Supplementary material

WRF-Chem simulated surface ozone over South Asia during the pre-monsoon: Effects of emission inventories and chemical mechanisms

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S1. Brief evaluation of precursors (NOx, ethane and ethene)

We include an evaluation of modeled NOx, ethane and ethene against recent measurements (Table S1). Significant differences are seen in NOx mixing ratios at Delhi in the north with only INTEX-RADM2 being within 1 standard deviation of the observed value. Ozone production at Delhi is VOC limited in all simulations in the present study (seen from CH2O/NOy ratio in Fig. 5). This makes it important to conduct observations of NMVOCs in the Delhi region. At Kanpur also NOx from INTEX-RADM2 compares better with the observed values. At Mt. Abu in the west, NOx from HTAP-RADM2 compares better with observed values, however it should be noted that the site is also impacted by transported ozone during spring (Naja et al., 2003). At Udaipur, all simulations tend to underpredict NOx. At Haldia in the east, NOx from S4RS-RADM2 compares better with observed value which is also in line with the results for ozone in the east region in this study. At Nainital, modelled NOy is evaluated and is seen to be within 1 standard deviation variability of the observed value in all simulations.

Modelled ethane mixing ratios are quite similar in all simulations and agree well with observed values at Mt. Abu but are underpredicted at Nainital by a factor of about 2. On the other hand, modelled ethene mixing ratios at both Mt. Abu and Nainital agree relatively well with observed values in INTEX-RADM2 and S4RS–RADM2 as compared to HTAP-RADM2.

We would like to mention that the observations of precursors are very sparse in the south Asian region and it is important to have an evaluation over a network of observations, as we present for ozone in this study, to understand their contribution into ozone formation and also the budget of NMVOCs over the region. However this does not affect the conclusions of the present study.

Table S1. Comparison of modeled monthly average (for April) precursor mixing ratios (in ppbv) with observations at several stations.

Specie	Site	Reference	Observations ±1 σ std	HTAP-RADM2	INTEX-RADM2	S4RS-RADM2	HTAP-MOZ
NOx	Delhi	SAFAR data	59.8±27.5	208.7	64.4	187.2	188.9
	Kanpur	Gaur et al. (2014)	5.0	10.2	6.5	30.5	9.1
	Mt. Abu	Naja et al. (2003)/ Kumar et al (2012b)	2.1	1.7	1.1	1.1	1.4
	Udaipur	Yadav et al. (2014)	8.7±4.2	2.1	1.6	1.5	2.0
	Haldia	Purkait et al. (2008)	12.6	4.4	3.5	8.2	4.6
NOy	Nainital	Sarangi et al. (2014)	1.8±1.6	3.2	2.7	2.9	2.6
NMVOC (*ethane*)	Nainital	Sarangi et al. (2016)	2.3	1.2	1.2	1.1	1.0
	Mt. Abu	Sahu and Lal (2006)	1.3	1.1	1.1	1.1	1.0
NMVOC (*ethene*)	Nainital	Sarangi et al. (2016)	0.9	1.2	0.9	0.8	0.9
	Mt. Abu	Sahu and Lal (2006)	0.3	0.7	0.5	0.5	0.6

Table S2. Diurnal mean biases (model-observations) in ppbv for all runs at different sites.

Site	HTAP-RADM2	INTEX-RADM2	S4RS-RADM2	HTAP-MOZ
Mohali	-3.4	-5.5	-9.4	1.0
Nainital	0.6	-2.0	-1.2	8.3
Pantnagar	17.8	15.1	16.1	24.7
Delhi	-6.5	-5.4	-12.3	-16.0
Dibrugarh	21.6	20.6	16.8	39.1
Kanpur	18.2	11.4	10.0	17.8
Mt. Abu	4.7	3.0	2.0	3.9
Udaipur	11.8	10.3	9.7	11.4
Jabalpur	-10.2	-15.1	-12.4	-2.1
Ahmedabad	8.7	8.1	7.8	3.7
Haldia	13.0	10.0	9.9	25.7
Bhubaneshwar	6.4	4.8	4.8	13.1
Joharapur	9.0	5.1	8.0	12.0
Pune	5.1	2.2	4.7	6.9
Anantpur	-5.3	-12.9	-11.2	4.8
Gadanki	7.1	3.2	3.8	11.6
Kannur	15.7	13.4	12.7	13.6
Thumba/Trivendrum	11.7	9.9	6.1	12.0

Table S3. Noontime (1130-1630 IST) average biases in ppbv for all runs at different sites.

Site	HTAP-RADM2	INTEX-RADM2	S4RS-RADM2	HTAP-MOZ
Mohali	-17.6	-20.8	-24.8	-13.5
Nainital	8.1	5.8	6.2	17.0
Pantnagar	10.5	7.4	8.5	20.4
Delhi	-12.3	-21.5	-26.0	-11.3
Dibrugarh	28.8	31.5	24.6	40.7
Kanpur	23.5	12.5	15.7	28.7
Mt. Abu	9.7	7.3	6.5	10.6
Udaipur	9.4	7.5	6.8	10.7
Jabalpur	0.9	-8.0	-2.5	8.8
Ahmedabad	10.7	9.4	6.9	10.7
Haldia	19.4	14.8	14.0	29.3
Bhubaneshwar	10.2	12.2	7.3	19.9
Joharapur	16.5	11.1	14.4	23.1
Pune	10.5	4.4	10.5	15.2
Anantpur	0.2	-10.0	-8.7	13.5
Gadanki	14.6	7.9	8.7	25.2
Kannur	22.5	14.9	15.0	27.4
Thumba/Trivendrum	24.0	19.9	11.1	35.8

Table S4. Diurnal mean biases in ppbv over different regions in all model runs.

Region	HTAP-RADM2	INTEX-RADM2	S4RS-RAMD2	HTAP-MOZ
North	5.4	2.7	0.6	9.4
East	13.7	11.8	10.5	21.4
West	7.9	5.8	6.4	6.4
Central	-10.2	-15.1	-12.4	-2.1
South	7.3	3.4	2.9	10
Overall	7.0	4.2	3.6	10

Table S5. Quantitative assessment of similarity between HTAP-RAMD2, INTEX-RADM2 and SEAC4RS-RADM2 scenarios for 24 h average and noontime (1130-1630 IST) average for simulated surface ozone mixing ratios.

24 h average	**HTAP-RADM2 (a) vs INTEX-RADM2 (b)**	**HTAP-RADM2 (a) vs S4RS-RADM2 (b)**	**INTEX-RADM2 (a) vs S4RS-RADM2 (b)**
r^2	0.98	0.98	0.99
variance of the residual (b-a)	4.61	5.32	2.05
Noontime average			
r^2	0.96	0.96	0.98
variance of the residual (b-a)	18.26	21.24	11.70

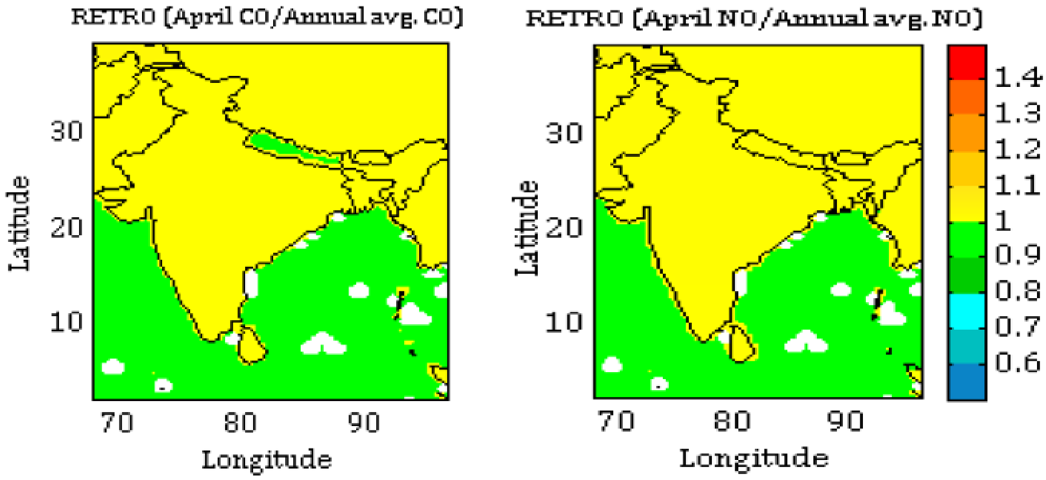

Figure S1. Spatial distribution of ratio of April emissions to annual average emissions for CO (left) and NO (right) derived from the RETRO inventory (http://accent.aero.jussieu.fr/RETRO_metadata.php).

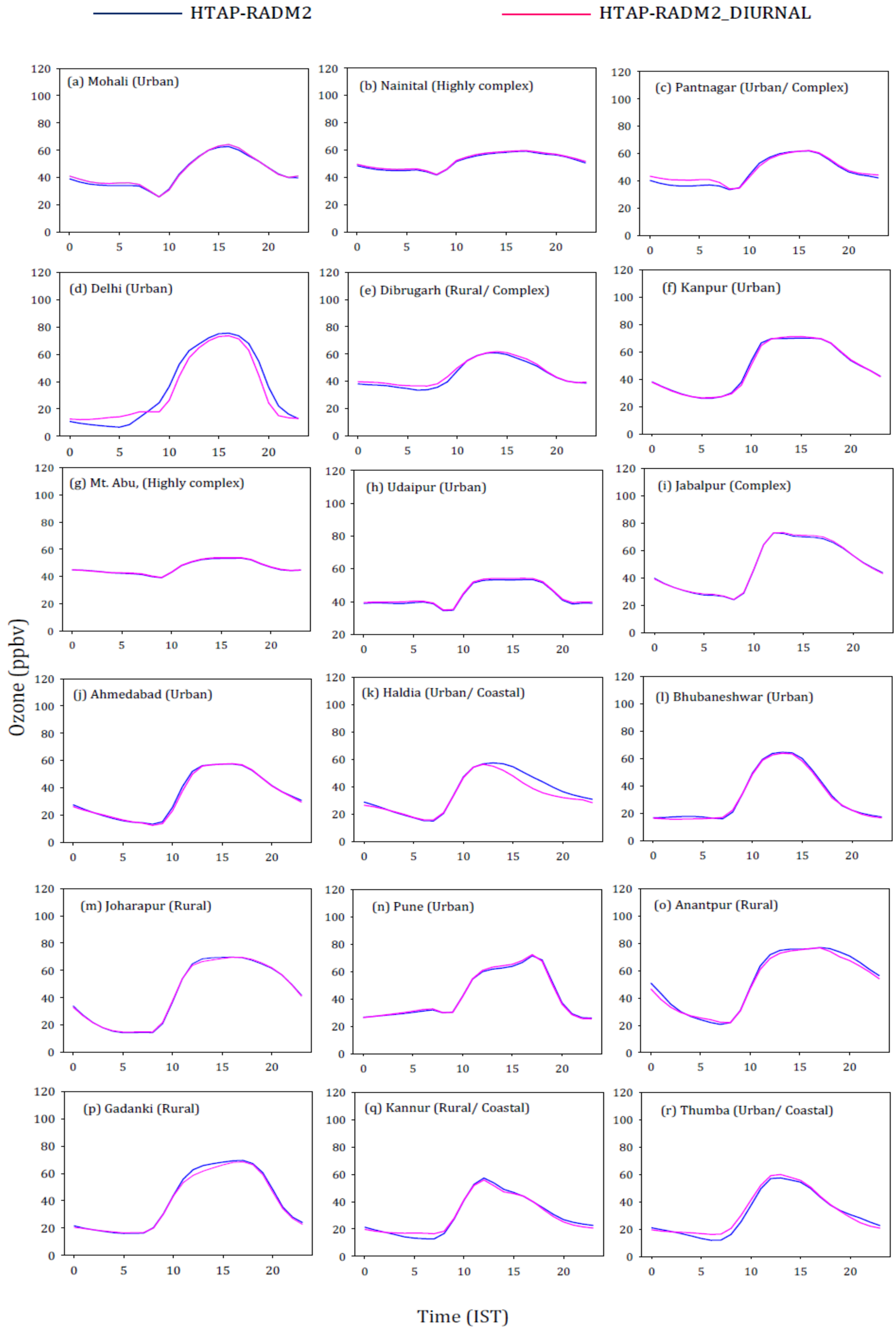

Figure S2. Comparison of 15 day average (01st April, 2013 – 15th April, 2013) diurnal variation of surface ozone simulated for anthropogenic emissions with and without the incorporation of diurnal profile of emissions at various observation sites. Both model simulations are with the HTAP inventory and RADM2 chemistry option.

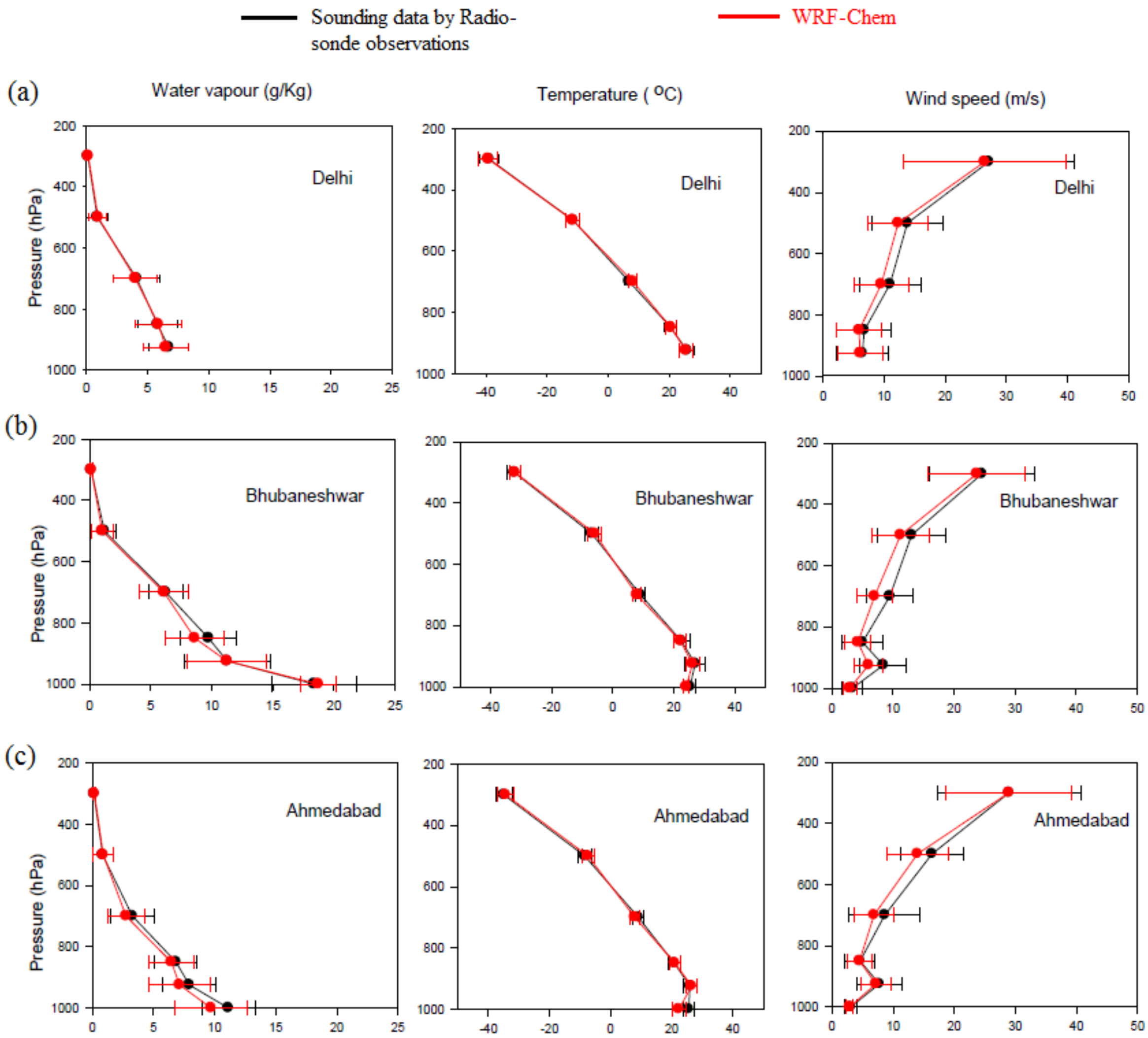

Figure S3. Vertical profiles of monthly average (April 2013) water vapour mixing ratio (g/Kg), temperature (^{o}C) and wind speed (m/s) from WRF-Chem (in red) and sounding data (in black) at (a) Delhi (in north India); (b) Bhubaneshwar (in east India); and, (c) Ahmedabad (in west India). Horizontal bars represent temporal standard deviation of monthly averages.

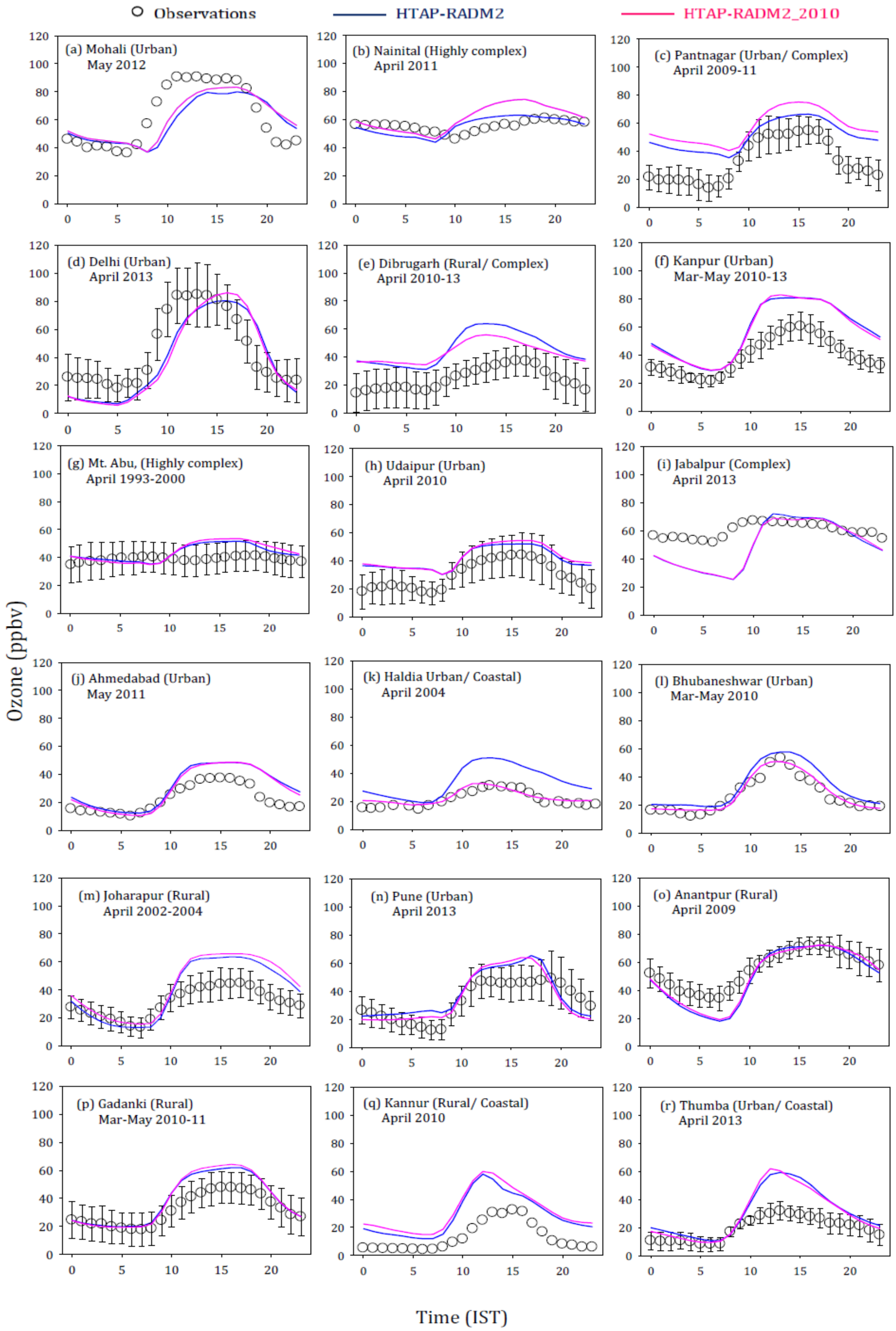

Figure S4. Comparison of monthly average diurnal variation of surface ozone simulated for the years 2010 and 2013 at various observation sites. The observational data is available for the period indicated in the figure. Error bars represent the temporal standard deviation of the monthly averages. All model simulations are with the HTAP inventory and RADM2 chemistry option.

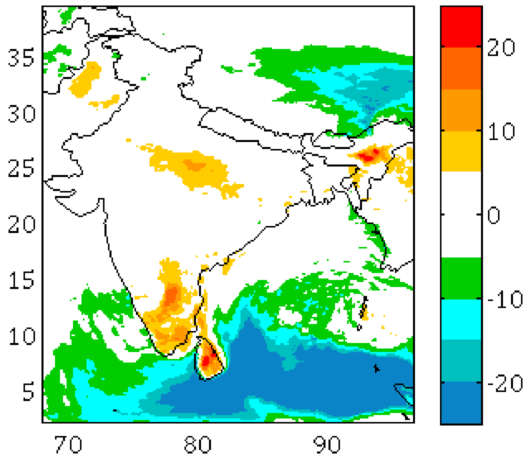

Figure S5. Percentage difference in monthly average surface ozone mixing ratio (ppbv) during April between S4RS-RADM2_kf run (using Kain-Fritsch cumulus parameterization scheme) and S4RS-RADM2 i.e. base run (using Grell 3D scheme). The percentage difference is calculated relative to base run.

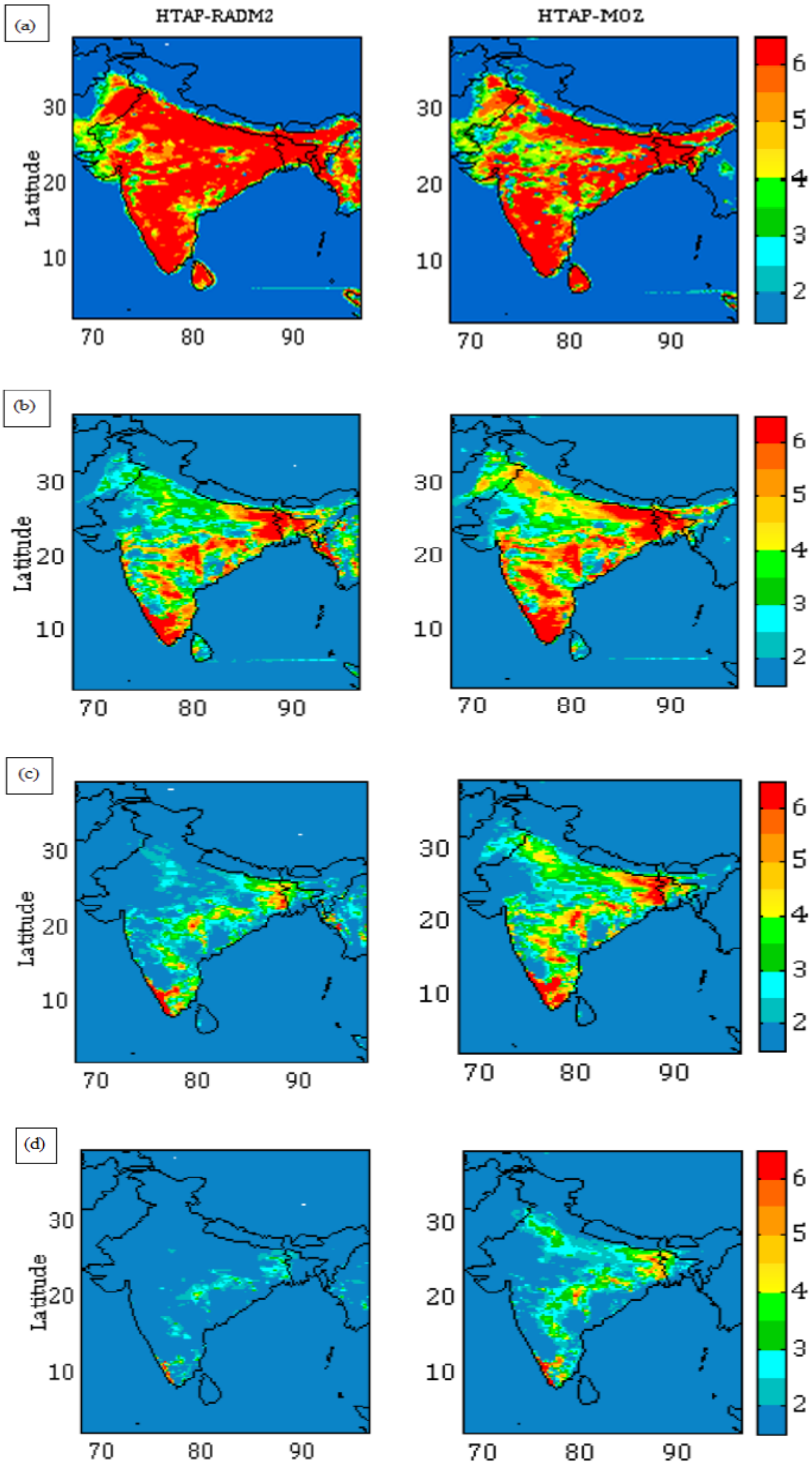

Figure S6. Spatial distribution of net daytime ozone chemical tendency (in ppbv h^{-1}) at model (a)level 1 - surface; (b) level 4; (c) level 5; and (d) level 6 during 0630-1230 IST. The pressure distribution at these model levels is shown in figure S9. Note the colour scale difference with respect to Fig. 9 in the manuscript.

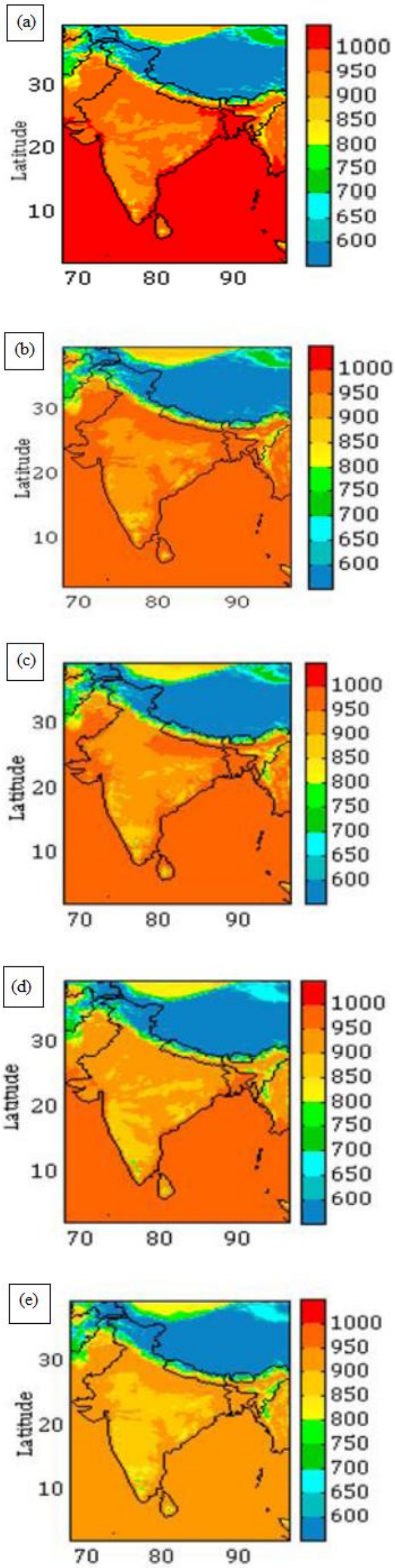

Figure S7. Spatial distribution of atmospheric pressure (in hPa) at model (a) level 1 - surface; (b) level 3; (c) level 4; (d) level 5 and (e) level 6.

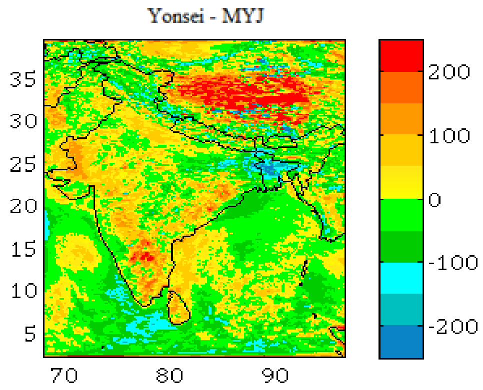

Figure S8. Difference in monthly average (in April) PBL height in meters between simulations with Yonsei and MYJ parameterization (i.e. base run) with HTAP-RADM2 setup.

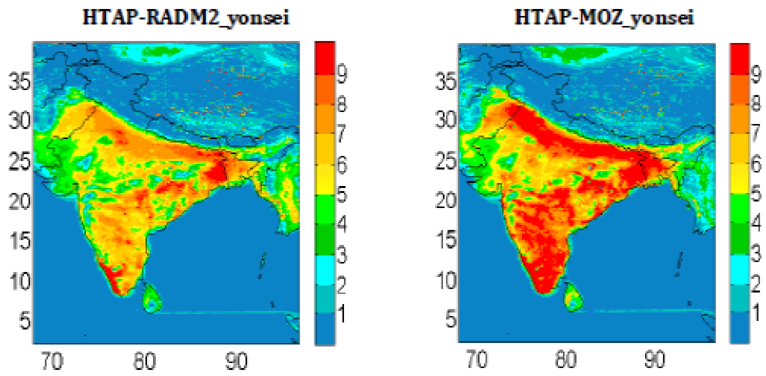

Figure S9. Average net daytime surface ozone chemical +vertical mixing tendency (in ppbv h^{-1}) for April during 0630-1230 IST for HTAP-RADM2 and HTAP-MOZ setup but with the Yonsei PBL scheme.

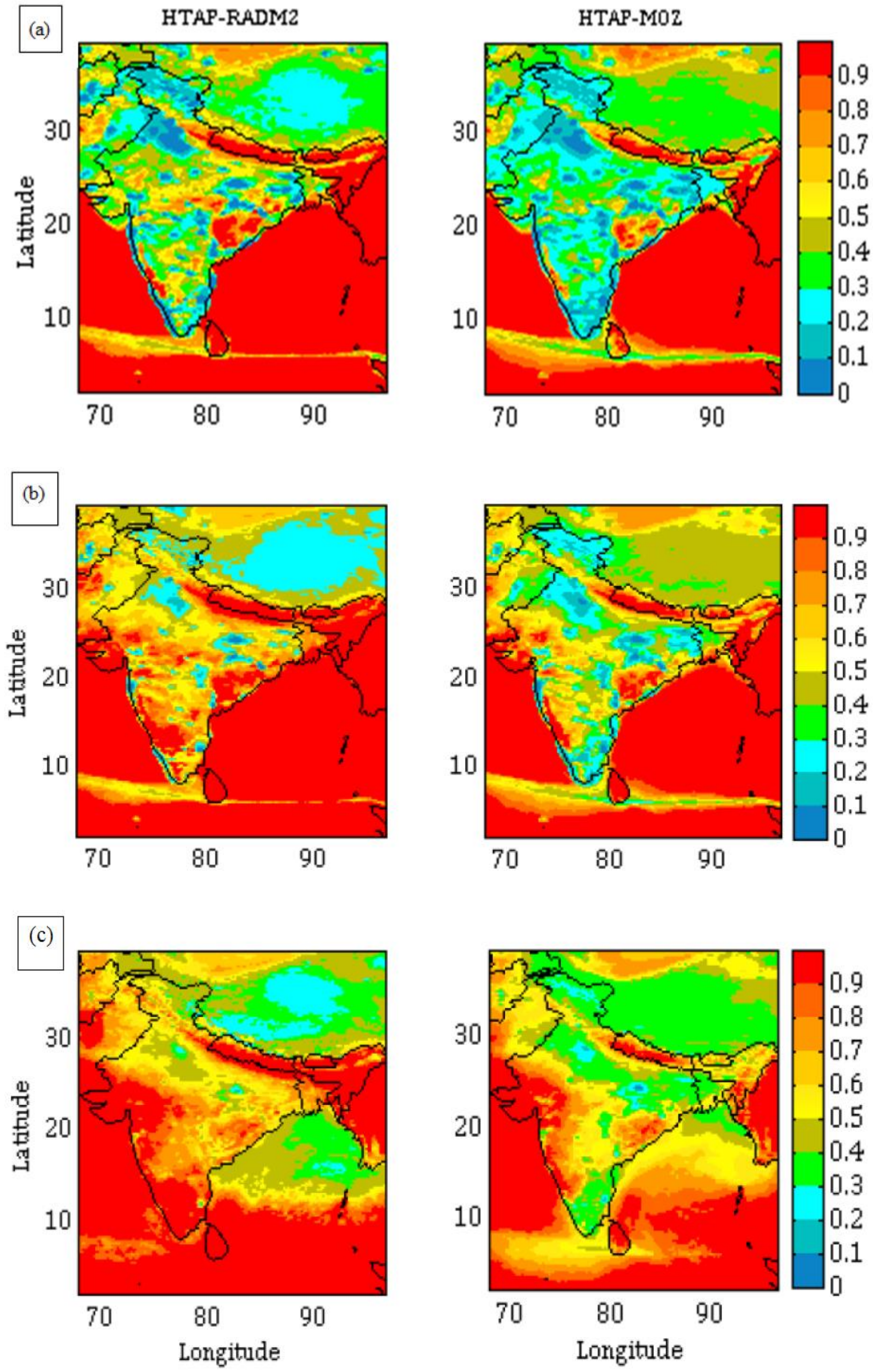

Figure S10. Spatial distribution of daytime (0630-1230 IST) average CH_2O/NO_y ratio comparing simulations with the MOZART and RADM2 chemical mechanisms at model (a) level 1- surface (b) level 3; and (c) level 6 during April. Note the colour scale difference with respect to Fig. 10 in the manuscript.

References

Gaur, A., Tripathi, S. N., Kanawade, V. P., Tare, V., and Shukla, S. P.: Four-year measurements of trace gases (SO2, NOx , CO, and O3) at an urban location, Kanpur, in Northern India, J. Atmos. Chem., 71, 283–301, 2014.

Naja, M., Lal, S., and Chand, D.: Diurnal and seasonal variabilities in surface ozone at a high altitude site Mt Abu (24.6N, 72.7E, 1680 m asl) in India, Atmospheric Environment 37, 4205-4215, 2003.

Purkait, N. N., De, S., Sen, S., and Chakrabarty, D. K.: Surface ozone and its precursors at its two sites in the northeast coast of India, Indian Journal of Radio and Space Physics, 38, 86–97, 2009.

Renuka, K., Gadhavi, H., Jayaraman, A., Lal, S., Naja, M., and Rao, S.: Study of Ozone and NO2 over Gadanki – a rural site in South India, J. Atmos. Chem., 71, 95–112, doi:10.1007/s10874-014-9284-y, 2014.

Sahu, L. K., and Lal, S.: Characterization of C2-C4 NMHCs distributions at a high altitude tropical site in India, J. Atmos. Chem., 54, 161–175, http://doi.org/10.1007/s10874-006-9023-0, 2006.

Sarangi, T., Naja, M., Ojha, N., Kumar, R., Lal, S., Venkataramani, S., Kumar, A., Sagar, R., and Chandola, H. C.: First simultaneous measurements of ozone, CO and NOy at a high altitude regional representative site in the central Himalayas, J. Geophys. Res.-Atmos., 119, 1592–1611, doi:10.1002/2013JD020631, 2014.

Sarangi, T., Naja, M., Lal, S., Venkataramani, S., Bhardwaj, P., Ojha, N., Kumar, R., and Chandola, H. C.: First observations of light non-methane hydrocarbons (C2–C5) over a high altitude site in the central Himalayas, Atmos. Environ., 125, 450-460, 2016.

Sharma, S. K., Datta, A., Saud, T., Saxena, M., Mandal, T. K., Ahammed, Y. N., and Arya, B. C.: Seasonal variability of ambient NH3, NO, NO2 and SO2 over Delhi, J. Environ. Sci, 22 (7), 1023–1028, 2010.

Yadav, R., Sahu, L. K., Jaaffrey, S. N. A., and Beig, G.: Distributions of ozone and related trace gases at an urban site in western India. J. Atmos. Chem. 71, 125–144, 2014.