Evaluation of emission source contributions for tropospheric ozone over East Asia based on HDDM and OSAT on CAMx model

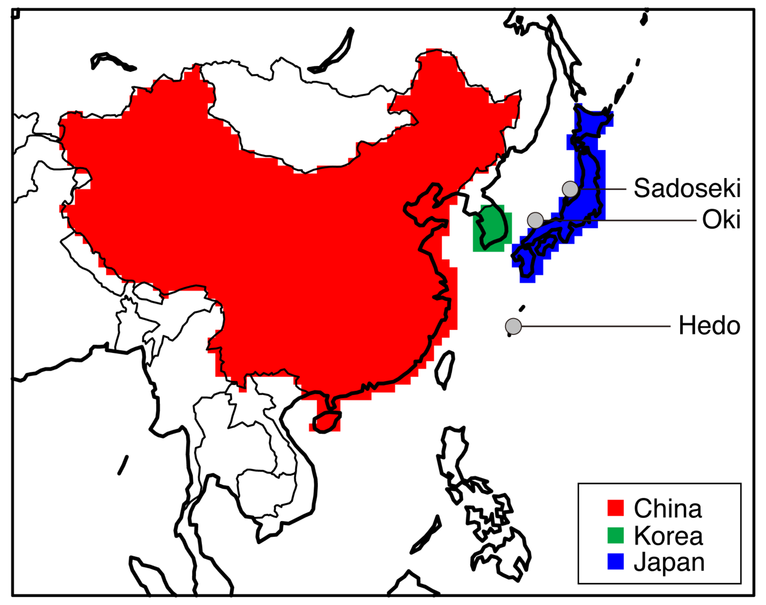
* **Syuichi Itahashi**, Hiroshi Hayami — Environmental Science Research Laboratory, Central Research Institute of Electric Power Industry (CRIEPI), Chiba, Japan

Itsushi Uno — Research Institute for Applied Mechanics, Kyushu University, Fukuoka, Japan

Introduction

The response of tropospheric ozone (O_3) to emissions reductions at various levels in mainland China, Korea, and Japan were comprehensively investigated by higher-order decoupled direct method (HDDM) for sensitivity analysis and the ozone source apportionment technology (OSAT) for mass balance analysis.

Model design



- Comprehensive air-quality model with extentions (CAMx) regional model
  - ✓ 80 km horizontal grid resolution
  - ✓ 37 vertical layers upto 50 hPa
  - ✓ SAPRC99 gas-phase chemistry
- Severe pollution event during 1-16 May 2009

Fig. 1. Model domain with source regions (China, Korea, and Japan) and receptor sites (Hedo, Oki, and Sadoseki).

● Brute force method (BFM)

The chemical concentration with input parameter A varied is calculated as C_A.
The impact of the parameter A is evaluated by substituting these concentrations.

$$\Delta C = C_{\text{base-case}} - C_{\text{A}}$$

● Higher-order decoupled direct method (HDDM)

The seminormalized first- and second-order sensitivity coefficients in HDDM can be defined with a scaling factor of ε with a nominal value of 1.

$$S_i^{(1)} = \frac{\partial C}{\partial \varepsilon_i} \quad , \quad S_{i,j}^{(2)} = \frac{\partial^2 C}{\partial \varepsilon_i \partial \varepsilon_j}$$

Projected concentration with the fractional perturbation from the base-case simulation can be estimated by a Taylor-series expansions.

$$C(p_i, p_j) \approx C_{\text{base-case}} + S_i^{(1)} \Delta\varepsilon_i + \frac{1}{2} S_{i,i}^{(2)} \Delta\varepsilon_i^2 + S_j^{(1)} \Delta\varepsilon_j + \frac{1}{2} S_{j,j}^{(2)} \Delta\varepsilon_j^2 + S_{i,j}^{(2)} \Delta\varepsilon_i \Delta\varepsilon_j$$

● Ozone source apportionment technology (OSAT)

The tracer for O_3 formed from NOx and VOC in OSAT are respectively expressed as $\mathrm{O3N}$ and $\mathrm{O3V}$, and the sums of tracers satisfy the mass consistency equations.

$$\sum_{k=1}^{m} \mathrm{O3N}_k + \sum_{k=1}^{m} \mathrm{O3V}_k = \mathrm{O_3}$$

Projected concentration with the fractional perturbation from the base-case simulation can be estimated by simple linear interpolation of tracers concentration.

$$C(p_k) = C_{\text{base-case}} + (\mathrm{O3N}_k + \mathrm{O3V}_k)\Delta\varepsilon_k$$

Results and Discussion

Modeling reproducibility was statistically examined. MB was 1-5 ppbv, ME was 7-10 ppbv, and R is 0.7-0.9 at Hedo, Oki, and Sadoseki during whole event. The modeling system captured the observed O_3 features. The averaged O_3 concentration during 10-11 May 2009 exceeded the AQS in Japan broadly (Fig. 2).

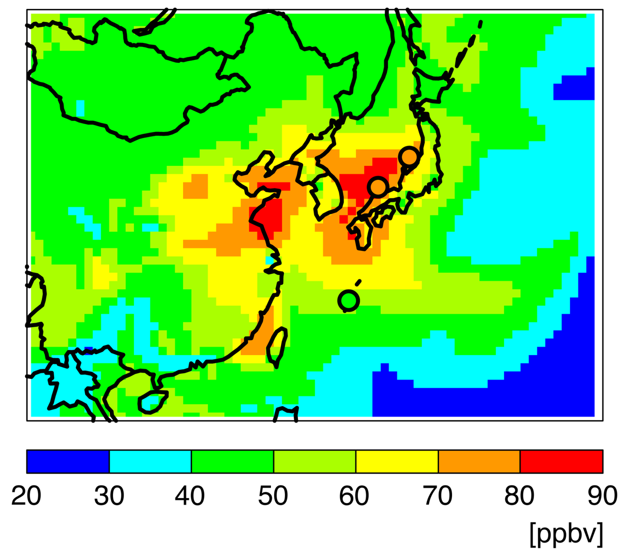


Fig. 2. Averaged O_3 concentration.

The source contributions of O_3 were evaluated against emissions from China, Korea, and Japan based on BFM, HDDM, and OSAT. BFM was configured with emission reduction of 10%, 30%, 50%, 70%, and 100%. Absence of the negative sensitivities on OSAT (Fig. 3) and the increasing errors as reduction became larger (Fig. 4, left), importance of the treatment of NO titration ($NO + O_3 \rightarrow NO_2 + O_2$) in OSAT was suggested; hence "potential" O_3 (PO) was introduced as: $PO = O_3 + NO_2$.

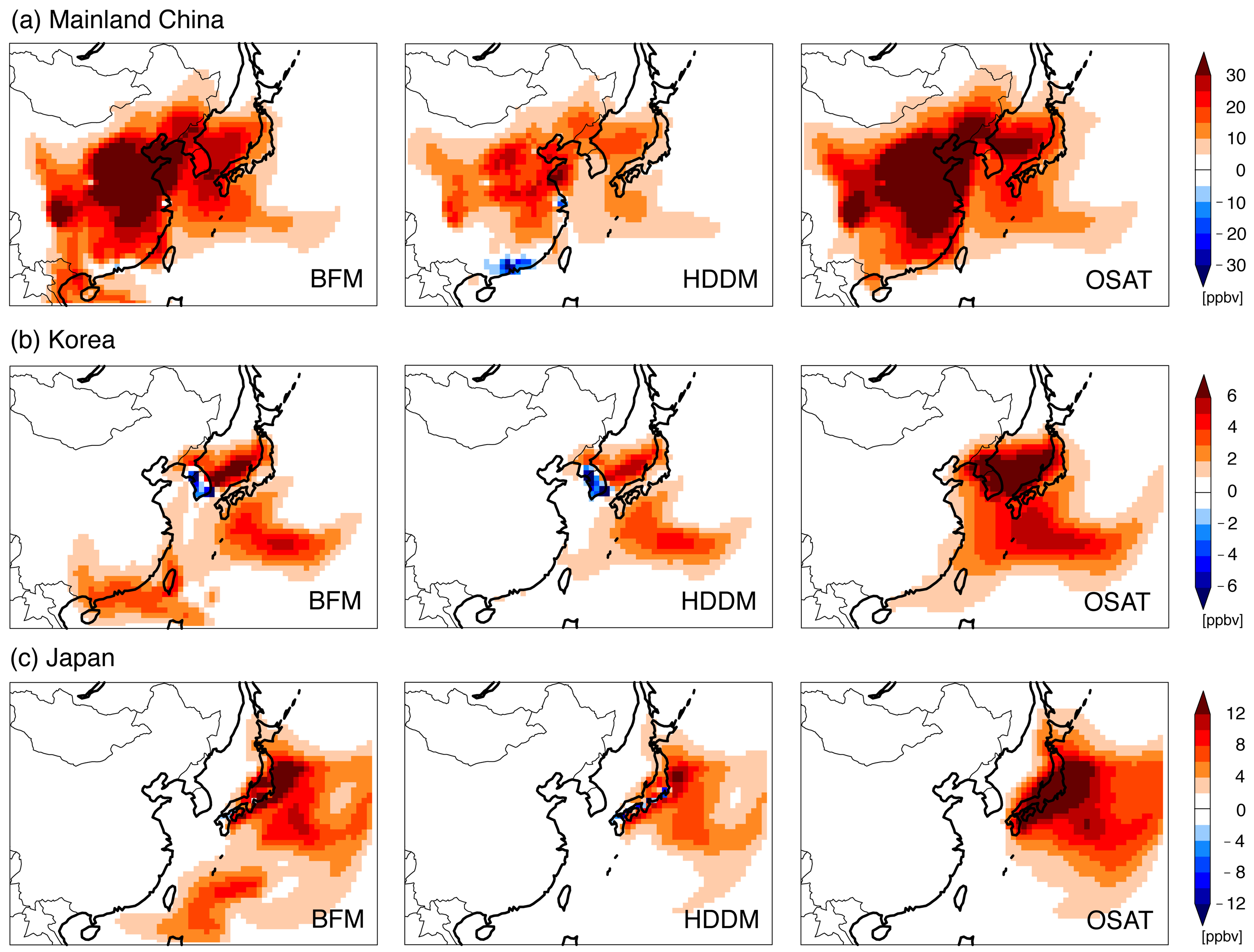


Fig. 3. Spatial distribution of O_3 source apportionments to (a) China, (b) Korea, and (c) Japan estimated by (left) BFM, (center) HDDM, and (right) OSAT averaged over 10-11 May 2009.

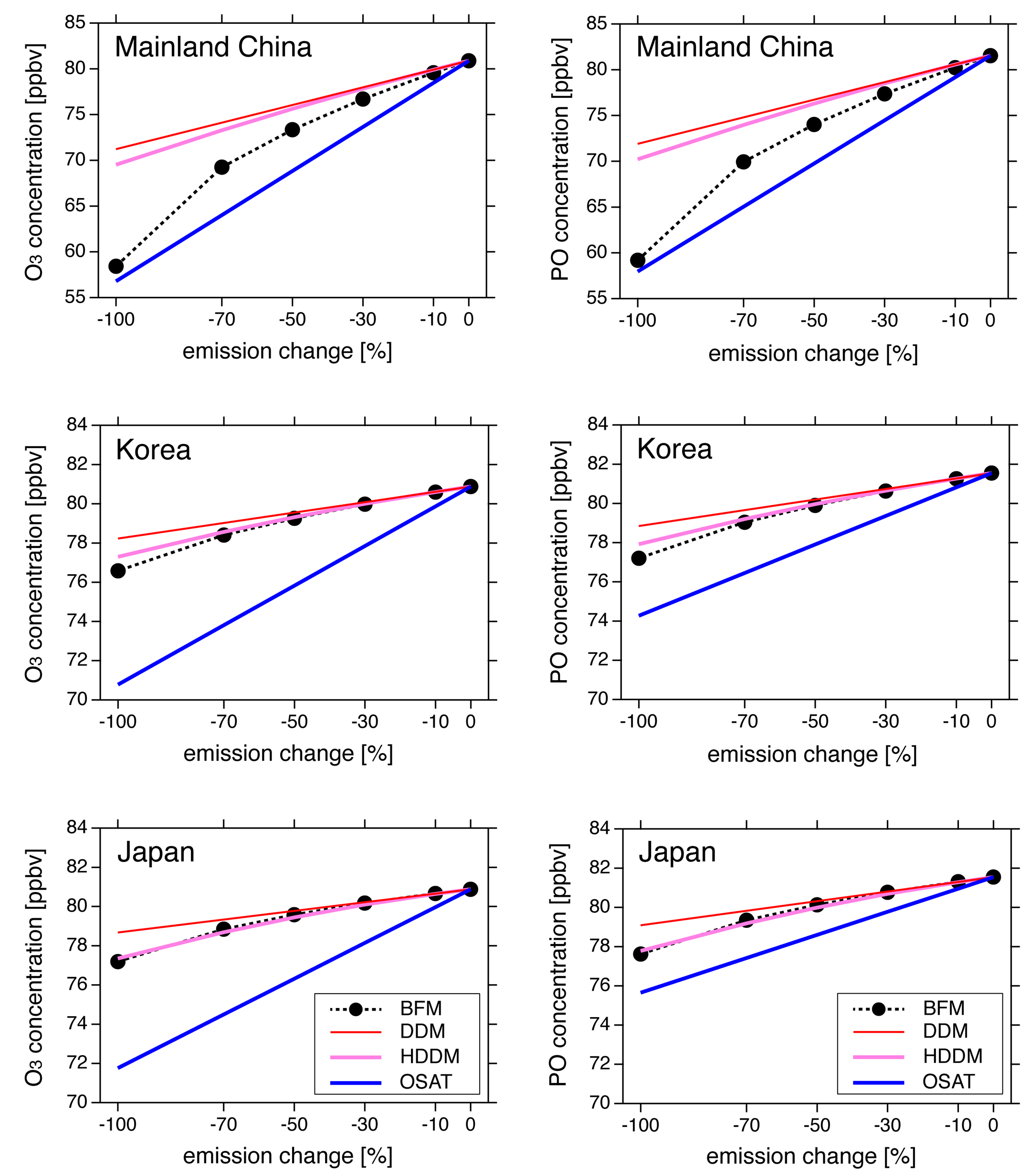


Fig. 4. Response of (left) O_3 and (right) PO concentration to reduction of emissions from China, Korea, and Japan.

By introducing PO instead of O_3, the source contributions of PO were evaluated (Fig. 4, right). The PO responses estimated by HDDM was comparable with that of O_3, but PO responses to emissions reduction from Korea and Japan estimated by OSAT have improved. The ability of proving tools against BFM were summarized (Table 1).

Table 1. Summary of the ability of DDM, HDDM, and OSAT to estimate source contributions for O_3 and PO.

Source region	Reduction rate	Ability for O_3			Ability for PO		
		DDM	HDDM	OSAT	DDM	HDDM	OSAT
China	10-30%	****	*****	****	****	*****	****
	50-70%	**	****	**	**	****	**
	100%	*	*	***	*	*	**
Korea	10-30%	*****	*****	****	*****	*****	*****
	50-70%	*****	*****	***	*****	*****	****
	100%	****	*****	**	****	*****	****
Japan	10-30%	*****	*****	****	*****	*****	*****
	50-70%	*****	*****	***	*****	*****	****
	100%	***	****	**	***	****	****

Note: Ability is assessed by averaging the NMB at Hedo, Oki, and Sadoseki.
*****, <1%; ****, 1-3%; ***, 3-5%; **, 5-10%; * >10% bias.

Summary

Emission source contributions for O_3 was comprehensively evaluated, and to address the limitation of the treatment of NO titration in OSAT, PO was introduced. The proposed approach with PO refined OSAT ability and did not degrade HDDM performance.

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Contact: isyuichi@criepi.denken.or.jp (Syuichi ITAHASHI)

CRIEPI Central Research Institute of Electric Power Industry