"A 2-year record of atmospheric mercury species at a background Southern Hemisphere station on Amsterdam Island" by H. Angot et al.

Response to referee comments by Referee #1.

We thank this anonymous referee for very insightful questions and comments. We provide below a point-by-point reply to the comments (points raised by the referee in bold, changes made in the manuscript in italic).

1. A general area for improvement

The work needs to include a more critical discussion of the limitations of the RGM and PBM measurements in the context of the identified sampling biases and artifacts (Gustin et al., 2013;Huang et al., 2013;Lyman et al., 2010;Malcolm and Keeler, 2007;Talbot et al., 2011). At this point, the weight of evidence is clear – RGM and PBM suffer from serious biases and interferences (Jaffe et al., 2014). The authors acknowledge that the KCl denuder has problems with collection efficiency, but do not discuss what this means for reliability and interpretation of the Amsterdam Island dataset. There is probably meaningful information embedded in the RGM and PBM that can be defensibly interpreted (e.g., elevated PBM associated with African biomass burning), but not all aspects of the RGM and PBM data are reliable and these need to be more openly acknowledged. For example, the RAMIX intercomparison conclusively demonstrated Tekran RGM concentrations are biased very low (Gustin et al., 2013;Huang et al., 2013). And Rutter et al. (2008), Talbot et al. (2011), and Malcolm and Keeler (2007) have all published studies suggesting PBM is biased as well. All of this work suggests we cannot trust the absolute magnitudes of Tekran RGM and PBM. This then places serious limitations on how useful RGM and PBM data are for model evaluation and development, so I'd like to see the authors be more thoughtful about how exactly they suggest modelers (or policy makers) use the Amsterdam Island data.

We fully agree with the referee that RGM and PBM suffer from serious biases and interferences and that we cannot trust the absolute concentrations. We have added a more critical discussion in the revised manuscript: "*There is growing evidence that RGM and PBM measurements might suffer from significant biases and interferences (Lyman et al., 2010;Gustin et al., 2013;Jaffe et al., 2014). Several studies highlighted the inefficient collection of gaseous oxidized mercury compounds with a KCl-coated denuder in the Tekran technique (Gustin et al., 2013;Huang et al., 2013), leading to an underestimation of reactive mercury concentrations by a factor 1.3 to 3.7 (Huang et al., 2013). Other studies suggested sampling artifacts for PBM measurements due to temperature or sampling duration (Malcolm and Keeler, 2007;Rutter et al., 2008). Moreover, the upper size cut-off diameter at 2.5 µm raises concerns about mercury associated with large (> 2.5 µm) particle fractions (Kos et al., 2013), especially in the marine environment where mercury is likely mainly contained in coarse sea salt aerosols (Talbot et al., 2011;Feddersen et al., 2012). There is no robust calibration technique of the Tekran speciation unit and no certified reference material available. The precision of RGM measurements - shown to be of 15% under given conditions (Landis et al., 2002) – should be assessed in various sampling environments (e.g., varying ozone/relative humidity conditions). Given the limitations of the RGM and PBM measurements, data reported in this study should thus only be directly compared with the existing Tekran-based literature, as suggested by Wang et al. (2014). An*

extensive dataset has been gathered worldwide using the Tekran speciation technique, which is the best available automated method. Future interference and calibration tests are fundamental to validate measurements and quantify uncertainties (Kos et al., 2013), and might enable us to correct RGM and PBM data. Until then, orders of magnitude and variability in time and space of Tekran-based RGM and PBM concentrations can be used as first estimates by policy makers or to evaluate atmospheric models."

2. Specific comments

Title: Should there be a hyphen in "2-year". If so, please correct here and elsewhere in the text.

Corrected.

Page 14440

Lines 2-3: "Scarcity of mercury species records in the Southern Hemisphere is a critical weak point for the development of appropriate modeling and regulation scenarios". It is debatable whether or not the lack of Hg speciation measurements in the SH is actually a "critical weak point" for modelers and regulators. For example, I would content that uncertainty in the atmospheric chemistry or anthropogenic emission inventories are more serious weak points. The second half of the sentence is also not helpful because it's unclear what you mean by "appropriate modeling and regulation scenarios". I strongly suggest revising or replacing this sentence.

Agreed. In the revised manuscript we have changed that sentence to: *"Although essential to fully understand the cycling of mercury at the global scale, mercury species records in the Southern Hemisphere are scarce".*

Lines 14-15: "Lowest concentrations of GEM" is grammatically incorrect.

Corrected.

Line 17: I suggest deleting "for further modeling studies". First, it's ambiguous what sort of modeling studies the author refer to and, second, the Amsterdam Island don't are helpful more than just models and so why limit yourself to just supporting model studies?

We agree. "for further modeling studies" has been deleted.

Line 23: "were" should be "have been" and "exposition" should be "exposure".

Corrected.

Line 26: "However, research gaps for mercury control policies at regional and global scale still remain such as our understanding of mercury sources, atmospheric chemistry or deposition processes (Selin et al., 2007)". First, the grammar of this sentence needs to be corrected. Second, what's being said isn't helpful because it's so general. What would be helpful is a more specific statement about what knowledge gaps are really limiting Hg regulations? You may find Selin (2011) or Selin (2014) helpful for ideas and references.

We agree with the referee. In the revised manuscript we have changed that sentence to: *"However, research gaps limiting mercury reduction policies at regional or global scale remain. For example, the policy effectiveness at reducing deposition of mercury requires a better knowledge of the chemistry of atmospheric mercury species (Selin, 2014)."*

Page 14441

Lines 5: Ocean upwelling is not a primary source of Hg. Please delete.

Deleted.

Lines 3-8: The 10-30-60 partitioning is an original result from Amos et al. (2013) and not UNEP (2013).

Corrected.

Lines 19-22: "Nevertheless, the cycling of mercury at the global scale is not fully understood and the role of the oceans still remains unclear, mainly due to the lack of long-term records of atmospheric mercury in the Southern Hemisphere (Pirrone et al., 2013)". This sentence needs to clarify that atmospheric observations place an important constraint on the global Hg cycle, but atmospheric measurements in the SH are not the largest uncertainty in ocean cycling. Uncertainties in air-sea exchange and carbon dynamics/particle scavenging play a much larger role in the ocean.

That is correct. We have changed the text: *"To better understand the cycling of mercury at the global scale a coordinated global monitoring network is needed (Pirrone et al., 2013), along with long-term records of atmospheric mercury species in the Southern Hemisphere and at background sites (Sprovieri et al., 2010)."*

Page 14443

Lines 19: Please rephrase or delete "most relevant". Tekran GEM, RGM, and PBM are operationally defined and not objectively the most relevant forms to measure, rather they are the forms we can measure current instrumentation.

Corrected.

Page 14444

Lines 10: I'd recommend saying just "4h" instead of "3 to 4h" since you later say that the sampling period is 4 hours.

Corrected.

Lines 22: Is checking the permeation source frequent enough? How does it compare to other long-term measurement sites, such as Alert?

Steffen et al. (2012) recommend calibration of the permeation source at least once per year and Landis et al. (2002) when the permeation rate calibration and standard addition are off by more than 5%. At Amsterdam Island, after one year the manual injections were within 3% of the expected mercury concentrations during the manual injection checks.

Page 14445

Lines 1: How does the GMOS QA/QC protocol compare to the protocols of AMNet/CAMNet? I'm just curious for the sake of intercomparaison.

The quality control software used in this study includes a lesser number of automatically generated flags than protocols of AMNet/CAMNet (Steffen et al., 2012; see Annex), requiring additional interpretation before validating/invalidating data. To ensure uniformity across the network, GMOS is currently developing a QC software. An intercomparaison of GMOS and AMNet QC softwares will be undertaken.

Lines 3-13: It's confusing to state the detection limits as "better than X". If possible, please provide the actual detection limit or your best estimate.

We have provided our best estimate of the detection limit in the revised manuscript.

Lines 10-11: Landis et al. (2002) reported 15% precision for RGM and PBM. Lyman et al. (2010) and Gustin et al. (2013) have suggested that RGM interferences vary with O3 and RH, which would suggest the precision reported in one sampling environment does not apply to sampling environments with different O3 and/or RH levels. Please provide a justification for why the Landis et al. (2002) can apply to RGM and PBM at Amsterdam Island.

We agree with the referee that the precision/background noise might be impacted by ozone and relative humidity. However, to the best of our knowledge, the precision of replicated RGM measurements has never been studied under varying ozone/RH conditions. Our statement has been nuanced in the revised manuscript: *"…the precision of RGM measurements – shown to be of 15% under given conditions (Landis et al., 2002) – should be assessed in various sampling environments (e.g. varying ozone/relative humidity conditions)"*.

Page 14446

Lines 8: "till" should be "until".

Corrected.

Section 3.2: It would be appropriate to include a citation to (Sprovieri et al., 2010) for their review on Southern Hemisphere atmospheric Hg data. Also, please provide a rationale for why you directly compare TGM and GEM. Lastly, I suggest providing the months instead of seasons. It will be easier for Northern Hemisphere readers (likely the bulk of your readership) to follow the text.

The citation has been added in the revised manuscript: *"GEM data are lower than concentrations reported in the Northern Hemisphere but well within the expected range for a remote marine site in the Southern Hemisphere (Sprovieri et al., 2010)"*.

RGM concentrations in the marine boundary layer are usually <10 pg/m^3 (Soerensen et al., 2010). The difference between GEM and TGM concentrations is then less than 1%, which is insignificant when comparing orders of magnitude.

Months have been provided in the revised manuscript, thank you for the suggestion.

Page 14447:

Line 11: "did follow" should be "followed".

Corrected.

Line 28: Should "this assumption" be "this observation"?

Yes, corrected.

Page 14448: Is Amsterdam Island really representative of background "tropospheric conditions"? Does the marine boundary layer where you're sampling really represent the free troposphere as well?

You are right, this is not correct. Amsterdam Island is only representative of background marine boundary layer conditions. This has been corrected in the revised version.

Page 14450:

Section 3.2.3: You might consider adding a table with summaries of the data (e.g., montly mean, median, std dev, max, min, n) to make the Amsterdam Island more accessible for modelers. Alternatively, you could provide the QA/QC's data online as an Excel spreadsheet in the SI.

A table with the monthly means, medians and standard deviation has been added in the revised manuscript.

Based on your analysis, can you comment on the existing estimates for biomass burning Hg emissions (Friedli et al., 2009;Holmes et al., 2010)? Are they consistent with what you're finding?

It would have been interesting to investigate the correlation of Hg^0 to carbon monoxide during winter months/biomass burning season but CO is no longer monitored at Amsterdam Island. We are currently investigating fluxes of mercury in the marine boundary layer around Amterdam island using GEOS-Chem. Our study partly focuses on biomass burning mercury emissions from Southern Africa.

Page 14451:

Line 6: Is it a good assumption everywhere that RGM can only be transported <100s kilometers? What about RGM formed in situ in the dry upper troposphere/lower stratosphere where wet scavenging in infrequent?

The distance RGM can be transported depends on meteorological conditions. Our aim was to emphasize the fact that, due to its physico-chemical properties, RGM is quickly deposited compared to Hg^0.

We already mention that RGM can originate from entrainment from the free troposphere.

Page 14452:

Line 3-12: Please connect this paragraph more to RGM. Its relevance is not obvious. Also, please conclude with the take home message for RGM. It's not entirely clear what the bottom line is for RGM and what are the most important controlling variables at Amsterdam Island.

This paragraph has been reorganized in the revised manuscript: "*More frequent RGM events between December and March could also be in line with an enhanced biological activity in summer. The production of halogen species, photochemically oxidizing GEM, could be driven by biological activity (Gschwend et al., 1985). Unlike the oceanic region surrounding Amsterdam Island, an area located in a southwest upwind sector covering the subtropical front (see Figure 8) is highly productive, with a marine productivity (characterizes by chlorophyll-a concentration) peaking from December to January and sometimes in March-April (Sciare et al., 2009), in agreement with peaks of RGM events. Similarly, marine organic aerosol concentrations at Amsterdam Island have been shown to be directly related to the seasonal cycle of chlorophyll-a (Sciare et al., 2009) and dimethylsulfide (DMS) concentrations peaking in summer have been reported on the island, in line with an enhanced biological activity (NGuyen et al., 1990;Sciare et al., 1999).*

While enhanced photochemistry and biological activity in summer might explain more frequent RGM events at Amsterdam Island between December and March, further field studies are needed to fully understand divalent mercury formation pathways".

Line 14: Why would GEM adsorb to particles? Does that make sense based on its vapor pressure?

We have corrected this sentence in the revised manuscript: *"PBM is associated with airborne particles – e.g., dust, soot, sea-salt aerosols or ice crystal -, or originates from the adsorption of reactive mercury onto particles (Lu and Schroeder, 2004)"*.

Line 16: Rutter and Schauer (2007) and Amos et al. (2012) should be included as references in addition to Steffen et al. (2014).

Done.

Section 3.3.2: Are RGM and PBM decoupled at Amsterdam Island? Is there any influence from marine sea salt?

Yes RGM and PBM are decoupled at Amsterdam Island.

According to Feddersen et al. (2012), Holmes et al. (2009) and Talbot et al. (2011), most of divalent mercury in the marine boundary layer is present in coarse sea salt aerosols rather than gas-phase. Marine sea salts influence the marine boundary layer chemistry by scavenging soluble gases or outgassing compounds such as Cl_2 and BrCl (Hedgecock et al., 2003).

Page 14453:

Lines 22-23: Please be more specific about how your work "opens the way for new avenues in future modeling studies".

We provide new measurement constraints on the mercury cycle.

Why are future efforts at Amsterdam Island focused on lower detection limits? Huang et al. (2013) suggest RGM is biased low by 1.3-3.7x. The detection limit issue is moot if what Huang et al. (2013) found is true at Amsterdam Island. Why go after the detection limit and not aim improve oxidized Hg measurements by addressing the biases, interferences, and lack of calibration.

We fully agree. This sentence has been changed in the revised manuscript: *"Further studies involving wet deposition, simultaneous measurements of other trace gases, and interference and calibration tests of the Tekran speciation unit are needed to improve our understanding of deposition processes and oxidation mechanisms."*

Figure 3: Consider adding the month to the x-axis to help orient the reader.

Done.

Figure 4: The asterisk (*) to denote statistical significance looks like an outlier.

Corrected.

Figure 6: Please put dGEM under Rn 222 and align the x-axis. Then the reader can track the temporal evolution of the two together.

Done.

Figure 8: You might consider merging this with Figure 5. It would be easier to see the relationship between fire counts and PBM. Also please define "events" in the caption.

Done.

Figure 9: Please provide a bit more of a description in the figure caption, so that the figure can stand alone from the text. Right now, if I was a reader scanning the figures I wouldn't know why Figure 9 was important without digging into the main text.

Done.

3. References

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4. Annex

Flag comparison for GEM and speciation parameters. NA indicates no common flag for the program.

Flag Code This study	**Flag Code CAMNet**	**Flag Code AMNet**	**Flag description**
GEM parameters			
NA	V01	B2	Baseline voltage change
NA	V02	NA	Hg concentration high
NA	V03	E1	Hg concentration low
E5	V04	E5	Same cartridge difference > 10%
A1	V05	A1	Cartridge A/B difference
A2	I05	A2	Cartridge A/B difference
BL1	V06	B1	Baseline voltage low
NA	NA	B0	Baseline voltage low
BL2	V07	B3	High baseline deviation
NA	NA	B5	High baseline deviation
NA	V08	NA	Below detection limit
NA	V09	M2	Multiple peaks detected
NA	NA	M3	Multiple peaks detected
NA	I09	NP	No peak detected
V1	V10	V5	Questionable sample volume
NA	NA	V7	Sample volume
NA	V19	NA	Time gap in sampling records
NA	V99	NA	Standard addition recovery questionable
NA	NA	F1	Calibration interval
NA	NA	F2	Invalid flag – calibration interval
NA	NA	R_1	Detector sensitivity
NA	NA	R_2	Invalid flag – detector sensitivity
NA	NA	C5	Calibration change
NA	NA	C0	Calibration change
NA	NA	Z1	Calibration blanks
NA	NA	Z2	Invalid flag – calibration blanks

NA	NA	C1	Calibration trap bias
NA	NA	C2	Invalid flag – calibration trap bias

Speciation parameters

P1	V22	P1	PBM desorption questionable
G1	V23	G1	RGM desorption questionable
NA	V24	S0	High desorption zero value
S1	NA	S1	High desorption zero value
NA	V25	NA	Leak check
NA	V26	P0	No PBM observed
NA	V27	G0	No RGM observed
NA	I98	L1	Invalid desorption cycle
V1	I98	NA	Incorrect sample volume
E0	I19	E0	After desorption