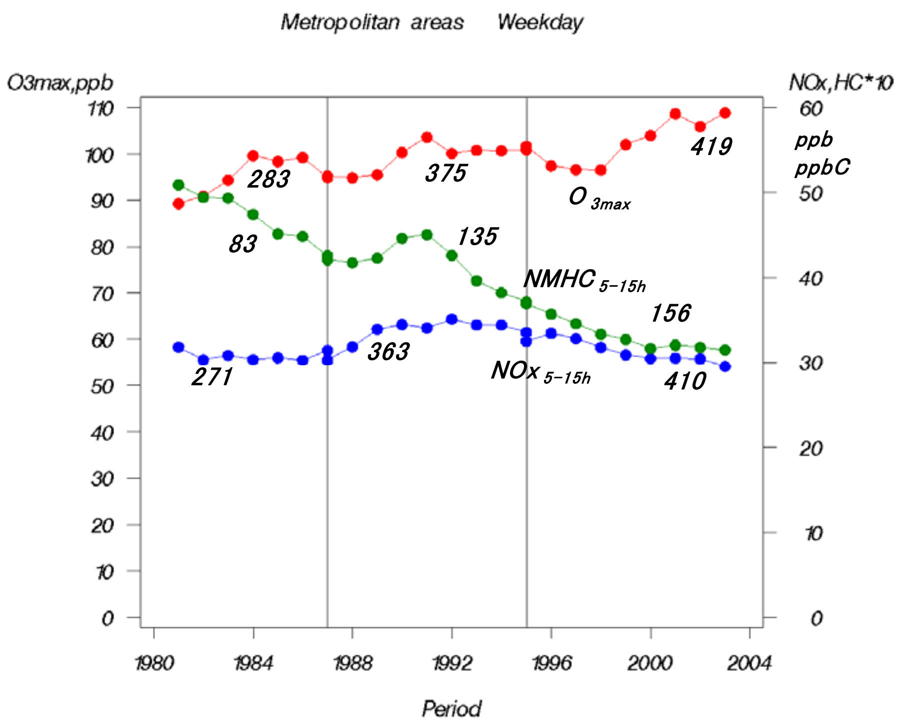
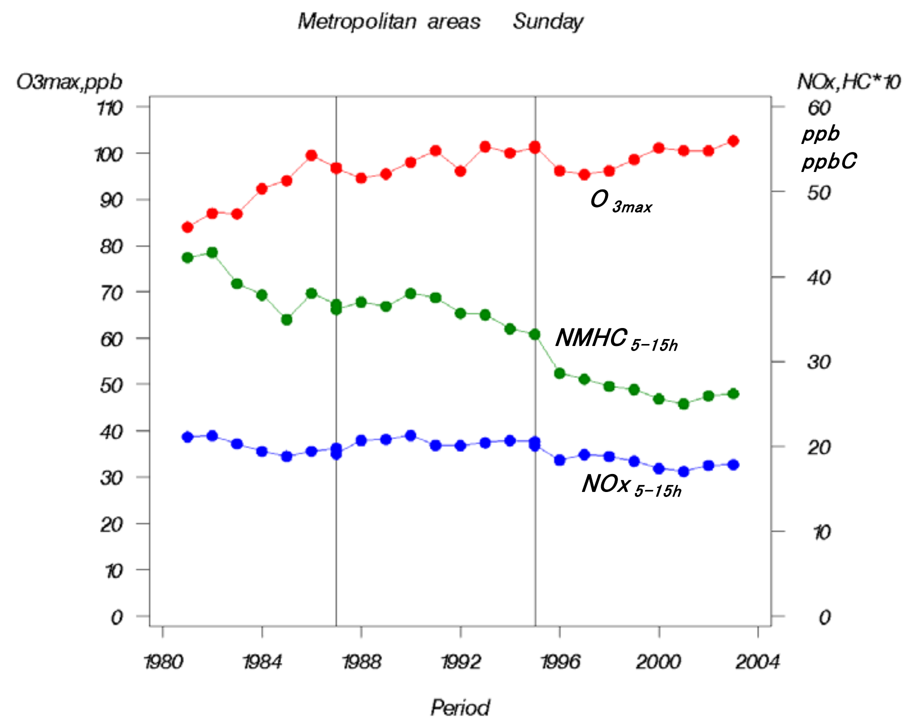
MICS-Asia, 10th Workshop
February 18-19, IIASA

A long term trend in VOC's photochemical reactivity in Japan

Akiyoshi Kannari/ Independent researcher,
Visiting researcher , NIES
Toshimasa Ohara/ National Institute for
Environmental Studies (NIES)

Recent problems on surface O_3 concentrations around mega-cities in Japan





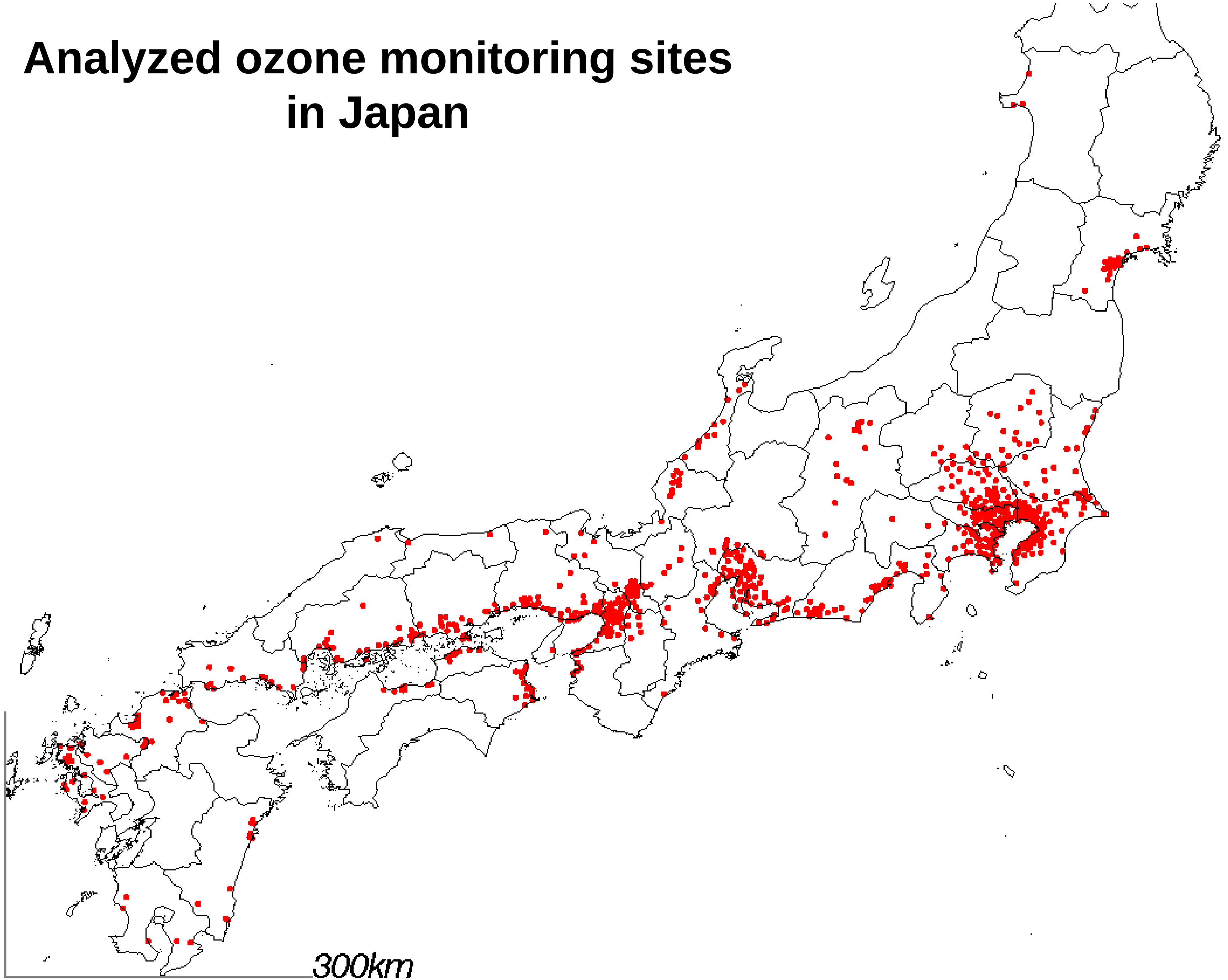
*1 Plots are three years moving averages

*2 Numbers in the figure indicate monitoring site numbers by time stage

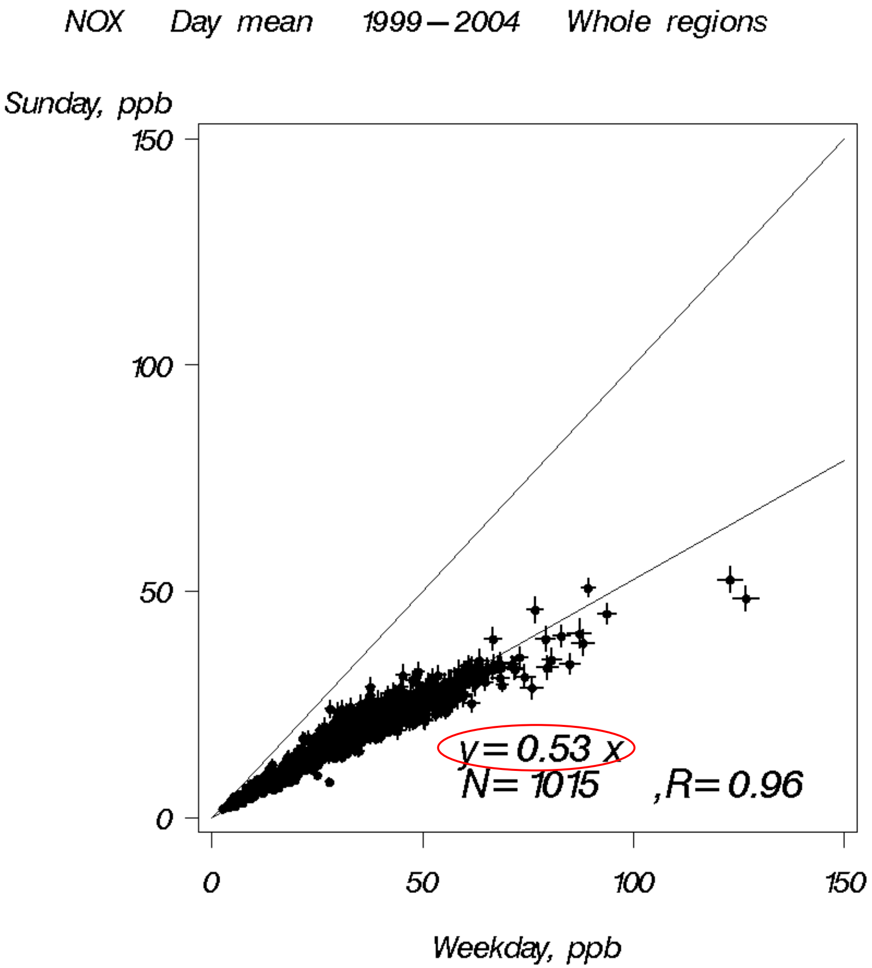
O3max=Highest 5% days averages of daily maximum one hour concentration
$NMHC_{5\text{-}15h}$, $NOx_{5\text{-}15h}$ =Composite means on the O_{3max} highest 5 % days

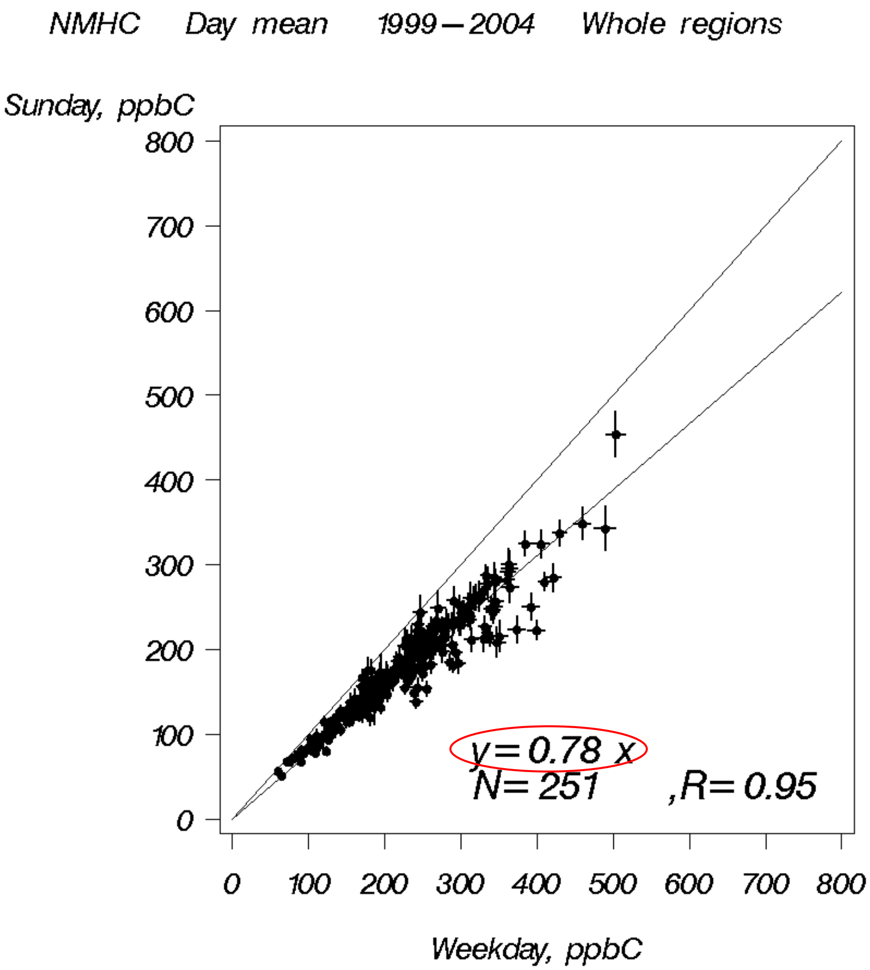
1. Analyses on the “ozone weekend effects”

Analyzed ozone monitoring sites in Japan



Weekend changes of NMHC and NOx concentrations



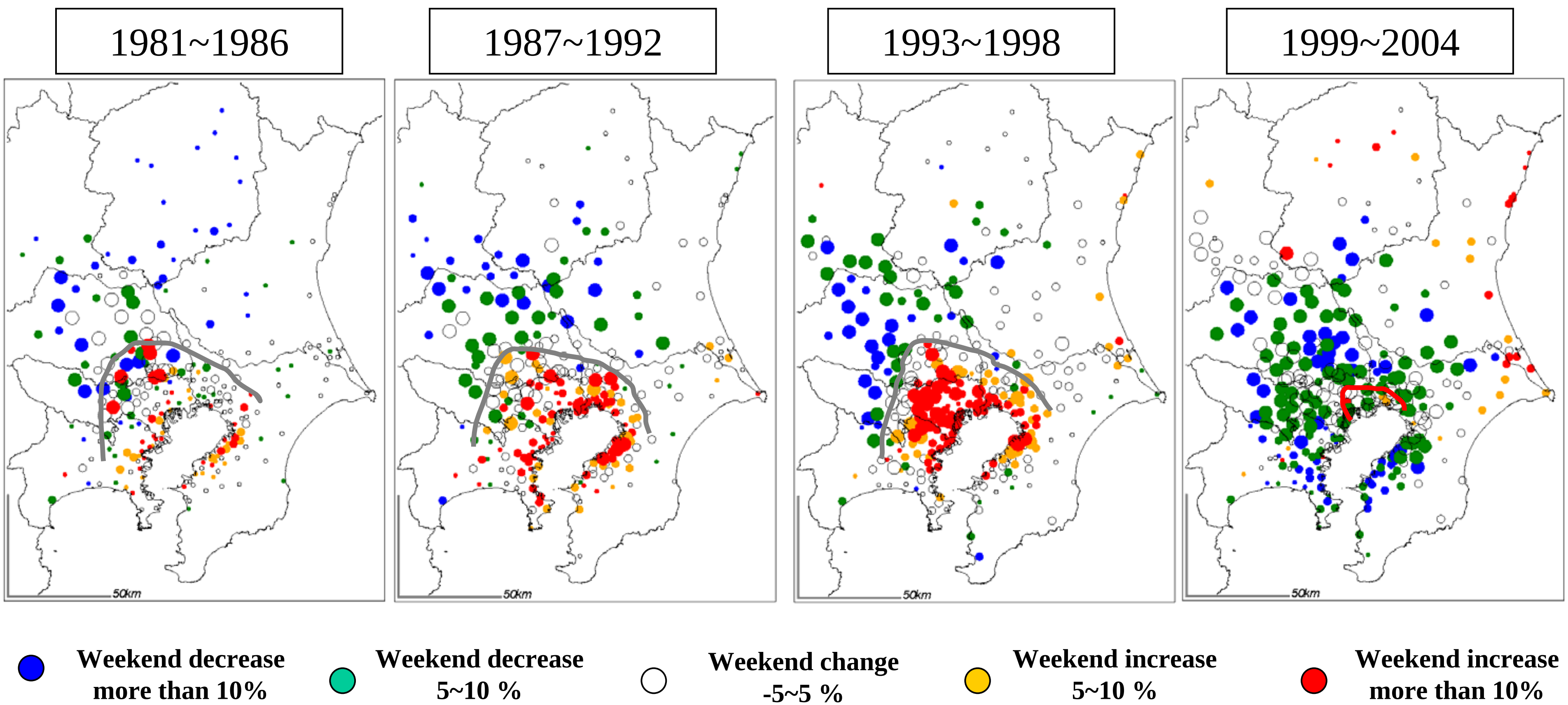


One of the results: Spatial changes of "ozone weekend effects"

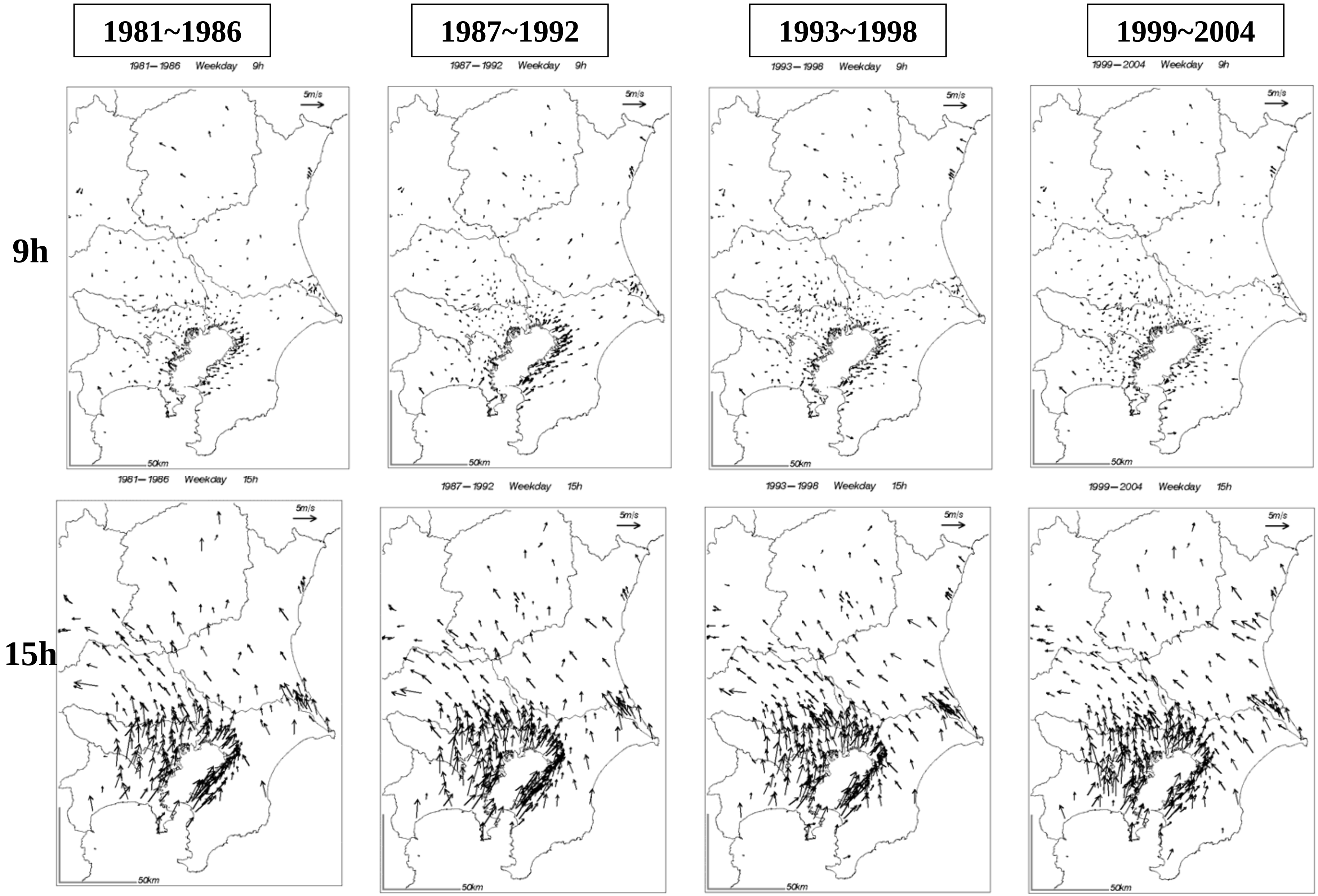
(An example in the Tokyo metropolitan area, highest 5% days averages)

1. ***Weekend increase*** at the source areas is changed to ***weekend decrease*** in the inland areas

2. Location of the boundary line changes by period

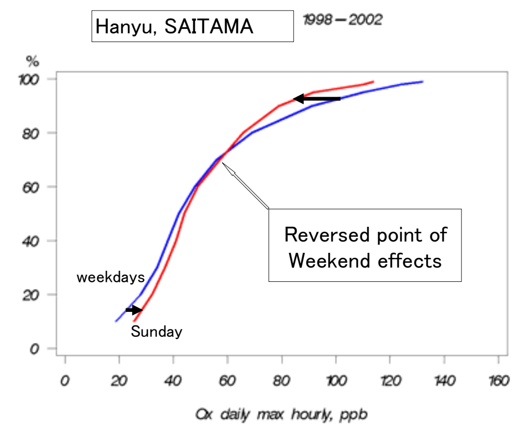


Surface wind system on the highest 5% days of O_{3max} is stable through the 4 periods



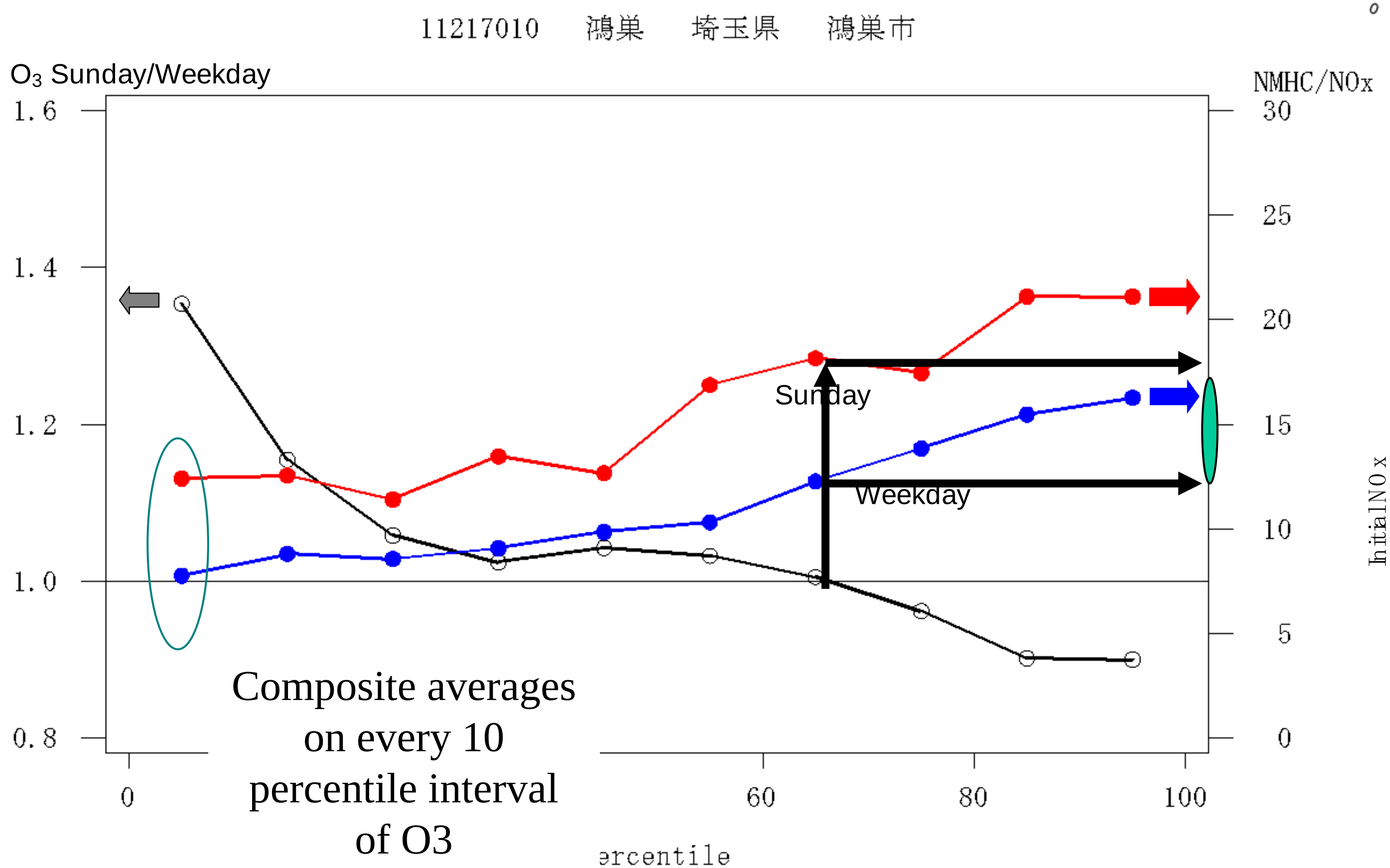
(Observed mean surface wind by time)

One of the results: Temporal changes of "ozone weekend effects"

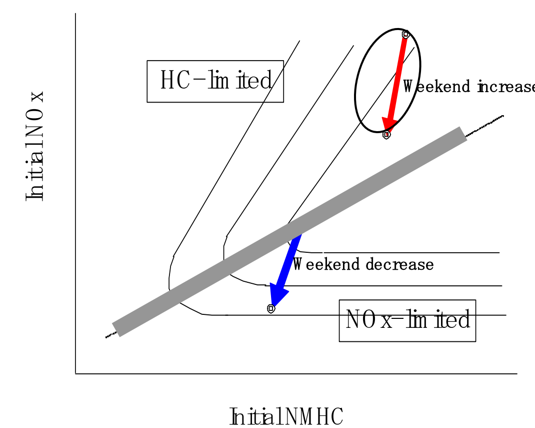


Boundary NMHC/NOx calculable under the following conditions

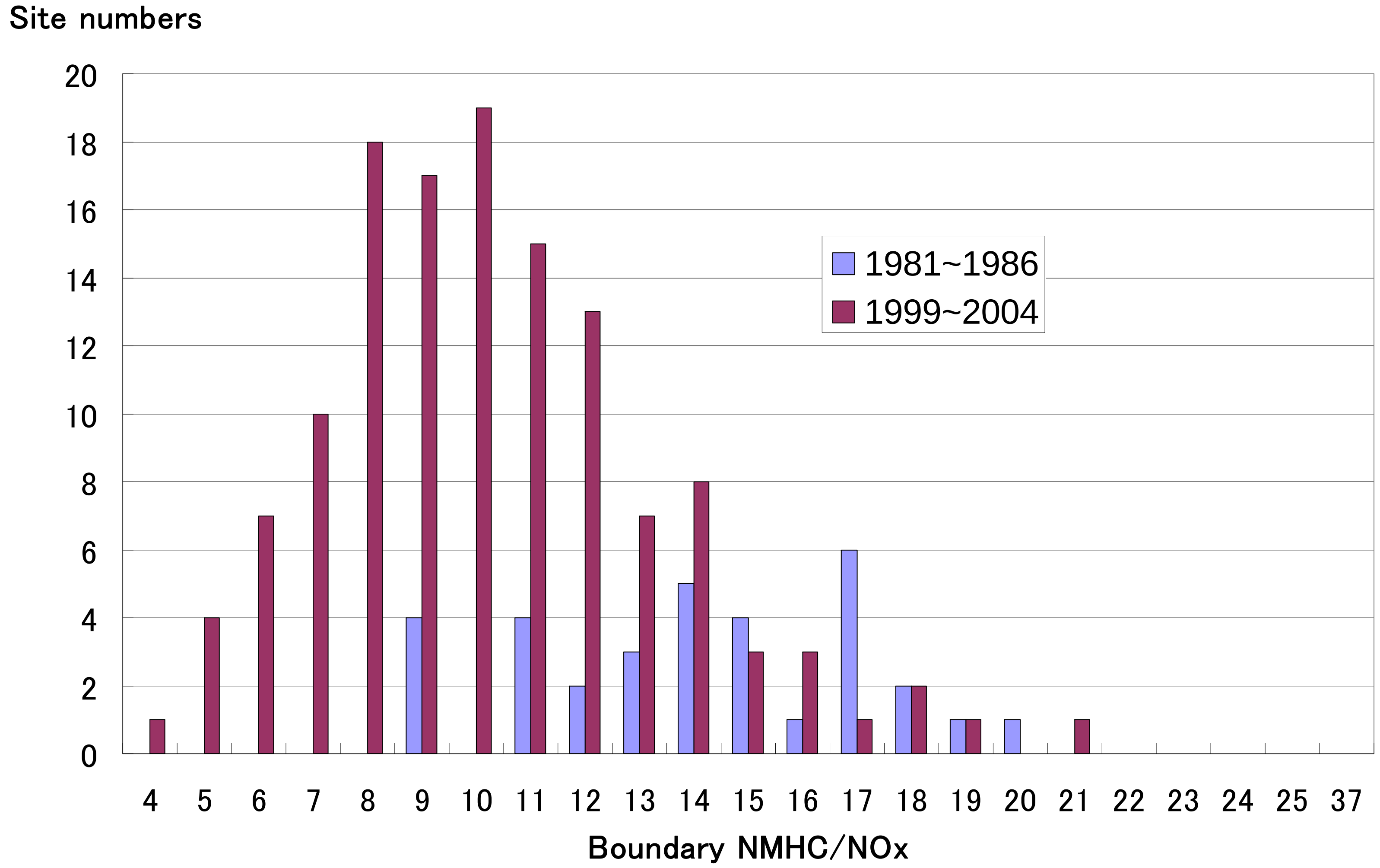
1.Ozone weekend change varies once from increase to decrease. 2. At that time, NMHC/NOx must increase along the percentile both weekdays and weekends. 3. NMHC/NOx on weekend must be higher than weekdays.



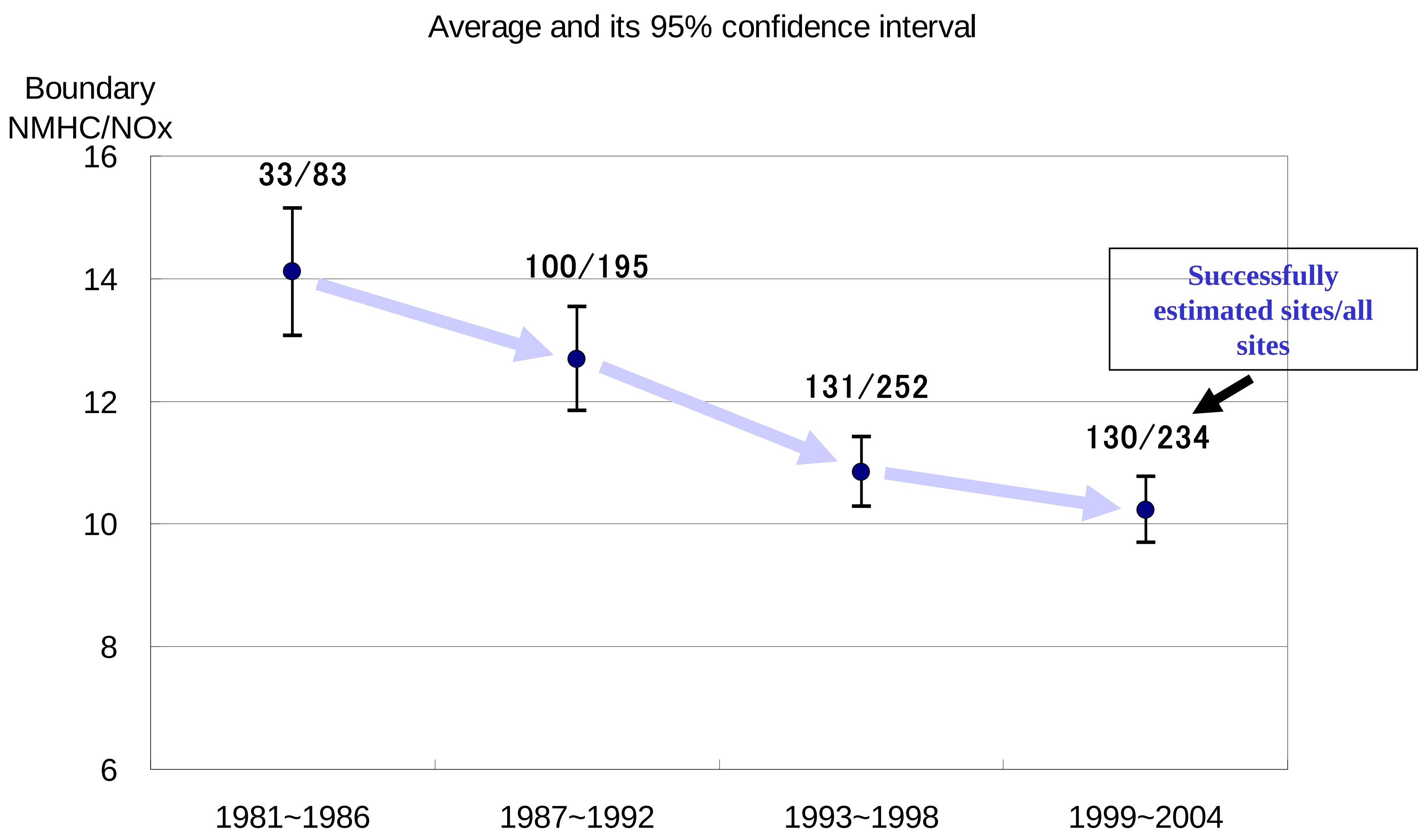
Composite averages on every 10 percentile interval of O3



Site numbers' distribution of estimated ***boundary NMHC/NOx*** in the two periods

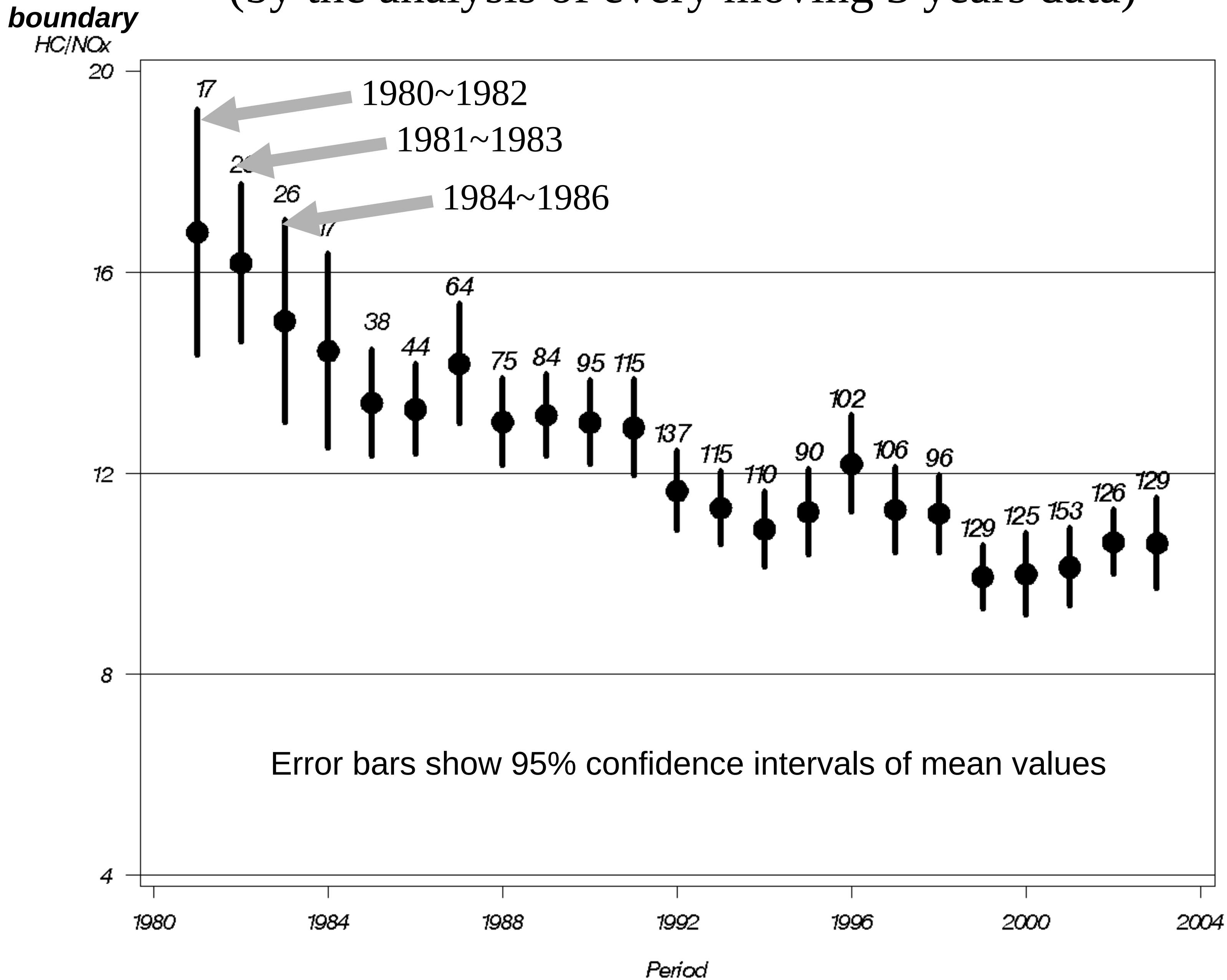


Estimation of boundary NMHC/NOx of O3 formation regimes
(for the whole domain in Japan)



Another estimates by the different time durations

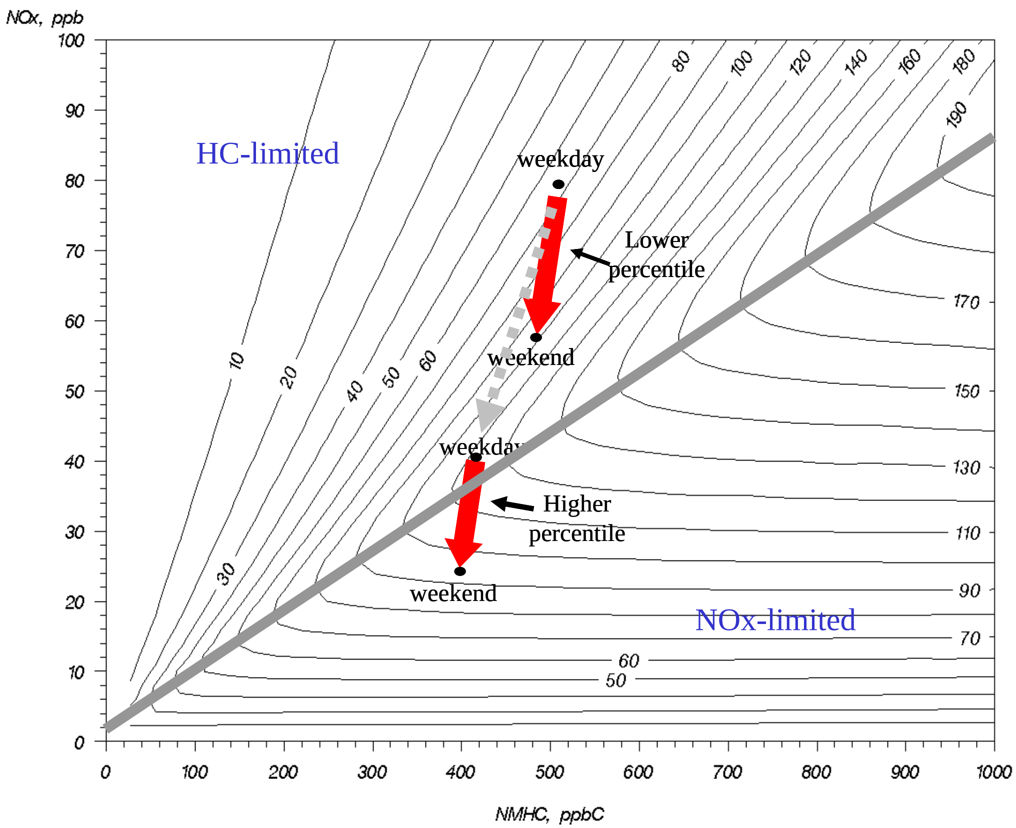
(by the analysis of every moving 3 years data)



Boundary of “direction of ozone weekend change”

↓ ↑ equivalent to

Boundary of “ozone formation regimes”



2. Consideration of ‘boundary’ using ozone isopleth diagrams

Estimation of ozone isopleth diagrams

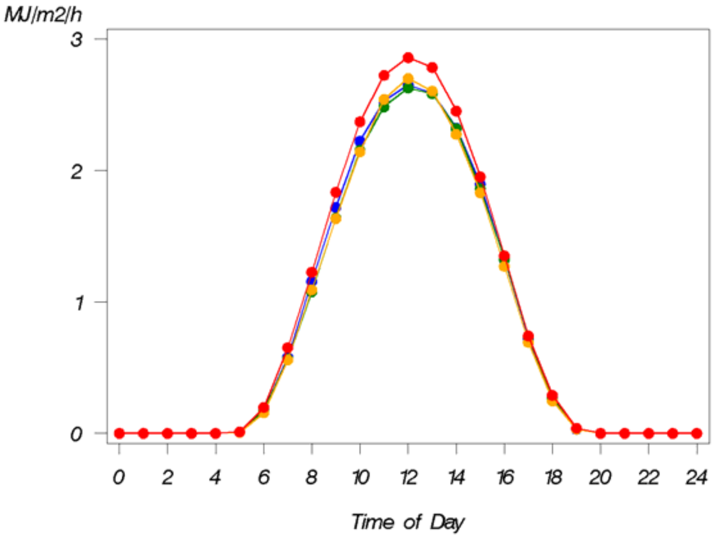
- Numerical simulation based on the CB4 chemical mechanism
- Giving NMHC, NOx initial concentration to closed box system without emission, ventilation and deposition
- Solar radiation, Temperature, Humidity

→ mean diurnal variation in the highest 5 % days of O_{3max}

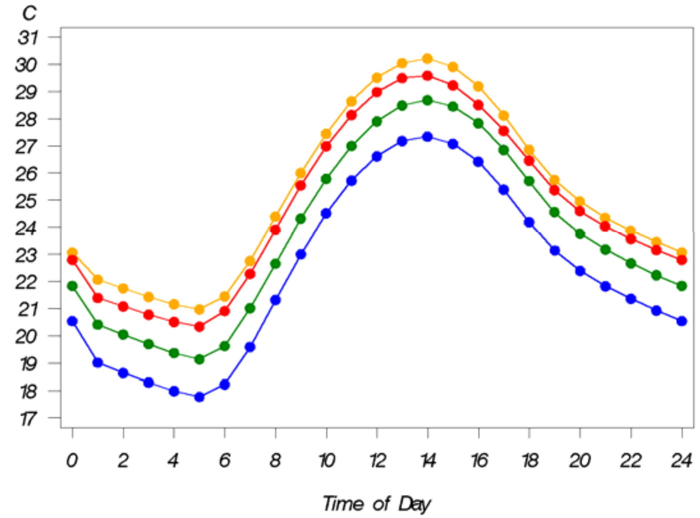
- Calculate a peak ozone concentration during 5-19h
- 50 x 50 cases of NMHC, NOx initial concentrations for a diagram

Meteorological condition on the highest 5 % days of O_{3max} in the Tokyo metropolitan area

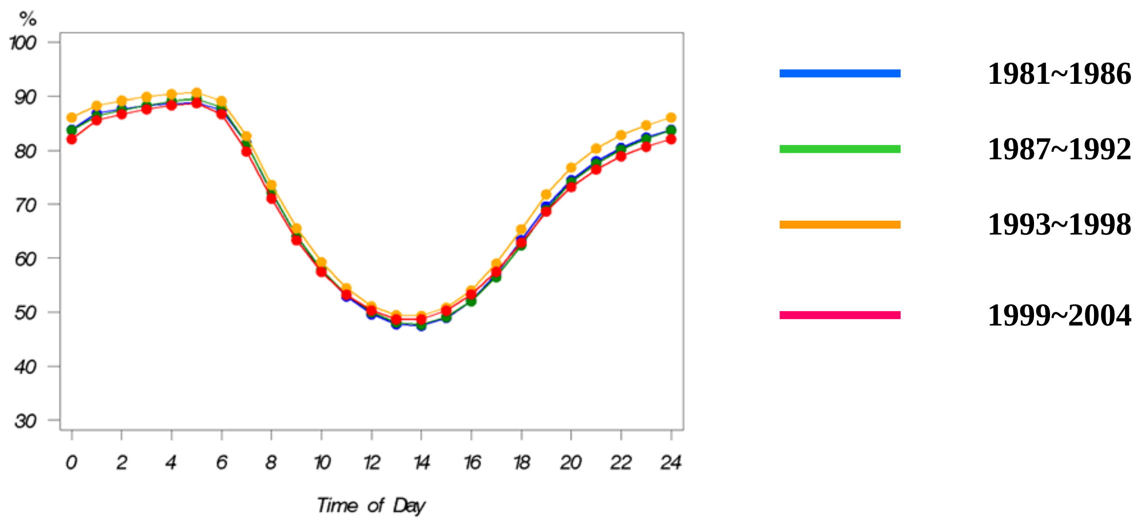
Solar radiation



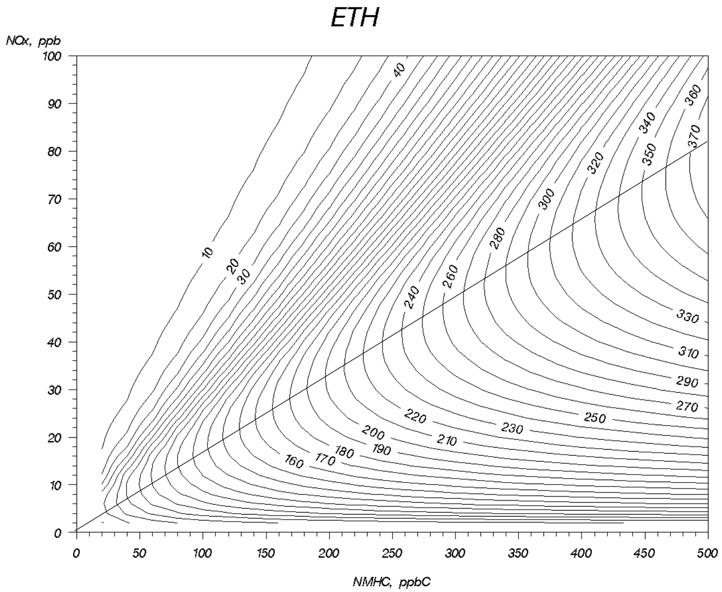
Temperature

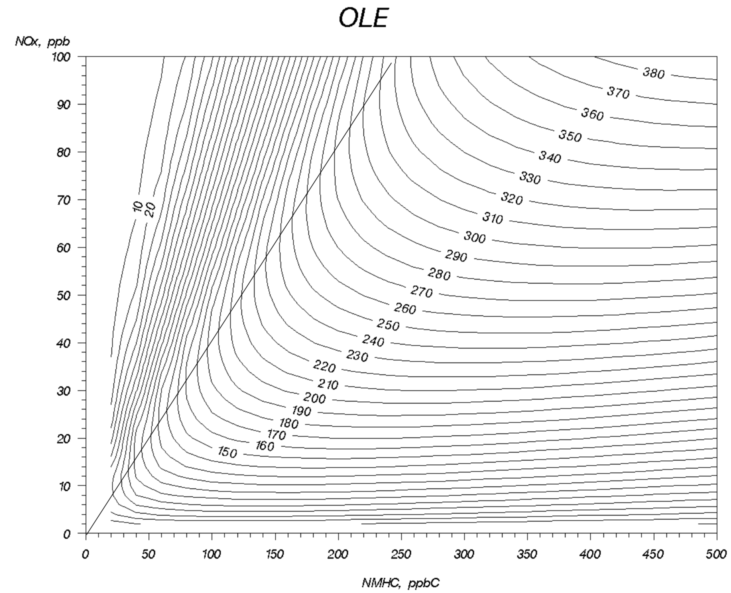


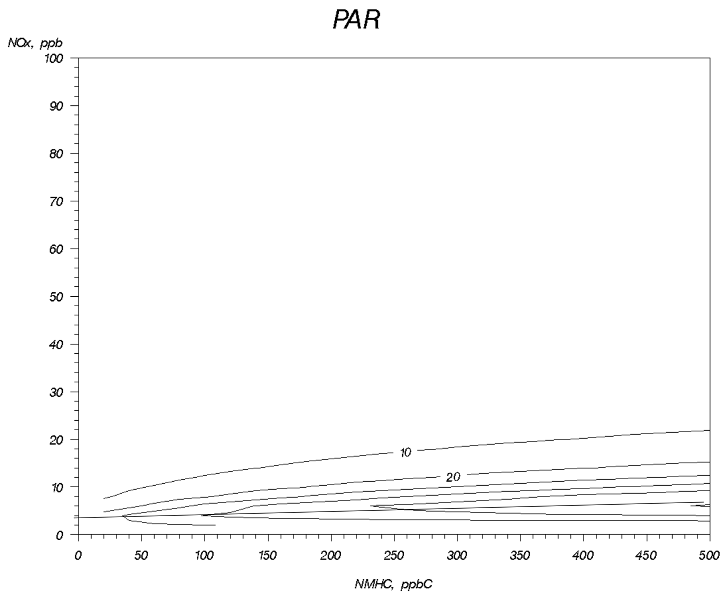
Humidity

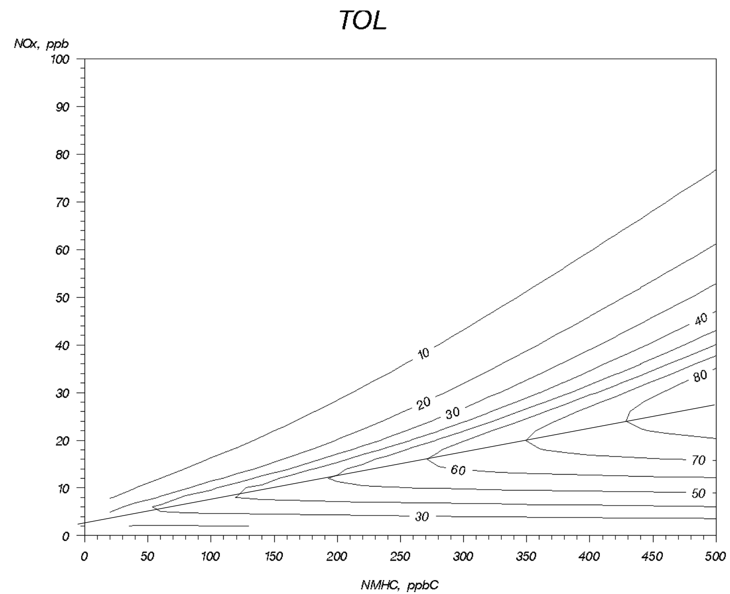


O_{3max} isopleth diagrams obtained from the single CB4 lumped species simulations (1)

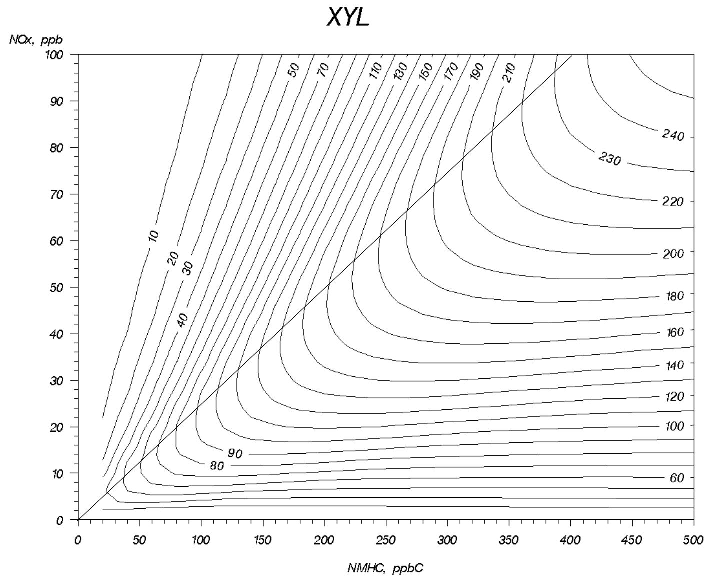


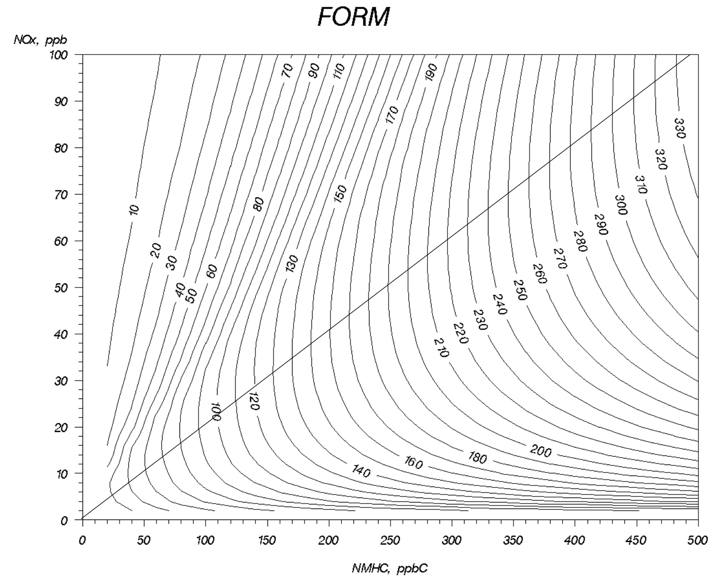


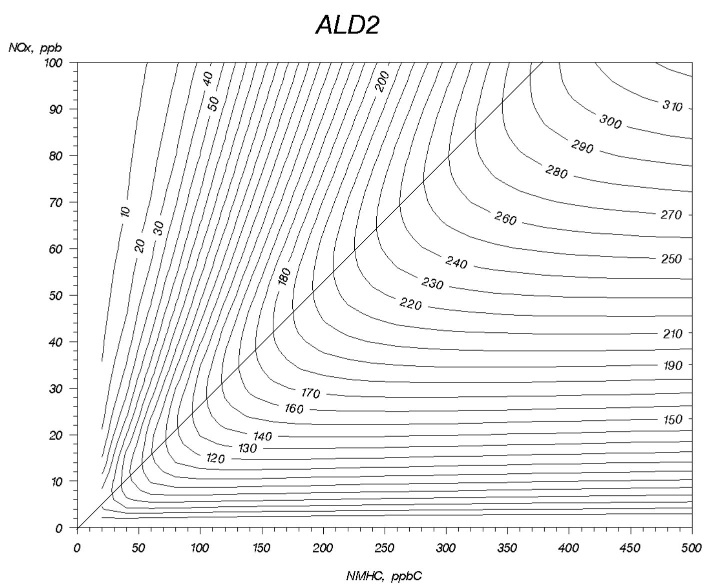


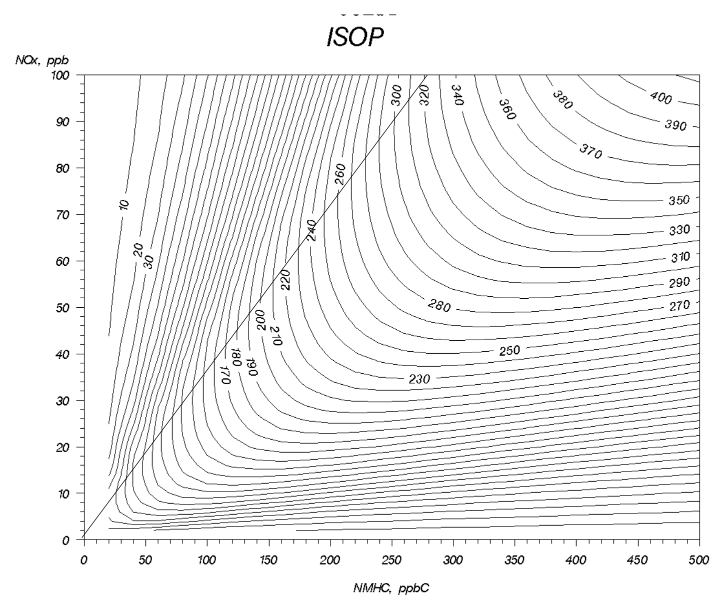


O_{3max} isopleth diagrams obtained from the single CB4 lumped species simulations (2)



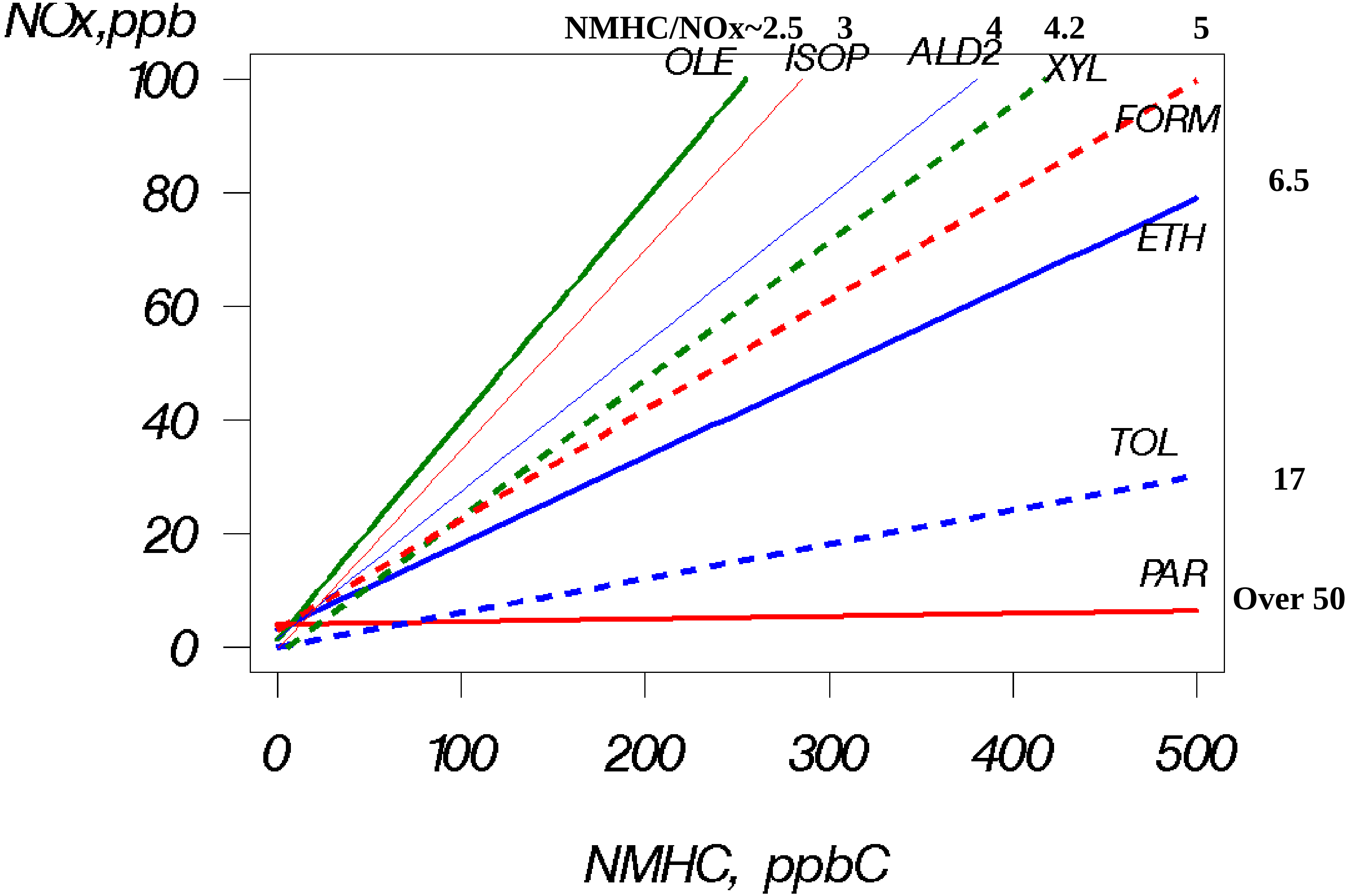




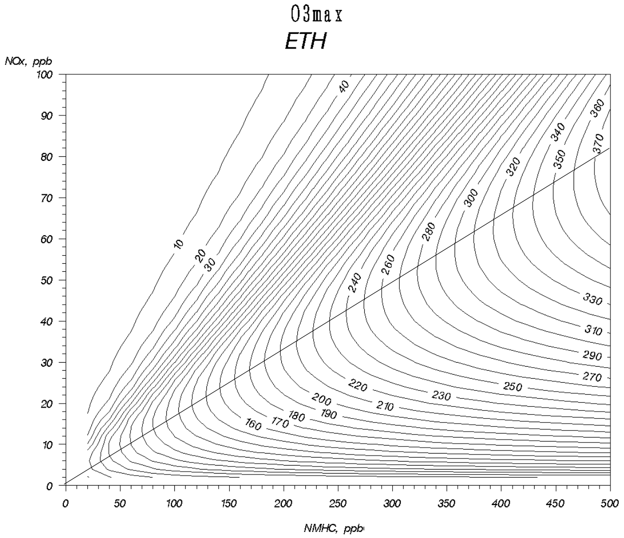


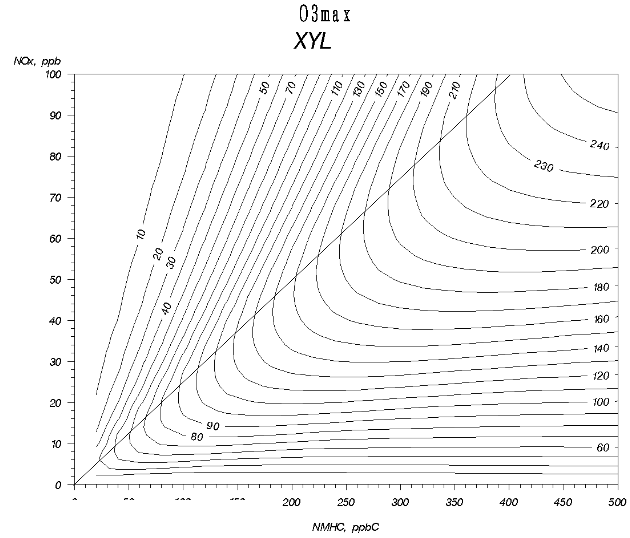
Boundary NMHC/NOx of ozone formation regimes for CB4 lumped species

More reactive species has lower boundary NMHC/NOx

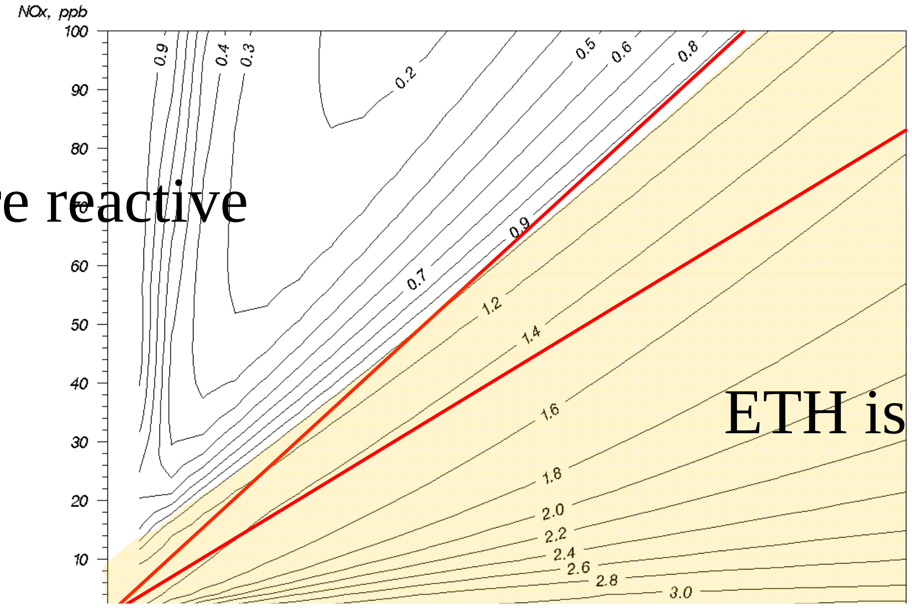


An example of comparison on the relative reactivity of lumped species





ETH/XYL

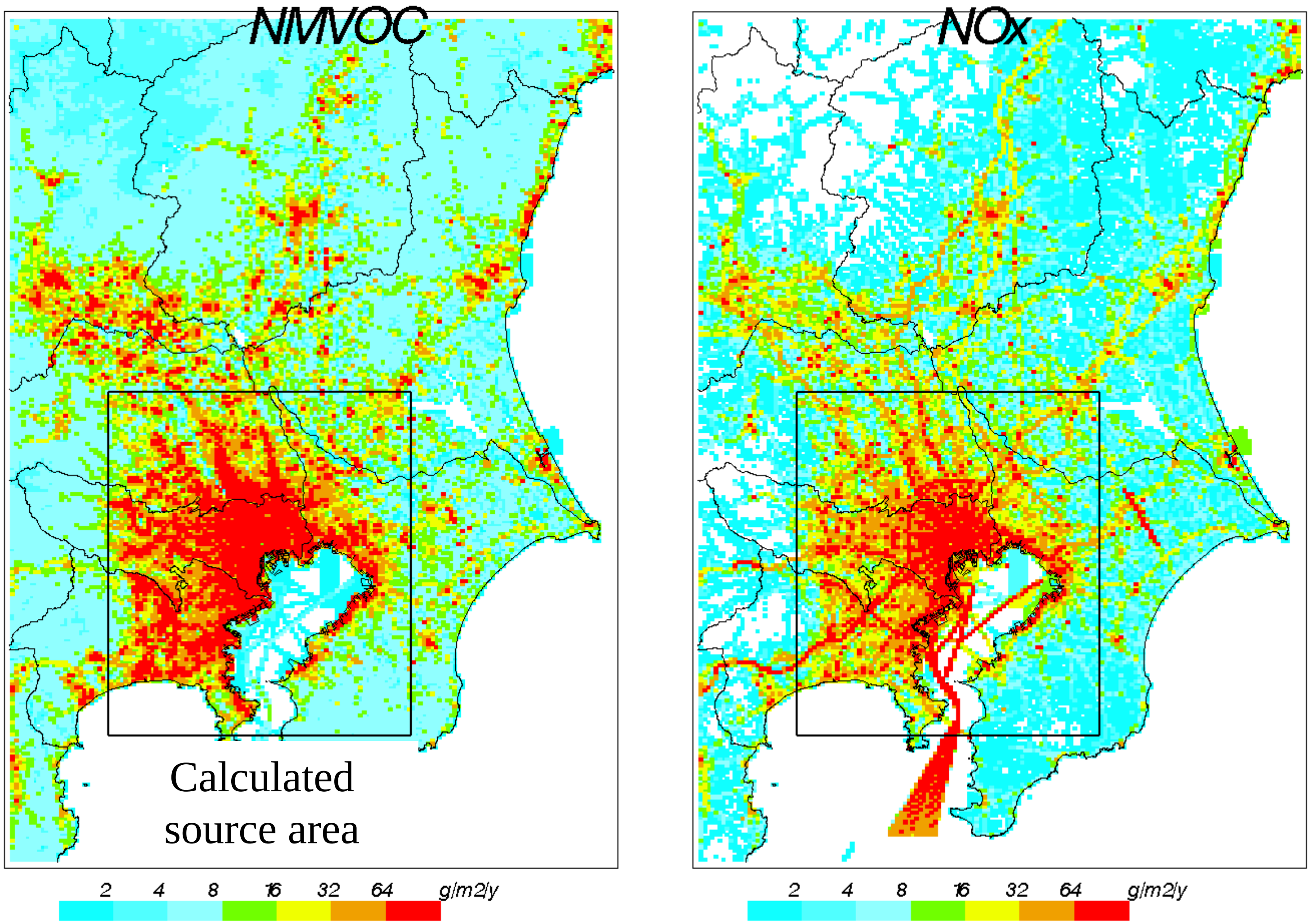


XYL is more reactive

ETH is more reactive

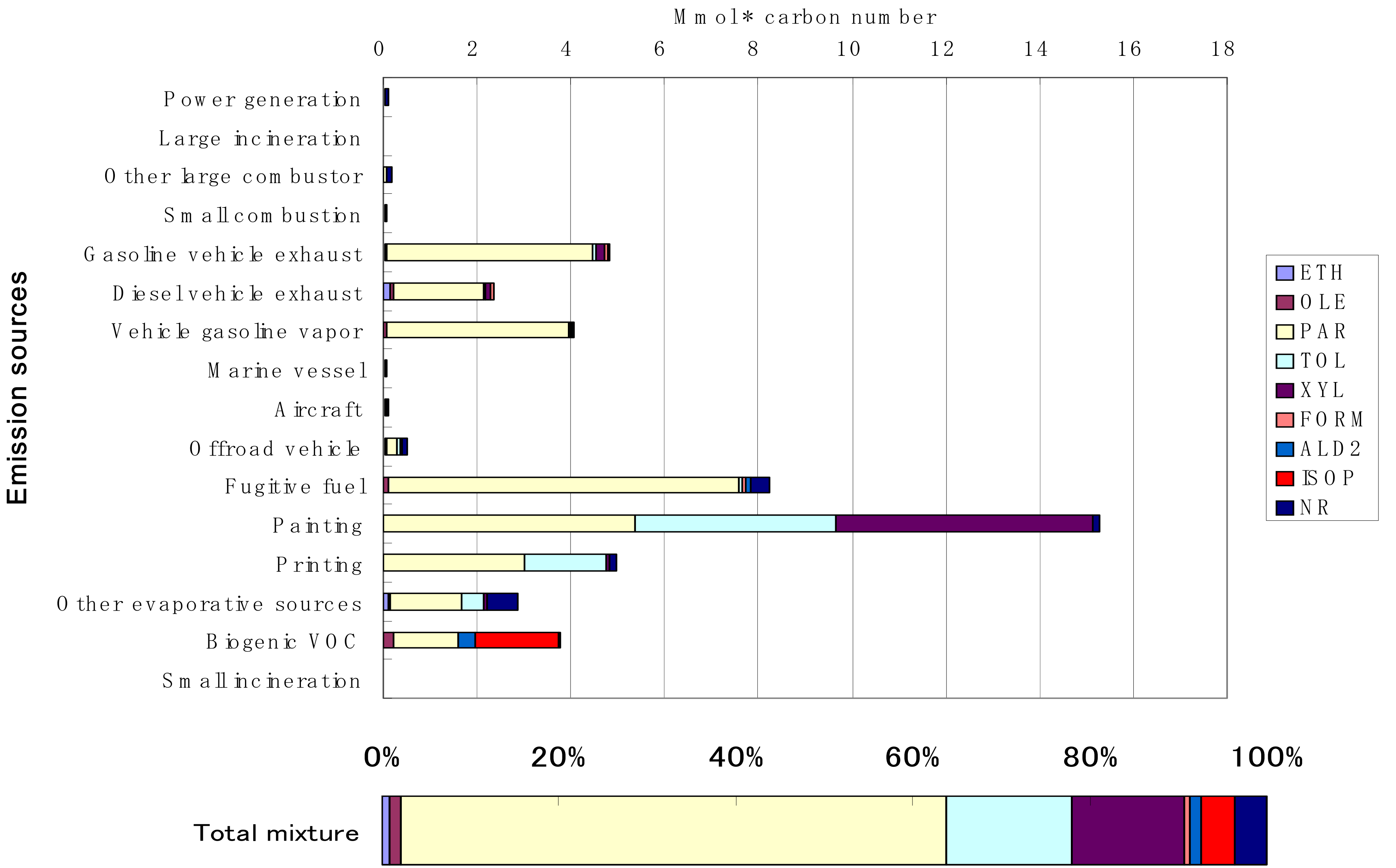
Relative reactivity is different in different regime position

Geographical distribution of NOx, NMHC daytime emissions in August, 2000



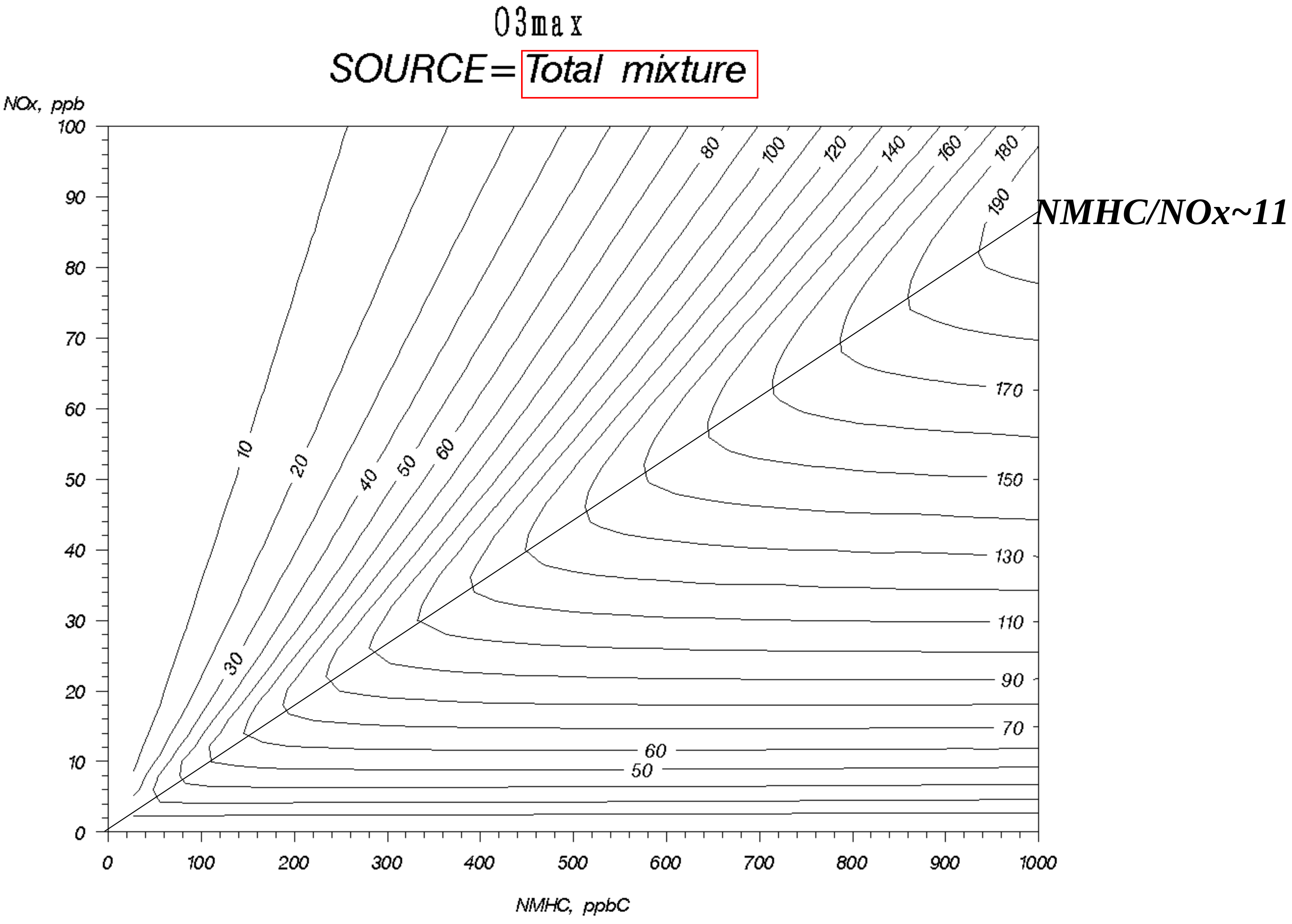
(EAGrid2000-JAPAN)

VOC composition (summary) in the Tokyo metropolitan source area in August, 2000 (Day time emissions from 5 to 15 o'clock)



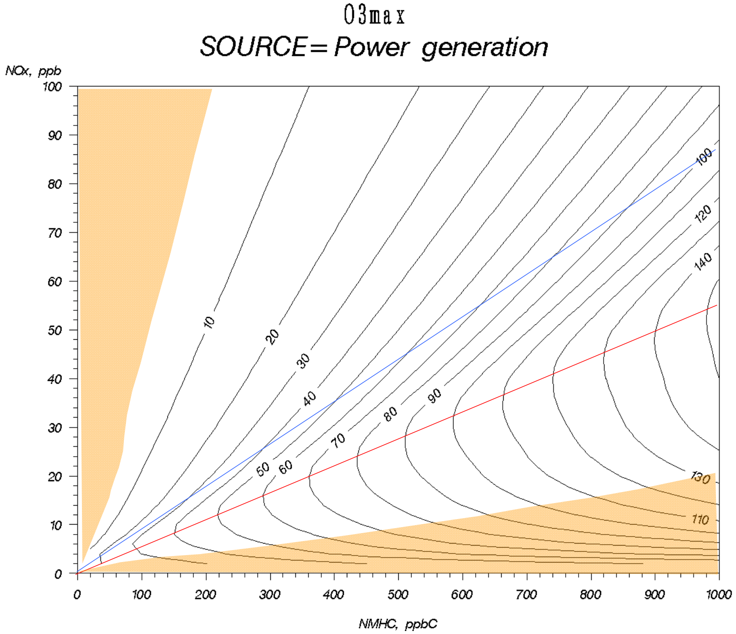
(Estimated from the EAGrid2000-JAPAN Emission database using the SPECIATE, JCAP composition profiles and the governmental solvent data)

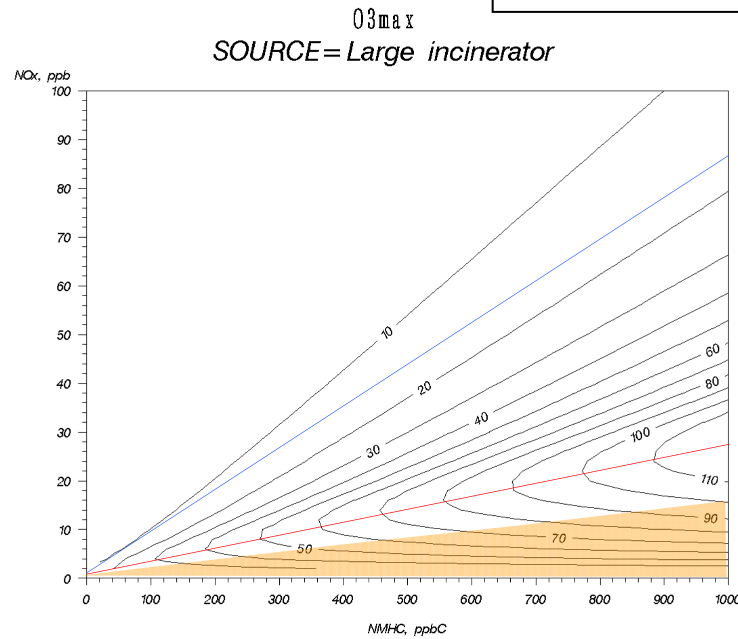
Ozone isopleth diagram based on the really emitted VOC composition in the Tokyo metropolitan area, August, 2000

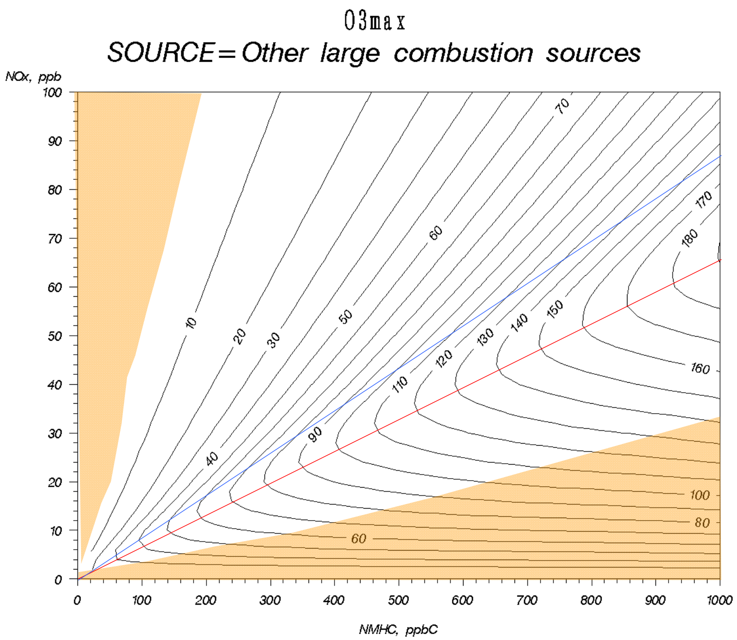


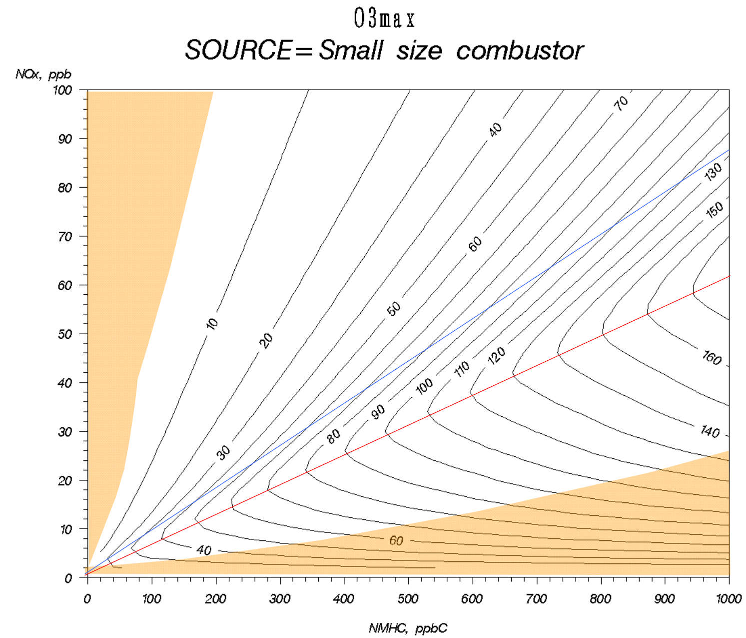
Source specific O_{3max} isopleth diagrams based on the VOC composition by source (1)

Shaded area= more reactive than the 'total mixture'



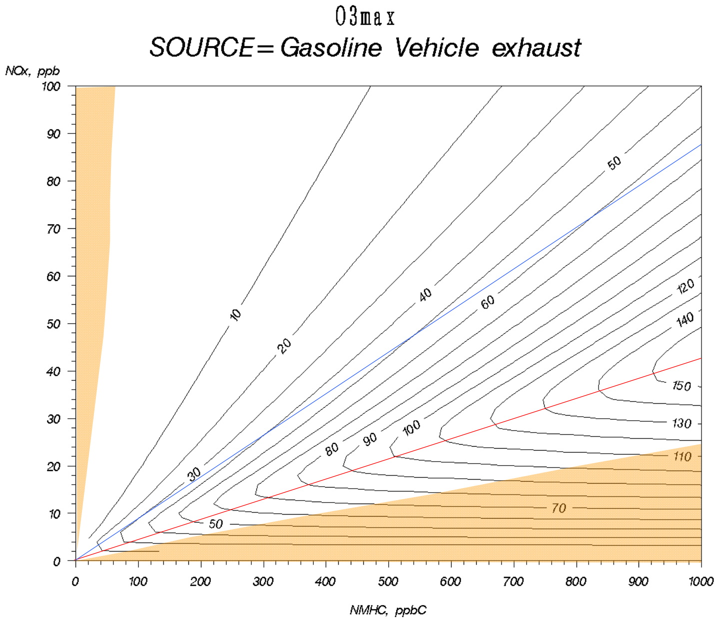


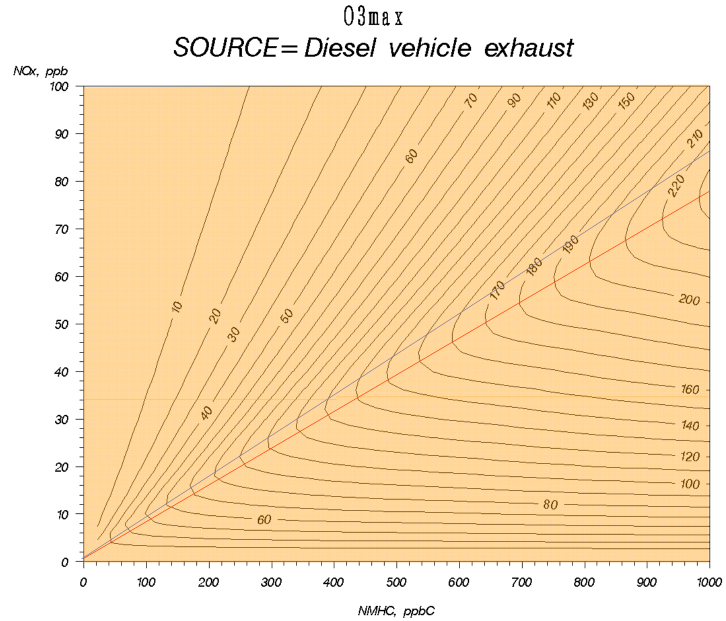


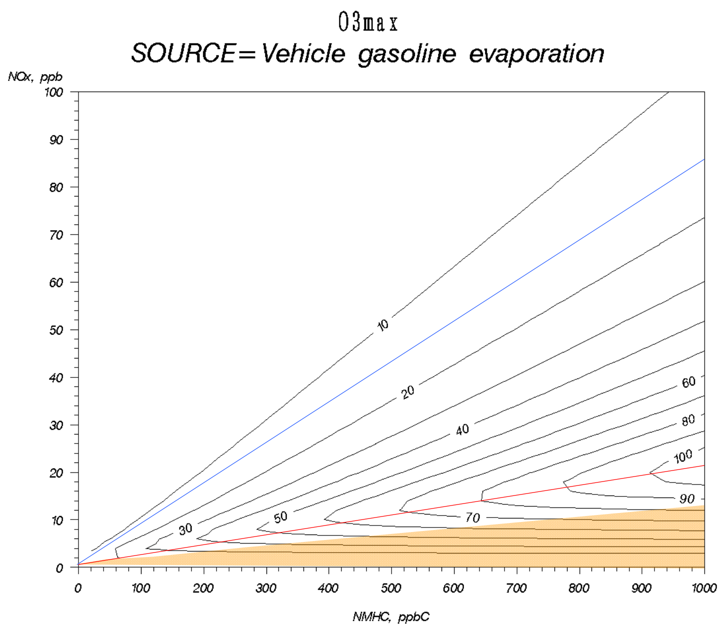


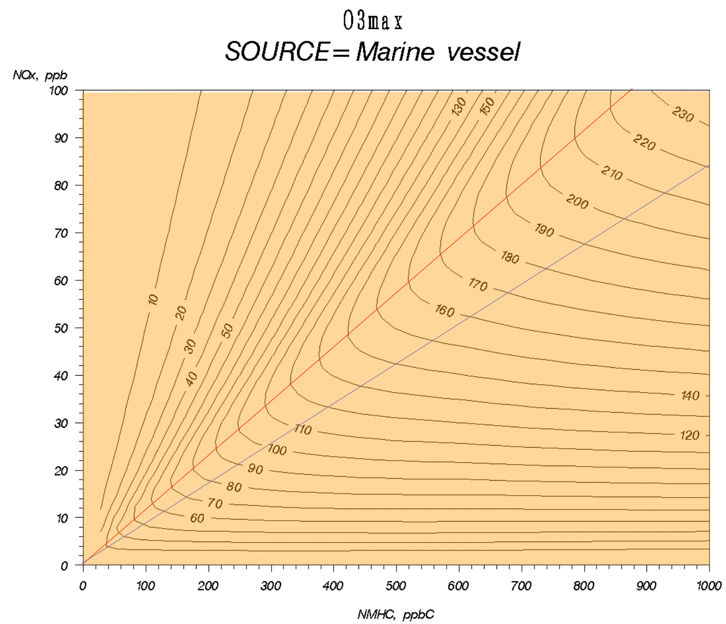
Source specific O_{3max} isopleth diagrams based on the VOC composition by source (2)

Shaded area= more reactive than the 'total mixture'



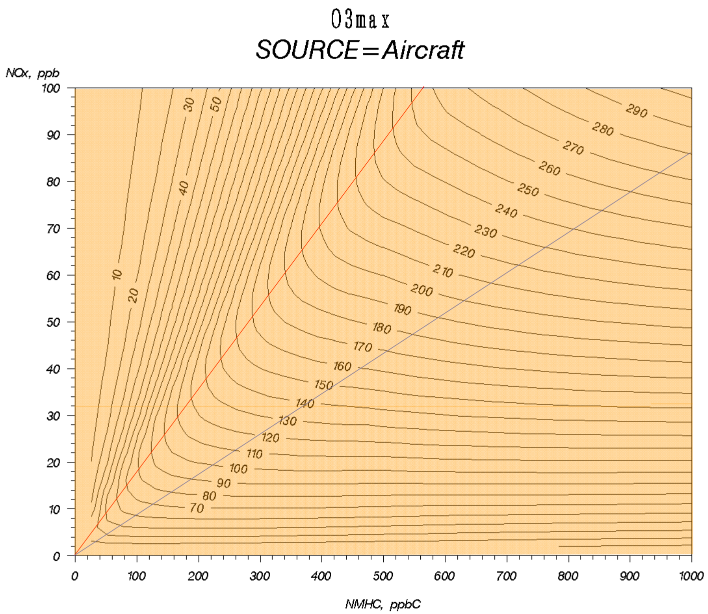


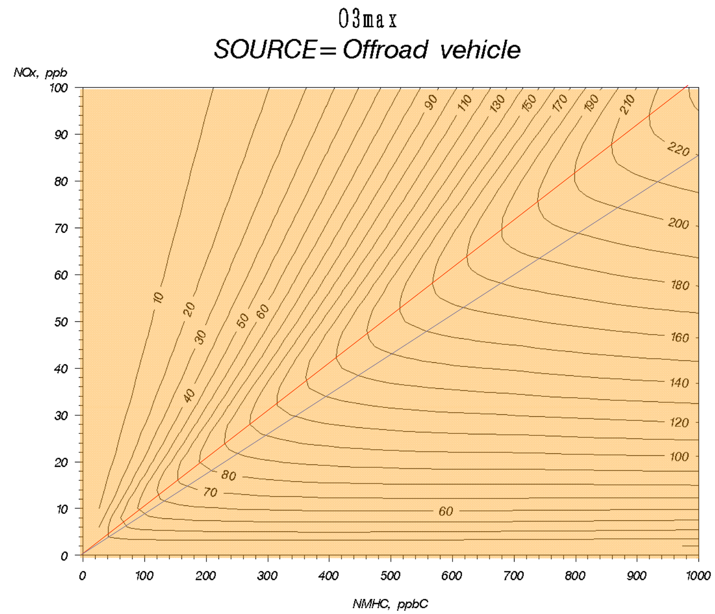


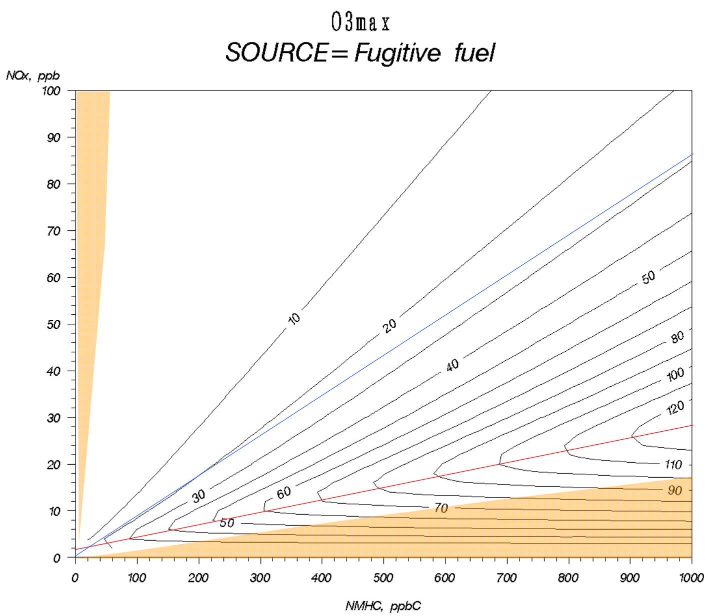


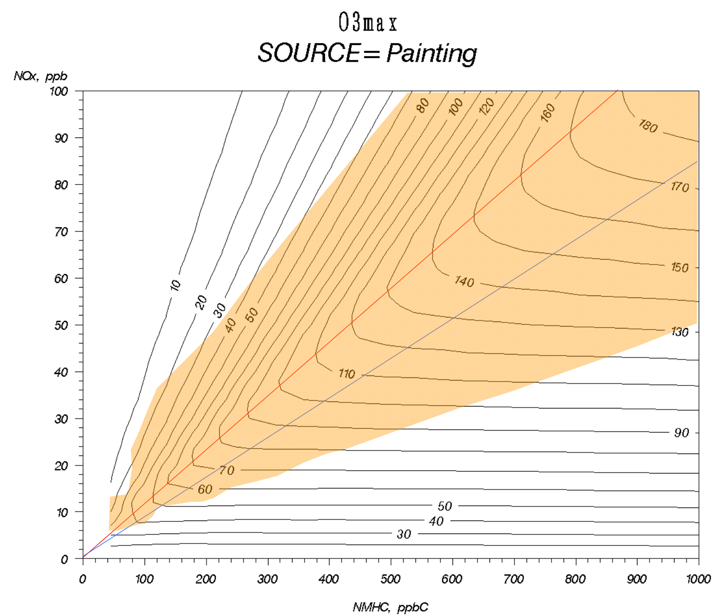
Source specific O_{3max} isopleth diagrams based on the VOC composition by source (3)

Shaded area= more reactive than the 'total mixture'



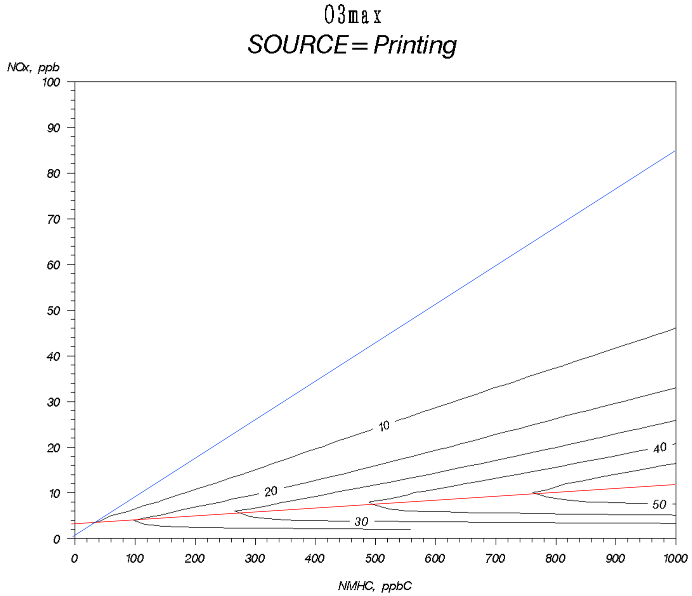


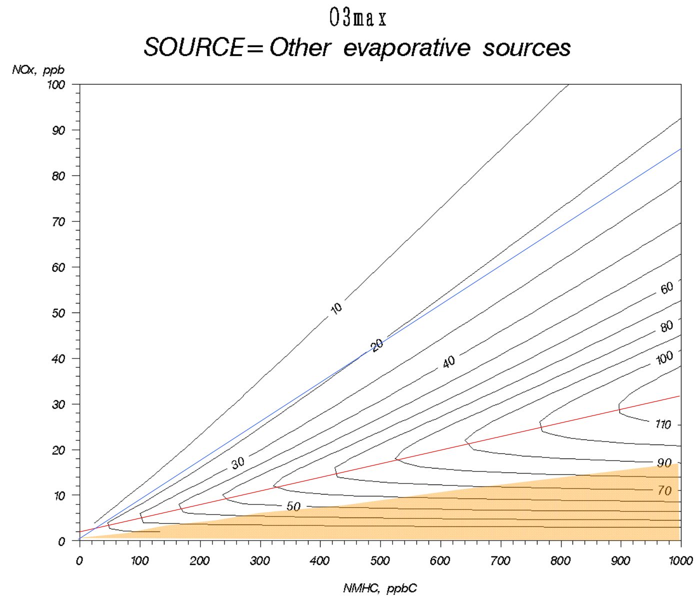


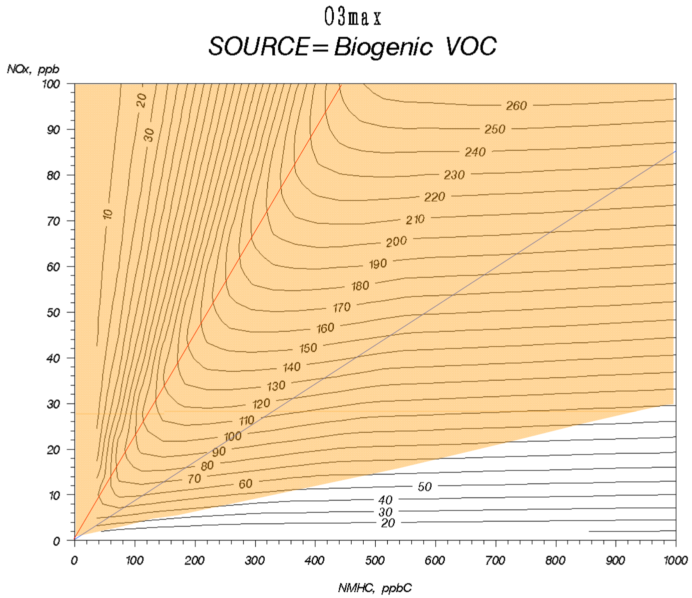


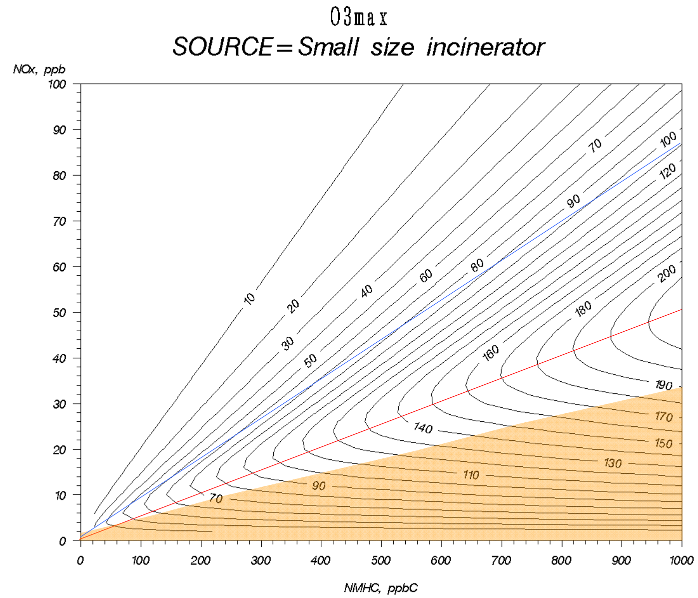
Source specific O_{3max} isopleth diagrams based on the VOC composition by source (4)

Shaded area= more reactive than the 'total mixture'



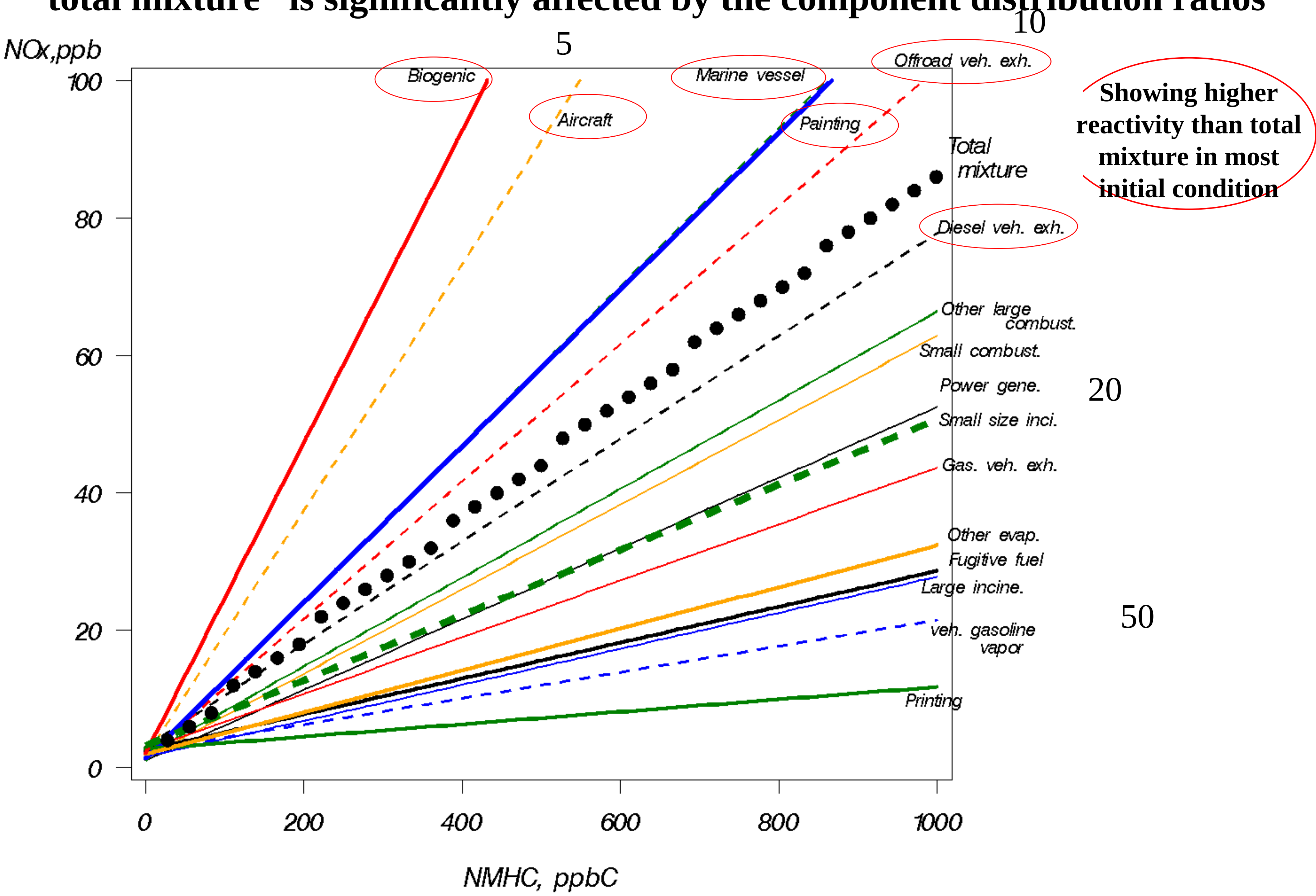






Comparison of boundary NMHC/NOx between the sources

"total mixture" is significantly affected by the component distribution ratios



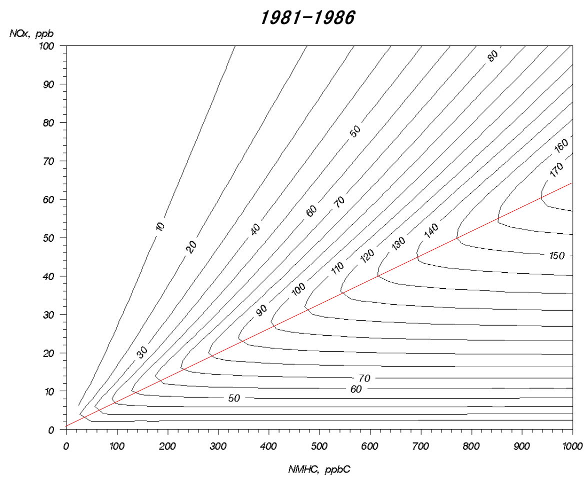
3. Cause of the trends

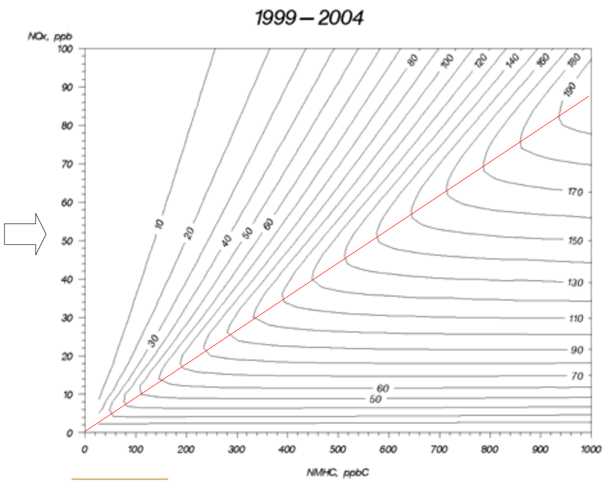
(1) Cause of NMHC decrease
(2) Cause of decreasing boundary NMHC/NOx of ozone formation regime

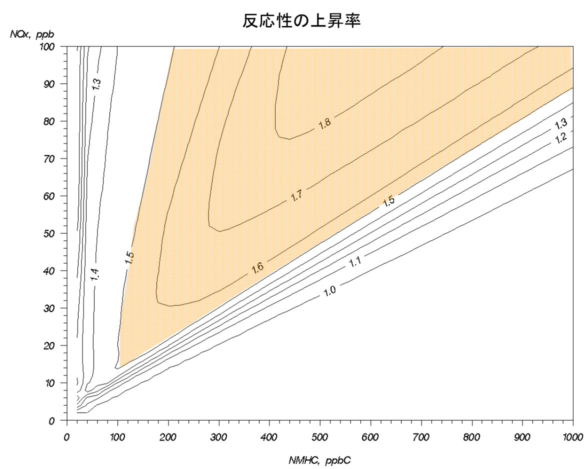
Hypothesis 1. Changes on vehicles emissions

80% reduction of gasoline exhaust and vapor
50% growth of diesel exhaust

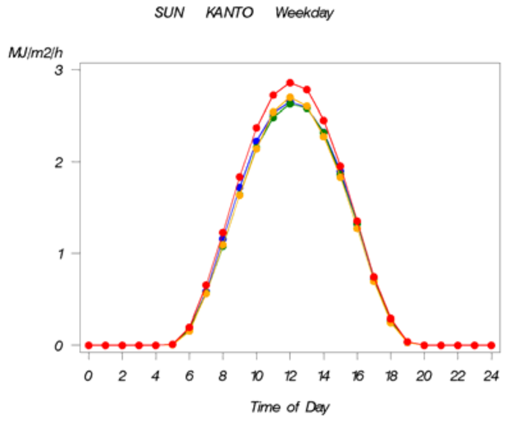
Reactivity→ 1.8 times, max
NMHC concentration→0.6 times

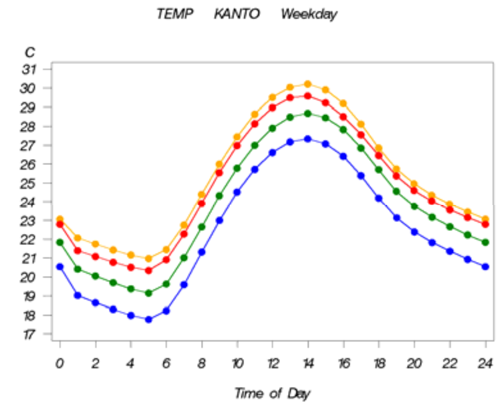






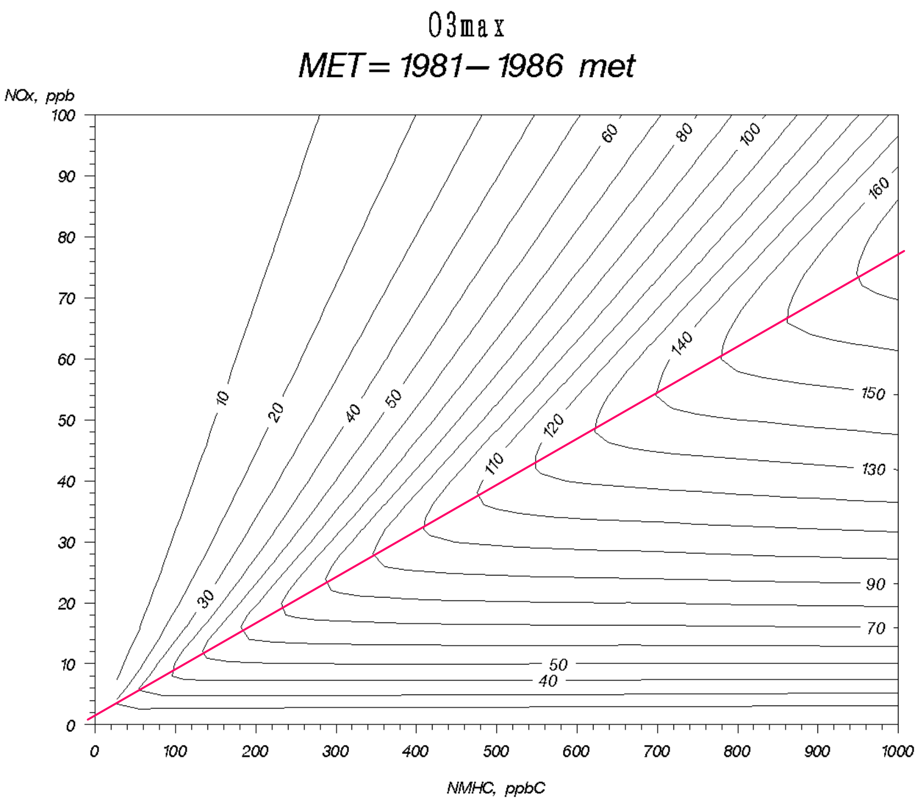
Hypothesis 2. Change of meteorological condition

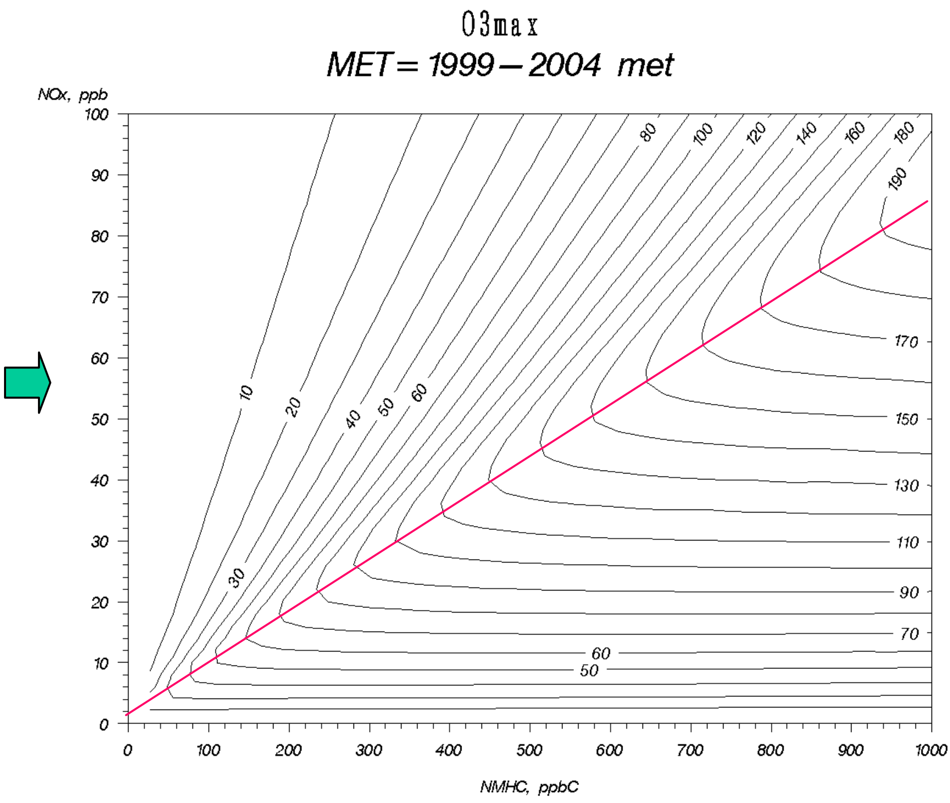




1981~1986 to 1999~2004

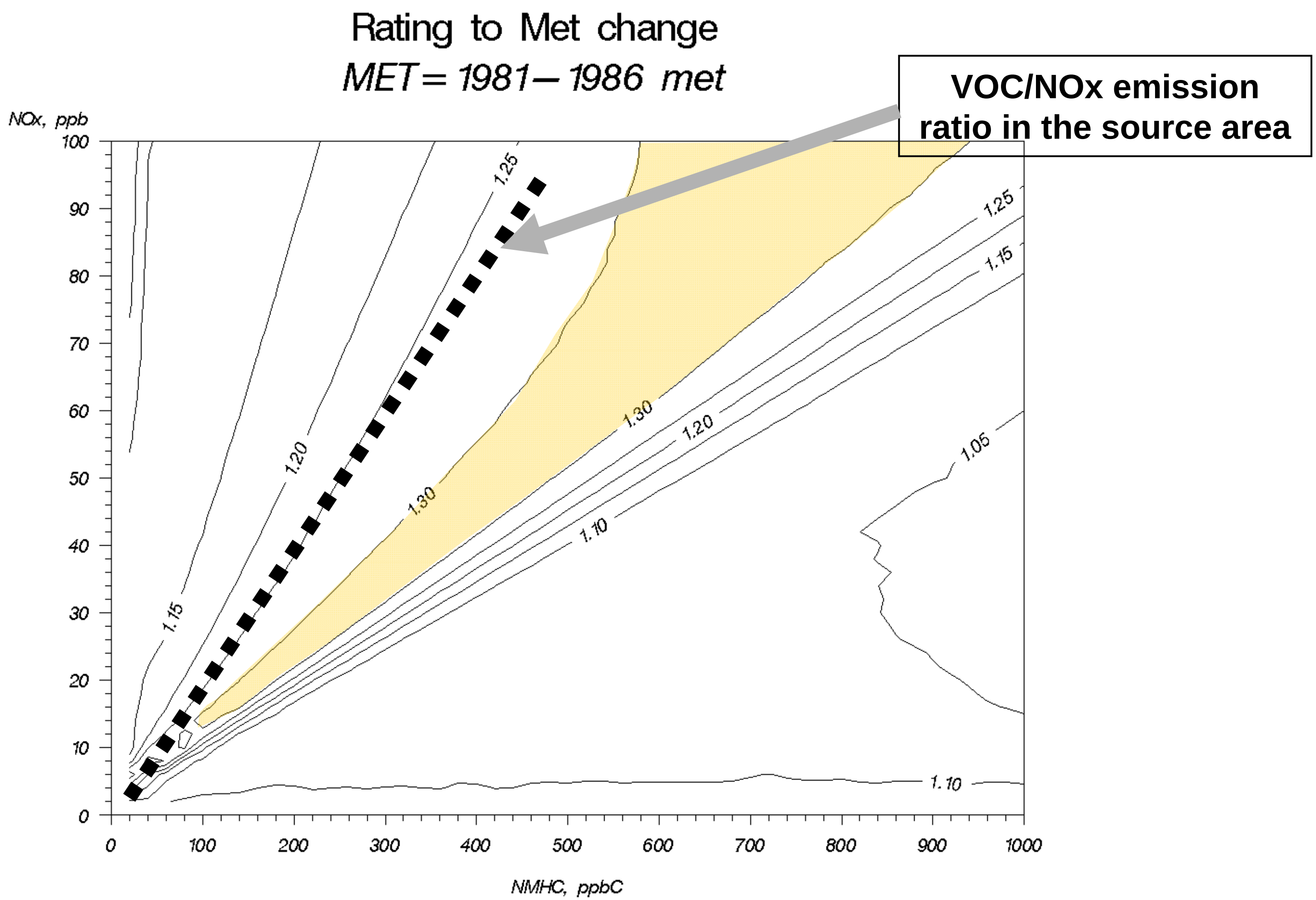
Solar radiation	7% increase
Temperature	2.3 ℃ rise





Reactivity→1.3 times, max

(different by position in the regime)



Conclusions

1. Boundary NMHC/NOx between “ozone weekend increase” and “ozone weekend decrease” is historically descending from 1980’s to 2000.
2. Boundary of “ozone weekend change” is thought to be equivalent to the boundary of “ozone formation regimes”.
3. Modeled ozone isopleth diagrams suggest relationship between the boundary NMHC/NOx of regimes and reactivity of the compounds;

   ‘lower NMHC/NOx’ means ‘more reactive’
4. Estimated historical increase of VOC reactivity is a part of the cause of peak ozone increase despite of the decreasing of concentration itself.

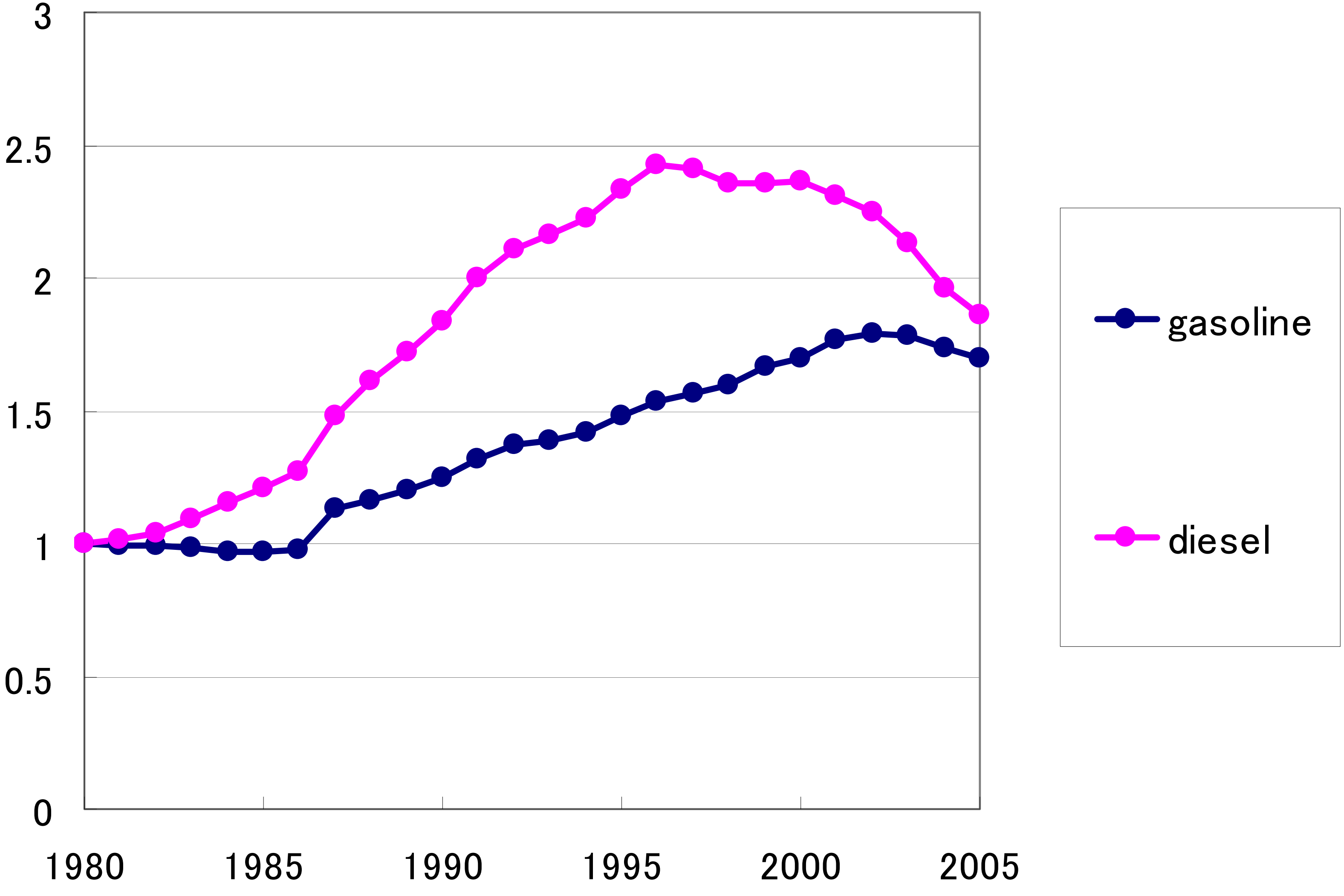
Future tasks

- Developing historical emission inventories for verification of our estimation
- Precise information of source specific VOC composition in detail, including seasonal variation
- How are the other regions in the world?

↓

- Strategy of more efficient VOC source measures for ozone reduction

Fuel consumption trends of gasoline and diesel oil in Japan



(Statistical Report on Motor Vehicle Transport)