remote sensing

Supplementary Information

Interdecadal changes in aerosol optical depth over Pakistan based on the MERRA-2 reanalysis data during 1980-2018

S1. Spatial Correlation between MERRA-2 and MODIS AOD

Figure S1 show the difference in AOD between MERRA-2 and MODIS-Aqua (DB) data sets during (2002–2018). Here MERRA-2 (0.5° × 0.625°) pixel sizes were converted to a grid of 1° × 1° to obtain the same spatial resolution of MODIS when comparing both the data sets. Since, the positive pixel value reflects that the MERRA-2 annual AOD is lower than that of MODIS, and vice versa. The low (0.05–0.1) to moderate (over 0.25) positive pixel values has been found in the central regions of Pakistan. However, the negative values were reported in the East and West regions of the study domain, with the highest in the Indus basin (> −0.2), suggested that both the sources have varying retrieval algorithms for AOD, which is generally associated with the uncertainties in the algorithm, all sky conditions etc. Besides, the discrepancy found between MODIS and MERRA-2 data sets over the study domain is attributed to affiliated uncertainty with surface reflectance by the MODIS, measurement precision, resolution, sampling time, and system calibration stability

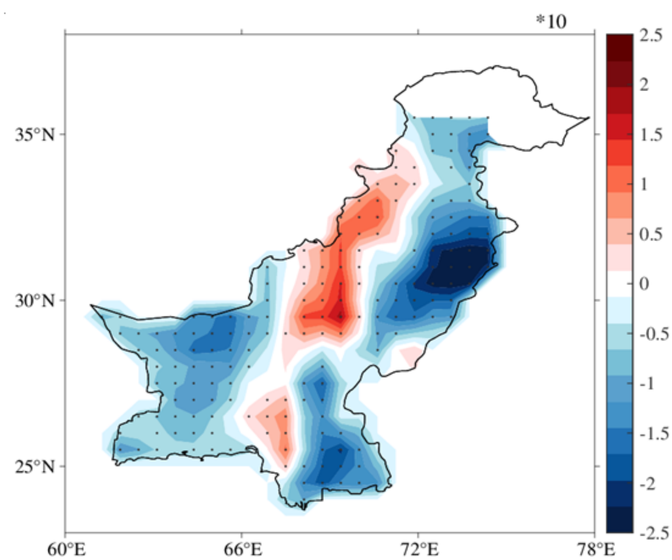

Figure 1. Correlation between MODIS-Aqua and MERRA-2 derived data set for the study period (2002-2018). The colored scale is common for both AODs retrieved from MODIS and MERRA-2.

S2. Sequential Mann-Kendall (Seq-MK) test

The Seq-MK test developed by Sneyers., (1990) is used to determine the beginning of a significant trend in a time series (Kumar, 2018). The potential trend turning point in long term datasets are estimated by progressive (UF) and retrograde (UB) series. In this test, UF is a standardized variable with zero mean and unit standard deviation. Therefore, its sequential behavior fluctuates around zero. The nature of UF is the same as the Z values that are starting from the first to last data point. Similarly, the value of UB is computed backward, starting from the endpoint to the first point of time series.

UF is the same in nature as the Z values that range from the first to the last data point. Similarly, UB's value is calculated backwards, beginning from the endpoint to the first time series point. This test takes into account the relative values of the time series for all terms (x_1, x_2,…,x_n). In computing the Seq-MK check the following steps were added.

1. First, we compared the magnitudes of x_j annual mean time series (j =1, …, n) with x_k, (k=1, …,j −1). The number of cases $x_j>x_k$ was counted and the outcome was denoted by n_j.
2. The test statistic t was obtained using Eq. (S1)

$$t_j = \sum_i^j n_j \tag{1}$$

3. The mean and variance of the t were computed using Eqs. (S2) and (S3):

$$E\,(t) = [n(n\text{-}1)]/4 \tag{2}$$

$$\mathrm{Var}\,(t_j) = j(j\text{-}1)(2j\text{+}5)/72 \tag{3}$$

4. In the end, the sequential values of UF and UB were calculated using Eq. (S4)

$$UF(t) = \frac{[t_j - E(t)]}{\sqrt{var}}(t_j)^{-1} \tag{4}$$

If $|UF\,(t)| \leq UF\,(t)_{1-\beta/2}$, where $UF\,(t)_{1-\beta/2}$ is the critical value of the standard normal distribution with a probability value exceeding β/2, the null hypothesis would be accepted at a significance level in the trend test. In this study, we set β value as 0.05. The decreasing values of UF and UB indicate a negative trend, while the increasing values represent a positive trend in the time series (Bari et al., 2016). The intersection points of the UF and UB curves indicate the possible turning year of the trend within a time series (Hamed et al., 1998). The trend turning point is considered significant at the corresponding threshold level (i.e., ± 1.96 for the 95% significance level).

S3. Mutations in seasonal and annual changes of AOD

Figures S2 and S3 show the results of Seq-MK non-parametric statistical analysis applied to detect fluctuations in the long-term monthly mean AOD over the study domain during 1980–2018 for the MERRA-2 reanalysis data. The reason behind the use of this approach is to recognize change points in long-term time-series data. Generally, this test does not need the data to be linear/normally distributed, but does require that there is no autocorrelation in the data (AOD) used, and is statistically helpful in calculating the monotonic upward or downward trend in time series values (Hamed et al., 1998). The progressive (UF) and retrograde (UB) series were attained at a statistical significance of 95% during the study period. On the annual scale, the variation in AOD over six different subdomains can be classified into different patterns (Figure S2).

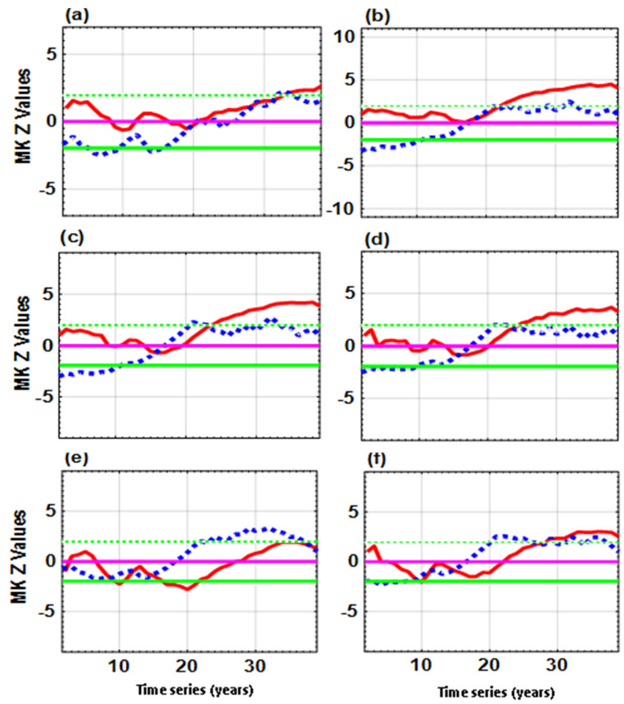

Figure 2. Abrupt variation in annual mean AOD values during 1980–2018; over the study regions **a)** NDR, **b)** Katawaz Basin **c**) Indus Basin **d)** Balochistan Plateau **e)** Coastal region and **f)** KDR covering the study domain of Pakistan at a significance level of 95%. The statistics UF (red solid line) represent the trend variability in forward direction whereas UB (blue dot line) represent the trend in the backward direction with zero mean and unit standard deviation.

An obvious increase in AOD is prominent over the NDR (relatively higher altitude region) with a steep negative shift detected during 2002, followed by twin positive mutations in the onward trend. Whereas, the trends in the regions of NDR, Katawaz Basin, Indus Basin, and Balochistan Plateau were found negative during 1980–2003, indicating a clean environment. However, an increase in the later years till 2018 is depicted with the most leading features that appeared during winter and summer seasons (Figure S3a–d, m–p) indicating increasing air pollution in the recent decade. The results are attributed to local and regional hydro-meteorological cycle, long-range natural aerosol fluxes, orographic factors, and temperature inversion.

Besides, the declining shift in AOD is conspicuous in the low (high) elevation zone of coastal region (KDR) till late years 2010 (2007), with an increase in the trend in the following years. The pattern is obvious during autumn (Figure S3w,x) attributed to the fact that the areas are mostly affected by the Asian monsoon circulation in the West and frequent dust storms during the season. The second reason is the shift of population from rural areas to these industrially developed regions after the economic development started in early 2002 in Pakistan. However, the seasonal variation in spring appears with significant sharp fluctuation paces throughout the study domain (Figure S30g–i) attributed to regional climatology (Khan et al., 2019), and dynamics played by different meteorological factors. Adding, the sudden changes usually indicate the urban air quality and transition of climate from one stable state to another in recent decades.

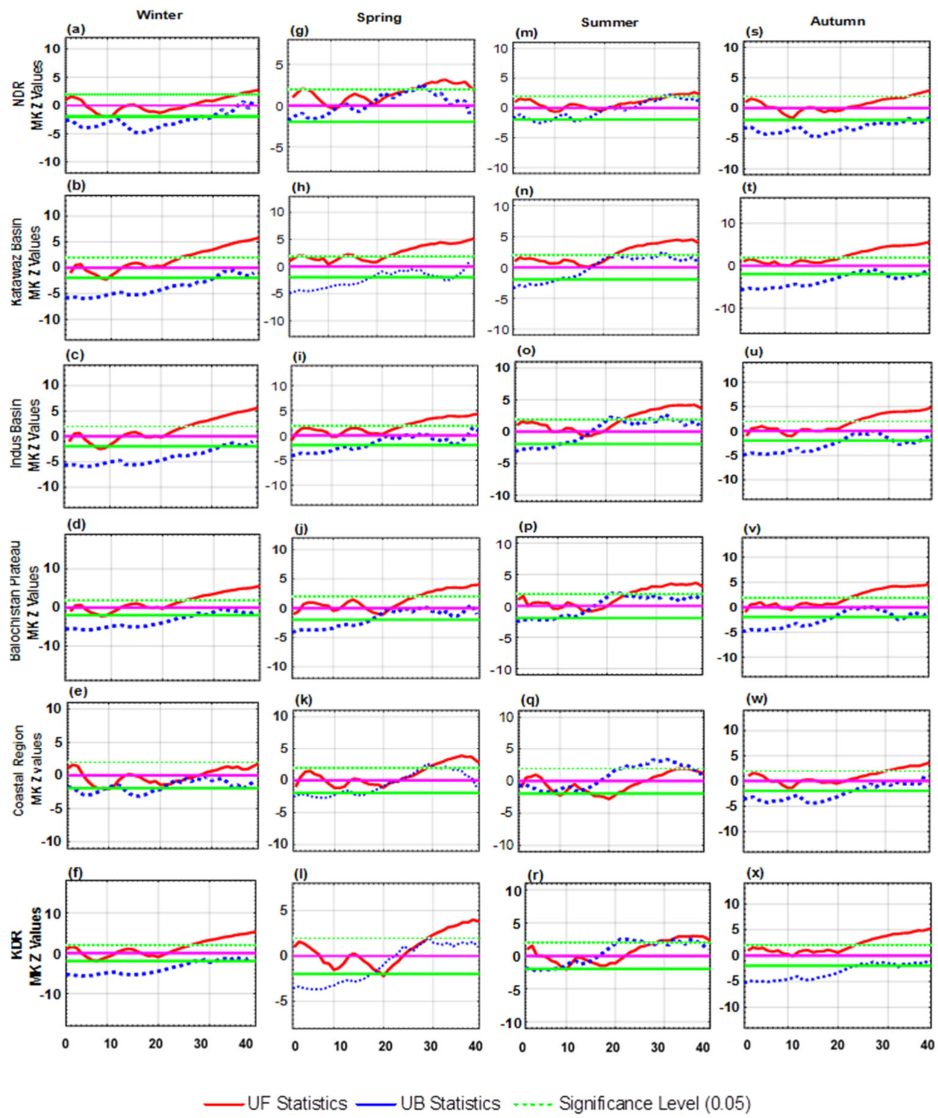

Figure 3. Same as Figure. S2, but for the seasonal variations.

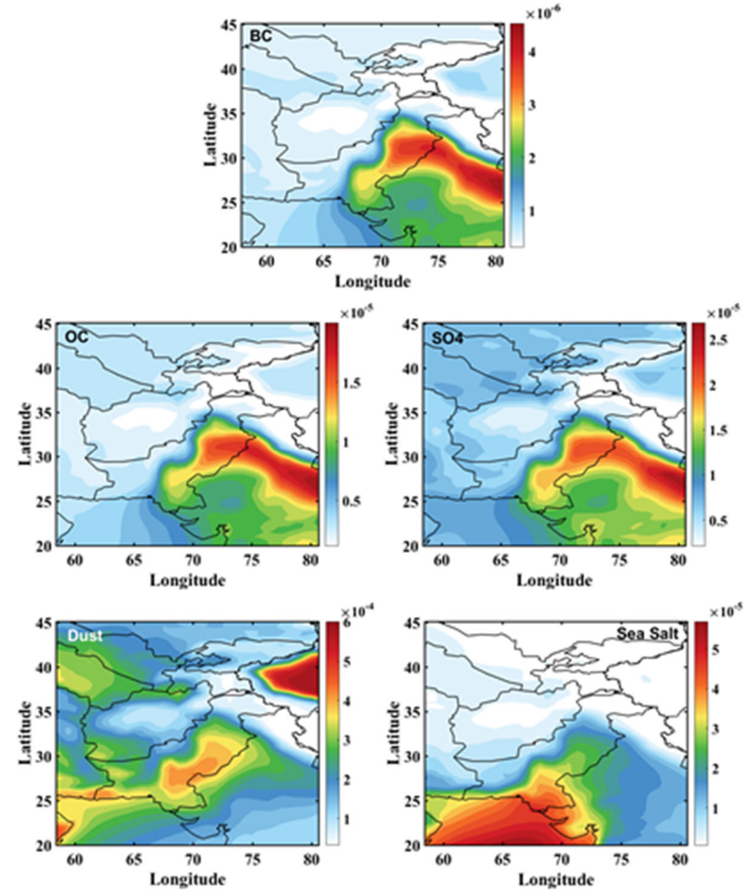

Figure 4. Spatial variation of total natural/anthropogenic emission of black carbon (BC), organic carbon (OC), SO_4, dust and sea salt particles (unit: kg m^{-2}) retrieved from MERRA-2 reanalysis during the study period 1980–2018 observed in Pakistan and the surrounding areas.