Supplementary Information

Table S1. PM$_{2.5}$ aerosol concentrations (in µg/m^3) at the boundaries of the domain.

Species	Boundaries[1]			
	North	South	West	East
OA[2]	1	1	1	1
Sulfate	1	1	1	1
Ammonium	0.37	0.37	0.37	0.37
Nitrate	0.06	0.015	0.007	0.06
Sodium	0.001	0.003	0.05	0.018
Chloride	0.001	0.006	0.06	0.009

[1] "North", "South", "West", and "East" here represent the top, bottom, left, and right edge, respectively, of our modeling domain.
[2] organic aerosol

Table S2. Summary of the evaluation of MODIS vs. AERONET AODs at 550nm for land, per satellite and Quality Confidence Flag (QAC).

Satellite	QAC	Collocated Points	Regression equation[1]	Correlation Coefficient	% points in expected error envelope
Aqua and Terra	123	8,331	y = 1.046 x - 0.0001	0.81	72.7
	23	7,226	y = 1.027 x - 0.003	0.82	74.0
	3	5,544	y = 0.976 x - 0.004	0.83	75.5
Aqua	123	3,721	y = 1.057 x + 0.002	0.79	71.1
Terra	123	4,592	y = 1.039 x - 0.002	0.82	73.9

[1]x is AERONET AOD and y is MODIS AOD

Table S3. Expected error values which describe MODIS AOD uncertainties (land and ocean algorithms) with respect to AERONET. The monthly mean value of AERONET AOD is employed for $AOD_{AERONET}$.

Region	$\pm(0.05+0.15AOD_{AERONET})$ Land	$\pm(0.03+0.05AOD_{AERONET})$ Ocean
UK and Ireland	±0.09	-
Central Europe	±0.07	-
North Europe	±0.06	-
Spain and Portugal	±0.07	-
East Europe	±0.07	-
Balkans	±0.08	-
Russia, Belarus, and Ukraine	±0.08	-
Turkey and Nothern Africa	±0.08	-
Mediterranean Sea	-	±0.04
North Atlantic Ocean	-	±0.04
South Atlantic Ocean	-	±0.04
Black Sea	-	±0.04
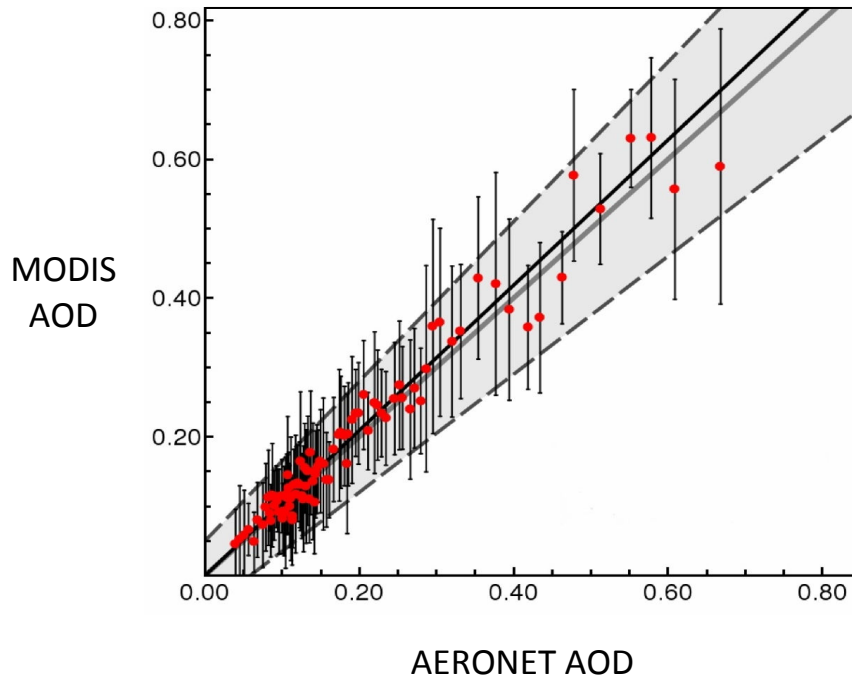
Fig S1 Comparison of MODIS AODs over land at 550 nm from Aqua and Terra (QAC $\geq$ 1) with AERONET AODs. The mean AERONET and MODIS values for each bin are denoted by the red dots. The collocated MODIS data standard deviation, for each bin, is illustrated by the vertical error bars. The solid black line is the regression equation y=-0.0001+1.046x calculated prior to data partitioning, along with the 1:1 solid gray line. The dashed gray lines denote the land expected error envelope of $\pm(0.05 + 0.15\text{AOD}_{\text{AERONET}})$.
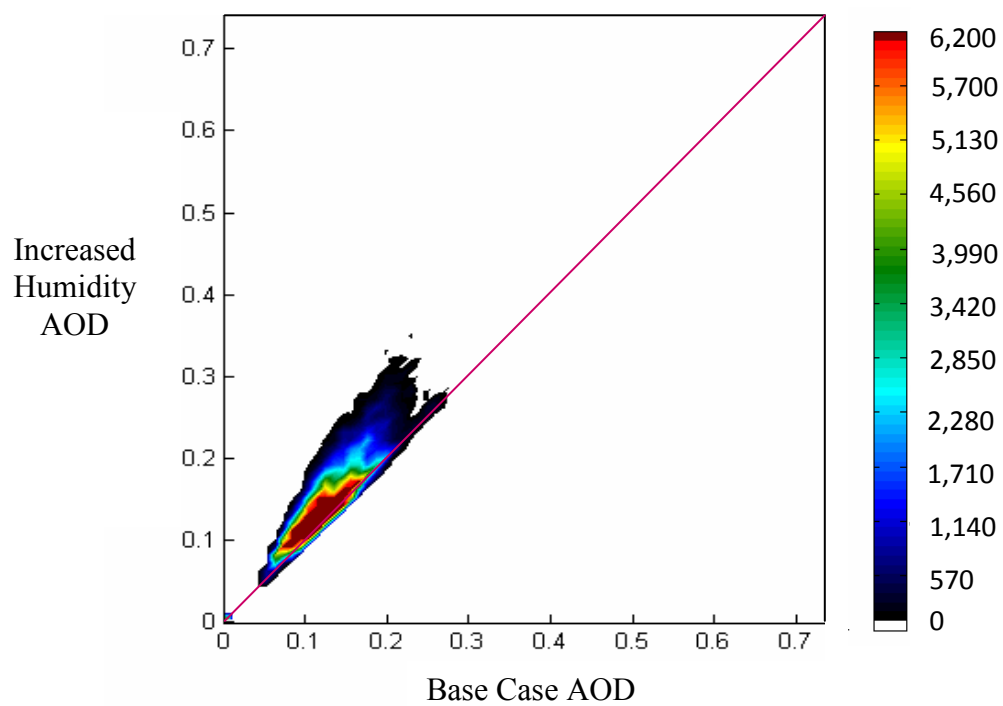
Fig S2 Comparison of the base case PMCAMx with the humidity test PMCAMx monthly mean AODs. The different colors indicate density.