#### CHANGES IN EMISSIONS AND AMBIENT CONCENTRATIONS OF NITROGEN DIOXIDE: THE U.S. AND CALIFORNIA EXPERIENCE

#### **Robert Harley (harley@berkeley.edu)**

Department of Civil and Environmental Engineering University of California, Berkeley

May 4, 2015

Health Effects Institute Annual Meeting

# Acknowledgments

UC Berkeley: Tim Dallmann, Tom Kirchstetter, Brian McDonald, Chelsea Preble, Ivy Tao

#### Research funding:

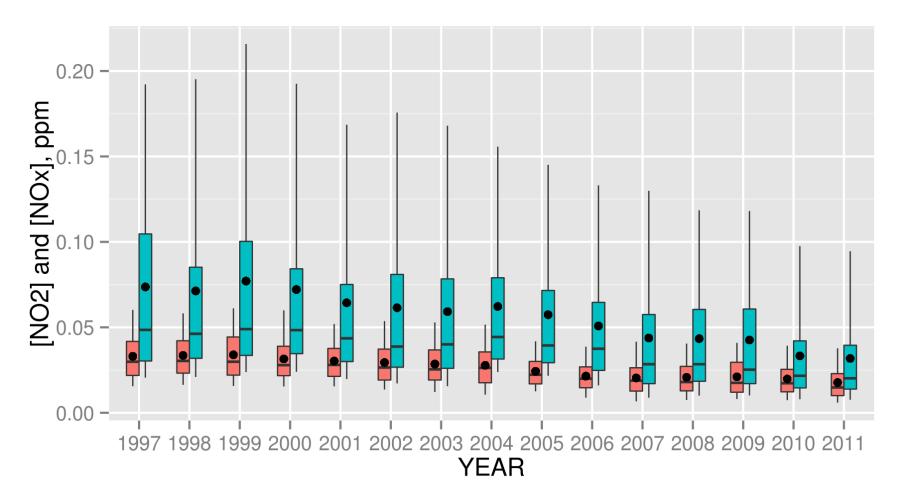
- California Air Resources Board (Contract no. 09-340)
- U.S. EPA (STAR grant no. RD834553)
- National Science Foundation Fellowship (Chelsea Preble)

## Introduction

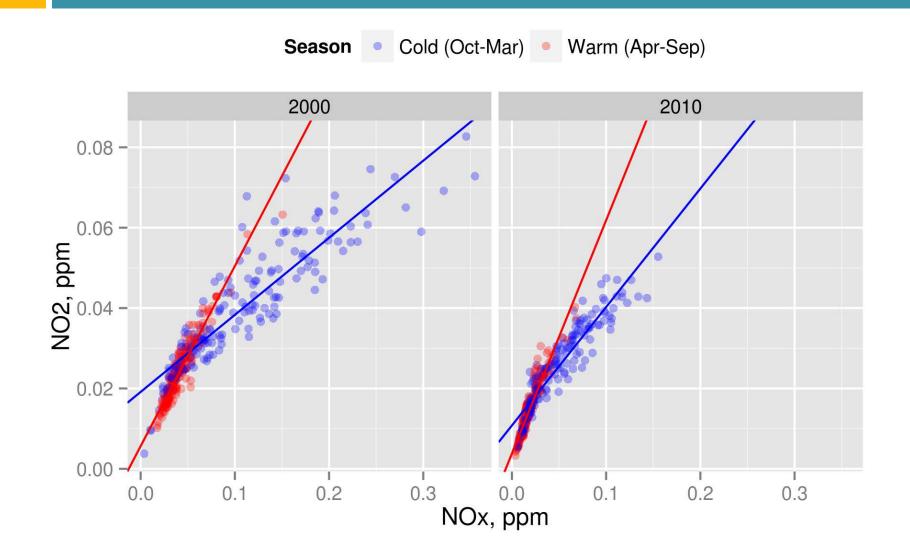
- NO<sub>2</sub> typically thought of as a secondary pollutant
  Forms via atmospheric oxidation of primary NO<sub>x</sub> emissions which are mostly NO (nitric oxide)
- Emissions and ambient concentrations of NO<sub>x</sub> have been decreasing in the U.S. since the late 1990s
- Ambient NO<sub>2</sub> is not decreasing as rapidly as NO<sub>x</sub>
  due in part to increased primary NO<sub>2</sub> emissions?

### **Trends in Ambient NO<sub>2</sub> and NO<sub>x</sub>** (Daily Averages from Long Beach, CA)

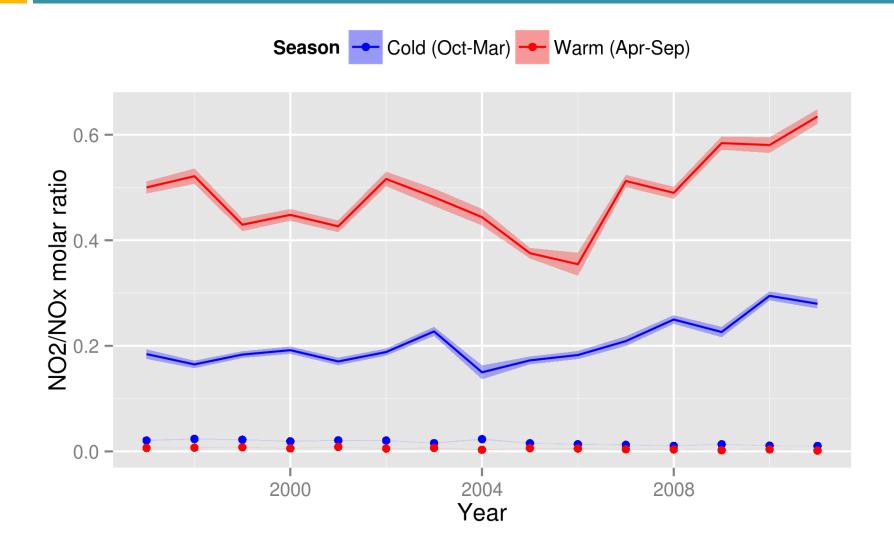




#### Correlation Between Ambient NO<sub>2</sub> & NO<sub>x</sub> (Daily Averages from Long Beach, CA)



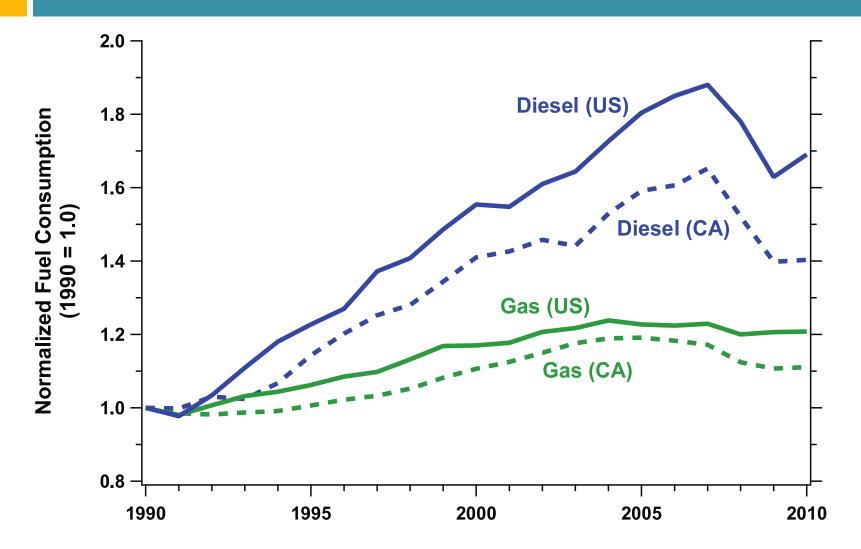
#### **Trends in Ambient NO<sub>2</sub>/NO<sub>x</sub> Ratio** (Separate Results for Winter and Summer)



# $NO_x$ and $NO_2$ Emission Trends

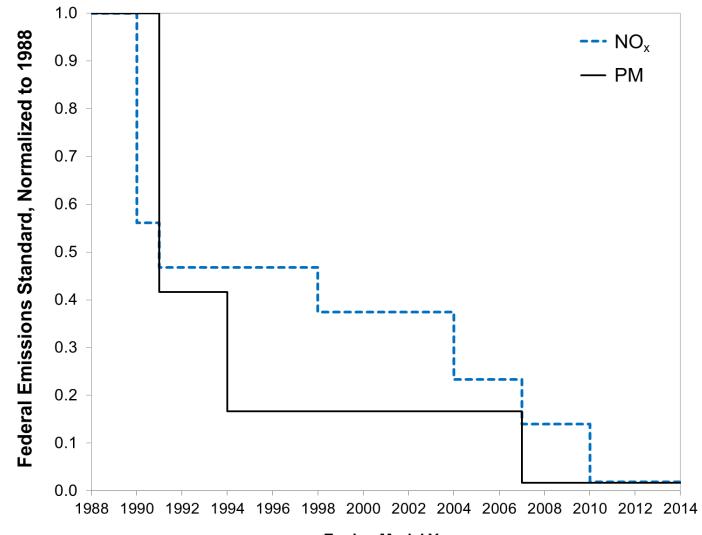
- Mobile sources (gasoline and diesel engines) are major emitters of NO<sub>x</sub>
  - Diesel contribution rising as emissions from gasoline engines and stationary sources are controlled
  - First round of diesel NO<sub>x</sub> control efforts in the 1990s failed to achieve expected emission reductions
- Use of diesel particle filters to control PM emissions has led to increased primary emissions of NO<sub>2</sub>

#### Trends in Fuel Use by On-Road Vehicles (From State and National Fuel Excise Tax Data)



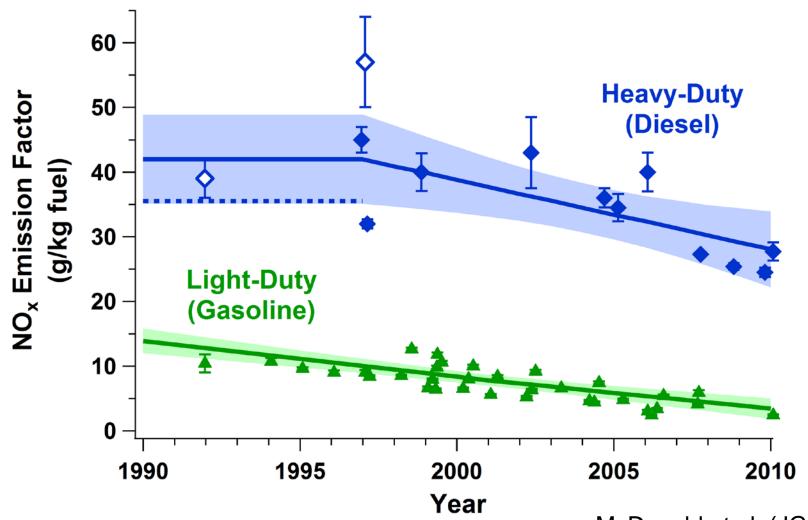
### **Heavy-Duty Diesel Emission Standards**

(For New On-Road Engines -- by Model Year)

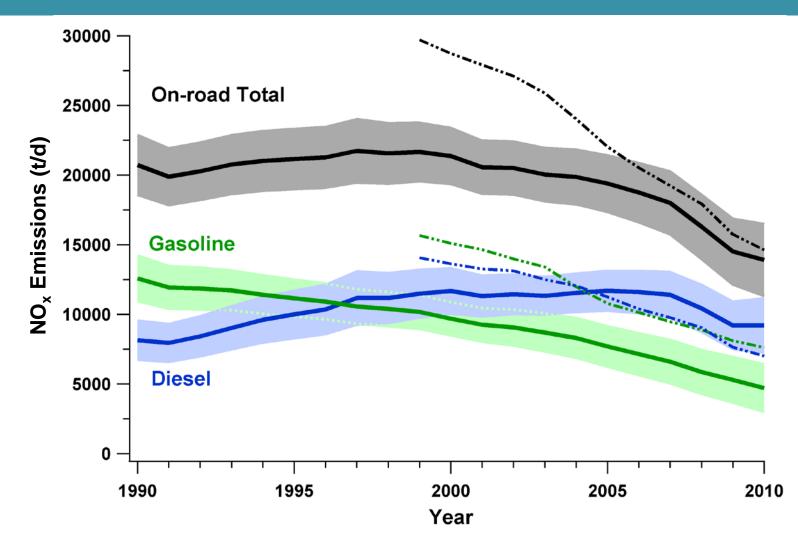


**Engine Model Year** 

#### **NO<sub>x</sub> Emission Factor Trends** Tunnel and Roadside Measurements of In-Use Emissions



### **National NO<sub>x</sub> Emission Trends** (On-Road Gasoline and Diesel Engines Only)



# National NO<sub>x</sub> Emission Trends

(Including Stationary and Off-Road Mobile Sources)

#### California Ports: Preview of Future Changes in Diesel Emissions

Accelerated retrofit / replacement of older diesel engines is required in California

Near-universal PM and NO<sub>x</sub> emission control required by 2016 and 2023, respectively

Early action item: drayage trucks in short-haul freight service to / from ports and rail yards "fixed" by 2013 SF-Oakland Bay Bridge

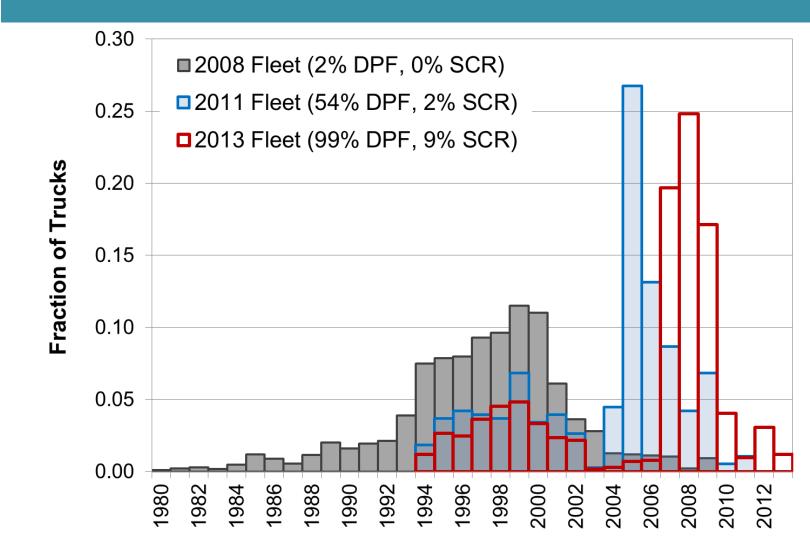
Port of Oakland

IP and BNSF Rail Yards 200

1

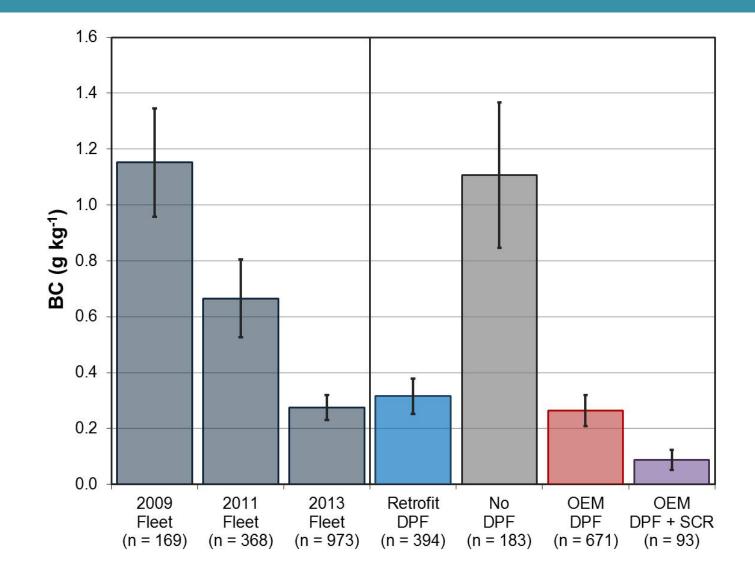


### **Port Truck Age Distribution**

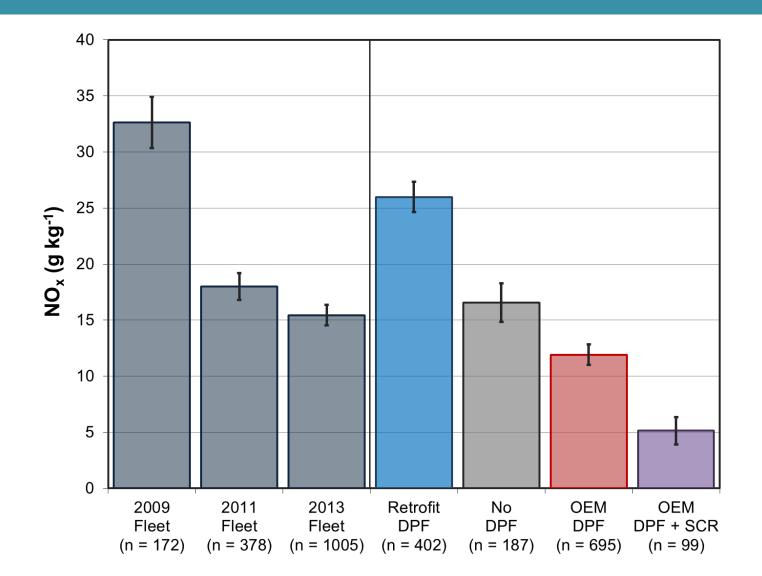


**Engine Model Year** 

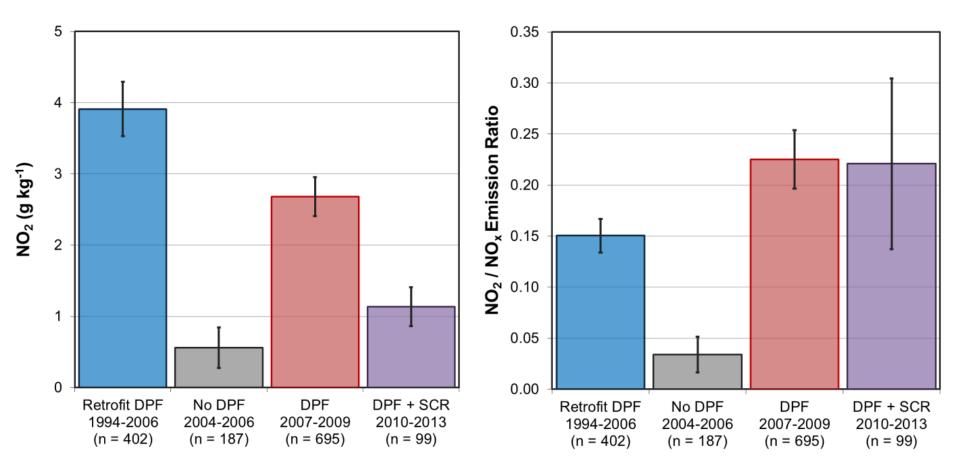
#### Black Carbon Emission Factors Decreased by 76 ± 22% between 2009 and 2013



#### NO<sub>x</sub> Emission Factors Decreased by $53 \pm 8\%$ between 2009 and 2013



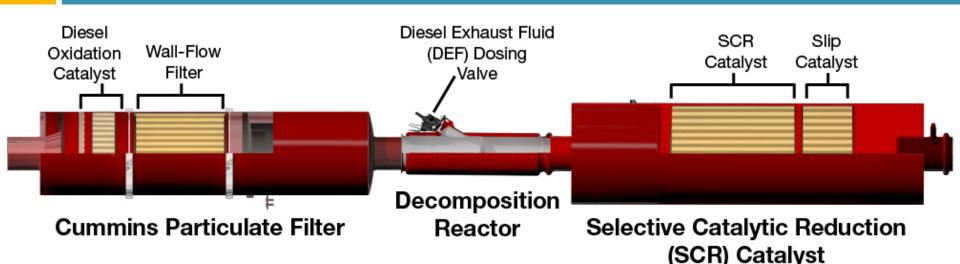
### NO<sub>2</sub> Emissions and NO<sub>2</sub>/NO<sub>x</sub> Ratio NO<sub>2</sub> increased from 3 to 18% of total NO<sub>x</sub> emissions



# Summary

- In summer, NO<sub>2</sub> appears to be mostly formed via atmospheric oxidation of NO rather than directly emitted
   Winter situation may be more affected by increased primary NO<sub>2</sub>
- Rapid changes in diesel emissions at Port of Oakland:
  - Black carbon (BC) decreased by 76 ± 22%
  - **D** NO<sub>x</sub> decreased by  $53 \pm 8\%$  (due to fleet modernization)
  - NO<sub>2</sub> increased from 3 to 18% of total NO<sub>x</sub> emissions (due to DPF)
- Advanced NO<sub>x</sub> controls (e.g., selective catalytic reduction) will help to mitigate increases in primary NO<sub>2</sub> emissions associated with widespread use of diesel particle filters

#### Diesel Particle Filter & Selective Catalytic Reduction (DPF) (SCR)



Used on 2007 & newer engines (DPF retrofits possible on older engines)

PM from engine exhaust trapped on filter

NO<sub>2</sub> oxidizes trapped carbon particles (this helps to regenerate the filter)

Used on 2010 & newer engines (SCR is difficult to add as a retrofit)

DEF = mixture of urea + water Urea converted to  $2 \text{ NH}_3 + \text{CO}_2$ 

 $NH_3$  reacts with  $NO_x$  to form  $N_2$