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PRODUCTS OF INCOMPLETE COMBUSTION* (O_x, CO_x, HO_x, NO_x, SO_x, RO_x, MO_x, AND PO_x)

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Summary

Perceived needs to identify easily measured surrogates for products of incomplete combustion (PICs) from hazardous wastes incinerators have spawned a spirited scientific debate. Can any simple surrogates be correlated with PICs? What surrogates can be measured and at what levels? How should surrogates be normalized, averaged, and reported?

Carbon monoxide (CO) and total hydrocarbons (THC) are two possible candidates. Interim guidelines of 100 ppm for CO and 20 ppm for THC have been suggested by the U.S. EPA for use by permit writers. Formal proposal, public comment, and promulgation as regulatory standards are being considered. Although CO, and possibly THC, have some utility as parameters of operation, both have recognized limitations as parameters of performance. There are also shortcomings in their measurement, normalization and statistical averaging.

Correlations of CO or THC with PICs, or with each other, are not quantitative. Continuous emission monitoring (CEM) is difficult. Interpretation of results is equivocal. Alternatives to technology-forcing "guidelines" are needed. Flexible control strategies, further development of CEM monitoring technology, valid statistical analysis of emissions, and consideration of composite parameters are encouraged.

Introduction

Increased attention is being directed at proper management of hazardous materials and hazardous wastes. This had led to increased recognition of a hierarchy of recovery, treatment, and disposal [1]. Shifts of emphasis from landfilling to incineration have created newly perceived needs to insure protection of public health and the environment. Attempts have been made to identify valid and easily measured surrogates for Products of Incomplete Combustion (PICs) that may be emitted from hazardous wastes incinerators. A spirited scientific debate had arisen [2]. Can simple surrogates be correlated with PICs? What surrogates can be measured and at what levels? How should surrogates be normalized, averaged, and reported?

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The search for surrogates has led to indiscriminant transfer of parameters used in existing combustion technology that were developed for purposes other than destruction of trace constituents. The consequences of transfers of parameters and parameter levels from combustion to destruction processes have not necessarily been fully interpreted. Various combustion and destruction processes need to be distinguished.

Carbon monoxide (CO) and total hydrocarbons (THC) are two minor volatile fractions of emissions that have been most often mentioned as possible surrogates for PICs. Interim guidelines for one or both are being used currently by the U.S. EPA to set operating conditions in permits for hazardous waste incinerators. The guideline for CO is 100 ppm normalized to 7 vol.% O_2 and arithmetically averaged over one hour. The guideline for THC of 20 ppm on a similar basis is a "variance" to CO. There has been limited scientific peer review of these guidelines by the regulated community. No formal proposal, public comment period, or promulgation of regulatory standards have occurred for CO of THC for hazardous waste incinerators. Revisions have not appeared that reflect comments and reservations on the setting of parallel CO standards for industrial boilers and furnaces. Final regulations have not been promulgated.

Although CO, and possibly THC, have utility as parameters of operation, both have recognized limitations as parameters of performance. Correlations of CO or THC with PICs, or POHCs, or with each other, are not quantitative at the low levels being suggested as guidelines [3–5]. Continuous emission monitoring (CEM) is not an easy matter on a real-time basis. Statistical quality control should be applied to avoid equivocal interpretations [6]. Alternatives to technology-forcing "guidelines" should reflect real-world conditions [7]. Flexible control strategies, further development of CEM monitoring technology, valid statistical analysis of emissions, and consideration of composite parameters are encouraged.

Distinctions among thermal processes

Thermal processes encompass a variety of oxidation reactions conducted at high temperatures in enclosed devices. In thermal processes, "reactants" are converted to "products", reaction rates are hastened, and some desirable equilibria are favored by applied or generated heat, The primary process may be: (1) recovery of energy from fuels, (ii) production or recovery of materials from byproducts, or (iii) treatment of wastes to reduce volume and hazard [1]. Physical and chemical conditions may be similar, but there are major differences, however, in the types of reactants and products, and in how each process is monitored, controlled, and evaluated. For boilers, the reactant is a fuel, the product is energy. For furnaces, the reactant is a raw material or process intermediate, the product is a usable material. For incinerators, the reactant is a waste, the "products" are treated gaseous emissions, liquid wastewaters, and solid residues.

"Gaseous" emissions are released by all "combustion" processes. Emissions may be true gases or vapors, liquid droplets, and airborne solid "particulates". Individual constituents in all phases may include: Products of Incomplete Combustion (PICs) and Products of Complete Combustion (PCCs) [3]. In conventional combustion, carbon is converted to carbon dioxide, hydrogen to water, and other elements to their terminal oxidation products. Air pollution control is applied to cool emissions to ambient temperature and to reduce combustion products of potential environmental or health concern to acceptable levels before release. This may require removal of acid gases, water droplets, and fine particulates. Treated emissions are then released to the atmosphere.

"Combustion" is not synonymous with "destruction". Combustion Efficiency (CE) is a measure of completeness of energy release from carbonaceous fuel and its relative conversion to a PIC (CO) and a PCC (CO_2). Destruction and Removal Efficiency (DRE) is a measure of how completely an original constituent in a feed is destroyed and/or separated for subsequent treatment before any residual is released in an emission. PICs found in emissions are not usually the same compounds identified as Principal Organic Hazardous Constituents (POHCs) in feeds during trial burns.

Performances of hazardous waste incinerators are determined during trial burns to insure performances during normal operations [8]. Performances of energy or material recovery facilities are demonstrated fairly directly, since raw material usages or fuel consumptions are relatively constant. Performances of incinerators having variable feeds and feed rates are more difficult and less direct to demonstrate [9]. Complex interactions among multiple trace constituents in emissions must be estimated.

Combustion and destruction: Reactants and products

In both combustion and destruction processes, some distinctions need be made to tell what 'goes in' (reactants) from what 'goes out' (products) [10,11]. A series of mnemonics helps to categorize constituents: O_x, CO_x, HO_x, NO_x, SO_x, RO_x, MO_x, and PO_x; i.e. oxygen (O_x), carbon oxides (CO_x), water (HO_x), nitrogen oxides (NO_x), sulfur oxides (SO_x), solid particulates (RO_x) ("rocks"), and metals and metallic oxides (MO_x). Particular organic compounds (PO_x) (né PICs and POHCs) are of special interest. Not all products include oxygen. They also may be unreacted O_2 or N_2, residual reactants (POHCs), or products (PICs and PCCs).

Relative concentrations of emission constituents depend upon types of reactants, conditions of thermal reaction, and treatment of products [12]. Constituents can be ranked in approximate relative order of abundance:

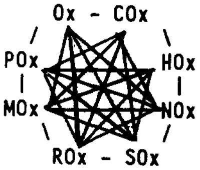

Fig. 1. Interactions among emission constituents.

$O_x > CO_x > HO_x > NO_x > SO_x > RO_x > MO_x \gg PO_x$

Concentrations may range over more than twelve orders of magnitude. Major constituents (N_2, O_2, CO_2, H_2O) may be several percent; minor constituents (CO, NO_x, SO_x, HCl) from fractions of a percent to parts per million; and trace constituents (POHCs, PICs) at lower levels, i.e. $>$ppb$<$ppt. Interactions that might occur among species can be pictorialized (Fig. 1). Some species may be simply linked to molecular oxygen. Other links may exist to various degrees among all species.

The validity of CO as a single "stand-alone" surrogate for composite PICs or DRE of POHCs is limited [13]. Complex interactions occur among CO and other trace constituents [14]. Heterogenous, high-temperature, unsteady-state interactions under both reductive and oxidative conditions make practical interpretation difficult. These include: (i) formation of both CO and THC from CH_4, (ii) formation of CO from carbon particulates, (iii) decreased NO_x and increased CO upon addition of H_2O, (iv) formation of CO from thermal degradation of CO_2, and (v) thermal "quenching" of CO upon rapid cooling emissions [3, 12, 15, 16].

Normalization of aerometric data

Once the limitations of monitoring discrete emission constituents are recognized, the resulting data must be properly assessed. "Normalization" of the concentration of one emission constituent to a reference level of some other constituent or condition is done only for comparative purposes. It should not be construed as a "correction" to some more desirable level! Concentrations are routinely adjusted from a wet-gas to a dry-gas basis, and from stack conditions to ambient temperature and pressure. Such normalizations, however, do not impose unforeseen "penalties", or lead to erroneous comparisons [8].

Rotary kiln incinerators operate optimally at slight negative pressures and at relatively high residual O_2 levels (11–14 vol.%). Sufficient, but not excessively high, O_2/fuel ratios insure optimal POHC and PIC destruction. Potential explosions which might occur at low O_2 levels are prevented. Addition of too much air is unrealistic, since high O_2 levels lead to undesirable thermal quenching and flameouts, and consumption of excessive fuel. In contrast, boil-

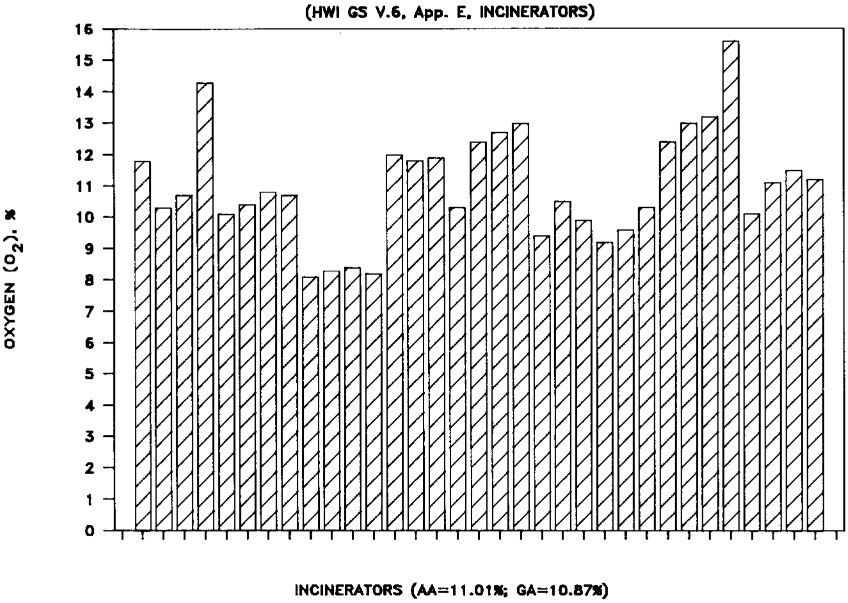

Fig. 2. Comparison of O_2 levels.

ers may operate at O_2 levels closer to stoichiometry ($O_2 < 3$ vol.%). CEM of O_2-sensitive parameters, such as CO and THC, have only recently been addressed for incinerators. Increased levels of CO, THC and other PICs are formed at both low and high O_2 extremes with optimum operation at some intermediate O_2 level [12,14]. Statistical variability has precluded exact determination that transients are enhanced by low O_2 [17] or dampened by high O_2 [18]. The consequences of normalization of CO to an artificially low level of O_2 has not been sufficiently reviewed, and thus is not considered to be a standard practice for incinerators.

If an arbitrary level of 7 vol.% O_2 intermediate between boilers and incinerators, is chosen for normalization, then a factor of ($21 - O_2$ chosen)/($21 - O_2$ observed) is applied. Any CO value normalized to 7 vol.% O_2 is affected by a "hidden" factor which varies from 2/3 @ 0 vol.% O_2 (stoichiometric), 1 @ 7 vol.% O_2, 2 @ 14 vol.% O_2, 3 @ 17.5 vol.% O_2, and infinity @ 21 vol.% O_2. A "standard" of 100 ppm CO at 7 vol.% O_2 becomes 150 ppm @ 0 vol.% O_2, 50 ppm @ 14 vol.% O_2, 33 ppm @ 17.5 vol.% O_2, and 0 ppm @ 21 vol.% O_2.

Background CO from combustion of supplemental fuels in the absence of waste at levels over 7 vol.% O_2 can approach 100 ppm. Little room remains for control within normal operating limits of fuel/air modulation. The upper control limit may rise above the "guideline"; the lower control limit may drop below "background". It is not unusual, during short shutdowns for flameouts or long shutdowns for maintenance, for O_2 to rise toward 21 vol.%, while both

CO and CO_2 drop toward 0 vol.%. The restriction of a 7 vol.% O_2 normalization for CO or THC is seen in the technical data for nine hazardous waste incinerators (Fig. 2) [19]. The average of 11 vol.% O_2 is coincidently the same level used by countries other than the US for normalization of aerometric data other than CO. Normalization to 7 vol.% CO_2 (~ 14 vol.% O_2) has also been used. Levels of 11–14 vol.% O_2 are more realistic for normalization.

Limits of correlation among CO, THC, and PICs

Carbonaceous compounds react with molecular oxygen at high temperatures to form carbon dioxide, elemental carbon, and carbon monoxide [20]. Both CO and CO_2, are used to calculate Combustion Efficiency ($CE = 1 - CO/CO_2$). CE is not a quantitative measure of Destruction and Removal Efficiency ($DRE = 1 - POHC_{in}/POHC_{out}$) of Principal Organic Hazardous Constituents (POHCs), which are selected on the basis of perceived abundance and incinerability. CE includes a ratio of two different products; DRE includes a ratio of a single constituent as both reactant and product.

Both CO and THC are relatively minor PICs that are formed from various POHCs or other PICs during the burning of natural gas, fuel oil, and coal, as well as hazardous wastes. A major intermediate is particulate elemental carbon [21,22]. "THC" is a broad category of volatile hydrocarbons, including methane (CH_4), ethane, and propane [23,24]. CO and CH_4 are not usually thought

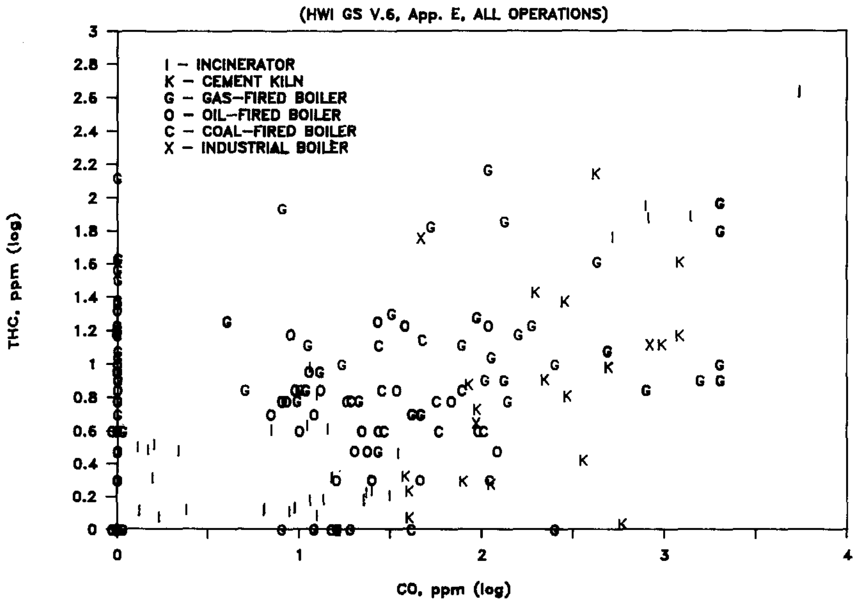

Fig. 3. Comparison of CO and THC emissions.

of as either "PICs" or POHCs", but they can often comprise the bulk of volatile emissions [19,25]. Both CO and CH_4 are relatively thermally stable compounds compared to most organics [23].

The largest fractions of total volatile PICs are compounds with low molecular weights, such as CO and CH_4 [23]. These are of relatively minor environmental consequence in the context of incinerator emissions. Non-volatile PICs and/or those with high molecular weights are associated with particulates [21,22,26,27]. An alternative PIC indicator may be unburned carbon particulate. This parameter cannot be directly measured with currently available techniques. It must be determined indirectly from CO, CO_2 and O_2 [13].

Use of single operating parameters, such as CO or THC, to estimate incinerator performances have not been quantitative [12,15]. Such single surrogates only qualitatively approximate overall destruction/removal of POHCs or formation of PICs [7,28,29]. CO has some utility as an operating parameter, but it is decidedly site-specific and unit-related. CO is unsuitable as a quantitative indicator of specific POHC destruction or specific PIC formation. Very high levels ($CO \gg 1000$ ppm) only approximate reduced destruction of more easily burned organic species. THC may appear to offer a better correlation to total PICs, but correlation between CO and THC, and between CO or THC and total PICs are imperfect.

Complete quantification is impossible, but the fraction of identified/total PICs could be better focussed. A limited number of constituents could be monitored by using a cost-effective risk-driven strategy [30]. The limits of CO and THC as valid surrogates for PICs can be seen by reviewing data available for hazardous waste incinerators, cement kilns, and boilers (Fig. 3) [19]. Considerable scatter of data is shown on the logarithmic plot of CO vs. THC. Approximate groupings are made of similar types of units and units burning similar fuels. The tentative guidelines of 100 ppm CO and 20 ppm THC lie serendipitously in the middle of the plot.

Statistical analysis of environmental data

Numerical representations and statistical analyses profoundly affect interpretations. Logarithmic scales are convenient to express extreme ranges of data otherwise obscured when using arithmetic scales. Examples include: seismic disturbances (Richter units), loudness of sounds (decibels), and hydrogen ion concentrations (pH units). Statistical distributions are not adequately described by the familiar symmetrical bell-shaped curve if arithmetic scales are used. Some values cluster about an arithmetic mean with a "tailing off" of values to one side. For a lognormal distribution, the logarithms of concentrations rather than the concentrations themselves are normally distributed about a geometric mean [31]. Concentrations of many environmental parameters, e.g. radionuclides in soil, metals in biological tissues, discharges from wastewater treatment plants, and indoor and ambient air constituents, follow logarithmically normal distributions [6,32–35]. Aerometric parameters exhibiting lognormality include: CO, THC, NO_x, SO_x, and particulates (RO_x) [13,15,36,37].

The ubiquity of lognormality of environmental parameters has several possible explanations [38]. Assumption of a constant source subject to a succession of random dilutions is one possibility [11]. This might be the case where both source and dilution factors are relatively independent. An alternative explanation is necessary assuming a variable source that is the product of several sequential and/or simultaneous events [3]. This might be the case for the apparent generation of CO in a hazardous waste incinerator.

In a rotary kiln incinerator, air flow is relatively constant, but both types and rates of fuel and waste flows may vary. In addition, perturbations in residence time, combustion temperature, and mixing turbulence. (the "3 T' s" of incineration) may induce regular and random transients in the concentrations of emitted constituents. These perturbations can be caused by routine and unscheduled maintenance, as well as by controlled responses to variables affecting operation and performance [17,39]. Transients of super imposed "events" occur during normal operations of large variable feed incinerators [3,7].

Environmental assessments of incinerators are influenced by differing interpretations between normal and lognormal distributions of CO [8]. This does not mean that the data are abnormal or atypical or nonrepresentative, but simply that different statistics may be more appropriate [40]. Commonly used "arithmetic" averages are not appropriate for data extending over orders of magnitude. Excessive statistical "weighting" of a few extreme values leads to an apparent "average" well removed from the majority of the data. In such cases, the "geometric average" is more valid. Example calculations of arithmetic and geometric averages are included in the appendix. Recognition of lognormal distributions and use of geometric averages are encouraged.

Interpretation of CO data from a rotary kiln incinerator

EPA permit writers have previously used CO as an operating parameter, but only for guidance purposes, and based solely on trial burns [41,42]. Any regulatory reproposal [43], public comment period, Agency review, and assessment of recommendations of EPA's Science Advisory Board [44] may extend well into 1990. In the meantime, EPA has indicated an intention to begin to permit facilities operating under interim status. Omnibus authority will be used to apply preliminary guidelines for CO and THC [41], and metals and HCl [45]. These guidelines as applied to incinerators have received no formal peer review by the regulated community. They also depend upon subsequent

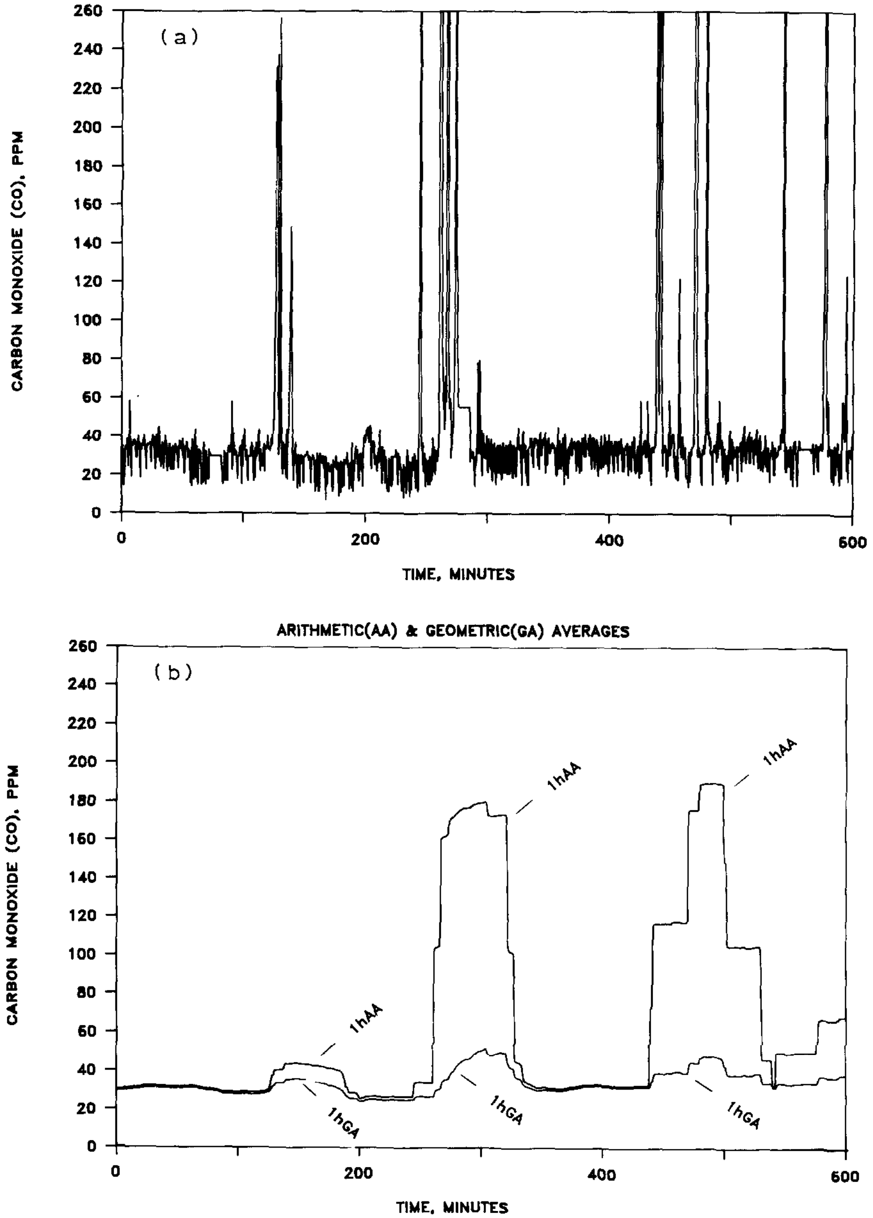

Fig. 4. Continuous emission monitoring of CO; 9a) 30 s values, (b) arithometic (AA) and geometric (GA) averages.

interpretations of what is a "hazardous" waste, a "mixture", and a "residue" [46].

"National Standards" have been informally suggested for CO and THC levels of 100 and 20 ppm, respectively, adjusted to 7 vol.% O_2, and arithmetically averaged over a one-hour period [41]. The limitations of the CO guideline have been previously discussed [3,13]. The limitations of the more recently conceived THC-guideline are less apparent. The hypothetical composite THC is assumed to include 115 compounds, including 72 carcinogens and 43 noncarcinogens, with an average molecular weight of 45.34 g/mol. These include: 37 measured at some HWI; 75 assumed to be present at 0.1 ng/l; formaldehyde from a municipal incinerator; and methane and ethane from fossil fuel burners. Over half of the assumed risk (rounded to 10^{-5}) is associated with only 0.33% by weight of the assumed PICs, many of which are not true VOCs and would be undetected as THC! Almost 90% of the assumed risk is associated with compounds assumed to be present at <0.1 ng/l which cannot be quantitated by THC. About 68.5% of the 20 ppm total is attributed to methane and ethane which contribute nothing to the assumed risk.

Continuous emission monitoring (CEM) of large rotary kiln incinerators includes CO, CO_2, and O_2 [3,9,12,15], Details have been provided in workshops [10,48], cooperative industry–university research [43], and comments to Federal and State regulatory agencies [13,49,50]. Example CO monitoring data (Fig. 4) include "instantaneous" (30-s) values together with one-hour geometric and arithmetic "rolling averages" normalized to 7 vol.% O_2. Calculations of rolling averages induce three effects: decreases of peaks, increases in valleys, and shifts forward in time. The arithmetic average overly magnifies the effect of a short duration spike. The geometric average yields better statistical interpretation.

Conclusion

Preferable alternatives to assuming fixed numerical levels for "National standards" for CO and THC include: (i) use of flexible site-specific control strategies to reduce apparent CO levels, (ii) assessment of CEM monitoring technology, (iii) expansion of the emissions database, and (iv) development of composite parameters.

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