Authors' reply to RC #1

SPECIFIC POINTS

1. Page 1, Abstract: Line 15: as this is the first discovery of hi-Si boninite in the Zambales ophiolite, this should be explicitly stated.

5 Sentence modified \rightarrow "We report for the first time..."

2. Page 1, Abstract: Line 18: place "the" before "Zambales ophiolite"; this has to be done in many instances throughout the ms

We reviewed the manuscript and placed "the" in places we've missed.

3. Page 1, towards bottom of the abstract: Perhaps should state that work on the Coto Block

10 was done by others and not by this study; I had to subsequently look through the ms to see if work was done on both the Coto and Acoje blocks

Reference added

4. Page 1, Line 31: would insert "and vice versa" at end of first sentence "and vice versa" added

15 5. Page 2, Line 2: after "plume-induced subduction initiation" should reference Whattam and Stern (2015) and Gerya et al. (2016) (I believe Whattam and Stern (2015) were the first to specifically coin this)

Added Whattam and Stern (2015). Gerya et al. (2016) is probably Gerya et al. (2015)? which was cited previously

20 6. Page 2, Line 6: Don't understand what "Challenge No. 11" means

As explicitly stated in the IODP (International Ocean Discovery Program) Science Plan for 2013-2013 "Challenge No. 11" is "How do subduction zones initiate, cycles volatiles, and generate continental crust?"

7. Page 2, Line 7: Would replace "including" with "with the exception of"

25 replaced with "with the exception of"

8. Page 2, Line 23: replace "verified" with "suggested"; we believe this to be the case, yes, but can't outright verify it

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replaced "verified" with "suggested"

9. Introduction focuses almost exclusively of subduction initiation (SI) at the IBM; relevant, but SI has also been discussed elsewhere; as well, different ideas of how SI transpires \rightarrow e.g., spontaneous vs. induced (Stern, 2004)

This is briefly discussed in Page 2 Lines 24-31

5 10. Page 3, Line 27: don't understand what "transitional" MORB means; transitional_to MORB and IAT? If so, state this. As well, the IBM FAB which may be analogous to_Coto Block MORB-like lavas, have characteristics intermediate to and which overlap_MORB and IAT (e.g., Whattam et al., submitted). For example, whole-rock chemistry_documents an arc-like Ti-V ratio <20 and evidence of melting of a source more_oxidized than MORB (higher Fe3+/FeT, 10 Brounce et al., 2015). More on relation

between Coto Block lavas and FAB later.

Sentence modified

Addressed fully in #56

11. Page 4, Line 7: sentence ending with "transition zone" needs references.

15 References added

12. Page 4, Line 24: I believe boninitic basalts was also mentioned earlier? These need to be defined at first instance (i.e., lavas which record MgO >8 wt. % and TiO2 <0.5 wt. % as per IUGS boninite definition but SiO2 <52 wt. %)

Sentence modified

13. Page 5, Line 17: Again, confused as to whether paper included Coto Block; Maybe state in first sentence of this paragraph that study was conducted on Acoje Block (only) Sentence changed to "For this study, a subset of forty-four (44) samples located along NW-SE transects of the Acoje Block volcanic sequence at Barlo were selected for whole rock geochemical analyses and screened through visual and microscopic assessment of secondary

25 alteration."

14. Page 5, Line 24: change "lost weight"→ weight lost changed to "weight lost"
15. Page 5, Line 31: Spell out GSJ/AIST

now "Geological Survey of Japan/National Institute of Advanced Industrial Science and Technology"

16. Page 6: Section 5.1: This section is very "dense" and difficult to read. I suggest making a table showing the different lithologies and their mineralogy and textures, and then significantly shortening the written description here

Seems ok to us

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17. Page 6, Line 15: Should of probably brought this up earlier, but similar to point 12 above, perhaps all the different categories of boninite (low-Si, high-Si, basaltic etc.) should be explained in the introduction or at least at an earlier point in ms

10 18. Page 6, Line 20: I think this is the first mention of Ogasawara? Mention where this is-Japan → part of IBM forearc?

Sentence changed to "This assemblage corresponds to Type II boninite of Umino (1986) in samples described from the type locality at Chichijima island, Ogasawara (Bonin) Archipelago in southern Japan."

15 19. Page 6, Line 24: change is → are changed
20. Page 7 Line 6: Insert "A" before peculiar changed

- 20 21. Page 7, Line 15: These LOI are very high. And what rock types exhibit these values? All or just high-Si boninites? These should be in the Table in Supp. Doc., correct? So put Supp. Doc. X at end of sentence. Maybe a plot of LOI versus various potentially mobile elements (e.g., MgO, K2O, Na2O, Ba) is warranted? Or at least some sort of statement like "though the LOI are high our petrologic arguments are based primarily on trace elements known to be
- 25 immobile up to greenschist-facies conditions". Are any filters being applied to your samples? For example, using only samples which yield 98-102 wt.% oxides or ones with <3 % LOI? Yes, indeed the LOI values are a bit high. We note that in general boninites have LOI values that are greater than 3 wt%.

IODP Expedition 352 shipboard boninite samples have LOI values that are much higher (mostly between 5-11 wt%) and samples from Haugen, 2017 which are the "cleanest, least weathered pieces" of the shipboard cores have LOI values that are just as much as the samples from this study.

5 Concerning element mobility, this is addressed in Page 7 lines 15-19.
Filters have already been applied in the current dataset. The current dataset was screened not only based on LOI values but primarily based on element immobility judged using diagrams such as CaO vs. Na₂O, major oxide vs LOI, and major oxide/trace element vs. immobile element (e.g. Zr). Samples with mobile element enrichment (e.g. total alkalis, Rb) followed a
10 vertical vector and were excluded.

22. Page 7, Line 18: What is primary? And I note here that primary is used later on but not defined. Do you mean primary lavas such that exhibit high MgO, high Mg# (≅65), high Cr and high Ni?

Sentence modified.

15 23. Page 7, Line 22: I think a reference is needed after "boninitic basalts". Maybe Pearce and Robinson, or Reagan et al. (2017)? Not sure.

Reagan et al. (2015) added

24. Page 7, Line 23: As mentioned at the beginning, Figure 6 is very "busy". Would suggest plotting only samples from this study.

20 We slightly modified Figure 6. We removed objects that are not discussed in the text such the primary PAB and PAB fractionation. However, we've chosen to still include the Ogasawara and Expedition 352 boninite and boninite series datasets. Our point is to compare Zambales boninite with the dataset from the IBM forerac. Another point is to show the major element variation vis-à-vis the modelled fractional crystallization path. We believe these are better

25 addressed if our data are shown with the Ogasawara and Expedition 352 datasets. The panels are now marked as "a, b, c"

25. Page 7, Line 24: change second "and" \rightarrow or changed

26. Page 7, Line 29: pristine? You mention above LOI values of 4-7 wt.% Addressed fully in #21 27. Page 7, Line 31: ug/g? usually reported in ppm Indeed, trace elements are traditionally reported in ppm but as per Wiedenbeck, (2017) ug/g is 5 better 28. Page 8, Line 3: replace within with \rightarrow between changed to between 29. Page 8, Line 15: Change so reads: Compositions of Zambales boninite. are marked by 10 low incompatible trace element abundances... changed to "Compositions of Zambales boninite and boninite series volcanics are marked by low incompatible trace element abundances relative to MORB" 30. Page 8, Line 23: replace times with $\rightarrow x$ replaced with x 15 31. Page 8, Line 25: insert boninite between Zambales and ophiolite now "The effect of fractionation in Zambales boninite samples" 32. Page 8, Line 32: descending order is unclear; perhaps describe from base \rightarrow top which is probably standard convention now "The volcanic sequence at Barlo, based on observed local stratigraphic relationships, is 20 LSB series volcanics-boninitic basalt-LSB-HSB from base to top." 33. Page 9, Line 6: Is unclear how can be classified as moderate-Fe tholeiites without the Miyahsiro plot overlain by Arculus' low-med-high Fe series fields Figure will be added as Supplementary Figure 1 34. Page 9, Line 20: Haugen (2017): Is this a MSc or PhD thesis (not indicated in references). 25 References have been updated.

35. Page 10, Line 4: I think a paragraph at least is warranted to explain how the modelling was done using MELTS (supplementary document probably appropriate).

The modelling is fairly straightforward and the parameters are given in the figure captions. Figure 6 captions is now modified

36. Page 10, Line 5: Ghiorso and Gualda (2015) not in references Ghiorso and Gualda (2015) is in the references

5 37. Page 10, Line 16: change "in the base" to → at the base 5 change to "at the base"
38. Page 10, Line 17: break sentence; add ";" after "at depth" now "at depth; influx"

10 39. Page 10, Line 20: change slightly deviate \rightarrow deviate slightly changed to "deviate slightly"

40. Page 10, Line 24: change does \rightarrow do changed

41. Page 10, bottom of page: Would change Section 6.2 title to \rightarrow Slab contributions

15 now "6.2 Slab contributions to a less depleted mantle source"

42. Page 10, bottom of page: Would include more up-to-date references for boninite petrogenesis

additional references added

43. Page 11, Line 7-8: OK, but they are equally LREE-depleted. What is the explanation for

20 the spoon-shaped REE patterns? I think for the "classical" U-shaped signatures that the explanation is high-degree partial melting (which produces low MREE) which is subsequently slab-fluid modified to produce LREE enrichment; not sure of explanation for the high HREE less depleted source

44. Page 11, Line 13: Maybe explain at beginning of section that Ba/Th is a marker/gauge of

25 shallow slab-contributions and reference (Pearce et al., ?) Why Ba liberated at shallow conditions? Low temperature (I think); low P as well?

45. Page 11, Line 14: insert "increasing" after "mirrored by" now "mirrored by increasing"

46. Page 11, Line 15: reference Fig. 8b after Th/Yb Fig. 8b added

47. Page 11, Line 16: what is decoupled?

48. Page 11, Line 18: Insert "A" before "high U/Th ratio"

5 now "A high U/Th ratio"

49. Page 11, Line 19: change ratio ! ratios; change by ! if; add "to source" at end of sentence changed

50. Page 11, Line 21: the La/Th vs. Sm/La is not shown so have to indicate this "not shown" now indicated

10 51. Page 11, Lines 30-31: Why mention slab melts? This is not mentioned previously and I don't think anyone familiar with boninite petrogenesis would consider slab melts as part of the equation.

Sentence now modified

52. Page 11, Line 2: change so reads: transitional between MORB and IAT

15 changed

53. Page 12, Line 3: Change "in" \rightarrow on the basis of

changed

54. Page 12, Lines 3-4: Confusing sentence; why mention distinct from Mariana BAB?

20 Mariana BAB is mentioned because originally Hawkins and Evans, 1983 characterized Coto Block as back-arc basin oceanic crust. By comparing certain immobile elements (e.g Hf, Ta, Th), Geary et al., 1989 was able to show that it is distinct from Mariana BAB and that is has composition transitional to MORB and IAT.

55. Page 12, Line 5: Another confusing sentence; have to get point across that depletion in
REEs, TiO2, Zr and Y of Acoje relative to Coto documents the progressive depletion of. . .what about LILE enrichments? These should increase from Coto ! Acoje

LILE enrichments are not discussed for two simple reasons- (1) paucity of reliable LILE analyses in the compiled dataset (Hawkins and Evans, 1983; Geary et al., 1989; Yumul, 1990,

Evans et al., 1991, Tamayo, 2001), in fact no samples from the Coto Block crustal section have been analyzed by ICP-MS and (2) the altered nature of most samples, with most samples lacking petrographic characterization, makes it difficult to assess the primary nature of LILE enrichment.

- 5 56. Page 12, Line 10: Can't readily see where Coto lavas plot in Ti/V space but this is a very important point as FAB can be distinguished by MORB on the basis of Ti/V which is arc-like (>20) and by virtue of elevated Fe3+/FeT indicative of a more oxidized (arc-like) source. Suggest you state what the Ti/V ratios of the Coto lavas are and compare these with those of IBM. Are they similar or not?
- 10 We increased the symbol size of Coto volcanics in Fig 6.

Coto Block volcanics (n=9) have variable Ti/V ratios, mostly between 20-26 (Fig. 6f). Recognizing the small sample size, this range is higher than proto-arc basalts (PAB) and overlaps with MORB (Supplementary Fig. 2a); suggesting a less oxidized source compared to IBM PAB.

We also added the primitive mantle normalized immobile element pattern for Coto Block volcanics together with IBM PAB (Supplementary Figure 2b) to highlight its depleted nature.
57. Page 12, Section 6.4. See the Presentation and Scientific Interpretations section
58. Page 13, Line 12: insert "above a west-dipping subduction zone" after (Ishizuka et al., 2011)

20 changed

59. Page 13, Lines 15-20: sounds perhaps like a plume-induced SI scenario addressed in first-half of response

60. Page 14, Lines 6-8: Why feasible? No explanation for this (doubly-vergent subduction) addressed in first-half of response

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25 61. Page 14, Line 11: Change north-verging → NE-verging changed to NE-verging

In addition, we slightly modified Fig. 7 because Nepoui and Cape Vogel HSB were mislabeled.

Fig. 7 is now updated

Authors' reply to RC #2

- ⁵ Page 1 Line 13: Delete "uniquely" and just say "predominant." We feel that it is indeed unique, especially the occurrence of high-silica boninite. This point is raised by Reagan et al., (2017) as well.
- Page 2 Lines 2-4: The statement that Cenozoic subduction initiation (SI) requires pre-existing weak zones or lithospheric collapse is erroneous. All models of SI require a weak zone, even lithospheric collapse.

This statement is a general characterization to differentiate spontaneous and induced SI from plume-induced SI.

Page 2 Lines 9-10: I wouldn't say that the IBM is "the most appropriate locality" to test models of subduction initiation. It might be the most appropriate place to test the Stern and Bloomer-type model, but there are other models as well. Maybe say "one of the most appropriate..." Sentence modified → "...one of the most appropriate..."

20

- Page 2 Lines 24-25: Cite references for "spontaneous" (e.g., Leng and Gurnis) and "induced" (e.g., Hall et al.) SI that have been replicated by numerical modeling. These references were cited in the following sentences (original MS page 2 lines 25-29.
- 25 Page 2 Line 28: delete semi-colon Semi-colon deleted.

Page 2 Line 30: Maybe change "unequivocally be attributed" to "uniquely explain." Sentence modified.

30

Page 3 Line 1: In discussing the basement complexes of the Philippines, Encarnacion (2004) might be an appropriate paper to cite. (Encarnacion, 2004, Multiple ophiolite generation preserved in the northern Philippines. Tectonophysics.) Additional references added.

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Page 3 Line 17: Change "structurally bound" to "fault-bound" (if that is what is meant by "structurally bound"). Changed to "fault-bound"

5 10	Page 3 Lines 21-22: In reference to "terrane docking," I presume the Zambales ophiolite is the terrane. But what is it docking with? (Indeed, what do you mean by "docking"?) Here terrane docking is used in a way similar to Pubellier et al., (2004). We mean that by the Pliocene its transport from equatorial positions to its present location probably have ceased. The identity of what Zambales ophiolite "docked with" is still unknown. It may be a composite of the following (1) an ocean basin that predates the South China Sea (pre-2016 there is consensus that this is proto-South China Sea), the vestiges of which are the Jurassic-Cretaceous cherts in the West Luzon Shear Zone of Karig, (1983) and complemented by recent radiolarian studies of Queaño et al., (2017a, 2017b) and (2) the northern extension of the Palawan microcontinental block (based on seismic data, Arfai et al., (2011) speculates that West Luzon Basin west of Zambales ophiolite is partly underlain by continental basement).	Field Code Changed
15	Page 3 Line 23: Change "in the westernmost margin" to "just west" of the ophiolite. The Mesozoic cherts are not found in the ophiolite itself, but in mélange-type shear zone material west of the ophiolite. The limited distribution of these cherts maybe more aptly described with "in the westernmost margin" rather than "just west" of the ophiolite.	
20	Page 3 Line 23-25: Besides Queano et al. 2017, you should also cite Hawkins and Evans (1983) who first described these cherts and Encarnacion (2004) who provides additional description of the shear zone. Additional references added - Hawkins and Evans (1983), Karig et al., (1986) and Encarnacion (2004).	
25	Page 3 Line 24: The shear zone is not "buried;" it is well-exposed in several places. Sentence modified	
30	Page 4 Lines 2-4: You might want to note that the idea that the San Antonio massif is a displaced block from the north is disputed by Encarnacion et al. (1999) for lack of convincing evidence. I re-visited the Encarnacion et al., (1999) paper but I can't seem to find the lines that dispute idea that the San Antonio massif is a displaced block from the north. In Encarnacion (2004) page 119, it is stated that "Except for some possible tectonic displacements of locally derived blocks (Yumul et al, 1998) the ophiolite is a coherent slab".	
35	Page 4 Line 18: You may want to add that the arc affinity of the Acoje block is also consistent with radiogenic isotope data (Pb, Sr, and Nd), which indicate hydrous fluid enrichment (Encarnacion et al., 1999). This reference is cited in the discussion section 6.2	
40	Page 4 Line 24: What rock types were the "bedding planes" measured on? Bedding planes were measured primarily on pillow lavas and tuff breccias.	
	10	

Page 4 Line 26: Change to "... NW-plunging anticline just south." Seems ok to us. 5 Page 4 Line 28: What does "conjugate intrusive directions" mean? (Intruded into conjugate fractures sets?). Please clarify. (Dikes with) conjugate intrusive directions pertain to obliquely intruding dikes. Line 32: Change to "subaqeous fall-out deposits" (?) 10 Changed. Page 5 Lines 3-4: Do the terms "Strombolian to Hawaiian fire fountaining" apply to subaqeous/submarine volcanics? I would think not(?) Are these deposits in fact subaerial? If so, that would be a very important finding! It would imply that portions of the Zamabales 15 ophiolite in fact contain what might be actual subaerial island arc crust, which would substantially change the paleogeography of Luzon. Please clarify. Here we are referring to submarine Strombolian to Hawaiian-type fire fountaining. Sentence is modified and a reference discussing deep submarine pyroclastic eruptions (Head Field Code Changed and Wilson, 2003) is added. 20 Page 5 Line 12: What are the "upright structures"? Sedimentary/volcaniclastic bedding? Please clarify. "Upright structures" include flattened flow lobes in the summit of the recognized pillow volcano shown in Fig3d (inset). "Upright" is used in page 5 line 12 to describe the primary igneous structures (emplacementrelated) in the synclinal area in a broad sense. It is used in contrast to post-ophiolite emplacement folding affecting the volcanic section and overlying sedimentary sequences. Page 6 Line 1: Change "12" to "Twelve." (Avoid numerals at the beginning of a sentence.) Sentence modified. 30 Page 6 Line 2: Add "Standard JB-2 and JB-3 ..." Sentence modified. Page 7 Line 15: Change to "The Cs, Rb, ... and Mn contents in ..." 35 Sentence modified. Page 8 Line 16: Change to "...MORB for most trace elements..."

40

I think this is not necessary.

Page 10 Line 32: Please add a few words summarizing the general consensus on boninite genesis. ("...there is general consensus on boninite petrogenesis, namely that..."

Sentence modified.

Page 12 Line 3: Change semi-colon to a period. Sentence modified.

Line 7: Change "Ma" to "Myr." (You are referring to differences in ages, not a point in time.) Sentence modified.

Page 13 Lines 2-5: Regarding the paleomagnetic data, do the declinations support a
 Philippine Sea Plate connection as well? The Philippine Sea Plate has been inferred to have rotated clockwise. There are additional previous paleomagnetic studies (for example, Fuller et al., 1991, J. Asian Earth Sciences) that appear to demonstrate counterclockwise rotation for Luzon.

Declination-based interpretations of the Cenozoic rotation history of Luzon and Zambales are contentious; Fuller et al., (1991) argues for CCW rotation while McCabe et al., (1987) calls for CW rotation.

As discussed by the Queano et al., (2007), the use of declination data in tectonic models of Luzon and Zambales might be of limited use since the CW and CCW directions cannot be unambiguously ascribed to either local or major plate rotations.

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Page 13 Line 20: By "rapid emplacement" do you mean "rapid formation"? If not, what do you mean by "rapid emplacement"?

We do not mean rapid formation. Rapid emplacement is inferred based on the reasoning that juvenile arc magmatism progressed only up to boninite; that it is a case of "failed' subduction initiation or arrested arc development. On the other hand, the IBM forearc which can be

described as a case of "successful" subduction initiation that produced an intra-oceanic arc with calc-alkaline lavas (41-35 Ma) after boninite (48-44 Ma) and proto-arc basalt (52-48 Ma).

Page 13 Line 20: Change to "The timing of the proposed subduction inititation..." 30 Sentence modified.

Page 13 Lines 21-35 to lines 1-3 of page 14: This section discusses the relationship between the Zambales ophiolite and Eocene (Angat) and Cretaceous ophiolites in the east Luzon area. The authors state that the similarity in ages of the Zambales and Angat ophiolites presented in Encarnacion et al. (1993) "does not necessarily prove and affinity" between the two. But

- 35 Encarnacion et al. (1993) "does not necessarily prove and affinity" between the two. But Encarnacion et al. (1993) (and Encarnacion et al., 1999, and Encarnacion, 2004) did not use the ages alone in arguing that the Zambales and Angat ophiolites (and Cretaceous ophiolites) are contiguous (not exotic/allochthonous to each other). The geology and stratigraphy as well are consistent with the Zambales-Angat ophiolite forming adjacent to the Cretaceous ophiolite
- ⁴⁰ in the east, which was the main point of Encarnacion et al., 1993 (and reiterated/amplified in Encarnacion, 2004).

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-	Field Code Changed
	Field Code Changed
	Field Code Changed

	We disagree with the interpretations of Encarnacion et al. (1993).		
5	In particular, we invite the reviewer to place the west dipping Cretaceous-Eocene subduction zone east of Sierra Madre that produced a late Cretaceous arc and an Eocene arc-backarc basin pair (presumably the Zambales ophiolite) shown in Fig.12 of Encarnación, (2004) in current global and regional plate reconstructions of SE Asia and the western Pacific region (Wu et al., 2016; Zahirovic et al., 2014, 2016).		Field Code Changed Field Code Changed
10	Indeed, age constraints were not the sole parameter used in arguing that the Zambales and Angat ophiolites (and Cretaceous ophiolites) are contiguous. But the Eocene volcaniclastic sequence, as shown in Fig.6 of Encarnación, (2004), overlies the Angat (Eocene) and Montalban (Cretaceous) ophiolites and not the Zambales ophiolite. Bachman et al., (1983) notes that there is marked lithelescal difference between the cast and weet flanks of the	*****	Field Code Changed Field Code Changed
	Luzon Central Valley Basin (CVB); he further characterized the CVB as a forearc basin.		
20	Regarding the issues mentioned above, I think suggesting that the Zambales is exotic to east Luzon causes greater problems with the proposed model, because the east Luzon ophiolites and arc crust are in-between the Zambales and the Philippines Sea Plate. In other words, if one wants to separate the Zambales ophiolite from east Luzon, shouldn't it also be separate from the Philippine Sea Plate?		
25	On the contrary, we find no problem if east Luzon ophiolites and arc crust are placed in- between the Zambales ophiolite and the Philippine Sea Plate. With regards to Zambales ophiolite being "separated" from the Philippines Sea Plate, this is the case in earlier tectonic reconstructions which follows the arc- backarc basin scenario where Zambales ophiolite is alternatively placed east of the Celebes Sea Basin in contact with the western margin of the Philippine Sea Plate (Fig.5 of <u>Rangin et al.</u> , 1990a and Plate 2 of Rangin et al., 1990b).		Field Code Changed Field Code Changed
30	In its present form, the G-Plates based reconstruction in Fig 10b using the unfolded slab data, plate polygons and rotation file of Wu et al. (2016) is actually compatible with Encarnacion (2004) in a broad sense. If Zambales ophiolite was formed by the proposed incipient subduction on the western margin of the Philippine Sea Plate, one can argue that Zambales ophiolite can be continuous with basement of Luzon and that it is relatively autochthonous		
35	(with respect to Luzon) (Encarnación, 2004); albeit highly displaced (relative to its present		Field Code Changed
	position) caused by transport along a plate boundary (Pubellier et al., 2004). A back-arc origin for Eocene ophiolites associated with Cretaceous arc in eastern Philippines is compatible with Fig. 10b as well.	*****	Field Code Changed
40	Arguments against a backarc basin origin for Zambales are given in section 6.3; the only contention would then be the relationship of Angat to Zambales ophiolite.		

Page 14 Lines 5-7: Why is doubly-vergent subduction "feasible"? Please elaborate.

We speculate that the location of Philippine Sea Plate (PSP) in the nexus of Pacific, Indo-Australian and Eurasian plates and their long-term Cenozoic plate motion makes doublyvergent subduction initiation along its margins feasible. The northwestward translation and

5 clockwise rotation of the Philippine Sea Plate starting in the early Eocene had to be accommodated by the adjoining oceanic domain east of southern Eurasia (e.g. East Asian Sea); hence, its interaction with the oceanic leading edge of the Philippine Sea Plate is expected (Wu et al., 2016; Zahirovic et al., 2016) and likely led to incipient subduction (Fig. 10).

Page 14 Line 29: Change to "By studying the Zambales ophiolite..." Sentence modified.

Page 14 Line 29: Delete "(SI)" 15 Deleted.

Page 14 Line 29: It is stated that subduction initiation is a "plate-scale process." I'm not sure what the purpose of this statement is. When is it not a plate-scale process? Please clarify. The purpose of the statement is to emphasize that subduction initiation may not be localized in the purpose of the Philips of Plate Philips of Plate Plate and Plate Plat

- 20 the eastern margin of the Philippine Sea Plate. The doubly-vergent SI configuration presented here is distinct from current SI scenarios solely based on the IBM forearc which mainly focuses on the problem of whether subduction initiation is spontaneous or induced (e.g. Arculus et al., 2016; Keenan and Encarnación, 2016).
- 25 Figure 2: The caption should include the references for the ages shown in the figure. References added.

Figure 2: Page 5 says the 44.1 Ma age is from a sill. This isn't clear or indicated in the stratigraphic column.

14

30 It is shown in the stratigraphic column.

Figure 10: In panel "b", what are the diamonds, inverted triangles, and squares? Captions modified.

35

10

Field Code Changed

Boninite and boninite-series volcanics in northern Zambales ophiolite: Doubly-vergent subduction initiation along Philippine Sea Plate margins

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Abstract. A key component of subduction initiation rock suites is boninite, a high-magnesium andesite that is uniquely predominant in Western Pacific forearc terranes and in select Tethyan ophiolites such as Oman and Troodos. We report for
 the first time the discovery of low-calcium, high-silica boninite in the middle Eocene Zambales ophiolite (Luzon island, Philippines). Olivine-orthopyroxene microphyric high-silica boninite, olivine-clinopyroxene-phyric low-silica boninite and boninitic basalt occur as lapilli fall deposits and pillow lava flows in the upper volcanic unit of the juvenile arc section (Barlo locality, Acoje Block) of the Zambales ophiolite. This upper volcanic unit in turn overlies a lower volcanic unit consisting of

basaltic andesite, andesite to dacitic lavas and explosive eruptives (subaqueous pahoehoe and lobate sheet flows, agglutinate,

- 20 and spatter deposits) forming a low-silica boninite series. The overall volcanic stratigraphy of the extrusive sequence at Barlo resembles Holes U1439 and U1442 drilled by IODP Expedition 352 in the Izu-Ogasawara (Bonin) trench slope. The presence of <u>depleted</u> proto-arc basalts in Coto Block (45 Ma) (Geary et al., Tectonophysics 168(1), 43-63, 1989), boninite and boninite series volcanics in Barlo, Acoje Block (44 Ma) and simultaneous and post-boninite moderate-Fe arc tholeites in Sual and Subic, Acoje Block (44-43 Ma) indicate that the observed subduction initiation stratigraphy in the Izu-
- 25 Ogasawara-Mariana forearc is present in the Zambales ophiolite as well. Paleolatitudes derived from tilt-corrected sites in the Acoje Block place the juvenile arc of northern Zambales ophiolite in the western margin of the Philippine Sea Plate. In this scenario, the origin of Philippine Sea Plate boninites (IBM and Zambales) would be in a doubly-vergent subduction initiation setting.

1 Introduction

30 As the surface manifestation of a convecting mantle, subduction zones play a fundamental role in material transfer from the surface to the deep interior and vice versa. Early in Earth's history, the transition from a stagnant lid regime to the present mode dominated by plate forces necessitates subduction initiation. Numerical models show that plate tectonics or mantle

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overturn in terrestrial planets can be triggered by narrow plumes impinging on weakened crust inducing gravitational instability and slab descent (Crameri and Tackley, 2016; Gerya et al., 2015). Internally driven mechanisms such as plume-induced subduction initiation (Whattam and Stern, 2015), however, are fundamentally different from end-member models of Cenozoic subduction initiation that rely on pre-existing zones of weakness such as fracture zones and transform faults or

- 5 require lithospheric collapse (Stern, 2004). How subduction begins in the oceanic domain is one fundamental issue yet to be fully deciphered, thus is regarded as a high priority scientific target (Challenge No. 11) of the International Ocean Discovery Program (IODP) for 2013 to 2023. Understanding this volcanic and tectonic process is hampered by its transient nature, with few examples in the geologic record with the exception of submarine active and paleo-forearcs and on land as suprasubduction ophiolites.
- 10 In testing subduction initiation models, the Izu-Ogasawara (Bonin)-Mariana (IBM) forearc region remains as <u>one</u> the most appropriate locality having a temporally distinct volcanic record of oceanic accretion and juvenile arc magmatism immediately following the onset of subduction. After the subduction of Pacific Plate beneath the Philippine Sea Plate commenced at 52 Ma, magmatism progressed from 52-48 Ma mid-ocean ridge basalt (MORB)-like proto-arc basalts (or forearc basalts) to 48-44 Ma boninite and boninite-series volcanics followed by 45-35 Ma arc tholeiites and calc-alkaline
- 15 lavas (Ishizuka et al., 2011; Kanayama et al., 2012; Reagan et al., 2010). The widespread occurrence of subduction initiation rock suites along the eastern margin of the Philippine Sea Plate is summarized in Fig.1a. Basalts of similar composition and age range to proto-arc basalts dredged along the Mariana and Bonin trench slopes were found at IODP Expedition 351 Site U1438 in the Amami Sankaku Basin (Arculus et al., 2015; Ishizuka et al., 2018). This indicates that the Oligocene Kyushu-Palau Ridge (KPR) intra-oceanic arc is built upon an igneous basement that formed part of the earliest seafloor spreading
- 20 event related to subduction initiation. The subsequent late Oligocene opening of the Shikoku-Parace Vela Basin resulted in its present configuration with Site U1438 in the backarc side of KPR. Based on dredging and dive observations in the forearc trench slope, proto-arc basalts are found at deeper depths relative to boninites which occur upslope and subaerially at Chichijima (type locality) and Mukojima island groups (Ishizuka et al., 2011; Kanayama et al., 2012; Umino & Nakano, 2007). The hypothesis that proto-arc basalt lies stratigraphically below boninite has been <u>suggested</u> by IODP Expedition 352
- 25 based on four sites drilled in the trench-side slope of the Bonin Ridge (Reagan et al., 2015, 2017). Both spontaneous and induced initiation have been replicated in numerical models of incipient subduction at the IBM. Earlier models with imposed plate velocities generally focused on initiation across a pre-existing weak zone, and combined with melting models could reproduce the observed proto-arc basalt-boninite stratigraphy (Hall et al., 2003; Leng et al., 2012). Subsequently, spontaneous subduction initiation has been shown to be plausible with compositional density contrast being provided by the
- 30 thickened middle crust of relic Cretaceous arc terranes (Leng and Gurnis, 2015). So far, neither spontaneous nor induced initiation models can <u>uniquely explain subduction initiation that produced the IBM</u> forearc and the mechanism remains elusive (Keenan and Encarnación, 2016).

Here an alternative approach to exploring subduction initiation processes in the Western Pacific region is presented. Following Stern et al. (2012), this study focuses on the middle Eocene Zambales ophiolite in the Philippines as it offers Deleted: including

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~3,200 km² exposure of Eocene supra-subduction oceanic lithosphere. Most of the Jurassic to Cretaceous ophiolites that form basement complexes in eastern Philippines are increasingly recognized as complementary features to Cretaceous terranes in the northern Philippine Sea Plate (e.g. Amami plateau, Daito Ridge) possibly sharing a common history as the overriding plate prior to initiation of subduction (Billedo et al., 1996; David et al., 1997; Deschamps and Lallemand, 2002;

- Lallemand, 2016). Therefore, juxtaposed Eocene ophiolites are potential targets to investigate subduction initiation as a 5 plate-scale process. Regional tectonic reconstructions predict the interaction of a pre-Eocene ocean basin located east of Eurasia and the western margin of the Philippine Sea Plate (Wu et al., 2016). Coincidentally, aside from the IBM forearc region, the Dasol-Barlo locality in northern Zambales ophiolite is where middle Eocene boninitic rocks are also reported (Evans et al., 1991; Florendo & Hawkins, 1992; Hawkins & Evans, 1983). The significance of Zambales ophiolite as an
- 10 analogue of the IBM forearc has been recognized since the early 1990s (Pearce et al., 1992) yet this connection remains unexplored. Motivated by how the ophiolite concept progressed through studies of the IBM forearc, we present field, petrologic, and geochemical characterization of a juvenile arc section in northern Zambales ophiolite to evaluate subduction initiation and boninite petrogenesis in a regional geodynamic context.

2 Geologic background

- 15 Located in western Luzon, the structurally coherent Zambales ophiolite is a representative supra-subduction ophiolite of the Western Pacific and the largest in the Philippines (Fig. 1b). The ophiolite spans the entire Zambales mountain range from 16° N and 14.7° N where it partly underlies stratovolcanoes of the Luzon Arc including Mt. Pinatubo. It consists of three fault-bound massifs, namely Masinloc, Cabangan and San Antonio, that each preserve complete ophiolite internal stratigraphy. The ophiolite is flanked by the Central Valley Basin (CVB) to the east and West Luzon Basin to the west. Late
- 20 Eocene to late Miocene Aksitero and Moriones Formations of the CVB record the transition from a hemipelagic, deep marine setting to shallower environment dominated by ophiolite-derived sedimentation signaling the uplift of the ophiolite. While sedimentary sequences west of the ophiolite, Cabaluan and Sta. Cruz Formations, indicate that terrane docking and large-scale uplift have transpired by the Pliocene (Karig et al., 1986; Schweller et al., 1984). Sporadic chert slivers with late Jurassic-early Cretaceous radiolarian fauna are found in the westernmost margin of the ophiolite (Hawkins and Evans, 1983;
- Karig et al., 1986; Queaño et al., 2017b). Similar Mesozoic cherts associated with disrupted ophiolitic fragments in northern Luzon highlights the existence of a shear zone (West Luzon shear zone) west of the ophiolite (Encarnación, 2004; Karig, 1983; Queaño et al., 2017a)

Petrologic, geochemical and isotopic studies reveal that the Zambales ophiolite represents two distinct mantle-crust sequences - the 45.1 Ma Coto Block with affinity transitional to mid-ocean ridge basalt (MORB) and island arc tholeiite

30 (IAT) and the 44.2 - 43.7 Ma Acoje Block with island arc characteristics (Encarnación et al., 1999; Encarnación et al., 1993; Geary et al., 1989; Hawkins & Evans, 1983; Yumul, 1990). The existence of a NNE-SSW trending structural boundary previously characterized by Hawkins and Evans (1983) as a left-lateral fault between Acoje and Coto Blocks within the

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Masinloc massif is supported by gravity and magnetic data with anomaly contrasts which extend at depth (Salapare et al., 2015). Although the juvenile arc character of Acoje Block and the supra-subduction origin of Zambales ophiolite are clear, the crustal nature and tectonic environment of Acoje and Coto blocks remain unresolved. Acoje Block as defined by Yumul et al. (1990) consists of the northern Masinloc massif and the San Antonio massif while the Coto Block consists of the

- southern Masinloc massif and the Cabangan massif (Fig. 1b). The present disposition of Acoje Block is postulated to be the 5 result of southward translation of San Antonio massif with respect to northern Masinloc massif (Yumul et al., 1998). Crustal thickness estimates, based on SW-NE transects of the northern Masinloc massif, are up to 9.5 km for the mantle section and 7 km for the lower crustal section. The fertile to moderately depleted Acoje mantle section is comprised of harzburgite and lherzolite with spinel Cr# [Cr/(Cr+Al)] ranging from 0.18 to 0.56, generally increasing towards the mantle-crust transition
- zone (Evans and Hawkins, 1989; Yumul, 1990). Harzburgites from the uppermost mantle section are strongly depleted in light and middle rare earth elements (LREEs and MREEs) with equilibration temperature and oxygen fugacity estimates of 730° C and 1.9 Alog(fO2) units above the FMQ buffer, respectively (Tamayo, 2001), From the residual mantle section, the mantle-crust transition zone passes through interlayered ultramafic cumulates and dunite to layered cumulate gabbronorite. The Acoje transition zone hosts podiform chromite deposits (Cr# = 0.71-0.77) and Ni-Cu sulfides both of which are PGE-
- bearing (Bacuta et al., 1990). Refractory transition zone dunites, characterized by spinel Cr# greater than 0.6, are interpreted 15 to be of cumulate origin (Abrajano et al., 1989). The Acoje Block lower crustal section is dominated by cumulate gabbronorites with olivine-clinopyroxene-orthopyroxene-plagioclase crystallization sequence. Coexisting plagioclase and $clinopyroxene \ in \ gabbronorite \ are \ calcic \ (anorthite \ content \ [Ca/(Ca+Na+K)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ magnesian \ (Mg\# \ [Mg/(Mg+Fe)] = 89-94) \ and \ (Mg\# \ [Mg/(Mg+Fe)]$ 0.80-0.87). In addition to petrological criteria discussed above, the arc affinity of Acoje Block has been demonstrated using
- 20 immobile element-based geochemical fingerprinting of volcanic and hypabyssal rocks, as well as constituent clinopyroxene (Yumul, 1996). On a regional scale, the ophiolite appears to have a domal structure with a north-south trending axis. The volcanic section of Acoje Block in northern Masinloc massif dips northwest and is unconformably overlain by middle to late Miocene sedimentary sequences

3. Volcanic geology

Based on observations around the Barlo massive sulfide mine, two volcanic units with distinct lithofacies are distinguished in the Acoje Block extrusive sequence-a dominantly pyroclastic lower basaltic andesite-andesite unit overlain by an upper boninite unit with minor boninitic basalts. Here and in the succeeding sections, we use the term boninite as per the recommended IUGS criterion (Le Maitre, 2002). Pillow lavas that qualify as boninite based on MgO and TiO2 contents but with < 52 wt% SiO₂ are classified as boninitic basalts, equivalent to the basaltic boninite of Reagan et al., (2015). Structural

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measurements of bedding planes in 136 locations constrain the gross structure of the crustal sequence, consisting of a NE-SW trending doubly-plunging anticline forming a dome structure and NW-plunging anticline (Fig. 2). Estimated thickness of the upper unit and lower volcanic units are 680 and 520 meters, respectively,

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South of Barlo mine, boninite dike swarms with conjugate intrusive directions change into a succession of submarine explosive volcanic deposits. In order of ascending stratigraphic level, the lower basaltic andesite-andesite unit consists of tuff breccia with block-sized pyroclasts, subaqueous pahoehoe lava flows with ropy wrinkles and stretched vesicles on lobe crust (Umino et al., 2002; Umino et al., 2000) (Fig. 3f), moderately welded agglutinate, welded scoria and spatter deposits

- 5 marginal to a NW-SE trending fissure vent, and an uppermost <u>subaqueous</u> fall-out deposit of glassy lapilli tuff (Fig. 3e). In places, tuff breccia is cut by NE-dipping boninite dikes. The dominant occurrence of flat pahoehoe lobes suggest the pre-existence of subhorizontal topography (Umino et al., 2002). The volcanic facies recognized in the upper section of the lower volcanic unit (agglutinate, scoria and spatter deposits surrounding fissure vent) are characteristic of intermittent <u>submarine</u> Strombolian to Hawaiian_style fountaining.(Head and Wilson, 2003).
- 10 NW-dipping boninite tuff breccia and pillow lavas of the upper unit (Fig. 3b) overlie hyaloclastite, spatter deposits and pillow lavas (Fig. 3c) of the lower basaltic andesite-andesite unit at an exposure north of Mt. Sol. A sill directly above lower unit pillow lavas has a whole-rock K-Ar age of 44.1 ± 3.0 Ma (Fuller et al., 1989). Pillow lavas of the upper and lower unit show distinct morphological features based on vertical (V) and horizontal (H) axes measurements after the method of Walker (1992). Pillow lavas of the lower unit are marked by lower aspect ratios (n=92, median H=0.53 m, median V=0.34
- 15 m) compared to boninitic basalts of the upper unit which are elongate (n=15, median H=0.74 m, median V=0.32 m). SW of Barlo mine, NE-dipping pillow lavas of the upper unit are overlain by middle-late Eocene radiolarian-bearing claystone (Schweller et al., 1984). Further southwest in a synclinal area with upright structures preserved, a ~30 meter-high boninite pillow volcano is recognized (Fig. 3d). It consists of flattened flow lobes in the summit with pillow lavas dipping downslope along its flanks. This volcanic construct is one of several discrete boninitic volcanic edifices rising above the lower unit.
- 20 Elsewhere, these pillow lavas are cut by SW-dipping dikes with glassy margins and overlain by glassy boninite lapilli tuff fall deposit (Fig. 3a).

4 Sampling and analytical methods

A total of one hundred fifty-two (152) samples were collected during the 2016 mapping campaign. For this study, a subset of forty-four (44) samples located along NW-SE transects of the Acoje Block volcanic sequence at Barlo were selected for
whole rock geochemical analyses and screened through visual and microscopic assessment of secondary alteration. Location of samples is shown in the geologic map in Fig. 2. Rock slabs were cut to remove altered surfaces. Initially, slabs were crushed coarsely to manually separate low temperature secondary minerals such as quartz and calcite. Coarse crushing step was followed by rinsing with deionized water and drying in an oven at 110 °C for at least 12 hours. Iron mortar and agate mill were used for fine crushing and grinding, respectively. Ignition loss is taken as the normalized weight lost after ignition
of ground rock powder at 900 °C for four hours. Major element compositions were determined by x-ray fluorescence (XRF) spectrometry using a PANanalytical Axios spectrometer at the Geological Survey of Japan/National Institute of Advanced Industrial Science and Technology. Glass beads were fused using a Tokyo Kagaku bead sampler (TK-4500) at 1:10 dilution

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ratio as mixtures of 0.5 g (±0.0002) rock powder and 5 g (±0.001) lithium tetraborate alkali flux (Li₂B₄O₇, Merck Spectromelt A10). Precision, measured as percent relative standard deviation (%RSD), is better than 2% based on repeated measurements of JB-2, BIR-1 and BHVO-2; accuracy is better than 2% as well. Trace element (REE, V, Cr, Ni, Li, Be, Rb, Sr, Y, Zr, Nb, Cs, Ba, Hf, Ta, Pb, Th and U) concentrations were determined by inductively coupled plasma mass

- 5 spectrometry (ICP-MS) using an Agilent 7900 instrument also at the Geological Survey of Japan, Samples were digested for at least 48 hours on a hotplate using screw top Teflon beakers with a mixture of HF and HNO3 at 5:1 ratio. Dissolution procedures were performed in a Class 1000 clean laboratory. <u>Twelve international standards (BIR-1, BRR-1, JB-2, JB-3, JA-</u> 1, JGb-1, JA-2, BCR-1, AGV-1, JB-1a, BHVO-2 and BE-N-1) were used to construct calibration lines. <u>Standard JB-2</u> and JB-3 solutions at similar dilution levels were used as external standards. Reproducibility is better than 2 % for REEs and
- 10 better than 3 % for the rest of the trace elements except for Ta (6.5 %) and Be (3.6 %). Mean percent error is generally within 5–10 % relative to the preferred values of Jochum et al. (2016) and Dulski (2001). Major element compositions of constituent mineral phases were determined with a JEOL-8800R electron microprobe at

Kanazawa University using 3 µm probe diameter, 20 nA probe current and 20 kV accelerating voltage. Natural (Kurose olivine and clinopyroxene) and synthetic mineral standards were used for calibration and data were corrected using the ZAF method.

5 Results

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Representative mineral chemistry data are listed in Supplementary Table 1. Results of whole-rock major and trace element analyses are given in Supplementary Table 2 with total Fe represented as Fe₂O₃. In succeeding discussions, all samples are normalized to 100 % volatile free with total Fe recalculated as FeO* to enable comparison with compiled datasets.

20 5.1 Textures and phase chemistry

Zambales high-silica boninite (as defined in 5.2) is mostly aphyric and consists of subhedral olivine microphenocrysts (Mg# $[Mg/(Mg + Fe^{2^+})] = 0.88-0.91$), abundant elongate enstatite microphenocrysts (Mg# = 0.86-0.89, Wo_{2.6-4.9}En₈₂₋₈₆) with augite \pm pigeonite overgrowth (Mg# = 0.67-0.84, Wo₃₁₋₄₄En₃₄₋₅₇), and chromian spinel (Cr# [Cr/(Cr+A1)] = 0.69-0.84, Mg# = 0.53-0.69) set in a glassy groundmass with feathery quench clinopyroxene (Figs. 4a-b). This assemblage corresponds to

25 Type II boninite of Umino (1986) in samples described from the type locality at Chichijima island, Ogasawara (Bonin) Archipelago in southern Japan. Unlike low-Ca, high-silica boninites from Ogasawara, clinoenstatite has not been identified so far. Comparative mineral chemistry of Zambales and Ogasawara boninite is shown in Fig. 5. Olivine, orthopyroxene and spinel compositions of Zambales and Ogasawara boninite are almost identical except for a limited range of olivine Mg#, which is most likely due to a small number of samples analyzed (two for olivine and four for spinel), and lower spinel Cr# in 30 Zambales boninite (Taylor et al., 1994; Umino, 1986; Yajima & Fujimaki, 2001). NiO contents of olivine range from 0.18–

0.41 wt% and plot on fractionation trends from the mantle olivine array. Subhedral olivine microphenocrysts in high-silica

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boninite (ZM2-100A) are zoned with magnesian cores. The range of spinel Cr# of Zambales boninite overlap with Acoje podiform chromitites and transition zone dunites. Enstatite microphenocrysts with sieve texture and reverse zoning are recognized together with embayed quartz xenocrysts (Figs. 4c-e). Reversely zoned enstatite has oscillatory-zoned magnesian rims (Mg# = 0.86-0.87) with resorbed and dissolved cores (Mg# = 0.68-0.77). Similar xenocrystic quartz and sieve-textured

- 5 enstatite have also been noted in Ogasawara boninite (Watanabe and Kuroda, 2000). Glassy olivine-phyric and olivineclinopyroxene-phyric low-silica boninite varieties are also present (Fig. 4f). Hypocrystalline boninite pillow lavas consist of olivine, usually altered and replaced by chlorite and carbonate minerals, subhedral clinopyroxene phenocrysts, elongate orthopyroxene and plagioclase microphenocrysts.
- Highly-phyric boninitic basalt has a phenocryst assemblage of olivine replaced by calcite, zeolite and clay minerals, euhedral
 to subhedral augite (Mg# = 0.82–0.90, Wo₄₁₋₄₃En₄₈₋₅₃) with oscillatory and sector zoning, and rare corroded enstatite (Mg# = 0.85–0.88, Wo₃En₈₂₋₈₄) with reverse zoning. Chromian spinel (Cr# = 0.62–0.71) occurs near phenocryst margins and as inclusions in olivine. <u>A peculiar zoning relationship of augite and enstatite demonstrates that some pyroxenes are xenocrysts</u> in nature. Groundmass consists of < 0.3 mm long plagioclase microlites with swallow-tail terminations, spherulitic clinopyroxene and altered interstitial glass totally replaced by clay minerals. Acicular zeolite occurs as vesicle fillings. We
 note that such textures have been described in samples reported as boninite by Evans et al. (1991).
- Lower-unit basaltic and esite and andesite pillows and lapili tuff are marked by the presence of abundant plagioclase laths as glomerocrysts and sparse cpx \pm opx microphenocrysts with two-pyroxene intergrowths in a glassy groundmass with plagioclase microlites and magnetite (Fig. 4g).

5.2 Whole-rock geochemistry

- 20 The Acoje Block extrusive section at Barlo is composed predominantly of basaltic andesite and andesite. Loss on ignition (LOI) values are generally between 4–7 wt% and total alkalis are < 3 wt% (Supplementary Table 2). Cs, Rb, K, Li, Ba and Mn_contents in some dikes and pillow lavas, especially those collected nearest to the massive sulfide mine, are highly scattered and plot away from general fractionation trends shown by the majority of samples. The rest of the samples show variations in the major element and trace element compositions consistent with fractional crystallization; thus, the</p>
- 25 geochemical characteristics, are deemed as primary. Pillow lava, tuff breccia and lapilli tuff of the upper volcanic unit and dikes from the uppermost section of the dike-sill complex are primitive with > 8 wt% MgO and < 0.5 wt% TiO₂, satisfying the IUGS criteria for boninite (Le Maitre, 2002), Following the classification of Kanayama et al. (2013) and Reagan et al. (2015) both high-silica (HSB) and low-silica (LSB) boninite subtypes are recognized (Fig. 6). This compositional division is consistent with petrographic observations, with HSB having olivine + orthopyroxene microphenocrysts and LSB with
- 30 olivine or olivine + clinopyroxene phenocrysts. Boninitic samples previously described at Barlo are actually from the dike section and lower section of the upper unit and with sampling limited to Balincaguin River and the periphery of Barlo VMS mine (Evans et al., 1991; Hawkins & Evans, 1983; Tamayo, 2001; Yumul, 1990). Besides the low-silica boninite and boninitic basalt samples from previous studies with altered equivalents having >3.5 wt % Na₂O+K₂O, 3–5 wt % LOI and

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possibly up to 4 wt% SiO₂ enrichment, this is the first report of pristine high-silica boninite in <u>the</u> Zambales ophiolite. Boninite from Zambales (n=14) have CaO/Al₂O₃ ratios less than 0.76, mostly within 0.63–0.71 with FeO*/MgO ratios <1 (Mg# values from 63 to 73). Ni and Cr contents are high, ranging from 72 μ g/g to 292 μ g/g and 145 μ g/g to 727 μ g/g, respectively. Boninitic basalts (n=3) are primitive as well with 12.4–13.6 wt% MgO and 712–1113 μ g/g Cr.

- 5 Pillow lava and lapilli tuff of the lower volcanic unit and dikes from the deeper section of the dike-sill complex are andesitic in composition with 55.5–63.5 wt% SiO₂, 2.4–7.1 wt% MgO and 0.31–0.47 wt% TiO₂ (Fig. 6). These andesites (n=26) are magnesian with Mg# mostly between 0.47 to 0.54, CaO/Al₂O₃ ratios < 0.63 and FeO*/MgO ratios > 1.16. Ni and Cr contents are low at 8–58 µg/g and 3–47 µg/g. MgO and SiO₂ compositions of lower unit volcanics and corresponding dikes lie above the island arc basalt (B)-basaltic andesite (BA)-andesite (A)-dacite (D) discriminant line of Pearce and Robinson (2010) and
- 10 follow a curvilinear fractionation trend from low-silica boninite; thus, is recognized here as forming a low-silica boninite differentiation series (Fig. 6). Dacitic pillow lavas represent the differentiated end of the LSB series. A gap between andesitic boninite-series volcanics with < 60 wt% SiO₂ and dacite (62–63 wt%) is present. A single dike (B-47 d4) that cross cuts the LSB-series dikes is the only sample with > 0.5 wt % TiO₂. Although the pervasive clay alteration of samples from the Barlo massive sulfide mine hinders the direct characterization of the host rocks, the massive sulfide deposit is most likely
- 15 hosted by the lower volcanic unit and possibly up to the lowermost section of the upper unit based on stratigraphic relationships and comparison of field and core descriptions (Paringit, 1977). The occurrence of massive sulfide-bearing horizons at Barlo is also consistent with massive sulfide mineralization in Oman, Troodos and Bonin Ridge which occur either below or within the boninitic volcanic section (Ishizuka et al., 2014; Umino et al., 2009).
- Compositions of Zambales boninite and boninite series volcanics are marked by low incompatible trace element abundances
 relative to MORB (Fig. 7). Extended trace element patterns arranged after Pearce and Parkinson (1993) show the dominance of slab-derived components with notable enrichment in fluid-mobile elements (Cs, Rb, Ba, U, K, Pb, Sr, Li, Na) and depletion of high field strength elements (Nb, Ta). Hf shows a positive anomaly relative to Sm while Ti shows a negative anomaly relative to Y. In contrast with samples from Ogasawara that exhibit characteristic U-shaped chondrite-normalized rare earth element (REE) patterns, Zambales boninite and boninite-series volcanics exhibit spoon-shaped REE patterns with
- 25 heavy REEs (HREE) at 5–15x chondrite values and weak negative Eu anomaly (Eu/Eu* = 0.9). Boninitic basalts are more depleted in middle and heavy REEs with just 3–5x the chondrite values. Excluding moderately altered samples, chondrite normalized La/Ce ([La/Ce]_N) ratios of Zambales and Ogasawara boninite are both <1. The range of [Sm/Yb]_N ratios, as measure of middle REE depletion relative to heavy REE, however, are distinct at 0.06–0.09 for Zambales and 0.02–0.05 for Ogasawara. The effect of fractionation in Zambales boninite samples is shown as increasing trace element abundances
- 30 relative to a glassy olivine-phyric LSB (ZM2-107) with 12.4 wt% MgO. The evolved dacitic samples does not differ significantly with just twice the normalized REE abundances of the most primitive sample. Trace element pattern of a single dike (B-47 d4) with > 0.5 wt% TiO₂ is visibly different with muted fluid-mobile element enrichment without a negative Ti anomaly, and smooth, concave downward LREE-depletion (Fig. 7).

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The volcanic sequence at Barlo, based on observed local stratigraphic relationships, is <u>LSB series volcanics-boninitic basalt-LSB-HSB from base to top</u> (Fig. 3). This volcanic stratigraphy is remarkably similar to Holes U1439 and U1442 of IODP Exp. 352. Both the Barlo extrusive section and Holes U1439 and U1442 are marked by abundance of LSB relative to HSB (Reagan et al., 2015). In contrast, the onshore boninite sequence at Ogasawara is dominated by 48-46 Ma HSB and HSB

- 5 series volcanics interbedded with minor LSB (Maruberiwan Formation), which are overlain by 45 Ma LSB and calc-alkaline andesite-dacite (Mikazukiyama Formation) (Kanayama et al., 2012; Taylor et al., 1994; Umino, 1986). Several dikes in the Subic crustal section in the southern San Antonio massif have compositions like Barlo boninitic basalts (Yumul et al., 2000), albeit their petrographic characteristics have not been described. This raises the possibility that boninite is present in other volcanic sections of the Acoje Block as well. The rest of the volcanic sections of the Zambales ophiolite (Coto volcanic
- 10 section in Tarlac, Sual dikes north of Barlo, and Subic dikes and pillow lavas in the south) are dominated by basalts and basaltic andesites with > 0.5 wt% TiO₂ that can be classified as moderate-Fe tholeiites (Supplementary Fig. 1). The tholeiite dike (B-47 d4) that cross cuts low-silica boninite dikes in the deeper dike swarm section at Barlo is similar in composition and correlatable with 44.0±3.0 Ma dikes in the Sual crustal section located NE of Barlo, suggesting that tholeiites are coeval or slightly younger than the Barlo extrusive section.

15 6 Discussion

6.1 Low-pressure fractionation and magma mixing

Early experimental studies of primitive boninite produced andesitic primary liquids and led to the initial characterization of boninite as a primary magma (Kuroda et al., 1978). Subsequent detailed petrologic analyses resulted in the recognition of the importance of low-pressure fractionation and magma mixing processes to account for compositional variation of boninite

- 20 and boninite series volcanics (Dobson et al., 2006; Taylor et al., 1994; Umino, 1986). Meijer (1980) introduced the concept of the boninite series to describe cogenetic bronzite andesite, dacite and rhyolite formed through differentiation of boninite, analogous to tholeiitic and calc-alkaline magma series. Figure 6 shows oxide vs. SiO₂ variation diagrams of Zambales boninite and boninite series volcanics. Also shown are boninite and boninite series volcanics from Ogasawara (Kanayama et al., 2012; Nagaishi, 2008; Taylor et al., 1994; Umino & Nakano, 2007; Yajima & Fujimaki, 2001) and Holes U1439 and
- 25 U1442 of IODP Expedition 352 (Haugen, 2017; Reagan et al., 2015). MgO, TiO₂, CaO and Al₂O₃ contents of HSB and LSB form subparallel trends suggesting distinct parental magma compositions and fractionation paths. Parental HSB is characterized by higher MgO, lower TiO₂, Al₂O₃ and CaO at a given SiO₂ than LSB. HSB is marked by the abundance of orthopyroxene and, in some localities, the presence of clinoenstatite. Umino (1986) describes the crystallization sequence at Chichijima as chromian spinel, olivine, clinoenstatite, enstatite and augite. HSB differentiation series is best exemplified by
- 30 samples from Ogasawara and Site 786, ODP Leg 125 (Arculus et al., 1992). Boninite from other notable Western Pacific localities such as the Mariana Trench (Bloomer & Hawkins, 1987; Dietrich et al., 1978), Cape Vogel, Papua New Guinea (Jenner, 1981; König et al., 2010), Nepoui, New Caledonia (Cameron, 1989; Cluzel et al., 2016) and Hahajima seamount (Li

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- 5 ODP Leg 60 (Meijer, 1982; Sharaskin, 1982). Other LSB and LSB series localities include Facpi Formation in Guam (Reagan & Meijer, 1984), Tonga Trench (Falloon et al., 2007; Falloon and Crawford, 1991), Troodos ophiolite (Cameron, 1985; König et al., 2008; Osozawa et al., 2012; Pearce & Robinson, 2010) and Oman ophiolite (Ishikawa et al., 2002; Kusano et al., 2017; Kusano et al., 2014; Nagaishi, 2008).
- Using spinel-hosted melt inclusions reported by Umino et al., (2017) as parental magma compositions, isobaric fractional crystallization paths of residual liquids at +1 Δlog(FMQ) are modelled using rhyoliteMELTS v.1.2.0 (Ghiorso and Gualda, 2015). The compositions of primary spinel-hosted boninitic melt inclusions from Ogasawara (2.1–3.4 wt% H₂O) and Guam (3.8 wt% H₂O) are in accord with experimental glasses formed by melting synthetic Troodos harzburgite and depleted Tinaquillo Iherzolite at 1.5 GPa (Falloon and Danyushevsky, 2000) (Fig. 6). The range of whole-rock HSB compositions can be reproduced by low pressure (1–2 kb) fractional crystallization of parental Ogasawara HSB magmas at lower water content
- 15 (1 wt%). TiO₂ and CaO contents of Zambales HSB are in the upper end of Ogasawara HSB compositions and alternatively can be produced by fractionation from a parental composition straddling between the HSB and LSB boundary of Reagan et al. (2015). Given a primary LSB magma composition identical to a spinel-hosted melt inclusion from Guam, low pressure (1–2 kb) fractional crystallization at lower water content (1 wt%) can likewise account for the major element variation of Zambales LSB and LSB series volcanics. The modelled early crystallization of cpx relative to opx and the onset of
- 20 plagioclase crystallization following cpx at ~57 wt% SiO₂ is reflected in the modal compositions of Zambales LSB and LSBseries volcanics (Fig. 4). The occurrence of differentiated LSB series volcanics at the base of the volcanic pile suggests that initial LSB magmas underwent protracted crystal fractionation in magma chambers at depth; influx of primitive HSB perturbed sub-volcanic magma chambers and likely induced magma mixing. The presence of reversely zoned enstatite microphenocrysts in Zambales HSB, attest to high-level magma mixing between an olivine+orthopyroxene saturated HSB
- 25 magma and a pre-existing LSB series andesitic magma. Zambales HSB deviate slightly from the modelled Ogasawara HSB fractionation paths and lie on a mixing line with potential HSB and LSB-series end member components having SiO₂ contents that do not vary significantly at 58 and 57 wt % respectively.

The markedly divergent fractionation paths followed by boninite and boninite series volcanics from Barlo and basalts from various volcanic sections of the Zambales ophiolite are shown in the MgO, TiO₂ and FeO*/MgO vs SiO₂ variation diagrams

30 (Fig. 6). Zambales LSB and LSB series volcanics follow fractionation paths of hydrous primitive arc magmas (Grove et al., 2012). Moderate-Fe tholeiitic basalts and basaltic andesites from the rest of the volcanic sections of Zambales ophiolite (Evans et al., 1991; Geary et al., 1989; Hawkins & Evans, 1983; Tamayo, 2001; Yumul, 1990) and proto-arc basalts from the Izu-Ogasawara-Mariana forearc (Arculus et al., 2015; Ishizuka et al., 2011; Reagan et al., 2010, 2015) exhibit large

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FeO*/MgO ratio variation at a restricted SiO_2 interval and define a slope steeper than the discriminant line of Miyashiro, (1974).

6.2 <u>Slab</u> contributions to a less depleted mantle source

Although there is a general consensus on boninite petrogenesis (Bedard, 1999; Crawford et al., 1989; Pearce et al., 1992), <u>namely that it involves shallow flux melting of refractory peridotite more depleted than MORB sources</u>, the requisite sub-arc conditions ideal for boninite generation span a range that may involve heterogeneous mantle wedge sources (depleted lherzolite to harzburgite) and addition of diverse slab-derived components (Cluzel et al., 2016; Kusano et al., 2017; Umino et al., 2015, 2017). We highlight the peculiar trace element characteristics of Zambales boninite; geochemical modelling will be presented elsewhere. Fig. 7 shows MORB and chondrite-normalized trace element patterns of Zambales boninite and

- 10 boninite-series volcanics together with boninite suites from Ogasawara, Holes U1439 and U1442 of IODP Expedition 352, Troodos and Oman ophiolites, Nepoui (New Caledonia) and Cape Vogel (Papua New Guinea). Boninite from Zambales, Oman and Troodos ophiolites all show spoon-shaped chondrite normalized REE patterns with heavy REEs that are 5–9 times chondrite values. Based on HREE abundances and spinel Cr#, Zambales boninite is derived from a mantle source that is less depleted than Ogasawara boninite (Fig. 7).
- 15 Superimposed on the depleted character of boninite are slab-derived components whose nature can be explored using normalized trace element diagrams and trace element ratios with varying compatibilities. The magnitude of hydrous fluid mobile element enrichment in boninite from Ogasawara, Zambales, Oman and Troodos ophiolites are comparable. LREE– enrichment is most prominent in Nepoui and Cape Vogel boninites. All boninite suites except for Nepoui and Cape Vogel have elevated Ba/Th ratios (Fig. 8a). Ba is strongly partitioned into aqueous fluids relative to Th, thus is a good proxy for the
- 20 addition of subduction components released at shallow depths sz1, Ba-only component of Pearce et al. (2005) (Keppler, 2017; Ribeiro et al., 2015). Increasing Ba/Th ratio is mirrored by increasing Sr/Nd ratio for Zambales, Oman and Troodos boninite (Fig. 8b). Ogasawara, Cape Vogel and Nepoui follow a vertical Th/Yb vector (Fig. 8b) towards sediments the component 1 of Elliott (2003). We interpret this decoupling as an indicator of increasing contributions from a deep subduction component that is usually characterized either as sediment melt and/or siliceous fluid (Pearce et al., 2005).
- 25 Zambales boninite and boninite-series volcanics have U/Th ratios (0.62–0.77) much higher than Cape Vogel boninites (0.20–0.29). <u>A high U/Th ratio is associated with saline and oxidizing fluids</u>, while low U/Th ratio point to transport of silica-rich fluids to source (Keppler, 2017). Zambales and Cape Vogel boninites both have a uniform La/Th ratio of 0.2. In the La/Th vs. Sm/La diagram of Plank (2005) (not shown), Zambales, Oman, and Troodos boninites lie on a mixing line between sediments with Th/La ratio of 0.30–0.35 and a mantle source more refractory than MORB with a Sm/La ratio of 1.75.
- 30 Ogasawara, Cape Vogel and Nepoui boninites may lie on the mixing line between sediments with Th/La ratio of 0.1–0.2 and a highly-depleted mantle source with Sm/La greater than 2. An alternative explanation is that low Sm/La ratios of Ogasawara, Cape Vogel and Nepoui boninites dominantly reflect LREE-enrichment that masks mixing relationship. The shared characteristics of Zambales, Oman, and Troodos boninites are further exemplified by subchondritic bulk Earth Nb/Ta

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5 volcanics, This is corroborated by enriched Pb and Sr isotopic compositions of Acoje Block uppermost crustal differentiates and cumulate ultramafic samples that do not show an inverse relationship with Nd isotopic compositions (Encarnación et al., 1999).

6.3 Subduction initiation origin of the Zambales ophiolite

- The 45 Ma crustal section of the Coto Block has a composition transitional to mid-ocean ridge basalt (MORB) and island arc
- 10 tholeiite (IAT), On the basis of immobile element-based trace element discrimination diagrams, Coto Block dikes and lavas from Tarlac are robustly characterized as distinct from Mariana Trough back-arc basin basalts (Geary et al., 1989). Decreasing REEs, TiO₂, Zr and Y in Coto volcanics, dikes in the Coto mantle section to Acoje volcanics document the progressive depletion of the mantle source in Zambales mantle wedge (Yumul, 1990). Succeeding zircon U-Pb geochronology established the 1–2 Myr, age difference between Coto and Acoje blocks (Encarnación et al., 1993). Thus,
- 15 combined geochemical and age constraints preclude a back-arc basin origin for Coto Block as forwarded by Hawkins and Evans, (1983), and are more compatible with a proto-forearc setting as originally suggested by Geary et al., (1989). <u>Coto Block volcanics (n=9)</u> have variable Ti/V ratios, mostly between 20-26 (Fig. 6f). Recognizing the small sample size, this range is higher than proto-arc basalts (PAB) and overlaps with MORB (Supplementary Fig. 2a); suggesting a less oxidized source compared to IBM PAB (Brounce et al., 2015; Hickey-Vargas et al., 2018; Mallmann and O'Neill, 2009). <u>The</u>
- 20 corresponding Ti₈ and Si₈ values of the Coto Block are comparable with proto-arc basalts from the Bonin Ridge, Mariana Trench slope, Hole U1430 of IODP Exp. 351 and Hole 1440 of IODP Exp. 352 (Arculus et al., 2015; Ishizuka et al., 2011; Reagan et al., 2010, 2015) (Fig. 9). Si₈ and Ti₈ values of Coto Block basalts are also distinct from Celebes Sea Basin and West Philippine Basin MORB (Pearce et al., 2005; Savov et al., 2006; Serri et al., 1991; Spadea et al., 1996; Zakariadze, 1981). In addition, the primitive mantle-normalized fluid immobile trace element abundances of Coto Block volcanics are
- 25 exceptionally low as well and more depleted compared to MORB (Hickey-Vargas et al., 2018) and Celebes Sea Basin basalts (Supplementary Fig. 2b). From observations in the field, oceanic accretion can be inferred from the Coto crustal section based on a well-developed sheeted dike complex, in contrast to the mutually intrusive dikes of the Acoje crustal section. With the recognition of boninite and boninite-series volcanics (Barlo), simultaneous and post-boninite moderate-Fe arc tholeiites (Barlo-Sual, Subic) in the crustal section of Acoje Block, it is apparent that the subduction initiation
- 30 chemostratigraphy (Ishizuka et al., 2011; Whattam & Stern, 2011) is present in <u>the</u> Zambales ophiolite as well. Although stratigraphic relationships of individual volcanic sections may be obfuscated by emplacement-related tectonic deformation, by considering available age constraints, the crustal sections of the Zambales ophiolite represent proto-arc accretion and

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juvenile arc sections supplied by separate magma plumbing systems as observed in IODP Exp. 352 sites (Reagan et al., 2015).

6.4 Doubly vergent subduction initiation

Previous attempts to ascertain the provenance of Zambales ophiolite focused on its characterization as a marginal basin and

- 5 thus linked it to the surrounding Eocene marginal basins such as the Celebes Sea Basin and Molucca Sea Basin. In the most comprehensive petrological and geochemical study of Philippine ophiolites, Tamayo et al. (2004) notes the strong subduction geochemical imprint of Zambales ophiolite and placed it as a marginal basin between Celebes Sea Basin and the western margin of the Philippine Sea Plate. The difficulty in reconstructing the Paleogene tectonic configuration of the southeast Asian region, specifically the spreading history of the oceanic domain east of Sundaland and west of the Philippine
- 10 Sea Plate, stems from the fact that this region has been fully consumed along convergent margins east of southeastern Eurasia (Zahirovic et al., 2014). This gap was addressed by Wu et al. (2016) through the novel use of structural unfolding of subducted slabs imaged by seismic tomography. In their preferred model (model 1b), unfolding of subducted slabs beneath southern Eurasia reveals that the Philippine Sea Plate was bordered by Cretaceous oceanic crust (the East Asian Sea) in its western margin. Paleomagnetic data derived from sites in the Philippine Sea Plate shows northward translation concomitant
- 15 with clockwise rotation (Hall et al., 1995; Richter & Ali, 2015; Yamazaki et al., 2010) (Fig. 10a). Eocene to Miocene sites from Luzon indicate similar northward migration; thus, the island can be regarded as the western promontory of the Philippine Sea Plate (Queano et al., 2007). The subequatorial position of Luzon during the middle Eocene is further supported by paleomagnetic data from pillow lavas forming the basement of Luzon Central Cordillera (K-Ar age: 45.49±2.27 Ma) that yield a paleolatitude of 6.3±3 °S (Queaño et al., 2009; Sajona, 1995). East of Luzon and forming the
- 20 greater part of Philippine Sea Plate, the West Philippine Basin (WPB) is characterized as a back-arc basin formed between opposing subduction zones (Deschamps and Lallemand, 2002). Pre-Eocene subduction south of WPB possibly generated the Mesozoic arc terranes in the Philippine Sea Plate including Amami Plateau and Daito ridge as well as the Cretaceous ophiolites in eastern Philippines (Lallemand, 2016). Initiation of a long-lived subduction east of WPB produced the Izu-Ogasawara (Bonin)-Mariana (IBM) forearc above a west-dipping subduction zone (Ishizuka et al., 2011). It is in this context that we re-evaluate the origin of Zambales ophiolite.

Tilt-corrected inclination data from sites in the 44 Ma Acoje Block equates to equatorial paleolatitudes (Fuller et al., 1991). Adopting southerly paleolatitudes in consonance with the modelled Luzon translation history of Wu et al., (2016), we make the case for the subduction initiation origin of the Zambales ophiolite in the leading edge of the northwestward-moving, clockwise-rotating Philippine Sea Plate. In this scenario, the origin of Philippine Sea Plate boninites (IBM and Zambales)
would be in a doubly-vergent subduction initiation setting (Fig. 10b). In contrast with IBM, incipient subduction that produced Zambales ophiolite proto-arc basalts, boninites and arc tholeiites within 3 Ma is rather short-lived and alludes to rapid emplacement. The timing of the proposed subduction initiation in the western margin of Philippine Sea Plate is yet to be constrained. The late Oligocene to early Miocene onset of true basinal sedimentation in the Central Valley Basin provides

a minimum docking age between Zambales ophiolite and eastern Luzon (Bachman et al., 1983). The GPlates-based reconstruction presented here, using unfolded slab data (Wu et al., 2016), is essentially similar to the Philippine Sea Plate evolutionary models of Deschamps and Lallemand (2003). Although it is beyond the scope of this study to identify the nature of a pre-existing structural boundary as a transform fault or strike-slip extension of a subduction zone, we note that

- 5 both Zambales and IBM are orthogonal to the West Philippine Basin spreading center and the Cretaceous arc terranes in the overriding plate. Contrary to a subduction initiation origin, Lallemand (2016) posits that proto-arc basalts (or forearc basalts) formed in short lived Eocene oceanic basins (e.g. Kita-Daito and Minami-Daito basins) bounded by the Cretaceous arc terranes. We believe that this might be the case for Eocene ophiolites in eastern Philippines (e.g. Angat ophiolite) but not for the Zambales ophiolite. The early Eocene age (48.1±0.5 Ma) of Angat ophiolite does not necessarily prove an affinity with
- 10 Zambales as suggested by Encarnación et al. (1993), rather it simply demonstrates that basement complexes in the Philippine Mobile Belt consists of juxtaposed geochemically distinct Eocene and Cretaceous ophiolites (ophiolite belts 1B and 1C of Tamayo et al., 2004) (Fig. 1a). Slightly older Eocene ages (e.g. 50.83±3.35 Ma) were obtained by subsequent whole-rock K-Ar and SHRIMP zircon U-Pb dating of ophiolitic samples from eastern Luzon (Sajona, 1995; Tani et al., 2015). In addition, these Cretaceous and Eocene ophiolites are associated with Eocene volcano-sedimentary arc sequences
- 15 (Caraballo, Maybangain, Anawan and Payo Formations)_(Billedo et al., 1996; David et al., 1997). This differs sharply with the deep-marine, isolated environment deduced from the Eocene sedimentary carapace of <u>the</u> Zambales ophiolite_(Schweller and Karig, 1982).

Although current terrestrial subduction is dominantly asymmetric, it is interesting to note that two-sided subduction is what is essentially produced in 2-D and 3-D models of mantle convection (Gerya et al., 2008; Wada & King, 2015). Likewise, we

- 20 discern that doubly-vergent SI geometry or subduction initiation with oppositely-dipping subduction zones along its margins is characteristic of 3-D thermo-mechanical models of plume-induced subduction initiation (Baes et al., 2016; Gerya et al., 2015). Plume-induced subduction initiation was first recognized by Whattam and Stern, (2015) to describe the temporal association of late Cretaceous plume-related oceanic plateaus in Central America that was followed by are volcanism with oppositely dipping slab dispositions. In the case of the Eocene Western Pacific, a mantle plume centered on the Manus Basin
- 25 had originally been invoked by Macpherson and Hall, (2001) to account for an inferred thermal anomaly in IBM boninite mantle source, implicitly connecting the initiation of the IBM arc to the presence of a mantle plume. Isotopic studies are in agreement that a mantle plume is likely associated with the oceanic plateaus and bathymetric highs emplaced at the same time as the opening of the West Philippine Basin (Hickey-Vargas et al., 2006; Ishizuka et al., 2013); however, thermal anomalies in excess of the ambient mantle are not reflected in mantle potential temperature estimates of proto-arc basalt
- 30 from the IBM forearc (Umino et al., 2017). A causative link between a mantle plume and the doubly-vergent SI configuration along Philippine Sea Plate margins is yet to be established. We speculate that the location of Philippine Sea Plate (PSP) in the nexus of Pacific, Indo-Australian and Eurasian plates and their long-term Cenozoic plate motion makes doubly-vergent subduction initiation along its margins feasible, The northwestward translation and clockwise rotation of the Philippine Sea Plate starting in the early Eocene had to be accommodated by the adjoining oceanic domain east of southerm

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Eurasia (e.g. East Asian Sea); hence, its interaction with the oceanic leading edge of the Philippine Sea Plate is expected (Wu et al., 2016; Zahirovic et al., 2016) and likely led to incipient subduction (Fig. 10). The dynamics of sustained doublevergent subduction is examined by Holt et al. (2017) but doubly-vergent subduction initiation is yet to be explored by numerical modelling. Field and petrologic data presented here demonstrate that models of subduction initiation based on the

- 5 IBM forearc are currently simplistic. Geodynamic models of subduction initiation should be based on robust geologic data and a doubly-vergent SI configuration based on the Zambales ophiolite provides another boundary condition for refined models of SI along PSP margins. We advocate that geodynamic models of subduction initiation along Philippine Sea Plate margins incorporate a pre-Eocene, <u>N/NE-dipping</u>, subduction zone (the proto_West Philippine Trench of Faccenna et al., 2010) associated with Cretaceous terranes forming the overriding plate, <u>doubly-vergent</u> subduction initiation along its margins as well as the interplay of plate forces and mantle upwelling (e.g. Oki-Daito plume of Ishizuka et al. 2013) during
- 10 margins as well as the interplay of plate forces and mantle upwelling (e.g. Oki-Daito plume of Ishizuka et al. 2013) during incipient subduction (Fig. 10b).

6 Conclusions

 The extrusive sequence in the juvenile arc section of Zambales ophiolite in Luzon Island, Philippines consists of boninite and boninite-series volcanics. This makes northern Zambales ophiolite the lone middle Eocene boninite locality in the Philippine Sea Plate apart from the Ogasawara Islands, Guam and their submarine equivalent in the IBM forearc.

2. Zambales boninite and boninite series volcanics are marked by low trace element abundances relative to mid-ocean ridge basalt with hydrous fluid mobile element enrichment and heavy rare earth element depletion comparable with boninite from Troodos and Oman. Trace element ratios suggest that aqueous fluids released from shallow sub-arc depths are the dominant slab components in Zambales boninite.

20 3. Subduction initiation stratigraphy deduced from diving, dredging and drilling programs in the Izu-Ogasawara (Bonin)-Mariana forearc is recognized in the Zambales ophiolite as well. The temporal variation from proto-arc basalt to boninite and post-boninite arc tholeiite is represented by the Coto (45 Ma), Barlo (44 Ma) and Sual-Subic (44–43 Ma) volcanic sections of Zambales ophiolite. Incipient subduction associated with the juvenile arc in northern Zambales ophiolite likely involved the unfolded East Asian Sea crust as subducting slab and the westernmost margin of Philippine Sea Plate as the overriding 25 plate.

4. By studying the Zambales ophiolite, it can be shown that subduction initiation is a plate-scale process. To address Eocene Western Pacific subduction initiation, it is essential to consider not just the Izu-Ogasawara-Mariana forearc but geologic constraints from the entire Philippine Sea Plate as well. Doubly-vergent subduction initiation deduced from the rock record is a testable mechanism that can be explored by geodynamic modelling.

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Data availability

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The data associated with this publication are available in the Supplement.

Supplementary Table 1: Representative microprobe analyses of olivine, pyroxene and spinel in Zambales boninite.

Supplementary Table 2: Whole rock compositions of boninite and boninite-series volcanics from northern Zambales ophiolite.

Supplementary Figure 1. Supplementary Figure 2.

Competing interests

10 The authors declare that they have no conflict of interest.

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Figure 1: (a) Distribution of subduction initiation rock suites in the Philippine Sea Plate and spreading histories of associated marginal basins (Deschamps and Lallemand, 2002; Pearce et al., 1992; Reagan et al., 2017). (b) Regional geological map of <u>the</u> Zambales ophiolite after Yumul et al., (1990) with whole rock K-Ar and zircon U-Pb ages from Fuller et al., (1989) and Encarnacion et al., (1993). The 44-43 MA Acoje Block consists of the northern Massifiand the southernmost San Antonio massif. <u>The enclosed region (red box)</u> is the Barlo extrusive section mapped in this study.

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Figure 2: Geological map of Barlo and environs. Closed symbols are samples with whole-rock geochemical data. Enclosed areas (panel A and B) are shown in Figure 3. Geologic cross-section is given along a NW-SE section line. Geologic column shows stratigraphic relationships and estimated thickness of the upper and lower volcanic unit. <u>Biostratigraphic</u>, whole rock K-Ar and zircon U-Pb ages are from Schweller et al., (1984) Fuller et al., (1989) and Encarnacion et al., (1993).

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Figure 3: Representative outcrops showing distinct volcanic facies and stratigraphic relationship of the upper boninite unit and lower basaltic andesite-andesite unit. (a) High-Si boninite lapilli tuff. (b) High-Si boninite pillow lava with fresh glassy margins. (c) Basaltic andesite pillow lavas cut by boninite dike. (d) Boninite pillow volcano. (e) Fissure vent with fluidal, flattened basaltic andesite-andesite spatter deposits and lapilli tuff. (f) Basaltic andesite pahoehoe flow with ropy wrinkles (inset) and transverse spreading cracks on lobe crust indicating flow direction. Rock hammer for scale (0.42 m). Orientation poles of bedding planes (dip azimuth) are shown in lower hemisphere stereographic projection (Vollmer, 2015). Bedding plane orientations in GoogleEarth (Digital Globe, CNES/Astrium) images, viewed from southeast direction, are visualized using the S2K macro (Blenkinsop, 2012).





Figure 4: Microphotographs showing textures of Zambales boninite and boninite-series volcanics. (a) High-silica boninite (HSB) with altered olivine and abundant elongate enstatite microphenocryts with augite rims (plane polarized-light). (b) Characteristic feathery clinopyroxene in groundmass glass, HSB (plane polarized-light). (c) Oscillatory zoning in magnesian rim of enstatite 5 xenocryst with dissolved core, HSB (BSE). (d) Reverse zoning in enstatite xenocryst with resorbed core, HSB (BSE). (e) Embayed quartz xenocryst, HSB (crossed polars). (f) Low-silica boninite (LSB) with fresh olivine phenocrysts in glassy groundmass (plane polarized-light). (g) Plagioclase, othropyroxene and clinopyroxene microphenocrysts in LSB-series basaltic andesite tuff breccia matrix (plane polarized-light).

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Figure 5: Comparative mineral chemistry of constituent phases (olivine, orthopyroxene, clinopyroxene and spinel) in Zambales and Ogasawara boninites. Mantle olivine array is from Takahashi, (1986).











Figure 7: Extended MORB and chondrite normalized trace element patterns. Normalizing values are from Barrat et al. (2012) and Gale et al. (2013).



Figure 8: Trace element ratios discriminating the nature of slab-derived components in boninite and boninite-series volcanics. Zambales, Oman and Troodos boninites have distinct LILE and HFSE enrichments compared to Ogasawara, Cape Vogel and Nepoui. Data sources: Ogasawara spinel-hosted boninitic melt inclusions (Umino et al., 2017), altered oceanic crust (AOC) fluids,
melts and sediments (Carter et al., 2015), aqueous fluids released at shallow depths (Ribeiro et al., 2015), subducting sediments and GLOSS II (Plank, 2014). Chondritic bulk Earth Nb/Ta and Zr/Hf ratios are from Munker et al., (2003). DMM and MORB compositions from Jenner and O'Neill, (2012) and Workman and Hart, (2005), respectively. Symbols are the same as in Fig. 6 except those shown in the diagram.



10 Figure 9: 8wt% MgO-normalized SiO₂ (Si₈) and TiO₂ (Ti₈) values of the crustal sections of Zambales ophiolite and IBM forearc showing temporal evolution from proto-arc basalt, boninite to arc tholeitte associated with subduction initiation. Also shown are Si₈-Ti₈ values of other notable boninite localities (Oman, Troodos, Tonga Trench). MORB and IAB fields are from Pearce and Robinson (2010). Data sources are given in the text. Symbols are the same as in Fig. 6 except those shown in the diagram.





Figure 10: (a) Paleolatitudes derived from Philippine Sea Plate, Luzon and Zambales inclination data (Fuller et al., 1991; Hall et al., 1995; Queano et al., 2007; Queano et al., 2009; Richter and Ali, 2015; Yamazaki et al., 2010) (b) Tectonic reconstruction of the western Pacific region at 44 Ma using GPlates (Boyden et al., 2011) and unfolded slab data, plate polygons and rotation file (model 1b) of Wu et al. (2016). Inverted triangles, diamonds and squares previously shown in Fig1b. refer to Cretaceous arc terranes in northern Philippine Sea Plate and Cretaceous-Eocene ophiolites in eastern Philippines, respectively.

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Supplementary Figure 1: FeO*-MgO diagram characterizing basalts and basaltic andesites from volcanic sections of the Zambales ophiolite apart from Barlo (Coto volcanic section in Tarlac, Sual dikes north of Barlo, dikes in the Coto Block mantle section, and Subic dikes and pillow lavas in the south) as moderate-Fe tholeites. Moderate and high-Fe boundaries after <u>Pearce and Robinson</u>, 5 (2010). Data sources: Zambales ophiolite dataset (Evans et al., 1991; Geary et al., 1989; Hawkins and Evans, 1983; Tamayo Jr., 2001; Yumul Jr., 1990).



Supplementary Figure 2: (a) Ti/V ratio histograms of proto-arc basalts, MORB and Coto Block volcanics (b) Primitive mantle normalized immobile element abundances of IBM proto-arc basalts (PAB) and Coto Block volcanics are similarly depleted
 compared to MORB. Data sources: IBM PAB (Haugen, 2017; Hickey-Vargas et al., 2018; Ishizuka et al., 2011, 2018; Reagan et al., 2010), MORB (Inner and O'Neill, 2012), all MORB mean (Gale et al., 2013), Coto volcanic section (Geary et al., 1989) and Celebes Sea Basin (Spadea et al., 1996).

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