# **Usage of passive air samplers in exposure assessment – a look at pesticides**

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**Layman’s summary**

People all over the world are coming into contact with an increasing number of chemicals over the years. One of those categories of chemicals are the pesticides. Exposure to pesticides is known to be associated with increased risk of certain diseases. Apart from pesticides on food, it is also important to look at other ways of how humans come into contact with pesticides. Residents of rural areas have a higher risk when dealing with pesticides via air because they live closer to the agricultural fields, which get sprayed with pesticides. The amount of pesticides in air can be determined with two main ways an active air sampler or a passive air sampler. The active air sampler is a small station using a pump to measure the contents in the air. The passive air sampler is a sponge-like device where the air leaves parts of its chemical contents in the matrix when wind passes through the device. Because of the cheap and easy way of using, the passive sampler is useful. A disadvantage however is that the accuracy of the method has to improve. The amount of air that passes through the passive air sampler has to be estimated. This study hopes to do that improvement by looking at new calculations to do these determinations. The information is then used to establish the amount of chemicals in the air. The method that showed most promise during the study is a model which uses data on the environment and deployment duration to estimate pesticide amounts in the air. In the future the model can be improved by adding new kinds of information on the environmental conditions which affect the amount of chemicals the air sampler absorbs. That will then improve the ability to check air quality in a cheap and easy way.

***Abstract***

The increasing amount of chemicals used around the globe has also increased the risk of disease through exposure to these compounds. A subcategory are pesticides, which are the subject of this study. The focus on monitoring pesticide exposure through food and/or occupational risk leaves exposure through inhalation understudied. Therefore a risk assessment is performed for residents of rural areas in the Netherlands, living near agricultural fields treated with pesticides. To monitor air concentrations of pesticides two main methods are available: Active and Passive air samplers (AAS & PAS). AAS allow for precise measurements but have disadvantages in cost, ease of deployment and noise. PAS do not suffer from these disadvantages however the measurements are still imprecise. The cause is that the precise sampling rate (R) of the PAS has to be estimated. This study utilizes three methods to extrapolate PAS measurements towards ng per cubic meter of air. I) A set value of R = 4, II) windspeed-derived individual sampling rate per PAS, and III) a linear model using environmental conditions. The results of predicted concentrations of each method were compared and varied in precision. For pendimethalin the methods resulted in a r-squared of; I) 0.18, II) 0.18, III) 0.40. The chlorpropham results were; I) 0.31, II) 0.31, III) 0.71. Taking environmental conditions like temperature and humidity into account does show promise. This can enable future research to build further on these kind of models and allow for more accurate estimations using PAS.

**Key Words**

Pesticides, Passive Air Sampler, Extrapolation, Exposure, Risk Assessment.

# **Introduction**

At this moment there are almost 500 pesticides approved for use in the European Union. (EC, 2020). These chemicals allow the agricultural sector to greatly increase its output. This is one of the essential developments that helps to produce sufficient food for the world's growing population. However, there are known negative effects associated with the introduction of pesticides into our environment. These include i) ecosystem deterioration and ii) increased risks for certain diseases for both animals (Gili et al, 2018) and humans (Dahiri et al, 2021). For humans, exposure to pesticides has been linked to several health problems, like certain types of cancer and asthma (Kim et al, 2017).

There are 3 main routes of exposure to pesticides: ingestion, inhalation and dermal contact. The most studied exposure route is ingestion (oral intake) via food and drink consumption. This is relevant for all consumers of products where at some point in the production pesticides were used.

Exposure through inhalation and dermal contact is studied less in comparison. The studies that do focus on these exposure routes are mostly focused on agricultural occupational exposure (Dhananjayan & Ravichandran, 2018). For agricultural workers the inhalation route accounts for about 10% of their exposure with dermal contact and ingestion forming the remaining 90% (Ye et al, 2013).

The risk for rural communities living close to the fields where pesticides are applied remain understudied (Dereumeaux, 2020). Some studies have been performed recently but results are inconclusive (Bukalasa, 2018, Shaw, 2018). Air is an important matrix for the spreading of pesticides away from target sites. This can be via spray drift (Figueiredo, 2021), wind erosion from ground particles (Bento *et al*, 2017) and volatilization from sources where the pesticides ended up after spraying (Bedos *et al*, 2002). Although the general population is exposed to a lesser dose of pesticides compared to occupational exposure, residential exposure is still a great matter of concern. Therefore, it is important to better understand exposure of populations living close to treated fields.

Each exposure route can be measured in a different way. Dermal contact can be measured by using silicone wristbands (Hamzai et al, 2022). For ingestion, food items can be analysed to determine their pesticide content (Curl *et al*, 2015). Finally, pesticides present in the air can be analysed with either active (AAS) or passive air samplers (PAS) (Harner *et al*, 2004). The air matrix is the focus of this study.

The AAS is a more accurate compared to the passive PAS. However using an AAS sampler also has some disadvantages. It requires a source of electricity and trained personnel to use, furthermore the installation is also larger and more noisy. Limitations which can hamper their use in certain study areas. The PAS is a smaller and cheaper alternative which does not suffer from the aforementioned limitations. The polyurethane passive air sampler consists of two metal disks with a matrix of polyurethane foam in between. In the centre of the Polyurethane foam passive air sampler (PUF-PAS) a part of the matrix is exposed to the air. Chemicals can partition from the air into the matrix at a certain rate. After a certain deployment time the PUF-PAS can be retrieved and the contents of the matrix can be analysed in the lab (Huang et al, 2018). Hence, the PUF-PAS provides a cheap method to measure the air concentrations of pollutants.

The biggest limitation regarding the use of the PUF-PAS is less precision due to the sampling rate being uncertain. It is not known how much air passes through the disks of the PUF (diffusion rate) and the partitioning coefficient between the air and the matrix of the PUF-Pas is also unavailable for a large amount of chemicals. The standard value of R=4m3 a day has been used to do calculations before and appears to work for rough calculations, however, for more precise calibrations there is a need for situation specific calculations. Taking meteorological conditions and spatial features into account should be able to deliver a more accurate sampling rate and thus a more accurate air concentration of the studied compound. Improving the calibration of PUF-PAS will be the focus of this study.

The aims of this study are i) to quantify pendimethalin and chlorpropham concentration in both active and passive air samplers; ii) to assess different approaches to convert the concentration of these pesticides found in the PUF-PAS to air concentrations (concentration per cubic meter of air). We achieve this by measuring the actual air concentrations with the AAS. A comparison can then be made with the concentrations found in the PUF-PAS. This way a standard way of calibration can be found to convert the PUF-PAS data towards real air concentrations which also can be used when no AAS are present.

1. **Method**
   * 1. *Study design*

This study uses data obtained during the OBO study, the relevant parts took place during March until June 2017. This study was designed to detect pesticide concentrations in the air. The study is performed in- and around rural homes near agricultural fields. The minimum distance to the actual field were pesticides were applied had to be at least 100 meters, and maximal distance was 250 m. With varying deployment periods Polyurethane Passive Air Samplers were deployed, alongside Active Air Samplers. PAS were deployed for up till three months, AAS were deployed for up till two days. This way two methods can be used to measure the pesticide concentrations in same position. On 6 of the measurements points HOBO-loggers were deployed to determine temperature and humidity of the exact environment.

* + 1. *PUF-PAS Characteristics*

The passive air samplers used within the OBO study are two types made by Tisch Environmental, the T-200 outdoor and T-300 indoor. Within the samplers there is the polyurethane disk for absorption of the pesticides in the air (14 cm long, 1.35 cm thick, 0.0213 g/cm-3, surface area of 365 cm2). Before deployment the disks were precleaned using Soxhlet extraction with acetone, for 24 hours. After drying under the fume hood (6-7 hours) the disks are stored in aluminium foil (freezer, -20°C). Houses selected for sampling preferably had both an indoor and outdoor sampler deployed. The actual PUF-disk is protected with a stainless steel “flying saucer” design of a 24cm top bowl and a 20cm lower bowl. Below in Figure 1, a schematic overview is shown of the outdoor PUF-PAS.

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**Fig. 1** Schematic of the passive air sampler with a polyurethane disk. (Tuduri, 2006)

* 1. *Lab analysis*

The pesticide contents in the PUF-disk were analysed using liquid chromatography tandem mass spectrometer (LC MS/MS). The contents were targeted for 46 unique pesticides. Techniques used were Solid Phase Extraction (SPE) to deal with the solids in the sample, Gel Permeation Chromatography (GPC) for separation of the polymers, silica columns to clean up the samples and LC MS/MS for quantification. For a detailed description of the analysis in the laboratory see the full report on the OBO study (Gooijer *et al*, 2019).

*2.3.1 Scoping review of existing extrapolation methods*

There are a multitude of calculations and models to extrapolate concentrations found within the PAS matrix towards the actual amount in a cubic meter of air. To assess what possibilities where available a scoping review of existing literature was performed. Emphasis was placed on a combination of accuracy and viability within the scope of this study. Below in subsection 2.3.2 possibilities that were considered and those that were selected for the study are described.

*2.3.2 Extrapolation of data*

The choice was made to proceed using the chlorpropham & pendimethalin data, because of both were detected on almost all PAS and AAS measurements. Only a minor percentage (6%) of measurements were below limit of detection (LOD). The data below LOD was imputed using single imputation to estimate the concentrations found there. Single imputation was performed using survival regression with maximum-likelihood estimator. Both the imputation and the calculations for the other methods of extrapolation were performed within the R software (R for Windows 4.1.2, Rstudio 2021.09.2 Build 382, R core team 2021).

The results measured by the active air samplers were taken as the actual concentrations of the pesticides in the air at that moment. After a scoping review of the available literature, 3 methods, that were deemed most suitable for the study were selected. These methods were used to extrapolate concentrations found within the PUF disk towards an estimated air concentration.

During the exploration of the existing literature on methods to determine sampling rate, polyparameter linear free energy relationships (PP-LFERs) was seen as an option. This empirical method uses equations with the mechanical characteristics of the compound of interest to determine the partition coefficient (Endo & Goss, 2014). However required data was only available for pendimethalin and could not be found for chlorpropham, therefore making it unsuitable to use in this study. Another option is the use of depurination compounds. These are not naturally occurring compounds which are added to the PAS at the start of the deployment period. The rate of loss of these compounds out of the PUF-matrix is determined by the air layer adjacent to the matrix. After the deployment time the rate of loss is analysed in the lab, this info gives insight into the amount of air passing through the air sampler. Using the air volume, the sampling rate can be established. The depurination compounds where not present in the PUF-PAS of the OBO study and the method could therefore not be utilized. Instead of combing PAS & AAS Herkert and colleagues combined a modelling approach together with depurination compounds to verify the results of the model. The modelled approach was performed using multiple regression models including known data on Koa of the category in question (PCB variants) (Herkert *et al*, 2018). The modelled approach is relevant and also takes environmental conditions into account but was focused on PCB variants. Also the model by Herkert and colleagues requires detailed meteorological data inputs which was no available for the exact location. These are alternative options for extrapolating PUF-PAS data towards ng/m3 in the air but were found unsuitable for the scope of this study.

*2.3.3 Set value for sampling rate*

The first method is commonly used in other studies, it involves using a set value for sampling rate, at R = 4 m3/day. Air concentrations could then be estimated by using the formula; Cair = CPUF / Vair, with air volume determined by; Vair = R \* t. Concentrations are measured in ng/puf and ng/m3, air volume in m3 , time in days and sampling rate in m3 per day

*2.3.4 Windspeed-derived individual sampling rate*

The second methods took individual windspeed into account for each measuring point, using this data to calculate the individual sampling rate. For the windspeed derived sampling rate, windspeed measurements by KNMI station 235 De Kooy were used. Because the weather stations measure at a height of 10 meters, the data has to be adjusted. Using the Wind profile power law, calculations can be adjusted towards the deployment height of both PAS and AAS. The formula is: u = ur (z/zr)α , windspeed (u), reference(r), height (z), neutral stability coefficient (α). Using the adjusted windspeed together with the surface area of the polyurethane disk a the sampling rate can be calculated. Rs (m3d-1dm-2)= 0.922 exp (1.489 \* air velocity (m sec-1) (Tromp *et al*, 2019). The outcome was adjusted by multiplying with the surface area of the PUF-PAS (365 cm2). The new sampling rate per PUF-PAS is fed back into the first formula described in this section to predict air concentrations.

*2.3.5 Linear model with environmental conditions*

The third extrapolation method is based on a linear model. The outcome here are the concentrations measured by the active air sampler (g/m3). As predictors we included the passive air samplers and environmental factors. All models were adjusted for deployment time of the PAS and sampling rate adjusted for windspeed (from method II, section 2.3.4). Environmental factors were added to the model because of the effect on sampling rate (Klanova *et al*, 2008). The code for RStudio can be found in the supplementary files. To compare the predicted concentrations of the three methods the R-squared and Root Mean Square Error (RMSE) are determined.

* 1. *Sensitivity analysis for temperature*

A common feature of previous studies (Klanova *et al*, 2008) is using far-off weather stations for data on environmental circumstances. As the measurements of the far-off stations may differ from the on-site conditions an analysis was performed. In this case the study investigates differences in data on temperature. An comparison was made between data provided by HOBO-loggers deployed alongside the air samplers and data originating from the weather station. KNMI weather station 235 De Kooy is the least distance away from the study area. KNMI weather data is open access, data from station 235 was used for the entire duration of the study(KNMI, 2017). Within literature examples were found that outside placed weather station data is sometimes used for environmental conditions within buildings.

1. **Results**
   1. *Pesticide concentrations*

The measured values from the air samplers (n=46) used within the OBO study can be found within Table 1 and 2. Out of the 46, 6 were controls and 8 were used as test samples, leaving 33 samples for detecting air concentrations. In short, values like the average pesticide concentrations are within the same order or magnitude, this is also visible for the range of the concentrations. These pesticides are often applied together which is why that range was expected. It would be predicted that this would also be true for the ranges of the AAS results. Except for the max value the values are indeed as expected, in the same order of magnitude. There is however a major difference in the maximum value found within the AAS values, because it is only a single value within the dataset, it may be an outlier.

**Table 1**: Descriptive of concentrations of the passive air samplers (in ng/PAS)

|  |  |  |
| --- | --- | --- |
| Descriptive PAS | Chlorpropham | Pendimethalin |
| Min | 34.24 | 44.47 |
| 1st Quartile | 156.78 | 183.90 |
| Median | 573.86 | 361.19 |
| Mean | 838.65 | 643.10 |
| 3th Quartile | 1500.26 | 817.26 |
| Max | 3196.23 | 4003.01 |
| SD | 768.05 | 821.65 |

**Table 2**: Descriptive of raw data from active air samplers (in ng/m3)

|  |  |  |
| --- | --- | --- |
| Descriptive AAS | Chlorpropham | Pendimethalin |
| Min | 0.10 | 0.03 |
| 1st Quartile | 1.18 | 0.74 |
| Median | 2.70 | 2.03 |
| Mean | 37.75 | 10.84 |
| 3th Quartile | 19.00 | 13.10 |
| Max | 1180.24 | 86.68 |
| SD | 148.29 | 19.12 |

* + 1. *Comparison between active and passive air samplers*

Before extrapolation, the difference between concentrations in AAS and PAS amounts to around two orders of magnitude. This is the case for both chlorpropham and pendimethalin(See Figure 2 and 3.

**Figure 2:** log-transformed pendimethalinconcentration distribution found within the air samplers (Pesticide conc. in ng).

**Figure 3:** log-transformed chlorpropham concentration distribution found within the air samplers (Pesticide conc. in ng).

* 1. *Statistics on methods of extrapolation*

To quantify the explained variability by the used extrapolation methods r-squared was calculated (see Table 3). Also to quantify the error made within the methods, the root mean square error (RMSE) was determined (see Table 4). Both the R-squared and the RMSE indicate that an individual derived sampling rate results in twice the error compared to the set value of the sampling rate within our study. For pendimethalin the environmental factors that resulted in the highest adjusted R-squared which is visible here were: temperature used in interaction with humidity. For chlorpropham these factors were temperature + humidity, so without using the interaction term in the model. All linear models were adjusted for passive air sampler deployment time. The result of the linear model for pendimethalin may be affected by outliers.

**Table 3**: Results of R-squared calculations for three extrapolation methods

|  |  |  |
| --- | --- | --- |
| Method | Pendimethalin | Chlorpropham |
| Set Value of R | 0.18 | 0.31 |
| Windspeed derived individual sampling rate | 0.18 | 0.31 |
| Linear Model | 0.40 | 0.71 |

**Table 4**: Results of Root mean square error calculations for three extrapolation methods (in ng)

|  |  |  |
| --- | --- | --- |
| Method | Pendimethalin | Chlorpropham |
| Set value of R | 15.79 | 13.11 |
| Windspeed derived individual sampling rate | 28.40 | 22.58 |
| Linear Model | 466.87 | 12.62 |

* 1. *Sensitivity analysis*

A sensitivity analysis between outside weather stations and HOBO-loggers deployed on the study sites was performed. Figure 4 shows the temperature variation across time. The temperature measured between both methods is quite different and this most likely caused by the fact that outside temperatures can not directly be used as values for inside homes. HOBO-loggers were not deployed outside of the residents homes during the OBO study, which could therefore not be included in the sensitivity analysis.

**Figure 4**: Relation between temperature found in KNMI weather station vs 6 HOBO-loggers inside homes

* 1. *Risk assessment using PUF-PAS data*

Daily average Inhalation rate adjusted for bodyweight was taken from the US governments Environmental Protection Agency (US EPA, 2011) The adjusted inhalation rate is combined with average air concentrations calculated using the PUF-PAS data to do a risk assessment. The risk assessment does not include other exposure rates such as through food or during occupational activities, given that the interest was on knowing how much pesticide inhalation contributes for the acceptable daily intake (ADI). Spray drift through air is one of the main exposure routes of residents living near agricultural areas (Holterman & van de Zande, 2010). Measured concentrations fell below the ADI of both age groups that were taken into account. Below in table 5 the outcomes of the risk assessment for both pesticides are visible. The 95th percentile is also shown in the table giving an indication of the highest section of the spectrum. Also on the 95th percentile data points, the level is below the ADI.

**Table 5**: Risk assessment results adjusted for inhalation rate and bodyweight (in mg/kg bw per day)

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  | Pendimethalin |  | Chlorpropham |  |
|  | Mean | 95th perc | Mean | 95th perc |
| Child (3-6 yrs) | 3.3\*10-4 | 4.0\*10-4 | 1.7\*10-5 | 2.2\*10-5 |
| Adult (21-31 yrs) | 1.0\*10-4 | 1.4\*10-4 | 5.0\*10-6 | 7.2\*10-6 |

*ADI Pendimethalin: 0.125 mg / kg bw per day*

*ADI Chlorpropham: 0.05 mg / kg bw per day*

1. **Discussion**

*4.1 Pesticide description*

The pesticides being investigated in this study were pendimethalin and chlorpropham. Chlorpropham was banned in 2020 within the EU, but could still be used during the study period which was in 2017. Together with pendimethalin chlorpropham was detected most often in the samplers used by this study. This is in line with the fact that these pesticides represent the largest market share in the bulb fields (OBO ref). Finally the fact that these two pesticides where found in the highest concentrations can be linked to the spraying season of the nearby bulb fields which occurred during the study period. Chlorpropham and pendimethalin are herbicides and also used as growth regulators to prevent sprouting.

In a similar French study, in the rural community of Oysonville, slightly lower concentrations were found. The range of pendimethalin concentrations found was: 0.46 – 3.98 ng/m3. The French study used PAS while also using AAS to verify measurements done with the passive samplers. Chlorpropham levels detected within this study were negligible.

*4.2 Evaluation of extrapolation methods*

At the moment there is no “golden” method on how to extrapolate concentrations found within the PAS towards the values in the air (cubic meter of air). To select suitable extrapolation methods a scoping review was performed. The three methods of calculation were selected, based on: I) they are commonly used in literature, II) are suspected to improve the precision, III) are suitable for incorporating the environmental circumstances (Diefenbacher *et al*, 2016). After determining predicted concentrations according to all methods deviations could be determined by comparison with results of active air samplers placed alongside the PAS.

*4.3 Sensitivity analysis*

The sensitivity analysis performed in our study shows the difference between the real environmental conditions and the proxies that may be used. Although this example is clear because of the conditions being in- and outside. The dataset used by this study did not include HOBO-loggers deployed alongside the samplers that were outside of the homes. Therefore this is an interesting opportunity for new studies to determine the gap when truly comparable data is used with both sources being in the outdoors. Further opportunities may lay in the modelling of barriers near the deployed samplers, as these could affect windspeed. The goal in mind would be a model using environmental circumstances and partition coefficients to be able to accept the complete array of semi volatile compounds.

*4.4 Risk Assessment*

A risk assessment, focusing on the inhalation pathway, was performed during this study using PUF-PAS data extrapolated to ng/m3 in air. The 95th percentile was determined to do a robust estimation using the pesticide concentrations, in combination with adjustments for the average adult or child body weight. Calculated exposure was always below ADI, even for the higher 95th percentile dose on children.

*4.5.1 Strengths*

For every deployed PUF used within this study there is a corresponding active sampler deployed, which was useful for the precision when comparing concentrations from both sources. Only two PUFS had pendimethalin & chlorpropham concentrations below the limit of detection (LOD). Therefore imputation only had to be performed for these data points limiting the amount of added statistical error. Humidity and temperature within homes could be determined precisely by using the HOBO loggers deployed within the houses on 6 locations. This indoor climatic conditions during the time that the samplers were deployed.

*4.5.2 Limitations*

Windspeed data was collected from KNMI weather stations, this involved having to convert the windspeed towards PUF deployment height (10m to 2m) using the wind profile power law. This is however a limitation that is common to studies that rely on weather stations. Another issue with the lack of on-site data on wind speed was the possible impact by physical barriers that could impact the wind speed. Barrier may also reduce air pollution by increasing deposition (Lee *et al*, 2018). One important limitation was that the sensitivity analysis could only performed by comparing weather station data with the data provided by the HOBO-loggers deployed inside homes of residents. Since no HOBO-loggers were deployed outside the homes of residents. Finally there is only a single type of passive air sampler tested in this study, another commonly used type is the polystyrene–divinylbenzene copolymeric resin-based (XAD) PAS. The difference in chemical structure of the matrix within the PAS will affect the partitioning coefficient and thus the sampling rate of the sampler(Harner *et al,* 2004).

*4.6 Future research on extrapolation methods*

For future evaluations of extrapolation methods for passive air samplers, another detailed take on the linear model using environmental conditions would be useful. An increase in sample size may reduce the effect of outliers that probably have skewed the data for pendimethalin during this study. When more suitable data on the environment is added using HOBO-loggers beside the outdoor-placed samplers there accuracy might be very high. The RMSE of chlorpropham was only 13ng/m3, which was promising. Possibly the error for the pendimethalin model could be reduced by the suggestions above. While the windspeed and set value calculations are useful for rough estimation, these methods can probably not match the accuracy of models using elaborate data. A new study evaluating multiple models under the same circumstances could prove interesting. A suggestion could be I) Mathlab model by Herkert, II) a PP-LFER based model, III) improved linear model.

Another interesting angle into the exposure of residents of agricultural areas was performed by Curchod *et al.* (2020) were an analysis was done of pesticides in the water in agricultural areas. If both air and water could be analysed in the same study, a better view could be obtained on the interaction of pesticides between water and air. This would add information on the fate of the degradation productions of the chemicals of interest.

**Conclusions**

This study examined the performance of PAS compared with AAS by studying pendimethalin and chlorpropham in agricultural areas. Concentrations found within the matrix of the PAS were extrapolated to ng/m3 in the air using rough calculations and models resulting in varying error rates compared to AAS derived air concentrations. Most promising was a linear model fitted with relative humidity and temperature. The precision of extrapolation methods may be enhanced by incorporating spatial features that could impact the windspeed and direction.

**Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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