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# An application of HDDM sensitivity analysis over East Asia

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# Outline

- Description of HDDM sensitivity analysis
- Results and Discussion
  - Large-scale trans-boundary episode on spring 2007
  - Seasonality of source-receptor relationship
- Conclusions and Future perspective

reference)

Cohan and Napelenok, 2011, *Atmosphere*

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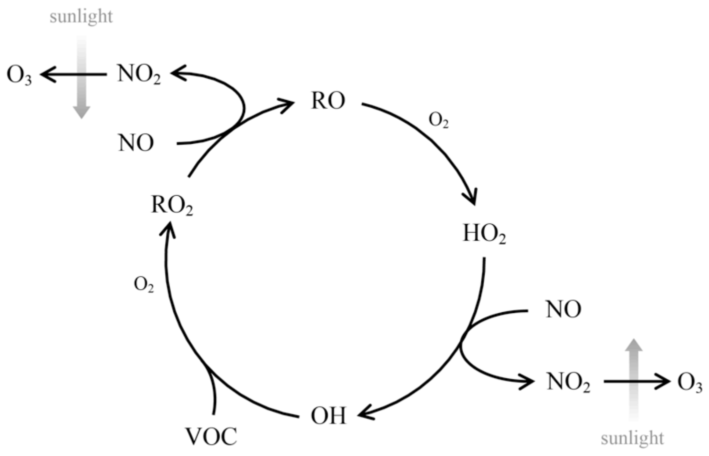
# Tropospheric Ozone

- Mechanism

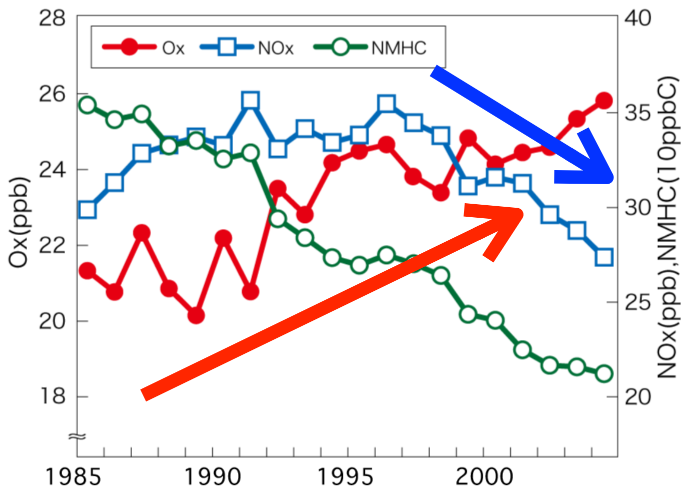



NOx = NO + $NO_2$

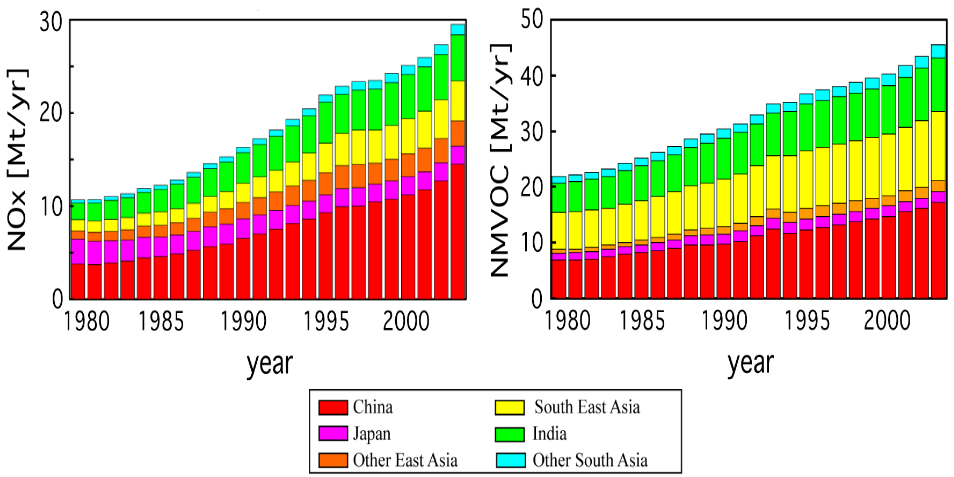
VOC = NMHC + R-OH + R-CHO + …

Ox = $O_3$ + HCHO + PAN + …

- Recent trend in Japan




- Emission trend over East Asia



# Source-Receptor relationship

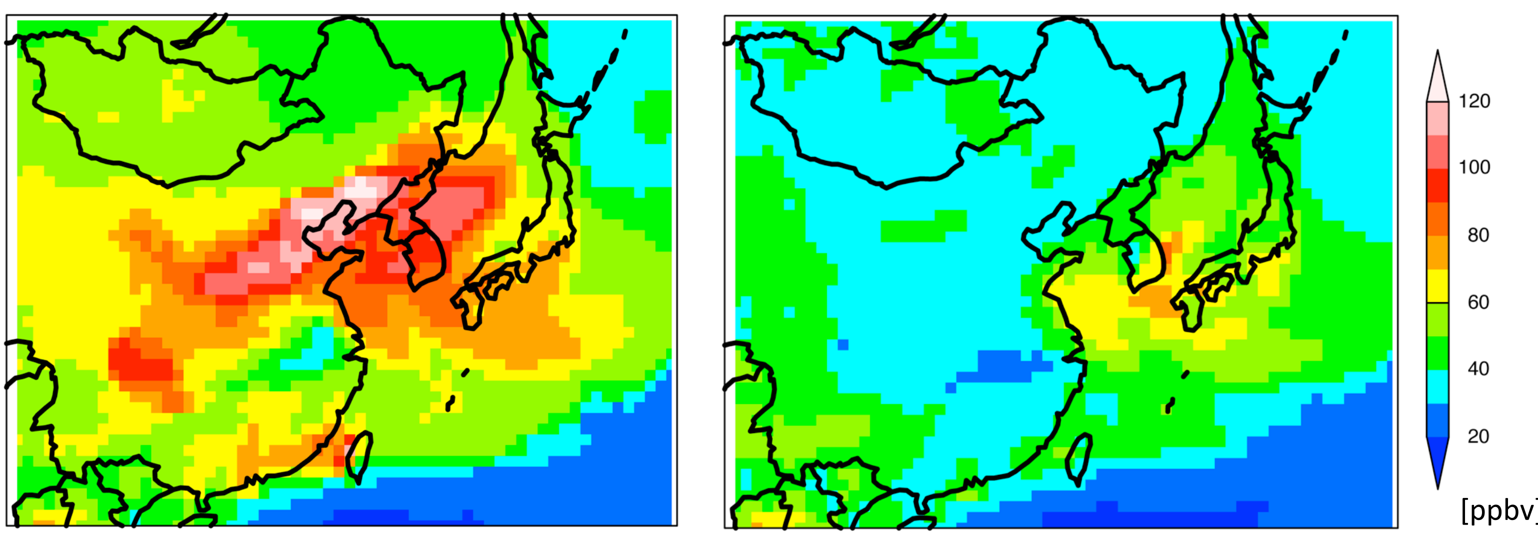
## ✓ Emission Sensitivity Approach

determines the changes of $O_3$ concentrations
due to emission perturbations on different sources/regions

- Zero-out run

base case simulation and switching off specific emission source/region

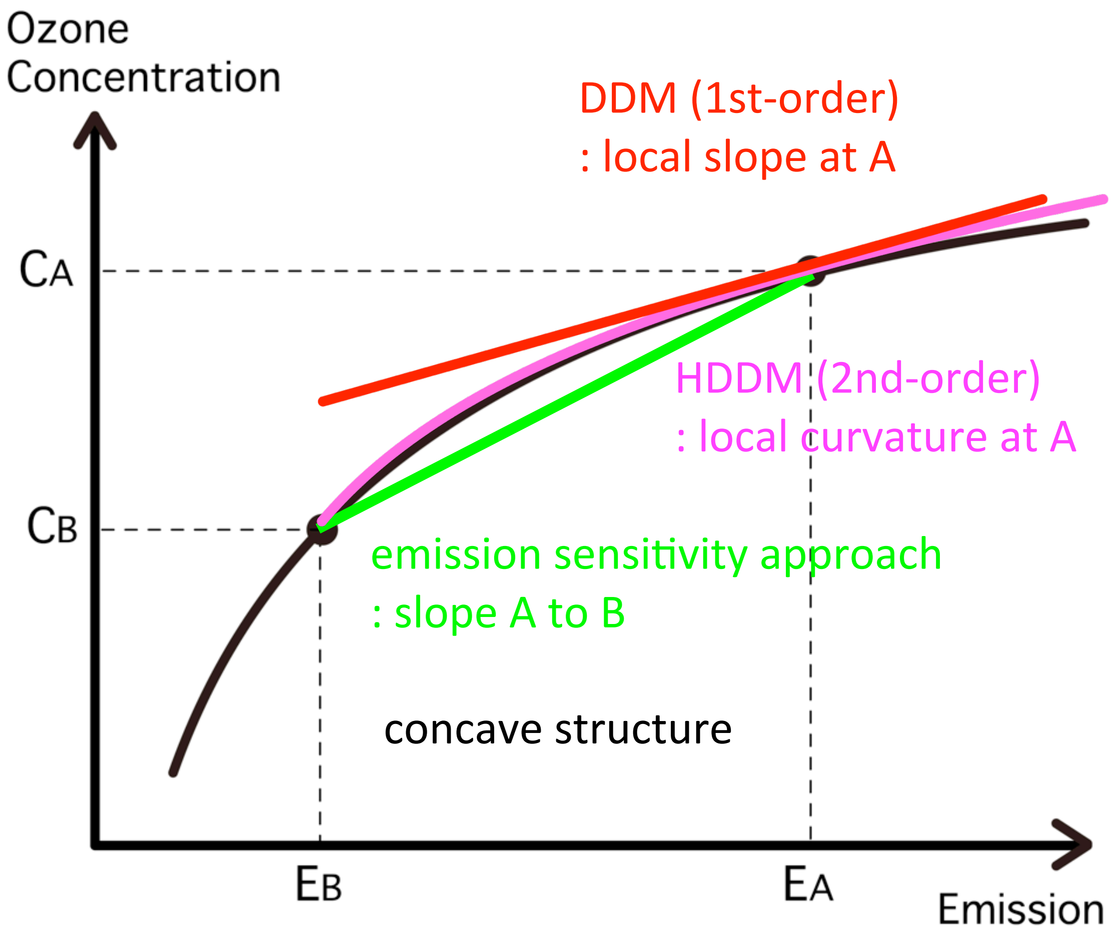
base case simulation        sensitivity simulation




Conclusions would depend on the magnitude of the perturbation range

# HDDM sensitivity analysis

## Higher-order Decoupled Direct Method （HDDM）




calculate the local slope and local curvature in one-time simulation

first-order sensitivity:

$$s_{i,j}^{(1)} = \frac{\partial C_i}{\partial p_j} > 0$$

second-order sensitivity:

$$s_{i,\{j,k\}}^{(2)} = \frac{\partial}{\partial p_j}\left(\frac{\partial C_i}{\partial p_k}\right) < 0$$

emission sensitivity approach : emissions are varied one-at-a-time

Dunker et al. (1984)

Yang et al. (1999)

Hakami et al. (2003)

Cohan et al. (2005)

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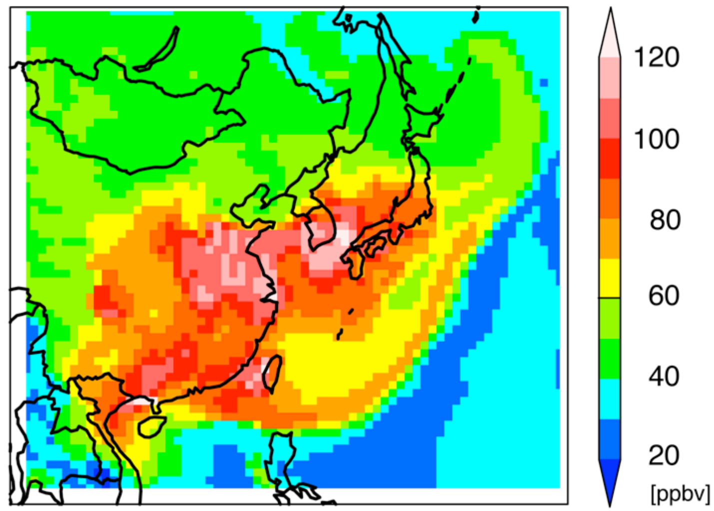
# Results of HDDM sensitivity analysis

ozone concentration
during severe air pollution
episode on 6-9, May, 2007

$O_3$ concentration

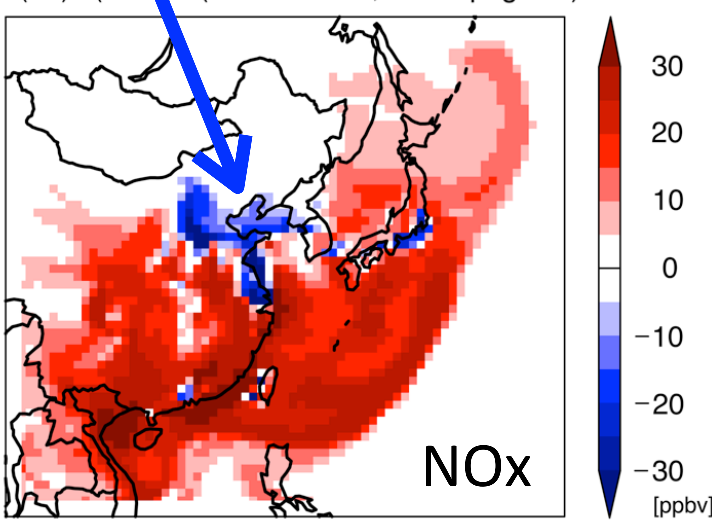



positive sensitivity:
ozone will increase
if NOx/VOC emis. increase

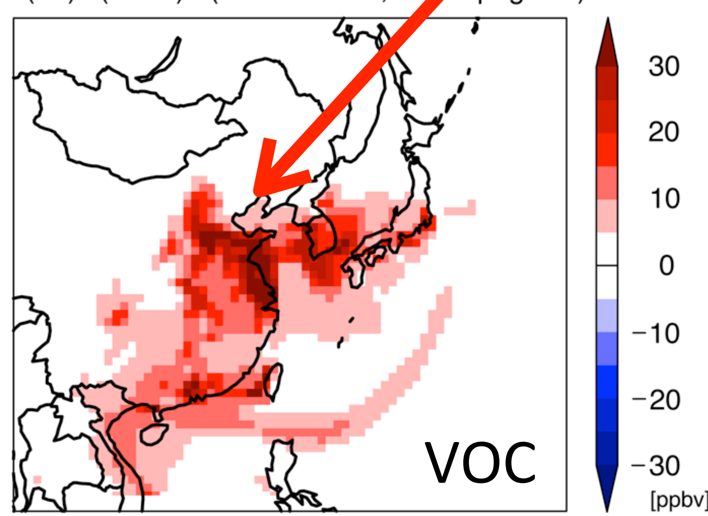
due to the NOx titration

$S_j^{(1)}$

$\partial(O_3)/\partial(E_{NOx})$ (domain-wide, anthropogenic)




$\partial(O_3)/\partial(E_{VOC})$ (domain-wide, anthropogenic)



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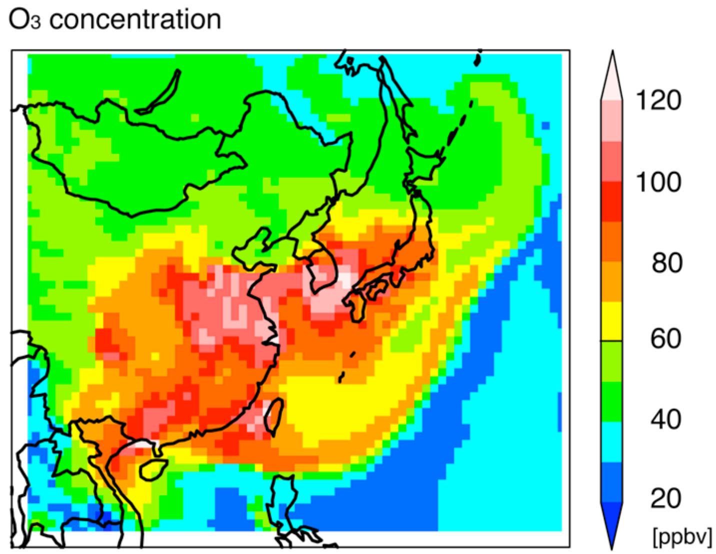


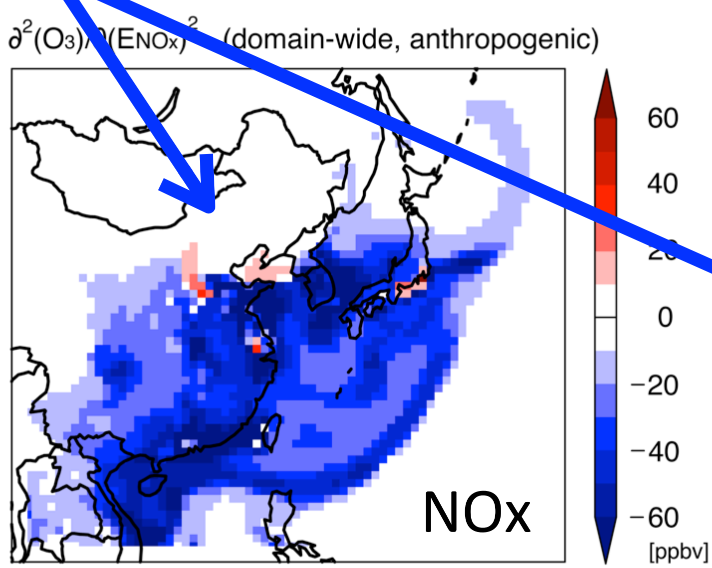

small sensitivity than that of NOx emissions

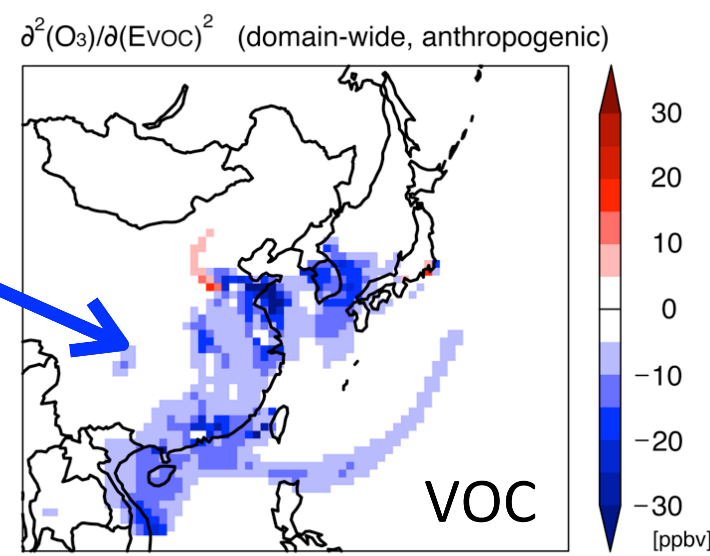
↓

nonlinearity of ozone to VOC is lower

concave structure

$S^{(2)}_{j,j}$



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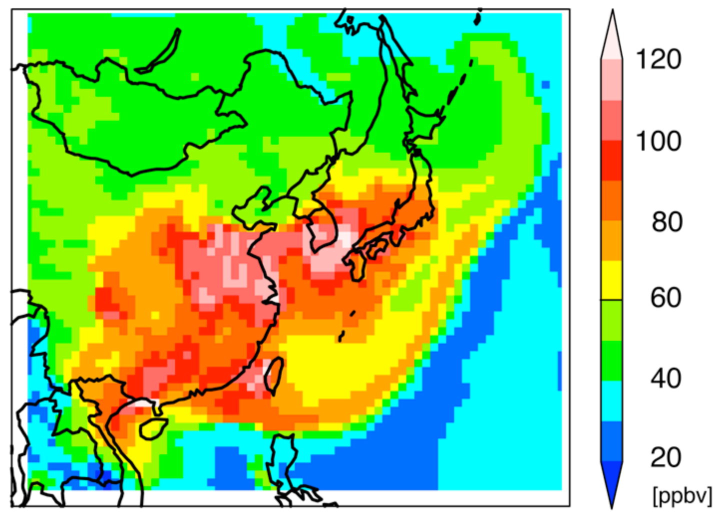
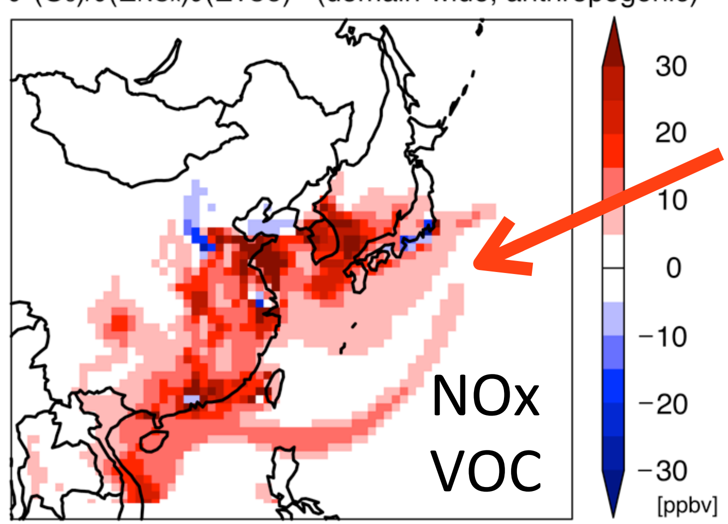



$S^{(2)}_{j,k}$

$\partial^2(O_3)/\partial(E_{NOx})\partial(E_{VOC})$ (domain-wide, anthropogenic)




ozone will become less
responsive to NOx emis.
under the decline of
VOC emis. (and *vice versa*)

# Validation of HDDM

## HDDM

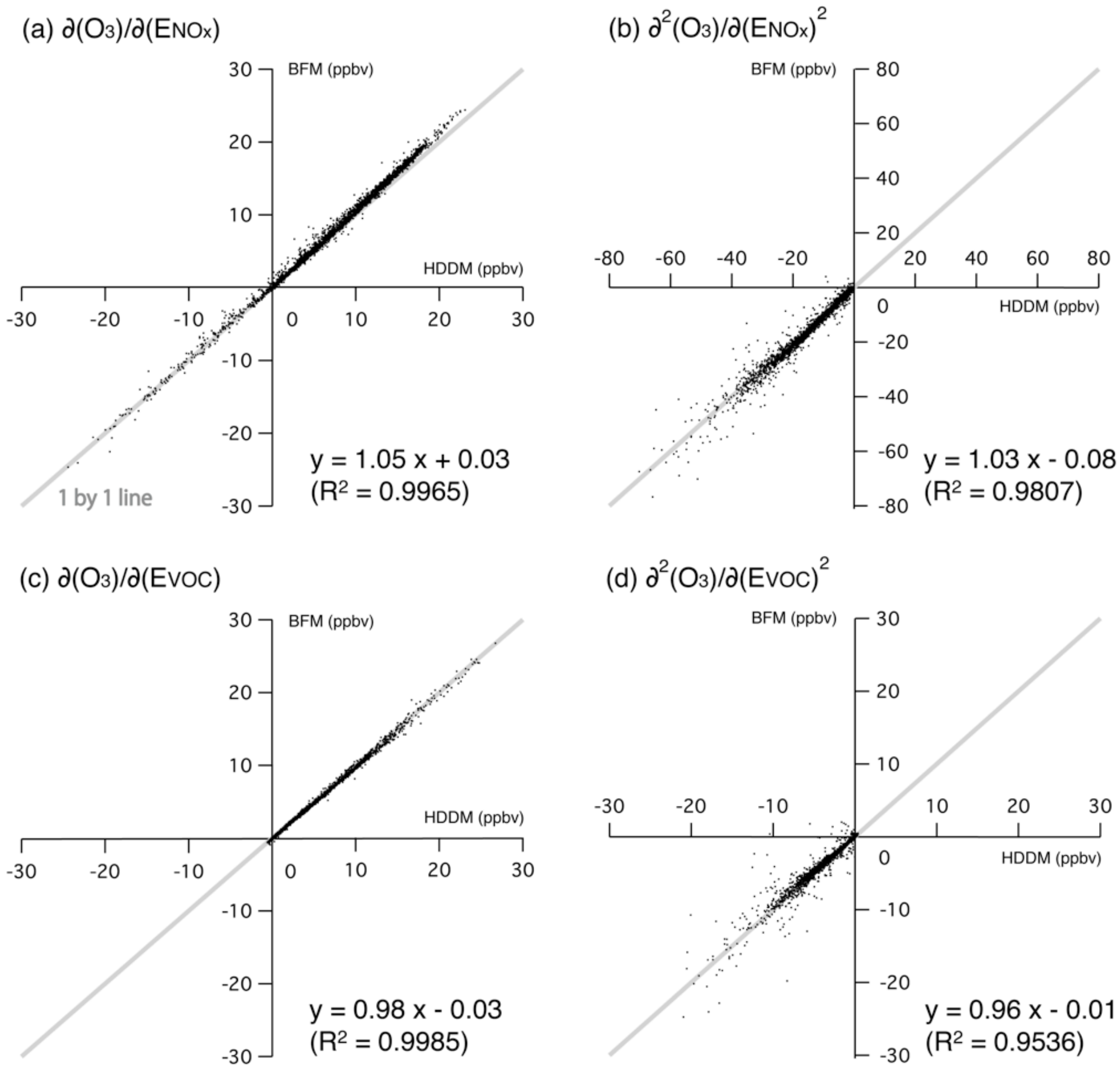
$$S_j^{(1)} = \frac{\partial C}{\partial \varepsilon_j}$$

$$S_{j,j}^{(2)} = \frac{\partial^2 C}{\partial \varepsilon_j^2}$$

## emission sensitivity approach

$$S_j^{(1)} \approx \frac{C^{+\Delta\varepsilon_j} - C^{-\Delta\varepsilon_j}}{2\Delta\varepsilon_j}$$

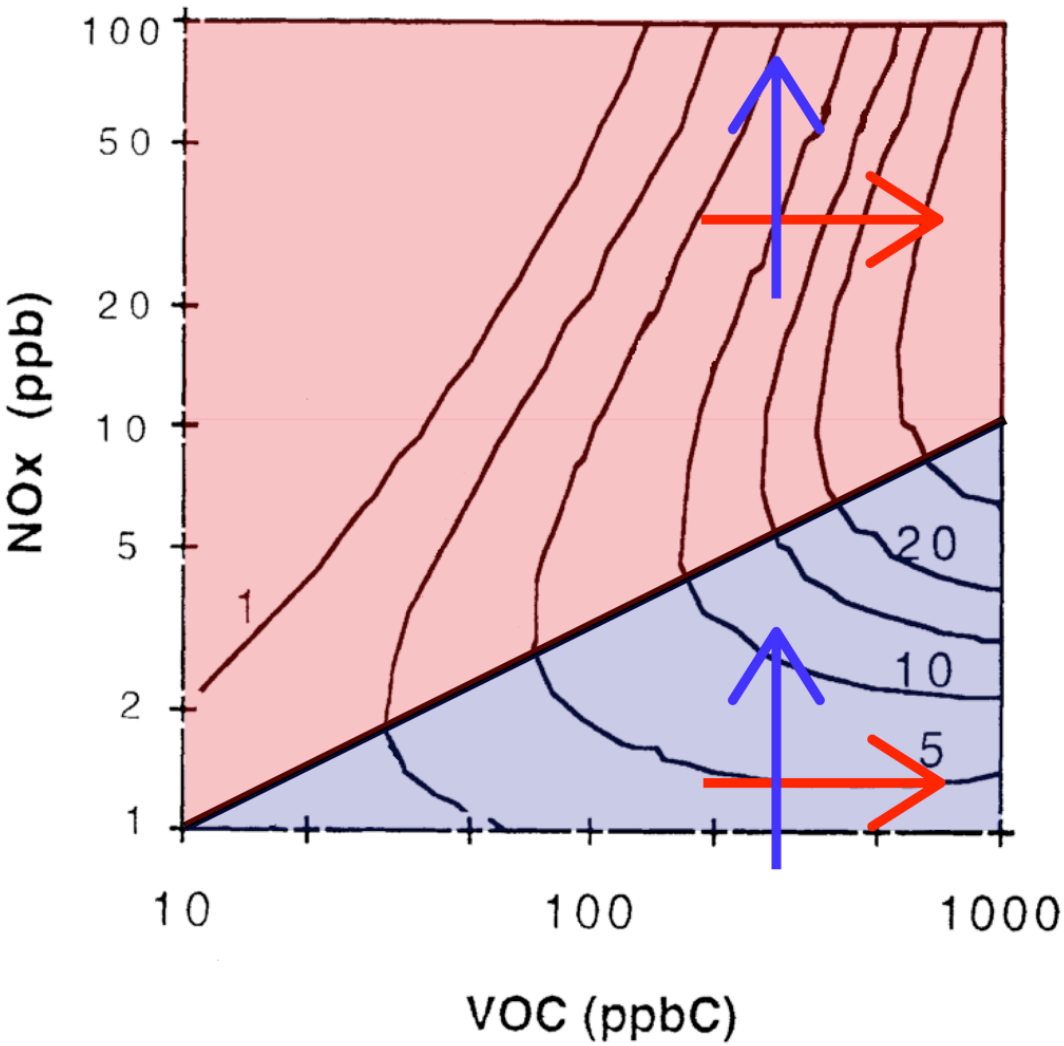
$$S_{j,j}^{(2)} \approx \frac{C^{+\Delta\varepsilon_j} - 2C_0 + C^{-\Delta\varepsilon_j}}{(\Delta\varepsilon_j)^2}$$




## Application of HDDM

- Ozone sensitive regime
- Ozone isopleth
- Source contribution

# Ozone sensitive regime




after Sillman (1999)

NOx → $\Delta O_3 < 0$
VOC → $\Delta O_3 > 0$
VOC-sensitve regime

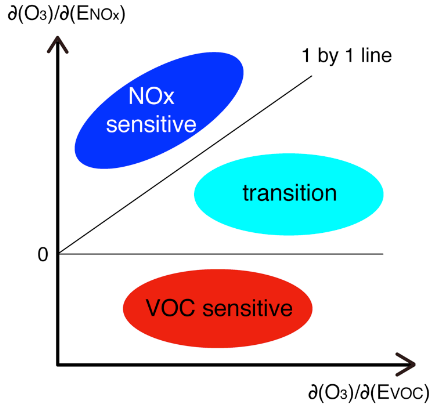
NOx → $\Delta O_3 > 0$
VOC → $\Delta O_3 = 0$
NOx-sensitve regime

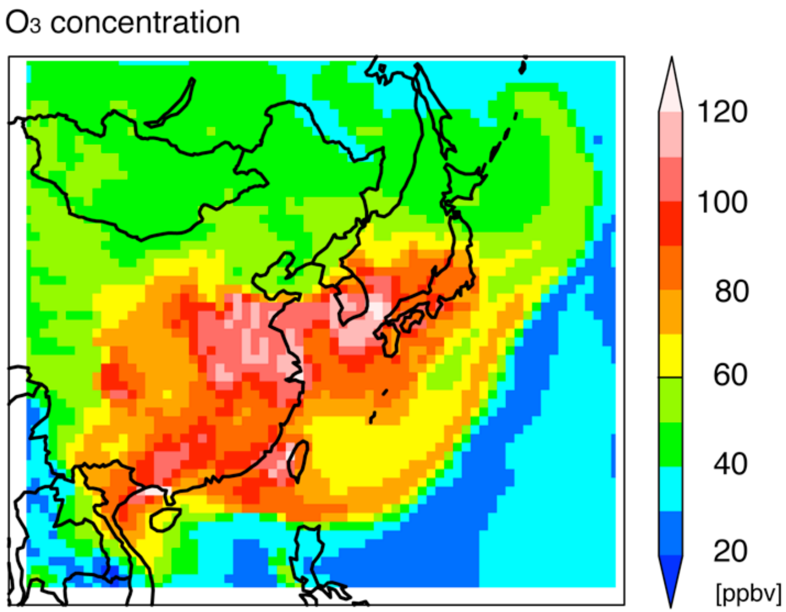
# Ozone sensitive regime

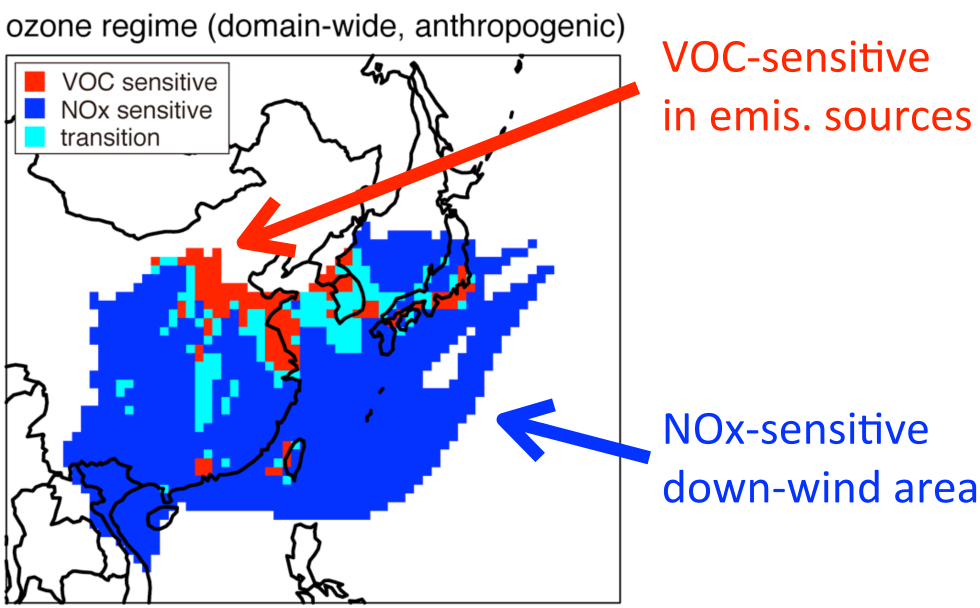
$$\frac{\partial[O_3]}{\partial\varepsilon_{\mathrm{ENOx}}} < 0, \qquad \text{VOC control}$$

$$\frac{\partial[O_3]}{\partial\varepsilon_{\mathrm{ENOx}}} > \frac{\partial[O_3]}{\partial\varepsilon_{\mathrm{EVOC}}} > 0, \qquad \text{NOx control}$$

$$0 < \frac{\partial[O_3]}{\partial\varepsilon_{\mathrm{ENOx}}} < \frac{\partial[O_3]}{\partial\varepsilon_{\mathrm{EVOC}}}, \qquad \text{transition}$$



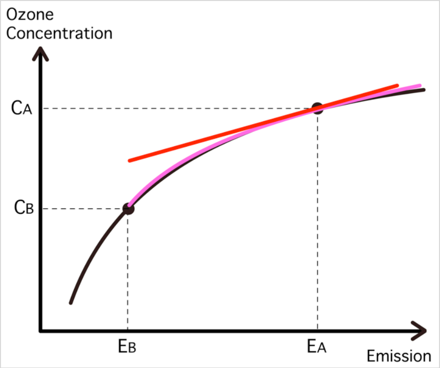



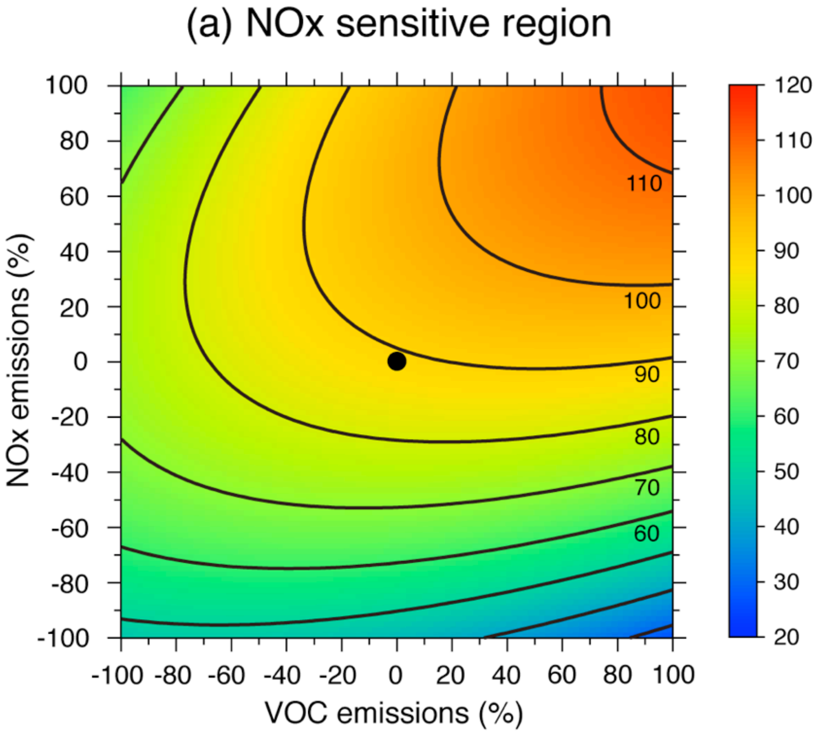
# Ozone isopleth

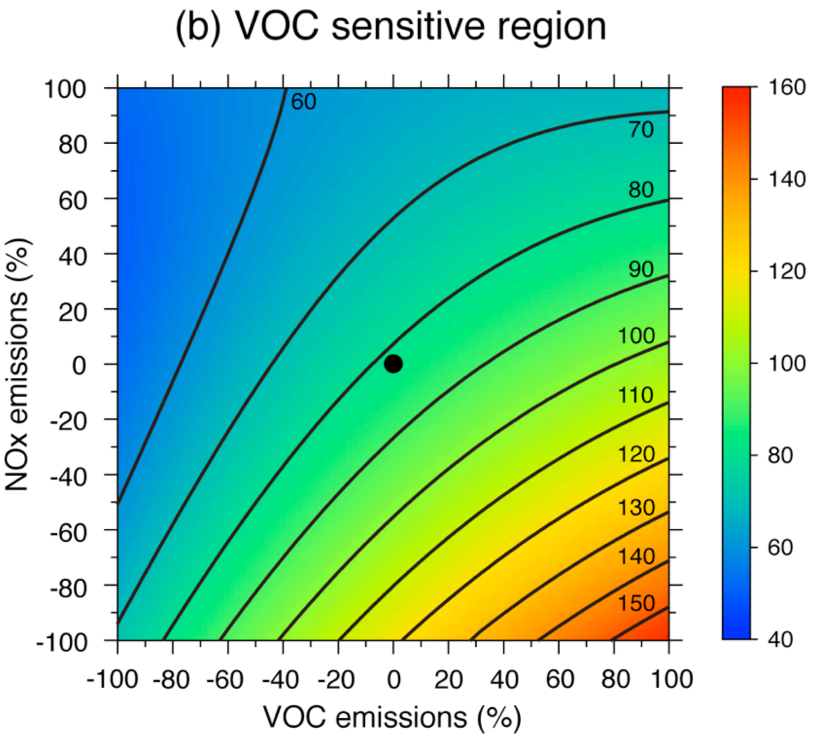
## Parametric Scaling Method

$$C(p_j) = C(P_j) + S_j^{(1)} \Delta\varepsilon_j + \frac{1}{2!} S_{j,j}^{(2)} \Delta\varepsilon_j^2 + \cdots$$

: Ozone response from base case simulation, C (Pj), against any fractional perturbation



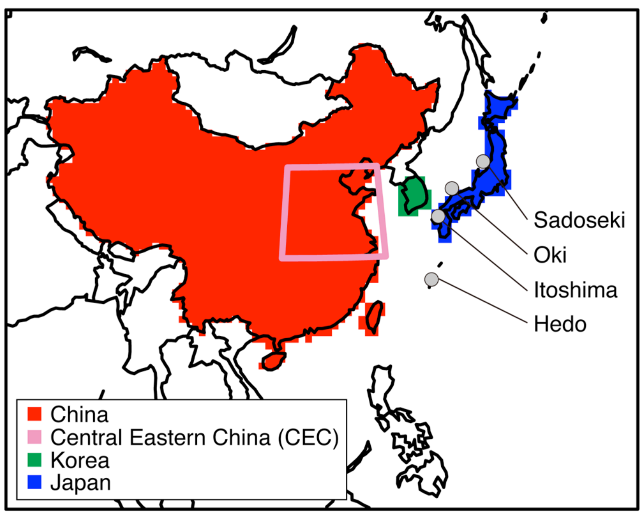
# Source contribution

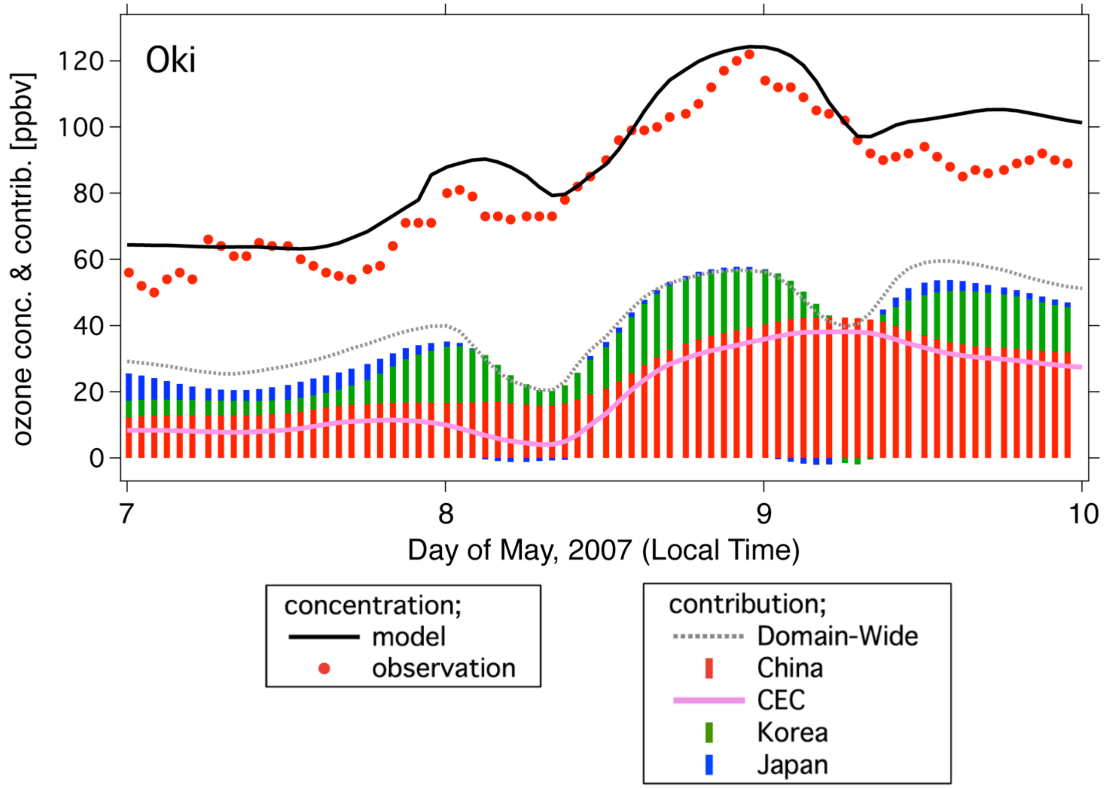
## Zero-Out Contribution (ZOC)

$$\mathrm{ZOC}(p_j, p_k) \approx \left(S_j^{(1)} - \frac{1}{2}S_{j,j}^{(2)}\right) + \left(S_k^{(1)} - \frac{1}{2}S_{k,k}^{(2)}\right) - S_{j,k}^{(2)}$$

j: anth. NOx emis.
k: anth. VOC emis.

: examine the source contributions including non-linear response of ozone



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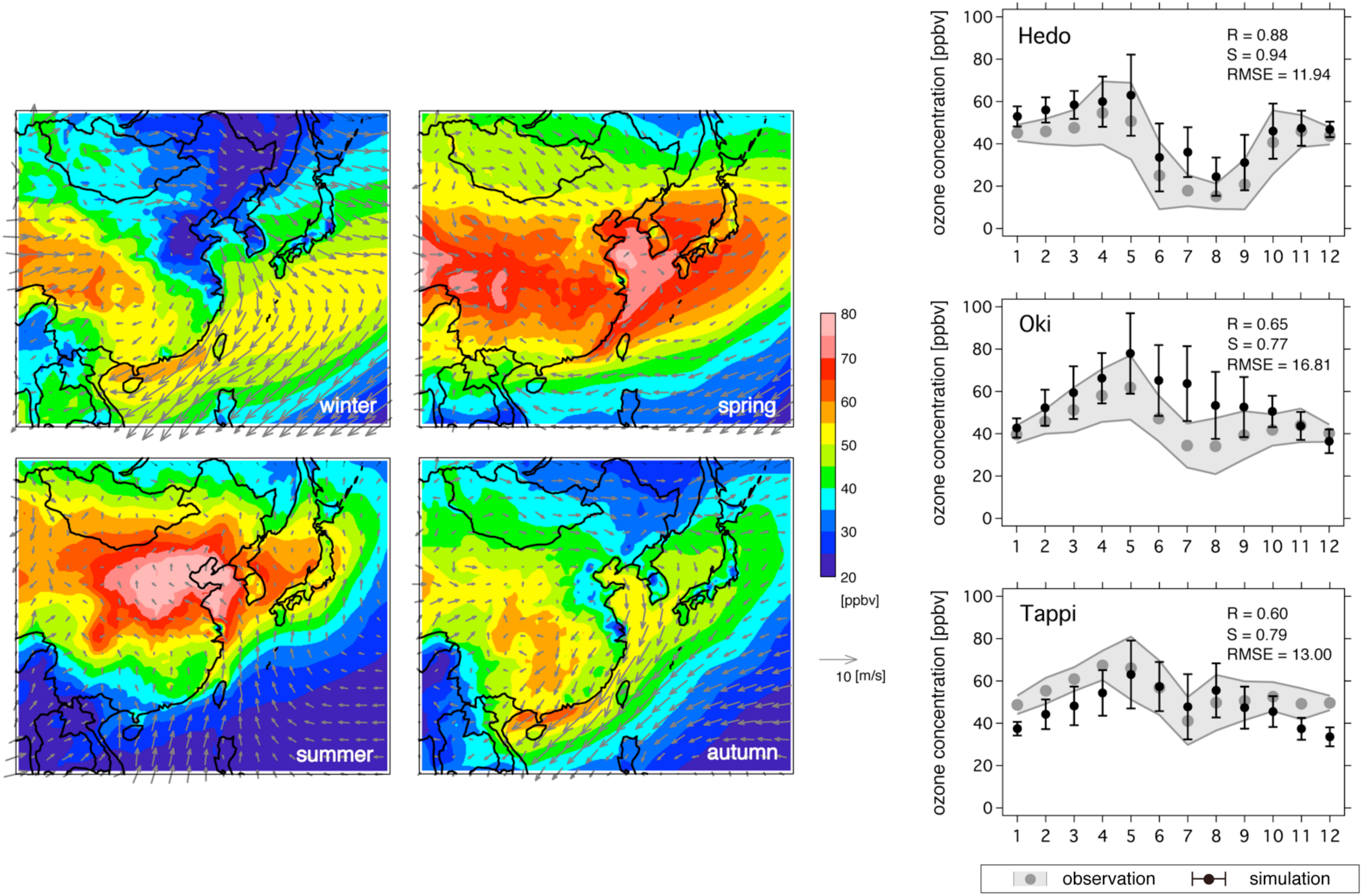
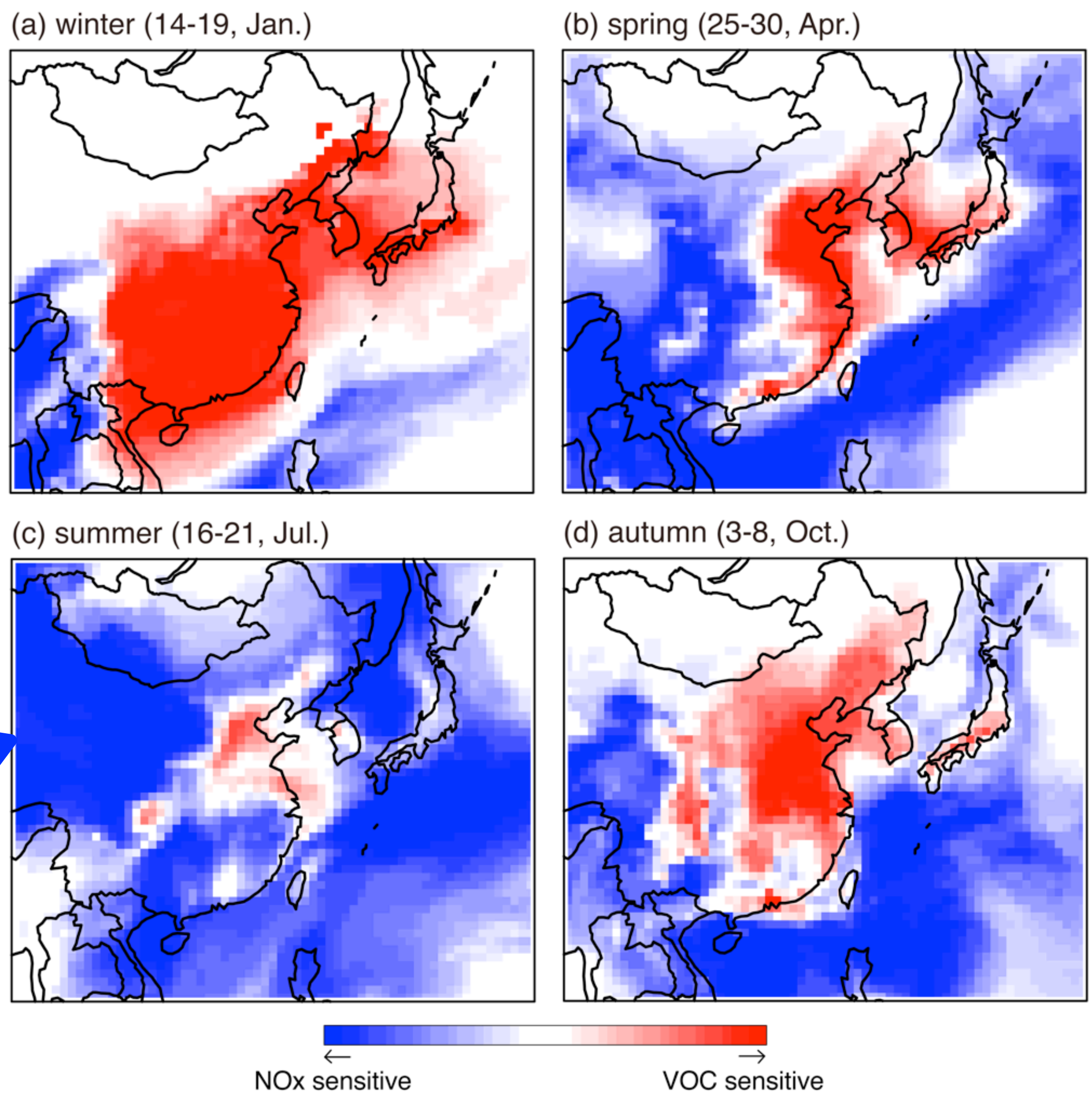
# Seasonal variation of tropospheric ozone



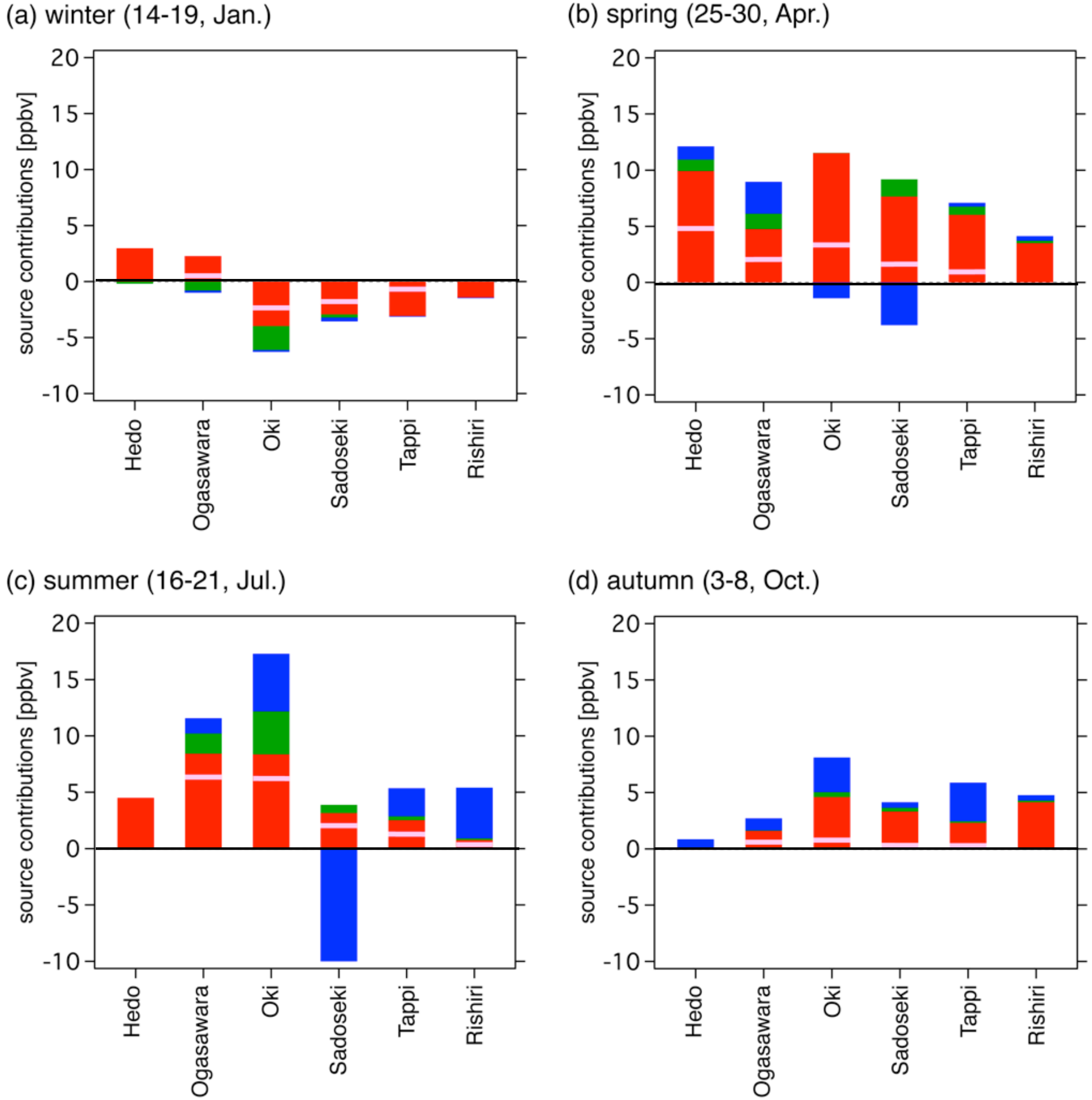
# Seasonal variation of ozone regime

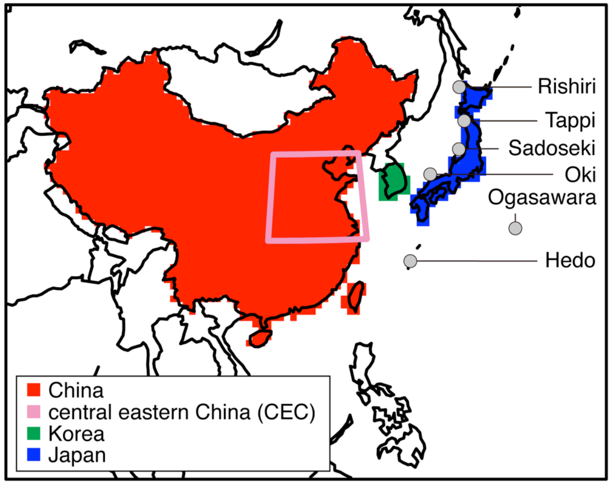
strong VOC-sensitive condition in winter

NOx-sensitive in summer



# Seasonal variation of source contribution




- spring; impact of trans-boundary air pollution
- summer, autumn; some local-scale contrib.
- winter; slight negative contrib.
- The contrib. of CEC to total-China is largest in summer

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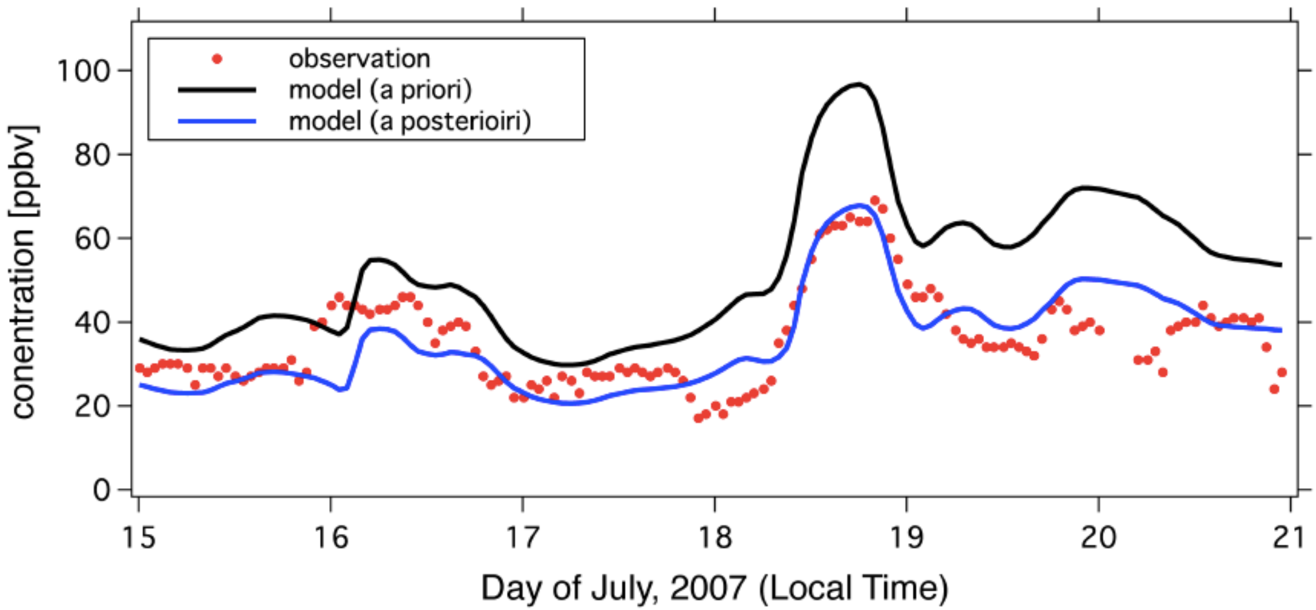
# Conclusion

- HDDM sensitivity analysis is a sophisticated way in emission sensitivity approach
  - calculate sensitivities in one-time simulation

- The application of HDDM results is also useful
  - provides the information of ozone sensitive regime
  - gives time- and location-dependent ozone isopleth
  - assesses source contributions

# Future perspective

The sensitivities are combined with the Green's function method (collaborated work with Dr. Yumimoto)




| Parameter | Uncertainty | Inversed solution |
|---|---|---|
| domain-wide precursor emissions | ±40 % | - 12.5 % |
| $NO_2 + hv \rightarrow NO + O$ | ±20 % | - 30.2 % |
| $NO_2 + OH + M \rightarrow HNO_3 + M$ | ±30 % | + 27.2 % |
| $NO + O_3 \rightarrow NO_2 + O_2$ | ±10 % | + 14.2 % |