Evidences of volcanic unrest on high-temperature fumaroles by satellite thermal monitoring: The case of Santa Ana volcano, El Salvador

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Title: The effects of environmental parameters on diffuse degassing at Stromboli volcano: insights from joint monitoring of soil CO2 flux and radon activity

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Abstract: Soil CO_2 flux and ^{222}Rn activity measurements may positively contribute to the geochemical monitoring of active volcanoes. The influence of several environmental parameters on the gas signals has been substantially demonstrated. Therefore, the implementation of tools capable of removing (or minimizing) the contribution of the atmospheric effects from the acquired timeseries is a challenge in volcano surveillance. Here, we present four years-long continuous monitoring (from April 2007 to September 2011) of radon activity and soil CO_2 flux collected on the NE flank of Stromboli volcano. Both gases record higher emissions during fall-winter (up to 2700 $Bq*m^{-3}$ for radon and 750 g m^{-2} day^{-1} for CO_2) than during spring-summer seasons. Short-time variations on ^{222}Rn activity are modulated by changes in soil humidity (rainfall), and changes in soil CO_2 flux that may be ascribed to variations in wind speed and direction. The spectral analyses reveal diurnal and semi-diurnal cycles on both gases, outlining that atmospheric variations are capable to modify the gas release rate from the soil. The long-term soil CO_2 flux shows a slow decreasing trend, not visible in ^{222}Rn activity, suggesting a possible difference in the source depth of the of the gases, CO_2 being deeper and likely related to degassing at depth of the magma batch involved in the February-April 2007 effusive eruption. To minimize the effect of the environmental parameters on the ^{222}Rn concentrations and soil CO_2 fluxes, two different statistical treatments were applied: the Multiple Linear Regression (MLR) and the Principal Component Regression (PCR). These approaches allow to quantify the weight of each environmental factor on the two gas species and show a strong influence of some parameters on the gas transfer processes through soils. The residual values of radon and CO_2 flux, i.e. the values obtained after correction for the environmental influence, were then compared with the eruptive episodes that occurred at Stromboli during the analysed time span (2007-2011) but no clear correlations emerge between soil gas release and volcanic activity. This is probably due to i) the distal location of the monitoring stations with respect to the active craters

and to ii) the fact that during the investigated period no major eruptive phenomena (paroxysmal explosion, flank eruption) occurred. Comparison of MLR and PCR methods in time-series analysis indicates that MLR can be more easily applied to real time data processing in monitoring of open conduit active volcanoes (like Stromboli) where the transition to an eruptive phase may occur in relatively short times.

Cover letter

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Prof. A. Aiuppa
Editor at JVGR

Dear Editor,
We are submitting the revised copy of the manuscript "The effects of environmental parameters on diffuse degassing at Stromboli volcano: insights from joint monitoring of soil CO_2 flux and radon activity".

As suggested by the Referees, the paper has been reorganised: Chapter 4 is now including four subchapters investigating more deeply the relationships between gases and environmental parameters.

Two new figures have been added: Fig. 5 showing the influence of wind (direction and speed) on soil CO_2 flux, and Fig. 6 which summarizes the results of spectral analyses on raw data (1 year of hourly measurements) for soil CO_2 flux, ^{222}Rn activity, air T and atmospheric P.
Figure 10 has been modified to include results of both the statistical treatments used.

We added in Table 3 the main parameters of the MLR analysis and the Table 4 has been modified to better explain the results of PCR statistical method. We also provided additional technical information of the two monitoring stations in the S1 Table for Supplementary Material.

Abstract, Discussion and Conclusions have been revised, as suggested by the Referees, to make immediately clear the scope of the paper and the obtained results.

We hope that the paper can now better satisfy the JVGR standards.

Sincerely,
Marco Laiolo
& co-workers

Torino February 03, 2016

Response to Reviewers (cited lines refer to the New Manuscript)

Referee # 1

- From a methodological point of view, I suggest the authors to add a table with the technical characteristics of the various sensors of the two stations (it can be added as supplementary material). Precision and accuracy of the measurements should be added.

As suggested, we added in Supplementary Material a Table (S1) with the technical characteristics, accuracy and sensitivity of the sensors of the two fully-automated stations.

- From an organizational point of view, I would suggest that chapter 4 was named "Results" with sub—chapters.

As suggested we changed the title of Chapter 4 and inserted four sub-chapters [line 205 – line 379].

- In page 12 authors mention that daily temperature variations have a weak influence on the gas flux, highlighting the correlation between wind and CO2 fluxes (also based on results obtained in previous works). However, looking at table 2, the correlation coefficient between CO2 flux and air temperature is higher than the coefficient between CO2 flux and wind speed. Authors should revisit these sentences and better discuss this aspect.

We rewrote this point trying to clarify this aspect (see Chapter 4.1 and 4.2). The relationship between CO_2 flux and wind has been more properly discussed (line 261 - 267 and Fig. 5). We performed a spectral analysis on both gases and on the main atmospheric variables (Air T and P) in order to outline the diurnal and semi-diurnal cycles of the analysed gas [line 278 – line 287].

- Pages 13 and 14 - Figure 6 is quite interesting and shows variations on the coefficients of correlation for different seasons. I would suggest the authors to add a shadow area in Figure 6 highlighting the first subset that authors refer "is somehow different compared to all the others". I suggest to better specify to which period authors refer to and to write the main differences (page 14, 3rd and 4th lines), as they are not easily observed just looking at the figure. Other aspect from Figure 6 is associated with the correlation established between CO2 flux and wind direction. Since the wind direction is a "circular" variable, how do the authors define the various wind quadrants? Do you define intervals or use the absolute values? In this last case 0 and 359 are almost the same, but will have a different weight for the correlation and for the regression and principal components models. Authors need to clarify how do they used this variable.

As suggested we outlined the subset *SS 2007* with a grey field in Fig. 6 (now Fig. 8). We added a new figure (Fig. 5), in Chapter 4.1., to show the wind influence (both speed and direction) on soil CO_2 flux. Moreover, we removed the variations of correlation coefficient between soil CO_2 flux and wind direction and speed. The question of wind influence is now more clearly discussed in chapter 4.1 (line 261 - 267).

- Authors analyse data obtained in the soil surface (soil CO2 flux station) and at about 1 m depth (soil radon concentration station), and this can result in different effects of the environmental parameters,

just depending on the site of the measurement (for instance the soil water content). I suggest the authors to add some discussion about this aspect.

We have inserted some sentences discussing the difference of the two stations [line 187 – 198] and the related influence of environmental parameters [line 233 – 267]. We have explained that the applied methods represent the state of the art for measuring soil CO_2 flux and ^{222}Rn concentration.

- On the chapter related with the statistical treatment, I suggest the authors to add some references that support the mathematical modelling. The reference "Hernandez et al., 2004" (page 15, line 2) refers to a work where this methodology was applied but do not explain the mathematical criteria. In addition, a previous work of Granieri et al. (2003 - EPSL) is, as far as I know, the first that applied regression analyses to the soil CO2 flux time series and should be mentioned. Those authors also discuss why selected regression models instead of the principal components analyses, aspect that should be here considered and discussed.

Following the suggestions of the referee, we inserted in Table 3 the variables used and the retrieved MLR equation by using the appropriate independent variables (i.e. environmental parameters) for both gases. Text has been adjusted accordingly [line 335 – 340]. Also Table 4 has been modified to better explain the PCR method.

- One question that was not clear to me was if the authors applied the statistical analyses to the raw data (hourly measurements) or to the daily values? This aspect needs to be clarified. If the statistical approach is done to the raw values, which in my opinion is the more adequate, did authors check also if there are daily variations on the gas time series? For the other side, if the statistical approach is applied to the daily averages (removing automatically the existence of daily cycles) I would suggest authors to add a sentence specifying that and to give some explanation how do they use the information of the wind speed for instance. Is it the average value selected, or the maximum speed of the day?

The statistical treatment was applied to the hourly raw values (wind included), as supposed by referee, and we have explained this aspect in the revised version of the paper [line 321 -323].

- As already mentioned authors used two different analyses to filter external influences from the gas data. Authors try also to discuss some differences between the two approaches used. In what concerns MLR authors mention (page 15) that MLR takes into account only the factors that are "more correlated" with CO2 and radon. What was the criterion used? Granieri et al. (2003) applied the procedure of Garside to select the variables; Viveiros et al. (2008; 2015 - JVGR and GSL) and Silva et al. (2015 - EPJ) selected only the variables that caused an increment on the R2 higher than 1%. A sentence about the criteria used need to be added. In what concerns the statistical methodology, I strongly suggest the authors to add the final regression models for the CO2 fluxes and radon concentrations. Same comment to the PCR models that need to be shown. The created variables (PCR method) that will correlate with the gas data are requested. These models can be added as tables or as equations, and are mandatory to understand which variables were added to the model, and which was the influence of each one.

We accepted the suggestions of the referee and adjusted the paper accordingly (see revised chapter 4.4; line 320 - 379).

- One aspect that also called my attention and I would like to see some discussion about is the fact that authors show that correlation factors change depending on the seasons. Would it be wise to produce

different regression models considering different seasons of the year? One additional point is related with the fact that some variables may have different influences on the gas data depending on different intervals of values, i.e., variables may work as second order polynomial and not as "simple" linear variables. Authors did not consider this possibility and evaluated only the linear correlations, what is acceptable, however, I would suggest to be more careful when in page 15 they refer: "…major fluctuations that obviously cannot be due to environmental variations" (line 17). Considering that some environmental parameters may not have only a straight linear influence on the gas flux, some of the explanation may be attributed to that aspect, thus I would recommend the authors to use the word "suggest" instead of "obviously".

We changed the sentence as suggested and added some sentences in chapters 4.3 and 5 [see line 383 – 385]. Additionally, regarding variations exhibited by radon residuals, we included a short discussion on the sensitivity of the radon sensor [line 389 – 395].

- Considering that MLR explains more gas variations than PCR (table 3), I do not understand why authors decided to use the standard residuals from the PCR to compare with the volcanic activity (Fig. 8). I would expect exactly the opposite. Authors should better explain this decision that has impact in the final discussion/observations. Authors mention that "PCR ensures a more accurate estimation of warning thresholds…" (pags 18, line 9), but in my opinion this sentence needs to be better explained.

Following the Referee suggestion, in the revised text we included the analyses of MLR residuals as well (see modified Fig. 10) and tried to better explain the reasons for preferring MLR for a quick monitoring application and PCR for a more accurate post-processing of data [see Chapter 5 and line 474 -481].

- In the abstract authors mention that no clear correlation emerges between soil gases and volcanic activity, which I agree. In fact, from the analyses of Fig. 8, some periods with anomalous CO2 or radon values are contemporaneous with the volcanic activity, but other periods do not show any relation. A pattern in the bahaviour of the CO2 fluxes - radon activity and volcanic activity is not established. Authors should better emphasize this, since in some paragraphs of sub-chapter 6.2, as well as in the conclusions, authors seem to highlight some correlations. Potential explanations for the differences observed between CO2 flux and radon data would also be welcome, as well as some attempt to explain why alternation between positive and negative anomalies is observed (essentially in the radon time series). Maybe authors have some idea why this occur and could be an interesting step forward to understand the subsuperficial phenomena.

The abstract, discussion and conclusions have been adequately reviewed following the suggestions in order to improve the comparison between our results and the Stromboli volcanic activity during the analysed period. We have also included an inference on a possible difference depth for Rn and CO_2 sources [line 396 – 401; line 433 – 437 and line 453 – 458].

I suggest the authors to read the recent work of Viveiros et al. (2014 - JGR), where similar seasonal trends were highlighted in the soil CO2 flux time series of data recorded at Furnas Volcano (S. Miguel Island). In addition, those authors, together with Rinaldi et al. (2012 - JGR), identified diurnal variations on the CO2 flux time series and modelled these variations with a geothermal simulator. In my opinion the results obtained by those authors can be useful to explain some of the observations here presented; in addition, those authors applied quite a similar filtering process to the data that is also confirmed in this *work (similar to Fig. 8). I suggest the authors to read the modelling done considering the thermal gradients, what is explained in the present work as a major cause for the fluctuations observed.*

We acknowledge the referee for this suggestion that it allowed to describe more properly the capability of atmospheric variables to modulate gas release from soil [see subchapter 4.2].

<u>Reviewer #2:</u> General comments

In this manuscript, the authors show a long time data series for two different gas species, consisting of continuous monitoring of Rn concentration and soil CO2 flux at Stromboli volcano between April 2007 and September 2011. The knowledge of the degassing processes in active volcanoes are of great interest for the volcanology community. In my opinion, the topics covered might be considered for a new contribution to the journal, however, some concerns regarding the manuscript should be considered before publication.

First, I suggest a revision of both the abstract and conclusion in order to make the novelty of the work and the findings immediately clear. Moreover, much of the original data considered in the paper are still lacking in clarity and scope. Specifically, section 4 is often redundant and the narrative explaining the correlation between the meteorological parameters and the Rn concentration and CO2 fluxes is confusing, despite the important role the authors seem to give this in the paper. Finally, I strongly recommend reviewing the conclusion. In my opinion, rather than focusing on the general literature published on Stromboli, the authors could be much more focused in demonstrating and underlining the relevance and importance of the results of their work.

We carefully revised in detail abstract, discussion and conclusions in order to focus better on the results obtained in this work and, specifically, highlighting the contribution that our results can give to the improvement of geochemical monitoring of active volcanoes.

I, therefore, recommend a major revision before publication, including a thorough proofreading in terms of use of English and general text cohesion.

Following the recommendations and previous suggestions, the text has been deeply revised and partly reorganized, avoiding repetitions and giving attention to its cohesion. We hope that it can now be accepted.

The following are specific comments for the improvement of the manuscript and requests for better explanation:

Pag. 3 "Due to its short half-life (t1/2=3.82 days)"

It should be specified that this refers to 222Rn

We changed the sentence according to this comment [line 60].

Pag 3 "In volcanic areas, the combined surveys of soil ^{222}Rn emissions and CO_2 flux give us the opportunity to track diffuse degassing and related processes (Giammanco et al., 2007)."

I suggest exploring this specific aspect in your work much more as it seems to be relevant to the topics within this paper and could also be reasonably considered one of the main arguments justifying the work.

The combined measurements of soil CO_2 flux and ^{222}Rn activity and specifically the $^{222}Rn/^{220}Rn$ ratio are mainly employed in order to reveal sites characterized by deep magmatic degassing (Giammanco et al., 2007). Following the suggestion we presented a more deep analysis of the CO2 flux and Rn activity reaching the conclusion that the two gases have likely a different source [line 396 – 401; line 433 – 437 and line 453 – 458].

Pag 6 "Since then, the scientific community acquired a tremendous amount of geophysical, geochemical and geodetic data were collected during the most recent eruptions 2007 and 2014 (Barberi et al., 2009; Rizzo et al., 2014)."

Grammar: Please insert "which"

The sentence was modified according to this comment [line 134 – 138].

Pag 8 "Therefore, two additional soil CO2 flux and multiparametric fully automated stations were installed along the ENE flank of the volcano (respectively at Rina Grande and Nel Cannestrà) where gaseous emissions are higher (Carapezza et al., 2009) (Fig. 2b)."

I suggest including in the Fig 2 b an inset showing the location of the stations on the island in order to identify the positions of the stations. In addition, it would be interesting to indicate the scale in order to recognize the "order of density" of the sampling shown in the maps both in fig. 2a and 2b.

In Figure 2a and 2b the location of the automated ^{222}Rn (2a) and soil CO_2 flux (2b) stations is shown. We added scale in both sub-Figures, as suggested.

Pag 8 "The gas automated stations consist of two units, one for measuring the isotope of the radon progeny …"

I suggest that the authors make a link with the previous paragraphs. It seems that there are four monitoring stations in operation, but in the following text you consider only one. It would be useful to understand why and what has motivated the choice. In addition, it is not clear which monitoring station is used by the authors; is it the station Nel Canestrà (as it seems indicated in tab 2), or at Rina Grande?

We clarified this point [line 163 – 165 and line 173 – 175] and clearly outlined (Figs. 1 and 2 and Tab. 1) the dataset analysed in this work.

Pag 9 "A preliminary analysis of the trend of environmental parameters, supported by the correlation factors reported in Table 2 …"

The authors should make clear what kind of "correlation factors" are considered between the parameters investigated (r, R2, CV …), also in table 2. In addition, in table 2, second line, the names in the column related to the CO2 monitoring station are not visible.

We changed correlation factors with correlation coefficients [line 226] and remarked in the text that in Table 2 we show the coefficient R [line 303].

Pag 10 "The wind speed signal is … (Fig. 4b)."

It should be Fig. 4e.

This sentence has been removed.

Pag 10 "Radon concentrations (see Fig. 3a) show relatively low values (Cigolini et al., 2009)"

For the sake of clarity, the authors should supply some more information in this sentence. Is a different regime of radon activity for Stromboli stated in Cigolini et al., 2009? If it is not, then it is preferable not to add self-reference in the paper.

We clarified this sentence and in order to better understand the mean of "relatively low values" we compare the discussed values with other measures performed in different areas [line 215 and line 219 – 221].

Pag 10 "Radon emissions are strongly negatively correlated to soil temperature, as already pointed out by Cigolini et al. (2009) and Laiolo et al. (2012). "

This is redundant.

The sentence has been removed.

Pag 11 "soil and air temperatures, … Therefore, a marked difference between …"

This is redundant.

The sentence has been removed.

Pag 11 "… an increase in soil moisture is capable of raising the 222Rn emanation coefficient of one order of magnitude …"

Do the authors intend perhaps to say rate?

We properly changed this sentence in order to clarify this point [line 237 – 240].

Pag 12 "…does not appear on radon long-term signals that, instead, show an excellent stability around the average of 2000 Bq/m3 (see straight line in Fig. 3a…"

Excellent? This adjective is very strong and I question its appropriacy.

The sentence has been modified.

Pag 12 "…It is important to outline that, according to this view, the concurrent measurements of CO2 and 222Rn can be considered as an important tool for detecting the variation in time of the deep vs. shallow contribution to the soil gas, as it was found in other volcanoes …"

This is an aspect that should be explored more, and the authors should explain in what way the data shown can contribute to make that link more evident. Perhaps it is also better to move this discussion to a different section of the paper, providing space for a wider argumentation.

The revised text includes a more carefully analysis of the temporal variations of CO_2 flux and ^{222}Rn activity leading to infer the deep vs shallow contribution of the two gases.

Pag 12 "…However, during a short term period, it is clear that…"

I suggest not introducing the phrase with "However" (here but also in other parts of the text and also "nevertheless"), because the contrast it introduces is not immediately obvious; i.e. the contrast refers to the topic sentence of the preceding paragraph and not its concluding sentence.

The comment has been considered in the English revision of the text.

Pag 13 "…This behaviour is evident by calculating the mean value of the whole data on each specific day of the year. The annual trend from Jan-1 to Dec-31 (Fig. 5) remarks the inverse relation between temperature and soil gas release, as well as the correlation between the two gas species…"

It would be preferable to make this sentence clearer rather than generate confusion amongst the relationship between daily variation of temperature and wind speed (previous sentence); and with the annual variation of temperature. In addition, I suggest adding the temperature in the figure 5.

We clarified the main steps of this analysis, which was applied on the raw dataset to investigate the annual trend affecting both gases [line 288 – 292]. Moreover, in the related Figure we added the trend of soil and air temperature (Fig. 7).

Pag 13 "Is also evident that the most significant day-by-day variations occur du…"

It is also ...

We corrected this sentence [line 294].

Pag 13 "In Fig. 6 we report … both CO2 flux and radon activity display a more distinctive negative …"

These arguments have already been considered in part, therefore the discussion in this section of the text becomes redundant and risks muddling the reader. The information included in fig. 6 should be re-articulated in a revised and synthesized text of the entire section 4.

This section of the paper has been reviewed taking into account the comments of both referees (see subchapter 4.3).

Pag 14 "In addition, seasonal variations in geochemical data and environmental correlation coefficients are somewhat different in the first subset compared to all the others."

This is not clear. Please explain better and introduce a description about what is meant by subset.

A specific comment has been introduced in subchapter 4.3 [line 315 – 319] and the first subset has been marked on Fig. 8.

Pag 14 "In this time span , there"

Please delete space

We deleted space.

Pag 15 "Particularly, soil and air temperature were considered by the regression for both gas species, together with wind conditions for CO2 …"

Please be specific with wind "conditions": speed; directions; or both?

The question has been clarified [line 339] and we added also a new Fig. 5.

Pag 15 "Results show that the atmospheric variables taken into account for this analysis are able to predict 45% and 51% (R=0.67 and R=0.71) …"

Is it R or R2? In table 3 it is indicated as "Determination Coefficient"

We clarified this point in the text [line 341 - 342] and in the related Tables (Table 3 – 4), in order to avoid misunderstanding.

Pag 15 "Moreover, soil CO2 flux and 222Rn values show low dispersions, respectively 4.0% and 4.8% of the computed residuals exceed the average ±2 standard deviation (Table 3)"

This should be inverted: … respectively 4.8% and 4.0%

We corrected the sentence [line 344].

Pag 15 "By looking at the residuals, it can be seen that…"

The bell shape in the last two years does not seem to have disappeared exactly. Indeed, the high CO2 flux fluctuation in the previous years masks the bell shape in the later years (it could be made better visible using a different scale). In addition, the CO2 residual shown in the graph appears to be significantly negative for most of the time. Can you give an interpretation/explanation for this; is there an error in the right scale of the graph?

We can assure the referee that the "bell-shape" of CO_2 flux does not depend on the scale. There was actually an error on the right scale of the graph now corrected in Fig. 9 and we are grateful to the referee for allowing this correction.

Pag 16 "So, we created one dataset for radon with soil temperature and atmospheric pressure of radon station and added the other variables from the CO2 station.…"

I think the authors should also indicate in the text why it is reasonable to use meteorological parameters measured from a monitoring station sited at distance; are they not far?; is there significant soil homogeneity… ?

As shown in Figure 1 and 2 the two stations are actually very close, at a distance of only 30 m. Actually, only soil moisture is extrapolated for ^{222}Rn station.

Pag 17 "However, it can be noted that the radon treatment provides many significant negative residual values ≥ 2σ, whereas only one negative residual is recorded for soil CO2 flux (see Fig. 8)…"

The graph shows an Rn time series that seems to vary without a specific connotation but, in some way, appears as a sort of noise. Please could you be more specific about the volcanological information that we can deduct from this signal?

In the Discussion and Conclusions we addressed this topic and suggested an explanation for the radon behaviour [line 387 – 395].

Pag 18 "Comparison of the standard residuals (by PCR) for CO2 soil flux and 222Rn activity versus time (Fig. 8) shows that … considered, only radon shows frequent positive anomalies whereas anomalous CO2 flux values are rarely recorded.…"

It is hardly possible to recognize if there is any similar behaviour in the graph, both for the length of the data series and the difficulty in overlapping the two signals (fig 8a and b). In order to state that there is, and how significant the correlation may be, it would be better to divide the graph into smaller windows of time. It would help also to overlap the signals and, if possible, add some numerical correlation values (like matching positive and negative cases).

The Discussion (and figure) has been modified to address this question.

Pag 18 "Finally, the comparison between … compare our emerging results with the 2007-2011 Stromboli volcanic activity"

It is not clear why:

- *MLR seems to be a more adequate method for getting the quick results needed in near-real time volcano monitoring.*

- *whereas PCR ensures a more accurate estimation of warning thresholds (which thresholds ?)*

Please explain

The chapter has been revised to better explain the characteristics of the two methods and their possible use [Chapter 5 and line 474 -481]. Results of both treatments have been considered for the comment on the residual values.

Pag 18 "…are slightly influenced by volcanic activity that, in the time-span considered, did not produce any relevant explosions or major lava effusion.

This section should be improved with more details in the interpretation. The authors stress that in the first two years there is a good correlation between the data recorded and the volcanic activity. But they should explain or interpret the scarce correlation from 2009 to 2011, where actually 7 major explosions and 3 lava overflows definitely confirm that Stromboli volcano was fairly active.

We modified the Discussion, as well as Conclusions and Abstract in order to remove any misunderstanding regarding the relationship between volcanic activity and gas signals obtained by using the PCR and MLR analyses. We enphasize that in the considered period volcanic activity was related only to changes in the upper plumbing system without involvement of deep gas-rich magma [line 416 – 421 and line 442 – 443].

Pag 19 "… are slightly influenced by volcanic activity that, in the time-span considered, did not produce any relevant explosions or major lava effusion."

This contradicts what is stated in the previous section and shown in fig. 8. In addition, in the recent literature and also in the references quoted in the text (Calvari et al. 2014; Rizzo et al. 2015) it is pointed out that Stromboli, in the time considered, has shown "dynamic" volcanic activities.

In the deep revision on the Discussion and Conclusions sections we carefully considered the referee comments.

Pag 19 "…As recently stressed by De Gregorio et al. (2014) … Thus, combining CO2 flux and 222 Rn concentration measurements gave us the opportunity to better investigate the changes in volcanic activity associated with magma rise."

The link is not clear between these two sentences. Moreover, it does not seem that there has been enough investigation into what way the combination of CO2 and Rn provided information in this work regarding the volcanic activity and the "associated magma rise".

We modified this sentence [line 451 – 453].

Pag 20 "We finally emphasize that the monitored area has been characterized by marked anomalous radon emissions prior and during the paroxysmal explosion of March 15, 2007 (Cigolini et al., 2013). Therefore, it is not excluded that multiparametric geochemical monitoring may play ..."

It is surely acceptable the statement that geochemical monitoring may play a very important role in order to forecast volcanic activities, but data and argumentations supporting this aspect in this paper are still weak and necessitate major deepening.

We changed this sentence remarking the contribution given by continuous measurements in the analysed sites and also considering previous observations [line 481 – 486].

Figure captions

Fig 1 Please correct CO2 in CO_2

We changed the figure according to this comment.

Fig 2 Please correct CO2 in CO_2 *flux. Please add some information in relation to the structural features shown in the maps.*

We changed the figure according to this comment.

Fig 3 It seems that the circles are the raw data; please check.

As specified in the caption, we confirm that circles are the raw data.

Fig 5 Please correct CO2 in CO_2

We changed the figure (now Fig. 7) according to this comment.

Fig 7 All the graphs are in different grey gradations, but CO_2 *in the legend b is in red. Please check.*

We changed the figure (now Fig. 10) according to this comment.

*Highlights (for review)

Highlights

CO_2 flux and Rn activity were continuously measured for 4 years on Stromboli NE flank

CO_2 flux and Rn are affected by environmental parameters (soil H and T, wind, air T)

Two statistical methods were used to identify and remove atmospheric effects on gas

In the reference period no deep gas-rich magma was involved in the eruptive activity

No clear correlation was found between volcanic activity and CO_2 and radon emissions

The effects of environmental parameters on diffuse degassing at Stromboli volcano: insights from joint monitoring of soil CO_2 flux and radon activity

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ABSTRACT

~~We~~ Soil CO_2 flux and ^{222}Rn activity measurements may positively contribute to the geochemical monitoring of active volcanoes. The influence of several environmental parameters on the gas signals has been substantially demonstrated. Therefore, the implementation of tools capable of removing (or minimizing) the contribution of the atmospheric effects from the acquired timeseries is a challenge in volcano surveillance. Here, we present ~~a~~ four years-long ~~period of~~ continuous monitoring (from April 2007 to September 2011) of radon activity and soil ~~carbon dioxide~~CO_2 flux ~~acquired~~collected on the NE flank of Stromboli volcano~~, from April 2007 to~~

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September 2011. Previous soil degassing surveys on this sector allowed us to decode the volcano-tectonic structures where diffuse degassing is actively taking place. Radon and CO_2 stations were installed in a distal area from the active vents, at ~520 m a.s.l. (named Liscione or Nel Cannestrà). Collected time series define a rather good correlation between the two . Both gases and both record higher emissions during fall-winter periods (up to 2700 Bq/m^3*m^{-3} for Rnradon and 750 g m^{-2} day^{-1} for CO_2). The ^{222}Rn concentration shows a remarkable steady state) than during spring-summer seasons. Short-time variations on ^{222}Rn activity are modulated by changes in soil humidity (rainfall), and changes in soil CO_2 flux that may be ascribed to variations in wind speed and direction. The spectral analyses reveal diurnal and semi-diurnal cycles on both gases, outlining that atmospheric variations are capable to modify the gas release rate from the soil. The long-term trend whereas soil and air temperatures define a typical sinusoidal curve and soil humidity variations strictly control short-time changes. Soil CO_2 flux exhibits a similar behaviour with drastic changes ascribed to wind conditions. However, the long-term datasets show a soil CO_2 flux shows a slow decreasing trend, not visible in ^{222}Rn activity, suggesting a reduced CO_2 contribution from a deep source (i.e. magmatic) within the most recent years.

Correlation coefficients outline that radon variations are essentially modulated by soil temperature and humidity, whereas CO_2 fluxes mainly depend on soil-air temperature and wind speed and direction. In order topossible difference in the source depth of the of the gases, CO_2 being deeper and likely related to degassing at depth of the magma batch involved in the February-April 2007 effusive eruption. To minimize the effect of the environmental parameters on the ^{222}Rn concentrations and soil CO_2 fluxes, we applied two different statistical treatments were applied: the Multiple Linear Regression (MLR) and the Principal Component Regression

(PCR). These approaches allow to quantify the weight of each ~~atmospheric~~ environmental factor on the two gas species~~. Both methods~~ and show ~~the~~a strong influence of some ~~of the environmental~~ parameters on the gas transfer processes through ~~the soil.~~

~~Residual~~soils. The residual values of radon and CO_2 flux, i.e. the values obtained after correction for the environmental influence, were then compared with the ~~volcanic~~eruptive episodes that occurred at Stromboli during the ~~analyzed~~analysed time span (2007-2011) but no clear ~~correlation emerges~~correlations emerge between soil gas release and volcanic activity. This is probably due to *i)* the distal location of the monitoring stations ~~in~~with respect to the active craters and to *ii)* ~~to~~ the fact that during the investigated period ~~is characterized by the “ordinary” Strombolian activity (occurring between two eruptions, i.e., 2007 and 2014) during which~~ no major eruptive phenomena (paroxysmal ~~explosions has been identified.~~

~~Results obtained by~~explosion, flank eruption) occurred. Comparison of MLR and PCR ~~have been evaluated~~methods in ~~terms of “cost-benefit” for improving~~ time-series ~~analyses. The methodology hereby tested~~analysis indicates that MLR can be more easily applied ~~in near-~~to real time ~~measurements to improve geochemical~~ data processing in monitoring ~~in volcanic areas~~of open conduit active volcanoes (like Stromboli) where the transition to ~~mitigate volcanic risk~~an eruptive phase may occur in relatively short times.

1 – Introduction

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~~Geochemical measurements and real~~Real-time monitoring of gas ~~species are~~release (output and composition) at active volcanoes is useful to ~~identify~~forecast changes in volcanic activity~~ and magma degassing. It is well known that diffuse and concentrated soil degassing may release high amount of gases at active volcanoes~~. Active volcanoes are characterized by persistent huge gas emissions from craters, fumaroles and also diffusively from soils (Allard et al., 1991; Burton et al., 2013; Inguaggiato et al., 2013~~; Burton et al., 2013), thus~~) and systematic gas monitoring may help to detect precursory signals of incoming eruptions (e.g., Aiuppa et al., 2009; Padrón et al., 2013). In recent years, this approach was applied at several volcanoes to record geochemical changes during volcanic activity~~,~~ and to investigate their role before, during and after major eruptive episodes (including flank instabilities; e.g., Carapezza et al., 2004 and 2009; Alparone et al. 2005; Cigolini et al., 2005). Another open and debated issue is the role of degassing prior the onset of earthquakes (Toutain and Baubron, 1999; Salazar et al., ~~2002~~2001) and during earthquake-volcano interactions including seismic-volcanic unrest (Cigolini et al., 2007; Padilla et al., 2014).

Carbon dioxide, after water, is the most abundant volatile dissolved in magmas and, because of its relatively low solubility in magmatic liquids, it is essentially released at higher depths and before other gas species (Pan et al., 1991, Papale et al., 2006). Notably, measurements ~~on~~of soil CO_2 fluxes or CO_2 concentrations in volcanic plumes, are critical for detecting degassing processes related to changes in the plumbing system of the volcano.

Radon is a noble gas, a daughter decay product of ^{226}Ra and belongs to the ^{238}U decay chain. Due to its short half-life ($t_{1/2}$= 3.82 days~~) it~~), ^{222}Rn can be used as a tracer of both diffuse and localized degassing since it can substantially be measured everywhere. ~~However, radon concentration in~~Radon concentrations may be moderate during diffuse degassing~~ magmas may be low~~, but during fracture opening ~~it~~they may reach extremely high values (~~well over 10^{6}Bq~~higher than 10^6 Bq/m^3~~ within the~~, as measured in Stromboli crater area; Cigolini et al., 2013). Its ascent towards the surface is strictly ruled by the mobility of other gas phases, such as CO_2 and H_2O defined as “carrier gases” (Gauthier and Condomines, 1999). ~~In volcanic areas, the~~

combined surveys of soil ^{222}Rn emissions and CO_2 flux give us the opportunity to track diffuse degassing and related processes (Giammanco et al., 2007). However, it is well known that environmental parameters are critical in modulating gas release from soils (including radon and CO_2, The joint measurements of soil CO_2 flux and ^{222}Rn activity have been used to search possible volcanic and seismic precursors (Makario Londoño, 2009), as well as to track fluid migration and outgassing along active faults, fractures or fumaroles (Baubron et al., 2002; Faber et al., 2003; Zimmer and Erzinger, 2003). Moreover, combined surveys of ^{222}Rn, ^{220}Rn and CO_2 give us a clue to discriminate distinct gas sources (i.e. rock fracturing, hydrothermal, magmatic) (Giammanco et al., 2007; Siniscalchi et al., 2010) and, to track the evolution of a volcanic unrest phase (Padilla et al., 2013). Pinault and Baubron, 1996; Carapezza and Granieri, 2004; Pérez et al., 2007; Cigolini et al., 2009).

AutomaticContinuous and real-timeautomatic measurements substantially increase the potential role of these gases in forecasting eruptionspossibility to identify precursory signals, since the data are easily collected, transferred, and processed and filtered thus minimizing the effects of environmental parameters on soil degassing in near real-time (Brusca et al., 2004; Viveiros et al., 2008; Cigolini et al., 2009; Carapezza et al., 2009). Environmental parameters are critical in modulating gas release from soils, including radon and CO_2 (Pinault and Baubron, 1996; Carapezza and Granieri, 2004; Pérez et al., 2007; Cigolini et al., 20102009) and their effects must be considered during continuous geochemical monitoring.

In this respect, a promising challenge is to establish a fully-automated data processing able to minimize the effects of environmental factors on the acquired data. In this way, data obtained by the geochemical monitoring networks can be easily transferred to the authority responsible of volcano surveillance. The statistical treatment or the spectral analysis of the data are the mostly used methods to recognize and remove the contribution of the atmospheric factors (e.g. Carapezza et al., 2009; Laiolo et al., 2012~~). ~~; Rinaldi et al., 2012; Silva et al., 2015; Viveiros et al., 2008; 2014). Particularly, the spectral analysis may be positively applied to recognize diurnal to seasonal cycles and to investigate the processes ruling the release of gases from soils (Rinaldi et al., 2012; Martin-Luis et al., 2015).

Radon concentrations can be diluted by major fluxes of CO_2 and water vapor (e.g., Giammanco et al., 2007; Siniscalchi et al., 2010). Recently, Girault et al. (2014) and Girault and Perrier (2014) have shown, at the Syabru-Bensi hydrothermal system (Central Nepal), that radon is ~~essentially incorporated into the released CO_2~~generated from a shallow ~~radium sources~~source (a rock thickness of 100 m is sufficient to account for the observed radon discharge~~). An alternative model with a deeper source of CO_2 (carrying~~) and incorporated into upraising CO_2. In active volcanoes radon can be carried to the surface from ~~greater depth) is also possible~~great depths along major faults~~ (Girault and Perrier, 2014).~~. Cigolini et al. (2013) have shown that ~~extremely~~ high radon emissions ~~at Stromboli volcano ~~can be related to the ascent of ~~hot to supercritical ~~CO_2 -bearing hot fluids ~~(up to 270-410 °C) ~~along the fractures (200-300 m deep) ~~that surround~~surrounding the crater rim~~.~~ of Stromboli volcano (at about 700-720 m a.s.l.) and well correlate with the estimated depth of the source region of VLP events (e.g., (Chouet et al., 1997; Marchetti and Ripepe, 2005). Previous investigations ~~showed~~have shown that CO_2 fluxes and ^{222}Rn concentrations at Stromboli are within the range of those measured in other open-conduit active volcanoes (~~Inguaggiato~~Cigolini et al., 2013; ~~Cigolini~~Inguaggiato et al., 2013).

In this paper, we present ~~a ~~four years~~ long period (from April 2007 to October 2011)~~ of continuous monitoring of ^{222}Rn activity and soil CO_2 flux ~~recorded~~collected by two automatic stations located on the north-eastern upper flank of Stromboli Island (Fig. 1). ~~These~~The measurement sites have been chosen in the light of previous surveys~~. In fact,~~: anomalous radon values were recorded in this site during periods of sustained volcanic activity and before, during and after the paroxysmal explosion of March 15, 2007 (Cigolini et al., 2013). Similarly, systematic measurements of soil CO_2 flux revealed anomalous degassing ~~zones~~areas on the volcano slopes and this site has been identified ~~this site~~ as a potential target for continuous monitoring (Carapezza et al., 2009). ~~Collected data on both gas species were analyzed to define statistical parameters (background, threshold and anomalies; Carapezza et al., 2009; Cigolini et al., 2009; 2013). Here, we applied two different statistical methods (Multiple Linear Regression and Principal Component Regression) for a critical evaluation on the effects of some environmental parameters on the acquired gas species. Both methods allow to minimize the contribution of the atmospheric factors on the CO_2 and radon signal and attempt to correlate gases anomalous variations with the volcanic regimes operating at Stromboli during the recorded time span. Finally, we briefly discuss the future application of the statistical treatments on near real-time measurements adopted in monitoring volcanic activity.~~

2 – Stromboli volcano

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Stromboli is the north-~~eastern~~easternmost island of the Aeolian archipelago and ~~it~~ reaches an elevation of 924 m a.s.l. (Fig. 1). It is a composite stratovolcano consisting of lava flows alternated with abundant tephra deposits. The emerged part of the volcanic edifice was built ~~within~~in the last 100 ky (Francalanci et al., 1989; Hornig-Kjiarsgaard et al., 1993). The morphology of the island results from periods of extrusive growth alternated to lateral collapses, in turn related to ~~the~~ dyke intrusions, magma upwelling and regional tectonics (Tibaldi, 2003 and 2004; Corazzato et al., 2008). The volcano is well known for its typical persistent explosive activity called Strombolian, that started approximately 2 ky ago (Rosi et al., 2000; Arrighi et al., 2004). Strombolian activity is characterised by continuous degassing with the emission, on average every 15-20 minutes, of juvenile material (glowing scoriae, lapilli and ash) ejected from the active vents located within the crater terrace at ~700 m. a.s.l. This mild explosive activity is episodically interrupted by lava flows, major and paroxysmal explosions (Barberi et al., 1993 and 2009) that can be accompanied by flank failure and collapses, which may also generate tsunamis, like in 1930 and recently in December 2002~~. These events may affect the South-Central Tyrrhenian sea~~ (Tinti et al., 2006). Paroxysmal events, such as the ones occurred on April 5, 2003 and March 15, 2007, are the most violent volcanic explosions of Stromboli and are characterized by the ejection of the so-called “golden pumices” (nearly aphyric, phenocrysts < 10 vol%, highly vesicular > 50 vol%, low viscosity K-basaltic pumiceous materials; Métrich et al., 2005 and 2010). These ejecta are generally mixed with degassed ~~scoriaes~~scorias (the latter also ejected during the typically mild Strombolian activity) and with ballistic solid blocks. The CO_2 and H_2O contents measured in primitive melt inclusions, found within forsteritic olivines of the golden pumices, indicate that these materials represent the undegassed magma residing in the deeper part of the Stromboli plumbing system (Bertagnini et al., 2003; Francalanci et al., 2004; Métrich et al., 2005; Cigolini et al., 2008; ~~2015~~2014).

Soon after the 2002-2003 effusive event, a great improvement of the monitoring system was undertaken under the coordination of the Italian Civil Protection Department. ~~Since then,~~This advance on ground-based monitoring allowed the scientific community ~~acquired~~to acquire a ~~tremendous~~great amount of geophysical, geochemical and geodetic data ~~were collected~~ during the most recent ~~eruptions 2007 and 2014 (~~effusive episodes, as well as during the span of time characterized by low to high explosive activity (cf. Barberi et al., 2009; Ripepe et al., 2009;

Calvari et al., 2014; Rizzo et al., 2014). Recent investigations ~~show~~have also shown that ~~in~~within the latter years there ~~has been~~was an increase in ~~thermal activity~~ the radiative heat power associated to several minor lava overflows within the summit area (Coppola et al., 2012; Calvari et al., 2014).

~~In recent years, geochemical~~Geochemical monitoring at Stromboli has involved the following activities: soil radon ~~concentrations~~concentration (Cigolini et al., 2009~~;~~ and 2013), soil CO_2 flux (Carapezza et al., 2004 and 2009; Federico et al., 2008; Rizzo et al., 2009 and 2014), ~~fully-automated~~ SO_2 plume ~~measures~~measurements by COSPEC (Burton et al., 2009), continuous measurements of CO_2/SO_2 ratios within the volcanic plume (Aiuppa et al., 2009 and 2011). These methods were tested to eventually forecast paroxysms and major explosions~~. However~~; in addition, the continuous monitoring of low-temperature fumaroles ~~has also been found as~~was useful ~~tool~~ to detect short-time changes in volcanic activity (Madonia and Fiordilino, 2013). The extension and structure of the complex hydrothermal system of the volcano has ~~also~~ been investigated by multidisciplinary studies (involving electrical resistivity, soil CO_2 ~~concentrations~~concentration, temperature and self-potential measurements~~, cf.,~~; Finizola et al., 2006 and 2009; Revil et al., 2011). ~~Nevertheless systematic data collection~~Geochemical studies on the geothermal aquifer ~~(e.g. chemical-physical parameters, dissolved gas, isotopes) has been very useful~~at the periphery of the volcano is an additional tool to detect precursory signals of an impending eruption (Carapezza ~~and Federico 2000~~et al., 2004; Capasso et al., 2005).

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3 – Methods and Techniques

Preliminary radon and ~~CO_2~~carbon dioxide surveys were conducted to find the most appropriate sites for continuous monitoring. A network of 21 radon stations has been operative at Stromboli since 2002 (~~e.g.~~ Cigolini et al., 2005; 2009; 2013). Systematic measurements were undertaken by using LR115 track-etches alpha-detectors exposed from two to six weeks (Bonetti et al., 1991), in order to obtain continuous time series on ^{222}Rn emissions. Additionally, periodic short-term measurements has been performed by means of EPERM® electretes (Kotrappa et al., 1993) that allowed ~~us~~ to better correlate radon emissions with the variations of volcanic activity (Cigolini et al., 2005 and 2007). These periodic measurements demonstrated that diffuse degassing occurs at Stromboli mainly along the main structural discontinuities ~~(Fig. 2).~~. After the February-April 2007 effusive-explosive event, a real-time station for radon measurements was first installed at 520 m a.s.l. at Liscione, on the northeastern side of the cone (see Fig. 1 and Fig. 2a). Similarly, a soil CO_2 flux survey first outlined the main sectors of gas emanation (Carapezza and Federico, 2000) and two automatic soil CO_2 flux stations (and environmental parameters) were installed at Stromboli: one at the summit (Pizzo sopra La Fossa) in 1999, and the second one near the sea-shore in 2001 (Pizzillo). In the following years, CO_2 soil concentration surveys, within the crater terrace and surrounding areas, were performed to identify the sectors of major degassing and higher hydrothermal activity (Finizola et al., 2002 and 2003). Furthermore, Carapezza et al. (2009) performed a wide detailed survey of soil CO_2 flux on the island and sectors of anomalous degassing were detected. Therefore, two ~~additional~~ soil CO_2 flux and multiparametric fully-automated stations were installed along the ENE flank of the volcano (respectively at Rina Grande and Nel Cannestrà) where ~~gaseous~~anomalous gas emissions ~~are higher~~were found (Carapezza et al., 2009) (Fig. 2b). ~~The area is confined between two major structural~~

~~discontinuities (the N40E fault, and the N60E fault) that cross-cut the northeastern sector of Stromboli.~~

~~The gas~~Dataset analysed in this paper refer to the ^{222}Rn and CO_2 measurements acquired by fully automated stations located in the Nel Cannestrà sector (see Fig. 1 and 2). The area is confined between two major structural discontinuities (the N40°E fault, and the N60°E fault) that cross-cut the north-eastern sector of Stromboli. These automated stations consist of two units, one for measuring the isotope of the radon progeny (together with soil temperature and atmospheric pressure) and the other for measuring soil CO_2 flux (by accumulation chamber) together with environmental parameters (atmospheric temperature, humidity and pressure; soil humidity and temperature; wind direction and horizontal speed). The radon unit provides near real-time measurements of ^{222}Rn concentrations (by using a DOSEMan, Sarad Gmbh, Germany) connected to an electronic board able to acquire and transfer the collected data to a radio-modem that sends, by means of a directional antenna, the signal to the COA volcano observatory ~~(Cigolini et al., 2009). Specifically, data are acquired~~. The station acquires data every 30 minutes and the radon concentration and soil temperature are measured at 1 m depth~~ (Cigolini~~. The DOSEMan radonmeter measures α-particles within a 4.5-10 MeV energy window, including both ^{218}Po and ^{214}Po peaks (Gründel and Postendörfer, 2003). An exhaustive description of the radon dosimeter and of the real-time ^{222}Rn station can be found in Cigolini et al. (2009) and Laiolo et al~~., 2009~~. (2012). Soil CO_2 flux and environmental parameters are measured hourly with a fully equipped automated station produced by West Systems (see Carapezza et al., 2009 for method description). Soil temperature and humidity are measured at 50 cm depth; air CO_2 concentration is measured 30 cm above the soil/air interface (Carapezza et al., 2009). Data are stored on a non-volatile memory and can be retrieved by means of a telemetry system at the volcano observatory (COA, see Fig. 1). The main technical characteristics of the sensors used in both stations are reported in the supplementary materials (Table S1).

The timespan investigated in this work (April 2007 - September 2011) matches the period in which both instrumentations were mostly ~~operatives~~operative. In fact, the ^{222}Rn station was installed in early April 2007 and is still operative whereas the automated CO_2 flux station, installed in mid March 2007, was dismissed in late September 2011.

4 – ~~Continuous monitoring~~ Results

4.1 – Time series of ~~radon activity and~~ soil CO_2 flux ~~from the soil~~and ^{222}Rn activity

~~The time series for ^{222}Rn activity and soil CO_2 flux (April 2007-September 2011) together with those of the environmental parameters, are reported in Figures 3 and 4, respectively. The main descriptive statistics are reported in Table 1.~~

The overall behaviour of the CO_2 and ^{222}Rn signals is somehow similar in the first two years: they both show bell-shaped profiles, strongly ruled by ~~season~~seasonal trend, that reach their lower and stable values during summer and the higher values ~~and~~, with a wider variation range, in winter. A similar behaviour is ~~shown~~observed also in ~~terms of~~ multi-modal distributions (see histograms in Fig. 3). Both trends display several marked spikes within each time series (Fig. 3a and 3b~~), with~~) and numerous peaks of ~~both~~the two gases ~~being~~are essentially concordant~~, thus indicating~~. In the last two years, soil CO_2 flux shows a ~~common origin.~~ decreasing trend, whereas radon activity maintains nearly the same annual average, or simply increase (see Table 1).

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A preliminary analysis of the trend of environmental parameters, supported by the correlation factors reported in Table 2,Compared with other active volcanic areas, radon shows that both the ^{222}Rn and CO_2 signals are inversely correlated to soil and air temperature, and positively correlated to soil humidity (especially radon). The decoupling of soil temperature from the radon signals is well known: convective cells that rule gaseous transfer toward the surface do not reach the surface due to the inversion of the thermal gradient within surface soils due to summer heating (Mogro Campero and Feischer, 1977; Cigolini et al., 2001; 2009; Laiolo et al., 2012). It is interesting to note that this phenomenon is affecting CO_2 fluxes as well.

The wind speed signal is rather variable and fluctuating. However, it seems to be more stable during late spring-summer, when winds are substantially below 7 m/sec (Fig. 4b).

Radon concentrations (see Fig. 3a) show relatively low values (concentration (cf. Cigolini et al., 2009),2013 and references therein). Values are essentially below 2000 Bq/m^3 for a large part of the year, and may exhibit a short-term variability. In fact,; the average radon activity in the four years is around 2000 Bq/m^3 with a standard deviation of 1200 Bq/m^3 (Table 1). During winter (November-February), radon typically exhibits higher average values (Fig. 3a) with peaks up to 7900 Bq/m^3. We remind that the ^{222}Rn activity in the summit area, close to the active vents, shows significantly higher average values reaching 12,500 Bq/m^3 (± 4,200; see Laiolo et al., 2012; Cigolini et al., 2013).

Radon seasonal minima refer to late spring-summer periods (March-October) with average values of ~1200 Bq/m^3 and hourly minima close to 200 Bq/m^3. This behaviour isThe ^{222}Rn long-term stability on low values seems closely related to the absence of marked weather changes during this season.

Radon emissionsThe time-series of environmental parameters are strongly negativelyshown in Fig. 4; a preliminary analysis of their effects, supported by the correlation coefficients (Table 2), shows that both the ^{222}Rn and CO_2 signals are inversely correlated to soil and air temperature, as already pointed out by Cigolini et al. (2009) and Laiolo et al. (2012). In turn, soil humidity (higher after rainfalls) is somehowand positively correlated with(especially radon emissions (Table 2).) to soil humidity, in turn depending on rainfall. It is interesting to point out that recorded data show that sudden variations in radon concentrations normally occur within few hours of continuous raining and/or temperature drops. Notably, aA similar phenomenon has been already observed at Furnas volcano (Azores archipelago) during continuous monitoring of CO_2 flux measurements (Viveiros et al., 2008) as well asand radon measurementsactivity (Silva et al., 2015).

The relation between temperature and ^{222}Rn activity is ascribed to the local thermal gradient (between soil and air temperatures) that affects the efficiency of the in-soil convective cells and, consequently, the migration of gasesgas toward the surface (Mogro-Campero and Fleisher, 1977; Cigolini et al., 2001). Therefore, a marked difference between soil and air temperatures, typical of the fall-winter season, causes an increase in the measured radon activities. Conversely, theThe entire dataset shows a clear positive correlation (R= 0.74; Table 2) between soil moisture and radon emissions. Indeed; indeed, an increase in soil moisture is capable of raisingmay increase the ^{222}Rn emanation coefficient of(i.e. exhalation rate) by one order of magnitude (Nazaroff, 1992; Sakoda et al., 2010; Girault and Perrier, 2012).

However, to evaluate the relation between ^{222}Rn activity and soil gas concentration and humidity in the specific site of radon station, we have also to consider the confinement of the box containing the radon detector. The device is placed in an impermeable polycarbonate case (permeable to ^{222}Rn but not to water) at a depth of about ~~80 cm~~1 m. Thus, if the soil matrix surrounding the case is affected by water saturation, the preferential pathway for radon migration will follow the interface soil-bottom of the case, leading to an increase in α decay counts. ~~An alternative explanation may be~~Another possibility is that, during a rainfall episode, only the higher portions of soil (down to about 10-30 cm) undergo water saturation that, in turn, temporarily inhibits the free motion of the radon particles toward the surface. Consequently, radon will preferentially be confined at lower levels (i.e., where water content is low or absent) so that decay counts will be drastically higher in the portion of soil where the case (containing the detector) is inserted. ~~Notably, as~~As the relation between soil humidity and radon activity essentially depends from soil permeability, the observed behaviour can be inhomogeneous over a given sector of the volcano (Perrier et al., 2009).

Soil CO_2 flux measurements ~~were~~ acquired by the automatic station ~~starting~~started on mid~~-~~-March 2007. ~~Average values~~The four years average value for CO_2 ~~fluxes are~~flux is ~600 (±643) g m^{-2} day^{-1} ~~with~~1 with a rather high standard deviation (as for radon) (Table 1). The maximum values were reached in January 2008 with fluxes ~~up to about~~slightly exceeding 7000 g m^{-2} day^{-1}. ~~Also in this case~~As already observed for ^{222}Rn activity, minima in soil CO_2 fluxes occur during summer-early fall when values ~~approaching to zero were recorded (or~~ well below 100 g m^{-2} day^{-1} were recorded. A similar trend outlines that the variation of the local thermal gradient is capable to affect also the CO_2 flux from the soil (Viveiros et al., 2014).

Surprisingly, there is a noticeable correlation between soil CO_2 flux and wind, both speed (positive) and direction (negative) (Table 2). In Fig. 5 it is clear how winds blowing toward SE at speed > 8 m/s are able to produce an efficient gas escape from the soil, causing an increase of the CO_2 flux. Such a behaviour is mainly related to a Venturi effect due to a local condition of the

Nel Cannestrà station site, that cannot be extrapolated to other sectors of the volcano, considering that it was not observed in the Rina Grande station (see Fig. 2b for location) (Carapezza et al., 2009).

It is interesting to note that the four years-long dataset exhibits a declining long-term trend (Fig. 3b and annual average in Table 1) which can be likely viewed as a decreasing supply of CO_2-rich magma from the deeper to the upper plumbing system. This hypothesis is supported by the long-term trend observed in the CO_2 emissions from the plume, retrieved by combining CO_2/SO_2 ratioratios and SO_2 flux measurements (see Aiuppa et al., 2011). The decreasing trend of the soil CO_2 flux, marked by the annual average shifting from 920 (in 2007-2008) to 310330 g m^{-2} day^{-1}, does (in 2010-2011), is not appear on evident in the radon long-term signals that, instead, show an excellent stability around the average of 2000a slight increase from 1777 to 2264 Bq/m^3 in annual averages (see straight linelines in Fig. 3a and 3b, respectively). It is important to outline that, according to this view, the concurrent measurements of CO_2 and ^{222}Rn can be considered as an important tool for detecting the variation in time of the deep vs. shallow contribution to the soil gas, as it was found in other volcanoes (i.e. Etna; Giammanco et al., 2007Table 1).

However, during a short term period, it is clear that relative increases in CO_2 flux coexist with and/or are immediately followed by similar trends in ^{222}Rn activity. Generally, CO_2 flux shows a more stable signal during short-term periods, indicating that daily temperature variations have a feeble influence on the gas flux. Conversely, as pointed out by Carapezza at al. (2009), there is a definite correlation between winds (including both speed and direction) and soil CO_2 fluxes (Table 2). Winds toward the SE blowing at speed higher than 8 m/s cause a more efficient escape of the gas from soil. This is related to a Venturi effect due to a local condition that cannot be extrapolated to other sectors of the volcano (Carapezza et al., 2009).

~~This behaviour is evident by calculating the~~

4.2 – Short-term periodicity and long-term trends

In order to identify diurnal and semidiurnal cycles affecting the gas signals, we performed a spectral analysis (Power Spectral Density) over a one year subset of data (sample time = 1 hour), using the method suggested by Viveiros et al. (2014). The analysis identified the 12h and the 24h frequency peaks in both CO_2 flux and ^{222}Rn activity (Fig. 6), confirming previous findings (Perrier et al., 2009 and 2012; Rinaldi et al., 2012). In our case, the ^{222}Rn signal seems to be modulated by temperature and barometric changes, although we do not exclude that this periodicity could be related with solar tides (e.g., Steinitz et al. 2011). On the other hand, the soil CO_2 flux reveals a main 12h period. It is worth noting that such a behaviour slightly differs from previous results suggesting a major influence of temperature rather than pressure (Rinaldi et al., 2012).

By analysing the long-time series of soil CO_2 flux and ^{222}Rn activity, we performed a calculation of the mean value of the whole data ~~on~~for each specific day of the year~~. The~~ and the same computation was carried out also on soil and air temperature data. The emerging annual trend ~~from Jan 1 to Dec 31 (~~(see Fig. ~~5) remarks~~7a, b) highlights the inverse relation between temperature and soil gas release, as well as ~~the~~an apparent correlation between the two gas species. Overall, we observe a 100% increment of the mean values comparing the spring-summer with the fall-winter period. ~~Is~~It is also evident that the most significant day-by-day variations occur during the fall and spring season, when the likelihood of drastic atmospheric changes (i.e. heavy rainfall or windstorm) is higher than during summer~~ or winter~~.

The long-period behaviour is ruled by soil and air temperature (i.e. thermal gradient), whereas short-time~~/high frequency~~ oscillations are modulated by soil humidity (e.g~~.~~., rainy events) and wind conditions (speed and direction), respectively. The ~~value of the~~ correlation coefficients (R) shown in Table 2, ~~allows~~allow to better ~~evaluate~~assess the ~~effects~~effect of the environmental parameters that actively modulate the trends of ^{222}Rn activity and soil CO_2 flux ~~trends.~~.

~~In Fig. 6 we report~~

4.3 – Variation of the correlation coefficients

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The seasonal ~~time variation~~variations of average correlation coefficients of ~~some~~the main environmental parameters ~~and compare them~~ with ^{222}Rn activity and soil CO_2 flux~~. Here~~ are reported in Fig. 8, where seasons are gathered and simply subdivided in spring~~/~~-summer and fall~~/~~-winter subsets. It can be seen that ~~^{222}Rn shows positive correlation with soil water content and negative correlation with air and soil temperatures. Similarly, soil CO_2 flux is mostly correlated positively to wind speed and negatively to air and soil temperatures. However, as shown in Fig. 6, the correlation factors are~~ the correlation coefficients are not so stable throughout the investigated ~~period~~time span, but appear slightly modulated by seasonal effects. For example, both soil CO_2 flux and radon activity display a more distinctive negative correlation with air and soil temperatures during the spring~~/~~-summer subsets. ~~In addition, seasonal variations in geochemical data and environmental correlation coefficients are~~ This behaviour is likely due to the lack of drastic variations in weather conditions during the "dry" season at Stromboli Island. Hence, the correlation between temperature and gas flux and concentration is not perturbed by other atmospheric factors (e.g. soil humidity). Correlation coefficients seem somewhat different in the first subset (spring-summer 2007) compared to all the others~~. This~~; this subset (grey field in Fig. 8) was ~~sampled~~obtained just after the March 15, 2007 explosive paroxysm when the effects of environmental conditions on degassing dynamics, even in relatively distal areas, ~~were~~seem somehow weaker. In ~~fact, the~~ April-June 2007 ~~period~~

represents a time span characterized by the interruption of the lava effusion ceased and the Strombolian activity and by the ceasing of lava effusion.was not resumed, or more precisely, the source of explosions was too deep to allow glowing scoriae to reach the crater surface. In this time span -, there was still a remarkable degassing rate at the crater vents likely related to a continuous supply of undegassedcraters from a relatively deep-seated magma to the upper feeding system of the volcano (Burton et al., 2009; Carapezza et al., 2009; level (Aiuppa et al., 2011; Cigolini2009; Barberi et al., 20132009).

54.4 – *Statistical Treatmenttreatment*

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Two different statistical methods have been applied toon the entire datasetsraw dataset (sample time = 1 hour) of soil CO_2 flux and ^{222}Rn concentration in order to identify and remove gas variationsthe effects due to environmental parameters.

54.4.1 - Multiple Linear Regression Statistics (MLR)

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The data setsdatasets of radon concentration, soil CO_2 flux and environmental parameters have been analyzedanalysed by the Multiple Linear Regression (MLR) which is a simple and largely applied method used to identify the contributions of several independent variables and model the fluctuations observed in the investigated signal. The analysis has been performed to predict the values of a dependent variable (Y) given a set of predictor variables (X_1, X_2, …., X_n). The relationship between the dependent variable (Y_{calc}) and the predicted variables is expressed as

$$Y_{calc} = Y_0 + b_1X_1 + b_2X_2 + \ldots + b_nX_n \quad (1)$$

Y_0 is the intercept, X_n are the acquired variables and b_n the calculated regression coefficients (Granieri et al., 2003; Hernandez et al., 2004). Moreover, in; and references therein). In order to simplify and restrainreduce the number of predictor variables, MLR takes into account only the factors that are more correlated (positively or negatively) with CO_2 flux and ^{222}Rn activity (Table 2).3). By considering previous research, we selected only the environmental factors (i.e. independent variables) causing an increment of the R^2 greater or equal to 1% (Viveiros et al., 2008; Silva et al., 2015). Particularly, soil and air temperature were consideredindicated by the regression for both gas species, together with wind conditionsspeed and direction for CO_2 and soil humidity for ^{222}Rn. This statistical approach can be quickly performed, thus it can be easily applied on near real-time measurements.In Table 3 we report the main parameters for both gases by the MLR analysis.

Results show that the atmospheric variables taken into account for this analysis are able to predict 45% and 51% (R= 0.67 and R= 0.71) of the variations observed in soil CO_2 flux and ^{222}Rn concentration, respectively. Moreover, soil CO_2 flux and ^{222}Rn values show low dispersions, as respectively the 4.08% and 4.80% of the computed residuals exceed the average ±2 standard deviation (Table 3).range. Predicted values by MLR both for radon concentration and soil CO_2 flux, together with the observed values and the calculated residuals, are plotted in Fig. 7a9a onto the recorded time series. By looking at the residuals, it can be seen that *i)* the bell shape of the CO_2 flux is somehow smoothed due to the removal of the seasonal trend andbut it disappears totally in the last two years; whereas the bell shape still persists for radon, and *ii)* the residuals of both gases show peaks and major fluctuations that obviously cannot be duerelated to environmental variations. Moreover, in both cases the signals are still characterizedcharacterised by noisy components (similar conditions were also previouslyas reported by Viveiros et al., 2008; Carapezza et al., 2009; Laiolo et al., 2012 and Viveiros et al., 2014).

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~~Notably, this~~This statistical approach has been used at different volcanoes in the attempt to detect short-term variations in volcanic activity (e.g. major explosions at Stromboli; Laiolo et al., 2012), as well as the effects of seismic sequences (due to stress/strain structural changes) on the shallow part of a volcanic edifice (e.g., Masaya as ~~analyzed~~analysed by Padilla et al., 2014).

~~5~~4.4.2 - Principal Component Regression (PCR)

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The second statistical treatment applied ~~to ^{222}Rn~~to ^{222}Rn activity, soil CO_2 flux and environmental parameters is the Principal Component Regression (PCR). This method differs from the previous one in how predictors are treated: first~~,~~ a factor analysis is performed on the environmental dataset (*X*); then a forward step-wise linear regression of measured soil CO_2 flux and radon activity (*Y*) is performed on the estimated factors. The goal of this approach is firstly to obtain a reduction in the *X* data set in a way that maintains the maximum amount of information (i.e. largest possible variance), and secondly to perform regression of *Y* on orthogonal (uncorrelated) components. The aim is to ensure that highly correlated principal components are not overlooked (Vandeginste et al., 1998).

We ~~had to perform~~performed factor analysis on two separate datasets because the radon station measures soil temperature and air pressure, whereas soil CO_2 flux station also measures air and soil humidity, wind speed and direction. ~~So~~Therefore, we created one dataset for radon with soil temperature and atmospheric pressure ~~of~~ measured at the radon station and added the other variables ~~from~~measured at the CO_2 station. The factor analysis of the two datasets ~~show~~shows that three eigenvalues are higher than 1.0 and these three factors can explain the 73% of the total variance~~.~~ (Table 4). In the second step, we performed forward step-wise regressions of ^{222}Rn concentration and soil CO_2 flux on the first three factors. We obtained two theoretical models

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which explain the 25% and the 47% (R= 0.50 and R= 0.68) respectively of the soil CO_2 flux and ^{222}Rn concentration measurements variance.

However, as (Table 4b). As in the MLR model, residual values show low dispersion, being less than 5% the portion of the data that exceeds the mean ±2σ range (4.11% and 4.89% for CO_2 and ^{222}Rn, respectively) of the data that exceed the mean standard deviations (±2σ; cf. Table 3).). The timeseriestime series of observed, predicted and residual values of radon concentration and soil CO_2 flux are reported in Fig. 7b9b. An overall comparison of the latter with Fig. 7a9a shows that the two statistical treatments provide basically the same results.

65 – DISCUSSION

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*6.1 - Considerations on*For ^{222}Rn, the applied statistical *treatment*

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On the whole, the results obtained by applying the two methods are apparently very similar (Fig. 7) but they provide some significant differences. In fact, theindicate nearly the same percentage of the data that can be attributed to environmental variations is quite similar for ^{222}Rn, , whereas the predicted soil CO_2 flux values differvary from 45% in MLR to 25% in MLR and PCR methods, respectively (see Table 2). Variations in. The residual computed values are related to processes that bypass meteorological factors and are likely related to the volcanic system and occur either within the shallow hydrothermal aquifer or in the deep magmatic plumbing system.

However, itIt can be noted that the radon treatment provides treatments provide many significant negative residual values ≥ ≤ -2σ, whereas only one negative residual is recorded for soil CO_2 flux (see Fig. 810). This indicates that, according to the statistical treatments, the measured radon concentrations are frequently lower than those calculated (that include the effects of by filtering the effects of the environmental factors. We explain the high fluctuations showed by the residuals of radon signal with the relative low sensitivity of the radon dosimeter (Table S1) when settled with a high sampling rate (1 hour) in areas characterised by general low emissions (< 2000 Bq/m^3). In fact, such a noisy signal was not observed in the datasets acquired where the radon emissions are higher (Laiolo et al., 2012). Moreover, a trend characterised by positive and negative fluctuations in the calculated ^{222}Rn residual values, has been already observed in a non-volcanic area (Hayashi et al., 2015).

The residual time series (Fig. ~~7a, 7b~~9) retrieved for ~~CO_2~~ soil CO_2 flux ~~show~~shows in both treatments a decreasing trend for the first two years (visible also in the raw data) followed by a nearly steady-state signal close or below the zero value. This behaviour supports the hypothesis that the supply of CO_2-rich magma from the deep plumbing system (that started before the 2007 eruption, ~~cf.,~~ Aiuppa et al., 2011~~) likely increased~~), besides increasing CO_2 ~~emissions not only within~~emission in the gas plume itself, ~~but~~induced also ~~created~~ a higher CO_2 flux in ~~the~~more distal zones, which lasted for nearly two years.

Comparison of the standard residuals (both by PCR and MLR) for CO_2 soil flux and ^{222}Rn activity versus time (Fig. ~~8~~10) shows that in the initial two years of monitoring (up to May 2009) the two gases display a similar behaviour with nearly synchronous alternation of periods with anomalous emissions and periods with few or no anomalies (such as the summer of 2007 and 2008). In the last two years considered, only radon shows frequent positive anomalies whereas anomalous CO_2 flux values are only rarely recorded.

~~Finally, the~~The comparison between the two statistical treatments suggests that ~~the~~ MLR ~~seems to be the~~is more adequate ~~method~~ for getting the quick results needed in near-real time volcano monitoring, whereas PCR ensures a more accurate ~~estimation~~estimate of ~~warning thresholds~~data, which might be eventually useful for a more accurate post-process ~~analyses~~analysis and for a more reliable monitoring. ~~In this view, because of the more reliability of the PCR treatment, in the next section we briefly compare our emerging results with the 2007-2011 Stromboli volcanic activity.~~

~~*6.2 Diffuse degassing vs. volcanic activity*~~

In order to ~~test~~assess the reliability of radon activity and ~~CO_2~~soil CO_2 flux, measured in the distal site of Liscione/Nel Cannestrà, as possible ~~geochemical~~ precursors of major changes in the volcanic activity, the residuals time-series ~~of both species~~ obtained by PCR ~~method~~and MLR and exceeding 2σ, have been compared with the main volcanic and seismic events occurred at Stromboli ~~volcano during~~in the ~~considered timespan~~same time span (Fig. ~~8). During the~~ 10).

The 4.5 years of gas monitoring (April 2007 - September 2011) represent a phase of ordinary volcanic activity of Stromboli. In this period, twelve major explosions, four minor lava overflows and a local earthquake (with M_L= 2.2) were recorded at Stromboli by the INGV monitoring system (~~cf.~~ Calvari et al., 2014). ~~It~~No explosive paroxysm or effusive eruption occurred. There is no clear correlation between our data and the recorded volcanic events, but some useful considerations can be done.

From Fig. 10, it can be seen ~~from Fig. 8~~ that in the first two years (following the 2007 explosive ~~paroxysm), periods of~~ and effusive eruptive phase), frequent and high CO_2 flux and ^{222}Rn anomalies ~~are essentially concordant~~were recorded in coincidence with ~~periods of~~some anomalous volcanic ~~activity~~episodes. Actually, the high number of positive residuals, from October 2007 to June 2008 and from November 2008 to May 2009, coincide with five major explosions and one lava overflow. ~~In~~During the ~~summer of~~ 2007, 2008 and 2009~~, though no~~

radon data are summers, neither major explosion/lava overflow nor residual CO_2 flux peaks were recorded (very few for radon, apart from 2009 summer when data were not available, no anomalous gas emissions and no major explosions were recorded. However in the). In summer of 2010, two major explosions occurred in a period of no anomalous gas release. Finally, during September During December 2010 August – February 2011, a major explosion and threetwo lava overflows occurred during in concurrence with isolated peaks of CO_2 flux and during a phase of anomalous ^{222}Rn emission. Also the most-recent lava overflow in September 2011 coincided with an anomalous emission, with only few isolated peaks of soil CO_2 fluxradon activity value.

7Conversely, we have to consider that the eruptive events that occurred in the above time span seem to be connected to minor changes associated to the dynamics of the upper part of the conduit (e.g., Barberi et al., 1993 and 2009) without any involvement of the deep seated gas-rich magma pockets (typically occurring during major effusive-explosive cycles of Stromboli volcano, such as those of 2002-2003 and 2007).

6 - CONCLUSIONS

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The presented data show that therefer to more than four years of soil gas measurements (^{222}Rn concentration and soil CO_2 flux) at relatively distal sites from the active vents (Liscione and Nel Cannestrà sites, located on the NE flank of Stromboli) are slightly influenced by volcanic activity that, in, Fig. 1) during a the time-span considered, did not produce any relevant explosions or(April 2007–September 2011) without major lava effusion. effusions and paroxysmal explosions.

The long-time averages for CO_2 flux and radon concentration exhibit relatively low values (585 g m^{-2} day^{-1} and 2050 Bq/m^3, respectively) when compared to those measured at the summit crater area (Carapezza et al., 2009; Cigolini et al., 2009 and 2013). This means that the advective processes, able to enhance the gas release from soil, are considerably reduced moving away from the crater area. However, theThe long term declining trend observed for the soil CO_2 flux (Fig. 9 and Table 1) suggests that the large supply of CO_2-rich magma associated with the 2007 eruption (and invoked to explain the exceptional CO_2 emissions from the plume; Aiuppa et al., 2009 and 2011) affected also the soil gas release in relatively distal areas. So, as already stressed by De Gregorio et al. (2014) for Etna volcano, the soil CO_2 flux measurements represent a key tool to infer the magma supply dynamics and to evaluate the local degassing regime. Furthermore, the combination of soil CO_2 flux and ^{222}Rn concentration measurements can better constrain the gas source in relation to changes in volcanic activity (Faber et al., 2003; Perez et al., 2007; Padilla et al., 2013). , e.g., Aiuppa et al., 2011) affected the soil gas release in distal areas as wellIn the last two years (2010 - 2011) of our monitoring, anomalous radon concentrations have been frequently recorded in periods with rare or absent soil CO_2 flux anomalies; this likely indicates a different source for the two gases, deeper for CO_2 and somehow shallower for radon.

As recently stressed by De Gregorio et al. (2014), measuring the soil CO_2 flux at active volcanoes is a valued tool to infer the magma supply dynamics. Thus, combining CO_2 flux and ^{222}Rn concentration measurements gave us the opportunity to better investigate the changes in volcanic activity associated with magma rise.

The four years monitoring of both gas species at Stromboli provided the opportunity of better decoding how gaseous transfer toward the surface is ruled by environmental changes. Our data show that both gases are affected by seasonal temperature variations that givegiving to the time series a trend with a bell shaped profile. In particular, higher emissions occur during fall-winter, because fluid convection is promoted by the higher soil-air temperature gradient. Conversely, during summer, this gradient is reversed and near-surface convection is inhibited. Moreover, soil CO_2 flux is locally influenced also by wind speed, and(>8 m/s in the SE sector), while radon activity by soil humidity.

In summary, the combined effect of decreasing surface temperaturestemperature, eventually coupled with increases in soil moisture, seems the main factor that controls the variations of radon emissions. The effect of soil humidity on radon activity probably reflects the adopted measurement techniques. In fact the radon measurements at 1 m depth are likely affected by soil humidity (particularly during the raining falls) which affects the radon diffusion and exhalation rates (i.e., Papachristodoulou et al., 2007).

We have shown that the influence of environmental parameters on gaseous timeseriestime series can be removedminimised by means of linear statistics and give us the opportunity to better track the evaluate possible variations related to changes in volcanic activity. The statistical analysesmethods presented in this paper can be adopted for different purposes. Particularly,; the Principal Component Regression (i.e. Factor Analysis) appears to be the better suitedmore suitable for an accurate analysis of large datasets following major changes in volcanic activity (post-event data processing): in fact, the application of this method carefully evaluates the contribution of each independent factor by means of precise cross correlations. Conversely, Multiple Linear Regression analysis can be more quickly and easily applied duringto a nearly real--time soil gas monitoring. This useful in volcano surveillance since it gives us the possibilityopportunity to efficiently track anomalies inanomalous gas concentrations, or fluxes, that are not necessarily related to environmental factors. We thus emphasize that the reported datasets represent a rather unique case, at the global scale as well, of geochemical and environmental data acquired in a very active volcanic area for such a long time. The monitored area represents an anomalous degassing zone (Carapezza et al., 2009; Cigolini et al., 2013) and our results show that a multiparametric geochemical monitoring may play a key role in decoding precursory signals related to major changes of Stromboli volcanic activity.

We finally emphasize that the monitored area has been characterized by marked anomalous radon emissions prior and during the paroxysmal explosion of March 15, 2007 (Cigolini et al., 2013). Therefore, it is not excluded that multiparametric geochemical monitoring may play, in the future, a key role in decoding precursory signals related to major explosive events that affect the NE sector of Stromboli.

Acknowledgments

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Figure Captions

Fig. 1. Digital Elevation Model of Stromboli island (Island (from Baldi et al., 2005) with the overprint of the major faults and collapsed sectors (simplified from Finizola et al., 2002 and Tibaldi et al., 2009). The locationLocations of the Volcano Observatory (COA) and of ^{222}Rn activitythe radon and soil CO_2 flux stations is also indicatedare reported.

Fig. 2 (a) Map of radon activity measured in March 10-18, 2007 on the NE flank of Stromboli. The pointsFull circles indicate measurement sites; the contour lines of radon emissions have been obtained by kriging (Cigolini et al., 2013). TriangleThe triangle indicates the location of the ^{222}Rn automatic station. Dotted white rectangle marks the area of the CO2 soil CO_2 flux map of March 2007 reported in b) and where stars). Stars are the sites of the automatic CO_2 stations (Carapezza et al., 2009).

Fig. 3. Time-series of daily averages of radon activities^{222}Rn activity (a) and soil CO_2 fluxesflux (b) recorded hourly from April 2007 to September 2011. (grey dots). Black curves and the straight dotted lines represents the daily and the annual average, respectively. Histograms show the multi-modal distributions.

Fig. 4. Time-series of the daily mobile average of the main environmental parameters measured hourly from April 2007 to September 2011 at NC station. (grey dots). Black curves are the daily average.

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Fig. 5. Soil CO_2 flux vs. wind direction. Red and blue circles refer to data acquired with wind speed above or below 8 m/s respectively. Note that most of the high soil CO_2 fluxes are recorded for wind speed >8m/s in the SE sector.

Fig. 6. Spectral amplitude for soil CO_2 flux (a), ^{222}Rn concentration (b), atmospheric pressure (c) and air temperature (d). The analyses were made over one year of hourly data (see text for details).

Fig. 7. Bulk annual trend of ~~Radon~~radon concentration and soil CO_2 flux (a) retrieved from the mean values measured each day in the 4 years monitoring. Results are compared with the annual trend of soil and air temperatures (b).

Fig. ~~6~~**8**. Seasonal variations of the average correlation coefficients (R) between main atmospheric factors and ^{222}Rn concentration (above) and soil CO_2 flux~~.~~ (below). Air T: air temperature; Soil T: soil temperature; Air P: barometric pressure; Soil H: soil humidity; ~~Wind Sp: wind speed; Wind Dir: wind direction.~~Air H: air humidity. SS and FW refer to Spring-Summer and Fall-Winter period, respectively.

Fig. ~~7~~**9**. Results from Multiple Linear Regression (a) and Principal Component Regression (b) for radon activity (above) and soil CO_2 flux (below) during the 4 ½ years of monitoring. Data are reported as daily ~~average~~averages. The observed, calculated and residual values are ~~shown~~indicated.

Fig. ~~8. Plot~~**10.** Time series of the ~~hourly calculated~~ residuals of ^{222}Rn ~~(a)~~concentration and soil CO_2 flux ~~(b)~~ obtained by MLR (a) and PCR (b) methods with indication of the main volcanic and seismic events ~~recorded~~occurred at Stromboli from April 2007 to September 2011~~ (see legend). The residuals were obtained by the results of PCR outcoming from about 95% of the dataset.~~. Vertical axes express the standard deviation from the mean; only values ≥ 2 ~~sigma~~σ are plotted. Grey ~~filed~~field marks the time span ~~where ^{222}Rn~~when no radon data ~~are not available.~~

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Table
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Table 1. Descriptive statistics of the main parameters acquired by the ^{222}Rn and CO_2 stations from April 2007 to September 2011

		average	**SD**	**max**	**min**	**data no.**
Rn station (LSC site)						
^{222}Rn	*Bq m^{-3}*	2051.00	1196.00	7879.00	0	35,178
Soil T (-1m)	*°C*	17.44	5.70	30.09	5.90	35,178
Air P	*hPa*	951.38	6.40	974.37	925.16	35,178
CO_2 station (NC site)						
Soil CO_2 flux	*g m-^{2} day^{-1}*	585.45	643.17	7021.72	10.92	39,412
Air RH	*%*	77.62	16.02	100.00	12.36	39,412
Air T	*°C*	16.45	6.55	38.78	0	39,412
Air P	*hPa*	954.63	5.66	973.45	921.69	39,412
Soil T (-0,5m)	*°C*	19.87	5.04	28.86	11.89	39,412
Soil RH (-0,5m)	*%*	12.30	2.11	21.38	8.63	39,412
Wind direction	*°N*			359	0	39,412
Wind speed	*m s^{-1}*	3.67	2.96	21.91	0.02	39,412

Annual averages	***Apr. 2007 - Mar. 2008***	***Apr. 2008 - Mar. 2009***	***Apr. 2009 - Mar. 2010***	***Apr. 2010 - Mar. 2011***
^{222}Rn activity (Bq m^{-3})	1777.4	1878.1	2158.4	2264.0
Soil CO_2 flux (g m-^{2} day^{-1})	919.9	658.0	406.9	329.5

Table
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Table 2. Correlation coefficients (R) between the daily average values of the parameters measured at the monitoring stations from April 2007 to September 2011

		^{222}Rn station (LSC site)			**CO_2 station (NC site)**							
		^{222}Rn	Soil T	Air P	Air RH	Air T	Air P	CO_2 flux	Soil T	Soil RH	Wind direction	Wind speed
^{222}Rn	*Bq m^{-3}*	1.000	-0.736	-0.282	0.387	-0.749		0.346		0.737	-0.104	0.225
Soil T	*°C*	-0.736	1.000	0.320	-0.360	0.874		-0.532		-0.745	0.171	-0.195
Air P	*hPa*	-0.282	0.320	1.000	-0.305	0.330		-0.204		-0.447	0.222	-0.251
CO_2 flux	*g m-^{2} day^{-1}*	0.346			0.187	-0.541	-0.090	1.000	-0.566	0.370	-0.298	0.431
Air RH	*%*	0.387			1.000	-0.489	-0.228	0.187	-0.304	0.517	-0.089	0.021
Air T	*°C*	-0.749			-0.489	1.000	0.155	-0.541	0.867	-0.772	0.083	-0.231
Air P	*hPa*	-0.114			-0.228	0.155	1.000	-0.090	0.125	-0.295	0.205	-0.227
Soil T	*°C*	-0.694			-0.304	0.867	0.125	-0.566	1.000	-0.713	0.198	-0.193
Soil RH	*%*	0.737			0.517	-0.772	-0.295	0.370	-0.713	1.000	-0.120	0.211
Wind direction	*° N*	-0.104			-0.089	0.083	0.205	-0.298	0.198	-0.120	1.000	0.148
Wind speed	*m s^{-1}*	0.225			0.021	-0.231	-0.227	0.431	-0.193	0.211	0.148	1.000

Table
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Table 3. Summary of the Multiple Linear Regression analysis (MLR) on the acquired dataset

	Coefficient B	Standard Error of B	Coefficient β	t-test	p-level
Dependent variable = soil CO_2 flux; Data n°. = 39,412					
Intercept	2284.44			67.27	0.00
Air T	-27.51	0.07	-0.43	-36.31	0.00
Soil T	-35.27	0.05	-0.44	-37.91	0.00
Soil RH	-48.43	0.02	-0.45	-26.20	0.00
Wind direction	-1.47	0.98	-0.40	-52.63	0.00
Wind speed	81.41	0.03	-0.31	96.98	0.00
Dependent variable = ^{222}Rn activity; Data n°. = 35.178					
Intercept	-6190.63		-1.79	-7.93	0.00
Air T	7.66	0.03	0.44	-21.47	0.00
Soil T	-38.02	0.03	0.42	-45.24	0.00
Atmospheric P	-48.60	0.03	0.42	10.01	0.00
Soil RH	198.10	0.01	0.51	53.95	0.00

Table 4. Application of the PCR statistical method

A. Environmental variable weights on factors

Variable	**Factor1**	**Factor2**	**Factor3**	**Factor4**	**Factor5**	**Factor6**	**Factor7**
Air RH	**0.59**	-0.16	-0.05	0.55	0.43	-0.07	-0.07
Air T	**-0.92**	-0.03	-0.25	0.05	0.04	0.17	-0.26
Air P	-0.34	-0.06	**0.82**	-0.19	0.39	0.10	0.00
Soil T	**-0.85**	0.08	-0.22	0.34	0.13	0.20	0.22
Soil RH	**0.89**	-0.03	0.03	0.00	-0.12	0.44	-0.01
Wind dir.	-0.06	**0.77**	0.40	0.39	-0.28	-0.02	-0.04
Wind speed	0.27	**0.72**	-0.36	-0.34	0.41	0.02	0.00
Eigenvalue	2.900	1.147	1.076	0.854	0.621	0.276	0.126
Proportion	0.414	0.164	0.154	0.122	0.089	0.039	0.018

B. Summary of the Principal Component Regression analysis (PCR) on the acquired dataset

	Coefficient B	**Standard Error B**	**Coefficient β**	**t-test**	**p-level**
Dependent variable = soil CO_2 flux; Data n°. = 39,412					
Intercept	520.71	2.63		198.05	0.00
Factor1	296.44	2.63	0.49	112.75	0.00
Factor2	61.69	2.63	0.10	23.46	0.00
Factor3	-35.21	2.63	-0.06	-13.39	0.00
Dependent variable = ^{222}Rn concentration; Data n°. = 35,178					
Intercept	1979.91	4.20		470.97	0.00
Factor1	779.48	4.20	0.68	185.42	0.00
Factor2	-230.39	4.20	-0.20	-54.80	0.00
Factor3	94.85	4.20	0.08	22.56	0.00

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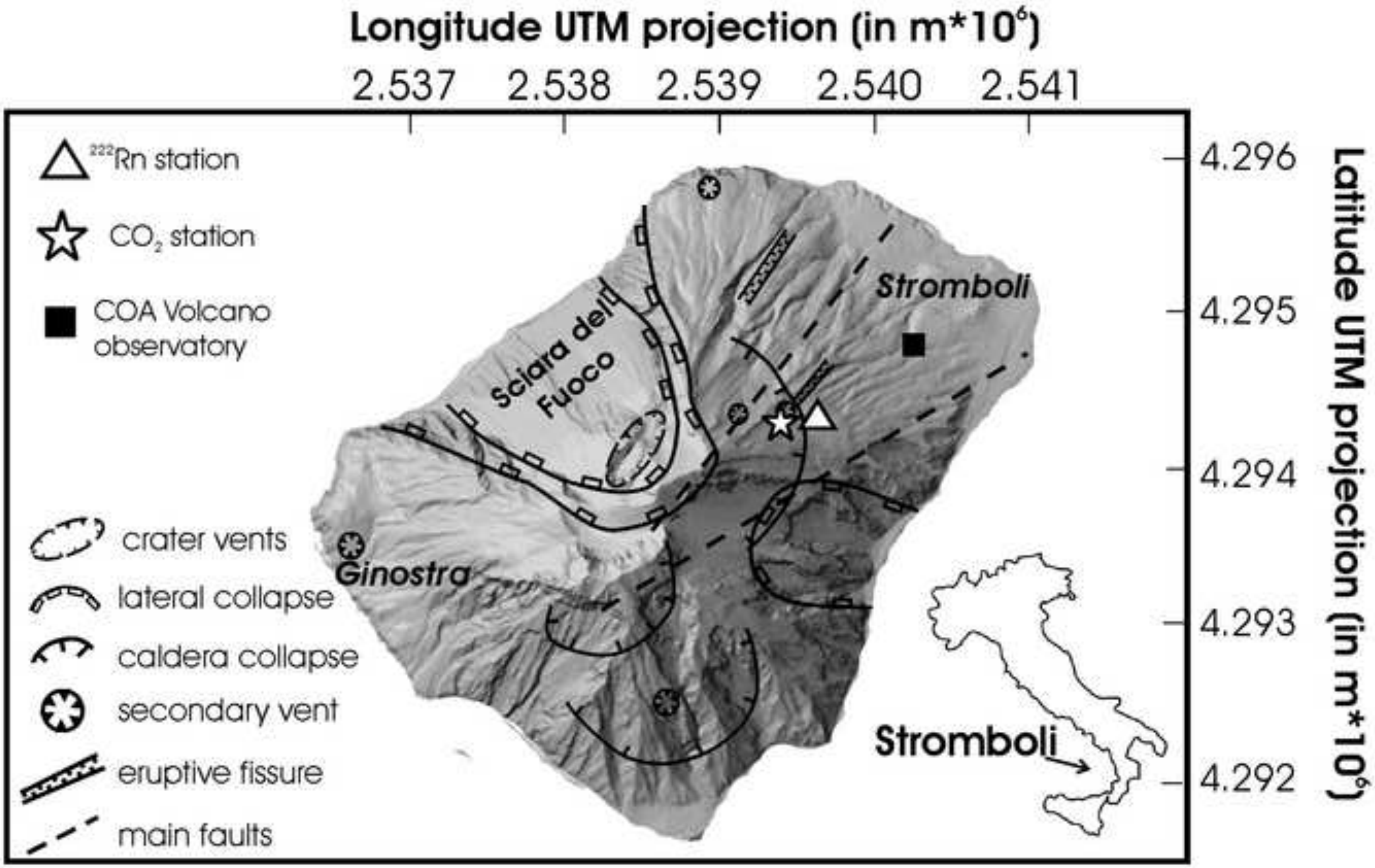

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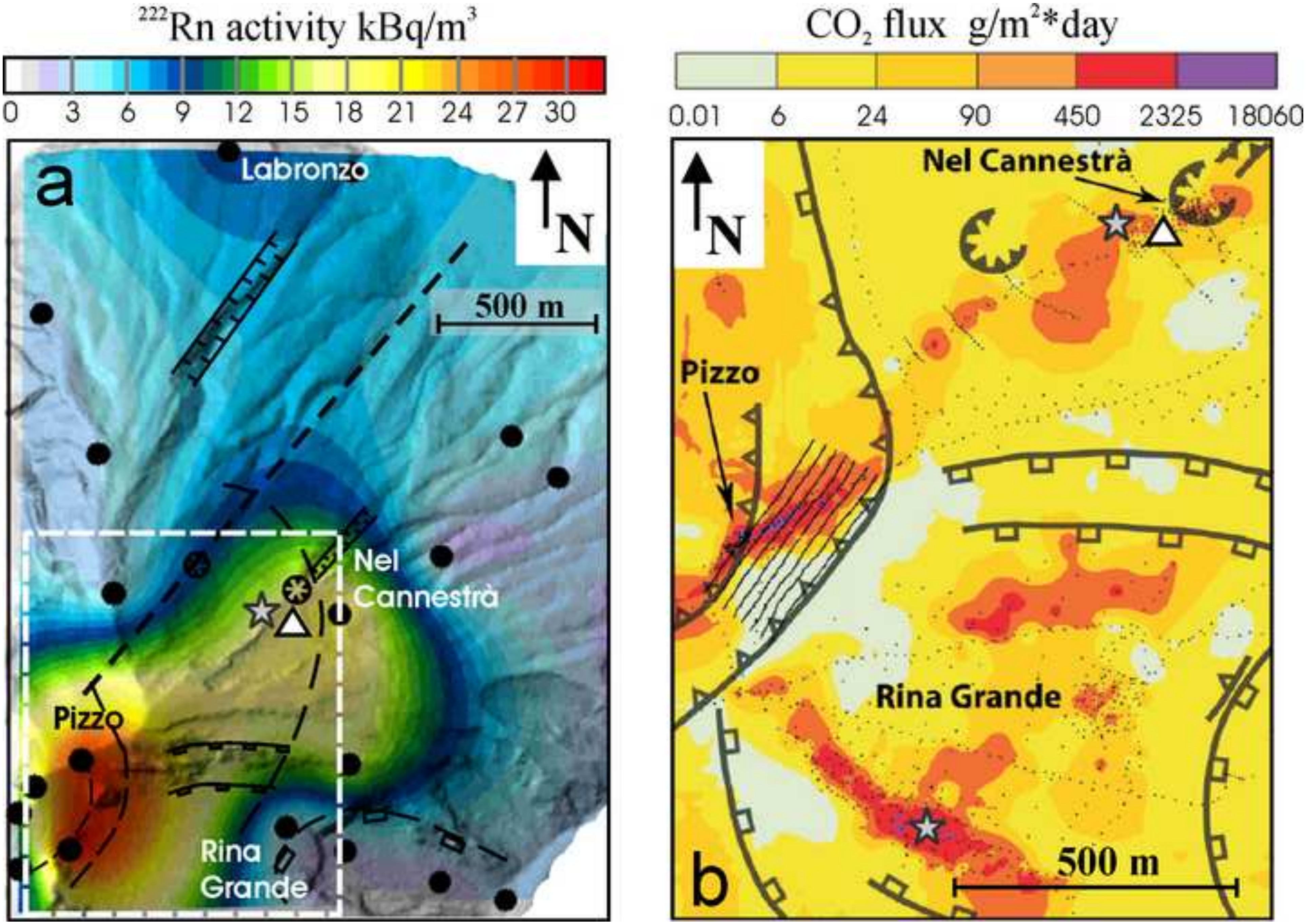

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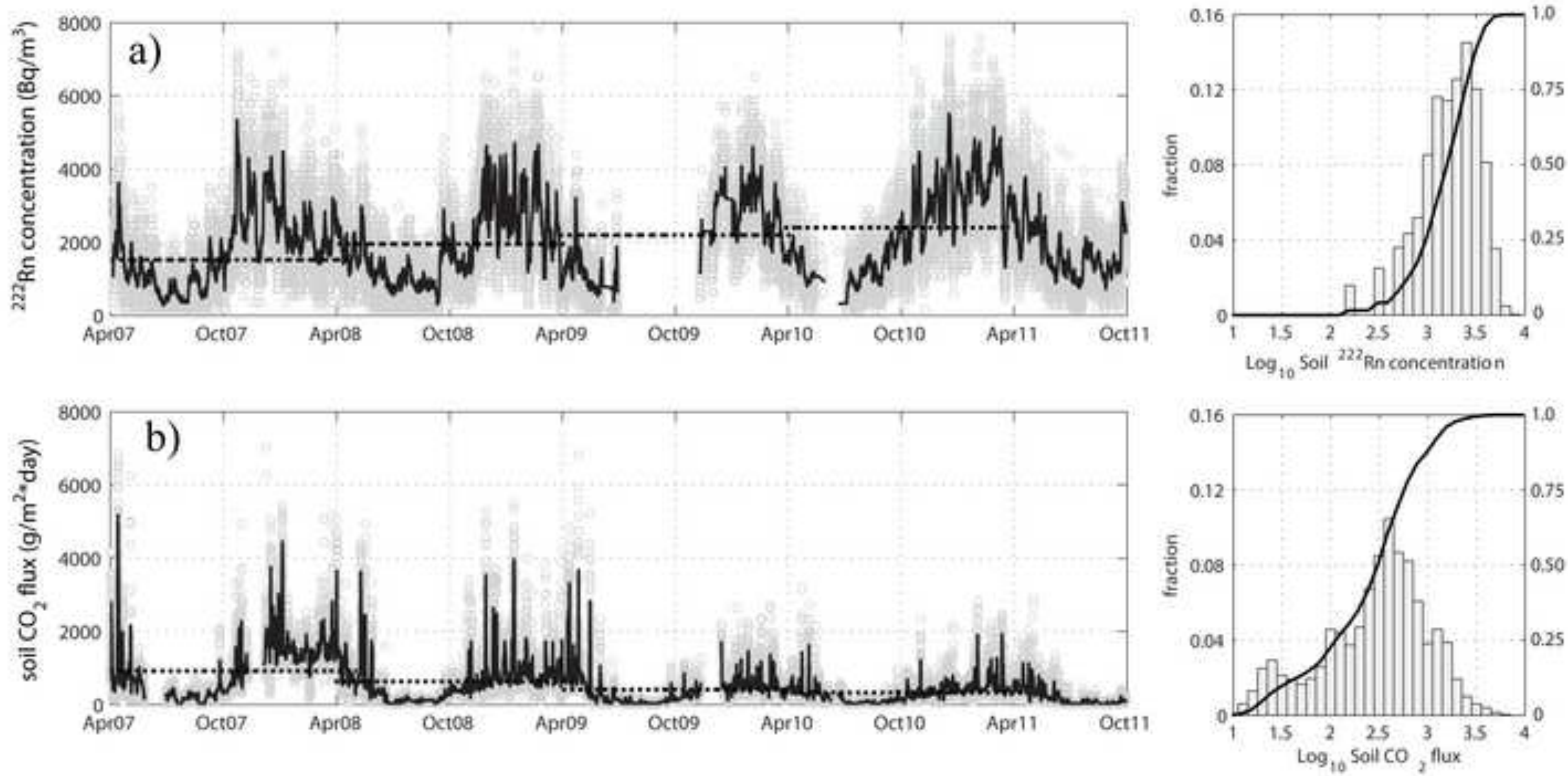

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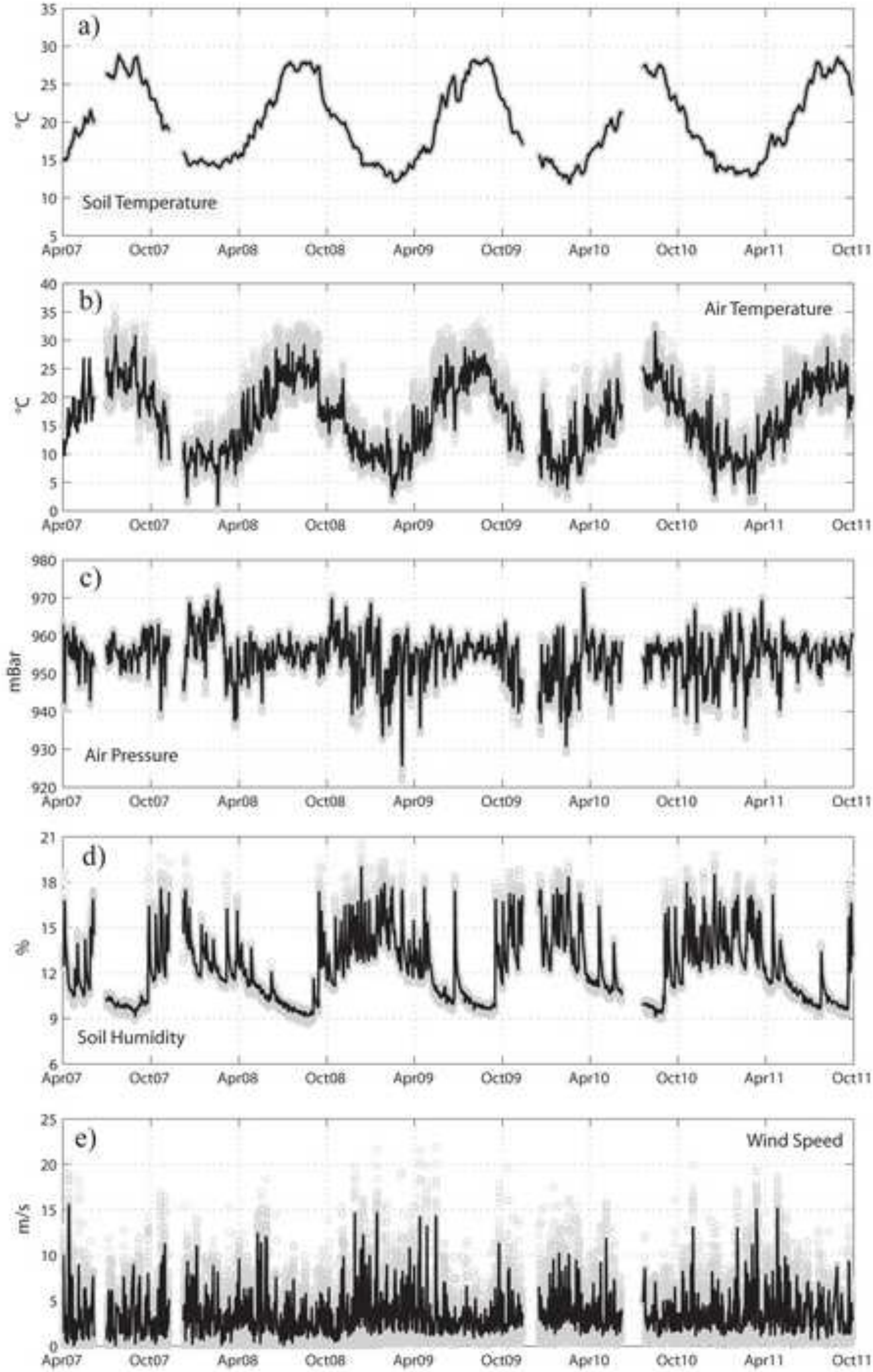

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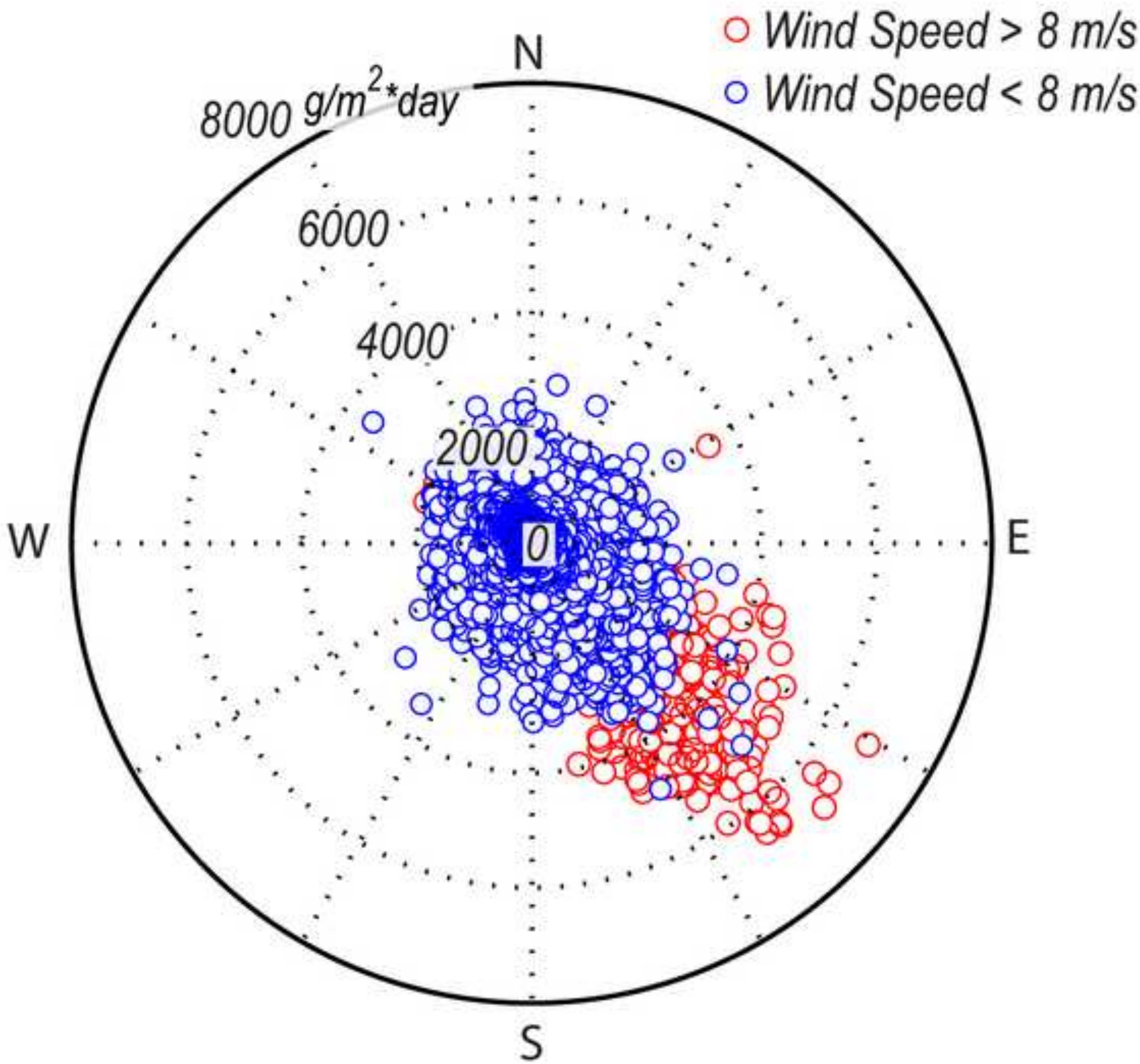

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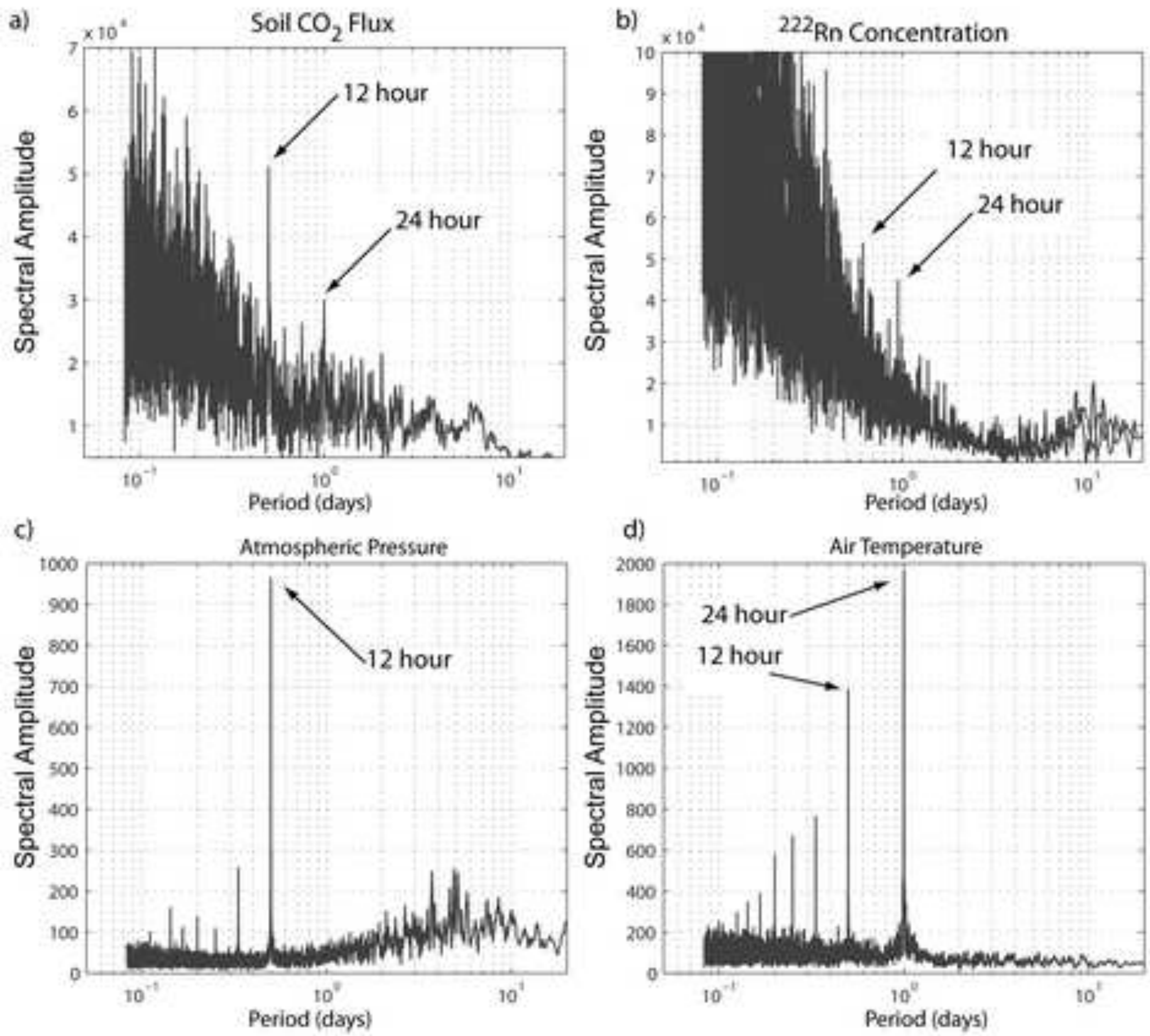

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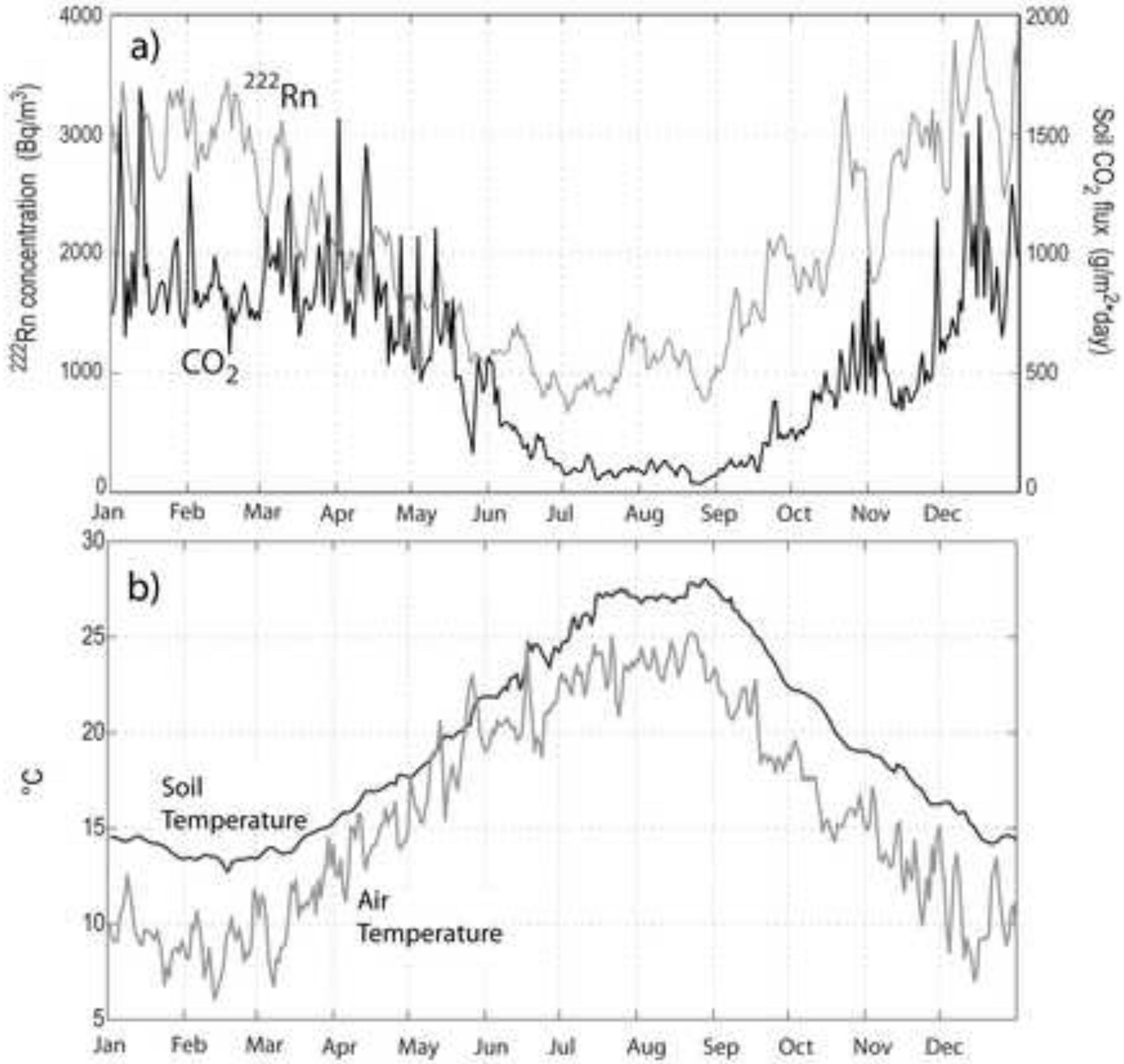

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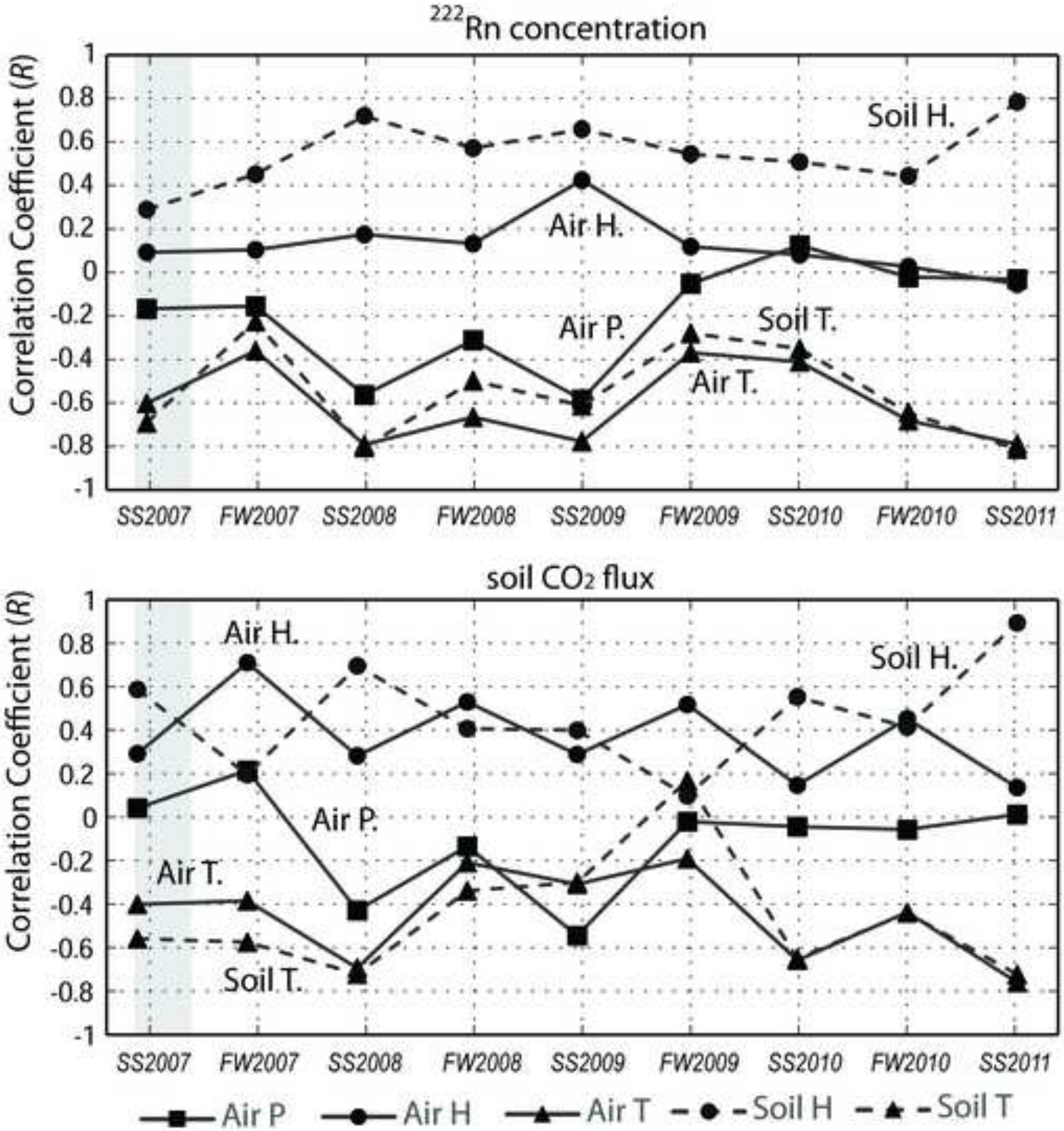

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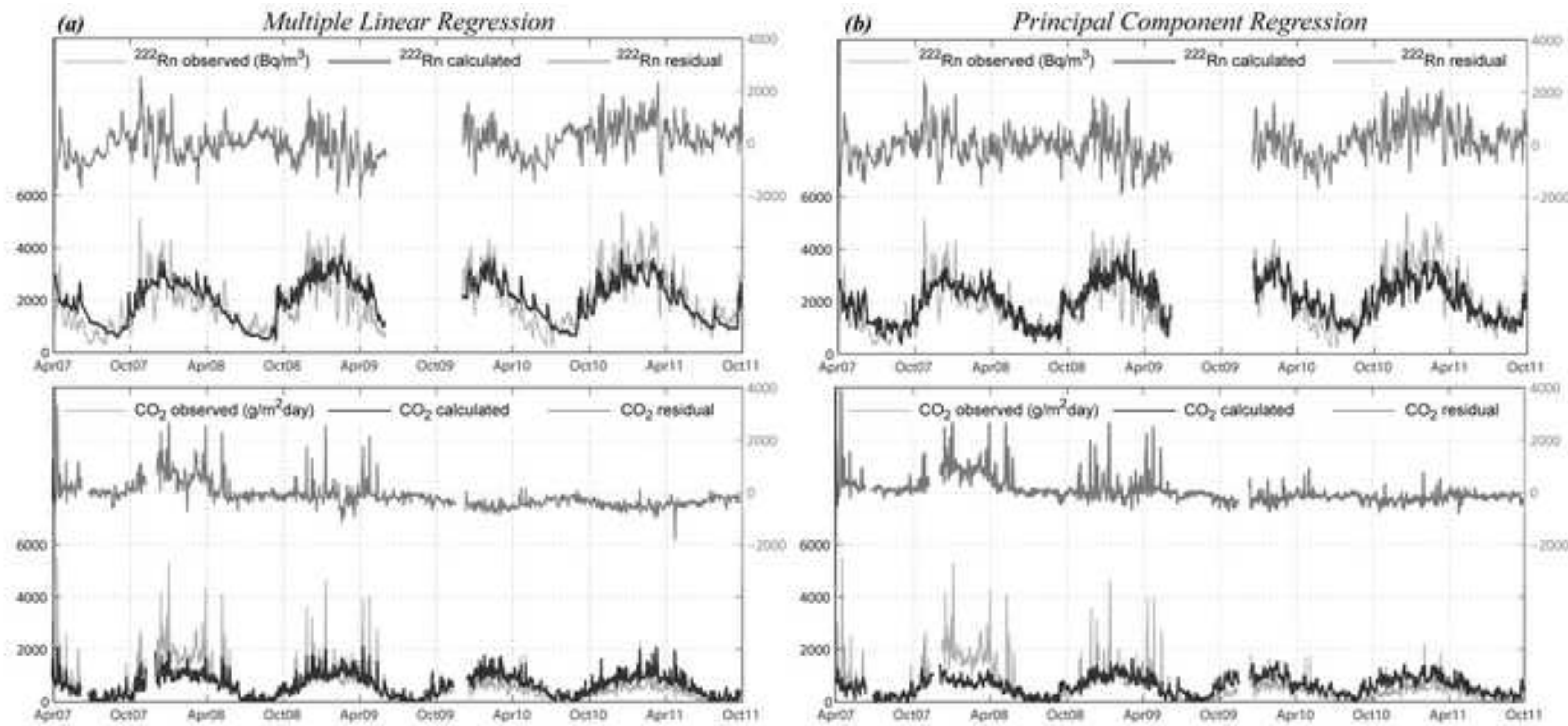

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