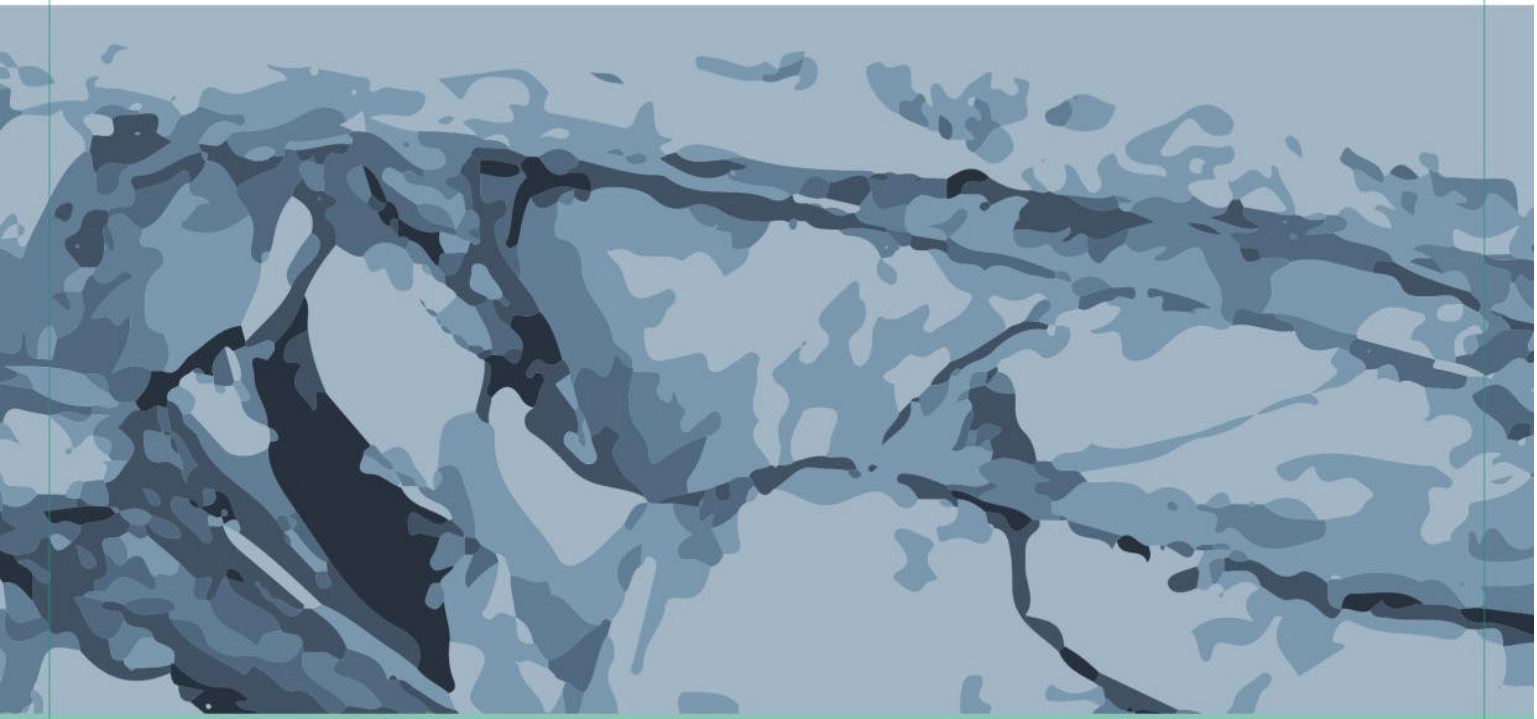
Review report 3

ChiNorBC

The review report for impact assessment of BC/OC on air quality and climate change



Chinese-Norwegian Project on Emission, Impact, and Control Policy for Black Carbon and its Co-benefits in Northern China

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The review report for impact assessment of BC/OC on air quality and climate change

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About this report

This report is part of a series of outputs produced under the Chinese-Norwegian Project on Emission, Impact, and Control Policy for Black Carbon and its Co-benefits in Northern China (ChiNorBC). The project is jointly implemented by the Chinese Research Academy of Environmental Sciences (CRAES) and the Norwegian Environment Agency (NEA), in partnership with the Chinese Academy of Environmental Planning (CAEP), the Norwegian Institute of Public Health (NIPH) and CICERO Center for International Climate Research, with financial support from the Norwegian Ministry of Foreign Affairs.

There is no internationally agreed definition of black carbon (BC) and organic carbon (OC). BC is the light-absorbing component of fine particles and is produced by incomplete combustion of fossil fuel, biofuel and biomass. BC is always co-emitted with OC. Emissions of BC and OC affects the climate and have adverse health effects. Reductions of BC and OC will have co-benefits for climate, air quality and health.

ChiNorBC will develop improved emission inventories for BC- and OC- emissions in China using the most recent, best available national statistics and measurements obtained in the project. Based on this, new estimates of effects of BC/OC on climate, air quality, and health will be provided. The project will further raise scientific, governmental, and public awareness and enhance the understanding of the positive impacts of BC/OC emissions reductions. Ultimately the ChiNorBC will provide Chinese policy makers with policy solutions for reducing BC/OC emissions in China which maximizes the co-benefits.

The project has six outputs. This report is a result of Output 3, Review of impact assessment of BC/OC on air quality and climate change. For a more comprehensive description of the project, and to get access to all the project reports, please visit the project web site http://chinorbc.net/.

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Findings and opinions expressed in this paper are not necessarily shared by those contributing to the work, and any errors and omissions are the responsibility of the authors and partner institutions.

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The cover design is non-figurative and created by Eilif Ursin Reed, Communication Advisor, CICERO, and is inspired by melting glaciers and polluted snow.

About the partner institutions in the ChiNorBC-project

Chinese Research Academy of Environmental Sciences

The Chinese Research Academy of Environmental Sciences (CRAES), founded in 1978, is a leading institute in environment-related studies in China, including studies on short-lived climate pollutants and its impacts. There are more than 1000 employees whose backgrounds cover all important areas of environmental sciences, including atmospheric science. One of the main responsibilities of CRAES is to provide technical and scientific support for decision making of Ministry of Ecology and Environment (MEE).

Norwegian Environment Agency (NEA)

The Norwegian Environment Agency (NEA) is an advisory and executive body under the Ministry of Climate and Environment (MCE), fully funded by the Norwegian Government. It has about 700 employees in Trondheim and Oslo as well as in local offices throughout the country. NEA was established 1st July 2013 after a merge of the former Directorate for Nature Management (est. 1965) and the Climate and Pollution Directorate (est. 1974). The Norwegian Nature Inspectorate (SNO) is organized as a department within NEA. The primary tasks of NEA are to reduce greenhouse gas emissions, manage Norwegian nature, and prevent pollution.

Chinese Academy of Environmental Planning (CAEP) is a public institution with independent legal status founded in 2001. Its missions are: Carrying out strategic research on national ecological civilization, green development and beautiful China, and undertaking technical support for the preparation and implementation of national medium and long-term ecological environment planning, key river basins and regions planning, and environmental planning in key fields, so as to meet the major needs of the country.

The Norwegian Institute of Public Health (NIPH) is a Norwegian government agency and research institute and is Norway's national public health institute. NIPH acts as a national competence institution placed directly under the Ministry of Health and Care Services, with approximately 1400 employees in Oslo and Bergen. It is responsible for knowledge production and systematic reviews for the health sector and provides knowledge about the health status in the population, influencing factors and how it can be improved.

Center for International Climate Research (CICERO) is a private foundation that for over thirty years has delivered interdisciplinary research of high scientific quality on climate science, economics, and policy. CICERO's mission is to conduct research and provide reports, information and expert advice about issues related to global climate change and international climate policy with the aim of acquiring knowledge that can help mitigate the climate problem and enhance international climate cooperation. CICERO has approximately 80 employees situated in the Oslo Science Park.

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Abbreviations and concepts

BTH	Beijing-Tianjin-Hebei
BC	Black carbon
CO	Carbon monoxide
CTM	Chemical transport model
GTP	Global temperature change potential
GWP	Global warming potential
NO_x	Nitrogen oxides. Comprises nitric oxide (NO) and nitrogen dioxide (NO_2)
OC	Organic carbon
$PM_{2.5}$	Fine particles with a diameter of 2.5 micrometers or less
PM_{10}	Inhalable particles with a diameter of 10 micrometers or less
SLCFs	Short-lived climate forcers
SO_2	Sulfur dioxide
ME	Mean Error
RMSE	Root Mean Square error
NMB	Normalized Mean Bias
NME	Normalized Mean Error

Summary

This review is a delivery from Output 3 of the ChiNorBC project and contains a literature survey and summary of the impact of black carbon (BC), organic carbon (OC) and other pollutants on climate change and air quality in recent years. BC and OC play an important role in air quality and climate change in China, and high concentrations of carbonaceous aerosols may exacerbate the severity of smog and increase the complexity of air pollution.

BC, OC and other aerosols affect the climate through their interactions with solar radiation and clouds. Most aerosol species have a cooling climate impact, while BC stands out by absorbing solar radiation and warming the atmosphere. There are significant uncertainties, but the best estimate of the global aerosol effect on climate is a net cooling that has masked a notable fraction of the anthropogenic warming induced by greenhouse gas emissions to date. Additionally, aerosols influence local and regional climate and weather, such as the precipitaiton, haze events, and extreme weather, although the exact nature and extent of their role is still a topic of ongoing research. The significant range in estimates of aerosol's climate effects stem from uncertainties in their distribution, atmospheric processing, and optical and microphysical properties, as well as uncertainties in their sources. Despite progress over the past decades, representing aerosols remain a challenge for global and regional models of the atmosphere, with validation in many cases hampered by lack of observations.

China is currently the world's largest emitter of BC due to its heavy use of solid fuels and high population density. OC emissions are also higher in China compared to BC emissions, which were 2.69Tg and 1.66Tg respectively in 2015, accounting for 50.0% and 39.8% of emissions from Asia. Although there have been no official measurement program of environmental BC and OC in China in the past decade, many recent measurements about BC/OC concentration have been reported in the literature. According to these measurements, the spatial distribution of BC/OC concentrations shows a trend of high concentrations in and around Beijing, Tianjin and Hebei (BTH) and low concentrations in the southeastern regions. The seasonal average concentration of BC in cities in The BTH was generally less than 2μg/m^3 in spring and summer, and up to 10μg/m^3 in autumn and winter. The average concentration of OC in autumn and winter is generally above 20μg/m^3, and some cities of BTH can reach 40μg/m^3.

In the literature, emission trends in China over the last decade have been compared with observations, and the BC concentration simulated by using most emission inventories are basically consistent with the observed results. Despite the significant advantages of modelling results over short time series in some cities, accurately modelling long-term regional spatial and temporal variations in BC/OC concentrations remains a challenge, and the accuracy of the emission inventory is an important basis for obtaining reliable BC/OC concentrations.

Different air quality transport models require different validation parameters (e.g. ME, NMB, NME, RMSE) to evaluate their performance in different ways. Therefore, there is no universal measurement standard that applies to all conditions. In the ChiNorBC project, the main objective of Output 3 is to

evaluate the performance of the model using current and updated emission inventories and to compare the atmospheric pollutant concentrations simulated by the model with the observed values in the base year. The indicators we finally selected included correlation coefficient (R), normalized mean deviation (NMB), normalized mean error (NME), etc.

1.1 Impact assessment of BC/OC on air quality and climate change

1.1.1 Status of Knowledge about BC/OC Impacts on Climate

Aerosols play a key role in shaping regional climate and air quality. Depending on type, they scatter and absorb solar radiation, causing a direct (i.e. aerosol-radiation) radiative forcing (RF_{ari}), and affect the properties of clouds (indirect, aerosol-cloud, radiative forcing, RF_{aci}). Globally, the combined effect of aerosols is an effective radiative forcing (ERF[1]) estimated to be -1.3 W m^{-2} over the industrial era (year 1750-2014 (Forster et al. 2021)), but with considerable uncertainty [-2.09 to -0.6 W m^{-2}]. Considering the period 1750-2019, the total aerosol ERF is assessed to be smaller in magnitude –1.1 [–1.7 to –0.4] W m^{-2} due to recent emission changes, but aerosols have nevertheless masked a significant fraction of the greenhouse gas induced warming to date (e.g., Samset et al. 2018a).

BC and OC are two of the key aerosol species that receive attention in the science and policy arena due to their dual impacts on climate and air quality. Originating from incomplete combustion of fossil fuels, biofuels, and biomass, OC is the fraction of carbonaceous aerosols which contain compounds of carbon and BC is soot made almost purely of carbon. Here we outline some key points and remaining knowledge gaps related to their atmospheric distribution and climate effects. Note, however, that this is not a systematic review. For a more comprehensive overview, we refer to existing assessments, e.g., Bond et al. (2013) and Lee et al. (2013).

The impact of BC aerosols on climate is a complex interplay of many mechanisms, including absorption of solar radiation, changes in cloud properties and cover, darkening of snow covered surfaces and rapid adjustments in the atmosphere (Bond et al. 2013). While most aerosols have a cooling impact on the climate, BC acts to warm the atmosphere by absorbing solar radiation, causing a positive RF_{ari}. There is a significant spread in estimates of the forcing of BC aerosols: the IPCC Sixth Assessment Report (AR6) reported a best estimate ERF of 0.11 W m^{-2} due to BC emissions, with a range from -0.20 to 0.42 W m^{-2}. This value is lower than the RF_{ari} (over the year 1850-2011) of 0.4 (0.05 to 0.8) W m^{-2} assessed in the previous IPCC report (Myhre et al. 2013a). The reason is partly the metric used, i.e. ERF versus RFari, where the former also includes the rapid adjustments to the BC forcing. Recent work suggests that these rapid adjustment, due to changes in clouds, lapse rate and atmospheric water vapor, act to offset a substatinal portion of the positive RF_{ari} of these aerosols, resulting in a weaker net ERF and relatively weak global-mean surface temperature response (Stjern et al. 2017; Takemura et al. 2019). BC aerosols contribute further to radiative forcing through depostion on snow and ice, darkening the surface and increasing absorption and snow melt. Considerable uncertainties exist in estimates of BC concentrations of in the snowpack (Dou and Xiao 2016) and in the processes and snow characteristics

[1] The change in the net energy flux at the top of the atmosphere of the Earth system due to an imposed perturbation, such as changes in greenhouse gas or aerosol concentrations, after allowing for rapid adjustments in e.g. cloud and vertical temperature (Forster et al. 2021).

that determines the subsequent albedo change, although scientific progress has been made over the years (Forster et al. 2021). In AR6, the best estimate of the global instantaneous RF (IRF) due to BC on snow deposition is assessed to be 0.04 [0.00 to 0.09] W m^{-2} (Forster et al. 2021), similar to AR5. To account for a higher efficacy (i.e. stronger temperature response per unit RF than for a correponsing RF by CO_2) of the BC-on-snow effect, the IRF estimate is doubled, giving an overall assessed ERF of +0.08 [0.00 to 0.18] W m^{-2}.

While the impact of BC on global mean surface temperature has been assessed to be lower than previously thought, BC still plays in an important role for regional climate, including through modulating the hydrological cycle. The sensitivity of the regional climate to reductions in aerosol emissions has been found to be especially high in the Asian region (Samset et al. 2018b).

The wide spread and high uncertainty in BC forcing estimates stem from high variability in atmospheric distribution and sign of rapid adjustments in global models, which in turn is related to uncertainties in emission inventories and model parameterizations of the processes that affect the subsequent transport and depositon of BC in the atmosphere. After being emitted, the aerosols get transported away from the source regions while undergoing chemical aging and scavenging processes along the way. Insufficient process understanding and model representation, as well as limited observations for validation, leads to uncertianties in the long-range aerosol transport and atmospheric lifetime. For instance, BC emitted in Asia has been shown to reach the Arctic where the aerosols can be deposited on snow and contribute to further warming (Ikeda et al. 2017; Qi and Wang 2019). However, the exact role of Asian emissions for Arctic BC levels isnot sufficiently constrained due to uncertainties in, and different model treatment of, the atmospheric lifetime, aging, and wet and dry removal of the aerosols (Bourgeois and Bey 2011; Browse et al. 2012; Liu et al. 2011; Lund et al. 2012) .

The vertical distribution of BC is also a critical factor for quantifying the subsequent climate impcact, as the BC efficacy (i.e., radiative forcing per kg BC) increases strongly with altitude. Global models have had a tendency to have overestimate high-altitude BC concentration, in particular in remote oceanic regions (e.g. Schwarz et al. 2010; Katich et al. 2018), which in turn may mean they overestimate the BC impact (Ban-Weiss et al. 2011; Samset et al. 2015). Several studies have pointed to a too long lifetime of BC in many current models as the main cause of this discrepancy and suggested that a global mean BC lifetime of less than approximately 5 days is required for reasonable agreement between modeled and measured vertical BC profiles (Samset et al. 2014; Lund et al. 2018b; Hodnebrog et al. 2014; Wang et al. 2014). Over recent years, considerable work has been undertaken to understand the processes underlying the model-measurement discrepancies and improve the model performance (e.g. Fan et al. 2012; Kipling et al. 2013; Zhang et al. 2012; He et al. 2016; Lund et al. 2017; Xu et al. 2019; Liu and Matsui 2021). Nevertheless, as summarized by the IPCC AR6, *"the lack of global scale observations of carbonaceous aerosol, its complex atmospheric chemistry, and the large spread in its simulated global budget and burdens means that there is only low confidence in the quantification of the present-day atmospheric distribution of individual components of carbonaceous aerosols.*"

Further uncertainty in the climate impact of BC arises from uncertianties in the optical properties, how efficiently the aerosols absorb radiation, and how that absorption is affected when BC becomes mixed

with other aerosol in the atmosphere. Studies also indicate differences between urban and rural environment. A recent review by Samset et al. (2018c) provide the current status of knowledge about aerosol absorption, the main uncertainties and needs for future improvements.

OC is co-emitted with BC but is predominantly scattering (i.e., a negative RF_{ari}). There is also a non-negligible absorbing component, at least for certain sources. These absorbing organic aerosols, often referred to as brown carbon (BrC), is a source of significant uncertainty in estimates of total aerosol absorption. OC also contribute to aerosol-cloud interactions, and hence to a negative RF_{aci}. The most recent best estimate ERF of OC emissions is -0.21 [-0.44 to +0.02] W m^{-2} (Naik et al. 2021). Global modelling and understanding of OC is confounded by many of the same uncertainties as BC.

1.1.2 BC/OC pollution Status in concerned regions in China

BC and OC plays an important role in air quality and climate change in China (Bahner et al, 2014; Ji et al, 2019; UNEP/WMO, 2011). BC and OC are always emitted together, but the proportion varies depending on the source. Zhang et al. (2014) has highlighted the increasing role of OC in air pollution as well as in climate change in China generally. Their results show that anthropogenic aerosols present in $PM_{2.5}$ (including BC and OC) were in high concentration in some Chinese cities. Hence, a high concentration of carbonaceous aerosols could strengthen the seriousness of haze and increase the complexity of air pollution (Han et al., 2011; Yang et al, 2017).

Although its per-capita emission of BC is generally not high compared to that of developed countries, China is presently the highest emitter of BC globally (Wang et al., 2012; Zhang et al., 2013; Hoesly et al., 2018; Kurokawa & Ohara, 2019; Choi et al., 2020). A recent study by Wang et al. (2014) suggests that the BC emissions in China could even be 2 or 3 times higher than current estimates**.** Due to the heavy use of solid fuels and high population density (Huang et al. 2015), the OC emissions in China are also high compared to BC emissions, it was reported that anthropogenic emissions of OC in Asia were 8.88Tg, among which 2.56Tg were from China in 2000 (Ohara et al. 2007). In 2015, emissions of BC and OC in China were 1.66Tg and 2.69Tg respectively, accounting for 50.0% and 39.8% emissions in Asia.

Although no routine ambient BC and OC measurement are available in China for the last decade, many measurements of the recent status are reported in the literature (Zhang et al., 2019). Statistics and calculation of long-term averages (2013-2017) of the results of studies on the characteristics of changes in BC/OC concentrations in general show that the concentration of BC/OC in the northern and southwest China (Hu et al. 2015; Yang et al. 2016; Wang et al. 2011; Huang et al. 2014) is higher than that in the southeast and northwest China (Chen et al. 2016; Feng et al. 2014; Zhuang et al. 2014; Tao et al. 2017). Pronounced seasonality of BC and OC concentration existed over Northern China. For cities over the Beijing-Tianjin-Heibei area, the seasonal mean concentration of BC was generally no more than 2μg/m^3 in spring and summer, while it increased to 3-10μg/m^3 in fall and winter seasons. For some cities the seasonality was even more pronounced. For example, the monthly mean BC concentration reached above 18μg/m^3 in January in Xi'an, a city in the Northwest of China, and it was

only 2μg/m^3 in June or July. However, the seasonal variation of BC concentration was not significant in cities such as Beijing, Nanjing (Zhang et al. 2019), Shanghai (Ming et al. 2017; Chang et al. 2017). In terms of seasonality in OC, spring and summer concentration were significantly lower than in fall and winter. Summer OC was the lowest among the seasons, where OC concentration never reached levels higher than 20μg/m^3. In fall and winter, the seasonal mean OC was generally above 20μg/m^3, with some cities in BTH being up to 40μg/m^3. The seasonality was more pronounced in Xi'an as well, where OC reached close to 60μg/m^3 in January and down to below 10μg/m^3 in June or July (Zhang et al. 2019).

In addition to the characteristics of BC/OC concentration changes and reductions obtained from literature studies, official formal monitoring studies of BC/OC are gradually being carried out, and regular measurements of $PM_{2.5}$ components have started in the "2+26" key cities in and around BTH and the surrounding areas after 2016, along with the "Pollution Prevention and Control Campaign". We attempted to obtain an overall picture of BC and OC pollution in northern China by integrating all available data. By collating the raw monitoring data from 2013-2017 obtained from the literature and the official observations of BC/OC from BTH and surrounding areas, we found that the measurements were most abundant in Beijing, Tianjin, Hebei and surrounding areas. However, the data are only representative of the main urban areas as the measurements were made at urban sites. Since most of the measurements from the literature were sporadic, and not designed systematically and operated routinely, the full picture of the status of BC and OC pollution could not be obtained at this stage. Along with establishment of a planned monitoring network for $PM_{2.5}$ components (including BC and OC), the pollution status of BC and OC in a specific part of Northern China, such as over BTH, could be obtained in near future. To obtain the full picture of Northern China, more time and effort should be taken.

1.2 Regional air quality over northern China

1.2.1 Summary of regional air quality simulation research activities

Chemical Transport Models (CTMs) such as CAMx, CMAQ, and WRF-Chem have been applied to modelling studies. Those classic CTMs have been widely employed for estimating the formation and transport of air pollutions (Chen et al.,2020), and simulate the impact of pollutant emissions on air quality (Gu et al.,2018).

The results of the simulation study and air quality monitoring data indicate that $PM_{2.5}$ air quality in northern China has gradually improved, but surface ozone pollution has increased in recent years (Li et al.,2019a, Cheng et al.,2019). The approximately40% decrease of $PM_{2.5}$ over the 2013–2017 period in the North China Plain (Li et al.,2019b) is mainly attributed to reductions in anthropogenic emissions (Silver et al.,2020) rather than meteorological variations (Zhang et al.,2019), moreover, natural emissions also have a significant impact on the increase in air quality (Yang et al.2017). Therefore, accurate and up-to-date emission inventories (Zheng et al.,2019, Zhou et al.,2017 Zhou et al.,2018) are essential for better performance on air quality modelling.

In addition to air quality effects, long-range transport of pollutants from Asia has been shown to exert large impacts on the formation and dissipation of haze (An et al.,2019, Zhang et al.,2019). Long-range transport of local air pollutant emissions from cities, such as fine particulate matter, can also influence air quality in rural areas surrounding or downwind the city (Ding et al.,2016), and long-range transport is also another source of secondary particulate matter (Wang et al.,2016) or fine aerosols (Chuang et al.,2003). Source apportionment tools in CAMx and precursor tagging methodology in CMAQ has been used to calculate the local and distant source contributions (Lin et al.,2016) to ozone, precursor, the primary and secondary PM species concentrations among the selected source groupings (Mailroom,2010). The sources of air pollutants vary widely across China, the major contributors to $PM_{2.5}$ concentrations are industrial or residential over northern China (Zhang et al.,2017b, Qiao et al.,2018, Shi et al.,2017, Liu et al.,2020), the high emission rates of BC and OC emissions from open burning of biomass (Qin and Xie, 2011) and $PM_{2.5}$ emissions from straw combustion (Zhang et al., 2016) are a considerable primary contribution to $PM_{2.5}$ concentration in northern China (Zhang et al.,2017a). Nevertheless, motor vehicles and regional sources are the most important sources of ozone formation, followed by power plant and biogenic sources (Li et al., 2012, Streets et al.,2007). A large proportion of O_3 concentration can be attributed to external contributions (Wang et al.,2019, Gao et al., 2016, Qu et al.,2014).

The O_3 source apportionment attributes in-situ O_3 formation to a NO_x or VOC source based on the index values that determine the sensitivity of O_3 formation to the NO_x or VOC-limit (Wang et al.,2019a), and CTMs can also determine the source sensitivities of modelled chemical species to emission (Wang et al.,2019b). The urban centers and the developed industrial areas of northern China are predominantly VOC-sensitive conditions, while both VOCs and NO_x-limited sensitive and NO_x-sensitive conditions predominate in suburban and remote areas, respectively (Xie et al.,2014, Han et al.,2018, Xu et al., 2019). Furthermore, CTMs has been used in different recent studies to study regional photochemical pollution (Tang et al. 2017). It is also possible to investigate what physicochemical processes that govern pollutant concentrations at different times and locations through the process analysis tools in CTMs. This can reveal, for example, that atmospheric processes can counteract the scavenging effect of chemical reactions on O_3 (Wang et al., 2019a), the relationship between visibility and aerosol optical depth (Lin et al.,2016, Li et al.,2019), and chemical formation and transformation (An et al. 2019) in China.

A comparison of the performances on simulating the $PM_{2.5}$ over East Asia in 2010 among the twelve CTMS (WRF-CMAQ (v4.7.1 and v5.0.2), WRF-Chem (v3.6.1 and v3.7.1), GEOS-Chem, NHM-Chem, NAQPMS, and NU-WRF) can be found in Tan et al. (2019). The study shows that the models behave very differently in terms of particle concentration simulations, with gas particle conversion being one of the main reasons for the different performance of the models in terms of particle concentration simulations. Ma et al. (2019) applied and evaluated four regional air quality models (WRF-Chem (version 3.9.1), CHIMERE (version 2017r4), CMAQ (version 5.2), and CAMx (version 6.50)) to simulate dust storms in East Asia. They found large differences in the model results of simulated PM_{10} concentrations. It is also evident from numerous model performance evaluation studies of CTMs that differences in the physical and chemical mechanisms used in the models affect the simulation results,

but the model performance evaluation results from the studies that appear in this paper all achieve the 'goal' range of the benchmark, which is the model performance that most studies are able to achieve (Emery et al., 2017; Huang et al., 2021).

In the National Key Research and Development Project "Research on Regulatory Air Quality Modeling Technology System"(2018-2021), a database with regional CTM validation has been established, including nationwide routine monitoring data of O_3, NO/NO_2, CO, SO_2, PM_{10}, $PM_{2.5}$ from 2017-2019 for 338 cities from China's national air quality monitoring network. $PM_{2.5}$ components (including BC/OC), VOCs and radicals of 17 severe ozone, $PM_{2.5}$, and sand-storm pollution cases from super research stations, and $PM_{2.5}$ composition data of 2017/18-2018/19 winters of Beijing-Tianjin-Hebei and Surrounding Cities (2+26 cities). The validation of regional CTM models of CMAQ model (version 5.02 and 5.3.2), CAMx model (version 6.20 and 7.1), NAQPMS model, and RegAEMS model have been conducted and the targeted air pollution species include SO_2, NO_2, CO, O_3, $PM_{2.5}$, PM_{10} and $PM_{2.5}$ component of sulfate, nitrate, ammonium, BC and OC.

1.2.2 Status of BC/OC simulation

Currently, regional air quality can be simulated using the Community Multiscale Air Quality (CMAQ) model, the Comprehensive Air Quality Model with extensions (CAMx), Weather Research and Forecasting model with Chemistry (WRF-Chem), as well as other air quality models. The anthropogenic emission inventory currently used in studies are the Multi-resolution Emission Inventory for China (MEIC), the Regional Emission inventory in Asia version 2 (REAS2), the Regional Emission inventory in Asia version 3.1 (REASv3.1), Emissions Database for Global Atmospheric Research (EDGAR), the Community Emissions Data System (CEDS), transport and Chemical Evolution over the Pacific (TRACE-P), the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B), etc. Most anthropogenic emissions inventories are slow to update (REAS2 (Kurokawa et al., 2013), EDGAR (Crippa et al., 2018), CEDS (Hoesly et al., 2016)) or have been discontinued (TRACE-P (Streets et al., 2003), INTEX-B (Zhang et al. 2009)). Therefore, continued efforts to update and improve the anthropogenic emissions inventory in China based on the latest fuel consumption statistics and emission factors are essential.

According to different anthropogenic emission inventories, the annual BC emissions increased gradually from 1949 (341 Gg/yr) to 1996 (2189 Gg/yr), with a fluctuating upward trend from 1996 to 2007 (1850 Gg/yr) (Wang et al., 2012). Another study shows that the annual emissions of BC in China have shown a decreasing trend in recent years from 2009 (1650 Gg/yr) to 2019 (1100 Gg/yr) (Kanaya et al., 2019). Similarly, the annual OC emissions increased gradually from 1990 (341 Gg/yr) to 2000 (2189 Gg/yr) (Huang et al., 2015), with a fluctuating upward trend from 2000 (2738 Gg/yr) to 2014 (4297 Gg/yr) (Hoesly et al., 2017). Yet another study shows the annual emissions of OC in China have shown a decreasing trend in recent years from 2015 (2500 Gg/yr) to 2017 (2100 Gg/yr) (Silver et al., 2020). Emissions of BC and OC from different sources vary considerably in published inventories, but the underlying trends are consistent (Zhang et al., 2009, Kurokawa et al., 2019, Crippa et al., 2018, An et

al., 2019, Chang et al., 2018). Spatially, China's BC and OC emissions are consistent with regional economic development and rural population density distribution, showing a trend of high in the east and low in the west (Zhang et al., 2013, Cao et al., 2006, Hoesly et al., 2016, Huang et al., 2015). In northern China, there are strong seasonal variations in BC and OC emissions due to residential heating and agricultural open burning, with peaks in winter and lower emissions in spring and summer (Cao et al., 2006, Liu et al., 2016); in southern China, BC is mainly emitted from mobile emission, so seasonal variations are not significant, while OC emissions are relatively high in winter, due to the important contribution of crop residue combustion (Zheng et al., 2012).

An essential component of all modelling studies is the evaluation of model performance for the simulation. There have been several recent regional studies of BC/OC simulation and evaluation using chemical transport models (CTMs) in China. Some of these studies simulated ground-level BC and OC concentrations in China using CMAQ model system with different emission inventories (Zhang et al. 2004, Zhang. 2005, Zhang et al. 2007, Wang et al. 2010, Chen et al. 2019 and Hu et al. 2017). The simulated values of BC and OC are generally in good agreement with the observed ones. In some regions, the simulated values are slightly lower than the observed values, with the normalized mean bias between model simulation and observation concentrations within -50% (Wang et al. 2010). Underestimation of pollutant concentrations is largely due to underestimation of primary emissions (Hu et al.,2017).

There are relatively few modelling studies on BC and OC compared with $PM_{2.5}$ and O_3 over China so far. One reason should be attributed to the unavailable long-term data of BC and OC via routine measurement at sites representing regional characteristics. Long-term observations of BC mass concentrations in East Asia (2006-2015) and assessment of model performance and uncertainties in Asia has been obtained in the MICS-Asia project (Han et al. 2010, Chen et al. 2019). The accuracy of the model simulations was improved when Wang et al. (2016) simulated the emissions using the monthly average BC measurements after inversion. It should be noted that the above evaluation work on BC and OC simulation was based on limited observations at some specific sites. However, along with the implementation of a $PM_{2.5}$ component network over Northern China and the detailed compilation of BC and OC emission inventory, further modelling works on BC and OC will be carried out in the project.

1.3 Impact Evaluation Methodology

This section outlines the methodology, models and tools used in the ChiNorBC project to assess and study the changes and distribution of BC/OC in the atmosphere, and the impact on climate, resulting from emission in China.

1.3.1 General methodology

The impact of BC/OC emissions in the study region on air quality, radiative forcing, and climate will in the ChiNorBC project be quantified with a stepwise approach using a suite of modelling tools of varying degrees of complexity. A critical component will be the use of updated national/regional emission estimates.

To determine the impact of the emissions on the atmospheric distribution of aerosols, we will perform simulations with two state-of-the-art chemistry-transport models, the regional CMAQ (https://www.cmascenter.org/cmaq/) and the global OsloCTM3 (https://www.cicero.oslo.no/en/osloctm3). Chemical transport models (CTMs) are numerical models that simulate emissions, transport, formation, deposition, and the fate of multiple air pollutants in the atmosphere (Brasseur, Jacob 2017) and may give air quality forecasts. CTMs simulate these processes to describe the spatiotemporal distribution of aerosols and trace gases, using meteorological information as input (Schaap et al. 2012). CTMs can be classified according to their methodology as Eulerian and Lagrangian models (Jacob et al. 1999), where Eulerian CTMs include weather research and forecasting model with chemistry (WRF-Chem), community multi-scale air quality (CMAQ), comprehensive air quality model with extensions (CAMx), global three-dimensional chemical transport model (GEOS-Chem) and model for ozone and related chemical tracers (MOZART).

CTMs have been applied in a variety of capacities, such as various atmospheric chemistry and air-pollution issues, all over the world, including sensitivity analysis for ozone (Simon et al., 2013; Sandu and Zhang. 2008), air quality forecasting for atmospheric pollutants (Odman et al., 2007), atmospheric chemistry and model evaluation research (Brune et al., 2016; Travis et al., 2016), aerosol-radiation-cloud feedbacks on meteorology and air quality (B. Zhang et al., 2015; Qiu et al., 2017); acid deposition, visibility, and haze pollution issues (Han et al., 2014; Fan et al., 2015); the source sector contribution, long-range transport for ozone and aerosol concentrations (K. Li et al., 2016; Zhu et al., 2017) and exposure studies (Bravo et al., 2012; Di et al., 2016). Although CTMs have been applied more frequently in the past decades to solve scientific problems related to air quality in China, the simulation results obtained vary due to the different input data of CTMs, model configurations, and the mechanisms of the models themselves. To determine the CTM model performance for BC/OC simulations in the Chinese context and in studying climate change, methods to objectively and accurately assess model performance are very important in air quality management applications.

1.3.2 Models and evaluation methods

- Global CTM simulations

The OsloCTM3 is an offline global 3-dimensional chemistry-transport model driven by 3-hourly meteorological forecast data from the European Center for Medium-Range Weather Forecast (ECMWF) OpenIFS model (Lund et al. 2018a; Søvde et al., 2012). The model is run in 2.25°x2.25° horizontal resolution, with 60 levels in the vertical, from the surface to 0.1 hPa. The model treats full tropospheric chemistry, as well as all the main climate-relevant aerosol species (BC/OC, sulfate, secondary organic aerosol, nitrate, dust and sea salt). Aerosols are transported in the atmosphere by advection and convection and are deposited through dry and wet scavenging. Detailed descriptions about the individual parameterizations of aerosols can be found in Lund et al. (2018a). I The OsloCTM3 (and its predecessor version OsloCTM2) is well documented and has been used in a large number of studies of global and regional anthropogenic emissions, as well as in detailed process studies. It is also one of the participating models in the multi-model experiments AeroCom and HTAP2.

Recently, Lund et al. (2018a) performed a comprehensive evaluation of the model performance in terms of present-day aerosols distribution using the most recent published global emission inventory, the Community Emission Data System (CEDS) (Hosely et al. 2018). Global models typically have trouble representing the vertical distribution of BC aerosol compared to observations (Samset et al. 2014). Work to constrain the treatment of BC in the model (Lund et al. 2017; 2018b) has resulted in significant improvements in many regions. Overall, the OsloCTM3 also performs well for surface concentrations and total aerosol optical depth. However, aerosol concentrations are underestimated in broader East Asian region. While using the CEDS emissions give better agreement with BC surface measurements than using older emissions data, the results indicate significant remaining uncertainties in also the recent global inventories. It is also a known issue that some global inventories do not fully capture the recent observed declining trends in SO_2 emissions in China. The evaluation of the model over China is also complicated by lack of available data. T

Through the ChiNorBC project, we will perform simulations with updated present-day emissions, comparing the results to those using recent global inventories, and perform an updated model evaluation with additional observational data provided by the CRAES collaborators. In addition to BC/OC, emissions of SO_2, NOx, CO, VOCs, and NH_3 are needed to run the model and will be updated with results from this project where available. Simulations will also be performed with new scenarios for 2035 developed as part of the project, keeping emissions from the rest of the world fixed at current levels to quantify the effect of projected emission changes in China on global and regional atmospheric composition and the associated radiative forcing.

- Regional CTM simulations

The Reginal CTM used in this project is the CMAQ model system (version 5.0.2, https://www.cmascenter.org/index.cfm), it is a core part of the third generation of the Air Quality Forecasting and Assessment System (Models-3) developed by the U.S. Environmental Protection

Agency, which comprehensively considers pollution issues such as particulate matter, photochemical oxidation, and acid deposition (Zhang, 2004; Zhang et al., 2005). CMAQ model adopts the concept of "one atmosphere", which includes complex gas-liquid-aerosol chemistry and simple non-homogeneous chemical processes, and has good simulation capability for atmospheric secondary pollutants (O_3, $PM_{2.5}$, and PM_{10}). Such model systems can be used for a variety of purposes such as simulation, assessment, and decision studies of air quality at multiple scales and with multiple pollutants. In this study, CMAQ is configured with the chemical mechanism of the Regional Acid Deposition Model version 2 (RADM2). The aerosol chemical species considered within CMAQ (Binkowski et al., 2003) include fine sulfate, nitrate, ammonium, biogenic organic carbon, elemental carbon, and other unspecified material of anthropogenic origin, etc.

Anthropogenic emission inventories used for modelling in this research are generally large-scale national (MEIC) and even continental (TRAC-P, INTEX-B, REAS, MIX) inventories (Zhang et al., 2009; Streets et al., 2003; Ohara et al., 2007; Li et al. 2017, etc.). Kanaya et al. (2019) simulated surface BC concentrations for China from 2009 to 2019 with different bottom-up emission inventories (e.g., MEIC1.3, ECLIPSE v5a, and v6b, REAS updated and CEDS) and compared decadal trend with observations, it was found that the decadal trend in the modelled BC concentrations for most emission inventory simulations, except CEDS, are generally in good agreement with the observed ones. Simulation results differ slightly between different emission inventories.

Although significant advantages can be found in CTMs, how to accurately simulate the concentrations of BC/OC is still a challenge, with the problems of inaccurate emission inventories and other imperfect physical and chemical parameterizations (Carmichael et al., 2008, Chen et al., 2019). To obtain a reliable BC/OC concentrations along with model simulations, we need more accurate bottom-up emission inventories.

- Radiative transfer calculation

We will calculate the instantaneous top-of-the atmosphere radiative forcing due to aerosol-radiation interactions using the Oslo RTM, which is an offline radiative transfer model. The same model has been used in earlier studies of RFari (Myhre et al., 2017; Myhre et al., 2013b) with recent updates to aerosol optical properties (Lund et al., 2018). The radiative forcing of aerosol-cloud interactions (RFaci) (earlier denoted the cloud albedo effect or Twomey effect) will be calculated using the same radiative transfer model. To account for the change in cloud droplet concentration resulting from anthropogenic aerosols, which alter the cloud effective radius and thus the optical properties of the clouds, the approach from Quaas et al. (2006) is used. This method has also been applied in the above-mentioned earlier studies. Opportunities to estimate the RF using the high-resolution CMAQ output will be explored. The radiative forcing due to deposition of BC on snow and ice is not calculated for this project.

- GWP and GTP assessment

Quantifying the impact of individual regions, sectors, or other emission sources on surface temperature is challenging using a coupled climate model due to the small signal of climate response relative to the

natural variability of the system. Instead, more simplified emulators are commonly used. One approach is to use so-called emission metrics. Emission metrics were developed to allow a simple comparison of the impacts of emissions of different species on a common scale and provide a basis for multi-component climate policies. Emission metrics can be used for simplified calculations of the global RF and temperature response over time following global, sectoral, or regional emissions. For the short-lived climate forcers, the RF and global temperature can be dependent on where the emission occurs. This can, at least to some extent, be accounted for by calculating region-specific emission metrics. This has been done in several previous studies, including recently by Lund et al. 2018. Using the output form simulations with OsloCTM3, we will in the ChiNorBC project produce updated region-specific values of the two most commonly used emission metrics, the Absolute Global Warming Potential (AGWP) (IPCC 1990) and the Absolute Global Temperature change Potential (AGTP) (Shine et al. 2005), using them to assess the impacts of present and future emissions in China on global climate over different time horizons. The method from Lund et al. (2020) will be used.

The AGWP and AGTP are both measures of the global mean impact and do not allow for an assessment of the local climate impacts. However, by calculation of region-specific metric values, we account for the fact that emissions in China can have a different effect on global climate than the same emission magnitude in another region. By combining the AGTP with the emission scenarios developed in the project, we will provide a first-order estimated of the global temperature response to present-day and changing Chinese emissions. The emission metrics can also be used more broadly to assess the climate implication of different policy packages for emission reduction in the near- and long-term, including potential trade-offs and synergies between e.g. long-lived greenhouse gases and short-lived climate forcers. We note that in addition to being a simplified climate impact assessment tool, there are important limitations, or caveats, when using emission metrics for comparing short-lived species, like aerosols, with the long-lived greenhouse gases on a common CO_2-equivalent scale due to their very different temporal behaviour. Ultimately, it is a user-choice which metric and time horizon to use, but these issues can be highlighted by by evaluating impacts at different time horizons and with different metrics.

1.3.3 Model evaluation and the common problems

Different CTMs need different validation parameters to evaluate their performance in different ways. Therefore, there is no general measurement standard suitable for all conditions (Chang and Hanna, 2004). Some of the most common metrics, such as Root mean square error (RMSE), the coefficient of determination(R^2), the correlation coefficient(R), the mean bias (MB), mean error (ME), normalized mean bias (NMB), normalized mean error (NME), etc. were discussed inYu et al., 2006; US EPA, 2007 and Wang et al., 2010, and some other more uncommon statistical metrics for different CTMs in recent studies (Appel et al., 2007; Appel et al., 2008;).The evaluation metrics are so varied that it is difficult to judge the overall performance of the model (Karroum et al., 2020). Before we decide on the evaluation metrics to be used in the ChiNorBC project, we make clear that the objective of our research is primarily to assess the performance of the model and compare the simulated atmospheric levels of air pollutants

from CTMs using current and update emission inventories against observations under the base-year within specific relatively clean areas and severely polluted areas. Our final selection of metrics includes NMB, NME and Pearson's r.

Relative measurement is particularly useful when comparing model performance between different CTMs (US EPA, 2007). The common metrics of relative measurement are normalization or fractional form, i.e., NMB and NME, meanwhile, MFB and MFE are frequently used in model performance evaluation (Zhang et al., 2014, Zhang and Ying, 2010). Depending on the variance of the frequency distribution of error magnitudes, we also can combine both RMSE and MAE to determine the model performance (Chai et al., 2014). Pearson's correlation coefficient, which passed a significance test (p-value), was used to examine the strength and direction of the linear relationship between the two continuous variables, the model and the monitoring data (Shimadera et al., 2016, Syrakov et al., 2016). Our performance goals and criteria for NMB, NME, and R refer to Yamaji et al. (2020) and Emery et al. (2017), while the performance goals and criteria for MFB and MFE refer to Boylan and Russel. (2006) and US EPA. (2007). RMSE and MAE are negatively-oriented scores, which means lower values are better.

In summary, there are some degrees of uncertainty in model simulations. In terms of $PM_{2.5}$ simulation, regional CTMs generally captured $PM_{2.5}$ magnitude in most cities and the trend of variation in daily average over Northern China (Gao et al., 2016, Chen et al., 2017, Liu et al., 2020). However, due to the complexity of each meteorology (Wang et al., 2010, Chen et al., 2020) and chemistry mechanism (Liu et al., 2019, An et al., 2019) involved and the uncertainty in emission inventories (Hoesly et al., 2017), accurately predicting $PM_{2.5}$ remains a challenge. For instance, the diurnal variation of $PM_{2.5}$ was not reproduced well (Petersen et al., 2019). And, predicting the $PM_{2.5}$ level in severe haze events remains a challenge (Li et al.2019; Zhang et al. 2012; Zhang et al. 2017), as meteorology simulation in calm weather was still a problem that needs to be solved (Hu et al., 2016, Bartholdy, 2000). Choi et al. (2019) compared surface $PM_{2.5}$ chemical components simulated by the WRF-Chem and CMAQ, the results show that the simulation results of different models using similar emission inventory had significant inconsistencies.

In terms of $PM_{2.5}$ components, although $PM_{2.5}$ concentration was well reproduced, each $PM_{2.5}$ species was not so good when compared with measurement (Qiao et al., 2019, Zhao et al., 2016). For instance, some secondary inorganic components were significantly underestimated, especially in severe pollution cases in winter (Hayami et al., 2008, Chen et al., 2016, Wang et al., 2014), when sulfate and nitrate experienced an abrupt increase in several hours (Li et al., 2017, Xu et al., 2019). Some new theories or reaction pathways (Cheng et al., 2017, Wang et al., 2018, He et al., 2014) were proposed to fill the gap and related research still went on. Secondary organic aerosols simulation was another problem (Hu et al., 2016, Huang et al., 2015).

Another issue was that air quality simulations require high-performance computational resources and modeling expertise (El-Harbawi, 2013). Improvements in computational efficiency have received increasing attention from researchers in the field of atmospheric pollution modeling in recent years.

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