

Response to Referee #2:

General comments

The work of Li et al. deals with the comparison of the ECLIPSE and MIX emission inventories over China focusing on SO₂ and NO_x sector- and region- specific emissions. Bottom up emissions are then compared with top down estimates from OMI. The paper is overall well written and I recommend it for publication after developing the following points:

The authors should clarify the aim of their work, since comparing two emission inventories (even at sector level) and top down vs. bottom up estimates comparison are not new topics in literature. It is not completely clear the novelty of this work compared to literature studies dealing with top down and bottom up estimates such as Wang et al. 2011 and other works. The authors state that “To our knowledge, it’s the first emission inventory assessment work where parameter-level comparison and remote sensing evaluations are combined”, however, there are several literature works comparing top down and bottom up estimates, even over China (e.g. Wang et al., 2011; R. J. van der A, 2017 etc.). Therefore, the authors should clarify the relevance of their study compared to former works.

Response: We thank reviewer #2 for the careful reading and constructive comments, which are crucial to improve the manuscript.

As stated by the referee, several work conducted the emissions comparisons and top-down validations (such as Wang et al., 2011 from the same group). Extensive comparisons among emission inventories were important to illustrate the effect of emission inventory on the model simulation results and atmospheric component analyses (e.g., Ding et al., 2017; Saikawa et al., 2017). Although they provide important indications on the extent of discrepancies, there are still gaps for applying the comparison results to improve the inventory accuracy:

- (1) Comparisons have been conducted for the total anthropogenic sources, instead of by sectors/subsectors/sources. Inconsistency of source categories included in inventory models were not overviewed or analyzed;
- (2) Few studies go into the comparisons on specific parameter level because the technology-based framework for each inventory was not publicly available;
- (3) Top-down and bottom-up comparisons have not been comprehensively combined to infer the potential uncertain parameters for all key sectors.

Through international collaborations between IIASA and Tsinghua University, we compared the ECLIPSE and MIX emissions over China parameter by parameters at a detailed activity-source level. What we focused on in this manuscript is the bottom-up comparison detailed to specific parameter contributing to the differences between the two widely used gridded emission inventories (ECLIPSE and MIX), combined with top-down validations from the satellite observations.

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 impact of air pollution.

We clarified the aim of this work in the revised manuscript.

The responses to the specific comments are provided below.

Specific comments

- In the introduction the authors list several emission inventories covering China, however, several other emission inventories have been developed for that region (e.g. Liu et al., 2015; the EDGAR database, etc.). The authors should explain why they provide only that list of references.

Response: In the revised manuscript, we complemented the reference list by including all emission inventories that developed gridded emissions of SO₂ or NO_x from anthropogenic sources over China as follows:

To support chemical transport modeling and provide scientific basis for policy-making, several emission inventories covering China have been developed (Streets et al., 2003; Ohara et al., 2007; Zhang et al., 2007, 2009; Lu et al., 2010, 2011; Kurokawa et al., 2013; Klimont et al., 2009, 2013; Wang et al., 2014; Li et al., 2017; EDGAR v4.2 (available at <http://edgar.jrc.ec.europa.eu>)).

- 2.1 The ECLIPSE and MIX emission inventory: This paragraph describes the two inventories later compared in the paper. To facilitate such comparison, it would be good to have a summary table listing for the two inventories the data sources for each sector (activity data and emission factors), the temporal and spatial resolution, the reference years, compounds, etc. The authors should highlight the independence of the two inventories in terms of statistics, EFs, proxies, etc. before doing the comparison.

Response: Thanks for the suggestions. We add a table listing the data sources of activity data, emission factors, and key features of ECLIPSE and MIX, to facilitate the comparison. We insert a paragraph before the detailed comparison by sectors in Sect. 3.1 as below:

“As shown in Table 1, the activity rates were assigned independently by two inventories. As a global emission inventory, ECLIPSE mainly relies on international statistics of IEA. Differently, MIX obtains the official statistics of energy consumption and industrial output from NBS (National Bureau of Statistics) or MEP (Ministry of Environmental Protection) of China. We can expect high independency for the determination of emission factors between ECLIPSE (GAINS model) and MIX (MEIC model). As two independently developed inventory model, the source classification, technology penetration, removal efficiencies in GAINS and MEIC are expected to be different although they both refer to up-to-date measurements and peer-reviewed

data. Different methods were developed in two inventory models for specific sectors, including power plants, transportation, and agriculture. For power plants, the spatial proxies were essentially consistent between ECLIPSE and MIX. For other sectors, emissions were gridded independently by two emission inventories (see Table S1).”

Table 1. Key features of ECLIPSE v5a and MIX emission inventories.

Item	ECLIPSE v5a	MIX
Year	1990-2010 at a 5-year interval	2005 ^a , 2008, 2010
Domain	Global	Asia
Spatial resolution	0.5°×0.5°	0.25°×0.25°
Temporal resolution	Monthly	Monthly
Activities included for each sector		
Energy / Power	Power plants (including CHP), energy production/conversion (including district heating plants), fossil fuel distribution	Power plants (including CHP)
Industry	Industrial combustion and processes	Industrial combustion (including industrial heating plants) and industrial processes
Residential	Residential combustion sources	Residential combustion sources (including residential heating plants)
Transportation	On-road and off-road transport sources ^b	On-road and off-road transport sources ^b
Agriculture	Livestock and fertilization	Livestock and fertilization
Data sources of activity rates		
Power	International Energy Agency (IEA)	CPED (Liu et al., 2015)
Industry	International Energy Agency (IEA)	Provincial industrial economy statistics (NBS)
Residential	International Energy Agency (IEA)	Provincial energy statistics (NBS)
Transportation	International Energy Agency (IEA)	Provincial energy statistics (NBS); Zheng et al. (2014)
Agriculture	UN Food and Agriculture Organization ^c	Provincial statistics (NBS, Huang et al., 2012)
Emission factors and technology	GAINS model (Klimont et al., 2017)	MEIC model ^d , Process-based model for NH ₃ (Huang et al., 2012)
Data Access	http://www.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv5a.html	http://www.meicmodel.org/dataset-mix

^a developed following the same methodology

^a International air and international shipping are not included.

^c FAO, <http://www.fao.org/faostat/en/#home>.

^d Zhang et al., 2009; Lei et al., 2011; Zheng et al., 2014; Liu et al., 2015

- 2.3 Top-down emission inventory: The authors should explain why the methodology presented is applied only to NO_x and not to SO₂ columns. It would be interesting to see the same procedure applied also to SO₂ since the paper focuses on both compounds.

Response: Thanks for the suggestions. We add more illustrations of top-down validations for SO₂ emissions. Firstly, we compared the SO₂ concentrations between modeled results and OMI SO₂ columns. Although OMI data tend to overestimate the concentrations due to the overlap in signals of SO₂ and O₃ during retrieval, good correlations are found between models and satellite (R=0.633-

0.667, Slope=0.842-0.863, general consistent by sensitivity cases), confirming the high accuracies of the priori SO₂ spatial emission patterns.

Secondly, following the method of NO₂ top-down inversion, we developed the top-down emissions for SO₂. The spatial distributions, emission amount and trend of the priori emission inventory were further evaluated. Given the large uncertainties involved in the OMI SO₂ 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 decrease trend from 2005 to 2010 were captured by both bottom-up and top-down inventories.

Figures, tables and discussions of top-down evaluations for SO₂ were added in the revised manuscript.

- Page 6, line 4: please clarify how the sectors “power”, “industry”, “residential” and “transportation” are defined for each inventory. As described at lines 7-10, sectors are different for the two inventories. Please clarify how emissions from heating plants are re-distributed (line 9) in MIX to match the ECLIPSE sectors.

Response: We clarified the definition of each sector in Table 1 (see above). We aggregated the heating emissions from the “industry” and “residential” sectors in MIX to the “power” sector, to match the defined ECLIPSE sectors. We clarified the procedure in the revised manuscript.

-page 10, line 8: “emission factors on mass base are converted to energy base with heating value of 43.1 MJ/kg”. Did the authors use the same heating value both for gasoline and diesel?

Response: Yes. These values are extracted from the GAINS model, comparable to the reported heating values of 44-46 MJ/kg for gasoline and 45 MJ/kg for diesel fuel by the World Nuclear Association (<http://www.world-nuclear.org/information-library/facts-and-figures/heat-values-of-various-fuels.aspx>). We revised the note for clarification.

-page 10, line 10: although only 3% difference is found in total gasoline consumption, big differences in gasoline use by vehicle are observed for the two inventories.

Response: We add more illustrations by vehicle types as follows:

“The consistency in the total gasoline consumption between ECLIPSE and MIX is attributed to the consistency in statistics. As shown in Table 3, the gasoline consumptions by vehicle types show large differences between ECLIPSE and MIX, indicating different vehicle fleet assumption in two inventory models. Detailed data is not known and each of the inventories (or research groups developing them) relied on own assumptions about fuel consumption per vehicle, mileage travelled, and combined those with the available data on the number of vehicles, their sales and retirement rate. Owing to the above reasons, the results can differ significantly. Light duty vehicles are the largest gasoline consumer (> 77%) in both inventories, with 18% higher gasoline consumption estimated in ECLIPSE than those of MIX in 2010.

Accordingly, ECLIPSE estimates less gasoline consumed in high duty vehicles (74%) and motorcycles (32%) than MIX. These differences reduced from 2005 to 2010.”

-page 10, line 11: huge differences are observed not only for light duty vehicles but also for HDV-G and MC.

Response: We focused on the comparison for light duty vehicles because they dominate the total emissions of gasoline-fueled vehicles, with emission contributions of 63% - 91% (estimated by two inventories) in 2010. For HDV-G and MC, we extended the discussion in the revised manuscript as below:

“Emission estimates of HDV-G (high duty vehicles) and MC (motorcycles) also show large differences between two inventories. For HDV-G, ECLIPSE estimates lower emissions than MIX (66% in 2010), as a result of less fuel consumption while higher emission factors in ECLIPSE. For MC, emissions of ECLIPSE are 64% lower than MIX, contributed by both fuel consumption and emission factors, as shown in Table 3.”

-3.1.3 Gridded emissions: Figure 3b shows the difference of the ECLIPSE-MIX gridded emissions. Did the authors compare the proxy data used by the two inventories to grid the emissions? A mismatch in the location of large point sources as well as the application of weighting factors to redistribute the emissions could strongly affect this type of calculation. Please develop this topic.

Response: The differences of gridded emissions illustrated in Fig. 3 are attributed to the discrepancies in emission estimates nationwide and by provinces (Sect. 3.1.1, Sect. 3.1.2), and also method and data in emission spatial allocations (see Sect. 2). We add a table (Table S1) summarizing the spatial proxy data used in both inventories in the revised manuscript. For power plants which were treated as point sources, emissions are gridded based on the locations verified by Google Earth (Liu et al., 2015), consistent between ECLIPSE and MIX. For other sectors, ECLIPSE gridded the provincial emissions according to the source-specific layers, and MEIC used two-step allocation method (province to county, county to grid). The data sources of spatial proxies also differ between two inventories. In general, as illustrated in Figure 4, the spatial distributions of emissions within provinces show quite good consistency, even by sectors. We revised Sect. 3.1.3 to clarify it.

- page 15, lines 7-9: “The different trends of transportation emissions are attributed to the different assumptions on legislation effect on pollution control in two inventory systems”. The authors should demonstrate the aforementioned statement.

Response: We complemented the discussion as follows:

“For Beijing, the differences of emissions trend in the transportation sector are mainly caused by diesel vehicles. In ECLIPSE, 47% increases are estimated for diesel fueled vehicles, compared to 28% emission decreases in MIX. Fuel consumptions show large discrepancies in

trend from 2005 to 2010, where +54% compared to -20% for high duty vehicles, and +45% compared to +3% for light duty vehicles, as estimated in ECLIPSE and MIX respectively. The emission factors of light duty vehicles increase by 5% in ECLIPSE, while decrease by 34% in MIX, attributed to the different assumptions on emission control effects. As a pioneer in pollution control of China, Beijing carried out Euro III standard in 2005 and Euro IV standard in 2008 for light duty vehicles. For Beijing, the Euro IV penetrations in 2010 are assumed around 12% in ECLIPSE, while more than 60% in MIX, which might be too optimistic and should be verified with local surveys.

For the PRD region, gasoline and high duty diesel vehicles contribute to the differences of emission trend. 22% emission growth for LDB-G (light duty gasoline buses) is estimated in ECLIPSE, compared to 12% emission reduction in MIX. For high duty diesel vehicles, trend of fuel consumption (+55% in ECLIPSE, compared to -11% in MIX) and technology distribution (21% of Euro III in 2010 for ECLIPSE, compared to >50% in MIX) are the main contributors to the different emissions trend. In summary, survey data are urgently needed to validate the fuel consumptions, effect of legislation effect and trend for diesel vehicles in pioneering regions such as Beijing and PRD.”

- page 18, line 17: “It can be concluded that ECLIPSE and MIX are consistent with the top-down estimates over China.” The authors should discuss why it is useful to compare bottom up and top down estimates. In their work they discuss the differences (sometimes not negligible) between two bottom up inventories over China and then through the comparison with top down estimates they find that the two inventories are consistent with these independent estimates. How is that possible? How can top down estimates help in constraining the bottom up emission inventories? How can this work reduce the uncertainty of emission inventories? Can the authors explain if the uncertainty of bottom up and top down estimates are larger, smaller or within the range of model uncertainties?

Response: Top-down emission estimates can provide independent third-party constraints on the bottom-up emissions on the emissions amount, spatial distribution and trend (Martin et al., 2003; Lamsal et al., 2011; Lee et al., 2011; Liu et al., 2016; Cooper et al., 2017). In this work, we compared the detailed parameters for bottom-up emission inventory development in the previous sections, and found general consistent emission estimates for the whole China (differ within 16%) and gridded emissions (slope ≥ 0.8 , $R \geq 0.9$), while large variations for provincial emissions and specific sectors. These detailed comparisons are important for guiding the emission inventory community to put more efforts on parameters that are quite uncertain in current inventory system, including the real-world running status of pollutant abatement facilities, statistics of diesel consumption, vehicle fleet, and emission factors of industrial boilers.

Gridded emissions are the direct inputs to atmospheric models. Through comparing the gridded emissions from bottom-up and top-down emissions derived from satellite observations, the model-ready emissions input can be overall constrained, given the comparable uncertainties in two inventories (as illustrated in Sect. 3.3.2). Compared to the top-down emissions, both ECLIPSE and MIX show high correlations ($R \geq 0.87$), supporting the conclusion that both

inventories are generally consistent with the top-down estimates in spatial emission patterns. We also found moderate negative biases (-21% - 39%) for the bottom-up inventories, indicating that ECLIPSE and MIX may underestimate NO_x emissions in 2010, which indicates the direction of verifying the uncertain parameters. Furthermore, the emission trends were validated based on the top-down retrievals. The general consistent emission trend proves the “fitness” of our inventories for policy relevant discussion and projections.

- page 19, lines 1-2: *“Through sensitivity test analyses, treating sources as point sources can significantly reduce the uncertainties in emission gridding process”. The authors should better explain how it is possible to reduce the uncertainties in emission gridding process through sensitivity tests. Sensitivity tests can help understanding the uncertainties due to the gridding procedure using e.g. different proxy data, but not necessarily to reduce the corresponding uncertainty.*

Response: We refer to the conclusions of Geng et al. (2017) here. In the work of Geng et al. (2017), a set of sensitivity test was conducted to evaluate the impact of spatial proxies on model performance. It’s proved that determining the exact locations of large emission sources will significantly strengthen the correlation with modeled and satellite retrieved NO₂ columns (Geng et al., 2017). To avoid misunderstanding, we revised the sentence to “Through sensitivity test analyses, it’s concluded that treating sources as point sources can significantly reduce the uncertainties in emission gridding process (Geng et al., 2017)”.

- *It would be interesting to see Figure S1 also in absolute terms. The authors should also better explain the different sectorial share for the various provinces. Why Tibet has only SO2 emissions from the transportation sector in the MIX inventory, while they are negligible for ECLIPSE? Large sector specific differences are also observed for NOx. Please discuss in a more comprehensive way the differences in sector specific emissions at province level.*

Response: The sectorial distributions of provincial emissions are overall consistent (within 30% difference on sector level) given the differences in source classification between two inventory systems. For Tibet, the emissions can be neglected (e.g., for NO_x, around 30 Gg/year, 0.1% of the national total) and unreliable for both emission inventories because the real-world energy consumption statistics are quite uncertain. We add the emissions by provinces in Figure S1 to give a more comprehensive reference to readers. The absolute values of sectorial emission share for each province are labeled in Figure S1.

Technical corrections

- *Figure 5a shows empty maps for the SO2 trend from transportation sector of both inventories. Please check them.*

Response: The SO₂ emissions from the transportation sector are ignorable compared to other sectors under the same color scale. We revised Figure 5a with different color scale for the

transportation sector.

-Figure S3: Please change the Figure caption with “NO_x emission changes. . .” instead of “Emission changes. . .”.

Response: Revised as suggested.

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