# Response to the Anonymous Referee#1's comments (acp-2016-680-RC1)

The authors have dealt with my suggestions made during preliminary review. Thank you for helping me better understand your study. I had missed that improvement of RMSE was large in my first read through. Overall the paper is well written and referenced. In regards to the choice of region, I think the discussion does a good job of making the case for South Asia being the most challenging test case for new methods.

**Authors:** We express our sincere thanks to the anonymous referee for his/her insightful and constructive comments and suggestions on this study. The comment/suggestions were to-the-point and very valuable for us to improve the scientific and technical clarity and quality of the manuscript. In the following, we itemize our point-by-point response to each of the concerns raised by the referee.

**Comment:1** Line 133: I think there is a typo on this line.

**Authors:** The sentence is rewritten in the revised manuscript.

**Authors change:**

**Line 144:** *Quaas et al., (2008)* have adopted the *Loeb (2004)* approach for the estimate of planetary albedo.

**Comment:2** Line 168: How much of an impact does the simplified surface albedo have? I would have thought that the calculation would have some sensitivity to whether it was looking over a forest or farmland. Maybe it averages out, but it seems like it might be enhancing the authors' calculation error. Sea surface albedo also varies a lot depending on meteorological conditions [Jin et al., 2011].

**Author:** In the present study, the surface albedo is used to simulate RFaci using the RT model, therefore the simulated values of $RF_{aci}$ are sensitive to the choice of the surface albedo but not the one computed using the statistical relationship between satellite based measurements. To estimate the sensitivity of the simulated $RF_{aci}$ to surface albedo in response to the reviewer's remark, we used different plausible values of surface albedo in sensitivity simulations with radiative transfer model to assess its impact on the simulate the $RF_{aci}$ and to compute the uncertainty statistics. These statistics are now reported in the revised manuscript and presented as supplementary material.

**Authors change:**

**Line 330-340:** In addition to above error budget, there are uncertainties involved in the RT simulated $RF_{aci}$ due to various parameters as shown above. In this regard, the surface albedo plays a major role in the simulation of $RF_{aci}$. In the standard approach, we have considered a surface albedo value 0.15 for land and the predefined option for the ocean surface albedo is used for the oceanic regions in the present study. To quantify the uncertainties involved due to assumptions about the surface albedo, we have simulated $RF_{aci}$ with different plausible surface albedo values and computed statistics as shown in **Table S3(a) and S3(b)**. The statistics shows that the considered values of surface albedo are suitably representative of the study regions. In addition, RT simulation have their own limitations and uncertainties e.g. inherent code accuracy, overestimate in calculated RF due to plane-parallel bias, 3-D radiative transfer effect etc. It would be useful to explore these issues in the future.

**Comment:3** Line 230: There might be a typo on this line discussing RMSE reduction.

**Author:** The sentence is revised and rewritten in the revised manuscript.

**Authors change:**

**Line 265-266:** The nonlinear fit increases the correlation by 21%-23% and reduce the RMSE by 0.007-0.011 W $m^{-2}$ compared to the multilinear approach.

**Comment:4** Line 262: There is a typo in the last sentence. I have no other major suggestions relating to this paper and find it acceptable for publication pending minor revisions and grammar corrections.

**Author:** The sentence is rewritten in the revised manuscript.

**Authors change:**

**Line 327-328:** Limitation involved in this approach or uncertainties in the satellite retrievals contribute to the overall uncertainty, which is difficult to quantify.

## Response to the Anonymous Referee#3's comments (acp-2016-680-RC2)

***General Comments:***

By extending the method of Quaas et al. (2008) to estimate $RF_{aci}$ and developing and employing a new evaluation scheme for it, this work contributes a useful analysis to the field of aerosol-cloud interactions. Some additional clarification is necessary, though, in order to document how exactly this work complements Quaas et al. (2008) and what advantages it introduces. Furthermore, the manuscript requires extensive copy- editing; as written, some results are hard to understand due to typographical errors and the manuscript is hard to follow at times.

**Authors:** We express our sincere thanks to the referee for his/her insightful and constructive comments and suggestions on this study. The comment/suggestions were to-the-point and very valuable for us to improve the scientific and technical clarity and quality of the manuscript. In the following, we itemize our point-by-point response to each of the concerns raised by referee.

**Comment:1** I strongly recommend that the authors request copy-editing services from Copernicus to improve the quality of the manuscript. In the Specific Comments section I have tried to document typographical and grammatical errors which produce confusion in interpreting the results, but overall there are many such corrections that should be made throughout the document.

**Authors:** The given specific comments are seriously considered and the typographical and grammatical errors are corrected throughout the document with the help of an expert to improve the quality of the manuscript. The suggested specific comments are seriously responded point-by-point in the revised manuscript.

**Comment:2** The authors estimate $N_d$ using an adiabatic liquid water cloud assumption. However, this assumption is invalid outside of stratiform clouds in the marine boundary layer, and similar estimates like Bennartz (2007) clearly indicate that this assumption is highly uncertain outside this type of regime. The authors should discuss the limitations of using $N_d$ in their Central India (CI) region, and in seasons dominated by non-stratiform clouds (such as the monsoon one they analyze).

**Authors:** The limitation of the computation of $N_d$ from satellite retrievals, and the associated uncertainty along with its contribution to the total uncertainty is discussed in the revised manuscript as suggested by the reviewer. This also takes into account new results on the retrievals of $N_d$ from satellites.

**Authors change:**

**Lines 137-144:** A limitation of this assumption is that it applies rather well for the stratiform clouds in the marine boundary layer, but less so for convective clouds. A detailed explanation and uncertainty assessment are described in *Bennartz, (2007) and Rausch et al., (2010)*. Recently, *Bennartz and Rausch, (2017)* show that the uncertainties in the CDNC climatology from 13-years of AQUA-MODIS observations are in the order of 30% in the stratocumulus regions and 60% to 80% elsewhere and its contribution to the total uncertainty for this study is discussed in the following section.

**<u>Comment:3</u>** Several clarifications should be made regarding the non-linear fitting technique. First, it isn't clear on line 135 why $\alpha_5$ should ever be set to 1; Quaas et al. (2008) does not seem to make this assumption - contrary to the assertion in lines 146-147 - and if the authors are suggesting this as an alternative formulation of equation (2), then some justification is necessary. For instance, in all except one of the nonlinear fits provided in Table S1, $\alpha_5$ is an order of magnitude smaller than 1. Second, the authors should clarify what method is used to perform the non-linear fits with a citation if possible, even if it's something standard such as non-linear least squares, for the sake of reproducibility.

**<u>Authors</u>:** The assumption of $\alpha_5$ set to be 1 was not clearly indicated in Quaas et al., (2008), apologies for this by the authors of our manuscript! - but a subsequent study, Ma et al., (2014) reassesses the same study by employing updated satellite products, where it is clearly indicated that for this study $\alpha_5$ was set to 1. This second paper is now referenced in the present study for clarity about this point. Second, as per the reviewer's suggestion, more details about the nonlinear least square method are included in the revised manuscript.

**<u>Authors change:</u>**

**Lines 175-177:** In the present study, instead of considering $\alpha_5$=1 in the multiple regression, as in *Quaas et al. (2008) and Ma et al., (2014),* we obtained the values of all six fitting parameters using a nonlinear fitting approach (L-M algorithm) for each month and region.

**Lines 163-175:** For that purpose, we adopted the new statistical nonlinear least square fitting approach to obtain the six fitting parameters in Eq. (3). Nonlinear least square methods involve an iterative improvement to parameters values in order to minimize the residual sum of squares between the observed values and the predicated value of the dependent variables. We used the Levenberg-Marquardt (L-M) algorithm (*Levenberg, 1944*) in the nonlinear least square approach to adjust the parameter values in the iterative procedure. This algorithm combines the Gauss-Newton method and the gradient descent method. In the gradient descent method, the sum of the squared errors is reduced by updating the parameters in the steepest descent direction. In the Gauss -Newton method, the sum of the squared errors is reduced by assuming the least squares function is locally quadratic, and finding the minimum of the quadratic. The L-M algorithm acts more like a gradient descent method when the parameters are far from the optimal value and acts more like to Gauss-Newton method when the parameters are close to their optimal value. More detail of this method is given in the literature (*Levenberg, 1944; Transtrum et al., 2010; Transtrum and Sethna, 2012*).

**<u>Comment:4</u>** In Section 3.2, the authors present an independent estimate of $RF_{aci}$ for validation purposes using a radiative transfer code. The authors should include some discussion of how this approach differs from those in the literature, such as Bellouin et al. (2013), and what its limitations are given the dataset and methodology employed. Furthermore, if the use of the radiative transfer code is so readily evaluated in conjunction with satellite data, then what advantage does equation (2) offer in terms of developing constraints for $RF_{aci}$?

**<u>Authors</u>:** As per the reviewer's suggestions the difference between the RT simulation of Bellouin et al., 2013 and in the present study is now explained in detail in the revised manuscript.

Following Quaas et al., (2008), Bellouin et al., (2013) performed a similar study (in terms of radiative forcing due to aerosol-cloud interactions) making use of the MACC aerosol reanalysis data and they used a radiative transfer code including a Monte-Carlo method to obtain the standard deviation for the analysis of uncertainty. However, in the present study, $RF_{aci}$ is simulated using a radiative transfer code (SBDART) to evaluate the performance of the statistical approach used to compute the $RF_{aci}$. The use of a simple, analytical expression to assess the aerosol-cloud radiative forcing has the advantage of being very easily understandable, accessible and applicable.

**Authors change:**

**Lines 190-195:** Following the study by *Quaas et al., (2008)* study, *Bellouin et al., (2013)* performed a similar study with MACC reanalysis data, in which RT simulations, using a Monte - Carlo method, were carried out to obtain the standard deviation for the uncertainty analysis. However, in the present study, $RF_{aci}$ is simulated using an RT model (SBDART) to validate the performance of both the statistical approaches used to compute the $RF_{aci}$ using the statistical relationship between satellite measurements.

**Comment:5** In equations (3-4) the authors require estimates of $d \ln N_d / d \ln \tau_\alpha$ but do not state where these come from. If they use the regression approach of Quaas et al. (2008), then this should be indicated.

**Authors:** We are thankful to reviewer to point out the lack of sufficient information. The values of $d \ln N_d / d \ln \tau_\alpha$ is obtained here are the same as Quaas et al., (2008); derived using a linear regression. This is now clarified in the revised manuscript.

**Authors change:**

**Lines 151-154:** $d \ln N_d / d \ln \tau_\alpha$ is the sensitivity of cloud droplet number concentration ($N_d$) to a relative change in AOD. It is computed as the slope of the linear regression fit between the natural logarithm of $N_d$ and AOD (*Quaas et al., 2008*). This value is calculated on a month-by-month basis and is unique to each region studied.

**Comment:6** The discussion of uncertainty in the estimates of $RF_{aci}$ in Section 4.2 does not seem to follow from the results presented earlier in the manuscript. On lines 258-259 the authors suggest that the nonlinear fitting approach reduces uncertainty by 20%-25%, but it is not clear where this estimate is coming from. The authors' analysis of the reduction in RMSE of planetary albedo compared to the radiative transfer simulations is not a measure of uncertainty, if that's what this statistic refers to. This estimate should be removed, and the authors should instead expand their error-propagation analysis to justify the estimate of -0.08 $W/m^2$. For instance, in relation to the previous comment, how does uncertainty in the regional and seasonal estimates **of** $d \ln N_d / d \ln \tau_\alpha$ influence the estimate of $RF_{aci}$?

**Authors:** We are thankful to reviewer for his/her suggestion to expand the error analysis. According to reviewer's suggestion, we introduce the uncertainties involved in the present study due to different parameters considered in this study. The detailed information, including a table about the uncertainty budget is now included in the revised manuscript.

The reduction in RMSE of planetary albedo compared to RT simulations is not a measure of uncertainty in the present study. It is the reduction in RMSE of $RF_{aci}$ due to nonlinear least square approach compared to multilinear regression. Additionally we incorporated the mean relative difference in $RF_{aci}$ due to both statistical approaches.

**Authors change:**

**Lines 266-272:** The relative difference between the RT-simulated and the statistically computed $RF_{aci}$ are computed for both the statistical methods. The mean relative difference in $RF_{aci}$ for anthropogenic fraction of AOD is 0.021 W $m^{-2}$ in the nonlinear and 0.033 W $m^{-2}$ in the multilinear statistical approach, whereas, for $RF_{aci}$ of natural fraction of AOD, it is 0.032 W $m^{-2}$ in nonlinear and 0.053 W $m^{-2}$ in multilinear statistical approach. This suggests that the use of the nonlinear fitting approach reduces the uncertainty by 36%-39% compared to the multilinear regression.

**Lines 298-309:** The uncertainties due to sensitivity of $N_d$ to a relative change in AOD ($d \ln N_d / d \ln \tau_a$) contribute most to the total uncertainty. For $N_d$ sensitivities to changes in AOD, standard deviations are derived from minimum and maximum values obtained for each season. Following the study by *Bellouin et al., (2013)*, the standard deviations are derived from minimum and maximum values by defining 4-sigma interval, which covers the large range of sensitivities and spatio-temporal variabilities. To define the standard deviations in $RF_{aci}$ due to variation in $d \ln N_d / d \ln \tau_a$, $RF_{aci}$ is recomputed using those standard deviations of $N_d$ sensitivities to changes in AOD. Table 2 shows the seasonal and regional sensitivities of $d \ln N_d / d \ln \tau_a$ along with their statistical standard deviation, which is computed from the minimum and maximum values for each season. The associated range in $RF_{aci}$ both for anthropogenic and natural fraction of AOD is also shown in Table 2, where the standard deviation of $RF_{aci}$ shows the variation due to change in $d \ln N_d / d \ln \tau_a$, which finally contribute to the total uncertainty.

**Lines 314-324:** In the present study, except for the statistical fitting method, all the variables and methodologies are same for both the statistical approach. Therefore, we used the relative difference between the RT-simulated and statistically computed $RF_{aci}$ as an uncertainty due to the choice of the statistical fitting approach for both the statistical fitting methods. As shown in section 4.1, the mean relative differences for the nonlinear and multilinear approaches are 0.021 W $m^{-2}$ and 0.033 W $m^{-2}$, respectively, in $RF_{aci}$ for anthropogenic fraction, whereas, for the $RF_{aci}$ of the natural fraction of AOD, these are 0.032 W $m^{-2}$ and 0.053 W $m^{-2}$ for nonlinear multilinear statistical approaches, respectively. Table 3 lists the uncertainty due to different parameters involved in the satellite-based estimate of $RF_{aci}$. We quantify the relative error as the square root of the sum of the squared relative errors for all individual contributions. This yields an influence of these relative uncertainties in the input quantities on the computed $RF_{aci}$ of ~±0.08W$m^{-2}$.

***Specific Comments:***

**Comment:1** Lines 12-15: This sentence is very awkward and partially repeats itself halfway through.

**Authors:** The sentence is rewritten in the revised manuscript.
**Authors change:**
**Lines 11-14:** Here we employ a new statistical approach to obtain the fitting parameters, determined using a non-linear least square statistical approach, for the relationship between planetary albedo and cloud properties and, further, the relationship between cloud properties and aerosol optical depth.

**Comment:2** Lines 18-20: Sentence needs to clarify what is being compared against with the correlation and error statistics.
**Authors:** The sentence is rewritten as per the reviewer's suggestion.
**Authors change**:
**Lines 14-20:** In order to verify the performance, the results from both statistical approaches (previous and present) were compared to the results from radiative transfer simulations over three regions for different seasons. We find that the results of the new statistical approach agree well with the simulated results both over land and ocean. The new statistical approach increases the correlation by 21%-23% and reduce the error, compared to the previous approach.

**Comment:3** Lines 37-38: Following McComiskey et al. (2009), $d \ln N_d / d \ln \tau_\alpha$ is not computed using partial derivatives and is not calculated with LWP held constant; please remove this statement, or clarify how this relationship differs from the other ACI metrics that could be considered.
**Authors:** We are sorry to introduce the misleading information in the sentence and according to reviewer's suggestions, the sentence is removed in the revised manuscript.
**Authors change:**
**Lines 34-39:** *Feingold et al. (2001, 2003); McComiskey et al., (2009)* proposed a metric to quantify the microphysical component of the cloud albedo effect ($ACI = -d \ln N_d / d \ln \alpha$), where $N_d$ is the cloud droplet number concentration and α in some proxy for the aerosol burden. A variety of proxies has been used to represent the cloud response to the change in aerosol, e, g., cloud optical depth ($\tau_c$), cloud drop number concentration ($N_d$) and cloud droplet effective radius ($r_e$).

**Comment:4** Line 39: Need to define re as "droplet effective radius"
**Authors:** $r_e$ defined as "cloud droplet effective radius" in the revised manuscript.
**Authors change:**
**Lines 37-39:** A variety of proxies has been used to represent the cloud response to the change in aerosol, e, g., cloud optical depth ($\tau_c$), cloud drop number concentration ($N_d$) and cloud droplet effective radius ($r_e$).

**Comment:5** Lines 40-41: Because they are column integrals, metrics like aerosol optical depth do not necessarily represent just the particles impacting clouds - just the total ambient aerosol burden, particularly with respect to larger particles. Please rephrase accordingly.
**Authors:** The sentence is revised according to reviewer's suggestion in the revised manuscript.
**Authors change:**

**Lines 39-41:** Similarly, various proxies have been used to represent the total ambient aerosol burden, including aerosol number concentration ($N_a$), aerosol optical depth ($\tau_a$) and aerosol index (AI).

**Comment:6** Lines 68-74: The first sentence is something of a non-sequitur and could be removed entirely. The second sentence is awkwardly phrased; it would be better to point out that the aerosol mixture in this region is very heterogeneous in time and space with respect to size distribution and chemical composition.
**Authors:** The suggested sentence is removed and others are rewritten in the revised manuscript.
**Authors change:**
**Lines 68-76:** The rapid socio-economic development in the recent past has increased the anthropogenic emissions in the South Asian region along with several parts of the world. The South Asian ones are among the potential sources of a variety of aerosol species; both natural and anthropogenic, and extensive investigations are being made in the past years (e.g., *Chin et al., 2000; Di Girolamo et al., 2004; Moorthy et al., 2013*). These densely populated regions with the increasing power demand, fuel consumption and equally diverse geographical features are also vulnerable to the impacts of atmospheric aerosols to the climate (e.g. *Liu et al., 2009*).

**Comment:7** Lines 82-85: It would be extremely helpful to the reader if you included a figure that outlined where these regions are on a map.
**Authors:** As per the reviewer's suggestions, a figure is included in the revised manuscript, which shows the study regions on a map.
**Authors change:**
**Lines 86-91and Figure-1 :** Therefore, we discuss the $RF_{aci}$ for both anthropogenic and natural fraction of aerosol for a period of six-years (2008-2013) for three different regions of south Asia (**Fig. 1**, Arabian Sea (AS; 63°E-72°E, 7°N-19°N), Bay of Bengal (BOB; 85°E-94°E, 7°N-19°N) and Central India (CI; 75°E-84°E, 20°N-30°N)), having significantly distinct aerosol environments as a result of variations in aerosol sources and transport pathways (*Cherian et al., 2013; Das et al., 2015; Tiwari et al., 2015*).

**Comment:8** Lines 91-93: This sentence should be flipped with the following and the beginning of the paragraph re-written to emphasize that your data comes predominantly from MODIS and CERES; then you should dive into the details of which data product (and citation) you use for each specific derived quantity.
**Authors:** The sentence is revised as per reviewer's suggestions.
**Authors change:**
**Lines 95-103:** Data acquired by MODerate Resolution Imaging Spectroradiometer (MODIS) and Clouds and the Earth's Radiant Energy System (CERES) mounted on Aqua (*Parkinson, 2003*) and Ozone Monitoring Instrument (OMI) onboard Aura (*Schoeberl et al., 2006*) are used in this study. We use the broadband shortwave planetary albedo ($\alpha$) (*Wielicki et al., 1996; Loeb, 2004; Loeb et al., 2007*) as retrieved by the CERES in combination with cloud properties from the MODIS (*Minnis et al., 2003*) and AOD ($\tau_a$) and fine mode fraction (FMF) as retrieved by the MODIS onboard Aqua (*Remer et al., 2005*).

**Comment:9** Lines 128-131: Pursuant to the general comment about $N_d$, the authors should discuss the limitations of this method for estimating $N_d$.

**Authors:** The limitation of $N_d$ along with its contribution to the total uncertainty is discussed in the revised manuscript as per suggestion.

**Authors change:**

**Lines 137-143:** A limitation of this assumption is that it applies rather well for the stratiform clouds in the marine boundary layer, but less so for convective clouds. A detailed explanation and uncertainty assessment are described in *Bennartz, (2007) and Rausch et al., (2010)*. Recently, *Bennartz and Rausch, (2017)* show that the uncertainties in the CDNC climatology from 13-years of AQUA-MODIS observations are in the order of 30% in the stratocumulus regions and 60% to 80% elsewhere and its contribution to the total uncertainty for this study is discussed in the following section.

**Comment:10** Line 132: Where does this particular value for γ come from?

**Authors:** The reference for the value of γ is added in the revised manuscript.

**Authors change:**

**Line 137:** Where, a constant value of $\gamma=1.37\times10^{-5}$ $m^{-0.5}$ (*Quaas et al., 2006*) is used in this study.

**Comment:11** Lines 144-152: At a minimum, this paragraph needs additional detail on what nonlinear fitting approach was used (non-linear least squares? some other method?) with a citation if applicable.

**Authors:** As per reviewer's suggestion, the detail about nonlinear method is discussed in the revised manuscript.

**Authors change:**

**Lines 162-175:** For that purpose, we adopted the new statistical nonlinear least square fitting approach to obtain the six fitting parameters in Eq. (3). Nonlinear least square methods involve an iterative improvement to parameters values in order to minimize the residual sum of squares between the observed values and the predicated value of the dependent variables. We used the Levenberg-Marquardt (L-M) algorithm (*Levenberg, 1944*) in the nonlinear least square approach to adjust the parameter values in the iterative procedure. This algorithm combines the Gauss-Newton method and the gradient descent method. In the gradient descent method, the sum of the squared errors is reduced by updating the parameters in the steepest descent direction. In the Gauss -Newton method, the sum of the squared errors is reduced by assuming the least squares function is locally quadratic, and finding the minimum of the quadratic. The L-M algorithm acts more like a gradient descent method when the parameters are far from the optimal value and acts more like to Gauss-Newton method when the parameters are close to their optimal value. More detail of this method is given in the literature (*Levenberg, 1944; Transtrum et al., 2010; Transtrum and Sethna, 2012*).

**Comment:12** Line 164: Before this sentence, it would be useful if the authors list the variables required to perform their SBDART computations.

**Authors:** Following this suggestion, a list of input variables and their sources are tabulated as table-1 in the revised manuscript.

**Authors change:**

**Lines 200-201: Table 1** shows the list of input parameters and their source provided to the RT model for the estimate of $RF_{aci}$.

**Comment:13** Line 184-185: Please clarify the difference between $\boldsymbol{\tau_\alpha}$ and $\tau_\alpha{}^{ant/nat}$. Presumably the first is the total AOD and the second is just the anthropogenic/natural contribution to AOD?

**Authors:** Yes, it is correct. The difference between both the terms $\tau_\alpha$ and $\tau_\alpha{}^{ant/nat}$ are clarified in the revised manuscript.

**Authors change:**

**Lines 154-155:** $\tau_a$ is the total AOD, whereas, $\tau_a^{ant/nat}$ are the anthropogenic and natural AOD, respectively, derived from the FMF and UV-AI as estimated above.

**Comment:14** Line 185 and Equation 5: I would recommend writing out explicitly $N_d' = N_d + \Delta N_d$ in both locations.

**Authors:** Terms are modified as per reviewer's suggestions.

**Authors change:**

**Lines 220-221:** The perturbed value of $N_d'$ ($N_d + \Delta N_d$) is used to obtain a perturbed value of $r_e$ using Eq. (5) for constant liquid water content because $r_e$ is used as an input to the radiative transfer code.

**Equation -5**

$$N_d' = q_l / (\frac{4}{3} \pi r_e{}^3 \rho_w) \quad (1)$$

**Comment:15** Lines 202-203: "Weight" is the wrong word; according to Table S1, it's simply that the magnitude of the coefficients are different.

**Authors:** The word "weight" is replaced by "magnitude of the coefficient" as per reviewer's suggestion.

**Authors change:**

**Lines 237-239:** The magnitude of the coefficients $\alpha_4$ and $\alpha_6$ is larger in the nonlinear fit than the multilinear regression fitting, which may reduce the magnitude of the coefficient $\alpha_5$.

**Comment:16** Lines 225-227: Rephrase to avoid using terms like "satisfactory results" in preference for neutral language.

**Authors:** The term "satisfactory results" is replaced in the revised manuscript.

**Authors change:**

**Lines 260-261:** The analysis showed good statistical agreement with Pearson's correlation coefficient r=0.82 and 0.75 and RMSE=0.037 $Wm^{-2}$ and 0.042 $Wm^{-2}$ for the anthropogenic and natural fraction of aerosols, respectively.

**<u>Comment:17</u>** Lines 229-231: The phrasing "... decreases RMSE by from 0.007 to 0.011 ..." is clearly a mistake; please delete whichever word is wrong and be clear about how the RMSE is changing.

**<u>Authors</u>:** The sentence is revised and rewritten in the revised manuscript.

**<u>Authors change:</u>**

**Lines 264-272:** The nonlinear fit increases the correlation by 21%-23% and reduce the RMSE by 0.007-0.011 W $m^{-2}$ compared to the multilinear approach. The relative difference between the RT-simulated and the statistically computed $RF_{aci}$ are computed for both the statistical methods. The mean relative difference in $RF_{aci}$ for anthropogenic fraction of AOD is 0.021 W $m^{-2}$ in the nonlinear and 0.033 W $m^{-2}$ in the multilinear statistical approach, whereas, for $RF_{aci}$ of natural fraction of AOD, it is 0.032 W $m^{-2}$ in nonlinear and 0.053 W $m^{-2}$ in multilinear statistical approach. This suggests that the use of the nonlinear fitting approach reduces the uncertainty by 36%-39% compared to the multilinear regression.

# A new statistical approach to improve the satellite based estimation of the radiative forcing by aerosol- cloud interactions

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## Abstract

In a previous, study of *Quaas et al., (2008)* the radiative forcing by anthropogenic aerosol due to aerosol-cloud interactions, $RF_{aci}$, was obtained by a statistical analysis of satellite retrievals using a multilinear regression. Here we employ a new statistical approach to obtain the ~~six~~ fitting parameters, determined using a non-linear least square statistical approach~~to obtain the six fitting parameters~~, for the relationship between planetary albedo and cloud properties and, further, the relationship ~~of the~~between cloud properties and aerosol optical depth. ~~The statistical approach is~~In order to verify the performance, the results from both statistical approaches (previous and present) were compared to the results from radiative transfer simulations over three ~~different~~ regions ~~and~~ for different seasons. We find that the results of the new statistical approach agree well with the simulated results both over ~~both~~ land and ocean. The new statistical approach increases the correlation ~~coefficient of the fitted to the satellite-retrieved albedo~~ by 21%-23% and ~~decreases~~reduce the error, compared to the previous approach.

## 1 Introduction

Aerosols are considered to have a large effect on climate, both through aerosol radiation interactions, and through aerosol-cloud interactions by serving as cloud condensation nuclei (CCN), therefore increasing $N_d$ and thus cloud albedo (*Twomey, 1974*), as well as rapid cloud adjustments (*Boucher et al., 2013*). Much work has been done to quantify the radiative forcing by aerosol-cloud interaction ($RF_{aci}$), yet it remains highly uncertain. The annual radiative forcing from aerosol induced changes in cloud albedo were reported as -0.7 $Wm^{-2}$ with an uncertainty range -1.8 to -0.3 $Wm^{-2}$ (*Boucher et al., 2013*); this effect could offset much of the warming from greenhouse gases (*Huber and Knutti, 2011*), emphasizing the need to understand the effect so that we can better predict the future climate.

In this study, we concentrate on the $RF_{aci}$, the change in cloud albedo with increasing aerosol. An increasing aerosol at constant cloud water content is supposed to decrease droplet size, which in turn increases the cloud albedo due to the increase scattering of the smaller, more numerous cloud droplets. *Feingold et al. (2001, 2003); McComiskey et al., (2009)* proposed a metric to quantify the microphysical component of the cloud albedo effect ($ACI = -d \ln N_d / d \ln \alpha$), where $N_d$ is the cloud droplet number concentration and α in some proxy for the aerosol burden. ~~Note that the partial derivatives must be calculated at constant liquid water path (LWP).~~ A variety of proxies has been used to represent the cloud response to the change in aerosol, e, g., cloud optical depth ($\tau_c$), cloud drop number concentration ($N_d$) and cloud droplet effective radius ($r_e$~~.~~). Similarly, various proxies have been used to represent the total ambient aerosol ~~particles affecting the cloud~~burden, including aerosol number concentration ($N_a$), aerosol optical depth ($\tau_a$) and aerosol index (AI). An overview about published relationships and their biases due to mismatches between process- and analysis scales are discussed in *McComiskey and Feingold, (2012)*. Values for ACI metrics from observations often differ significantly from model-based values (*Quaas et al., 2008, 2009; Bellouin et al., 2008; Penner et al., 2011, 2012*). For example, the observational-based values of $RF_{aci}$, often in the range of -0.2 to -0.6 Wm$^{-2}$ (*Quaas et al., 2008; Bellouin et al.,2013*), is tend to be weaker than the modeled values in the range of -0.5 to -1.9 Wm$^{-2}$ (*IPCC, 2007*). The differences in model and observational-based $RF_{aci}$ have to be reconciled. *Penner et al., (2011)* reported that the lower sensitivities of cloud droplet number concentration, when considering aerosol optical depth (AOD) compared to aerosol index as aerosol quantity may lead to a significant underestimation in satellite-based $RF_{aci}$. However, *Quaas et al., (2011)* pointed out the weaknesses in the approach used by *Penner et al., (2011)*.

Clearly, further study is needed to reduce the uncertainties in both observational- and model-based estimates of aerosol RFaci and to reconcile the differences.

*Quaas et al., (2008)* derived the anthropogenic aerosol $RF_{aci}$ based on satellite retrievals of aerosol and clouds properties using statistical relationships between cloud properties and anthropogenic aerosols without the use of radiative transfer model. They developed a statistical relationship between planetary albedo and cloud properties using a multilinear fit, and further, the relationships of cloud properties and aerosol optical depth. *Quaas et al. (2008)* suggested that uncertainties in the statistical relationship and fitting parameters introduced uncertainty in the estimate of $RF_{aci}$. Therefore, it is useful to reassess the estimated $RF_{aci}$ by using a new statistical fitting approach. The main objective of this study is to explore the uncertainty in the satellite-based quantification of $RF_{aci}$. This study differs from previous studies by introducing new statistical fitting approach to obtain the fitting parameters for the estimates of $RF_{aci}$, determined using a nonlinear fit between planetary albedo and cloud properties. To verify the present approach, the results from both statistical approaches are compared with the results from a radiative transfer model.

~~Recent studies suggest that~~The rapid socio-economic development in the ~~south Asian~~ recent past has increased the anthropogenic emissions in the South asian region ~~is one of the world's most populous (~24%~~along with several parts of the world~~ population) region with growing industrial~~. The South Asian ones are among the potential sources of a variety of aerosol species; both natural and ~~transport sectors. A large~~anthropogenic, and extensive investigations are being made in the past years (e.g., *Chin et al., 2000; Di Girolamo et al., 2004; Moorthy et al., 2013*). These densely populated regions with the increasing power demand, fuel consumption~~,~~ and equally diverse geographical features ~~make this region among~~are also vulnerable to the ~~global hotspots~~impacts of atmospheric aerosols~~.~~ to the climate (e.g. *Liu et al., 2009*). The complex geography of this region contributes significant amounts of natural aerosols (desert dust, pollen, sea-salt etc) into the atmosphere, which mix with anthropogenic ones, making the aerosol environment one of the most complex in the world (*Moorthy et al., 2015*). The large spatial heterogeneity of the sources coupled with the atmospheric dynamics driven by topography and contrasting monsoons, make South Asia's aerosol very difficult to characterize and to model their implications on radiative and climate forcing. While tropospheric perturbations would produce strong regional signatures, their global impacts still remain marginally above the uncertainty levels (*IPCC, 2013*). In the recent years, several studies are carried out on the aerosol characterization and its direct effect over south Asia, but there have been very few studies reported on the aerosol indirect effect using ground- and satellite-based measurements due to complex aerosol and cloud environments. Therefore, we discuss the $RF_{aci}$ for both anthropogenic and natural fraction of aerosol for a period of six-years (2008-2013) for three different regions of south Asia (**Fig. 1,** Arabian Sea (AS; 63°E-72°E, 7°N-19°N), Bay of Bengal (BOB; 85°E-94°E, 7°N-19°N) and Central India (CI; 75°E-84°E, 20°N-30°N)), having significantly distinct aerosol environments as a result of variations in aerosol sources and transport pathways (*Cherian et al., 2013; Das et al., 2015; Tiwari et al., 2015*) Additionally, we also discuss the uncertainties of the results in the following sections.

## 2 Data

We combine measurements of aerosol, cloud and radiative properties to derive the top-of-the atmosphere (TOA) $RF_{aci}$ offor both anthropogenic and natural aerosols. Data acquired by sensorsMODerate Resolution Imaging Spectroradiometer (MODIS) and Clouds and the Earth's Radiant Energy System (CERES) mounted on Aqua (*Parkinson, 2003*) and Ozone Monitoring Instrument (OMI) onboard Aura (*Schoeberl et al., 2006*) are used in this study. We use the broadband shortwave planetary albedo (α) (*Wielicki et al., 1996; Loeb, 2004; Loeb et al., 2007*) as retrieved by the Clouds and the Earth's Radiant Energy System (CERES) in combination with cloud properties from the MODerate Resolution Imaging Spectroradiometer (MODIS; (*Minnis et al., 2003*) and AOD ($\tau_a$) and fine mode fraction (FMF) as retrieved by the MODIS onboard Aqua (*Remer et al., 2005*). Albedo and cloud properties are from the CERES Single-Scanner-Footprint (SSF) Level-2 Edition-3A data set at 20×20 $km^2$ horizontal resolution and aerosol properties (AOD and FMF) at 550nm from the MYD04 level-2 collection-5.1 dataset at 10×10 $km^2$ horizontal resolution are used. We used UV-aerosol index (UV-AI; *Torres et al., 1998*) measured by Ozone Monitoring Instrument (OMI; AURA (*Levelt et al., 2006*) onboard Aura from the OMAERUVG level-2 version 003 dataset at 0.25°×0.25° grid, which is a gridded dataset containing retrievals from the OMAERUV (*Torres et al., 2007*) algorithm. The data from CERES and MODIS level-2 products are interpolated to a 0.25°×0.25° regular longitude-latitude grid to separate the aerosol and cloud properties for anthropogenic and natural aerosols using UV-AI. Daily data, taken at roughly 13:30 local time, cover the 2008-2013 period.

## 3 Methods

All statistics between aerosol and cloud properties are computed separately for 3 regions and for each month of data at 0.25°×0.25° grid resolution. To avoid the greater uncertainty that exists in a clear distinction between aerosols and clouds and accurate retrieval of cloud properties, only single-layer cloud with liquid water path (LWP) > 20 $gm^{-2}$ are taken into account. $RF_{aci}$ for anthropogenic and natural aerosols are calculated using the methods outlined by *Quaas et al., (2008)* with the new statistical approach. As a part of this process, the method by *Kim et al., (2007)* MODIS-OMI algorithm (MOA) is employed to classify the aerosol types into one of four types sea-salt, carbonaceous, dust and sulfate using MODIS FMF and OMI UV-AI data. FMF provides information on the representative size of the aerosol. FMF is close to 1 for mostly small aerosol particles, which implies an anthropogenic origin and FMF becomes small for non-anthropogenic aerosol like dust. UV-AI allows to detect the absorption due to the presence of an aerosol layer by utilizing the sensitivity of absorptive aerosol in UV. Under most condition, UV-AI is positive for absorbing aerosols and negative for non-absorbing aerosols. Using these two independent data sets, aerosol can be classified. Details for the aerosol classification are discussed in *Kim et al., (2007)*. For the purpose of this research, the combination of dust and sea-salt AOD considered as a natural AOD and an anthropogenic AOD contains the combination of carbonaceous and sulfate. Further, the $RF_{aci}$ is estimated for both anthropogenic and natural aerosols.

### 3.1 Satellite-based estimate of $RF_{aci}$

$RF_{aci}$ is a function of the relationship between AOD and $N_d$ in a cloud. $N_d$ is not directly provided by satellite product and must be computed using cloud optical thickness ($\tau_c$) and effective droplet radius ($r_e$) for liquid water clouds assuming adiabaticity (*Brenguier et al., 2000*~~)~~.; *Schüller et al., 2005; Quaas et al., 2006; Bennartz, 2007; Rausch et al., 2010*).

$$N_d = \gamma \tau_c^{1/2} r_e^{-5/2} \qquad (2)$$

~~Where, $\gamma$=1.37×10$^{-5}$ m$^{-0.5}$ in this study.~~

Where, a constant value of $\gamma$=1.37×10$^{-5}$ m$^{-0.5}$ (*Quaas et al., 2006*) is used in this study. A limitation of this assumption is that it applies rather well for the stratiform clouds in the marine boundary layer, but less so for convective clouds. A detailed explanation and uncertainty assessment are described in *Bennartz, (2007) and Rausch et al., (2010)*. Recently, *Bennartz and Rausch, (2017)* show that the uncertainties in the CDNC climatology from 13-years of AQUA-MODIS observations are in the order of 30% in the stratocumulus regions and 60% to 80% elsewhere and its contribution to the total uncertainty for this study is discussed in the following section.

*Quaas et al., (2008)* ~~is~~have adopted the *Loeb (2004)* approach for the estimate of planetary albedo. Albedo ($\alpha$) of a cloud scene can be well described by a sigmoidal fit as

$$\alpha \approx (1-f)[a_1 + a_2 ln\tau_a] + f[a_3 + a_4(f\tau_c)^{a_5}]^{a_6} \qquad (3)$$

Where, $\alpha_1$- $\alpha_6$ are fitting parameters obtained by a multilinear regression, where $\alpha_5$ is set as 1~~.~~ (*Ma et al., 2014*). Dependency of $\tau_a$ is introduced to include the clear part of the scene in the above equation and f is the cloud fraction. The satellite-based estimate of $RF_{aci}$ for anthropogenic and natural aerosols can be expressed as

$$\Delta F_{ant/nat}^{RF_{aci}} = f_{liq}.A(f,\tau_c)\frac{1}{3}\frac{d\ln N_d}{d\ln \tau_a}\left[\ln\tau_a - \ln(\tau_a - \tau_a^{ant/nat})\right] S \qquad (4)$$

$$where, A(f,\tau_c) = a_4 a_5 a_6 \left[a_3 + a_4(f\tau_c)^{a_5}\right]^{a_6-1}(f\tau_c)^{a_5}$$

$d \ln N_d / d \ln \tau_a$ is the sensitivity of cloud droplet number concentration ($N_d$) to a relative change in AOD. It is computed as the slope of the linear regression fit between the natural logarithm of $N_d$ and AOD (*Quaas et al., 2008*). This value is calculated on a month-by-month basis and is unique to each region studied. $\tau_a$ is the total AOD, whereas, $\tau_a^{ant/nat}$ are the anthropogenic and natural AOD, respectively, derived from the FMF and UV-AI as estimated above. $A(f, \tau_c)$ is the empirical function relating albedo to f and $\tau_c$. S is the daily mean solar incoming solar radiation. $RF_{aci}$ is calculated separately for the anthropogenic and natural aerosols for all three regions for each month. ~~$A(f, \tau_c)$ is the empirical function relating albedo to f and $\tau_c$. $\tau_a^{ant/nat}$ is aerosol optical depth for anthropogenic and natural aerosol, respectively, S is the daily mean solar incoming solar radiation.~~

A goal of the present study is to assess the uncertainty in the satellite-based estimate of the $RF_{aci}$. For that purpose, we adopted the new statistical nonlinear least square fitting approach to obtain the six fitting parameters in Eq. (3). ~~Instead~~Nonlinear least square methods involve an iterative improvement to parameters values in order to minimize the residual sum of squares between the observed values and the predicated value of the dependent variables. We used the Levenberg-Marquardt (L-M) algorithm (*Levenberg, 1944*) in the nonlinear least square approach to adjust the parameter values in the iterative procedure. This algorithm combines the Gauss-Newton method and the gradient descent method. In the gradient descent method, the sum of the squared errors is reduced by updating the parameters in the steepest descent direction. In the Gauss -Newton method, the sum of the squared errors is reduced by assuming the least squares function is locally quadratic, and finding the minimum of the quadratic. The L-M algorithm acts more like a gradient descent method when the parameters are far from the optimal value and acts more like to Gauss-Newton method when the parameters are close to their optimal value. More detail of this method is given in the literature (*Levenberg, 1944; Transtrum et al., 2010; Transtrum and Sethna, 2012*). In the present study, instead of considering $\alpha_5$=1 in the multiple regression, as in *Quaas et al. (2008) and Ma et al., (2014)*, we obtained the values of all six fitting parameters using a nonlinear fitting approach (L-M algorithm) for each month and region. To get an impression of the performance of our statistical approach, we correlate α and $RF_{aci}$ at TOA obtained from both statistical fitting methods (multilinear and nonlinear) vs. α and $RF_{aci}$ simulated by radiative transfer model for all three regions. The following section describes the detail information about the simulation of α and $RF_{aci}$ using the radiative transfer model.

### 3.2 Simulation of planetary albedo (α) and $RF_{aci}$

In order to verify both the statistical approaches, we performed a radiative transfer simulation to obtain α and $RF_{aci}$ for all three regions. Radiative transfer calculations are performed with the SBDART [Santa Barbara DISORT Atmospheric Radiative Transfer; *Ricchiazzi et al., 1998*] that is a plane-parallel radiative transfer code based on the DISORT algorithm for discrete-ordinate-method radiative transfer in multiple scattering and emitting layered media (*Stamnes et al., 1988*). The discrete ordinate method provides a numerically stable algorithm to solve the equations of plane-parallel radiative transfer in a vertically inhomogeneous atmosphere. Simulations are carried out for the solar spectrum (0.2-4.0μm) for all three regions. Following the study by *Quaas et al., (2008)* study, *Bellouin et al., (2013)* performed a similar study with MACC reanalysis data, in which RT simulations, using a Monte -Carlo method, were carried out to obtain the standard deviation for the uncertainty analysis. However, in the present study, RFaci is simulated using an RT model (SBDART) to validate the performance of both the statistical approaches used to compute the $RF_{aci}$ using the statistical relationship between satellite measurements.

In the present study, simulations are carried out to simulate first α and later $RF_{aci}$ for the given inputs. Here α is evaluated as the ratio of broadband outgoing (or upwelling) shortwave flux to the incoming (or downwelling) solar flux. Inputs to the model include profiles of temperature and water vapor which are resolved into 32 layers extending from 1000 to 1 mbar and come from European Centre for Medium-range Weather Forecast (ECMWF) reanalysis data. **Table 1** shows the list of input parameters and their source provided to the RT model for the estimate of $RF_{aci}$. Total columnar amount of atmospheric ozone is provided from OMI-AURA. Surface albedo is set to 0.15 to represent a typical land cover value for CI and, predefined option of the ocean surface is used for the oceanic regions (AS and BOB). In the SBDART model, the cloud parameter inputs are effective droplet radius ($r_e$), liquid water path (LWP) and the cloud fraction, all of which are taken from MODIS retrievals reported in the CERES-SSF product. The geometrical thickness of cloud (CGT) is computed as a difference between cloud top and bottom heights. Cloud top height is taken from CERES-SSF product and cloud base height is evaluated using the geopotential height profile from ECMWF data. Only liquid water clouds are considered in the estimation of $RF_{aci}$. The upwelling and downwelling fluxes are computed individually computed for all three regions at satellite (MODIS-Aqua as a reference) overpass time.

The local radiative forcing associated with the $RF_{aci}$ is estimated as the difference between the perturbed and unperturbed radiative fluxes caused by perturbation in $N_d$ due to the addition of aerosols while keeping the same meteorology. $RF_{aci}$ is diagnosed by making two calls to the radiative transfer code: the first call used the unperturbed satellite-derived $N_d$ and the second used perturbed $N_d$ due to anthropogenic and natural aerosols. The numerical evaluation of radiative flux for the perturbed case starts by determining the finite perturbation of cloud droplet number concentration ($\Delta N_d$), calculated as follows:

$$\Delta N_d^{ant/nat} = \frac{d\ln N_d}{d\ln \tau_a}\,[\ln\tau_a - \ln(\tau_a - \tau_a^{ant/nat})] \qquad (5)$$

The finite perturbation in $N_d$ are evaluated separately for anthropogenic and natural aerosol to estimate the radiative flux for the perturbed case. The perturbed value of ~~$\acute{N}_d$~~ $N'_d$ ($N_d + \Delta N_d$) is used to obtain a perturbed value of $r_e$ using Eq. (5) for constant liquid water content because $r_e$ is used as an input to the radiative transfer code.

~~$\acute{N}_d = q_l/(\frac{4}{3}$~~
$$N'_d = q_l/(\frac{4}{3}\pi {r_e}^3\rho_w) \qquad (6)$$

Where, $\rho_w$ is the liquid water density, $q_l$ the liquid water content ($q_l$=liquid water path / geometrical thickness). $RF_{aci}$ is diagnosed as $RF_{unperturbed}$ - $RF_{perturbed}$ radiative fluxes at the top of the atmosphere, because increased concentrations of aerosol reduce the effective radius of cloud particles and smaller cloud particles reflect more radiation back to space. The following section describes the details of regression analysis of α and $RF_{aci}$ performed between values from statistical-approaches and simulated values.

# 4 Results

## 4.1 Regression analysis

As stated in section 3.1, the satellite-based estimates of $RF_{aci}$ are dependent on the fitting parameters $\alpha_1$-$\alpha_6$, obtained here from the two different statistical fitting approaches (multilinear and nonlinear). The parameters obtained from these two approaches are listed in **Table- S1** for all three regions investigated in this study. These parameters vary with months since we conducted both the fitting approaches for each month, but only the mean seasonal parameters are shown here. The main differences in fitting parameters from both methods are found in the values of $\alpha_4$, $\alpha_5$ and $\alpha_6$. The ~~weight given to~~magnitude of the coefficients $\alpha_4$ and $\alpha_6$ is larger in the nonlinear fit ~~is larger~~ than ~~for~~ the multilinear regression fitting, which may reduce the ~~weight~~magnitude of the coefficient $\alpha_5$.

To accomplish the objective of this study, we correlate α and $RF_{aci}$ at TOA obtained from both statistical fitting approaches (multilinear and nonlinear) with estimates obtained from radiative transfer model for all three regions. **Fig. ~~1~~2** shows scatter density plots of comparison between model-simulated albedo and the one computed from satellite measurements at 0.25°×0.25° grid resolution using both statistical methods for all three regions. This regression analysis suggests that the albedo fitted by the new statistical approach (nonlinear fit) agrees well with the model-simulated albedo over both land and ocean. The scatter of the results from the nonlinear fit around the 1:1 line is much smaller compared to multilinear fit, which is also reflected in the coefficients of determination ($R^2$) ranging from 0.74 to 0.79. However, a reduction in over and underestimation at very large and very small albedos, respectively, is found in the nonlinear fit compared to the multilinear statistical approach. This is also clearly reflected in the values for the root mean square error (RMSE), which reduces from 0.042-0.065 to 0.010-0.017, supporting the expectation that the new statistical method is more reliable. Additionally, a comparison between the planetary albedo computed using both statistical fits and the CERES retrieved albedo is shown in **Fig. ~~S3~~S1** for all three regions. Similar to the results discussed above, the analysis shows a good agreement between the CERES derived albedo and the one calculated using the nonlinear fit.

In addition, we performed a comparison of $RF_{aci}$ obtained from satellite measurements using both statistical approaches with the one simulated by SBDART for each season and for each region. **Fig. ~~2~~3** illustrates the linear regression of $RF_{aci}$ from the two statistical approaches plotted against the one obtained from the radiative transfer model for both anthropogenic and natural aerosols for all seasons and all three regions. The analysis showed ~~satisfactory results~~good statistical agreement with Pearson's correlation coefficient r=0.82 and 0.75 and RMSE=0.037 $Wm^{-2}$ and 0.042 $Wm^{-2}$ for the anthropogenic and natural fraction of aerosols, respectively. An examination of **Fig.** ~~2~~**3** reveals that the nonlinear fitting approach reduces the scatter seen for the multilinear fit and the improvement in correlation with the simulated forcing. ~~Using the~~The nonlinear fit increases the correlation ~~coefficient~~ by 21%-23% and ~~decreases~~reduce the RMSE by ~~from~~ 0.007~~ $Wm^{-2}$ to~~ -0.011 ~~Wm~~W $m^{-2}$ compared to the multilinear approach. The relative difference between the RT-simulated and the statistically computed $RF_{aci}$ are computed for both the statistical methods. The mean relative difference in $RF_{aci}$ for anthropogenic fraction of AOD is 0.021 W $m^{-2}$ in the nonlinear and 0.033 W $m^{-2}$ in the multilinear statistical approach, whereas, for $RF_{aci}$ of natural fraction of AOD, it is 0.032 W $m^{-2}$ in nonlinear and 0.053 W $m^{-2}$ in multilinear statistical approach. This suggests that the use of the nonlinear fitting approach reduces the uncertainty by 36%-39% compared to the multilinear regression.

### 4.2 $RF_{aci}$ and Uncertainties

~~Aerosol~~Aerosols and clouds vary substantially as a function of time in all regions; thus~~,~~ it is interesting to ~~analyze~~analyses aerosol-cloud interactions as a function of season. **Fig.** ~~3~~**4** shows the seasonal variability of six-year averaged radiative forcing by aerosol-cloud interaction for the three regions as defined above. The maximum anthropogenic $RF_{aci}$ is found over oceanic regions (AS: -0.15$Wm^{-2}$, BOB: -0.16$Wm^{-2}$), instead of regions over land (CI: -0.12 $Wm^{-2}$) with high anthropogenic emissions. This is because maritime clouds are more susceptible to changes in concentration of anthropogenic aerosols (*Quaas et al., 2008*). In contrast, the natural $RF_{aci}$ is generally stronger over land (-0.15 $Wm^{-2}$) than over oceanic regions (AS: -0.098 $Wm^{-2}$, BOB: -0.07$Wm^{-2}$). It is seen that the anthropogenic $RF_{aci}$ is strongest during winter over AS and BOB, with values near -0.19 $Wm^{-2}$ and -0.22$Wm^{-2}$, whereas it is strong (-0.2 $Wm^{-2}$) during pre-monsoon over CI (land). The dominance of natural aerosols in pre-monsoon results a large natural $RF_{aci}$ both over land (-0.15 $Wm^{-2}$) and ocean (-0.098 $Wm^{-2}$ and -0.07 $Wm^{-2}$).

~~It~~A direct comparison of the satellite to simulations-based $RF_{aci}$, shows a good correlation. However, both satellite estimated and simulated $RF_{aci}$ are subject to errors and it is useful to compute the associated uncertainties in the above results due to various parameters. Uncertainty involves the ones due to satellite retrievals of AOD which can be highly biased in the vicinity of cloud due to swelling (*Koren et al., 2007*), and also due to 3D effects (*Wen et al., 2007*). Since both biases may be particularly high for thick clouds, our estimate of the $RF_{aci}$ could be still be overestimated. The uncertainty in MODIS retrievals of AOD from validation studies (*Levy et al., 2007*) was quantified at $0.03+0.05\tau_a$ over ocean and $0.05+0.15\tau_a$ over land. However, since we use the MODIS-OMI algorithm (*Kim et al., 2007*) to estimate the anthropogenic and natural fraction of AOD, uncertainty in this is given as 1σ standard deviations as per Table~~-~~ S2. From satellite intercomparison, the uncertainty in radiative flux retrievals by CERES is estimated at 5% (*Loeb, 2004*), and uncertainty in cloud optical depth is 21% (*Minnis et al., 2004*). ~~The computed $RF_{aci}$ in this study is closely~~The uncertainties due to sensitivity of $N_d$ to a relative change in AOD ($d \ln N_d / d \ln \tau_a$) contribute most to the total uncertainty. For $N_d$ sensitivities to changes in AOD, standard deviations are derived from minimum and maximum values obtained for each season. Following the study by *Bellouin et al., (2013)*, the standard deviations are derived from minimum and maximum values by defining 4-sigma interval, which covers the large range of sensitivities and spatio-temporal variabilities. To define the standard deviations in $RF_{aci}$ due to variation in $d \ln N_d / d \ln \tau_a$, $RF_{aci}$ is recomputed using those standard deviations of $N_d$ sensitivities to changes in AOD. Table 2 shows the seasonal and regional sensitivities of $d \ln N_d / d \ln \tau_a$ along with their statistical standard deviation, which is computed from the minimum and maximum values for each season. The associated range in $RF_{aci}$ both for anthropogenic and natural fraction of AOD is also shown in Table 2, where the standard deviation of $RF_{aci}$ shows the variation due to change in d ln $N_d$ / d ln $\tau_a$, which finally contribute to the total uncertainty. In addition to this, the computed $RF_{aci}$ in this study is associated with the statistical fitting approach as described in section 3. As mentioned earlier, two different statistical fitting methods are used to obtain the regression coefficients for the estimates of $RF_{aci}$. The study showed that the new nonlinear fitting approach reduces by ~20%-25% the uncertainty from the statistical relationship and fitting parameters. The propagation of errorestimate of $RF_{aci}$. In the present study, except for the statistical fitting method, all the variables and methodologies are same for both the statistical approach. Therefore, we used the relative difference between the RT-simulated and statistically computed $RF_{aci}$ as an uncertainty due to the choice of the statistical fitting approach for both the statistical fitting methods. As shown in section 4.1, the mean relative differences for the nonlinear and multilinear approaches are 0.021 W $m^{-2}$ and 0.033 W $m^{-2}$, respectively, in $RF_{aci}$ for anthropogenic fraction, whereas, for the $RF_{aci}$ of the natural fraction of AOD, these are 0.032 W $m^{-2}$ and 0.053 W $m^{-2}$ for nonlinear multilinear statistical approaches, respectively. Table 3 lists the uncertainty due to different parameters involved in the satellite-based estimate of $RF_{aci}$. We quantify the relative error as the square root of the sum of the squared relative errors for all individual contributions. This yields an influence of these relative uncertainties in the input quantities on the computed $RF_{aci}$ of ~±0.08W$m^{-2}$. It should be noted that we refer here to the published quantifiable uncertainties in the satellite retrievals. Limitation involvesinvolved in this approach or uncertainties in the satellite measurementsretrievals contribute to the overall uncertainty but cannot be quantified, which is difficult to quantify.

# 1 Conclusion

In addition to above error budget, there are uncertainties involved in the RT simulated $RF_{aci}$ due to various parameters as shown above. In this regard, the surface albedo plays a major role in the simulation of $RF_{aci}$. In the standard approach, we have considered a surface albedo value 0.15 for land and the predefined option for the ocean surface albedo is used for the oceanic regions in the present study. To quantify the uncertainties involved due to assumptions about the surface albedo, we have simulated $RF_{aci}$ with different plausible surface albedo values and computed statistics as shown in **Table S3(a) and S3(b)**. The statistics shows that the considered values of surface albedo are suitably representative of the study regions. In addition, RT simulation have their own limitations and uncertainties e.g. inherent code accuracy, overestimate in calculated RF due to plane-parallel bias, 3-D radiative transfer effect etc. It would be useful to explore these issues in the future. However, in the present study, RT simulation is used to evaluate the results computed with satellite- based measurements. There is a scope to improve the present study with the upcoming data set retrieved from spaceborne active remote sensing instruments, with the improved satellite products and with the new statistical relationship.

# 5 Conclusions

In this study, we employed a new nonlinear statistical fitting approach to develop the statistical relationship. A satellite-based algorithm is used to quantify the anthropogenic and natural fraction of aerosol optical depth for the computation of $RF_{aci}$ from satellite retrievals. In order to verify, α and $RF_{aci}$ estimates using the new statistical approach (nonlinear) along with the previous statistical approach (multilinear fit), these are compared with the results obtained from radiative transfer simulations. The results show a better agreement between model-based estimates and the one estimated using the nonlinear approach compared to the multilinear approach. The nonlinear approach relatively increases by 21%-23% the correlation coefficient and decreasesreduce RMSE by 0.007W$m^{-2}$ to 0.011

$Wm^{-2}$ ~~the RMSE~~ compared to multilinear approach. The nonlinear fitting approach reduces the relative difference by 36%-39% compare to the multilinear regression method. The $RF_{aci}$ is found to be consistent with the value found by statistical relationship between aerosol and cloud properties from MODIS and CERES, respectively, and radiative transfer calculations. Further studies using the data retrieved from ~~active remote sensing~~advanced instruments ~~(~~e.g. lidar and radar~~)~~ may be useful to test the assumption made in the present study concerning the proxy of aerosol column, the overestimation of AOD over land and deal with the multi-layer clouds.


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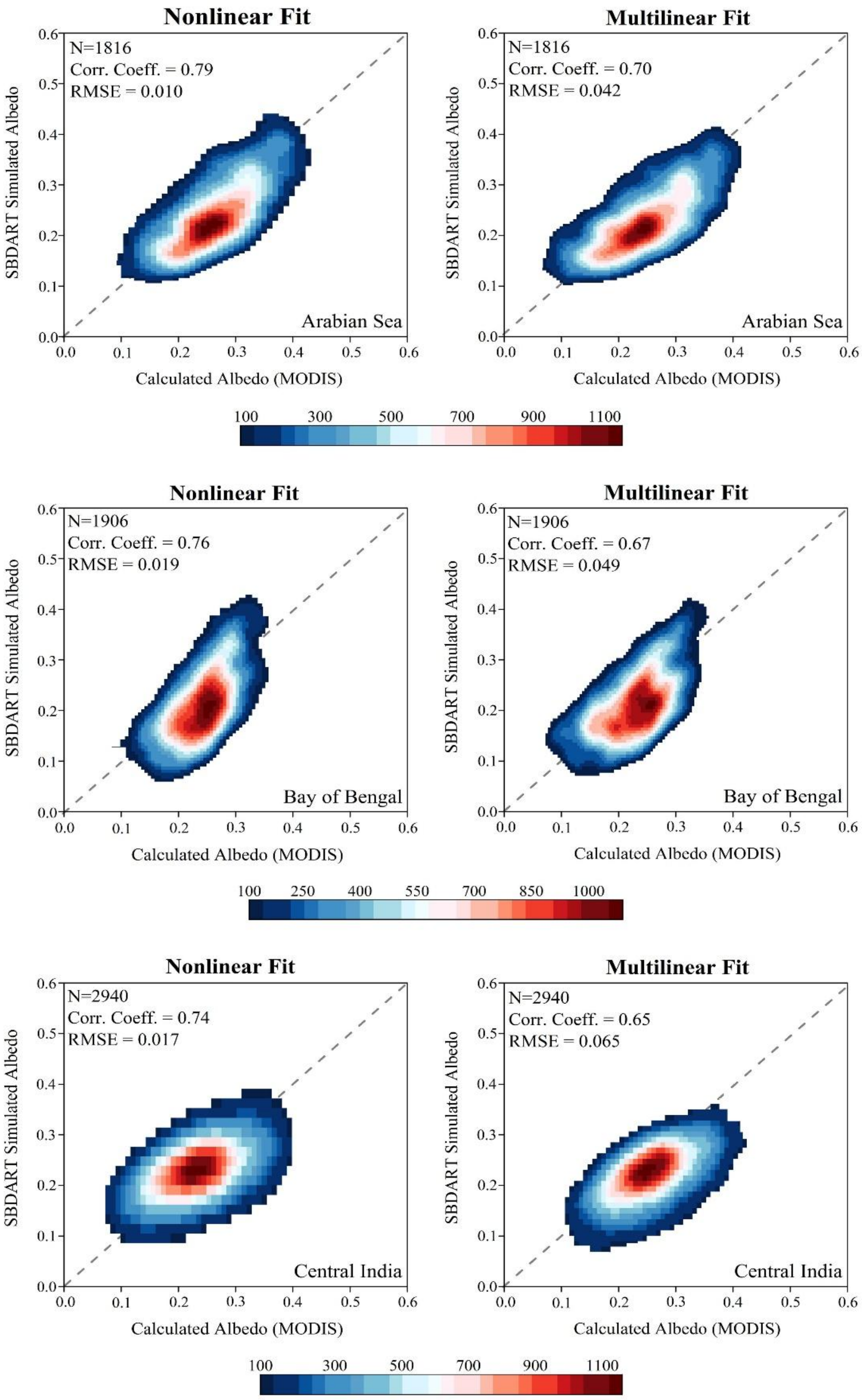



~~Figure 1.~~

**Table 1**: The list of parameters and their sources used as an input to the SDBART model for the simulation of $RF_{aci}$.

| Input parameters | Source |
|---|---|
| Temperature and Water vapor (for 32 layers extending from 1000 to 1 hPa) | ECMWF reanalysis |
| Total Columnar ozone | OMI-AURA |
| Surface Albedo | For land - 0.15<br>For ocean - default value of “ocean” (given in SBDART) |
| cloud effective droplet radius<br>cloud liquid water path<br>cloud fraction | MODIS retrievals reported in CERES-SSF product |
| geometrical thickness of cloud | Computed from MODIS and ECMWF data |

**Table 2:** Seasonal and regional sensitivities d ln $N_d$/d ln $\tau_a$ of cloud droplet number concentration $N_d$ to changes in aerosol optical depth used in this study. The given standard deviation is derived from minimum and maximum values for a particular season. The associated range in $RF_{aci}$ is also estimated where the standard deviation of $RF_{aci}$ shows the variation due to change in d ln Nd / d ln $\tau_a$.

| | Region | Winter | Pre-Monsoon | Monsoon | Post-Monsoon |
|---|---|---|---|---|---|
| $\frac{d \ln N_d}{d \ln \tau}$ | AS | 0.384 ± 0.146 | 0.408 ± 0.189 | 0.272 ± 0.131 | 0.18 ± 0.102 |
| | BOB | 0.314 ± 0.136 | 0.414 ± 0.15 | 0.194 ± 0.104 | 0.148 ± 0.088 |
| | CI | 0.214 ± 0.107 | 0.178 ± 0.105 | 0.107 ± 0.069 | 0.122 ± 0.071 |
| $Rf_{aci}$ for Anthrophonic Fraction | AS | -0.19 ± 0.036 | -0.14 ± 0.056 | -0.08 ± 0.036 | -0.16 ± 0.036 |
| | BOB | -0.22 ± 0.062 | -0.16 ± 0.036 | -0.07 ± 0.02 | -0.2 ± 0.036 |
| | CI | -0.13 ± 0.02 | -0.2 ± 0.036 | -0.05 ± 0.034 | -0.16 ± 0.036 |
| $Rf_{aci}$ for Natural Fraction | AS | -0.12 ± 0.036 | -0.18 ± 0.036 | -0.03 ± 0.04 | -0.06 ± 0.036 |
| | BOB | -0.08 ± 0.026 | -0.11 ± 0.026 | -0.04 ± 0.039 | -0.06 ± 0.017 |
| | CI | -0.16 ± 0.027 | -0.22 ± 0.055 | -0.1 ± 0.027 | -0.14 ± 0.036 |

**Table 3:** Lists the sources of uncertainties and their values involved in the satellite-based estimate of $RF_{aci}$ in the present study.

| **Source of uncertainty** | **Values** |
|---|---|
| Total AOD | $0.03\pm0.05.\tau_a$ over ocean<br>$0.05\pm0.05.\tau_a$ over land |
| MODIS-OMI algorithm (for the estimate of anthropogenic and natural fraction of aerosol) | 1σ standard deviation as per below table-S2 |
| Flux retrieval from CERES | 5% |
| Cloud optical depth retrieval from CERES | 21% |
| Cloud droplet number concentration | See table-2 |
| Statistical fitting approach | 0.021 W $m^{-2}$ in nonlinear for anthropogenic<br>0.032 W $m^{-2}$ in nonlinear for natural<br>0.033 W $m^{-2}$ in multilinear for anthropogenic<br>0.053 W $m^{-2}$ in multilinear for natural |

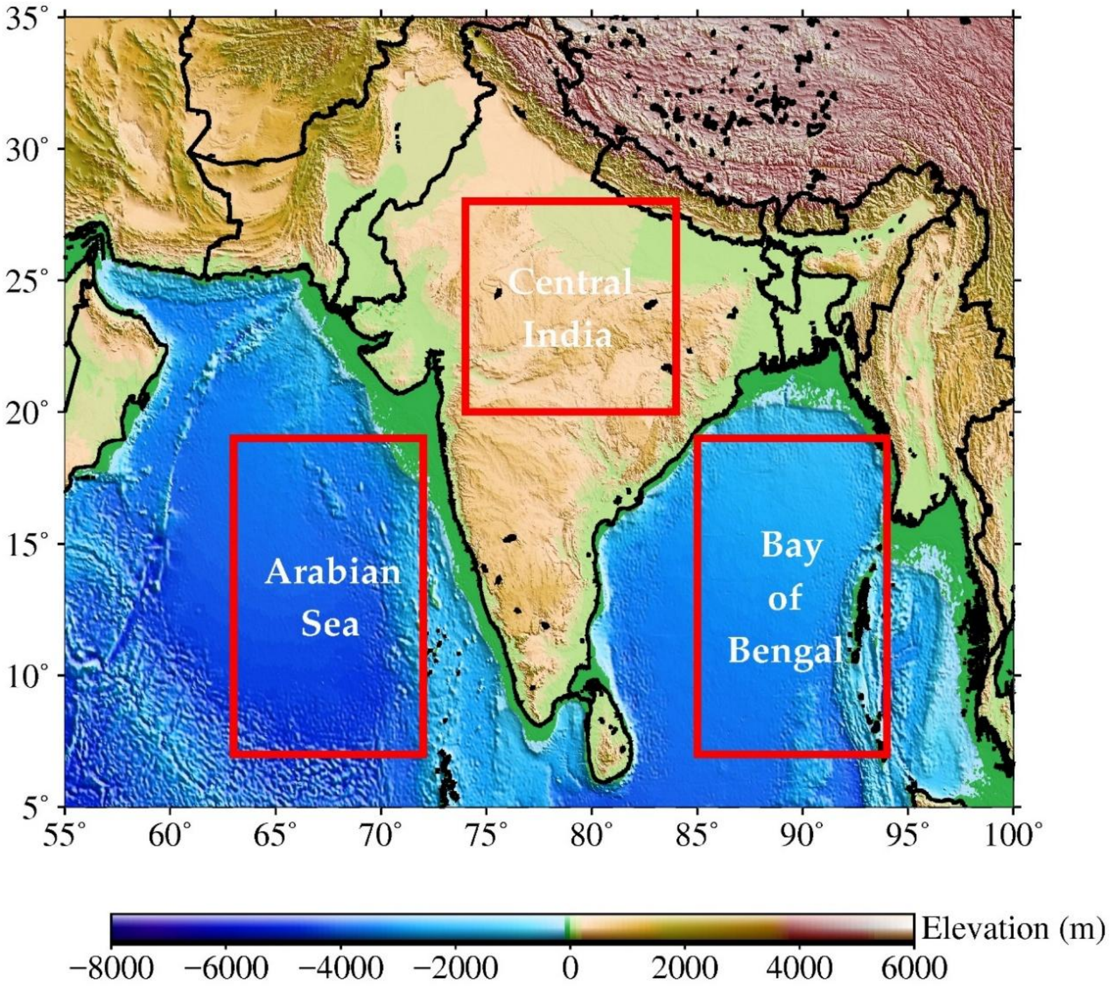



**Figure 1**: Map of India and surroundings showing the study regions. The regions covered by red box represent the study locations (Arabian Sea, Bay of Bengal and Central India).

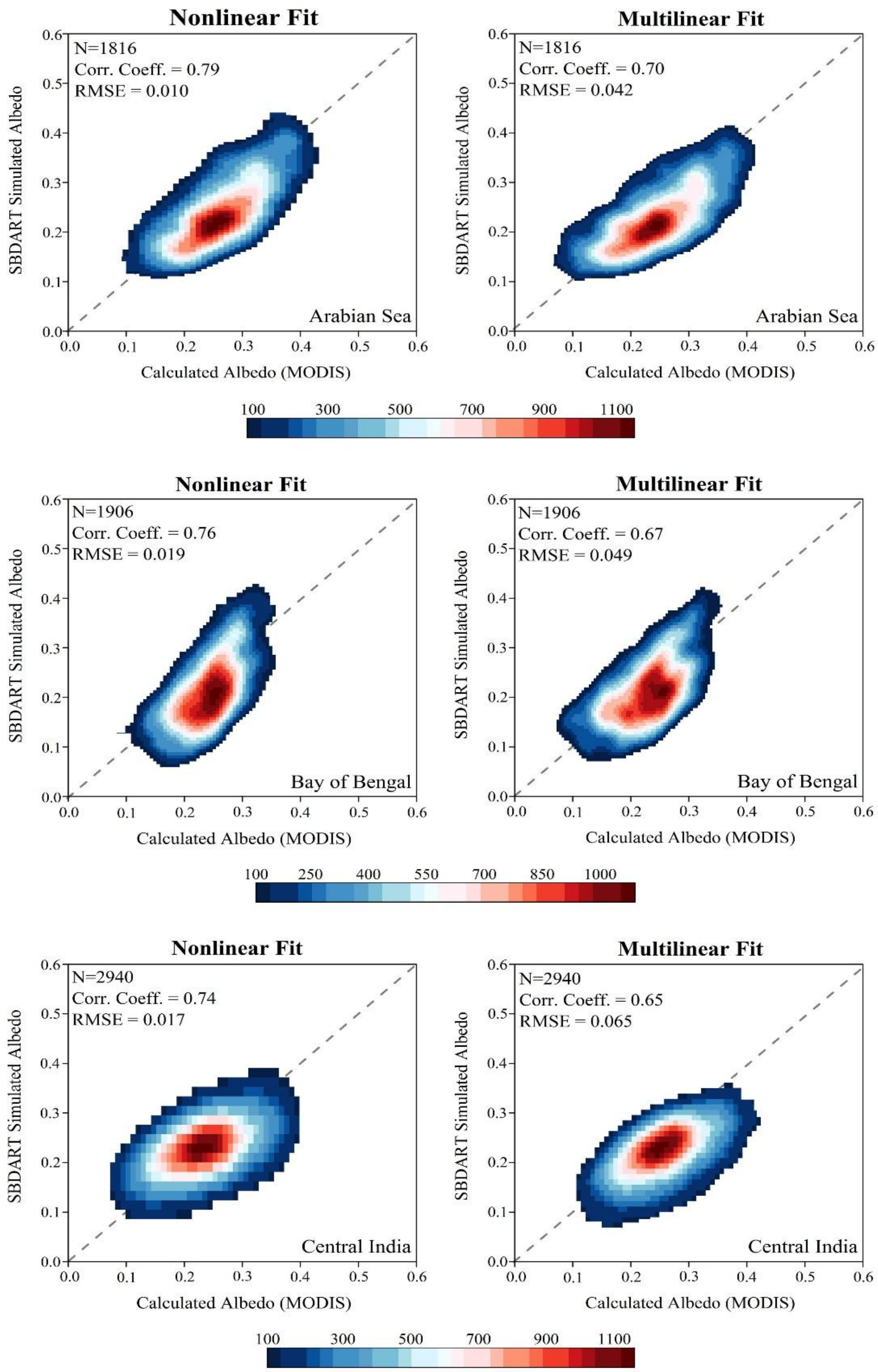
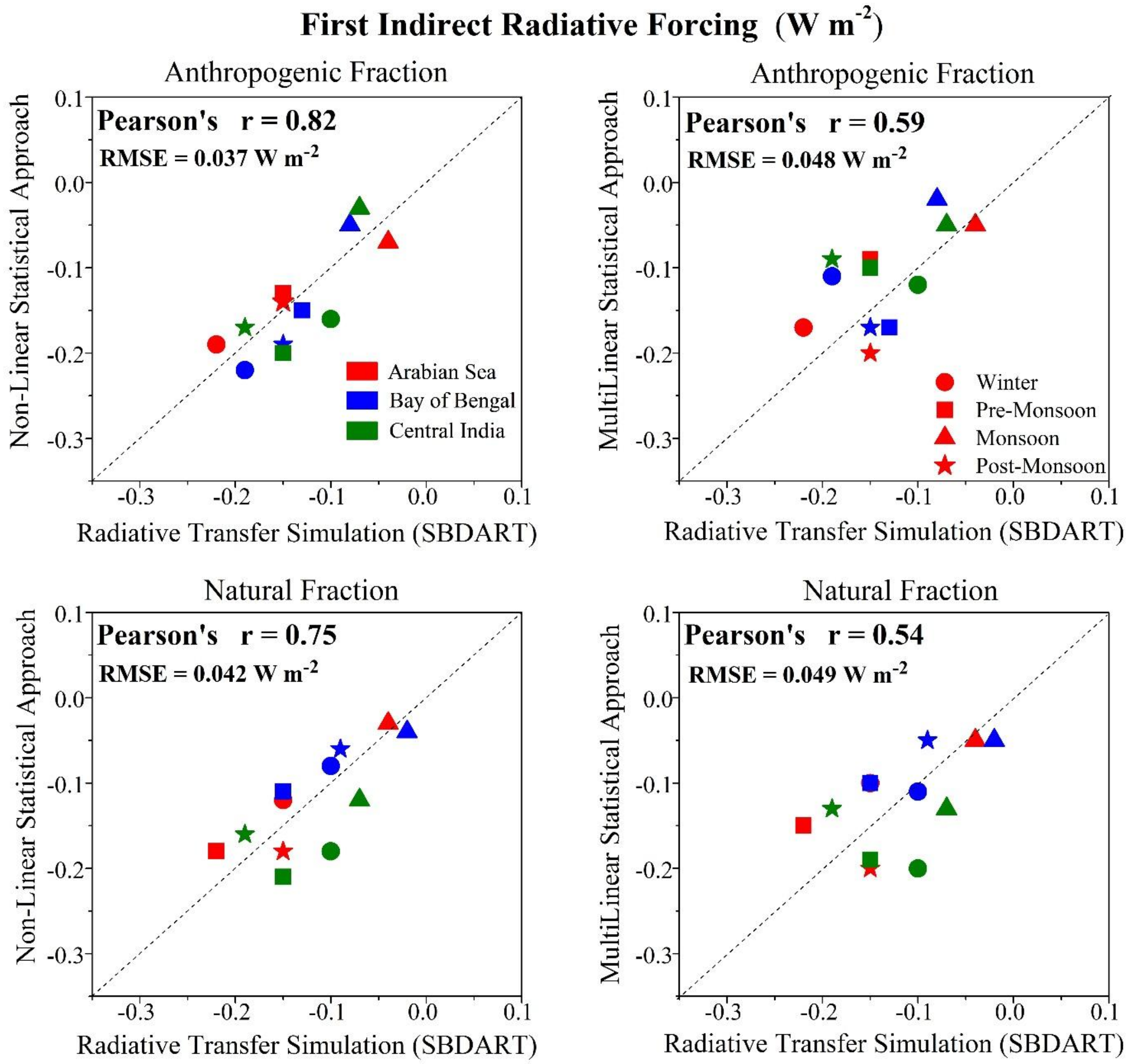



**Figure 2:** Scatter density plots of model-simulated albedo and the one computed using both statistical fitting method (nonlinear and multilinear fit) using satellite measurements for all three regions.


First Indirect Radiative Forcing (W m-2)
Anthropogenic Fraction
Pearson's r = 0.82
RMSE = 0.037 W m-2
Non-Linear Statistical Approach
Radiative Transfer Simulation (SBDART)
Arabian Sea
Bay of Bengal
Central India
Anthropogenic Fraction
Pearson's r = 0.59
RMSE = 0.048 W m-2
MultiLinear Statistical Approach
Radiative Transfer Simulation (SBDART)
Winter
Pre-Monsoon
Monsoon
Post-Monsoon
Natural Fraction
Pearson's r = 0.75
RMSE = 0.042 W m-2
Non-Linear Statistical Approach
Radiative Transfer Simulation (SBDART)
Natural Fraction
Pearson's r = 0.54
RMSE = 0.049 W m-2
MultiLinear Statistical Approach
Radiative Transfer Simulation (SBDART)

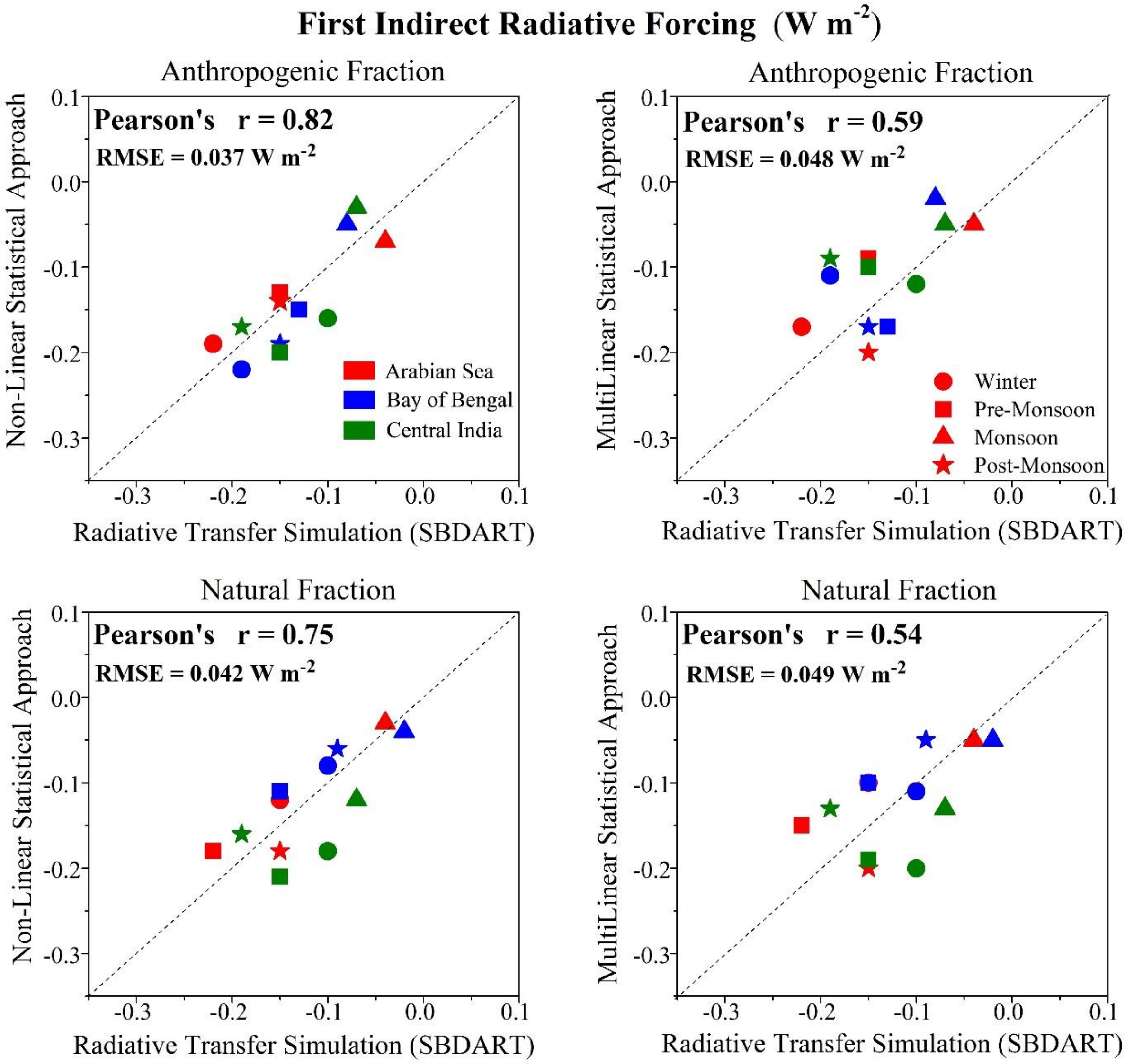



**Figure 3:** Comparison between satellite-based RFaci using both statistical fits and the one simulated by the SBDART model for all three regions and for all seasons. The different color indicates the regions, whereas the different symbols indicates the different seasons. Note that the fit is separately performed for each season and each region.

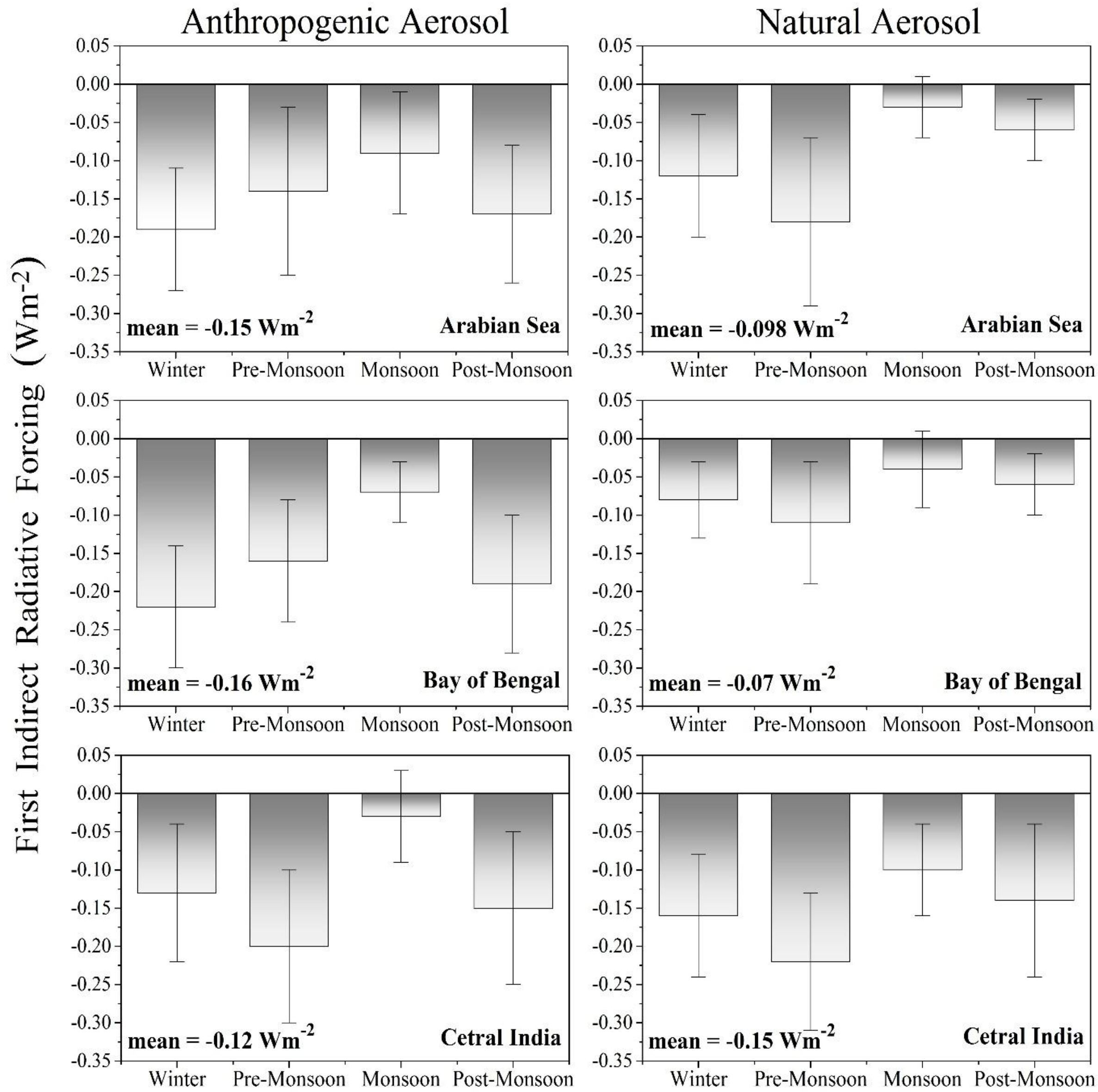



***Figure 4*.:** Seasonal variability of six-year averaged RF*aci* obtained using the nonlinear fit for all three regions for both anthropogenic and natural aerosols along with mean values.

**Table S4.:** The seasonal and regional variation of fitting parameters α1-α6 obtained from both multilinear and nonlinear fitting approaches.

| Area | Season | Nonlinear fit | | | | | | Multilinear regression fit | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | α1 | α2 | α3 | α4 | α5 | α6 | α1 | α2 | α3 | α4 | α5 | α6 |
| Arabian Sea | Winter | 0.158 | 0.023 | 0.376 | 0.232 | 0.199 | 0.943 | 0.098 | 0.101 | 0.002 | 0.005 | 1.000 | 0.303 |
| | Pre-Monsoon | 0.136 | 0.021 | -0.046 | 0.205 | 0.201 | 0.888 | 0.089 | 0.067 | 0.008 | 0.010 | 1.000 | 0.416 |
| | Monsoon | 0.109 | 0.029 | -0.046 | 0.395 | 0.201 | 0.888 | 0.092 | 0.049 | 0.009 | 0.012 | 1.000 | 0.422 |
| | Post-Monsoon | 0.154 | 0.026 | 0.108 | 0.010 | 1.192 | 0.172 | 0.091 | 0.097 | 0.044 | 0.024 | 1.000 | 0.558 |
| Bay of Bengal | Winter | 0.158 | 0.024 | -0.084 | 0.209 | 0.140 | 0.652 | 0.100 | 0.084 | 0.345 | 0.088 | 1.000 | 0.136 |
| | Pre-Monsoon | 0.127 | 0.012 | -0.043 | 0.081 | 0.081 | 0.474 | 0.092 | 0.060 | 0.004 | 0.002 | 1.000 | 0.324 |
| | Monsoon | 0.126 | 0.011 | -0.398 | 0.415 | 0.006 | 0.473 | 0.095 | 0.046 | 0.011 | 0.005 | 1.000 | 0.414 |
| | Post-Monsoon | 0.150 | 0.020 | 0.331 | 0.311 | 0.119 | 1.097 | 0.100 | 0.071 | 0.014 | 0.007 | 1.000 | 0.364 |
| Central India | Winter | 0.215 | 0.010 | -0.278 | 0.685 | 0.105 | 1.236 | 0.187 | 0.026 | 0.079 | 0.084 | 1.000 | 0.269 |
| | Pre-Monsoon | 0.183 | 0.003 | -0.259 | 0.662 | 0.099 | 1.339 | 0.171 | 0.018 | 0.020 | 0.009 | 1.000 | 0.278 |
| | Monsoon | 0.187 | 0.007 | 0.322 | 0.390 | 0.098 | 0.555 | 0.168 | 0.024 | 0.002 | 0.005 | 1.000 | 0.319 |
| | Post-Monsoon | 0.210 | 0.016 | -0.291 | 0.684 | 0.105 | 1.444 | 0.174 | 0.042 | 0.000 | 0.005 | 1.000 | 0.253 |

**Table S2**: Mean seasonal variation of anthropogenic and natural fraction of aerosol optical depth over all three regions estimated using methodology by *Kim et al., (2007)*.

| | Arabian Sea (AS) | | Bay of Bengal (BOB) | | Central India (CI) | |
|---|---|---|---|---|---|---|
| | Anthro* | Nat* | Anthro | Nat | Anthro | Nat |
| Winter | 0.146±0.029 | 0.112±0.022 | 0.16±0.032 | 0.142±0.028 | 0.376±0.055 | 0.279±0.056 |
| Pre-Monsoon | 0.305±0.061 | 0.347±0.069 | 0.309±0.062 | 0.327±0.065 | 0.407±0.081 | 0.46±0.092 |
| Monsoon | 0.113±0.023 | 0.346±0.069 | 0.16±0.032 | 0.276±0.055 | 0.435±0.087 | 0.655±0.101 |
| Post-Monsoon | 0.309±0.062 | 0.298±0.059 | 0.305±0.061 | 0.234±0.046 | 0.627±0.095 | 0.505±0.089 |

*Anthro represents anthropogenic and Nat represents natural

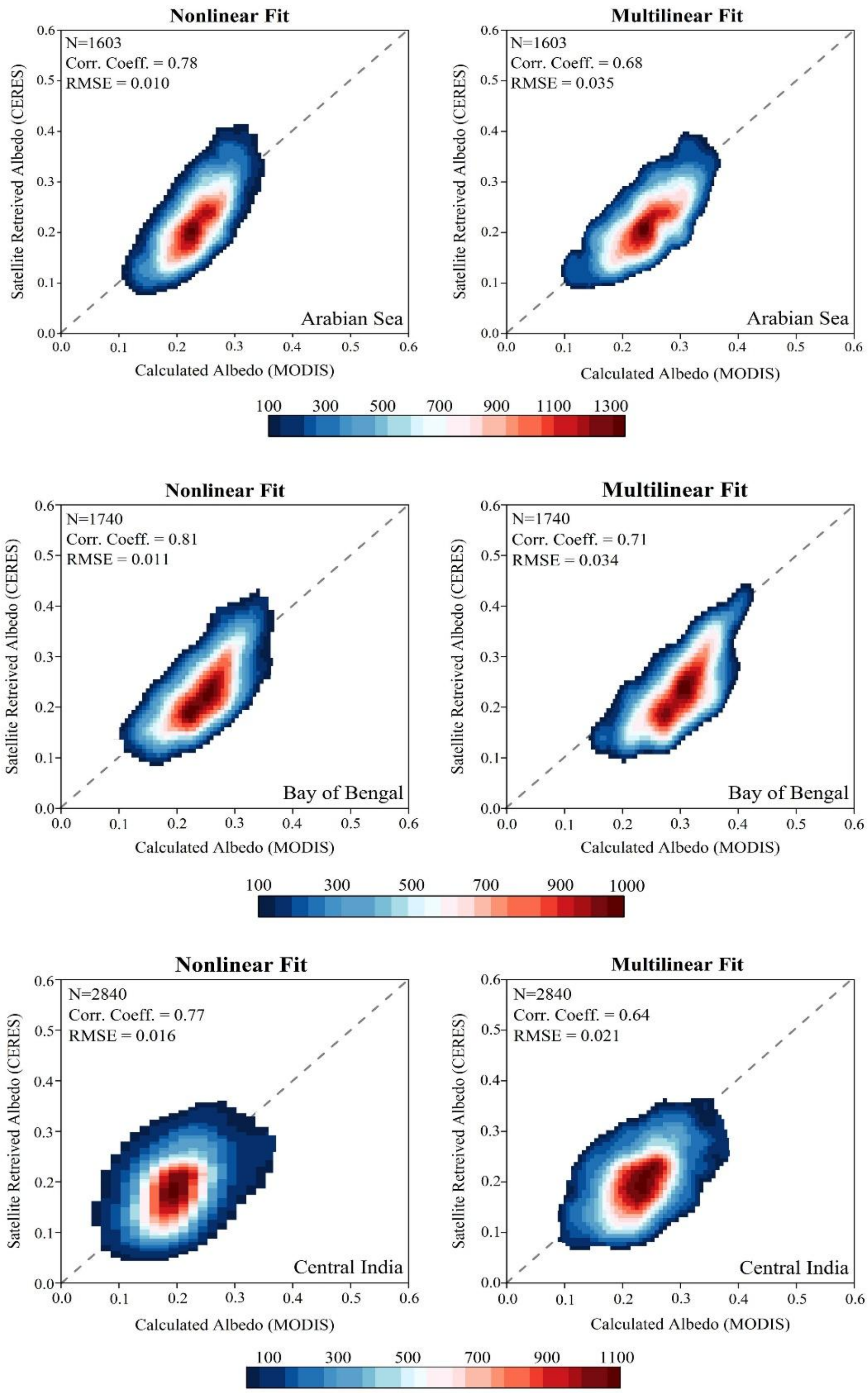



**Table S3(a):** The statistics calculated for a different plausible values of surface albedo over land.

| Calculated (MODIS) Vs. Simulated (SBDART) | Land type | Land Surface Albedo | Nonlinear fit | | Multilinear fit | |
|---|---|---|---|---|---|---|
| | | | R | RMSE | R | RMSE |
| Planetary albedo | Present study | 0.15 | 0.74 | 0.017 | 0.65 | 0.065 |
| | Forest | 0.14 | 0.72 | 0.019 | 0.63 | 0.067 |
| | Cropland | 0.20 | 0.69 | 0.023 | 0.59 | 0.071 |
| | Grass land | 0.21 | 0.67 | 0.025 | 0.56 | 0.074 |
| | Barren land | 0.38 | 0.62 | 0.033 | 0.50 | 0.079 |
| First Indirect Forcing by Anthropogenic fraction | Present study | 0.15 | 0.83 | 0.037 | 0.62 | 0.048 |
| | Forest | 0.14 | 0.73 | 0.039 | 0.55 | 0.050 |
| | Cropland | 0.20 | 0.69 | 0.043 | 0.50 | 0.055 |
| | Grass land | 0.21 | 0.66 | 0.044 | 0.49 | 0.057 |
| | Barren land | 0.38 | 0.60 | 0.050 | 0.45 | 0.062 |
| First Indirect Forcing by Natural fraction | Present study | 0.15 | 0.77 | 0.042 | 0.54 | 0.049 |
| | Forest | 0.14 | 0.71 | 0.043 | 0.50 | 0.050 |
| | Cropland | 0.20 | 0.66 | 0.045 | 0.48 | 0.051 |
| | Grass land | 0.21 | 0.62 | 0.047 | 0.47 | 0.053 |
| | Barren land | 0.38 | 0.59 | 0.052 | 0.45 | 0.060 |

**Table S3(b):** The statistics calculated for the different plausible values of surface albedo over ocean.

| Calculated (MODIS) Vs. Simulated (SBDART) | Ocean Surface Albedo | Nonlinear fit | | | | Multilinear fit | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | R | | RMSE | | R | | RMSE | |
| | | AS | BOB | AS | BOB | AS | BOB | AS | BOB |
| Planetary albedo | Present study | 0.79 | 0.76 | 0.010 | 0.019 | 0.70 | 0.67 | 0.042 | 0.049 |
| | 0.13 | 0.69 | 0.67 | 0.021 | 0.029 | 0.61 | 0.59 | 0.059 | 0.059 |
| | 0.11 | 0.75 | 0.73 | 0.013 | 0.022 | 0.66 | 0.63 | 0.047 | 0.052 |
| | 0.08 | 0.71 | 0.7 | 0.017 | 0.026 | 0.63 | 0.61 | 0.053 | 0.055 |
| | 0.06 | 0.68 | 0.65 | 0.023 | 0.03 | 0.59 | 0.58 | 0.056 | 0.061 |
| First Indirect Forcing by Anthropogenic fraction | Present study | 0.89 | 0.87 | 0.035 | 0.037 | 0.68 | 0.68 | 0.047 | 0.048 |
| | 0.13 | 0.83 | 0.8 | 0.041 | 0.043 | 0.60 | 0.59 | 0.060 | 0.061 |
| | 0.11 | 0.87 | 0.85 | 0.037 | 0.04 | 0.64 | 0.64 | 0.055 | 0.054 |
| | 0.08 | 0.81 | 0.83 | 0.039 | 0.041 | 0.60 | 0.61 | 0.059 | 0.057 |
| | 0.06 | 0.78 | 0.78 | 0.042 | 0.045 | 0.58 | 0.56 | 0.065 | 0.067 |
| First Indirect Forcing by Natural fraction | Present study | 0.86 | 0.85 | 0.039 | 0.038 | 0.61 | 0.65 | 0.049 | 0.051 |
| | 0.13 | 0.74 | 0.76 | 0.046 | 0.047 | 0.51 | 0.57 | 0.055 | 0.060 |
| | 0.11 | 0.83 | 0.83 | 0.040 | 0.041 | 0.58 | 0.62 | 0.051 | 0.053 |
| | 0.08 | 0.80 | 0.79 | 0.042 | 0.044 | 0.54 | 0.60 | 0.054 | 0.057 |
| | 0.06 | 0.72 | 0.75 | 0.049 | 0.051 | 0.50 | 0.55 | 0.058 | 0.061 |

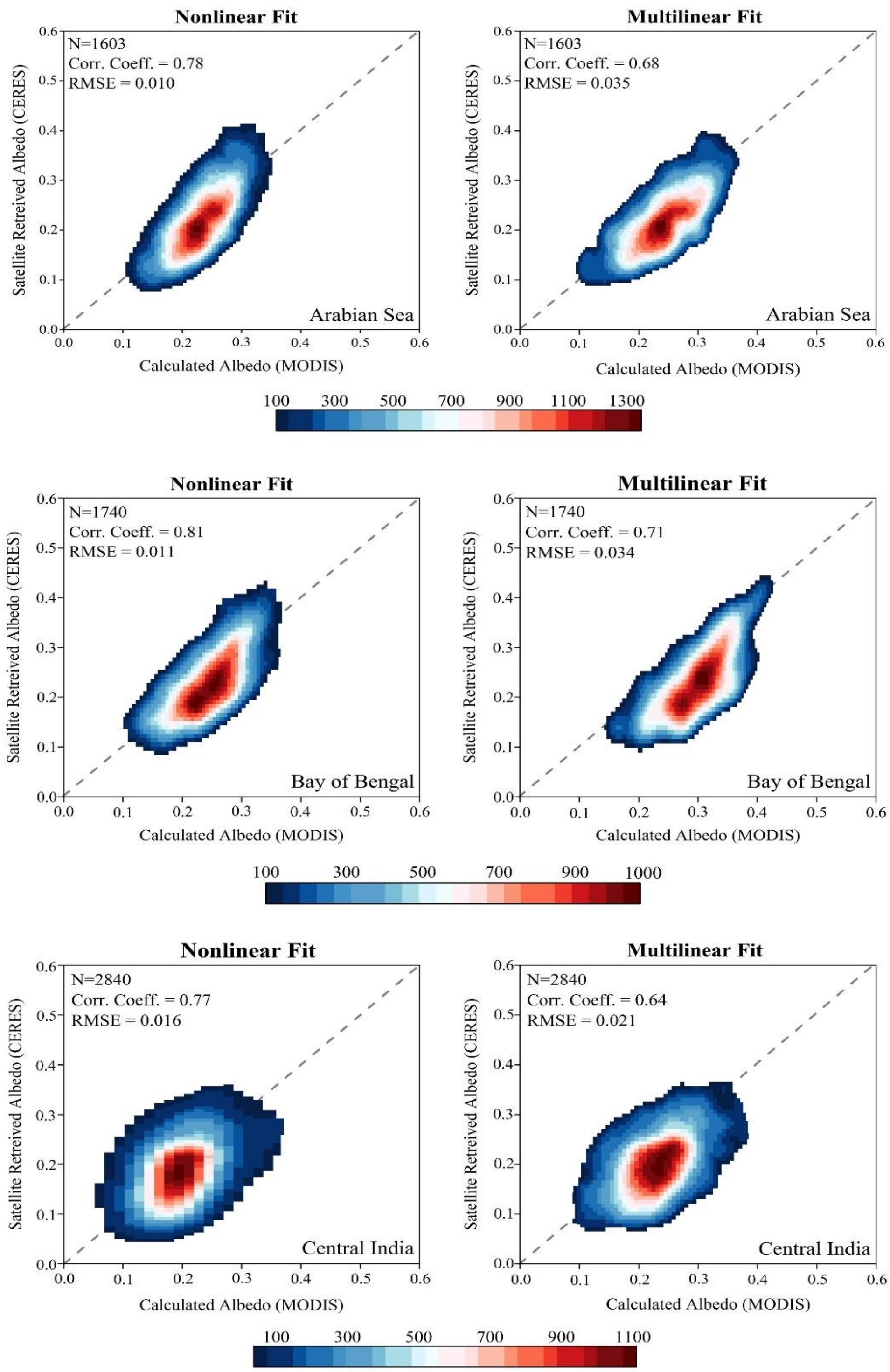



**Figure** ~~S3.~~**S1:** Scatter density plots CERES-retrieved α vs. calculated α for all three regions.