Inter-provincial Reliance for Improving Air Quality in China: A

Case Study on Black Carbon Aerosol

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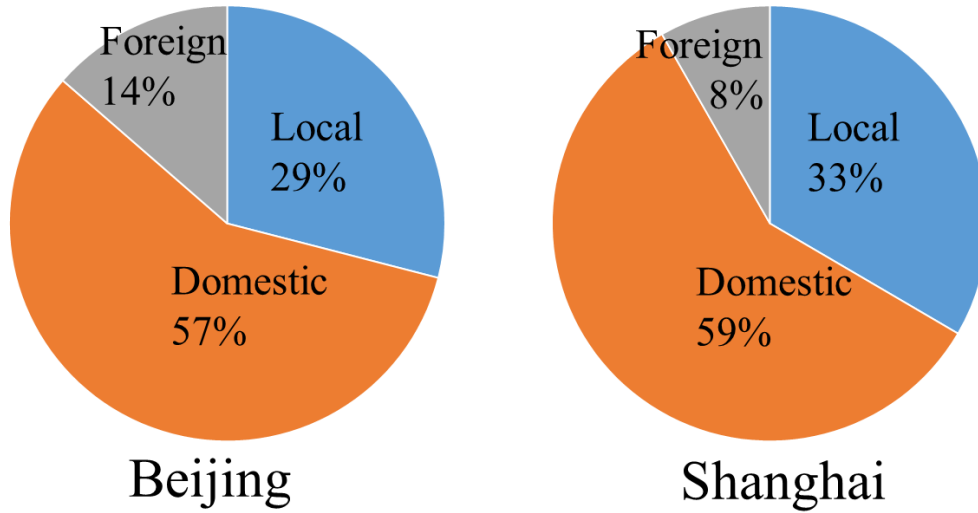[″] Y. Li and J. Meng contributed equally to this paper.

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ABSTRACT

Black carbon (BC) is of global concern because of its adverse effects on climate and human health. It can travel long distances via atmospheric movement, and can be geographically relocated through trade. Here, we explored the integrated patterns of BC transport within 30 provinces in China from the perspective of meteorology and inter-provincial trade using the Weather Research and Forecasting with Chemistry (WRF/Chem) model and multi-region input-output analysis. In general, cross-border BC transport, which accounts for more than 30% of the surface concentration, occurs mainly between neighboring provinces. Specifically, Hebei contributes 1.2 $\mu g \cdot m^{-3}$ BC concentration in Tianjin. By contrast, trade typically drives virtual BC flows from developed provinces to heavily industrial provinces, with the largest net flow from Beijing to Hebei (4.2 Gg). Shanghai is most vulnerable to domestic consumption with an average inter-provincial consumption influence efficiency of 1.5×10^{-4} ($\mu g \cdot m^{-3}$)/(billion Yuan$\cdot yr^{-1}$). High efficiencies ($\sim 8 \times 10^{-5}$ ($\mu g \cdot m^{-3}$)/(billion Yuan$\cdot yr^{-1}$)) are also found from regions including Beijing, Jiangsu and Shanghai to regions including Hebei, Shandong and Henan. The above source-receptor relationship indicates two control zones—Huabei and Huadong control zones. Both mitigating end-of-pipe emissions and rationalizing the demand for pollution-intense products are important within the two control zones to reduce BC and other pollutants.

Keywords: black carbon, inter-provincial transport, source-receptor relationship, input–output analysis

Consumption-induced BC concentrations

Foreign
14%

Local
29%

Domestic
57%

Beijing

Foreign
8%

Local
33%

Domestic
59%

Shanghai

35

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Word count for text:	**5182**
Word count for **5** figures:	**1500**=300×5
Total word count:	**6682**

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INTRODUCTION

Black carbon (BC), which is generated by the incomplete combustion of carbonaceous fuels,[1, 2] is an important combustion component of fine particulate matter (PM$_{2.5}$).[3, 4] Moreover, the scientific community has been increasingly concerned about its adverse impact on climate change, air quality and human health.[5-7] BC aerosols influence climate both regionally and globally by absorbing solar radiation, which reduces the atmospheric lapse rate and burns off cloud droplets.[3, 7] BC level varies consistently with carbon monoxide (CO), nitric oxide (NO) and other traffic-related gaseous pollutants and occupies roughly a fixed proportion of particulate matter (PM) concentration in summer and autumn.[8, 9] Additionally, pollution containing BC has been proven to have a robust epidemiological association with many types of mortality, particularly cardiovascular.[10, 11] Thus, it is acknowledged that BC may serve as an effective indicator of air quality and its health effects in helping to mitigate air pollution including PM, CO and NO.[4, 11, 12] Once emitted into the atmosphere, BC has a lifetime of 2-10 days and can be transported long distances by atmospheric movement,[13-16] indicating its well-mixed condition in lower troposphere and regional, rather than local, character.[17]

China has been the world's largest emitter of anthropogenic BC, organic matter (OM) and other PM$_{2.5}$ precursors.[18-20] In 2014, approximately 90% of the major cities in China failed to meet the national air quality standard for PM$_{2.5}$.[21] Emissions from the industrial and transport sectors have been identified as the major sources of BC and other combustion PM$_{2.5}$,[22] which has led to the need for serious emissions control in China. Recently, the "Law of the People's Republic of China on the Prevention and Control of Atmospheric Pollution" has been revised to emphasize the national target of air quality improvement from a concentration-based perspective and the supervision of pollution sources using emissions-based strategies.[23] This law also calls for collaborative efforts across administrative boundaries for emissions control and air quality improvement. Consequently, a quantitative understanding of the inter-provincial source-receptor relationship of air pollution transport and the underlying

68   economic drivers is of great importance.[24, 25]

69   Previous studies have evaluated the possible sources of air pollution for a target

70   region.[13, 26] For example, Guo et al. analyzed the observational data collected at

71   Changdao Island north of Shanghai and found that a significant amount of BC is

72   transported from Shandong and Jiangsu provinces.[27] Xue et al. utilized particulate

73   source apportionment technology (PSAT) in the CAMx model and found that

74   approximately 40% of ambient $PM_{2.5}$ concentrations in Beijing, Shanghai and Jiangsu

75   are contributed by cross-boundary transport.[28] If the source-receptor relationship were

76   accepted by the relevant provincial governments, it might be the basis for promoting

77   multi-province cooperation on emissions control.[29]

78   Apart from observed atmospheric transport, domestic trade also affects the

79   distribution of emissions to a large extent and thus changes air pollution levels

80   geographically.[30, 31] The production of traded products increases local emissions while

81   reducing the emissions in consuming regions. This trade-induced virtual emissions

82   transfer among provinces in China has been well documented for carbon dioxide (CO_2),

83   sulfur dioxide (SO_2), primary $PM_{2.5}$ and other atmospheric pollutants, which

84   demonstrates that some developed provinces shift emissions to less-developed

85   provinces by importing products.[25, 30] As with atmospheric transport, this virtual

86   transfer of emissions and the degradation of air quality via trade also lead to a source-

87   receptor relationship. When considering the location disparity between consumers and

88   producers, the emissions generated in one province might be significantly driven by

89   final consumption in a different province. Thus, for purposes of cross-provincial action

90   on air pollution control, it is equally important to identify both the sources and the

91   drivers of air pollution.

92   In this study, we used BC as a proxy to establish the source-receptor relationship

93   involving the atmospheric transport and trade-induced virtual transfer of primary

94   pollution among 30 provinces in mainland China in 2007 (excluding only Tibet, where

95   reliable data are unavailable). BC was chosen as a representative for cross-regional air

96   pollutant mitigation because of its impact on environment, regional character and

97   representative for other fine aerosol species. The model simulation of BC transport was

98 undertaken using the Weather Research and Forecasting with Chemistry (WRF/Chem)

99 model. An explicit tagging method was implemented in the WRF/Chem model to

100 efficiently track the pathways of BC transport.[32] We also used multi-region input-output

101 (MRIO) analysis to examine the virtual transfer of BC emissions resulting from trading

102 goods and services.[33] By combining both physical and virtual transfers of BC emissions,

103 we quantified the direct and indirect inter-provincial linkages in terms of pollution

104 transport. This quantification leads to feasible suggestions on the priority of BC

105 reduction and the possibility of cooperative responsibility for pollution mitigation in

106 China.

107 **METHODOLOGY AND DATA**

108 **Emission inventory and data sources.**

109 A production-based emissions inventory was developed by multiplying the energy

110 consumption data and BC emission factors.[34, 35] Energy consumption data for 30

111 provinces in China (as listed in Table S1 in the supporting information) were derived

112 from provincial statistical yearbooks and energy balance tables from the 2008 Chinese

113 Statistical Yearbooks (Data were based on the investigation of year 2007) for each

114 province.[36] We aggregated the provincial BC emissions into 17 sectors (listed in Table

115 S2 in the supporting information) to conform with the Chinese MRIO Table.[37] Emission

116 factors for 8 types of energy (i.e., coal, coke, gasoline, kerosene, diesel, fuel oil,

117 liquefied petroleum gas and natural gas) were obtained from previous studies (listed in

118 Table S3 in the supporting information).[34, 35] The derivation of production-based BC

119 emissions attributable to energy consumption for province f is expressed as:

120 $$C_p^f = \sum_{i=1}^{17} \sum_{m=1}^{8} E_{i,m}^f \times EF_{i,m} \tag{1}$$

121 where $E_{i,m}^f$ is the energy consumption of fuel m in sector i, province f; $EF_{i,m}$ is the

122 emission factor of fuel m in sector i.

123 Here, we considered only industrial BC emissions from all 17 aggregated

124 economic sectors, because industrial emissions can "flow" in inter-provincial trade.

125 According to the Chinese MRIO Table, every particular sector has a more or less

126 monetary output to support non-local industry. By contrast, residential energy

127 consumption cannot "flow" in trade, and was thus excluded in our analysis. We used

128 the industrial BC emissions in WRF/Chem modeling for consistency with the MRIO

129 analysis by mapping the emissions with high spatial resolution.[34] We also conducted

130 additional simulations using revised all-source anthropogenic BC emission inventory

131 from Wang et al. for model evaluation.[34]

Model description and configuration.

133   WRF/Chem is a meteorological model that enables the simulation of atmospheric

134 phenomena across scales ranging from meters to thousands of kilometers.[38]

135 WRF/Chem includes chemical processes such as emissions, gas/aqueous phase

136 chemistry and dry/wet deposition.[39] WRF/Chem has been widely applied to simulate

137 the transport of BC and its radiative impact.[38, 40, 41] Real meteorological data are used

138 as the initial and lateral boundary condition input for the WRF/Chem model to simulate

139 the physical transport of BC aerosols. Here we applied a data set from the NCEP FNL

140 Operational Model Global Tropospheric Analyses, which provide data every six hours

141 for the period from December 16st, 2006, to December 31st, 2007 for model simulation.

142 The first two weeks of the simulation were used for model spin-up.

143   To quantify source-receptor relationships among the 30 provinces, an explicit

144 tagging technique was used in WRF/Chem to avoid modifying BC emissions.[42] This

145 method differs from the traditional sensitivity approach to avoid reducing BC emissions

146 that may strongly disturb the local climate. Similar approaches have been previously

147 applied in global models to estimate the long-range transport of BC, OC and PM$_{2.5}$

148 between continental regions.[13, 32, 43] In this tagging approach, two classes of BC tracers

149 are used for each "tagged" region. One is for hydrophobic BC, which represents freshly

150 emitted BC species, and the other is for hydrophilic BC, which represents aged BC and

151 has sufficient soluble coating to behave as cloud condensation nuclei (CCN). Therefore,

152 30 non-overlapping geographical regions were tagged individually with additional

153 variables to track their transportation and transformation until deposition. Tagged BC

154 has the same physical and chemical properties as untagged BC, and the model thus

155 accurately predicts the pathways of BC dispersion and its influence on surface

156 concentration.

157   We use the WRF/Chem model to track the inter-provincial source-receptor

158 relationships for BC in 2007 with a 0.2°×0.2° horizontal resolution. In general, the

159 model agreed within a factor of 2 with the observations (As shown by Figure S1 in the

160 supporting information - observational data were collected from published literatures.[44-

161 57] The spatial distribution of the data is shown in Figure S2). The output results were

162 archived hourly and used to calculate the average surface concentrations for a province

163 over a given period of time for analysis.

164 **Multi-region Input-output (MRIO) Analysis.**

165   Originating from Leontief,[58] input-output analysis has been widely used to link

166 global and regional environmental issues with final consumption.[31, 33] In the past decade,

167 environmental MRIO analysis has been developed to quantify emissions transfer via

168 inter-regional trade.[33, 59, 60] Here, we used the Chinese MRIO Table from 2007 that was

169 developed by Liu et al. to quantify BC emissions embodied in traded products.[37] The

170 MRIO table consists of three parts. Part One is the intermediate input/output for 17

171 sectors in 30 provinces. Part Two consists of provincial final consumption (i.e., urban

172 household consumption, rural household consumption, government consumption and

173 investment) and international export. Part Three consists of production-based BC

174 emissions for 17 sectors in 30 provinces.

175   For the entire system covering all provincial economies, we have the following

176 balance of monetary flows:

177      $X = AX + Y$ (2)

178 where X is a vector representing total monetary output for every province, A is a matrix

179 with its elements defined as intermediate input to produce a unit output, and Y is a

180 vector representing the total output of final consumption and international export in

181 each province.

182   Consumption-based BC emissions can be obtained by introducing emission

183   intensity *EI*:

$$C_c = EI(I - A)^{-1}Y_c \tag{3}$$

185   where *EI* is a vector with its elements defined as the direct BC emissions per unit of

186   economic output, $(I - A)^{-1}$ is the Leontief inverse matrix and Y_c is the final

187   consumption.

188   This basic formula can be further used to quantify emissions from the production

189   of traded products. For instance, BC emissions embodied in the products exported from

190   province *f* to province *s* can be calculated as

$$C_c{}^{fs} = EI^f (I - A)^{-1}Y_c^s \tag{4}$$

192   where EI^f is a vector of BC emission intensity for province *f* but zero for all others

193   and Y_c^s is the final consumption of province *s*.

RESULTS

Physical Transport of BC via Atmospheric Movement.

196   Figure 1 shows the major cross-boundary influence pattern of the area-weighted

197   annual mean surface BC concentration caused by industrial emissions. The annual

198   mean surface BC concentrations range from 0.025 µg·m⁻³ (Qinghai) to 5.7 µg·m⁻³

199   (Shanghai). Shanghai and Tianjin (4.2 µg·m⁻³) have the highest BC concentration.

200   Major local sources of pollution for these two coastal megalopolises are traffic and

201   transport sectors (as suggested by Figure S3), while emissions in their contiguous

202   provinces also exert considerable influence. Industry-dominant provinces including

203   Shandong (2.8 µg·m⁻³), Henan (2.9 µg·m⁻³) and Liaoning (2.0 µg·m⁻³) also have heavy

204   BC concentrations. Moreover, provinces with heavier BC pollution are likely to be

205   located along or near the coastline.

206   Provincial BC concentrations are profoundly influenced by trans-boundary

207   transport. The reciprocal effect between two contiguous provinces whose emissions

208   share resemblances is generally comparable. It is particularly noticeable between Hebei

209   and Shandong, Shandong and Henan, Jiangsu and Anhui, and Jiangsu and Shanghai,

210   where approximately 10% of the BC concentrations in these provinces are contributed

by one another. The northern provinces tend to be net contributors to the pollution load of the more southerly provinces in eastern China. Remarkably, Hebei is responsible for 0.59 $\mu g \cdot m^{-3}$ (24%) and 1.2 $\mu g \cdot m^{-3}$ (28%) surface BC concentrations in Beijing and Tianjin, respectively. It is also responsible for 0.13 $\mu g \cdot m^{-3}$ (7%) and 0.17 $\mu g \cdot m^{-3}$ (6%) of the BC concentration in Shanxi and Henan, respectively. Whereas Shanxi contributes 0.20 $\mu g \cdot m^{-3}$ (22%) BC in Shaaxi, and Henan contributes 0.23 $\mu g \cdot m^{-3}$ (19%) of the BC concentration in Hubei and 0.17 $\mu g \cdot m^{-3}$ (8%) in Anhui.

Virtual Transfer of BC via Inter-provincial Trade.

Figure 2(a) shows the comparison of total production-based and consumption-based BC emissions in 2007 for 30 Chinese provinces. Total industrial BC emissions amount to 894 Gg in China in 2007, which is consistent with previous studies.[34, 61, 62] From the production perspective, Shandong ranks first with emissions of 79.7 Gg, followed by Henan (73.5 Gg), Shanxi (61.1 Gg) and Hebei (60.3 Gg). Provincial consumption-based BC emissions present a different distribution pattern, with 741 Gg (83%) emissions induced by domestic demand. This percentage is comparable to previous results on primary $PM_{2.5}$ and gaseous pollutants including SO_2 and NO_x.[25, 63] Except for Shandong (contributing 64.7 Gg emission), the southern provinces, including Zhejiang (57.2 Gg), Jiangsu (55.0 Gg) and Guangdong (51.2 Gg), hold the top positions. Remarkably, the Yangtze River Delta contributes 20% of the total embodied BC emissions, although its domain area is less than 2% of the total area. Consumption-based emissions consist of emissions from four types of final consumption. Investment is the dominant motor driving industrial BC emissions for 29 provinces (the exception is Xinjiang), contributing approximately 40%−70% of the total consumption-based BC emissions. Urban household consumption is the second largest driver of BC emissions, ranging from 15% in Shanxi to 43% in Tianjin. Government consumption and rural household consumption account for the remaining 15%.

The difference between production-based and consumption-based BC emissions indicates that emissions are transferred via trade. Figure 2(b) illustrates net emissions

240 transfer through trade (only the largest fluxes between provinces are shown). 13 of 30

241 provinces are net emissions importers, and the other 17 provinces are net exporters. Net

242 importers are mainly industry-dominant provinces such as Hebei (23.2 Gg), Shanxi

243 (18.3 Gg), Liaoning (17.3 Gg) and Henan (15.7 Gg). Their industrial activities and

244 associated emissions enhanced by trade support consumption across the country,

245 particularly for a few developed provinces. Conversely, Zhejiang (24.1 Gg), Shanghai

246 (16.4 Gg), Beijing (14.5 Gg) and Guangdong (13.8 Gg) are major BC exporters. They

247 behave as exporters in trade with almost all other provinces, whereas the larger flows

248 more often end up in Hebei and Henan.

249 The pattern of major flows is from southeastern China to the North China Plain

250 (NCP) geographically and from developed to less-developed provinces economically.

251 The demand-driven flows can be categorized into three types based on the economic

252 strength of emission exporter and importer, with the dominant pattern being from a

253 province with abundant capital to a province owning heavy industry (Figure 2b). First,

254 the largest BC emissions transfer occurs from Beijing to Hebei (with 4.2 Gg BC

255 emissions being relocated), followed by a flow of 4.1 Gg emissions from Zhejiang to

256 Hebei. Second, shifts in emissions between contiguous provinces of comparable

257 economic strength are also noticeable. They occur noticeably within the Yangtze River

258 Delta and northeastern provinces, including Heilongjiang, Jilin and Liaoning, which

259 indicate intimate economic relationships between contiguous provinces. The typical

260 emission flows in this category are from Shanghai to Jiangsu (1.2 Gg) and from Jilin to

261 Liaoning (2.7 Gg). The third type of flow is from industrial provinces to resource-rich

262 but less-developed provinces, suggesting the need for inputting fundamental raw

263 materials for industrial activities. For example, 1.4 Gg of emissions are transported

264 from Hebei to Shanxi. In general, trade-induced emission flows across China occur

265 from south to north and coastal to inland, exhibiting a reversed source-receptor pattern

266 to BC dispersion via atmospheric transport.

267 Apart from revealing emission flows from a regional aspect, MRIO can also

268 explore the sectors that undertake the transfer of BC emissions in trade.[31] Figure 3

269 shows the sector-specific BC emission transfer embodied in inter-provincial trade. At

270 the national level, nearly half of these trade-relevant emissions are caused by the

271 production of intermediate products. This ratio is even higher in Shanxi, Gansu,

272 Qinghai, Xinjiang and Yunnan, where more than 80% of the emissions are caused by

273 the massive quantities of low-value-added raw materials and energy that are produced

274 for export. By contrast, Zhejiang and Jiangsu have the highest proportion of

275 intermediate goods from the import aspect because these provinces lack natural

276 resources but are advanced in processing capabilities.

277 Moreover, final use accounts for the remaining 15%–55% of trade-embodied

278 emissions, in which heavy industry (including the petro-chemical, nonmetallic mineral

279 products, and metallic mineral products) plays a dominant role in most provinces due

280 to its intensive energy consumption. In addition to heavy industry, exporting

281 agricultural products induces salient BC emissions in Hubei and Sichuan provinces;

282 mining and washing are responsible for most trade-relevant BC emissions in Shanxi

283 and Liaoning provinces. However, for Jiangsu and Zhejiang, light industry such as

284 textile and timber processing is key to generating BC emissions. With regard to

285 imported products, less-developed provinces tend to have a higher proportion of

286 emission output in high-value products of light industry, whereas the provinces scarce

287 in energy and raw materials are likely to depend on products of mining and washing in

288 other provinces.

289 **Surface BC Concentrations from a Production and Consumption Perspective.**

290 By combining atmospheric BC transport and emission flow in trade, the source of

291 surface BC concentrations can be classified according to their on-site emission region

292 and the final consumer of the relevant products. With regard to whether BC is emitted

293 locally or from other provinces, BC concentration can be classified as either local or

294 domestic production. Meanwhile, from a consumption-based perspective, BC

295 concentration can be referred to as local, domestic or foreign consumption. We specify

296 "domestic consumption" as the consumption from the other 29 provinces, and "foreign

297 consumption" as the internationally-exported products from the province where

298 production-based BC is generated. From an on-site emission perspective, 9 provinces

299 (i.e., Hubei, Anhui, Jiangxi, Hainan, Guangxi, Chongqing, Guizhou, Shaanxi and

300 Qinghai) have a share of domestic-production concentration (i.e., originated from the

301 other 29 provinces, namely the green, yellow plus light grey portions in Figure 4) over

302 50%. Others are typically within 30%−50%, with the lowest (3%) being Xinjiang. This

303 indicates the importance of local emissions, but trans-boundary transport is also a

304 noteworthy contributor. With regard to the key provinces for air pollution mitigation,

305 Beijing has a proportion of domestic-production BC surface concentration equal to 42%,

306 Tianjin 45%, Shanghai 22%, Jiangsu 39%, Zhejiang 41% and Guangdong 40%. BC

307 concentration originating from other provinces but induced by local consumption (i.e.,

308 the green portion in Figure 4) is very small (<3%) and mainly driven by neighboring

309 provinces.

310   From the consumption perspective, final demands within mainland China on

311 average induce 82% of provincial surface BC concentration across China. Domestic-

312 consumption-induced concentration (i.e., the red plus yellow portions in Figure 4) is

313 greater than local-consumption-induced concentration (i.e., the blue plus green portions

314 in Figure 4) for all provinces, which indicates the profound influence of inter-regional

315 trade and trans-boundary transport on concentration. The proportion of BC

316 concentration generated locally but induced by domestic consumption ranges from 4%

317 in Hainan to 50% in Xinjiang and reflects the comparative industrial scales for

318 supporting local living standards or for securing economic growth via exports. This

319 proportion is above 30% for typical emission importers. Particularly, 43% and 41% of

320 the concentration in Hebei and Liaoning, respectively, is contributed by the local

321 emissions induced by domestic consumption. Meanwhile, for emissions-exporting

322 provinces, such as Beijing, Zhejiang and Guangdong, this proportion falls to near 20%.

323 However, Shanghai has a higher percentage (40%) of local-domestic concentration

324 because of its considerable share of industry to support domestic consumption.

325 **Source-Receptor Relationship from Combined Production and Consumption**

326 **Perspectives.**

327   To combine the influence of atmospheric transport and inter-provincial trade on

328   surface BC concentrations and emissions, which is informative for judging the priority

329   of cooperative action for pollution mitigation, we introduce three source-receptor

330   indicators shown in Figure 5.

331   Figure 5(a) shows the emission-concentration relationship that is defined as the

332   annual averaged surface BC concentration over a receptor resulting from a unit of

333   emission in a source region (in $(\mu g \cdot m^{-3})/(Gg \cdot yr^{-1})$).[29] This atmospheric transport

334   efficiency is calculated by dividing the source-produced concentration in a receptor by

335   total annual emission in the source region. The colored chart shows a pattern in which

336   a receptor is more sensitive to its own emissions than to domestic emissions and to

337   upwind contiguously located sources than to remote ones. Normalized BC

338   concentrations resulting from local emission range from 0.0047 $(\mu g \cdot m^{-3})/(Gg \cdot yr^{-1})$ in

339   Xinjiang to 0.20 $(\mu g \cdot m^{-3})/(Gg \cdot yr^{-1})$ in Shanghai, which is mainly determined by the

340   emission density. Non-local contributions to BC concentration from neighboring

341   provinces is typically 1-2 orders of magnitude smaller than local emissions but more

342   than 1-2 orders of magnitude larger than emissions from remote provinces. Provinces

343   within Jing-Jin-Ji and the Yangtze River Delta share a close relationship in BC

344   concentration through atmospheric transport. For instance, 1 Gg annual emission in

345   Beijing and Hebei can increase the surface BC concentration in Tianjin by 0.022 and

346   0.019 $\mu g \cdot m^{-3}$, respectively. Similarly, the bilateral influence between Jiangsu, Shanghai

347   and Zhejiang ranges from 0.003 to 0.013 $(\mu g \cdot m^{-3})/(Gg \cdot yr^{-1})$.

348   Figure 5(b) shows the trade-induced consumption-emission relationship that is

349   defined as the production-based BC emissions of a receptor associated with a unit of

350   domestic consumption from a source (in tons of BC per billion Yuan), in which

351   consumption includes the sum of four final consumption.[30] This BC intensity is

352   calculated by dividing the source-induced BC emission in a receptor by total annual

353   final consumption in the source region. Unlike Figure 5(a) in which the pattern of

354   atmospheric transport exhibits a diagonal distribution, the consumption-emission graph

355   reflects a column-like distribution, which indicates that massive amounts of BC

356   imported from almost all other provinces via trade into some industry-dominant

357   provinces (e.g., Hebei, Henan, and Liaoning). Normalized production-based BC

358   emissions induced by local consumption range from 3.4 tons per billion Yuan in Tianjin

359   to 53.4 tons per billion Yuan in Shanxi. Generally, this value is higher in provinces with

360   massive energy consumption than in developed provinces with strict environmental

361   laws. However, those developed provinces relocate significant amounts of emissions to

362   other provinces. The proportion of emissions caused in other 29 provinces is

363   comparable to the proportion of local on-site emissions in developed provinces. Every

364   billion Yuan of consumption in Tianjin produces 5.6 tons of BC emissions in Hebei, 1.6

365   times its own local emission intensity. Similarly, Hebei and Henan receives 2.4 and 2.1

366   tons of BC emissions respectively for every billion Yuan of consumption in the Yangtze

367   River Delta. Heilongjiang and Jilin shift 7.2 and 8.4 tons of BC emissions, respectively,

368   to Liaoning for every one billion Yuan of consumption.

369   Figure 5(c) shows the consumption-concentration relationship, i.e., annual

370   averaged surface BC concentration in a receptor resulting from a unit of domestic

371   consumption in a source (in $(\mu g \cdot m^{-3})/(billion\ Yuan \cdot yr^{-1})$), which considers the joint

372   influence of trans-boundary transport and inter-regional trade on surface BC

373   concentration together. This consumption influence efficiency is calculated by

374   concentration in a receptor caused by on-site emissions in 30 provinces that are induced

375   by a unit of annual consumption in a source. The pattern of the graph is a combination

376   of Figure 5(a) and (b), showing both aggregated groups along the diagonal and column-

377   like distribution of eminent contributions. The bilateral influence of domestic

378   consumption within Jing-Jin-Ji and the Yangtze River Delta is more than 8×10^{-5} $(\mu g \cdot m^{-}$

379    $^{3})/(billion\ Yuan \cdot yr^{-1})$. Northeastern provinces also show intimate internal relationships

380   regarding both atmospheric transport and inter-provincial trade. Surprisingly, Tianjin

381   and Shanghai are the two provinces most vulnerable to consumption-based emissions

382   in other provinces despite being net BC exporters in inter-provincial trade. The average

383   surface BC concentration resulting from a unit of consumption in a non-local source

384   region in Shanghai and Tianjin is 1.5×10^{-4} $(\mu g \cdot m^{-3})/(billion\ Yuan \cdot yr^{-1})$ and 1.4×10^{-4}

385    $(\mu g \cdot m^{-3})/(billion\ Yuan \cdot yr^{-1})$, respectively. This phenomenon can be attributed to the

386   considerably high BC concentration resulting from a unit of local-production emission

387   and a prevailing proportion of local-production emission induced by domestic

388 consumption than by local consumption. Although they are densely urbanized

389 metropolises, Tianjin and Shanghai have a considerable scale of secondary industry,

390 which induces a massive emission import (see S3 in the supporting information). In

391 addition, the source-receptor relationship between consumption and concentration is

392 also noticeable within some emission exporters and importers in inter-provincial trade.

393 Per-billion Yuan annual consumption from a source province can lead to an

394 approximately 8×10^{-5} $\mu g \cdot m^{-3}$ increase in BC concentration in a receptor province. In

395 particular, BC concentrations in Liaoning and Hebei are largely affected by domestic-

396 consumption emissions particularly from the Jing-Jin-Ji area and Shanxi, whereas the

397 concentrations in Anhui and Jiangsu are affected by emissions from the Yangtze Delta

398 area and Anhui. In Henan and Shandong, BC concentration is sensitive to consumption

399 in all the source provinces mentioned above. These provinces may show a close

400 relationship for air pollution control.

DISCUSSION

402 Using WRF/Chem modeling and environmental MRIO analysis, we quantified the

403 source-receptor relationship of atmospheric transport and trade-induced geographical

404 relocation of BC emissions. By combining the dual effects, we traced the influence

405 from both producer and consumer perspectives on BC surface concentration for each

406 province in China. The results can provide insights into collaborative efforts on air

407 pollution control for policy makers.

408 The source-receptor relationship of physical BC transport among provinces is

409 largely determined by both the amount of BC emissions and the direction of prevailing

410 winds. Depending on locations, more than 20% of surface BC concentration may

411 originate from a neighboring province, particularly for provinces such as Tianjin and

412 Hubei, which are located downwind contiguously of major BC source provinces (such

413 as Hebei and Henan). By contrast, provinces such as Hebei, Shanxi, Henan, Zhejiang

414 and Guangdong transport substantial BC pollution to their neighbor provinces. Notably,

415 in inland China, where the north wind is dominant particularly in autumn and winter

416 because of the influence of the Siberian High, BC transport typically occurs more

417   usually from north to south. In southeastern coastal China, however, where airflow is

418   driven by the Hawaiian High, BC transport occurs mainly northwestward during the

419   summer. Combining these two factors, the proportion of surface BC concentration

420   caused by the other provinces varies from 3% to 71%. Provinces such as Hubei, Shaanxi,

421   and Hainan are vulnerable to non-local emissions because of their relatively small scale

422   of industry compared with their neighbors with massive industrial production. Beijing,

423   Shandong, Jiangsu and other provinces with on-site emissions comparable to those of

424   their neighbors are also sensitive to domestic-production emissions. Several provinces,

425   such as Xinjiang, Liaoning and Shanghai, have a percentage above 75% of the local-

426   production surface BC concentration owing to their location and the total amount of

427   BC emissions, which indicates a local-production dominant situation.

428   Unlike atmospheric transport, which is driven by natural forces, domestic trade

429   relocates BC in a different way. Beijing, Tianjin, Guangdong and the Yangtze River

430   Delta are more likely to outsource BC emissions via inter-provincial trade to industrial

431   provinces including Hebei, Henan, Shanxi and Liaoning. For developed provinces such

432   as Beijing and Shanghai, consumption-based BC emissions can be double the

433   production-based BC emissions, whereas in industry-dominant provinces such as Hebei

434   and Shanxi, net BC emissions transferred via trade amount to approximately 30% of

435   their production-based emissions. Surface BC concentration generated by local

436   emission but induced by domestic consumption can account for more than 30% of the

437   total concentration in these provinces. In addition, three northeastern provinces show

438   tight economic connections, with Liaoning playing the major role of emissions importer.

439   This imbalance in inter-provincial trade may be due largely to the enormous disparity

440   in wealth and economic structure among provinces.[64] Although emission transport via

441   trade is bilateral, developed provinces are more likely to import low value-added

442   commodities in the heavy industry, mining and washing, and agricultural sectors from

443   less-developed provinces according to their mainstay industry, while exporting

444   technology-containing commodities from light industry.

445   Combining these two aspects, it is reasonable to say that the patterns of

446   atmospheric transport and inter-provincial trade are the opposite of one another.

447 Emission flows from Beijing and Tianjin to Hebei are transported in reverse to

448 influence the local BC concentration. Similar patterns can also be observed from

449 Shanghai to Jiangsu and Zhejiang. Moreover, consumption in developed provinces

450 located along the southeastern coastline increases BC concentration and exerts adverse

451 influence on air quality in those emission input industry-dominant provinces, mainly

452 on NCP via trade. These phenomena may serve as a major motivation for the close

453 cooperation between provinces on air pollution control as advocated in the "Law of the

454 People's Republic of China on the Prevention and Control of Atmospheric Pollution."

455 By introducing advanced technology from developed provinces to industry-dominant

456 provinces and by taking cross-regional governance into consideration, the supportive

457 provinces benefit in that their surface pollutant concentration caused by domestic-

458 production emissions is reduced. Meanwhile, cooperation contributes to reducing

459 surface BC concentration in the emission source, which may compensate for the

460 pollutant transferred via inter-provincial trade. Overall air quality in China can also be

461 enhanced because downwind provinces suffer less from trans-boundary emissions.

462 Generally, neighboring provinces have a more intimate relationship concerning both

463 trans-boundary transport and inter-provincial trade, which indicates their optimal

464 prospects for joint efforts on mitigating air pollution. Two partly overlapping control

465 zones for collaborative action on air pollution mitigation with prior concern are

466 promoted according to our study. The Huabei control zone, led by the Jing-Jin-Ji area,

467 together with Shandong, Shanxi, Liaoning and Henan has a close relationship with

468 respect to both trans-boundary transport and virtual transfer of emissions. Liaoning is

469 also intimate with Jilin and Heilongjiang and may implement multilateral supervision

470 action. Meanwhile, for Shandong and Henan, cooperation with the Yangtze River Delta

471 (i.e., Jiangsu, Shanghai and Zhejiang) and Anhui can also achieve enhanced efficiency

472 in emission mitigation. These 6 provinces comprise the Huadong control zone.

ASSOCIATED CONTENT

473 **ASSOCIATED CONTENT**

474 **Supporting Information.** Tables S1-S4, Figures S1-S3 include provinces and sectors

475 information, emission factors of BC for 8 fuel types, sectoral distribution of provincial

production-based BC emissions and the evaluation of model simulation with observations. Supporting data contain simplified Chinese MRIO Table (2007) for 30 provinces and 17 sectors. This material is available free of charge via the Internet at http://pubs.acs.org.

ACKNOWLEDGEMENTS

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Notes

The authors declare no competing financial interest.

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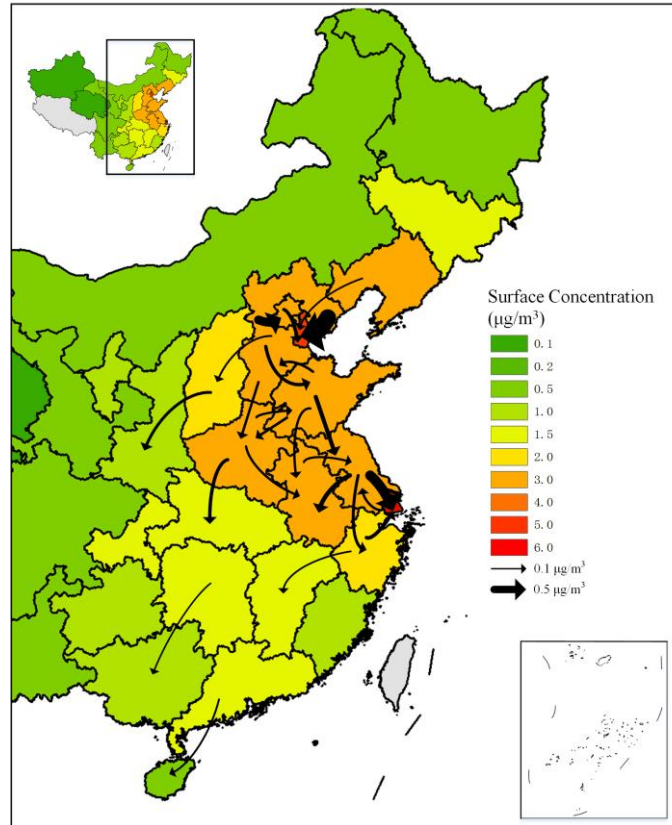
Figure 1. Largest surface BC concentration contribution via atmospheric transport within 30 provinces. The colors in the map indicate annual mean surface BC concentration. Arrows on the map reflect a typical contribution above 0.1 μg·m⁻³. The thickness of the arrows indicates the relative magnitude of the absolute inter-provincial contribution of surface concentration.

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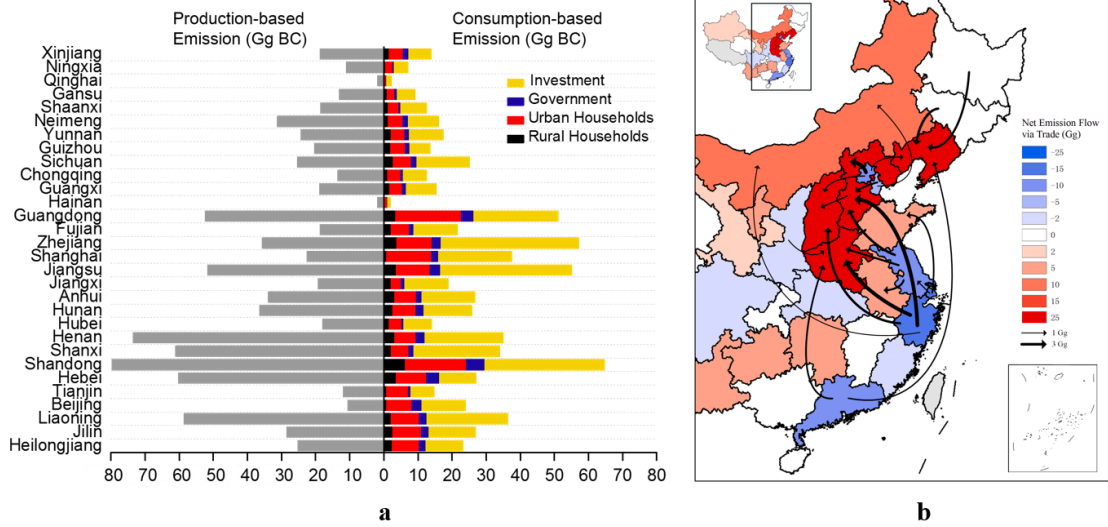
Figure 2. (a) A comparison between production-based and consumption-based BC emissions. Consumption-based BC emissions are categorized into 4 types based on final consumption; (b) Largest net fluxes in "traded" BC emissions among 30 provinces. Color in the map indicates total net emission budget (emission imports minus exports) via trade. Red indicates an emission importer, i.e., more BC is emitted due to the inter-provincial trade. Blue indicates an emission exporter. The arrows reflect typical cross-border net emission flows above 1 Gg in inter-provincial trade. The thickness of the arrows indicates the relative magnitude of the net BC emissions transferred between provinces.
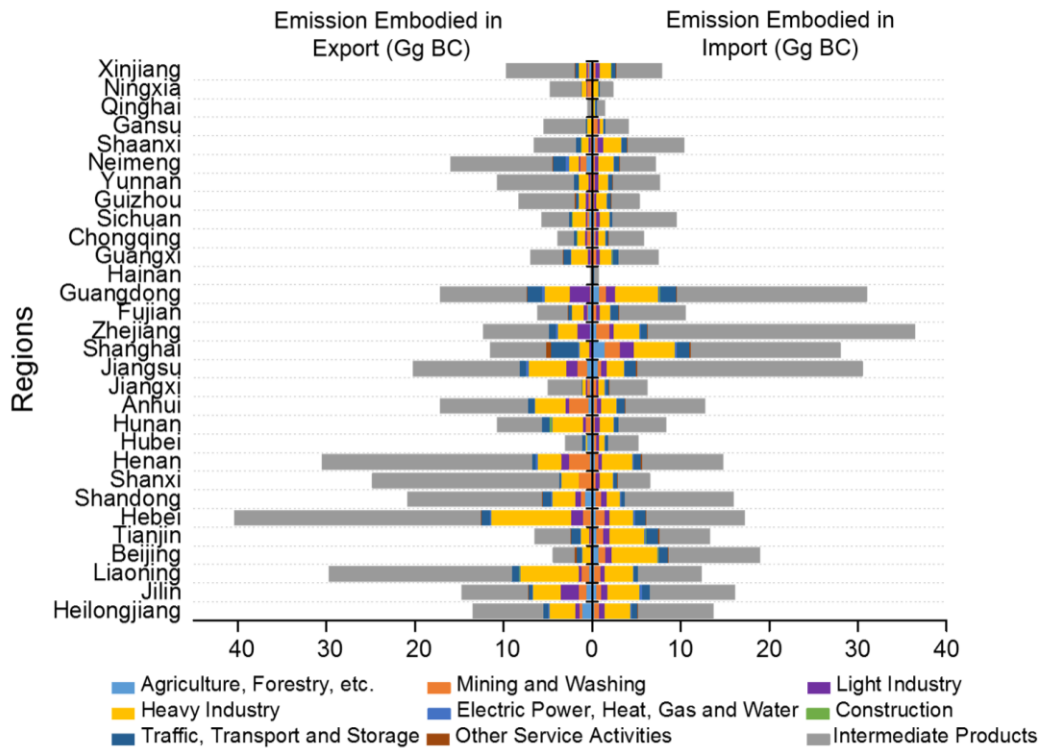
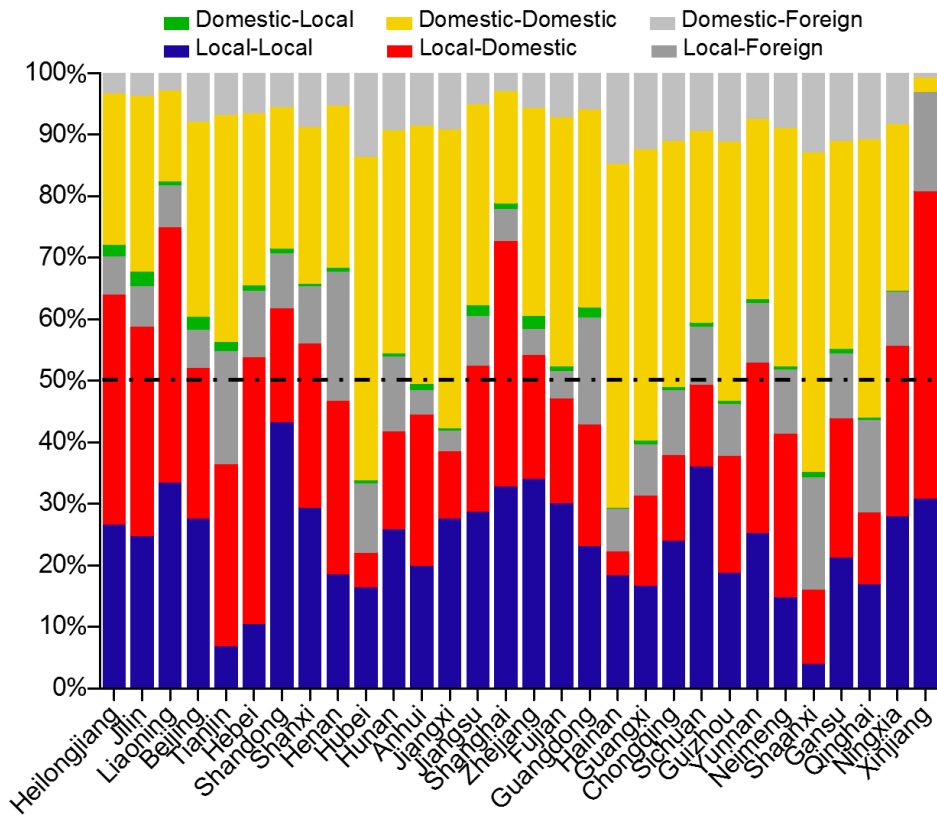**Figure 3.** Sectoral BC emissions embodied in exported and imported products via inter-provincial trade for 30 provinces; the 17 sectors in MRIO are sorted into 8 for clearer presentation (listed in Table S2). Intermediate products (dark gray) are embodied BC emissions used by industry.

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Figure 4. The contribution of surface BC concentration in 30 provinces from both production and

consumption perspectives. Blue (Local-Local), red (Local-Domestic), dark grey (Local-Foreign)

columns indicate the percentage of surface concentration in a province that is contributed by its own

on-site emissions but induced by consumptions from its own province, the rest 29 provinces, and

foreign countries, respectively. Comparably, green (Domestic-Local), yellow (Domestic-Domestic),

and light grey (Domestic-Foreign) columns, respectively, indicate the percentage that is contributed

by the emissions released in the other 29 provinces but induced by local, domestic and foreign

consumptions. The dashed line at 50% marks the comparison between surface concentrations

contributed by local and non-local on-site emissions.

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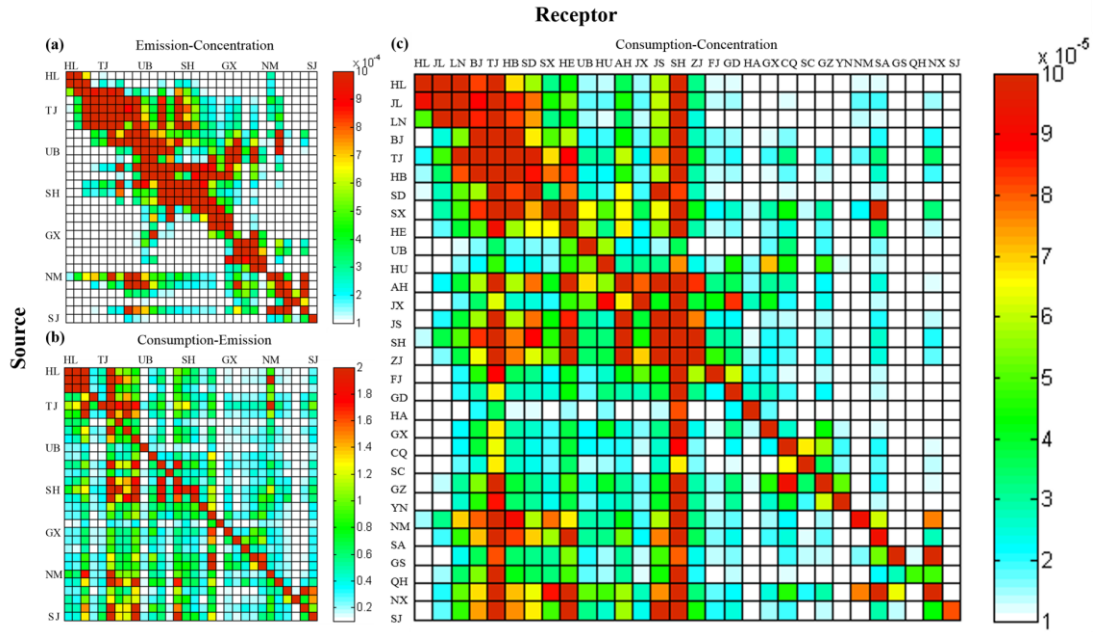
Figure 5. (a) Source-receptor relationship between provincial emission and BC concentration (area-weighted at the surface) among 30 provinces (unit: $(\mu g \cdot m^{-3})/(Gg \cdot yr^{-1})$)); (b) Source-receptor relationship between provincial final consumption and on-site emission (unit: tons per billion Yuan); (c) Source-receptor relationship between provincial final consumption and surface BC concentration (unit: $(\mu g \cdot m^{-3})/(billion\ Yuan \cdot yr^{-1})$)). Province abbreviations (or see Table S1 in the supporting information) are: HL-Heilongjiang, JL-Jilin, LN-Liaoning, BJ-Beijing, TJ-Tianjin, HB-Hebei, SD-Shandong, SX-Shanxi, HE-Henan, UB-Hubei, HU-Hunan, AH-Anhui, JX-Jiangxi, JS-Jiangsu, SH-Shanghai, ZJ-Zhejiang, FJ-Fujian, GD-Guangdong, HA-Hainan, GX-Guangxi, CQ-Chongqing, SC-Sichuan, GZ-Guizhou, YN-Yunnan, NM-Neimeng, SA-Shaanxi, GS-Gansu, QH-Qinghai, NX-Ningxia, SJ-Xinjiang.

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