

# Air Quality Monitoring At Historic Sites

## Redefining an Environmental Classification System for Gaseous Pollution

By **Evandro Sacchi, Prof., Ing.**, Associate/Life Member ASHRAE, and **Chris Muller**, Member ASHRAE

Ten years ago environmental reactivity coupons (ERCs) were used in a study (Museum Campaign) to assess air quality in and around many cultural heritage sites in Italy. Analysis of the resulting reactivity monitoring database showed that outdoor air quality did not (generally) meet specified acceptance criteria for gaseous pollutants. Also, in a significant number of locations, indoor air was deemed unacceptable for these environments.

About the same time a project by the Istituto Centrale per il Restauro (Central Institute for Restoration [ICR]), The Risk Map of Cultural Heritage, was being completed. It is well known that air pollution and other factors were responsible for degradation of many items of historic significance. A correlation was observed between locations with the highest risk factors and those not meeting reactivity monitoring standards.

Since the original Museum Campaign, reactivity monitoring has continued in many of the same sites. These results have shown higher average reactivity levels indicating that, if anything, air pollution levels are becoming worse instead of better. If these trends continue, many cultural treasures may be lost or irreparably damaged.

Closer examination of past and present reactivity data revealed many instances where this correlation was not as strong

as expected when compared to the Risk Map. It was suspected that the standard practice of normalizing reactivity monitoring results may have artificially increased the reported severity level.

This article summarizes these findings and presents a new classification scheme for use with reactivity monitoring. The intent is to anticipate its use as a standard gage of outdoor and indoor air quality for museums, libraries, and archives as well as making recommendations for its use as a general monitoring tool.

### Reactivity Monitoring

For the last 15 years, conservators have worked to develop and refine techniques to accurately gage the destructive potential of their institution's environments towards

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#### About the Authors

**Evandro Sacchi, Prof., Ing.**, is a university professor and engineer at the Politecnico di Milano, in Milan, Italy. **Chris Muller** is technical services manager at Purafil in Doraville, Ga.



*Sistine Chapel painting showing the damaging effects of air pollution (left) and after restoration (right).*

materials and artifacts. Because little definitive information exists relating to specific levels of gaseous pollutants and the damage they cause to paper, artwork, and historic artifacts, many have turned to environmental classification via reactivity, or corrosion, monitoring. This air-monitoring technique is valid because many of the pollutants targeted for control are corrosive and, therefore, can be effectively measured using this technique.

Reactivity monitoring can be used to characterize the destructive potential of an environment by analysis of the corrosion that forms on specially prepared copper and silver coupons. It provides an excellent indication of the type(s) and level(s) of essentially all corrosive chemical types present in the local environment. Both passive and real-time reactivity monitors are in use.

**Environmental reactivity coupons.** ERCs are passive monitors exposed to the environment and then analyzed for the type and amount of corrosion that has formed. This technique can provide cumulative reactivity rates, an assessment of average environmental conditions over time, and an indication of the type(s) and relative level(s) of corrosive gaseous pollutants in the local environment.

ERCs can be used to indicate the presence of sulfur dioxide ( $\text{SO}_2$ ), ozone ( $\text{O}_3$ ), nitrogen dioxide ( $\text{NO}_2$ ), chlorine ( $\text{Cl}_2$ ), and many other corrosive materials that can cause deterioration of metals, cellulose, and organic materials. They also can detect

levels of gaseous pollutants at or below one parts per billion (*Table 1*). ERCs originally used only copper reactivity to establish environmental classifications. However, copper cannot directly detect the presence of chlorine, a particularly dangerous contaminant to metals. Therefore, the use of copper and silver reactivity monitoring is now standard. Silver is sensitive to chlorine, and, when used with copper reactivity monitoring, can provide for differentiation between various types of contaminants.

The corrosion reported from ERC analysis is actually the sum of individual corrosion films as detected by electrolytic (or cathodic) reduction. Each coupon is analyzed as to the type and amount of film present and its relative contribution to the total amount of corrosion produced.<sup>1,2</sup> For copper coupons, sulfide ( $\text{Cu}_2\text{S}$ ) and oxide films ( $\text{CuO}$ ,  $\text{Cu}_2\text{O}$ ) are most prevalent. For silver coupons, sulfide ( $\text{Ag}_2\text{S}$ ), chloride ( $\text{AgCl}$ ), and oxide ( $\text{Ag}_2\text{O}$ ) films may be observed. Reactivity



*Figure 1: Environmental reactivity coupons (ERCs) showing corrosion.*

Chemical Class	Chemical Types	Detection Limits	Corrosion Films Detected
Inorganic Chlorine Compounds	Cl <sub>2</sub> , HCl	< 1 ppb	AgCl
Strong Oxidants	O <sub>3</sub> , ClO <sub>2</sub> , HNO <sub>3</sub>	< 2 ppb	CuO, Cu <sub>2</sub> O, Ag <sub>2</sub> O
Active Sulfur Compounds	H <sub>2</sub> S, Mercaptans, Elemental Sulfur	< 3 ppb	Cu <sub>2</sub> S, Ag <sub>2</sub> S
Sulfur Oxides	SO <sub>2</sub> , SO <sub>3</sub> , (Sulfurous Acids)	< 10 ppb	Ag <sub>2</sub> S
Nitrogen Oxides	NO, NO <sub>2</sub> , N <sub>2</sub> O <sub>4</sub>	< 50 ppb	CuO, Cu <sub>2</sub> O, Ag <sub>2</sub> O

Table 1: Environmental reactivity coupon (ERC) sensitivities.

monitoring results traditionally are reported as the amount of corrosion film thickness in angstroms (Å, 1 angstrom = 10<sup>-10</sup> m) that forms (normalized to a 30-day exposure).<sup>3</sup>

Another reason for using silver coupons for reactivity monitoring is that the copper corrosion rate is sensitive to humidity changes, whereas silver corrosion is not. Small changes in humidity can significantly increase or decrease the amount of corrosion that forms on copper. Looking at corresponding pairs of copper and silver coupons can provide information as to whether the amount of corrosion formed was due more to the presence of gaseous pollutants or to humidity effects alone. This can aid in determining whether improvements to the HVAC system should be implemented as a first step or whether direct pollutant control is required.

### Control Specifications

Some research has been done to determine what pollutant levels actually cause deterioration of historical artifacts and archival materials.<sup>4-13</sup> Still, most experience has come from the determination of the background levels of these pollutants to which these materials have been exposed over the years. Normal background pollutant levels measured in nonindustrial vs. industrial areas today frequently show differences of two orders of magnitude (Table 2).

Just as wide variations exist between background and peak pollutant levels, similar variations exist in exactly what is considered acceptable levels for these pollutants. The British Museum specifies that SO<sub>2</sub>, NO<sub>2</sub>, and O<sub>3</sub> are to be removed completely. Others recommend levels from fractional parts per billion up to the low parts per million.<sup>14</sup> Although considerable variation still exists in the recommended levels, at least attempts have been made to set standard levels (Table 3).

Based on research<sup>13,15-18</sup> reactivity monitoring using copper and/or silver corrosion rates has been accepted as a preferred alternative to direct gas monitoring. It has become the standard for air quality monitoring in Dutch government archives<sup>14</sup> and is being considered as an ISO standard.<sup>19</sup>

This environmental analysis method is currently being used by many institutions and international government agencies.<sup>16, 20-27</sup>

Contaminant Measured	Normal Background Concentrations	Peak Concentrations (Urban Areas)
Sulfur Dioxide	6–30 ppb	100–750 ppb
Ozone	0.4 ppb	20–40 ppb
Nitrogen Dioxide	1.0–1.5 ppb	40–100 ppb
Chlorine	0.06–0.6 ppb	20–130 ppb
Hydrogen Chloride	20–50 ppb	200–450 ppb
Acetic Acid	4–10 ppb	20–100 ppb
Formaldehyde	3–15 ppb	10–40 ppb

Table 2: Common levels of gaseous pollutants using data from the U.S. Environmental Protection Agency and the European Environment Agency Web sites.

Contaminant/Parameter Measured	Levels Specified		Reactivity
	ppb	µg/m <sup>3</sup>	
Sulfur Dioxide	≤0.35–1.0	≤1–2.85	—
Ozone	≤2.65	≤1.8–24.5	—
Nitrogen Dioxide	≤0.94–12.5	≤5	—
Chlorine	≤1–3	≤3–9	—
Hydrogen Chloride	≤1–3	≤1.5–4.5	—
Acetic Acid	<4	<10	—
Formaldehyde	<4	<5	—
Copper Reactivity <sup>14</sup>	—	—	<90 Å/30 days <sup>a</sup>
Silver Reactivity <sup>16</sup>	—	—	<40 Å/30 days <sup>b</sup>

a—No Sulfur Corrosion Evident; b—No Chloride Corrosion Evident.

Table 3: Control specifications for conservation environments.

**Environmental Classifications.** Where direct control (e.g., air cleaning) is used to maintain indoor concentrations of gaseous pollutants as low as possible, corrosion rates ≤15–20A/30 days can be achieved. Subsequent gas monitoring has indicated pollutant levels to be at or below the limits of detection for the analytical techniques used. This “no detectable pollutants” scenario is the basis of an environmental classification system using reactivity monitoring. It is believed that if an environment exhibits corrosion rates less than or equal to Class S1/C1 (Table 4), little else can be done, economically, to improve the environment.

This classification scheme is used to correlate corrosion rates to environmental classifications directly. These classifications—both in terms of the severity level and the corresponding silver/copper reactivity rates—are being refined based on the results of testing, input from conservators, as well as the specific needs of each institution. Air purity recommendations developed based on facility and material types are as follows:

- Archives, Metal Collections, Rare Books: Class S1/C1
- Museums, Museum Storage, Libraries: Class S2/C2
- Historic Houses: Class S3/C3
- Short Term Acceptable: Class S4/C4
- Not Acceptable: Class S5/C5

In general, the reactivity rates should be Class 2 (C2/S2) or better unless otherwise agreed upon. Individual corrosion films can be used to further characterize the environment and determine proper control strategies. Based upon recommended gaseous pollutant control levels, general acceptance criteria have been determined (Table 5). These criteria account for total corrosion as well as the relative contribution of each individual corrosion film. They are more general in their application than those in Table 4 and often are used to characterize an environment prior to the implementation of pollutant control measures.

If the total corrosion and each corrosion film meet recommended criteria, the local environment in which that particular coupon has been exposed is deemed acceptable. Any of the criteria that are not met indicate that the local environment may not be sufficiently well controlled to minimize decay of artifacts and materials due to the presence of gaseous pollutants. Steps should be taken to determine what problems exist and what corrective actions may be appropriate.

### The Italian Museum Campaign

For the Museum Campaign, 187 ERCs were placed in 32 museums in 14 cities throughout northern, central, and southern Italy. The ERCs monitored outdoor and indoor air and were placed in areas exhibiting or storing (among other items) frescoes, tapestries, stone and wooden artifacts, metals, coins, jewels, mosaics, mummies, papyrus, sarcophagi, glazed ceramics, parchment, photographic negatives, and various other cultural assets.

Analysis of the resulting monitoring data showed outdoor air quality (generally) did not meet the specified acceptance criteria for gaseous pollutants. A significant number of locations were identified where indoor air quality was deemed as unacceptable for conservation purposes.<sup>22</sup>

In the 10 years since the original Museum Campaign, reactivity monitoring has continued for many of the same sites as well as some new sites. Air quality data was obtained from 150 ERCs placed in 26 museums in 15 cities. These results showed higher average reactivity levels than before indicating that, if anything, air pollution levels in Italy are worsening (Figure 2). For instance, the amount of corrosion due to reactive sulfur ( $\text{Cu}_2\text{S}$ ) and sulfur oxide ( $\text{Ag}_2\text{S}$ ) contamination has essentially doubled during this period. If these trends continue, many cultural treasures may be lost or irreparably damaged.

Silver Corrosion			Copper Corrosion		
Class	Air Quality Classification	Corrosion Amount	Class	Air Quality Classification	Corrosion Amount
S1	Extremely Pure	<40Å/30 days	C1	Extremely Pure	<90Å/30 days
S2	Pure	<100Å/30 days	C2	Pure	<150Å/30 days
S3	Clean	<200Å/30 days	C3	Clean	<250Å/30 days
S4	Slightly Contaminated	<300Å/30 days	C4	Slightly Contaminated	<350Å/30 days
S5	Polluted	≥300Å/30 days	C5	Polluted	≥350Å/30 days

Table 4: Environmental classifications for conservation environments.

Silver Reactivity		Copper Reactivity	
Silver Corrosion Reaction Products	Corrosion Film Thickness	Copper Corrosion Reaction Products	Corrosion Film Thickness
Silver Chloride, $\text{AgCl}$	0Å/30 days	Copper Sulfide, $\text{Cu}_2\text{S}$	0Å/30 days
Silver Sulfide, $\text{Ag}_2\text{S}$	<50Å/30 days	Copper Oxide, $\text{Cu}_2\text{O}$	<150Å/30 days
Silver Oxide, $\text{Ag}_2\text{O}$	<50Å/30 days	Copper Unknowns	0Å/30 days
Total Silver Corrosion	<100Å/30 days	Total Copper Corrosion	<150Å/30 days

Table 5: General reactivity monitoring acceptance criteria.

### Risk Map of Cultural Heritage

At the same Museum Campaign was being completed, ICR was completing what would become the largest databank on cultural heritage in Italy.

The ICR started implementation of a geographic information system (GIS), called “MARIS” (MAppa RISchio, i.e., Risk Map), a territorial information system devised by the ICR as a support instrument for decisions concerning the preservation of Italy’s cultural heritage.<sup>28,29</sup>

At present, the Risk Map provides information from the areas delimited by the administrative boundaries of the 8,100 Italian municipalities and, for some factors, by the boundaries of the 300,000 census sections. The main users of this information are the Departments of the Ministry for the Assets and Cultural Activities who are charged with the safeguarding, conservation, and maintenance of archaeological, architectonic, artistic and historic assets in Italy.

The information contained within the geographic information system of the Risk Map makes it possible to calculate the severity of the risk to which each monumental and historical artistic asset of the Italian cultural heritage is subject. It also provides the opportunity to become familiar with their distribution throughout Italy via thematic cartographic representations that can be constantly updated.

The primary damage phenomena are divided into three risk categories: static (structural) risk, environmental-air risk, and risk from human factors (anthropic risk). The data collected have been processed which, on the basis of the phenomenon

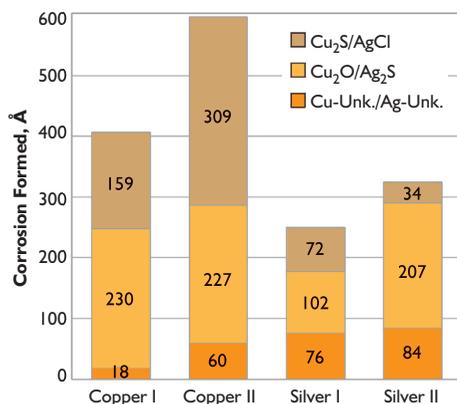


Figure 2: Summary of ERC data (average values) from the Museum Campaign (Copper I/Silver I) and in the years since (Copper II/Silver II).

presence or absence, and of its extent and intensity, permits a danger value (index) to be assigned to each single municipality.

For a definition that encompassed as much of the environmental air risk as possible, three different independent chemical-physical indicators were identified: the erosion indicator, the blackening indicator, and the physical stress indicator. Of these, the blackening indicator takes into account the air quality data relevant for use with reactivity monitoring.

To calculate the blackening index, all available regional and municipal air monitoring data was reviewed and, in particular: municipal SO<sub>2</sub>, NO<sub>x</sub> and suspended particulate matter (PST) emissions, sulfur deposition, acid rain mean values, and meteorological and climatic variables. This data was obtained from existing, historical data banks and through mobile and fixed monitoring stations. Monitoring data included that for PST, black smoke, sulfur dioxide, nitrogen oxides, rain pH, and ozone.

#### Risk Map vs. ERC Data

Comparing the ERC results with the environmental air data from the Risk Map resulted in a correlation between locations with the highest risk factors and those not meeting the specified reactivity monitoring standards. This was expected as those areas showing the highest risk factors were also those with the poorest air quality.

A closer examination of the reactivity monitoring data revealed that at some locations existed where this correlation was not as close as would have been expected when compared to the Risk Map. It was suspected that the practice of normalizing the reactivity monitoring results might artificially increase the reported severity level, especially for exposure periods longer than 60 days.

Copper corrosion data is normalized to a 30-day exposure period using the following equation.<sup>3</sup>

$$x_1 = x(t_1/t)^A$$

where

- $x_1$  = the equivalent film thickness after one month
- $x$  = the measured film thickness after time  $t$
- $t_1$  = thirty days
- $t$  = actual test time (days)
- $A$  = 0.3 for <300Å, 0.5 for <1,000Å, or 1 for >1,000Å

This equation comes from research that established copper corrosion is nonlinear<sup>1,2</sup> and that the main corrosion products

City	ERC Exposure		Normalized Copper Data		Measured Copper Data		Changes in Severity Level Based on New Risk Index
	Time, Days		Total	Class	Total	Class	
Bologna	91		44	1	62	1	0
Florence	95		44	1	62	1	0
Genoa	90		192	3	333	2	-1
	90		238	3	413	2	-1
Mantova	112		41	1	62	1	0
	112		41	1	62	1	0
	112		41	1	62	1	0
Milan	110		168	3	322	1	-2
	110		142	2	210	1	-1
	91		270	4	469	3	-1
Minori	95		44	1	62	1	0
Paestum	95		44	1	62	1	0
	95		44	1	62	1	0
Palermo	35		159	3	167	2	-1
	89		161	3	223	1	-2
	89		157	3	217	1	-2
	89		200	3	345	2	-1
	92		143	2	200	1	-1
	89		161	3	223	1	-2
	92		160	3	224	1	-2
	89		157	3	218	1	-2
	92		190	3	333	2	-1
89		200	3	344	2	-1	
Pontecagnano	95		44	1	62	1	0
Rome	63		610	5	884	5	0
	61		135	2	167	1	-1
	89		185	3	319	2	-1
	93		397	5	699	3	-2
	63		359	5	520	3	-2
	63		3,125	5	6,563	5	0
Vatican City	95		180	3	254	1	-2
Venice	86		207	3	284	2	-1
	86		567	5	960	4	-1
	86		347	4	588	3	-1
	86		63	1	86	1	-0
	91		1,072	5	3,252	5	-0

Table 6: Sample data from Museum Campaign compared to values obtained using this new classification scheme. Data is from selected cities where the ERCs had been exposed for at least 60 days.

readily discernable were sulfides and oxides. Silver corrosion is considered to be (essentially) linear with the main corrosion products being chlorides, sulfides, and oxides.

**An Updated Environmental Risk Index for Reactivity Monitoring.** It had been postulated by some of those reviewing the ERC data from the original Museum Campaign that a classification system using values over a given range could cause the results near the boundaries to overestimate or underestimate the actual risk depending on exposure time. Using reactivity monitoring, the actual corrosion risk may be difficult to delineate and “Classes” have to account for ana-

lytical error, background corrosion, etc. Thus, the question “Is the difference between 99Å and 100Å (S2 vs. S3) really enough to warrant a more severe environmental classification?” is valid.

Using the same reactivity monitoring classifications and acceptance criteria shown in Table 4, an environmental assessment scheme was introduced using the actual corrosion amounts and exposures time instead of the normalized data.<sup>22,25,26</sup> Reviewing the available ERC data including that from 1996 to the present has resulted in a proposal to switch from the current acceptance criteria based on stepwise classifications (e.g., C1, S2) to one based on a continuous numerical risk index (Figure 3). With these new criteria, air quality problems are evaluated in a continuum with more accuracy and relevance. It is generally agreed that this numerical index is a better representation of the actual corrosion risk.

Reactivity rates that fall below or right of the shaded areas indicate an environment not considered to be at risk from gaseous pollutants with the exception being those areas housing or storing archives, metal collections, and/or rare books (see air purity recommendations earlier). Those values that fall within the shaded areas indicate the potential exists for damage or deterioration from exposure to the environment and that continued monitoring is indicated. Values above or to the left of the shaded areas would be considered to be at high risk of damage and direct control of contamination is indicated.

Using this index with the results from the Museum Campaign revealed that the copper reactivity rates were overstated by at least one classification level more than 35% of the time with long (>60 days) exposure periods and more than 28% of the time for exposure times between 30 and 60 days. These rates were being overstated by two levels more than 10% of the time. Using this new risk index meant that 80% of these coupons would be reclassified as Class 1 or 2. The silver reactivity data was not affected.<sup>24</sup>

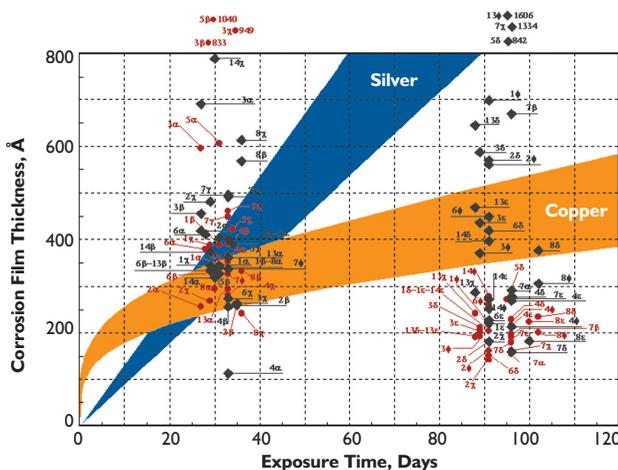
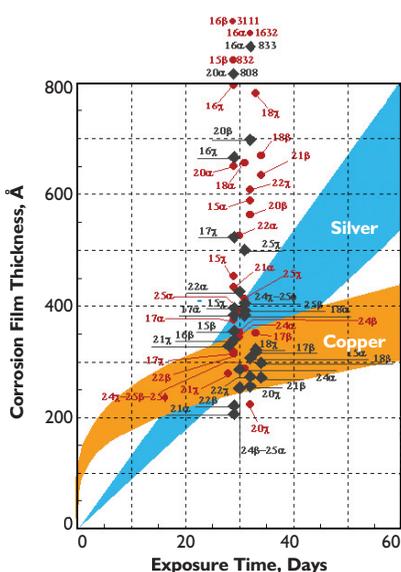


Figure 4 (left): ERC data for northern Italy (normalized to 30-day exposure).

Figure 5 (right): ERC data for northern Italy (based on numerical risk).

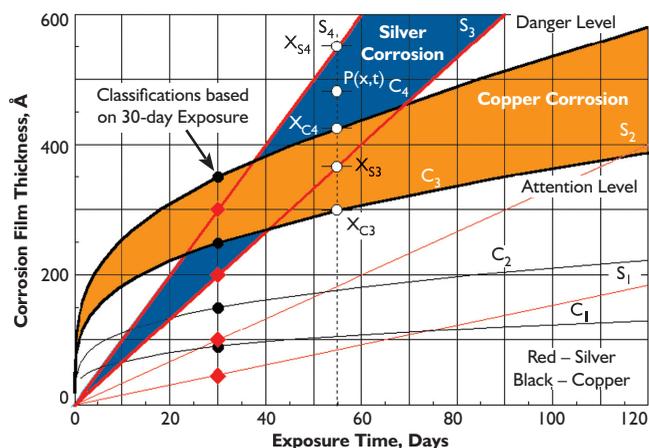


Figure 3: Environmental classifications based on numerical risk index.

Table 6 shows normalized copper ERC data from the original Museum Campaign compared to values obtained using this new classification scheme. Data shown is from selected cities where the ERCs had been exposed for at least 60 days.

Examples of ERC data plotted according to this new risk index are shown in Figures 4 and 5. Figure 4 shows how the data would appear when normalized to a 30-day exposure. Figure 5 shows actual corrosion data without normalizing for exposure periods. Note that the silver data is not affected as corrosion on silver is assumed to be linear.

This becomes significant for indoor locations using direct control of gaseous pollution. Due to the typically lower level of pollutants encountered indoors, ERC exposure times averaged 60–70 days. Normalizing the data makes it appear in some cases that the air cleaning systems were not providing the specified air quality. Looking at the data in a continuum provides a more representative picture of air quality with respect to gaseous pollutants and even shows some areas that may have been considering the application of some form of air cleaning technology may be able to meet their specific air quality criteria without direct control of gaseous pollutants.

## Summary and Conclusions

Reactivity monitoring can be a primary indicator of how well controlled an environment is with regards to gaseous pollutants. If an environment exhibits an S1/C1 classification, little can be done economically to improve the environment. If the general acceptance criteria of S2/C2 are met, with the possible exception of certain papers and metal collections, this generally indicates an environment sufficiently well controlled as to prevent damage to objects and artifacts. Where appropriate air cleaning technologies are used, reactivity levels well within specified acceptance criteria can be readily attained.

A new standard classification system has been proposed that establishes a continuous numerical risk index to convey information on environmental air quality to conservators. With this new index, there is good correlation between the results of reactivity monitoring and the environmental air risk for the same municipalities as determined by the Risk Map.

Recommendations are being made to the ICR for using reactivity monitoring (i.e., ERCs) and this environmental classification scheme as a low-cost ambient air quality monitoring tool. The use of ERCs also can provide information on the presence of chlorine, which is a most dangerous contaminant for metals. Their use allows an expansion of the type of data available for the determination of danger (risk) factors as well as for continuous and relevant updates to the Risk Map. All of this with the intention of helping to protect and preserve Italy's and the world's historic and artistic assets.

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