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Archaeometry of Air Pollution: Urban Emission in Italy during the 17th Century

Dario Camuffo

CNR-ICTIMA, Corso Stati Uniti 4, I-35127 Padova, Italy

Claudia Daffara and Mirca Sghedoni

Consorzio Padova Ricerche, Corso Spagna 14, I-35127 Padova, Italy

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Past sources of air pollution in the cities of Northern Italy are investigated by a critical analysis of a 17th-century treatise written by B. Ramazzini, a medical doctor interested in the associations between work, environmental pollution and health. In this paper, past emissions due to domestic and craftsman's activity have been recognised and classified according to the potential damage to cultural heritage. The indoor environment experienced concentrations of smoke due to bad ventilation and domestic combustion for lighting and heating. High indoor concentrations of sulphur dioxide were common from the burning of sulphur for domestic and workshop activities. The outdoor environment experienced smoke for the combustion necessary for several workshop activities and uncontrolled dangerous emissions. The urban pollution was not homogeneous; the craftsmen's activities were organised in different specific areas so that the environmental deterioration potentials changed from site to site inside the same town.

Keywords: ARCHAEOMETRY, POLLUTION HISTORY, AIR POLLUTION, CULTURAL HERITAGE.

Introduction

Historical cities include many monuments that have deteriorated as a result of the cumulative effects of natural factors and anthropic pollution. Understanding monument decay requires knowledge of environmental archaeometry, including domestic and craftsman's pollution. Knowledge of the materials used, details about the working practices and the documentation, when available, of the noxious effects on people and artefacts constitute valuable information to the interpretation of the decay mechanisms that have affected our cultural heritage.

The history of air pollution can be divided into two main periods: before and after the industrial revolution, and the aim of this paper is to investigate the former period. This is the less studied and concerns the main part of the monuments life. The activity of craftsmen remained mainly the same for a long time prior to the industrial revolution (Uccelli, 1944; Singer *et al*., 1958; Mondadori, 1980) and can be found described in some interesting writings. Among them, we especially mention a medicine treatise "*De morbis artificum*" (i.e. Craftsmen Occupational Diseases), written in 1700 by Bernardino Ramazzini (1633–1714), and the French encyclopedic opera "*The Spectacle of the Nature*" (Anonymous, 1740). The first book is very technical and detailed; the second sometimes adds new light to this period. They are not only useful in order to understand the degree of past craftsmen pollution, but also the domestic style of life, the use and preparation of different substances burnt for lighting houses, the use of sulphur to light fires in fireplaces or to make white washing, as can be seen in the following.

Ramazzini was a physician who for a long time studied workers diseases in Modena and Padova and in his book he listed a long series of illness, typical of workers and craftsmen working with poisonous substances such as sulphur, tin, mercury, lead, or typical of jobs where the operators had to stay for a long time in direct contact with kilns and hearth thus inhaling large quantities of combustion gas. Extracts from the sections, which give new light to our knowledge, will be divided into classes showing indoor and outdoor problems, and classified according to the severity of the environmental contamination.

It is useful to point out that in Italy, until the mid 1800s, the main fuels for either domestic or craftsmen use were timber and charcoal. Different to Northern Europe (Cipolla, 1983; Brimblecombe, 1987), the relevant use of coal in Italy was only made during the Industrial Revolution at the end of nineteenth century. Problems related to the quality of urban air were different from the case of London:

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> In London the air is full of "*sulphur spirits*" for the burning of large quantity of coals. Clothes you had on, for a period of time, in this city, keep the smell of sulphur through the years (Anonymous, 1740).

As already stressed for the early scientific literature (Camuffo, 1992), the information extracted from Ramazzini has to be analysed with criticism according to the scientific knowledge of the period. In the 17th and first half of the 18th century, the theoretical concepts used in chemistry were, at the beginning, still related to the atomistic theory of Democritus, the theory of the opposites of Aristotle, sometimes with some influence of Medieval alchemy and many individual interpretations, often with contradictory aspects.

The "fire" (i.e. the father of the phlogiston and ancestor of the modern heat) has been for a long time considered the property of heating, melting, and vaporising. According to Aristotle's theory, the substance antagonistic to the fire was the "cold", which had the opposing properties. In addition, the "fire" was also considered a "matter" on the grounds of two basic ideas: (i) the essential characteristic of the matter was not the mass, but the volume and the actual fire has a space extension; and (ii) the success of the atomistic theory under development. Two known atoms were associated with the heat and the cold, i.e. the sulphur and nitrogen, respectively. Philosophical reasons to identify the "sulphur" with the substance able to allow combustion in all the inflammable bodies were found by Philip Theophrast Von Hohenheim, called Paracelsus (1525–27). A convincing practical reason was that sulphur was used in lighting fires; the nitrogen for the discovery of the refrigerating mixtures that employed salts derived from nitrogen. The nitrogen atoms were supposed to be so acuminate that they could enter the materials and remain wedged between atom and atom, hindering any movement: this was the simple explanation for the freezing of liquids. Thunder and lightning were claimed to be due to burning of the hypothetical sulphur dissolved in the atmosphere (Boerhaave, 1749). After the experience with gunpowder (which is composed of sulphur, potassium nitrate and carbon), the combustion in general was related to some reaction between sulphur and nitrogen compounds.

This theory is found in theoretical treatises of the time, and here a clear explanation of the physicist–meteorologist Giovanni Poleni (1683–1761) is reported, referring to the cause of exceptional cold periods, as follows: "the excessive hot is generated when the air becomes mixed with a much greater quantity than usual of sulphur particles by which more intense storms are set off. The air is filled with such a large quantity of nitrous particles that are the cause of such severe cold. In fact, that the cold is born, or anyway its intensity is increased above all because the nitre is ignited, is demonstrated in the first place by the ice made artificially which freezes more and more (as demonstrates by various and convincing experiments) or because of the increased percentage of nitre mixed with the ice, or by the use of more concentrated nitre" (Poleni, 1740).

Only in the second half of the 18th century did the development of physics and chemistry lead to more mature ideas. For example, in 1789 A. L. Lavoisier published his "*Traité élémentaire de Chimie*" (Elementary Treatise of Chemistry) in which he listed the so-called "simple substances" including light, caloric, some 30 known atoms and a few minerals or radicals. The caloric was a scientific development of the "fire", and sulphur and nitrogen, among others, were conceived in terms of the modern chemical elements. In particular, Lavoisier discovered the properties of the oxygen, which radically changed the old theory about combustion.

It is evident that a quantitative microanalysis of the quality of the air or water to measure the concentration of sulphur or nitrogen was not possible in the early period given the experimental limitations, and that mention to sulphur particles or nitre compounds were not related to an effective pollution in the atmosphere. The above interpretation is essential for an appropriate comprehension of the old scientific literature especially because in the past some scholars of environmental history have misinterpreted for early measurements of air pollution the simple statement that the weather was cold (i.e. air rich of nitrogen) or hot (air rich of sulphur).

Domestic Indoor Pollution (Heating and Lighting)

Concerning the problem of the conservation of artworks in historical buildings, i.e. the problem of indoor pollution, the main problems can be identified in: (i) the domestic burning of sulphur; (ii) the smoke originating from domestic heating in a period in which the ventilation and the early chimneys were inefficient, and (iii) light sources which used smoky fuels. The main effects were fading colours, tarnishing silver and bronzes, deteriorating tissues, blackening surfaces in general.

Burning sulphur

The uncontrolled domestic burning of sulphur, to light fireplaces or other aims, was common, although rarely mentioned by historians of air pollution. This combustion, made in relatively small rooms with no ventilation, certainly led to severe and dangerous indoor sulphur dioxide concentrations (which is considered the primary factor for artwork deterioration or, at least, one of the hazardous substances), as is evident from the following description:

> A baker once found small sulphur cylinders in his stove, which were used as fire starters, in order to avoid a fire propagation he squashed them with a foot and almost died. [. . .]

> The powerful effect generated by burning sulphur was well known by those women that whiten their clothes by exposing them to the exhalations. Once, a man had hidden below the bed when the husband of the lady he was courting arrived. However, the sheets were recently washed and then treated with sulphur smoke for a better whitening, so that the man had a whooping cough and was discovered (*Diseases of people working with sulphur*, in Ramazzini, 1700).

This gives an orientative idea about the elevated concentration of sulphur dioxide in a room of a house. However, in addition to the domestic use, craftsmen also burned sulphur. In 1404 in England, the king, Henry IV, prohibited the alchemistical practice for the excessive smell of burning sulphur. In 1418 in Venice, the Great Council made the same prohibition against sorcery, but craftsmen could continue their activity.

> In fact, sulphur exhalations can whiten even purple roses (*Diseases of dyers*, in Ramazzini, 1700).
>
> The sulphur is much used in the wool production to whiten woollen clothes. It is powerful in purifying all that is infected by pestilential air (Anonymous, 1740).

The production of wool and silk was important in Venice. The silk production flourished especially between the 13th and 15th centuries; the wool production culminated between 1550 and 1630.

> Burning sulphur was also used to whiten the purple roses. [. . .] the situation was as in the ancient Rome times, when the poet Martial reported that eye diseases often affected the matches merchant (*Diseases of people working with sulphur*, in Ramazzini, 1700).

Heating

The smoke was not well eliminated from rooms, where braziers were placed in the middle of rooms, or fireplaces had inefficient natural draught. To stay for a long time in a heated room was not recommended for health:

> It is more advisable to study in a large room, being covered with cloths, than stay in a small, or in a heated room, as also Hypocrites suggested (*Diseases of literary men*, in Ramazzini, 1700). We talk about the typographers typical diseases. [. . .] As well as eyes disease, they can have fevers and pneumonia and others lung disease. This arose because during the winter they have to stay inside well insulated and well heated rooms, in order to let the printed sheets dry (*Diseases of typographers*, in Ramazzini, 1700).

Also, artworks were exposed to risk and the most immediate consequence was blackening of the surfaces.

Lighting

To a lesser extent, the use of candles and oil lamps also caused thick smoke and incompletely burned fats that blackened paintings and decorations. Large rooms were preferred to small ones, where the smoke was more concentrated:

> It is advisable that the men of letter stay in large and not small rooms. The oil-lamp smoke is very dangerous [. . .] and also the tallow candles whose bad smell and smoke are noxious [. . .] (*Diseases of literary men*, in Ramazzini, 1700).

Candles were of two kinds: made of tallow or wax (pure or stearic or with paraffin), the former being cheaper with a foul smell and were very smoky, the latter more expensive and cleaner. Pure wax was used since the ancient times; stearin was for the first time extracted from the animal fat by Chevreul in 1826, and the paraffin was firstly extracted from lignite by Buchner in 1819; the first industrial production was made in Glasgow in 1845 and after 1890 paraffin was commonly extracted from petrol (Graziani, 1973).

During the 17th century, the oil was mainly made from olive, walnut or linseed; the mineral oil was rarely used for its unpleasant odour:

> Nuts are very much used, here where they are grown, to produce the oil that everyone uses for the night lighting; they prefer the nuts oil rather than the olive oil because this one is too costly (*Diseases of oil makers, tanners and people working with dirty matter*, in Ramazzini, 1700).

Recommendations were made to avoid the types of candles or oil which caused heavy pollution, and to ventilate rooms to diminish the smoke concentration, as follows:

> I urge the scholars to not use the tallow candles during their studies; and if they can not afford the wax candles, they have to use the olive oil lamps, as the ancient scholars used to do. People who spend some hours in a room without ventilation, reading, writing or doing something else with the light of a nut oil lamp, know very well how much this smoke is harmful; they get out from the room with heavy headache, dizziness, and the room is full of smoke (*Diseases of oil makers, tanners and people working with dirty matter*, in Ramazzini, 1700).

Craftsmen Pollution

The most significant parts of Ramazzini's work for craftsmen activity, and then responsible for both indoor and outdoor pollution, can be divided as follows.

Important emission of substances noxious to people and artefacts, in addition to smoke produced by combustion

> I strongly admire the work of chemists, in fact in order to accurately prepare the drugs they have to closely follow each operation exposing themselves to the fire and the smoke. Few years ago there was a legal fight between a citizen of a village called Finale and the owner of a factory. He was claiming that during the transformation of the vitriol the entire neighbourhood was contaminated. During the trial a physician, showing that the deaths were mostly of lung diseases due to the vitriol gas reached some medical evidence. Also the list of deaths recorded by a

priest was significantly higher in the neighbourhood of the plant than in the rest of the village (*Diseases of chemists*, in Ramazzini, 1700).

At that time, the word vitriol was used to denote a mineral composed of metal sulphates, mainly copper or iron sulphate (Testi, 1980). Therefore, the problem was the pollution generated by calcinating sulphur-containing minerals with a furnace inside the village centre. Vitriol containing iron was much used in the production of every type of dye and ink (Anonymous, 1740).

Important combustion, without other emissions dangerous to works of art

Other diseases affect also the chalk and lime workers. Lime is not as dangerous as chalk, but when taken out of the oven it is highly inflammable; Paolo Zachia is surprised by the fact that in some towns lime ovens are allowed, since the lime gases are very dangerous to the lungs (*Diseases of chalk and lime workers*, in Ramazzini, 1700).

It should be noted that in the preparation of chalk and lime, the carbonatic stone roasting was dangerous for the pollution due to the fuel combustion, and that the additional particulate emission was dangerous for the workmen, less for the monuments. In several parts of Europe, where charcoal, wood and limestone were abundant, lime was prepared near the building sites and constituted an important source of pollution. However, in the case of some Italian towns, it was not convenient to transport into the site both fuel and stones. For instance, in the case of Venice, lime was produced inland (mainly in the Treviso area) and then transported with boats along rivers and across the Lagoon. Therefore, the periods of intense city building, e.g. when many Baroque buildings were erected after the end of the Black Death in 1630–31, did not constitute periods of more intense city pollution.

Glass workers are the wisest. They work only six months a year, in winter and spring, and they rest on the other seasons. They retire very wisely in their forties. I was often surprised by the high toxicity of borax and antimony mixed with the glass. According to Prof. Giuseppe de Grandi, well-known physician and anatomist in Venice, in the island of Murano, home of several mirror factories, the workers have poor health conditions (*Diseases of glass worker and mirror makers*, in Ramazzini, 1700).

The indoor environment was dramatic for workmen. The furnaces were indoors, extremely hot, emitting large amounts of infra-red radiation, heat and pollution; several small windows were made all around the furnaces, which allowed the free outlet of smoke that was stagnant in the workshop. The only remedy was to use a piece of cloth over the mouth. Glass workshops are clearly described and illustrated with figures in Georgius Agricola's treatise "*De Re Metallica*" (1556). Agricola was in Italy from 1524 to 1526 and lived in Bologna, Venice and probably Padova, so he knew the Venetian furnaces later described in his treatise and remained unchanged for centuries.

Nobody ignores the terrible diseases that occur to the goldsmiths that do silver and copper gold plating. This work involves combining gold and mercury and the subsequent evaporation of the mercury eventually poisons the workers who are affected by paralysis, dizziness and assume a deathly pale appearance (*Diseases of goldsmiths*, in Ramazzini, 1700).

As it is well known, gold was applied to metals either by means of a mercury amalgam, or making pressure on a gold leaf which adhered to the hot metal. In the first case, here described, the volatile mercury was very dangerous for human health and for forming new amalgam on other metal surfaces:

Pottery craftsmen use lead based paints they manufacture melting and burning lead. During the preparation and the painting those men are exposed to the highly toxic lead gas. The painting process consisted of the following steps: pottery was first fired in the oven, then painted with a mixture of burnt lead, crushed stones and water, and after the pottery was put into the oven again for the final paint fixing phase (*Diseases of pottery makers*, in Ramazzini, 1700).

Blacksmiths are often affected by eyes diseases provoked by exposure to the white hot iron emissions of sulphur gases that irritate the eye membranes (*Disease of blacksmiths*, in Ramazzini, 1700).

In reality, although sulphur gases are explicitly mentioned, according to the scientific knowledge of that time people claimed that hot emissions and infra-red radiation were composed of sulphur, and the sulphur was considered the substance responsible for hot. In this case, workmen were affected by infra-red emission from wood or carbon combustion and hot iron as well as for some SO_2, CO_2 and CO:

Tin was harmful to manufacturers during melting and refining, to miners, during the extraction, and also to those that were melting old plates or those that were scraping the plates for cleaning (*Diseases of tinsmiths*, in Ramazzini, 1700).

Bakers sift and knead the flour, they cook the bread in the oven; those are heavy works that often cause different kind of aliments. And they work during the night; when other people sleep, they are working and they try to sleep during the day, as animals scared by the light. I noticed that bakers become ill more often in big cities, were people prefer to buy bread rather than make it by themselves; it is different in small towns or in the country, where everyone is baker himself. Pliny says that there were no bakers in Rome until the year 530 [after the foundation of Rome, i.e. 223 BC]; originally the bread was home-made. Then, when the city became too crowded, public slaves became bakers. In Rome there was a big number of mills; every district had a precise number of them, as P. Victor mentions (*De Urbis regionibus*) (*Diseases of bakers and millers*, in Ramazzini, 1700).

Jam production is a joy for people who eat it, but it can cause many ailments to the workers who prepare it. A big brass boiler is hanged on the ceiling, at the right distance from a brazier full of burning coal. [. . .] In Venice, where they make many jams, two workers shake the boiler in order to cook the fruits and let them cover with a white crust. [. . .] The coal smoke has a surprising poisoning property, which almost instantly can kill a man if there is not a way out for it. [. . .] Van Helmont, in *Jus Duumviratus*, says that, in the middle of the winter, he has writing in a small closed room heated with a brazier full of coal; he could escape from the room only with difficulty, and he fell unconscious just outside the door (*Diseases of jam makers*, in Ramazzini, 1700).

No or little combustion, but emissions dangerous to works of art

In order to dye clothes red, the dyer made large use of human urine. In a nice short poem by Martial there are several examples of stinking things, but above all the twice dyed purple wool. The use of human urine was widespread since the ancient times to clean wool and clothes and is still generally adopted in fact in the wool's factories the human urine is collected in bottles for the subsequent dye process (*Diseases of dyers*, in Ramazzini, 1700).

Actually, some modest fires were used to prepare hot urea for whitening wall and silk. Also bacteria may grow on areas contaminated by urea:

[. . .] a volatil acid comes out from the deposits of drain along the streets. This conjecture is confirmed by the following aspects: tarnishing of bronze coins that drain operators keep in their pocket, painting getting dirty when exposed to this esalation [. . .] (*Diseases of drain workers*, in Ramazzini, 1700).

What Ramazzini supposed can be identified in the hydrogene sulfide produced in the degrade of organic components:

Cervia, on the Adriatic Sea, supplies the salt to our areas and to almost all Italy. [. . .] I knew that over there the air corrodes iron; it becomes soft like wax and it ends in powder. [. . .] Probably there is so much salt in the air that it can corrode iron [. . .]. The work in the saltworks is very damaging for workers; I noticed that the walls of the warehouse where the salt from Cervia is stocked are corroded until the half of them. Between brick and brick there are cracks, due to the salt corrosive property (*Diseases of salt workers*, in Ramazzini, 1700).

In this case the damage is due to windborne salt particles, whose mechanism is well known, or accurately studied by a large number of scientists.

Unpleasant smell is the most outstanding product, but with risk for biological contamination

Nut oil is prepared as olive oil is. Kernels are crushed with a millstone, and when they are soft they are cooked on fire in a big copper pan; after this, they are pressed and the oil is extracted. During this operation black and nauseating smokes are produced, that cause the rancid exhalations that workers have to inhale. [. . .] The same problem is to be found in workshops where tanners worked; Martial (*Epigrams* liber XIII) lists all the bad odor from Taide, and remembers the one from the dogs skins macerated in Trastevere.

Also cheese makers have their troubles because of the bad smells that are typical of their work. [. . .] In Italy it is rare to find a cheese producer workshop inside the city walls: the cheese is produced for the most part in the country. In Modena however Jewish, who for their religion can eat only food prepared by themselves, take the milk from the suburbs and during the summer they produce the cheese in their district.

Nothing is stinker than the tallow candles factories. These buildings are confined to the most miserable town districts and close to the city walls, because their stink troubles not only the workers but also the houses around [. . .]. When goat, ox and pork fat begin to boil, the stench is so bad that the entire neighbourhood is affected (*Diseases of oil makers, tanners and people working with dirty matter*, in Ramazzini, 1700).

Conclusions

After Ramazzini, the first relevant conclusion is that craftsmen were not distributed everywhere in the town, but concentrated in the outskirts, divided by different categories, in order to reduce the space extension of the unpleasant effects. An example is given for the unpleasant smells:

Those are the places where work oil makers, tanners, strings makers for musical instruments, butchers, fishmongers, cheese makers, tallow candles makers. [. . .] The law prohibits doing these types of work at home; they are allowed only close to the city walls or in the suburbs (*Diseases of oil makers, tanners and people working with dirty matter*, in Ramazzini, 1700).

Therefore, an urban monument, depending upon its topographical position, if it was located far from these craftsmen centres, was practically unaffected by their emissions; if it was close, it experienced the dangerous effects of all the workshops concentrated there.

It is evident that concentrations of sulphur dioxide were commonly found for the use of burning sulphur for several domestic or workshop activities, i.e. lighting fireplaces, whitening cloths, discolouring wool and silk, disinfesting the washing or whitening roses. Particularly dangerous was the elevated SO_2 concentration reached indoor, especially in small and badly ventilated rooms, or also outdoor, when craftsmen burned important sulphur quantities. As far as air pollution is concerned, it was much more relevant the number of times people lit fires (with sulphur candles) than the total amount of timber burned.

Given the bad ventilation, the lighting made burning materials smoky, and the insufficient drought of fireplaces and the elevated indoor smoke concentration, easily led to blackening paintings, tissues, decorations and other artefacts. Candles were only rarely made of bees wax because of their high price, and in general were made of tallow; only after the mid 1800s were they made of stearin and after the end of the past century they were also made of paraffin.

The outdoor environment was also affected by the combustion necessary for several workshop activities, but other dangerous uncontrolled emissions were present, ranging from the products burned or boiled by chemists during their preparations, to the wind-borne particles of sea salt removed from the open sky deposits.

Finally, the several unpleasant craftsmen's activities, concentrated in some areas dispersed in the outskirts, where excessively bad smells were produced, also dispersed organic substances in the atmosphere, with the elevated risk of biological contamination. Dangerous emissions of N_2O, NO and ammonia are expected from the deposits of urine of dyers. The urea that reached marble or a carbonatic stone transformed the calcium carbonate into a patina of calcium oxalate, in the mineral hydrated forms of whewellite and/or weddellite (Del Monte & Sabbioni, 1983).

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