

1 **Effect of water vapour on the determination of Aerosol Direct Radiative Effect based on the**
2 **AERONET fluxes**

3 **J. Huttunen^{1,2}, A. Arola¹, G. Myhre³, A.V. Lindfors¹, T. Mielonen¹, S. Mikkonen², J. S. Schafer⁴, S.**
4 **N. Tripathi⁵, M. Wild⁶, M. Komppula¹ and K. E. J. Lehtinen^{1,2}**

5 [1]{Finnish Meteorological Institute (FMI), Kuopio Unit, Finland.}

6 [2]{Department of Applied Physics, University of Eastern Finland, Kuopio, Finland.}

7 [3]{Center for International Climate and Environmental Research - Oslo, Norway.}

8 [4]{NASA/Goddard Space Flight Center (GSFC), Biospheric Sciences Branch, Greenbelt, MD, USA.}

9 [5]{Department of Civil Engineering, Indian Institute of Technology, Kanpur, India.}

10 [6]{Institute for Atmospheric and Climate Science, ETH Zurich, Switzerland.}

11 Corresponding author: J. Huttunen, Finnish Meteorological Institute, Kuopio Unit, P.O. Box
12 1627, FI-70211 Kuopio, Finland. (jani.huttunen@fmi.fi)

13

14 **Abstract**

15 The Aerosol Direct Radiative Effect (ADRE) is defined as the change in the solar radiation flux, F , due
16 to aerosol scattering and absorption. The difficulty in determining ADRE stems mainly from the need
17 to estimate F without aerosols, F^0 , with either radiative transfer modelling and knowledge of the
18 atmospheric state, or regression analysis of radiation data down to zero aerosol optical depth (AOD), if
19 only F and AOD are observed. This paper examines the regression analysis method by using modeled
20 surface data products provided by the AEROSOL ROBOTIC NETWORK (AERONET). We extrapolated F^0 by
21 two functions: a straight linear line and an exponential nonlinear decay. The exponential decay
22 regression is expected to give a better estimation of ADRE with a few percents larger extrapolated F^0
23 than the linear regression. We found that, contrary to the expectation, in most cases the linear

24 regression gives better results than the nonlinear. In such cases the extrapolated F^0 represents an
25 unrealistically low water vapour column (WVC), resulting in underestimation of attenuation caused by
26 the water vapour, and hence too large F^0 and overestimation of the magnitude of ADRE. The nonlinear
27 ADRE is generally 40-50 % larger in magnitude than the linear ADRE due to the extrapolated F^0
28 difference. Since for a majority of locations, AOD and WVC have a positive correlation, the
29 extrapolated F^0 with the nonlinear regression fit represents an unrealistically low WVC, and hence too
30 large F^0 . The systematic underestimation of F^0 with the linear regression is compensated by the positive
31 correlation between AOD and water vapour, providing the better result.

32

33 **1. Introduction**

34 Significant uncertainties exist in the current estimates of aerosol effects on climate (IPCC, 2013). This
35 holds also for the aerosol direct radiative effect (ADRE) and aerosol direct radiative forcing (ADRF).
36 The ADRE defines the attenuation of the (cloud free sky) surface solar radiation flux (F) due to aerosol
37 scattering and absorption. Herein, we consider the solar radiation flux at the surface, although ADRE
38 applies also for the longwave flux and above the atmosphere. In the definitions of ADRE and ADRF,
39 effects relate to both anthropogenic and natural aerosol particles, while forcing refers to the impact of
40 anthropogenic aerosol particles. Although, e.g., Myhre (2009) recently showed an increment of the
41 consistency between observation based and global aerosol model estimates, with a reduction in the
42 uncertainty of this effect, other studies (e.g., Loeb and Su, 2010) highlight that considerable
43 uncertainties are still associated with ADRE, mainly due to the uncertainties in single scattering albedo
44 (SSA). Satheesh and Ramanathan (2000) employed a method in which ADRE is estimated using the
45 aerosol direct effect efficiency (ADREE), which is the ADRE normalized by the aerosol optical depth
46 (AOD), and it is estimated by fitting a straight line into surface solar flux and AOD observations. A

47 linear dependence between aerosol attenuation and AOD has been commonly assumed when estimating
48 ADRE (e.g., Kaufman et al., 2002; Bush and Valero, 2002, 2003; Dumka et al., 2006; Roger et al.,
49 2006; di Sarra et al., 2008; Garcia et al., 2009; Satheesh et al., 2010). Typical attenuation of radiation
50 intensity, however, implies nonlinear decay, as considered by e.g. Conant et al. (2003), Markowicz et
51 al. (2008) and Kudo et al. (2010). Thus, a linear fit to F and AOD data may result in an incorrect
52 extrapolation of F_0 .

53 The aim of this paper is to examine the uncertainties involved in estimating ADRE, both using
54 the linear fitting method and a nonlinear approach if F and AOD data are available from surface or
55 satellite measurements. For this, we use Aerosol Robotic Network (AERONET) products
56 (<http://aeronet.gsfc.nasa.gov/>) from all available AERONET stations, which cover different aerosol
57 types and surface reflectance properties and provide modelled surface solar radiation fluxes also. We
58 conducted our analysis using these modeled fluxes since they represent realistically enough the aerosol-
59 induced relative changes in F and furthermore give an estimate for F^0 , which is self-consistent within
60 the selected F (AOD) data set. As AERONET provides an estimation of F^0 , we can compare the
61 estimations immediately with the baseline (AERONET). Special attention is paid to the possible effect
62 of water vapour on estimating ADRE.

63

64 **2. Methods and data**

65 AERONET is a ground-based remote-sensing global network of Cimel sun photometers (Holben et al.,
66 1998) including the AERONET inversion code with radiative transfer code implementation. The
67 inversion strategy, described in Dubovik and King (2000), provides a group of parameters, e.g. AOD,
68 Ångström exponent (AE) and water vapour column (WVC) from the sun measurements and e.g. SSA,

69 asymmetry parameter (ASYM) and size distribution from the sky measurements. AOD is provided with
70 wavelength channels 340, 380, 440, 500, 670, 870, 1020 and 1640 nm (all or some of these, depending
71 on site of AERONET), WVC from 940 nm and e.g. SSA and ASYM from 440, 670, 870 and 1020 nm.
72 The Discrete Ordinates (DISORT) provides broadband fluxes (both at the top of atmosphere and at the
73 surface, with and without aerosols), calculated with the correlated-k distribution in the Global
74 Atmospheric Model (GAME) code from 200 nm to 4000 nm. The ozone is based on monthly averaged
75 climatology by the Total Ozone Mapping Spectrometer (TOMS). Moreover, the US standard 1976
76 atmosphere model sets the atmospheric gaseous profile. The surface reflectivity is approximated by the
77 Bidirectional Reflectance Distribution Function (BRDF) and observations from the Moderate-
78 Resolution Imaging Spectroradiometer (MODIS). More details about the AERONET description from
79 e.g. García et al. (2012). The uncertainty of AOD is 0.01-0.02 depending on the wavelength (Eck et al.,
80 1999), the uncertainty in SSA is approximately 0.03 (Dubovik et al., 2000), and the uncertainty in
81 WVC of 12 % (Holben et al., 1998). We used broad-band modeled surface shortwave fluxes from this
82 data set. In this study, level 1.5 sky AERONET data are divided into groups by station, season
83 (December-February, March-May, June-August and September-November) and by solar zenith angle
84 (SZA) (3° steps in the range 0° - 80°). A dataset was included in the analysis if it had at least 20
85 observations and the data contained AOD 550 nm values above 0.3 and below 0.1. We chose to use
86 level 1.5 data because using level 2.0 would leave out all quality-assured data with AOD 440 nm < 0.4
87 (including e.g. quality assured SSA and F calculations). The drawback of this choice is that at these low
88 values of AOD, there are significant uncertainties in the optical properties retrieved. This is especially
89 true for SSA, which is an important parameter. Thus, we applied all other level 2 criteria except for
90 AOD (and SZA) limit, in order to enhance the accuracy of the data set selected. Moreover, we have
91 imposed an additional data flagging criterion, removing those SSA points at the AOD 440 nm < 0.4 ,
92 which are outside the average SSA \pm standard deviation, defined for the AOD 440 nm > 0.4 .

93 ADRE at the surface is the difference between the solar flux with and without aerosols: ADRE
 94 $= \Delta F = F^{aer} - F^0$ (F^{aer} is flux with aerosols). The major challenge obviously is the determination of F^0 .
 95 The methodology for its estimation employed in this study is illustrated in Fig. 1, in which F^{aer}
 96 (+symbols) is plotted as a function of AOD (from now on 550 nm) for the AERONET site in Kanpur
 97 station (26° N, 80° E) for the spring months March-May with $SZA = 69^\circ \pm 1.5^\circ$ (F^{aer} values were
 98 normalized for the average earth-sun distance and cosine correction of F^{aer} was done within SZA
 99 ranges to its midpoints). F^0 represents the case AOD = 0, but with measurements only at AOD above
 100 ca. 0.15, we have to extrapolate down to 0. In Fig. 1 we show two such extrapolations: a linear fit
 101 (dashed line) and an nonlinear decay fit (solid line) with the data.

102 We chose this data subset since it represent a case in which the F^{aer} and AOD data exhibit the
 103 natural nonlinear behavior of radiation intensity decay. Thus the resulting intercepts of the two curves
 104 at AOD = 0 are quite different, 317 Wm^{-2} with linear extrapolation and 349 Wm^{-2} with nonlinear
 105 regression, with a difference of 32 Wm^{-2} when estimating ADRE. Also, for each F^{aer} we show the
 106 corresponding AERONET F^0 (circles), based on the retrieved WVC and surface albedo, and calculated
 107 with a radiative transfer model (e.g., Garcia et al., 2008; Derimian et al., 2008). We use the ADRE
 108 obtained by averaging these F^0 (circles) values (bar at $F = 325 Wm^{-2}$ on the y-axis) as the benchmark
 109 against which the extrapolation methods are evaluated.

110 Mathematically, our analysis can be summed up as a comparison between the extrapolated
 111 ADRE

$$112 \quad ADRE_{extrapol} = \frac{1}{n} \sum F_i^{aer} - F_{extrapol}^0 \quad (1)$$

113 and the AERONET ADRE

$$114 \quad ADRE_{AERONET} = \frac{1}{n} \sum F_i^{aer} - \frac{1}{n} \sum F_i^0, \quad (2)$$

115 in where F_i^{aer} and F_i^0 is F^{aer} and F^0 , respectively, with i varying from one to the number of dataset, n .

116 Notably, the extrapolated F^0 ($F^0_{extrapol}$) derived with fits represents a single value for a dataset, but in the
 117 AERONET, F^0 is determined side-by-side with each F^{aer} . $F^0_{extrapol}$ is calculated using fits as follows

$$118 \quad F_i^{nonlin} = x_1 + x_2 * \exp(-x_3 * AOD_i); F_{extrapol}^{0,nonlin} = x_1 + x_2, \quad (3)$$

$$119 \quad F_i^{lin} = x'_1 + x'_2 * AOD_i; F_{extrapol}^{0,lin} = x'_1, \quad (4)$$

120 in where F_i^{nonlin} and F_i^{lin} is estimated F^{aer} derived for each AOD with the nonlinear and linear method,
 121 respectively. Constants of fits are x_1, x_2, x_3, x'_1 and x'_2 , and $F_i^{0,nonlin}$ and $F_i^{0,lin}$, thus $F^0_{extrapol}$ of the nonlinear
 122 and linear fits, are provided with the constants.

123 Our decision to use the modeled F from AERONET, instead of pyranometer measurements, was
 124 based on two different aspects. First, this allowed us to include a multiple number of sites, with very
 125 different and varying aerosol conditions. Second, AERONET data provided interesting ancillary
 126 measurements to support and better understand our analysis, WVC being the most crucial one. In
 127 addition, the AERONET F s agree with pyranometer measurements with a correlation better than 99%
 128 and the relative difference varies from 0.98 to 1.02 (Garcia et al., 2008). Moreover, we tested the
 129 analysis in two sites, Alta-Floresta and Goddard Space Flight Center (GSFC), by using pyranometer
 130 measured fluxes F and found no significant difference of the results in these two sites, if compared to
 131 the corresponding analysis using the AERONET-modeled fluxes instead (see Supplement Appendix A).

132

133 3. Results

134 As further examples of determining ADRE using regression analysis, we show F^{aer} and AOD data from
 135 four sites in Fig. 2. In addition, the linear (dashed line) and nonlinear decay (solid line) fits to the data
 136 are shown. The bar on the vertical axis represents the average (with STD) value for F^0 . GSFC (39° N,
 137 77° W) (SZA = 70°) (Fig. 2a) and Rio-Branco (10° S, 68° W) (SZA = 70°) (Fig. 2b) represent cases in
 138 which the data are of sufficient quality for estimating ADRE: AOD values reach close zero with only

139 minor changes in WVC, aerosol optical properties and surface reflectance for a given AOD, resulting in
140 a narrow spread in the data. In these cases, since the nonlinear decay represents a more realistic decay
141 of radiation intensity (based on squared values of residuals), the intersection of the nonlinear fit with
142 the AOD=0 axis (y-axis) is within the STD of the baseline value. Dhadnah (26° N, 56° E) (SZA = 70°)
143 (Fig. 2c) and GSFC at SZA = 22° (Fig. 2d) are examples of more challenging cases: in Fig. 2c only
144 data points with AOD > 0.2 exist so that a more extensive extrapolation is needed, and in Fig. 2d there
145 is significant scatter in the points.

146 Perhaps the most interesting feature shown in Fig. 2, which also significantly affects the quality
147 of ADRE estimation, is the correlation of F^0 with AOD. In Fig. 2a-d there is a negative correlation
148 while in 2b the correlation is positive. The negative correlation between F^0 and AOD is indirectly
149 caused mainly by a positive correlation of AOD with WVC due to humid airmasses with large aerosol
150 concentration. Only in some cases, where airmasses are dominated by dust aerosols, the correlation is
151 negative. With increasing AOD and WVC, the WVC dims an increasing fraction of the radiation
152 intensity – resulting in a smaller F^0 . The opposite occurs if AOD and WVC have a negative correlation.
153 Increase in the AOD as a function of WVC is presumably partly due to hygroscopic growth (e.g.,
154 Kitamori et al., 2009), although probably a major part of the correlation can be attributed to a large
155 variance in atmospheric conditions of aerosol properties and air humidity during seasons.

156 The intersections of the nonlinear decay fits (solid lines in Fig. 2) with the AOD = 0 axis –
157 313.5 W/m² (Fig. 2a), 295.9 W/m² (2b), 327.4 W/m² (2c) and 1008.9 W/m² (2d) – approximate the F^0
158 value at AOD = 0. This is clear from the figure, if one imagines straight line fits through the circles and
159 extrapolates fits down to AOD = 0. This approximation is, however, not necessarily a good one for the
160 mean F^0 , if F^0 and AOD correlate (through the AOD-WVC-correlation). For the negative correlation
161 cases (2a-d) the intersections of the nonlinear decay fits with the AOD = 0 axis tend to therefore over-
162 estimate the mean baseline F^0 (307.3 W/m² for 2a, 312.9 W/m² for 2c, and 972.1 W/m² for 2d) – as the

163 majority of F^0 values are below the extrapolated F^0 . Typically, for the positive correlation cases (2b,
164 mean of $F^0 = 303.4 \text{ W/m}^2$) the opposite occurs. As the linear fit obviously results in a lower estimation
165 of F^0 , the linear regression method can result often in a better estimation of the mean F^0 , as is clearly
166 the case in Fig. 2c (mean $F^0 = 306.7 \text{ W/m}^2$) and Fig. 2d (mean $F^0 = 973.0 \text{ W/m}^2$) – even if the nonlinear
167 regression is physically more correct.

168 The performance of the two different regression methods and, in particular, the WVC and AOD
169 correlation effect on the performance, is illustrated as scatter plots in Fig. 3. In Fig. 3a all data are
170 presented in ADRE (nonlinear decay method) and ADRE (AERONET $\Delta F^{average}$, Eq. 2) form. The colour
171 of the single points indicates the correlation of the WVC and AOD. In Fig. 3b the same is shown for the
172 linear regression case. Evidently a majority of the cases are such that WVC and AOD have a strong
173 positive correlation (red colored points). In addition, it seems that for most of these cases, the linear
174 regression method (Fig. 3b) results in a better ADRE estimation than the nonlinear decay regression
175 method (Fig. 3a). This means that the inaccuracy inherent in the linear regression cancels out errors
176 caused by the WVC and AOD correlation. For a weak WVC and AOD correlation, the nonlinear decay
177 method appears to be clearly better. Other parameters as surface albedo, ASYM or SSA do not play as a
178 crucial role as WVC. We classified the ADRE estimates of the both methods against the baseline in
179 respect of AOD, albedo, ASYM, SSA and WVC. It was evident that only WVC can explain the
180 observed differences of both methods when compared against the baseline (see Supplement Appendix
181 B). Moreover, we confirmed, by modeling a short wavelength range (310 nm -500 nm), that this WVC-
182 effect vanishes, if some other wavelength band as e.g. the visible range of 400-700 nm containing no
183 significant water vapour absorption is under consideration, instead of the broadband wavelength range
184 of F^{aer} (see Supplement Appendix C).

185 Next we investigated possible geographical features of this correlation. Figure 4 shows the
186 WVC and AOD correlation (in the color scales) at all the sites available from AERONET, in this case

187 for seasons; December-February (DJF, Fig. 4a), March-May (MAM, Fig. 4b), June-August (JJA, Fig.
188 4c) and September-November (SON, Fig. 4d)(all years available). Most of the points are colored either
189 green or red, indicating an absent or a positive correlation. The strongest positive correlation is for the
190 stations in Europe and eastern USA, presumably due to aerosol hygroscopic growth. This holds
191 especially for the JJA and SON- seasons The DJF and MAM- seasons provide weaker positive
192 correlation, indicating that the linear method can then provide there somewhat underestimated ADRE.
193 Interestingly, the strongest negative correlation appears during the JJA-season in the west Sahara's
194 region and Central-America, probably caused by a strong desert dust domination and low WVC in the
195 Saharan outflow region (Marsham et al., 2008). During those particular cases, the linear method can
196 significantly underestimate ADRE, as indicated by the points of largest negative WVC vs. AOD
197 correlation in Fig. 3b, while the nonlinear decay provides then a better estimate.

198 Finally, the ADRE estimations of all data are grouped together in numerical form in Table 1. As
199 already evident from the figures, the nonlinear decay regression method overestimates (mean = -57.2
200 Wm^{-2}) while the linear method underestimates (mean = -39.4 Wm^{-2}) the magnitude of ADRE
201 (AERONET value = -46.1 Wm^{-2}). Overall, the linear method yields better results than the nonlinear
202 decay method.

203 Previous studies have shown that the AERONET WVC agrees well with radiosonde sounding
204 data (e.g., Prasad and Singh, 2009; Bokoye et al., 2007). Also, Smirnov et al. (2004) indicates that the
205 AERONET WVC provides the root mean square difference of 7.0 % in a multiyear comparison with
206 WVC measurements derived from GPS. We also compared AERONET WVC measurements against
207 radiosonde data from five sites (Alta-Floresta, Cuiaba-Miranda, Niamey, Thessaloniki and Wallops)
208 and observed similarly high correlations between these two data sources. However, we wanted to
209 assess in particular whether there exists any systematic dependence between WVC from these two data
210 sources as a function of AOD, which could affect our ADRE analysis based on the modeled F . We

211 found that while the ratio between the AERONET and radiosonde WVC is essentially constant for
212 AODs (at 500nm) larger than about 0.1, in many sites WVC can deviate for the cases of smallest AOD
213 (below 0.1). We estimated how our ADRE values (based on the F and AOD relation) would change if
214 we normalized the AERONET-modeled fluxes to incorporate the WVC from the radiosonde
215 measurements instead of AERONET-measured WVC. We found that the increased WVC uncertainty at
216 the lowest AOD values introduces an insignificant change in our ADRE estimates.

217

218 **4. Conclusions**

219 Determining the ADRE at the Earth's surface from radiative flux, F , measurements is not
220 straightforward because it involves the estimation of the flux without aerosols F^0 . This requires either
221 radiative transfer modelling or an extrapolation of F down to AOD = 0.

222 We have evaluated two such extrapolation methods: i) a linear fit and ii) an nonlinear decay fit
223 to the F and AOD data. As a reference we used the AERONET ADRE data in which F^0 (and F) is
224 calculated with radiative transfer modelling. Radiation attenuation due to multiple scattering and
225 absorption results typically in a near nonlinear decay of the intensity, and thus the nonlinear decay
226 regression is expected to give a better estimation of ADRE. This would be the case if the typically
227 positive correlation of WVC and AOD would not affect the dependency. F^0 represents an
228 unrealistically low WVC, resulting in an underestimation of attenuation caused by the WVC, and hence
229 a too large F^0 . This leads to an overestimation of the magnitude of ADRE. For stations and data series
230 in which there is no correlation between WVC and AOD, the nonlinear decay fit is superior.

231 As the WVC effect was found to be of such importance, we also investigated the geographical
232 correlation of WVC and AOD. The positive correlations clearly dominate, and clear negative
233 correlations occur predominantly in desert dust dominated data series, such as the regions at the

234 Saharan outflow. The strongest positive correlation was found in in stations in Europe and Eastern
235 USA. Our results indicate that the regression method, either linear or nonlinear, can readily produce a
236 significant error due to the correlation of WVC and AOD. Since for a majority of locations, AOD and
237 water vapour column (WVC) have a positive correlation, the linear method gives somewhat better
238 results in general than the nonlinear approach, for the reasons discussed above. However, there are
239 specific regions of strong negative WVC and AOD correlation, most notably in the Saharan dust
240 outflow region, where the opposite takes place and nonlinear approach results in better estimate for
241 ADRE. Therefore, based on our results we recommend that when the surface ADRE is estimated by
242 using pyranometer and AOD measurements, the site-specific correlation between WVC and AOD
243 should be also estimated to deduce whether linear or nonlinear approach is more suitable. We moreover
244 recommend to take a one step forward and additionally attempt to correct for the possible bias due to
245 WVC and AOD correlation. If the data for the WVC is available, then better ADRE accuracy is likely
246 achieved if the flux measurements are normalized to constant WVC amount with simple scaling
247 obtained from RT modeling.

248

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345 Table 1. The estimated ADRE(F^{aer}) with standard deviations compared with the AERONET value.

346 MAD = Mean Absolute Deviation. Units are in Wm^{-2} , except for the correlation coefficient (CC).

Parameter	AERONET	Method	Estimate	Est. - AERONET	CC	MAD
ADRE	-46.1 ± 20.4	Exp. decay	-57.2 ± 23.4	-11.1	0.75	13.4
		Linear	-39.4 ± 16.9	+6.7	0.89	8.9

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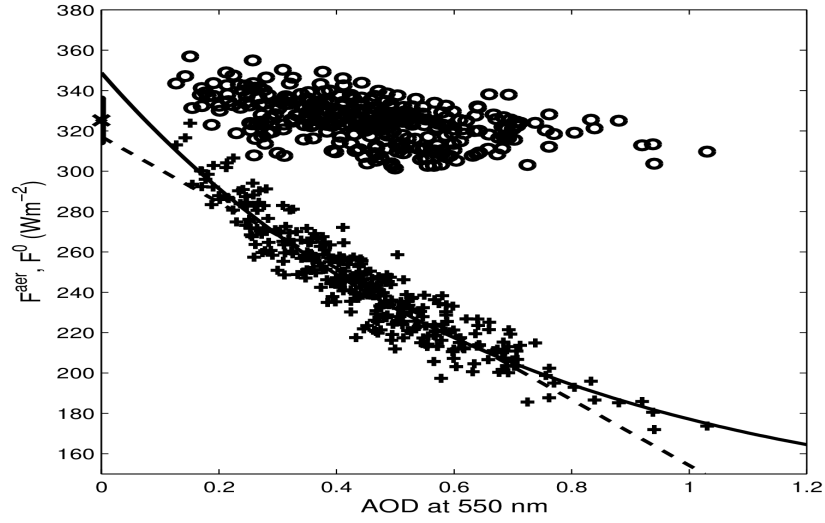
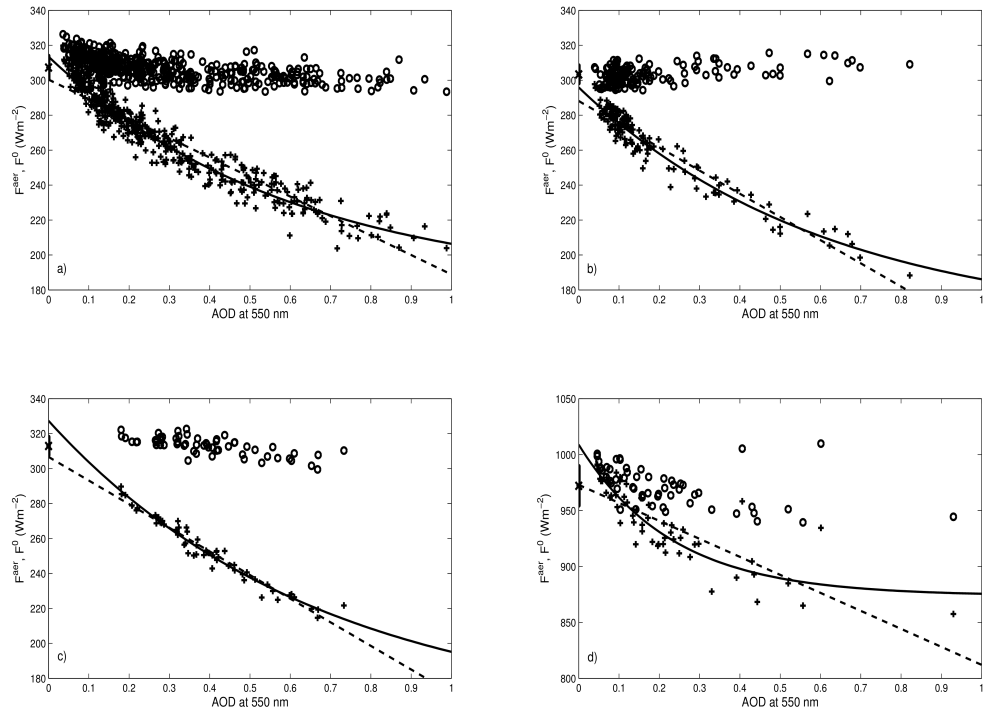


Figure 1: Radiative flux with aerosols F^{aer} (plusses) and without aerosols F^0 (circles) as a function of AOD for the AERONETsite in Kanpur in March-May and with $SZA = 69^\circ \pm 1.5^\circ$. The bar on the vertical axis represents the mean value of the estimated F^0 (all circles). The solid and dashed lines represent the exponential and linear fits to the data, respectively.



376 Figure 2: Same as Fig. 1, but for the June-August season in a) GSFC (SZA=70°), b) Rio-Branco (SZA
 377 = 70°), c) Dhadnah (SZA = 70°), d) GSFC (SZA = 22°).

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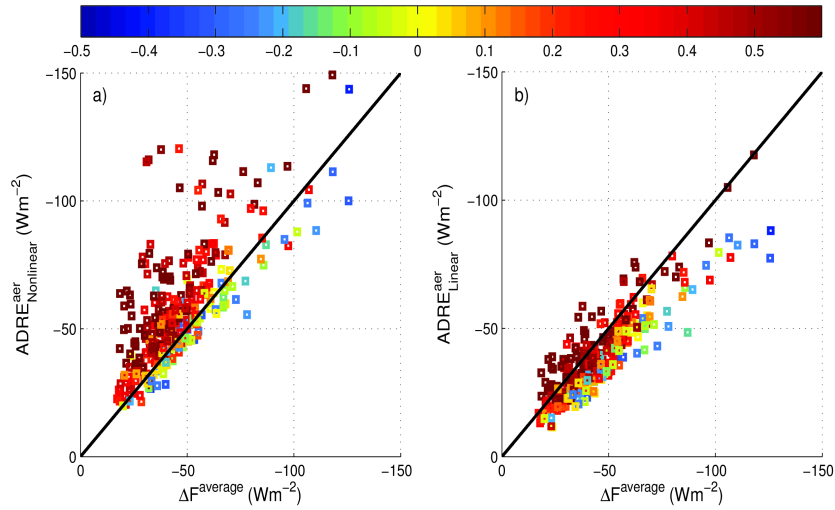
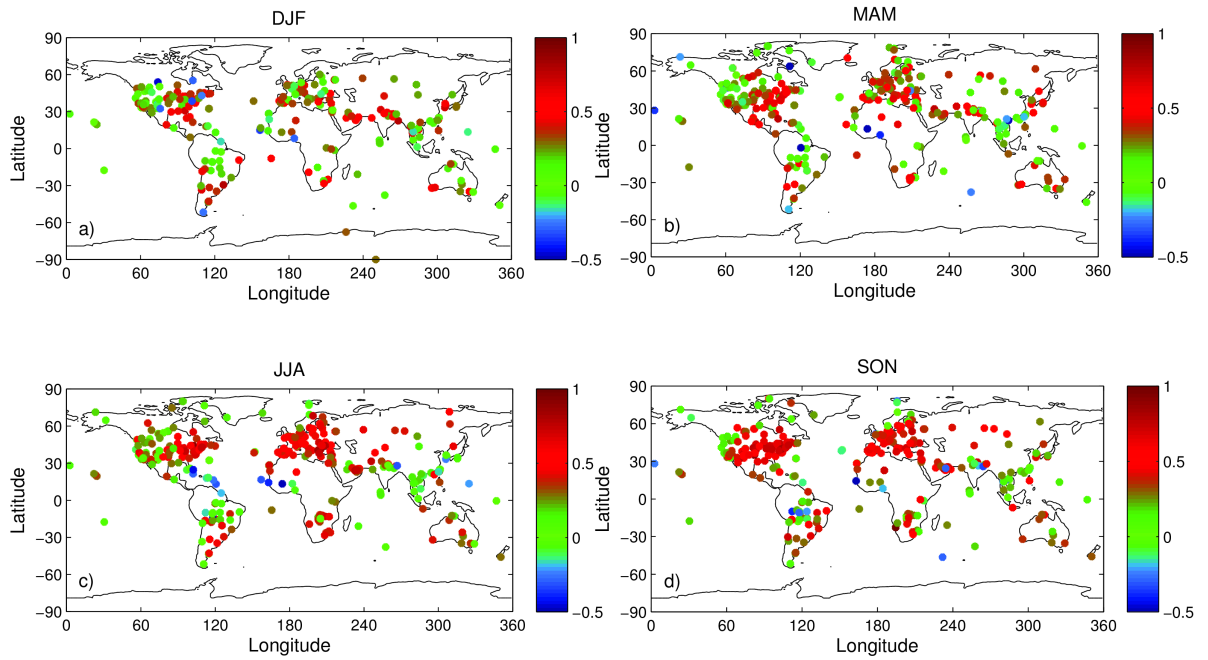


Figure 3: ADRE predicted with exponential decay (a) and linear (b) regression methods (equation 1), compared with AERONET values (equation 2). The color of the data points represents the correlation coefficient of the AOD and WVC correlation, with red color indicating positive and blue color negative correlation.



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440 Figure 4: Geographical distribution of the AOD and WVC correlation, at all AERONET stations
 441 considered in this study for a) December-February, b) March-May, c) June-August and d) September-
 442 November (all available years).