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Assessment of transboundary ozone contribution toward South Korea using multiple source–receptor modeling techniques

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HIGHLIGHTS

- We assessed the S–R relationships for ozone using three methods over East Asia.
- We evaluated S–Rs for not only anthropogenic emissions but biogenic emissions.
- Three methods showed similar S–Rs for anthropogenic emission sources.
- OPTM estimated higher biogenic contributions for ozone formation then HDDM.
- NO_x limited regime was formed over South Korea for July periods.

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ABSTRACT

Ozone concentrations in East Asia were simulated using the Community Multi-scale Air Quality (CMAQ) model, and its source contributions were estimated by multiple source–receptor modeling techniques. To study relationships between ozone concentrations and precursor emission sources, three approaches were applied to four months (January, April, July, and October 2009) to represent seasonal characteristics and compare results, with a particular focus on South Korea. Brute force (BF) is a traditional sensitivity analysis method used to estimate model output response to an input change. The high-order decoupled direct method (HDDM), a computational method, is an efficient and accurate alternative to the BF method for sensitivity. The Ozone and Particulate Precursor Tagging Methodology (OPTM) provides contribution information quantified by tracking emissions from selected sources throughout the simulation period. The approaches generally show that most of the receptor regions were substantially influenced by emissions from central China, which is the largest anthropogenic emissions source region in East Asia. Local emissions were still major contributors, especially South Korea and Japan during July 2009. On the other hand, a case study of maximum 8-h ozone concentrations derived from CMAQ–OPTM on April 9 in South Korea shows that the NO_x and VOCs emissions from China contributed approximately 82% and 91%, respectively, to maximum 8-h ozone in Region 4 (South Korea) without boundary inflow, which indicates that Chinese emissions are the dominant contributor in this episode. A comparison study of the three approaches shows that HDDM tends to estimate biogenic source contributions lower than that from OPTM in China but similar to OPTM in South Korea and Japan. When comparing the BF method and HDDM, the sensitivity results show a reasonably good agreement during a given period. The location- and time-dependent maximum 8-h ozone isopleths over South Korea as a receptor region created by HDDM suggest that most ozone was being transported from central China, whereas almost no ozone was formed locally during April 2009, and local conditions were heavily VOC limited. On the other hand, local emissions were the dominant contributor during July 2009, and every source region showed a NO_x-limited regime, which indicates that ozone concentrations in South Korea strongly depend on NO_x emissions during this month.

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1. Introduction

Both nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the troposphere can have an impact on human health, either directly or because of their oxidation. This oxidation can lead to a variety of secondary oxidized products such as ozone, many of which are potentially more harmful than their precursors (Jenkin and Clemitshaw, 2000). Tropospheric ozone damages natural vegetation, agricultural crops, and human health, and is a greenhouse gas.

Previous research suggests that tropospheric ozone has increased over East Asia in the past decades and that the rate of increase is higher than in other regions (Akimoto et al., 1994; Oltmans et al., 1998; Lee et al., 1998; Meigen et al., 2004). Recently, NO_x, which is a critical precursor of ozone, has rapidly increased over East Asia, chiefly because of emission increases in China during the last decade (Yamaji et al., 2008). Zhang et al. (2007) reported that China's NO_x emissions increased from 10.9 Tg in 1995 to 18.6 Tg in 2004, a 70% increase. Long-range transport of ozone and its precursors can influence other regions located downwind of emission source regions. For instance, Asian pollution enhanced surface ozone concentrations by 5–7 ppb over western North America in spring 2006 (Zhang et al., 2008).

The future ozone concentrations over East Asia will be significantly influenced by anthropogenic ozone precursor emissions, which in turn depend on future economic growth and environmental policies in East Asia (Yamaji et al., 2008). To support effective air quality management plans, contribution assessments of source regions as well as emission source categories are needed to better understand contributions from the East Asian countries. In the region, Lin et al. (2008) have estimated source–receptor (S–R) relations for sulfur and reactive nitrogen deposition, and Zhang et al. (2004) and Yamaji et al. (2008) have studied ozone over East Asia.

The Community Multi-scale Air Quality (CMAQ) model (Byun and Ching, 1999) is one of the photochemical grid models being used to investigate air quality degradation and to predict the effectiveness of emission control measures. Photochemical grid models such as CMAQ have also been used to estimate source contributions. To assess source contributions, various techniques have been developed based on this model to inform policy and scientific applications, such as control strategy development and source apportionment review. Here, we apply multiple S–R modeling techniques, including Brute Force (BF), High-Order Decoupled Direct Method (HDDM), and the Ozone and Precursor Tagging Methodology (OPTM), to assess source contributions and compare results from multiple modeling techniques over the East Asia region. The S–R modeling techniques used in this study are described in chapter 2.1.

The main objectives of this study were to assess contributions from specific emission source regions to ambient ozone concentrations in receptor regions and investigate ozone sensitivities toward South Korea. Source apportionment information for ozone has been investigated mainly by using source-tagging methodology, and the effects of emission perturbations on regionally averaged surface ozone have been derived by HDDM. The traditional BF method was applied to compare both of the alternative approaches. The three approaches were used to examine the contributions of five source regions (South Korea, Japan, and three regions in China), and both anthropogenic and biogenic emissions in East Asia were applied to our modeling experiments.

2. Methodology

2.1. Source–receptor analysis methodologies

Sensitivity analysis generally measures how air concentrations respond to emissions perturbations at sources. In general, photochemical grid models have been used to estimate source contributions by performing a sensitivity case minus base case simulation. This approach is easily used for source contribution assessment when the relationship between model input and output is linear, such as sulfur transport. If emission perturbations have a nonlinear effect on concentrations because of nonlinearities in the atmospheric chemistry, such as ozone pollution, this method will underestimate source contribution. Sensitivity methods such as the BF method and the decoupled direct method (DDM) will not provide source apportionment if the relationship between model input and output is nonlinear (Yarwood et al., 2007).

On the other hand, source apportionment seeks to quantify contributions of various emission sources from specific geographic areas or of emissions sources to pollutant levels at particular locations (Cohan and Napelenok, 2011). This approach typically tracks target species separately from base model simulations and apportioned fractions of emission sources. However, the approach does not provide sensitivity results by an emission control scenario, because source apportionment seeks to determine the total contribution of each emission source to ambient concentration (Cohan et al., 2005). Source apportionment is most suited for identifying sources responsible for conditions present in a model.

The sensitivity approaches the BF method and HDDM were used in this study. The BF method calculates differences between concentrations in simulations with base case and perturbed emission scenarios from each source region or source category. In this respect, BF is one of the common methods used to estimate S–R relationships because of simplicity and ease of application in any model. However, the method is not always practical because computational cost increases linearly with the number of perturbations examined, and smaller concentration changes between simulations may be strongly influenced by numerical errors (Koo et al., 2009).

Several approaches have been attempted to use as substitutes for the traditional BF method to reduce the time needed for BF simulations within a photochemical grid model. DDM provides the same type of sensitivity information as the BF method but uses a computational method that is directly implemented in the host model (Dunker, 1981; Yarwood et al., 2007). Disadvantages of DDM include large computer memory requirements (Yarwood et al., 2007) and the nonlinearity of ozone formation. Instead, HDDM, which is an extension of DDM, is used to assess nonlinearity of ozone response to a variety of perturbations in emission rates (Cohan et al., 2005). Both DDM and BF methods are more suitable for looking at response rather than contribution to emission sources (Dunker et al., 2002; Cohan and Napelenok, 2011).

In this study, we applied HDDM (Hakami et al., 2003, 2004; Cohan et al., 2005) to assess nonlinearity of ozone response to a variety of emission perturbations. The sensitivity results calculated by HDDM have been compared with BF results to validate the HDDM estimates. HDDM was also used to explore comprehensive relationships between ozone and its perturbed precursor emissions over the S–R regions.

As the source apportionment approach, the CMAQ–OPTM source apportionment tool (Douglas et al., 2009) was used to estimate regional contributions to ozone in this study. OPTM was developed by the U.S. Environmental Protection Agency (EPA) as an applied tagging methodology for the CMAQ model (Arunachalam, 2009). OPTM provides contribution information for each emission source under unmodified simulated conditions. This method is a tracer-based technique that allows extra tagged species to be added to a grid model to track ozone or its precursors from specific sources. For ozone, aggregate modeled species are defined to track oxidants, NO_x, and VOCs (Douglas et al., 2009). These techniques are useful evaluation tools in identifying which source categories or