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Consumption Based Source Apportionment Indicates Different Regional Contributions to O_3 Concentrations and Health Effects

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ABSTRACT

China is confronting aggravated ozone (O_3) pollution, leading to adverse health impacts. This study quantifies the regional contributions to O_3 in China using two approaches; estimating ① where goods are produced (the production method), and ② where goods are consumed (the consumption method). The production method predicts higher local source contribution than the consumption method; this difference can be attributed to exports. Occurrence of high-O_3 episodes suggests a major contribution to O_3 concentration as a result of trade activities. Based on the consumption method, 9219 out of 18 532 daily premature mortalities were caused by local sources in north China, while it increased to 14 471 of the production method when neglecting contributions due to export and consumption in other regions. This study suggests that O_3 control should consider both where goods are consumed and emissions are emitted, especially taking account of international trade activities.

1. Introduction

Ozone (O_3) pollution is a severe environmental problem in China, especially in northern regions with maximum daily 8 h average (MDA8) O_3 levels up to 75 parts per billion (ppb) during the summer of 2013 [1], and even higher levels recorded later [2]. Exacerbated O_3 pollution leads to adverse health impacts [3,4] and secondary particulate matter formation [5]. The annual respiratory disease (RD) mortality due to O_3 pollution ranged from 180 000 to 320 000 during the 2010–2015 period in China [6–8], which represents a significant contribution to the annual global burden of RD mortality (ca. 6 300 000) [9].

Ground level O_3 is formed through nonlinear photochemical reactions of emitted nitrogen oxides (NO_x = NO + NO_2) and volatile organic compounds (VOCs) [10]. As meteorological conditions vary uncontrollably, reducing anthropogenic emissions is crucial in controlling O_3 formation [2,11]. Designing effective control measures is however hampered by the unknown roles of local emissions and regional transport. Although emissions occur at the source of production, responsibility should also lie with the parties who consume the products [12]. To ensure equality in pollution control, regional source apportionment of O_3 emission based on consumption rather than production is crucial, combined with a consideration of transport between regions. Recent studies have illustrated that traditional bottom-up inventories assign emissions to locations where pollutants are generated (or produced), resulting in production-based accounting [13,14]. Air pollutant emissions should be redistributed to take account of interprovincial trade.

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A considerable level of emissions are associated with the imports of eastern regions from northern and central regions, and significant emissions are outsourced to inland provinces in the import of goods by coastal provinces [13]. Recent research [15] has shown that China's international trade has a considerable impact on global air quality; export-related emissions from China contributed 12%–24% of sulfate over the western United States. However, current understanding of trade impacts on O_3 pollution is limited as the complex formation of O_3 cannot be source resolved by the brute-force methods often used for $PM_{2.5}$ [16–18].

Recently, a new O_3 source apportionment method was developed based on NO_x–VOC–O_3 sensitivity regimes [19], and used for O_3 formation attributed to local sources and regional transport [3,20]. Atmospheric transport influences O_3 pollution as O_3 and precursors emitted or produced in other regions can be readily transported across regional boundaries [21,22]. For instance, recent work reported O_3 transport from central and eastern China made a 36% contribution to the increased concentrations in north China in summer during 2014–2018 [23]. Another study has revealed extremely high O_3 levels (up to 286 ppb) at rural sites downwind site in Beijing as a consequence of atmospheric transport [24]. However, due to the complexity of O_3 formation, the comprehensive effects of trade and atmospheric transport and related health impacts are not well established.

In this study, we have applied a source-oriented chemical transport model to compare regional contributions to O_3, based on where goods are produced (the production method) and where goods are consumed (the consumption method) during summer 2013 in China. The O_3 related health impacts are also discussed. This study provides an in-depth investigation of the regional contributions of O_3 in China, which may help policy makers better understand the overall responsibility for O_3 pollution from the perspective of goods production and consumption.

2. Material and methods

2.1. The Community Multiscale Air Quality (CMAQ) model set-up and validation

A modified CMAQ model v5.0.2 with an expanded Statewide Air Pollution Research Center (SAPRC-99) photochemical mechanism was applied to simulate O_3 levels and attribute O_3 to NO_x and VOCs sources based on NO_x–VOC–O_3 sensitivity regime. The regime indicator R is defined in Eq. (1):

$$R = \frac{P_{H_2O_2} + P_{ROOH}}{P_{HNO_3}} \tag{1}$$

where $P_{H_2O_2}$ is the formation rate of hydrogen peroxide (H_2O_2), P_{ROOH} is the formation rate of organic peroxide (ROOH), and P_{HNO_3} is the formation rate of HNO_3 in each chemical step. The threshold value for the transition regime is 0.047 (change from VOC-limited to transition regime) and 5.142 (change from transition regime to NO_x-limited regime) in this study [19]. The formed O_3 is entirely attributed to NO_x or VOC sources, where O_3 sensitivity is either NO_x-limited ($R > R_{te}$) or VOC-limited ($R < R_{ts}$) regime. Moreover, O_3 is attributed to both NO_x and VOC sources when the O_3 sensitivity is transition regime ($R_{ts} < R < R_{te}$). In addition, O_3N and O_3V represent O_3 formed from NO_x and VOC, respectively. The details of this source-oriented scheme and the calculation methods of O_3N and O_3V have been described in previous studies [19,20].

The model domain includes China and its surrounding countries (Fig. S1 in Appendix A), with a horizontal resolution of (36 × 36)

Table 1
Model performance for pollutants concentration in China from June to August 2013.

Item	June	July	August	Criteria
O_3-1h (ppb)				
OBS	76.70	77.24	78.79	–
PRE	88.29	74.74	80.22	–
MNB	**0.17**	−0.01	0.04	[−0.15, 0.15]
MNE	**0.55**	**0.33**	**0.36**	≤ 0.3
MFB	−0.01	−0.11	−0.06	–
MFE	0.53	0.36	0.37	–
O_3-8h (ppb)				
OBS	63.61	67.45	97.57	–
PRE	68.35	69.23	72.84	–
MNB	**0.53**	**0.57**	**0.35**	[−0.15, 0.15]
MNE	**0.72**	**0.77**	**0.73**	≤ 0.3
MFB	0.11	0.11	0.02	–
MFE	0.24	0.26	0.30	–
NO_2 (ppb)				
OBS	15.83	14.37	14.47	–
PRE	11.82	11.20	11.32	–
MNB	−0.12	−0.14	−0.11	–
MNE	0.69	0.67	0.68	–
MFB	−0.49	−0.49	−0.46	–
MFE	0.79	0.78	0.77	–
SO_2 (ppb)				
OBS	8.90	7.82	8.88	–
PRE	11.90	11.11	11.43	–
MNB	1.19	1.09	1.04	–
MNE	1.68	1.58	1.54	–
MFB	0.09	0.08	0.05	–
MFE	0.84	0.83	0.83	–

OBS is the mean observation value and PRE is the mean prediction value; MNB: mean normalized bias; MNE: mean normalized error; MFB: mean fractional bias; MFE: mean fractional error. Data in bold exceed the criteria.

km^2 ((127 × 197) grids). The simulation was conducted from June 6 to August 23 in 2013. The meteorological inputs for the CMAQ model were generated by Weather Research and Forecasting (WRF) model v4.2, driven by the National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Model Global Tropospheric Analysis dataset (USA) (please see the Data Availability Section in Appendix A for details). The anthropogenic emissions in China including the regional production and consumption of final goods were generated by the multi-regional input–output (MRIO) model. The MRIO model contains a total of 30 regional sectors representing the 30 provinces/areas in China [12,25]. These sectors were lumped into seven regions based on where goods were ultimately consumed (consumption method, Table S1 in Appendix A). A control experiment was also conducted, where the sectors were lumped (total 7) according to regions where goods were produced (production method). The detailed information concerning the emission processes is described by Zhao et al. [12]. Emissions from other countries were obtained from the Emissions Database for Global Atmospheric Research (EDGAR) v4.3.1 (USA) [26] for both consumption and production simulations. Biogenic emissions were generated by the Model of Emissions of Gases and aerosols from Nature (MEGAN) v2.1 [27]. Open burning emissions were obtained from the Fire Inventory from National Center for Atmospheric Research (NCAR) (FINN) [28].

The WRF model performed well, though it slightly underestimated the temperature and overestimated the wind speed in this study. The gross error (GE) of temperature at 2 m height and the mean bias (MB) of wind speed exceeded the benchmark by approximately 20% and 10% (Tables S2 in Appendix A), respectively. The WRF model performance was comparable to previous studies [29-31] that provided robust meteorological inputs to the CMAQ model. The CMAQ results were evaluated against O_3, NO_2, and SO_2 observations from the national air quality monitoring network[†] (Table 1). The model predicted hourly O_3 concentrations, with mean normalized bias (MNB) values of −0.01 to 0.17. The performance satisfied suggested criteria and was similar to previous studies [11,30], although MDA8 O_3 levels were slightly overestimated, which might be caused by the limited observation data in 2013 [32]. Overall, our simulation was suitable for an O_3 study in China.

2.2. Health impact estimation

The O_3-related daily mortalities, due to the effects of atmospheric transport and trade, from non-accidental causes, cardiovascular disease (CVD), respiratory disease (RD), hypertension (Hyper), and strokes and chronic obstructive disease (COPD), were calculated based on previous studies [9,33], as shown in Eq. (2):

$$M = y_0[1 - \exp(-\beta \Delta X)] \times \text{Pop} \tag{2}$$

where M is the O_3-related daily premature mortality; y_0 is the daily baseline mortality rate, provided by the China Health Statistical Yearbook 2018 [3]; β is the concentration-response function (CRF), representing the increase in daily mortality with each 10 $\mu g \cdot m^{-3}$ increase of MDA8 O_3 [34]; ΔX is the incremental concentration of O_3 based on the threshold concentration (35.1 ppb) [35,36]; Pop is the population exposure data, provided by the LandScan global population database [37]. In this study, the population data were taken from all age groups, which may induce higher daily mortality than expected [36]. Due to the data limitation, y_0 was constant over the simulation periods with the assumption that the cases took place equally each day [38]. This may induce errors as there is spatial heterogeneity in mortality estimation [39,40], but this approach has been adopted in previous related studies [3,31,38].

[†] http://www.cnemc.cn/.

3. Results and discussion

3.1. Regional source apportionment of O_3

As shown in Fig. 1(a), the high O_3 pollution areas were mainly in north, northeast, and central regions up to 80 ppb. O_3 increased from June to August. The most prominent enhancement in north regions at rate of about 10% (from 74.80 to 81.90 ppb), which is consistent with the observation. In general, the export (international export) and inter-provincial trade play an important role in these high-O_3 regions on the basis of consumption methods (Fig. 1(b)). During the high-O_3 period (August 5 to 15), export contributed 6.39, 2.80, and 2.72 ppb MDA8 O_3 to north, central, and northeast regions. In addition, the average contribution from inter-provincial trade activities, by applying the consumption method, was ~50% higher than that using the production method (Fig. S2 and Table S3 in Appendix A). The local source is predicted to be the major contributor in both production and consumption methods. It plays a more important role in the production approach with a significant difference (up to 37%) in Yangtze River Delta (YRD) regions. In north and central regions, according to the production method, 78% and 80% of O_3 are from local sources compared with 50% and 53% from consumption (Fig. 1).

The spatial variations of MDA8 O_3N and O_3V show similar results to MDA8 O_3 (Figs. S3 and S4 in Appendix A) for both consumption and production methods. The high O_3N and O_3V areas were mainly in central and north regions, which is consistent with previous studies [19]. The local source is a more important contributor in the production method even with respect to the O_3 source apportionment results. In the case of O_3N and O_3V, the local source (from the consumption method) contributed 14.14 (49%) and 1.59 (53%) ppb, respectively, compared with 22.06 (76%) and 2.62 (87%) ppb in the production approach in the north region (Tables S4 and S5 in Appendix A). Trade activities, both export and inter-provincial, play a vital role in O_3 pollution and the conventional production method may overestimate the contribution from the local source in O_3 formation. The regional source apportionment differences in terms of O_3 precursors (NO_2 and HCHO) during summer 2013 based on production and consumption method are presented in Fig. S5 in Appendix A. It should be noted that the production method overestimated the contributions of the local source of emitted NO_2 and HCHO, especially in the YRD and north regions. The highest rates of emitted NO_2 (~0.5 $mol \cdot s^{-1}$) and HCHO (~0.3 $mol \cdot s^{-1}$) were found in YRD regions. The emission distributions of NO_2 and HCHO were higher even for the consumption method. The regional source apportionment of emitted NO_2 and HCHO was consistent with that of O_3 based on production and consumption methods.

3.2. Differences in consumption and production results

Considering the O_3 pollution periods, export and local sources are the major contributors in the consumption method, and the higher contribution from the local source are simulated in the production method regardless of the O_3 level in all key areas (Fig. 2). The periods of high and low O_3 pollution in different regions are illustrated in Fig. S6 in Appendix A. Based on the results of the consumption method, both international and inter-provincial trade play important roles in O_3 pollution. The average contributions from the export sector using the consumption method is 7.9 ppb (16%) during periods of high-O_3 in the north region, indicating the importance of trade activities. In the YRD and north regions where the average MDA8 O_3 was over 80 ppb (Fig. 1(a)), more than 50% of O_3 is attributed to trade activities. In contrast, the local source is the dominant

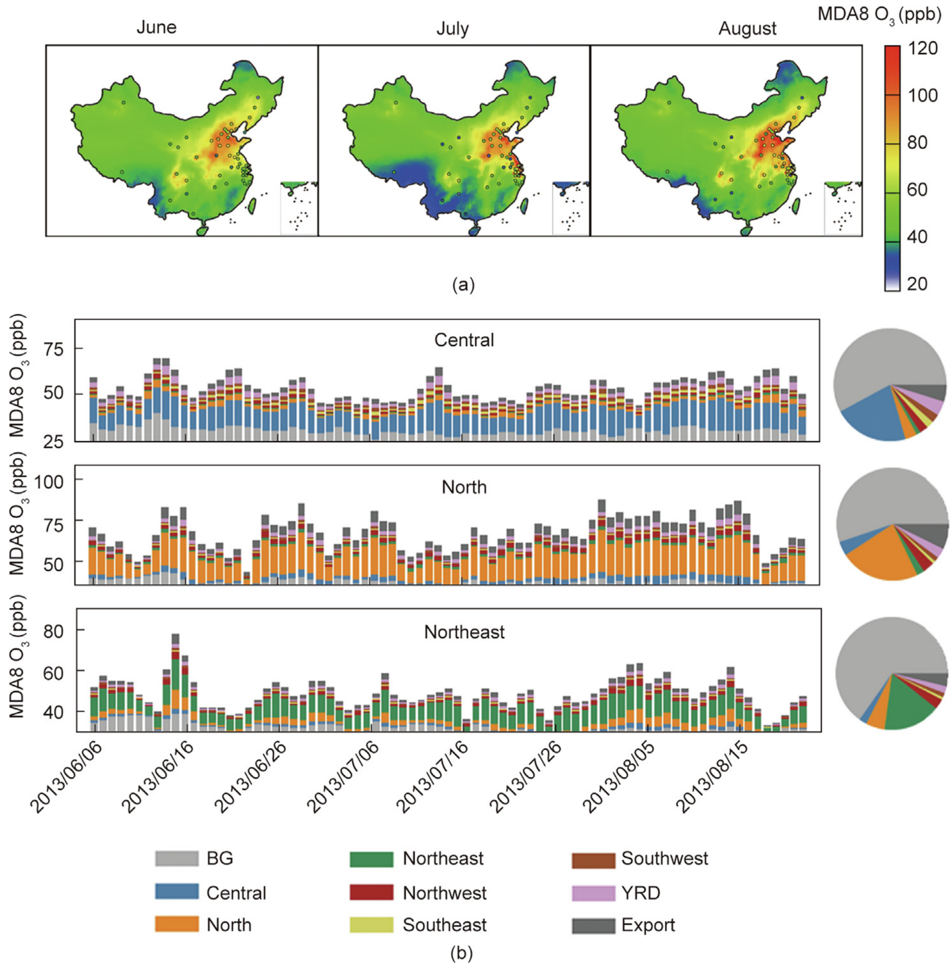

Fig. 1. (a) Observed and predicted MDA8 O_3 across China, and (b) source apportionment results of the consumption method in central, north, and northeast regions during the summer of 2013 (June, July, and August). The dots in panel (a) represent the observed MDA8 O_3 values. The pie charts in panel (b) represent the averaged regional sector contributions. YRD: Yangtze River Delta. Export represents international export. BG represents background concentrations of MDA8 O_3. The unit is ppb.

contributor in the production method. Even as high as 93% of O_3 is attributed to the local source when using the production method in the southeast region, which is 1.9 times than that of the consumption method. Reducing the emissions from trade activities may benefit a synergetic control of O_3 and $PM_{2.5}$, since previous studies have demonstrated that trade activities aggravate $PM_{2.5}$ pollution in China [41,42].

The O_3 source apportionment results show remarkable changes during the high-O_3 episodes when compared with the averaged results for both methods in different regions. Taking the consumption method, the most remarkable change is predicted in the north region with 11% decrease in the contribution from the local source. The simulation for the central region applying the production method reveals a decrease in the local source by up to 12%. It should be noted that a higher contribution due to the export sector is found during the periods of high-O_3 for most parts of China with the exception of the southeast and YRD regions. Export contributes 7.9 ppb in north regions during high-O_3 episodes, compared with 2.7 ppb during periods of lower O_3. Similar trends are found in other sectors during the high-O_3 episodes using the consumption method. In particular, the contribution of central to north regions increased from 0.1 to 4.7 ppb during high O_3 pollution. During periods of low-O_3, the contribution from local sources in the production method is still higher than that of the consumption method. In the north and central regions that have higher non-background O_3 (Fig. 2), 74% and 68% of O_3 is attributed to local source but these values drop to 39% and 46% in the consumption method.

Compared with previous reports, this study underscores the importance of formulating different strategies to control O_3 pollution in different regions as the percentage of contributions due to local sources vary greatly from region to region [11,43]. Moreover, the international and inter-provincial trade should be taken into careful consideration due to the significant contributions to O_3 pollution.

3.3. Health risks related to O_3 regional source apportionment

With the impact of inter-provincial and international trade activities, O_3 pollution causes serious health risks from COPD, CVD, Hyper, and strokes as presented in Fig. S7 in Appendix A. Our results show that the spatial distributions of premature mortality from five different diseases are similar as the health risks are mainly determined by the uneven population density and

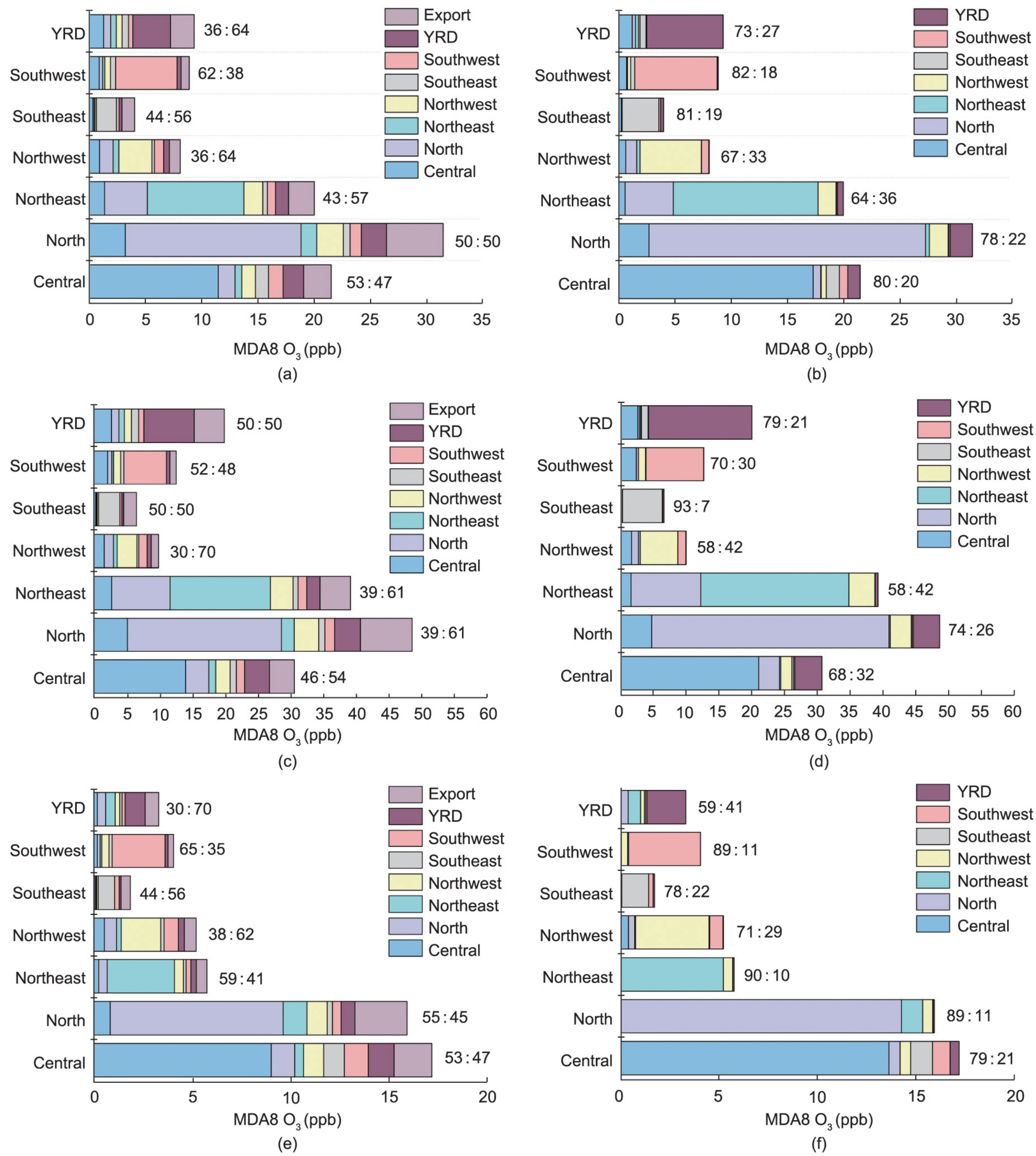

Fig. 2. (a, b) The averaged regional source apportionment of MDA8 O_3 during the entire summer period, (c, d) high-O_3 pollution periods, and (e, f) low-O_3 pollution periods based on consumption and production methods. The results given in the left (a), (c), and (e) and right columns (b), (d), and (f) are obtained from consumption and production methods, respectively. Datasets at the end of each bar represent the percentage of MDA8 O_3 attributed to local source and percentages attributed to other regions.

MDA8 O_3 levels [3,44]. The areas of high health risks are primarily in eastern China, especially in north and YRD regions with an increase in daily premature mortality from all non-accidental causes (by 200). Premature mortality from COPD, CVD, Hyper, RD, and strokes shows an increase by 50 in north and YRD regions. In addition, a high premature mortality in the Sichuan Basin can be linked to the high levels of MDA8 O_3 shown in Fig. 1. This is consistent with previous studies [4,36,45]. Our work has revealed the areas of highest health risks located in central and north regions, with premature mortality from all non-accidental causes up to 68 868 and 45 265, respectively. In addition, premature mortality from COPD, CVD, Hyper, RD, and strokes could be up to 5530, 17 798, 5457, 5650, and 589, respectively, in the central region where the population density (Fig. S7) and MDA8 levels are both high. The similarly high O_3-related premature mortality in north regions can also be explained on this basis, in line with previous studies [44,46].

Moreover, our regional source apportionment results show an obvious difference between the consumption and production methods, as presented in Table 2. Local source contributions to premature mortality amounted to 13 303 and 9219 of all non-accidental causes in central and north regions based on consumption methods (Fig. 3 and Table 2), which are considerably lower than the premature mortality of 20 046 (+51%) and 14 471 (+57%) based on production methods. The results also reveal that export and inter-provincial trade activities play important roles in serious health risks due to O_3 pollution. Exports contribute 2822, 2975, and 608 premature mortality cases in central, north, and YRD regions. In addition, inter-provincial trade activities from north, northeast, and northwest are responsible for 163, 93, 264 non-accidental causes in southwest regions based on consumption methods. The contributions to premature mortality in the southwest regions are only 38 (−77%), 10 (−89%), and 143 (−46%) based on production methods, which suggest an overestimate of local sources to health risks caused by O_3 pollution reported in the previous study [31]. We propose that control of the emissions of O_3 precursors and related health risks should be re-considered based on inter-provincial and international trade activities. In particular, the consumption activities in the YRD region induced large emissions of O_3 precursors in central and north regions (Fig. S5), which contributed to premature mortality in these regions (2146 and 1319, respectively). This phenomenon has been neglected in previous studies based on production methods [3,47]. The consumption in other locations, such as north and central regions, resulted in a much higher emission of O_3 precursors in YRD regions, causing higher premature mortality in YRD. It is necessary to pay close attention to the trade export of YRD to other regions. Similar results were found in north regions. The trade export of the north region to central and northeast regions elevated emissions of O_3 precursors and associated health risks in north regions. The central and northeast regions contributed 1881 and 820 premature mortalities to north regions. Accordingly, we should control the local emissions in the north by optimizing the trade export of north to central and northeast regions as a means of controlling O_3 levels in north regions. Detailed information regarding health risks of COPD, CVD, Hyper, RD, and strokes can be found in Tables S6–S11 in Appendix A. Based on the above analysis, we have devised a conceptual scheme to enable the comprehensive perspectives of production and consumption methods control O_3 pollution (Fig. 4). The conceptual scheme highlights differences in source apportionment of O_3 between the production and consumption methods. It also demonstrates that the neglected inter-provincial trade has resulted in unexpected increases in O_3 precursor emissions in other regions, compounding deleterious pollution through the synergistic action of atmospheric transport.

4. Conclusions and implications

In this study, the aggravated O_3 pollution was addressed by considering possible contributions due to inter-provincial and international trade activities in China during the summer of 2013 using a source-oriented chemical transport model. The conventional production method approach overestimates contributions from local sources to MDA8 O_3, and related health risks are caused by emissions due to consumption in other regions which have been neglected in previous studies. Exports play an important role in contributing 5.08 (7%), 2.30 (4%), 2.45 (4%), and 2.11 (4%) ppb in north, northeast, central, and YRD regions. The results also point out that overestimated levels vary region-by-region during periods of heavy and light O_3 pollution. The differences in daily mortality caused by illnesses related to O_3 release based on the consumption and production methods indi-

Table 2
Regional contributions to premature mortality from non-accidental causes attributed to regions where goods are consumed (consumption), and the differences (as a percentage) of regional contributions to premature mortality from non-accidental causes between the consumption and production methods. The difference is calculated by (production − consumption)/consumption.

Type	Region	Central	North	Northeast	Northwest	Southeast	Southwest	YRD	Export	Total
Consumption	YRD	370	184	143	151	162	113	978	608	2 709
	Southwest	455	163	93	264	237	2836	180	377	4 604
	Southeast	90	30	23	39	507	74	69	319	1 150
	Northwest	214	279	120	684	53	196	116	226	1 888
	Northeast	190	530	1198	233	59	101	164	320	2 795
	North	1 881	9219	820	1388	355	575	1319	2975	18 532
	Central	13 303	1709	684	1392	1363	1502	2146	2822	24 922
Difference	YRD	−3%	−55%	−45%	−75%	−3%	−85%	102%	N/A	0
	Southwest	−16%	−77%	−89%	−46%	−23%	34%	−68%	N/A	0
	Southeast	−14%	−73%	−87%	−77%	84%	−39%	7%	N/A	0
	Northwest	−28%	−18%	−43%	86%	−91%	−24%	−91%	N/A	0
	Northeast	−55%	13%	50%	−4%	−90%	−88%	−56%	N/A	0
	North	−14%	57%	−76%	−29%	−90%	−87%	−12%	N/A	0
	Central	51%	−52%	−93%	−63%	0	−43%	−41%	N/A	0

N/A: not applicable.

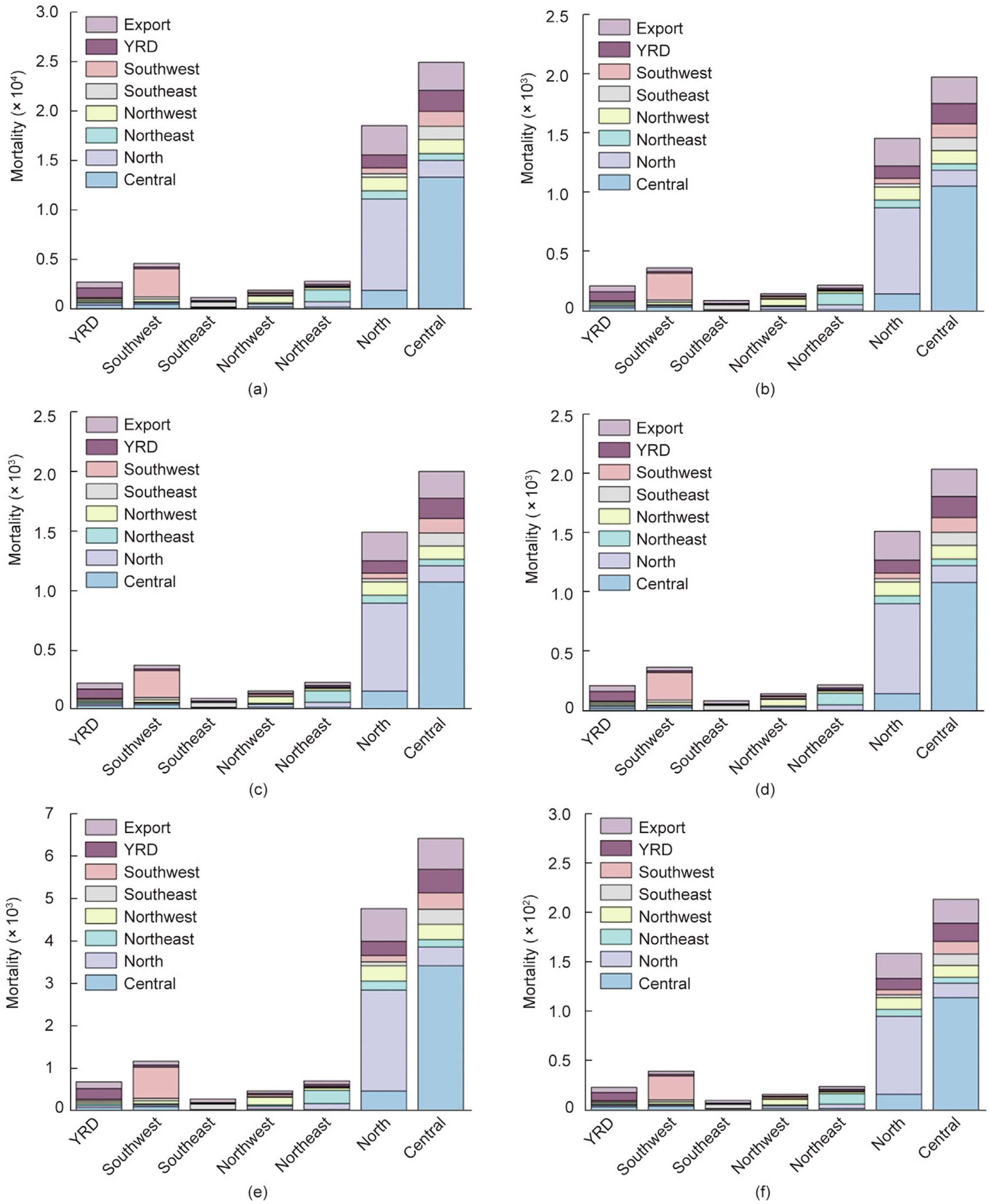

Fig. 3. Regional contributions to premature mortality from (a) all non-accidental causes, (b) hypertension, (c) COPD, (d) RD, (e) CVD, and (f) strokes based on the consumption method in seven regions during June, July, and August 2013.

cate that the health risks should be reassessed with a full consideration of trade activities. We propose that O_3 control policies should be comprehensively formulated from the perspective of both goods production and consumption, rather than assigning responsibility arbitrarily to production, especially when dealing with international trade.

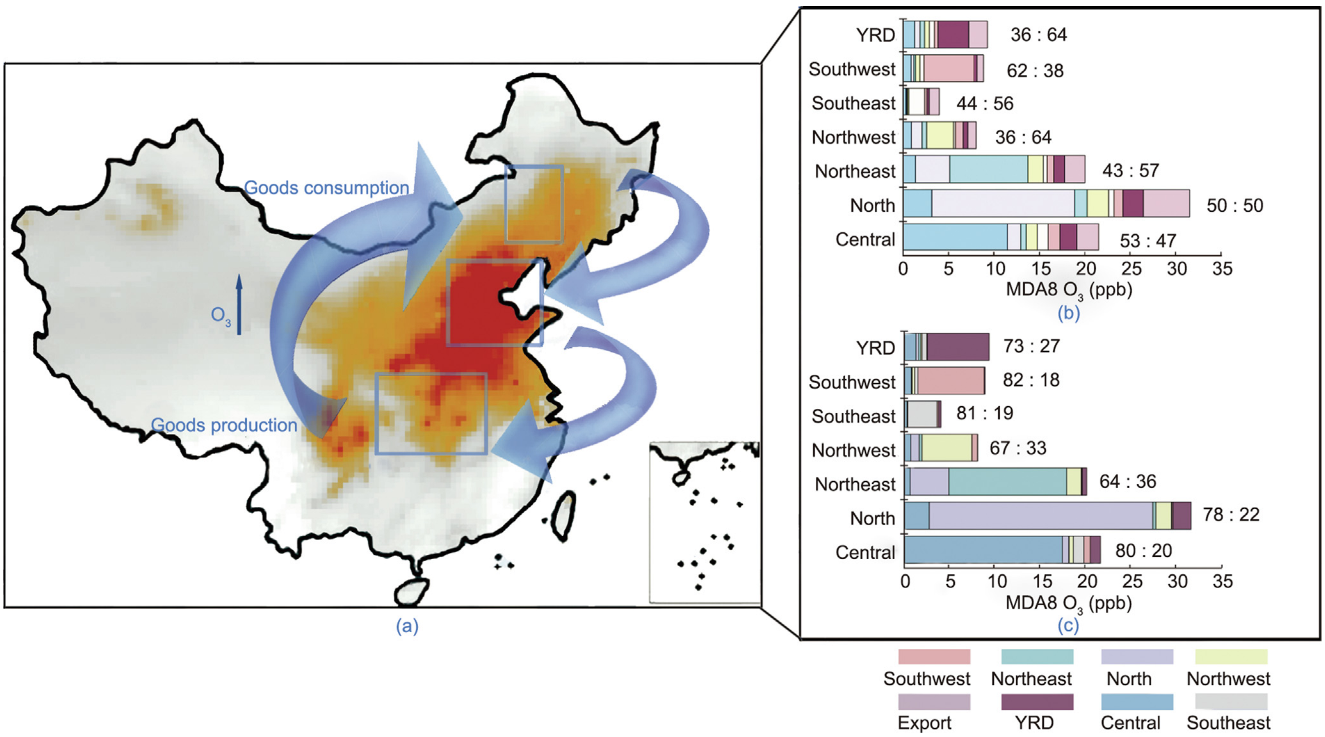

Fig. 4. (a) Conceptual framework representing the comprehensive perspectives of goods production and consumption to control O_3 pollution. The right panel presents the results of O_3 source apportionment based on (b) consumption and (c) production methods, respectively.

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Compliance with ethical guidelines

Shengqiang Zhu, Peng Wang, Siyu Wang, Guannan Geng, Hongyan Zhao, Yuan Wang, and Hongliang Zhang declare that they have no conflicts of interest or financial conflicts to disclose.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.eng.2022.11.011.

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