



# Trend analysis in aerosol optical depths and pollutant emission estimates between 2000 and 2009

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## ABSTRACT

We evaluated global and regional aerosol optical depth (AOD) trends in view of aerosol (precursor) emission changes between 2000 and 2009. We used AOD data products from MODIS, MISR and AERONET and emission estimates from the EMEP, REAS and IPCC inventories. The trends in global monthly AOD of MODIS (L3), MISR (L3) and AERONET (L2) are significantly negative over Europe and North America, whereas over South and East Asia they are mostly positive. The calculated 2000–2009 trends from the monthly L3 products correspond well with the more detailed daily MODIS L2 AODs for three selected regions (Central Mediterranean, North-East America and East Asia). Furthermore, daily and monthly AERONET L2 AOD trends agree well. The trends in AOD are compared to estimated emission changes of SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub> and black carbon. We associate the downward trends in AOD over Europe and North America with decreasing emissions of SO<sub>2</sub>, NO<sub>x</sub> and other pollutants. Over East Asia the MODIS L2 trends are generally positive, consistent with increasing pollutant emissions by fossil energy use and growing industrial and urban activities. It appears that SO<sub>2</sub> emission changes dominate the AOD trends, although especially in Asia NO<sub>x</sub> emissions may become increasingly important. Our results suggest that solar brightening due to decreasing SO<sub>2</sub> emissions and the resulting downward AOD trends over Europe may have weakened in the 2000s compared to the 1990s.

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## 1. Introduction

The changing abundance of aerosols perturbs the atmospheric radiation budget (radiative forcing). The concentration changes or modifications of the aerosol type lead to variations of the induced radiative forcing. Aerosols scatter (e.g. sulfate) and absorb (e.g. black carbon) sunlight, generally cooling the Earth's surface and warming the atmosphere, depending on the level of absorption. Recent studies focusing on the northern hemisphere (Wild et al., 2005, Wild, 2010, special issue J. Geophys. Res., and references therein) have shown that since the late 1980s the decline in solar radiation at the Earth's surface due to aerosol pollution (dimming) has reversed. This change from dimming to brightening has important consequences for climate change, affecting the hydrological cycle, cloud formation and surface temperatures, possibly intensifying the warming trend caused by CO<sub>2</sub> and other

greenhouse gases. Changes in the Aerosol Optical Depth (AOD) are critical in the solar dimming-brightening discussion, and trends in the emissions have been shown to be largely anthropogenic (Streets et al., 2009).

Several studies have investigated the global and regional surface solar radiation budget using data of the Global Energy Balance Archive (GEBA), e.g. Ohmura (2009), Gilgen et al. (1998), Long et al. (2009), Norris and Wild (2009). Pinker et al. (2005) and Hinkelman et al. (2009) analyzed long-term variations in solar radiation at the Earth's surface using satellite data. Mishchenko et al. (2007) and Mishchenko and Geogdzhayev (2007) used the AOD from the Advanced Very High Resolution Radiometer (AVHRR) satellite to study the dimming and brightening tendencies. They found a decrease of 0.033 (~0.024/decade) of the global tropospheric AOD. Zhao et al. (2008) studied the long-term (nearly 25 years) AOD trends over the global oceans using the AVHRR Pathfinder Atmosphere extended (PATMOS-x) data set and their results support the conclusion by Mishchenko and Geogdzhayev (2007). Thomas et al. (2010) evaluated the Global Retrieval of Along Track Scanning Radiometer (ATSR-2) Cloud Parameters Evaluation (GRAPE) AOD product over the ocean by comparing against

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measurements of the AErosol RObotic NETwork (AERONET) and AOD of the AVHRR of the Global Aerosol Climatology Project (GACP). They concluded that the GRAPE AODs corroborate the AERONET observations, but some discrepancies are apparent with the GACP data. Remer et al. (2008) analyzed Moderate resolution Imaging Spectroradiometer (MODIS) Terra and Aqua Level2 (L2) and Level3 (L3) aerosol products over land and ocean for 2002–2006. They uncovered increasing AODs over India and East Asia and decreasing AODs over North America and Europe. Chylek et al. (2007) analyzed Multi-angle Imaging Spectro Radiometer (MISR) AODs for 2000–2006 and found a decrease over the USA of  $-0.073/\text{decade}$  and over the Northern Hemisphere of  $-0.014/\text{decade}$ . Lu et al. (2010) studied the  $\text{SO}_2$  emission trend in China after 2000 and found a positive relation with observed L3 AODs of the MODIS and MISR instruments. A modeling study by Streets et al. (2006) indicated that the trends in surface radiation are correlated with the trends in calculated emissions of sulfur dioxide ( $\text{SO}_2$ ), black carbon (BC) and AODs. Zhang and Reid (2010) analyzed global and regional trends in AOD of MODIS and MISR over the oceans, using L2 products. For both instruments they indicated a statistically insignificant global trend of  $0.003/\text{decade}$ , with stronger regional trends over the Bay of Bengal ( $0.07/\text{decade}$ ), the east coast of Asia ( $0.06/\text{decade}$ ) and the Arabian Sea ( $0.06/\text{decade}$ ). To our knowledge comparisons of the AOD trends of MODIS and MISR measurements with AERONET data for the period 2000–2009 (comparing L3 with L2 products), and also linking to aerosol (precursors) emissions have not yet been reported. We did not use products of the MODIS-Aqua instrument, because Aqua was launched in May 2002, and a comparison with MISR data based on 10 years is therefore not possible. This study has three main objectives. The first is to compare trends in global monthly L3 AOD products of MODIS, MISR and AERONET (L2) for the period 2000–2009. A selection was made of the AERONET stations for which ample AERONET monthly L2 data is available ( $>50$  months), which resulted in 62 stations, see Fig. 1. We identified within the MODIS and MISR L3 products the geographical pixel location of the AERONET station and extracted the AODs. However, it is not

recommended to draw strong conclusions based on L3 products, because the sampling of actual retrievals is highly non-uniform in space and time, even at the resolution of these products (MODIS  $1^\circ \times 1^\circ$  and MISR  $0.5^\circ \times 0.5^\circ$ , [Kahn et al., 2009]). Therefore our second objective is to also analyze MODIS L2 AODs between 2000 and 2009 for three selected regions over land with good data coverage and compare the trends with MODIS L3 data. The advantage of L2 is that it provides information about the sampling time and has a better spatial representation ( $10 \times 10 \text{ km}$ ), which is relevant for the comparison to AERONET observations. For MODIS L2 we extract from the overpasses the AODs within the geographical range of  $\pm 0.1^\circ \times 0.1^\circ$  around the AERONET station. The three regions are (i) the Mediterranean basin, (ii) the eastern part of North America and (iii) the north eastern part of China, i.e. densely populated areas with intense industrial activities.

Our third objective is to compare the trends in AODs with the trends in emissions of  $\text{SO}_2$ , nitrogen oxides ( $\text{NO}_x$ ), ammonia ( $\text{NH}_3$ ) and black carbon, which are expected to contribute substantially to the global AOD (Streets et al., 2006, 2009). For this purpose we analyzed estimated source strengths for the years 1990, 1995, 2000, 2005 and 2010 from global and regional emission inventories; from the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants (EMEP) for Europe, the Region Emission Inventory for Asia (REAS) and the IPCC (Representative Concentration Pathway [RCP] 3-Peak and Decline [PD] for North America and the entire globe. Unfortunately, it is difficult to define error margins for the emission estimates, so that this comparison is qualitative rather than quantitative, though nevertheless useful to study the consistency of different data sources.

In this work we use AOD observation of MODIS (version MOD04 for L2 and MOD08 for L3, collection 005), MISR (CGAS-F15) and AERONET. Both MODIS and MISR are aboard the Terra platform which was launched in December 1999. More descriptions of the remote sensing instruments, MODIS MISR and AERONET are given in the electronic supplement, together with more details on the three emission inventories EMEP, IPCC and REAS.

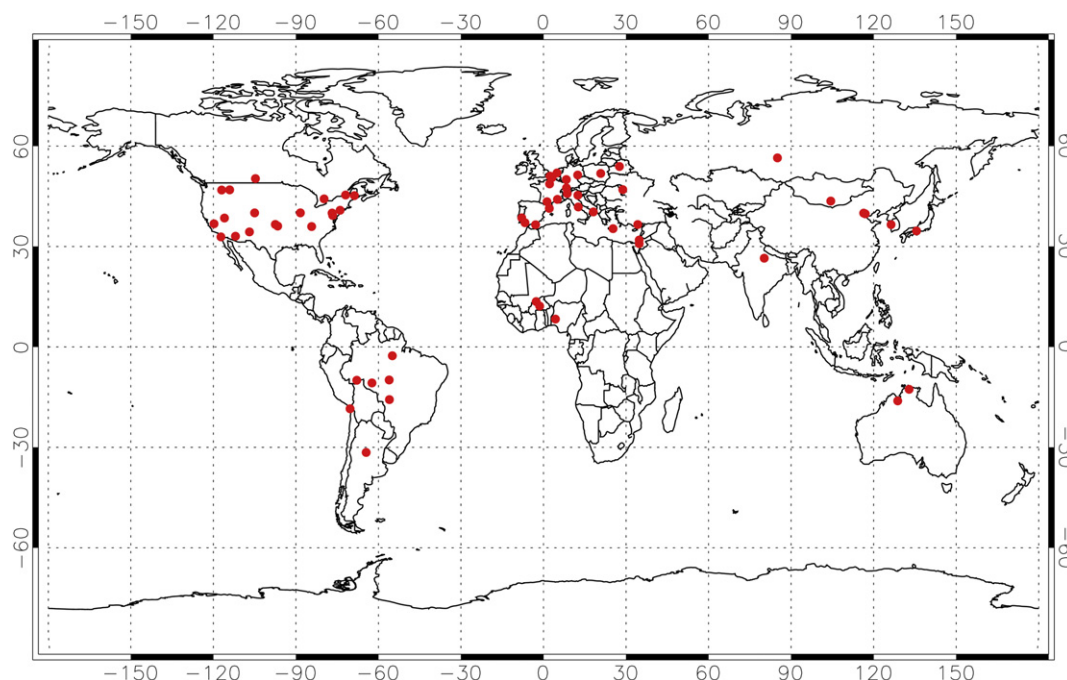


Fig. 1. Overview of the AERONET stations for which monthly mean AODs are compared with MODIS-Terra and MISR AOD, for the period 2000–2009.

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