties due to hygroscopic effects of humidity, surface complexity, and columnar parameters should be quantified.



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APPENDICES

Appendix A: Tables

Appendix B: Figures



Appendix A

Table A.1: Daily mean AOD for values considered as extreme aerosol events ($AOD \geq 0.5$). The numbers 0 and 1 reveal as the extreme event was detected by the MODIS and MERRA-2 or not, respectively.

Station (I) ATHENS-NOA				
AERONET	MODIS	MERRA-2	MODIS C6.1	MERRA-2
0.722	0.841	0.552	1	1
0.621	0.632	0.228	1	0
1.210	1.171	1.107	1	1
0.810	0.707	0.167	1	0
0.575	0.591	0.422	1	0
0.694	0.833	0.549	1	1
0.532	0.676	0.264	0	0
0.607	0.592	0.614	1	1
Number of extreme events with respect to AERONET (in %)			88	50
Station (II) METU-ERDEMLI				
0.527	0.295	0.255	0	0
0.672	0.703	0.271	1	0
0.860	0.891	0.372	1	0
0.657	0.354	0.292	0	0
0.658	0.707	0.456	1	0
0.681	0.201	0.712	0	1
0.501	0.447	0.331	0	0
0.515	0.443	0.338	0	0
0.525	0.616	0.307	1	0
0.604	0.317	0.211	0	0
0.644	0.439	0.252	0	0
0.531	0.282	0.170	0	0
0.521	0.445	0.131	0	0
0.530	0.336	0.282	0	0
0.510	0.177	0.161	0	0
0.536	0.324	0.312	0	0
0.507	0.495	0.245	0	0
0.514	0.508	0.315	1	0
0.567	0.388	0.280	0	0
0.527	0.295	0.255	0	0
Number of extreme events with respect to AERONET (in %)			25	10
Station (III) CUT-TEPAK				
0.613	0.607	0.292	1	0
0.684	0.687	0.506	1	0
0.512	0.521	0.416	1	1
0.977	0.979	0.721	1	0
0.768	0.700	0.936	1	0
0.642	0.692	0.505	1	1
0.529	0.400	0.280	0	0
Number of extreme events with respect to AERONET (in %)			86	29

Table A.2: Descriptive statistics of the used parameters according to the season for the Marmara Region during 2013–2017. Parameters were daily averaged as in Table 3.2.

Parameter	Mean	Median	Min. ^a	Max. ^b	STD ^c	RMSE ^{rel d}	Confidence Interval	
							5%	95%
Winter								
PM2.5 ($\mu\text{g}/\text{m}^3$)	28.765	27.290	6.544	68.079	13.194	45.87	1.407	0.045
AOD 550	0.160	0.132	0.013	1.277	0.116	72.50	0.015	0.000
BLH (m)	470.742	405.434	23.209	1381.518	317.614	67.47	33.860	1.083
RH (%)	75.331	75.648	44.086	93.049	8.928	11.85	0.952	0.030
SLP (mbar)	1.016	1.016	0.995	1.037	0.007	0.69	0.00073	2.33E-05
T ($^{\circ}\text{C}$)	5.944	6.038	-4.421	17.351	4.029	67.78	0.430	0.014
TP(mm)	0.476	0.083	0.000	5.942	0.944	198.32	0.101	0.003
WS (ms^{-1})	3.665	3.342	0.859	12.534	1.858	50.70	0.198	0.006
Spring								
PM2.5 ($\mu\text{g}/\text{m}^3$)	21.528	20.370	8.880	54.526	7.821	36.33	0.719	0.023
AOD 550	0.251	0.216	0.042	1.396	0.162	64.54	0.016	0.001
BLH (m)	412.309	379.327	20.682	1482.508	268.395	65.10	24.661	0.789
RH (%)	68.055	68.060	43.020	95.000	10.523	15.46	0.967	0.031
SLP (mbar)	1.020	1.021	0.995	1.039	0.007	0.69	0.00065	2.08E-05
T ($^{\circ}\text{C}$)	13.364	13.349	3.307	24.592	4.516	33.79	0.415	0.013
TP(mm)	0.445	0.108	0.000	4.983	0.718	161.35	0.066	0.002
WS (ms^{-1})	2.839	2.683	0.568	8.180	1.369	48.22	0.126	0.004
Summer								
PM2.5 ($\mu\text{g}/\text{m}^3$)	15.543	14.978	6.069	32.932	4.383	28.20	0.405	0.013
AOD 550	0.252	0.234	0.047	0.696	0.114	45.24	0.011	0.000
BLH (m)	497.358	510.447	32.347	1319.747	254.672	51.20	23.530	0.753
RH (%)	66.899	67.328	43.866	89.325	7.708	11.52	0.712	0.023
SLP (mbar)	1.015	1.014	0.999	1.033	0.006	0.59	0.00053	1.69E-05
T ($^{\circ}\text{C}$)	23.715	24.168	16.500	28.571	2.334	9.84	0.216	0.007
TP(mm)	0.338	0.092	0.000	3.742	0.585	173.08	0.054	0.002
WS (ms^{-1})	3.330	3.347	0.276	7.072	1.439	43.21	0.133	0.004
Autumn								
PM2.5 ($\mu\text{g}/\text{m}^3$)	20.769	17.317	6.924	70.027	10.613	51.10	0.967	0.031
AOD 550	0.194	0.167	0.023	0.733	0.110	56.70	0.011	0.000
BLH (m)	647.372	664.604	82.486	1530.021	269.869	41.69	24.582	0.786
RH (%)	71.323	71.710	47.708	95.731	10.017	14.04	0.912	0.029
SLP (mbar)	1.013	1.013	1.000	1.031	0.004	0.39	0.00041	1.30E-05
T ($^{\circ}\text{C}$)	15.826	15.740	2.337	27.115	5.313	33.57	0.484	0.015
TP(mm)	0.198	0.008	0.000	3.083	0.435	219.70	0.040	0.001
WS (ms^{-1})	3.169	2.961	0.612	9.342	1.516	47.84	0.138	0.004

Table A.3: Summary of the utilized datasets in the study.

Variable	Unit	Temporal Resolution	Spatial Resolution	Source
AOD550 nm	Unitless	1 day	10 km	MODIS C6.1
AOD550 nm	Unitless	1 hour	50 x 62.5 km	MERRA-2
AOD440 nm	Unitless	15 minutes	Point Data N/A	AERONET
AE440-675	Unitless	15 minutes	Point Data N/A	AERONET
PM2.5	$\mu\text{g}/\text{m}^3$	1 hour	Point Data N/A	UHKIA
RH	%	1 hour	30 x 30 km	ECMWF ERA-5
BLH	meter	1 hour	30 x 30 km	ECMWF ERA-5
SLP	mbar	1 hour	30 x 30 km	ECMWF ERA-5
T	C°	1 hour	30 x 30 km	ECMWF ERA-5
TP	mm	1 hour	30 x 30 km	ECMWF ERA-5
VWS	ms^{-1}	1 hour	30 x 30 km	ECMWF ERA-5
HWS	ms^{-1}	1 hour	30 x 30 km	ECMWF ERA-5

Appendix B

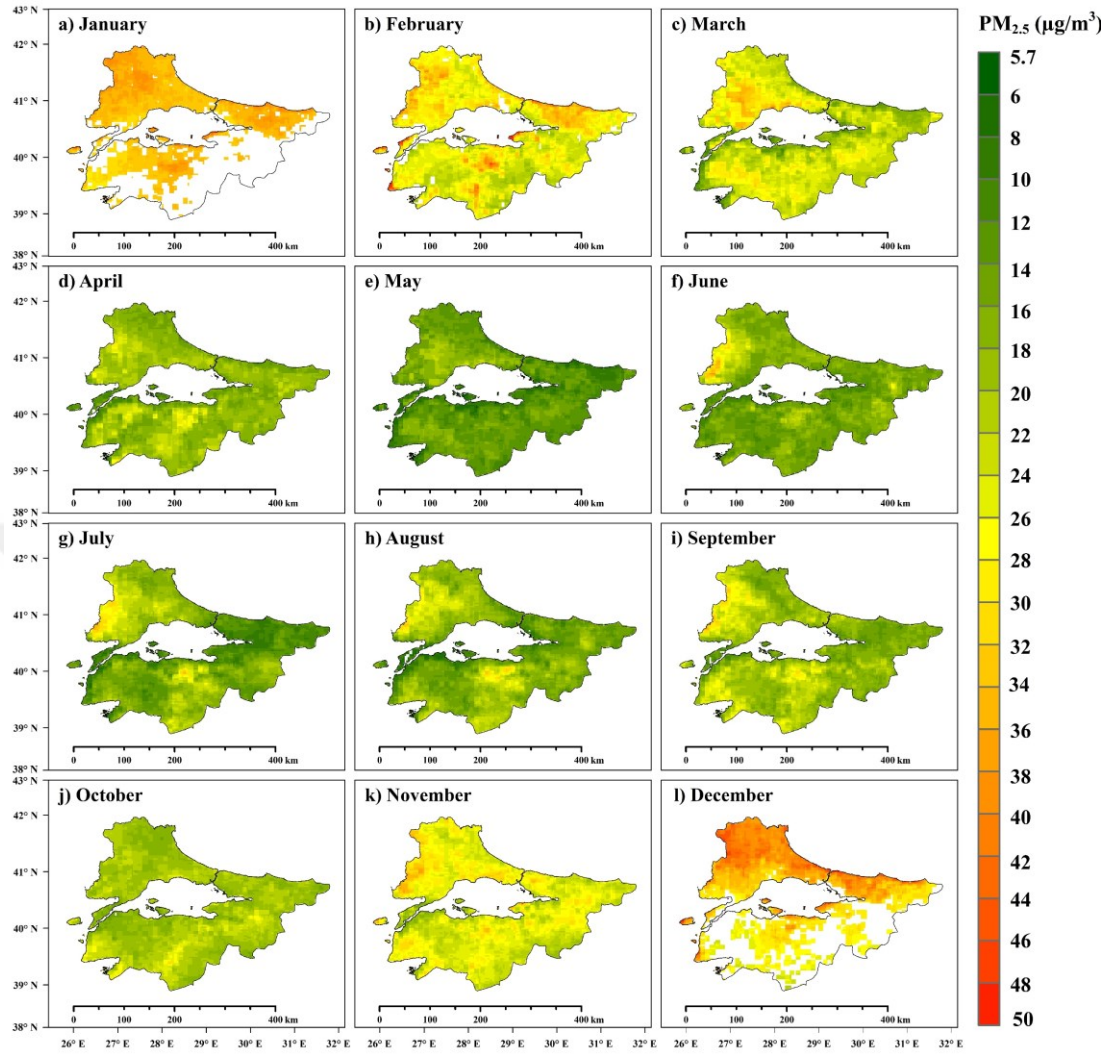


Figure B.1: Maps of estimated monthly average PM_{2.5} concentrations for the Marmara region during 2013.

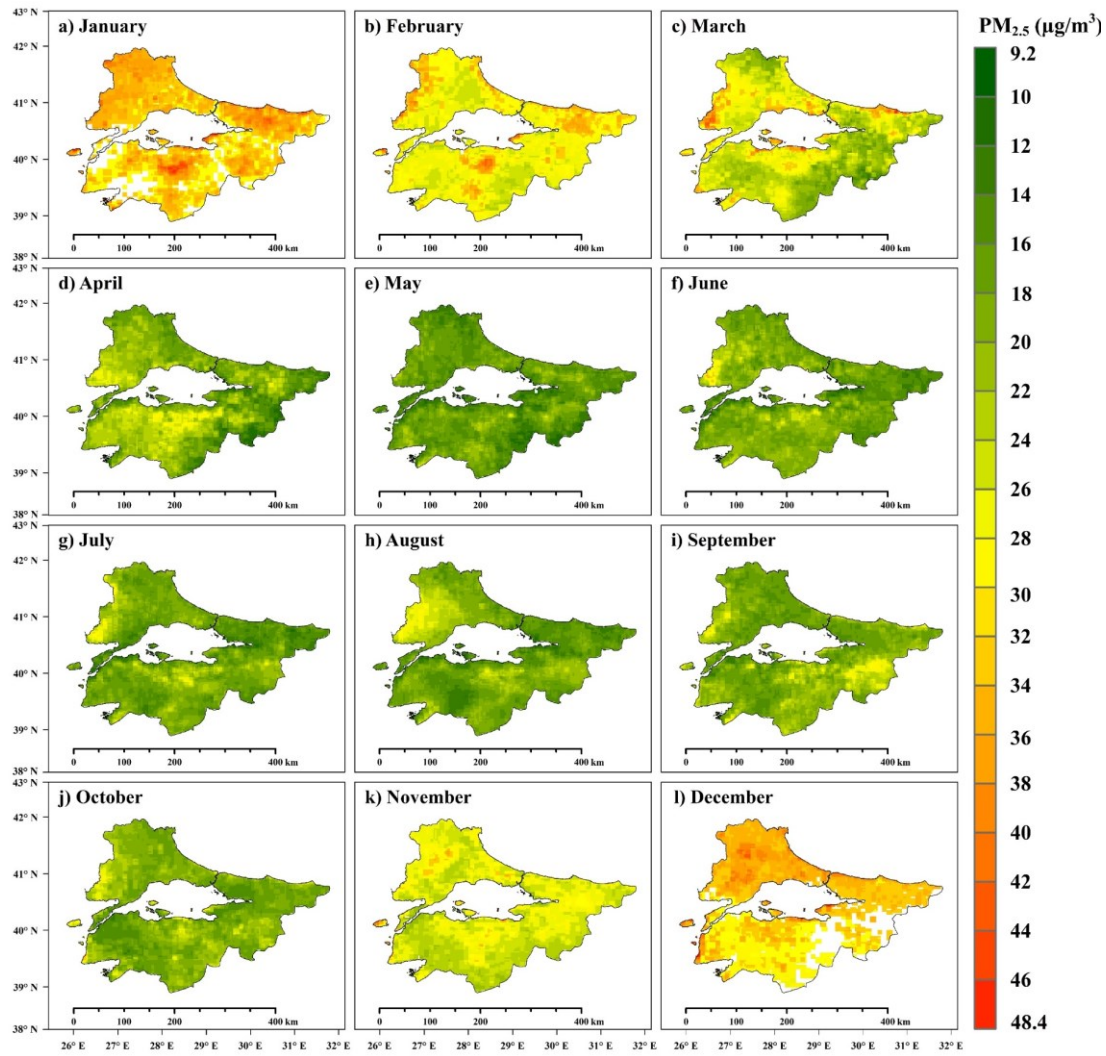


Figure B.2: Maps of estimated monthly average PM_{2.5} concentrations for the Marmara region during 2014.

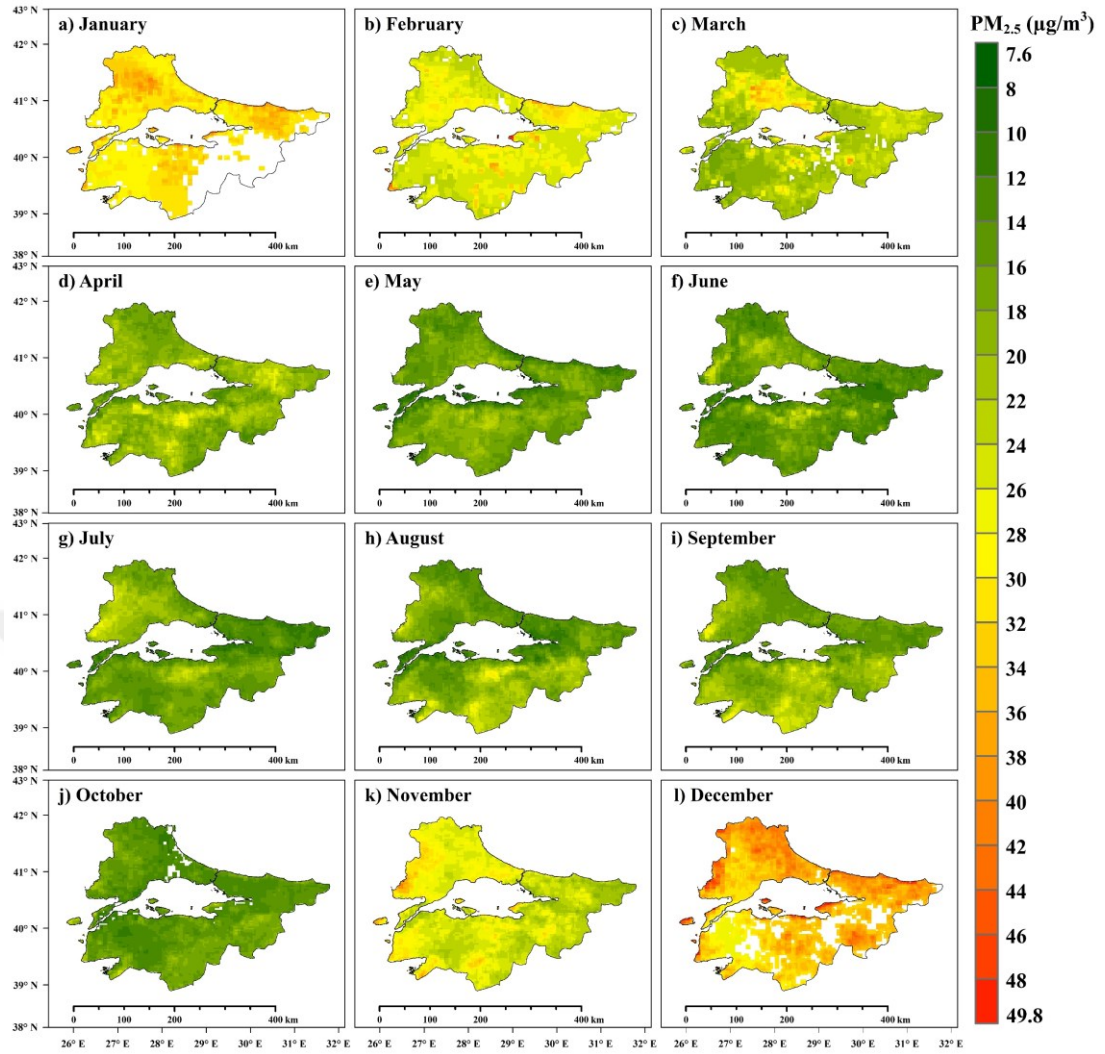


Figure B.3: Maps of estimated monthly average PM_{2.5} concentrations for the Marmara region during 2015.

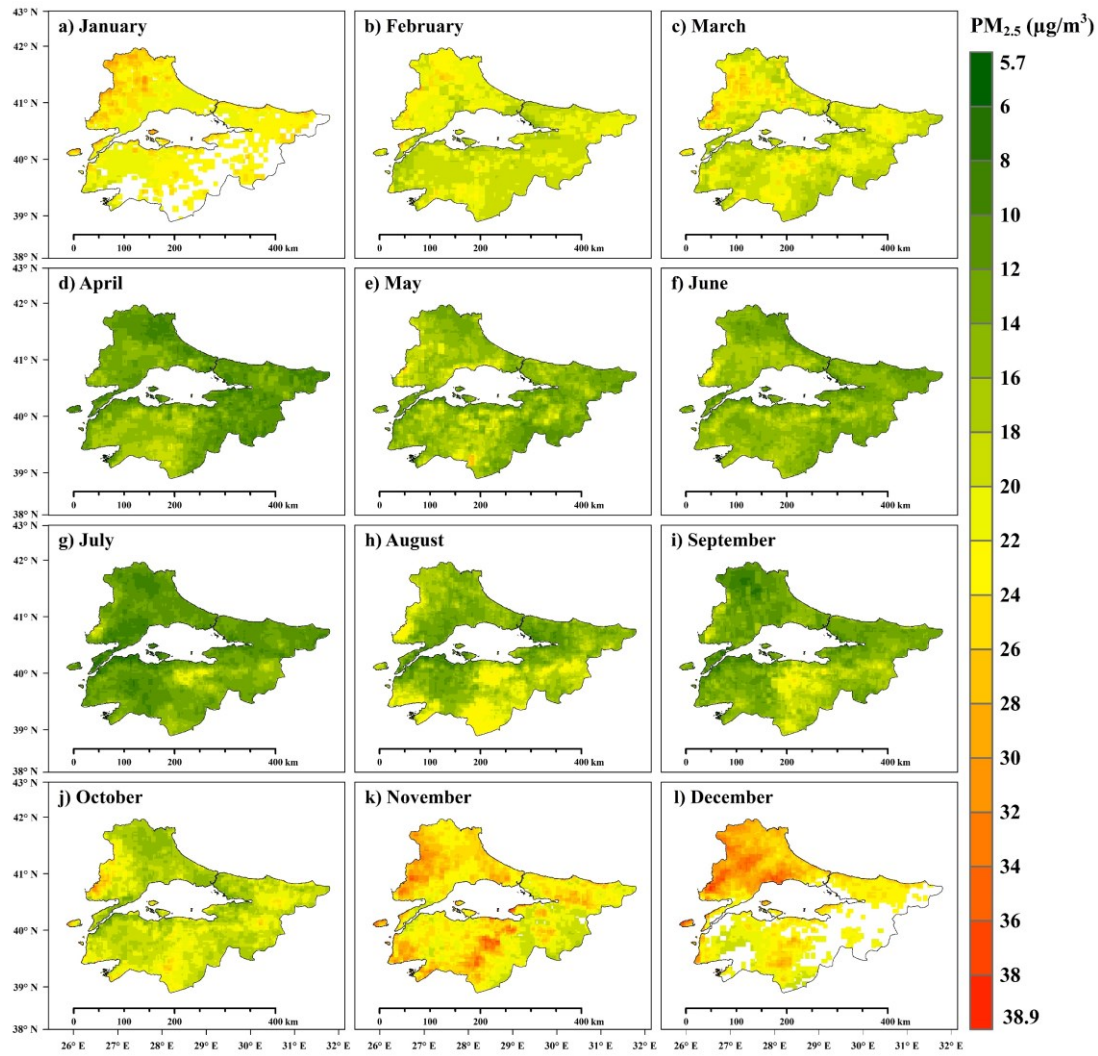


Figure B.4: Maps of estimated monthly average PM_{2.5} concentrations for the Marmara region during 2016.

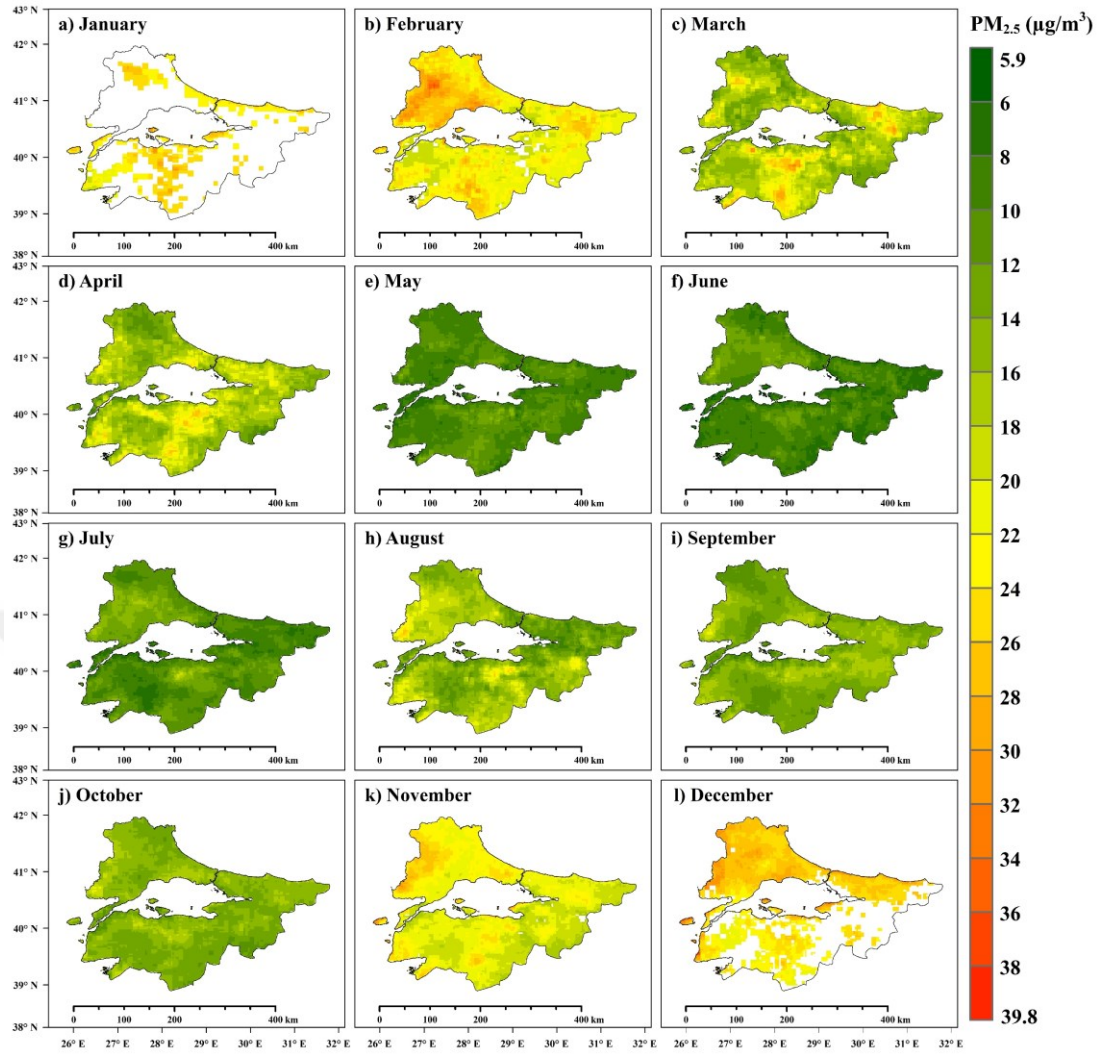


Figure B.5: Maps of estimated monthly average PM_{2.5} concentrations for the Marmara region during 2017.

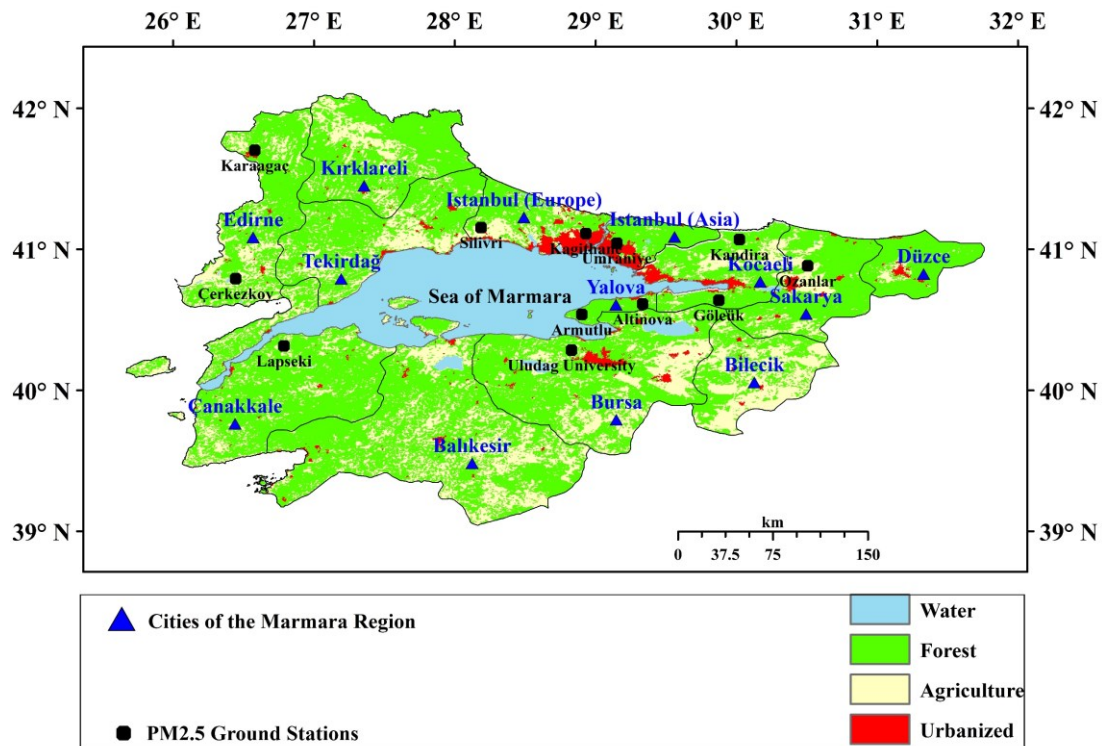


Figure B.6: Locations of the PM_{2.5} ground stations and cities of the Marmara Region with respect to the LULC of the Marmara Region. LULC data were obtained from (URL-10).

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