Global surface ozone trends, a synthesis of recently published findings

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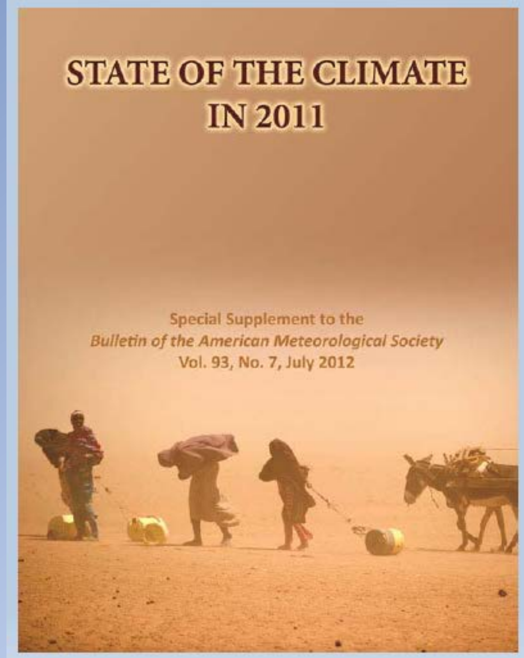
The results in this presentation were inspired by, and in partial fulfillment of the goals of:

First International Workshop on Tropospheric Ozone Changes
Boulder, Colorado, USA, October 2009

Second International Workshop on Tropospheric Ozone Changes
Toulouse, France, 11-14 April 2011

The results also support the new section on ozone as a short-lived greenhouse gas in the upcoming State of the Climate in 2012 Report



This presentation summarizes ozone trends reported by:

Ding, A. J., et al., 2008: Tropospheric ozone climatology over Beijing: analysis of aircraft data from the MOZAIC program. *Atmos. Chem. Phys.*, **8**, 1–13.

Logan, J.A. et al. (2012), Changes in Ozone over Europe since 1990: analysis of ozone measurements from sondes, regular Aircraft (MOZAIC), and alpine surface sites, *J. Geophys. Res., 117(D09301*)

Cooper, O.R., et al (2012), Long-term ozone trends at rural ozone monitoring sites across the United States, 1990-2010, *J. Geophys. Res., 117(D22307).*

Helmig, D., S. J. Oltmans, D. Carlson, J.-F. Lamarque, A. Jones, C. Labuschagne, K. Anlauf, and K. Hayden, 2007: A review of surface ozone in the polar regions. *Atmospheric Environment*, **41**, 5138–5161.

Hess, P.G. and Zbinden, R., 2013. Stratospheric impact on tropospheric ozone variability and trends: 1990–2009. Atmos. Chem. Phys., 13(2): 649-674.

Lee, H.-J. et al. (2013), Transport of NOX in East Asia identified by satellite and in-situ measurements and Lagrangian particle dispersion model simulations, *J. Geophys. Res*, submitted.

Lelieveld, J., van Aardenne, J., Fischer, H., de Reus, M., Williams, J., Winkler, P., 2004: Increasing ozone over the Atlantic Ocean. *Science*, **304**, 1483–1487.

Li et al., 2010: Meteorologically adjusted long-term trend of ground-level ozone concentrations in Kaohsiung County, southern Taiwan. *Atmos. Environ*., **44**, 3605-3608.

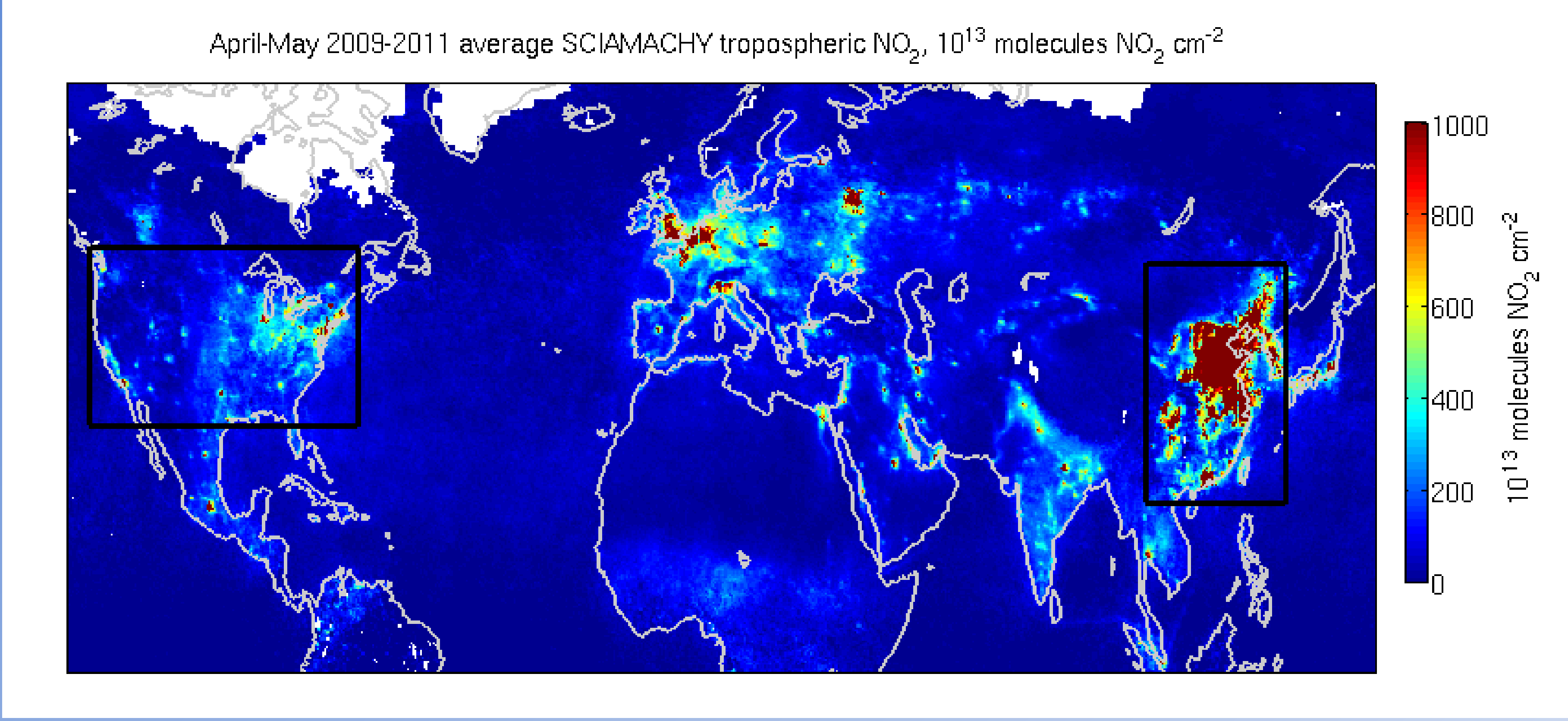
Lin et al., 2010: The changes in different ozone metrics and their implications following precursor reductions over northern Taiwan from 1994 to 2007. *Environ. Monit. Assess*., **169**, 143–157, DOI 10.1007/s10661-009-1158-4

Parrish, D.D., et al. (2012), Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes, *Atmos. Chem. Phys., 12(23): 11485-11504.*

Oltmans, S.J., et al. (2013), Recent tropospheric ozone changes – A pattern dominated by slow or no growth, *Atmos. Environ., 67: 331-351.*

Tarasova O. A., et al., 2009: Surface ozone at the Caucasian site Kislovodsk High Mountain Station and the Swiss Alpine site Jungfraujoch (1990-2006). *Atmos. Chem. Phys*., **9**, 4157-4175.

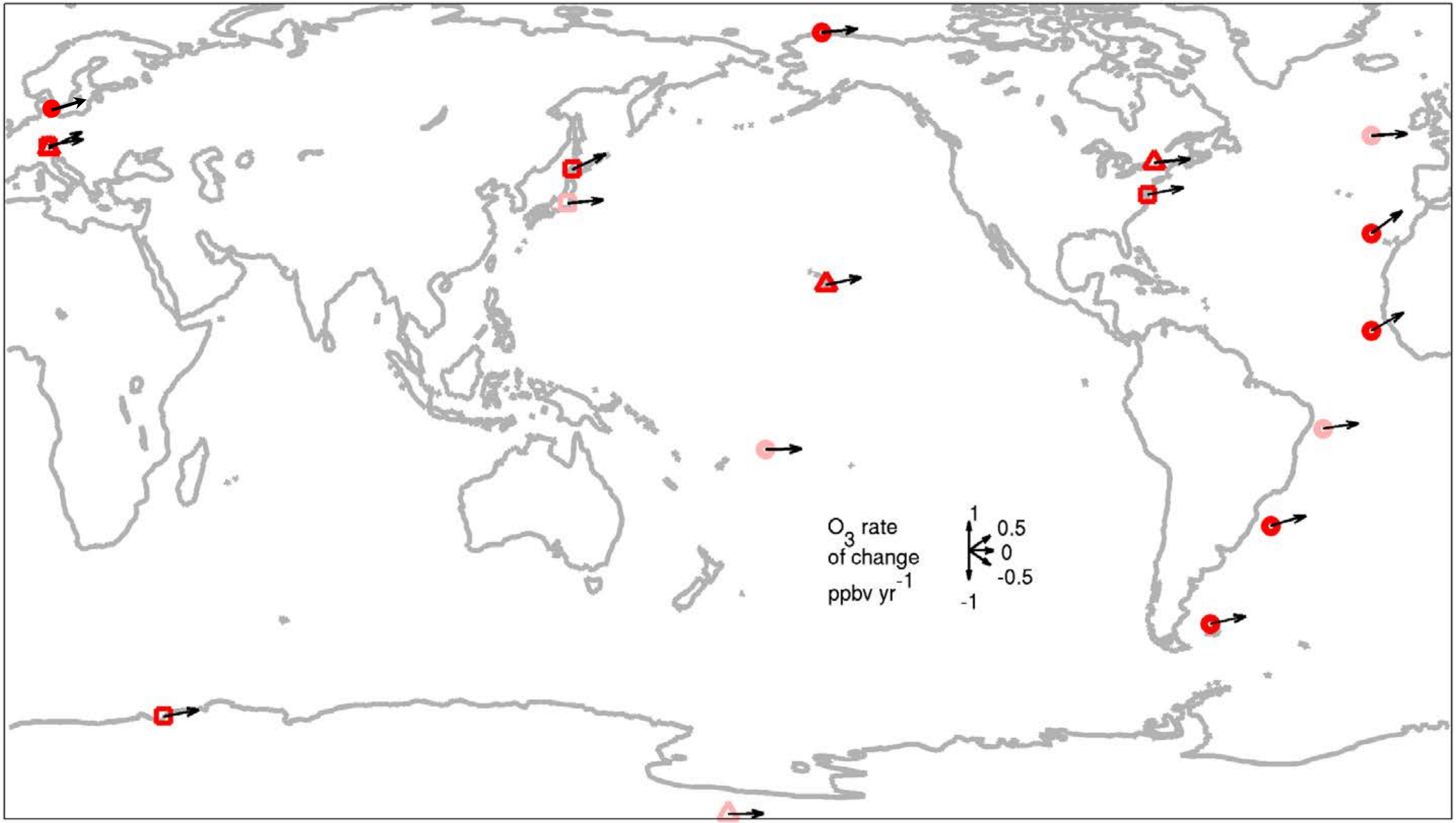
Wang, et al., 2009: Increasing surface ozone concentrations in the background atmosphere of Southern China, 1994-2007. *Atmos. Chem. Phys*., **9**, 6217-6227.



Tropospheric NO_2 column data from the GOME and SCIAMACHY sensors were freely downloaded from: www.temis.nl
For methodology see:

Boersma, K. F., et al. (2004), Error analysis for tropospheric NO2 retrieval from space, J. Geophys. Res., 109, D04311,
Richter, A., et al.(2005), Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437

Annual Surface ozone trends: 1970s through 2002-2010 (from the peer-reviewed literature)



significant increase

insignificant increase

significant decrease

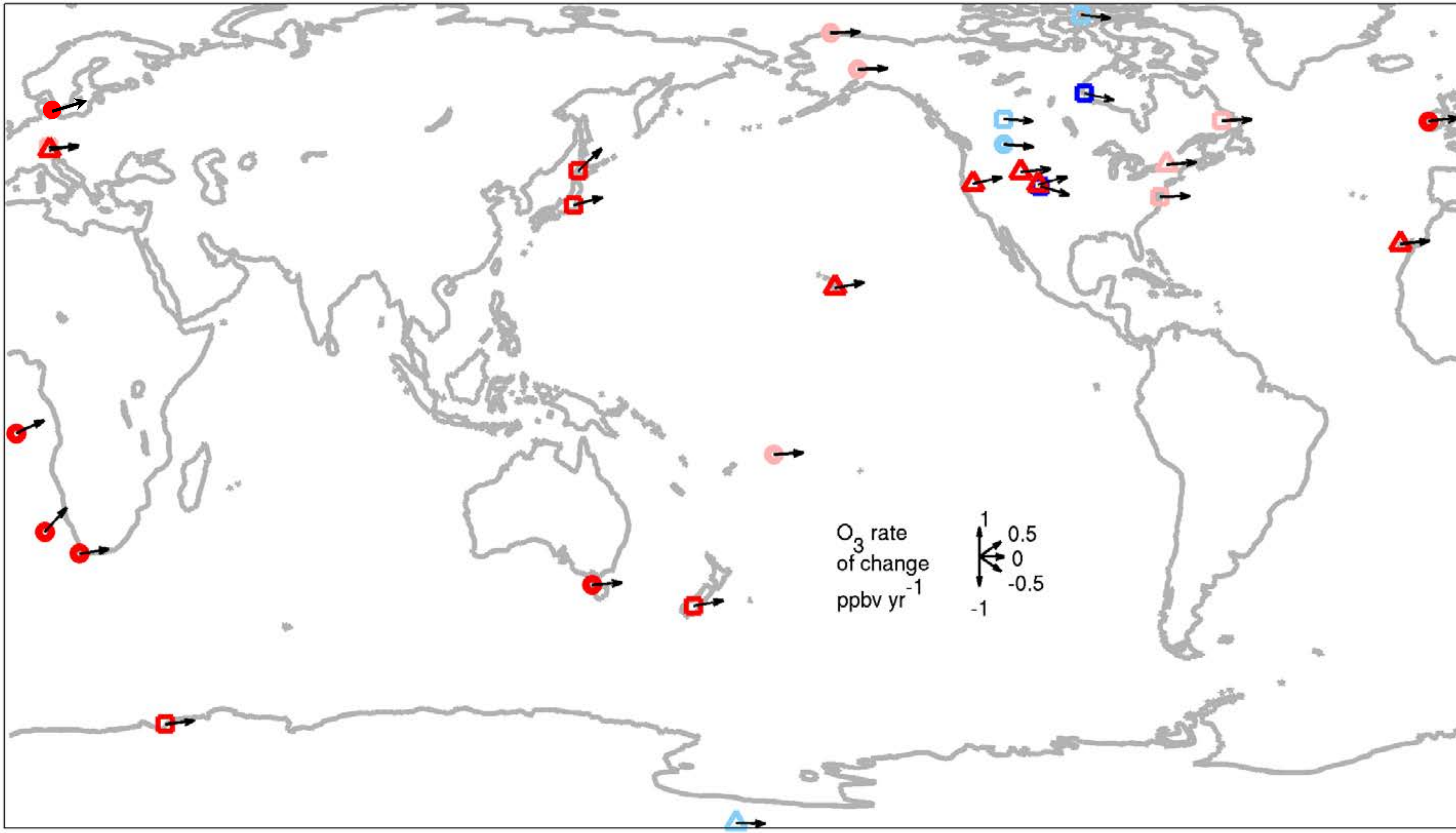
insignificant decrease

□ ozonesonde site

O low elevation site

Δ high elevation site

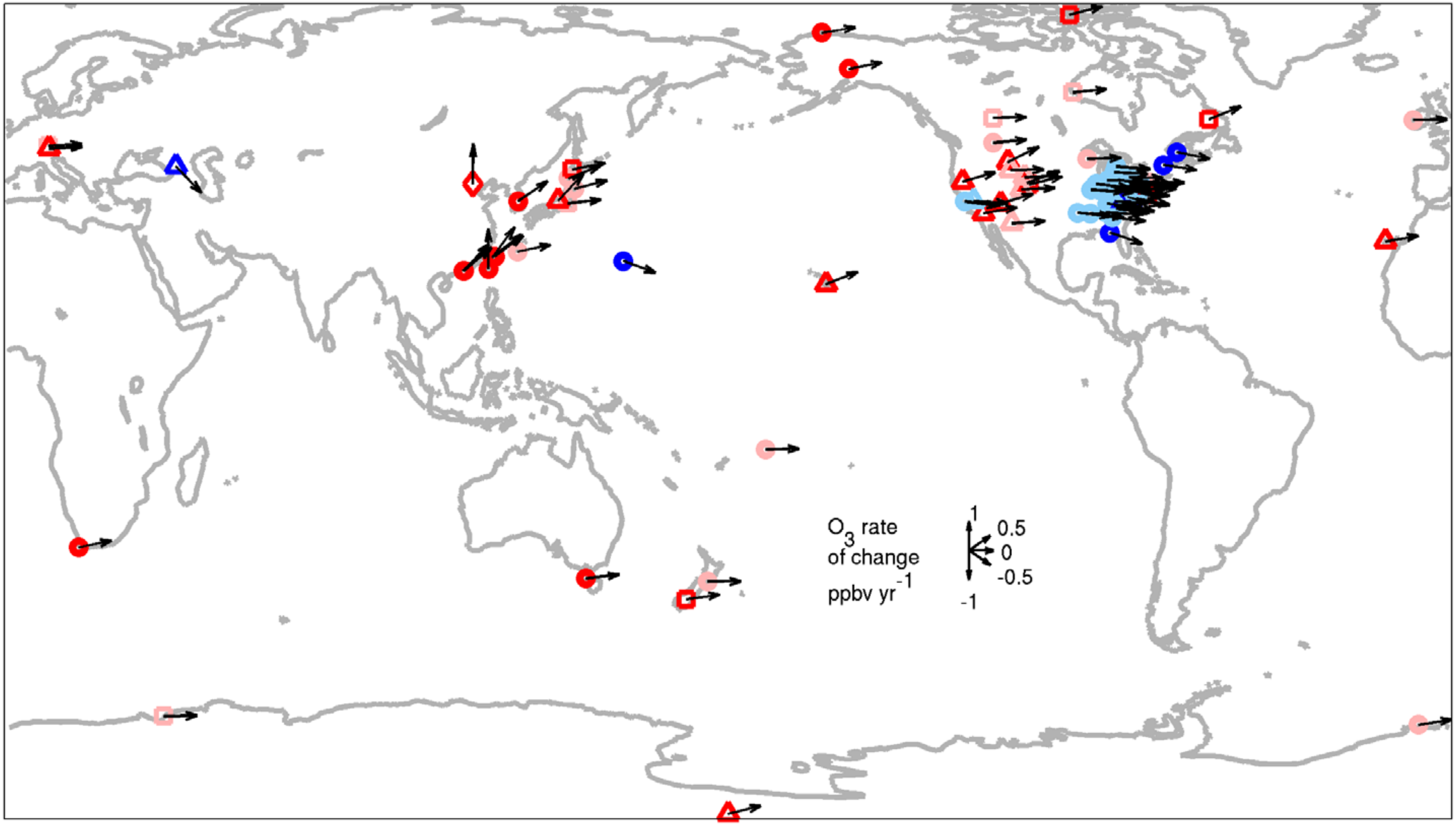
Annual Surface ozone trends: 1980s through 2002-2010 (from the peer-reviewed literature)



significant increase | **insignificant increase** | **significant decrease** | **insignificant decrease**

□ ozonesonde site | O low elevation site | Δ high elevation site

Annual Surface ozone trends: 1990s through 2005-2010 (from the peer-reviewed literature)



significant increase

insignificant increase

significant decrease

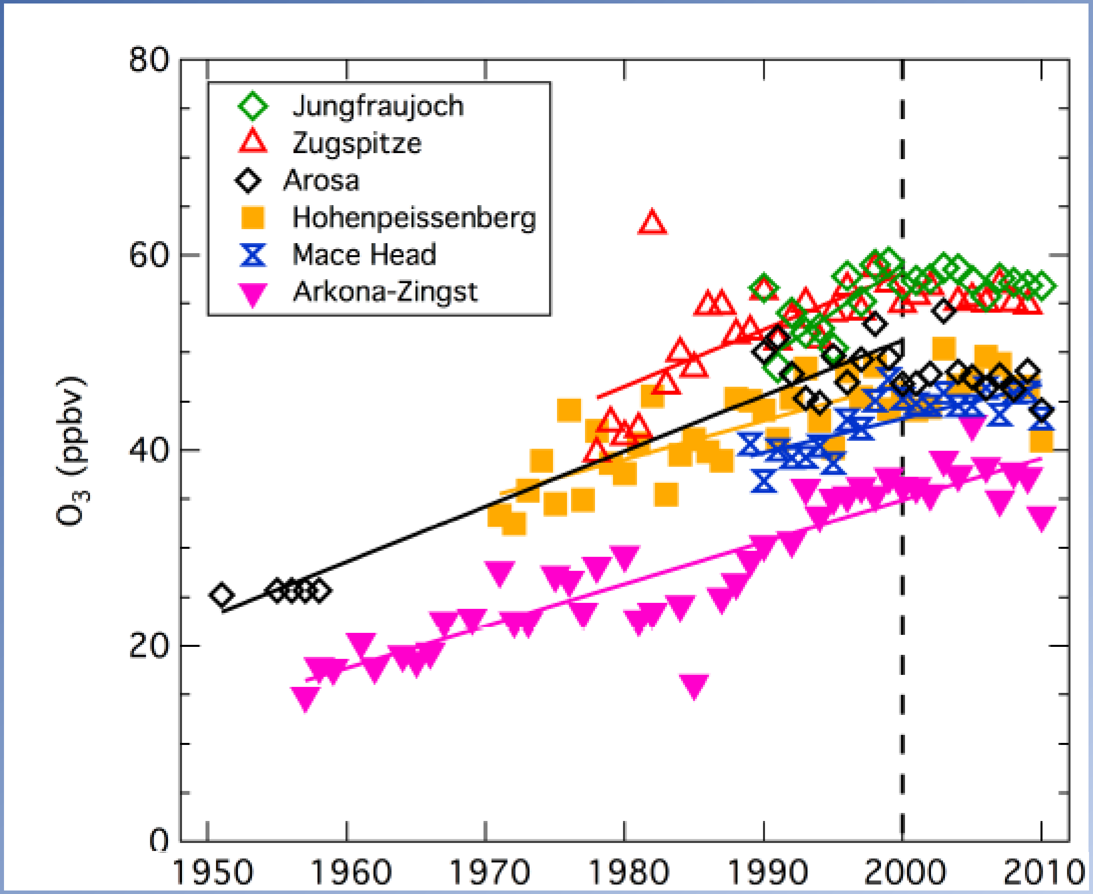
insignificant decrease

□ ozonesonde site

O low elevation site

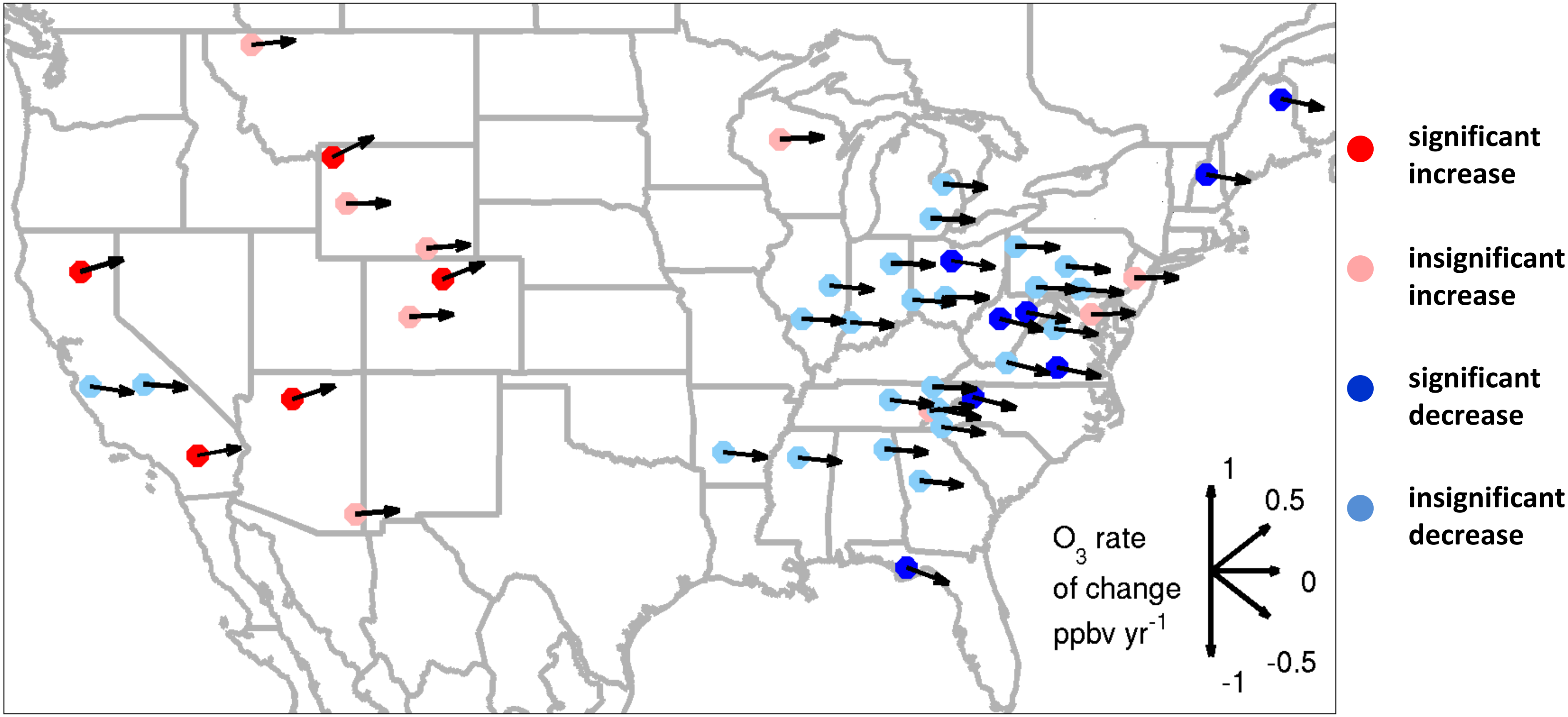
Δ high elevation site

Springtime ozone trends at regionally representative European sites



Parrish, D.D., Law, K.S., Staehelin, J., Derwent, R., Cooper, O.R., Tanimoto, H., Volz-Thomas, A., Gilge, S., Scheel, H.E., Steinbacher, M. and Chan, E. (2012), Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes, *Atmos. Chem. Phys., 12(23): 11485-11504.*

Annual average ozone trends at rural sites, 1990-2010 (data from all 24-hours)

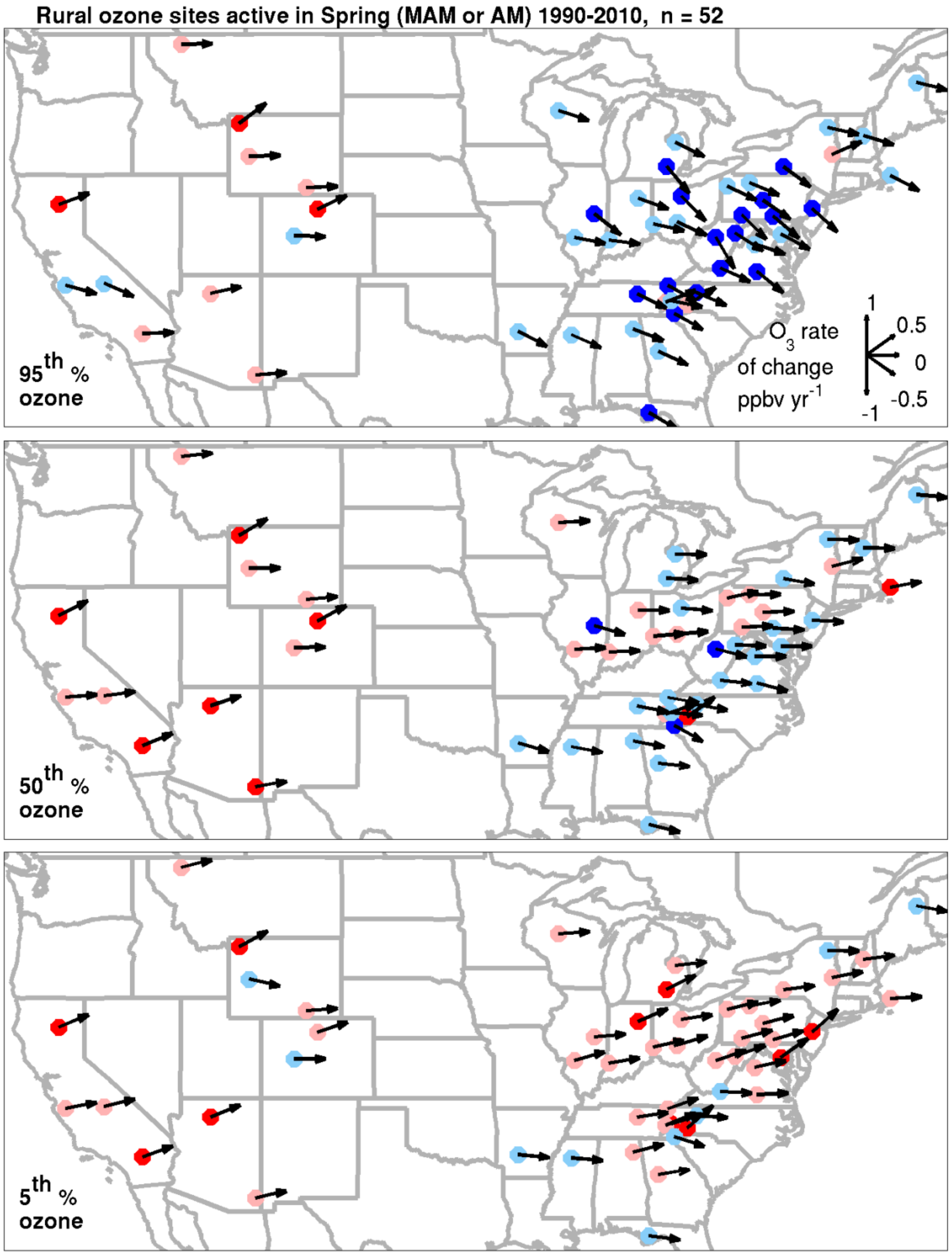


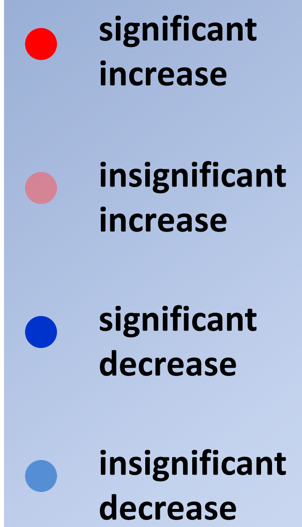
Domestic ozone precursor emissions decreased by 50% during 1990-2010

In the west ozone increased significantly at 42% of rural sites.

In the east ozone decreased significantly at 24% of rural sites.

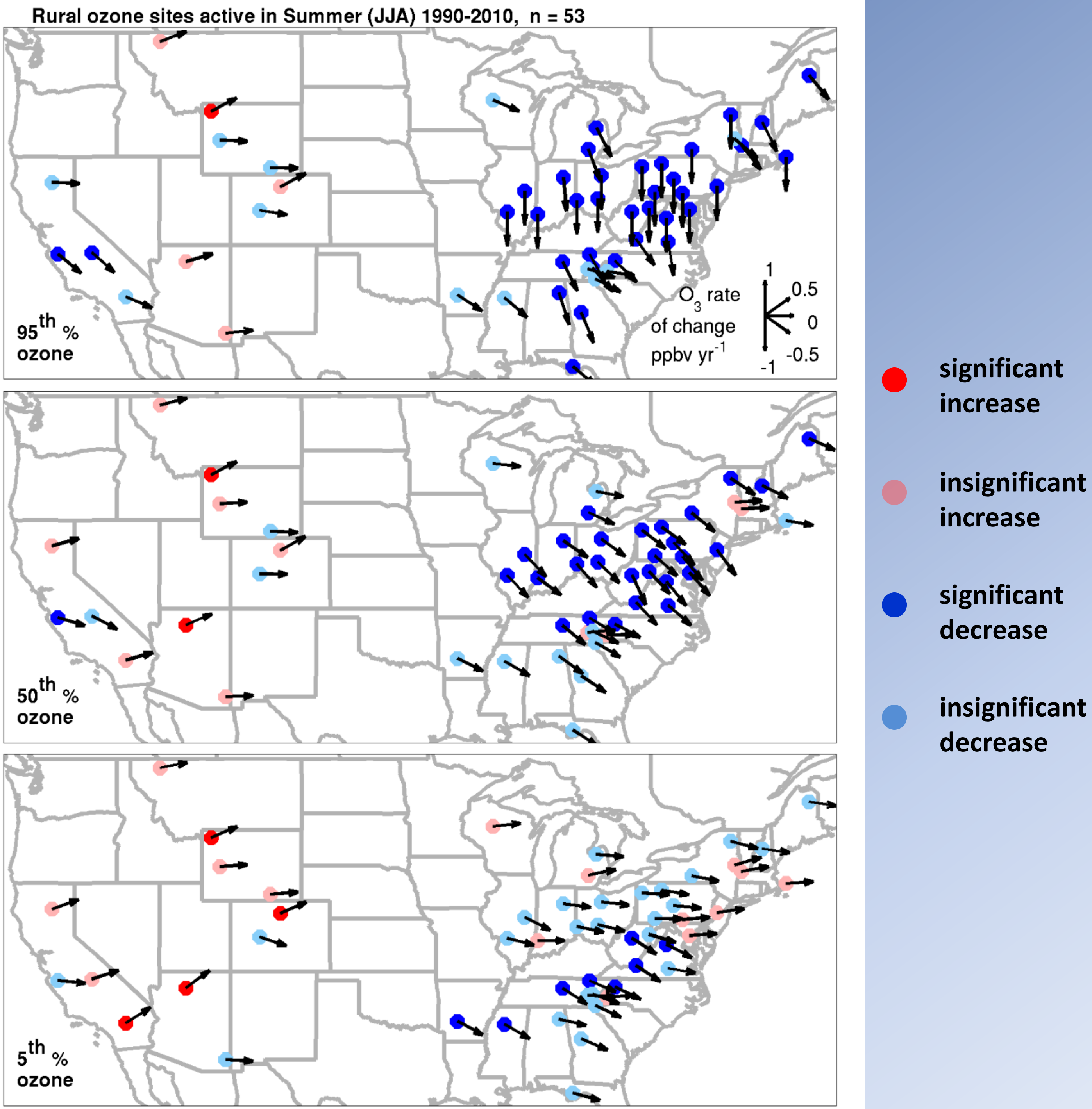
Spring 1990-2010 ozone trends, daytime only





Cooper et al. (2012), Long-term ozone trends at rural ozone monitoring sites across the United States, 1990-2010, *J. Geophys. Res., 117(D22307).*

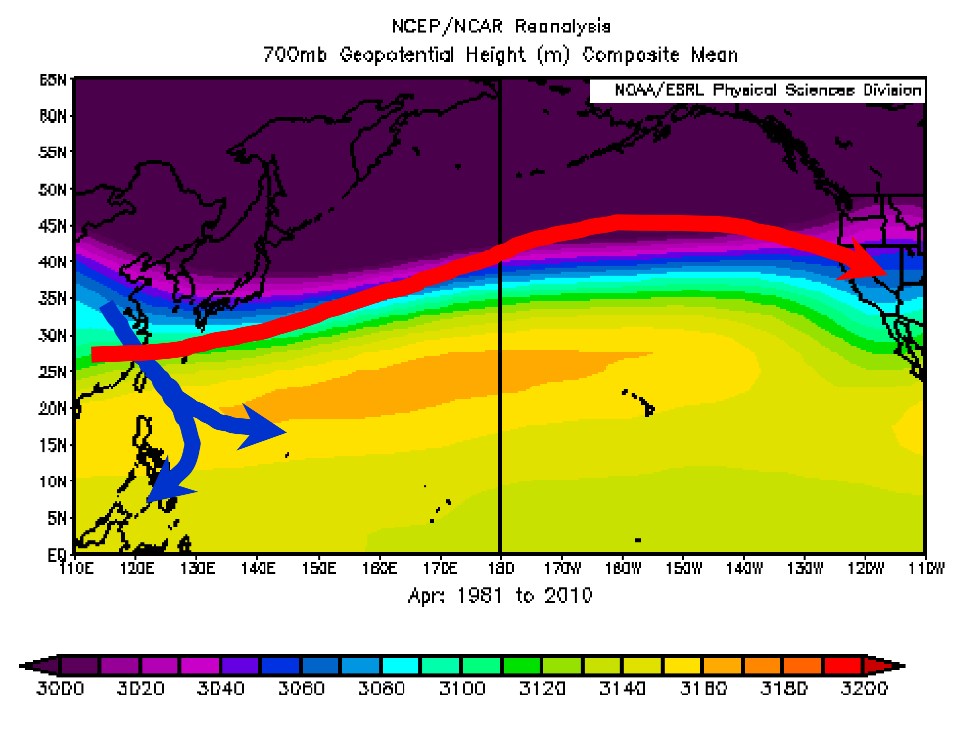
Summer 1990-2010 ozone trends, daytime only



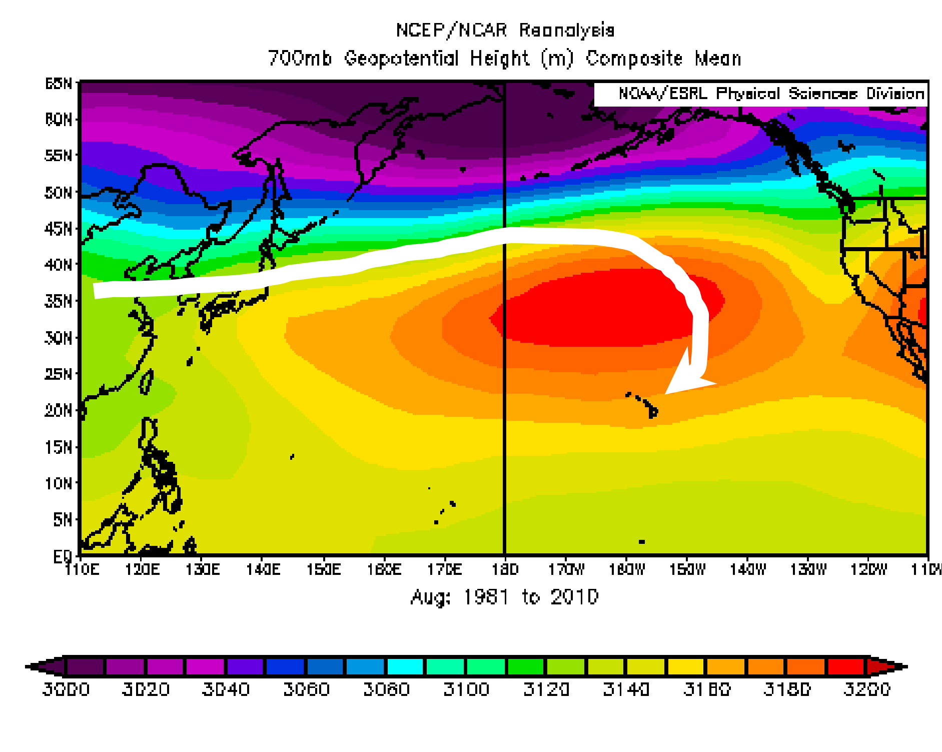
Cooper et al. (2012), Long-term ozone trends at rural ozone monitoring sites across the United States, 1990-2010, *J. Geophys. Res., 117(D22307).*

Transport pathways of Asian outflow in relation to Mauna Loa Observatory (3.4 km above sea level)

April



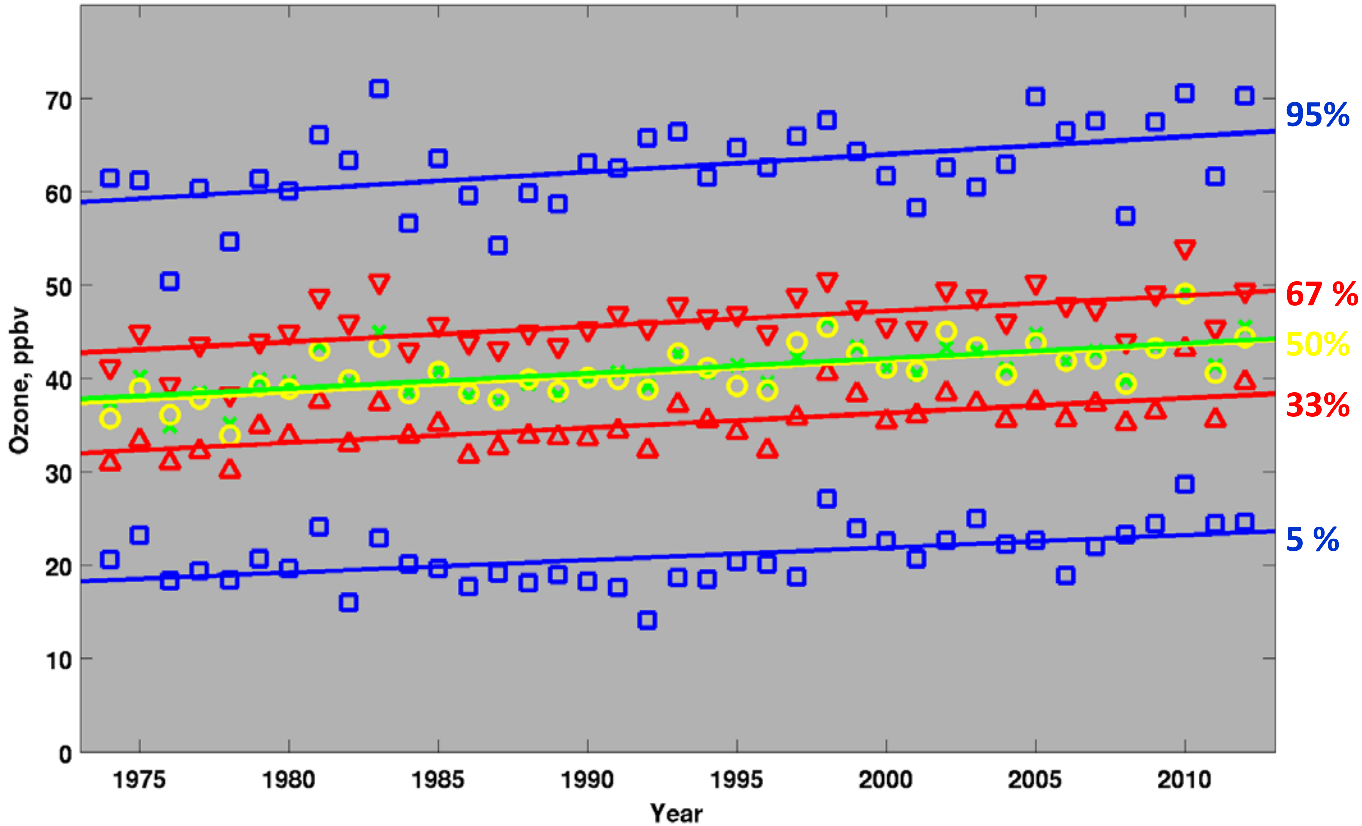
August



Ozone trend at Mauna Loa Observatory, Hawaii, 3.4 km above sea level

Data from years: 1974 - 2012

Data from months: 1 2 3 4 5 6 7 8 9 10 11 12



Green - mean

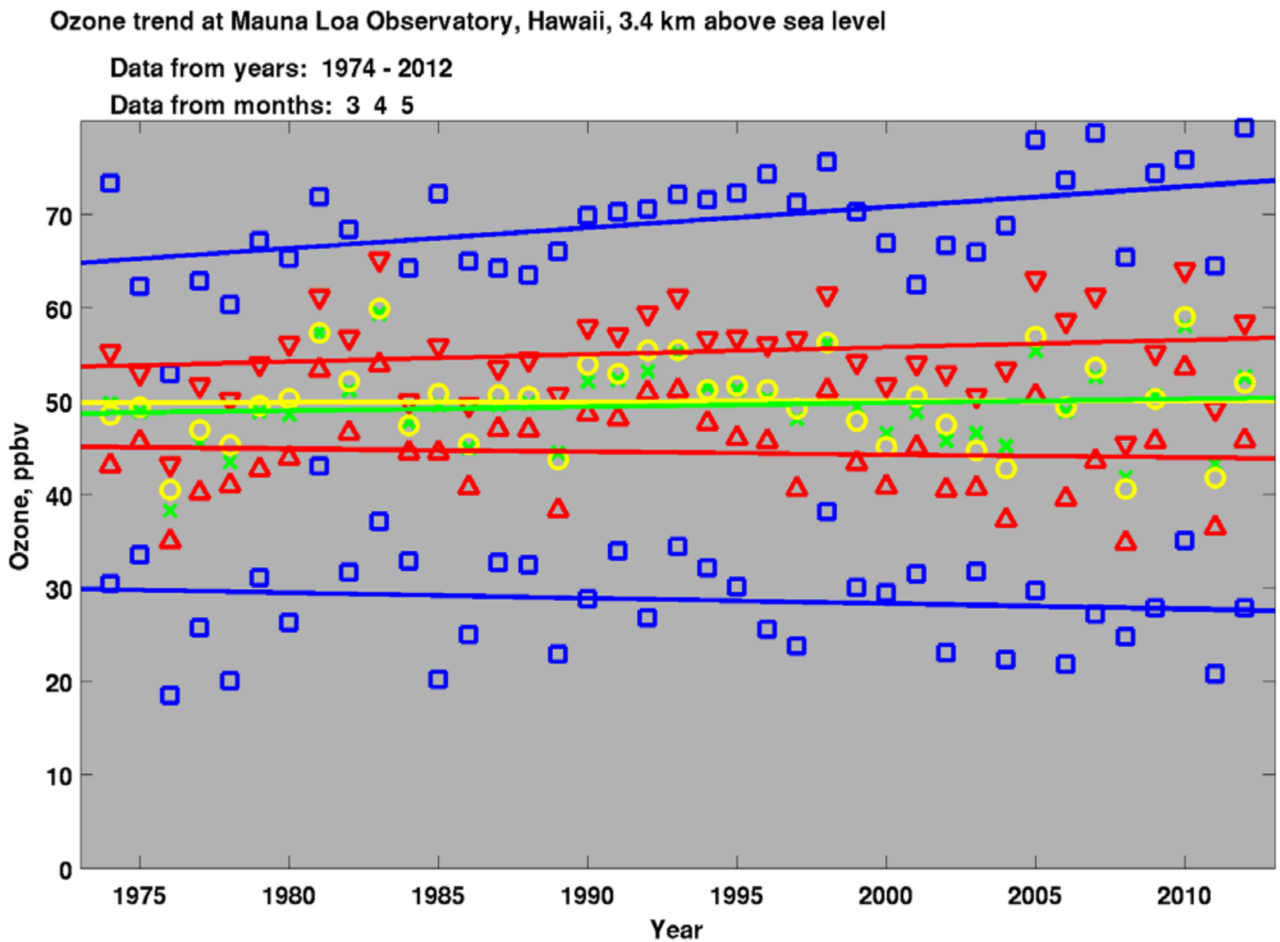
Yellow - median

Blue - 5th & 95th percentiles

Red - 33rd and 67th percentiles

O_3 percentile	increase ppbv yr^{-1}	p value
95^{th} %:	0.19	0.00
67^{th} %:	0.17	0.00
50^{th} %:	0.17	0.00
33^{th} %:	0.16	0.00
05^{th} %:	0.13	0.00

Annual median ozone increased by 17% during 1974-2012.



Green - mean

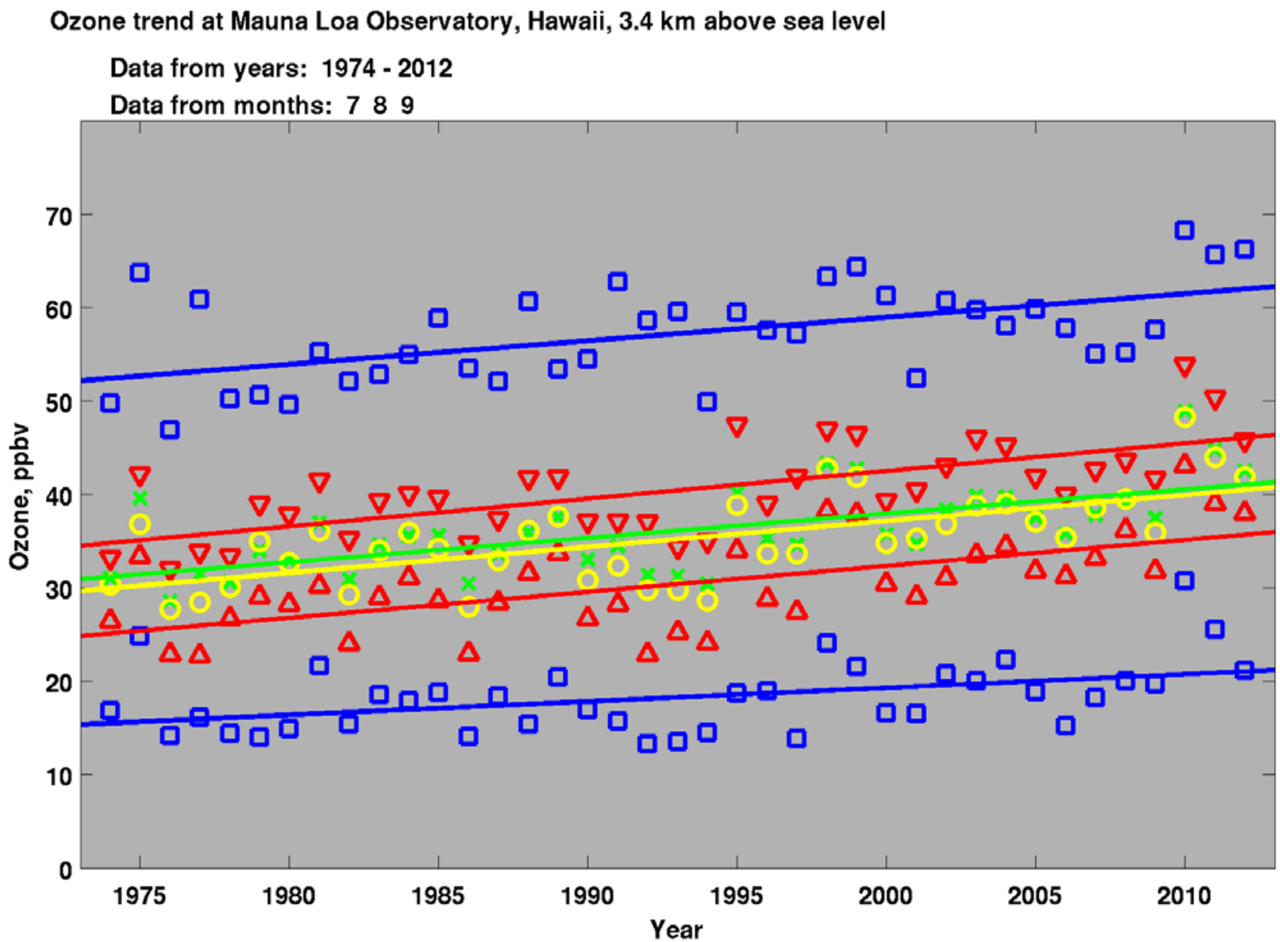
Yellow - median

Blue - 5th & 95th percentiles

Red - 33rd and 67th percentiles

O_3 percentile	increase ppbv yr^{-1}	p value
95^{th} %:	0.22	0.01
67^{th} %:	0.08	0.27
50^{th} %:	0.01	0.93
33^{th} %:	-0.03	0.67
05^{th} %:	-0.06	0.47

Springtime median ozone was unchanged during 1974-2012.



Green - mean

Yellow - median

Blue - 5th & 95th percentiles

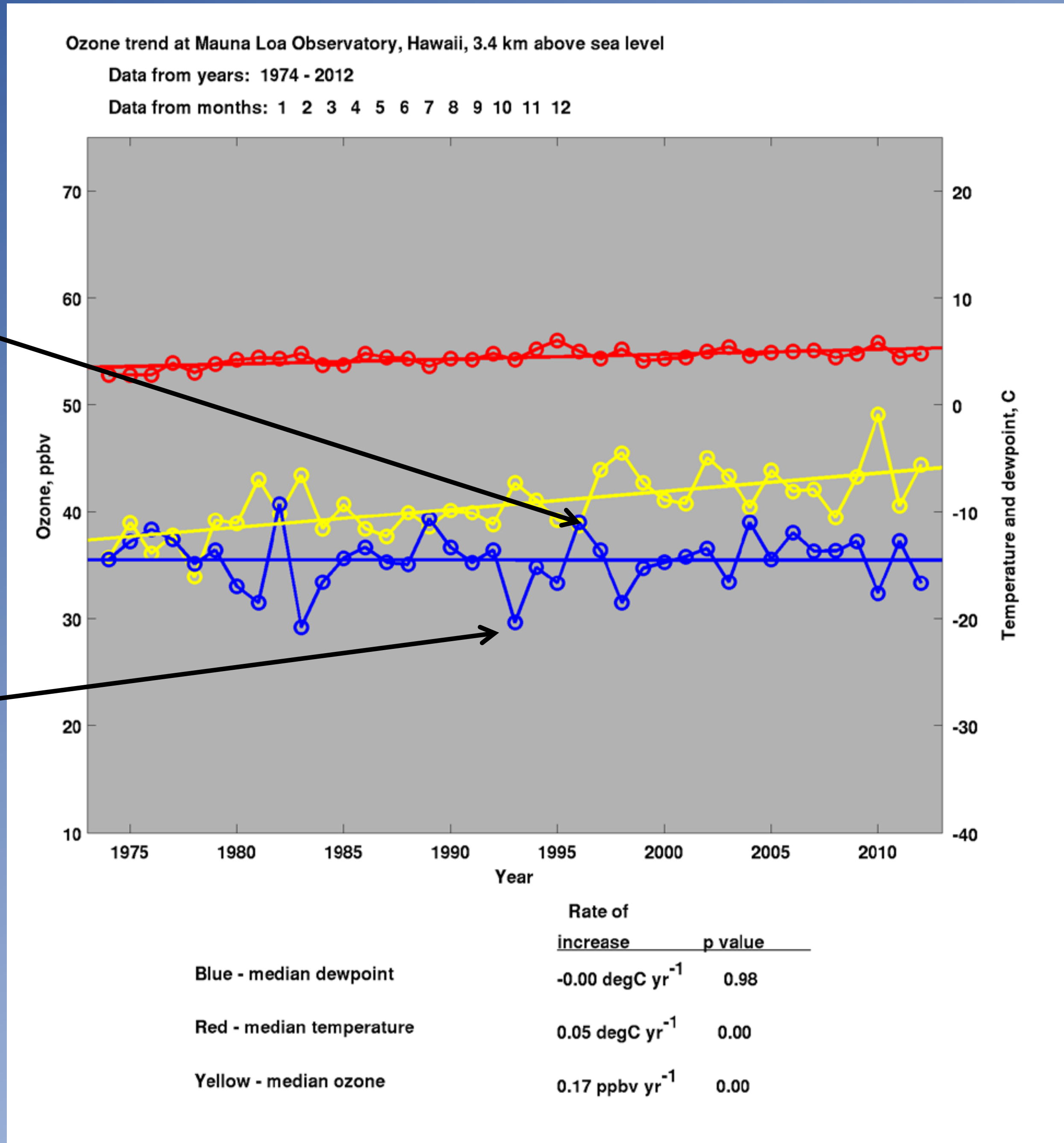
Red - 33rd and 67th percentiles

O_3 percentile	increase ppbv yr^{-1}	p value
95^{th} %:	0.25	0.00
67^{th} %:	0.30	0.00
50^{th} %:	0.28	0.00
33^{th} %:	0.28	0.00
05^{th} %:	0.14	0.01

Late summer median ozone increased by 35% during 1974-2012.

Higher dewpoints:

- Greater transport from the tropics
- Lower ozone



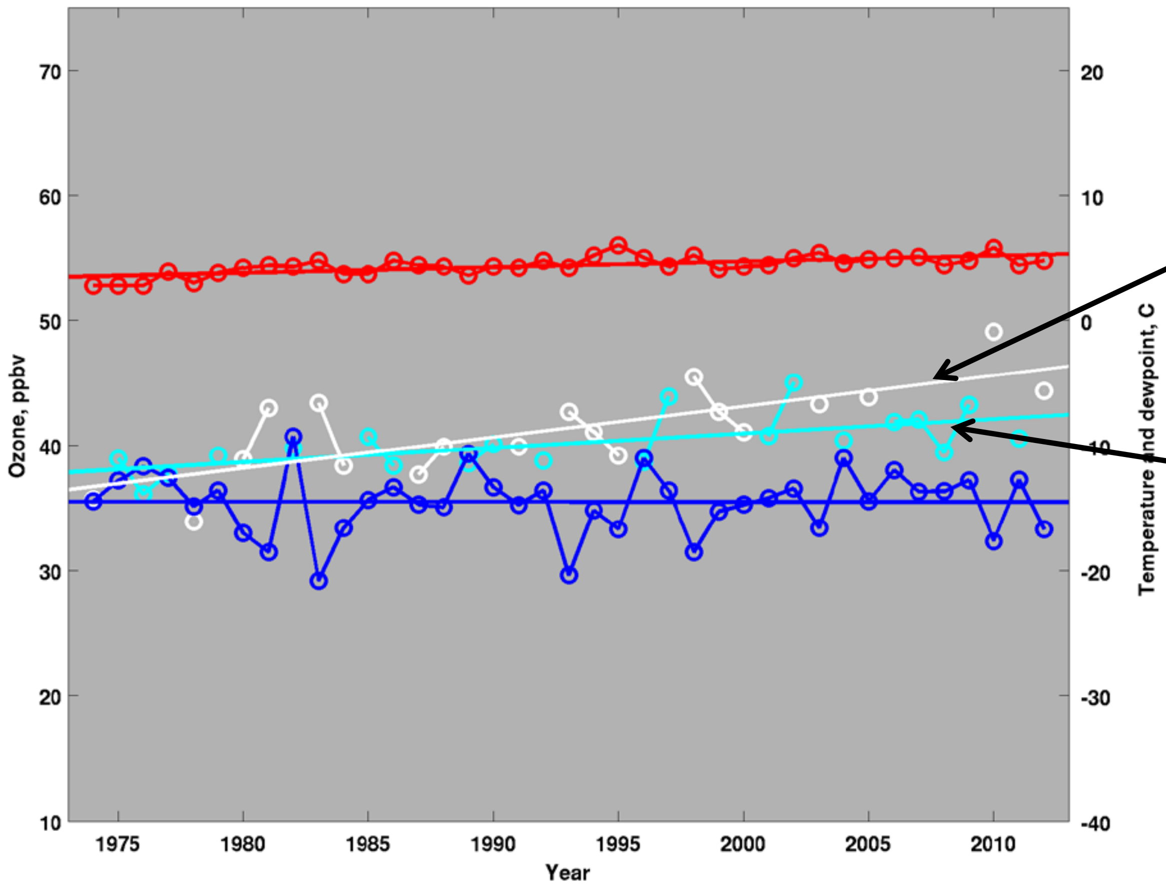
Lower dewpoints:

- Greater transport from mid-latitudes
- Higher ozone

Ozone trend at Mauna Loa Observatory, Hawaii, 3.4 km above sea level

Data from years: 1974 - 2012

Data from months: 1 2 3 4 5 6 7 8 9 10 11 12



Stronger mid-latitude influence

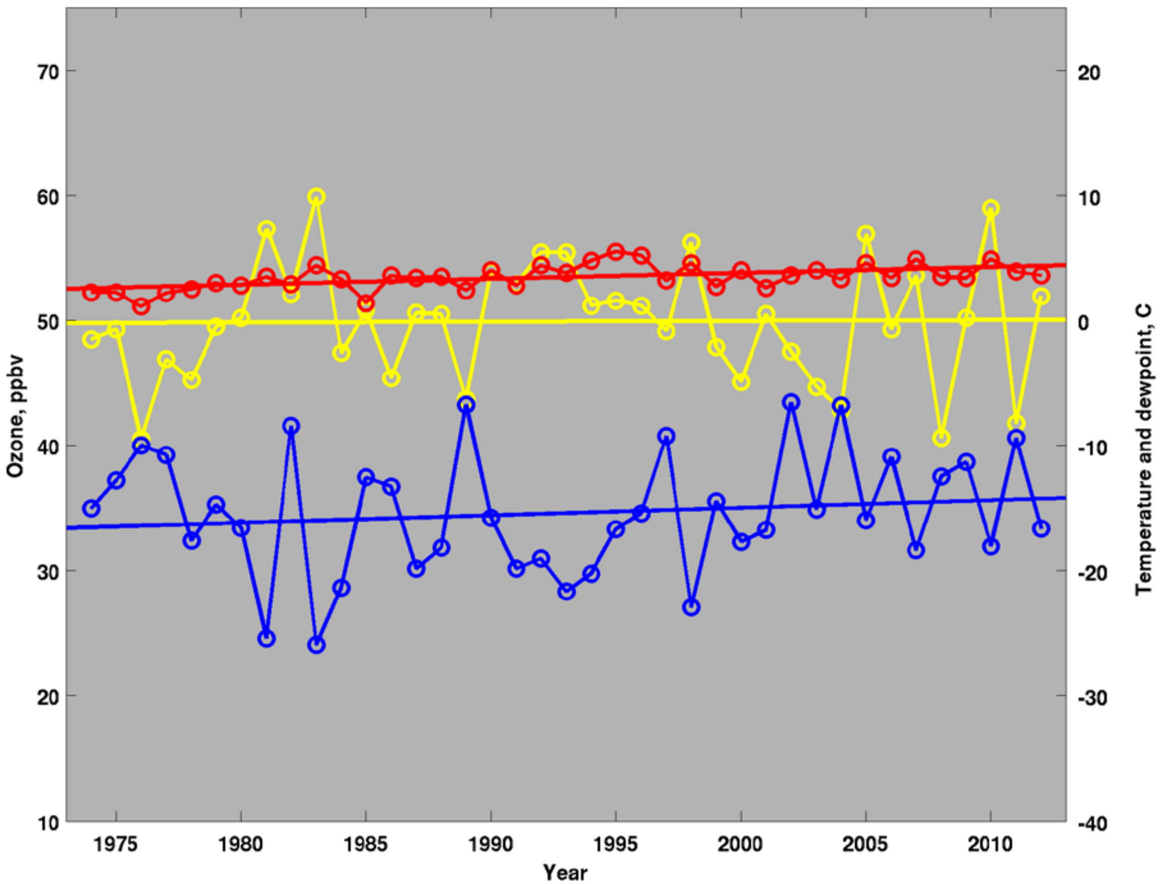
Stronger tropical influence

	Rate of increase	p value
Blue - median dewpoint	-0.00 degC yr^{-1}	0.98
Red - median temperature	0.05 degC yr^{-1}	0.00
White - median ozone, dry	0.25 ppbv yr^{-1}	0.00
Cyan - median ozone, moist	0.11 ppbv yr^{-1}	0.00

Ozone trend at Mauna Loa Observatory, Hawaii, 3.4 km above sea level

Data from years: 1974 - 2012

Data from months: 3 4 5

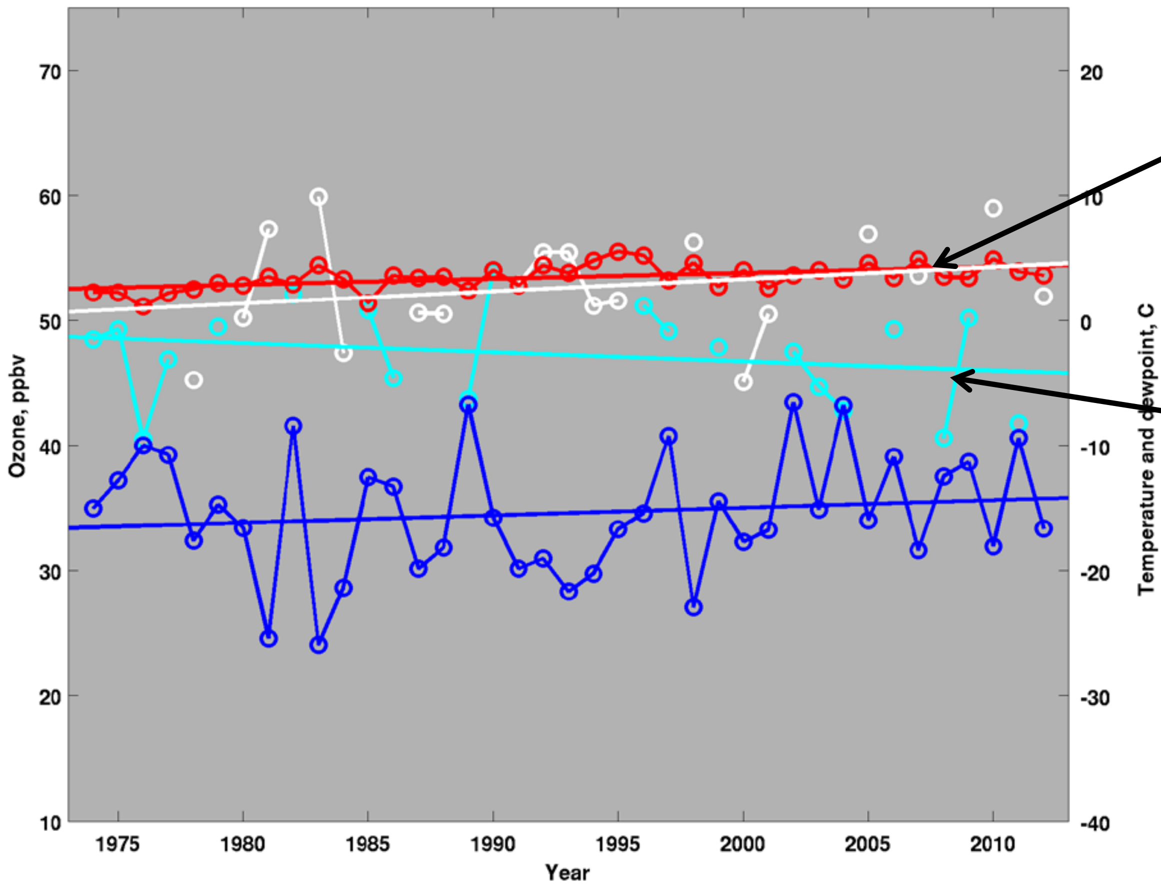


	Rate of increase	p value
Blue - median dewpoint	0.06 degC yr^{-1}	0.40
Red - median temperature	0.05 degC yr^{-1}	0.00
Yellow - median ozone	0.01 ppbv yr^{-1}	0.93

Ozone trend at Mauna Loa Observatory, Hawaii, 3.4 km above sea level

Data from years: 1974 - 2012

Data from months: 3 4 5



Stronger mid-latitude influence

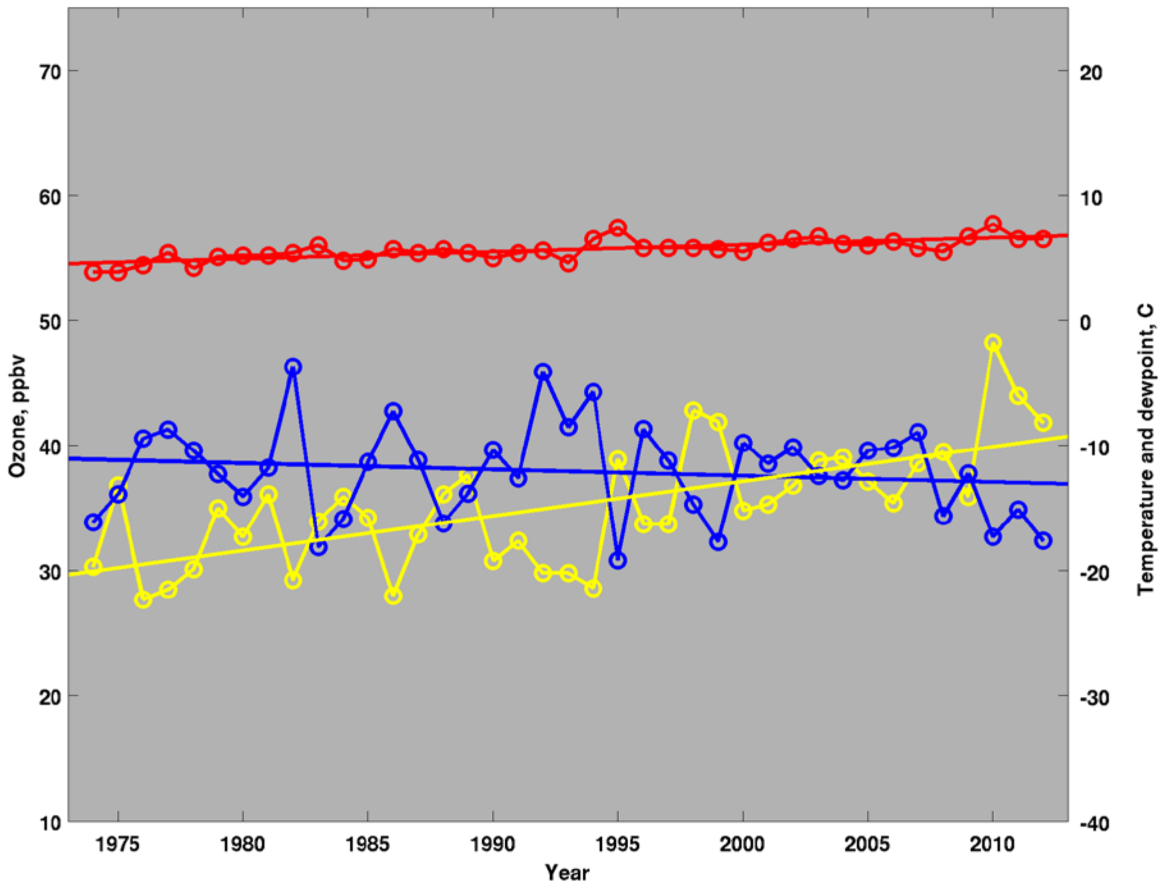
Stronger tropical influence

	Rate of increase	p value
Blue - median dewpoint	0.06 degC yr^{-1}	0.40
Red - median temperature	0.05 degC yr^{-1}	0.00
White - median ozone, dry	0.10 ppbv yr^{-1}	0.33
Cyan - median ozone, moist	-0.07 ppbv yr^{-1}	0.32

Ozone trend at Mauna Loa Observatory, Hawaii, 3.4 km above sea level

Data from years: 1974 - 2012

Data from months: 7 8 9

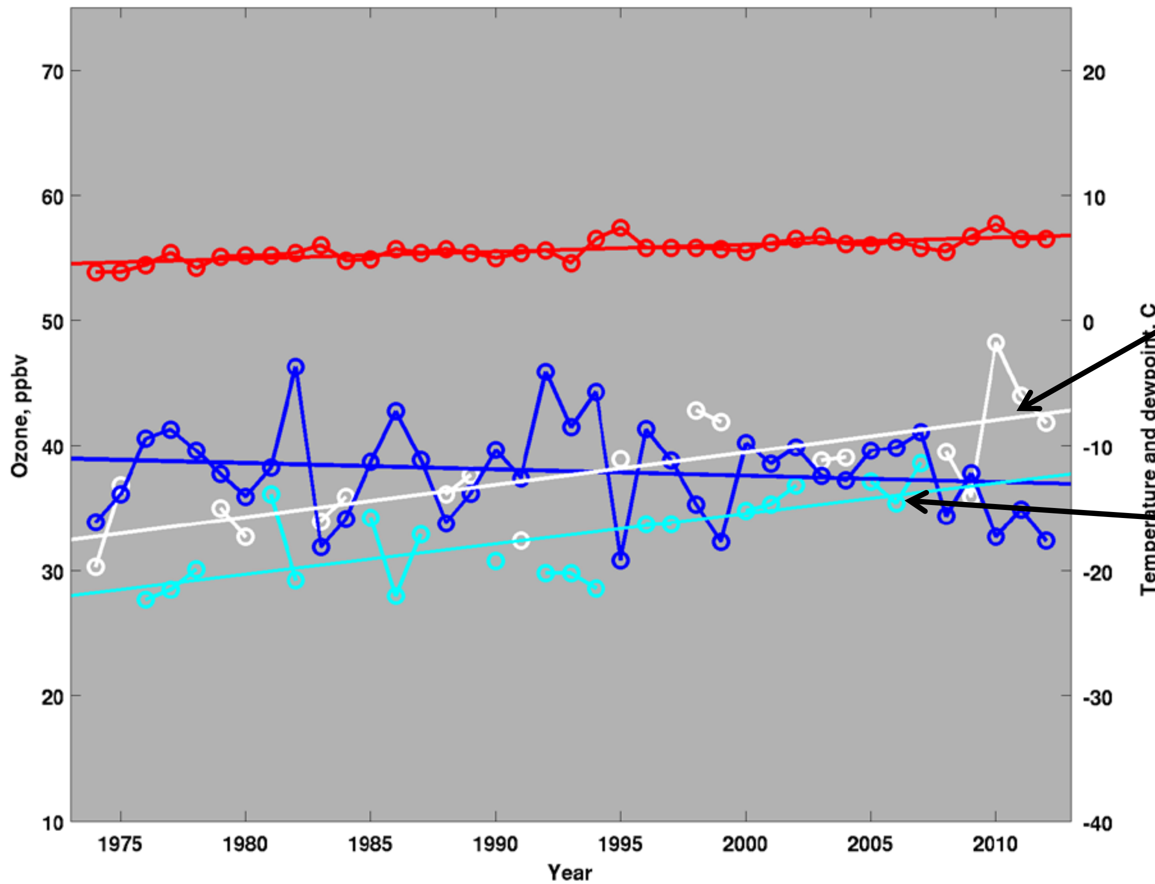


	Rate of increase	p value
Blue - median dewpoint	-0.05 degC yr^{-1}	0.36
Red - median temperature	0.06 degC yr^{-1}	0.00
Yellow - median ozone	0.28 ppbv yr^{-1}	0.00

Ozone trend at Mauna Loa Observatory, Hawaii, 3.4 km above sea level

Data from years: 1974 - 2012

Data from months: 7 8 9



	Rate of increase	p value
Blue - median dewpoint	-0.05 degC yr^{-1}	0.36
Red - median temperature	0.06 degC yr^{-1}	0.00
White - median ozone, dry	0.26 ppbv yr^{-1}	0.00
Cyan - median ozone, moist	0.24 ppbv yr^{-1}	0.00

Stronger mid-latitude influence

Stronger tropical influence

Conclusions

Surface ozone has generally increased across the northern hemisphere since the 1970s

Fewer data are available from the Southern Hemisphere but ozone appears to have also increased since the 1970s.

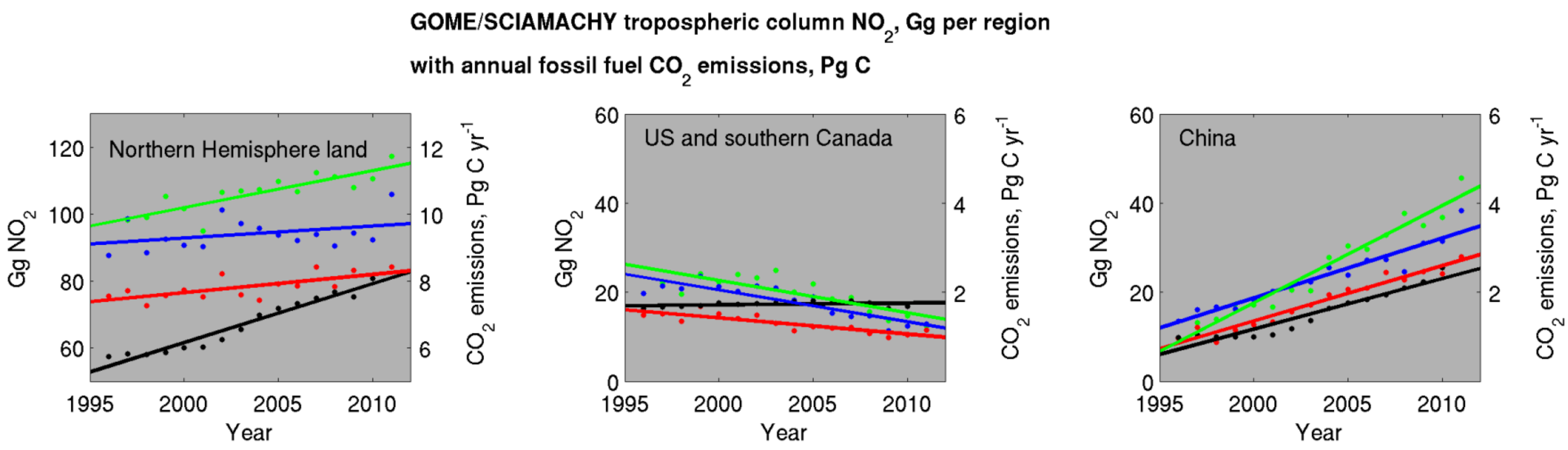
Since 1990 surface ozone has leveled off in Europe, decreased somewhat in the eastern US and increased in East Asia.

Free tropospheric monitoring sites downwind of Asia are limited to the western North America free troposphere during spring and Mauna Loa. Both sites shows increasing ozone.

Ozone increases at Mauna Loa are strongest in late summer and absent in spring.

Mauna Loa is an excellent site for evaluating model performance due to its long record (39 years), free tropospheric characteristics, and contrasting spring and summer ozone trends.

EXTRA SLIDES



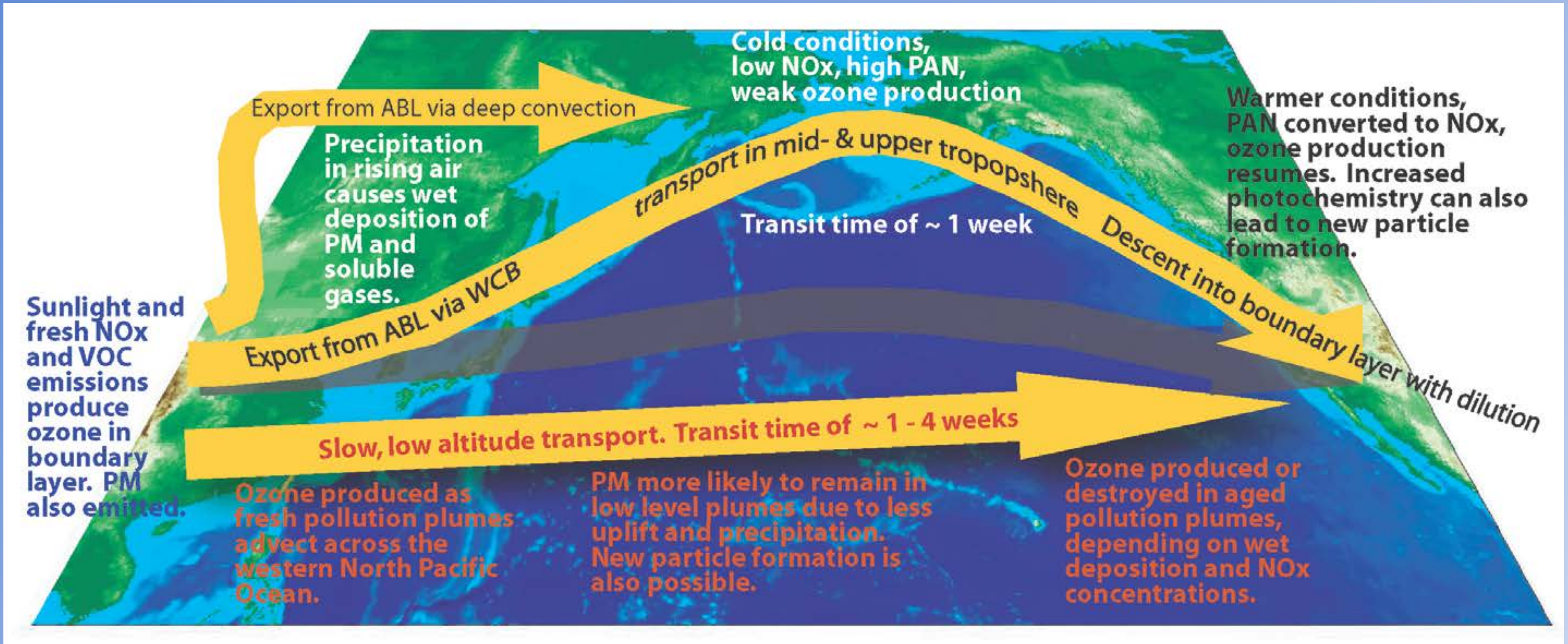
Green - NO_2 spring (Mar Apr May)
Red - NO_2 summer (Jun Jul Aug)
Blue - NO_2 autumn (Sep Oct)
Black - annual CO_2 emissions

NO_x emissions in China doubled from 1990-2005 and are currently increasing at the same relative rate as CO_2 emissions.

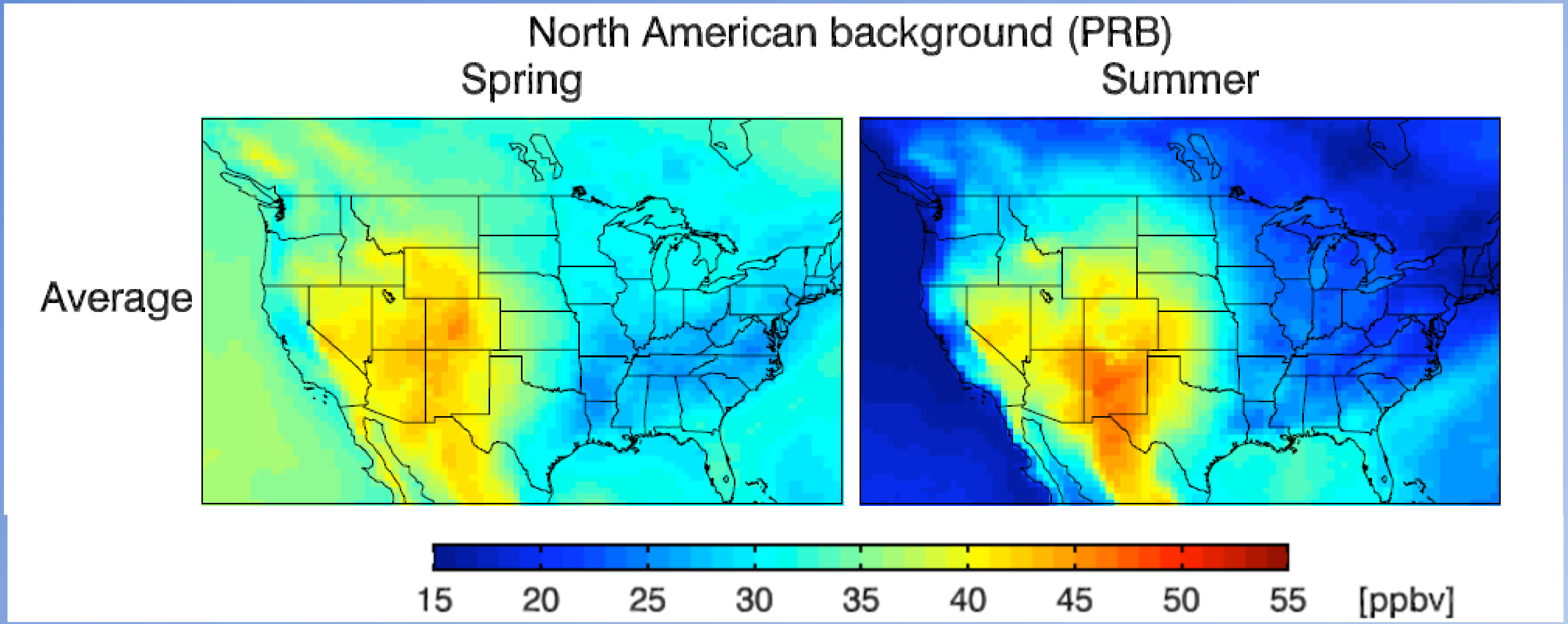
Tropospheric NO_2 column data from the GOME and SCIAMACHY sensors were freely downloaded from: www.temis.nl
For methodology see:

Boersma, K. F., et al. (2004), Error analysis for tropospheric NO2 retrieval from space, J. Geophys. Res., 109, D04311,
Richter, A., et al.(2005), Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437

General intercontinental transport processes.



Hemispheric Transport of Air Pollution 2010, Part A: Ozone and Particulate Matter, F. Dentener, T. Keating and H. Akimoto (eds.), Air Pollution Studies No. 17, United Nations, New York and Geneva, ISSN 1014-4625, ISBN 978-92-1-117043-6.

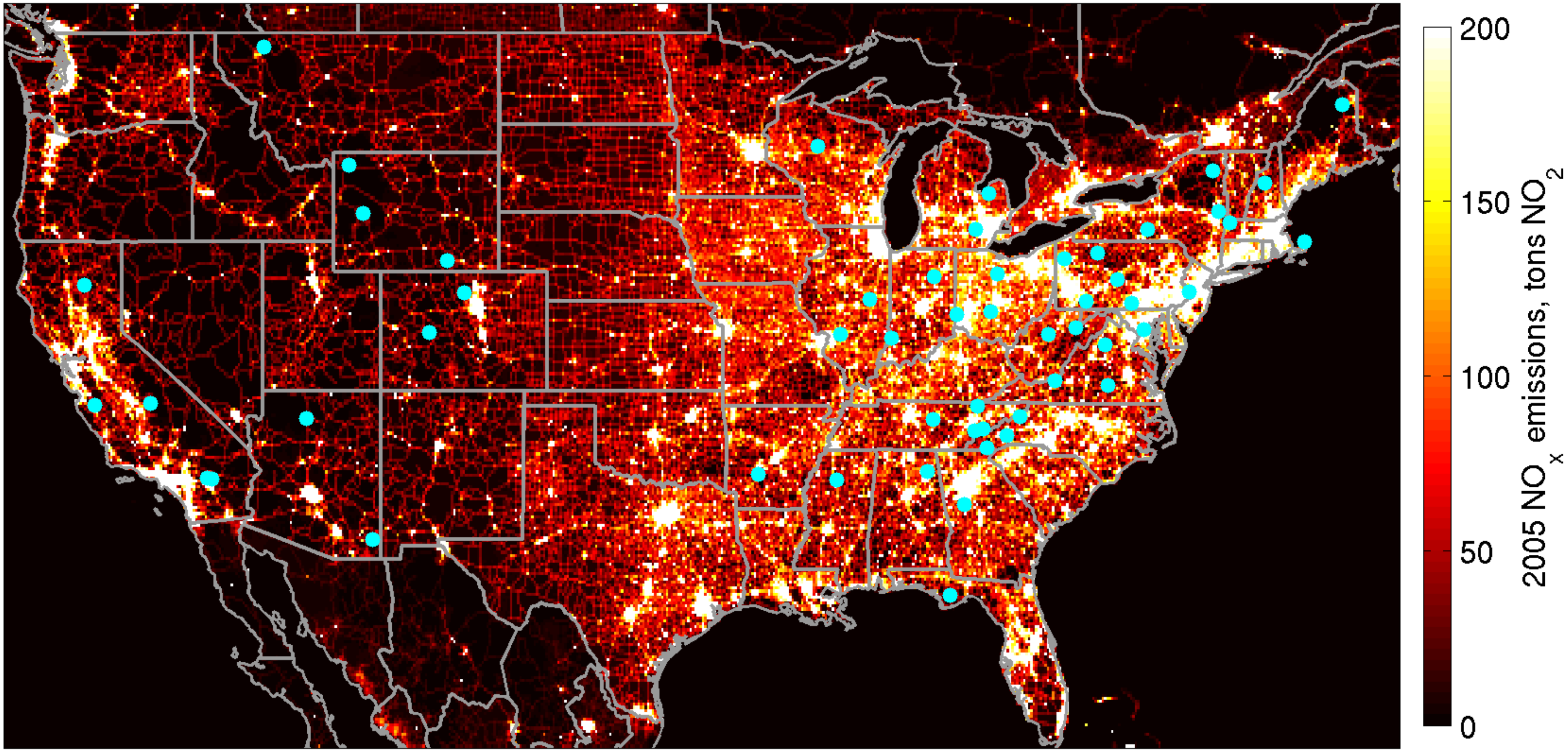


North American background (PRB) ozone concentration in surface air for spring and summer 2006.

From: Zhang, L., et al. (2011), Improved estimate of the policy-relevant background ozone in the United States using the GEOS-Chem global model with 1/2°x 2/3° horizontal resolution over North America, Atmos. Environ., in-press.

Locations of the 53 rural monitoring sites used in the study

- 12 in the west and 41 in the east



All sites have data from 1990-2010

Mid-day data only (11:00-16:00 local time)

Most eastern sites are below 1000 m a.s.l

Most western sites are above 1500 m a.s.l.

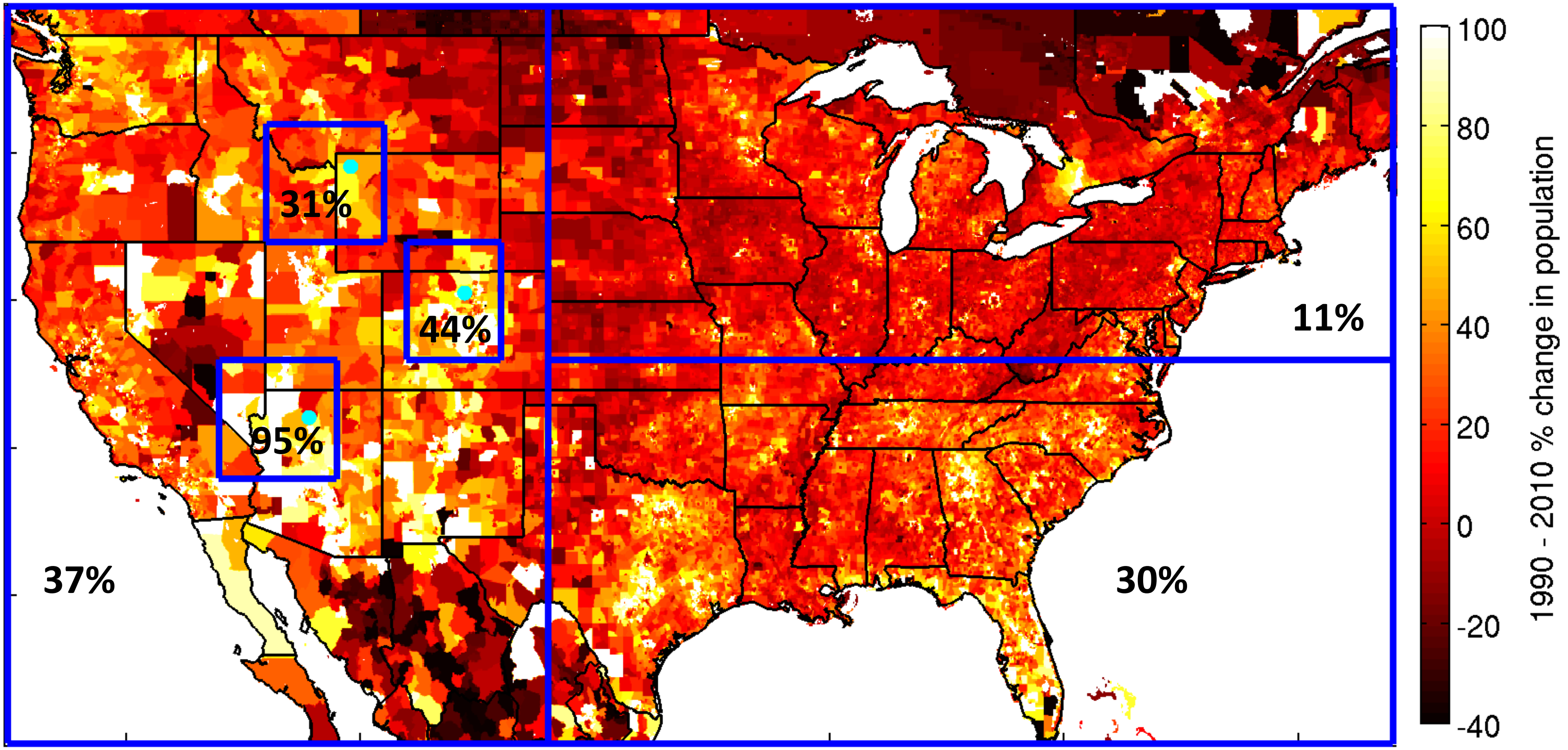
Data collected by:

National Park Service Air Resources Division

EPA Clean Air Status and Trends Network (CASTNET)

Whiteface Mtn. Summit, NY, data from U. of Albany

The US population increased by 22% from 1990-2010



1990-2010 US ozone precursor emission reductions (source: EPA)

NOx	-49 %
CO	-58 %
VOC	-44 %

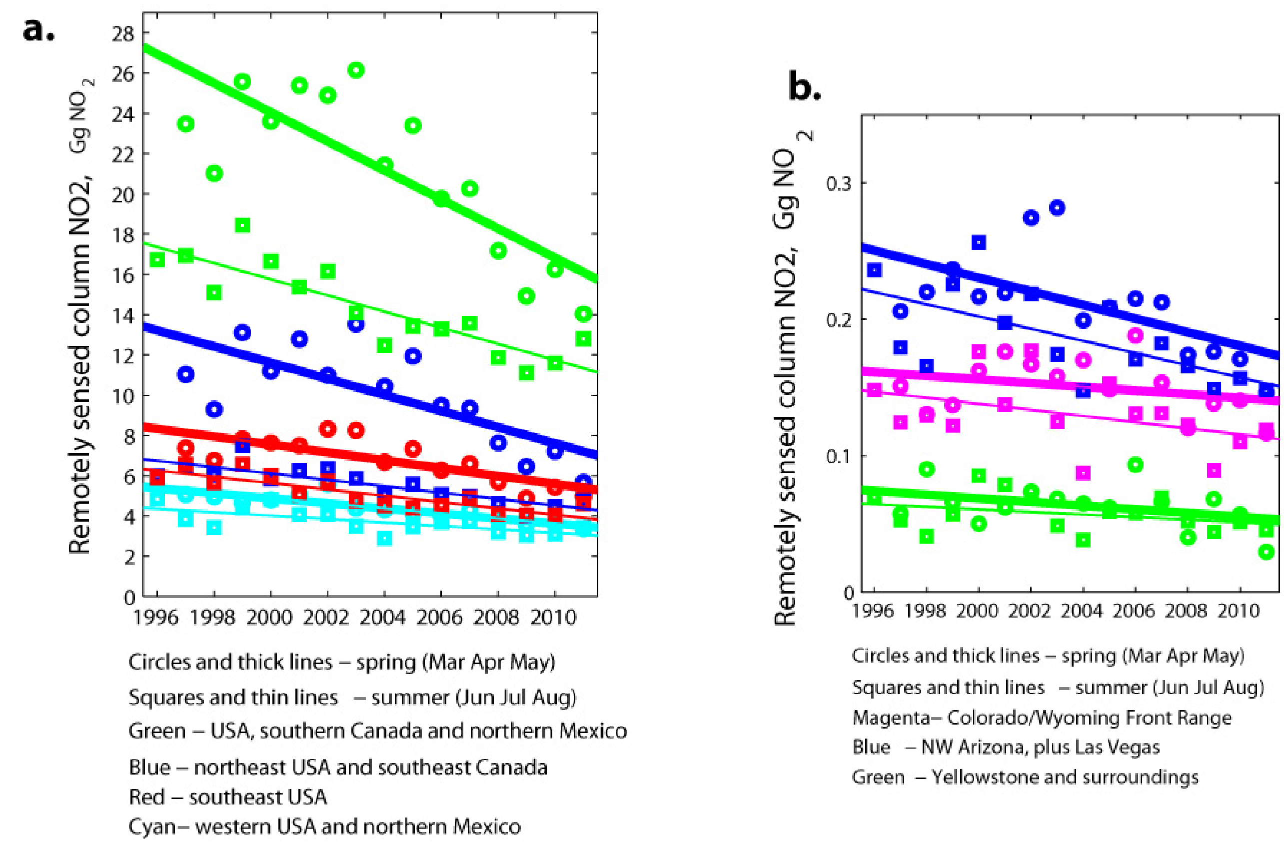
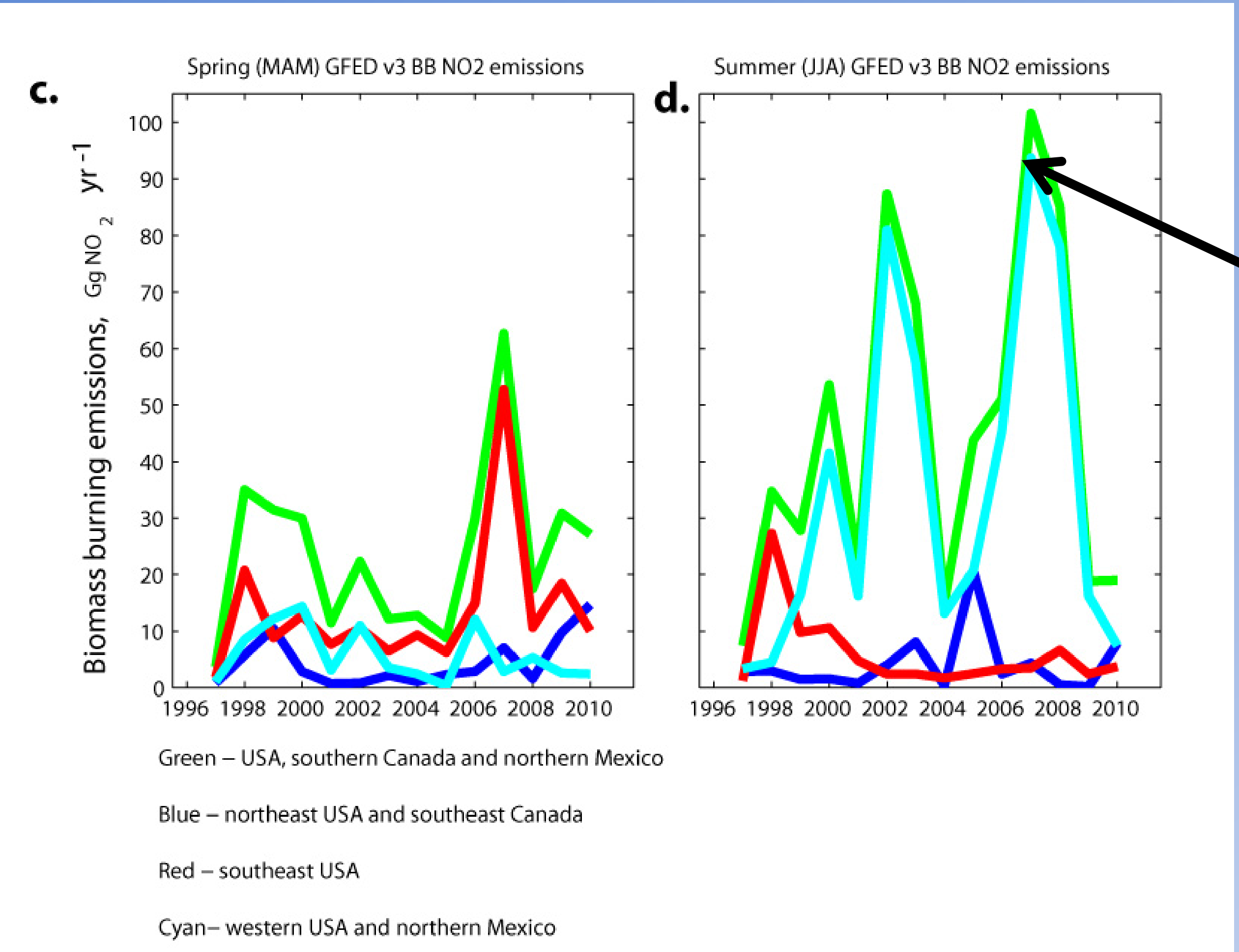


Figure 3.

a) Trends in tropospheric column NO_2, as detected by the GOME and SCIAMACHY instruments for spring (circles and thick lines) and summer (squares and thin lines) across the continental USA, and three sub-regions.

b) Tropospheric column NO_2 trends for three small areas in the western USA.

Tropospheric NO_2 column data from GOME /SCIAMACHY was freely downloaded from: www.temis.nl



2007 biomass burning emissions in the western USA were only 12 % of anthropogenic emissions in the same region.

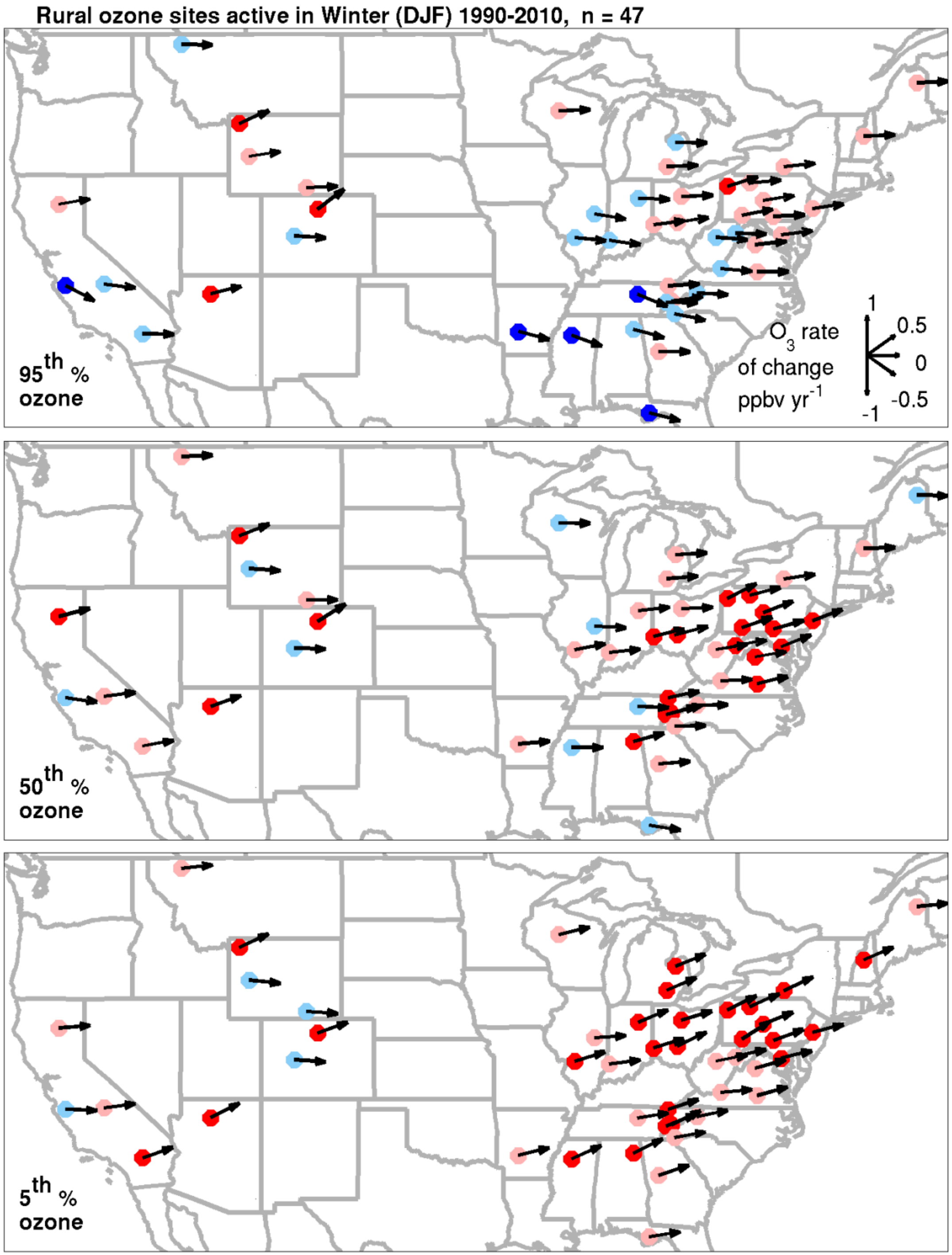
Figure 3.

c) NO_2 emitted by biomass burning across the USA in spring.

d) NO_2 emitted by biomass burning across the USA in summer.

Data from GFED v3 emission inventory:
van der Werf, et al. (2010), Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997-2009), Atmos. Chem. Phys., 10, 11707-11735.

Winter 1990-2010 ozone trends



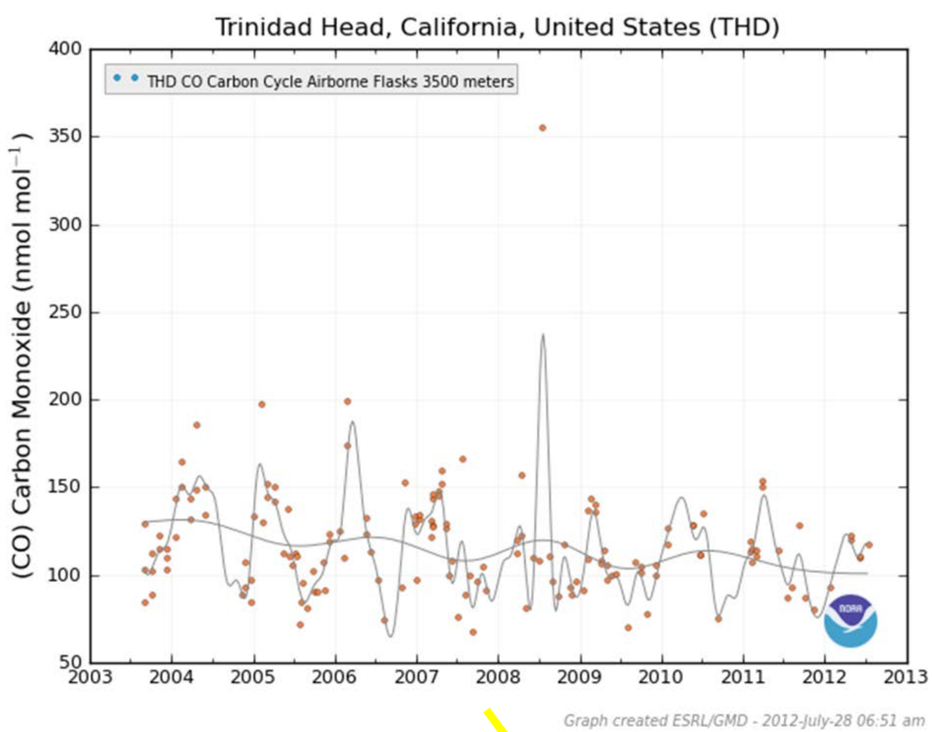
significant increase

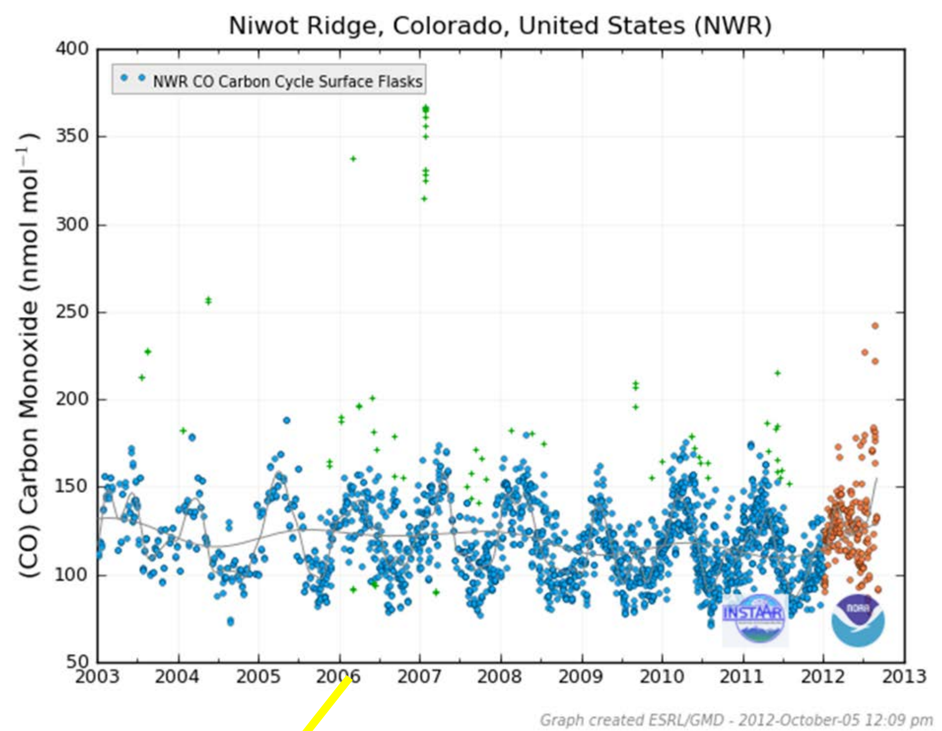
insignificant increase

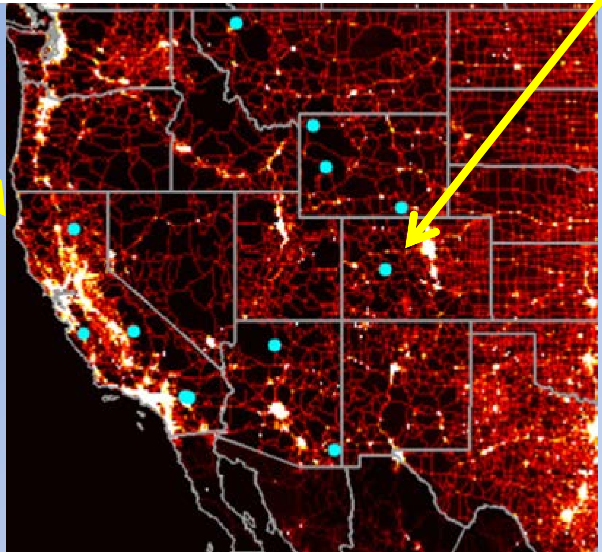
significant decrease

insignificant decrease

CO at 3.5 km above Trinidad Head is similar to Niwot Ridge







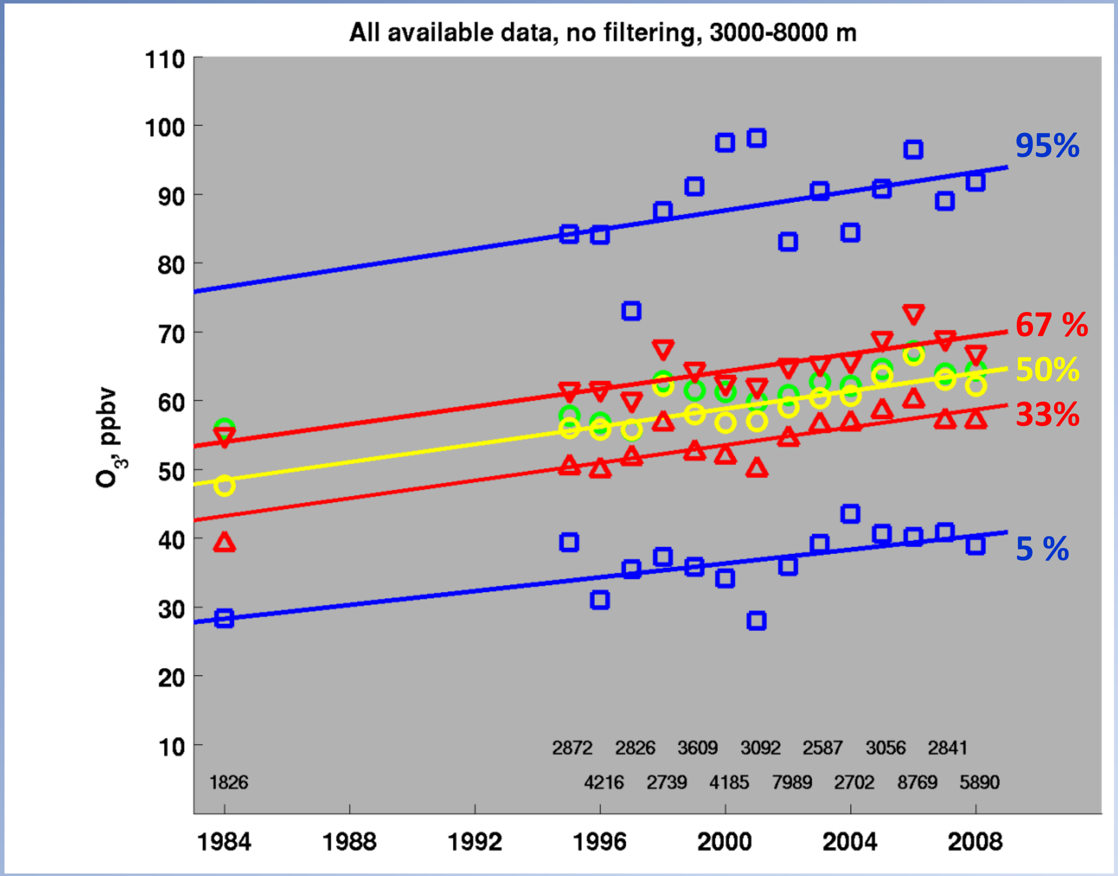
CO measurements from T. Head and Niwot Ridge, courtesy of NOAA Global Monitoring Division, Boulder.

Ozone trend in the free tropospheric above western North America

Cooper et al. [2010] used all available measurements above western North America to show that ozone is increasing significantly during spring.

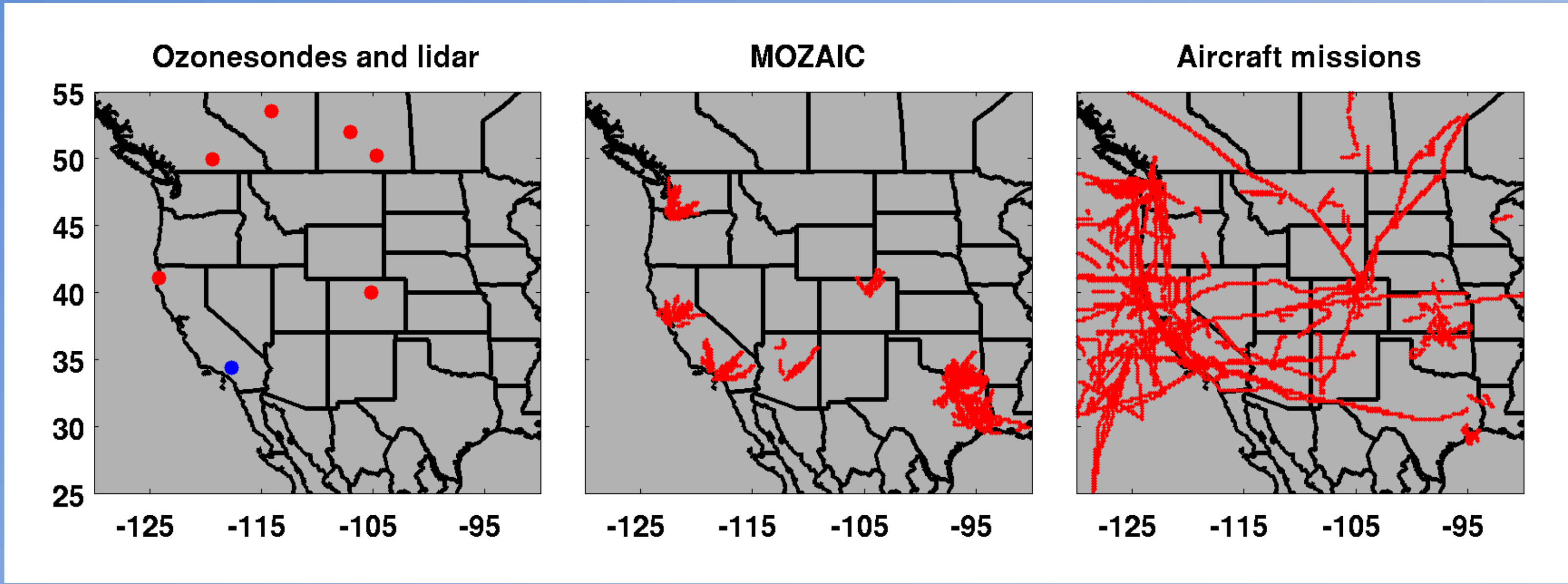
The analysis covered the period 1984-2008.

But what has happened since?



Cooper, O. R., et al. (2010), Increasing springtime ozone mixing ratios in the free troposphere over western North America, *Nature*, *463*, 344–348, doi:10.1038/nature08708.

Ozone measurements used to determine the free-tropospheric ozone trend:
April-May, 1984-2008, 3000-8000 m



Ozonesondes, 1995-2008

Trinidad Head, CA
Boulder, CO
Kelowna, BC
Edmonton/Stoneyplain, AB
Vanscoy, SA
Bratt's Lake, SA

Lidar, 2003-2008

Table Mountain, CA

MOZAIC, 1995-2008

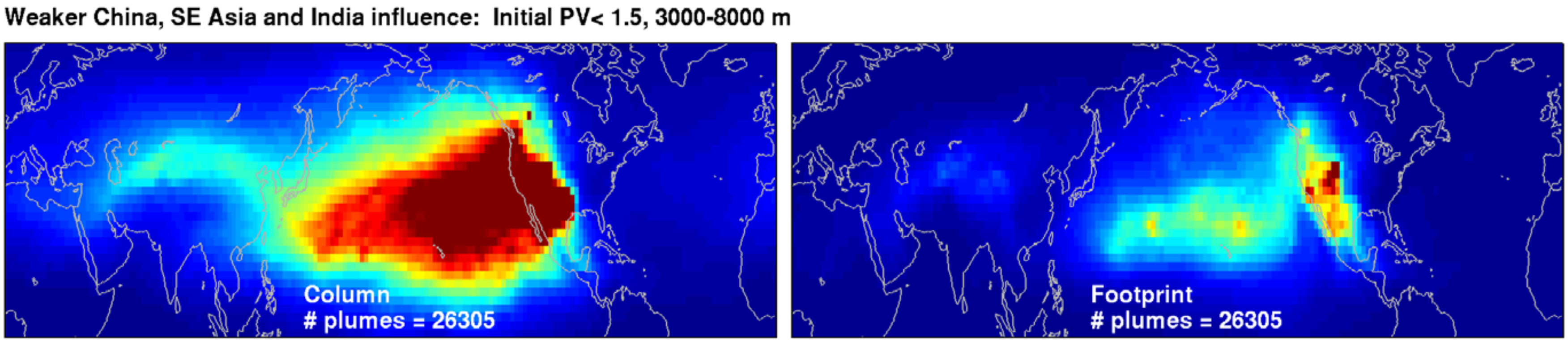
Portland, OR
San Francisco, A
Los Angeles, BC
Phoenix, AZ
Denver, CO
Dallas, TX
Houston, TX

CITE 1-C	1984
NO DATA	
STRAT	1995
SUCCESS	1996
POLARIS	1997
WAM	1998
PEM-TROPICS B	1999
PHOBEA 1999, 2001,	2003
TOPSE	2000
TRACE-P	2001
ITCT	2002
INTEX-B	2006
PACDEX	2007
ARCPAC	2008
START08	2008

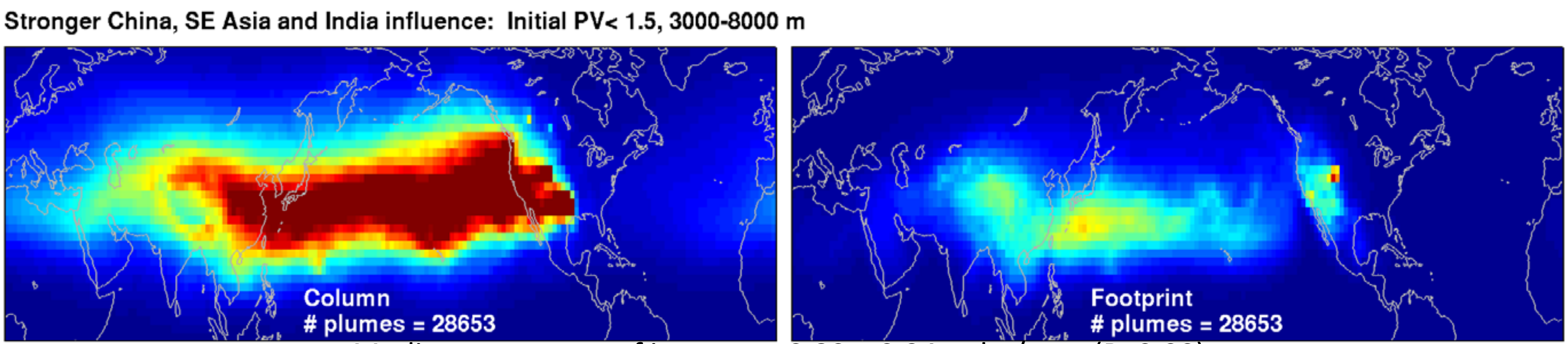
The FLEXPART Lagrangian Particle Dispersion Model was used to calculate the 15-day transport history, or retroplume, of each ozone measurement.

Using the retroplumes, the data set was split into two groups, measurements with stronger transport from South and East Asian emissions regions, and measurements with weaker influence.

1995-2008



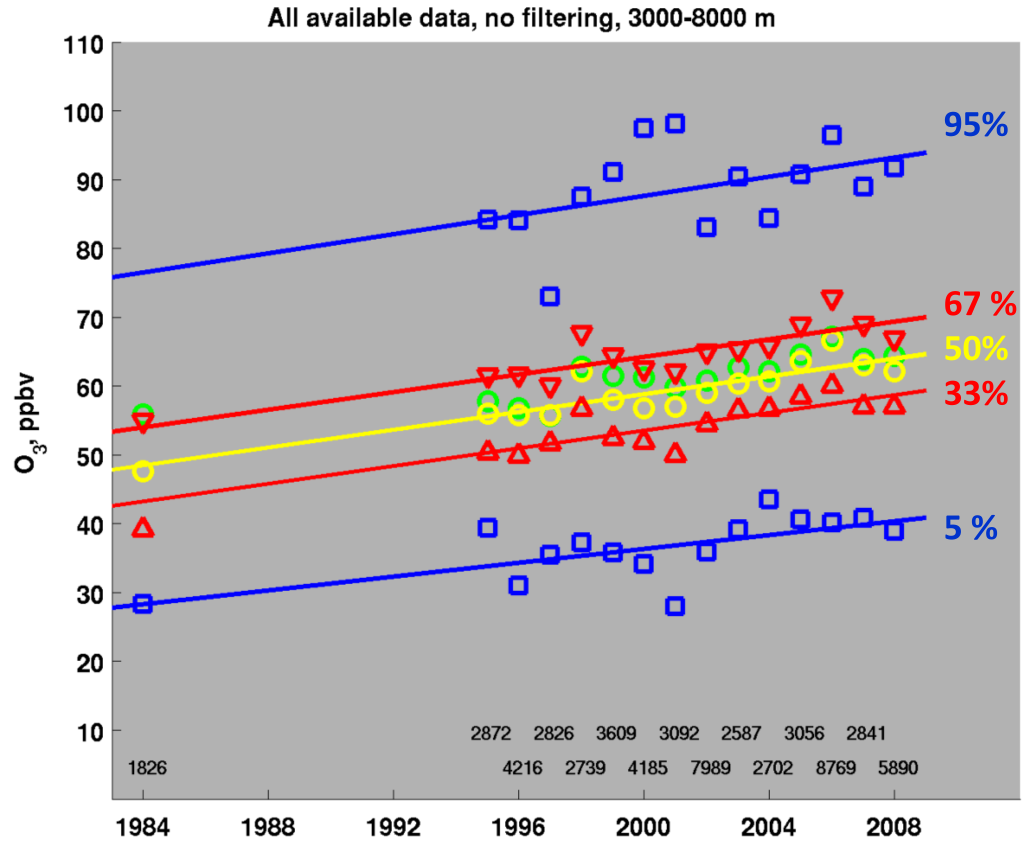
Median ozone rate of increase: 0.45 ± 0.32 ppbv/year (P=0.01)



Median ozone rate of increase: 0.80 ± 0.34 ppbv/year (P=0.00)

Free tropospheric ozone trend above western North America

All available data above western North America, regardless of transport history.



Yellow - median

Green - mean

Blue - 5th & 95th percentiles

Red - 33rd and 67th percentiles

1995 - 2008

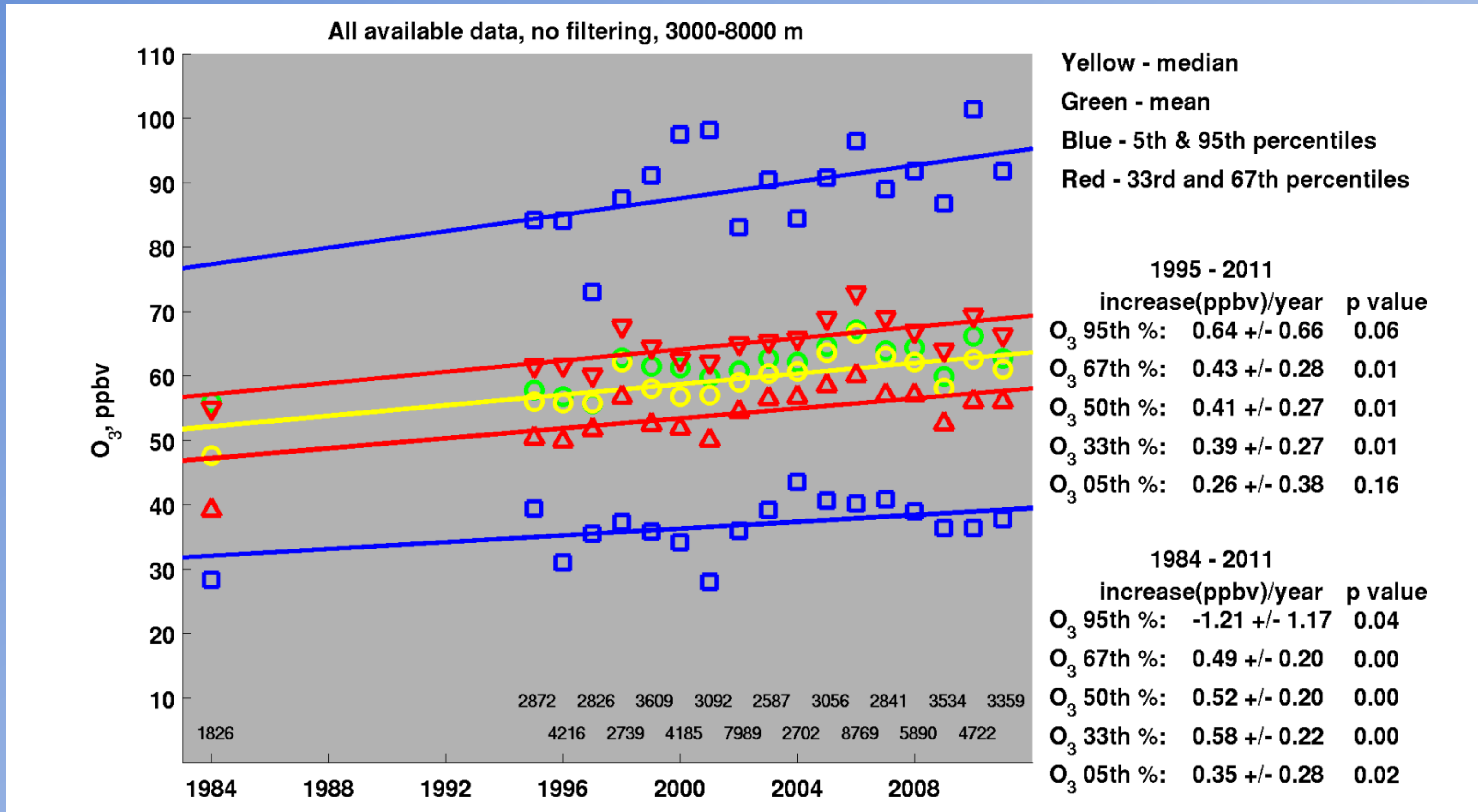
	increase(ppbv)/year	p value
O_3 95th %:	0.70 +/- 0.91	0.12
O_3 67th %:	0.64 +/- 0.35	0.00
O_3 50th %:	0.65 +/- 0.31	0.00
O_3 33th %:	0.64 +/- 0.31	0.00
O_3 05th %:	0.50 +/- 0.54	0.06

1984 - 2008

	increase(ppbv)/year	p value
O_3 95th %:	-1.78 +/- 1.45	0.02
O_3 67th %:	0.61 +/- 0.22	0.00
O_3 50th %:	0.67 +/- 0.20	0.00
O_3 33th %:	0.77 +/- 0.21	0.00
O_3 05th %:	0.50 +/- 0.34	0.01

Free tropospheric ozone trend above western North America

All available data above western North America, regardless of transport history.

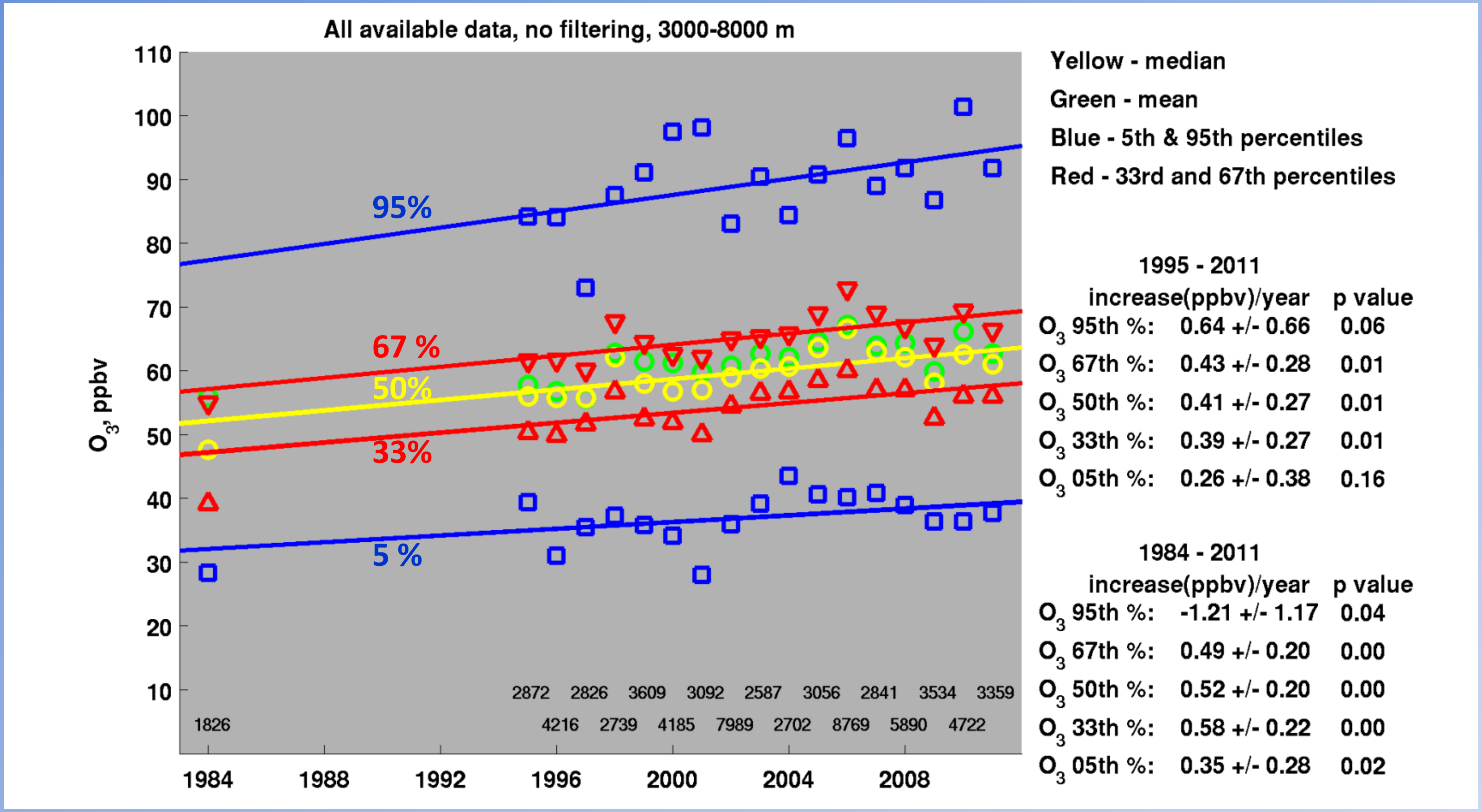


An extension of the 1995-2008 ozone trend, adding years 2009 – 2011.
Ozone has increased by 29% from 1984-2011.

FLEXPART retroplumes have not yet been calculated for 2009-2011, so data have not been filtered to remove stratospheric intrusions or to restrict by transport pathway.

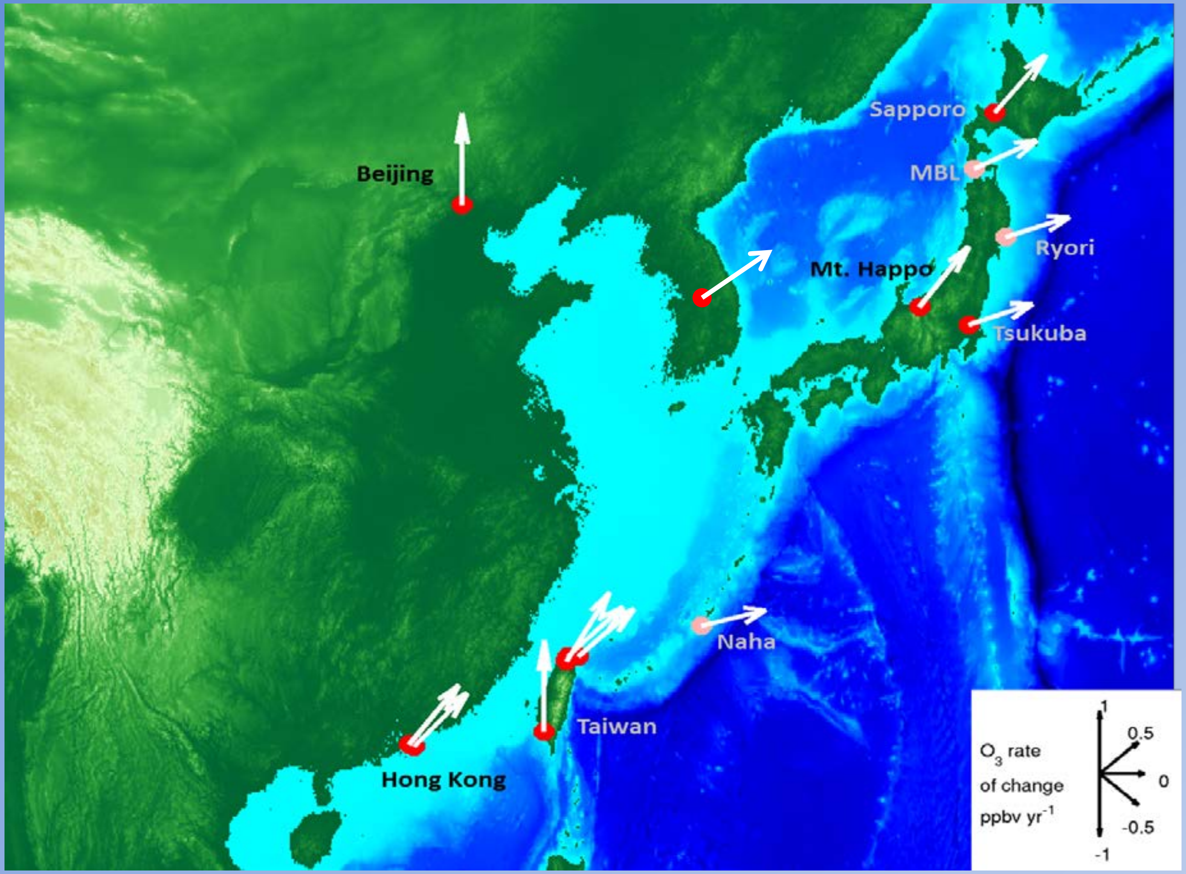
Free tropospheric ozone trend above western North America

All available data above western North America, regardless of transport history.



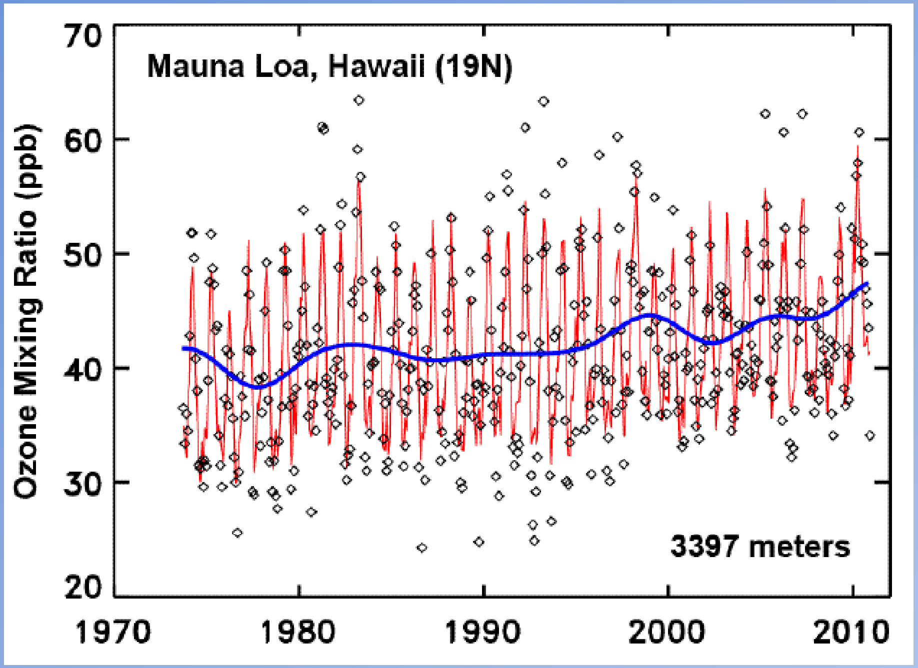
Ozone has increased by 29% from 1984-2011.

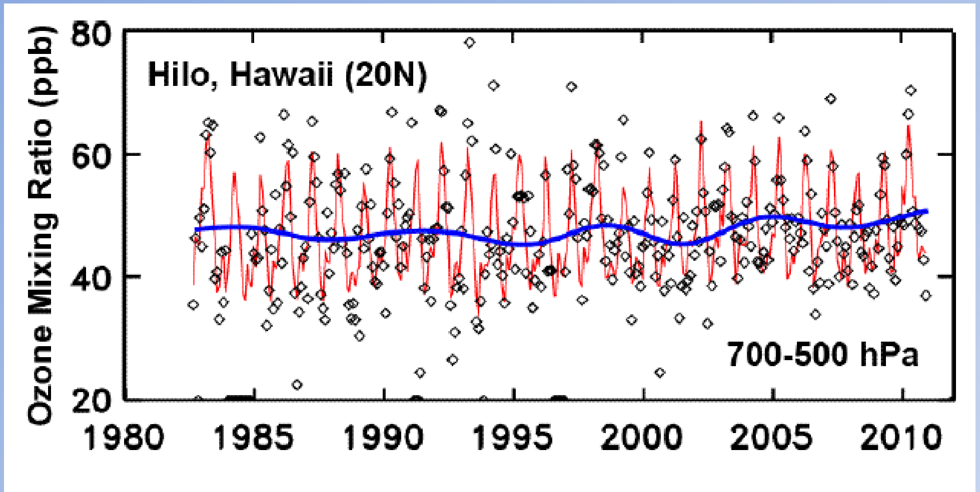
Cooper, O.R., Gao, R.S., Tarasick, D., Leblanc, T. and Sweeney, C. (2012), Long-term ozone trends at rural ozone monitoring sites across the United States, 1990-2010, *J. Geophys. Res., 117(D22307).*



Site and time period	Reference
Sapporo (1981-2010), Ryori (1991-2010), Tsukuba (1981-2010) & Naha (1991-2010), Japan:	Oltmans et al. (2012), *Atmos. Environ., in-press*
Beijing, MOZAIC profiles in boundary layer (1997-2004):	Ding et al. (2008), *ACP*
Mt. Happo (1991-2011), Japan:	Parrish et al. (2012), *ACPD*
Marine boundary layer, western Japan (1998-2011)	Parrish et al. (2012), *ACPD*
Northern Taiwan, urban, coastal and mountain (1994-2007):	Lin et al. (2010), *Environ. Monit. Assess.*
Southern Taiwan (1997-2006):	Li et al. (2010), *Atmos. Environ.*
Urban & Coastal Hong Kong (1994-2007):	Wang et al. (2009), *ACP*

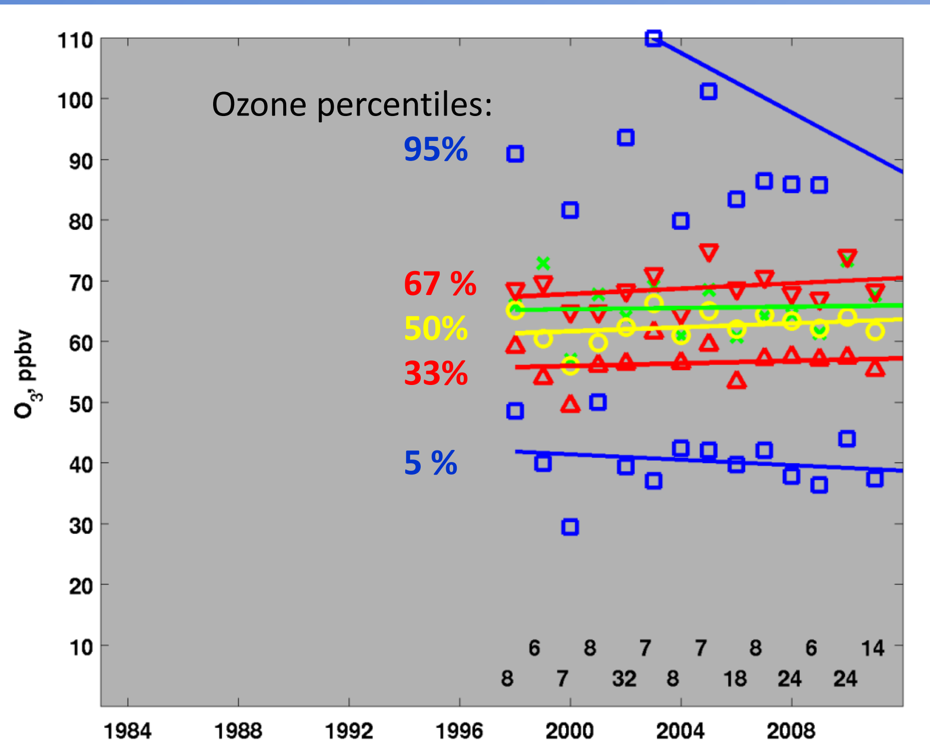
Mid-tropospheric ozone trends above Hawaii





Oltmans et al. (2012), Recent Tropospheric Ozone Changes – A Pattern Dominated by Slow or No Growth, Atmos. Environ., submitted.

April-May mid-tropospheric (3-8 km) ozone above Trinidad Head, CA

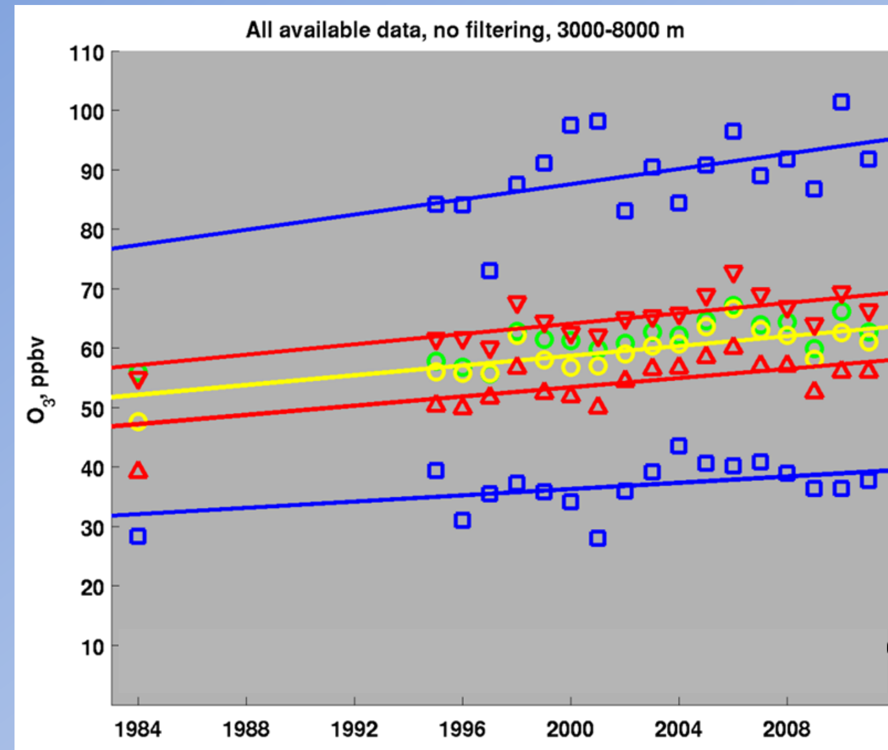


Most years have a sampling frequency of just 4 profiles per month.

Accurate characterization of monthly mean ozone requires at least 12 profiles per month at a given location.

Saunois et al. (2012), Impact of sampling frequency in the analysis of tropospheric O3 observations, *ACP, 12*, 6757-6773.

April-May mid-tropospheric (3-8 km) ozone above western North America



This composite of all available ozonesonde, aircraft and lidar profiles contains 75-350 profiles per season (April-May).

Cooper et al. (2012), Long-term ozone trends at rural ozone monitoring sites across the United States, 1990-2010, *JGR, submitted*.

Changes in ozone precursor emissions:

Bottom-up emission inventories
- Global anthropogenic NO_x emissions changed little during 1990-2005.

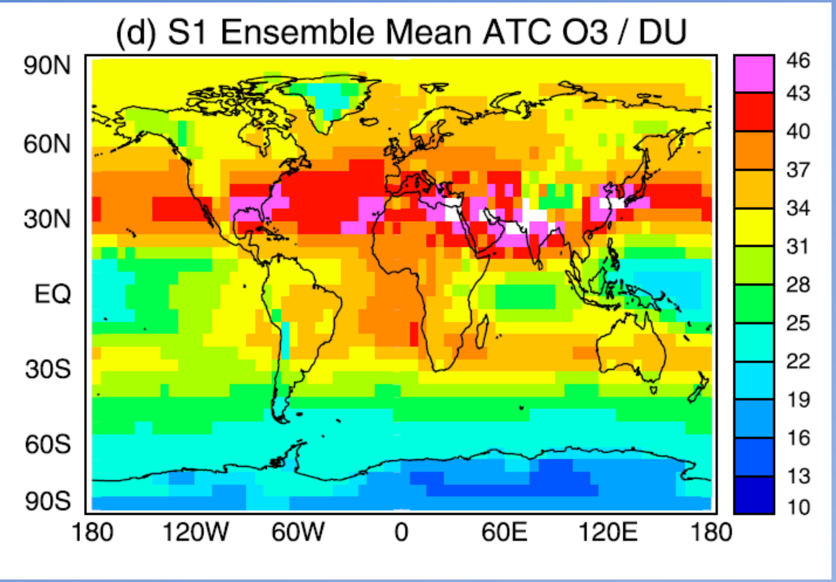
1990: 91 Tg NO_2 (Lamarque et al., 2010)
2000: 87 Tg NO_2 (Lamarque et al., 2010)
2005: 91 Tg NO_2 (EDGAR v4.1)

- but large regional shifts in NO_x emissions over the same period:

North America: -29%
Europe: -46%
Asia: +103%

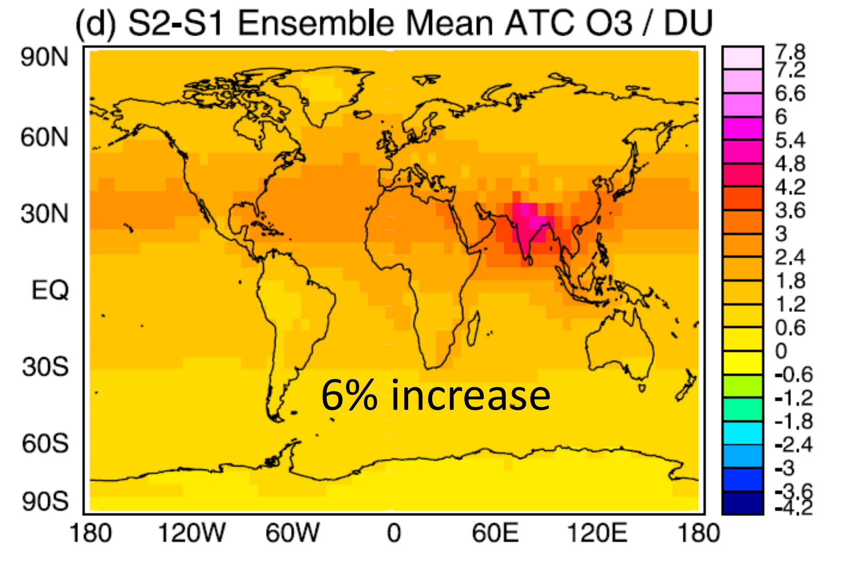
Lamarque et al. (2010), Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application, Atmos. Chem. Phys., 10, 4963-5019.

Tropospheric O_3, 2000



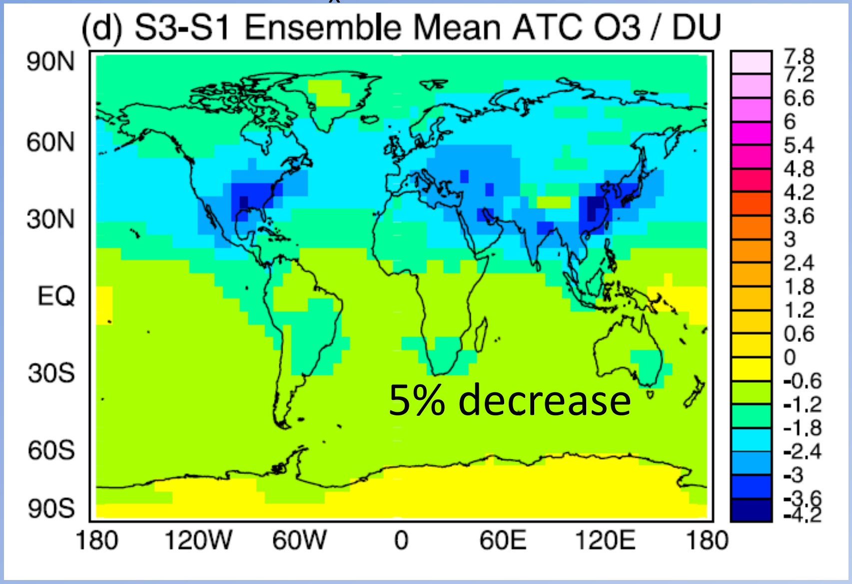
Current legislation emissions, 2030

Anthropogenic NO_x: + 18%



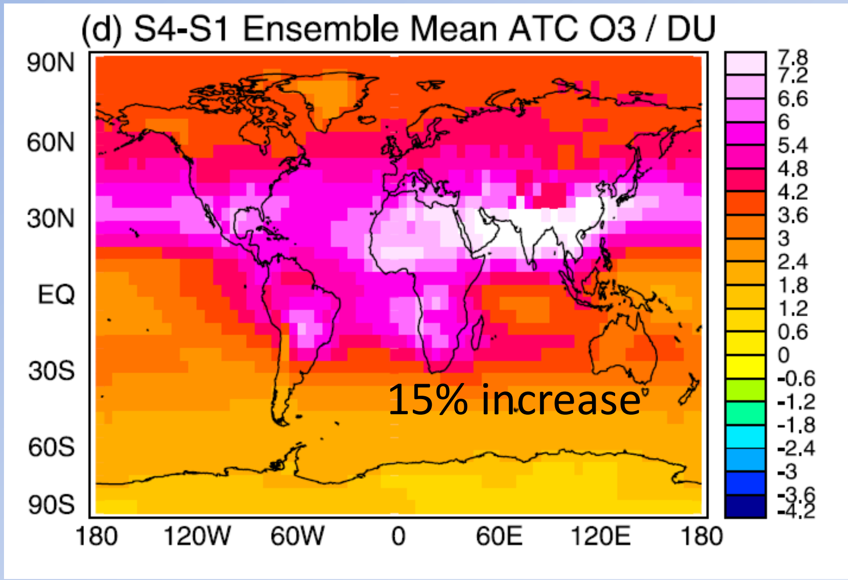
Max. feasible emission reductions, 2030

Anthropogenic NO_x: - 53%

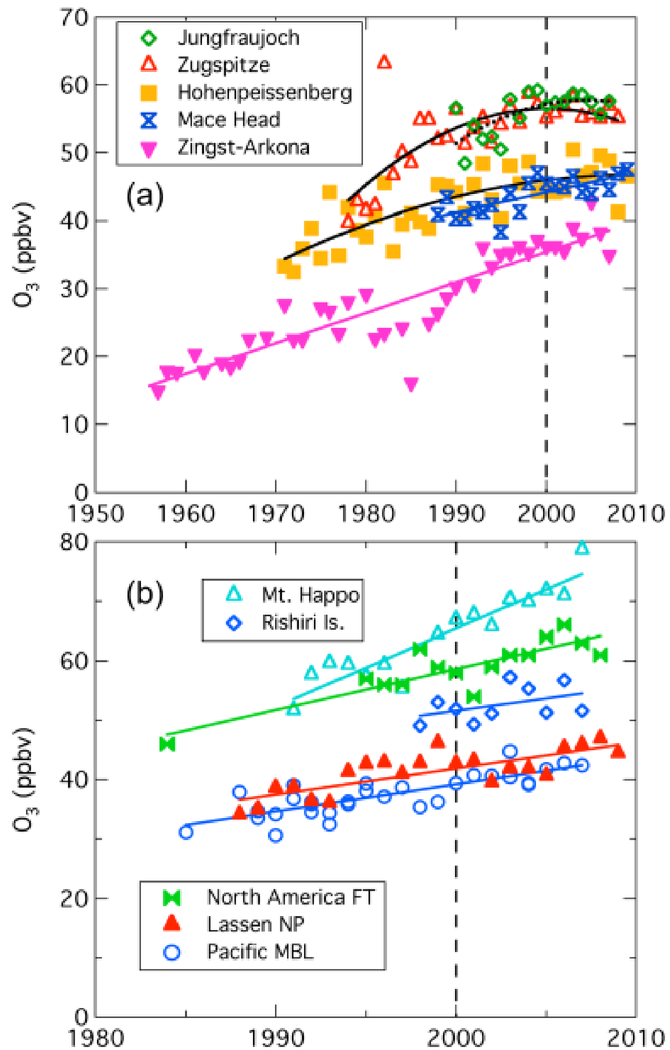


Strongly increased emissions, 2030

Anthropogenic NOx: + 96%



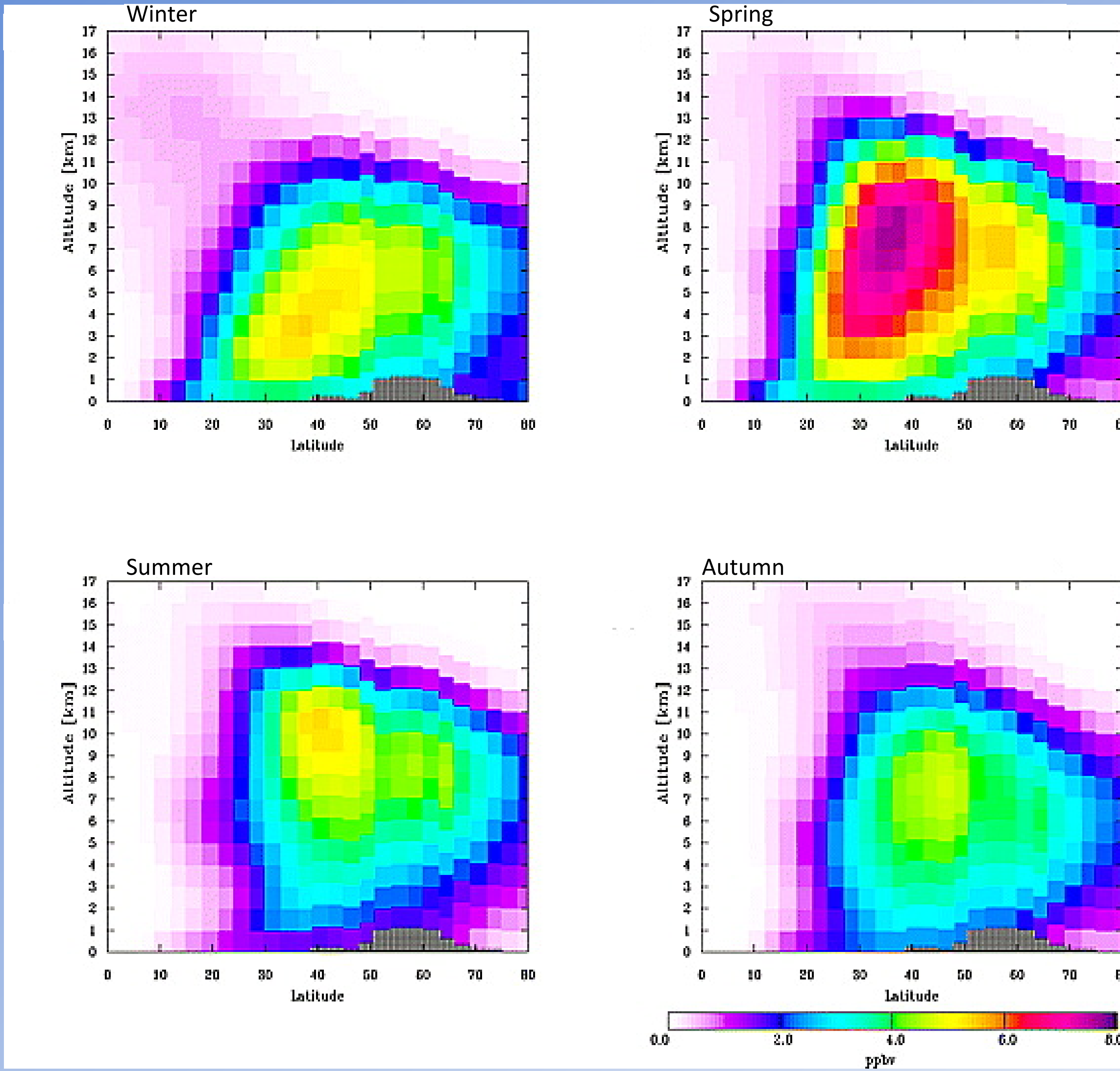
Stevenson et al., Multimodel ensemble simulations of present-day and near-future tropospheric ozone, J. *Geophys. Res., 111*, 2006.



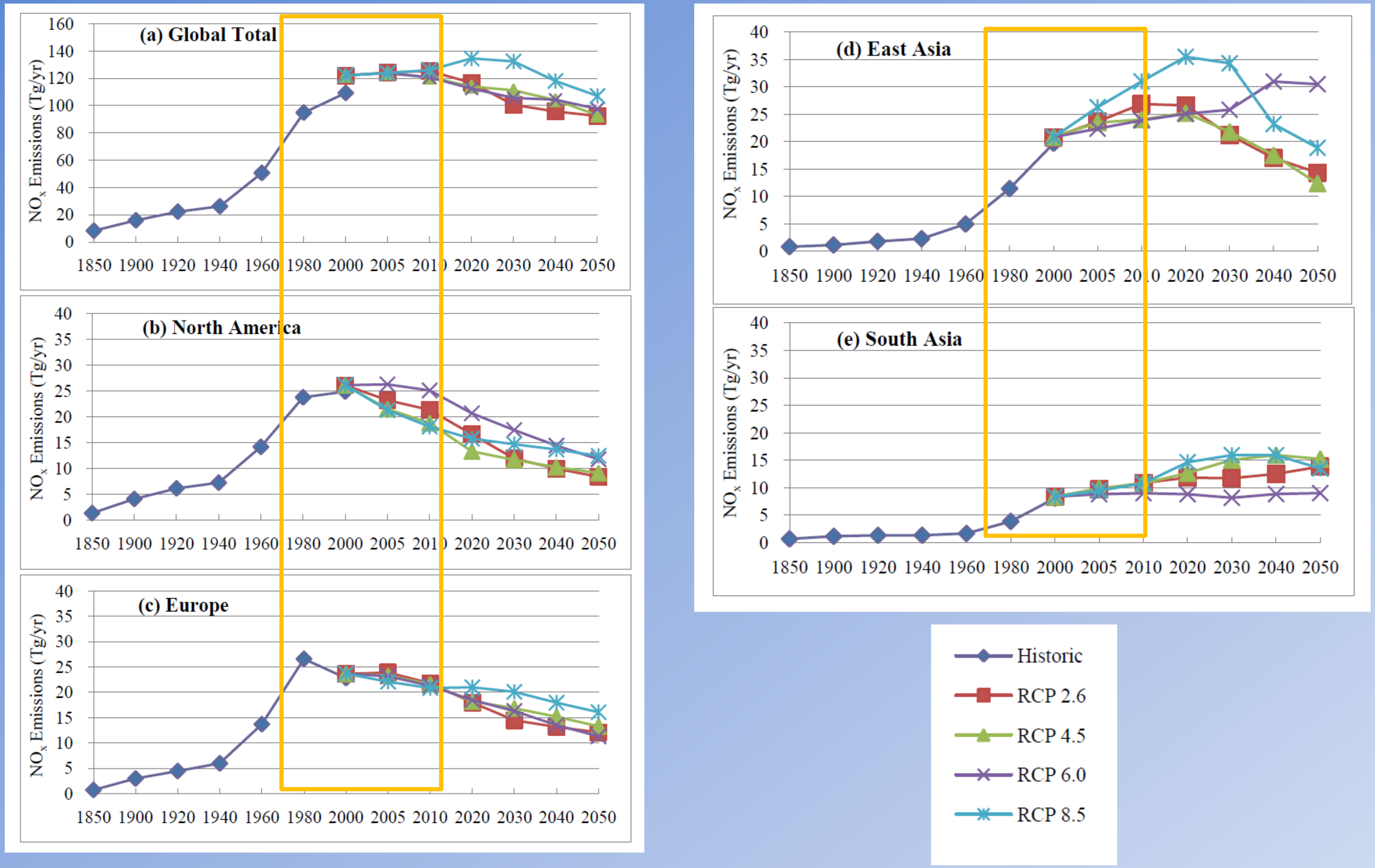
Source: HTAP 2010 report

Figure 2.7. Springtime trends in O_3 concentrations measured in (a) Europe and (b) western North America and Japan. The lines (in colour) indicate the linear regressions to the data, and the curves (in black) indicate quadratic polynomial fits to the three central European sites over the time span of the lines. Arkona and Zingst are two sites located close to the Baltic Sea. Mace Head is located at the west coast of Ireland. Hohenpiessenberg (1.0 km asl) and Zugspitze (3.0 km asl) are in southern Germany, and Jungfraujoch (3.6 km asl) is in Switzerland. The North American data are from several sea level Pacific coastal sites and Lassen National Park (1.8 km asl) near the west coast, and from the free troposphere over the western part of the continent. The Japanese data are from Mt. Happo (1.9 km asl) on the Japanese mainland and Rishiri, a northern (45N) sea level island site.

15-yr climatology of Asian anthropogenic CO tracer along the west coast of North America, by season.



Forster, C., et al. (2004), Lagrangian transport model forecasts and a transport climatology for the Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) measurement campaign, J. Geophys. Res., 109, D07S92.



Historic (1850-2000) global and regional anthropogenic NO_x emissions, with future RCP scenarios (2000-2050).

Figure 3.10 from: *Hemispheric Transport of Air Pollution 2010, Part A: Ozone and Particulate Matter, F. Dentener, T. Keating and H. Akimoto (eds.), Air Pollution Studies No. 17, United Nations, New York and Geneva, ISSN 1014-4625, ISBN 978-92-1-117043-6.*

Impact of Asia on springtime ozone across the N. Pacific Ocean and North America

Surface 500 hPa 300 hPa

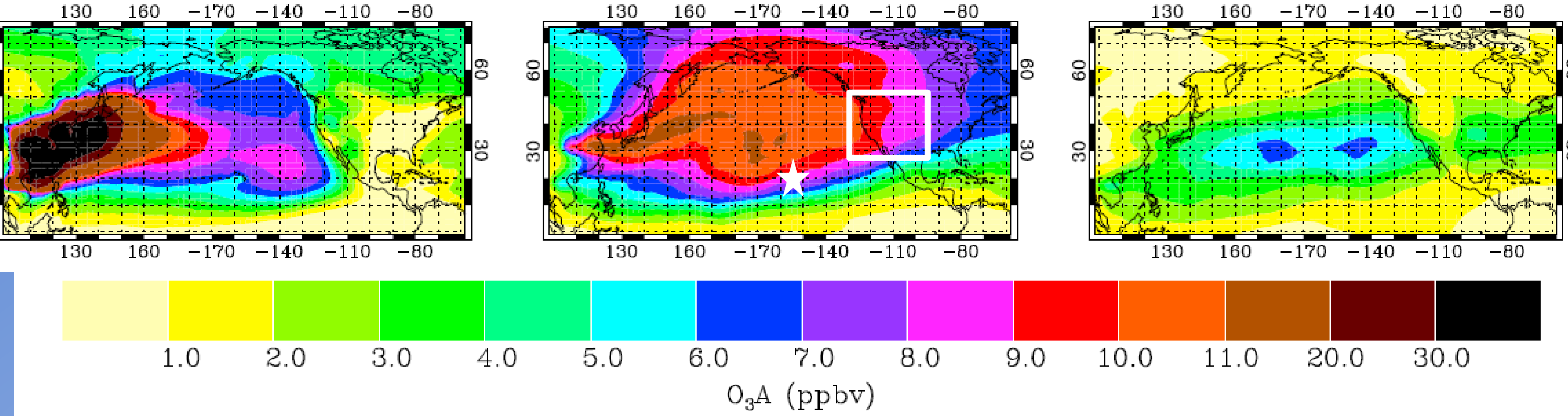


Figure 1. Seasonal five-year average of O_3A over the Pacific Basin and North America at the surface, 500 hPa (approximately 5 km), and 300 hPa (approximately 10 km) taken from the 2001–2005 model results.

Ozone transport from Asia is at a maximum during spring.

The "sphere of influence" of Asian ozone is strongest in the mid-troposphere and reaches Hawaii and western North America.

Only free tropospheric ozone datasets sites downwind of Asia:

- Mauna Loa and Hilo, Hawaii
- Springtime composite dataset above western N. America

Brown-Steiner, B., and P. Hess (2011), Asian influence on surface ozone in the United States: A comparison of chemistry, seasonality, and transport mechanisms, *J. Geophys. Res., 116*, D17309, doi:10.1029/2011JD015846.