References


CAAC: Twelfth five-year plan on air pollution prevention and control in key regions (China clean air policy briefing no. 1) (English Translation), Clean Air Alliance of China, Secretariat for Clean Air Alliance of China, suit 1705, Building 1, Park Avenue, 16 Jianguomenwai Street, Beijing 100022, 2013.


CPCB: NATIONAL AMBIENT AIR QUALITY STATUS & TRENDS IN INDIA-2010, Central Pollution Control Board, India, 2012.

CPCB: National Air Quality Standards, Central Pollution Control Board (CPCB), 2009.


References


Holland, M., Watkiss, P. and Pye, S.: Cost-benefit analysis of the Thematic strategy on air pollution for service contract for carrying out cost-benefit analysis of air...
quality related issues, in particular in the Clean Air for Europe (CAFE) programme, European Commission DG Environment, 2005.


Exploring the atmospheric chemistry of nitrous acid (HONO) at a rural site in Southern China, Atmos. Chem. Phys., 12, 1497-1513, doi:10.5194/acp-12-1497-2012, 2012.


Stohl, A., Klimont, Z., Eckhardt, S., Kupiainen, K., Shevchenko, V. P., Kopeikin, V. M. and Novigatsky, A. N.: Black carbon in the Arctic: the underestimated role of
References


Tripathi, O. P., Stephen, G. J., O'Dowd, C., O'Leary, B., Lambkin, K., Moran, E., O'Doherty, S. J. and Spain, T. G.: An assessment of the surface ozone trend in
References


Ware, J. H., Ferris, B. G., Dockery, D. W., Spengler, J. D., Stram, D. O. and Speizer, F. E.: Effects of ambient sulfur oxides and suspended particles on respiratory


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Study of Ozone and NO$_2$ over Gadanki – a rural site in South India

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Study of Ozone and NO$_2$ over Gadanki – a rural site in South India

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Abstract We have studied long-term changes in tropospheric NO$_2$ over South India using ground-based observations, and GOME and OMI satellite data. We have found that unlike urban regions, the region between Eastern and Western Ghat mountain ranges experiences statistically significant decreasing trend. There are few ground-based observatories to verify satellite based trends for rural regions. However, using a past study and recent measurements we show a statistically significant decrease in NO$_X$ and O$_3$ mixing ratio over a rural location (Gadanki; 13.48° N, 79.18° E) in South India. In the ground-based records of surface NO$_X$, the concentration during 2010–11 is found to be lower by 0.9 ppbv which is nearly 60 % of the values observed during 1994–95. Small but statistically significant decrease in noon-time peak ozone concentration is also observed. Noon-time peak ozone concentration has decreased from 34±13 ppbv during 1993–96 to 30±15 ppbv during 2010–11. NO$_X$ mixing ratios are very low over Gadanki. In spite of low NO$_X$ values (0.5 to 2 ppbv during 2010–11), ozone mixing ratios are not significantly low compared to many cities with high NO$_X$. The monthly mean ozone mixing ratio varies from 9 ppbv to 37 ppbv with high values during Spring and low values during late Summer. Using a box-model, we show that presence of VOCs is also very important in addition to NO$_X$ in determining ozone levels in rural environment and to explain its seasonal cycle.

Keywords Ozone · Trace-gases · NO$_X$ · Atmospheric chemistry · Rural India · Oxides of nitrogen

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1 Introduction

Ozone plays an important role in the earth’s atmosphere and has both good and bad effects on all lives on the earth either directly or indirectly. Some of the well known effects are: (1) its presence in the troposphere deteriorates air quality and thereby has harmful effects on health (National Research Council 1991; WHO 2003), while its presence in the stratosphere protects lives from harmful ultraviolet radiation (van der Leun et al. 1998); (2) it contributes to global warming by absorbing the terrestrial radiation in troposphere (Fishman et al. 1979; Forster et al. 2007); (3) High concentration of ozone in troposphere reduces crop yield and damages the natural ecosystems (Wang and Mauzerall 2004; Mohammed et al. 2012; Mohammed et al. 2013). Natural source of surface ozone is intrusion of stratospheric ozone (Ganguly and Tzanis 2011). However, large part of it is produced in-situ through complex photochemical reactions by oxides of nitrogen (NOX), which are also responsible for forming secondary aerosols and acid deposition (Seinfeld and Pandis 1998; Varotsos et al. 2012). This in turn has many negative effects such as corrosion or soiling of cultural and historical heritage buildings/structures (Tzanis et al. 2009; Tzanis et al. 2011).

Accordingly, the concentrations of ozone and NOX have been considered important for monitoring and controlling directly (Holland et al. 2005; Foster and Kumar 2011; Berman et al. 2012; CAAC 2013) and for assessing indirectly through transport processes linked with global warming and climate change related dynamics (Price et al. 1997; Walter and Heimann 2000; Ganzeveld 2002; Sanderson et al. 2003; Wiedingmyer et al. 2006). Production, loss and transport of ozone and NOX, however, are not homogeneous across the globe (Cartalis and Varotsos 1994; Chou et al. 2006; Jonson et al. 2006; Jaffe and Ray 2007; Tanimoto 2009; Tanimoto et al. 2009; Tripathi et al. 2012; Engardt 2008) and thus their monitoring from different parts of the globe, both urban and rural have assumed importance.

In the above context, several studies were initiated in India to study tropospheric ozone and its precursors (Naja and Lal 1996; Naja et al. 2003; Beig et al. 2007; Lal 2007; Ghude et al. 2008; Purkait et al. 2009; Kumar et al. 2010; David and Nair 2011; Ali et al. 2012; Ganguly 2012; Swamy et al. 2012; Ganguly and Tzanis 2013). However, very little has been done from rural India. One observational program, under the auspices of Indian Space Research Organisation’s Geosphere-Biosphere Program (ISRO-GBP), to measure surface ozone, NOX, CO and CH4 was initiated from Gadanki (13.48° N, 79.18° E), a rural site in South India and observations were made during 1993–96 (Naja and Lal 2002). Realising the importance of these measurements, we have re-initiated a long-term observational program by setting up the first comprehensive observatory of the ‘Indian Climate Observatory Network’ (known as ICON) over Gadanki in 2010.

In this paper, we present new observations on ozone and NOX concentrations, compare these with those observed in the past (1993–1996) and satellite based observations and discuss these results in the light of current understanding on the concentration of ozone from the rural environment of Gadanki. The paper is organised as follows. In section 2, we describe the rural site and the meteorological conditions prevailed during the observational period. In section 3, we describe instruments and data-sets used in this study. In section 4, we present the results and discuss the main findings in section 5.

2 Site description and meteorology

Observations presented in this work are from the National Atmospheric Research Laboratory (NARL) located at Gadanki (13.48° N, 79.18° E), a rural site in the Southern part of India.
NARL operates a large number of instruments, both ground-based remote sensing (viz. radio, optical, acoustic) and ground-based and balloon-borne sensors to measure atmospheric and ionospheric parameters. The laboratory is surrounded by hills having heights of ~750 m. Observations of trace-gases were carried out from Gadanki, the first climate observatory of ICON. The observatory is located on a hillock, a place whose altitude is the highest in the NARL campus. The analysers are kept on the top floor of the ICON building. There is no major industrial activity near the observational site except for the road connecting Tirupati and Bangalore, where the traffic is low. The aerial distance from the road to the observation site is 1.5 km and height difference between the road and the place of observation is about 50 m. The location is approximately 390 m above mean sea level.

Gadanki experiences tropical wet climate. Monthly mean relative humidity (RH), temperature, wind speed, wind direction and monthly total rainfall over Gadanki for 2010–11 are shown in Fig. 1. RH over Gadanki varies from 45 % in spring (pre-monsoon) to 90 % in winter (north-east monsoon period). Monthly mean temperature varies from 31ºC (from end of spring to beginning of summer) to 22ºC (in winter). Gadanki experiences two rainy seasons; one during south-west monsoon (June to September) and another during north-east monsoon (October to December). The period from January to May has minimum amount of rainfall. Total rainfall during 2010 and 2011 was 1,151 mm and 882 mm, respectively. The dominant wind directions close to surface are south-westerly during June to September, north-easterly during October to December and south-easterly during February to April. The wind speeds are generally higher during south-west monsoon with monthly mean values reaching as high as 2.3 m/s.

![Fig. 1](Fig. 1) Monthly mean temperature, relative humidity, wind speed, wind direction and monthly total rainfall over Gadanki for 2010–2011
3 Instruments and data

Surface concentrations of O$_3$ and NO$_X$ were measured continuously using on-line analysers (Model: 49i for O$_3$, and 42i for NO$_X$, Thermo Scientific, USA) since January 2010. Ambient air is drawn through a 293 cm long inlet running from the top of the building to a glass manifold located inside a room. The ozone analyser has in-built calibration unit for the span and zero checks. Ozone analyser works on the principle of Beer-Lambert-Baugher’s law, which relates absorption of light to the concentration of species. UV light of wavelength 254 nm is used as light source where ozone has strong absorption. Zero check for the ozone analyser is done once in a week. The span checks were done four times during the study period (2010–11). The changes in the consecutive calibrations have been very small. The zero offset reduced by 0.1 ppbv and span constant reduced by 2 % during the study period. The lowest detection limit of the analyser is 1 ppbv and the response time is 20 s. The accuracy of the ozone analyser is ± 5 % for an ozone concentration of 60 ppbv.

The NO$_X$ analyser works on the principle of chemiluminescence. The analyser uses molybdenum converter to convert NO$_2$ into NO and the intensity of light emitted in the reaction of NO with O$_3$ is related to total amount of NO$_X$. The response time is 60 s and the lowest detection limit is 0.4 ppbv for one minute time resolution. Molybdenum converter can also convert other nitrogen containing substances to NO and hence NO$_2$ measurement by this analyser should be regarded as upper limit. The analyser is equipped with facilities of zero and span calibrations. The zero calibration checks are done on weekly basis. Span calibration checks were done three times during the study period. Here too there has been very small differences in consecutive calibrations. Average differences of consecutive calibrations have been zero for NO$_X$ analyser and variabilities of calibration constants have been of the order of 7 %.

To a small extent we have used total irradiance data from Pyranometer operated from ICON terrace. In addition to our own data, GOME (Global Ozone Monitoring Experiment) and OMI (Ozone Monitoring Instrument) satellites’ tropospheric NO$_2$ data (Boersma et al. 2004; Boersma et al. 2007) have also been used to study NO$_2$ trend over South India. GOME instrument was launched onboard ERS-2 satellite of European Space Agency in 1995. It was in operation from 1996 to 2003. The tropospheric NO2 retrieval from GOME has precision of the order of 35 to 60 % depending upon location. OMI instrument was launched in 2004 on board of NASA’s Aura satellite. The OMI data are available globally at 0.125° X 0.125° grid resolution. OMI data has error of the order of 0.7 x 10$^{15}$ molec/cm² which is about 40 % over Southern India.

4 Results and observations

4.1 Ozone variability

Figure 2a shows diurnal variations of surface ozone for four seasons: spring (March-May), summer (June-August), autumn (September-November) and winter (December-February). The peak concentration is found to be in the afternoon and low concentration just before the sunrise. The time of peak concentration changes from 16:00 h in spring to 14:00 h in autumn. Observations also suggest that ozone mixing ratio builds up (at the rate 1 to 3.5 ppbv per hour on average depending on season) during 7-14/16 LT and decreases rather slowly during 14/16-7 LT. The diurnal range of mixing ratio is found to be in the range of 15–47 ppbv in spring, 14–31.5 ppbv in winter, 11.5 - 22.8 ppbv in summer and 11.4 - 20.8 ppbv in autumn. These results suggest that the ozone mixing ratio maximises in spring followed by winter, summer and autumn.
Figure 2b shows monthly mean ozone mixing ratios for the period January 2010-December 2011. Monthly mean mixing ratios varies from 9 ppbv to 37 ppbv and follows a clear seasonal cycle. It is generally higher during March-May and low during August-September. Variability of ozone mixing ratio represented by standard deviation is also found to be high during March-May and low during August-September.

4.1.1 Comparison of Ozone levels with other places

In order to compare these results with those observed from other locations in India, we show monthly average surface ozone for few places in Fig. 3. Inset figure depicts the map of locations from where observations were made. Sources of data for other stations are as follows: Nainital (Kumar et al. 2010), Ahmedabad (Lal 2007), Pune (Beig et al. 2007), Hyderabad (Swamy et al. 2012) and Thiruvananthapuram (also known as Trivendrum) (David and Nair 2011). Trivandrum has very different seasonal variation in ozone compared to Gadanki and other stations shown in the figure. Trivandrum experiences maximum ozone mixing ratio during winter whereas Gadanki and other stations are experiencing maximum [O₃] during spring. Trivandrum is located on west coast of India. Because of its geographic location, Trivandrum experiences minimum RH and rain-fall during February and soon after that RH starts building up due to air-masses coming from
Indian Ocean and Arabian Sea. Whereas, in-land stations including Gadanki experience minimum RH during spring. RH and ozone are known to be negatively correlated (Camalier et al. 2007). There is steep rise in ozone concentration soon after the minimum during monsoon season over most of the stations whereas in case of Gadanki, ozone concentration during monsoon and winter is more or less same. This is again could be linked to rain-fall and RH variation. Whereas other stations do not get rain-fall in winter, Gadanki receives large fraction of annual rain-fall during winter. Ozone mixing ratio over Gadanki is higher or comparable to some of the major cities of India for e.g. Kolkata (Purkait et al. 2009), Ahmedabad, Delhi (Lal 2007) and Thrivanthpuram (David and Nair 2011). However, these cities have 3 to 10 times higher [NOX] compared to Gadanki. The cities which have ozone mixing ratio higher than Gadanki are Hyderabad (Swamy et al. 2012) and Pune (Beig et al. 2007; Lal 2007; Londhe et al. 2008). High altitude remote sites like Mt. Abu (Naja et al. 2003) and Nainital (Kumar et al. 2010) have higher ozone mixing ratio compared to Gadanki. However, the diurnal variation of ozone over Gadanki is quite different from Nainital and Mt. Abu where noon-time peak is not very prominent. This is indicative of in-situ ozone production through photochemical reactions rather than being a place of high background ozone level.

4.2 NOX variability

NOX (NO+NO2) mixing ratio over Gadanki is relatively low. To have better insights in observational skewness and variance, we have plotted median values of [NOX] (circles) and its inter-quartile range (vertical bars). Diurnal variation with hourly median values is shown in Fig. 4a. NOX has high mixing ratio in morning and evening and low mixing ratio in afternoon. The morning peak mixing ratio is higher than evening peak mixing ratio. Monthly median variation of [NOX] is shown in Fig. 4b. It varies from 0.5 ppbv to 2 ppbv. Being very low values, it is not possible to see seasonal variation as clearly as for ozone nevertheless increase during winter and decrease during summer is discernible.
5 Results and discussion

5.1 Comparison between past and present observations

As noted earlier, (Naja and Lal 2002) have carried out observations of $O_3$, NOX, CO and CH4 over Gadanki between 1993 and 1996. The observations of $[O_3]$ were made from November 1993 to December 1996 while observations of [NOX] were made from January 1994 to December 1995. For the sake of brevity, observations reported in (Naja and Lal 2002) are referred as past observations and the observations reported in preceding section are referred as present observations.

Average noon time maximum $[O_3]$ is $30 \pm 15$ ppbv for the period 2010–11 and it is $34 \pm 13$ ppbv for the period 1993–96. The difference is significant at confidence level of 99.9% though the satellite based tropospheric $[O_3]$ trend is insignificant. The comparison of monthly mean $[O_3]$ for 2010–11 and 1993–96 is shown in Fig. 5. The difference between monthly mean of all data as well as monthly mean of noon-time peak concentration is statistically significant at confidence level of 99% except for the March. Overall, the present $[O_3]$ is less than the past and the difference between two periods is not uniform across all the months. Winter months are having bigger differences than spring and summer months. April and May have present $[O_3]$ higher than the past $[O_3]$. The high values of present $[O_3]$ during April and May are mainly due to night-time high $[O_3]$ during these months as can be seen in the Fig. 6b.
A comparison of diurnal variations of $[O_3]$ for the past and present is shown in Fig. 6 for different seasons. Night-time $[O_3]$ have relatively less seasonal variation and small differences between past and present values. In case of day-time mixing ratios, past $[O_3]$ values are significantly higher than the present $[O_3]$ for Winter and Autumn. The amplitude of diurnal cycles for all the seasons are more in past observations compared to present observations. Winter season has highest amplitude difference of 13.4 ppbv between past and present followed by autumn (12.7 ppbv), spring (6.6 ppbv) and summer (0.33 ppbv). This indicates there is a decrease in photochemically produced ozone during recent years. The morning rate of change of ozone is high (7 ppbv per hour) during 1993–96 compared to 2010–11 (4 ppbv per hour) as shown in Fig. 7. Asymmetry between morning and evening rate of change of ozone is characteristics of rural environment (Naja and Lal 2002).

Fig. 5 Comparison of monthly mean ozone mixing ratios between past (1993–1996) and present (2010–2011) observations

Fig. 6 Comparison of seasonal diurnal patterns of surface ozone over Gadanki for the past and present observations
There has been very few studies that report ozone trend over regions of India. Naja and Lal (1996) have reported a significant increase in [O$_3$] over Ahmedabad – a major city in western part of India. Ali et al. (2012) have reported increase in [O$_3$] over Delhi but decrease over Pune for the period between 1990 and 1999. As noted earlier, ozone trends are not uniform over globe. Cartalis and Varotsos (1994) have reported increasing trend of ozone over Athens, Greece during last century. Chou et al. (2006) have reported increasing trend of ozone over Taipei, Taiwan during 1994 to 2003 in spite of decrease in volatile organic compounds, NO$_X$ and non-methane hydrocarbon. They attribute increase in ozone to reduced titration of ozone by NO. Jaffe and Ray (2007) have studied ozone trends for period from 1987 to 2004 over 11 rural and remote sites in north and western US including Alaska. They have found increasing trend over seven out of eleven sites. (Tanimoto 2009; Tanimoto et al. 2009) have studied ozone trends over several remote sites in Japan. They have found overall increasing trend over all the sites with mountain sites having higher increasing trends. They attribute increasing trend to increase in atmospheric pollution in Asian countries and their long-range transport to Japan. Fiore et al. (1998) studied ozone trends over 549 sites across contiguous United States and found no significant increase in ozone during 1980 to 1995 period. Decreasing trends are predominantly observed over urban regions attributable to decrease in NO$_X$ and VOCs. Jonson et al. (2006) have studied trends of ozone and its precursor gases using observations and model. They have found significant decrease in summer time ozone over Europe during 1990–2002, however, the decrease in annual averages is less than expected from reduction in precursor gases. Tripathi et al. (2012) have studied ozone trends over eight sites across Ireland from 1994 to 2009. They have found either negative trend in peak ozone concentration or no increasing trend. In majority of cases ozone trends when decreasing attributed to new pollution control norms to reduce precursors like NO$_X$ and when increasing attributed to increase in precursor gases or long-range transport of pollutants. In one instance increase in [O$_3$] is linked to decrease in [NO] (Chou et al. 2006).

The comparison of monthly median [NO$_X$] for the two periods is shown in Fig. 8. The present [NO$_X$] are nearly half of the past values. Statistical significance was determined using $t$-test and it is found that the difference is statistically significant at the level higher than 99.9% for all the months. Absolute value of difference is of the order of 0.9 ppbv except for October, November, December and February. The difference during October, November, December and February is 0.5, 0.5, 0.6 and 0.6 ppbv respectively. No comparison could be made for...
September. There are several places around the world e.g. the USA, Europe, etc. where a decreasing trend in NOX is observed (Akimoto 2003). However, these changes are mostly in the western world. In Asia, mostly increasing trends in NOX or NO2 are reported (Akimoto, 2003; Sheel et al. 2010; Hilboll et al. 2013). Main reason for increase in NOX or NO2 mixing ratios over Asian region is increase in industrial and vehicular emissions. However, rural regions have different economic activities compared to urban regions and far less industrialization and vehicular emissions. Over a rural location, the major sources of NOX are soil emission, lightning activity, nitrogen-based fertilizer and biomass-burning. It is difficult to say at this stage whether the decrease in concentration is part of continuous decreasing trend or a difference between anomalous years. Nevertheless, one can conclude with great degree of confidence that NOX mixing ratio is not increased over this region in contrast to the trends observed over urban regions of Asia.

To look into the aspect whether the ‘decrease’ in NOX is a localized phenomenon or a phenomenon applicable to a larger region, we have used GOME and OMI satellite data (Boersma et al. 2004; Boersma et al. 2007). GOME and OMI are two different sets of satellite sensors and there is a difference in spatial resolution of two data-sets as well. This requires special care in calculating trends from combined data set. Different sensor characteristics and spatial resolution can produce differences in mean and variability. We have followed the method described in (Mieruch et al. 2008) and (Hilboll et al. 2013) to calculate bias and trends. While the decrease in NO2 values is not seen over Gadanki as one might have expected from ground-based data of NOX, satellite data show statistically significant decreasing trend in west of Gadanki and increasing trend in East and South of Gadanki with Gadanki in transition zone (Fig. 9).

The trend analysis for entire South India is shown in Fig. 9b. Grid-boxes having a trend, which is statistically significant at confidence level 95 % or more are shown with black dots. There are few hotspots with very high increasing trends viz: Chennai, Vizag, Bellari, Madurai. This may be due to recent industrialisation of these places. Besides hot-spots, there are regions of increasing trend. The spatial pattern of increasing trends follows geographical pattern of river basins in South India known for intense agricultural activities. Increase in use of fertilizers can be one of the reasons for increasing trend over these regions. However, more interesting is the decreasing trends observed between mountain ranges known as Eastern and Western Ghats. The decrease in tropospheric NO2 over this region is found in-spite of increase in population and vehicular traffic. Possibly the decrease over this region is linked with changes in land-use pattern since soil emissions are one of the major source of NO2 in the atmosphere. Sheel et al. (2010) and Hilboll et al. (2013) have reported increasing trend over urban centres in India. Ghude et al. (2008) studied NO2 hotspots and trends over five regions of India using GOME and SCIAMACHY data between year 1996 to 2006 and have reported increasing trends. They attribute increase in NO2 to rapid industrialisation and vehicular traffic.

![Fig. 8](Author's personal copy)  
Comparison of monthly median of NOX mixing ratios during 1994–95 with that observed during 2010–11. Vertical bars represent inter-quartile range.
growth. However, there exist high degree of heterogeneity in NO$_2$ trends over sub-regional scale. Out of all the regions studied by Ghude et al. (2008), the minimum trend was found over South India, 1.38±0.12 % per year. They have also found far less number of hot-spot over

Fig. 9 (a) Monthly mean tropospheric column NO$_2$ concentration observed over Gadanki using GOME (1996–2003) and OMI (2005–2011). (b) Trends in tropospheric NO$_2$ over South India calculated for the period between 1996 to 2011 after combining GOME and OMI data. Grid-boxes with black dots show statistically significant trend at confidence level 95 % or more. Trends are calculated using method described in (Hilboll et al. 2013) and (Mieruch et al. 2008)
South India compared to other parts of India. Since regulations and population dynamics are not significantly different from one region to other region in India such heterogeneity in trends, we believe is driven by geographic condition of the place and factors such as land-use change.

Though there is a significant decrease in surface \([\text{NOX}]\), decrease is less for surface \([\text{O}_3]\). Photochemical production of ozone depends on \([\text{NO}]\) and \([\text{NO}_2]\) in complex manner. In such a low \([\text{NOX}]\) environment, other factors such as presence of VOCs play very important role in determining ozone-production-efficiency of \(\text{NOX}\) molecules. Using numerical simulations, we try to understand seasonal variation of \([\text{O}_3]\), its high mixing-ratio in spite of low \([\text{NOX}]\) and level of decrease in \([\text{O}_3]\) expected based on \([\text{NOX}]\) decrease.

5.2 Numerical simulation of Ozone at Gadanki

Our objective in carrying out the numerical simulations is to understand seasonal variation of \([\text{O}_3]\), the high ozone values over rural areas of India in spite of low \([\text{NOX}]\) in comparison to big cities and the role of \([\text{NOX}]\) decrease on \([\text{O}_3]\) with respect to past values. It is not expected that the numerical simulation will exactly reproduce observed \([\text{O}_3]\) since VOC values are not based on observations over the place, cloudiness is over simplified, the \([\text{NO}]\) are near detection limit of the instrument and hence not constrained for diurnal variation, and transport processes are not considered. Nevertheless the simulations are helpful in understanding the role of \(\text{NOX}\) in diagnostic manner and to identify gaps in observational strategy to better understand the trends. Numerical simulation of ozone concentration over Gadanki are carried out using NCAR Master Mechanism (NCAR-MM) Box (0D) model (Madronich and Calvert 1990; Aumont et al. 2000; Stroud et al. 2003; Madronich 2006). The model consists explicit and detailed gas phase chemistry combined with box model solver. Photolysis rates are calculated using TUV (Tropospheric Ultraviolet and Visible) radiation model included with NCAR-MM. The model has ability to simulate 5,000 reactions among 2,000 species (Madronich 2006). The model is developed at National Center for Atmospheric Research (NCAR), USA and can be downloaded from NCAR/UCAR Community data portal [http://cdp.ucar.edu/].

We have run the model for various sets of inputs and simulated the \([\text{O}_3]\) and \([\text{NO}]\) on diurnal cycle. Each simulation is done for 48 h and the values from the second cycle of 24 h are reported in this study. In Fig. 10, noon-time peak ozone mixing ratios from various model runs are compared with observations made during (a) 2010–11 and (b) 1993–96. Inputs for various model runs are described in Table 1. The model run M-01 has set of inputs which are very close to observed values during 2010–11 or climatological values over Gadanki and assuming zero VOCs. \([\text{NO}_2]\) for M-01 is fixed to monthly mean diurnal variation during 2010–11. Initial \([\text{O}_3]\) is set to 21 ppbv which is close to observed night time \([\text{O}_3]\) over Gadanki. Diurnal variation of boundary layer height (BLH) over Gadanki is taken from (Basha and Ratnam 2009). Four different sets of BLH variations are used for the four seasons. In order to simulate effect of cloudiness, we have used pyranometer data but in indirect way. Clouds affect surface \([\text{O}_3]\) because of their ability to control amount of surface reaching solar radiation. The daily mean surface reaching solar energy is maximum in April with value 6.6 kWh/m². Simulations are carried out assuming clear sky fraction equal to ratio of daily mean solar energy for a given month to daily mean solar energy received in April. This approach makes April month simulations as fully clear sky day simulation and for other months the simulations are for partially cloudy sky in relation to April simulation. To understand the role of VOCs for tropical climate, in the model run M-02, ozone simulations are carried out keeping all the inputs same as M-01 except for VOCs. Since no observations of VOCs are available for this region, we have used diurnally varying isoprene values reported in (Karl et al. 2007). Karl et al. (2007) have studied diurnal variation of isoprene and monoterpenes over
Central Amazonian which has the tropical climate. Model run M-03 is similar to M-02 but the Isoprene values are reduced by factor 10. Model runs M-04 to M-06 are similar to the model runs M-01 to M-03 but the [NO] and [NO$_2$] are scaled by factors such that [NO$_X$] matches with values reported in (Naja and Lal 2002) for year 1994 to 1995. The multipliers are of the order of 2. Besides these model runs, we have done several runs to study sensitivity to various input parameters. It is found that the noon-time peak [O$_3$] is highly sensitive to [NO], [NO$_2$], isoprene and cloudiness. It is moderately sensitive to CO, aerosol properties, boundary layer height and columnar ozone concentration.

**Fig. 10** Comparison of monthly mean noon-time peak ozone mixing ratios between box-model (NCAR-MM) and observations for year (a) 2010–11 and (b) 1993–96. For all the model runs NO$_2$ concentration is constrained to monthly mean observations. Model run M-01 and M-04 are without any VOCs. M-02 and M-05 are with Isoprene values based on (Karl et al. 2007) M-03 and M-06 are with Isoprene values equal to 1/10 of the values used in M-02 and M-05. (c) Difference between past and present noon-time peak ozone mixing ratios for observations and models.
Presence of isoprene in the atmosphere is very important in determining peak [O₃]. In the absence of isoprene, simulated peak [O₃] is significantly smaller than the observed peak [O₃] for both the present and the past [NO₂] scenarios. In the absence of isoprene, it is also not reproducing the observed seasonal cycle of [O₃]. When the diurnally varying but seasonally fixed isoprene concentration as reported in Karl et al. (2007) is included, the simulated peak [O₃] is moderately higher than the observed peak [O₃] for the first half of year but significantly higher than the observed peak [O₃] for second half of the year in case of present day scenario (M-02 in Fig. 10). This is because inclusion of isoprene in simulations leads to seasonal cycle.

### Table 1  Inputs for numerical simulations of ozone concentration over Gadanki

<table>
<thead>
<tr>
<th>Model Runs</th>
<th>Inputs</th>
<th>Input Sr. No.</th>
<th>Values or basis of values</th>
</tr>
</thead>
<tbody>
<tr>
<td>M-01</td>
<td>Ozone (Initial conc.)</td>
<td>1</td>
<td>4.9628 x 10⁻¹¹ molec./cm⁻³ (21 ppbv)</td>
</tr>
<tr>
<td></td>
<td>Ozone columnar</td>
<td>2</td>
<td>247 DU</td>
</tr>
<tr>
<td></td>
<td>Ozone background</td>
<td>3</td>
<td>Zero</td>
</tr>
<tr>
<td></td>
<td>CO</td>
<td>4</td>
<td>8.2115 x 10⁻¹² molec./cm⁻³ (352 ppbv)</td>
</tr>
<tr>
<td></td>
<td>CH₄</td>
<td>5</td>
<td>4.0473 x 10⁻¹³ molec./cm⁻³ (1,736 ppbv)</td>
</tr>
<tr>
<td></td>
<td>Temperature</td>
<td>6</td>
<td>Obs. mean diurnal variation for given month</td>
</tr>
<tr>
<td></td>
<td>H₂O</td>
<td>7</td>
<td>Obs. mean diurnal variation for given month</td>
</tr>
<tr>
<td></td>
<td>N₂</td>
<td>8</td>
<td>1.8415 x 10⁻¹⁹ molec./cm⁻³</td>
</tr>
<tr>
<td></td>
<td>O₂</td>
<td>9</td>
<td>4.8951 x 10⁻¹⁸ molec./cm⁻³</td>
</tr>
<tr>
<td></td>
<td>Aerosol Optical Depth</td>
<td>10</td>
<td>0.3</td>
</tr>
<tr>
<td></td>
<td>Single scattering albedo</td>
<td>11</td>
<td>0.92</td>
</tr>
<tr>
<td></td>
<td>Angstrom Exponent</td>
<td>12</td>
<td>1.13</td>
</tr>
<tr>
<td></td>
<td>Surface albedo</td>
<td>13</td>
<td>0.08</td>
</tr>
<tr>
<td></td>
<td>NO (initial conc.)</td>
<td>14</td>
<td>Zero. Diurnal variation during 2010–11 in their respective months.</td>
</tr>
<tr>
<td></td>
<td>NO₂ (diurnally constrained)</td>
<td>15</td>
<td>Obs. monthly mean diurnal variation during 2010–11 in their respective months.</td>
</tr>
<tr>
<td></td>
<td>Boundary Layer Height (BLH)</td>
<td>16</td>
<td>Diurnally varying and season dependent. Based on (Basha and Ratnam 2009).</td>
</tr>
<tr>
<td></td>
<td>Clear Sky Fraction</td>
<td>17</td>
<td>Diurnally fixed but varies monthly based on Fig. 12.</td>
</tr>
<tr>
<td>M-02</td>
<td>Inputs 1 to 17</td>
<td>18</td>
<td>Same as in M-01</td>
</tr>
<tr>
<td></td>
<td>Isoprene (diurnally constrained)</td>
<td></td>
<td>Diurnally fixed to the values reported in (Karl et al. 2007); Same for all months</td>
</tr>
<tr>
<td>M-03</td>
<td>Inputs 1 to 17</td>
<td></td>
<td>Same as in M-01</td>
</tr>
<tr>
<td></td>
<td>Isoprene</td>
<td></td>
<td>One tenth of the values used in M-02</td>
</tr>
<tr>
<td>M-04</td>
<td>Inputs 1 to 13 and 16 to 18</td>
<td></td>
<td>Same as in M-01</td>
</tr>
<tr>
<td></td>
<td>NO (initial conc.)</td>
<td></td>
<td>M-01 NO input value being scaled such that (NO+NO₂) will match (Naja and Lal 2002).</td>
</tr>
<tr>
<td></td>
<td>NO₂</td>
<td></td>
<td>- as above -</td>
</tr>
<tr>
<td>M-05</td>
<td>Inputs 1 to 17</td>
<td></td>
<td>Same as in M-04</td>
</tr>
<tr>
<td></td>
<td>Isoprene</td>
<td></td>
<td>Same as in M-02</td>
</tr>
<tr>
<td>M-06</td>
<td>Inputs 1 to 17</td>
<td></td>
<td>Same as in M-04</td>
</tr>
<tr>
<td></td>
<td>Isoprene</td>
<td></td>
<td>One tenth of the values used in M-05</td>
</tr>
</tbody>
</table>
with two maxima in simulated values, one during spring and another during autumn. However, in-case of observations, only spring-time maxima is observed. This could be because of seasonally fixed isoprene but seasonally varying $[\text{NO}_2]$ and solar radiation. It is worth noting that spring is the season of biomass burning (agricultural waste burning in open and forest fires) in South India which can be a potential source of various VOCs that may participate in ozone production but in winter there is very little biomass burning activity. Hence, one can expect a seasonally varying VOC concentration over South India.

The simulated peak $[\text{O}_3]$ is very high in comparison to observed values for the past $[\text{NO}_2]$ when isoprene concentration at the same level as in reported in Karl et al. (2007) are used (M-05 in Fig. 10b). When isoprene concentration is scaled down to 1/10 of values being used in the run M-02 and M-05, the simulated peak $[\text{O}_3]$ is also reduced but still higher than the past observations.

The differences in simulation output, which include isoprene are considerably greater than observed differences (Fig. 10c). At the same time, absence of isoprene in simulation inputs (model runs M-01 and M-04) is also not able to explain differences as they are found to be reverse way that is past low and present high $[\text{O}_3]$. There exists a possibility that isoprene or VOC concentrations may not have been same in past and present. In absence of observations of VOC it will be difficult to quantify role of $[\text{NO}_X]$ and effect of its decrease on ozone concentrations, nevertheless the simulations bring-out the importance of VOCs in rural atmospheric chemistry and their role in high ozone level in spite of low $[\text{NO}_X]$.

6 Summary

Surface level $\text{NO}_X$ and $\text{O}_3$ observations are carried out over a rural location in Southern India. Though the $[\text{O}_3]$ is not very low compared to major cities of India, concentration of $\text{NO}_X$ is quite low and typical of a rural background site. The ground-based measurements of $[\text{NO}_X]$ from period 2010–11 are nearly half of the past values (1994–95) in relative terms and about 0.9 ppbv low in absolute terms. A statistically significant decreasing trend in tropospheric columnar $[\text{NO}_2]$ is also found in GOME and OMI satellite data over several parts of the South India. This is in contrast to urban places and fertile river basins where these satellite sensors have detected increasing trend. A small but statistically significant decrease is also observed in $[\text{O}_3]$ compared to past. The decrease is observed mostly in day-time ozone concentration indicative of decrease in photochemically produced ozone. The decrease is high during winter. A likely cause of decrease of $[\text{O}_3]$ is a decrease of $[\text{NO}_X]$, however magnitude of decrease of $[\text{O}_3]$ is less than expected from decrease in $[\text{NO}_X]$.

Presence of VOCs is very important to explain not only the high $[\text{O}_3]$ in spite of low $[\text{NO}_X]$ but also the observed seasonal cycle to some extent. Though we have used only isoprene, it fairly outlines the importance of VOCs for rural atmospheric chemistry. Overall our results indicate that the atmospheric chemistry and trends of trace-gases such as $\text{O}_3$ and $\text{NO}_X$ in rural India where a major bulk of population lives are distinctly different from urban India.

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References


CAAC: Twelfth five-year plan on air pollution prevention and control in key regions (China clean air policy briefing no. 1) (English Translation), Clean Air Alliance of China, Secretariat for Clean Air Alliance of China, suit 1705, Building 1, Park Avenue, 16 Jianguomenwai Street, Beijing 100022 (2013)


National Research Council, U., Rethinking the ozone problem in urban and regional air pollution, (1991)


Evaluation of black carbon emission inventories using a Lagrangian dispersion model – a case study over southern India

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Abstract. We evaluated three emission inventories of black carbon (BC) using Lagrangian particle dispersion model simulations and BC observations from a rural site in southern India (Gadanki; 13.48°N, 79.18°E) from 2008 to 2012. We found that 93 to 95 % of the BC load at the observation site originated from emissions in India and the rest from the neighbouring countries and shipping. A substantial fraction (33 to 43 %) of the BC was transported from northern India. Wet deposition is found to play a minor role in reducing BC mass at the site because of its proximity to BC sources during rainy season and relatively short rainy season over western and northern parts of India. Seasonally, the highest BC concentration (approx. 3.3 µg m⁻³) is observed during winter, followed by spring (approx. 2.8 µg m⁻³). While the model reproduced well the seasonal cycle, the modelled BC concentrations are significantly lower than observed values, especially in spring. The model bias is correlated to fire radiative power – a proxy of open biomass burning activity. Using potential emission sensitivity maps derived using the model, we suggest that underestimation of BC mass in the model during spring is due to the underestimation of BC fluxes over southern India (possibly from open-biomass-burning/forest-fires). The overall performance of the model simulations using three different emission inventories (SAFAR-India, ECLIPSE and RETRO) is similar, with ECLIPSE and SAFAR-India performing marginally better as both have about 30 % higher emissions for India than RETRO. The ratio of observed to modelled annual mean BC concentration was estimated as 1.5 for SAFAR, 1.7 for ECLIPSE and 2.4 for RETRO.

1 Introduction

Black carbon (BC) is a component of soot, which is responsible for the absorption of visible light (Yasa et al., 1979). It is emitted into the atmosphere as a consequence of incomplete combustion processes like biofuel burning, running of inefficient diesel engines, forest fires, etc. Unlike other aerosols, BC aerosols are responsible for positive radiative forcing which is comparable to forcing by major greenhouse gases (Haywood and Ramaswamy, 1998; Jacobson, 2001; Bond et al., 2013). Presence of BC in the atmosphere also affects the hydrological cycle of Earth and regional climate (Ackerman et al., 2000).

Understanding the sources of BC, their geographical distribution and future changes is therefore important to improve climate modelling and would support development of policies exploring climate co-benefits of air pollution regulation controlling sources of BC. However, global BC emissions estimates are highly uncertain. Dickerson et al. (2002) estimated BC emissions of South Asia at between 2 and 3 Tg in year 1999 using the BC/CO ratio – a factor of 2 to 3 higher than bottom-up BC inventories, suggesting significant underestimation of BC sources in South Asia. The range of global BC emissions has been reported as 4 to 13 Tg yr⁻¹ (Bond et al., 2013). Emissions from India contribute 7 to 14 % of global BC emissions (Bond et al., 2004; Schultz and Rast, 2007; Klimont et al., 2009, 2015a, b) and observed BC concentrations over India are significantly higher than in other regions (Suresh Babu and Moorthy, 2002; Suresh Babu et al., 2002; Ganguly et al., 2005, 2006a, b; Jayara-
Figure 1. Monthly precipitation amounts over Gadanki during 2009 and 2011.

Figure 2. Monthly median fire radiative power values obtained from MODIS satellite for whole India and peninsular India (south of 18° N latitude).

Open biomass burning has a well characterized seasonal cycle over India (Joseph et al., 2009). Fire radiative power (FRP) is a measure of radiative energy emitted per unit time in a fire event. Its value is proportional to the amount of material being burnt in the fire event. Fires detected using MODIS satellite sensor are characterized by FRP values using an empirical formula based on difference in brightness temperature at 4 µm with respect to non-fire pixels in the vicinity (Giglio et al., 2003; Justice et al., 2006; Davies et al., 2009). In Fig. 2, long-term (2000–2013) monthly median FRP values over the southern part of India (south of 18° N latitude; henceforth referred as Peninsular India) and over the whole of India are shown. FRP is high during February to May and low during June to September. The largest differences in the seasonal variation of FRP between Peninsular India and whole India occur during October to November. As mentioned before, Peninsular India where the observations are carried out experiences two rainy seasons whereas North, West and Central India experiences only one rainy season. The northeast monsoon brings rain over Peninsular India during winter and reduces the number of fire events and hence FRP whereas absence of rain results in high FRP over other parts of India.

2 Site description

Observations of BC have been carried out at the climate observatory of the National Atmospheric Research Laboratory in Gadanki. Gadanki (13.48° N and 79.18° E, 365 m a.s.l.) is a typical rural site in southern India, with no major industrial activities in the near vicinity. Gadanki has tropical wet climate and experiences a prolonged rainy season from both southwest and northeast monsoons unlike the northern and western parts of India. Monthly rainfall patterns over Gadanki for the years 2009 and 2011 are shown in Fig. 1. February to May is mostly dry. The rainy season starts in June and goes on until December with short lulls in between. The maximum rainfall over Gadanki in the year 2009 was observed during November whereas in the year 2011 it occurred during August with a comparable rain amount in November. The year 2009 was officially declared as a drought year for the state of Andhra Pradesh (in which Gadanki is located), whereas 2011 was a normal year.

In this paper we examine the emission inventories RETRO (Schultz et al., 2007; Schultz and Rast, 2007), ECLIPSE (Klimont et al., 2013, 2015a, b) and SAFAR-India (Sahu et al., 2008) using the particle dispersion model FLEXPART (Stohl et al., 1998, 2005) driven by observed meteorological fields and suggest possible causes of the underestimation of BC concentrations by models over India.

3 Instrumentation and data

Equivalent BC (EBC) concentrations are measured using an aethalometer (Model AE31; Magee-Scientific, USA), which has seven wavelength channels centred at 370, 470, 520, 590, 660, 880 and 950 nm. In this study, we report EBC values based on 880 nm channel data as it has minimum interference from other species and is considered to be the standard channel for BC measurement with this technology (Hansen, 2005). Details of the instrument and the typical set-up used at Gadanki are reported in an earlier study (Gadhavi and Jayaraman, 2010). The ambient air is drawn with a typical flow rate...
of 2.9 L min$^{-1}$ for five minutes and passes through a quartz fibre filter fitted in an optical chamber. Changes in transmission of light through filter paper is monitored which is affected by accumulated deposition of light-absorbing particles on the filter paper. The changes in absorption coefficient of filter paper are converted to equivalent BC mass by dividing it with mass absorption cross-section 0.166 cm$^2$ µg$^{-1}$ (at 880 nm). Assuming that most of light absorption is due to BC at 880 nm, for the convenience of comparisons with the model simulations, we refer to these measurements as BC. The error in estimating BC concentration is expected to be less than 10% (Hansen, 2005; Gadhavi and Jayaraman, 2010 and references therein).

### 3.1 Emission inventory data

We have considered three emission inventories namely ECLIPSE, RETRO and SAFAR-India. The ECLIPSE (Evaluating the CLimate and air quality Impacts of Short-lived pollutants) global emission inventory has been developed using the GAINS Model (Greenhouse gas – Air pollution Interactions and Synergies Model; Amann et al., 2011). The sources considered range from wick lamps to thermal power stations, including residential combustion, transport, shipping, large combustion installations, industrial processes, waste and open burning of agricultural residues. This inventory does not include emissions from open biomass burning other than agricultural waste burning. Hence forest fire emissions are included from GFEDv3 (Global Fire Emissions Database; van der Werf et al., 2010). The ECLIPSE emission data set has been developed for the period from 1990 to 2050; the inventory extends to 2010 while the baseline projection until 2050 assumes implementation of existing environmental legislation and draws on the energy projection of the IEA – International Energy Agency’s Energy Technology Perspective 2012 (ETP2012) (Klimont et al., 2015a, b). In this work, emission values for the year 2010 from version 5 of the inventory are used. Version 5 was recently released and has about 44% higher emissions than version 4a inventory over India, mainly due to the addition of sources which were not previously considered (e.g. wick lamps). The original data set is available at 0.5$^\circ$ × 0.5$^\circ$ resolution including monthly resolution for several key source sectors; however in this study, the grid resolution has been reduced to 1$^\circ$ × 1$^\circ$. Emission fluxes from the ECLIPSE + GFED inventory are shown in Fig. 3a. Hereafter, if not specifically mentioned, reference to ECLIPSE inventory implies ECLIPSE + GFED. The total BC emissions of India in 2010 are estimated at 1233 Gg yr$^{-1}$ of which 52 Gg yr$^{-1}$ are from forest fire emissions based on GFED. The major contribution originates from the Indo-Gangetic Basin (IGB) in the north and in a few pockets on the western coast of India. In contrast, in South and Central India BC emissions are relatively low. Within IGB, emissions are higher in Bihar, West Bengal and Haryana states of India and Bangladesh (a map of India with state names is provided in the Supplement).

The RETRO emission inventory is the outcome of the project REanalysis of the TROpospheric (RETO) chemical composition over the past 40 years. The emission inventory for BC has two parts – one for anthropogenic emissions which includes biofuel burning, industrial combustion and
agricultural residue burning. BC emissions from forest fires over India are accounted for separately based on the RegFIRM model (Schultz et al., 2008). Schultz et al. (2008) had to reduce the literature values for carbon emissions per unit area over India to achieve consistency with reported emissions from the subcontinent which highlights inherent problems in the bottom-up inventory approach for emissions from biomass burning. The emission fluxes are monthly averages of BC in kg m$^{-2}$ s$^{-1}$ for each grid-box. Annual total BC emissions of India based on this inventory for the year 2010 are 697 Gg yr$^{-1}$ out of which 31 Gg yr$^{-1}$ are from forest fires. In Fig. 3b, differences between ECLIPSE and RETRO (ECLIPSE – RETRO) are shown, i.e. RETRO emission fluxes are lower than ECLIPSE emissions in all of South Asia. The difference is particularly high over the Bihar, West Bengal states of India, and Bangladesh and Myanmar.

Finally, we have considered the regional emission inventory SAFAR (System of Air quality Forecasting and Research)-India (Sahu et al., 2008). The SAFAR-India includes only anthropogenic emissions from fossil fuel, fuel wood, dung combustion and agricultural waste burning using district-level statistics on activities, population, farming, etc. In preparation of the inventory, Sahu et al. (2008) have used emission factors for biofuel combustion from Venkataraman et al. (2005), and emission factors for fossil fuel combustion are based on Cooke et al. (1999) for their “under-developed-countries” category. The inventory was updated after publication of Sahu et al. (2008). The latest inventory contains annual emissions for the years 1991, 2001 and 2011 at a spatial resolution of 1° × 1°. In this work, we have used emission values for 2011. Total BC emissions of India based on this inventory are 1119 Gg yr$^{-1}$. Although ECLIPSE and SAFAR inventory have comparable total emissions for India, their spatial and source distribution are significantly different. In Fig. 3c, spatial allocation differences between ECLIPSE and SAFAR (ECLIPSE – SAFAR) are shown. SAFAR inventory has comparable or marginally higher emissions in central, southern and western parts of India. Regions close to big cities like Mumbai, Delhi, Ahmedabad and Kolkata have significantly higher emissions in SAFAR compared to ECLIPSE. The opposite is true over Bihar, West Bengal and northeastern parts of India, where the ECLIPSE inventory is significantly higher than SAFAR. With respect to source distribution, the key difference is between large combustion plants (power plants and industrial boilers) and the residential sector. SAFAR estimates large BC emissions from power plant boilers while this source is very small in ECLIPSE. This is linked to the emission factors used, i.e. SAFAR uses values from Cooke et al. (1999) who suggested high emission factors for large industrial boilers but Bond et al. (2004) concluded that there is no evidence for such high values. ECLIPSE relies on smaller values as discussed in Bond et al. (2004) and Kupiainen and Klimont (2007). For the residential sector, ECLIPSE includes specific calculation of emissions from diesel generators and wick lamps; particularly the inclusion of the latter source resulted in additional BC emissions leading to higher estimates in version 5 of ECLIPSE.

3.2 Model description

We have used the Lagrangian particle dispersion model (LPDM) FLEXPART v9.0 (Stohl et al., 1998, 2005). The LPDM computes the trajectories of a large number of particles (infinitesimally small air parcels). Unlike ordinary air back-trajectory models, FLEXPART includes several processes important for aerosol dispersion and removal like diffusion by turbulence in the boundary layer and aloft, deep convective mixing, dry deposition and wet deposition. The representation of narrow plumes is not possible in Eulerian models whereas in LPDM one can track the particles correctly at subgrid scale. Furthermore, FLEXPART can be run in both forward- and backward-in-time modes. The output of the forward modelling from emission sources are simulated concentration fields, whereas a backward run of the model initialized from a receptor point (typically, a measurement location) provides source–receptor (S–R) relationships or potential emission sensitivity (PES) fields. A detailed description of the FLEXPART-based S–R relationship can be found in Seibert and Frank (2004). It is related to the residence time of particles in output grid cells. The S–R relationship describes the sensitivity of receptor $y$ to source $x$. In the present case, the receptor $y$ is a vector of 24 h average BC concentrations at Gadanki for different days, and source $x$ is a vector of area-averaged BC emissions in different grid-boxes at different time intervals. In case of FLEXPART based S–R relationship, the S–R relationship is a matrix $M$ whose elements $m_{ij}$ are defined by $m_{ij} = \frac{x_j}{y_i}$ (Seibert and Frank, 2004). Once the matrix $M$ is known for a given source vector (emission inventory), receptor vector (BC concentrations at measurement site) can be obtained by a simple matrix-vector multiplication. The backward (also known as retroplume) runs are particularly useful to understand the regional distribution of sources contributing to pollution at the observation site and the corresponding transport pathways and for evaluating emission inventories using point observations.

We have used the NCEP Global Forecast Systems Final – GFS-FNL; NCEP (2000): hereinafter referred to as FNL data – meteorological analysis data to drive FLEXPART. GFS-FNL data are available at 1° × 1° spatial resolution and at 6-hourly temporal resolution. Vertically, the data are available at 26 pressure levels extending from the surface to 10 hPa.

We have used backward runs of FLEXPART to simulate the BC concentrations at Gadanki to understand the relative merit of various inventories for the comparison of modelled values with observations. Various settings for the model runs are summarized in Table 1. In the backward runs, BC particles were traced backward in time from the receptor site (Gadanki) for 10 days. The simulations were carried out for every day of the years 2009 and 2011. Since the
Table 1. FLEXPART model set-up for retroplume runs from Gadanki.

| Input meteorological data | NCEP-GFS data at $1^\circ \times 1^\circ$ global  
| Tracer | Black carbon aerosol  
| Point of origin for retroplume | Gadanki (13.48° N, 79.18° E, 365 m a.s.l.), altitude: 0–100 m  
| Output grid | Horizontal: $1^\circ \times 1^\circ$ global; Vertical: 0–100, 100–3000, 3000–5000 m a.g.l.  
| Mode | Backward runs  
| Number of days backward for each release | 10 days  
| Dry deposition | Enabled for 2009 and 2011  
| Convection | Enabled for 2009 and 2011  
| Wet deposition | Enabled only for year 2011  
| Dry deposition parameters | Density (rho) = 1400 kg m$^{-3}$  
| Mean Diameter ($d_p$) = 0.25 µm  
| Sigma of log-normal distribution (dsig) = 1.25  
| Below-cloud scavenging parameters | Scavenging coefficient at rainfall rate  
| $1 \text{mm h}^{-1} (A) = 2 \times 10^{-7} \text{s}^{-1}$  
| Dependency factor ($B$) = 0.62  

FNL data do not include precipitation values for the year 2009, the model particles were subjected to only dry deposition in the year 2009 whereas the particles were subjected to both dry and wet deposition in the year 2011. To calculate dry deposition, the particle density, aerodynamic diameter and standard deviation of a log-normal distribution were assumed to be 1400 kg m$^{-3}$, 0.25 µm and 1.25, respectively following Stohl et al. (2013). Below-cloud scavenging is modelled using a wet scavenging coefficient defined as $\lambda = A I^B$, where $A$ is the wet scavenging coefficient at precipitation rate ($I$) equal to 1 mm h$^{-1}$, and $B$ is the factor dependency (McMahon and Denison, 1979). We have set values of $A$ equal to $2 \times 10^{-7} \text{s}^{-1}$ and $B$ equal to 0.62 following Stohl et al. (2013). The in-cloud scavenging is simulated using a scavenging coefficient defined as $\lambda = (1.25 I^{0.64}) H^{-1}$, where $H$ is cloud thickness in metres (Hertel et al., 1995). The PES values in the bottom-most layer (so-called footprint layer; 0–100 m a.g.l.) were multiplied by the emission fluxes to calculate the BC concentration at the receptor.

4 Results and discussion

4.1 Observations

Daily mean measured BC concentrations at Gadanki from April 2008 to October 2012 are shown in Fig. 4. BC concentrations at Gadanki vary strongly with season, with high values during late winter and spring and low values during monsoon months. The daily mean values varied from 6.8 ± 3.1 µg m$^{-3}$ (February) to 0.3 ± 0.2 µg m$^{-3}$ (November). Although the data period is not sufficient to do a thorough trend analysis, for the available data, no trend is observed. Also, there are no major differences in seasonal peak and low concentrations from 2008 to 2012. Hence, keeping computational time constraints in mind, the numerical simulations were carried out only for the relatively dry year 2009 and the normally wet year 2011.

4.2 Potential emission sensitivity

The model output is PES values on a three-dimensional grid. Since BC is mainly emitted near surface, we focus here only on the PES of the bottom-most layer from 0–100 m a.g.l. (the so-called footprint layer) and refer to this simply as PES. PES maps for 5 different days representing different meteor..
Selected examples of footprint potential emission sensitivity (PES) maps (also known as source–receptor relationships) using 10 days of backward runs (retroplumes) of FLEXPART from Gadanki. Panels (e) and (f) are PES maps of 14 October 2011 local time with and without wet deposition respectively. See Supplement for the PES maps for other days.

orological situations are shown in Fig. 5a–f. PES maps for all the days during 2009 are provided in the Supplement. PES values are represented in a logarithmic colour scale defined at the side of the figure panels. The median height of the retroplume in daily intervals is shown using grey-shaded dots. Depending on the season, Gadanki receives air coming from different regions. Generally, during winter air parcels are either from the Indo-Gangetic Basin (northern India) or Central Bay of Bengal (e.g. Fig. 5a and b). During summer or the southwest monsoon period the air comes from the northern Indian Ocean and Arabian Sea (e.g. Fig. 5c). During the transition period, the air travels over western and central India before reaching Gadanki (e.g. Fig. 5d). It is rare that significant PES values occur over Southeast Asian countries or China, though in a few instances trajectories came from Myanmar, Southeast Asian countries and South China (e.g. Fig. 5b). The advantage of a dispersion model vis-à-vis a simple air trajectory model can be seen in Fig. 5c. The median trajectory shown with grey dots is found to pass over the Arabian Peninsula, though surface PES values are not significant over the Arabian Peninsula, but are substantial over the northern Indian Ocean. In such circumstances, simple air back-trajectory analysis may ascribe observed concentration to emissions over Arabia whereas in reality it is not being influenced by surface emissions over that region. To demonstrate the effect of wet deposition on PES, PES maps for 14 October 2011 are shown with and without wet deposition in Fig. 5e and f respectively. The week preceding 14 October
2011 had large rainfall over southern India and the Bay of Bengal. Precipitation maps for 6 days from TRMM satellite are provided in the Supplement. Wet deposition is generally the most important removal process for aerosol and its effect on PES can be seen by the reduction of the high PES area especially over the ocean. However, the highest PES values over India close to the observation site remain almost unaffected by precipitation. Simulated BC concentrations for this case with and without precipitation are 1.0 and 1.4 µg m$^{-3}$, respectively.

### 4.3 Modeled BC concentrations

BC concentrations are determined by multiplying the footprint PES values with emission fluxes from the various inventories for every grid point and then integrating over the whole globe. The method implies that BC emissions are uniformly distributed in a grid cell of height 100 m (height of footprint PES layer). For a surface source, the footprint PES layer should be as small as possible. However, a very shallow footprint layer is not ideal from statistical point of view, as the PES is calculated based on the mass (and, thus, approximately the number) of particles in the footprint layer. With a very shallow height (say, 10 m), one would need to release 10 times more particles (number of trajectories) than with a 100 m height of the footprint layer to arrive at the same statistical error for the footprint PES, whereas increasing the height of the PES layer will not introduce significant error as long as the boundary layer height is higher than the footprint PES layer. BC concentrations are calculated with the three emission inventories ECLIPSE, RETRO and SAFAR-India. The SAFAR-India emission inventory is available only for the Indian region, hence inventory values outside India are set to zero. In the case of the ECLIPSE inventory, emissions outside India including shipping are found to contribute on average 6% of the total modelled BC concentrations over Gadanki. There were only 36 days in the year 2009 that had more than 15% of the BC originating from emissions outside India. Note that for the year 2009, wet deposition was not simulated. In the case of 2011, for which the wet deposition was simulated, emissions outside India contributed 5% on average and there were only 24 days when their contribution was more than 15%. In Fig. 6, seasonal and annual averages of source contribution maps are shown. During winter the emissions from IGB region (North India) dominate the BC concentrations at Gadanki, whereas during spring, emissions from southern India dominate. During summer, the source region is very small resulting in low concentration of BC as shown later. Autumn is a transition period from southwest monsoon to northeast monsoon and hence BC concentrations at Gadanki are due to both northern and southern India emissions. On average, India north of 18° N latitude contributes 43% of simulated BC mass and the part north of 22° N latitude contributes 33% at Gadanki. The contribution increases to 67 and 57% during winter from the two regions, respectively.

A comparison of observed and model-estimated BC concentration for the year 2009 is shown in Fig. 7a. There are no big differences between the three emission inventories. BC estimates based on RETRO are a little lower than for the other inventories, as expected, since total BC emissions of India (697 Gg yr$^{-1}$) in RETRO are significantly lower than in the other two inventories. Overall, the seasonal pattern is well reproduced in the model runs. Several submonthly-scale variations of observed BC concentrations are also well reproduced by the model, confirming its ability to simulate the influence of short-term changes in the meteorological conditions. During autumn and winter, the observed values are reproduced by the model within around 30% but the model underestimates the observed BC concentrations during spring and summer quite substantially. In Table 2, values of annual and seasonal averages, observation to model ratio, mean biases, root mean square differences (RMSD) and correlation coefficients ($R$) between observation and model for different inventories are shown. Overall, SAFAR has the smallest bias (0.8 µg m$^{-3}$ and least RMSD (1.4 µg m$^{-3}$ in 2009 and 1.1 µg m$^{-3}$ in 2011) with comparable values for ECLIPSE. The bias is small during autumn in general. In fact, with the SAFAR inventory, the model overestimates the observed concentrations during autumn of 2009 by a small amount (0.104 µg m$^{-3}$). The largest bias and RMSD are found during spring. Note that seasonal variations in model values are purely due to meteorology and transport as emission fluxes are constant within a month in the ECLIPSE and RETRO inventories and throughout the year for SAFAR inventory. Although the SAFAR inventory has seasonally fixed emission fluxes, the model’s performance using the SAFAR inventory is not very different compared to using the ECLIPSE inventory. This is because BC emissions in ECLIPSE inventory has very small seasonal variation. Monthly BC emissions of India in ECLIPSE (excluding GFED) inventory vary from 93.7 Gg in September to a maximum 104.2 Gg in July mainly due to seasonal variation of agricultural waste burning, which varies from 1.2 to 10.5 Gg. Together with GFED, there is less than 4.1% monthly variation of total BC emissions in a year in India.

As mentioned before, simulations for the year 2009 were carried out without including the wet deposition process. When including wet deposition for the year 2009, the underestimation may even be larger than that reported here. However, in Fig. 7b and c, we show a comparison for the year 2011 without and with wet deposition, respectively. It can be seen that the wet deposition has very little effect and hardly produces perceptible differences between Fig. 7b and c. Overall, wet deposition reduces modelled BC values by only 8% when using the ECLIPSE inventory. Seasonally, the wet deposition is found to be reducing modelled BC values by 5% in winter, 6% in spring, 14% in summer and 15% in autumn. Such seasonal influence is expected as
Figure 6. BC source contribution (per $1^{\circ} \times 1^{\circ}$ grid cell) maps based on FLEXPART retroplume calculations and the ECLIPSE inventory. Values are for seasonal averages, i.e. (a) winter, (b) spring, (c) summer, (d) autumn and (e) annual average for year 2009.

the maximum rain over Gadanki is received during summer and autumn (see Fig. 1). There were about 76 days in the year 2011 when wet deposition reduced the BC concentrations by more than 15%. In summary, wet deposition is not a major factor that causes underestimation of model BC values over Gadanki. This is a result of the relatively short rainy season over major parts of India and the short transport times during the rainy season for a major fraction of the BC between its emission and the arrival at Gadanki, rendering precipitation scavenging an ineffective process for this particular site. This result is site specific and does not imply that wet deposition is globally of minor importance. On the contrary, it is the main removal mechanism for BC in the model.

In Fig. 8, the average age spectra (measuring the time between BC emission and BC arrival at Gadanki) of modelled BC values estimated using the ECLIPSE inventory are shown for the full year as well as for the four seasons. One can see that on average about 30 to 40% of BC mass is of age 4 days or more. During winter this value increases to 65%. In other words, a large fraction of BC mass during winter is due to long-range transport of BC particles from northern India. If the dry deposition process is the reason for underestimation then one may expect larger model biases in winter. Instead,
Table 2. Average, ratio, bias, RMSD and correlation coefficient between modelled and observed BC concentrations when using different inventories for the years 2009 and 2011.

<table>
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<td>–</td>
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Winter: December to February; spring: March to May; summer: June to August; autumn: September to November. RMSD: root mean square deviation

* Note: calculations for 2011 are with wet deposition; calculations for 2009 are without wet deposition. ECLIPSE inventory includes ECLIPSE v5, GFED v3 and shipping emissions.

Hence, our analysis suggests that underestimation is due to underestimation of emissions over southern India; however, it is
difficult to pinpoint sectors that are being underestimated for BC emissions. Contextual information such as underestimation being correlated to FRP suggests that BC emissions from open biomass burning may be the main sector responsible for underestimation of BC concentration at Gadanki. Gustafsson et al. (2009) and Sheesley et al. (2012) apportioned carbonaceous aerosols using radiocarbon technique over two locations influenced by air masses from India and found biomass burning contributing to the extent of 70% of total mass of carbonaceous aerosols. Pavuluri et al. (2011) studied the correlation of BC with levoglucosan and non-sea-salt K\(^+\) at Chennai (a major city in southern India) and found that biomass burning is the major source of them during winter and summer. Lelieveld et al. (2001) estimated the contribution of biomass burning in CO in the range of 60 to 90% using correlation with CH\(_3\)CN and radiocarbon technique during Indian Ocean Experiment (INDOEX). However, underestimation of open biomass burning as a source cannot explain fully the underestimation of BC concentration by the model during summer when biomass burning activity is low. A possibility exists that the aethalometer may overestimate BC concentration during summer. The aethalometer relates absorption by particles on filter paper to BC mass. During summer when wind speeds and direction are conducive for dust aerosol, atmosphere may have a high level of dust amount. Dust is a weakly absorbing type of aerosol. The mass absorption cross-section of dust is 9 times lower than the BC mass absorption cross-section (Zhang et al., 2008). However, during summer, when BC concentrations are low,
absorption by dust particles can be a significant part of total absorption by the particles on the filter paper and may be wrongly attributed to BC mass. In the absence of chemical analysis, we rely on the spectral signature of the absorption coefficient for qualitative information on aerosol type. Zhang et al. (2008) have found an inverse wavelength ($\lambda^{-1}$) dependence of the absorption coefficient for BC particles and no significant wavelength dependence for dust particles observed in China. The exponent of wavelength (in power-law form of the relation between absorption coefficient and wavelength) is reported as between $-1.5$ and $-3$ for BC particles emitted in biomass burning (Bergstrom et al., 2004, 2007; Kirchstetter et al., 2004; Clarke et al., 2007). The spectral characteristics of dust particles vary from place to place and the wavelength exponent is reported as between $-2$ and $-3$ for some of the places in Asia and Africa (Bergstrom et al., 2004, 2007; Fialho et al., 2005). Monthly median values of wavelength exponent based on seven wavelengths of aethalometer at Gadanki are found to vary between $-0.98$ to $-1.18$ for year 2009 (Figure absorption_alpha_2009.png in the Supplement). However, the variability as indicated by interquartile range is found to have increased significantly during summer, indicative of an increase in heterogeneity of absorbing aerosol types during summer. Hence, dust aerosols may be a factor but it may not account for all the difference between model and observation during summer.

This suggests that not only biomass burning but also other anthropogenic emissions in South India are underestimated. BC emission ratios vary within a country due to different stages of economic development (power plant and auto-mobile technology, environmental regulations enforcement). Changes, particularly environmental regulations and their implementation, can be highly region/place specific. Moreover, the changes can be non-linear in time. southern states in India are comparatively more industrialized than northern states but have lower population growth. If emission ratios are generalized for whole country or linear growth is assumed based on population, it may introduce errors in emission inventory. In addition, small-scale anthropogenic biomass burning can be significant. While satellite-based fire detection is low during summer, radiocarbon-based and levoglucosan-based BC apportionment suggest significant contribution of BC from biomass burning during summer (Pavuluri et al., 2011; Sheesley et al., 2012). Indoor biomass burning and small-scale agricultural waste burning will go unnoticed in satellite data due to increased cloudiness during summer. When overall BC concentrations are low, underestimation of these sources may cause significant fractional error in the estimated BC concentrations.

Overall, although underestimation of anthropogenic emissions cannot be ruled out, underestimation of BC concentration at Gadanki is likely related to underestimation of BC emission fluxes from forest fires and/or agricultural waste burning over southern India, particularly during spring.

4.3.1 Case studies

Since emission inventories for the years 2009 and 2011 are kept the same, the difference in model values between these two years is purely due to meteorology. In the rest of the paper, we focus on the year 2009. As noted in the previous section, the model underestimates BC values during spring and summer. The underestimation of BC concentrations may be related to underestimation of biomass burning activity during the dry season and subregionally incorrect anthropogenic emission fluxes. Here we discuss a few cases from the year 2009 which provide insight into these aspects.

In Fig. 9, a comparison of modelled and observed BC concentration over Gadanki for the year 2009 is shown. It is similar to Fig. 7a, but zoomed-in for three different periods. Note the sudden decrease in BC concentration for both observation and model on 8 January. This is possible if emission changes can be non-linear in time. southern states in India are comparatively more industrialized than northern states but have lower population growth. If emission ratios are generalized for whole country or linear growth is assumed based on population, it may introduce errors in emission inventory. In addition, small-scale anthropogenic biomass burning can be significant. While satellite-based fire detection is low during summer, radiocarbon-based and levoglucosan-based BC apportionment suggest significant contribution of BC from biomass burning during summer (Pavuluri et al., 2011; Sheesley et al., 2012). Indoor biomass burning and small-scale agricultural waste burning will go unnoticed in satellite data due to increased cloudiness during summer. When overall BC concentrations are low, underestimation of these sources may cause significant fractional error in the estimated BC concentrations.

Overall, although underestimation of anthropogenic emissions cannot be ruled out, underestimation of BC concentration at Gadanki is likely related to underestimation of BC emission fluxes from forest fires and/or agricultural waste burning over southern India, particularly during spring.

During February and to the middle of March, observed BC concentrations are increasing whereas model values are systematically decreasing. During this period the high PES region has moved away from India towards the BoB, whereas the PES region in the immediate vicinity of Gadanki has moved southward over Tamil Nadu state of India. The large divergence between observation and model is an indication that the inventoried emission fluxes are significantly underestimated over southern India. This is also a period of high biomass burning activity in the region of the high PES (see Fig. 2). Hence, underestimation may be related specifically to underestimation of open biomass burning in southern India.

From 22 April, the high PES region moves to the west of the observation site, over Karnataka state of India and the Arabian Sea (see Fig. 5d), occasionally moving south of the observation site over entire Tamil Nadu. During later part of May, and June and July months, the high PES region is mostly over the Arabian Sea with a very small region over land due to strong winds (see Fig. 5c). Both model and observations have low values during this period. However, the model systematically underestimates the observations by a factor of 2 to 3 (see Fig. 9b).

From 5 September onward the PES region covers Andhra Pradesh, Madhya Pradesh and Gujarat states of India. On 22 September, the model significantly overestimates the observation and for some days it remains higher than observations (see Fig. 9c). The high PES region during this period lay over Andhra Pradesh and Tamil Nadu border and over southern Karnataka. From 16 October onwards the PES pattern moves entirely north of the observation site. At the beginning the pattern covers central and western India, but on 1 November the PES pattern is similar to that found during January–February and extends all the way up to the northwest border of India covering the entire Indo-Gangetic region (see Fig. 5a). On this day, the concentration is the highest in model with similar values in observations. During September–December, differences between observation and model estimates are small.

Summarizing the above description, observed and modelled BC values are high when winds are from northern and western India, with relatively small differences between model and observations, indicative of relatively small errors in the emission inventories over this region. When winds are from South or South Central India, the observed values are high but the model values are substantially lower. Coincidentally, this is also the period of high biomass burning activity over southern India. The differences between the model and observation thus suggest that open biomass burning emissions over southern India have been underestimated in all the three inventories.

5 Conclusions

Several field studies over India and in adjoining oceans have found high amount of absorbing aerosols. However, models are found to underpredict the observed high concentrations of absorbing aerosols. Using the Lagrangian particle dispersion model FLEXPART and three emission inventories, we compared the simulated BC concentrations with BC measurements at a rural site in southern India. As for the other models, FLEXPART underestimates the observed BC concentrations. We found that 93 to 95 % of the model BC concentration is the result of emissions from India. Northern India is a major source of anthropogenic BC particles, but southern India also has significant BC emissions. This study identifies a potentially significant underestimate of emissions in southern India, which is reflected in a large difference in the observed and modelled BC values in Gadanki during spring when the winds are predominantly from the south. We suggest that the key source for which the emission fluxes may be
underestimated is open biomass burning. This is not to rule out the possibility that anthropogenic emissions may also be underestimated.

In the three emission inventories that we evaluated, the ECLIPSE inventory has the highest emissions (1.2 Tg yr$^{-1}$), with similar emissions in the SAFAR-India inventory (1.1 Tg yr$^{-1}$). This is also reflected in the comparison between the modelled and observed BC concentration over Gadanki. Modelled BC values based on ECLIPSE and SAFAR-India are higher than the values based on the RETRO inventory. However, they are not high enough to resolve underestimation in most of the seasons. The overall ratio of observation to model is found to be 1.5 for the SAFAR inventory, 1.7 for ECLIPSE inventory and 2.4 for RETRO inventory. Although the ECLIPSE inventory has the highest emissions over India, it is the SAFAR-India inventory that has the lowest ratio because of differences in spatial distribution in emission fluxes. The SAFAR-India inventory has higher emission fluxes over southern India compared to ECLIPSE.

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Edited by: R. Krejci

References


Cooke, W. F., Lioussse, C., Cachier, H., and Feichter, J.: Construction of a 1°×1° fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in


Remarks from the typesetter

TS1  Please check title.
TS2  Please check if all vectors are bold italic and matrices bold.
TS3  Do you mean “Gisci. Remote Sens.”?
TS4  Please update if possible.
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