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# ASSOCIATION OF CARBON DIOXIDE WITH INDOOR AIR POLLUTANTS AND EXCEEDANCE OF HEALTH GUIDELINE VALUES

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#### SUMMARY

This paper focuses on the correlations between concentrations of carbon dioxide ( $CO_2$ ) and pollutants in two large-scale measurement campaigns, namely formaldehyde, acetaldehyde, acrolein, benzene and particulate matter. In both campaigns, the relation between  $CO_2$  and pollutant weekly average concentrations was studied. With high concentration of  $CO_2$  in dwellings and schools, the probability to exceed the pollutant health guideline values for long term exposure increased. However, dealing with  $CO_2$  is not enough to prevent exceedances to occur.  $CO_2$  as a tracer of some pollution sources cannot be considered as a unique indoor air quality indicator.  $CO_2$  is also often considered as a surrogate of ventilation rate. Hence, this study confirms that even with good ventilation conditions (i.e. low  $CO_2$  level), the reduction of pollutant sources remains necessary to achieve a satisfactory indoor air quality.

#### **INTRODUCTION**

Several studies have reported associations between  $CO_2$  indoor air concentration and health symptoms (Seppänen et al., 1999; Simoni et al., 2010; Sundell et al., 2011; Wargocki, 2013) or human performance (Satish et al., 2012). These associations were observed at  $CO_2$ concentrations around or above 1,000 ppm, far below known  $CO_2$  toxicity values which are above 10,000 ppm (Guais et al., 2011), and may indicate the presence of other pollutants. A guideline value for  $CO_2$  is provided in French regulation, i.e. 1,300 ppm in non-smoking and non-residential buildings (RSDT, 1978). However, this guideline is not health-based. This study is part of a larger French study looking at the opportunity to recommend a health-based guideline value for  $CO_2$  in indoor air (ANSES, 2013a). This paper aims to test if  $CO_2$ concentrations of  $CO_2$  and priority air pollutants in French dwellings and schools, namely formaldehyde, acetaldehyde, acrolein, benzene and particulate matter. In particular, the probability that these pollutants exceed health guideline values was calculated according to  $CO_2$  concentration classes.

#### **METHODS**

The pollutants considered in this study were previously defined as priority pollutants in indoor air according to a health-based ranking (ANSES, 2011). Two sets of data were used to study the statistical relations between  $CO_2$  and indoor air pollutant concentrations:

- A national survey on 567 French dwellings carried out between 2003 and 2005 (Kirchner et al., 2007, 2008). The measurements were performed in the main bedroom during 7 days either during heating or non-heating season. The following measured pollutants were considered in this study: CO<sub>2</sub> monitored every 10 minutes, formaldehyde, acetaldehyde, acrolein and benzene measured with diffusive sampling over 7 days, and particulate matter fractions PM<sub>2.5</sub> and PM<sub>10</sub> with active sampling on evenings and nights from 5 pm to 8 am and week-ends;
- A national survey on 310 French nurseries, kindergartens and schools carried out between 2009 and 2011 (Michelot et al., 2013; Ramalho et al., 2013). The measurements were performed during 4.5 days from Monday to Friday during both heating and non-heating season, in a total of 896 rooms. Formaldehyde and benzene were measured in the air of classrooms or playrooms by di ffusive sampling, and CO<sub>2</sub> was monitored every 10 minutes.

Associations between 1-week average concentrations of  $CO_2$  and pollutant were expressed as linear correlations (Pearson). The observed correlations were confirmed using rank correlations (Spearman) and Pearson correlations on log-transformed data. Thus, the non-normality of data distribution has little incidence on the observed correlations.

The long-term health-based guideline values for inhalation of formaldehyde, benzene (for an excess individual lifetime cancer risk of  $10^{-5}$ ) and acrolein were defined by the French Agency for Food, Environmental and Occupational Health & Safety (ANSES), respectively as  $10 \ \mu g/m^3$ ,  $2 \ \mu g/m^3$  and  $0.8 \ \mu g/m^3$  (Mandin et al., 2010; ANSES, 2013b). For acetaldehyde, the reference concentration for chronic inhalation exposure (RfC, US EPA, IRIS) of  $9 \ \mu g/m^3$  was adopted as guideline value. For PM<sub>2.5</sub> and PM<sub>10</sub>, the ambient air guidelines defined by the World health Organization (WHO) for long-term exposure, were taken into account, 10 and 20  $\mu g/m^3$  respectively.

In order to assess the probability of exceedance of the guideline values, the  $CO_2$  concentration averaged on the weekly occupancy period was separated in two classes according to a given threshold. Four different thresholds were tested:

- 750 ppm: 350 ppm default value above outdoor level (400 ppm) for the highest indoor air quality category recommended in standard EN 15251 (2007)
- 1,000 ppm: guideline value provided in French regulation for non-residential buildings
- 1,500 ppm: guideline value for schools in the United Kingdom.
- 2,000 ppm: high limit value for average CO<sub>2</sub> level observed in French schools during occupancy (Ramalho et al., 2013).

The concentration of  $CO_2$  in different classes was considered as a diagnostic test to detect cases where the concentration of a given pollutant exceeds its guideline value. The quality of this test is defined by different statistical parameters (Pepe, 2003) as presented in Table 1.

Table 1. D iagnostic test for exceedance detection of a guideline value for a pollutant according to a given  $CO_2$  concentration threshold.

	[Pollutant] > Guideline	$[Pollutant] \leq Guideline$	
[CO <sub>2</sub> ] > threshold	True positive (TP)	False positive (FP)	Positive predictive value (PPV) = $\frac{TP}{TP + FP}$
$[CO_2] \le threshold$	False negative (FN)	True negative (TN)	Negative predictive value (NPV) = $\frac{TN}{FN + TN}$
	Sensitivity (TP Rate) =	Specificity (TN Rate) =	Test accuracy =
	TP		TP + TN
	$\overline{TP + FN}$	FP + TN	$\overline{TP + FP + FN + TN}$

The gain of using the concentration level as a mean to predict exceedance of guideline value for a given pollutant was also calculated in several ways. The positive diagnostic gain was determined as the difference between PPV and prevalence, where prevalence represents the effective exceedance rate in the sample. It represents the gain in probability to use the  $CO_2$ level as a test to detect exceedances compared to the initial or pre-test probability. The negative diagnostic gain is defined as the difference between NPV and (1-prevalence) and represents the gain in probability to detect "safe" results (below guideline) when  $CO_2$  level is lower than the threshold. The overall gain in accuracy is calculated as the ratio between the test accuracy and the random accuracy. The later is the sum of the squared fraction of positive and negative cases in the dataset. The overall gain in accuracy varies between 0 (complete opposite result), 1 (complete randomness) and 2 (perfect accuracy).

Receiver operating characteristic (ROC) curves were also used to characterize the diagnostic test (using the area under curve parameter AUC) and also to detect the optimum  $CO_2$  threshold concentration that offers the best sensitivity and specificity. The ROC curve plots the true positive rate *versus* the false positive rate (1-specificity) at various thresholds. The AUC criterion represents the probability that an actual exceedance case is classified as an exceedance case within all the range of possible thresholds (i.e. the complete range of  $CO_2$  concentration). An AUC value of 0.5 means complete randomness whereas a value of 1 is ideal. The model is assumed to be good when the AUC value is above 0.7.

#### **RESULTS AND DISCUSSION**

#### Correlations between CO<sub>2</sub> and indoor pollutant concentrations

Carbon dioxide average concentration was found to be correlated with average concentration of formaldehyde, acetaldehyde, acrolein, benzene,  $PM_{2.5}$  and  $PM_{10}$  in both campaign studies. In figure 1, the correlation between  $CO_2$  and formaldehyde average concentrations is quite similar in dwellings and in schools (respective Pearson correlation coefficient of 0.229 and 0.276, p < 0.0001). However, in both cases, the correlation is weak (5% and 8% of explained variance respectively). Significant correlations between formaldehyde and  $CO_2$  were also observed during both the heating and the non-heating season, and in smoking and non-smoking dwellings. The correlation was found to be equally significant (p < 0.0001) in nurseries and schools.

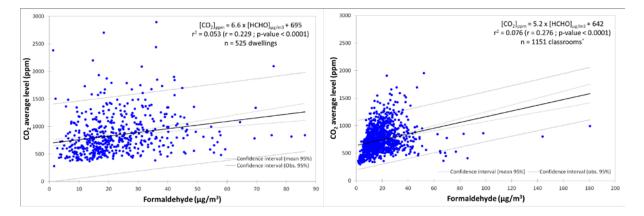


Figure 1. A ssociation between  $CO_2$  and formaldehyde 1-week average concentrations in dwellings (left) and in classrooms (right). The classrooms include playrooms from nurseries.

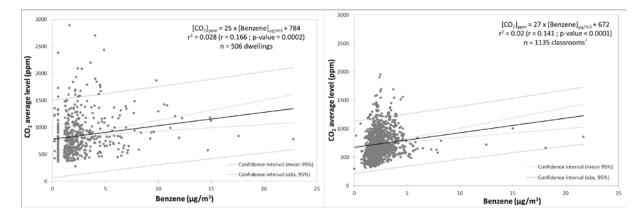


Figure 2. Association between  $CO_2$  and benzene 1-week average concentrations in dwellings (left) and in classrooms (right). The classrooms include playrooms from nurseries.

For benzene (figure 2), the correlation with  $CO_2$  concentration was also found to be very weak (3% and 2% explained variances) but statistically significant. Interestingly, as with formaldehyde, the slope was similar between dwellings and schools. When the sample of dwellings is split according to the heating season, smoking habit or the presence of active combustion appliance, the correlation between benzene and  $CO_2$  concentrations is not significant anymore, except in dwellings where occupants smoke or use combustion appliances. In nurseries, kindergartens and schools, the observed correlation completely disappears when the classrooms are split in various categories (summer/winter, type of school). However, the correlation between benzene and  $CO_2$  remained significant in elementary schools (p < 0.0001). In winter, the indoor concentration of benzene was negatively correlated with  $CO_2$  concentration (p = 0.03). When taking into account the difference between indoor and outdoor benzene concentrations, significant negative correlations with  $CO_2$  concentration are observed in nurseries (p = 0.009), and in kindergartens and schools (p < 0.0001).

Among the pollutants and the indoor environment studied, acetaldehyde in dwellings is the most correlated with  $CO_2$  concentration with 20% of the variance explained, despite this correlation still being weak (figure 3). The correlation remains significant (p < 0.0001) in all sub-categories of dwellings (heating/non-heating, use or no us e of combustion appliances, smoking or non-smoking habit). The level of acrolein is in the same way correlated with  $CO_2$  level with an explained variance of 9%.

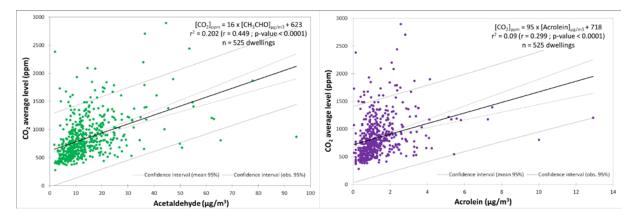


Figure 3. Association between 1-week average concentrations of  $CO_2$  and acetaldehyde (left) and acrolein (right) in dwellings.

The average CO<sub>2</sub> level during occupancy is also correlated with  $PM_{10}$  (r = 0.209, p = 0.0004) and  $PM_{2.5}$  (r = 0.123, p= 0.044) concentrations in dwellings. Within the different groups of dwellings, the correlation remains significant in the heating season (but not in the non-heating season), when combustion appliances are used (but the correlation is lost when they are not used) and in non-smoking dwellings (compared to smoking dwellings). As particulate matter was collected in the living room and the other pollutants in the main bedroom, spatial heterogeneity may have affected the correlation.

#### Detection of health guideline value exceedances using CO<sub>2</sub> concentration

The prevalence of exceedance of health guideline values varies from 50% (benzene >2  $\mu$ g/m<sup>3</sup>) to 92% (PM<sub>2.5</sub> >10  $\mu$ g/m<sup>3</sup>) in dwellings. For formaldehyde (figure 4), the prevalence of exceedance is almost constant (more than 80%) whatever the CO<sub>2</sub> concentration in dwellings, while it increases from 44% to 97% when the CO<sub>2</sub> concentration increases in schools, kindergartens and nurseries.

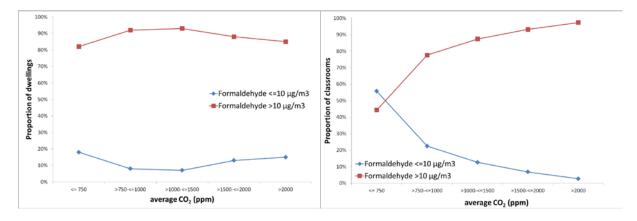


Figure 4. Proportion of dwellings (left) and classrooms (right) that exceeds the formaldehyde guideline value according to  $CO_2$  concentration averaged on the occupancy period.

For benzene (figure 5), the trend is almost identical in dwellings and in schools, with a slight increase from 40% to 70% of exceedances when the  $CO_2$  concentration increases as well. The prevalence of exceedance for the other pollutants is presented in table 2.

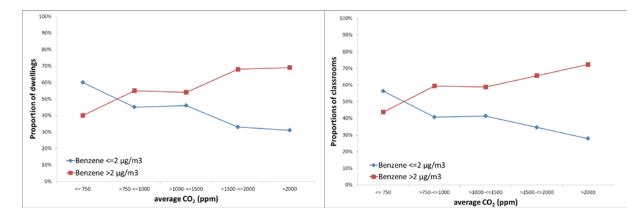


Figure 5. P roportion of dwellings (left) and classrooms (right) that exceeds the benzene guideline value according to CO<sub>2</sub> concentration averaged on the weekly occupancy period.

In order to quantify the real gain in using the CO<sub>2</sub> concentration as a diagnostic test to better detect exceedances of a health-based guideline value, the parameters described in table 1 were calculated and presented in table 2 for a given threshold of 750 pp m. Increasing the CO<sub>2</sub> threshold systematically reduces the sensibility (and NPV) and increases the specificity (and PPV). The optimum CO<sub>2</sub> threshold that maximizes the sum of sensibility and specificity varies according to the pollutant between 661 and 892 ppm. As such, the threshold of 750 ppm represents fairly well the performance of the test to detect exceedances.

and Gain -) on exceedance of health-guideline values of indoor pollutants according to a threshold $CO_2$ concentration of 750 ppm.												
-				11					Accur.		Opt. CO <sub>2</sub>	
Dwellings	TPR	TNR	PPV	NPV	Prev	1-Prev	Gain +	Gain -	Gain	AUC	threshold	
Formaldehyde > 10	0.61	0.60	0.92	0.18	0.88	0.12	0.04	0.05	0.78	0.62	701	
Acetaldehyde > 9	0.73	0.64	0.76	0.60	0.61	0.39	0.15	0.21	1.33	0.73	801	
Acrolein > 0.8	0.70	0.64	0.80	0.52	0.67	0.33	0.13	0.19	1.23	0.72	701	
Benzene > 2	0.67	0.49	0.57	0.60	0.50	0.50	0.07	0.10	1.16	0.61	771	
$PM_{10} > 20$	0.60	0.67	0.90	0.26	0.83	0.17	0.07	0.08	0.85	0.69	678	
$PM_{2.5} > 10$	0.58	0.67	0.95	0.12	0.92	0.08	0.03	0.04	0.68	0.67	661	
Nurseries/ Schools	TPR	TNR	PPV	NPV	Prev	1-Prev	Gain +	Gain -	Gain	AUC	Opt. CO <sub>2</sub>	
Formaldehyde > 10	0.88	0.51	0.86	0.56	0.78	0.22	0.09	0.33	1.23	0.80	892	
Benzene > 2	0.84	0.27	0.61	0.56	0.57	0.43	0.04	0.14	1.17	0.58	680	

Table 2. Sensitivity (TPR), specificity (TNR), positive and negative diagnostic gain (Gain +

Overall, PPV are high and as such the occurrence of exceedances is increased when CO<sub>2</sub> concentration is above the 750 ppm-threshold. However, the initial prevalence is already high. As a consequence, the real positive gain of the test is low (max +15% for acetaldehyde). NPV is generally lower than PPV, but the negative gain is higher than the positive gain (max +33% for formaldehyde in schools). This means that when the  $CO_2$  concentration is below the 750 ppm-threshold, the probability that the formaldehyde concentration remains below  $10 \,\mu g/m^3$ is increased by 33%. The highest overall gain in accuracy of using the  $CO_2$  concentration as a diagnostic test with a 750 ppm -threshold is obtained for acetaldehyde (+33% over randomness), acrolein (+23%) and benzene (+16%) in dwellings and for formaldehyde (+23%) in schools.

When considering the AUC parameter, the test is discriminative for acetaldehyde and acrolein in dwellings (AUC >0.7) and for formaldehyde in schools (AUC = 0.8). However, misclassification may always occur, e.g. when  $CO_2$  concentration is under 750 pp m, the guideline value of formaldehyde is exceeded 44% of the time.

Therefore, the use of  $CO_2$  concentration to better predict exceedance of health guideline values is an interesting tool due to its ease of measurement, but is not sufficient as the chance of misclassification may be high.

#### CONCLUSIONS

Carbon dioxide is most of the time significantly and positively correlated with indoor air pollutants in two large measurement campaigns. However, the strength of the correlation is weak. The probability that the concentration of a pollutant exceeds health guideline value increases and decreases with  $CO_2$  concentration. Nevertheless, this probability still remains high at low  $CO_2$  concentrations, meaning a high chance of misclassification. Using the  $CO_2$  level as a diagnostic test to detect exceedance of guideline values allowed us to quantify the real gain in detection probability (at best +33% for acetaldehyde compared to a guideline value of 9  $\mu$ g/m<sup>3</sup>). The results of the detection test vary according to the pollutant, but also according to the indoor environment. As  $CO_2$  is often considered as a surrogate of ventilation rate, this study confirms that even with good ventilation conditions (i.e. low  $CO_2$  level), the reduction of pollutant sources remains necessary to achieve a satisfactory indoor air quality.

The results from this study were used in a collective expertise that looked at the opportunity to recommend a health-based guideline value for  $CO_2$  in indoor air (ANSES, 2013a). It was concluded that the concentration of  $CO_2$  cannot be considered as a unique indoor air quality indicator. This expertise also concluded from an extensive literature review of the epidemiological studies that the current knowledge is too limited to define a  $CO_2$  guideline value that protects against health effects in indoor environments. In particular, the definition of a  $CO_2$  protective threshold is prevented by the lack of analysis on the shape of the  $CO_2$  concentration-risk relationships and by the lack of French large-scale epidemiological studies. Alongside this, further research is required on the adverse effects intrinsic to  $CO_2$  on human psychomotor performance (decision-making, problem-solving) and on sensitive populations such as children, pregnant women, and people with lung or heart diseases.

For mitigation purposes, the measurement of  $CO_2$  is still recommended as an indicator of air stuffiness. In schools, where higher  $CO_2$  levels are observed compared to dwellings and offices, the awareness of technical services, school directors and teachers should be improved on the measures to effectively reduce the effects of closed spaces on health, performance and perceived comfort, including ventilation through opening windows and doors. The ventilation strategy must also take into account other risk factors such as noise pollution, transfer of outdoor air pollution, thermal comfort, etc.

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