Reply to Comments from Referee #2

1. Section 2.2.4 seems a bit out of place. You are going through the figure and the APCD's, and this section focuses on industrial boilers, where the sections before and the section after is for CF power plants. Consider making this one last of Section 2.2.X.

Response:

This sub-section has been moved to the last of this section as suggested.

2. Section 2.2.5 isn't in the figure. Perhaps it is not appropriately put there? I was just expecting it to be there.

Response:

Section 2.2.5 is indeed in the figure. We used "FGD" which is not consistent with the text. We have changed it to "WFGD". Please see the modified Fig. 1.

3. Page 32899, line 13; bonds instead of bounds.

Response:

Modification has been made.

4. Page 32900, DCA isn't defined or in the figure.

Response:

The DCA towers mean dehydration tower, conversion tower and absorption tower, all of which are units in the acid plant. Therefore, we have changed it to "acid plant" in the revised manuscript.

5. Page 32901, Line 8. "FGS uses…". Again, I am no engineer, but I thought these used calcium carbonate and the like to scrub sulfur out of these streams in wet scrubbers.

Response:

Unlike WS or WFGD for SO_2 control in coal combustion, FGS in non-ferrous metal smelters uses diluted sulfuric acid to capture SO_2 and SO_3. The yield from FGS is waste acid, which will be treated to acid sludge. This point has been added to this part. Please see Lines 322-325 on Page 12 in the revised manuscript.

6. Page 32903, line 3, reference Wu et al., 2006 isn't in the references.

Response:

This reference has been added to the reference list.

7. Page 32910, line 11-12, sentence ", and found that the mercury" is unclear to me what the release rate means. Do you mean that the emission is 70-90% and the remainder is in the ash? Just unclear.

Response:

We have changed the expression of this sentence. Please see Lines 587-588 on Page 21 in the revised manuscript:

"… and found that the mercury emission rate during biomass burning is 78–99% while the remainder stays in the residue"

8. Section 4 is very difficult to follow. You should definitely point people to the figure (#4) at the beginning of the discussion for better understanding by the reader. I agree to understand the system, much discussion is needed. However, an alternative is to shorten the discussion, making the broad points that: 1. Hg in the raw materials comes out in the roasting process, 2. by using the flue gas to preheat the raw materials and coal, the Hg is recycled and is enriched along the way, and 3. that the operational modes vary dramatically and the emissions and ratios will be highly different and have to be accounted for in emission databases. I agree it is complicated and so variable that the process takes a bit of discussion to explain it.

Response:

We thank the reviewer for the valuable suggestion. We have revised Sections 4.1,

4.2 and 4.3 in our manuscript according to this comment. The introduction of cement production process and the mercury behavior have been simplified. The three points advised by the reviewer have been emphasized and highlighted in the manuscript. Figure 4 has also been introduced in the beginning of our discussion. Please see the updated Section 4 in the revised manuscript:

"Precalciner process is usually composed of the raw mill system, the coal mill system, the kiln system and the kiln head system. Raw materials are ground and homogenized in the raw mill system. The fuel, usually coal, is prepared in the coal mill system including coal mill and FF. The kiln system for the production of cement clinker includes the preheater, the precalciner and the rotary kiln. The prepared raw materials, namely raw meal, enter the kiln system from one end of rotary kiln (kiln tail), and the coal powder is brought into the kiln system by air from the other kiln end (kiln head). The solid materials flow in opposite direction with the flue gas. The flue gas from kiln tail is used to preheat raw materials in raw mill and coal in coal mill. The flue gas from kiln head is de-dusted and then emitted into the atmosphere. All the dust collected by dust collector is recycled to kiln system."

"The mercury behavior in cement production process is summarized as three stages: vaporization, adsorption and recycling (Sikkema et al., 2011) (see Fig. 4). At the vaporization stage, mercury in raw materials and fuel is vaporized into flue gas in the kiln system. Then part of the mercury in flue gas is captured by raw materials in the raw mill and coal in the coal mill when the flue gas is used to preheat solid materials, and part of the mercury in flue gas is also collected in the dust collector with dust. This process is called the adsorption stage. Finally, the mercury is cycled back into the kiln system with raw materials, coal and collected dust, which is the recycling stage."

The first two sentences of Section 4.3 have been deleted.

9. Further, in figure 4, I would suggest: a. Add a "clinker out" yellow arrow b. Add a box on the green arrow of mercury from the coal mill and collector that shows it is preheated as described in the text.

Response:

Figure 4 has been revised as suggested by the reviewer.

10. I would restate in Section 4 at the end, that these process are so highly variable that the emissions inventories are likely to be wrong, or something to that effect.

Response:

We have added some discussion about the point commented by the reviewer. Please see Lines 501-506 on Page 18 in the revised manuscript:

"As discussed above, the mercury speciation and emission are largely variable because of the complicated mercury cycling and operational modes of the cement clinker production process. Previous estimates of mercury emission and speciation from cement clinker production have large uncertainties. More studies including field tests should be conducted to further understand the mechanism of mercury speciation and transformation in cement production."

11. Section 5; many readers will not know what sintering is. I would use a short parenthetical to define.

Response:

We have explained the sintering process clearer as suggested by the reviewer:

"…roasted in the sintering machine, namely the sintering process."

12. References: a. I did not find Eriksen used. b. Lopez-Anton is referenced as 2007 in the text; check this one c. Takahashi is referenced as 2010 in the paper; check this one.

Response:

The reference of Eriksen et al. has been removed. Both López-Antón et al. (2007) and López-Antón et al. (2011) have been used in the paper. López-Antón et al. (2007) has been added to the reference list. The year of Takahashi et al. should be 2012, which has been modified in the text.

13. Table 1: I would add to the title Table 1. "Average (Range)" Speciation profile of mercury emissions from coal combustion "By Boiler Type and Control Technology (%)".

Response:

Modification has been made accordingly.

14. Table 2: same comment.

Response:

Modification has been made accordingly.

15. Table 3: same comment.

Response:

Modification has been made accordingly.

16. Table 4: I would add lines between the countries or regions so that they are more easy to compare. Hard with no dividing lines.

Response:

The original table has lines in between. It is probably the requirement of the editorial office.

17. Figure 1. a. APH isn't defined, and not sure what it is b. The influence of mercury by the FGD was not discussed in the paper. Perhaps there is none, but you have the chemical transformations in the table, and would imply that there are implications to the mercury reduction (or addition), and the fractionation between the three Hg types.

Response:

APH means air preheater. We have replaced it with full name. The "FGD" should be "WFGD" as explained in Comment 2. We have changed it in the figure.

18. Figure 2. Minor points here a. The arrows between the boxes; is there any meaning to the different sized arrows. If yes, describe in a caption. If no, make them all the same size. b. Same comment with blue arrows and size. c. Just a question: why

is the liquid phase in the Flue Gas Purification box not represented the same way? Is it water droplets? Should it be represented the same way? d. Same box. Does Hg0 really get removed by the spray? Is this why the arrow size is so small? too little affinity for water? e. Bottom box: in other boxes the Hg0 arrow goes both ways. Is it different here? Only one way?

Response:

We have revised all the arrows to the same size and deleted the sprayers to make it consistent.

Previous studies have observed liquid Hg^0 in the removed waste acid (Wang, 2011). Researchers deduce that the decreased temperature in the flue gas purification system is one important reason for gaseous Hg^0 condensation and removal by water spray. However, according to the saturated vapor pressure of Hg^0, this phenomenon happens when the gaseous mercury concentration is very high, approximately 29–62 mg/m^3 in the flue gas purification system. Currently, gaseous mercury concentration in this system in most of the Chinese nonferrous smelters can't reach this value. Therefore, we believe the Hg^0 removal by spray is not a predominant mechanism in the flue gas purification system, but we keep this mechanism in the figure. This point has also been added to the text.

Studies on the mercury behavior in the FGD system of nonferrous metal smelters are limited. According to studies in coal-fired power plants, Hg^{2+} in the FGD gypsum might be reduced to Hg^0 in the FGD system. We have added this mechanism into this figure.

Reference:

Wang, Q. W.: New technology for treatment of mercury-containing acidic wastewater from gas washing process in lead and zinc smelting by biologics, Ph.D. thesis, Central South University, Changsha, China, 2011.

19. Figure 4: I would add "and flow" to the caption after transformation.

Response:

Modification has been made accordingly.