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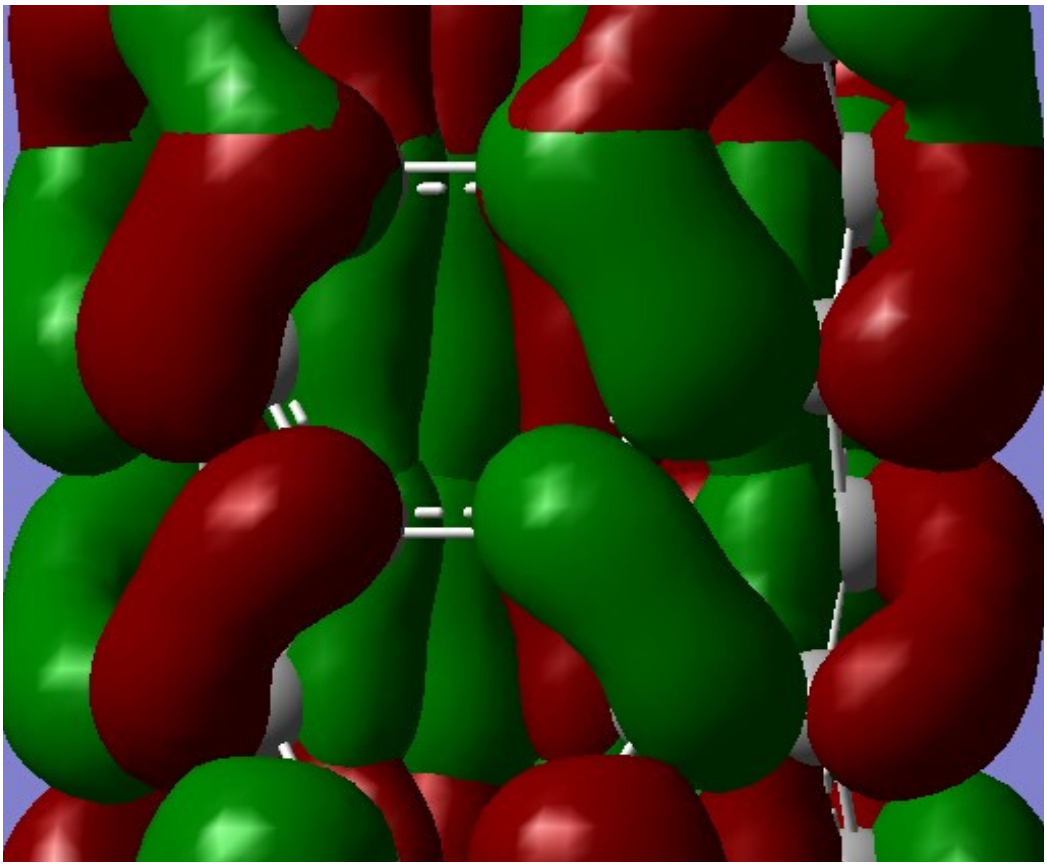

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Himalayan Physics

Aerosol optical properties of size segregated aerosol particles and radiative forcing over Pokhara valley

Research Article

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Abstract: The AERONET data from sun/sky radiometer over Pokhara (2017) was analyzed to observe aerosol optical depth (AOD) of size segregated particles and radiative forcing. Fine mode particles have over seventy percent of contribution to AOD in all months with the maximum, ninety-four percent, on November and February, and minimum on July, seventy-nine percent. The monthly mean top of atmosphere (TOA) forcing are negative $-23.225 \pm 4.71\, Wm^{-2}$ in March, $-28.958 \pm 4.71\, Wm^{-2}$ in April and $-19.616 \pm 4.71\, Wm^{-2}$ in May. The negative value of TOA during all the months in pre-monsoon season indicates net cooling. Whereas surface forcing (BOA forcing) was found to be positive with a maximum value of $111.18 \pm 27.63\, Wm^{-2}$ during April and a minimum of $56.22 \pm 27.63\, Wm^{-2}$ during May. The resultant atmospheric forcing is the absorption due to aerosols within the atmosphere and found to be $+267.57\ Wm^{-2}$ during April and $+228.55\ Wm^{-2}$ during May; indicating significant heating of the atmosphere.

Keywords: Radiative Forcing • Aerosol Optical Depth • Coarse Mode/Fine Mode • Atmosphere (TOA and BOA)

1. Introduction

The radiation balance of the incoming and outgoing energy in the Earth-Atmosphere system is affected by external factors such as greenhouse gases and aerosols. Radiative forcing (RF), which is the change in the radiation budget received by the planet and energy radiated back to space, leads to either cooling or warming based on negative and positive RF [1]. Alternatively, the flux changes at the top of the atmosphere (TOA) is radiative forcing and is a better indicator of the global mean surface temperature [2]. Aerosol particles can influence climate directly by scattering and absorbing (in the case of black carbon) solar radiation [3] . Since a portion of the scattered radiation will be reflected back into space and leads to a cooling of the atmosphere. Fine particles of diameters less than one micron (μm) are most effective for governing the strength of the aerosol optical properties [4].

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The characteristic of chemical components of aerosols particles indicate for the strength of absorption and scattering of solar radiation by aerosol particles. Black carbon absorbs solar radiation, which leads to a warming of the atmosphere [5]. Black carbon deposition on snow and ice could decrease the surface albedo, leading to additional warming. Mineral dust also contributes for the absorption of radiation. Aerosol compounds such as sea salt, nitrate and sulfate scatter the light significantly than absorption resulting the cooling of the Earths atmosphere and therefore are also known as cooling agents. The indirect effect of aerosols is on the Cloud Microphysics that alters the cloud albedo by forming cloud condensation nuclei (CCN). The aerosols are responsible to reduce surface albedo upon deposition on snow, resulting in positive radiative forcing [6–8]. The aerosols can have both warming or cooling of the atmosphere called aerosol radiative forcing [9].

The deviation in radiation energy due to the interaction with atmospheric components is called attenuation. Absorption and Scattering of radiation are two processes included in attenuation. The radiation-matter interaction depends on wavelength of radiation, physical and chemical properties of the particle interacting with the radiation. It is known that more attenuation is possible in a region which has large numbers of such attenuators. The interaction and attenuation of radiation is determined by the optical depth (τ) and is given by,

$$\tau = \int \sigma \ \rho \ \mathrm{dx} \tag{1}$$

where ρ is the density of the attenuator present in the path length dx in the direction of propagation and σ is the mass extinction cross section of the matter present within dx. In general, the intensity of radiation decreases exponentially with optical depth. If radiation with initial intensity I_o traverses a thickness dx with matter of optical depth ρ then the intensity after crossing this distance is given by;

$$I\,(\lambda) = I_0 \ \ (\lambda) \ \ e^{-\tau} \tag{2}$$

This is called Beers law and sometimes it is referred to as Beer Lamberts law. The optical depth depends upon wavelength and number density of the attenuator. In order to get total attenuation of radiation, optical depth of all matters should be taken into account. Hence;

$$\tau(\lambda) = \tau Rayleigh(\lambda) + \tau Ozone(\lambda) + \tau Aerosol(\lambda) + \tau Cloud(\lambda) \tag{3}$$

where $\tau Rayleigh(\lambda)$, $\tau Ozone(\lambda)$, $\tau Aerosol(\lambda)$ and $\tau Cloud(\lambda)$ are optical depths caused by molecular, ozone, aerosols and clouds respectively [10].

The larger the optical thickness at a particular wavelength, the less light of that wavelength reaches Earths surface. Measurement of aerosol optical depth (AOD) provides important information of concentration, size distribution, and variability of aerosols in the atmosphere which are essential parameters for climate studies and to understand the overall effects of aerosols.

Aerosol optical depth (AOD, $\tau(\lambda)$) is a quantitative measure of the extinction of solar radiation by aerosol scattering and absorption and is expressed in terms of Angstrom's parameters, α and β [11, 12] as;

$$\tau(\lambda) = \beta\lambda^{-\alpha} \tag{4}$$

where α is the wavelength exponent that represents columnar aerosol size distribution in the atmosphere and used as a qualitative indicator of aerosol particle size and chemical components. The coefficient β is equal to $\tau(\lambda)$ at a wavelength of 1 μm and depends on the concentration of particles. Typical values of β vary from 0 to 0.5 indicating the higher the amount of aerosol present in the atmosphere with higher the values of β [13]. Size segregated monthly averaged AOD, which are automatically cloud cleared and manually inspected data, over an AERONET site of Nepal, Pokhara (28.18 N, 83.97 E) were received from AERONET webpage (https://solrad-net.gsfc.nasa.gov/networks.html) and analyzed. Comparing these data with RF gives a clear picture of aerosol characteristics. Coarse mode size distribution is being mechanically cut off at 0.6 μm separating fine mode as smaller particles with radius smaller than 0.6 μm. For air quality applications, this paper focuses on the analysis of total AOD , fine/coarse mode aerosol contributions in the AOD, comparison of RF with AOD [14]

The aerosol radiative forcing at the top of the atmosphere (TOA) or at the surface is defined as the difference in the net (down minus up) solar flux (solar plus long wave; in Wm^{-2}) with and without aerosol, i.e..

$$\Delta F = (F_{a\downarrow} - F_{a\uparrow}) - (F_{0\downarrow} - F_{0\uparrow}) \tag{5}$$

where ΔF denotes the irradiance (down-welling or upwelling, Wm^{-2}) and $(F\downarrow - F\uparrow)$ denotes the net irradiance (down-welling minus upwelling) computed with aerosol (F_a) and without aerosol (F_0) at either the TOA or the surface [15].

The aerosol radiative forcing efficiency is defined as the rate at which the atmosphere is forced per unit of aerosol optical depth at 0.55 μm both at BOA and TOA:

$$\Delta F^{eff}_{BOA/TOA} = \Delta F_{\frac{BOA}{TOA}}/\tau(0.55) \tag{6}$$

It gives an evaluation of the direct radiative effect for each type of aerosol, characterized by absorption and size distribution [16]. Forcing efficiency depends upon nature of aerosol type. It is higher for absorbing aerosols like black carbon.

2. Methodology

Aerosol Robotic Network (AERONET)

AERONET is a globally distributed network of automatic sun and sky scanning radiometers. Aerosol data from an AERONET site, Pokhara (28.187N, 83.975E), is retrieved for this study. AERONET uses CIMEL sun

and sky radiometer, which operates in two modes, direct sun measurements at 340 nm, 380 nm, 440 nm, 500 nm, 675 nm, 870 nm, 1020 nm, 1640 nm wavelengths and sky measurements at 440 nm, 675 nm, 870 nm and 1020 nm [17, 18]. These solar extinction measurements were used to compute aerosol optical depth (AOD), which were automatically computed by using a software and are available in the AERONET website. Holben et al. [17] presents the detailed estimation of uncertainty in computed AOD of approximately ± 0.010 to ± 0.021, which is spectrally dependent with higher errors in the UV region.

AERONET Data selection

In our analysis, we included fine-mode AOD, coarse mode and total AOD at 500 nm. First, we obtained daily means aerosol data from the website and were then used to calculate the monthly averages, by requiring at least 10 days per month. For the seasonal means it was required that all the months had sufficient amount of measurements. Eck et al. [19] included several example cases to show how the AERONET data have meaningful information for our study as well. These Level 2 size segregated daily averaged aerosol optical depth data also used to calculate the standard deviation while obtaining the monthly averaged data by using Microsoft excel math function. The percentage contribution of fine mode particles on total Aerosol optical depth is calculated and total AOD is compared with RF.

RF data analysis

The sky radiance is measured along the solar principal plane (i.e., at constant azimuth angle, with varied scattering angles) up to nine times a day and along the solar almucantar (i.e., at constant elevation angle, with varied azimuth angles) up to six times a day. The approach is to acquire aureole and sky radiances observations through a large range of scattering angles from the sun through a constant aerosol profile to retrieve size distribution, phase function and aerosol optical depth. Sky radiance measurements are inverted with the Dubovik and Nakajima inversions to provide aerosol properties of size distribution and phase function over the particle size range of 0.1 to 5 μm. We retrieved Version 3, Level 2.0 data of radiative forcing, from inversion products of AERONET site which assures high accuracy and quality-controlled data. Version 3 inversion description document describes the input data sets, scalar to vector computations, and additional retrieval products including sensitivity to input uncertainties.

Radiative Forcing (ΔF) at the BOA (Bottom of Atmosphere) and at the TOA (Top of Atmosphere)

The study on the effect of atmospheric aerosols on the climate system is managed by AERONET by computing the direct radiative forcing at the BOA (Bottom of Atmosphere) and at the TOA (Top of Atmosphere). The aerosol radiative forcing strongly depends on the total aerosol extinction (AOD), the solar geometry and the surface type. Solar geometry is especially critical in estimating the ΔF at the TOA, since a clear decrease of

the ΔF absolute values has been documented as surface reflectivity increases, even changing the sign of the radiative forcing. It is found that over dark surfaces atmospheric aerosols always cool the Earth-atmosphere system, regardless of aerosol type [20, 21]. But, over the brightest surfaces, the total radiative effect depends on the aerosol absorption properties and on the SR values. It means a consistent comparison of the net aerosol effect requires the analysis for two ranges of surface reflectivity: SR $\leq$ 30% and SR $\geq$ 30%.

3. Results and Discussion

Contribution of Fine and coarse mode particles on AOD

The fractional contribution of Fine mode on AOD is shown in Figure 1. The fine mode particle contribution over all AOD is over seventy percentage in all months with the maximum, ninety-four percent, on November and February with minimum on July, seventy-nine percent. Particle size distribution is one of the most important parameters for characterizing the aerosol particles in the atmosphere. Seinfeld (1998) reports that the size distribution of particles in the atmosphere strongly indicates for the sources of aerosols. The coarse mode particles, diameters greater than 1 micrometer, are mainly produced by mechanical processes and injected into the atmosphere directly from anthropogenic and natural processes. On the other hand, fine mode particles, diameters smaller than 1 micrometer, are the particulate matters which are injected into the atmosphere through the process of combustion of wood, oil, coal, gasoline and other fuels. Because of the nature of their sources fine particles generally contain substantial amounts of organic material as well as soluble inorganics such as ammonium, nitrate and sulfate [22].

As a result, it is not surprising to see that the fine mode fractional AOD, percentage contribution of fine mode aerosols (Fig. 1) are strong in the months of pre-monsoon and post-monsoon seasons in comparison to months of summer monsoon. The post monsoon season (Oct-Nov) begins with a slow withdrawal of the monsoon. This retreat leads to an almost complete disappearance of moist air by October and resumes cool, clear, and dry weather. The summer monsoon, a strong flow of moist air from the southwest, follows the pre-monsoon period, which changes the particle composition and increases water contents in the atmosphere. Previous study shows that Pokhara has a significant Elemental Carbon contributed by biomass burning and fossil fuel combustion [3]. This shows that the concentration of fine mode particles stays abundantly in atmosphere for a long time and are significantly important for the attenuation of solar radiation over the Pokhara during the pre-monsoon and post-monsoon season in comparison to rainy season.

Fig. 2 shows that the aerosol impact on solar radiation elevated from post-monsoon season towards pre-monsoon seasons. This period must be impacted by anthropogenic aerosols and in the summer time the overall AOD decreases sharply and indicate for the impact of rainfall due to monsoon. The overall AOD in two months July and August is only about 12% of total average AOD of months February to April. This shows intense aerosol plumes in pre-monsoon period.

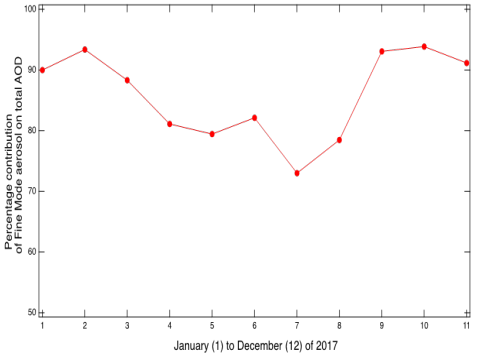

Figure 1. Percentage contribution of fine mode aerosol on total columnar AOD over Pokhara.

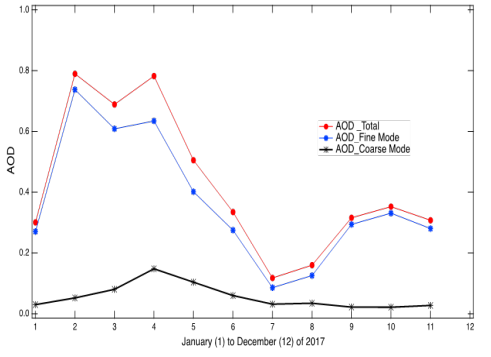

Figure 2. Monthly average total AOD and AODs due to fine and Coarse Mode particles.

Variation of Radiative forcing

The Table 1 shows the radiative forcing in the season where the attenuation of solar radiation by aerosols are significantly high in comparison to other months. TOA forcing is found to be negative during all the months in pre-monsoon season which indicates net cooling as shown in figure 3 (c) and (d). The monthly mean TOA forcing was found to be $-23.225 \pm 4.71\ Wm^{-2}$ in March, $-28.958 \pm 4.71\ Wm^{-2}$ in April and $-19.616 \pm 4.71\ Wm^{-2}$ in May whereas surface forcing (BOA forcing) was found to positive with a maximum value of $111.18 \pm 27.63\ Wm^{-2}$ during April and a minimum of $56.22 \pm 27.63\ Wm^{-2}$ during May. The forcing efficiency was found to be maximum $-65.619 \pm 6.504\ \mathrm{Wm}^{-2}$ in March indicating the abundant burning of biomass.

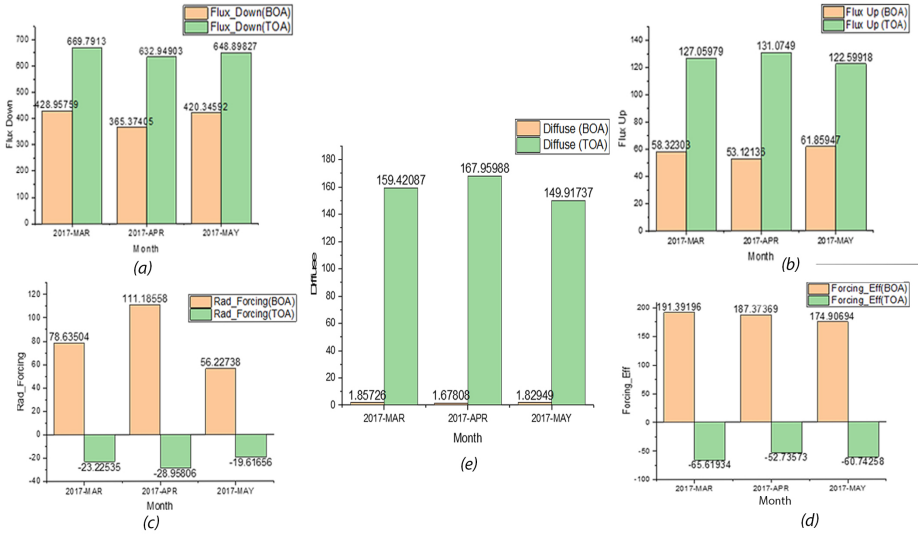

Figure 3. Variation of Radiative Forcing during Pre-Monsoon Period of 2017

Fig. 3(a) and Fig. 3(b) shows a net flux down and net flux up are positive in all the months of pre monsoon season. The resultant atmospheric forcing is the absorption due to aerosols within the atmosphere and found to be $+267.57\ Wm^{-2}$ during April and $+228.55\ Wm^{-2}$ during May indicating significant heating of the atmosphere. The forcing efficiency (ΔF) at the surface was found to be $-65.6, -52.7$ and $-60.7\ Wm^{-2}$ during March, April and May respectively as seen in Fig. 3(d). Large aerosol induced negative surface forcing during pre-monsoon season is due to the mixing of anthropogenic aerosol pollution with transported natural dusts [23]. Figs. 4, 5 and 6 shows the daily variation of flux down, flux up and radiative forcing respectively at BOA and TOA. They reveal the significant variation of radiation pre and post monsoon days but it is almost washed out due to heavy precipitation during monsoon period.

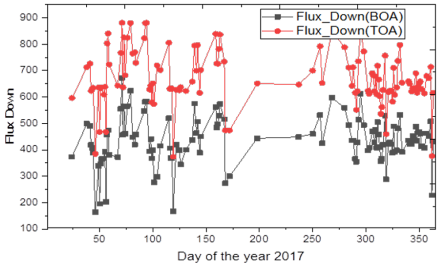

Figure 4. Daily variation of flux down at the top of atmosphere (TOA) and bottom of atmosphere (BOA).

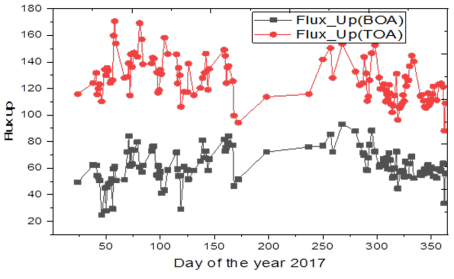

Figure 5. Daily variation of flux up at the top of atmosphere (TOA) and bottom of atmosphere (BOA).

The variety of components of solar radiation at the top of atmosphere and bottom of atmosphere are given in the Table 1 and graphs are presented in Figs. 3 to 7 [24]

Table 1. Variation of Radiative Forcing during Pre-Monsoon Period of 2017

Month of 2017	Flux Down (BOA)	Flux_Down (TOA)	Flux_Up (BOA)	Flux_Up (TOA)	Rad_Forcing (BOA)	Rad_Forcing (TOA)	Forcing_Eff (BOA)	Forcing_Eff (TOA)	Diffuse (BOA)	Diffuse (TOA)
MARCH	428.96	669.79	58.32	127.06	78.63	-23.22	191.39	-65.62	1.86	159.42
APRIL	365.37	632.95	53.12	131.07	111.19	-28.96	187.37	-52.74	1.68	167.96
MAY	420.35	648.90	61.86	122.60	56.23	-19.62	174.91	-60.74	1.83	149.92
STDV	34.49	18.48	4.39	4.24	27.63	4.71	8.60	6.50	0.10	9.02

Relationship between AOD and Radiative Forcing

The amount of cloud cover and duration of bright sunshine hour makes a difference in the amount of solar irradiance. Fig. 7 shows the monthly variation of diffuse radiation (BOA) and AOD_500. It is seen that there is an increasing trend in AOD with decreasing value of diffuse solar radiation. Diffuse solar radiation is found to be more during winter than that of summer and consequently the value of AOD varied.

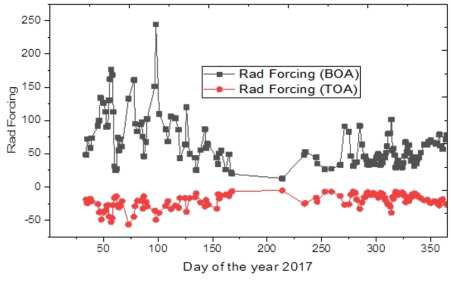

Figure 6. Variation of Radiative forcing and day of the year 2017.

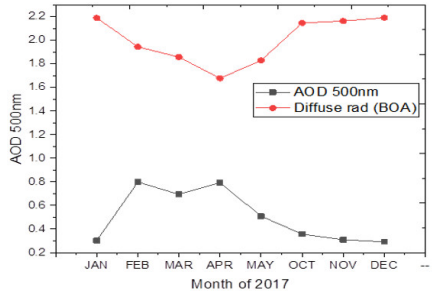

Figure 7. Monthly averaged Aerosol Optical Depth at 500 nm with the months of 2017.

4. Conclusions

Our finding based on the percentage contribution of fine mode aerosol particles on total AOD data suggests that using total AOD as a single predictor will not give a clear picture of aerosol components of the atmosphere. AOD weighs all types of aerosol components equally and does not consider the impact of size segregated particle composition. Presenting the contribution of different size of particles on overall AOD will indicate for the aerosol sources and origin of air particulate matter over the atmosphere. During the transition months (AprilMay and SeptemberNovember) the aerosol patterns are dominated by fine particles, however in summer months the pattern is the hybrid mix of the two sizes of particles. Along with climatic and weather impacts, aerosol also leads to negative effects on public health and ecosystem. The study of contribution of aerosol on overall radiative forcing in this paper formulates the fundamental reasons for resulting the climate change from human induced changes in atmospheric composition. The results also suggest for the importance of further study by comparing aerosol chemical composition and size segregated aerosol particles for identifying the origin aerosol particles over Pokhara.

5. Acknowledgments

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