Development of a High-Resolution Integrated Emission Inventory of Air Pollutants for China

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20 Abstract. Constructing a highly-resolved comprehensive emission dataset for China is challenging due to limited availability 21 of refined information for parameters in a unified bottom-up framework. Here, by developing an integrated modeling 22 framework, we harmonized multi-source heterogeneous data including several up-to-date emission inventories at national and 23 regional scale, and for key species and sources in China, to generate a 0.1 ° resolution inventory for 2017. By source mapping, 24 species mapping, temporal disaggregation, spatial allocation and spatial-temporal coupling, different emission inventories are 25 normalized in terms of source categories, chemical species, and spatiotemporal resolutions. This achieves the coupling of 26 multi-scale, high-resolution emission inventories with the MEIC (Multi-resolution Emission Inventory for China), forming a 27 high-resolution INTegrated emission inventory of Air pollutants for China (i.e., INTAC). We find that the INTAC provides 28 more accurate representations for emission magnitudes and spatiotemporal patterns. In 2017, China's emissions of SO_2 , NO_3 , 29 CO, NMVOC, NH₃, PM₁₀, PM_{2.5}, BC, and OC are 12.3, 24.5, 141.0, 27.9, 9.2, 11.1, 8.4, 1.3 and 2.2 Tg, respectively. The 30 proportion of point source emissions for SO₂, PM₁₀, NO_x, PM_{2.5} increases from 7–19% in MEIC to 48–66% in INTAC, 31 resulting in improved spatial accuracy, especially mitigating overestimations in densely populated areas. Compared to MEIC, 32 INTAC reduces mean biases in simulated concentrations of major air pollutants by 2–14 µg/m³ across 74 cities against ground 33 observations. The enhanced model performance by INTAC is particularly evident at finer grid resolutions. Our new dataset is 34 accessible at http://meicmodel.org.cn/intac, and it will provide a solid data foundation for fine-scale atmospheric research and 35 air quality improvement.

36

37 **1 Introduction**

In recent years, China has achieved remarkable progress in improving air quality and public health through the active implementation of clean air policies (Liu et al., 2020; Xiao et al., 2022; Zhang and Geng, 2019; Zhang et al., 2019a). To further unlock the potential of targeted clean air actions, there is an urgent need for an accurate and detailed depiction for emissions, encompassing their magnitudes and spatial-temporal patterns. Developing a reliable highly-resolved emission inventory for China is also crucial for studies of atmospheric chemistry and climate change (Cheng et al., 2021a; Geng et al., 2021; Zhang et al., 2019a).

The construction of high-resolution emission inventories for China poses significant challenges due to the diversity and complexity of emission sources and technology distributions. Additionally, the limited availability of localized measurements for emission factors (EFs) and source profiles, along with exact location of the emission facilities, further compounds the difficulties (Li et al., 2017a). The widely-used bottom-up approach involves the establishment of a unified framework that encompasses source categories, chemical speciation processes, spatial-temporal allocation profiles and emission estimation methods (An et al., 2021; Huang et al., 2021). However, achieving both wide coverage and high accuracy in compiling an emission inventory for China through this approach remains a formidable task for individual research institutions.

51 Comprehensive national-scale emission inventories developed using the unified framework typically provide extensive 52 coverage of space, species and sectors (Li et al., 2017a; Li et al., 2023b), but tend to exhibit limitations in spatial accuracy (Wu 53 et al., 2021; Zhao et al., 2015; Zheng et al., 2021; Zhou et al., 2017b). Previous studies have indicated that the spatial allocation 54 in large-scale emission inventories rely on spatial proxies (e.g., population, road networks) rather than latitude-longitude 55 coordinates of emission sources due to the unavailability of extensive spatial information (Li et al., 2017b; Zhang et al., 2009). 56 The assumption of a linear correlation between emissions and spatial proxies might lead to an overestimation of emissions in 57 urban areas, especially at scales finer than 0.25 ° (Wu et al., 2021; Zheng et al., 2021; Zheng et al., 2017). Biases introduced 58 by the proxy-based method are found to be propagated as the grid size diminishes, resulting in uncertainties for chemical 59 transport models (CTMs) (Zheng et al., 2021; Zheng et al., 2017).

60 Emission inventories focused on a specific region (An et al., 2021; Huang et al., 2021; Liu et al., 2018), sector (Chen et al., 61 2016; Deng et al., 2020; Zhou et al., 2017a) or key species (Huang et al., 2012b; Li et al., 2021; Wang et al., 2023) under the 62 aforementioned unified framework demonstrate enhanced accuracy, but fail to achieve comprehensive coverage. These 63 inventories assimilate substantial detailed foundational data from various statistical dataset, on-site measurements or surveys 64 to represent real-world emission magnitudes, including energy consumption, removal efficiencies, and localized speciation 65 profile (An et al., 2021; Huang et al., 2021; Liu et al., 2018). Innovative data, such as measurements from continuous emission 66 monitoring systems (Bo et al., 2021; Tang et al., 2023; Wu et al., 2022), or methodologies like process-based models (Kang 67 et al., 2016; Zhao et al., 2020) are implemented to enable a more accurate characterization of complex emission dynamics. 68 Facility-level geographic location is incorporated to optimize the representation of spatial patterns (Liu et al., 2015a; Wang et al., 2019; Wu et al., 2023). The reliability of these local-scale, sector- or species-specified inventories has been validated
against satellite and ground-based measurements (Liu et al., 2016a; Zhang et al., 2021; Zheng et al., 2019).

71 The other strategy for developing bottom-up emission inventories is commonly known as the integrated method. This method 72 consolidates multiple emission datasets for specific regions, species or sectors into a unified product, ensuring extensive 73 representation (Li et al., 2017b). Taking advantage of existing inventories derived from localized data and advanced methods, 74 the integrated method facilitates the efficient generation of highly-resolved emission inventories at large scales. However, the 75 heterogeneity of different emission datasets presents challenges for the fusion, manifested in diverse data formats, sector 76 categories, species, spatial-temporal resolution. In recent years, there has been growing interest in adopting the integrated 77 approach to enrich inventories with local insights, particularly at the global (Crippa et al., 2023; Janssens-Maenhout et al., 78 2015) and Asian scales (Kurokawa et al., 2013; Li et al., 2023a; Li et al., 2017b; Zhang et al., 2009). Researches on establishing 79 integrated inventories for China are constrained due to the inherent complexity and challenging accessibility of the data. These 80 efforts are concentrated in specific regions, such as the Yangtze River Delta (YRD) (An et al., 2021).

In this work, with the support of several research institutions, we use an emission integration model to construct a highresolution integrated emission inventory at a spatial resolution of 0.1 ° for China in 2017, denoted as INTAC. The challenges associated with coupling multi-source heterogeneous data are addressed through the implementation of an inventory integration framework. Then, leveraging the strengths of inventories enriched with local knowledge, we compile a comprehensive highly resolved emission product to enhance the accurate representation of emissions from crucial regions, sectors and species. Finally, the improved accuracy of emission magnitude and spatial distribution is evaluated using atmospheric chemistry models.

88 2 Methodology and data

89 Figure 1 illustrates the schematic diagram of the integration process of INTAC. We collect seven emission inventories—MEIC 90 developed by Tsinghua University (Li et al., 2017a; Zheng et al., 2018), the industrial point source emission inventory for 91 China by the MEIC team (Zheng et al., 2021; Zheng et al., 2017), the YRD air pollutant emission inventory led by Nanjing 92 University (An et al., 2021; Zhou et al., 2017b), the Pearl River Delta (PRD) emission inventory by Jinan University (Huang 93 et al., 2021; Sha et al., 2021), the open biomass burning emission inventory in China by Peking University (Huang et al., 2012a; 94 Liu et al., 2015b; Song et al., 2009; Yin et al., 2019), the shipping emission inventory in East Asia by Tsinghua University 95 (Liu et al., 2016b; Liu et al., 2019), and the high-resolution ammonia emission inventory in China (PKU-NH₃) by Peking 96 University (Huang et al., 2012b; Kang et al., 2016). The details of these inventories and the rationale for choosing them will 97 be described in Sect. 2.1.



98 Figure 1: Methodology framework of the INTAC inventory development.

An integration model is then established to merge together emission inventories which have different sectors, species, spatialtemporal resolution and formats (i.e., point, area, and gridded forms). The integration process consists of five steps: source mapping, species mapping, temporal disaggregation, spatial allocation, and spatial-temporal coupling, as detailed in Sect. 2.2. Based on the priority order, multi-source emission inventories are assembled at the standardized species, sector, and grid levels, yielding a standardized data cube. Ultimately, the integrated emission inventory INTAC is created for China, featuring a resolution of 0.1 ° on a monthly scale and covering nine air pollutants (i.e., SO₂, NO_x, CO, NMVOC, NH₃, PM₁₀, PM_{2.5}, BC, OC).

106 2.1 Components of the integrated emission inventory INTAC

107 Table 1 lists the essential details about the seven inventories and priority order utilized for integration. Given MEIC's extensive 108 coverage across species, sectors, and spatial domains, it functions as the default inventory in our integration, supplementing 109 the missing data in other inventories. The remaining six inventories can be categorized into three types in sequence: point-110 source-based inventory (ranked sixth), regional inventories (ranked fifth and fourth), and process-based inventories (ranked third to first). The point-source-based inventory can directly correct the spatial misallocation of industrial emissions in MEIC 111 112 at fine scales (Zheng et al., 2021; Zheng et al., 2017). The regional inventories further enhance local investigations of individual emission sources and simultaneously refine estimation methods for mobile and area sources (Gu et al., 2023; Zhao et al., 2018; 113 Zhou et al., 2017b). Process-based inventories typically adopt advanced methods to improve the characterization for emission 114 115 processes and parameters specific to particular sectors or species, thereby providing emission totals and distributions that are 116 more in line with measurements (Huang et al., 2012a; Huang et al., 2012b; Kang et al., 2016; Liu et al., 2016; Liu et al., 2019; 117 Liu et al., 2015b; Song et al., 2009; Yin et al., 2019).

Priority ranking	Emission inventory and developer	Year	Resolution	Region	Resolution	Species
1	PKU-NH ₃ (Peking University)	1980– 2017	Monthly	Mainland China	0.1 °	NH ₃
2	The shipping emission inventory for East Asia (Tsinghua University)	2017	Annually	East Asia	0.1 °	SO ₂ /NO _x /CO/NMVOC/ PM _{2.5} /BC/OC
3	The open biomass burning emission inventory for China (Peking University)	1980– 2017	Daily	Mainland China	~1km	SO ₂ /NO _x /CO/NMVOC/ NH ₃ /PM ₁₀ /PM _{2.5} /BC/OC
4	The PRD emission inventory (Jinan University)	2017	Monthly	PRD	0.05 °	SO ₂ /NO _x /CO/NMVOC/ NH ₃ /PM ₁₀ /PM _{2.5} /BC/OC
5	The YRD emission inventory (Nanjing University/Shanghai Academy of Environmental Sciences/Jiangsu Provincial Academy of Environmental Science)	2017	Annually	YRD	0.1 °	SO ₂ /NO _x /CO/NMVOC/ NH ₃ /PM ₁₀ /PM _{2.5} /BC/OC
6	The industrial point source emission inventory for China (Tsinghua University)	2012– 2018	Monthly	Mainland China	~1km	SO ₂ /NO _x /CO/NMVOC/ NH ₃ /PM ₁₀ /PM _{2.5} /BC/OC
7	MEICv1.3 (Tsinghua University)	2008– 2017	Monthly	Mainland China	0.25 °	SO ₂ /NO _x /CO/NMVOC/ NH ₃ /PM ₁₀ /PM _{2.5} /BC/OC

119 **2.1.1 MEIC**

The integrated inventory INTAC is built upon MEIC, a comprehensive database with extensive coverage across time periods, space, species, and sectors. Developed by Tsinghua University since 2010 (http://meicmodel.org.cn) (Li et al., 2017a; Zheng et al., 2018), the MEIC provides monthly emissions for air pollutants and CO₂ in China from 1990 to the present at a resolution of $0.25^{\circ} \times 0.25^{\circ}$. It caters to the demand for timely and accurate estimates of atmospheric emissions, gaining widespread adoption by both domestic and international research institutions. We use 2017 emissions from MEICv1.3 in this study.

MEIC employs several strategies to improve emission estimation parameters. This includes categorizing emission sources across ~800 sectors, utilizing a technology- and big-data-driven approach for dynamic emission characterization, and

- 127 employing a localized emission factor database (Li et al., 2017a; Zheng et al., 2018). Emission estimates for power, on-road,
- 128 and residential sources are enhanced through the use of unit-level data (Liu et al., 2015a), county-level emission estimates
- 129 (Zheng et al., 2014), and integration of extensive household surveys (Peng et al., 2019), respectively. MEIC builds an database
- 130 encompassing temporal allocation profiles (ranging from yearly to monthly, daily, and hourly) (Li et al., 2017b), spatial
- 131 allocation proxies (from province to county, and further to grids) (Geng et al., 2017; Li et al., 2017b; Zheng et al., 2017), and
- 132 a speciation framework for NMVOC involving five mechanisms (CB-IV, CB05, SAPRC-07, SAPRC-99, and RADM2) (Li et
- al., 2014) (Li et al., 2014) to support the development of model-ready gridded emissions.
- 134 Among the seven inventories, MEIC has the lowest priority, and is only considered when the other six cannot provide necessary
- 135 emissions for a specific city and source.

136 2.1.2 The industrial point source emission inventory for China

137 The proxy-based method used for spatial allocation in MEIC introduces biases in emission mapping, especially at kilometer 138 scale (Zheng et al., 2021; Zheng et al., 2017). To significantly reduce the uncertainty, we merged an industrial emission 139 inventory with detailed information on ~100,000 facilities into INTAC.

140 Compiled by the MEIC team at Tsinghua University for the year 2013 (Zheng et al., 2021) and updated by them for 2017, this 141 point-based inventory combines three databases investigated under the guidance of the Chinese government, offering a 142 comprehensive overview of industrial facilities. It includes details on the locations, activity rates, production technology, end-143 of-pipe pollution control devices, and other parameters. It is worth noting that the facility-level activity data were corrected 144 using provincial activity data from MEIC as a total constraint to ensure consistency with national totals from statistics (Zheng 145 et al., 2021). The facility-level, technology-based approach allows for dynamic tracking of emission fluctuations resulting from technological advancements and tightening emission regulations. Crucially, the use of facility geolocations rather than 146 147 relying on spatial proxies like urban population enables the derivation of gridded industrial data at a resolution of ~ 1 km. This 148 approach significantly avoids misallocating emissions from rural to urban areas at fine grids, as supported by previous studies 149 demonstrating its effectiveness in mitigating simulated biases in air pollutant concentrations within densely populated regions 150 (Zheng et al., 2021). For temporal variations, it employs the same monthly profiles as MEIC, including the production of 151 various industrial goods or Gross Domestic Product (GDP), as outlined in Li et al. (2017b). The NMVOC speciation also 152 aligns with the MEIC model. This inventory takes priority over MEIC, indicating that only few industrial sources not covered 153 in this inventory are substituted with MEIC.

154 2.1.3 The YRD air pollutant emission inventory

155 Regional emission inventories within YRD provide a more accurate representation of emissions compared to the national-

- 156 scale MEIC, as proven by ground and satellite observations (Yang and Zhao, 2019; Zhang et al., 2021; Zhao et al., 2017a;
- 157 Zhao et al., 2018; Zhao et al., 2020; Zhou et al., 2017b). This improvement is attributed to the avoidance of outdated or non-
- 158 localized emission calculation parameters, commonly present in large-scale inventories like MEIC. Here, we merge the 2017

- YRD air pollutant emission inventory into INTAC to achieve state-of-the-art estimates for rapidly changing emissions over
 this core area (An et al., 2021; Gu et al., 2023; Zhou et al., 2017b).
- 161 Localized field surveys and measurements greatly enhance the reliability of calculation parameters within the YRD inventory. 162 Highly-resolved emissions for the power sector are acquired through on-site monitoring with high temporal resolution (Zhang 163 et al., 2019b), rather than relying on static and outdated average emission factors. Facility-level information (e.g., the removal 164 efficiencies) obtained from local investigation and a segment-based industrial process method enhances the understanding of 165 both the quantity and spatial patterns of industrial emissions. Considering meteorological factors and land use conditions 166 during agricultural processes results in more accurate seasonal and spatial distributions of NH₃ emissions. (Zhao et al., 2020). An investigation of in-use machinery is conducted to capture the seasonal emission patterns from off-road machines (Zhang 167 et al., 2020). Real-world surveys are performed to determine grain straw ratios and household burning proportions, facilitating 168 169 the quantification of emissions from biomass-fueled stoves. The PM_{2.5} and NMVOC speciation profiles are updated based on 170 multi-instrument sampling and analysis in both current and previous studies (Huang et al., 2018; Zhao et al., 2017a), satisfying 171 the needs for simulating $PM_{2.5}$ chemical components and O_3 . The YRD inventory is collected with a spatial resolution of 0.1 172 degree and an annually temporal resolution in this study. Only CB05 VOC species are collected.

173 2.1.4 The PRD emission inventory

The regional emission inventories within the PRD region have demonstrated enhanced reliability compared to previous studies (Huang et al., 2021; Sha et al., 2021; Zheng et al., 2012). The PRD emission inventory developed by the Jinan University captures spatial and temporal variations within the PRD region under emission control policies, serving as a foundation for supporting air quality modeling (Huang et al., 2021; Sha et al., 2021).

178 The PRD inventory exhibits notable accuracy improvements, achieved by means of big data-driven estimation methods, 179 updated spatial-temporal allocations, and localized NMVOC speciation profiles. Gridded hourly open biomass burning 180 emissions are quantified by fusing the fire radiative power data from three satellites, and hourly shipping emissions are 181 estimated using high-frequency Automatic Identification System (AIS) records. Thirty-one monthly profiles and ten spatial 182 proxies are updated to reflect spatial-temporal patterns of emissions influenced by economic growth and energy consumption 183 structural adjustment. Approximately 90% of industrial emissions are disaggregated using exact locations, and novel proxies 184 (e.g., farmland production potential) have been developed for several sectors. The NMVOC speciation is carried out through 185 massive localized measurements and literature reviews, manifested as a collection of 480 NMVOC source profiles across eight 186 sectors and 380 species. The species relevant to the SAPRC-07 chemical mechanism are collected in this work. Additionally, 187 the inventory encompasses 800 source categories, placing particular emphasis on incorporating new sectors relevant to VOC 188 emissions. Activity rates are improved by utilizing extensive field surveys and data mining efforts, involving investigations of 189 production data for 10,000 industrial plants and the gathering of activity-relevant information for 50 million vehicles. Emission 190 factors that reflect local context are obtained or revised based on source measurements and latest research findings. These 191 updates help mitigate uncertainties in emission estimates for the PRD region. The PRD inventory is initially collected at a monthly resolution and a spatial resolution of 0.05° , with detailed spatial-temporal allocation proxies outlined in Huang et al. (2021).

194 2.1.5 The open biomass burning emission inventory in China

As a significant source of CO₂, BC, OC and other pollutants, open biomass burning profoundly influences air quality, climate change, and human health (Reisen et al., 2013). A case study in summer 2011 for the YRD region revealed that during a severe haze episode, open biomass burning contributed to 37%, 70%, and 61% of PM_{2.5}, OC, and EC emissions, respectively (Cheng et al., 2014). To address the absence of this source in MEIC, we integrate a high-resolution open biomass burning emission inventory by Peking University into INTAC (Huang et al., 2012a; Liu et al., 2015b; Song et al., 2009; Yin et al., 2019). The inventory applies satellite observations to tackle considerable uncertainties associated with provincial statistical data and

overcome the coarse resolution found in previous studies (Ni et al., 2015). The estimation of biomass consumption in the inventory is based on the fire radiative energy (FRE) approach, which depends on the energy emitted by fires. This approach helps reduce the biases introduced by burned areas algorithms, especially for small-scale fires. The inventory utilizes the high spatial resolution land cover dataset GlobeLand30 derived from multispectral images to classify biomass fuel types. Eventually, daily emissions from forest, grassland, cropland and shrubland are calculated at a 1-kilometer resolution. The reasonableness is validated by comparing with other datasets, such as the fourth version of the Global Fire Emissions Database (GFED). The initially collected inventory lacks model-ready VOC species.

208 2.1.6 The shipping emission inventory in East Asia

In recent years, maritime trade in the East Asian region has significantly increased (Trade and Development, 2014), resulting in a surge in shipping emissions with substantial impacts on air quality and climate. Previous studies have indicated that East Asian shipping emissions accounted for 16% of the global total in 2013. Shipping emissions made a growing contribution to the rise in annual mean $PM_{2.5}$ concentrations, reaching levels as high as 5.2 µg/m³ in 2015 (Lv et al., 2018). To address the omission of this emission source in the MEIC, we integrate the shipping emission inventory in East Asia for 2017 into INTAC (Liu et al., 2016b; Liu et al., 2019).

215 The inventory introduces an innovative approach based on comprehensive and dynamic ship activity data. A static dataset of 216 approximately 66,000 vessels is compiled as a foundation, using information from Lloyd's Register and China Classification 217 Society. This dataset encompasses various ship properties, including ship category, hull shape, engine rotational speed, engine capacity, maximum speed capability, build year, and more. High quality AIS data is used to capture ship activities, 218 219 incorporating the Maritime Mobile Service Identification identifier, geographical location, real-time speed, and time-related 220 information. The AIS data is also employed to generate gridded emissions from shipping at a spatial resolution of 0.1 °. The inventory enhances our comprehension of regional-level shipping emissions and significantly alleviates biases arising from 221 222 the misallocation of marine fuels, as observed in global studies (Endresen et al., 2007). The collected shipping inventory 223 provides emissions at an annual resolution for seven species, including SO₂, NO_x, CO, NMVOC, PM_{2.5}, BC, and OC.

224 2.1.7 PKU-NH3

225 As a prominent alkaline component in the atmosphere, ammonia plays a crucial role in atmospheric chemistry, terrestrial and 226 aquatic ecosystems through its participation in atmospheric reactions and deposition processes. This study integrates PKU-227 NH₃, a high-resolution ammonia emission inventory for China developed by Peking University, PKU-NH₃ is designed to track 228 the evolution of NH₃ emissions amid the rapid increase in grain and meat production in China over the past few decades 229 (Huang et al., 2012b; Kang et al., 2016). This inventory offers a better grasp on NH₃ emissions in China through the application 230 of a process-based method and more reliable emission factors, in contrast to previous studies (Kurokawa et al., 2013; Li et al., 231 2017b). Top-down NH₃ inversion through satellite observations provides additional validation for the accuracy of PKU-NH₃ 232 (Paulot et al., 2014).

233 Earlier studies of NH₃ emissions commonly used fixed EFs, overlooked some ammonia emission sources, and had coarse 234 resolutions (Ohara et al., 2007; Streets et al., 2003). Unlike previous approaches, the PKU-NH₃ incorporates dynamic and 235 multifactorial EFs and more comprehensive emissions sources. The determination of emission factors takes into account 236 various parameters related to local conditions and agricultural practices. When estimating NH₃ emissions of synthetic fertilizer 237 application, the model considers five types of fertilizers, as well as factors such as soil acidity, ambient temperature, fertilizer 238 application technique and dosage, wind speed, and in-situ measurements of NH₃ flux. For livestock waste, NH₃ emissions are 239 calculated using a mass-flow approach across four phases of manure management, considering variables such as animal rearing 240 types, temperature and wind speed. In addition, NH₃ emissions from other small sources are also quantified, including 241 agricultural soil, nitrogen-fixing crop, crop residue compost, excretion of rural populations, open biomass burning, waste 242 disposal, gasoline vehicles, diesel vehicles, and industrial processes. The NH₃ emissions are allocated from provinces into 0.1 $^{\circ}$ 243 grids based on spatial proxies such as land cover, rural population, and other relevant indicators. Monthly emission factors 244 shaped by meteorological conditions are used to calculate NH_3 emissions from fertilizer application and livestock source at a 245 monthly level.

246 2.2 The integration of multi-source heterogeneous data

In the integration process, seven heterogeneous inventories are first normalized in terms of emission sources, species, spatial temporal resolutions, and then integrated following a priority order to produce a standardized, highly-resolved data cube.

249 2.2.1 Source mapping

To merge inventories under a unified emission source classification system, the emission sources in the MEIC model are categorized into 88 standard sectors for mapping (Table S1). The first-level category comprises 10 subcategories, namely, stationary combustion, industrial process, mobile source, solvent use, agriculture, dust, biomass burning, storage and transportation, waste treatment and other sources. These are then further subdivided into 88 second-level sources, which take industrial classification for national economic activities for reference. For example, the industrial process sector encompasses 255 emission sources such as the manufacturing of non-metallic mineral products, manufacturing of chemical fibers, manufacturing

of foods, smelting and pressing of ferrous metals, and more. In the initial step of integration, the sectors in each emission inventory are mapped to the standardized two-level sources.

258 2.2.2 Species mapping

259 Then, non-methane volatile organic compounds (NMVOC), particulate matter (PM), and NO_x in each inventory are converted 260into model-ready species to support CTMs. The species mapping process is grounded in the chemical species mapping methods 261 in MEIC model (Li et al., 2017b; Li et al., 2014). The model supports aerosol chemical schemes such as AER05 and AER06. NO_x emissions are allocated to NO and NO₂ emissions based on ground observations. The step-by-step NMVOC speciation 262 263 framework developed in Li et al. (2014) is employed to generate emissions for various gas-phase chemical mechanisms commonly used in CTMs, including CB-IV, CB05, SAPRC-07, SAPRC-99 and RADM2. The framework incorporates an 264 265 explicit assignment approach and updated profiles based on both local measurements and the SPECIATE database v.4.5. The 266 sources abundant with oxygenated volatile organic compounds (OVOC) are identified, and the incomplete profiles with missing OVOC fractions are corrected. The accurate speciation mapping helps reduce uncertainties in model-ready emissions. 267 268 For inventories providing speciated VOC emissions for certain mechanisms (e.g., the YRD inventory for CB05, PRD inventory 269 for SAPRC-07), we directly use their emissions, or alternatively, utilize MEIC's speciation framework to generate model 270 species for the five chemical mechanisms.

271 2.2.3 Temporal disaggregation

272 The seven emission inventories are collected at different temporal resolutions (Table 1) and need to be temporally allocated to 273 a unified monthly scale for integration. Monthly emissions from PKU-NH₃, the PRD inventory, the industrial point source 274 inventory and MEIC can be directly used for data merge. Daily-level emissions from the open biomass burning inventory are 275 aggregated to monthly scales through summation. For annual inventory (e.g., the YRD inventory), sector-specific monthly 276 profiles derived from the MEIC model are used for disaggregation (Li et al., 2017b). For instance, monthly power generation 277 data from the National Bureau of Statistics describe variations in monthly power emissions. Industrial production or GDP from 278 the National Bureau of Statistics are employed to account for monthly emission fluctuations related to industrial heating, 279 boilers, cement, iron and steel, and other industrial processes. Monthly emission factors calculated by the International Vehicle 280 Emissions model are applied to on-road vehicles. Considering the insignificant monthly variations of Automatic Identification 281 System data for marine shipping, the annual shipping emissions are uniformly disaggregated across the months.

282 2.2.4 Spatial allocation

The seven inventories are in different data formats, including point source and gridded formats at varying resolutions, necessitating spatial harmonization for integration. Although the industrial point source inventory and the open biomass burning inventory can accurately pinpoint the specific geographic locations of emission sources, the other five inventories rely 286 on numerous spatial proxies to disaggregate emissions into grids, which inevitably introduce uncertainties at very fine 287 resolutions. Therefore, we re-grid the final product to 0.1 °to ensure high level spatial accuracy. Gridded emissions finer than 0.1° resolution are aggregated to 0.1° , which is performed in the open biomass burning inventory and the PRD inventory. For 288 289 the industrial point source inventory, latitude and longitude coordinates are employed to directly position them within grid 290 locations. Area sources in MEIC are allocated to grids using spatial proxies within the MEIC model (Li et al., 2017b). For 291 instance, industrial sources are assigned to grids based on urban population (Schneider et al., 2009). The road network (Zheng 292 et al., 2014) serves as a proxy for disaggregating emissions of on-road vehicles, while rural population (Schneider et al., 2009) is used as the proxy for fertilizer and livestock sources. It's important to mention that uncertainties may arise at city borders if 293 294 emissions from adjacent cities come from different inventories during the integration process. To mitigate biases introduced 295 by border issues, all emissions at 0.1 °resolution are first uniformly downscaled to 1 km in preparation for the spatial-temporal 296 coupling process, and then re-gridded back to 0.1 ° for the final product.

297 2.2.5 Spatial-temporal coupling

Finally, following the procedures outlined in Sections 2.2.1 to 2.2.4, all inventories are preprocessed to a standardized format, encompassing 88 sectors, various species, a spatial resolution of 1 km, and a monthly temporal resolution. This preprocessing prepares the inventories for merging, ultimately resulting in the generation of a standardized data cube.

301 The integration is carried out at source-by-source, species-by-species, and grid-by-grid levels, with the process guided by the 302 priority order of each inventory (Table 1). MEIC serves as the default inventory in our integration, offering extensive spatial 303 and species coverage, along with spatial proxies, temporal profiles, and NMVOC speciation methods within the model. The 304 remaining six emission inventories are assigned a predefined priority order. The industrial point source emission inventory for 305 China takes precedence over industrial emissions in MEIC, substituting proxy-based spatial allocation with precise 306 geographical coordinates. This extends the applicability of MEIC from a resolution greater than 0.25 °to finer scale (Zheng et 307 al., 2021; Zheng et al., 2017). To achieve fine-grained emission characterization in critical areas, the YRD and PRD emission 308 inventory enriched with localized data and advanced methods are incorporated to update emissions in these areas. While MEIC 309 comprehensively estimates emissions for ~800 source categories in China, there may still be omissions for certain emission sources. The inclusion of inventories for open biomass burning and East Asian shipping helps partially fill this gap. The PKU-310 311 NH₃, generated by a process-based model to provide a comprehensive understanding of China's NH₃ sources, is utilized to 312 replace all NH₃ emissions in other inventories. The prioritization is performed city by city. For emissions of a particular species from a specific emission sector, when multiple inventories overlap in city grids, the estimates from the highest-priority 313 314 inventory is selected as the final emissions. Through this step, the integrated inventories are developed based on the configured 315 output settings, such as map projection and spatial-temporal attributes.

316 2.3 Evaluation of the emission inventory using WRF/CMAQ model

We apply Weather Research and Forecasting Version 3.9 (WRFv3.9) and Community Multiscale Air Quality Version 5.2 (CMAQ5.2) as the air quality simulation systems. Two nested simulation domains with horizontal resolutions of 36 and 12 km are used (Fig. S1). The mother domain (172×127 cells) covers the entire Chinese mainland and parts of the neighboring countries, and the nested domain (226×241 cells) includes the heavily polluted Eastern China. Four-month (January, April, July, and October) simulations in 2017 are carried out, with a 7-day spin-up period preceded each month. The vertical resolution in WRF is set with 45 sigma levels ranging from the surface up to 100 hPa. Subsequently, it is collapsed into 28 layers through the Meteorology-Chemistry Interface Processor (MCIP) before being input into CMAQ.

324 The configuration of WRF and CMAQ model in this study follows Cheng et al. (2019). The meteorological initial and boundary 325 conditions for the simulation are provided by the final reanalysis data from the National Centers for Environmental Prediction 326 (NCEP-FNL, https://rda.ucar.edu/datasets/ds083.2/). The schemes for shortwave radiation, longwave radiation, land surface 327 processes, boundary layer, cumulus parameterization, and cloud microphysics are selected as the New Goddard scheme (Chou 328 et al., 1998), RRTM scheme (Mlawer et al., 1997), Pleim–Xiu surface layer scheme (Xiu and Pleim, 2001), ACM2 PLB 329 scheme (Pleim, 2007), Kain-Fritsch scheme (Kain, 2004), and WSM6 scheme (Hong and Lim, 2006), respectively. 330 Observational nudging and soil nudging are employed to enhance the meteorological simulation. Regarding CMAQ model, 331 the chemical mechanisms for gas-phase, aqueous-phase, and aerosol are configured as CB05, the Regional Acid Deposition 332 Model (RADM), and AERO6, respectively. Photolysis rates are calculated online using the simulated aerosols and ozone 333 concentrations. Anthropogenic emissions outside China are taken from MIX inventory (Li et al., 2017b). The integrated 334 inventory INTAC and MEIC are used for comparison within China. Biogenic emissions are calculated using the Model of 335 Emissions of Gases and Aerosols from Nature version 2.1 (MEGANv2.1), while dust and lightning emissions are not 336 considered in this study.

The performances of WRF for the meteorological parameters are evaluated against the Integrated Surface Database (ISD) from the National Climatic Data Center (NCDC) (ftp://ftp.ncdc.noaa.gov/pub/data/noaa/). Evaluation metrics include correlation coefficient (R), mean bias (MB), root mean square error (RMSE), normalized mean bias (NMB), and normalized mean error (NME). Table S2 demonstrates good agreement between WRF model results and ground-level observations. Similar configurations have been also validated in previous studies (Cheng et al., 2019; Cheng et al., 2021a; Cheng et al., 2021b). CMAQ modeling performance are assessed using hourly observed concentrations of air pollutants obtained from the China National Environmental Monitoring Center (http://www.cnemc.cn/).

344 3 Results

345 3.1 China's emission characteristics in 2017

We use the integrated emission inventory to analyze air pollutant emissions in China for the year 2017. Major air pollutant emissions are estimated as follows: 12.3 Tg SO₂, 24.5 Tg NO_x, 141.0 Tg CO, 27.9 Tg NMVOC, 9.2 Tg NH₃, 11.1 Tg PM₁₀, 8.4 Tg PM_{2.5}, 1.3 Tg BC, and 2.2 Tg OC. The emission data, organized into power, industry, residential, transportation, agriculture, solvent use, shipping, and open biomass burning sectors, are available for download from https://doi.org/10.5281/zenodo.10459198 (Wu et al., 2024) and http://meicmodel.org.cn/intac. The following sections will characterize emissions in detail across sectors, fuel types, and spatial distributions.

352 **3.1.1 By sectors**

353 Table 2 displays emissions specific to power, industry, residential, transportation, agriculture, solvent use, shipping, and open 354 biomass burning sectors in the INTAC. For pollutants primarily originating from fuel combustion and industrial processes (e.g., SO₂, NO_x, CO, PM₁₀, and PM_{2.5}), the power, industry, and transportation sources collectively contribute significantly to 355 356 their emissions, ranging from 56% to 83%. Industrial sources take a leading role in various atmospheric pollutants, contributing 357 more than 30% for SO₂, NO_x, CO, NMVOC, PM_{10} , and $PM_{2.5}$ emissions. Due to low combustion efficiency and a lack of 358 emission control measures, residential sources exhibit a high emission factor for products of incomplete combustion, leading 359 to 40% of CO emissions, 48% for BC, and 73% for OC. Solvent sources exclusively produce NMVOC emissions, constituting 360 33% to the overall emissions. The complexity of VOC emission origins is evident in the diverse range of contributing sources. Agricultural sources dominate NH₃ emissions, comprising an 83% share of total emissions. As described in Sect. 2.1.7, the 361 362 PKU-NH₃ incorporates a wide variety of NH₃ sources, providing a more comprehensive understanding of the sectors 363 contributing to NH₃ emissions. Insignificant sources may exert large influence in specific regions or periods, such as during large wildfires or in cities with heavy traffic. Additionally, the contribution of the supplemented open biomass burning source 364 365 cannot be overlooked, especially for OC (7%) and NMVOC (6%).

Figure 2 consolidates 88 standardized emission sources into 25 categories, allowing for a more detailed analysis of sectoral 366 367 emission patterns compared to Table 2. Owing to substantial coal use in industrial and power sectors, along with sulfur-rich 368 ship fuels, prominent contributors to SO₂ emissions include power, shipping, stationary combustion, and manufacture of non-369 metallic mineral products sources, accounting for 15%, 13%, 12%, and 12% respectively to total SO₂ emissions. This indicates 370 that achieving further reductions in SO₂ emissions requires the implementation of more energy-efficient end-of-pipe control 371 measures, and adoption of low-sulfur fuels. The dominant origins of NO_x emissions are from the freight truck, power 372 generation, and shipping sectors, representing 21%, 15%, and 13% of the total emissions. Both freight trucks and vessels 373 extensively use compression ignition engines, prone to generating NO_x emissions under high-temperature and oxygen-rich 374 conditions. Implementing strict vehicle standards is crucial to effectively reduce NO_x emissions from exhaust gases. Coatings, 375 other industrial processes, and passenger vehicle sources together account for 51% of anthropogenic NMVOC emissions. The 376 major contributors to primary PM_{2.5} emissions include biomass fuel, the manufacture of non-metallic mineral products, and

377 the smelting and pressing of ferrous metals source, making up 22%, 17%, and 10% of the total emissions, respectively. It's

- 378 noteworthy that the use of biomass fuels (e.g., rice straw, firewood) for cooking or heating in rural areas results in considerable
- 379 PM_{2.5} emissions, especially in provinces like Sichuan, Anhui, Shandong, and Heilongjiang.
- 380
- Table 2: Anthropogenic emissions of air pollutants by sectors in the 2017 INTAC inventory for China (Units: Gg). The shipping sector includes inland waterway sources and the marine vessels.

Sectors	SO ₂	NO _x	CO	NMVOC	NH ₃	PM10	PM _{2.5}	BC	OC
Power	1822	3790	4909	152	14	981	568	6	0
Industry	6066	8800	52828	8824	249	5603	3620	308	285
Residential	2361	861	55895	3676	629	3516	3088	606	1649
Transportation	341	7751	22597	4123	619	533	493	257	95
Agriculture	0	0	0	0	7609	0	0	0	0
Solvent	0	0	0	9255	0	0	0	0	0
Shipping	1642	3077	391	191	2	73	264	43	49
Open biomass burning	21	215	4403	1659	76	409	355	35	167
Total	12253	24494	141023	27881	9198	11117	8388	1255	2245



Figure 2: Sector-specific distributions of emissions in the 2017 INTAC inventory for China. (a), (b), (c) and (d) represent the sectoral contributions for SO₂, NO_x, NMVOC and PM_{2.5}, respectively. The figure only displays the top eight contributing sources, while sources

385 excluding these are categorized as "other sources".

386 3.1.2 By fuel types

387 Figure 3 illustrates the proportions of major air pollutant emissions in 2017 for each fuel type. Fossil fuel combustion significantly dominates the emissions of PM₁₀, PM_{2.5}, CO, BC, SO₂, NO_x, with proportion ranging from 38% to 80%. The coal 388 389 combustion accounts for 56% of SO₂ emissions, with power, residential activities and industrial production as the primary 390 emitter. Meanwhile, petroleum combustion, mainly from marine vessels, constitutes 20% of SO_2 emissions. For NO_x emissions, 391 petroleum combustion contributes 48% of the total, predominantly arising from freight trucks (5.2 Tg), marine vessels (3.1 392 Tg), and passenger vehicles (1.0 Tg). Coal combustion processes, such as power and industrial boiler also result in substantial 393 NO_x emissions (31%). The biomass fuel source causes 53% of OC emissions. Emissions of NMVOC and NH_3 are primarily 394 associated with non-combustion processes.



395 Figure 3: Fuel-specific distributions of major air pollutant emissions in the 2017 INTAC inventory for China.

396 3.1.3 Spatial distribution

We present the gridded emission maps of major air pollutants in Fig. 4. Emissions from anthropogenic sources in China exhibit significant spatial heterogeneity. Due to economic growth and industrial activities, air pollutant emissions are primarily concentrated in the central and eastern regions of China, especially in economically developed urban clusters such as the Beijing-Tianjin-Hebei (BTH) region, the YRD, the PRD, as well as in regions like Sichuan and Chongqing. These four key 401 areas, as depicted in Fig. S2, collectively account for 25%, 33%, 35%, 37%, 30%, 35%, 33%, 27%, and 29% of the national 402 emissions of SO₂, NO_x, CO, NMVOC, NH₃, PM₁₀, PM_{2.5}, BC, and OC, respectively. Moreover, the emission maps at a fine 403 spatial resolution of $0.1^{\circ} \times 0.1^{\circ}$ present the local variations in emission patterns, identifying numerous hotspots in small areas 404 and showcasing distinct gradients in emissions. Table 3 shows the provincial-level emissions (except Hong Kong, Macao, and 405 Taiwan), and a map depicting provincial boundaries is displayed in Fig. S2. The emission levels in specific provinces are 406 determined by factors such as resource endowments, industrial structure, energy consumption, and emission control measures. 407 Taking SO_2 as an example, the top five provinces are Shanxi, Shandong, Hebei, Guizhou, and Inner Mongolia, collectively 408 accounting for 36% of the national total SO₂ emissions. The Guizhou Province, located in the southwest of China, is 409 characterized by high-sulfur coal and a relatively gradual implementation of pollution control measures, which result in 410 elevated SO₂ emissions. In other four provinces, large scale heavy industries have led to substantial coal consumption and 411 correspondingly higher SO_2 emissions. Provinces with a less industry-focused economic structure and lower energy 412 consumption, including Tianjin, Hainan, Qinghai, Beijing, and Tibet, exhibit the lowest SO₂ emissions, accounting for 413 approximately 2% of the national total.



414 Figure 4: Spatial distributions of major air pollutant emissions in the 2017 INTAC inventory for China.

Table 3: Anthropogenic emissions of air pollutants by provinces in the 2017 INTAC inventory for China (Units: Gg). Emissions from

416	the shipping	emission	inventory in	ı East Asia	are not included.
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Sectors	SO ₂	NOx	СО	NMVOC	NH ₃	PM ₁₀	PM2.5	BC	OC
Anhui	315	846	5955	1089	341	596	443	49	114
Beijing	26	231	1394	516	36	61	49	7	16
Chongqing	396	375	2419	564	150	207	158	22	47
Fujian	161	530	2344	895	149	202	152	22	49
Gansu	189	351	2221	358	276	163	126	22	42
Guangdong	430	1566	6897	1268	351	783	356	17	67
Guangxi	265	434	3578	808	323	355	275	29	83
Guizhou	652	355	6629	508	236	459	347	76	125
Hainan	47	95	584	172	57	46	37	5	14
Hebei	667	1697	11731	1673	523	708	528	88	125
Heilongjiang	246	822	7034	1419	379	495	403	65	156
Henan	367	1256	7962	1500	678	620	459	79	108
Hubei	513	703	6341	1183	358	455	354	68	118
Hunan	518	633	6802	953	330	481	363	77	122
Inner Mongolia	594	1211	5747	831	562	459	340	56	89
Jiangsu	391	1217	8628	1529	498	667	496	50	105
Jiangxi	179	449	3676	646	209	273	195	28	52
Jilin	235	652	3973	847	207	307	237	39	76
Liaoning	459	1200	5835	1316	268	432	325	54	86
Ningxia	226	327	766	178	79	91	63	7	9
Qinghai	43	106	598	129	131	59	45	5	8
Shaanxi	334	549	3781	820	273	294	221	39	68
Shandong	946	2134	11469	2846	696	897	678	105	150
Shanghai	114	469	1130	342	29	104	86	15	6
Shanxi	977	964	6017	756	199	555	415	64	81
Sichuan	379	777	6362	1478	646	463	371	56	141
Tianjin	90	333	1434	573	33	81	61	9	12
Xinjiang	257	608	2639	632	516	218	158	23	32
Xizang	1	52	149	46	149	15	12	2	5
Yunnan	332	435	3823	576	398	302	230	38	75
Zhejiang	293	670	3009	1342	118	270	195	22	22

417 3.2 Improved accuracy of China's anthropogenic emissions by INTAC

418 3.2.1 Comparison of emission magnitudes in INTAC with MEIC across sectors and regions

419 The INTAC inventory improves the representation of anthropogenic air pollutant emissions by incorporating a large number 420 of industrial point sources, integrating high-resolution regional inventories, and supplementing missing emission sources in 421 MEIC. Remarkable differences between INTAC and MEIC are illustrated in Fig. 5 across regions and sectors. Compared to MEIC, the INTAC inventory shows higher level of 16.7%, 11.5%, 10.8%, 11.0%, and 9.1% for SO₂, NO_x, PM₁₀, PM₂₅, and 422 423 OC emissions, respectively. However, it indicates lower levels of 6.3% and 10.6% for NMVOC and NH₃. CO and BC 424 emissions exhibit good agreement between the two inventories, with differences lower than 3.9%. In comparison to MEIC, the 425 supplementary emission sources in INTAC-specifically, open biomass burning and marine shipping-account for the 426 majority of increased emissions, contributing 95%, 89%, and 74% for SO₂, CO, and PM_{2.5}, respectively. Additionally, the 427 incorporation of PKU-NH₃ in INTAC leads to a 21% decrease in NH₃ emissions from agricultural sources, while NH₃ 428 emissions from residential sources and transportation increase by 99% and 13.1 times, respectively. Such difference in 429 agricultural sources is mainly caused by the estimates of synthetic fertilizer (Kang et al., 2016), particularly concerning the 430 treatment of fertilizer types and corresponding emission factors.



Figure 5: Inter-comparisons of emission estimates between the INTAC inventory and MEIC. (a) shows the difference by sectors, and
 (b) presents the ratio of emissions in INTAC to those in MEIC.

Many discrepancies between MEIC and INTAC arise from the integration of regional emission inventories. As presented in 433 434 Fig. 5b, notable disparities are observed in the YRD and PRD region. Estimates for NO_x emissions in the YRD region are 435 approximately 88% of those derived from the MEIC model. This highlights an enhanced precision attributable to reliable 436 assessments of denitrification efficiency in power plants and the measured NO_x emission factors for both power plants and 437 boilers within the integrated YRD inventory, as supported by previous research studies (Zhao et al., 2018). INTAC's estimates for NMVOC emissions in the YRD region are 26% lower than estimates in MEIC. The overestimation in MEIC mainly results 438 439 from the uncertainties of solvent use source, particularly coating and printing and dveing processes. The integrated YRD 440 emission inventory employs more accurate calculation parameters for NMVOC, such as statistical data from local city 441 yearbooks, industry association reports, and apparent consumption of solvents. Furthermore, the speciation profiles of 442 NMVOC are localized and corrected based on the literature research and measurements. In the PRD region, The NO_x emissions from INTAC are 41% higher than MEIC estimates, with non-road sources and non-metallic mineral products contributing 45% 443 444 and 40% to this difference, respectively. The PRD inventory employs a detailed calculation approach for shipping emissions 445 based on AIS data, in contrast to the simplified approach for inland waterway sources in MEIC. The NO_x emissions from 446 industrial processes of brick and flat glass manufacturing are not considered in MEIC, which is a deficiency that is addressed 447 in the integrated PRD inventory. INTAC's NMVOC emissions are approximately 59% of those from MEIC. The disparity is 448 particularly notable in industrial and solvent use sources, contributing 49% and 35%, respectively, to the observed difference. 449 In INTAC, nearly half of the VOC emission factors for industrial solvent sources are based on local measurements, and a 450 preference for raw material-based calculations over product-based ones reduces uncertainty in the estimation. For significant 451 VOC-emitting sources like cleaning solvents, MEIC employs an emission factor of 1000 g/kg, whereas the PRD inventory 452 uses 850 g/kg. In the case of oil refineries, the emission factors are 2.76 g/kg for MEIC and 1.82 g/kg for the PRD inventory.

453 **3.2.2 Impact of point source contributions**

454 The most accurate method for obtaining emissions at finer-scale grids relies on spatial allocation based on precise geographical 455 coordinates. In MEIC, the majority of emission sources are represented as area sources and distributed onto grids using spatial 456 proxies such as urban population, except for power plants. In contrast, the increased proportion of industrial point source 457 emissions in INTAC significantly constrains the uncertainties associated with spatial proxies. Figure 6 shows the inter-458 comparisons of the percentage of point, on-road, and area source emissions between the INTAC and MEIC. Air pollutants, 459 especially those dominated by industrial combustion sources like SO₂, NO_x, PM₁₀, and PM_{2.5}, exhibit a significantly higher 460 proportion of point source emissions within INTAC compared to MEIC. In MEIC, the proportion of point source emissions for SO₂, PM₁₀, NO_x, and PM_{2.5} is 17%, 9%, 19%, and 7%, respectively. However, in the INTAC inventory, these percentages 461 462 substantially increase to 66%, 54%, 52%, and 48%, respectively, indicating a more accurate representation of spatial patterns. 463 For other species with emissions mainly from area sources (e.g., residential and transportation), there are limited improvements 464 in the proportion of point source emissions in INTAC.



465 Figure 6: Inter-comparisons of the percentage of point, on-road, and area source emissions between the INTAC inventory and MEIC.

466 To further assess the impact of point sources, Figure 7 takes SO_2 and YRD region as an example to compare the spatial 467 emission patterns between INTAC and MEIC. Figures 7c-e reveal that MEIC tends to overestimate emissions in urban centers and underestimate emissions in rural areas compared to INTAC. Amid economic growth and rapid urbanization, MEIC's use 468 469 of urban population as a proxy for spatial allocation becomes impractical as many factories relocate from city centers to rural 470 areas. To elucidate the difference between population-based and point-source-based allocation methods in emissions mapping, 471 we present the cumulative percentage of SO₂ emissions in MEIC and INTAC based on descending population orders in Fig. 472 7f. We use the grid groups where densely populated areas contribute 50% of SO_2 emissions in MEIC as an example, and 473 compare them with the cumulative percentage in INTAC across various grid sizes. The results show that at a resolution of 474 0.05° , INTAC only accounts for 17% of the emissions, while it reaches to 48% as the grid size increases to 1.0°. This suggests 475 that at a fine grid scale, MEIC tends to allocate more emissions to densely populated urban areas, while INTAC allocates a 476 larger proportion to suburban and rural areas, aligning better with the real-world emission spatial patterns. This mitigation of 477 bias through INTAC is especially notable at finer resolutions. The close cumulative percentage at 1.0° in the two inventories 478 can be attributed to the fact that urban and suburban areas often fall within the same grid, leading to a decreasing enhancement 479 in spatial accuracy achieved by INTAC. Figure 7g further presents the correlation between the spatial patterns of SO_2 emissions 480 in INTAC and various spatial proxies. At a resolution of 1.0°, the correlation coefficients between emission distributions and 481 factors (i.e., road networks, nighttime lights, total population, urban population, and rural population) fall within the range of 482 0.55 to 0.79. Nevertheless, at a resolution of 0.05° , the correlation coefficients range from 0.05 to 0.13. This indicates that at 483 higher spatial resolutions, INTAC substantially reduces the bias introduced by spatial proxies in MEIC.



Figure 7: Spatial pattern analysis of emissions in the INTAC inventory, using SO₂ emissions as an example. (a) and (b) display the spatial distributions of SO₂ emissions in MEIC and INTAC, respectively. MEIC emissions have been downscaled from 0.25 degrees to 0.1 degrees for comparison. To compare MEIC and INTAC in details, a zoom-in is applied to the YRD region. (c), (d), and (e) show spatial distributions of SO₂ emissions in MEIC, INTAC and their difference. Circles in (e) represent the center of a city. (f) compares cumulative percentage of SO₂ emissions in the INTAC inventory with those in MEIC across different spatial resolutions. The gridded SO₂ emissions, ranging from resolutions of 0.05 ° to 1.0 °, are cumulated in descending order of populations. The percentage annotations in different colors indicate the level of accumulated SO₂ emissions in INTAC at various spatial resolutions, when SO₂ emissions in MEIC reach 50%

491 accumulation. (g) shows correlation coefficient between SO₂ emissions in the INTAC inventory and multiple spatial proxies at different grid
 492 sizes.

493 3.3 Improvements on air quality modelling by INTAC

494 **3.3.1 Overall performance in key regions**

495 We conduct simulations using the WRF-CMAQ model driven by INTAC and MEIC separately to evaluate the improvements 496 in modeled air pollutant concentrations. Table 4 evaluates the simulated emissions in 74 major cities (locations depicted in Fig. 497 S2) against in-situ observations, with corresponding scatter plots shown in Fig. S3. The INTAC demonstrates an improved 498 agreement between modeled concentrations and ground-level observations, which benefits from the integrated high resolution 499 inventories. Compared to MEIC, INTAC leads to a decline in the mean bias of simulated major pollutant concentrations by 2– 500 $14 \,\mu g/m^3$, a reduction in the root mean square error by $4-19 \,\mu g/m^3$, and a decrease in the normalized mean error by 4-71%. 501 This finding indicates that INTAC produces a more accurate characterization of emissions in China overall. Furthermore, 502 given that atmospheric pollution monitoring stations are mainly located in urban areas in China, the observed differences 503 suggest that the INTAC can mitigate the overestimation of major pollutant concentrations in urban centers. As discussed in 504 Sect. 3.2.2, MEIC overestimates emissions in urban areas and underestimates them in rural and suburban areas, consequently 505 introducing uncertainties into air quality modeling. The improved accuracy in spatial distributions within INTAC significantly 506 contributes to enhancing the overall accuracy of air pollutant modeling.

Table 4: The discrepancies between simulated SO₂, NO₂ and PM_{2.5} concentrations and observed values for 74 major cities at a
 resolution of 12 km, using MEIC and INTAC as emission inputs. The statistical metrics used for comparison include R, MB, and RMSE.
 The bold font represents the difference of modeling performance between INTAC and MEIC.

Pollutants	Inventory	MB ($\mu g/m^3$)	RMSE ($\mu g/m^3$)	NME (%)
	INTAC	11	30	92
SO_2	MEIC	25	49	163
	Difference	-14	-19	-71
	INTAC	7	22	43
NO_2	MEIC	18	31	60
	Difference	-11	-9	-17
	INTAC	6	35	46
PM _{2.5}	MEIC	8	39	50
	Difference	-2	-4	-4

510 Figure 8 further compares the overall simulation performance between INTAC and MEIC in three key regions (BTH, YRD,

511 and PRD), with corresponding scatter plots shown from Fig. S4 to S6. Regarding PM_{2.5} and its precursors, MEIC shows a

512 considerable mean bias of up to 36 µg/m³ and a root mean square error of up to 59 µg/m³ in key regions. In contrast, INTAC

513 demonstrates the maximum MB values of 15 µg/m³ and RMSE values of 40 µg/m³. The correlation coefficients between 514 simulated and observed concentrations of the three air pollutants are generally lower in MEIC compared to those in INTAC. The modeling performance driven by INTAC, particularly for short-lived pollutants, experiences significant improvement due 515 to their strong correlation with spatial distributions of emission sources. Nonetheless, discrepancies between modeled and 516 observed surface concentrations still exist because of uncertainties from meteorological, physical, and chemical processes 517 518 within chemical transport models. Moreover, emission sources such as residential, transportation, agriculture in INTAC are 519 treated as nonpoint sources, and their allocation to grids using spatial proxies can introduce biases to air quality modeling. It is noteworthy that simulated ammonium concentrations by INTAC agree better with ground measurements than MEIC (Table 520 S3). While NH_4^+ concentrations are influenced by secondary chemical reactions, the improved model performance still reflects 521 522 the benefits from the integration of PKU-NH₃.



523 Figure 8: The Comparison of modeling performance across key regions (i.e., BTH, YRD, PRD) when using MEIC and INTAC as

524 emission inputs, respectively. The statistical metrics used for comparison include R, MB, and RMSE. The regions under comparison

525 comprise the BTH, YRD, and PRD.

526 **3.3.2 Improvements across different spatial resolutions**

527 To provide a more in-depth assessment of improved spatial patterns in INTAC, Figure 9 categorizes grid cells into different 528 bins based on their urban population and calculates the ratio of simulated pollutant concentrations to ground observations for 529 both INTAC and MEIC in each category. The results demonstrate that as urban population increases, the enhanced model 530 performance of INTAC over MEIC for SO₂, NO₂ and PM_{2.5} becomes more evident. Specifically, when the urban population is less than 50,000, both INTAC and MEIC exhibit a median range of simulated-to-observed concentration ratios close to 1. 531 532 However, as the urban population exceeds 550,000, the average range for MEIC widens to 1.4-5.2, whereas it remains within the range of 0.9–1.0 for INTAC. This indicates a significant improvement in mitigating the overestimation of emissions in 533 534 densely populated areas by INTAC. The incorporation of the industrial point source emission inventory for China, along with the YRD and PRD emission inventory significantly increases point source shares in INTAC, and thus producing better spatial representations for real-world emission distributions and smaller simulated deviations.

537 Model performance differences between MEIC and INTAC are influenced by grid size. Figure 10 presents the comparison 538 between modeled SO₂, NO₂ and PM_{2.5} concentrations against ground observations for 74 major cities at resolutions of 36 and 539 12 km. Increasing spatial resolution does not lead to a reduction in simulation errors, especially for MEIC. As the horizontal 540 resolution increases from 36 km to 12 km, the mean biases of simulated SO₂, NO₂, and PM_{2.5} concentrations using MEIC as 541 input show an increase from 37% to 143%, 11% to 46%, and -3% to 15%, respectively, when compared to in-situ observations. 542 In contrast, the simulation results using INTAC as input exhibit better agreement with ground observations, with mean biases for SO₂, NO₂, and PM_{2.5} increasing from 23% to 64%, -0% to 17%, and 2% to 11%, respectively. This is due to the fact that 543 544 the deviations in finer grid cells, whether overestimated or underestimated, tend to cancel out at a coarse spatial resolution. 545 The decoupling between emission spatial distributions with proxies at finer grids leads to more noticeable biases in air quality 546 modeling. Therefore, the findings suggest that the INTAC developed in this study can effectively constrain uncertainties in 547 emissions and the modeling bias, especially at fine spatial scales. The improvement will help tackle emerging challenges in 548 high-resolution air quality modeling in China.



Figure 9: Comparisons of modeling performance between INTAC and MEIC in different ranges of urban population. The 12 km grids are categorized to different bins according to the urban population residing within each grid. The ratio of simulated pollutant concentrations (Sim) to observed concentrations (Obs) for major pollutants (SO₂, NO₂, and PM_{2.5}) are calculated. The boxplot presents the upper quartile, median (red dot), and lower quartile of the ratios.



Figure 10: The comparison of modeled air pollutant concentrations and ground observations for 74 cities at 36 and 12 km resolutions, using MEIC and INTAC as emission inputs, respectively. The black dashed line represents the observational mean, and the annotations above the bar charts indicate the mean biases between simulated concentrations and the corresponding observed values.

556 4 Discussion

557 Both qualitative and quantitative uncertainty assessments are essential components of a comprehensive inventory for policy 558 or scientific purposes. Approaches such as error propagation and Monte Carlo simulation are commonly used for quantitative 559 uncertainty analysis in China's emission inventory (Lu et al., 2011; Streets et al., 2003; Zhao et al., 2011; Zhao et al., 2017b). 560 However, this study uses an integrated method rather than a unified framework to compile the high resolution emission 561 inventory for China. Collecting only emission quantities from the seven inventories without detailed calculation parameters makes it challenging to assess the overall uncertainties of INTAC here. We have summarized the estimated uncertainty range 562 563 for components of INTAC in Table 5, where such information is available. Although the uncertainties might be reported for a 564 year other than 2017, they still provide a rough representation of the uncertainty range in major air pollutant emission estimates 565 within INTAC. Species such as SO₂ and NO_x exhibit relatively low uncertainties, benefiting from well-established estimates 566 for large-scale combustion sources. The considerable uncertainties observed in BC and OC emissions may be attributed to 567 inaccuracies in the emission factors of the residential sector. Further details regarding the uncertainties of each component 568 inventory can be found in corresponding literature (An et al., 2021; Huang et al., 2021; Kang et al., 2016; Liu et al., 2016b; Yin et al., 2019; Zhao et al., 2011). 569

570 The uncertainties of INTAC also arise from the integrated process: (1) The emission sectors in all inventories need to be 571 mapped to the 88 standard sectors first. Due to limited foundational information for an aggregated sector's disaggregation, this 572 process may introduce biases for those who initially provide coarser source categories. For example, if an inventory only offers 573 one aggregated sector for power, which needs to be broken down into four subsectors (i.e., production of power, supply of 574 power, production of industrial heat power and production of residential heat power). We use the energy consumption for 575 corresponding sectors from the statistical yearbook as a reference basis for this allocation, which is a relatively reliable method 576 despite potential deviations. (2) To generate speciated VOC species, sectoral NMVOC emissions in each inventory need to be 577 matched to corresponding source profiles from the MEIC model. Discrepancies in emission source mapping can impact the 578 outcomes, which will be overcome by gathering more detailed sectoral information for each inventory or directly collecting 579 speciated species in future studies. (3) The INTAC is made publicly available at a monthly scale, given that the majority of its 580 components are gathered on a monthly or annual scale. The temporal disaggregation to finer resolutions for modeling is 581 achieved using empirically selected weighting factors in the MEIC model. However, it is noteworthy that the parameters 582 employed for allocating emissions to daily or hourly scales remain fixed and do not vary over time or region, introducing 583 additional uncertainties. In the future, we plan to incorporate more advanced data or method (e.g., real-time emission 584 measurements) to enhance temporal accuracy at finer scales, as indicated in the previous work for the power sector (Wu et al., 585 2022). (4) The border issue is inevitable when emissions for the same species in two adjacent cities are derived from different 586 inventories. A typical example is the cities located at the boundary of the YRD or the PRD regions. In the INTAC, we 587 downscale all emissions to 1 km before spatial-temporal coupling process, thereby mitigating this uncertainty to some extent.

Emission inventory	Reporting year	SO ₂	NOx	СО	NMVOC	NH3	PM10	PM2.5	BC	OC	References
PKU-NH ₃	2012					-26– 25					(Kang et al., 2016)
The shipping emission inventory for East Asia	2013	<u>+4</u>	<u>±</u> 4	±5	<u>+4</u>			<u>+4</u>	<u>+4</u>	<u>+</u> 4	(Liu et al., 2016b)
The open biomass burning emission inventory for China	2003– 2017	-67– 67	-78– 98	-54– 56		-44– 89	-74– 84	-65– 65	-75– 100	-74– 81	(Yin et al., 2019)
The PRD emission inventory	2017	-17– 20	-25– 28	-30– 39	-34–50	-50– 86	-45– 60	-43– 62	-53– 116	-54– 160	(Huang et al., 2021)
The YRD air pollutant	2017	-29– 36	-28– 33	-42– 75	-44–68	-58– 117	-36– 62	-30– 46			(An et al., 2021)
emission inventory	2005	-14– 13	-13– 37				-14– 45	-17– 54	-25– 136	-40– 121	(Zhao et al., 2011)

588 Table 5: Uncertainties in the inventory components of INTAC, contingent upon the availability of such information (Unit: %).

The INTAC for 2017 is subject to some limitations: (1) In the integrated method, emissions data for the same city and species across different sectors may come from different datasets. Similarly, emissions data for different species within the same city and sector may also originate from different datasets. The utilization of species ratios requires careful consideration in these cases. (2) Limited resources present a substantial challenge in gathering emission inventories over extended time series from 593 diverse research institutions within the scope of this study. Consequently, we exclusively present the INTAC for the year 2017,

594 with the possibility of extension to other years in subsequent research.

595 **5 Data Availability**

596 Data described in this manuscript can be accessed at Zenodo under https://doi.org/10.5281/zenodo.10459198 (Wu et al., 2024)
597 and http://meicmodel.org.cn/intac.

598 6 Concluding remarks

599 Compiling a comprehensive bottom-up emission inventory for China that achieves both extensive coverage and high resolution 600 poses a significant challenge. In this work, we construct a 0.1 ° resolution integrated inventory for 2017 through the fusion of 601 multi-source emission inventories. An integration model has been developed to effectively couple heterogeneous emission 602 datasets, aimed at generating a standardized data cube with consistent sectors, species, and spatial-temporal resolution. The 603 INTAC is created through source mapping, species mapping, temporal disaggregation, spatial allocation and spatial-temporal 604 coupling. Six representative emission inventories focusing on national and regional scales, as well as key species and sources 605 in China are merged with MEIC. This integration harnesses the strengths of each inventory, resulting in an improved depiction 606 of emission totals and spatial distribution patterns for China.

607 We find that the total emissions of SO₂, NO_x, CO, NMVOC, NH₃, PM₁₀, PM_{2.5}, BC, and OC in INTAC for 2017 are 12.3, 24.5,

608 141.0, 27.9, 9.2, 11.1, 8.4, 1.3 and 2.2 Tg, respectively. Industrial production serves as the main source of various atmospheric 609 pollutants. Residential sources contribute over 40% to CO, BC and OC emissions. Apart from agricultural sources, which 610 account for 83% of NH_3 emissions, the contributions from various minor emission sources cannot be overlooked. This study emphasizes the significance of shipping emissions, particularly in contributing to SO_2 (13%) and NO_x (13%). Fossil fuel 611 612 combustion dominates the emissions of PM₁₀, PM_{2.5}, CO, BC, SO₂, and NO_x, ranging from 38% to 80%. The enhancement in emission estimates for China in INTAC is demonstrated by the comparison with MEIC. For instance, the incorporation of 613 614 numerous point sources has notably addressed MEIC's tendency to overestimate emissions in urban centers, particularly at 615 higher spatial resolutions. In comparison to MEIC, INTAC exhibits a mean bias reduction in simulated concentrations of major 616 pollutants against ground observations across 74 cities, ranging from $2-14 \ \mu g/m^3$. The improvement in model performance 617 achieved by INTAC is particularly noticeable at finer spatial resolutions.

618 Our study offers an efficient framework for creating highly-resolved emission inventory on a large scale. This approach 619 integrates advantages from previous studies and holds the potential to support policymakers in making well-informed decisions 620 for improving air quality. In the future, we anticipate the ongoing incorporation of additional emission datasets to offer a more

621 reliable representation of emissions in China over extended time periods.

622 Supplement

623 The supplement related to this article has six figures and three tables.

624 Author contributions

Nana Wu, Guannan Geng and Qiang Zhang designed the study. Nana Wu developed the INTAC emission inventory and

626 conducted chemical transport modelling, and analyzed the emissions. Junyu Zheng, Yu Song, Huan Liu, Yu Zhao, Ying Zhou

and Qinren Shi provided the emission inventories for the integration. Ruochong Xu helped with the data analysis. Shigan Liu

628 compiled the chemical transport model. Xiaodong Liu contributed to the design of computer programmes for the integration

629 model. The manuscript was written by Nana Wu and Guannan Geng, and it was revised and discussed by all coauthors.

630 Competing interests

631 The authors declare that they have no conflict of interest.

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