SEASONAL AND SPATIAL VARIABILITY OF OZONE INFERRED FROM GLOBAL CHEMISTRY TRANSPORT MODEL SIMULATIONS OVER INDIA

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ABSTRACT:

In the present study, 3-D chemical transport model, MOZART-4 (Model for Ozone and Related chemical Tracers-Version-4) has been used to study the seasonal and spatial distribution of surface ozone (O₃) over India. To illustrate the capabilities of model, the simulations are compared with the ground-based observations. The model reproduces the principal features present in ground-based observations. However, model is unable to simulate very low concentration of O₃ during monsoon months. The model simulations are used to quantify the contribution of background ozone and Indian anthropogenic emissions to the variability of surface O₃. The spatial distribution of total O₃ (TO) shows maximum over Indo-Gangetic plains (IGP) and Eastern India while minimum over southern Indian region. The seasonal cycle of TO vary from 34.2±7.6 – 51.9±4.9 ppbv over the Indian landmass region due to changes in its topography and ozone lifetime. Annual mean total background O₃ (TBO) over India shows highest during spring (29.3±5.0 ppbv) and lowest during monsoon (19.6±3.4 ppbv). Both Natural background O₃ (NBO) and pollution background O₃ (PBO) shows a minimum in summer which are essentially following the seasonal changes of total ozone. Significant variation of India pollution O₃ (IPO) is found over India and the spatial variation of IPO follows the spatial variation of TO as well as spatial variation of emission of ozone precursors.

1. INTRODUCTION

The third most important anthropogenic greenhouse gas is the tropospheric O₃ after carbon dioxide (CO₂) and methane (CH₄) (Ehhalt and Prather, 2001) which plays a crucial role in the earth’s radiation budget (Gauss, 2003). Higher concentration of O₃ in the atmospheric boundary layer may adversely affecting vegetation, animals as well as humans (Adams et.al., 1989). Ozone is produced by rapid photochemical reactions of CO (carbon monoxide) and VOCs (volatile organic compounds) with OH (hydroxyl) radical in the presence of NOx (nitrogen oxides) and sunlight. Apart from this, downward transport of stratospheric O₃ into the troposphere is another source of the tropospheric O₃. The contribution of later is much smaller on an annual and global scale, but it can be accountable in the regions of subsidence (Cooper et al., 2002). Anthropogenic activities (such as combustion of biofuel and fossil fuel), biomass burning, biogenic sources, and lightnings are the major sources of O₃ precursor gases like CO, NOx, and VOCs, etc. in the atmosphere. During later half of the 20th century, the fast economic growth and rapid industrialization of the developing countries has led to a dramatic increase in emission of these gases in the atmosphere. This subsequently lead to increase in ozone concentration in the lower troposphere (Wang et al., 2006; Roy et. al., 2008; Kim et al., 2013).

Thus, tropospheric O₃ and its precursor gases play a crucial role in the degradation of air quality, atmospheric chemistry, and climate change (e.g. Stevenson et al., 2013; WHO, 2003; Monks et al., 2015). In recent years, numerous in-situ ground-based measurements (Lal et al., 2000; Kumar et al., 2010) and very few ship-based campaigns (Srivastava et al., 2012 references therein) have been initiated to account the spatio-temporal variation of O₃ and precursors over Indian region. However, most of these measurements are confined to the specific region and for short durations. These in-situ observations are unable to explain the different atmospheric processes accountable for high pollution events. Therefore, chemistry transport models are valuable for providing a large-scale view of the regional impact of these gases and are useful for interpretation of observations on regional to global scale (Yarragunta et al., 2017). In the present work, in-situ observations have been used to understand the variability of O₃ over the Indian subcontinent and to evaluate the chemistry transport model, MOZART-4. The MOZART-4 model simulations are used to investigate seasonal and spatial variation of surface ozone as well as to identify the contribution of various sources (natural and anthropogenic) and its horizontal distribution over the Indian sub-continent. Along with seasonal analysis of ozone is also presented over different geographical regions of India.

2. METHODOLOGY

2.1 The Model for Ozone and Related chemical Tracers-Version-4

The global chemistry transport model, MOZART-4 (Model for Ozone and Related chemical Tracers-Version-4) which is jointly developed by the NCAR (National Center for Atmospheric Research), GFDL (NOAA Geophysical Fluid Dynamics Laboratory) and MPI-Met (Max Planck Institute for Meteorology). MOZART-4 contained comprehensive tropospheric chemistry which includes more than 130 chemical and aerosol species and 157 chemical reactions (Emmons et al., 2010). MOZART-4 is driven by assimilated meteorological data from the GEOS-5 (Goddard Earth Observing System version 5) of the NASA Global Modelling and Assimilation Office (https://www.earthsystemgrid.org/dataset/ucar.cgd.cssm4.g

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3. RESULTS AND DISCUSSION

3.1 Comparison with ground-based observations

In order to find the performance of model in case of seasonal cycle, the comparison of monthly observed and simulated mixing ratios of $\text{O}_3$ have been compared in the follow section. The monthly distribution of $\text{O}_3$ have been reported in different observational sites over Indian region. The details of each site viz. latitude, longitude, elevation, period of study and reference are given in Table 2 and the geographical location of each site is shown in Fig. 1(a). The monthly distribution of $\text{O}_3$ mixing ratio has been taken from the following observational sites viz. Anantapur (14.7N, 77.6E) (Ahamed et al., 2000), Ahmedabad (23.0N, 72.6E) (Sahu and Lal, 2006), Delhi (28.6N, 77.2E) (Jain et al., 2005), Hyderabad (17.4N, 78.4E) (Surendran et al., 2015), Nainital (29.2N, 79.3 E) (Kumar et al., 2010), Pune (18.5N, 73.9E) (Beig et al., 2007), Kanpur (26.5N, 80.3E) (Gaur et al., 2014), Mount Abu (24.6N, 72.7E) (Naja et al., 2003), Chennai (13.1N, 80.4E) (Surendran et al., 2015), and Udaipur (24.6N, 73.7E) (Yadav et al., 2014). Fig. 2 represent the comparison of monthly mean observed surface $\text{O}_3$ mixing ratios with monthly simulated results at above said locations. Generally, model reproduced the observed seasonal cycle reasonably well over all measurement sites except Delhi, Kanpur, and Nainital where model fails to capture the minimum concentration of $\text{O}_3$ during monsoon months. Over Chennai, the systematic overestimation of $\text{O}_3$ mixing ratio by the model has been found throughout the year. The simulated $\text{O}_3$ mixing ratio showed maxima during spring over all the measurement sites except Delhi and Chennai where this feature is not clearly evident. The observed surface concentration of $\text{O}_3$ is between 7 ppbv and 60 ppbv whereas the simulated $\text{O}_3$ is between 27 ppbv and 53 ppbv among these observational sites. The observed $\text{O}_3$ is maximum during spring and minimum during summer over urban sites (Delhi, Kanpur, Ahmedabad, Udaipur, Chennai, and Hyderabad). The model is unable to reproduce this minimum concentration during summer. The observed $\text{O}_3$ mixing ratio over Nainital and Mount Abu is maximum during late spring and late autumn respectively. The simulated $\text{O}_3$ mixing ratio is maximum during late spring and spring/autumn respectively. Their range of absolute difference is -9.6 - 20.5 ppbv and -13.0- 10.8 ppbv respectively. Anantapur and Pune showed maximum observed $\text{O}_3$ mixing ratio during spring and minimum during monsoon months while simulated mixing ratio peaks during autumn and dips during the monsoon period. The quantitative difference is maximum during autumn and minimum during monsoon season over both the sites.

These observed seasonal changes are attributed to the changing wind pattern. Westerly winds during pre-monsoon and south-westerly or south-easterly winds during summer monsoon season prevail over Indian region. The strong westerly winds during pre-monsoon season bring pollution rich air masses to the observational sites resulting in higher levels of $\text{O}_3$ and its precursor gases. The higher observed $\text{O}_3$ mixing ratios during pre-monsoon season are also due to photochemical transformation, long-range transport of air pollutants (Lal et al., 2013; Kumar et al., 2010a) and atmospheric boundary layer dynamics (Lal et al., 2000). South-westerly or south-easterly winds during summer-monsoon bring marine air from the oceanic regions (Guttikunda, 2010). The cleaner air from marine regions reduce the pollutant concentration in the continental air. In addition, insufficient sunshine for photochemical ozone...
production, reduced impact of local emissions due to pronounced washout effect and strong convective activities are responsible for the minimum concentration of ozone in summer monsoon. The overall seasonality of ozone has been reproduced by model fairly well over all the sites. Improper representation of meteorological phenomenon, photochemical production, underestimation of dry deposition and wet scavenging of ozone precursors are some possible reasons of the overestimation of the observed $O_3$ concentration by the model. In addition, the local emissions are diluted in large grid boxes in the model due to its coarser resolution. These results are consistent with results presented by previous studies (Kumar et al., 2012; Surendran et al., 2015).

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Name</th>
<th>Lat.</th>
<th>Lon.</th>
<th>Elevation (m)</th>
<th>Site Type</th>
<th>Period of Study</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Anantapur (ANA)</td>
<td>14.7</td>
<td>77.6</td>
<td>331</td>
<td>Rural</td>
<td>2001-03</td>
<td>Ahammed et al., 2006</td>
</tr>
<tr>
<td>2</td>
<td>Ahmedabad (AHM)</td>
<td>23.0</td>
<td>72.6</td>
<td>49</td>
<td>Urban</td>
<td>2002</td>
<td>Saha &amp; Lal, 2006</td>
</tr>
<tr>
<td>3</td>
<td>Delhi (DEL)</td>
<td>28.6</td>
<td>77.2</td>
<td>220</td>
<td>Urban</td>
<td>1997-03</td>
<td>Jain et al., 2005</td>
</tr>
<tr>
<td>4</td>
<td>Hyderabad (HYD)</td>
<td>17.4</td>
<td>78.5</td>
<td>542</td>
<td>Urban</td>
<td>2010-13</td>
<td>Surendran et al., 2015</td>
</tr>
<tr>
<td>5</td>
<td>Nainital (NAI)</td>
<td>29.2</td>
<td>79.3</td>
<td>1958</td>
<td>High Altitude</td>
<td>2006-08</td>
<td>Kumar et al., 2010</td>
</tr>
<tr>
<td>6</td>
<td>Pune (PUNE)</td>
<td>18.5</td>
<td>73.9</td>
<td>559</td>
<td>Semi-Urban</td>
<td>2003-04</td>
<td>Beig et al., 2007</td>
</tr>
<tr>
<td>7</td>
<td>Kanpur (KAN)</td>
<td>26.5</td>
<td>80.3</td>
<td>125</td>
<td>Urban</td>
<td>2009-13</td>
<td>Gaur et al., 2014</td>
</tr>
<tr>
<td>8</td>
<td>Mount Abu (MT. ABU)</td>
<td>24.6</td>
<td>72.7</td>
<td>1680</td>
<td>High Altitude</td>
<td>1993-00</td>
<td>Noja et al., 2003</td>
</tr>
<tr>
<td>9</td>
<td>Chennai (CHE)</td>
<td>13.1</td>
<td>80.4</td>
<td>7</td>
<td>Urban</td>
<td>2010-13</td>
<td>Surendran et al., 2015</td>
</tr>
<tr>
<td>10</td>
<td>Udaipur (UDA)</td>
<td>24.6</td>
<td>73.7</td>
<td>598</td>
<td>Urban</td>
<td>2010-11</td>
<td>Yadav et al., 2014</td>
</tr>
</tbody>
</table>

Table 2. Monthly mean mixing ratios of ozone (ppbv) surface based observational sites are used for comparison in the present study.

3.2 Contributions of surface ozone from MOZART

The previous analysis established the confidence over model simulations. The model is able to simulating the key features of surface ozone over India. The model sensitivity simulations introduced in the section 2.2 can be used to quantify the contributions of background and anthropogenic emissions to the total ozone on the seasonal scale over Indian landmass region. Table 3 summarizes the mean and spatial variability (defined as the standard deviation of all model grids with in the region) of surface ozone over India as a whole by season. The annual mean of TO over India is 43.3±6.7 ppbv, with minimum in summer (35.3±7.8 ppbv) and maximum in spring (50.1±6.7 ppbv). The annual mean TBO over India is 24.6±4.3 ppbv which accounts 57% of total surface ozone. The TBO is maximum (29.3±5.0 ppbv) in spring and minimum (19.6±3.4 ppbv) in summer, accounting for 57% and 56% of surface ozone in these seasons respectively. In all the seasons, more than 50% of total surface ozone is from TBO while at least 70 % of total background ozone is of natural origin except spring where it is only 53 %. The annual mean NBO is 17.5±3.6 ppbv which ranges from 14.9±2.8 ppbv in summer to 20.1±3.5 ppbv in winter. The IPO ranges from 15.7±5.8 to 20.8±5.2 ppbv which follows the seasonal pattern of TO.
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summer season (15.7 ± 5.8 ppbv). The TBO and PBO also shows high during spring and low during summer while NBO shows high during winter season (20.1 ± 3.5 ppbv) while low in the summer season (14.9 ± 2.8 ppbv).

Fig. 5 Monthly variations of mean surface ozone (TO), total background ozone (TBO), natural background ozone (NBO), pollution background ozone (PBO), and India pollution ozone (IPO) for different Indian regions defined in Fig. 1(b). Results are presented as mean ± spatial variability (unit: ppbv).

Except NBO, all the components of ozone and TO are high during spring and low during summer. Table 5 shows the average surface concentration of ozone and its decomposed components during spring, 2008. TO, IPO and PBO shows maxima during spring over IGP while TBO and NBO shows maxima over NEI. The NBO contributes 63% to the TBO over NEI region where the forests are mainly sub-tropical evergreen with secondary successional fallow forests dominated by Bamboos. The NEI is the easternmost region of India and consists the seven hilly states of Meghalaya, Manipur, Arunachal Pradesh, Nagaland, Tripura, Mizoram and Assam. More than 4, 00,000 families practice slash and burn agriculture annually for cultivation during March-May which is just before the monsoon (Vadrevu et. al., 2013).

<table>
<thead>
<tr>
<th>Region</th>
<th>TO</th>
<th>TBO</th>
<th>NBO</th>
<th>IPO</th>
<th>PBO</th>
</tr>
</thead>
<tbody>
<tr>
<td>India</td>
<td>50.1±6.7</td>
<td>29.3±5.0</td>
<td>15.5±4.3</td>
<td>20.8±5.2</td>
<td>13.8±2.5</td>
</tr>
<tr>
<td>IGP</td>
<td>56.1±6.1</td>
<td>32.2±3.6</td>
<td>16.9±2.2</td>
<td>23.9±5.5</td>
<td>15.3±2.6</td>
</tr>
<tr>
<td>WI</td>
<td>46.2±2.6</td>
<td>27.4±1.1</td>
<td>16.6±1.7</td>
<td>19.1±3.2</td>
<td>10.4±2.4</td>
</tr>
<tr>
<td>EI</td>
<td>50.3±3.5</td>
<td>28.9±1.8</td>
<td>14.3±1.5</td>
<td>21.4±2.1</td>
<td>14.7±0.5</td>
</tr>
<tr>
<td>NEI</td>
<td>54.6±2.7</td>
<td>35.6±3.3</td>
<td>22.4±4.4</td>
<td>19.0±5.1</td>
<td>13.2±2.3</td>
</tr>
<tr>
<td>CI</td>
<td>49.1±2.4</td>
<td>29.4±1.6</td>
<td>14.9±1.3</td>
<td>19.7±1.8</td>
<td>14.5±1.2</td>
</tr>
<tr>
<td>SI</td>
<td>43.6±4.0</td>
<td>23.7±2.6</td>
<td>10.4±2.2</td>
<td>19.9±2.8</td>
<td>13.3±0.9</td>
</tr>
</tbody>
</table>

Table 5. Average surface ozone and its decomposition (TO): total surface ozone, TBO: total background ozone, NBO: natural background ozone, IPO: Indian Pollution ozone (IPO) and PBO: pollution background ozone) averaged over different regions of India during spring (Mar-Apr-May). Results are presented as mean ± spatial variability (unit: ppbv).

4. CONCLUSIONS

Daily simulations of tropospheric $O_3$ have been made using MOZART-4 during 2008. The model simulated $O_3$ concentrations are evaluated against ground-based observations. The comparison of model results with in-situ observations at ten surface sites revealed that the model successfully reproduced the observed seasonal cycle at these observational sites. The magnitude of observed $O_3$ is in the range of 7–60 ppbv whereas the magnitude of simulated $O_3$ is in the range of 27–53 ppbv. The high quantitative difference of $O_3$ is estimated over Delhi, Hyderabad, Kanpur and Nainital during monsoon season but it is estimated differently over remaining observational sites. Model systematically overestimates observed $O_3$ over different observational sites during various seasons.

The contributions of various source types (natural and anthropogenic) to the seasonal distribution of surface ozone have been studied using global chemistry transport model, MOZART-4. The annual mean of TO over India is 43.3±6.7 ppbv, with minimum in summer and maximum in spring. Annually 57% of total ozone is from TBO over India. The TBO is maximum during spring and minimum during summer, accounting 57% and 56% of surface ozone in these seasons respectively. In all the seasons, more than 50% of total surface ozone is from TBO while at least 70% of total background ozone is of natural origin except spring where it is only 53%. The IPO ranges from 15.7±5.8 to 20.8±5.2 ppbv which follows the seasonal pattern of TO. Spatial distribution of surface ozone shows approximately 60 to 70 ppbv over IGP and Eastern part of India whereas Central India shows 50 to 60 ppbv. The western and southern India shows minimum total surface ozone of 40 to 50 ppbv. Ozone seasonality shows that, except NBO, all the components of ozone and total ozone to be high during spring and low during summer. TO, IPO and PBO shows maxima during spring over IGP while TBO and NBO shows maxima over NEI during spring.

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