Introduction
The response of tropospheric ozone (O₃) 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 extensions (CAMx) regional model
- 80 km horizontal grid resolution
- 37 vertical layers up to 50 hPa
- SAPRC99 gas-phase chemistry
- Severe pollution event during 1-16 May 2009

Brute force method (BFM)
The chemical concentration with input parameter A varied is calculated as Cₐ.
The impact of the parameter A is evaluated by substituting these concentrations.

Higher-order decoupled direct method (HDDM)
The seminormalized first- and second-order sensitivity coefficients in HDDM can be defined with a scaling factor of 1 with a nominal value of 1.

Ozone source apportionment technology (OSAT)
The tracer for O₃ formed from NOx and VOC in OSAT are respectively expressed as O₃N and O₃V, and the sums of tracers satisfy the mass consistency equations.

By introducing PO instead of O₃, the source contributions of PO were evaluated (Fig. 4, right). The PO responses estimated by HDDM was comparable with that of O₃, 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).

Summary
Emission source contributions for O₃ were 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.

Table 1. Summary of the ability of DDM, HDDM, and OSAT to estimate source contributions for O₃ and PO.

<table>
<thead>
<tr>
<th>Source region</th>
<th>Reduction rate</th>
<th>Ability for O₃</th>
<th>Ability for PO</th>
</tr>
</thead>
<tbody>
<tr>
<td>China</td>
<td>10-30%</td>
<td>***</td>
<td>*****</td>
</tr>
<tr>
<td></td>
<td>50-70%</td>
<td>**</td>
<td>**</td>
</tr>
<tr>
<td></td>
<td>100%</td>
<td>*</td>
<td>*</td>
</tr>
<tr>
<td>Korea</td>
<td>10-30%</td>
<td>****</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>50-70%</td>
<td>**</td>
<td>**</td>
</tr>
<tr>
<td></td>
<td>100%</td>
<td>*</td>
<td>*</td>
</tr>
<tr>
<td>Japan</td>
<td>10-30%</td>
<td>****</td>
<td>***</td>
</tr>
<tr>
<td></td>
<td>50-70%</td>
<td>**</td>
<td>**</td>
</tr>
<tr>
<td></td>
<td>100%</td>
<td>*</td>
<td>*</td>
</tr>
</tbody>
</table>

Note: Ability is assessed by averaging the NMB at Hedo, Oki, and Sadoski.

***** (>1%); ****, 1-3%; ***, 3-5%; **, 5-10%; * >10% bias.

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

Fig. 2. Averaged O₃ concentration.

Fig. 3. Spatial distribution of O₃ 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) O₃ and (right) PO concentration to reduction of emissions from China, Korea, and Japan.

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