Supplementary Material

Table S1. Isoprene oxidation chemistry in AM3. T represents temperature (K).

Reactions	Reaction Rate	Note
Isoprene Daytime Chemistry		
ISOP + OH → ISOPO2	3.10×10^{-11}exp(350./T)	
ISOPO2→ 2.0*HO2 + CH2O + .333*MGLY + .5*GLYALD + 0.25*GLYX	4.07×10^{8}exp(-7694./T)	
ISOPO2 + NO → .90*NO2 + .90*HO2 + .9*CH2O + .55*MVK + 0.35*MACR + 0.1*ISOPNB	2.70×10^{-12}exp(350./T)	
ISOPO2 + HO2 → .937*ISOPOOH + .063*OH + .025*MACR + .038*MVK + .063*HO2 + .063*CH2O	2.06×10^{-13}exp(1300./T)	
ISOPO2 + ISOPO2 → 1.28*HO2 + .92*CH2O + .56*MVK + .36*MACR + .48*ROH + .5*HC5	1.54×10^{-13}	
ISOPO2 + CH3O2 → 1.1*HO2 + 1.22*CH2O + .28*MVK + .18*MACR + .3*HC5 + .24*CH3OH + .24*ROH	8.37×10^{-14}	
ISOPO2 + CH3CO3 → .887*HO2 + .747*CH2O + .453*MVK + .294*MACR + .14*HC5 + .113*DIBOO + {CO2} + CH3O2	1.68×10^{-12}exp(500./T)	
ISOPO2 + CH3CO3 → MEK + CH3COOH	1.87×10^{-13}exp(500./T)	
ISOPNB + OH → ISOPNBO2	2.40×10^{-12}exp(745./T)	
ISOPNBO2 + NO → .09*GLYALD + .09*HYAC + .69*CH2O + 0.88*NO2 + .44*MACRN + .69*HO2 + .26*MVKN + 0.21*DHDN	2.40×10^{-12}exp(360./T)	
ISOPNBO2 + HO2 → .06*GLYALD + .06*HYAC + .44*CH2O + .28*MACRN + .16*MVKN + .06*NO2 + .44*HO2 + .5*OH + .5*ISNP + 0.5*ROOH	8.70×10^{-14}exp(1650./T)	
ISOPNB + O3 → 0.05*HO2 + 0.05*OH + 0.11*MVKN + 0.32*MACRN + 0.16*HCOOH + 0.62*CH2O + 0.36*{CO2} + 0.21*CO + 0.06*C4NACID + 0.36*HPROPN + 0.1*MVKNOOH	3.70×10^{-19}	
ISNP + OH → .612*OH + .612*R4N1 + .386*ISOPNBO2	4.75×10^{-12}exp(200./T)	
ISOPOOH + OH → .387*ISOPO2 + .613*OH + .613*HC5	4.75×10^{-12}exp(200./T)	
ISOPOOH + OH → OH + IEPOX	1.90×10^{-11}exp(390./T)	
IEPOX + OH → IEPOXOO	5.78×10^{-11}exp(-400./T)	
IEPOXOO + HO2 → .725*HYAC + .275*GLYALD + .275*GLYX + .375*CH2O + .251*CO + .275*MGLY + 1.125*OH + .825*HO2	2.06×10^{-13}exp(1300./T)	
IEPOXOO + NO → .725*HYAC + .275*GLYALD + .275*GLYX + .375*CH2O + .074*HCOOH	2.70×10^{-12}exp(350./T)	

+ .251*CO + NO2 + .275*MGLY + .125*OH+ .825*HO2		
HC5 + OH → HC5OO	3.35×10^{-11}exp(380./T)	
HC5 + O3 → .6*MGLY + .1*OH + .12*CH2O + .28*GLYALD + .3*O3 + .4*CO + .2*H2 + .2*HYAC + .2*HCOOH	6.16×10^{-15}exp(-1814./T)	
HC5OO + NO → NO2 + .216*GLYX + .234*MGLY + .234*GLYALD + .09*RCHO + HO2 + .09*CO + .216*HYAC + .29*DHMOB + .17*MOBA	2.35×10^{-12}exp(350./T)	
HC5OO + NO → HNO3	3.50×10^{-13}exp(350./T)	
HC5OO + HO2 → .1*IAP + .9*OH + .9*MGLY + .9*GLYALD + .9*HO2	2.06×10^{-13}exp(1300./T)	
HC5OO + CH3O2 → .5*HO2 + .33*CO + .09*H2 + .18*HYAC + .25*C2H5OH + .5*HO2 + .13*GLYALD + .29*MGLY + .25*MEK+ .95*CH2O + .25*CH3OH	8.37×10^{-14}	
HC5OO + CH3CO3 → .216*GLYX + .234*MGLY + .234*GLYALD + .216*HYAC + .29*DHMOB + .17*MOBA + .09*RCHO + HO2 + .09*CO + CH3O2	1.68×10^{-12}exp(500./T)	
HC5OO + CH3CO3 → MEK + CH3COOH	1.87×10^{-13}exp(500./T)	
IAP + OH → .654*OH + .654*DHMOB + .346*HC5OO	5.31×10^{-12}exp(200./T)	
MOBA + OH → MOBAOO	2.79×10^{-11}exp(380./T)	
MOBA + O3 → OH + HO2 + MEK	2.00×10^{-17}	
MOBAOO + NO → RCHO + HO2 + NO2	2.35×10^{-12}exp(350./T)	
MOBAOO + NO → HNO3	3.50×10^{-13}exp(350./T)	
MOBAOO + HO2 → .5*OH + .5*HO2 + .5*RCHO + .5*C3H7OOH	2.06×10^{-13}exp(1300./T)	
MVK + OH → MVKO2	2.60×10^{-12}exp(610./T)	
MVK + O3 → .202*OH + .202*HO2 + .352*HCOOH + .535*CO + .05*CH3CHO + .95*MGLY + .05*CH2O	8.50×10^{-16}exp(-1520./T)	
MVKO2 + NO → .965*NO2 + .249*HO2 + .249*CH2O + .716*CH3CO3 + .716*GLYALD + .249*MGLY + .035*MVKN	2.70×10^{-12}exp(350./T)	
MVKO2 + HO2 → .38*MVKOOH + .62*OH + .37*GLYALD + .37*CH3CO3+ .13*MEK + .25*HO2 + .12*CH2O + .12*MGLY	1.82×10^{-13}exp(1300./T)	
MVKO2 + CH3O2 → .14*HO2 + .14*CH2O + .36*CH3CO3 + .36*GLYALD + .25*ROH + .5*HO2 + .14*MGLY + .25*MEK + .75*CH2O + .25*CH3OH	8.37×10^{-14}	
MVKO2 + CH3CO3 → .4*HO2 + .4*CH2O + .6*CH3CO3 + .6*GLYALD + .4*MGLY + CH3O2	1.68×10^{-12}exp(500./T)	
MVKO2 + CH3CO3 → MEK + CH3COOH	1.87×10^{-13}exp(500./T)	

MVKOOH + OH → .791*OH + .791*MEK + .209*MVKO2	8.78×10^{-12}exp(200./T)	
MVKN + OH → .65*HCOOH + NO3 + .65*MGLY + .35*CH2O	1.60×10^{-12}	
MACR + OH → .45*MAO3 + .55*MACRO2	8.00×10^{-12}exp(380./T)	
MACR + O3 → .261*OH + .202*HO2 + .326*HCOOH + .569*CO + .88*MGLY + 0.12*CH2O	1.40×10^{-15}exp(-2100./T)	
MACR + NO3 → MAO3 + HNO3	3.40×10^{-15}	
MACRO2 + HO2 → 0.42*MACROOH + 0.58*OH + 0.58*HYAC + 0.58*CO+ 0.58*HO2	1.82×10^{-13}exp(1300./T)	
MACRO2 + NO3 → NO2 + HYAC + CO + HO2	2.30×10^{-12}	
MACRO2 + NO → .97*NO2 + 0.97*HO2 + 0.97*CO + 0.97*HYAC + .03*MACRN	2.70×10^{-12}exp(360./T)	
MACRO2 → CO + HYAC + OH	0.5	
MACRO2 + CH3O2 → .595*HYAC + .255*MGLY + .595*CO + 1.255*CH2O + 1.7*HO2 + .15*ROH	8.37×10^{-14}	
MACRO2 + CH3CO3 → .85*HO2 + .143*MGLY + .857*HYAC + .857*CO + .143*CH2O + CH3O2	1.68×10^{-12}exp(500./T)	
MACRO2 + CH3CO3 → MEK + CH3COOH	1.87×10^{-13}exp(500./T)	
MACROOH + OH → MACRO2	1.84×10^{-12}exp(200./T)	
MACROOH + OH → HYAC + OH	4.40×10^{-12}exp(380./T)	
MACRN + OH → MACRNO2	3.20×10^{-12}	
MACRNO2 + NO → .08*CH3COOH + .08*CH2O + .07*MGLY + .85*HYAC+ 1.85*NO2 + 0.15*NO3 + .07*HCOOH	2.70×10^{-12}exp(350./T)	
MACRNO2 + HO2 → .08*CH3COOH + .08*CH2O + .15*NO3 + .07*HCOOH + .07*MGLY + .85*HYAC + .85*NO2 + OH	1.82×10^{-13}exp(1300./T)	
MAO3 + NO → NO2 + CH2O + .65*CH3O2 + 0.65*CO + .35*CH3CO3	8.70×10^{-12}exp(290./T)	
MAO3 + HO2 → .44*OH + .15*O3 + .44*CH2O + .29*CH3O2 + .41*MAOP + 0.15*CH3CO3 + 0.15*MACO2H + 0.29*CO	5.20×10^{-13}exp(980./T)	
MAO3 + NO3 → NO2 + 0.35*CH3CO3 + CH2O + 0.65*CH3O2 + 0.65*CO	4.00×10^{-12}	
MAO3 + CH3O2 → CH2O + HO2 + CH2O + CH3CO3	1.68×10^{-12}exp(500./T)	
MAO3 + CH3O2 → RCOOH + CH2O	1.87×10^{-13}exp(500./T)	
MAO3 + CH3CO3 → CH3O2 + CH2O + CH3CO3	2.50×10^{-12}exp(500./T)	
MAO3 + NO2 + M → MPAN + M	ko=9.00E-28*(300/T)$^{8.90}$; ki=7.70E-12*(300/T) $^{0.20}$; f=0.60; usr53	
MPAN → MAO3 + NO2	1.111×10^{28}exp(-14000./T)*usr53	
MPAN + OH → HYAC + CO + NO2	2.90×10^{-11}	

MPAN + O3 → NO2 + .6*CH2O + HO2	8.20×10^{-18}	
MACO2H + OH → 0.35*CH3CO3 + 0.65*CH3O2 + CH2O + 0.65*CO	1.51×10^{-11}	
MAOP + OH → MAO3	6.13×10^{-13}exp(200./T)	
MAOP + OH → MAOPO2	3.60×10^{-12}exp(380./T)	
MAOPO2 + HO2 → HYAC + 2*OH	1.82×10^{-13}exp(1300./T)	
MAOPO2 + NO → HYAC + OH + NO2	2.35×10^{-12}exp(350./T)	
MAOPO2 + MAOPO2 → 2*HYAC + 2*OH	8.37×10^{-14}	
MAOPO2 + CH3O2 → .7*HYAC + .7*OH + CH2O + .7*HO2 + .3*C2H5OH	8.37×10^{-14}	
MAOPO2 + CH3CO3 → HYAC + OH + CH3O2	1.68×10^{-12}exp(500./T)	
MAOPO2 + CH3CO3 → CH3COOH + MEK	1.87×10^{-13}exp(500./T)	
GLYALD + OH → 0.2*GLYX + HO2 + 0.8*CH2O + 0.8*CO	1.00×10^{-11}	MCM v3.3.1
GLYX + OH → HO2 + 2*CO	3.10×10^{-12}exp(340./T)	
GLYX + NO3 → HNO3 + HO2 + 2*CO	$k_1=1.4\times10^{-12}$exp(-1860./T); k_1*(M*0.21+3.5×10^{18}) /(M*0.42+3.5×10^{18})	
MGLY + OH → CH3CO3 + CO	1.50×10^{-11}	
MGLY + NO3 → HNO3 + CO + CH3CO3	1.40×10^{-12}exp(-1860./T)	
HYAC + OH → MGLY + HO2	frac=1.-23.7exp(-T/70.); 2.15×10^{-12}exp(305./T)*frac	
HYAC + OH → .5*HCOOH + OH + .5*CH3COOH + .5*CO + .5*CH3O2	2.15×10^{-12}exp(305./T)*(1-frac)	
Isoprene Nighttime Chemistry		
ISOP + NO3 → INO2	3.15×10^{-12}exp(-450./T)	
INO2 + NO → ISN1 + NO2 + HO2	2.70×10^{-12}exp(360./T)	MCM v3.2
INO2 + NO3 → ISN1 + NO2 + HO2	2.30×10^{-12}	MCM v3.2
INO2 + HO2 → 0.22*MVK + 0.015*MACR + 0.235*OH + 0.235*NO2 + 0.235*CH2O + 0.77*INPN	2.05×10^{-13}exp(1300./T)	Schwantes et al.(2015)
INO2 + INO2 → 2.0*ISN1 + 1.2*HO2	1.30×10^{-12}	
INPN + OH → ISN1 + OH	1.03×10^{-10}	
ISN1 + OH → 0.52*C510O2 + 0.48*ISNOO	4.16×10^{-11}	
ISN1 + NO3 → ISNOO + HNO3	5.95×10^{-12}exp(-1860./T)	
ISN1 + O3 → 0.555*NOA + 0.52*GLYX + 0.445*MGLY + 0.075*H2O2 + 0.445*HO2 + 0.89*CO + 0.89*OH + 0.445*NO2	2.40×10^{-17}	
ISNOO + HO2 → 0.15*NC4CO2H + 0.15*O3 + 0.41*NC4CO3H + 0.44*NOA + 0.44*CO + 0.44*HO2 + 0.44*OH	5.20×10^{-13}exp(980./T)	
ISNOO + NO → NOA + CO + HO2 + NO2	7.50×10^{-12}exp(290./T)	
ISNOO + NO2 → C5PAN1	$k_o=2.7\times10^{-28}$*M*$(300./T)^{7.1}$; $k_1=1.2\times10^{-11}$*$(300./T)^{0.9}$; fc=k_o/k_1; fcc=0.75-1.27*$\log_{10}$ 0.3;	

	$nfc=10^{\frac{\log_{10} 0.3}{(1+(\log_{10} fc/fcc)^2)}}$; $k_o k_1/(k_o+k_1)$*nfc	
ISNOO + NO3 → NOA + CO + HO2 + NO2	4.00×10^{-12}	
ISNOO + ISNOO → 0.6*NC4CO2H + 1.4*NOA + 1.4*CO + 1.4*HO2	1.00×10^{-11}	
C510O2 + HO2 → C510OOH	2.05×10^{-13}exp(1300./T)	
C510O2 + NO → NO2 + NOA + GLYX + HO2	2.70×10^{-12}exp(360./T)	
C510O2 + NO3 → NO2 + NOA + GLYX + HO2	2.30×10^{-12}	
C510O2 + C510O2 → 0.6*C510OH + 1.4*NOA + 1.4*GLYX + 1.4*HO2	9.20×10^{-14}	
C510OH + OH → NOA + GLYX + HO2	2.69×10^{-11}	
C510OOH + OH → C510O2	2.81×10^{-11}	
NC4CO2H + OH → NOA + HO2 + CO	2.16×10^{-11}	
NC4CO3H + OH → ISNOO	2.52×10^{-11}	
NOA + OH → MGLY + NO2	1.00×10^{-12}	
C5PAN1 → ISNOO + NO2	$k_o=4.9\times10^{-3}$*M*exp(-12100./T); $k_1=5.4\times10^{16}$exp(-13830./T); $fc=k_o/k_1$; fcc=0.75-1.27*$\log_{10}$ 0.3; $nfc=10^{\frac{\log_{10} 0.3}{(1+(\log_{10} fc/fcc)^2)}}$; $k_o k_1/(k_o+k_1)$*nfc	
C5PAN1 + OH → NOA + CO + CO + NO2	2.16×10^{-11}	
Isoprene Ozonlysis		
ISOP + O3 → .325*MACR + .244*MVK + .845*CH2O + .11*H2O2 + .27*OH + .128*C3H6 + .051*CH3O2 + .522*CO + .204*HCOOH + .199*CH3CO3 + .026*HO2	1.00×10^{-14}exp(-1970./T)	
Photolysis of major organic nitrates		
ISOPNB + hν → HC5 + NO2 + HO2	1.21×10^{-6} s^{-1}	Calculated based on 24hr average of modeled results
MACRN + hν → NO2 + HYAC + HO2 + CO	8.48×10^{-5} s^{-1}	
MVKN + hν → GLYALD + NO2 + CH3CO3	1.36×10^{-5} s^{-1}	
NOA + hν → CH3CO3 + CH2O + NO2	8.48×10^{-6} s^{-1}	
ISN1 + hν → NOA + 2.0*CO + 2.0*HO2	1.36×10^{-4} s^{-1}	
TERPN1 + hν → NO2	1.21×10^{-6} s^{-1}	
TERPN2 + hν → NO2	1.21×10^{-6} s^{-1}	

Table S2. Monoterpene oxidation chemistry in AM3. T represents temperature (K).

Reactions	Reaction Rates (molecule^{-1}cm^{3}s^{-1})	Note
$C10H16 + OH \rightarrow TERPO2$	$1.2\times10^{-11}\exp(440/T)$	Atkinson and Arey (2003)
$TERPO2 + NO \rightarrow 0.74*NO2 + 0.26*TERPN1$	$2.7\times10^{-12}\exp(360/T)$	Saunders et al. (2003)
$TERPO2 + HO2 \rightarrow$ products	$2.9\times10^{-13}\exp(1300/T)$	Saunders et al. (2003)
$TERPO2 + TERPO2 \rightarrow$ products	$1.68\times10^{-12}\exp(500/T)$	Tyndall et al. (2001)
$C10H16 + O3 \rightarrow$ products	$5.30\times10^{-16}\exp(-530/T)$	RCAM2
$C10H16 + NO3 \rightarrow 0.1*TERPN2 + 0.9*NO2$	$1.2\times10^{-12}\exp(490/T)$	Atkinson and Arey (2003)

Table S3. Hydrolysis reactions added in AM3.

Reactions	Reaction Rate or Uptake Coefficient	Note
GLYX → AGLYX	$\gamma_{glyx} = 1.0\times10^{-3}$	
MGLY → AMGLY	$\gamma_{mgly} = 1.0\times10^{-7}$	
IEPOX → AIEPOX	$\gamma_{iepox} = 1.0\times10^{-3}$	
ISOPNB → AONJ	$\gamma_{IN} = 5.0\times10^{-3}$	Fisher et al.(2016)
AONJ → HNO3 + ROH	$k = 9.259\times10^{-5}\ s^{-1}$	Pye et al.(2015)

Table S4. Lifetimes of major nitrates from isoprene and monoterpene in AM3.

Species in AM3	Formation Pathways	Lifetime[a]
ISOPNB[b]	C_5 β-hydroxy nitrate from isoprene oxidation by OH	1.2 (1.4)
ISN1	C_5 carbonyl nitrate from isoprene oxidation by NO_3	3.6 (3.5)
INPN	C_5 nitrooxy hydroperoxide from isoprene oxidation by NO_3	8.5 (7.8)
MVKN	Methyl vinyl ketone nitrate from ISOPNB oxidation by OH	9.6 (9.3)
MACRN	Methacrolein nitrate from ISOPNB oxidation by OH	1.5 (1.4)
ISNP	C_5 nitrooxy hydroperoxide from isoprene oxidation by OH	12 (12)
PROPNN	Propanone nitrate from oxidation of INPN and ISN1	17.6 (16.2)
DHDN[c]	C_5 dihydroxy dinitrate from ISOPNB oxidation by OH	11.4 (10.3)
TERPN1	Nitrate from monoterpene oxidation by OH	19.6 (18.8)
TERPN2	Nitrate from monoterpene oxidation by NO_3	14.1 (14.4)
PAN		0.72 (0.66)
HNO_3		16(16)

[a] Lifetimes (hr) are calculated based on the total chemical loss rate, dry and wet deposition flux of each. compound during July-August of 2004; values in brackets are from July-August of 2013. It should be noted that the lifetimes listed here are 24-h average, different from estimates by Müller et al. (2014).

[b] The lifetime of ISOPNB includes impacts of aerosol hydrolysis.

[c] The lifetime of DHDN is based on dry and wet deposition since it is not removed by chemical oxidation.

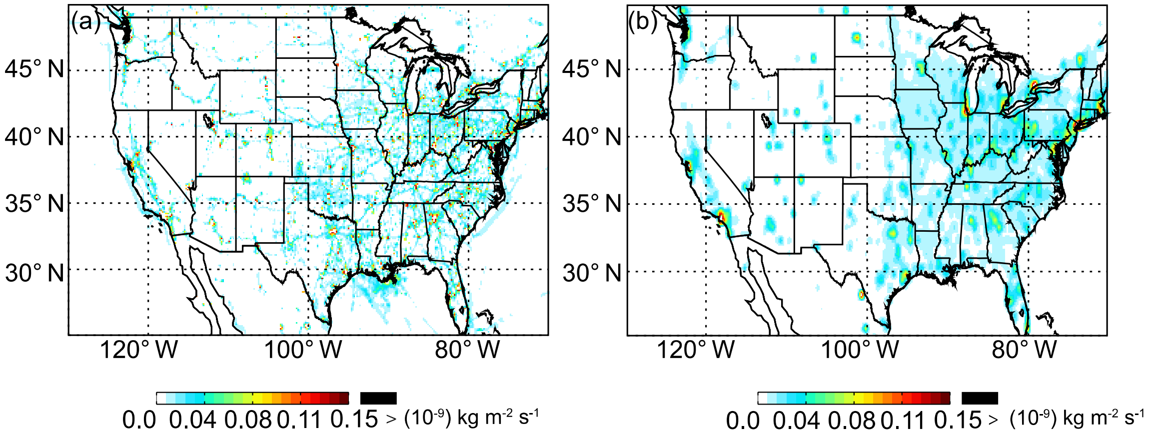

Figure S1. Anthropogenic NO_x emission rate during July-August 2013 of (a) NEI11v1 inventory and (b) RCP8.5.

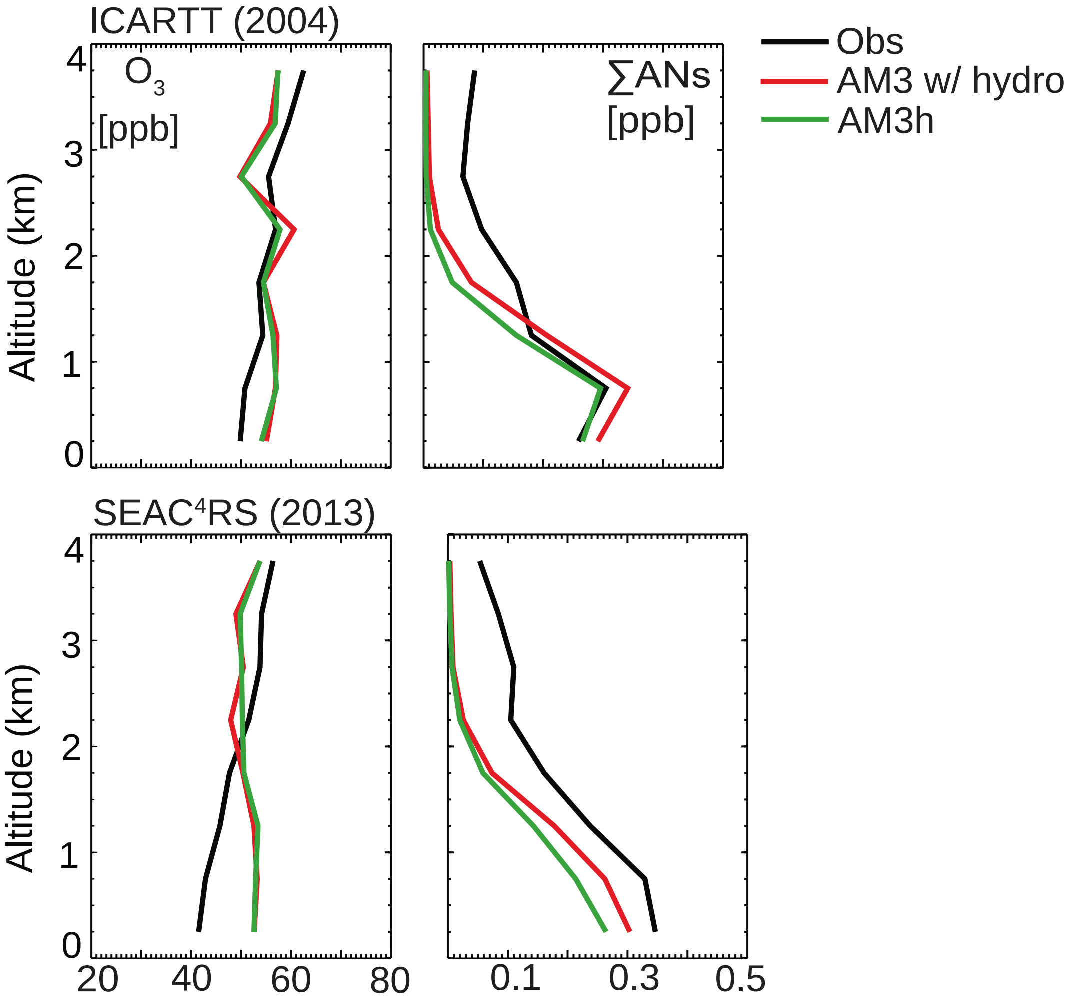

Figure S2. Mean vertical profiles of ozone and ∑ANs during ICARTT (top) and SEAC^{4}RS (bottom). Red lines are results of AM3 with ISOPNB hydrolysis only; green lines are from AM3 with hydrolysis of ISOPNB, DHDN and TERPN1.

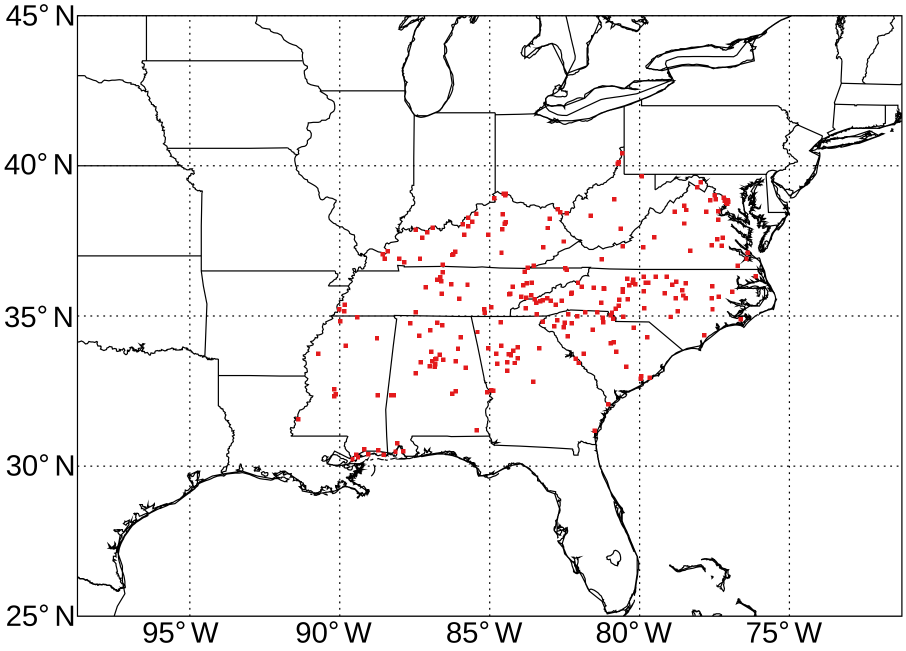

Figure S3. EPA AQS ozone monitoring sites in the Southeast U.S.A., from which EPA provides the MDA8 metric used in our study.

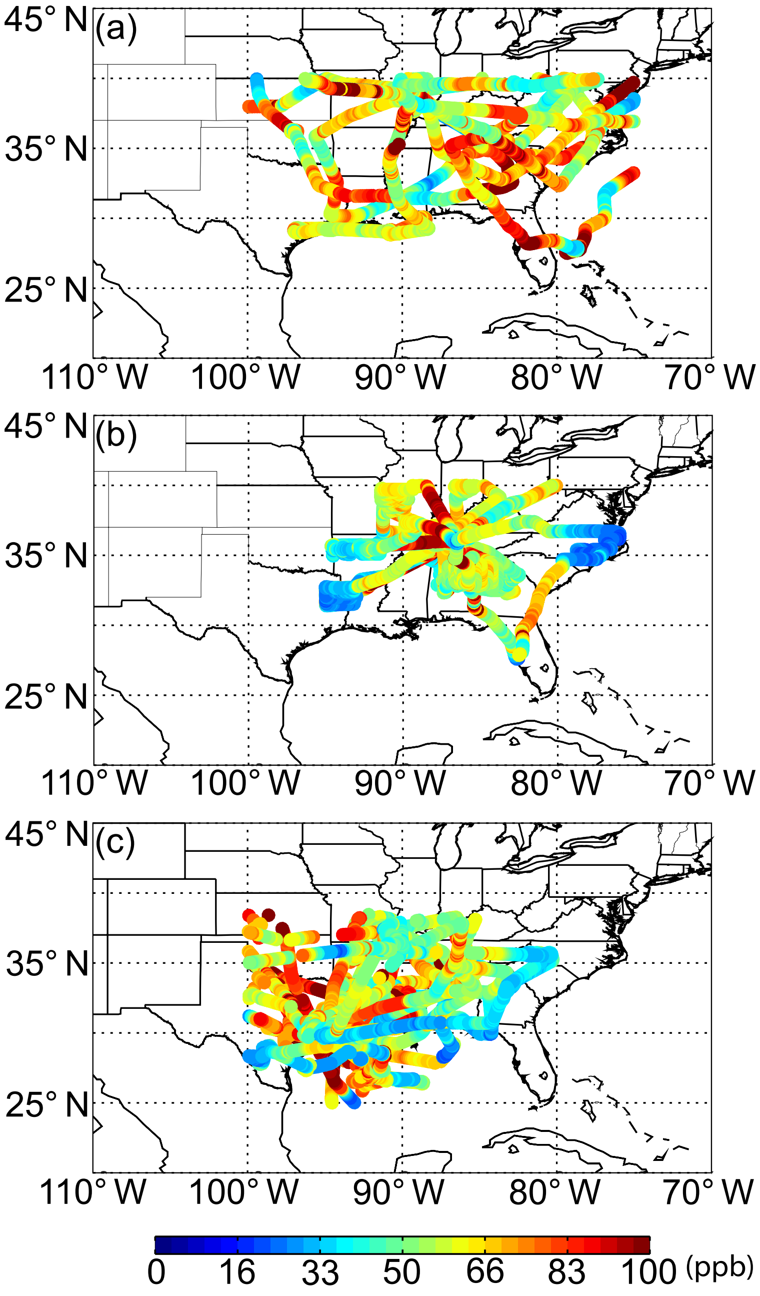

Figure S4. Ozone (ppb) along daytime flight tracks during (a) ICARTT (up to 12 km) (b) SENEX (up to 6 km) and (c) SEAC^{4}RS (up to 12.5 km). Data from biomass burning, urban plumes, stratospheric air and outside of the 25-40° N latitude and 100-75° W longitude region are excluded.

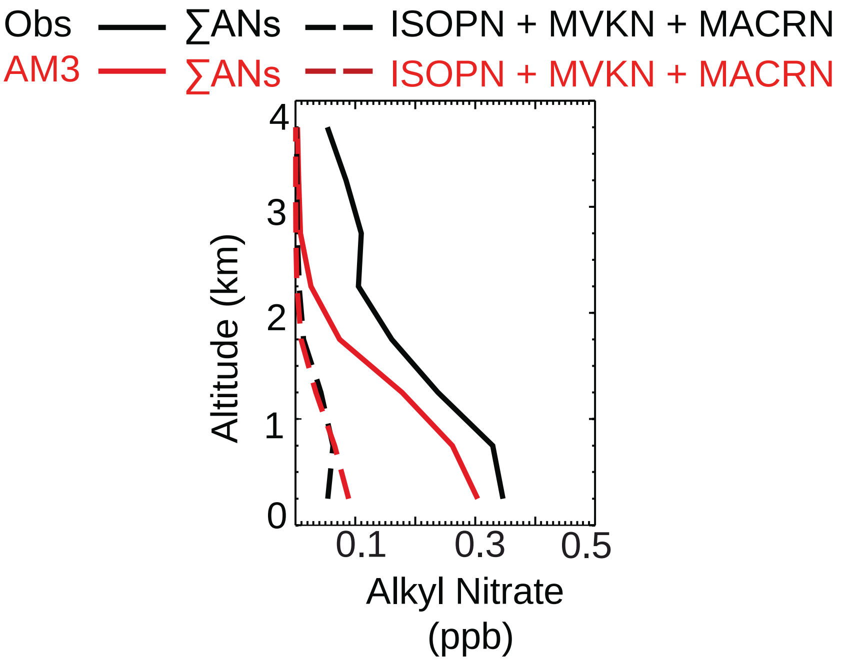

Figure S5. Mean vertical profiles of ∑ANs (solid lines) and sum of ISOPN, MVKN and MACRN (dashed lines) during SEAC^{4}RS from observations (black) and AM3 with hydrolysis of ISOPNB (red).

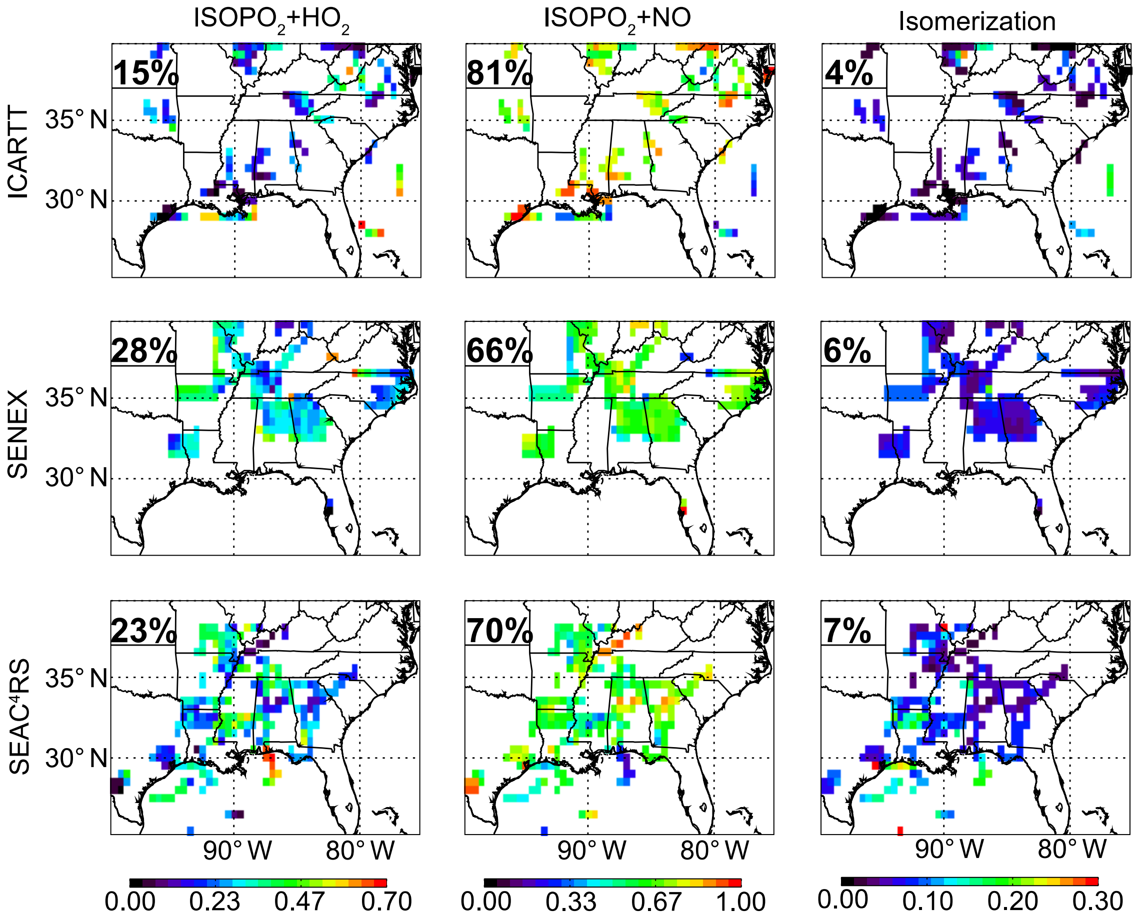

Figure S6. Ratio of major ISOPO$_2$ loss pathways to the total ISOPO$_2$ loss (sum of the three individual pathways) during ICARTT (July-August, 2004, top), SENEX (June-July, 2013, middle) and SEAC^{4}RS (July-September, 2013, bottom) in the boundary layer (below 1.5km). Bold values in each panel are the mean percentage of each pathway to the total ISOPO$_2$ loss.

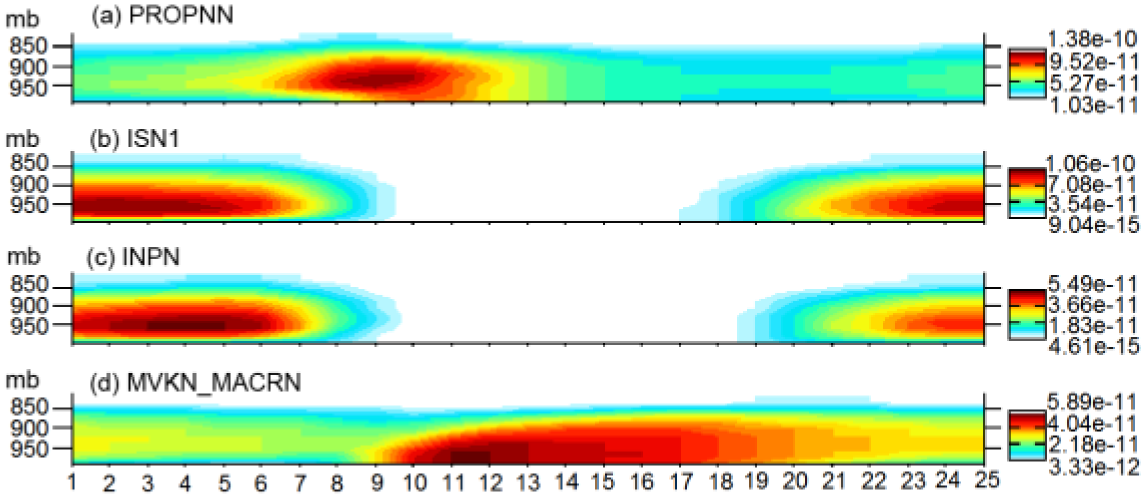

Figure S7. Modeled diurnal variation of PROPNN, ISN1, INPN and MVKN + MACRN (MVKN_MACRN) in the boundary layer of the Southeast US. The x-axis is the local time, and y-axis is the pressure. Color bar indicates the concentration in volume mixing ratio.

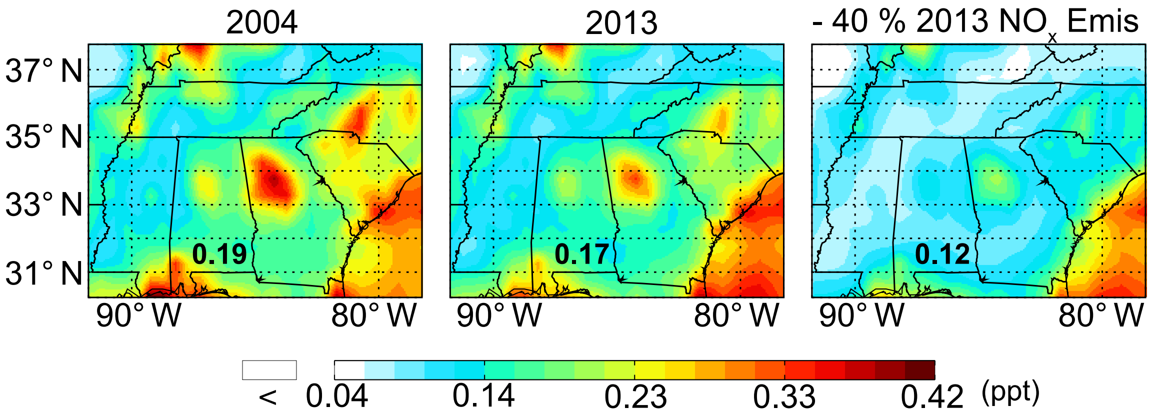

Figure S8. Averaged OH from 10:00 to 14:00 during July-August of 2004, 2013 in the model, and the scenario with 40% reduction in the anthropogenic NO$_x$ emissions of 2013. Bold number in each panel is the regionally-averaged OH concentration.

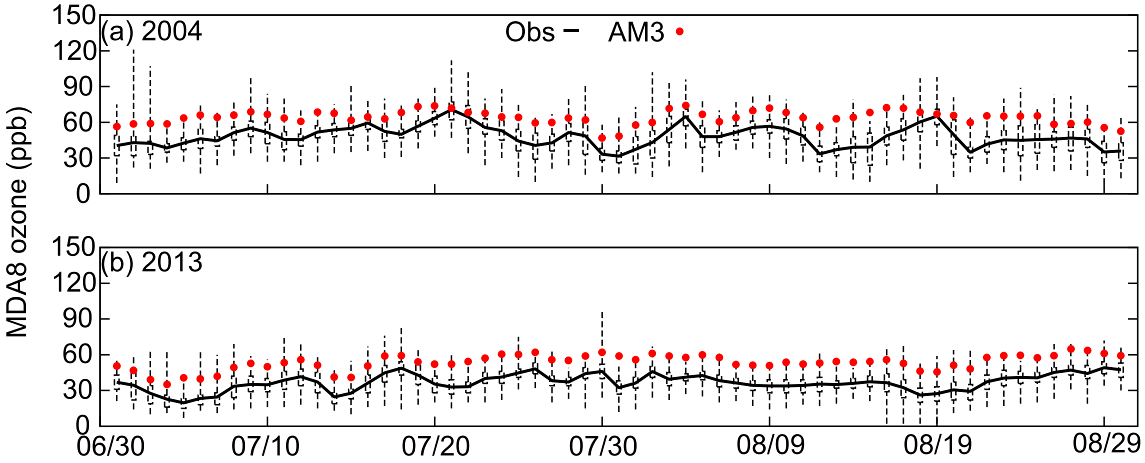

Figure S9. Daily MDA8 ozone during July-August of 2004 (a) and July-August of 2013 (b) averaged over 157 EPA AQS monitoring sites shown in Figure S1. Horizontal axis is month and day of the year. Black lines, dash boxes and whiskers are the mean, interquartile range and lowest and highest observations; red dots are the mean of AM3.

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