1. INTRODUCTION

The seasonally reversing monsoon circulation system due to strong thermal contrast between the land and the ocean over South Asia typically characterizes the episodes of wet summers (June to September) and dry winters (December to February) (Ramage 1971, Rao 1976, Webster 1987, Wang et al. 2009). The wet summers are associated with moisture-laden southwesterly winds from the tropical Indian Ocean to the Indian subcontinent, and the winters are associated with the dry northeasterly winds from Tibet to the south Indian Ocean. The precipitation during the wet summers or during the Indian Summer Monsoon (ISM), has a strong influence on the economy of the country.

Because of the seasonally reversing South Asian monsoon circulations, which flow from land to ocean and vice-versa, the moving air-masses provide different information about the natural and anthropogenic greenhouse gases (GHGs, e.g. CO$_2$) from various regions to receptor (station) locations over India. Despite the fact that any point measurements for GHGs may provide the temporal distribution of the relevant tracer elements, the sparse network of ground-based measurements still poses serious limitations to the spatial analysis of their sources and sinks.

The majority of the anthropogenic GHGs that are emitted to the atmosphere contribute to a positive radiative forcing (by trapping more heat) and lead to a secular trend in the global average temperature (IPCC 2001, 2007). For example, the global CO$_2$ concentrations in the atmosphere have increased by over ~30% compared to the pre-industrial era, and since then their contribution to the trapping of longwave...
radiation in the atmosphere has increased. India has the third highest \text{CO}_2 emissions, as calculated from the country’s total fossil fuel consumption. A significant (57\%) increase in the country’s \text{CO}_2 emissions since the early 1990s is associated with rapid economic growth (Boden et al. 2010). The localized increase in GHGs emissions in India, and their heterogeneous distribution in the atmosphere, highlights the need for more \text{CO}_2 monitoring stations at various locations in the country (Bhattacharya et al. 2009, Indira et al. 2011, Patra et al. 2011, Tiwari et al. 2011). Inversion methods using Lagrangian particle dispersion models are commonly employed techniques to assess the apportionment of regional sources and sinks of trace gases in space and time (Uliasz & Pielke 1990, Flesch et al. 1995, Enting 2002, Seibert & Frank 2004, Lowenthal et al. 2010, Koracin et al. 2011, Koyama et al. 2011). This method is analogous to adjoint modeling in the Eulerian perspective; however, the adjoint modeling approach is ill-conditioned (Enting 2002). Lagrangian diagnostics follow a linear source–receptor relationship, thus providing useful information about multiple source contributions that could reach the limited number of available receptors (e.g. Seibert & Frank 2004, Flesch et al. 2009, Koracin et al. 2011).

The seasonal cycle of \text{CO}_2 at Hanle (HLE; 32.77°N, 78.95°E) shows a linear growth at a rate of 1 ppm yr^{-1} with a maximum (minimum) emission in March (October) (Tiwari et al. 2011). A similar growth rate of \text{CO}_2 is seen at Hanle (HLE) (Indira et al. 2011). Recent studies have indicated that inversion estimates of \text{CO}_2 fluxes over South Asia are weakly constrained by surface \text{CO}_2 observations from southwestern India (Rayner et al. 2008, Bhattacharya et al. 2009, Patra et al. 2011).

As a first step, this study investigates the usefulness of the currently available \text{CO}_2 monitoring locations over India to the proximate and distant sources using a Lagrangian particle dispersion model in order to provide guidance for \text{CO}_2 monitoring over India.

2. METHODS

In order to investigate the influence of regional \text{CO}_2 fluxes, a Lagrangian particle dispersion model FLEXPART version 8.0 (Stohl et al. 1998, 2005, 2009) was used in this study. FLEXPART calculates the advection of multiple tracer particles backward in time using mean 3D winds and stochastic motions from the turbulence fluxes. That is, FLEXPART introduces discrete inert particles at each receptor location and moves them backward in time in the model domain using the 3-hourly meteorological fields (archived at 0.5° × 0.5° resolution in the horizontal direction and at 26 pressure levels in the vertical) from the National Center for Environmental Prediction (NCEP) Global Forecast System (GFS) model analyses interpolated to the location of the particle.

The 2 \text{CO}_2 monitoring stations considered in this study are (1) Hanle (HLE; 32.77°N, 78.95°E) located on the foothills of the Himalayas at 4517 m above sea level (a.s.l.) surrounded by a dry arid area with sparse population; and (2) the Cape Rama (CRI; 15.08°N, 73.83°E), a coastal site near Goa located at 50 m a.s.l. A topographical map and the location of CRI and HLE are shown in Fig. 1. The locations for these sites are chosen in such a way that they are fairly far from point sources so that they primarily monitor the background concentrations.

The seasonal cycle of \text{CO}_2 at CRI (see Fig. 3) compares well with the seasonal cycles of other similar sites from the GLOBALVIEW \text{CO}_2 data sets (Masarie & Tans 1995). In order to monitor \text{CO}_2 at a clean site (in the absence of point source emissions) CRI, ~80 km from the nearest city (Panaji Goa), was selected. CRI is on a flat rocky terrain, devoid of any vegetation over a scale of 50 m on all sides and a few 100 m away from any sparse habitation. Furthermore, database studies on \text{CO}_2 at CRI (e.g. Bhattacharya et al. 2009, Tiwari et al. 2011) discount the possibility of point source emissions at this site irrespective of the seasons. Being a high-elevation station, HLE is largely unaffected by local emissions (Indira et al. 2011), and the observations at HLE also show good agreement with global datasets.

FLEXPART simulates backward in time. Such simulations from the measurement sites HLE and CRI were made every 3 h. Every 3 h interval, 20,000 particles were released at the measurement point and followed backward in time for 7 d (168 h). The starting period of the back-trajectory analysis was fixed on the 15th of every month for the year 2009. The particles are released from a receptor location at 12:00 h UTC (measurement site) and FLEXPART calculates a 4D (space and time) response function (sensitivity) to emission input.

3. RESULTS

3.1. Atmospheric circulation patterns and \text{CO}_2 observations

Fig. 2 shows the atmospheric circulation patterns at 850, 500, and 200 hPa pressure levels during January.
and July of 2009 from the National Centers for Environmental Predictions/National Center for Atmospheric Research (NCEP/NCAR) 2.5° × 2.5° gridded reanalysis dataset (Kalnay et al. 1996). The salient features of the atmospheric circulation are (1) the pressure gradient between Mascarene high (a semi-permanent high located east of Madagascar in the South Indian Ocean) and monsoon trough (a region of strong low-level convergence over the central and northern parts of the Indian continent) drives a cross-equatorial circulation and southwesterlies over the Arabian Sea into the Indian landmass (Fig. 2b) during summer, and (2) the pressure gradient reverses from land to ocean resulting in northeasterlies during winter months (Fig. 2a). Both seasons have prevailing westerlies in the upper troposphere. As the wet monsoon gradually withdraws in September, the direction of the pressure contrast is reversed during winter months, i.e., the pressure gradient between Tibet (at ~35°N, 87°E; isolines of height >1540 m shown in Fig. 2a) and the South Indian Ocean facilitates northeasterly winds blowing from land towards ocean. It should be mentioned that the stations HLE and CRI are located north and south of the monsoon trough, respectively.

Fig. 3 shows the time series and de-trended mean seasonal cycle of atmospheric CO₂ (ppm) concentration measured at CRI during 1993 to 2002 (Tiwari et al. 2011). The time series in Fig. 3a clearly indicates the seasonality with a varied amplitude from year to year, and a monotonic increasing trend. Mean seasonal cycle (Fig. 3b) amplitude is the greatest (smallest) in March (November; see Tiwari et al. 2011) for more details. The CO₂ observing station at HLE has been operational since 2006 (Indira et al. 2011; the data is currently being processed and is not available for public use).

3.2. Back-trajectory analysis

Fig. 4 shows the particle back-trajectories reaching HLE at the surface level for January and July. One can clearly see that the air parcels predominantly originated from the Middle Eastern and northwest African regions in January, when the winter mon-
Fig. 2. Monthly mean winds (vectors) and geopotential height (km) at (a,b) 850, (c,d) 500, and (e,f) 200 hPa during (a,c,e) January and (b,d,f) July 2009 from the NCEP/NCAR reanalysis. Locations of Hanle (HLE) and Cape Rama (CRI) shown in (a).
Fig. 3. Measured surface CO₂ concentration at Stn Cape Rama (CRI) during February 1993 to October 2002 as (a) time-series and (b) de-trended mean seasonal cycle.

Fig. 4. Lagrangian back-trajectories of particles simulated by FLEXPART starting from Stn Hanle (HLE) during (a) January and (b) July. Colors: position of particles in the vertical (m). Daily position of particles indicated by numbers (d).
soon circulation is at its peak. Furthermore, the parcels descend from altitudes much higher than 8 km a.s.l. Particles remained within 4 to 6 km a.s.l. for most of the days on their path, and they apparently had longer residence time over the Indus Plains over northwestern India. The tracer transport from the source regions is apparently dominated by the strong subtropical westerlies in the mid-to-upper troposphere (Fig. 4b). A westerly upper level mean flow of 50 m s$^{-1}$ at $\sim$30°N primarily transports the tracers to HLE, and the wave structure of the trajectories can be attributed to the existence of cyclonic-anticyclonic wind anomalies between tropical and extra-tropical regions at these levels along the trajectory path. This feature continued until the pre-monsoon period in April in the presence of the anomalous anticyclonic circulation extending from the surface to 500 hPa level (figure not shown). The regional dynamics prevailing over northwestern India, the northern Arabian Sea and Arabia contribute to the longer residence of particles over these regions.

During the wet month of July, the back-trajectories indicate that a juxtaposition of particles originated from both the upper level westerlies and easterlies.

Fig. 5. Stn Cape Rama (CRI) back-trajectories. As in Fig. 4
The particles generally resided in the upper troposphere during their transport until their descent over Afghanistan, Pakistan, and over northwestern India. The particle directions are consistent with the northward migration of subtropical and mid-latitude westerlies in the mid-troposphere, as well as strong tropical upper-level easterlies above the monsoon trough. The anomalous cyclonic-anticyclonic circulations (figure not shown) at sub-tropical latitudes further facilitate a faster tracer transport at a time scale of 2 to 4 d. Following the monsoon, the upper level circulation in October (figure not shown) is purely aligned eastward over the latitude belt 20° to 60° N, with no easterlies in the tropical upper troposphere. The long-range transport of the particles during this time was primarily from northern Africa and southern Europe, and the transport from middle-Eastern countries to HLE is within 2 to 4 d.

Fig. 5 shows the particle back-trajectories reaching CRI at the surface level during January and July. Although the particles are transported from the western part of Asia in January, they are primarily transported by the northeasterly winds within the planetary boundary layer (PBL) over India as they reach CRI. During summer, the particles remained within 2 km a.s.l. throughout the entire course of back-trajectories. This suggests that the impact of marine layer CO2 fluxes is rather significant on the receptor at CRI. In contrast to the features seen at HLE, the particle transport to CRI is in good correspondence with the southwesterly monsoonal flow during summer. During the seasonal transitions in April and October, however, the particle trajectories remained primarily over India and northern Arabian Sea (figure not shown). This indicates that emissions both from the land and the ocean apparently reach CRI.

A sensitivity examination showed that the back-trajectories initiated from the end of each month showed similar signatures to the ones initiated from the middle of each month as used in this study (figure not shown). The trajectories to CRI indicated a rotation in a counter-clockwise manner during the post-monsoon and winter months, i.e. from October to April. That is, the tracers originate from the Arabian Sea reaching CRI during February and March, while they originate from the landmass of northern parts of India, Bangladesh, Myanmar, and Thailand between September and December. The notion of backward trajectory synthesis here is that tracer elements received at HLE or CRI pick up flux information along the entire pathway of the trajectory, and not just from its end points.

### 3.3. Surface sensitivity

Footprint emission sensitivity represents the sensitivity of mole fraction at the receptor site to upwind surface sources and sinks. For each sampling time at the receptor sites (HLE or CRI), we obtain the emission sensitivity in a surface footprint averaged over the surface layer from 0 to 100 m above the ground. Emission sensitivity (or surface sensitivity expressed in ppm µmol⁻¹ m⁻² s⁻¹) in a given grid cell is proportional to the particle residence time in that grid cell. Fig. 6 shows the surface sensitivity at CRI.
During January, higher surface sensitivity is seen over the central and east coast of India as well as over the foothills of the Himalayas (Fig. 6a). In contrast, the surface sensitivity magnitudes are significant over the central Arabian Sea in July (Fig. 6b). The concentrations can be more sensitive to local terrestrial (marine/oceanic) fluxes in January (July).

Recently Tiwari et al. (2011) investigated the sensitivity of surface fluxes on the CO₂ mixing ratios at CRI during 1993 to 2002 using the observational and forward model simulations (cf. Fig. 6 of Tiwari et al. 2011). They indicated that the concentrations at CRI were found to be more sensitive to the fluxes from the south Indian Ocean during wet months (such as June) while the sensitivity is significantly diluted due to local terrestrial influences during the dry winter months (January). Their findings are in reasonable agreement with the results obtained in this study from FLEXPART model surface sensitivity simulations. The surface sensitivity at HLE showed similar aforementioned signatures (figure not shown).

To summarize, the 2 receptors HLE and CRI on either side of the monsoon trough are able to capture the tropospheric tracer transport during summer months. HLE is strongly influenced by the air motion originating from the subtropical–mid-latitude belt irrespective of the seasonal wind reversals of the monsoon during most of the year. Although inert particles were used in examining the transport, one can say that CO₂ observations at HLE can be more sensitive to the background concentration from west Asia and from the upper troposphere for all the seasons, whereas the tracer concentration at CRI is influenced by the lower tropospheric transport coherent with the seasonal reversal of winds. This suggests that the monitoring stations CRI and HLE can substantially capture the regional local emissions and their short-(long-) range transport from the lower (upper) troposphere.

With reference to the northward movement of pressure systems (monsoon trough) over India, the 2 receptors located on either side of the monsoon trough are able to effectively capture the tropospheric tracer transport from different proximate and distant source regions. The winds from the lower troposphere associated with the monsoonal wind reversals appear to strongly influence the flux information at the CRI receptor. The tracer transport to CRI seems to be more representative of the boundary layer sources from continental India.

The tracer transport to HLE appears to be influenced by the source regions from west Asia and especially the emissions from the upper troposphere, irrespective of the seasons. Furthermore, they constitute both boundary layer sources as well as the background concentrations from the upper troposphere.

This preliminary investigation provides a formal guidance on the usefulness of the 2 receptor stations that can capture the tracer elements from the tropical continental/maritime source regions in addition to the background concentrations. It also showed indications of different transport time scales, as well as a variety of probable emission regions that can act as sources of CO₂ occurring over India in 1 seasonal cycle. The regional carbon cycle under a climate change scenario is likely to have serious implications in the regional climate assessment over India, owing to the prevalence of carbon sources and sinks from the neighbouring regions during different seasons, and the strengthening and weakening of monsoonal flows (Cherchi et al. 2010). Hence, a detailed further analysis of CO₂ observations, as well as the aid of full-suite chemistry transport models, are necessary for accurate regional identification of CO₂ and other tracer species to aid understanding of the regional carbon cycle over India.

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