Reply to Comments from Referee #3

1. "Environmental diplomatic affairs"?

Response:

We have revised it to "global environmental issues".

2. "Annex D for Article 8 of the Minamata Convention". Please provide Reference to this.

Response:

We have added the reference:

"United Nations Environment Programme (UNEP): Minamata Convention on Mercury, available at: http://www.mercuryconvention.org/Convention/tabid/3426/Default.aspx (last access: 1 February 2016), 2013b."

3. "Operationally defined chemical forms" Is this the right terminology?

Response:

We have modified it to "major chemical forms".

4. Authors present "The dust cake layer also facilitates oxidation of Hg0" How does dust cake layer facilitate oxidation? It should capture Hg instead! Needs clarification. Please provide reference of your argument.

Response:

The fly ash on the dust cake directly captures Hg^{2+} and facilitates the oxidation of Hg^{0} . We have added reference to this argument.

Reference:

Wang, F. Y., Wang, S. X., Meng, Y., Zhang, L., Wu, Q. R., and Hao, J. M.: Mechanisms and roles of fly ash compositions on the adsorption and oxidation of 5. 2.3 Reference to Table 1 is missing. And in the discussion part of the Table there are too many generalizations without supporting data. What are these data for, which coal types? Anthracite? Bituminous? Lignite?? What was the chlorine content? Those will have significant implications in Hg speciation. Actually, there are bulky data available in literature from field tests and Review paper should include those.

Response:

We have added all the references to Section 2.3. Section 2.3 is a summary of the ultimate speciation profiles for Section 2. The influence of coal quality (e.g. chlorine content, mercury content, etc.) on mercury speciation from coal combustion is discussed in Sections 2.1 and 2.2. The coal type does not directly influence mercury speciation. It is the coal quality of different coal types that takes the effect. We have included about 20 papers with field tests.

6. 2.2.5 Mercury transformation during wet flue gas desulfurization (WFGD): In this section, authors have mixed up information of all the plants together, without taking care of the upstream APCDs configuration. As authors have presented in the earlier section that APCDs configuration has important effect on speciation, authors have missed to explain the effect of upstream APCDs e.g. ESP and SCR in Hg removal in FGD. Needs careful review of these.

Response:

We have added the influence of SCR to this part. Please refer to Lines 216-218 on Page 8 in the revised manuscript:

"The applications of high-chlorine coal, SCR and halogen addition can increase the Hg²⁺ proportion in flue gas before WFGD, which will enhance the overall mercury capture efficiency of WFGD."

The influence of ESP is not clear. Because the proportion of either Hg^{2+} or Hg^{0} can increase after ESP due to the co-existence of Hg^{0} oxidation and Hg^{2+} reduction inside ESP, which has been discussed in Section 2.2.2.

7. Hg emission from MSW incinerators are one of the major sources, however, only little has been covered about it. No coverage on medical, hospital waste incineration? No discussion on hazardous waste incineration? This makes the review incomplete!

Response:

We have carefully modified this part and added results on mercury speciation in medical and industrial/hazardous waste incinerators. Please see Section 5.2:

"Waste incineration is a potential predominant source in the global mercury emission inventory. The major incineration types are municipal solid waste (MSW) incineration, medical waste incineration and industrial/hazardous waste incineration. A significant proportion of mercury (80-96%) in the MSW releases from the incinerator into the flue gas is in the form of Hg⁰ at 850–1000°C (Park et al., 2008). Grate furnace combustor (GFC) and circulation fluidized bed combustor (CFBC) are the two most commonly used incinerators. The flue gas from CFBC has a larger proportion of Hgp than that from GFC. Typical APCDs for incinerators are combinations of semi-dry or dry flue gas deacidification (SD-FGD or D-FGD) for SO₂ and HCl removal and dust controller (e.g., WS+ESP, FF, FF+WS, etc.). SCR is sometimes used as well for NO_x control. Activated carbon injection (ACI) is used for the control of persistent organic pollutants (POPs), which is required for incinerators in China. The overall mercury removal efficiency of the APCDs for MSW incineration ranges from 60% to over 99% (Zhang et al., 2008; Takahashi et al., 2012). Previous studies in Europe and the USA indicated that the Hg²⁺ proportion in the exhausted flue gas ranges from 75% to 85% (Pacyna and Münch, 1991; Carpi, 1997). A Korean study found the Hg²⁺ proportion in MSW incinerators to be in the range of 78-89%, and that in industrial waste incinerators are even as high as 96.3 - 98.7% (Park et al., 2008). Kim et al. (2010a) tested two medical waste incinerators with SD-FGD+FF+WS and got the Hg⁰ proportion to be 43.9% and 96.8% respectively. A Japanese study showed that an industrial waste incinerator with WS and wet ESP has the Hg⁰ proportion of 92.7% (Takahashi et al., 2012). Based on field measurements in eight MSW incinerators in China, Chen et al. (2013) found that average Hg²⁺ proportion in flue gas from the outlet of GFC+SD-FGD+ACI+FF is 96%, while that for CFBC+SD-FGD+ACI+FF is 64%. High chlorine content in the waste results in high Hg²⁺ proportion in the flue gas. Limestone slurry or powder sprayed in SD-FGD or D-FGD absorbs a large amount of Hg²⁺ and activated carbon adsorbs a large amount of both Hg⁰ and Hg²⁺. Particles from SD-FGD and ACI are captured by the

downstream FF. Hg_p is removed by all types of dust controllers. The high Hg²⁺ formation rate due to the oxidative condition in flue gas and the high Hg²⁺ removal rate by APCDs (especially SD-FGD, FF and ACI) cause the significant variation in mercury speciation profiles for incinerators."

8. Conclusion: "Hg0 is the predominant mercury species in exiting flue gases from coal-fired power plants due to the high Hgp removal efficiency of ESP or FF and the high Hg2+ removal efficiency of WS or WFGD." The predominant Hg0 is not only because Hgp removal in ESP or FF and the high Hg2+ removal in FGD, this is also because the emission of Hg0 is dominant in boiler outlet.

Response:

We have added one sentence to clarify this point and modified the expression of this sentence. Please see Lines 650-655 in the revised manuscript:

"The initial speciation of mercury after the boiler, smelter or kiln varies significantly because of the diverse qualities of coals or raw materials. Nearly all mercury in coal is released into the flue gas in the form of Hg^0 during combustion. Hg^0 is the predominant mercury species in exiting flue gases from coal-fired power plants mainly due to the high Hg_P removal efficiency of ESP or FF and the high Hg^{2+} removal efficiency of WS or WFGD."