



HAL
open science

Transport effects on the vertical distribution of tropospheric ozone over western India

Shyam Lal, S. Venkataramani, Naveen Chandra, Owen R. Cooper, Jérôme Brioude, Manish Naja

► **To cite this version:**

Shyam Lal, S. Venkataramani, Naveen Chandra, Owen R. Cooper, Jérôme Brioude, et al.. Transport effects on the vertical distribution of tropospheric ozone over western India. *Journal of Geophysical Research: Atmospheres*, 2014, 119 (16), pp.10012-10026. 10.1002/2014JD021854 . hal-01069991

HAL Id: hal-01069991

<https://hal.science/hal-01069991v1>

Submitted on 30 Sep 2014

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1 **Transport effects on the vertical distribution of tropospheric ozone over western India.**

2 S. Lal^{1*}, S. Venkataramani¹, N. Chandra¹

3 O. R. Cooper^{2,3} and J. Brioude^{2,3,4}

4 M. Naja⁵

5 ¹Physical Research Laboratory, Navrangpura Ahmedabad, India

6 ²Cooperative Institute for Research in Environmental Sciences, University of Colorado,

7 Boulder, USA

8 ³NOAA Earth System Research Laboratory, Boulder, USA

9 ⁴Laboratoire de l'Atmosphere et des Cyclones, UMR8105, CNRS-Meteo France-Universite,

10 La Reunion, La Reunion, France

11 ⁵Aryabhata Research Institute of Observational Sciences, Nainital, India

12 * Corresponding author

13

14

15

16

17

18

19

20

21

22

23

24

25

26 **Abstract**

27 In situ tropospheric ozone measurements by balloon borne electrochemical concentration cell
28 (ECC) sensors above Ahmedabad in western India from May 2003 to July 2007 are presented,
29 along with an analysis of the transport processes responsible for the observed vertical ozone
30 distribution. This analysis is supported by 12-day back trajectory calculations using the
31 FLEXPART Lagrangian particle dispersion model. Lowest ozone (~20 ppbv) is observed near
32 the surface during September at the end of the Asian summer monsoon season. Average mid-
33 tropospheric (5-10 km above sea level) ozone is greatest (70-75 ppbv) during April-June and
34 lowest (40-50 ppbv) during winter. Ozone variability is greatest in the upper troposphere with
35 higher ozone during March-May. The FLEXPART retrorplume results show that the free
36 tropospheric vertical ozone distribution above this location is affected by long-range transport
37 from the direction of North Africa and North America. Ozone levels are also affected by
38 transport from the stratosphere particularly during March-April. The lower tropospheric (<3
39 km) ozone distribution during the Asian summer monsoon is affected by transport from the
40 Indian Ocean via the east coast of Africa and the Arabian Sea. Influence from deep convection
41 in the upper troposphere confined over central Asia has been simulated by FLEXPART. Lower
42 ozone levels are observed during August-November than in any other season at 10-14 km
43 above sea level. These in situ observations are in contrast to other studies based on satellite
44 data which show that the lowest ozone values at these altitudes occur during the Asian summer
45 monsoon.

46

47 **1. Introduction**

48 Ozone in the troposphere plays crucial roles as an oxidant and as a greenhouse gas. It is the
49 major source of the highly reactive OH radicals in the troposphere, which control oxidation
50 processes as well as act as a cleaning agent by controlling the abundance of many trace gases,

51 particularly hydrocarbons [*Monks et al.*, 2009]. Ozone traps the outgoing long-wave radiation
52 at 9.6 μm , and its contribution to radiative forcing is significant next to CO_2 and CH_4 [*Gauss*
53 *et al.*, 2003; *Stevenson et al.*, 2012; *IPCC*, 2013]. The efficiency of this radiative forcing is
54 greater at higher altitudes in the troposphere [*Lacis et al.*, 1990; *Gauss et al.*, 2003]. Ozone in
55 the troposphere is also a pollutant that impacts air quality [*The Royal Society*, 2008;
56 *Ravishankara et al.*, 2012].

57

58 Ozone in the troposphere is either transported from the stratosphere or produced photo-
59 chemically from pollutants emitted by anthropogenic and natural processes [*Crutzen*, 1995].
60 The first source is mostly dominant in the upper troposphere while the latter plays a major role
61 in the lower troposphere. The lifetime of ozone varies from hours near the surface to months
62 in the free troposphere. Since its lifetime is high in the mid- and upper troposphere, ozone (and
63 some of its precursors) can be transported over long distances [*Lelieveld et al.*, 2002, *Liu et al.*,
64 2003; *Lawrence and Lelieveld*, 2010; *Dentener et al.*, 2011]. Tropospheric ozone above the US
65 is affected by south and east Asian anthropogenic emissions due to the prevailing westerly
66 winds at mid-latitudes [*Cooper et al.*, 2005a, 2010; *Lin et al.*, 2012]. However, Asia is also
67 influenced by European emissions [*Wild et al.*, 2004; *Naja and Akimoto*, 2004]. Particularly
68 relevant to this analysis, a study of ozone profiles in the lower troposphere shows that
69 Ahmedabad in western India is affected by transport from Southern Europe and North Africa
70 [*Srivastava et al.*, 2012].

71

72 South and East Asian countries are experiencing strong industrial and economic growth leading
73 to increasing emissions of pollutants, many of which are ozone precursors [*Akimoto*, 2003;
74 *Streets et al.*, 2003; *Gurjar et al.*, 2004; *Lal et al.*, 2004; *Olivier et al.*, 2005; *Ohara et al.*,
75 2007]. Because the tropical regions of Asia are strongly affected by deep convection [*Emanuel*,

76 1994] the region's ozone and ozone precursors can be lofted to the mid- and upper troposphere,
77 with subsequent advection across great distances [*Kley et al.*, 1997; *Kar et al.*, 2004; *Folkins*
78 *and Martin*, 2005]. During the last decade, enhanced pollution in the upper troposphere has
79 been observed over Asia and the Middle East due to wide-spread deep convection during the
80 summer monsoon period. The polluted convective outflow can be confined over this region by
81 the strong anticyclonic circulation associated with the Asian summer monsoon [*Gettelman et*
82 *al.*, 2004; *Randel and Park*, 2006; *Randel et al.*, 2010; *Park et al.*, 2007; *Worden et al.*, 2009;
83 *Liu et al.*, 2009, 2011; *Barret et al.*, 2011].

84

85 India is a developing country located in the tropics and sub-tropics with over one billion
86 citizens. Hence, the atmosphere over this region is strongly impacted by increasing emissions
87 of pollutants due to anthropogenic activity, along with intense sunlight and high water vapour
88 content. However, studies related to tropospheric ozone are very limited for the Indian region
89 despite its importance for the northern hemisphere budgets of ozone and particulate matter
90 [*Lawrence and Lelieveld*, 2010], with many of these studies based on satellite data [*Fishman*
91 *et al.*, 2003; *Saraf and Beig*, 2004; *Beig and Singh*, 2007; *Fadnavis et al.*, 2010]. Here we
92 present an analysis of tropospheric ozone based on in situ ozonesonde observations from May
93 2003 to July 2007 to improve the scientific community's understanding of the ozone
94 distribution above western India and the transport processes that modulate its abundance.

95

96 **2. Location and meteorology**

97 Ahmedabad (23.03°N, 72.54°E, 50 m amsl) (See Figure 1) is an urban centre in central west
98 India, with a population of 5.6 million that has increased by about 20% in the last decade
99 [<http://www.censusindia.gov.in>]. The Thar Desert is located to the north-west (about 500 km)
100 and the Arabian Sea to the south-west with the closest shores being 100 km to the south and

101 400 km to the west. Physical Research Laboratory (PRL), where the ozonesondes were
102 launched, is located on the western side of the city. Textile mills and pharmaceutical production
103 facilities are located in and around the city with most industries in the eastern and northern
104 regions, about 15-20 km from PRL. In addition, a 400 MW coal fired power plant is located
105 approximately 10 km to the northeast. Ahmedabad has a large number (about 2.5 million) of
106 automobiles, which are increasing at the rate of about 10% per year.

107

108 Ahmedabad has a hot semi-arid climate. The climate is dry except during the summer monsoon
109 season. The weather is hot from April to June, while winds during December-February bring
110 milder conditions. The monthly average surface temperature is in the range of 28 °C to 35 °C
111 during summer and 20 °C to 25 °C during the winter months of November to February. The
112 southwest Asian summer monsoon produces a humid climate from mid-June to mid-September
113 with an average annual rainfall of about 750 mm. Most rainfall over Ahmedabad occurs during
114 July (~43%) and August (~27%).

115

116 The average wind patterns based on NCEP reanalysis data for December-January- February
117 (DJF), March-April-May (MAM), June-July-August (JJA) and September-October-November
118 (SON) on the 925, 500 and 200 hPa surfaces are shown in Figure 1. The winds over
119 Ahmedabad at 925 hPa are north-easterly during winter (DJF), westerly during spring (MAM),
120 south westerly during the summer monsoon (JJA) and north-westerly during autumn (SON).
121 The streamlines at 500 hPa are south-westerly during DJF, westerly during MAM, north-
122 easterly during JJA and again westerly during SON. At 200 hPa, the winds are stronger and
123 south-westerly during DJF, westerly during MAM and again south-westerly during SON.
124 However, the winds are easterly during the summer monsoon months with an anticyclone
125 pattern centred over the Himalaya and extending to the Middle East.

126

127 3. Techniques

128 3.1 Ozone and meteorological soundings

129 Vertical profiles of ozone, pressure, humidity and temperature were measured using balloon
130 borne ozonesondes coupled to radiosondes. Each ozonesonde consists of a Teflon pump, an
131 ozone sensing electrochemical concentration cell (ECC) [Komhyr and Harris, 1971] and an
132 electronic interface board. The ECC is comprised of Teflon cathode and anode chambers
133 containing platinum electrodes immersed in KI solutions of two different concentrations
134 thereby providing a potential difference to collect the electrons produced in the cathode
135 chamber from the reaction between the KI solution and ozone present in the ambient
136 atmosphere. Its accuracy is ± 5 -10% up to 30 km altitude [Smit *et al.*, 2007]. The Vaisala RS-
137 80 radiosonde consists of capacitance based temperature, humidity and pressure sensors. The
138 temperature and pressure sensors have accuracies below 20 km of ± 0.3 °C, and ± 0.5 hPa,
139 respectively. The heights are calculated based on the observed pressure and are above mean
140 sea level (AMSL). However, the humidity sensor has an accuracy of about $\pm 2\%$ near the
141 ground which decreases to ± 15 -30% in the 5-15 km altitude range [Kley *et al.*, 1997].

142

143 Balloon soundings carrying an ozonesonde and a radiosonde were made from the terrace of
144 PRL once every two weeks between 9:30-10:30 AM IST (UTC+ 5.5 hrs). Some of these
145 sondes also carried a GPS, which provided winds in addition to the position information. The
146 exact launch time depended on clearance from the Air Traffic Control of the local airport. The
147 first flight was made on 7 May, 2003 and the last on 11 July, 2007. There were breaks in
148 between due to technical problems or due to other campaigns. A total of 83 ozonesondes were
149 launched during this period. The balloons reached a maximum altitude of 30 to 33 km with an
150 average ascent rate of about 250 ms^{-1} . The instruments fell mostly within 150 km around the

151 launch site and some of the ozone sensors were recovered and were flown again after proper
152 cleaning, charging and testing.

153

154 Typical vertical distributions of air temperature, relative humidity and ozone are shown in
155 Figure 2. The humidity profile is shown up to about 20 km but errors are large above 10 km.
156 The figure shows structures in the humidity profile and often corresponding changes in
157 temperature. The ozone profile also shows variability sometimes linked to changes in humidity
158 and temperature. The ozone partial pressure increases rapidly around 18 km, peaks around 28
159 km, and decreases above this altitude.

160

161 **3.2. Lagrangian Dispersion Model**

162 The FLEXPART Lagrangian particle dispersion model (version 8.1) [Stohl *et al.*, 2005] was
163 used to simulate the 12-day transport history of each ozonesonde profile at 200 m intervals.
164 The model calculates the trajectories of a multitude of particles and was driven by ECMWF
165 ERA-interim global wind fields, with a temporal resolution of 6 h (analyses at 0000, 0600,
166 1200, and 1800 UTC), horizontal resolution of $0.7^\circ \times 0.7^\circ$, and 37 pressure levels. Particles
167 are transported both by the resolved winds and parameterized sub-grid motions, including a
168 vertical deep convection scheme.

169

170 To determine the transport history of each ozone measurement, a retroplume was calculated
171 (see Cooper *et al.*, [2005a, b] for a detailed illustration of the method) consisting of 40,000
172 back trajectory particles released from a box surrounding the location and time of each
173 measurement (1 hour duration; 10 km x 10 km x 200 m box) and advected backwards in time
174 over a 12-day period. Retroplume distributions were output in 1-day intervals on a $2^\circ \times 2^\circ$
175 degree output grid covering the globe, with a $0.5^\circ \times 0.5^\circ$ nested grid over India. FLEXPART

176 outputs the retroplumes in units of $\text{s kg}^{-1} \text{m}^3$, which is the residence time of the plume per grid
177 cell divided by the air density. The residence of each retroplume is calculated for the 300 m
178 layer of the atmosphere adjacent to the Earth's surface where the air would pick up surface
179 emissions (known as the footprint layer). For simplicity this specific volume weighted
180 residence time is hereafter referred to as the retroplume residence time.

181

182 The dispersion of a retroplume backwards in time indicates the likely source regions of the
183 ozone precursors (or stratospheric intrusions) that contributed to the measured ozone, but over
184 the previous 12 days. This is especially true of the so-called footprint layer which is the 300 m
185 layer adjacent to the Earth's surface. For plumes passing through this layer the residence time
186 of the back trajectory particles is folded with an anthropogenic NO_x emission inventory to
187 quantify the amount of anthropogenic NO_x emitted into the air mass represented by the
188 retroplume [Stohl *et al.*, 2003]. The NO_x emission inventory used in this study is the
189 EDGARv4.1 2005 annual data set, which estimates anthropogenic NO_x emissions on land
190 [Olivier *et al.*, 2005]. International shipping NO_x emissions are from the University of
191 Delaware 2001 inventory [Corbett and Koehler, 2003]. Monthly biomass burning emissions
192 were provided by the Global Fire Emission Database (GFED) version 3
193 (<http://www.globalfiredata.org>) [van der Werf *et al.*, 2010]. With this technique the quantity
194 of NO_x emitted into each retroplume from several source regions (India, Southeast Asia, China,
195 Japan-Korea, Central Asia, Middle East, Africa, Europe, North America and South America)
196 was tabulated. The NO_x tracer has no chemical or depositional removal processes and is treated
197 as a passive tracer. The ECMWF ERA-Interim analyses contain stratospheric ozone values
198 above the tropopause. We use these gridded values to calculate the quantity of ozone
199 transported from the stratosphere to the location where a retroplume is released. This method
200 involves tagging all retroplume back trajectory particles that originated in the stratosphere and

201 scaling the mass of each particle by the ozone value at its highest altitude in the lower
202 stratosphere. The accuracy of the stratospheric ozone tracer was also assessed using 260
203 ozonesonde profiles above the USA during summer 2004 [Cooper et al., 2005b, 2006]. The
204 result is an upper limit of the estimated stratospheric ozone that was directly transported from
205 the stratosphere to a retroplume release point over the previous 12 days.

206

207 **4. Results and discussions**

208 **4.1 Temperature and humidity variations**

209 The observed average air temperature in the troposphere is shown in Figures 3a and 3 c. The
210 average seasonal temperature profiles show minimum surface temperatures of about 20 °C in
211 winter (DJF) at the time of morning balloon launches (9:30 – 10:30 am). Winter temperatures
212 are lower than in all other seasons up to about 15 km. The highest average seasonal surface
213 temperature (about 30 °C) is observed in spring (MAM). But at 3-15 km, spring temperatures
214 are lower than during JJA and SON. The highest temperatures at 3-15 km are observed during
215 the summer monsoon season (JJA). The lapse rates vary widely below about 4 km in the
216 planetary boundary layer but between 4 km and 15 km, they are mostly in the range of -8 to -
217 5 °C/km. The tropopause is observed to be in the 16-18 km range.

218

219 Relative humidity varies widely with altitude and from month to month and is at a maximum
220 during the summer monsoon (JJA) and lower during winter and spring (Figures 3b and c). The
221 average monthly minimum at the surface occurs in November (~25%) and the maximum occurs
222 in July (~69%). The greatest values extend up to about 10 km during the peak monsoon month
223 of July. Occasionally, higher relative humidity is also observed in December/January due to
224 the winter monsoon.

225

226 **4.2 Ozone distribution in the troposphere**

227 The average seasonal profiles of ozone mixing ratios are shown in Figure 4a. Ozone increases
228 rapidly with altitude in the lower troposphere (below about 1 km height) during winter (DJF).
229 Average ozone is about 28 ppbv near the surface but increases to about 56 ppbv near 1 km.
230 This could be due to a shallow boundary layer during this season and loss near the surface due
231 to dry deposition or destruction by NO. Above 1 km, ozone decreases to about 45 ppbv at 3.5
232 km; above this altitude ozone increases slowly throughout the entire troposphere. In the middle
233 troposphere (4 to 10 km, height), average ozone is highest in spring (MAM) and the summer
234 monsoon (JJA) seasons and lowest in the winter season (DJF). Ozone is lowest (~18 ppbv)
235 during the monsoon season (JJA) near the surface and remains low below 3 km. However, at
236 7 to 9 km ozone is slightly greater (~ 66 ppbv) in JJA than during MAM. Ozone during autumn
237 (SON) is similar to spring below 3.5 km but it is lower than all other seasons between 9 and 16
238 km. The ozone profiles in MAM and SON have sharper increases than in other seasons above
239 13 km. The average ozone in the upper troposphere (above 10 km height) is highest in spring,
240 particularly in March-April and lowest in autumn (SON). Measurements made over Hilo,
241 Hawaii (19.4°N) have a maximum during spring in the free troposphere above 3 km [Cooper
242 et al., 2011]. The present results also show that the highest ozone values in the free troposphere
243 above Ahmedabad occur in spring. The lowest ozone in the 10-12 km range is observed in
244 SON at both the locations. However, overall, ozone values are greater above Ahmedabad in
245 all seasons, when compared with observations at Hilo.

246

247 Variation of ozone in the troposphere based on average monthly values at 200 m intervals using
248 all balloon flights during 2003-2007 is shown in Figure 4b. Ozone mixing ratios are lowest
249 below about 2 km during April to September. This is the period when the winds are from
250 south-west, bringing air from the marine regions of the Indian Ocean and the Arabian Sea.

251 Higher ozone mixing ratios are observed from October to March below about 2 km. However,
252 lower ozone levels are observed during these months between 3 and 5 km. In the mid-
253 troposphere, ozone values as high as 70-80 ppbv are observed in May-June at 6-9 km. The two
254 higher ozone spots seen in May at 6 and 8 km heights are caused by relatively higher ozone
255 peaks observed on 30 May 2007 and 4 May 2005 respectively. The FLEXPART analysis
256 indicated that these higher ozone containing air came from higher latitudes on these two days.
257 Lower ozone in the upper troposphere above 10 km is observed during August to November.
258 High ozone in the upper troposphere is observed throughout the year except during August to
259 October. Stratospheric intrusions penetrate deeper into the troposphere during March- April.

260

261 The average seasonal profiles discussed above show little vertical variability. However, there
262 is large variability in the vertical tropospheric ozone distribution from flight to flight. Figures
263 5a,b show individual ozone profiles on several days during different months of 2005. The
264 annual average profile is also shown for comparison. Large variability occurs not only in the
265 upper troposphere but throughout the troposphere. Figure 5a shows ozone profiles during
266 March and May 2005. Surface level ozone was observed to be lowest (only about 6 ppbv) on
267 9 March, but ozone was highest above 11 km (greater than 100 ppbv at 11 km). On the
268 contrary, ozone was highest (~ 65 ppbv) around 2 km on 16 March, but lowest (~30 ppbv)
269 around 11 km. The ozone profile on 4 May shows very high ozone (~105 ppbv) around 8.5
270 km, while on 16 March it was only about 25 ppbv at the same altitude. Similar sharp peaks
271 with values of 100 and 115 ppbv were observed at 6-9 km on 8 June, 2005 (Figure 5b). Such
272 high ozone layers have been observed at other locations [*Newell et al.*, 1999; *Lal et al.*, 2013].

273

274 Figure 6 shows observed monthly average ozone at selected heights covering different regions
275 of the troposphere. The variation of the monthly average tropospheric column ozone estimated

276 from the observed profiles are also shown for a comparison. Ozone mixing ratios at 0.5 km
277 (average of values within ± 0.5 km) show large variation with decreasing ozone from 44 ppbv
278 in January to a minimum of 18 ppbv in September. There is a sudden increase in ozone at this
279 height in October with a maximum of 53 ppbv in November. The average ozone values at 4.5,
280 8.5 and 12.5 km show different patterns than that at 0.5 km. Ozone values at 4.5 and 8.5 km
281 have maxima during April-May with lowest values in January. The variation at 12.5 km shows
282 sharper changes but the maximum ozone (81 ppbv) is observed in early spring (March-April).
283 Additionally, higher ozone is also seen in winter (December-January). The tropospheric ozone
284 column (TOC) is greatest in April (~44 DU) and least (~34-35 DU) during July, August and
285 September. Sharp increases in TOC are also observed from September to October, as at 0.5
286 km.

287

288 **4.3 Residence times of air parcels in different global regions**

289 The FLEXPART retroplume technique [Stohl *et al.*, 2003; Cooper *et al.*, 2005a; 2010] has been
290 used to calculate residence times of air parcels above several regions including India, SE Asia,
291 China, Japan-Korea, Central Asia, the Middle East, Africa, Europe, N. America and S. America
292 (Figure 7).

293

294 Figures 8a,b,c,d show the average residence times of the retroplumes released from
295 Ahmedabad over India, SE Asia, Central Asia, the Middle East, the Arabian Sea and other
296 regions of the world for different seasons, expressed as the percent of the total global surface
297 residence time for a retroplume over the previous 12 days. During winter (Figure 8a) in the
298 lowest km of the troposphere (within the planetary boundary layer), the percent residence time
299 over India is about 50%, but decreases sharply with altitude. The other major contributions
300 below 4 km are from the Arabian Sea and the Middle East. The contribution from Africa

301 dominates above 4 km, albeit in the range of 20-30%. Other regions contribute in the range of
302 15-20% except East Asia which is less than 10% above about 8 km. The ozone distribution in
303 this season, after the sharp increase in the first one km, decreases up to about 4 km. This is also
304 the height where the residence time over India becomes much less and the contribution from
305 Africa becomes dominant. In fact, ozone is lower than in any other season in the 3-8 km range
306 but it is higher above 10 km than in other seasons, such as the summer monsoon and post-
307 monsoon. The dip seen in the monsoon and post-monsoon seasons around 12 km is not seen in
308 winter. This seems to be due to the dominance of air coming from the Africa.

309

310 The average residence time percentages for the pre-monsoonal spring season (March, April
311 and May) are shown in Figure 8b. Compared to winter the contribution from India (25-35%)
312 has increased in the entire troposphere except in the lowest one km. The contribution from the
313 Arabian Sea (mostly in the range of 20-30%) has also increased as compared to winter. On the
314 contrary, the contribution from Africa has decreased and it is less than 25% even in the 8-12
315 km region. Contributions from other regions have also decreased. Ozone levels have increased
316 at all altitudes above 3 km as compared to winter. This change in residence times, particularly
317 the increase in the contribution from India is associated with increased ozone levels above 3
318 km.

319

320 During the monsoon season (June, July and August), the percent residence time over India
321 increases sharply above 2 km to approximately 55% in the 6-10 km range (Figure 8c). This is
322 also the region of the troposphere with the highest ozone mixing ratios. The percent residence
323 time over the Arabian Sea is greatest only below 2 km and decreases above this height. Above
324 8 km, the contribution from East Asia is the second greatest but only reaches 20%. This pattern

325 indicates a strong re-circulation of air above India during the summer monsoon that also
326 extends to SE Asia, the Middle East, Central Asia and the Arabian Sea.

327

328 The percent residence times during the post-monsoon (fall) season (SON) are shown in Figure
329 8d. Residence times over India dominate throughout the troposphere. Residence times are
330 about 55% in the lower 2 km, much greater than during the monsoon season, but above 4 km,
331 the Indian residence times are much lower than during the monsoon season. The percent
332 residence time over the Arabian Sea decreased to 25% below 3 km but increased (to 25%)
333 above this height in comparison to the monsoon season. The vertical ozone distribution
334 between the monsoon and fall (post-monsoon) seasons is correlated with the residence time
335 above India. Below 4 km both ozone and the Indian residence time increase from the monsoon
336 to the post-monsoon seasons, with the opposite effect above 4 km.

337

338 **4.4 Residence times over different regions**

339 The average monthly residence times are shown in Figure 9 for several regions. The residence
340 time over India is always higher below 2 km except during the monsoon season, with maximum
341 values occurring during September-December (note the colour scale, which is different than
342 over the other regions). The residence time over India is also higher throughout the troposphere
343 from mid-May to mid-October. Note that while the winds in the lower troposphere (<2 km)
344 reach Ahmedabad directly from the Indian Ocean and the Arabian Sea during the peak
345 monsoon period, the weak upper level winds allow air to reside over the Indian region during
346 the previous 12 days. During this period, the retroplumes also show transport from East Asia
347 in the mid-troposphere (6-12 km). The East Asian plume is also relatively stronger in the 7-12
348 km layer and during the July to September period (Figure 9), when the rains over Ahmedabad
349 are heaviest.

350

351 Average ozone levels during the monsoon months (JJA) are lowest below about 3km height as
352 the winds are directly from the Indian Ocean via the Arabian Sea. Even though peak ozone
353 levels occur in May at 5-10 km, ozone remains high during the monsoon and fall months in
354 this height range, while residence times are much higher for the Indian region and East Asian
355 region. This may be due to higher levels of pollutants.

356

357 The residence time over the Arabian Sea is more-or-less higher in the lower troposphere
358 throughout the year, mainly due to proximity to the western Indian region. The residence time
359 over the Arabian Sea is higher during April to October below 2 km. However, there is a dip
360 during July, when the transport pathway from the Indian Ocean touches the African east coast.
361 Ozone is lower throughout this period in this height region. The residence time over this marine
362 region is also higher at the beginning and end of the monsoon period.

363

364 March to May experiences hot and dry winds from the Middle East (Figure 9) below about 6
365 km. This effect is also seen beyond October in the 2-10 km range. This transport pattern only
366 corresponds to lower ozone mixing ratios. As mentioned earlier, July experiences transport
367 from the east coast of Africa below about 2 km. Transport is also from this region above 4 km
368 during October to May. The residence times of retroplumes during October to May above 4
369 km is dominated by the African region. However, there is also transport from North America
370 especially during March-May in the 4 to 8 km region above Ahmedabad.

371

372 **4.5 Long range transport effects**

373 Many recent studies have documented inter-regional and inter-continental air pollution
374 transport such as from Asia to North America [*Cooper et al.*, 2010; *Lin et al.*, 2012] and from

375 Asia to Europe [Lawrence and Lelieveld, 2010]. There is also evidence to show that pollutants
376 from Europe, North Africa, etc., also reach East Asia [Newell and Evans, 2000; Naja and
377 Akimoto, 2004] and in particular the south Asian region [Srivastava et al., 2011; Lal et al.,
378 2013]. Transport of pollutants from India to the surrounding marine regions was well
379 documented during INDOEX [Lelieveld et al., 2001] and other campaigns [Srivastava et al.,
380 2011; Lal et al., 2013].

381

382 The FLEXPART retroplume analysis over Ahmedabad shows transport of air parcels from
383 nearby regions or from as far away as North America depending upon altitude and season.
384 Figure 10a shows the average transport pathways during winter (DJF), spring (MAM),
385 monsoon (JJA) and post-monsoon/fall (SON) at 0-1 km, 7-8 km and 12-13 km. The
386 corresponding average heights of the retroplumes during the 12 days prior to the release are
387 shown in Figure 10b. Within the boundary layer (0-1 km) ozone over Ahmedabad is affected
388 in winter mostly by regional transport from the northwest across India, Pakistan, Afghanistan,
389 etc. (Figure 10a). However, in the upper troposphere during this season, ozone can be affected
390 by transport from North Africa, the North Atlantic Ocean, the Central US and the North Pacific
391 Ocean. The corresponding average height of the plumes during this season and during the past
392 12 days shows descending air from the free troposphere (4-5 km) down to the 0-1 km layer
393 (Figure 10b). However, there is almost no change in the height of the retroplume in the mid-
394 troposphere (7-8 km). The pattern in the 12-13 km layer shows a marginal lifting of the air
395 masses. The ozone distribution in spring is affected by dynamics similar to winter at all heights,
396 except that the transport patterns extend further to the west.

397

398 Transport characteristics during the monsoon are very different (Figures 10a,b). Ahmedabad
399 is affected by monsoon winds in the 0-1 km range from the Indian Ocean/Arabian Sea via the

400 east coast of Africa and the western Gulf countries. The mid-troposphere is affected by
401 transport from India, East Asia and to a small extent from North Africa. However, the upper
402 troposphere over Ahmedabad is affected by convective transport from India, the Middle East
403 and East Asia only. The closed region of recirculation is due to the persistent anticyclonic
404 winds associated with the summer monsoon flow. As in the earlier two seasons, the vertical
405 transport of the average retroplumes shows descent to the 0-1 km layer, marginal lifting to the
406 7-8 km layer and stronger convective lifting from below 8 km up to the 12-13 km layer (Figure
407 10b). In the monsoon season, ozone levels are lower in the upper troposphere compared to
408 winter and spring. But the lowest ozone levels are observed during August-November than in
409 any season at 10-14 km above sea level. These in situ observations are in contrast to other
410 studies based on satellite data which show that the lowest ozone values at these altitudes occur
411 during the Asian summer monsoon due to deep convection [*Park et al.*, 2007, 2009; *Randel et*
412 *al.*, 2010].

413

414 The unique transport pattern during the summer monsoon also produces conditions most
415 conducive for photochemical ozone production in the mid- and upper troposphere above
416 India. Figure 11 shows the abundance of the FLEXPART anthropogenic NO_x tracer above
417 Ahmedabad. This passive tracer with a 12-day lifetime indicates that relatively fresh
418 anthropogenic emissions are most common in the free troposphere during the summer
419 monsoon. Approximately 80% of the NO_x tracer in the upper troposphere is originated from
420 the surface of India. This result suggests that future changes in Indian emissions would have
421 their greatest impact on the chemical composition of the atmosphere above India during the
422 summer monsoon, a phenomenon that deserves further research, both in terms of chemical
423 transport modelling and in situ observations of chemistry and radiation.

424

425 The post-monsoon/fall season is the transition from monsoon to winter conditions and the
426 transport characteristics also change. The vertical transport of the retroplumes at all three levels
427 is similar to those in winter. Ozone levels are lowest above 9 km in this season.

428

429

430 **4.6 Stratosphere-Troposphere Exchange**

431 The stratosphere is a significant source of ozone for the middle and upper troposphere as a
432 result of stratosphere-troposphere exchange processes in the extra-tropics [Stohl *et al.*, 2003].
433 This flux is greatest in spring [Crutzen, 1995 and references therein], and mainly associated
434 with stratospheric intrusions into the troposphere that form filamentary structures that appear
435 as laminae in ozone profiles [Holton *et al.*, 1995; Stohl *et al.*, 2003]. Stratosphere-to-
436 troposphere transport has also been observed in the tropics involving the transport of
437 stratospheric intrusions from mid-latitudes or wave breaking in the sub-tropics [Baray *et al.*,
438 1998; Cooper *et al.*, 2005b]. Satellite observations have revealed higher ozone in the upper
439 troposphere above India due to transport from the stratosphere during winter and the pre-
440 monsoon period [Fadnavis *et al.*, 2010]. Balloon-based measurements have shown
441 stratospheric ozone intrusions in the mid- and upper troposphere above the Indian Ocean during
442 February-March 1999 [Zachariasse *et al.*, 2001] and above the Arabian Sea during May, 2006
443 [Lal *et al.*, 2013] and over the central Himalayas [Ojha *et al.*, 2014].

444

445 Figure 12 shows the quantity of stratospheric ozone in the troposphere above Ahmedabad
446 according to FLEXPART. The monthly values are averages corresponding to the times of the
447 ozonesonde measurements during 2003-2007. These results show enhanced ozone in March,
448 August and December in the upper troposphere. Modelled stratospheric contributions are
449 located down to 10 km, with ozone values of about 15-20 ppbv. Extremely high ozone (about

450 150 ppbv) was observed on 11 April 2007 at about 11.5 km. FLEXPART indicated that this
451 air parcel was transported from the Pacific Ocean via North Africa. It is interesting to note that
452 even around 5-6 km, FLEXPART indicates stratospheric ozone intrusions during April-June
453 with enhancements up to 15 ppbv. Ozone was observed to be more than 100 ppbv on 30 May
454 2007 at 6 km, with transport from the upper troposphere and lower stratosphere above the
455 eastern US, followed by descent to Ahmedabad via the high latitude region of Europe.

456

457 *Livesey et al.* [2013] observed seasonally enhanced ozone at 215 hPa (~ 12 km height) over
458 India during March-April using Microwave Limb Sounder (MLS) satellite data. We also
459 observed these high ozone values above Ahmedabad, but in addition our ozone profiles
460 demonstrate that the enhanced ozone during spring extends down to 6-9 km, a region of the
461 troposphere not visible to MLS.

462

463 **5. Summary and conclusions**

464 The vertical distribution of ozone was measured above Ahmedabad using ECC ozonesondes
465 launched twice per month during May 2003 to July 2007, for a total of 83 soundings. The
466 variability of ozone in the troposphere has been studied using these data. The tropospheric
467 column content is at a maximum (44 DU) in April and at a minimum (35 DU) during the
468 monsoon season (July to September). The maximum contribution to this total is associated with
469 altitudes below 4 km. The seasonal average vertical profiles show different features in the
470 three regions of the troposphere: i. below 4 km (lower troposphere), ii. between 4 and 10 km
471 (middle troposphere) and iii. between 10 and 16 km (upper troposphere). In the lower
472 troposphere ozone is greatest during winter (DJF) and lowest during the summer monsoon
473 (JJA). But in the middle troposphere, ozone is at a maximum in spring (MAM) followed by
474 JJA, and lowest in winter (DJF). In the upper troposphere, ozone is at a maximum in summer

475 and lowest in the fall (SON). Even though the seasonal ozone profiles do not show much
476 variability, individual profiles show large variability in the middle and upper troposphere,
477 where ozone ranges from 30 ppbv to 110 ppbv.

478

479 The FLEXPART retroplume technique was used to calculate residence times of air parcels over
480 several regions such as India, SE Asia, China, Japan-Korea, Central Asia, Middle East, Africa,
481 Europe, N. America and S. America. The residence times and locations of the plumes show
482 very different characteristics. In the lower troposphere (particularly below 2 km) during the
483 summer monsoon, transport is from the Indian Ocean and Arabian Sea via the east coast of
484 Africa. But during winter, the transport pathway is across north-western India, Pakistan and
485 Afghanistan. It is interesting to note that in the upper troposphere, the transport pathway is
486 centred over the central Asia region during the summer monsoon but can circle the globe in the
487 zonal direction during winter. This transport pattern can advect stratospheric ozone from higher
488 latitudes to Ahmedabad.

489

490 High values of pollutants like CO and low values of ozone have been observed in the upper
491 troposphere - lower stratosphere (UTLS) region during the summer monsoon above North
492 Africa and the Middle East using satellite retrievals (AIRS and MLS) [*Randel and Park, 2006;*
493 *Park et al., 2007, 2009*]. These observations are believed to be due to convectively lifted air
494 from the surface source regions during summer that becomes trapped in the anti-cyclonic winds
495 in the upper troposphere. Since the convection over India is strong during the monsoon season,
496 lower ozone in the upper troposphere during this season could be due to convective lofting of
497 low ozone air from the lower troposphere.

498

499 We observe low ozone over Ahmedabad in the 10-14 km layer from July to November. As
500 mentioned in the previous paragraph, we hypothesize that the low ozone in the UT is due to
501 convective lofting of surface air that is depleted in ozone. Other satellite measurements based
502 on TES retrievals show higher ozone in summer in the middle troposphere above North Africa,
503 the Middle East and India [Worden *et al.*, 2009; Liu *et al.*, 2009, 2011]. They argue that this
504 mid-tropospheric enhanced ozone in summer is due to long-range transport of ozone and local
505 production due to enhanced pollutant levels. As mentioned earlier, we find higher ozone in the
506 6-9 km layer during April-June. Additionally, March-April ozone enhancements detected by
507 MLS are verified here. FLEXPART indicates long range transport from North America, Africa
508 and the Middle East to Ahmedabad for this season and altitude range. Hence, different altitudes
509 and seasons have different transport and chemistry in the troposphere.

510

511 This is the first detailed analysis of ozone in the full troposphere above western India. These
512 measurements will be useful for the validation of satellite retrievals and model simulations.
513 While this data set gives a broader view of variability of ozone over Ahmedabad and the effects
514 of transport and chemistry, there is a need to understand the various sources and photochemical
515 processes using chemical transport models.

516

517 **Acknowledgements:** We thank PRL and ISRO GBP for encouraging and supporting this
518 balloon program at PRL. SL is grateful to D. Kley and H. Smit for their help in the initial
519 planning of this balloon sounding program at PRL. SL is also grateful to A. R. Ravishankara,
520 former Director (now at Colorado State University) of the NOAA Earth System Research
521 Laboratory's Chemical Sciences Division, Boulder, USA for supporting his stay there to
522 initiate this analysis. We thank Shilpy Gupta, K. S. Modh, T. A. Rajesh and T. K. Sunilkumar
523 for their support in conducting these balloon flights from Ahmedabad. We thank the

524 anonymous reviewers for their fruitful comments and suggestions, which have greatly
525 improved the MS. We are also grateful to the Editor for his encouragement and support. The
526 EDGARv4.1 global NO_x emissions inventory was provided by European Commission, Joint
527 Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL): Emission
528 Database for Global Atmospheric Research (EDGAR), release version 4.1
529 <http://edgar.jrc.ec.europa.eu>, 2010. The international shipping NO_x emission inventory was
530 provided by James Corbett, University of Delaware. Fire NO_x emissions are from the Global
531 Fire Emissions Database version 3 (GFED3).

532

533 **References**

- 534 Akimoto H. (2003), Global Air Quality and Pollution, *Science* 302, 1717-1719.
- 535 Baray, J., L. Baray, G. Ancellet, F. G. Taupin, M. Bessafi, S. Baldy, and P. Keckhut, (1998),
536 Subtropical tropopause break as a possible stratospheric source of ozone in the tropical
537 troposphere, *J. Atmos. Solar Terr. Phys.*, 60, 27–36.
- 538 Barret B., E. L. Flochmoen, B. Sauvage, E. Pavelin, M. Matricardi, and J. P. Cammas (2011),
539 The detection of post-monsoon tropospheric ozone variability over south Asia using
540 IASI data, *Atmos. Chem. Phys. Discuss.*, 11, 10031–10068
- 541 Beig, G., and V. Singh (2007), Trends in tropical tropospheric column ozone from satellite data
542 and MOZART model, *Geophys. Res. Lett.*, 34, L17801, doi:10.1029/2007GL030460.
- 543 Cooper, O. R., et al. (2005a), A springtime comparison of tropospheric ozone and transport
544 pathways on the east and west coasts of the United States, *J. Geophys. Res.*, 110,
545 D05S90, doi:10.1029/2004JD005183.
- 546 Cooper, O. R., et al. (2005b), Direct transport of mid-latitude stratospheric ozone into the lower
547 troposphere and marine boundary layer of the tropical Pacific Ocean, *J. Geophys. Res.*,
548 110, D23310, doi:10.1029/ 2005JD005783.

549 Cooper, O. R., et al. (2006), Large upper tropospheric ozone enhancements above
550 midlatitude North America during summer: In situ evidence from the IONS and
551 MOZAIC ozone measurement network, *J. Geophys. Res.*, 111, D24S05,
552 doi:10.1029/2006JD007306.

553 Cooper et al., (2010), Increasing springtime ozone mixing ratios in the free troposphere over
554 western North America, *Nature* 463, 344-348.

555 Cooper, O. R., et al. (2011), Measurement of western U.S. baseline ozone from the surface to
556 the tropopause and assessment of downwind impact regions, *J. Geophys. Res.*, 116,
557 D00V03, doi:10.1029/2011JD016095.

558 Corbett, J. J. and H. W. Koehler (2003), Updated emissions from ocean shipping, *J. Geophys.*
559 *Res.*, 108(D20), 4650, doi:10.1029/2003JD003751.

560 Crutzen P. J. (1995), Ozone in the troposphere in *Composition, chemistry, and climate of the*
561 *atmosphere*, Editor H. B. Singh, Van Norstrand Reinhold, New York. Dentener, F., T.
562 Keating, and H. Akimoto (Eds.) (2011), *Hemispheric Transport of Air Pollution 2010:*
563 *Part A: Ozone and Particulate Matter*, *Air Pollut. Stud*, vol. 17, U. N., New York.

564 Dentener, F., T. Keating, and H. Akimoto (Eds.) (2011), *Hemispheric Transport of Air*
565 *Pollution 2010: Part A: Ozone and Particulate Matter*, *Air Pollut. Stud*, vol. 17, U. N.,
566 New York. Emanuel K. A. (1994) *Atmospheric convection*, Oxford university press.

567 Fadnavis S., T. Chakraborty, and G. Beig (2010), Seasonal stratospheric intrusion of ozone in
568 the upper troposphere over India. *Ann. Geophys.*, 28, 2149–2159.

569 Fishman J., A. E. Wozniak, and J. K. Creilson (2003) Global distribution of tropospheric ozone
570 from satellite measurements using the empirically corrected tropospheric ozone
571 residual technique: Identification of the regional aspects of air pollution. *Atmos. Chem.*
572 *Phys.*, 3, 893–907.

573 Folkins I. and R. V. Martin, (2005), The vertical structure of tropical convection and its impact
574 on the budgets of water vapor and ozone, *J. Atm. Sci.* 62, 1560

575 Gauss, M., et al. (2003), Radiative forcing in the 21st century due to ozone changes in the
576 troposphere and the lower stratosphere. *J. Geophys. Res.* 108(D9),4292.
577 doi:10.1029/2002JD002624

578 Gettelman, A., D. E. Kinnison, T. J. Dunkerton, and G. P. Brasseur (2004), Impact of monsoon
579 circulations on the upper troposphere and lower stratosphere, *J. Geophys. Res.*, 109,
580 D22101, doi:10.1029/2004JD004878.

581 Gurjar B. R., J. A. van Aardenne, J. Lelieveld, M. Mohan (2004), Emission estimates and
582 trends (1990–2000) for megacity Delhi and implications, *Atmos. Env.* 38, 5663–5681

583 Holton J. R., P. H. Haynes, M. E. McIntyre, A. R. Douglass, R. B. Rood, L. Pfister (1995),
584 Stratosphere-troposphere exchange, *Reviews of Geophysics*, 33, 403–439.

585 Intergovernmental Panel on Climate Change (IPCC) (2013), Working Group I contribution to
586 the IPCC Fifth Assessment Report "Climate Change 2013: The Physical Science
587 Basis", Final Draft Underlying Scientific-Technical Assessment, <http://www.ipcc.ch>

588 Kar, J., et al. (2004), Evidence of vertical transport of carbon monoxide from Measurements
589 of Pollution in the Troposphere (MOPITT), *Geophys. Res. Lett.*, 31, L23105,
590 doi:10.1029/2004GL021128.

591 Kley, D., H. G. J. Smit, H. Vomel, H. Grassl, V. Ramanathan, P. J. Crutzen, S. Williams, J.
592 Meywerk, and S. J. Oltmans (1997), Tropospheric water vapour and ozone cross
593 sections in a zonal plane over the central equatorial Pacific, *Q. J. R. Meteorol. Soc.*,
594 123, 2009–2040.

595 Komhyr, W.D. and T. B. Harris (1971), Development of an ECC ozonesonde, NOAA Tech.
596 Rep. ERL 200, APCL 18, , Boulder, CO USA.

597 Lacis, A. A., Wuebbles, D. J. and Logan, J. A., (1990), Radiative forcing by changes in the
598 vertical distribution of ozone. *J. Geophys Res.* 95, 9971–9981.

599 Lal S., D. Chand, S. Venkataramani, K. S. Appu, M. Naja, P. K. Patra (2004), Trends in
600 methane, and sulfure hexafluoride at a tropical coastal site, Thumba (8.6N, 77E), in
601 India, *Atmos. Env.* 38(8), 1145-1151.

602 Lal S., S. Venkataramani, S. Srivastava, S. Gupta, C. Mallik, M. Naja, T. Sarangi, Y. B.
603 Acharya and X. Liu (2013), Transport effects on the vertical distribution of tropospheric
604 ozone over the tropical marine regions surrounding India , *J. Geophys. Res.*, 118,
605 doi:10.1002/jgrd.50180.

606 Lawrence, M. G., and J. Lelieveld (2010), Atmospheric pollutant outflow from southern Asia:
607 A review, *Atmos. Chem. Phys.*, 10, 11,017–11,096, doi:10.5194/acp-10-11017-2010.

608 Lelieveld, J. et al. (2001), The Indian Ocean Experiment: Widespread Air Pollution from South
609 and Southeast Asia, *Science*, 291, 1031–1036.

610 Lelieveld et al. (2002), Global Air Pollution Crossroads over the Mediterranean, *Science* 25
611 October 2002: 794-799. DOI:10.1126/science.1075457

612 Lin, M., et al. (2012), Transport of Asian ozone pollution into surface air over the western
613 United States in spring, *J. Geophys Res.*, 117, D00V07, doi:10.1029/2011JD016961

614 Liu, H., D. J. Jacob, I. Bey, R. M. Yantosca, B. N. Duncan, and G. W. Sachse (2003), Transport
615 pathways for Asian pollution outflow over the Pacific: Interannual and seasonal
616 variations, *J. Geophys. Res.*, 108(D20), 8786, doi:10.1029/2002JD003102.

617 Liu, J. J., D. B. A. Jones, J. R. Worden, D. Noone, M. Parrington, and J. Kar (2009), Analysis
618 of the summertime build up of tropospheric ozone abundances over the Middle East
619 and North Africa as observed by the Tropospheric Emission Spectrometer instrument,
620 *J. Geophys. Res.*, 114, D05304, doi:10.1029/2008JD010993.

621 Liu, J. J., D. B. A. Jones, S. Zhang, and J. Kar (2011), Influence of interannual variations in
622 transport on summertime abundances of ozone over the Middle East, *J. Geophys. Res.*,
623 116, D20310, doi:10.1029/2011JD016188.

624 Livesey N. J., J. A. Logan, M. L. Santee, J. W. Waters, R. M. Doherty, W. G. Read, L.
625 Froidevaux, and J. H. Jiang, (2013), Interrelated variations of O₃, CO and deep
626 convection in the tropical/subtropical upper troposphere observed by the Aura
627 Microwave Limb Sounder (MLS) during 2004–2011, *Atmos. Chem. Phys.*, 13, 579–
628 598.

629 Monks, P. S., et al. (2009), Atmospheric Composition Change – Global and Regional Air
630 Quality, *Atmos. Environ.*, 43, 5268-5350.

631 Naja and Akimoto (2004), Contribution of regional pollution and long-range transport to the
632 Asia-Pacific region: Analysis of long-term ozonesonde data over Japan, *J. Geophys.*
633 *Res.*, 109, D21306, doi:10.1029/2004JD004687.

634 Newell, R., and M. Evans (2000), Seasonal changes in pollutant transport to the North
635 Pacific: The relative importance of Asian and European sources, *Geophys. Res. Lett.*,
636 27, 2509– 2512.

637 Newell R. E., V. Thouret, J. Y. N. Cho, P. Stoller, A. Marenco and H.G. Smit (1999), Ubiquity
638 of quasi-horizontal layers in the troposphere, *Nature*, 398, 316-319.

639 Ohara, T., H. Akimoto, J. Kurokawa, et al., (2007), Asian emission inventory for anthropogenic
640 emission sources during the period 1980–2020. *Atmos. Chem. Phys.*, 7, 4419–4444.

641 Ojha, N., M. Naja, T Sarangi, R Kumar, P Bhardwaj, S Lal, S Venkataramani, R Sagar, A
642 Kumar, HC Chandola, (2014), On the processes influencing the vertical distribution of
643 ozone over the central Himalayas: Analysis of yearlong ozonesonde observations,
644 *Atmospheric Environment*, 88, 201-211.

645 Olivier, J. G. J., van Aardenne, J. A., Dentener, F., Pagliari, V., Ganzeveld, L. N., and J. A.
646 H.W., Peters (2005) Recent trends in global greenhouse gas emissions: regional trends
647 1970–2000 and spatial distribution of key sources in 2000, *Environ. Sci.*, 2, 81–99,
648 doi:10.1080/15693430500400345.

649 Park, M., W. J. Randel, A. Gettelman, S. T. Massie, and J. H. Jiang (2007), Transport above
650 the Asian summer monsoon anticyclone inferred from Aura Microwave Limb Sounder
651 tracers, *J. Geophys. Res.*, 112, D16309, doi:10.1029/2006JD008294.

652 Park, M., W. J. Randel, L. K. Emmons, and N. J. Livesey (2009), Transport pathways of carbon
653 monoxide in the Asian summer monsoon diagnosed from Model of Ozone and Related
654 Tracers (MOZART), *J. Geophys. Res.*, 114, D08303, doi:10.1029/2008JD010621.

655 Randel, W. J., and M. Park (2006), Deep convective influence on the Asian summer monsoon
656 anticyclone and associated tracer variability observed with Atmospheric Infrared
657 Sounder (AIRS), *J. Geophys. Res.*, 111, D12314, doi:10.1029/2005JD006490.

658 Randel W. J., et al. (2010), Asian Monsoon Transport of Pollution to the Stratosphere, *Science*
659 328, 611; DOI: 10.1126/science.1182274

660 Ravishankara A. R., J. P. Dawson, D. A. Winner (2012), New Directions: Adapting air quality
661 management to climate change: A must for planning, *Atmos. Env.* 50, 387–389

662 Saraf N. and G. Beig (2004), Long-term trends in tropospheric ozone over the Indian tropical
663 region. *Geophys. Res. Lett.*, 31, L05101, doi:10.1029/2003GL018516.

664 Smit, H. G. J., et al. (2007), Assessment of the performance of ECC-ozonesondes under quasi-
665 flight conditions in the environmental simulation chamber: Insights from the Juelich
666 Ozone Sonde Intercomparison Experiment (JOSIE), *J. Geophys. Res.*, 112, D19306,
667 doi:10.1029/2006JD007308.

668 Srivastava S., S. Lal S. Venkataramani, S. Gupta, and Y. B. Acharya (2011), Vertical
669 distribution of ozone in the lower troposphere over the Bay of Bengal and the Arabian

670 Sea during ICARB-2006: Effects of continental outflow, *J. Geophys. Res.*
671 doi:10.1029/2010JD015298.

672 Srivastava S., S. Lal, M. Naja, S. Venkataramani and S. Gupta (2012), Influences of regional
673 pollution and long range transport to western India: Analysis of ozonesonde data,
674 *Atmos. Env.* 47, 174-182

675 Stevenson et al. (2012), Tropospheric ozone changes, radiative forcing and attribution to
676 emissions in the Atmospheric Chemistry and Climate Model Inter-comparison Project
677 (ACCMIP), *Atmos. Chem. Phys. Discuss.*, 12, 26047–26097.

678 Stohl, A., et al. (2003), Stratosphere-troposphere exchange: A review, and what we have
679 learned from STACCATO, *J. Geophys. Res.* 108, NO. D12, 8516,
680 doi:10.1029/2002JD002490.

681 Stohl, A., C. Forster, S. Eckhardt, N. Spichtinger, H. Huntrieser, J. Heland, H. Schlager, H.
682 Aufmhoff, F. Arnold, and O. Cooper (2003), A backward modeling study of
683 intercontinental pollution transport using aircraft measurements, *J. Geophys. Res.*,
684 108(D12), 4370, doi:10.1029/ 2002JD002862.

685 Stohl, A., C. Forster, A. Frank, P. Seibert, and G. Wotowa (2005), Technical note: The
686 Lagrangian particle dispersion model FLEXPART version 6.2, *Atmos. Chem. Phys.*, 5,
687 2461–2474.

688 Streets, D.G., Bond, T.C., Carmichael, G.R., Fernandes, S.D., Fu, Q., He, D., Klimont, Z.,
689 Nelson, S.M., Tsai, N.Y., Wang, M.Q., Woo, J., Yarber, K.F., (2003). An inventory
690 of gaseous and primary aerosol emissions in Asia in the year 2000. *J. Geophys. Res.*
691 108, 8809. doi:10.1029/2002JD003093. The Royal Society (2008), Ground-level
692 Ozone in the 21st century: Future Trends, Impacts and Policy Implications, Royal
693 Society policy document 15/08, RS1276,
694 http://royalsociety.org/Report_WF.aspx?pageid57924&terms5ground-level1ozone

695 The Royal Society (2008), Ground-level Ozone in the 21st century: Future Trends, Impacts
696 and Policy Implications, Royal Society policy document 15/08, RS1276,
697 http://royalsociety.org/Report_WF.aspx?pageid57924&terms5ground-level1ozone
698 van der Werf, G. R. et al., (2010), Global fire emissions and the contribution of deforestation,
699 savanna, forest, agricultural, and peat fires (1997-2009), *Atmos. Chem. Phys.*, 10,
700 11707-11735, doi:10.5194/acp-10-11707-2010.

701 Wild, O., M.J. Prather, H. Akimoto, J.K. Sundet, I.S.A. Isaksen, J.H. Crawford, D.D. Davis,
702 M.A. Avery, Y. Kondo, G.W. Sachse, and S.T. Sandholm (2004), CTM Ozone
703 Simulations for Spring 2001 over the Western Pacific: Regional ozone production and
704 its global impacts, *J. Geophys. Res.*, 109, D15S02, doi:10.1029/2003JD004041.

705 Worden, J. et al., (2009), Observed vertical distribution of tropospheric ozone during the Asian
706 summer time monsoon. *J. Geophys. Res.*, 114, D13304, doi:1029/2008JD010560

707 Zachariasse, M., H. Smit, P. van Velthoven, and H. Kelder (2001), Cross-tropopause and
708 interhemispheric transports into the tropical free troposphere over the Indian Ocean, *J.*
709 *Geophys. Res.*, 106(D22), 28441-28452.

710

711 **Figure Captions:**

712 Figure 1: Average streamlines based on NCEP reanalysis data during winter (DJF), spring
713 (MAM), monsoon (JJA) and fall (SON) seasons at 925, 500 and 200 hPa. The color bar code
714 at the bottom is wind speed (ms^{-1}).

715

716 Figure 2: Ozone, temperature and relative humidity profiles as observed from a balloon ascent
717 on 25 June, 2003 above Ahmedabad.

718

719 Figure 3: (a) Average temperature, (b) relative humidity during different months and (c)
720 average seasonal profiles of both, observed from the balloon ascents above Ahmedabad during
721 2003-2007.

722

723 Figure 4: (a) Average ozone mixing ratios (ppbv) during different seasons and (b) during
724 different months observed from the balloon ascents above Ahmedabad during 2003-2007.

725

726 Figure 5: (a) Ozone profiles observed above Ahmedabad on different dates during March-May
727 2005 and (b) during Jun-August 2005, along with the annual average profile.

728

729 Figure 6: Average ozone during different months and at different heights based on all the
730 balloon ascents made over Ahmedabad during 2003-2007. Variations in the monthly average
731 tropospheric ozone columns are shown.

732

733 Figure 7: Example of a FLEXPART retroplume released from 100 m above mean sea level
734 (amsl) at Ahmedabad, 4:00 UTC, 27 January, 2005. Shown are the retroplume pathway
735 throughout the entire atmospheric column (top left), and only within the 300 m footprint layer
736 (top right). The center of the 12-day retroplume is denoted by the white back trajectory with
737 black dots indicating the mean location every 24-hours and white labels indicating every
738 second day. Magenta boxes outline the regions for which NO_x emissions are totaled. Also
739 shown are the 2x2 degree EDGAR NO_x emission inventory (center left) and the quantity of
740 NO_x emitted into the retroplume from each of the regions of interest (center right). Similarly,
741 the emission inventory and NO_x emitted into the retroplume at 0.5x0.5 degree resolution across
742 South Asia are shown at bottom left and bottom right, respectively.

743

744 Figure 8: Percent residence times over different regions and for different seasons using the
745 average FLEXPART model results for all the balloon flights conducted from Ahmedabad
746 during 2003-2007. Average ozone profiles for each season are also shown.

747

748 Figure 9: Monthly average residence times (kSec) over major regions using the FLEXPART
749 model results for all the balloon flights conducted from Ahmedabad during 2003-2007. Please
750 note different scale used for India.

751

752 Figure 10: Composite diagrams showing FLEXPART 12-day retroplumes for 0-1 km
753 (Column-1), 7-8 km (Column-2) and 12-13 km (Column-3) and for Dec-Jan-Feb (Row-1),
754 Mar-Apr-May (Row-2), Jun-Jul-Aug (Row-3) and Sep-Oct-Nov (Row-4). Panel a shows
755 transport pathways while Panel b shows average vertical movement of the retroplumes during
756 the 12 days prior to release. Retroplumes correspond to the launch times of all balloon ascents
757 during the respective season.

758

759 Figure 11: Estimated NO_x (ppbv) over Ahmedabad using the FLEXPART model.

760

761 Figure 12: Monthly average contribution of ozone from the stratosphere based on FLEXPART
762 model calculations.

763

764

765

766

767

768

769

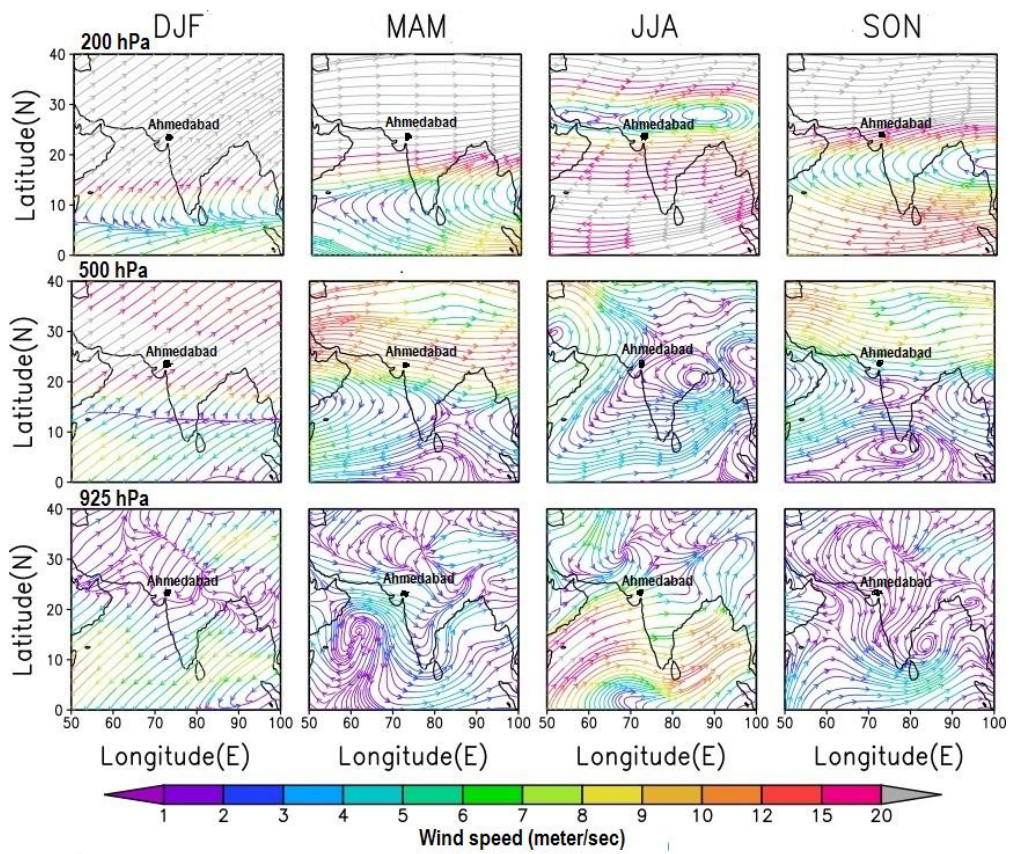
770

771

772

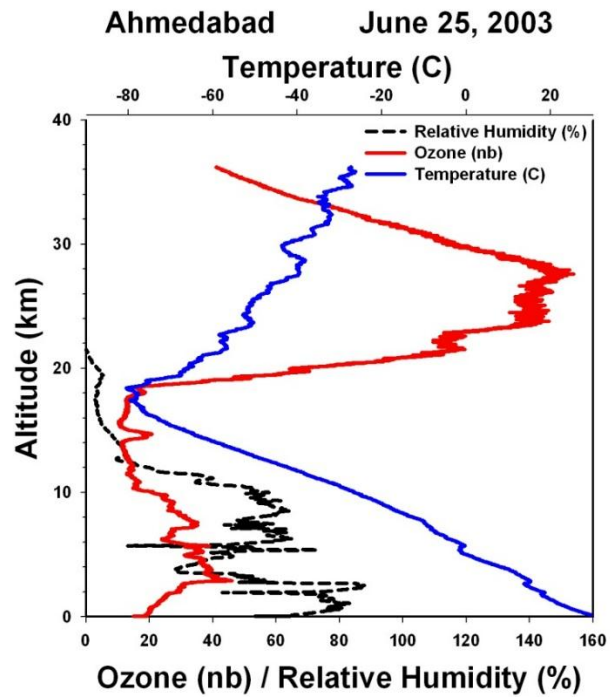
773 **Figures:**

774 **Figure 1:**



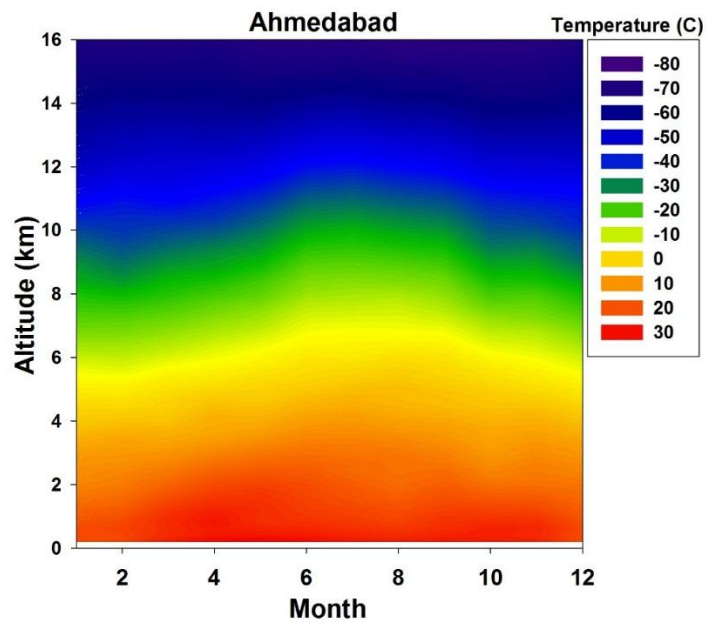
775

776 **Figure 2:**



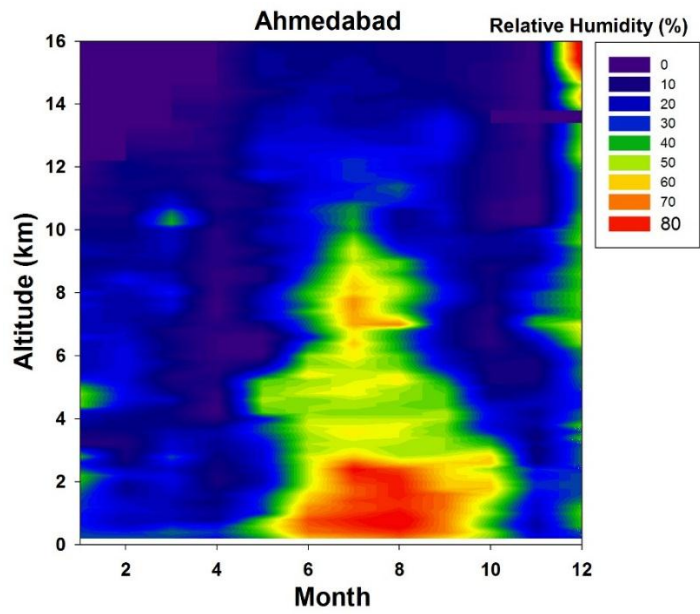
777

778 **Figure 3a:**



779

780 **Figure 3b:**



781

782

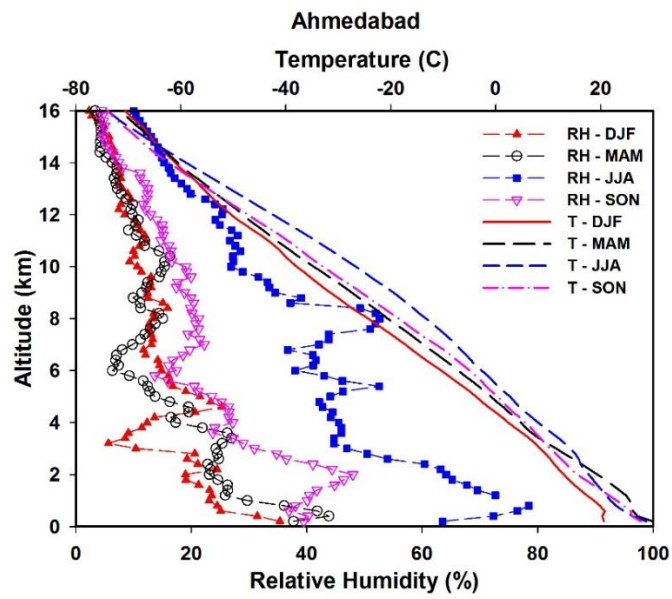
783

784

785

786

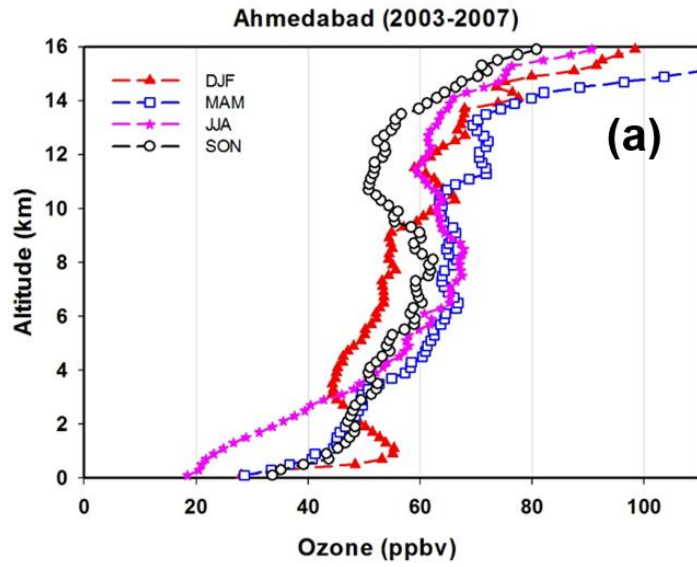
787 **Figure 3c:**



788

789

790 **Figure 4a:**



791

792

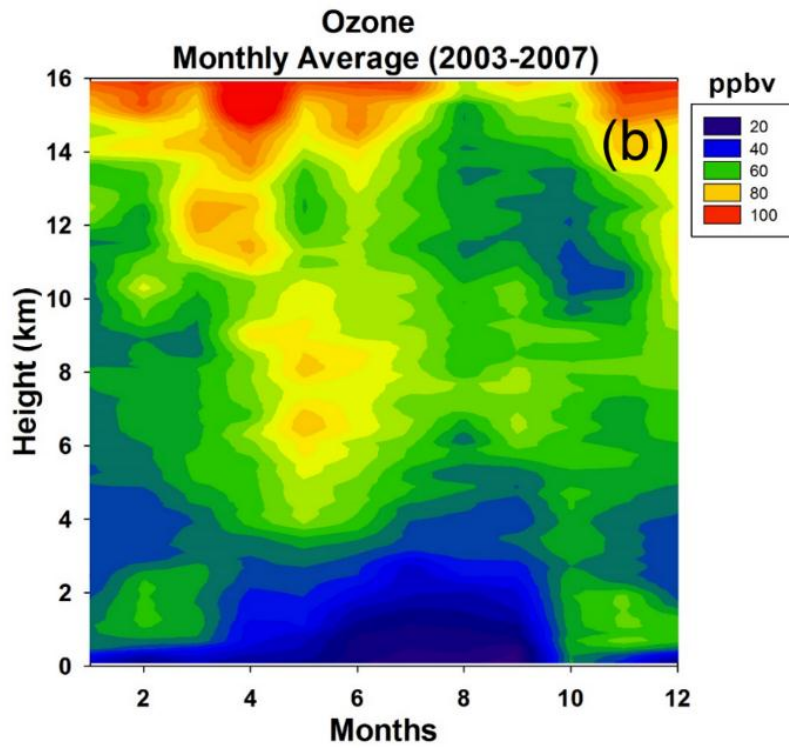
793

794

795

796

797 **Figure 4b:**



798

799

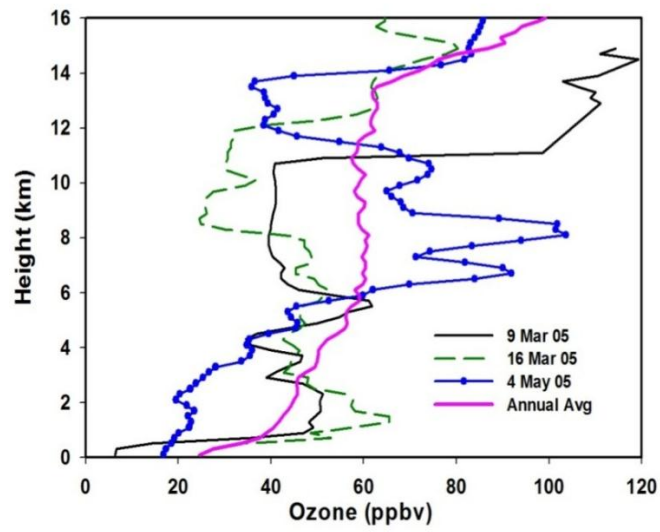
800

801

802

803

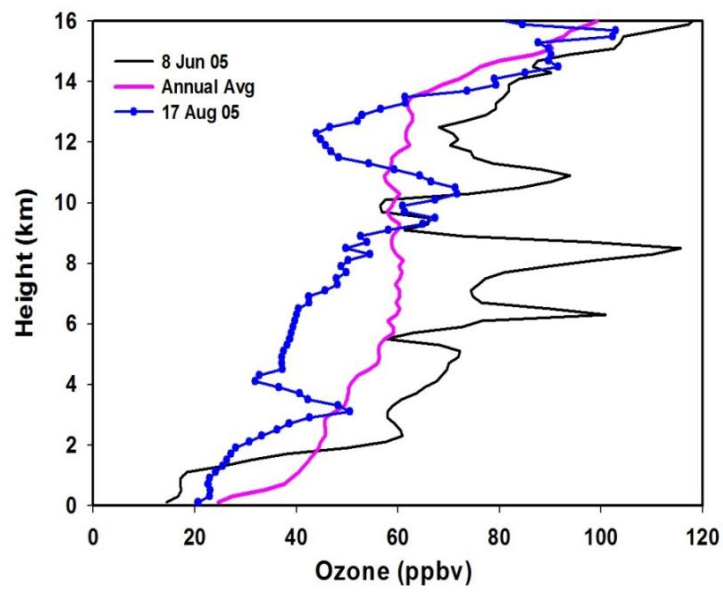
804 **Figure 5a:**



805

806 **Figure 5b:**

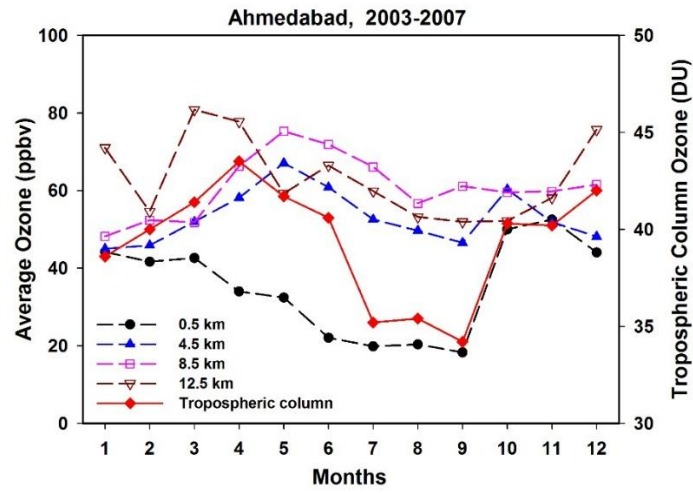
807



808

809

810 **Figure 6:**



811

812

813

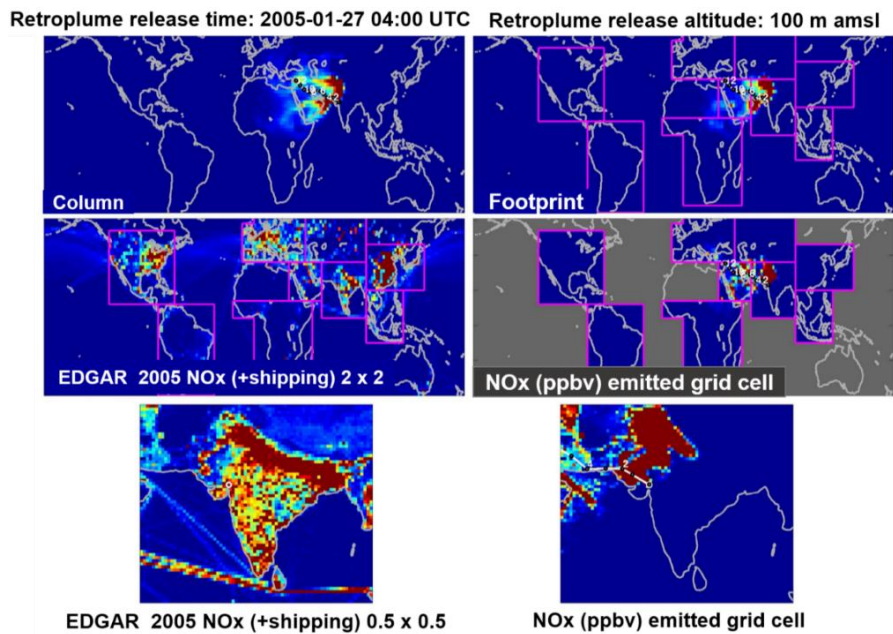
814

815

816

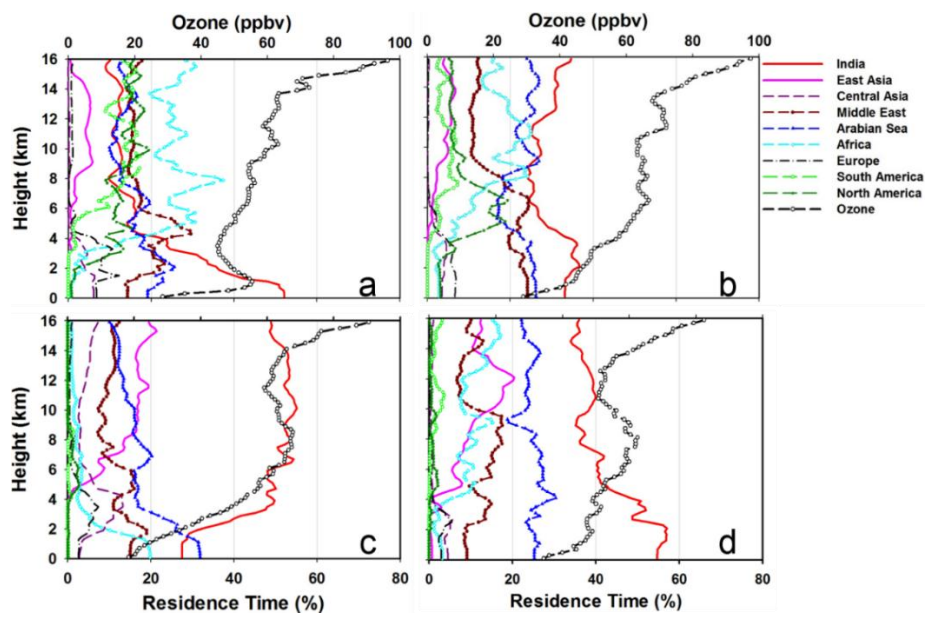
817

818 **Figure 7:**



819

820 **Figure 8:**



821

822

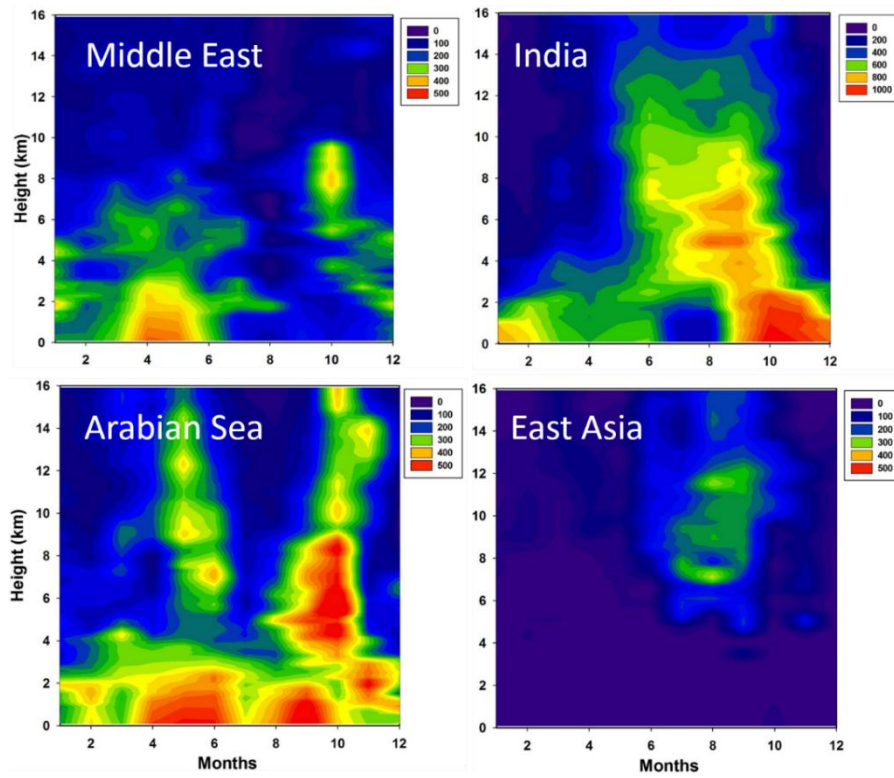
823

824

825

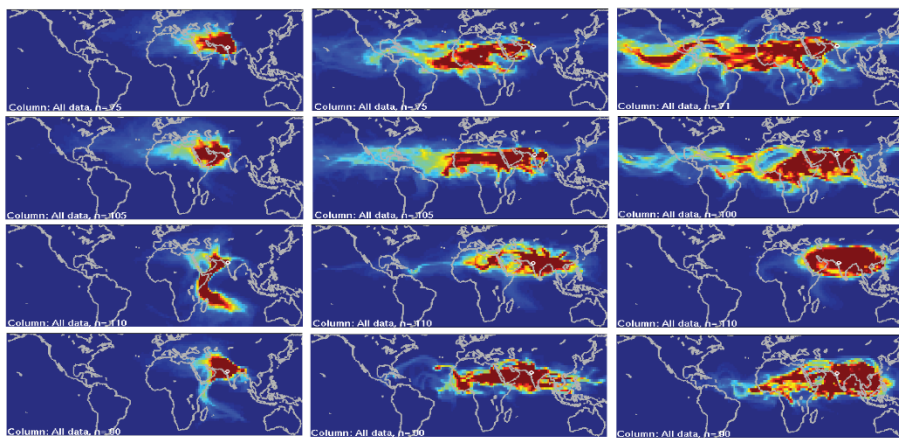
826

827 **Figure 9:**



828

829 **Figure 10 a:**



830

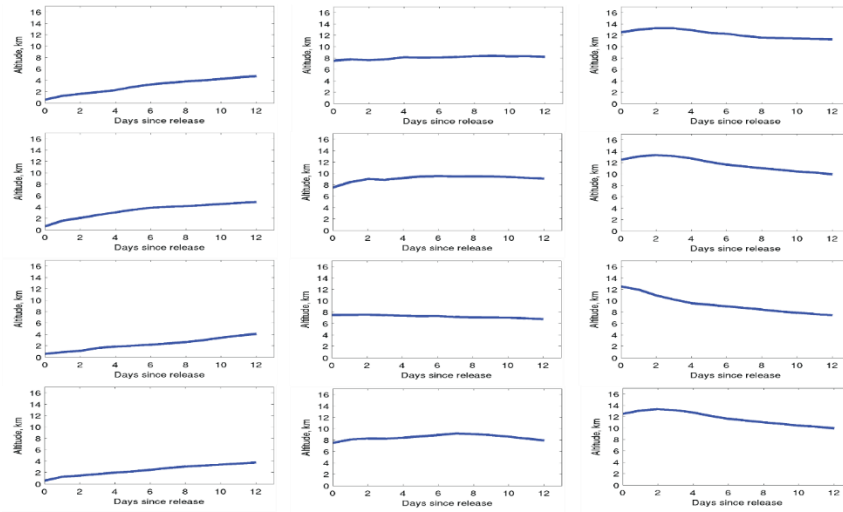
831

832

833

834

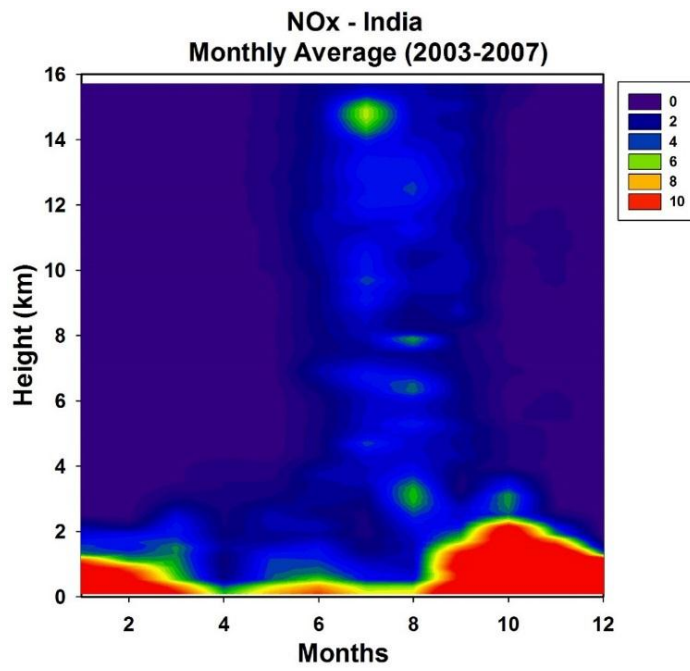
835 **Figure 10b:**



Row 1: Dec-Jan-Feb, 0-1 km, 7-8 km, 12-13 km
 Row 2: Mar-Apr-May, 0-1 km, 7-8 km, 12-13 km
 Row 3: Jun-Jul-Aug, 0-1 km, 7-8 km, 12-13 km
 Row 4: Sep-Oct-Nov, 0-1 km, 7-8 km, 12-13 km

836

837 **Figure 11:**



838

839

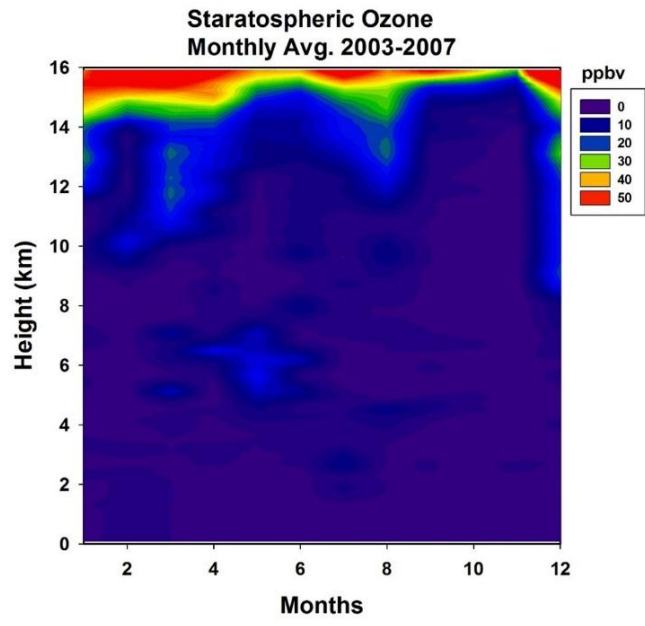
840

841

842

843

844 **Figure 12:**



845

846

847

848

849

850

851

852

853