

# Supporting Information

## Spatial distribution of atmospheric PCBs in Zurich, Switzerland: Do joint sealants still matter?

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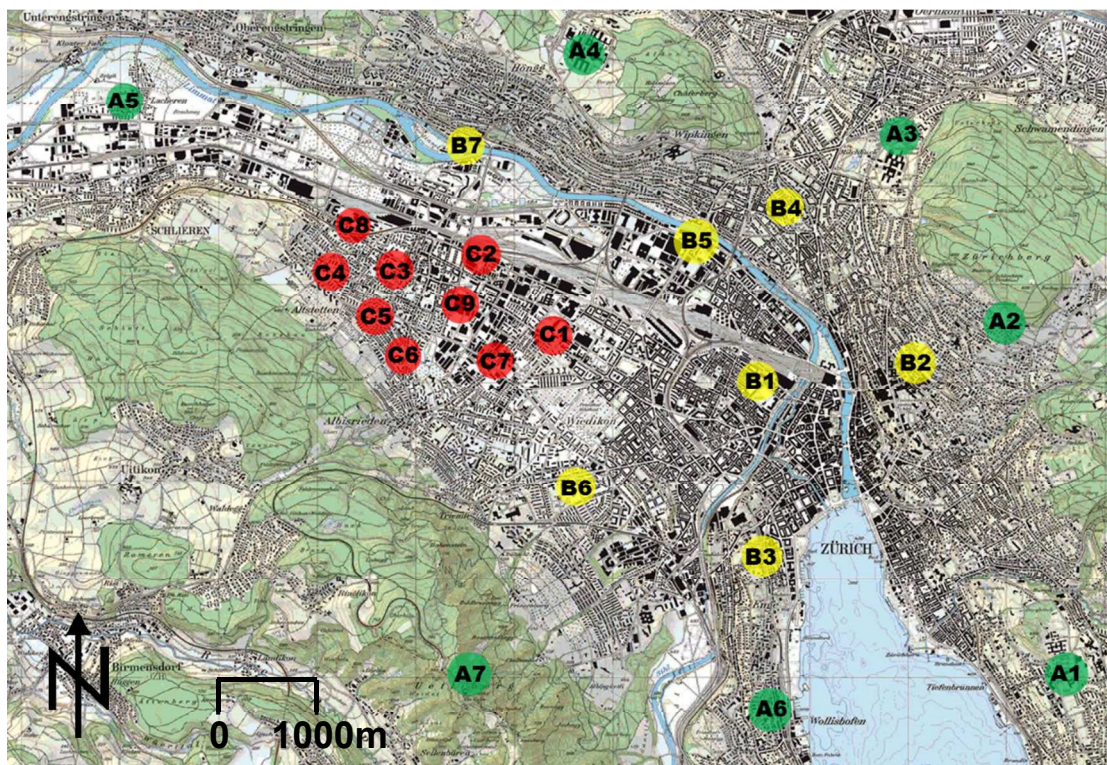
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**The Supporting Information contains 14 pages, and includes 7 figures and 4  
tables.**

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## 1. Sampling Sites



**Figure S1.** Map of the study area showing the sampling locations. Sites with initial A are located in the outskirts of the city; sites with initial B are randomly distributed within the urban area, whereas sites with initial C are located in a specific neighborhood (© 2014 swisstopo (JD100043)).

**Table S1.** *Description of the sampling sites*

<b>Name</b>	<b>Site characteristic</b>	<b>PAS installation</b>	<b>Deployment time 2011 (days)</b>	<b>Deployment time 2013 (days)</b>
A1	urban-residential	1.5 m above ground	32.9	43.2
A2	urban-residential	fixed to a rainwater gutter	33.0	42.0
A3	urban-residential	1.5 m above roof	33.0	43.2
A4	semi-rural	1.5 m above roof	33.0	42.9
A5	urban-commercial	1.5 m above roof	33.0	42.1
A6	urban-industrial	1.5 m above roof	36.7	43.0
A7	rural	1.5 m above roof	28.9	42.1
B1	urban-residential	1.5 m above ground	33.0	43.1
B2	urban-commercial	fixed to a railing	36.6	42.2
B3	urban-commercial	fixed to a railing	42.0	49.0
B4	urban-residential	1.5 m above roof	33.0	43.0
B5	urban-commercial	1.5 m above roof	35.0	42.0
B6	urban-residential	3 m above ground	32.9	42.1
B7	recreational area	1.5 m above ground	33.9	42.0
C1	urban-commercial	1.5 m above roof	35.1	40.0
C2	urban-commercial	1.5 m above roof	34.0	41.9
C3	urban-residential	1.5 m above roof	34.8	41.8
C4	urban-residential	1.5 m above roof	34.9	49.2
C5	urban-residential	1.5 m above roof	33.8	42.1
C6	urban-residential	1.5 m above roof	34.8	42.0
C7	urban-residential	fixed to a railing	34.9	42.0
C8	urban-industrial	fixed to a railing	34.9	41.1
C9	urban-industrial	1.5 m above roof	33.0	41.0

## 2. Quality Assurance/ Quality Control

**Quality control.** To account for loss of target analytes during sample preparation, an isotope labeled analogue was added as internal standard prior to the extraction. Considering the response of native and isotope labeled PCB congeners, results are corrected for recovery. Average absolute recovery rates are presented in Table S2 and are generally higher than 75%. Duplicate analyses of sample extracts provided satisfactory differences of not more than 25%.

**Table S2.** Monitored ions, recoveries, and method detection limit for iPCBs

	PCB-28	PCB-52	PCB-101	PCB-138	PCB-153	PCB-180
Molecular ions	255.9608	289.9218	325.8799	359.8409	359.8409	393.8019
monitored (m/z)	257.9578	291.9189	327.8769	361.8380	361.8380	395.8019
Absolute recoveries of isotope labeled standards (mean $\pm$ SD)	77 $\pm$ 30%	75 $\pm$ 22%	83 $\pm$ 27%	85 $\pm$ 24%	89 $\pm$ 27%	88 $\pm$ 27%
Absolute differences of duplicate analyses <sup>a</sup> (mean $\pm$ SD)	13 $\pm$ 8%	25 $\pm$ 25%	8 $\pm$ 5%	5 $\pm$ 4%	6 $\pm$ 6%	8 $\pm$ 8%
Method detection limit <sup>b</sup> [pg·m <sup>-3</sup> ]	0.05	0.04	0.04	0.04	0.04	0.04

<sup>a</sup> 5 duplicate analyses were performed in total

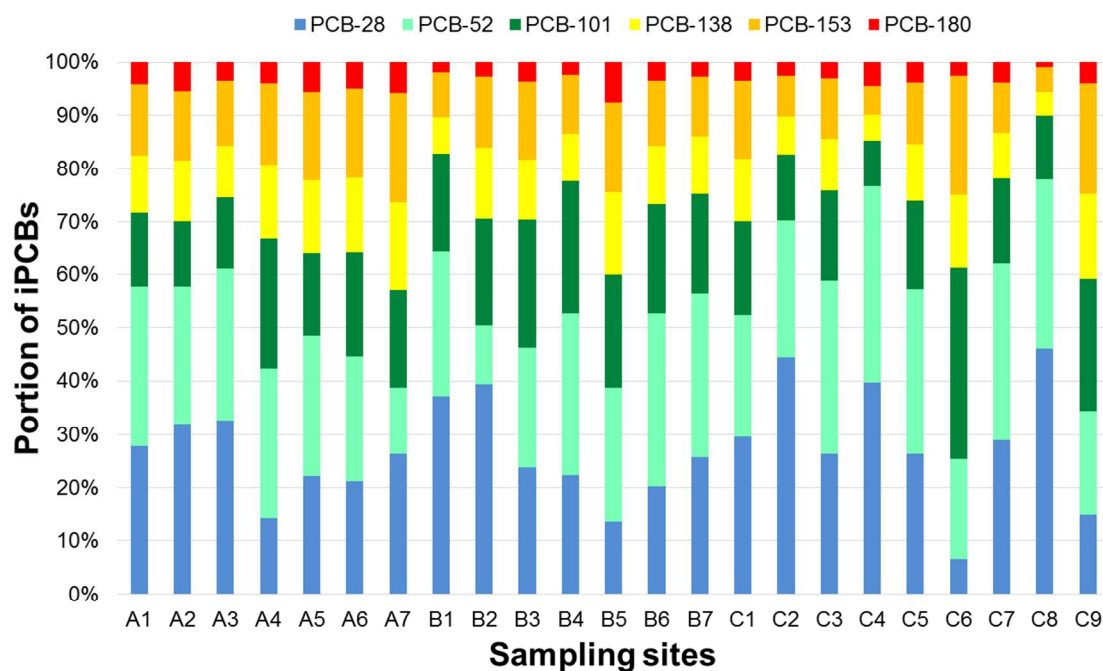
<sup>b</sup> Based on a sampling volume of 120 m<sup>3</sup>, LOD from Bogdal et al. (2014)<sup>1</sup> who used the identical GC/EI-HRMS device  
SD = standard deviation

Instrumental limits of detection (LOD) were derived by Bogdal et al. (2014),<sup>1</sup> who used the same instrumental set-up and are based on a signal-to-noise ratio >3 in the reconstructed ion chromatogram.

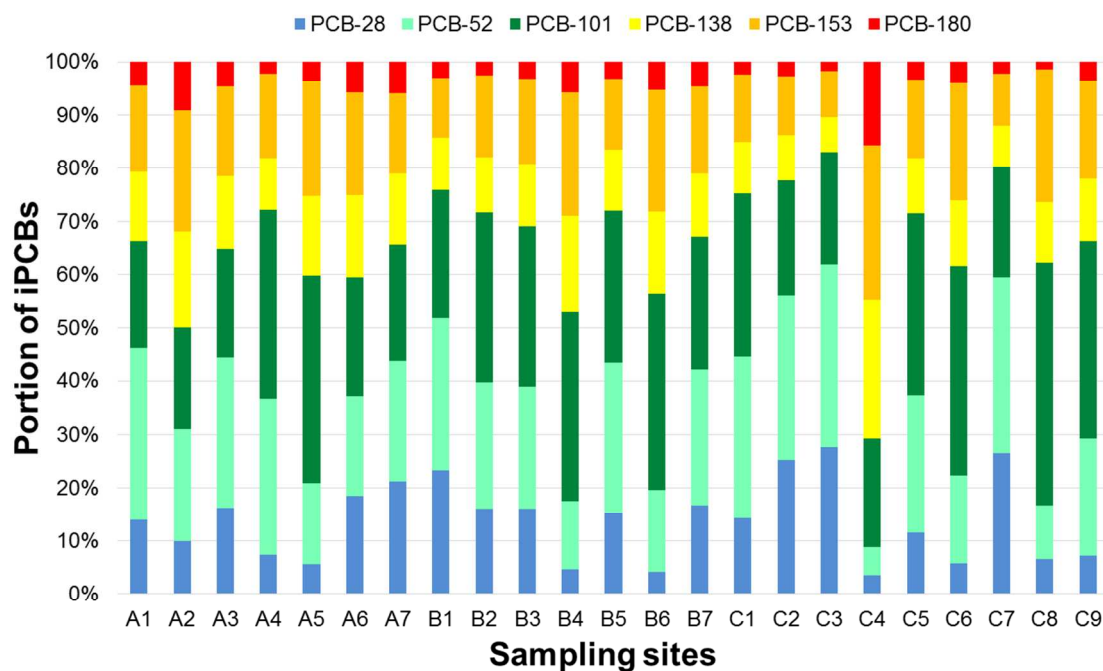
**Glassware preparation.** All glassware used in the analysis was cleaned in a glassware washer and immersed in a detergent bath for 12 hours. Further, the glassware was heated to 450 °C overnight. Prior to use, glassware was rinsed with acetone and dichloromethane.

**Chemicals and reference materials.** All solvents were of high purity grade for trace analysis. Reference native PCBs were purchased from AccuStandard Inc., New Haven, CT, USA and  $^{13}\text{C}_{12}$ -labeled PCBs were from Cambridge Isotope Laboratories, Andover, MA, USA.

### 3. PCB Congener Pattern



**Figure S2.** PCB congener pattern observed at the 23 sampling site in the city of Zurich in spring 2011

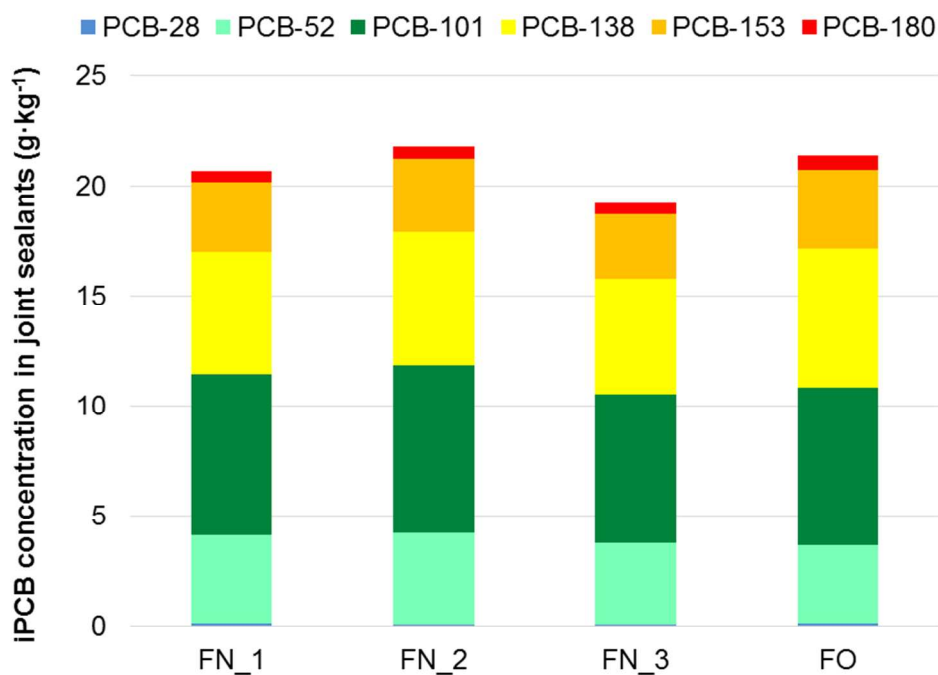


**Figure S3.** PCB congener pattern observed at the 23 sampling site in the city of Zurich in spring 2013

## 4. PCB Concentrations in Sealants

**Sampling.** Samples of joint sealants (approximately 5 g) were collected in June 2014 using a hollow-core drill. The first sample (FN) was collected at the northern outside wall of the investigated housing complex in Zurich, the second sample (FO) was collected at the eastern outside wall of this building.

**Analysis.** To investigate if a concentration gradient occurs within the joint sealants, the sample FN was subdivided into three subsamples that represent layers with different depths. Sample FO was not modified. Subsequently, approximately 100 mg of each sample was Soxhlet extracted with 100 mL dichloromethane/*n*-hexane (DCM/Hex) (1/1) for 12 hours. These extracts were diluted and spiked with  $^{13}\text{C}_{12}$ -labeled iPCB analogues as internal standards. iPCB concentrations were determined by gas chromatography coupled to electron ionization high resolution mass spectrometry (GC/EI-HRMS) on a Thermo Finnigan MAT 95 high resolution mass spectrometer.

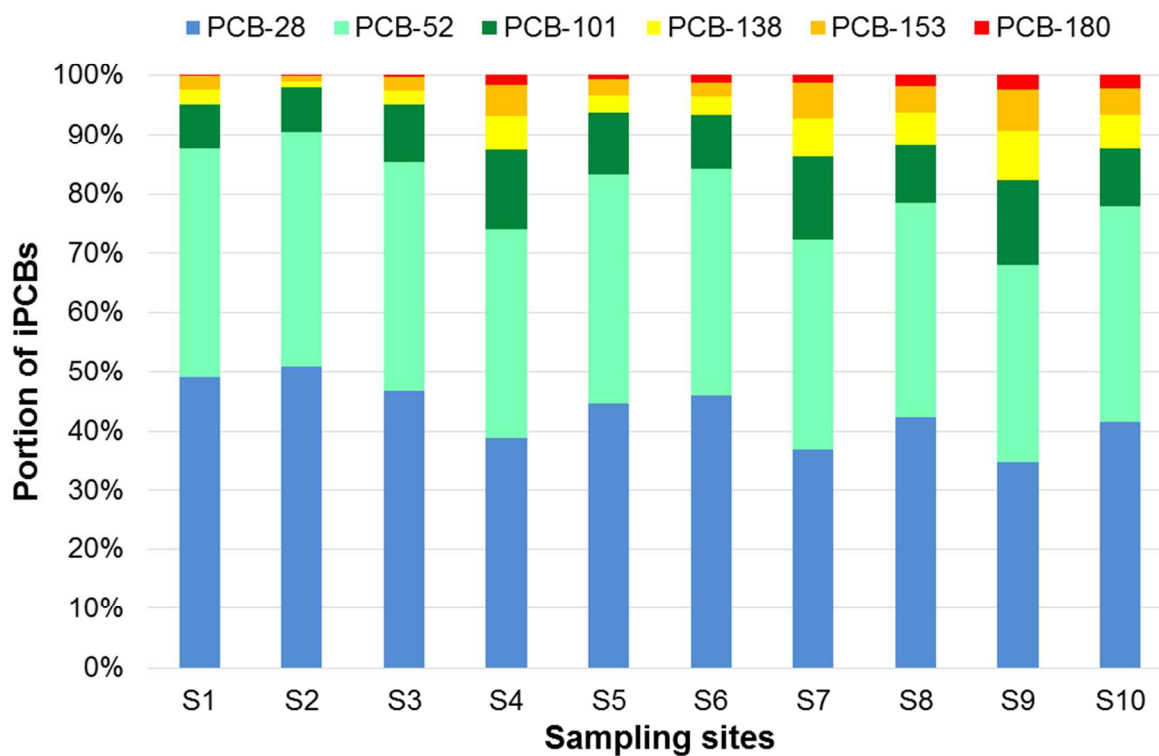


**Figure S4.** iPCB concentrations in joint sealants attached to the investigated housing complex. FN\_1 (0–4 mm), FN\_2 (4–7 mm), and FN\_3 (7–11 mm) represent the depth profile of one sample, FO shows the concentration in joint sealants at another outside wall of this housing complex.

**Results.** iPCB concentrations in the samples ranged from 19.2 to 21.8 g iPCB per kg joint sealant with an average of 20.8 g per kg. The comparison of the sample FN\_1, FN\_2 and FN\_3 shows that there exists no concentration gradient within the sealants. Further, the PCB pattern in all samples was very similar suggesting that the same commercial product was applied on the entire housing complex.

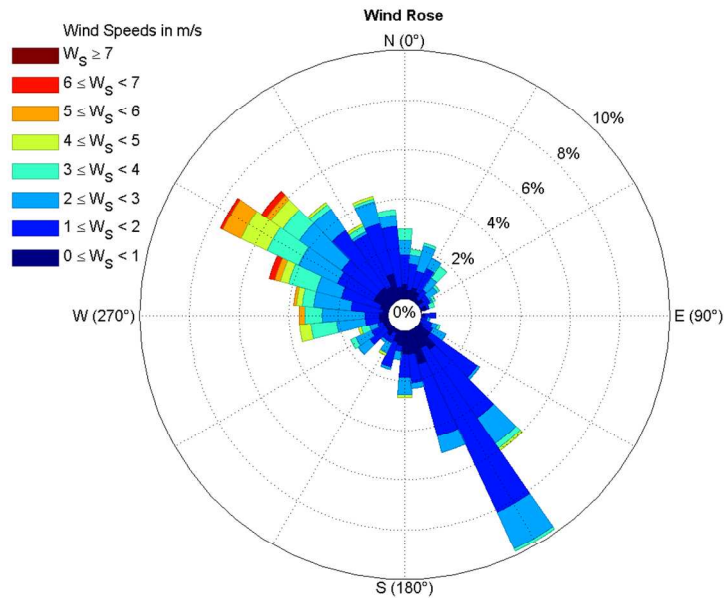


## 5. PCB Congeners in Atmospheric Samples

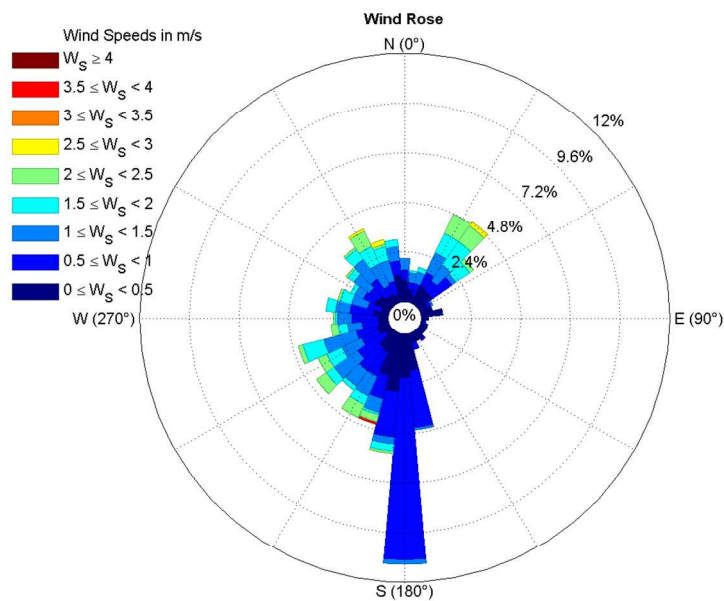


**Figure S5.** PCB congener pattern observed at sampling sites in the surrounding of the investigated housing complex.

## 6. Wind Roses



**Figure S6.** Wind rose that illustrates the distribution of wind speeds and wind directions in spring 2015 determined at the meteorological station Kaserne (M1). Data provided by the Swiss Meteorological Service (MeteoSwiss).



**Figure S7.** Wind rose that illustrates the distribution of wind speeds and wind directions in spring 2015 determined at the meteorological station Schlieren (M1). Data provided by Office of Waste, Water, Energy and Air of the canton of Zurich (WWEA)

## 7. Gaussian Plume Modeling

For continuous point sources, the spatial distribution of pollutants can be described by: <sup>3</sup>

$$C_{air}(x, y, z) = \frac{Q}{2\pi u \sigma_y \sigma_z} \exp\left(-\frac{y^2}{2\sigma_y^2}\right) \times \left[ \exp\left(-\frac{(z-h)^2}{2\sigma_z^2}\right) + \exp\left(-\frac{(z+h)^2}{2\sigma_z^2}\right) \right] \quad (1)$$

Where  $C_{air}(x,y,z)$  is the steady-state air concentration of a pollutant at the point (x,y,z) in  $g \cdot m^{-3}$ , x,y, and z are the coordinates for the downwind, crosswind, and vertical direction (m); Q is the source strength ( $g \cdot s^{-1}$ ); u is the average wind speed ( $m \cdot s^{-1}$ ), z and h are the heights of the source and the samplers at the sites S1-S10 (m); and  $\sigma_y$  and  $\sigma_z$  are the lateral and vertical diffusion coefficients (-), respectively.

The diffusion coefficients are a function of the downwind distance and depend on the atmospheric stability. For this urban location, the coefficients were calculated using urban Briggs sigmas that are also called McElroy-Pooler sigmas (Table S3).<sup>4</sup>

**Table S3.** Urban dispersion parameters. x represents the distance in downwind direction.

Pasquill stability	$\sigma_y$ (m)	$\sigma_z$ (m)
A - B	$0.32 x \cdot (1 + 0.0004 x)^{-0.5}$	$0.24 x \cdot (1 + 0.001 x)^{0.5}$
C	$0.22 x \cdot (1 + 0.0004 x)^{-0.5}$	$0.20 x$
D	$0.16 x \cdot (1 + 0.0004 x)^{-0.5}$	$0.14 x \cdot (1 + 0.0003 x)^{-0.5}$
E – F	$0.11 x \cdot (1 + 0.0004 x)^{-0.5}$	$0.08 x \cdot (1 + 0.00015 x)^{-0.5}$

Usually, Gaussian diffusion models are used with ideal point sources. In this study, the emissions originate from a housing complex consisting of separate buildings. We therefore modeled these buildings as volume sources with initial lateral and vertical distances according to the industrial source complex (ISC3) model that has been the US-EPA recommended air dispersion model for many years.<sup>5</sup> Usually, initial lateral dimensions of an individual building can be achieved by

dividing the length of its side by 4.3 and the initial vertical dimension can be calculated by dividing its height by 2.15.

As this housing complex consists of several buildings, we integrated the buildings as individual volume sources into our modeling approach. The contribution of each individual building to the source strength of the total housing complex was calculated by dividing its cubic volume by the total volume of this housing complex.

In the next step, the downwind and crosswind distances of the sampling sites were calculated. As these distances depend on the wind direction, they were determined for every hour of the sampling period as follows:

$$x = -(X(R) - X(S)) \sin(WD) - (Y(R) - Y(S)) \cos(WD) \quad (2)$$

$$y = -(X(R) - X(S)) \cos(WD) + (Y(R) - Y(S)) \sin(WD) \quad (3)$$

where  $x$  is the downwind distance;  $y$  is the crosswind distance;  $X$  and  $Y$  are the coordinates of the source (S) and receptor (R) and  $WD$  is the wind direction.

The hourly-resolved data on the atmospheric stability was provided by the Swiss Meteorological Service (MeteoSwiss), whereas the wind speed and wind directions were derived from two meteorological station that are located within the city of Zurich.

As the measurement of the wind speed at these meteorological stations were performed at heights of 15 m and 10 m, the wind speed had to be adjusted to the release height:

$$u_s = u_m \times \left( \frac{h_s}{h_m} \right)^p \quad (4)$$

where  $u_s$  is the wind speed at the release height (m);  $u_m$  is the reference measurement height wind speed ( $\text{m} \cdot \text{s}^{-1}$ );  $h_s$  is the release height (m);  $h_m$  is the measurement height (m); and  $p$  (-) is the wind profile exponent that depends on the atmospheric stability class. Default values are as follows:<sup>5</sup>

<i><b>Table S4.</b> Urban exponents (<i>p</i>) for vertical wind speed gradients.</i>	
<b>Pasquill stability class</b>	<b>p</b>
A	0.15
B	0.15
C	0.20
D	0.25
E	0.30
F	0.30

The release height wind speed was not allowed to be less than 1 m·s<sup>-1</sup> in this Gaussian plume model.

## References

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