Response to Referee #1:

This manuscript describes a comparison of two bottom-up SO2 and NOx inventories over China. The authors describe some of the input data, and explore the reasons for discrepancies between the two inventories. Satellite observations of NO2 are used with the GEOS-Chem model to produce top-down NOx emissions that are also used to evaluate the bottom-up inventories. The authors find that while differences in total emissions of SO2 and NOx from the bottom-up inventories are small, discrepancies at the sector level and provincial level are large. Compared to the top-down emissions, both bottom-up inventories are found to have negative biases, although uncertainty in the top-down approach cannot be ignored.

Response: We thank the constructive comments from reviewer #1 to improve our manuscript. Detailed responses to each comments are provided below.

General comment:

This study is written clearly for the most part, and brings attention to specific details/uncertainties about bottom-up inventories that should be considered when used in chemical transport model simulations. In general, the methods are technically sound and the conclusions are supported by the results. However, as a reader I was left with a larger question: What is the take-home message of this article? What is its importance to the atmospheric chemistry community? The authors do a sound job of pointing out differences between two (seemingly arbitrary) bottom-up inventories, but besides the obvious conclusion that some inventories will be different than others as a result of different methods/datasets, I'm not sure of the relevance here. The manuscript is quite technical, and in my opinion, misses the mark in terms of scientific significance. I encourage the authors to consider how their results and conclusions have larger impact. As written, it's not clear what substantial new concepts or methods have been advanced.

Response: Thanks for the reviewer's suggestions.

As for the aim of this work, as illustrated in the text, we want to better understand the reasons of differences between inventories for the currently widely used emission inventories (especially the two were picked because we have good access of the full inventory model, data and results), evaluate their effects on model simulation and their accuracies from the remote sensing perspective. The reasons of selecting ECLIPSE and MIX are detailed illustrated in the response to specific comments below. Although inventory comparisons have been conducted by several studies, there are still gasps to apply these results to improve the inventory accuracy. The comparisons cannot point to the systematic uncertainty of source, parameters, assumptions in inventories because of the lacking of the detailed technology-based inventory model. Initiated by this, we performed the comparisons and analyses of the two inventory model on parameter level to give more explicit indications for inventory developers. Our study should be important for inventory developers and modelers for understanding the potential uncertainties in the gridded emission inventory over China. For modelers, the comparisons and validations are important to understand the effect of emissions on model performance.

Another motivation of this work is to discuss the "fitness" of current developed inventories (specifically ECLIPSE and MIX) and modeling work done with them for policy relevant discussion. The inventories and the relevant modeling work is playing increasingly important role for policy discussion in Europe and most recently more and more in Asia at different scales. However, there is no systematic and officially approved methods and inventories but a variety of scientific products. While a lot of effort has been made to validate emission estimates with measurements, higher source and spatial resolution of inventories and projections will serve also discussion about the how to shape future policies to reduce the impact of air pollution. In fact, this discussion already takes place and science contributes to it. This work shows that our best inventories appear to be fit for evaluation of the policies at an aggregated or national level, more work is needed in specific areas in order to improve accuracy and robustness of outcomes at the finer spatial and also technology level.

The main take-home messages are illustrated in the "Concluding Remarks" section. Generally, this work shows that our best inventories are consistent on emission estimates at national level. The spatial distribution, emission amount and trend of current inventories are well captured by the satellite remote sensing. It appears that current inventories and relevant modeling work are fit for policy analyses and future projections at an aggregated level. More work is needed to improve the accuracy through verifying important while currently uncertain parameters on finer spatial units, including the real-world running efficiency and application rate of pollutant control facilities of FGD and LNB, diesel consumptions, vehicle fleet and emission factors for various vehicle types.

Specifically, the take-home messages are:

a) ECLIPSE and MIX estimates of SO_2 and NO_x are quite close in 2010, with 1% and 16% difference in China. The trends from 2005 to 2010 estimated by two inventories are both generally consistent with the OMI observations.

b) The emission differences on provincial and sector level are still high. On a sector level, 40% difference is found for power plants (higher in ECLIPSE), 24% for the industry sector (lower in ECLIPSE) for SO_2, and 15%~21% in power and transportation for NO_x (lower in ECLIPSE). Source classification, energy statistics, emission factors and assumption of technology penetrations are contributing parameters. Specifically, the FGD penetration rate and removal efficiencies, the LNB application rates and abatement efficiency in power plants, vehicle fleet and emission factors for various vehicle types in the transportation sector need further verification. Diesel consumptions are quite uncertain, which need more local surveys and validations for future inventory improvement.

c) The model case done with MIX show the best performance. Increasing the ECLIPSE emission estimates to MIX reduces the biases from -12.2% to -6.19% for NO_x. Negative biases in bottom-up gridded emission inventories are found compared to the top-down inversion.

We addressed the aim and importance of our work in the introduction section. We clarified the take-home message points in the concluding section, and revised the abstract to highlight the main findings.

<u>*Specific comments:*</u>

Abstract:

1) As written, the abstract seems to focus quite a bit on the methods, and very little on the results and relevance. I encourage the authors to consider editing their abstract to include the important results and conclusions.

Response: Thanks for the suggestions. We revised the abstract to include important results, relevance to the atmospheric community and main findings.

Methodology:

1) What is the reason for focusing on ECLIPSE and MIX? It comes across as an arbitrary choice of inventories. Are they the most recently developed? Are they the most popular in chemical transport models? Do they provide the most methodological de-tails? Why should the readers be interested in these two inventories specifically?

Response: We focused on the comparisons of ECLIPSE and MIX due to the following reasons:

a) Up to the time of manuscript preparation, ECLIPSE and MIX are the only publicly accessible gridded emissions dataset which include both SO_2 and NO_x covering China for the period of 2005 and 2010;

b) Both inventories have been widely applied in atmospheric modeling and policy discussions (e.g., Stohl et al., 2015; Duan et al., 2016; Galmarini et al., 2017; Rao et al., 2017);

c) The technology-based framework and compiling parameters by source categories are obtained for ECLIPSE and MIX through international collaboration, which is not accessible for other inventories over China. The methods and data were extensively described by a series of paper (Liu et al., 2015; Zheng et al., 2014; Klimont et al., 2017; Li et al., 2017), supporting us for explicit comparisons and analyses;

d) ECLIPSE (GAINS model) can be representative of the state-of-science global emission inventory covering China, and MIX (MEIC model) as the regional inventory compiled with advanced methods and local data. The methods, parameters and assumptions of GAINS and MEIC are always referred by inventory developers (e.g., Lu et al., 2010; Fu et al., 2013; Kurokawa et al., 2013; Zhao et al., 2013). The comparisons and validations are important to improve the accuracy of gridded emissions and model performance over China.

We clarified the reasons of comparing these two emission inventories in the revised manuscript.

2) The spatial proxies are mentioned very generally many times throughout the manuscript, but almost no detail is given the methods about the actual data used in each case. On Page 14 the authors state, "Proxies used .. are summarized in Section 2". But unless I missed it entirely, they are not summarized beyond some very general language. Further broad strokes are given on Page 14 ("for industry and residential sector, emissions are distributed mainly on population *data. Road networks and population are used as proxy for transportation emissions"), but I think at the very least, these details belong in the methods earlier on. I was frustrated by the number of times spatial proxy data are referred to with general language ("mainly"; "including"), but did not come away with a comprehensive understanding. Can the authors include a table that summarizes the source of all the spatial proxy data that is actually used in each inventory/sector? Or perhaps include maps of the different spatial data used in the Supplementary Information? A lot of attention is given to the spatial patterns, for them to be of such little importance in the methods.*

Response: Thanks for the comments on the spatial proxies. We add a table summarizing the spatial proxies by sub-sectors for ECLIPSE and MIX in the supplement to avoid repeated presentation with previous work (Klimont et al., 2017; Li et al., 2017), and enrich the elaborations in the main text for clarification:

"Emissions are distributed to grids at specific resolution (0.5°×0.5° for ECLIPSE, longitude×latitude) based on the percentages of spatial proxies located in grids by source category using GIS (Geographic Information System) techniques. Spatial proxies for both ECLIPSE and MIX are summarized in Table S1. For ECLIPSE, several layers were developed as spatial proxies in line with those used in the Representative Concentration Pathways (RCP) (Lamarque et al., 2010), i.e., locations of energy and manufacturing facilities, road networks, shipping routes, human and animal population density and agricultural land use. Spatial proxies were further developed within the Global Energy Assessment project (GEA project, Riahi et al., 2012), including improved population distribution, flaring in oil and gas production, smelters, and power plants for which provincial emission layers of MEIC (Multi-resolution Emission Inventory for China) were used.

For MIX, provincial emissions were firstly distributed to county, then further distributed to grids. The former process was based on statistics by county (i.e., GDP, IGDP, total population, urban population, rural population, agricultural activity, vehicle population at county level), and the latter was based on gridded maps as spatial proxies (i.e., population density map, road network). For power plants, locations were determined using Google Earth following the unit-based methodology."

Table S1. Spatial proxies used in the ECLIPSE and MIX emission inventories.

Sub-sectors	Spatial proxies	Data source
ECLIPSE		
Power plants	Plant locations	Google Earth (Liu et al., 2015)
Industrial combustion [a]	Total population, urban population, rural population	Lamarque et al., 2010; Riahi et al., 2012
Industrial processes [a]	Urban population, industrial plants	Lamarque et al., 2010; Riahi et al., 2012
Residential [a]	Total population, urban population, rural population	Lamarque et al., 2010; Riahi et al., 2012
On-road transportation [a]	Population, Road networks	Lamarque et al., 2010; Riahi et al., 2012
Off-road transportation [a]	Inland waterways, roads, railways, population, urban population	Lamarque et al., 2010; Riahi et al., 2012
Waste [a]	Population, urban and rural population	Lamarque et al., 2010; Riahi et al., 2012

Agriculture (fertilizer)	Cropland area	Potter et al., 2010
Agriculture (livestock)	Livestock map	FAO, 2007
MIX[b]		
Power	Plant locations	Google Earth (Liu et al., 2015)
Industrial heating	Industrial GDP[c], urban population[d]	NBS[e], LandScan[f], urban/rural extents[g]
Residential heating	Urban population	NBS, LandScan, urban/rural extents
industrial boiler	Industrial GDP[c], urban population[d]	NBS, LandScan, urban/rural extents
Residential combustion (fossil fuel)	Urban population	NBS, LandScan, urban/rural extents
Residential combustion (biofuel)	Rural population	NBS, LandScan, urban/rural extents
Iron and steel	Industrial GDP[c], urban population[d]	NBS, LandScan, urban/rural extents
Cement	Industrial GDP[c], urban population[d]	NBS, LandScan, urban/rural extents
Other industrial process	Industrial GDP[c], urban population[d]	NBS, LandScan, urban/rural extents
On-road vehicles	Vehicle population[c], road network[d]	China Digital Road-network Map (Zheng et al., 2014)
motorcycles	Vehicle population[c], road network[d]	China Digital Road-network Map (Zheng et al., 2014)
Off-road (agriculture machinery)	Machine power[c], rural population[d]	NBS, LandScan, urban/rural extents
Off-road (construction)	Total GDP[c] , urban population[d]	NBS, LandScan, urban/rural extents
off-road (others)	Total population	NBS, LandScan
Solvent use - industry	Industrial GDP[c], urban population[d]	NBS, LandScan, urban/rural extents
Solvent use - residential	Urban population	NBS, LandScan, urban/rural extents
Agriculture (fertilizer)	Fertilizer use[c], rural population[d]	NBS, LandScan, urban/rural extents
Agriculture (livestock)	Meat consumption[c] , rural population[d]	NBS, LandScan, urban/rural extents
Waste	Total population	NBS, LandScan

[a] Spatial proxies included were derived from the EDGAR emissions gridding manual, http://publications.jrc.ec.europa.eu/repository/bitstream/JRC78261/edgarv4_manual_i_gridding_pubsy_final.pdf

[b] derived from Li et al. (2017)

[c] Proxies used to distribute provincial emissions to county

[d] Proxies used to distribute county-level emissions to grids

[e] National Bureau of Statistics, http://www.stats.gov.cn/tjsj/

[f] LandScan Global Population database

[g] Urban / rural extents developed by Schneider et al. (2009)

3) The authors point out that OMI SO_2 observations have large uncertainties. Would the observations be at all valuable in a qualitative comparison of spatial emission patterns?

Response: Thanks for the suggestions. We add more illustrations of top-down validations for SO_2 emissions. Firstly, we compared the SO_2 concentrations between modeled results and OMI SO_2 columns. Although OMI data tend to overestimate the concentrations due to the overlap in signals of SO_2 and O_3 during retrieval, good correlations are found between models and satellite

(R=0.633-0.667, Slope=0.842-0.863, general consistent among sensitivity cases), confirming the high accuracies of the priori SO_2 spatial emission patterns.

Secondly, following the method of NO_2 top-down inversion, we developed the top-down emissions for SO_2. The spatial distributions, emission amount and trend of the priori emission inventory were further evaluated. Given the large uncertainties involved in the OMI SO_2 columns, the evaluated results were interpreted with caution. Compared to the top-down emission inventory, both ECLIPSE and MIX show relatively good correlations (R=0.722-0.896, Slope = 0.539-0.923). The national decreasing emission trend from 2005 to 2010 were captured by both bottom-up and top-down inventories.

Figures, tables and discussions of the top-down evaluations for SO_2 are added in the revised manuscript.

Results:

1) P 14 Line 2 mentions how the industrial and residential sectors show "clear administrative boundaries". But for someone who is not familiar with the administrative boundaries, this isn't obvious (Provincial? county?). Would it be useful to include some of the boundaries they are referring to?

Response: We add the provincial boundaries for all gridded emission maps to make it more clear.

2) P 14 Line 17 mentions how "other" proxies are population-based. Which proxies exactly are the authors referring to?

Response: "Other proxies" refer to the total population map (for some industrial sources), urban population map (for industrial heating, residential coal burning, etc.) and rural population map (for residential biofuel burning). We clarified the sentence in the revised manuscript.

3) P 14 Line 20 mentions the excellent correlations observed for all sectors, but then misses the most interesting question. What are the exact sources of the occasions when they are different? For example, residential NOx has a slope of 0.88, whereas the slopes for the other sectors are all very close to 1. What data has been used differently that causes this difference in the residential sector between the two?

Response: The excellent correlations for all sectors (with slope ≥ 0.87, R ≥ 0.94) are attributed to the overall similarity in spatial proxies (as summarized in the table of spatial proxies). The differences for specific sectors (e.g., residential with slope of 0.87) are slightly higher than others, mainly due to the different population dataset used for emission allocation of relevant sources in ECLIPSE and MIX. We clarified it in the revised manuscript.

4) P 15 Line 15 "In light of the bottom-up comparisons". Here, can the authors be specific about what issues they are referring to? Exactly what hypotheses are the sensitivity tests set up to test? This would help understand the importance and purpose of the sensitivity simulations.

Response: The bottom-up comparisons work done earlier show that differences exit for both the national emission estimates and gridded emission spatial patterns. In Sect. 3.2.1, the main purpose is to evaluate the effect of gridded emissions on model accuracy, and find out the effect of the national emission estimates and spatial distributions on the model simulated results, using satellite observations as a criterion. Therefore, we further set up four sensitivity tests, ECL-case0~case2 and MIX, where ECL-case0 and MIX are two basic inventory scenario, ECL-case1 replaced the total emissions of ECL-case0 with MIX's estimates, and ECL-case2 changed the spatial distributions of ECL-case0 to MIX. We clarified this in the revised manuscript.

5) P 18 Line 26: This is the first indication in the entire article that IGDP is used as a spatial proxy. This is a good example of why the discussion about spatial proxies became frustrating to me. Again, I encourage the authors to lay out or list the spatial proxies comprehensively in the methods. Perhaps these details are obvious to some, but they aren't obvious to me.

Response: We summarized the spatial proxies in Table S1 in the revised manuscript.

Figure 1: Might I suggest the authors include the totals for SO2 and NOx from each inventory in the figure (just as a number, somewhere in the panel)?

Response: Revised as recommended.

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