se-2019-53 - "**The acid-sulfate zone and the mineral alteration styles of the Roman Puteolis (Neapolitan area, Italy): clues on fluid fracturing progression at the Campi Flegrei volcano**" by Monica Piochi et al.

Dear Editor Kei Ogata,

we are submitting the se-2019-53 manuscript reviewed following the further indications by the review #2.

All the issues raised by reviewer has been considered and the text has been revised accordingly.

Below you will find our author's response and author's changes into manuscript for each comment from Referee.

We applied some editing to improve English and these are listed from page 7 to the end.

The uploaded files include a copy of both the new text and supplement with changes marked (in yellow for response to reviewer, and cyan for our editing), in order to facilitate your work.

We hope that the manuscript is now suitable for publication on Solid Earth.

Thank you very much for handling the manuscript.

Sincerely
Monica Piochi and co-Authors

Response to Anonymous Referee #2

Dear Reviewer,

thank you for your additional work and attention that improve our research.

We have considered all the issues you raised and we have reviewed the text of manuscript following your indications.

Here, you will find our reply to the three specific comments and related changes at specific lines and pages from the annotated pdf.

We hope that the manuscript will be now considered suitable for publication into SE.

Best regards
Monica Piochi and co-Authors

Specific points
1. there are XRD traces or patterns not spectra. There is no smectite present in figure S1f. The diffraction maxima at 25.5 °2θ and 30 °2θ belong to alunite (the authors should look carefully at the reference XRD pattern of alunite in S1e). The hump at low angle (~3 °2θ) corresponds to a d-spacing of ~30 Å. Such a d-spacing is not rational for this type of materials because it is indicative of a superstructure. Such a phase which corresponds to certain types of clay minerals is highly unlikely to form in this environment. The hump at this low angle is an artifact (i.e. continuous radiation). Also, in Fig. S1d all samples contain alunite. Illite/mica might be present only in sample MP-24-09-14 (Fig. S1d). I suggest that the authors look at their XRD traces again and modify the text accordingly.
Reply.
Spectra changed into patterns (line 42 pag 3; line 12 pag 8; line 24 pag 12; title of chapter 1 in the supplement) or traces (line 26 pag 4; line 21 pag 14; caption in Table S1; caption in Fig S1; words at various lines within the chapter 1 of the supplement).

The patterns in figure S1c-f are have been selected to show the multiphase samples, in addition to the clays attribution you asked in the first round of revision.
Smectites are now not reported for our samples. We agree that the peak at the low angle in Fig. S1e is very small and can be also considered an artefact, but it occurs in some (although few, as listed in Table S1) samples. In effect we initially did not considered the montmorillonite (see firstly submitted Table 3). However, the literature describes montmorillonite and illite/montmorillonite in the area. So, we want be careful and comprehensive, and, in agreement with your indication in the first revision, we prefer citing the possible occurrence of these clays, at least in the supplement. Specific studies on clays should be of interest and we hope to have the possibility to solve this issue in the future by adopting adequate sample preparation and additional techniques that were beyond our purposes and were not considered in this context.

Fig. S1d. Yes, all samples contain alunite; alunite dominates in these mud samples as also derived by FTIR data on Fig S2d. These muds restituted similar XRDP traces with additional peaks that can be attributed to the illite.

Based on your notes and our results, we modified the text and the caption as it is listed below.

Caption of Table S1 – supplement.
"Representative spectra are in Fig. S1."
changed to
"Selected traces are in Fig. S1."

Table S1 – Montmorillonite is indicated as phase to be validated.

Caption of Table S2 – supplement.
Montmorillonite has been erased.

At the first page of chapter 1 – supplement
"Figure S1 displays representative XRDP spectra obtained on different samples and the peaks intensity of the identified phases. Each panel allows appreciating the comparable patterns obtained from different samples collected at the same sites. In particular, the panels S1a,b show the well-defined XRDP traces obtained on exsiccated water pool samples. The remaining panels illustrate the clays attribution to illite (S1d), kaolinite (S1e) and montmorillonite (S1f) in muds. The clays are usually present in low abundance in the analyzed samples; the La Fangaia muds (Fig. S1c) are the poorest of clays. The presence of the different clay minerals (i.e. illite, montmorillonite and kaolinite) has been established in consideration of the specific reflection lines."

changed to

"Figure S1 displays selected XRDP traces obtained on different samples and the peaks intensity of the identified phases. Each panel allows appreciating the comparable patterns obtained from different samples collected at the same sites. In particular, the panels S1a,b show the well-defined XRDP traces obtained on exsiccated water pool samples abounding in mascagnite. The remaining panels illustrate the complex X-ray patterns and the clays attribution to illite (S1d), kaolinite (S1e) and, possibly, montmorillonite (S1f) in muds that contain alunite dominating on amorphous, feldspar, and other phases (e.g., sulfur, pyrite; Table S1). The clays are usually present in very low abundance in the analyzed samples; the La Fangaia muds (Fig. S1c) are the poorest of clays. The presence of the different clay minerals has been established in consideration of the specific reflection lines, although the both low abundance and sample complexity produced results requiring validation by adequate studies, particularly for montmorillonite (Fig. S1f and Table S1), the occurrence of which is however suggested by literature (Caprarelli et al., 1997; Valentino and Stanzione, 2003; 2004)."

"The XRDP spectra of the Solfatara new pool muds clearly indicate kaolinite and this is an important difference with the other muds containing smectites, and with most of the other samples…"

changed to

"The XRDP traces of the Solfatara new pool muds clearly indicate kaolinite and this is an important difference with the other muds, and with most of the other samples…".

At the beginning of the second page in chapter 1 – supplement
"….(2002), it is possible to reveal three strong bands (i.e., 1092-1097, 1026-1029, 1008-1012; Table S2) between 1100 and 1000 cm^{-1} (Fig. S2e) that lacks in the smectites-bearing muds …"

changed to

"…(2002), it is possible to reveal three strong bands (i.e., 1092-1097, 1026-1029, 1008-1012; Table S2) between 1100 and 1000 cm^{-1} (Fig. S2e) that lacks in the smectites-bearing materials …".

"Otherwise, the DRIFT-FT-IR spectra are not resolute to determine the illite or montmorillonite contribution, particularly when present in low amount into mixtures of clays (Madejova et al., 2002), as in our case. We observe that the Pisciarelli smectite-bearing samples have a few band/hamp at around 3620 cm^{-1} (Fig. S2d) and this band is stronger in illite with respect to montomorillonite (Clark et al., 2007). However, the illite should produce signals between 4260 and 4090 cm^{-1}. These signals lack also in the infrared spectra of our Pisciarelli mud samples, probably due their small amount of smectites. Nevertheless, in absence of details on clays, we cannot exclude that the flat infrared traces in this region can be indicative of the montmorillonite or the illite/montmorillonite interlayer that characterize several samples (Fig. S1g). The Solfatara muds (Fig. S2c) produced infrared spectra very similar to those of smectites-bearing samples, and in particular display the band at 3620 cm^{-1}. Although the XRDP spectra of these muds do not present smectites-related peaks (Fig. S1c), the infrared spectra suggest further evaluations."

changed to

"Otherwise, the DRIFT-FT-IR spectra are not resolute to determine the illite or montmorillonite contribution, particularly when present in low amount into mixtures of clays (Madejova et al., 2002), as in our case. We observe that the Pisciarelli samples have a few band/hamp at around 3620 cm^{-1} (Fig. S2d) and this band is stronger in illite with respect to montomorillonite (Clark et al., 2007). However, the illite should produce signals between 4260 and 4090 cm^{-1}. These signals lack also in the infrared spectra of our Pisciarelli mud samples, probably due their small amount of clays. The Solfatara muds (Fig. S2c) also produced infrared spectra very similar to those of Pisciarelli clays-bearing samples, and in particular display the band at 3620 cm^{-1}, although the XRDP traces of these muds do not present smectites-related peaks (Fig. S1c). Nevertheless, in absence of exhaustive investigations on clays, we cannot exclude that the flat infrared traces in this region can be indicative of the montmorillonite or the illite/montmorillonite interlayer that are described in the literature (Caprarelli et al., 1997; Valentino and Stanzione, 2003; 2004) and we suspect to be present in some samples (Fig. S1g; Table S1)."

At the end of the second and the beginning of the third page in chapter 1 – supplement

"The XRDP and FT-IR investigations on evaporates are very interesting when compared with their cognate muds. Based on XRDP spectra, NH_4-bearing species that characterize waters, seem absent in most of these muds. The muds decanted from the Pisciarelli pools are dominantly mixtures of alunite, pyrite, amorphous, variable amount of primary feldspars and illite/montmorillonite;"

changed to

"The XRDP and FT-IR investigations on evaporates are very interesting when compared with their cognate muds. Based on XRDP traces, NH_4-bearing species that characterize waters, seem absent in most of these muds. The muds decanted from the Pisciarelli pools are dominantly mixtures of alunite, pyrite, amorphous, variable amount of primary feldspars and illite;"

Caption Fig. S1 – "Figure S1 – Selected XRDP traces of NH_4- sulfates dominating the assemblage formed from drying the Pisciarelli water (a, b) and of various multiphase muds from Solfatara (c,e) and Pisciarelli (d,f). Each panel reports the sample name in Table S1. The muds show the large hump between 18 ° and 30 ° 2θ degree attributed to the amorphous phase and the signals from the dominant alunite, feldspars (except MS new 9/17) and other minor phases (sulfur, pyrite and clays; Table S1). The smaller panels evidence the reflection intensity in the most significant range useful to discriminate illite in d) and kaolinites in e). Some XRDP trace has a corresponding infrared spectrum in Fig. S2: the sample with mascagnite is the same of Fig. S2a, the Solfatara muds in c) produced the FT-IR spectra in Fig. S2b, the Pisciarelli mud MP 6_16 is in Fig. S2d, the samples in e) are the same of Fig. S2e. Abbreviations (c, f): S = Sulfur; Al = Alunite; Kfd =Alkali feldspar. Note: montmorillonite needs of validation by specific studies, as the very small signals (Fig. S2f)."

References – supplement Updated

2. FTIR results. The interpretations in the NIR region are erroneous. The bands in the area 4520-4610 cm^{-1} are combination modes i.e. neither OH-stretching nor OH-bending modes. Both bands in this region belong to kaolinite not alunite. This can be easily observed if one combines the OH-stretching modes at 3695 cm^{-1} and 3620 cm-1 with the bending mode at 915 cm-1. There is no solid evidence for the presence of other clay minerals in the FTIR spectra provided. On the other hand alunite is identified positively by the band sharp at ~3500 cm-1 as the authors correctly mentioned. The main text and the supplement should be modified accordingly.

Reply.

Done. See changes at pag. 5 of the main text that you have indicated and that are listed in the following point 4.

Table S2 – the bands in the area 4520-4610 cm^{-1} attributed to kaolinite.

3. The use of term realm: The definition provided by the authors for realm in the text is identical to what is known as alteration zone. I am not convinced that it should be used.
Reply.
realm replaced by sub-zone (line 37 pag 6; lines 4, 6, 9, 11, 26, 30, 36 pag 7; lines 4, 18 pag 8; line 4 pag 9; line 4 pag 10, line 30 pag 11)

4. A few minor points have been highlighted in the attached script.
Reply:

Lines 25-30 – pag 4.
Accepted; we also changed spectra into traces. The sentence
"Clays have a low relative abundance in the studied samples (Supplement). They are mostly smectite, and most common are illite/montmorillonite, while kaolinite seems least abundant (Table S1, Table 1), as derived by the XRDP spectra (see Fig. S1c,d,e) and supported by DRIFT-FT-IR study (see below; Fig. S2 and Supplement). In particular, the infrared technique is suitable to detect the kaolinite and the related bands in the OH-region, in agreement with Madejová et al. (2002). Illite/montmorillonite usually occurs in the muds at Pisciarelli (from geyser and around other emissive vents) and occasionally at Solfatara (Table 1, S1)."

is now changed into:

"Clays have a low relative abundance in the studied samples (Supplement). They are mostly kaolinite and illite (Table S1, Table 1), as derived by the XRDP traces (see Fig. S1c,d,e) and supported by EDS-BSEM and DRIFT-FT-IR study (see below; Fig. S2 and Supplement). In particular, the infrared technique is suitable to detect the kaolinite and the related bands in the OH-region, in agreement with Madejová et al. (2002). Illite usually occurs in the muds at Pisciarelli (from geyser and around other emissive vents) and occasionally at Solfatara (Table 1, S1)."

Lines 33-35 – pag 5.
Accepted and the sentence
"Accordingly, the spectra show vibrations at ca. 4605 cm^{-1} ascribed to the Al-OH stretching contribution in alunite; furthermore, it is possible recognizing the OH-deformation at ca. 915 and 938 cm^{-1}, the Si-O stretch at 1008 and 1026 cm^{-1}, and the Al-OH bending contribution at 4523 cm^{-1} from kaolinite."

is now changed into:

"Accordingly, it is possible recognizing the OH-deformation at ca. 915 and 938 cm^{-1}, the Si-O stretch at 1008 and 1026 cm^{-1}, and the Al-OH modes at ca. 4605 cm^{-1} and 4523 cm^{-1} from kaolinite.".

Lines 39-41 – pag 5.
Accepted and the sentence below has been erased:
"The IR spectra of samples for which XRDP confirmed illite/montmorillonite (Fig. S1, Supplement) lack of overtones at 4255 and 4081 cm^{-1}. Following the literature (Clark et al., 1990; Madejová; 2003), these overtones are characteristics of illite and their absence can support the presence of montomorillonite.".

Lines 4-5 pag 7
"Based on the presented dataset, we propose the existence of major realms, in which some (minor/peculiar) mineral phases can appear or disappear, in response to changing physical-chemical conditions mainly associated to weather circumstances, mostly humidity and water abundance. The realms are the geographical zones discriminated by their dominant and repetitive…"

changed to

"Based on the presented dataset, we propose the existence of major alteration sub-zones, in which some (minor/peculiar) mineral phases can appear or disappear, in response to changing physical-chemical conditions mainly associated to weather circumstances, mostly humidity and water abundance. These sub-zones are discriminated by their dominant and repetitive…"

Line 17 – pag 7.

Accepted; montmorillonite has been erased and the sentence is now:
"We recurrently detected an enrichment in pyrite, illite and feldspar at Pisciarelli and in native sulfur at Solfatara.".

Other changes in relation to your comments.

Line 30 – pag 8.
Montmorillonite has been erased and the sentence is below:
"Meteoric and surface waters can dilute the aggressive endogenous fluids determining alteration degree conditions low enough for the generation of illlite, or other clays (Pirajno, 2008) at Pisciarelli.".

Line 39 – pag 8.
Montmorillonite has been erased and the sentence below has been erased:
"Alunite plus kaolinite form in steam-heated environments at….".

Lines 8-9 – pag 9.
"in intermediate argillic alteration zones (Pirajno, 2008) and their widespread occurrence in the various studied realms is among the results that contrasts this high sulfidation environment."
changed to
"in intermediate argillic alteration zones (Pirajno, 2008) and their widespread occurrence in the various studied sites, as well as in the local subsurface (Valentino and Stanzione, 2003; 2004), is among the results that contrasts this high sulfidation environment."

Table 1 - Caption.
"Table 1 – Main hydrothermal minerals detected by XRPD with related ideal chemical formula and sites of occurrence (name as in Fig. 1a,d,e). The complete set of minerals is in Table S1."
changed to
"Table 1 – Main hydrothermal minerals detected by XRPD with related ideal chemical formula and sites of occurrence (name as in Fig. 1a,d,e). The complete set of minerals is in Table S1. Refer to the supplement for details."

Table 3 - Caption.
"Table 3 – Summary of the mineralogical and isotopical features at the acid sulfate area following Rye et al. (1992); Hedenquist and Lowerstern (1994). *highest ^{34}S and ^{18}O during bacteriogenic reduction of sulfates with maximum fractionation in dry-wet alternating conditions. ^ $\delta^{34}S$ reflects the H_2S/SO_2 and temperature of fluid. 1 always present, 2 may be associated. 3 from Valentino and Stanzione (2003; 2004), Gresse et al. (2017). 'halloysite is indicated in Montanaro et al. (2017) and included here for completeness."
changed to
"Table 3 – Summary of the mineralogical and isotopical features at the acid sulfate area following Rye et al. (1992); Hedenquist and Lowerstern (1994). *highest ^{34}S and ^{18}O during bacteriogenic reduction of sulfates with maximum fractionation in dry-wet alternating conditions. ^ $\delta^{34}S$ reflects the H_2S/SO_2 and temperature of fluid. 1 always present, 2 may be associated. 3 from Valentino and Stanzione (2003; 2004), Gresse et al. (2017). 'halloysite is indicated in Montanaro et al. (2017) and included here for completeness. "Montmorillonite needs of specific validation (supplement) and is reported considering its detection in the local subsurface (Rosi and Sbrana, 1987; Valentino and Stanzione, 2003; 2004)."

Table 3
At the "montmorillonite" line we indicated possible (very rare) into "Solfatara" column and possible (rare) into "Pisciarelli" column

Other minor changes to improve the English

Line 10 pag 1

"mineralization"

changed to

"mineralizations"

Line 24 pag 1

"settings"

changed to

"setting"

Lines 3-6 pag 2

"By contrasts several studies relate to bradyseism phenomena that concern with seismicity, ground deformation and outgassing (e.g., Corrado et al., 1976; Barberi et al., 1984; Chiodini et al., 2016; Cardellini et al., 2017; Moretti et al., 2017), life in these environments (e.g., Zillig et al., 1996; Glamoclija et al., 2004; Sgavetti et al., 2008), and a continuous interest into the thermal bath and medical care…."

changed to

"In contrast, several studies relate to bradyseism phenomena addressing the various aspects of seismicity, ground deformation and outgassing (e.g., Corrado et al., 1976; Barberi et al., 1984; Chiodini et al., 2016; Cardellini et al., 2017; Moretti et al., 2017), life in these environments (e.g., Zillig et al., 1996; Glamoclija et al., 2004; Sgavetti et al., 2008), and a continuous interest into the use of hydrothermal products as thermal bath and for medical care …"

Lines 9-10 pag 2

"Results derive from temperatures determination contextually to sampling, investigations…."

changed to

"Results derive from temperature determinations contextually to sampling, and investigations"

Lines 18-19 pag 2

"The Solfatara volcano (Fig. 1a,b,c) displays impressive and powerful hydrothermal activities with hot fumaroles, thermal springs, mud pools and diffuse degassing…"

changed to

"The Solfatara volcano (Fig. 1a,b,c) exhibits impressive and powerful hydrothermal activities with hot fumaroles, thermal springs, mud pools and diffuse outgassing …"

Line 26 pag 2

"), that is slowly on-going"

changed to

"), that are slowly on-going"

Line 29 pag 2

"The intense mining in Roman…"

changed to

"Intense mining during Roman…."

Line 36 pag 2

"Medieval periods"

changed to

"Medieval time"

Lines 39-40 pag 2

"springs, while somes ("de Pisis" and "Sprudel" springs in the Terme of Agnano; Fig. 1a) disappeared, although high temperatures can be still…."

changed to

"springs, while some ("de Pisis" and "Sprudel" springs in the Terme of Agnano; Fig. 1a) disappeared. Yet, high temperatures can be still…"

Line 11 pag 3

"a detectable (very low) abundance of gaseous SO_2"

changed to

"a detectable (yet very low) abundance of SO_2"

Lines 17-20 pag 3

"$\delta^{34}S$ values determined for shallow subsurface sulphur-bearing minerals range between −5.5 to 0.0‰, while the deep-seated pyrite has values from 3.3 to 7.4‰ (Piochi et al., 2015). $\delta^{18}O$ values for alunite are from 4.2 to 7.0‰ (Piochi et al., 2015). The pH of water pools and soils is neutral to acid, with values…"

changed to

"$\delta^{34}S$ values determined for shallow subsurface sulfur-bearing minerals range between −5.5 and 0.0‰, while the deep-seated pyrite shows values from 3.3 to 7.4‰ (Piochi et al., 2015). $\delta^{18}O$ values for alunite vary from 4.2 to 7.0‰ (Piochi et al., 2015). The pH of water pools and soils is neutral to acid, with pH values"

Line 25 pag 3

"and the possible anthropogenic contamination. This study intends to enlarge"

changed to

"and thus a possible anthropogenic contamination. This study enlarges"

Line 29 pag 3

"the observation period on the Puteolis sulfate"

changed to

"the observation period for the Puteolis sulfate"

Line 31 pag 3

"including degassing "magnitude""

changed to

"including outgassing "magnitude""

Line 34 pag 3

"on the field"

changed to

"in the field"

Line 36 pag 3

"order to define"

changed to

"order to assess"

Line 38 pag 3

"were crush"

changed to

"were pulverized"

Line 41 pag 3

“Appendix A reports details for analytical techniques.”

changed to

“Appendix A provides detailed information about analytical techniques.”

Line 5 pag 6

“sulphur-bearing phases at the different locations, reveal dominantly negative”

changed to

“sulfur-bearing phases at the different locations reveal a dominantly negative”

Line 10 pag 6

“Pisciarelli also include”

changed to

“Pisciarelli include”

Line 12 pag 6

“heavy oxygen, with except for the 2013 - 2014 data for”

changed to

“heavy oxygen isotopes, except samples from 2013 - 2014 for”

Line 17 pag 6

“5) a likely appearance of positive correlation between S isotopes from pyrite and sulfate”

changed to

“5) a likely appearance of a positive correlation between S isotope results for pyrite and for sulfate”

Line 25 pag 6

“and/or native S in”

changed to

“and/or the presence of native S in”

Line 27 pag 6

“Carbon is generally low (< 1.25 %), and always < 22 %. Notably, SiO_2”

changed to

“Carbon is generally low (< 1.25 wt%), and always < wt 22 %. Notably, the SiO_2”

Line 28 pag 6

“Also, MnO”

changed to

“MnO”

Line 33 pag 6

“mostly lies”

changed to

“mostly lie”

Lines 4-6 pag 7

“phases can appear or disappear, in response to changing physical-chemical conditions mainly associated to weather circumstances, mostly humidity”

changed to

“phases appear or disappear, in response to changing physical-chemical boundary conditions mainly associated to weather circumstances, i.e., mostly humidity”

Line 16 pag 7

“The mud is beige and fine”

changed to

"The mud is beige and fine grained"

Line 22-23 pag 7

"A slightly cooling can be also supposed by our measurements (Table S1) with respect those in the literature"

changed to

"A slight cooling is discernible when comparing our data (Table S1) with those in the literature"

Line 32 pag 7

"91 °C is the temperature on April 2019"

changed to

"a temperature of 91 °C was measured in April 2019"

Line 33 pag 7

"reminishent"

changed to

"reminiscent"

Line 37 pag 7

"relative"

changed to

"relatively"

Lines 4-3 pag 8

"important factors that influence the neogenesis of the alunite-dominated sub-zones."

changed to

"important factors affecting the mineral neogenesis at the alunite-dominated sub-zones."

Line 7 pag 8

"The NH_4^+ ions were present in the solution"

changed to

"The NH_4^+ ions were present in solution"

Line 26-28 pag 8

"Furthermore, periods of intense rainfall determine the areal extent and depth of the mud pools, the generation of secondary mud vents and the erosion in Pisciarelli and its periodic water puddle. Sicardi (1959) also described"

changed to

"Furthermore, periods of intense rainfall determine the timing areal extent and depth of the mud pools, as well as the generation of secondary mud vents and the erosion in Pisciarelli and its periodic water puddle. Sicardi (1959) already noted"

Line 7 pag 9

"support the high sulfidation - magmatic hydrothermal environment (Rye et al., 1992)."

changed to

"support the classification as high sulfidation - magmatic hydrothermal environment (Rye et al., 1992)."

Line 11 pag 9

"of the lower temperature (< 40 °C)"

changed to

"of lower temperatures (< 40 °C)"

Line 12 pag 9

"Only, the alunite coexisting with kaolinite in the new hole pool has the finest grain sizes."

changed to

"Only, the alunite coexisting with kaolinite in the new hole pool exhibits the finest grain size."

Line 13 pag 9

"ordered kaolinite forms that usually occurs at temperatures"

changed to

"ordered kaolinite forms that usually occur at temperatures"

Line 18 pag 9

"the clay-bearing muds can be ascribed to various supergene to hypogene alteration"

changed to

"the clay-bearing muds can be ascribed to variable supergene to hypogene alteration"

Lines 23-24 pag 9

"In contrast, this equilibrium cannot be accounted at Campi Flegrei and any reliable temperatures result because of S-isotope fractionation between sulfates and H_2S."

changed to

"In contrast, this equilibrium cannot be accounted for at Campi Flegrei and any reliable temperatures result from the S-isotope fractionation between sulfates and H_2S."

Line 26 pag 9

"sulphate"

changed to

"sulfate"

Line 27 pag 9

"that show high oxidation state"

changed to

"that show a high oxidation state"

Lines 31-33 pag 9

"Actually, Campi Flegrei lacks of the occurrence of enargite and luzonite, both diagnostic for high-sulfidation environments, and instead shows minor occurrences of realgar (AsS) as well as cinnabar (HgS) (Tables 1, S1), and orpiment has been described (Russo et al., 2017)."

changed to

"Actually, Campi Flegrei lacks the occurrence of enargite and luzonite, both diagnostic for high-sulfidation environments. Instead, it shows minor occurrences of realgar (AsS) as well as cinnabar (HgS) (Tables 1, S1), and also orpiment has been described (Russo et al., 2017)."

Lines 41 pag 9-1pag 10

"The analysed samples do not produce bands attributable to C=H ligands (Supplement) and the carbon content is <1.25 wt% (most common < 0.2 wt%; Table S3). Yet, some higher $\delta^{34}S$ and $\delta^{18}O$ values for sulfates could be indicative, particularly considering the dry-wet alternating conditions."

changed to

"The analysed samples do not exhibit bands attributable to C=H ligands (Supplement) and the carbon content is <1.25 wt% (most common < 0.2 wt%; Table S3). Yet, some higher $\delta^{34}S$ and $\delta^{18}O$ values for sulfates could be indicative of microbial sulfur cycling, particularly considering the dry-wet alternating conditions."

Lines 4-5 pag 10

"Merging all available information, it appears that observations concerning the both sub-zones' "stationarity" and contradictory classification environment reflect"

changed to

"Merging all available information, it appears that observations concerning the both an apparent "stationarity" sub-zones' and a seemingly contradictory classification environment reflect"

Line 10 pag 10

"degassing"

changed to

"outgassing"

Lines 30-31 pag 10

"inside the crater waters literature data <0.001 g/l (Aiuppa et al. 2006) and crystallization of alum instead than NH_4^+-sulfates in"

changed to

"inside the crater waters (<0.001 g/l; Aiuppa et al. 2006) and crystallization of alum instead of NH_4^+-sulfates has been obtained"

Lines 40-42 pag 10

"However, marine strata and volcano-clastic sequence intercepted by deep drillings (San Vito1, Mofete and CF23 wells; Rosi and Sbrana, 1987; Piochi et al., 2014) are the key sediments of the NH_4 species. An additional supply can be the swampy sediments encountered in shallowest boreholes"

changed to

"However, marine strata and a volcano-clastic sequence intercepted by deep drillings (San Vito1, Mofete and CF23 wells; Rosi and Sbrana, 1987; Piochi et al., 2014) are considered as the key sediments for the NH_4 species. An additional supply can originate from the swampy sediments encountered in shallowest boreholes"

Lines 11-15 pag 11

"surroundings (Astroni, Bruno et al., 2007).

In the model, we further speculate that the acid-sulfate alteration zone at the Campi Flegrei is actually evidence of a paleo-conduit. This idea comes from field observations that indicate the alteration deposits locally underlying the most recent eruptive"

changed to

"surroundings (Bruno et al., 2007).

In the model, we further speculate that the acid-sulfate alteration zone at the Campi Flegrei is actually evidence of a paleo-conduit. This is based on field observations showing that alteration deposits locally underlie the most recent eruptive"

Line 32 pag 11

"new mineral phases at the variable sub-mm- to dm- to m- scales."

changed to

"new mineral phases at the sub-mm- to dm- to m- scales."

Lines 35-36 pag 9

"Tl, are possibly attributes of the evolution of a paleo-conduit."

changed to

"Tl, are attributed to the evolution of a paleo-conduit."

Line 4 pag 12

"between as "hydrothermal" (Moretti et al., 2017) vs. "magmatic""

changed to

"debate existing between a "hydrothermal" (Moretti et al., 2017) vs. a "magmatic""

Line 13 pag 12

"What cause the presence of NH_4^+"

changed to

"What causes the presence of NH_4^+"

Lines 17-19 pag 12

"Finally, the Pisciarelli site appears suitable for the biota and the life evolution studies. Here, the water-dominance, nitrogen richness, ≤ 200 °C temperatures and supergenic conditions are all needed ingredients for the growth of the organic substance and organisms (Jaffe, 2000)."

changed to

"Finally, the Pisciarelli site appears suitable for studies related to biota and the origin and evolution of life. Here, the water-dominance, nitrogen richness, ≤ 200 °C temperatures and supergenic conditions are all considered important ingredients for the formation of organic substances and the ultimate source of organisms (Jaffe, 2000)."

Line 12 pag 13

"multi-phases"

changed to

"multi-phase"

Line 19 pag 13

"as silver sulfide precipitates"

changed to

"as silver sulphide and barium sulfate precipitates"