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Continental export efficiencies and delineation of sources for trace gases and black carbon in North-East India: Seasonal variability

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HIGHLIGHTS

- Export efficiency of BC, NO_x and SO_2 from the IGP to NER are estimated.
- BC and NO_x export efficiency is highest in winter and lowest in monsoon.
- BC, CO and NO_x annual variability is affected by transport from IGP.
- Pollutants from North-East India outflows to southeast China and East-Asia.

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ABSTRACT

The Indo Gangetic Plain (IGP) has been identified from back-trajectory analyses, as one of the most potential region affecting the species transport to the Northeastern region of India (NER). The continental export efficiency ($\varepsilon\varepsilon$) of BC, NO_x and SO_2 within the boundary layer is estimated in order to examine how efficiently these chemical species are transported towards the NER. For this the measurements carried out at Dibrugarh, a wet tropical location in NER during 2012–2013 have been used as the references in the estimation of the species enhancements above their background. CO is used as a passive tracer of transport due to its longer lifetime in the atmosphere. The emission estimates of BC, NO_x, SO_2 and CO in the IGP region are adopted from the emission inventories REAS and INTEX-B. The estimated export efficiency is highest in winter (DJF) for BC and NO_x, whereas SO_2 shows maximum efficiency in monsoon (JJAS). BC due to efficient transportation/removal from the IGP region exhibits highest $\varepsilon\varepsilon$ values compared to the other species. NO_x and SO_2 on the other hand get transformed to other chemical species shortly after emission into the atmosphere and hence are less efficiently transported towards the study region. The export of BC, CO, NO_x and SO_2 are expected to supplement the chemical atmosphere in NER, which is further studied through the annual variability in their distribution in Dibrugarh. Pearson correlation analyses of BC, NO_x and SO_2 with CO is carried out to examine the similarity or dissimilarity among the sources.

1. Introduction

Emissions of trace gases and aerosols are increasing due to the fast industrialization and rapid growth in urbanization and economy in Asia, particularly in the developing countries like China and India. This region, with 60% of the world's population turns out to be the most vulnerable region of the globe regarding emission of pollutants that have significant susceptibility to global climate change, regional air quality and human health. Due to the large variability of atmospheric processes as well as emissions and properties of the chemical species, the interactions among the species extend over a wide range of spatial and temporal scales that adversely affect the regional and intercontinental atmospheric environment. Examples of such chemical species include Black carbon (BC), carbon monoxide (CO), sulfur dioxide (SO_2) and nitrogen oxides (NO_x) etc. The emission of these species degrades the air quality and influence the oxidation potential of the atmosphere in ways that can alter the energy balance of the climate system (Houghton et al., 1992). Zhang et al. (2009) estimated that during 2001–2006, Asian anthropogenic emissions have increased by 33% for SO_2, 44% for NO_x, 18% for CO and 11% for BC. According to National Ambient Air Quality Monitoring Program (NAAQMP), nitrogen dioxide

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(NO_2) and SO_2 respectively show the second and third highest exceedance rate in India, after PM_{10} (particulate matter <10 μm) (CPCB, 2012). These species originating mainly from combustion are subsequently removed from the atmosphere by gas to particle conversion processes, dry deposition, wet scavenging and transport. Chemical conversion and loss processes of BC, CO, SO_2 and NO_x take place mostly within the boundary layer. The concentration of these species at any location depends on how efficiently these are dispersed from the atmosphere of the source region. Anthropogenic pollutants from South Asia leave the source region and affect distant areas via horizontal transport within the boundary layer or are uplifted to the free troposphere by convection.

The presence of carbonaceous aerosols like BC, a byproduct of combustion significantly perturbs the radiation budget of the atmosphere through the absorption of solar radiation in urban and biomass burning environments. BC contributes to radiative forcing by heating the lower atmosphere (Ramanathan and Carmichael, 2008) as well as to indirect forcing by acting as cloud condensation nuclei (Lohmann et al., 2000; Ackerman et al., 2000). According to IPCC (2014) aerosols including BC present the largest uncertainty to total radiative forcing estimates with a net negative radiative forcing of −0.9 [−1.9 to −0.1] W/m^2. In addition to this global effect, BC influences visibility, air quality and human health at regional as well as local scale. Tropospheric BC has a lifetime of approximately one week (Koch and Hansen, 2005) against dry and wet scavenging. As the transcontinental transport typically occurs on timescale of days to weeks, BC may be used as a tracer to investigate pollution sources. CO, another byproduct of incomplete combustion/oxidation emitted from both anthropogenic and biomass burning is also a good tracer for investigating long range atmospheric transport because of its long lifetime (30–90 days) in the boundary layer/troposphere (Dickerson et al., 2002). The largest contributor to CO emissions in South Asia is the residential combustion of bio-fuels (Dickerson et al., 2002 and references therein). BC and CO are found to be significantly correlated in a number of studies (Pan et al., 2011 and references therein). Therefore, $\Delta BC/\Delta CO$ is used as a good indicator for distinguishing different emission sources in case studies (Kondo et al., 2006; Spackman et al., 2008; Han et al., 2009; Subramanian et al., 2010) as well as for validating BC emission inventories for models (Derwent et al., 2001; Dickerson et al., 2002). NO_x (= NO + NO_2) are important atmospheric species that affect atmospheric chemistry, air quality and climate (IPCC, 2007), which dominantly control the tropospheric O_3 budget, the abundance of the hydroxyl radical (OH) and the formation of nitrate aerosols. The lifetime of NO_x varies from few hours in the boundary layer to about a week in the upper troposphere. NO_x and SO_2 can also influence the atmospheric environment by acid rain and corrosion (Finlayson-Pitts and Pitts, 1986). SO_2, the predominant anthropogenic sulfur containing air pollutant has an adverse impact on the environment. It is one of the major secondary pollutants that contribute to the formation of particulate matter in the atmosphere. Sulfate aerosols reflect sunlight into space and act as cloud condensation nuclei that tend to make clouds more reflective and change their lifetimes, causing a net cooling at the Earth's surface. The lifetime of SO_2 against its reaction with OH radical at typical atmospheric levels of OH is about a week and is efficiently removed by deposition at the surface and by cloud processes (Seinfeld and Pandis, 2006). SO_2 can be transported over long distances (beyond 500–1000 km) and associated with cross-country monsoon phenomena.

In India, there were no systematic and simultaneous measurements of surface O_3 and precursor gases until the Indian Space Research Organization's Geosphere Biosphere Program (ISRO-GBP) started the measurements of O_3 and other pollutant gases. Under ISRO- GBP land campaign I, Reddy et al. (2007) simultaneously measured the concentrations of CO and SO_2 over Southern India and studied their diurnal variations. Several reports on surface O_3 and its precursors are now available over the Indian region (e.g. Lal et al., 1998; Lal et al., 2000; Nair et al., 2002; Naja and Lal, 2002; Kumar et al., 2010; David and Nair, 2011; Bhuyan et al., 2014; Gaur et al., 2014). However, the focus of all these studies were confined to the analysis of spatio-temporal distribution of the species under the influence of meteorology, transportation etc. The spatio-temporal distributions of BC for various aerosol environments are also available in Indian subcontinent (Pathak and Bhuyan, 2014 and references therein). In view of the regional effects of BC and pollutant gases, it is worthwhile to examine the transportation or removal efficiency of these species at a particular location.

In the present study, we have estimated the continental transport or export efficiencies of transported BC, NO_x and SO_2 from the Indo Gangetic Plains (IGP) towards the Northeastern region of India (NER) within the boundary layer. The measurements of the above discussed species carried out at Dibrugarh (27.3°N, 94.6°E, 111 m amsl) during January 2012–December 2013 are used as reference data. The correlation of BC, NO_x and SO_2 with CO is also tested in order to examine the seasonal variability in their sources. Further, the probable outflow pathways of the trajectories carrying CO and other gases originating in NER are identified using forward trajectory analysis.

2. Approach

2.1. Methodology: export efficiency

In order to estimate the export or transport efficiency of BC, NO_x and SO_2 we have adopted the method followed by Koike et al. (2003) and Park et al. (2005). They have estimated (equation (1)) the altitude dependent export efficiencies of combustion generated NO_y, SO_x and BC over East Asia with reference to CO as an inert combustion tracer.

$$\varepsilon\varepsilon = \frac{1}{R_X}\left(\frac{\Delta[X]}{\Delta[CO]}\right) \quad (1)$$

where $\varepsilon\varepsilon$ is the export efficiency, X is the combustion derived species (here BC, SO_2 and NO_x), R_x is the emission ratio (X/CO), CO is being used as a passive tracer of transport due to its longer lifetime within the boundary layer and Δ is the concentration enhancement of X relative to background. Estimates of background concentrations in air upwind from the source region, especially for CO, which has a substantial background owing to its long lifetime is essential for the application in Equation (1) (Park et al., 2005 and references therein). Here we have assumed the minimum detection level of gases (NO_x = 0.4 ppb, CO = 0.04 ppm, SO_2 = 0.4 ppb) and BC (50 ngm^{-3}) as the background values. According to Park et al. (2005), the specification of background values is not critical especially for the species with short lifetimes.

2.2. Wind pathways: an indicator of species transport

The seasonal variability of aerosol and gases measured over a location cannot be explained solely based on local meteorology and boundary layer dynamics. For example, in Dibrugarh the boundary layer is shallow and precipitation is almost absent in winter. Nevertheless, aerosol loading is higher in the pre-monsoon season when about 40% of total rainfall occurs and the boundary layer is deeper. It is now well established that transport of aerosols from one region to another within a continent and across