Journal of Cleaner Production 234 (2019) 881–894

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Life cycle assessment of atmospheric emission profiles of the Italian geothermal power plants☆

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A R T I C L E I N F O

Article history:
Received 21 March 2019
Received in revised form
17 June 2019
Accepted 20 June 2019
Available online 25 June 2019

Handling Editor: Prof. Jiri Jaromir Klemeš

Keywords:
Geothermal energy
Atmospheric emissions
Life cycle assessment
Uncertainty analysis
Geo-thermoelectric power plants
AMIS abatement system

A B S T R A C T

After nearly a decade of only small development in capacity in deep geothermal sector in Europe, in recent years a resurgence of interest in geothermal power and the use of innovative technologies to increase and better exploit geo-thermoelectric generation has stolen the limelight from the scientific community. Differently from other types of energy sources, the environmental impacts determined by geothermal exploitation are extremely dependent on the geographical location. Life Cycle Assessment offers a powerful methodological approach for the investigation of the environmental footprint of power generation systems.

Focusing on an unprecedented system-modelling approach for the investigation of an environmental impacts analysis of geo-thermoelectric activity in the Tuscany Region, Italy, in this work we perform a comprehensive environmental impact assessment for the calculation of atmospheric emissions profiles connected with the operational phase of the power plants. A clustering of all the geothermal installations in operation nowadays is performed by considering geographical representativeness This allows the identification of regional geothermal subareas. Moreover, an extensive data processing analysis is implemented with the aim of reconciling the great variability found among data collected. Results demonstrate that the efforts undertaken by the operator of the geothermal power plants to limit the impact of emissions, through abatement systems like AMIS, are quite effective. Indeed, in areas where mercury and ammonia concentration in fluids constitute a problem to deal with, nowadays the emissive patterns result comparable to the other ones. Notwithstanding, mercury and ammonia emissions, mainly emitted through the cooling towers, still represent a critical problem for all the geothermal fields. On the basis of our findings we conclude that potential chemical interactions and environmental impacts related to the variety of the compounds emitted should be object of future research and a further effort to minimize them.

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1. Introduction

Geothermal energy has been perceived as a convenient source for electric energy production only on a local scale so far, as just few areas in the world have enough geothermal potential to exploit it. Italy, Iceland, some U.S. States, Indonesia, Philippines, New Zealand are some of the countries that have already benefited from its exploitation. In recent years things have changed, and geothermal energy is now considered as one of the most promising renewable energy sources for producing electricity and heating. This is also proven by significant investments that are being made at international level: in fact, new technologies could allow the exploitation of reservoirs that would have been impossible to use in a cost-effective way until now (very deep drilling, binary cycle for low temperature fields, Enhanced Geothermal System). So far, environmental concerns perceived by the community have been one of the important barriers especially for deep geothermal market development. In this context, nowadays decision-makers require

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☆ This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

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https://doi.org/10.1016/j.jclepro.2019.06.222
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Abbreviations

AMIS	Abatement System for Mercury and Hydrogen Sulphide
EGS	Enhanced Geothermal System
g/h	Grams per Hour
g/MWh	Grams per Mega Watt hour
g/y	Grams per Year
GHG	Greenhouse Gas
GWe	Giga Watt electricity
GWh/y	Giga Watt hour per year
LCA	Life Cycle Assessment
LCI	Life Cycle inventory
LCIA	Life Cycle impact Assessment
MWe	Mega Watt electricity
MWhe	Mega Watt hour electricity
NCG	Non-Condensable Gas
ORC	Organic Rankine Cycle
W/	With
W/O	Without

more reliability in the environmental performance assessment of the power plants. In fact, differently from other types of energy sources, the environmental impacts determined by geothermal exploitation are extremely dependent on the geographical location. Concerning the global panorama of the geo-thermoelectric market, traditional hydrothermal flash power plants still dominate in terms of installed capacity all over the world, because of the greater electrical production that such technology can generate compared to others. In fact, according to the World Geothermal Congress survey, in 2015 only 1.8 GWe of the total 12.6 GWe world installed capacity was represented by binary power plants, while innovative enhanced geothermal technologies (EGS) were just not representative. Moreover, concerning the produced electrical geothermal energy in that year, only 12% was obtained from binary power plants. (Bertani, 2016). Nevertheless, the multiple technological solutions available today have put geothermal energy into renewed attention by the scientific community. Many topics have been investigated, from countries' geothermal potential to technical innovations to the environmental impact of these power plants. This latter issue is the one that in Italy is becoming more explored and even more discussed for the social impact on the population involved (Borzoni et al., 2014; Pellizzone et al., 2019, 2017). Historically, Italy is the country that first exploited this renewable energy, in fact, it was the major geothermal produces in the world in 2005 (Bertani, 2011). Recently, many countries have invested in this energy source in Europe, sometimes overtaking Italy: for example, nowadays Turkey is the leader country for installed capacity with 1.3 GWe (EGEC Geothermal, 2018). Actually, the possibility to increase the geothermal production largely depends on the perception of the community and the determination of decision-makers requiring more reliability in the environmental performance assessment of the power plants. Differently from other types of energy sources, the impacts determined by geothermal exploitation are extremely dependent on the geographical location, especially for what concerns the operative phase and the reservoir exploited which determine the peculiarity of the power plant's emission profile.

Life Cycle Assessment (LCA) is acknowledged as the most powerful methodological tool for the evaluation of the environmental performances of power generation systems (Rossi et al., 2019; Peng et al., 2013; Turconi et al., 2013; Parisi et al., 2013; Bravi et al., 2010; Brown and Ulgiati, 2002) and for the investigation of potential impacts associated with new projects prior their construction, thus allowing definition of the best strategies for mitigation of environmental emissions or even annihilation. Indeed, there are many studies available in the scientific literature reporting detailed life cycle inventory data enabling for an accurate description of the investigated systems and allowing also for the development of sophisticated parametrized model and predictive LCAs (Pehl et al., 2017; Padey et al., 2013, 2012). In the field of geothermal energy, the scientific literature is lacking in LCA studies providing primary data. In fact, just few studies on geothermal power plants are available and the studies focused on the assessment of the environmental profile of working power plants are even fewer (Bravi and Basosi, 2014; Buonocore et al., 2015; Karlsdóttir et al., 2015; Parisi and Basosi, 2018). Most of the LCA studies on geothermal systems employ data coming from the literature or indirect and not pertinent secondary data (Marchand et al., 2015; Martínez-Corona et al., 2017). Such scarcity of specific information is also due to the fact that geothermal exploitation can be performed with different technologies (flash, dry steam, binary) and for different purposes (electricity, heat or both) (Martin-Gamboa et al., 2015; Ruzzenenti et al., 2014), making the collection of primary data much more difficult compared to other power generation systems. Several authors have also performed reviews (Bauer et al., 2008; Bayer et al., 2013; Menberg et al., 2016; Tomasini-Montenegro et al., 2017) and harmonisations (Asdrubali et al., 2015; Sullivan et al., 2012, 2010) of previous LCA studies on geothermal energy production in which they clearly underline the scarcity of accurate data and variability of information that prevent the definition of reliable eco-profiles of geothermal systems (Lacirignola et al., 2014, 2017).

The analysis proposed in this work tries to increase the knowledge and reduce data scarcity for the geo-thermoelectric activity in Italy by analysing the emission data available for all geothermal power plants operating in the Tuscany Region in a range of 10 years of analytical determinations collected by ARPAT (Tuscany Regional Agency for Environmental Protection). Focusing on an unprecedented system-modelling approach for the investigation of an environmental impacts analysis of geo-thermoelectric activity, we perform a comprehensive assessment of atmospheric emissions profiles representative of the actual situation in all the Tuscany geothermal areas. An extensive data processing analysis is implemented with the aim of reconciling the great variability found among data collected during the whole time series. Moreover, a clustering of geothermal installations in the Tuscany Region is performed by considering geographical representativeness This allows identification of regional geothermal subareas and calculation of environmental footprints connected to the operational phase of all the power plants in operation nowadays. As pointed out by the NREL report (Eberle et al., 2017) in which a systematic review of 180 papers on LCA of geothermal power plants worldwide reveals how the field location heavily influences the greenhouse gases' (GHGs) emissions, the large variety of environmental footprint calculated for geo-thermoelectric power plants is significant. Likewise, the technology implemented for the exploitation of geothermal energy deeply characterizes the eco-profiles of power plants, as showed in the same report by disaggregating the contributions to the various life cycle phases.

This study is in no way intended to be an ecotoxicological review, as results obtained from an LCA study are not suitable to be used for that purpose. The authors' goal is to evaluate the potential atmospherics environmental impact generated by the geo-thermoelectric activity in Tuscany, employing all the available information, thus extending the analysis published by Bravi and Basosi (2014) in terms of geographic dimension and data quality on the basis of the availability of larger amount of data in the historical series. To this aim, a rigorous statistical approach is adopted in order to obtain precise environmental profiles.

In addition, data presented here are a novel addition to the scientific literature of the geothermal field. The purpose is to obtain the most complete source of information about the emissions generated in atmosphere by deep geothermal exploitation of electric power generation plants in Italy, which nowadays is probably the most long-established region for geo-thermoelectric energy source in the EU. The different geochemical characteristics of the fields cause the impacts of this energy source strongly dependent on the location, in addition to the technology employed. Thus, it is hard to find estimated emissions which reflect the real emission profile and management activities of a geothermal power station. A current assessment of the concise and detailed emission profile of such productive systems is essential to ensure sustainable development of these technologies, especially considering the social aspects involved in the projects under development (Dumas and Angelino, 2016). Also, the aim of this study is to propose a protocol for the evaluation of the environmental impact related to the atmospheric emissions of geothermal exploitation that could be useful to build up a common framework for all the actors involved in the development of this energy source.

2. Materials and methods

In this work, the LCA approach is implemented according to the ISO 14040 (International Standards Organization, 2010) and ISO 14044 (The International Standards Organisation, 2006) standards, next to the more completely elaborated ILCD Handbook Guidelines (European Commission - Joint Research Centre - Institute for Environment and Sustainability, 2010). The methodology is composed of four phases:

- Definition of the goal and scope of the system: it includes the description of the model system and the purpose of the study, along with all the methodological key elements (functional unit, system boundaries, cut-off rules, data quality, etc) that characterize the analysis and a detailed explanation of all the assumptions made to guarantee clarity, transparency and reliability of the results;
- Life Cycle Inventory, LCI: it lists and quantifies all the input and output flows of energy and materials and releases to the environment;
- Life Cycle Impact Assessment, LCIA: impacts generated by the system are assessed through the application of an environmental impact calculation method that translate emissions, resources and energy use into a limited number of indicators;
- Life Cycle Interpretation: correlation among inventory results and impact analyses allows identification of the relevant technical information and critical points that can be employed to outline useful conclusions and recommendations to maximize the global energetic-environmental efficiency of the LCA case system in accordance with scopes and goals of the assessment.

2.1. Goal and scope definition

The objective of this study is the assessment, in a life cycle perspective, of the environmental impacts related to the exploitation of deep geothermal energy for electricity production in Italy. More specifically, the study is focused on the geothermal area located in Tuscany Region where the majority of the 916 MW Italian geo-thermoelectric plants are installed. Furthermore, the study considers all the currently operative power plants to outline subregional eco-profiles connected with the geo-thermoelectric activity. The findings of such an overarching study are intended to be used as a basic information for a sustainable development and exploitation of the Tuscan geothermal areas, while addressing the environmental issues concerning such kind of energy source.

2.1.1. System boundaries and functional unit

The life cycle of a geothermal power plant includes (i) the activity for the identification of the geothermal field, (ii) the drilling operations to obtain the production and injection wells, (iii) the building and commissioning of the power station and its connection to the wells through pipelines for the transportation of the geothermal fluid extracted as well as the fluid that needs to be reinjected after the utilization and (iv) the decommissioning of all the infrastructures (power plant and wells). The outcomes of a previous study (Buonocore et al., 2015), that was focused on the whole life cycle of a power station located in Tuscany, showed that the major environmental impacts are determined by the operational phase for Flash technology, unlike other Enhanced Geothermal System (EGS) and Organic Rankine Cycle (ORC) plants installed in other countries (for example in EU Germany, Belgium, Netherlands). The analysis performed in this work implements a gate-to-gate approach focused on the atmospheric emissions generated by the exploitation of fluids and produced during the operational phase of the geo-thermoelectric industry. In Fig. 1, a sketch of the system boundaries defined in this study is reported.

The atmospheric emissions generated by geothermal exploitation using flash power plants can be divided into two main fractions, one gaseous and the other dissolved into the geothermal fluid. The gaseous fraction is also identified as non-condensable gases (NCGs) as they cannot be condensed at the same conditions of the geo-fluid. These gases need to be extracted in order to avoid accumulation of NCGs within the condenser and progressive loss of vacuum conditions, as this is the fundamental state to keep the power plant in operation. Gases commonly extracted from geothermal fluids are carbon dioxide (CO_2), hydrogen sulphide (H_2S), methane (CH_4), ammonia (NH_3), hydrogen (H_2), nitrogen (N_2), argon (Ar) and radon (Rn) and gaseous mercury (Hg) (Bertani e Thain, 2002; Fridriksson et al., 2016). The quantity of these gases is extremely dependent on the field exploited and it is possible to observe very large variations among the World's geothermal reservoir. Furthermore, in the geo-fluid phase other chemical species are found such as arsenic (As), antimony (Sb), boric acid (H_3BO_3), lead (Pb), selenium (Se), chromium (Cr), cadmium (Cd), nickel (Ni), copper (Cu), manganese (Mn) and vanadium (V).

The impact connected to the maintenance operations of the power plant, such as the periodic substitution of the turbine or the change of the lubrication oil, were not considered. In the same way, processes concerning the maintenance of the wells, like the activities intended to recover the flow capacity lost over the year (stimulation), were not considered. The assumption is that all the burdens connected with these activities are virtually negligible compared to the environmental impacts determined by the direct emissions of a typical condensing flash power plant, like the ones operating in Tuscany. As the main product of the considered geo-thermoelectric power plants is not heat but electricity, we choose as the functional unit 1 Megawatt/hour (MWhe) generated in the various plants by conversion of the geothermal energy.

2.1.2. Data quality and collection

Data concerning the atmospheric emissions generated by all the 34 power plants currently operating in Tuscany have been collected from the geothermal areas monitoring annual reports published by ARPAT. The timeframe considered in this study ranges from the beginning of the sampling campaign started by ARPAT in 2002 up

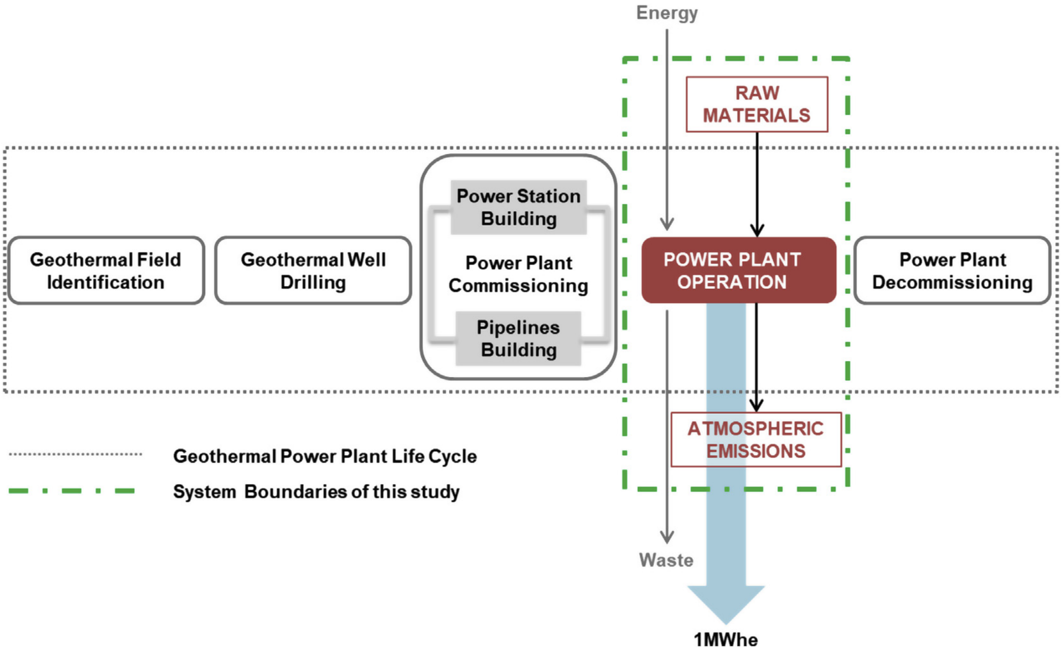

Fig. 1. Geothermal life cycle and system boundaries of this study.

to 2016, referring to the last report publicly available while this study was in preparation (ARPAT Tuscany Regional Agency for Environmental Protection (“in Italian”), 2018). Measurement data are based on sampling of the emission materials from the geothermal power plant's cooling towers in defined period of the year. The use of standardized methods for the analytical determination of substances (IGG-ICCOM, 2017; UNI EN, 2003; US EPA, 2017) ensure for the accuracy of the data. Moreover, in its reports, ARPAT provides emissions information concerning all the sampling points. This additional characterization allowed us to process and interpret data with higher accuracy, in respect to the knowledge of the aggregated data. The information was then carefully analysed to identify typical patterns and to elaborate a procedure ensuring the lowest error margin possible during the data rationalisation process.

2.1.3. Geography and configuration of power plants system

Nowadays, there are 34 power plants in Tuscany in an area of about 330 Km^2 displaced among the Provinces of Grosseto, Pisa and Siena. In 2018 the geothermal electricity production was about 6500 GWh. The geothermal geographic zones in Tuscany are usually dispersed in four areas as shown in Fig. 2: Larderello (South-East of Pisa Province), Lago (South of Pisa Province), Radicondoli (West of Siena Province) and the area of Mount Amiata in the southern Tuscany (East of Grosseto and South-West of Siena). The analysis of data has shown that the area of Mount Amiata presents two different geothermal fields with distinctive profiles in terms of atmospheric emissions. In fact, they are located on two sides of the mountain generating very different emission trends. Due to this, a further division of this subarea must be considered, namely Bagnore and Piancastagnaio, one on the Grosseto side and the other one on the Siena side, respectively.

Most of the power plants were built by ENEL and all of them are currently operated by ENEL GP (ENEL GreenPower) which developed a smart modular system to achieve the highest technical reliability. In fact, every power plant is composed by one or more standardized productive unit (of 20, 40 or 60 MWe each) which shares large part of the system component's (compressor, condenser, turbine, etc.). This approach allows the operator to use the same components for several reservoirs with different characteristics. The result is the reduction of the operating cost since the plant unavailability can be considerably reduced (DiPippo, 2015; Parri et al., 2013). From the methodological point of view, this technological configuration allowed our approach to reduce the variability of data among the geothermal areas considered, thus obtaining a more accurate analysis.

In this framework, usually one production well can serve different power stations, thanks to a very well-developed “steam network”. This allows the operator to direct the flow to the power station which presents higher efficiency or redirect the steam to the active power plants during maintenance operations of some others. All the reservoirs exploited are recharged by using brine reinjection wells to maintain the renewability of the resource over the years; this is also necessary for maintaining the pressure of the reservoir within certain values to avoid dangerous geological side effects connected with the geothermal sites' exploitation (seismic activity, subsidence). The success of this managing strategy is confirmed by the fact that the area of Larderello has been exploited for electric production since 1905, and more intensively since ’80s, without any significant loss (Cappetti et al., 1995; Minissale, 1991; Kaya et al., 2011). In recent years, power generation is even increased thanks to the implementation of new technological solutions allowing the exploitation of geothermal fluids that were impossible to use with previous systems because of their corrosive nature (Parri et al., 2013).

All the power plants present the same configuration if the capacity is the same. The power plant's working structure is mainly divided between the Non-Condensable Gases line (NCGs) and the fluids line, the samplings carried out by ARPAT were performed on both the lines. Fig. 3 shows the basic scheme of the ENEL power plants and some of the sampling points identified by ARPAT corresponding to the data used in this work.

The fluids coming from the production wells are directed to the turbine where they expand generating power. After this process the fluid is condensed in a direct contact condenser. In this component the already cooled geothermal fluid is used to cool down the fluid

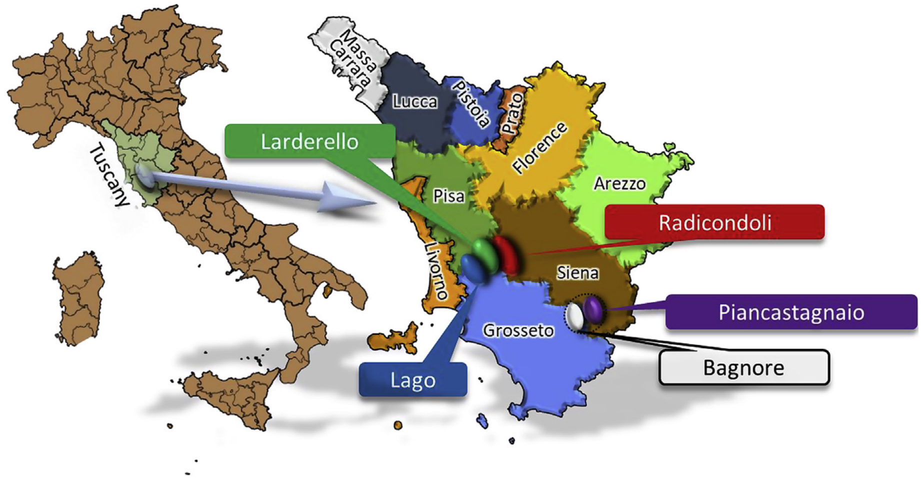

Fig. 2. Map of the Italian Regions and geothermal identified subareas in Tuscany.

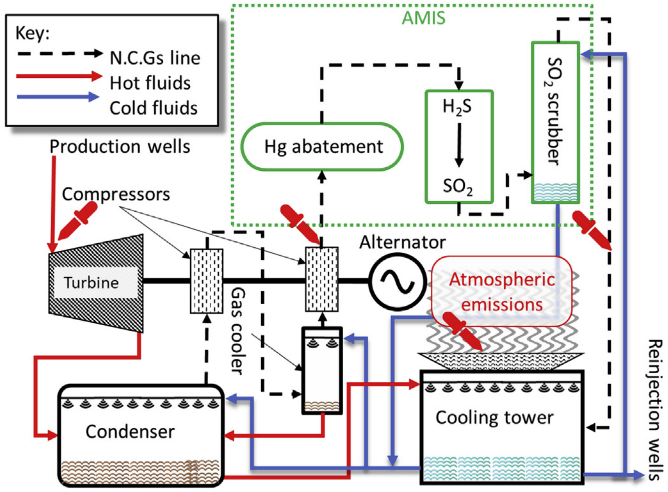

Fig. 3. Basic scheme of the geothermal power plant configuration implemented by ENEL. This configuration is employed in 20 MW and 60 MW productive units in operation nowadays. The red pipettes show the most important sampling points identified by ARPAT. As the recovery of heat is not an issue of this paper, the scheme has been simplified accordingly. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

overflowing from the turbine. Then, the condensate is pumped at high pressure to the cooling tower where it is sprayed in counter flow in respect to the air flow. The cooled fluid collected here is then employed in the condenser to cool the fluid overflowing from the turbine. The NCGs must be separated from the fluid to not compromise the process as they can accumulate in the condenser obstructing the cycle. Therefore, NCGs are extracted from the condenser by using compressors directly connected to the turbine and alternator axles: gases extracted in this way are sent to the AMIS (i.e. the abatement system for mercury and hydrogen sulphide) before dispersing them into the atmosphere through the cooling towers. The AMIS is composed by three main components: an absorber made of Selenium or activated Carbon, to remove the gaseous Hg, a catalytic reactor to oxidize the H_2S to SO_2, and a scrubber where the SO_2 produced by the redox reaction is washed from the gas by using the fluids collected in the cooling tower (Baldacci et al., 2005). Since the geothermal fluid naturally contains NH_3, the basic behaviour allows an efficient washing and neutralisation of the SO_2. The treated gas is then sent to the cooling tower where it is dispersed into the atmosphere together with the drift (small drops of geothermal water).

The direct emissions from these geo-thermoelectric plants to the atmosphere take place at the cooling tower and are differentiated into two distinct sources: the NCGs line and the drift. The atmospheric emissions connected with the geo-thermoelectric activity are then directly dependent on the chemical composition of the geo-fluid of the specific site, and thus depends on the geomorphological characteristics of the geothermal field. This is the reason why the emissions originating from different power plants - although located very close in a sub-regional area - can be very different from each other.

2.2. Life cycle inventory analysis

This study is focused on the potential environmental impact associated with the emission of NGCs that are found in greater concentration in the geothermal fluid (CO_2, CH_4, NH_3, H_2S) as well as gaseous Hg. In addition, also potential impacts associated to pollutants dissolved in the drift are investigated. This fraction is characterized by higher concentration of NH_3 and its salts, Hg, As, Sb, H_3BO_3, and other metals in traces (Pb, Se, Cr, Cd, Ni, Cu, Mn, V). All data regarding these chemical species were normalized with respect to the functional unit using the value of global electric production.

The need to process the original data reported by ARPAT arises from the fact that emissions detected over the years show an appreciable level of variability in their analytical determination (i.e. remarkable differences for some substances can be found in the various sampling campaigns). These differences are probably caused by technical difficulties related to some sampling procedures, such as the determination of Hg, due to the very low concentration involved and to the complex matrix present at the sampling point. Another source that determines the great variability observed could be linked to the technical characteristics of the different power stations. In fact, for different geochemical situations, the performances and characteristic emissions of the power plants appear very differentiated and largely affected by the geothermal field and, definitively, by their geographical positioning. For all these reasons, the intent of this work is to analyse the emissions of the geothermal power plants by identifying areas with common characteristics from a geographical point of view. Table 1 reports all the parameters collected and used to accomplish the analysis.

Table 1
List and description of all the parameters used to model the atmospheric emission scenarios of the geothermal power plants.

PARAMETER	SUBSTANCE	DEFINITION
Mass Flow	H_2S g/h	Power Plant Emission with AMIS
Mass Flow	H_2S g/h	Power Plant Emission without AMIS
Mass Flow	CO_2 g/h	Power Plant Emission
Mass Flow	SO_2 g/h	Power Plant Emission
Mass Flow	NH_3 g/h	Power Plant Emission with Abatement System
Mass Flow	NH_3 g/h	Power Plant Emission without Abatement System
Mass Flow	As g/h	Power Plant Emission
Mass Flow	Sb g/h	Power Plant Emission
Mass Flow	Hg g/h	Power Plant Emission with AMIS
Mass Flow	Hg g/h	Power Plant Emission without AMIS
Mass Flow	CH_4 g/h	Power Plant Emission
Mass Flow	CO g/h	Power Plant Emission
Central Parameter	MWe	Load during the sampling
Central Parameter	t/h	Supply Fluid Mass Flow during the sampling
Central Parameter	hour	Yearly Power Plant Out of Service
Central Parameter	hour	Yearly AMIS Out of Service
Electric production	MWh/y	Yearly electric production

The knowledge of all the information reported in Table 1 for each operating power plant allowed to draw a complete and detailed picture concerning the actual situation regarding the atmospheric emissions and the typical working parameters for each power plant. As mentioned above, the collection of data presented some problems regarding the expected uniformity over time. To overcome this problem, it was decided to create a typical scenario that might represent the most common emissive profile based on the consistent amount of data gathered.

This profile has been generated for each power station, then a geothermal field clustering criterion was selected (see related Data in Brief article).

This data processing allows to minimize the irregularities observed. Moreover, the impact analysis implemented in this way turns out to be not limited to a definite sampling campaign, as presented in previous studies (Bravi and Basosi, 2014; Buonocore et al., 2015), but it is representative of a typical outline accounting for all the variables involved in the geothermal energy exploitation. As for some power stations there is a lack of observed data, the scenarios are incorporated by subarea because data analysis shows good affinity among productive units in the same territory. Therefore, this process is suitable and reliable to use all the collected information. In addition, since there are several power stations installed on a relatively limited surface, it is essential to consider the whole area to obtain a correct evaluation and a good representation of the emissions profile.

The profile obtained for each plant is expressed as mass flow emission for each substance, multiplied by the yearly hours of operation. For pollutants processed by the abatement system (H_2S, Hg and NH_3 in some cases) the emission value is obtained considering the number of hours in a year in which the system is out of work. Thus, the final value, expressed in yearly mass flow (g/year), is composed by two portions: one comes from the determinations with the AMIS installed, the other is composed by the determinations without the AMIS installed, each weighted by the correspondent amount of operation. The logical steps of this procedure are sketched in Fig. 4. To average the values among the various samplings the median value is used in place of the average, due to the non-normal distribution of the values (Ferrara et al., 2019).

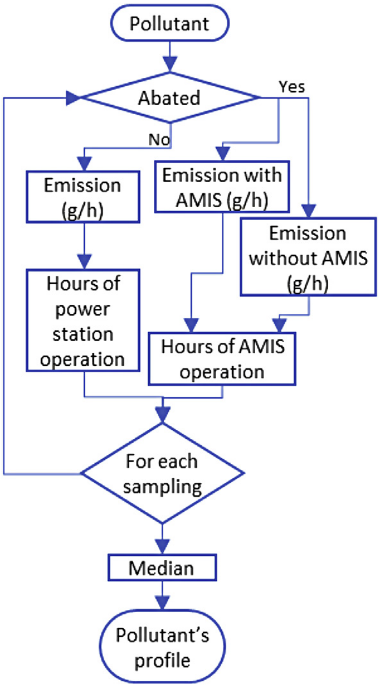

Fig. 4. Logical steps followed to obtain the emission profile for each pollutant.

2.2.1. *Scenario modelling*

According to the clustering criterion selected to identify geothermal fields, several scenarios describing each geothermal area were created. Such sub-regional environmental scenarios are intended to give an accurate description of the actual geothermal exploitation activity in Tuscany, by gathering information regarding both the geochemical profile of the field and the operating patterns of the power plants.

All the emissions information was collected year-by-year for each power plant and for each pollutant: those abated are treated separately in order to generate two different scenarios, one including the abatement due to the AMIS (actual scenario) and another which describes the emission like if no abatement system were in operation (W/O AMIS scenario). For each pollutant, the median value is calculated, excluding analytical determinations affected by human errors, as stated by ARPAT. This also allows to better fit the general reduction of emissions observed for some pollutants over the historical series obtained, thanks to technological improvements (Parri et al., 2013). Data concerning the amount of electricity produced every year were collected from the Market Report provided by EGEC (EGEC Geothermal, 2018). Also, in this case the median value was calculated. At this point the two scenarios were built: for the W/O AMIS scenario the emission (g/h) obtained from the previous step is multiplied by the on-order hours of the specific power station, obtaining a yearly emission value (g/y). Then this value is divided by the yearly electricity production (MWh/y), obtaining the emission value weighted by the typical electric production expressed as g/MWh for each power station. In case of the actual scenario, the number of the working hours of the abatement system is required, and it is collected from the ARPAT reports as well. The actual scenario is modelled as the scenario corresponding to the real emissions of the geothermal power plant analysed. In this case the abatement ratio caused by the AMIS is included. The resulting affected pollutants are Hg and H_2S, and in the case of Bagnore 3 and Bagnore 4, together with the AMIS, also the abatement due to the NH_3 treatment system is considered (Bonciani et al., 2013; Fedeli et al., 2016). The final emission value for these pollutants is then composed by two portions: one corresponding to the emission of the non-abated pollutant, multiplied by the number of hours in which the AMIS is out-of-order, while the other portion is composed by the emission value detected with the AMIS in function multiplied by the remaining hours. Finally, the emission is expressed as g/y and, following the same procedure explained above, the final value is expressed as g/MWh for each power station.

Each power station scenario is averaged accordingly to geographic and field distribution (see related Data in Brief article) in order to obtain the actual scenario and the scenario W/O AMIS for the five geothermal areas identified. Additionally, the global average scenario (average actual scenario) is computed to obtain the representation of the whole geothermal area.

Another scenario including the raw materials required during the operational phase of a geo-thermoelectric power plant has been implemented employing data published for the year 2016 (Enel Green Power, 2017). Such a scenario is useful to better judge the benefits connected with technological innovations that allow to use less chemicals to exploit the geothermal fluids. In fact, the amount of substances employed for fluid processing clearly decreased over the years. Moreover, it is noteworthy to specify that the operator cannot employ substances that are not naturally present into the fluid, according to the regional law.

In order to compare the sub-regional emissions profiles of geothermoelectric activity in Tuscany, a further scenario has been built using data concerning emissions generated by the electric production from natural gas. This last process is modelled starting from the dataset present in the Ecoinvent 3.4 database (Wernet et al., 2016), referred to a conventional power plant in operation in Italy (Treyer and Paul Scherrer Institute). Such a process has been conveniently customized to match the system boundaries defined for this study. Thus, the modified dataset is composed by the energy requirement for the extraction phase, the impact generated by the gas purification processes, the energy requirement, the gas leakage along the transportation phase and the atmospheric emissions due to the combustion process in a conventional natural gas power plant.

2.3. Life cycle impact assessment methods

In this study the ILCD 2011 Midpoint + normalized by EU27 2010, equal weighting, method v1.0.9, composed by sixteen impact categories, is applied to perform the analysis. As the purpose of this study is to provide eco-profiles connected with the geothermoelectric sector in several sub-regional areas, a midpoint (problem-oriented) approach was selected to characterize the environmental footprint on a large number of impact categories while maintaining accurate results.

Calculations were performed with the open source software OpenLCA version 1.7 LCIA package v2.0.3 (developed by Greendelta).

The choice of the ILCD 2011 Midpoint + method has been preferred because it allows to obtain single scores compared to other LCIA methods available in the OpenLCA software package. Furthermore, the ILCD 2011 Midpoint + method includes also the characterization of the particular matter formation potential connected to NH_3 emission. Finally, as the method used is developed by the Joint Research Centre of the European Commission, its application in this study looks even more justified.

Secondary data are taken from database Ecoinvent v3.4, eventually customized when necessary. Data uncertainty analysis is performed using the Monte Carlo tool included in the OpenLCA software.

3. Results and discussion

3.1. Atmospheric emissions

Table 2 reports the emissions expressed as g/MWh of electricity produced, the data presented are obtained following the process illustrated in Fig. 4 and the profiles obtained for each power station are then unified by area.

The emissions without the AMIS installed (W/O AMIS) is composed by the values detected before the AMIS was installed and, after the AMIS became operative, by the values detected at the gas extractor, where the sampling points identified by the ARPAT were located. We have included this scenario, even if it is a theoretical one and it is not representative of any actual emission of the geothermal plant, just to have an estimation, in terms of potential environmental impact, of the differences between the geothermal areas without and with technological improvements like the introduction of the AMIS system.

The actual scenario, instead, is the emission profile closer to the real situation of a geothermal area. Included in this scenario is the abatement obtained by using the AMIS and in the case of Bagnore, the ammonia abatement system is considered. Human errors in sampling activity, as registered by ARPAT, have been neglected.

3.2. Impact assessment

The emissions data obtained after the previously described processing are employed to compute the potential environmental impact associated to each scenario and each area. Results are shown in Table 3. The W/O AMIS scenario results show the differences among the areas considered but does not represent an environmental profile, rather it gives a description of the different geochemical characteristics of the several geothermal fields. Among all, it is evident the situation of the Piancastagnaio field: the much higher emission of Hg considerably influences the human toxicity and freshwater toxicity impact categories (together with antimony for the last category). The Bagnore field, even if quite geographically close to Piancastagnaio, shows less Hg but larger NH_3 emissions, in some cases also 10 times higher in respect to other fields. The release into the atmosphere of this compound has an impact on acidification, terrestrial eutrophication and particulate matter formation categories. All the remaining areas show more aligned results, in general lower compared to Bagnore and Piancastagnaio.

The actual scenario, instead, is intended to be considered the most similar and the one which better reflects the potential environmental impact produced by the geothermal power stations considered.

The presence of the AMIS abatement system has the effect to

Table 2
Emission values which outcome from the elaborated scenarios, expressed as g/h for each pollutant considered. The scenario without AMIS (W/O AMIS) is explanatory of the geochemical differences between the areas, it does not coincide to the emission detected in the area. The actual scenario (grey) represents the real emission currently present in each geothermal area.

Geothermal Area - Scenario	H_2S (g/MWh)	CO_2 (g/MWh)	SO_2 (g/MWh)	NH_3 (g/MWh)	As (g/MWh)	Sb (g/MWh)	Hg (g/MWh)	CH_4 (g/MWh)	CO (g/MWh)
Bagnore - W/O AMIS	3.62E+03	7.17E+05		1.09E+04	4.68E-02	4.62E-02	1.02E+00	1.96E+04	1.09E+02
Bagnore - actual scenario	9.24E+02	7.17E+05	1.17E+00	2.31E+03	4.66E-02	4.62E-02	2.03E-01	1.96E+04	1.09E+02
Lago - W/O AMIS	4.24E+03	2.59E+05		6.05E+02	5.98E-02	2.28E-02	4.14E-01	1.88E+03	4.32E+01
Lago - actual scenario	1.52E+03	2.59E+05	1.10E+00	6.05E+02	5.98E-02	2.28E-02	3.45E-01	1.88E+03	4.32E+01
Larderello - W/O AMIS	6.11E+03	3.43E+05		1.47E+03	5.30E-02	3.32E-02	6.97E-01	1.36E+03	1.97E+01
Larderello - actual scenario	1.62E+03	3.43E+05	7.73E-01	1.47E+03	5.30E-02	3.32E-02	4.87E-01	1.36E+03	1.97E+01
Piancastagnaio - W/O AMIS	1.02E+04	5.65E+05		1.81E+03	1.90E-02	4.58E-02	1.98E+00	7.81E+03	5.55E+01
Piancastagnaio - actual scenario	1.38E+03	5.65E+05	3.93E+00	1.81E+03	1.90E-02	4.58E-02	4.91E-01	7.81E+03	5.55E+01
Radicondoli - W/O AMIS	6.50E+03	5.32E+05		5.65E+02	2.14E-02	5.74E-02	5.94E-01	4.95E+03	2.11E+01
Radicondoli - actual scenario	1.26E+03	5.32E+05	2.99E+00	5.65E+02	2.14E-02	5.74E-02	3.32E-01	4.95E+03	2.11E+01

Table 3
Values of potential environmental impacts generated by the different geothermal areas for each scenario calculated with the ILCD Midpoint+ 2011 method. The grey rows represent the impact attributed to the actual scenario.

Impact category	Acidification	Climate change	Freshwater ecotoxicity	Human toxicity, cancer effects	Human toxicity, non-cancer effects	Particulate matter	Photochemical ozone formation	Terrestrial eutrophication
Bagnore - Scenario W/O AMIS	3.29E+01	1.21E+03	1.33E+01	7.38E-06	8.73E-04	7.27E-01	1.98E-01	1.47E+02
Bagnore - Actual scenario	6.98E+00	1.21E+03	4.03E+00	1.47E-06	1.73E-04	1.54E-01	1.98E-01	3.12E+01
Lago - Scenario W/O AMIS	1.83E+00	3.06E+02	6.05E+00	2.97E-06	3.52E-04	4.04E-02	1.90E-02	8.17E+00
Lago - Actual scenario	1.83E+00	3.06E+02	5.19E+00	2.47E-06	2.92E-04	4.04E-02	1.90E-02	8.17E+00
Larderello - Scenario W/O AMIS	4.44E+00	3.77E+02	9.34E+00	4.99E-06	5.91E-04	9.81E-02	1.37E-02	1.98E+01
Larderello - Actual scenario	4.44E+00	3.77E+02	6.88E+00	3.55E-06	4.20E-04	9.81E-02	1.37E-02	1.98E+01
Piancastagnaio - Scenario W/O AMIS	3.78E+00	7.65E+02	3.11E+01	1.43E-05	1.70E-03	8.34E-02	7.89E-02	1.69E+01
Piancastagnaio - Actual scenario	3.78E+00	7.65E+02	9.85E+00	3.54E-06	4.19E-04	8.34E-02	7.89E-02	1.69E+01
Radicondoli - Scenario W/O AMIS	3.81E+00	6.56E+02	7.62E+00	4.26E-06	5.05E-04	8.40E-02	4.99E-02	1.70E+01
Radicondoli - Actual scenario	3.81E+00	6.56E+02	4.42E+00	2.39E-06	2.82E-04	8.40E-02	4.99.E-02	1.70E+01
Unit	molc H^+ eq	kg CO_2 eq	CTUe	CTUh	CTUh	kg PM2.5 eq	kg NMVOC eq	molc N eq

reduce consistently the amount of Hg and H_2S released to the atmosphere, even though the LCIA method used for the analysis does not include a characterization factor for H_2S. As a matter of fact, the toxic effect of H_2S in geothermal field is not well modelled and documented yet in the literature, although the reduction of H_2S emission represents an important issue for the resident population. Indeed, the bad smell produced by this compound is quite effectively reduced by the AMIS, ensuring better wellness (Baldacci et al., 2005; International Programme on Chemical Safety (IPCS), 2003; Pertot et al., 2013).

Therefore, in this analysis, the AMIS only affects the impact categories related to Hg emission and, only for the Bagnore field, also the categories influenced by NH_3 emission.

The analysis of the impacts generated by the actual scenarios shows comparable values among all the areas thanks to the reduction of Hg, and NH_3 for Bagnore. In detail, the reduction of pollutants, and the consequent change of the indicators' values, in respect to the scenario W/O AMIS is very strong for Piancastagnaio and Bagnore fields. In fact, these territories are those where the Hg emissions are sizable due to the presence of cinnabar mines which heavily influence the chemical composition of the extracted fluids (Barazzuoli et al., 2008; Loppi and Bonini, 2000; Manzo et al., 2013). The profile of this scenario still shows the effects related to NH_3 emission for the Bagnore field: despite the presence of the abatement system devoted to NH_3 reduction, the residual value is still high compared to the other fields.

The impacts on climate change and photochemical ozone formation categories are determined by the gaseous fraction of the fluids, namely the amount of CO_2, CH_4 and CO. Even for these emissions, it is possible to observe differences among the areas: Bagnore shows the highest values followed by Piancastagnaio. For those pollutants there is no abatement system operating, therefore the values in the two scenarios are the same.

Analysing the single score indicator results, it is possible to better visualize the differences among the areas. The graphs in Fig. 5 show the effectiveness of AMIS in reducing the potential environmental impacts. In fact, for Bagnore and Piancastagnaio the reduction is 76% and 69%, respectively, while for other areas the advantage is below 50%, namely 14% for Lago, 24% for Larderello and 35% for Radicondoli. Indeed, the single score turns out to be

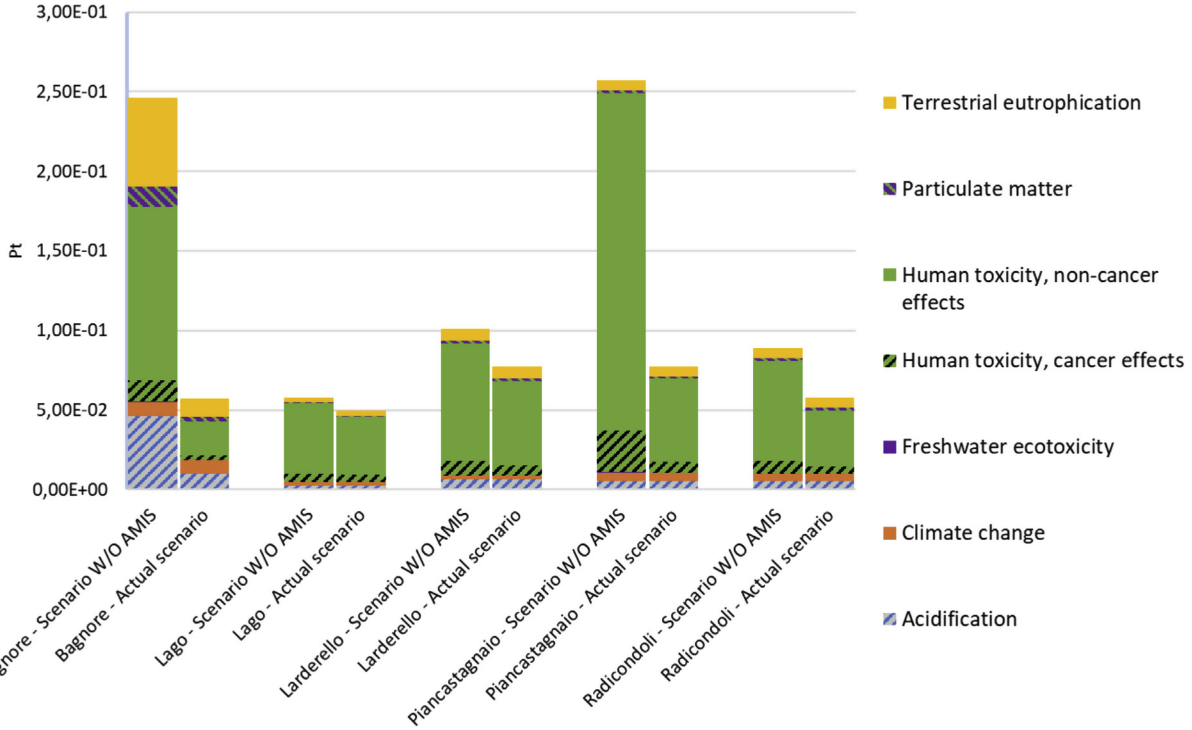

Fig. 5. Single score obtained for each geothermal area, the graph is divided into column for each area, the left bar refers to the scenario without the AMIS, while the right bar corresponds to the actual scenario. Cut-off rules were defined for impact categories giving a contribution below 2% in the eco-profiles. The large reduction of potential environmental impacts between the scenarios with and without the AMIS is clearly seen.

largely composed of categories related to toxicity themes that generally are affected by a quite high uncertainty (Pizzol et al., 2011). Since these categories are based on characterization factors derived from ecotoxicological evaluations, the LCIA methods cannot model peculiar regional situations. This is even more true when the impact is generated by a heavy metal. In fact, as stated in the USETox method documentation (Fantke P. et al., 2015): *"It should be stressed that the characterization factors are useful for a first-tier assessment. In case a substance appears to dominantly contribute to the impact scores for toxicity, it is recommended to verify the reliability of the chemical-specific input data for this substance and to improve the data whenever possible"*. The case considered here matches this condition, as Hg heavily influences the whole impact profile and, furthermore, almost totally accounts for the toxicity categories. Thus, to better understand the environmental burden caused by Hg, further investigation should be performed to properly model diffusion pathway and chemical transformations.

For a more general analysis of the geothermal power plants emissions, an average scenario among the areas was modelled. This is used to compare the geothermal exploitation with the natural gas electric production and to compare the emissions arising from different points of the plant (gas, fluids). Table 4 reports values obtained for each pollutant.

Since the AMIS can process the extracted gaseous phase of the fluid, the remaining part of pollutants dissolved in the drift is still emitted into the atmosphere through the evaporative tower. In fact, the power station abatement ratio (the efficiency of abatement in respect to the total emission, and not only in respect to the processed phase) among the areas where the Hg presence is higher (Piancastagnaio) and the others is very different (ARPAT, 2011; Barazzuoli et al., 2008; Manzo C et al., 2013). Piancastagnaio shows better results because the gaseous Hg concentration is high (see Table 2), thus more substance can be absorbed by the AMIS, but the Hg concentration dissolved in the drift is quite similar for all the areas. The results of the abatement process reflect the amount of Hg emitted with the drift, since the abatement ratio over the gaseous phase is more than 95% in most cases.

Comparing the results obtained by using the emissions detected at the gas extractor after the AMIS treatment (Gas Phase) and the total emissions of the actual scenario, it is evident how the most important source of impact is determined by the drift if the AMIS system is employed, results are reported in Fig. 6. The actual scenario is determined by the impact generated by the Hg dissolved in the drift, while the scenario W/O AMIS is composed by the last one in addition to the Hg emitted by the non-abated gas phase. Then, the impact generated by the processed gas is the one showed as Gas Phase.

This result is determined by the fact that no abatement system is employed for the liquid phase, so the contact between the geothermal fluid and the atmosphere in the evaporative tower generates the emission of the pollutants contained in the fluid extracted.

Other comparisons are made by evaluating the impacts generated by the geothermal electric production and those associated with electricity production from natural gas. In this case the emission information showed previously are integrated with raw materials required to treat the geofluids extracted from the wells (Gas Phase + Raw Materials)>. Results are reported in Table 5.

The substances needed are HCl used to avoid colloid formation of $CaSO_4$, and NaOH used as neutralising agent, as geothermal fluids used in some plants can contain high concentration of chlorides which might be very corrosive for the plant's elements. The NaOH is also used to control the silicate scaling formation (Brown, 2013; Parri et al., 2013). In the plants of Bagnore area, also H_2SO_4 is used to control and lower the atmospheric emission of NH_3: the acidification of the fluid maintains the NH_3 as a dissolved salt avoiding its

Table 4
Emissions calculated for the average scenario based on date collected from all the geothermal fields.

	Average - actual scenario (g/MWh)	*Average scenario - W/O AMIS (g/MWh)*
H_2S	1.34E+03	6.12E+03
CO_2	4.83E+05	4.83E+05
SO_2	1.99E+00	
NH_3	1.23E+03	3.07E+03
As	4.00E-02	4.00E-02
Sb	4.10E-02	4.11E-02
Hg	3.72E-01	9.42E-01
CH_4	7.10E+03	7.10E+03
CO	4.96E+01	4.96E+01

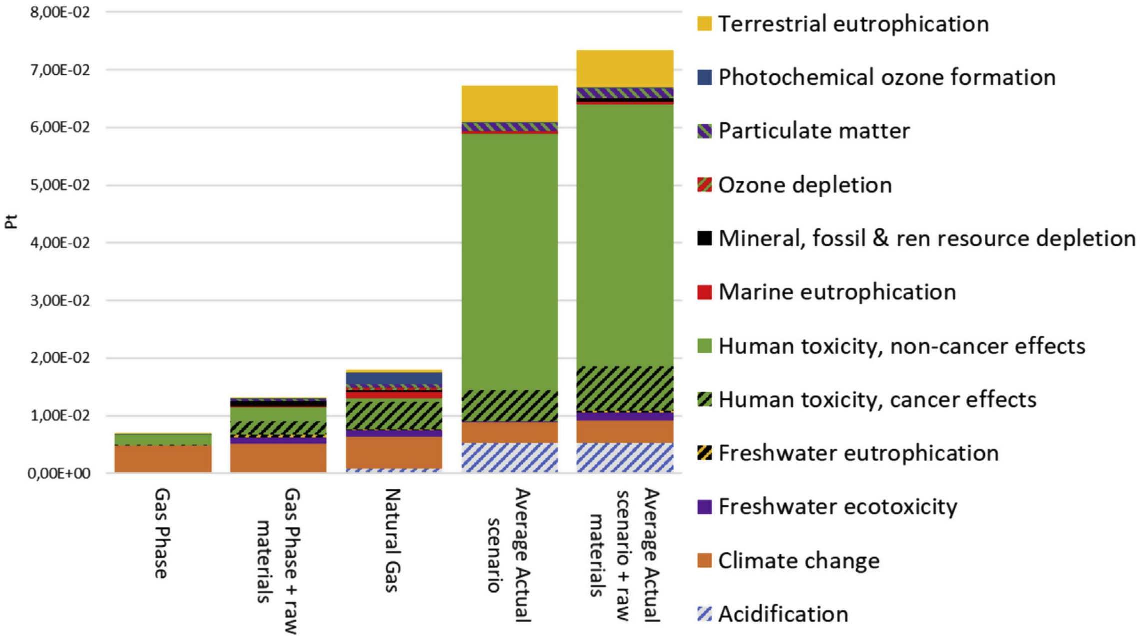

Fig. 6. Single score indicators showing the differences between several scenarios of geothermal exploitation and the electric production from natural gas. Cut-off rules were defined for impact categories giving a contribution below 2% in the eco-profiles.

Table 5
Percentage variation attributed to the inclusion of the raw materials use to the average scenario of geothermal exploitation.

Average Actual scenario	Average Actual scenario + raw materials	Variation %	IMPACT CATEGORY
5.24E-03	5.38E-03	2.65	Acidification
3.69E-03	3.81E-03	3.30	Climate change
6.66E-05	1.30E-03	94.89	Freshwater ecotoxicity
0.00E+00	4.26E-04	100.00	Freshwater eutrophication
5.43E-03	7.62E-03	28.73	Human toxicity, cancer effects
4.45E-02	4.54E-02	2.05	Human toxicity, non-cancer effects
4.46E-04	5.20E-04	14.18	Marine eutrophication
0.00E+00	5.69E-04	100.00	Mineral, fossil & ren resource depletion
0.00E+00	2.95E-05	100.00	Ozone depletion
1.44E-03	1.72E-03	16.38	Particulate matter
9.30E-05	1.96E-04	52.43	Photochemical ozone formation
6.29E-03	6.36E-03	1.03	Terrestrial eutrophication
6.72E-02	**7.34E-02**	**8.41**	**Single Score**

extraction as a gas.

The analysis carried out is based on the assumption that the raw materials employed to treat the geofluids do not generate any (local?) impact, as they are injected into the reservoir. The impact related to the raw materials is considered to be only associated to the upstream production processes involved. In this case the original Ecoinvent process (Althaus et al., 2007a, 2007b; Parada, 2017) is modified to account only for the use phase.

Table 5 shows that the greater burden is determined by the actual scenario integrated with the raw materials which increase the potential impact by 8.4% compared to the actual scenario without raw materials. Among the substances employed for fluid processing the greater impact is generated by the production process of the NaOH, while the H_2SO_4 and HCl production processes only account for less than 1% compared to the raw materials impacts. Table 5 also shows the percentage differences due to the inclusion of the raw materials on each impact category. Those presenting the higher variation are Freshwater ecotoxicity, Freshwater eutrophication, Mineral, Fossil & renewable resource depletion, Ozone depletion. Definitively, the significant amount of NaOH used to process the geothermal fluid is responsible for a sizable increase of potential environmental impact. In fact, for all the geothermal areas during the year 2016, a total of 75,388 t of NaOH have been employed, while only 280 t of HCl and 3640 t of H_2SO_4 (employed to reduce the NH_3 emission in the Bagnore field) were used.

The results presented above suggest that the only way to avoid emissions of pollutants would be the implementation of full reinjection of both fluid and gas phases. In fact, total reinjection only of the fluid would result in potential impact due to the gas (Gas Phase as in Fig. 6), if the abatement system for Hg and H_2S are employed. To avoid this residual impact total reinjection should be employed (Bruscoli et al., 2015; Bonalumi et al., 2017).

3.3. Uncertainty analysis

The large amount of data collected allowed to determine the error associated to each atmospheric emission, together with the information already present in the Ecoinvent database. As the reliability of data is crucial for assuring consistency of the study, a Monte Carlo analysis was performed in order to determine the variability and the confidence range of the scenario which represents the whole geothermal area.

The Monte Carlo simulation, performed over the main scenarios considered in this study as illustrated in Fig. 7, shows that most of the uncertainty is due to the atmospheric emission of the geothermal power plant, while the raw materials employed and the electric production from natural gas show much less uncertainty.

Monte Carlo simulations performed on the inventory's data show that the error associated to the measures of Hg and Sb present the largest uncertainty, compared to other pollutants (see Fig. 8). As shown in Fig. 9, comparing the uncertainty among impact categories expressed through normalized (adimensional) values, the human toxicity-non-carcinogenic effects shows the highest uncertainty results, responsible for the large uncertainty of the geothermal scenarios. This is a direct consequence of the characterization factor attributed by the ILCD method to the gaseous emission of Hg into the atmosphere. This compound has the highest score among the atmospheric emissions with a value of $8.5x10^{-1}$ and it occupies the second position in the list of compounds included in this impact categories. The highest score is attributed to polychlorinated biphenyls with a value of 25.5. For comparison, Sb has a factor of $1.55x10^{-4}$ while the value of As is $1.6x10^{-2}$. Thus, such a high uncertainty associated with Hg is directly connected with the high uncertainty of this impact category. A different output is obtained considering the human toxicity -cancer effects category for which Hg has a characterization factor of $7.2x10^{-2}$, while As is $2.42x10^{-4}$ and Sb has no effect at all. In addition, it should also be considered that the comparison presented in Fig. 9 is performed after the normalization step, which assigns to the human toxicity categories the highest ratios: $5.3x10^{-4}$ and $3.6x10^{-5}$ for non-carcinogenic and carcinogenic effect, respectively. Thus, the margin of error related to the inventory data for Hg is further increased by the characterization and normalization factors and produces a sort of magnification of the overall uncertainty as a result.

The large error connected with the measure of the effect of this compound could be minimized adequately increasing the frequency of sampling and has to be better understood employing different methodological tools. Indeed, LCA analysis results should be treated with awareness of the LCIA methods limitations. A more accurate ecotoxicological analysis should be performed to connect the emission of this heavy metal to real effects on the environment and residential population's health.

4. Conclusions

The objective of this study is the assessment of the environmental impacts associated to the atmospheric emissions connected with the exploitation of deep geothermal energy for electricity

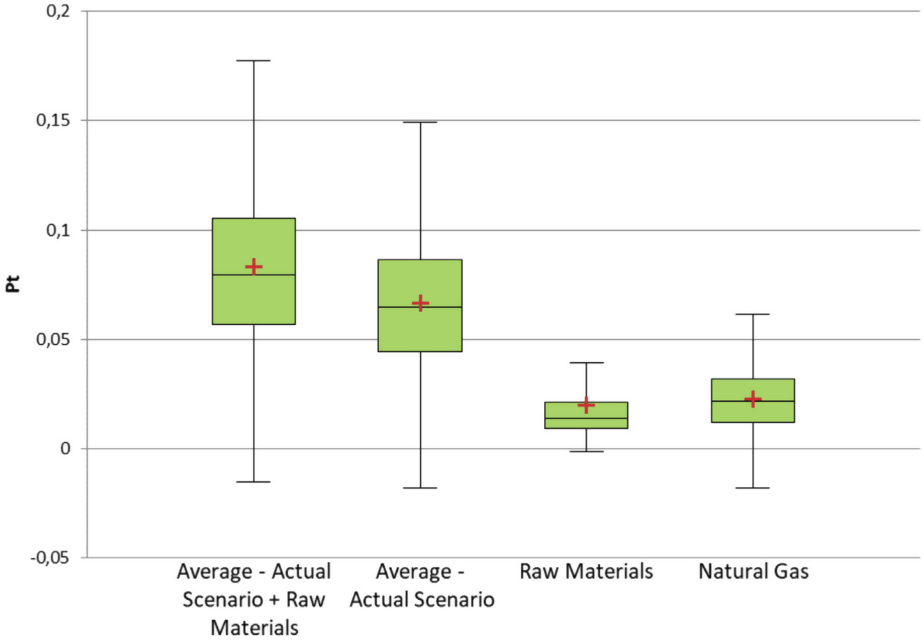

Fig. 7. Box plot resulting from the Monte Carlo analysis: result is shown as single score, cross corresponds to the mean value.

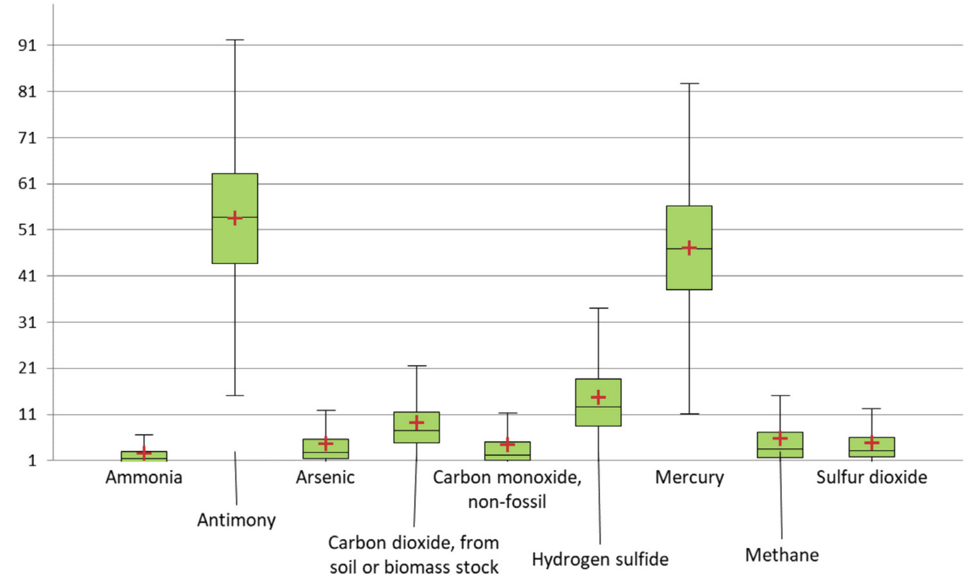

Fig. 8. Box plot of the Monte Carlo analysis performed on the inventory data employing the errors calculated taking into account the variability found; results are rescaled to 0–100 range in order to compare them.

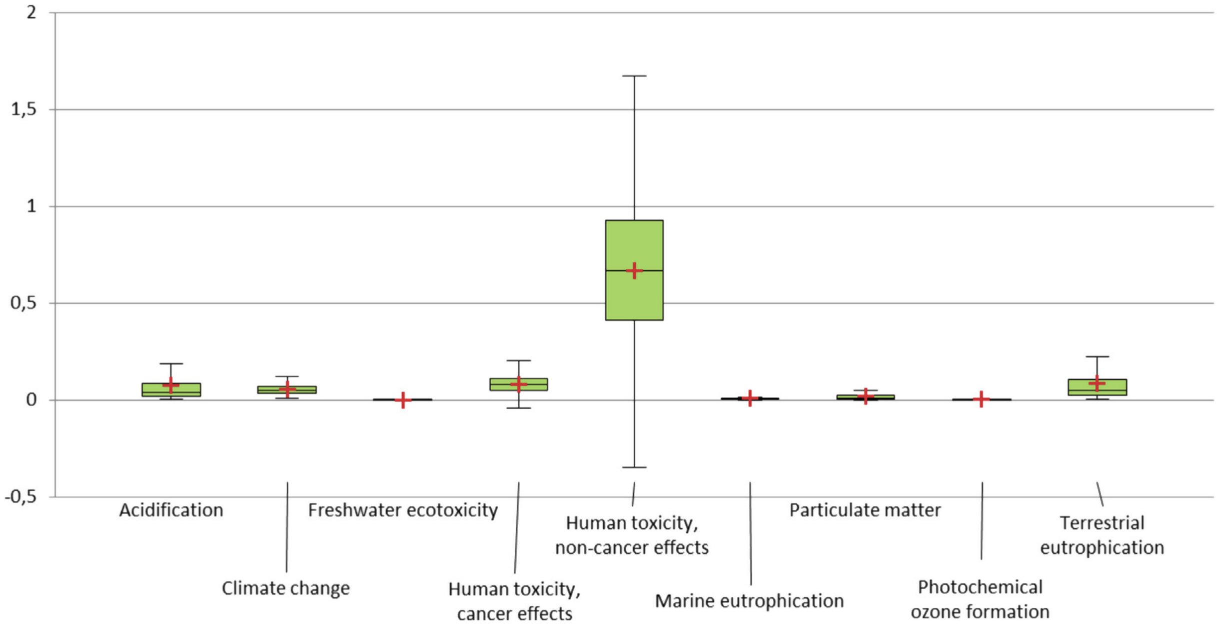

Fig. 9. Box plot associated to each impact category, the statistical descriptors are calculated considering the normalised results.

production in Tuscany (Italy). To this aim, the modelling of several scenarios is proposed in order to draw geothermal fields profiles that are independent on technological differences and time. At the same time the models must be accurate, and representative of the system analysed, reflecting the geographical location and the geochemical characteristics of the reservoir exploited. Comparing our present results with previous studies which were taking into account only a single power plant to represent an entire area is evident that the environmental profile obtained with the procedure proposed in this study is much more representative of the actuality. The needs to consider all the power plants in the impact analysis is required by the fact that not all of them present the same emissions and the same operating parameters (AMIS efficiency, operational times, etc), even if they are exploiting the same field. In conclusion, a crucial point stressed by this paper that should not be neglected in further research and discussion is that the emissions profile of a geothermal area need to be representative of all the productive units working in that space.

The analysis of data shows the effectiveness of the AMIS abatement system in reducing H_2S and gaseous Hg emissions. It is noteworthy that the potential environmental pollution of geothermal areas commonly associated to the highest emission of Hg (Bagnore and Piancastagnaio) has nowadays a comparable profile to those of the “traditional” fields (Larderello, Lago, Radicondoli). Indeed, the great geochemical differences among the geothermal fields can be considered virtually eliminated. Furthermore, in some cases, the results turn out to be even better, as in the field of Bagnore the acidification of the circulating geothermal water reduces the amount of NH_3 stripped into the atmosphere. This evidence allows to confirm that, the managing strategy adopted by the operator ENEL GP through the development of a smart steam network is successful in pursuing a consistent reduction of the environmental pollution associated with flash geothermal power plants.

Nevertheless, the efforts to solve the problem of the presence of NH_3 in the drift are not so successful, as the acidification system employed just allows to limit the NH_3 stripping. Different solutions should be engineered in order to obtain a proper abatement process than just a reduction obtained thanks to the pH variation.

Therefore, NH_3 still represents a problem to deal with, overall but not only in the Bagnore field. In fact, the interaction with H_2S and other elements could generate larger production of particulate matter but, in our opinion, at the moment, there is not enough research devoted to investigating such crucial aspect.

The problem could be overcome in perspective using technologies with a total reinjection of fluids applicable also in hybrid configuration to flash power plants or typical of binary cycle installations. As already mentioned above, another scientific problem arising from the findings of this paper which deserves further attention is about the potential environmental impact caused by Hg: the high uncertainty related both to measurements and to the LCIA method itself does not yet allows to identify the real dimension of the problems related to toxicity impact categories. A step forward could be the elaboration of an optimized LCIA method able to identify and compute the potential environmental impact due to the peculiar atmospheric emissions of flash power plants and, in general, of a variety of geo-thermoelectric installations.

Declarations of interest

None.

Author contributions

R.B. and M.L.P. designed research; N.F. and M.L.P. performed LCA analysis; all authors participated in the interpretation and discussion of results and in writing the paper.

Notes

The authors declare no conflict of interest.

Acknowledgment

Authors acknowledge MIUR Grant - Department of Excellence 2018–2022. NF thanks COSVIG for contributing to his PhD grant.

Authors would like to thank Prof. G.P. Manfrida (University of Firenze) and D. Bonciani (COSVIG) for fruitful discussion and Professor S. A. Loiselle (University of Siena) for his valuable suggestions concerning data processing. Collaboration of ARPAT in collecting data and careful reading and revising of the manuscript by Professor Emeritus Michael Rodgers, Bowling Green State University, is gratefully acknowledged.

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