Proposals for new spray drift exposure values in orchard and vineyards for residents and bystanders

Bystander Resident Orchard Vineyard (BROV) Project report

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Content

1 Abstract

New spray drift exposure values for residents and bystanders in orchards and vineyards have been developed for the assessment of plant protection products in the EU. They are based on drift trials undertaken on behalf of the Occupational and Bystander Exposure Expert Group (OBEEG) group of the European Crop Protection Association for the purposes of addressing the EFSA Working Group recommendations to further collect/produce data on relevant drift for residents/bystanders after application in high crops. Exposure values can be obtained for adults and children located at different distances up to 15m from the sprayer. Values were further refined to differentiate between orchard crops and vineyards and between early and late season application. For dermal exposure both potential dermal exposure (PDE) and actual dermal exposure (ADE) values for adults and children wearing light clothing (shorts and t-shirt) have been obtained. Potential inhalation exposure to spray is also included. Vapour concentration in the breathing zone of adults and children was monitored but detailed analysis is not included within this report. The trials were undertaken in a range of locations and conditions such that the data and associated models can be applied by all regulatory authorities as a harmonised approach for the evaluation and authorisation of plant protection products.

2 Summary

For the purposes of estimating exposure to residents and bystanders as a consequence of high crop spraying in orchards and vineyards the existing approach in the EFSA Guidance document[1] and associated calculator is based on a single study by Lloyd *et al* [2] in orchard crops using conventional nozzles (no drift reduction technologies) applying 470 L/ha for an 8m distance downwind from the middle of the tree trunk. Given these data are from a single study conducted in 1987, further modern data are required reflecting a range of operators and equipment under varying typographical and meteorological conditions.

The BROV bystander/resident exposure data base consists of 16 studies conducted in four EU countries, 8 in orchards and 8 in vineyards. For each crop 4 trials were undertaken at an early growth stage and 4 at a late growth stage. Potential and actual dermal exposures were measured using adult and child sized mannequins which were located at distances of 5m, 10m and 15m downwind of the crop. Three replicates of each mannequin at each distance were included. Potential inhalation to spray drift was measured during application via air samplers located within the adult / child breathing zone at heights consistent with BREAM[3] field trials. Potential inhalation of vapour over 7 days post application was also measured.

Two active substances were applied by orchard and vineyard growers using their own application equipment with minimal intervention to reflect real life application practices and a range of equipment. The data frame consists of 288 observations (16 studies x 18 adult/child mannequins per study) and includes detailed information relating to the

application equipment, application parameters, crop (including downwind vegetation) and meteorological conditions. Additional parameters were also derived to summarise some of the more complex variables and aide analysis. This included factors such as spray quality, sprayer type, leaf cover and mean wind direction.

For the purposes of direct comparison with the existing spray drift values from Lloyd *et al,* 75th and 95th percentiles were derived for orchards and vines at early and late stages of crop development at 5m, 10m and 15m downwind of the crop. Unlike the approach in the EFSA guidance document in which exposure to adults was extrapolated to children by a factor of 0.3, separate measured values were obtained for each. The new data provide measured PDE and ADE for individuals wearing shorts and a t-shirt compared to an assumed clothing reduction factor of 18% applied to the exposure values from Lloyd.

This paper presents 75th and 95th percentile exposure values for PDE, ADE and potential inhalation exposure (PIE) for adult and child residents and bystanders according to crop, leaf cover and distance from sprayer. Regression analysis was performed to identify and model key influencing parameters to attempt to determine an appropriate approach to use these data for pre-authorisation exposure estimates. It was concluded that a consistent and practical approach was to consider the variation in exposure with distance from the source. The vapour monitoring data are not reported in this paper. Many vapour measurements were <LOQ, however, further consideration of these data is required and could include a comparison of measured data with those derived using the BROWSE[4] vapour model.

3. Introduction

There are many factors that affect spray drift and it was the intention of the study sponsor to derive measured exposure levels reflecting a range of circumstances and typical everyday use. Because the trials were not constrained the variability associated with individual behaviours, range of application equipment and meteorological and typographical conditions are captured within the data. The studies undertaken on behalf of ECPA have been made fully available to regulators with no conditions imposed on their use. As such they represent a significant data set upon which to derive new dermal and inhalation exposure values.

4. Study design

All sixteen spray drift studies were conducted to the same study protocol and essentially fulfilled the same quality criteria as those included in the recent Agricultural Operator Exposure Model (AOEM)[5] database accepting some obvious differences between operator studies and those measuring spray drift onto residents / bystanders. GLP compliance certificates, compliance statements and QA statements were provided for both

field and analytical phases for each of the submitted studies. In addition to full GLP compliance and in line with the quality criteria specified for the AOEM database the submitted spray drift studies were deemed to fulfil the following quality criteria.

- Compliance with the principles of OECD Series No.9
- Data recording and observations according to current scientific knowledge
- Consistent field recovery (any outlying data must be explainable on a scientific basis)
- Suitable data form for model development (e.g. separately measured areas of the body)
- Whole body dosimetry for dermal exposure
- Inhalation exposure determined with appropriate inhalation fraction samplers
- Representative application methods and application techniques reflecting current agricultural application practices in Europe.

The studies were undertaken in 4 different EU countries and were balanced between active substance applied, crop type and growth stage.

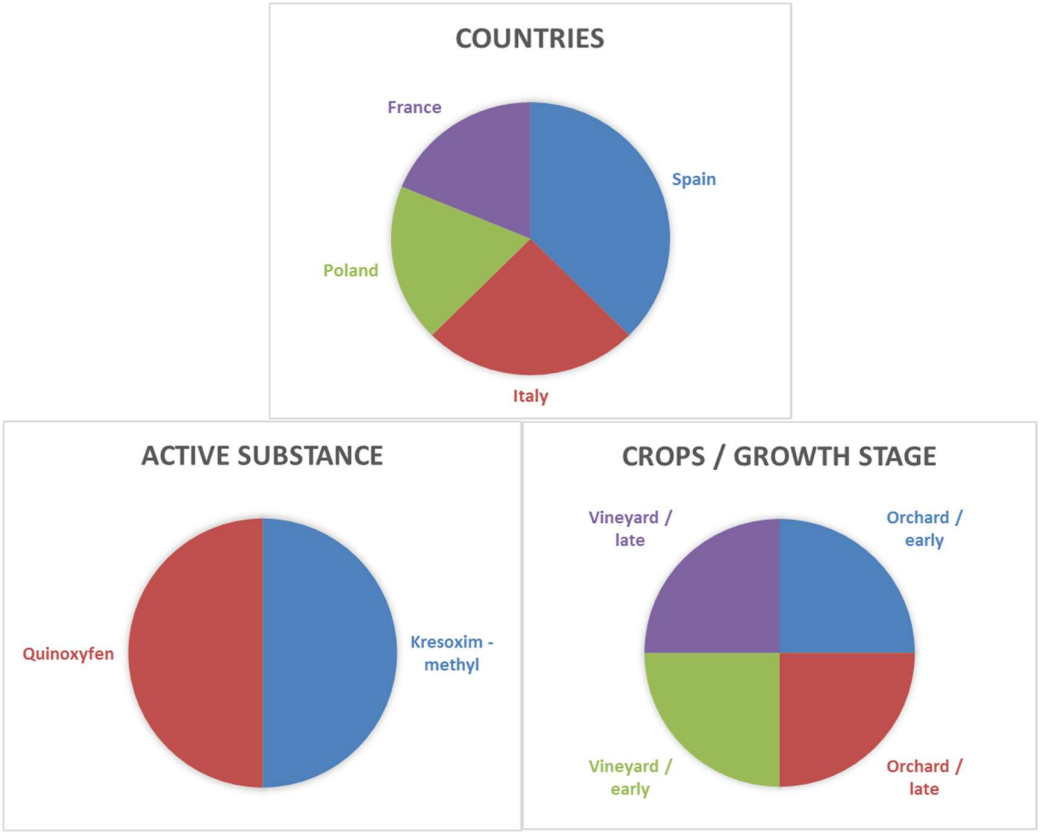


Figure 1: Study overview; the 16 trials were undertaken in Spain (6), Italy (4), Poland (3) and France (3); quinoxyfen was used exclusively in vineyards (8 trials) and kresoxim-methyl in orchards (8 trials); crops and growth stages were split equally with 4 trials conducted for each.

The studies were undertaken in 2016 to 2017 using a range of application equipment by operatives during the normal course of their work. The area treated ranged from 0.72 to 1.59 ha and the duration of spraying between 34 and 85 minutes. Full details of the test material (product, actives, formulation type, dose and application volume) were reported (see Appendix A). Each trial was set up the same in which six replicates (3x adult and 3x child mannequins) were placed at 5m, 10m and 15m from the zero metres position defined as half the row width from the base of the outermost tree or vine. The adult mannequins were 1.88m in height and the child mannequins 1.0m in height. PDE and ADE was measured using whole body dosimetry with shorts and t-shirt worn over inner dosimeters consisting of full length ≥95% cotton underwear garments (long sleeved vest and long johns) and ≥95% cotton head sleeve. Outer dosimeters consisted of 100% cotton shorts and T-shirt.

PIE of airborne spray was measured using IOM samplers located in the breathing zone of each mannequin for the duration of spraying. One sampling cassette per mannequin was positioned 1.5m (adult) and 0.7m (child) above the ground to represent the breathing zone. The pump drew air through the sampling media at 2.0 ±0.1 L/min. Potential inhalation of vapour was measured using XAD-2 OVS air sampling tubes placed on each side of the treated area and 10m from the perimeter at 0.7m and 1.5m above the ground. Sampling began after spray droplets on the dosimeters were deemed to have dried. The vapour sampling tubes were replaced every 8 hours for 7 days following application. Flow rate was 1.0 ±0.1 L/min.

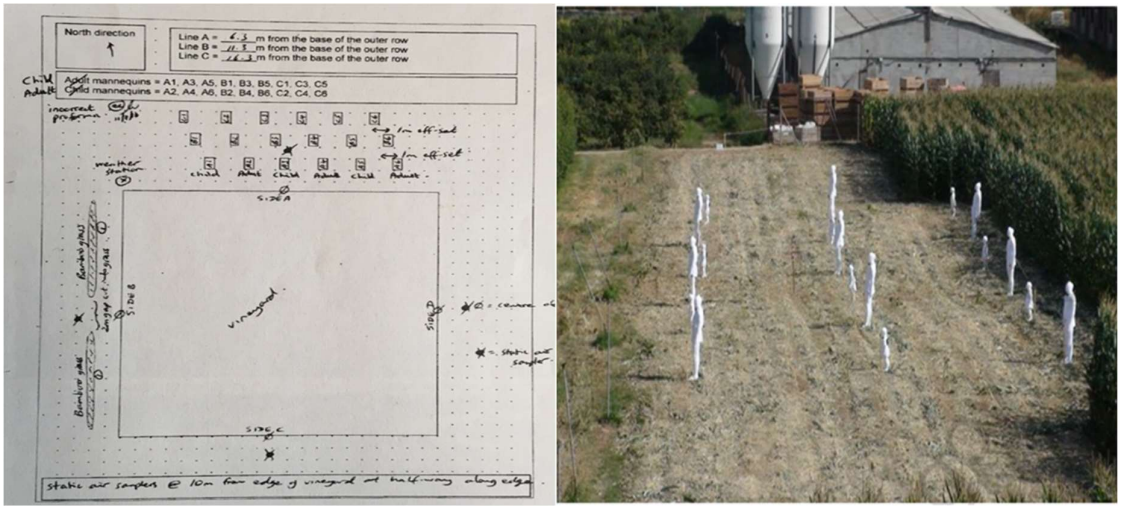


Figure 2: Field trial set-up; the location of the adult/child mannequins is downwind of the treatment area and mannequins at each of the three measurement distances were off-set to minimise any 'shadowing'

Sample handling

Full details of the removal and sectioning of dosimeters, the packaging (e.g. foil wrapping and double bagging) of the samples and subsequent placement in frozen storage (below -18 ºC) were reported.

Field recoveries

Three sets of field fortifications (x 2 concentrations) were undertaken for the cotton dosimeter (300 cm^2 patches) and IOM filters on the day of application. Fortified specimens were exposed to the same environmental conditions for the same period of time. XAD-2 OVS sampling tubes were fortified on each of the 7 sampling days (x 2 concentrations). One control sample for cotton dosimeter and IOM filter and one control for each day for XAD-2 OVS sampling tubes were included. Measurements for which the recoveries <95% were scaled up and measurements for which recoveries >100% were not adjusted. Only two fortification levels were used to measure field recoveries, these being 0.1 and 10 µg/sample for inhalation samplers (both IOM air filters and XAD-2 OVS air tubes) and 1 and 100 µg/sample for cotton dosimeters.

For inhalation air samplers most field measurements were < 0.1 µg/sample and all were <0.5 µg/sample, therefore the mean recovery (3 replicates) for the low level fortification was used in all cases where adjustment was necessary. For cotton dosimeters actual measured values ranged from LOD to 3000 µg/sample and were well outside the range tested. Ideally a third spiking level (e.g. 1000 µg/sample) would be necessary. However, recoveries for the 1 and 100 µg/sample fortifications were broadly similar and at a relatively high level such that the omission of a third field fortification level was not considered to create significant uncertainty in accepting the analytical results. For cotton dosimeter measurements <50 µg/sample the mean recovery (3 replicates) for the low level fortification was applied and for measurements ≥ 50 µg/sample the mean recovery (3 replicates) for the high level fortification was applied.

Travel recoveries

Travel recovery samples were undertaken for each sampling material (cotton dosimeters, IOM air sampling filter, XAD-2 OVS air sampling tubes) on the day prior to application. 1 x fortification for each material was undertaken. These should be shipped and stored with the field recovery and field monitoring samples but this is not clear from the report. With one exception mean recoveries for transit samples were within 70-120% of the fortification dose and mean RSD values <20% in all but two studies.

LOD/LOQ

Inhalation samplers: For both quinoxyfen and kresoxim methyl the LOQ was 0.01 µg/sample. The LOD for kresoxim methyl was 0.0001 µg/sample (spray and vapour) and for quinoxyfen the LOD was 0.007 µg/sample (spray) and 0.0032 µg/sample (vapour). Regarding the treatment of values below these levels, it was noted in the AOEM project the approach was to use ½ LOQ for values between LOQ and LOD and 0.01 µg/sample as a default value for the LOD. For this project a default LOD of 0.01 µg/sample is not appropriate given this is the same value as the LOQ for both substances. Also ½ LOQ < LOD for quinoxyfen therefore the ½ LOQ substitution approach is not appropriate. Therefore, in this case a slightly different approach was taken: measured values between LOQ and LOD were substituted with the LOQ and values reported as ND were substituted with the LOD relevant to each active.

Cotton Dosimeters: For both quinoxyfen and kresoxim-methyl the LOQ was 0.1 µg/sample and to be consistent with the approach for inhalation samplers the LOQ was used for values between LOQ and LOD. The LOD for kresoxim-methyl depending on the dosimeter ranged from 0.0058 to 0.027 µg/sample and for quinoxyfen ranged from 0.0035 to 0.0115 µg/sample and all cases values reported as ND were substituted with the relevant LOD.

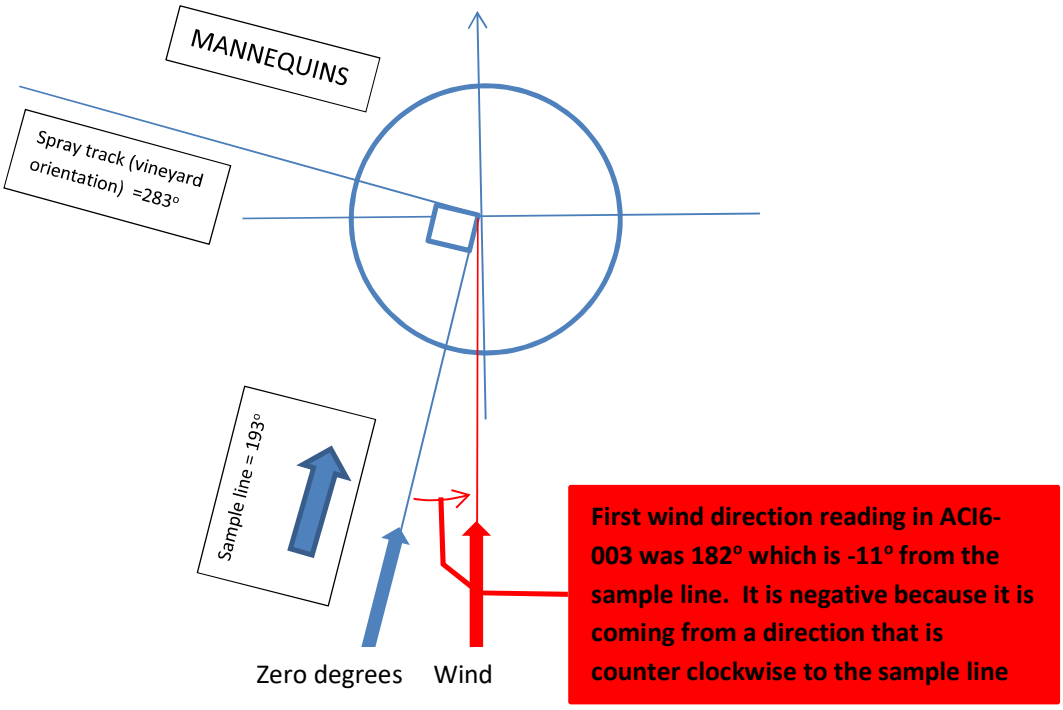
Environmental monitoring

Environmental parameters (air temperature, relative humidity, solar radiation, wind speed, wind direction and rainfall) were recorded on-site at 10 second intervals from prior to the application until completion of the application and then at 15 minute intervals until the last sampling at 7 days post application. With the exception of AC16-003 and AC16-005 the study reports contained mean values for humidity, temperature and radiation and these were cross checked with detailed observations obtained separately from the study author. For studies CEMR 8025, 8026 and 8028 temperature and relative humidity measurements were recorded on handheld anemometers due to malfunctioning met station equipment. Across all the studies the temperature ranged from 2 to 32^{o}C, humidity from 32 to 86% and average windspeed 1 to 4 m/s. See Appendix B

Wind direction

Ideally wind direction should be perpendicular to the direction of travel of the sprayer. Whilst this reflects the ideal situation in which to maximise measured spray drift onto downwind mannequins, shifts in wind direction are to be expected and subsequent measurements simply reflect the inherent variability of prevailing conditions. In the case of wind direction, it is not appropriate to simply report a mean value for angular or circular data (i.e. there is an issue where the direction passes through the discontinuity at 360/0 degrees). However, some instances of this erroneous approach were noted in the study reports so for this evaluation the wind data averaging was recalculated. Several approaches were considered but the simplest and preferred approach is to express the individual wind direction values as deviations from the sample line, this being the ideal wind direction of perpendicular to the direction of travel of the sprayer. The drift standard for classification of spray equipment ISO 22369[6] states the spray track as the datum, however, the sample line is considered a better one to use avoiding the confusion of which direction the spray track is (i.e. left to right or right to left). For comparison of wind direction against the ISO standard, the sample line is equivalent to 180° and the wind directions adjusted so that they are expressed as deviation from the sample line (i.e. the data are expressed as ± 180°). The direction of angle is measured clockwise from the sample line; a clockwise shift will result in a wind direction > 180° (i.e. positive value) and an anticlockwise shift will result in a wind direction < 180 (i.e. negative value). The resultant finite scale (-180° to +180°) avoids any crossover discontinuity in the circular data and makes the estimating the average direction angle straightforward. This makes it possible to give some indication of the variability using the absolute values.

Study AC16-003 Field trial set-up



	Direction	Notes
Measured wind direction	182°	
Spray track	283°	
Sample line	193°	±90 dependent on mannequin location
Default sample line	180°	
Required change in wind direction values	347°	MOD* (180° – 193°, 360°)
Wind direction data relative to default sample line	169°	MOD* (182° + 347°, 360°)
Variation from sample line	-11°	169° -180°

*MOD function in excel calculates the remainder after division of two numbers

Figure 3: Wind direction calculation; In study AC16-003 the vineyard row orientation or spray track is given as 283° with the unobstructed area (i.e. the area in which the mannequins are situated) being to the north. In this case the desired wind direction being perpendicular to the spray track and towards the mannequins is 193°. As the sample line is assumed to be equivalent to 180° then the wind directions are adjusted so that they are expressed as deviation from the sample line (i.e. the data are expressed as ± 180°)

Sprayer classification

The different sprayer types used in the trials have been classified into one of two groups in order to facilitate the analysis of this information. The two groups are identified as:

(i) R = Radial. Typically an axial fan sprayer in which spray is relatively undirected travelling in a downwards, sideways and upwards direction.

(ii) S = Sideways (with high release point). Typically a crossflow sprayer in which spray is predominantly directed sideways by comparison with the axial fan sprayer.

The Pulveman sprayer in Study CEMR 8028 included downward facing deflectors. For the Calvet Pneumatic sprayer, the downwards facing ducts were switched on when treating a full foliage vine in CEMR 8027 and switched off when treating a low foliage vine in CEMR 8026. Over the 16 studies there were 12 different types of sprayer 6 of which were classed as 'radial' and 6 as 'sideways' sprayers.

Figure 4: Sprayer classification; image on the left is 'Balleste' radial sprayer whilst those on the right are examples of sideways sprayers (e.g. 'Dragone', 'Dominiak', 'Pulveman' and 'Calvet Pneumatic')

Spray quality

Inclusion of spray quality criteria into the database was based on nozzle type and spray pressure according to nozzle manufacturer's charts. All trials were undertaken using either VF = very fine or F = fine spray quality which is typical for vineyard / orchard application.

The spray quality of the Calvet 10/10 pastilles (1mm diameter, flow rate = 0.65 L/min) as seen in Study CEMR 8027 and Calvet 16/10 pastilles (1.6 mm diameter, flow rate = 1.31 L/min) as seen in Study CEMR 8026 could not be identified.

Foliage density

The EFSA calculator currently has two selection options when considering high crop application these being 'early (without leaves)' and 'late (dense foliage)'. The canopy acts as a filter that may catch part of the passing spray plume and the selection of the late (dense foliage) option has the effect of reducing spray drift fallout onto horizontal surfaces. It does not currently affect the amount of spray drift onto vertical (adult or child) surfaces.

The BROV spray drift trials were set up to reflect a clear differentiation in foliage between early and late application with no trials undertaken in what might be described as an 'interim' stage of foliage development.

Figure 5: Foliage density, demonstrating the clear differentiation in a vineyard between early and late application.

The BROV database consisting of 16 trials is equally divided into 4 trials each for early and late stage application in vineyards and orchards respectively and the data can easily be segregated and interpreted in this way. However, given application can occur throughout the year further practical guidance is needed to help determine the circumstances under which it would be appropriate to assign the late stage scenario.

Scenario	BBCH	Description
Vineyard early	13 to 15	3 to 5 leaves unfolded
Vineyard late	81	Beginning of ripening: berries begin to develop variety-specific colour
Pome fruit early	53 to 57	Bud burst: scales separated, light green bud sections visible Sepals open: petal tips visible; single flowers with white or pink petals (still closed)
Pome fruit late	81 to 91	Beginning of fruit colouring Shoot growth completed; foliage still fully green

Figure 6: Growth stages; range of BBCH phenological plant growth stages under which the BROV spray drift trials were undertaken

A consideration of the relationship between growth stage in orchards and canopy density is included in a paper by Holterman *et al*[7] in which it is observed that whilst canopy density is a function of growth stage, the phenological BBCH scale is non-linear in time and it is more appropriate to relate this to the DOY or day-of-year. Figure 7 shows the estimated relationship between BBCH and DOY for apple trees in The Netherlands. Assuming a roughly similar relationship for orchards in other EU countries in which the BROV trials were undertaken, early stage applications appear to occur between DOY 60-100 and late stage applications between DOY 240-270.

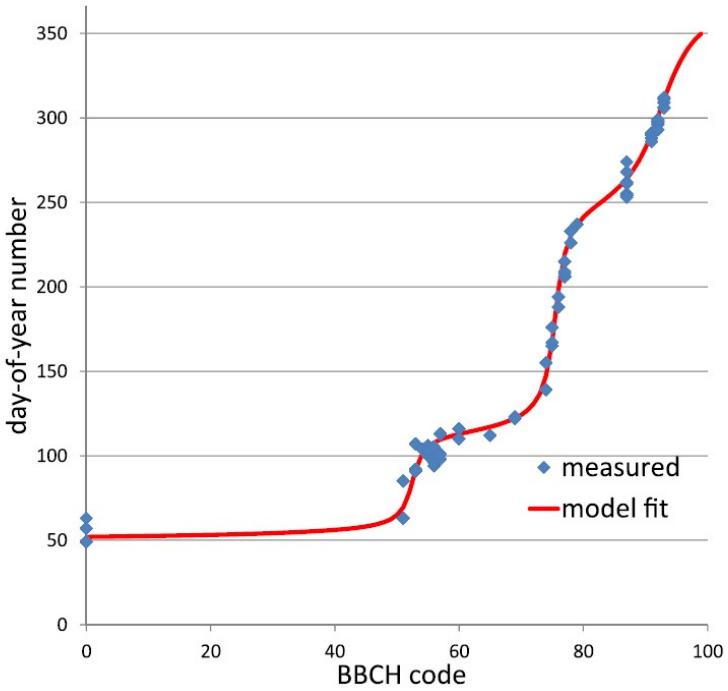


Figure 7: BBCH growth stage vs DOY; from expert judgement for the spray drift experiments in apple trees (dots). Solid line: fitted curve. From Holterman *et al*

Holterman *et al* describes a 'canopy density factor' β to quantify the effect of the tree canopy in which β=0 for bare trees and β>0 as the canopy develops. The canopy density factor is a function of growth stage and since the phenological BBCH growth scale is non-linear in time it is more practical to describe β as a function of z (where z = DOY expressed as a fraction of the year i.e. DOY/365).

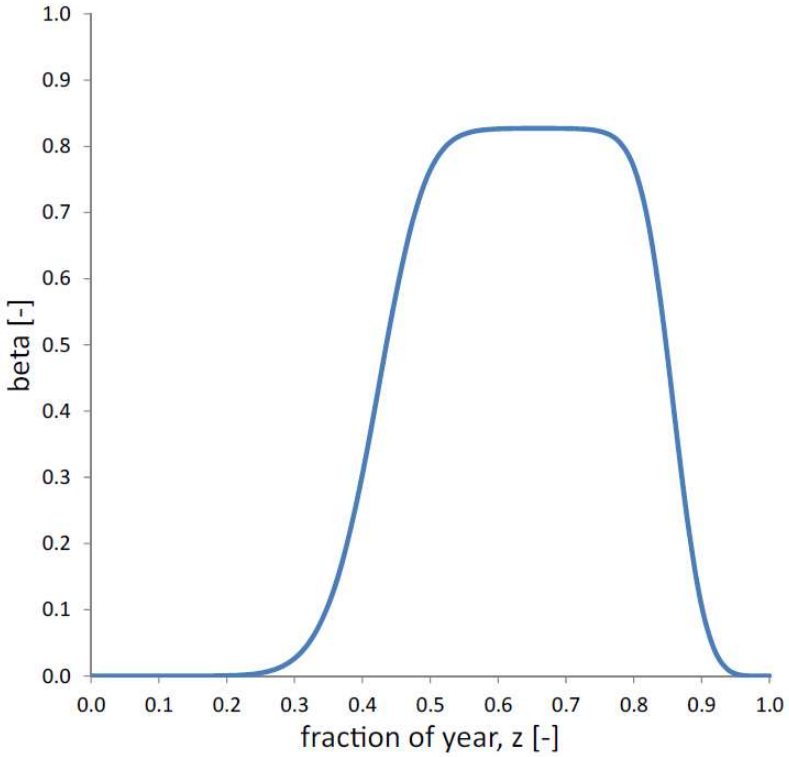


Figure 8: Canopy density factor β as a function of fraction of year (z). From Holterman *et al*

The early orchard trials for which the DOY is estimated to be 60-100 equates to a z fraction of 0.16 to 0.27 for which β is approximately 0 according to the curve above. The late trials for which estimated DOY is 240 to 270 equates to a z fraction of 0.66 to 0.74 for which a maximum β value of 0.8 is observed. This supports the growth stages selected in the BROV trials as being representative of early and late application. According to Figure 8, the maximum canopy density factor occurs between z = 0.5 to 0.8 approximately, which is equivalent to DOY 183 to 292. In growth stage terms (according to Figure 7) DOY 183 is roughly equivalent to the beginning of the BBCH growth stage 7 Development of fruit (which includes stages BBCH 71 to 79). For risk assessment purposes it is proposed that late (dense foliage) application can be assumed in orchards for which the BBCH growth stage is from 71 to 93 (beginning of leaf fall). For vineyards the later development stages are equivalent to orchards such that BBCH 71 also equates to the beginning of fruit development and BBCH 93 to the beginning of leaf fall so the same criteria could apply.

<u>Spray volume and amount of active substance applied</u>

Both the amount of product and water added to the spray tank were determined and measured by the operator and recorded in the field by the study team. The product was measured using a graduated jug or cylinder. The amount of kresoxim-methyl applied in orchards ranged from 89 to 127 g a.s./ha and for quinoxyfen in vineyards from 11 to 67 g a.s./ha.

Water was measured using the graduations on the side of the spray tank (on level ground) except for the 3 French vine studies where it was measured using an in-line flow meter. The amount remaining after treatment, where the volume was small, was determined by measuring cylinder. Where it was more than about 50 L it was determined from graduations on the spray tank. Whilst spray tank samples were taken none were analysed to confirm the in-use concentration. This has been calculated from the measured amounts of product and water. The accuracy of the spray concentration is important as it is the parameter required to convert back the amount of active substance measured to the amount of spray contamination expressed as ml of spray/person. The more dilute the spray the greater the spray volume required to achieve the same amount of active substance contamination. Measurement errors up to plus or minus 5% could occur through use of spray tank graduations or in line flow meters. This uncertainty could be incorporated into the calculation of spray concentrations; however, measurements could be both positive or negative and potentially cancel the other. No adjustment has therefore been made here. However, it is recommended that future studies should include analysis of spray solution concentration both in the spray tank and from spray solution collected from the nozzle.

Whilst the calculated spray concentration has been accepted without adjustment, there is greater uncertainty as to the amount of spray solution that was actually applied to the crop (and therefore to the mannequins) relative to the target dose. According to the study reports the volume of spray applied by the operators ranged from 122 to 872 L/ha in vineyards and 530 to 1091 L/ha in orchards. Spray equipment was not calibrated on the day of application in any of the trials although confirmation was sought that spray equipment was in compliance with the relevant testing requirements in each of the four countries in which the trials were undertaken.

The Silsoe Spray Applications Unit (SSAU) Ltd have undertaken a technical review of the content of the field trial reports, Butler Ellis C, 2019[8]. The report assesses the conduct of the trials (from the perspective of the application technique) and the meteorological data to determine whether the exposure data is appropriate for consideration in future regulatory risk assessments. A copy of the report is provided in Appendix C. As part of the analysis SSAU calculated the theoretical spray output of the spray equipment and compared this to the reported volume of spray applied.

Reported spray outputs were cross checked with theoretical spray output according to the following formula:

Spray output (L/ha) = (600 x nozzle flow rate (L/min)) / (forward speed (km/h) x W)

Where

W = row spacing (m) / number of nozzles

Figure 9 provides a comparison of reported and calculated spray volumes. In 5/16 trials the difference was estimated to be plus or minus 5% or less, 6% to 34% in 4/16 trials and ≥ 35% in 7/16 trials. The greatest uncertainty is associated with the vineyard trials in which 6/8 gave rise to large unaccountable differences in which all reported water volumes were much lower than those calculated.

Study ID	Study Code	Country	Crop	Water volume reported L/ha	Water volume calculated SSAU L/ha	% Difference
AC116-003	ECPA_1	Spain	Vineyard (low leaf cover)	346	274	21%
AC116-005	ECPA_2	Spain	Pome fruit (low leaf cover)	610	577	5%
CEMR-7089	ECPA_3	Italy	Pome fruit (full leaf cover)	1091	1069	2%
CEMR-7090	ECPA_4	Italy	Vineyard (full leaf cover)	597	691	-16%
CEMR-7091	ECPA_5	Spain	Pome fruit (full leaf cover)	962	633	34%
CEMR-7092	ECPA_6	Spain	Vineyard (full leaf cover)	872	1129	-29%
CEMR-7456	ECPA_7	Italy	Pome fruit (low leaf cover)	1055	1069	-1%
CEMR-7457	ECPA_8	Italy	Vineyard (low leaf cover)	150	1093	-629%
CEMR-7458	ECPA_9	Spain	Pome fruit (low leaf cover)	641	551	14%
CEMR-7459	ECPA_10	Spain	Vineyard (low leaf cover)	569	1491	-162%
CEMR-7500	ECPA_11	Poland	Pome fruit (full leaf cover)	605	636	-5%
CEMR-7501	ECPA_12	Poland	Pome fruit (full leaf cover)	824	792	4%
CEMR-8025	ECPA_13	Poland	Pome fruit (low leaf cover)	530	716	-35%
CEMR-8026	ECPA_14	France	Vineyard (low leaf cover)	140	718	-413%
CEMR-8027	ECPA_15	France	Vineyard (full leaf cover)	122	266	-118%
CEMR-8028	ECPA_16	France	Vineyard (full leaf cover)	213	452	-112%
	A small difference – some or all of which could be explained by inaccuracies in volume measurement; could be regarded as a good estimate of the application volume					
	A large difference, cannot be explained by inaccuracies in volume measurement alone; but could be explained by variable speeds or the point at which spray stopped on turns.					
	A large difference that cannot be explained by inaccuracies in volume measurement; or by speed or other events					

Figure 9: Percentage difference in reported vs calculated water volume
(adapted from Table 2, Butler Ellis C, 2019, Appendix C)

If it is assumed that row spacing and number and type of nozzles are likely to be accurately reported then errors could be a consequence of incorrect flow rate (possibly because of worn nozzles or incorrect operating pressure) or average forward speed. Further detailed investigation of the study data was unable to resolve these discrepancies in water volume and may impact on modelling the relationship between exposure and spray volume and/or amount of active substance applied.

5 Methods of analysis

The analytical methods for quinoxyfen and kresoxim-methyl were fully reported and were conducted in accordance with SANCO/3029/99 rev.4[9] a full consideration of which is provided in Appendix H.

6 Results

The BROV database for spray drift (BROV WG bystander database_V8) was compiled by UK HSE based on the original study reports and supplementary Excel files from the study authors (i.e. inhalation sampler flow rates and some meteorological data). The data frame was compiled by UK HSE therefore it was considered to be acceptable that validation was undertaken by ECPA and should comprise the following checks:

- all non-calculated values / information must accurately reflect the contents of the study reports or supplementary files (i.e. sampler flow rates and supplementary meteorological data);
- values reported below the LOQ or LOD were correctly substituted with the relevant LOQ and LOD values for each active substance/dosimeter type;
- all formula and calculated values are correct;
- the relevant recovery adjustments have been undertaken;
- the final calculated 75th and 95th percentile values for dermal and inhalation exposure are correct.

The data frame was validated and agreed as version 'BROV WG bystander database_V8' and a copy of the raw data relating to dosimeters and inhalation samplers is provided in Appendix D. Visualisation of the data was undertaken using 'R' version 3.4.1('single candle') and associated package 'ggplot_2'

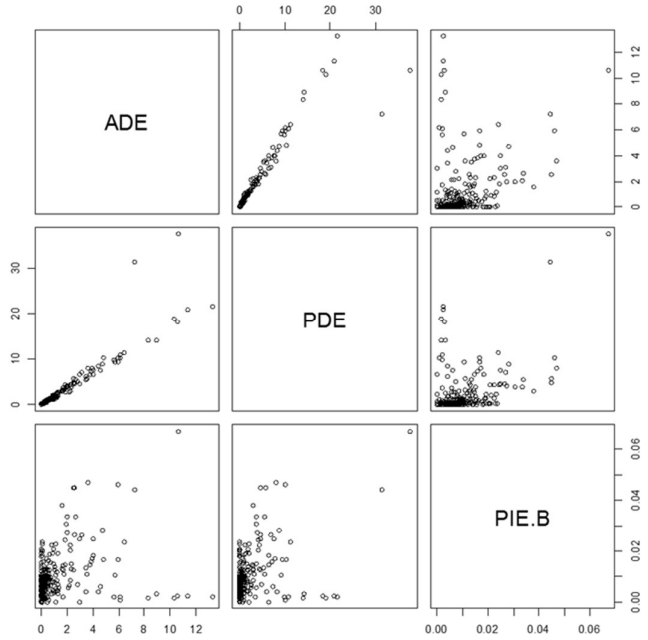


Figure 10: Relationship between PDE, ADE and PIE; As expected a clear correlation between PDE and ADE was observed but a similar relationship with PIE was not observed.

Appendix E provides a range of graphs comparing dermal and inhalation exposure to a range of variables. These show levels of exposure in vineyards to be significantly lower

than in orchards and levels of exposure as a consequence of late growth stage application to be significantly lower than when no leaf cover is present. As would be expected both dermal and inhalation exposure generally decrease with increasing distance from the sprayer although this relationship is not quite so clear cut with regard to inhalation exposure in vineyards.

Wind speeds above 2 m/s give rise to higher dermal exposures in orchards but not in vineyards. With regard to wind direction the '0' position equates to right angles to the direction of travel of the sprayer. Exposures in orchards were observed to be highest at this position but no such relationship is apparent in vineyards. Noting the uncertainty arising from reported and calculated spray volumes there appears to be no clear relationship between the volume of spray applied and dermal exposure in orchards or vineyards. The highest exposures arise from application of kresoxim-methyl in orchards for which rates of application are higher than quinoxyfen in vineyards.

A range of different application equipment was used in the trials and for simplicity these were placed in two main groups. These are (i) 'radial' axial fan type sprayers in which spray is relatively undirected and (ii) 'sideways' crossflow type sprayers in which the spray is predominantly directed sideways. In vineyards radial sprayers result in higher PDE than sideways sprayers, the relationship is not so marked in orchards but nevertheless is the same. Spray quality criteria according to nozzle manufacturers charts were defined as 'very fine' or 'fine'. Orchard applications predominantly used a very fine spray but the small number of trials using a fine spray quality appear to lead to higher levels of PDE. In vineyards the difference is not as marked but again the relationship is the same.

Further consideration of the uncertainties in the reported water volume applied has be undertaken in Appendix E6 by comparing the relationship between dermal and inhalation exposure estimates expressed as volume of spray compared to mass of active substance. The analysis showed a good correlation of the data between exposure volume and mass of active substance, and in conclusion the expression of exposure as a volume of spray is considered to be reasonable, especially to facilitate comparisons with other data. However, the exposure expressed as mass of active is considered to be the definitive value.

Appendix F and G summarise the 75^{th} and 95^{th} percentile values for PDE, ADE and PIE relating to residents and bystanders respectively. In the case of PIE, inhalation exposure was based on the adult/child breathing rates for residents and bystanders as stated in the 2014 EFSA guidance document.

Age Group	Daily inhalation rate			
	Bodyweight (kg)	Inhalation rate (m^3/day/kg)	Inhalation rate (L/min)	Flow sampler scaling factor
Adult	60	0.23	9.6	4.8
Child	10	1.07	7.4	3.7

Age Group	High intensity hourly inhalation rate			
	Bodyweight (kg)	Inhalation rate (m^3/h/kg)	Inhalation rate (L/min)	Flow sampler scaling factor
Adult	60	0.04	40.0	20.0
Child	10	0.19	31.7	15.8

Figure 11: Adult/child daily inhalation rates (for longer term exposures) and hourly inhalation rates (for acute exposures)

The measured values on IOM samplers were derived from a flow rate of 2 L/min and were scaled up based on the inhalation rates referred to in Figure 11. The sampling duration reflected the total time spraying took place which ranged from 34 to 85 minutes. The extent to which it would be realistic to assume that at a bystander would be exposed to and be able to maintain a high intensity breathing rate throughout the duration of spraying was considered. It was assumed to be unrepresentative or unrealistic for a bystander to remain in close proximity to the spray operation whilst maintaining a high intensity breathing rate for half an hour or more and therefore a 15 minute exposure duration was assumed to be more realistic. On this basis an additional set of values is presented in which bystander PIE is normalised to 15 minutes. Results are shown in comparison with the existing data from Lloyd *et al*.

7 Conclusions

Overall the BROV spray drift trials are considered to be a well conducted set of trials in accordance with quality criteria applied in the AOEM (Agricultural Operator Exposure Model) although the lack of adequate calibration introduces some uncertainty particularly when considering the relationship between exposure and active substance applied. However, 16 spray drift trials undertaken by different operatives using a range of equipment in varying 'real life' conditions within the EU provides a significant improvement in the data base supporting spray drift in high crops. Visualisation of the data show a clear differentiation between adult and child, crop type, leaf cover and distance from the sprayer supporting the approach of deriving indicative 75th and 95th percentile exposure values for each subset of data.

Levels of dermal and inhalation exposure are significantly lower in vineyards than in orchards and further analysis is required to understand the factors that may be influencing this. Also important is the relevance of leaf cover and when the crop can be considered to have 'dense foliage'. The proposal in this paper is that 'dense foliage' can be assumed between BBCH 71 and BBCH 93 for both orchards and vineyards.

Analysis of the associations between exposures and potential explanatory variables, such as wind speed, wind direction, sprayer type, spray quality, spray concentration and amount applied did not yield any robust model to describe the observed exposures that could serve as a predictive model for additional scenarios. Therefore, it is proposed that the observed percentiles should be regarded as representative exposure values for applications involving similar application rates to those in the trials. For vineyard applications the early stage trials were undertaken at 11-50 g a.s./ha and the late stage trails were undertaken at 47-75 g a.s./ha. For orchard applications the rates were 89.2-97 g a.s./ha and 96-127 g a.s./ha, for the early and late trails, respectively. As the relationships between amount of active substance applied and exposures were not robustly identified, it would be protective to assume the reported percentiles also apply to situations below the trial ranges. For cases where the application rates are above the range observed in the trails a pro rata assumption should be assumed and the reported percentile values adjusted upwards.

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9 Abbreviations

ADE	Actual Dermal Exposure
AOEM	Agricultural Operator Exposure Model
BREAM	Bystander and Resident Exposure Assessment Model
BROV	Bystander Resident Orchard Vineyard project
BROWSE	Bystanders, Residents, Operators and WorkerS Exposure models
ECPA	European Crop Protection Association
EFSA	European Food Standards Agency
EU	European Union
GLP	Good Laboratory Practice
IOM	Institute of Occupational Medicine
LOD	Limit of Detection
LOQ	Limit of Quantification
ND	Non Detect
OBEEG	Occupational and Bystander Exposure Expert Group
OECD	Organisation for Economic Co-operation and Development
PDE	Potential Dermal Exposure
PIE	Potential Inhalation Exposure
QA	Quality Assurance
RSD	Relative Standard Deviation
SSAU	Silsoe Spray Applications Unit

Appendix A: Application parameters

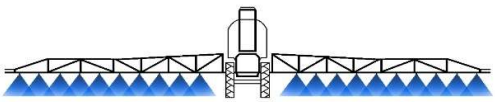
Study ID	Country	Sprayer type	Sprayer category	Nozzle type	Nozzle number	Nozzle pressure (bar)	Spray quality	Forward speed (km/hr)	Spray volume applied (L/ha)	a.s.applied (g a.s./ha)	Spray conc. (g a.s./L)	Crop	Growth stage	Leaf cover	Crop height (m)	Row spacing (m)	Area sprayed (ha)	Duration of spraying (min)
ACI16-003	Spain	SAHER AT800	R	Albuz ATR (red) hollow cone 80° angle. Size 12 (1.2 mm). Nominal flow rate 1.92 L/min. 20cm spacing.	4	10	F	6.48	346	26.47	0.077	vineyard	BBCH 13	early	0.7	2.6	0.78	58
ACI16-005	Spain	Berthoud Fructair 2000	R	Albuz ATR (red) hollow cone 80° angle. Size 12 (1.2 mm). Nominal flow rate 1.92 L/min. 20cm spacing.	12	10	F	6.3	610	89.2	0.146	pome fruit	BBCH 53 to 55	early	2.9	3.8	1.44	56
CEMR-7089	Italy	Nobili Geo 2000	R	Albuz Red ATR-80 (20 cm spacing)	10	22.5	VF	4	1091	127	0.117	pome fruit	BBCH 91	late	4	4	1.1	43
CEMR-7090	Italy	Dragone	S	Albuz Brown ATR 80 (20 cm spacing)	8	15	VF	1.9	597	75	0.125	vineyard	BBCH 81	late	2.1	3	0.72	84
CEMR-7091	Spain	Balleste 2000T	R	Albuz Orange ATR-80 (25 cm spacing)	14	7.5	VF	4.7	962	96	0.1	pome fruit	BBCH 87	late	3.5	3.5	0.78	70
CEMR-7092	Spain	Caffini Eurotech	R	Albuz Red ATI 80-04 (18 cm spacing)	8	8	F	3.7	872	65	0.075	vineyard	BBCH 81	late	1.9	3	1.09	70
CEMR-7456	Italy	Nobili Geo 2000	R	Albuz ATR-80 (20 cm spacing)	10	22.5	VF	4	1055	97	0.092	pome fruit	BBCH 53	early	4	4	1.1	54
CEMR-7457	Italy	Dragone Athos 1000	R	Albuz Red ATR-80 (20 cm spacing)	4	9	F	1.6	150	11	0.075	vineyard	BBCH 13-15	early	2.1	2.5	1.2	75
CEMR-7458	Spain	Balleste 2000T	R	Albuz Orange ATR-80 (25 cm spacing)	14	5.5	VF	4.7	641	92	0.143	pome fruit	BBCH 53	early	3.5	3.5	0.78	55
CEMR-7459	Spain	Caffini Eurotech	R	Albus Red ATI 80-04 (18 cm spacing)	8	8	F	2.8	569	43	0.075	vineyard	BBCH 13	early	1.9	3	1.09	57
CEMR-7500	Poland	Dominiak OS1500	S	Teejet/Conejet Green TXB800 15 VK (25 cm spacing)	22	20	VF	8.9	605	121	0.2	pome fruit	BBCH 81	late	3.5	3.5	0.83	56
CEMR-7501	Poland	Wulkan 2000 S	S	Lechler Blue hollow cone TR 80-03 (TK 80-01C) (25 cm spacing)	16	4.5	VF	5.3	824	118	0.143	pome fruit	BBCH 81	late	3	3.5	0.84	49
CEMR-8025	Poland	Dominiak 05 1500 Dual rotor	S	Teejet/Conejet 11x Yellow TXB800 02VK; 11x Green TXB800 15 VK (25 cm spacing)	22	20	VF	5.3	530	96	0.182	pome fruit	BBCH 53-55	early	2.5	4	1	44
CEMR-8026	France	Calvet Pneumatic	S	Calvet 16/10 pastilles (1.6 mm diameter, flow rate 1.31 L/min). Nozzles at 50 and 150 cm above the ground.	8	2.4	unknown	3.5	140	50	0.359	vineyard	BBCH 13-15	early	0.6	2.5	1.59	55
CEMR-8027	France	Calvet Pneumatic	S	Calvet 10/10 pastilles (1mm diameter, flow rate 0.65 L/min). Nozzles at 50, 150, 180 and 220 cm above ground.	8	2	unknown	4.7	122	47	0.386	vineyard	BBCH 81	late	0.6	2.5	0.74	34
CEMR-8028	France	Pulveman S21	S	Albuz 10x ATR 80 Lilac at bottom & 10x ATR 80 Brown at top (25cm spacing). Top 2 nozzles switched off so 18 used.	20	10	VF	5.5	213	67	0.313	vineyard	BBCH 81	late	0.6	2.5	0.75	43

Appendix B: Meteorological data

Study ID	Country	Mean wind deviation from 0° position	Mean wind speed (m/s)	Mean air temperature (°C)	Radiation (W.m-2)	Rainfall (mm)	Humidity (%)
ACI16-003	Spain	-24	3.3	18	529	0	49
ACI16-005	Spain	15	3.0	14	355	0	86
CEMR-7089	Italy	-8	1.1	17	421	0	56
CEMR-7090	Italy	-7	1.1	27	not recorded	40	64
CEMR-7091	Spain	-20	1.0	32	not recorded	0	52
CEMR-7092	Spain	-38	1.4	32	908	0	32
CEMR-7456	Italy	50	1.8	15	690	0	33
CEMR-7457	Italy	-3	1.6	15	196	0	46
CEMR-7458	Spain	-24	1.9	3	not recorded	0	85
CEMR-7459	Spain	-21	1.3	13	763	0	70
CEMR-7500	Poland	10	1.4	18	780	0	44
CEMR-7501	Poland	6	2.8	18	415	0	67
CEMR-8025	Poland	11	3.3	2	not recorded	0	62
CEMR-8026	France	-3	2.0	15	not recorded	0	65
CEMR-8027	France	-3	1.7	24	2935	0	65
CEMR-8028	France	80	4.1	24	not recorded	0	54

Appendix C: Silsoe Spray Unit Report

Staff at Silsoe Spray Applications Unit Ltd, who have expertise in spray application equipment and techniques, spray drift measurement and bystander exposure assessment, have undertaken a technical review of the content of the field trial reports. They have assessed the conduct of the trials (from the perspective of the application technique) and the meteorological data to determine whether the exposure data is appropriate for consideration in future regulatory risk assessments. A copy of their report is provided below.



Silsoe Spray Applications Unit

COMMERCIAL – IN CONFIDENCE

CONTRACT REPORT

S0182

A review of trials data for the evaluation of bystander exposure from pesticide applications to orchards and grapevines

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13th February 2019

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SUMMARY

A review of trials data for the evaluation of bystander exposure from pesticide applications to orchards and grapevines

16 field trials were conducted in 2016 and 2017 at locations across Poland, Spain and Italy to measure dermal and inhalation exposure of bystanders to spray, and inhalation exposure to vapour, during and following a pesticide application.

The content of the trial reports has been reviewed by staff at Silsoe Spray Applications Unit Ltd, who have expertise in spray application equipment and techniques, spray drift measurement and bystander exposure assessment.

The conduct of the trials (from the perspective of the application technique) and the meteorological data have been assessed to determine whether the exposure data is appropriate for consideration in future regulatory risk assessments.

A database containing the measured exposures and the contextual information surrounding the experiment has been made available to us, and some of this data has been compared with other available data.

The existing exposure assessment (EFSA, 2014) is based on data produced by Lloyd *et al* (1987). Further work was undertaken by Butler Ellis *et al* (2014) to provide supplementary data. These two datasets are therefore available for comparison with the new trials being reviewed. The comparisons suggest that the possible exposures are higher than previously measured, when a wider range of sites, conditions and sprayers are included. A review of the contextual data has identified no clear reason why these exposure values could be an overestimate, although the aim of the protocol was to provide worst case data only, whereas this is not necessarily true of the older studies.

When separating out grapes from pome fruit, exposures from applications to grapes are clearly much lower than pome fruit, and exposures from full leaf pome fruit are clearly much lower than exposures from dormant pome fruit applications.

The current regulatory exposure model is captured in the EFSA exposure calculator, and the new data generated by these studies are compared with the regulatory values. There is also a more sophisticated model - the BROWSE model - which can be used to more accurately represent each individual trial, by taking account of wind speed, sprayer type, tree growth stage. The new dermal exposure data are compared with the values generated by these models.

Comparison with predictions from the BROWSE model shows that BROWSE gives a much narrower range of exposures than were measured, and there appears to be little or no correlation between predictions and measurement. However, the level of conservatism of BROWSE appears to be consistent with the new data.

If the EFSA calculator is adjusted to take account of applied volume correctly, then the current approach is slightly over conservative at 75% and slightly under-conservative at 95% when compared with the new data.

The new trials also include inhalation exposure data, but these have not been compared with any existing models or data because of lack of resources. This could be addressed in future work.

Few of the trials fully met both the requirements of the protocol and our own criteria for a 'good' trial. However, we do not believe that there are any reasons for recommending that any of the new data should be eliminated from further analysis to support developments in exposure assessment. It is important, though, that any such analysis is done with the full knowledge of the contextual data and the limitations of the experiment, as well as using relevant expertise, so that the data are not extrapolated inappropriately.

Contents

A review of trials data for the evaluation of bystander exposure from pesticide applications to orchards and grapevines

Clare Butler Ellis

1. Introduction

16 field trials were conducted in 2016 and 2017 at locations across Poland, Spain and Italy to measure dermal and inhalation exposure of bystanders to spray, and inhalation exposure to vapour, during and following a pesticide application.

The content of the trial reports has been reviewed by staff at Silsoe Spray Applications Unit Ltd, who have expertise in spray application equipment and techniques, spray drift measurement and bystander exposure assessment.

The conduct of the trials (from the perspective of the application technique) and the meteorological data have been assessed to determine whether the exposure data is appropriate for consideration in future regulatory risk assessments.

A database containing the measured exposures and the contextual information surrounding the experiment has been produced, and some of this data has been compared with other available data.

The body of bystander exposure data in the public domain is small compared with other spray drift data, and the component that is relevant to applications to orchards is smaller still. We are unaware of any data relevant to applications to vineyards.

The existing exposure assessment (EFSA, 2014) is based on data produced by Lloyd *et al* (1987). Further work was undertaken by Butler Ellis *et al* (2014) to provide supplementary data that could relate bystander exposure to airborne spray concentrations from applications to orchards, since there is significantly more data available relating to airborne spray that could be used as part of an exposure assessment model. These two datasets are therefore available for comparison with the new trials being reviewed.

Lloyd *et al* (1987) also attempted to measure inhaled spray, although most of the data was below the limit of quantification. We have therefore not compared their results with the new data.

The current regulatory exposure model is captured in the EFSA exposure calculator, and the new data generated by these studies can be compared with the regulatory values. There is also a more sophisticated model - the BROWSE model (Butler Ellis *et al*, 2017a) – which can be used to more accurately represent each individual trial, by taking account of wind speed, sprayer type, tree growth stage. The new dermal exposure data are compared with the values generated by these models, recognising that neither of them aim to predict accurately the actual exposures that might occur in practice, but to provide a realistic worst case estimate for an idealised scenario. We would expect, therefore, that both models would over-predict compared with the data, although previous study suggested that the BROWSE model underestimates exposures from applications to orchards (Butler Ellis *et al*, 2017b)

The EFSA calculator and the BROWSE model also predict inhalation exposure and vapour exposures. However, no comparisons of these have been undertaken because of limited availability of resources. Further investigations could be conducted to establish how well these models are able to estimate exposures to inhalation exposure through both routes.

2. Objectives

The objectives of this study were to:

1. Identify the data required for a database for resident and bystander exposures
2. Review the experimental reports for the 16 trials
3. Contribute to checking the contents of the database
4. Conduct an analysis of the data in the context of other available relevant exposure data

3. Review of trials

3.1 Experimental protocols

There is an International Standard for spray drift measurements (ISO 22866:2005(E)). This has been successful in conducting experiments to compare two different parameters, as it provides a methodology for minimising variability (which is challenging in field trials) and prevents some of the potential pitfalls. It has been the basis of most field experiments for determining spray drift reduction from engineering controls, for example. The standard was not cited in the experimental protocol as the basis for the trial, although it has clearly influenced the experimental design, either directly or indirectly. The standard was not designed with any input from the risk assessment community, however, and therefore it provides a very restricted experimental scenario, rather than representing the full range of possible exposures. It is potentially a 'highly worst case' for exposure, because of the need for an open, flat, unobstructed field site and a very narrow range of wind directions. It also has some difficulties in being applied to orchards (Llorens *et al*, 2016) which have been identified by the spray drift research community.

Expert knowledge on conducting good spray drift trials has made significant advances in recent years and it is recognised that the protocol has to reflect the purpose of the experiment, therefore a good protocol will not be a carbon copy of a previous, different, study. There are a number of areas where the protocol could have been improved to:

- Better reflect the purpose of the experiment. In order to do this, however, a clear definition of the scenario(s) to be simulated is required
- More accurately and usefully capture the contextual data (i.e. application technique, meteorology and site characteristics)

While it is tempting to go through the protocol listing its many deficiencies, we recognise that there is no perfect experiment and it is always easier to be wise after the event. Our main recommendation is therefore, in future trials, that spray drift expertise – which is plentiful across the EU – is involved in both designing the protocol and in conducting the trials. This would have ensured much better quality of data and a much greater reliability in extrapolating measured exposure values to other situations. In our view, this is far more important than 'GLP' since this quality standard can only increase the probability that the protocol is followed, not that the protocol is appropriate.

3.1 Weather conditions

There is a number of meteorological parameters that can influence spray drift, the most important being wind speed, which will determine how far droplets of a given size, initial velocity and release height will travel downwind.

Wind direction is also important in a spray drift trial, because it is necessary to ensure that the drifting spray plume passes over at least some spray drift collectors. The protocol was designed to ensure bystanders were immediately downwind of the treated area, with the mean wind direction over the duration of the experiment having a tolerance of ±30°. More discussion of this is included later.

Evaporation of the drifting droplets can also potentially change the behaviour of those droplets – a droplet which reduces in size will travel further downwind, will be more concentrated, but will have a lower collection efficiency and therefore might contribute more or less to real exposures. Data is very limited on the extent to which evaporation influences spray drift. However, on the assumption that it could be an important parameter, the measured variables which determine evaporation are temperature and humidity. These can be combined into a single parameter – wet bulb depression – which is usually the single driver of evaporation in spray drift models.

Temperature itself might have a small impact by influencing spray droplet size, particularly where cold water is sprayed into warm air (Miller and Tuck, 2005), but this is unlikely to have a noticeable effect.

The measured temperatures and relative humidities were therefore used to determine the wet bulb depression (wbd) for each trial.

The range of wind speeds, wind directions and wet bulb depressions is given in Table 1. The wind speeds and direction are taken from the original data relating to the time period for spraying the first three rows. Mean wind direction was incorrectly calculated in several of the reports.

There is no information to establish whether this distribution of conditions is representative or typical of real conditions when spraying takes place across Europe.

	Temp, C	Wet Bulb Depression, C	Absolute deviation from sample line, deg	Wind speed, m/s
Mean	17.9	5.2	24	1.90
median	17.2	5.0	26	1.83
75th percentile	23.9	6.4	33	2.46
95th percentile	32.0	8.9	47	3.37
Minimum	2.2	1.0	1	0.30
Maximum	32.2	12.2	56	3.47

One trial, CEMR-7090, had a wind speed that was very low (0.3 m/s) which also resulted in a very variable wind direction. This was incorrectly reported. CEMR-7456 had a mean deviation of 56 degrees from the sample line, which suggests that the bystanders were not placed in an appropriate location to receive the highest level of exposures from that application.

3.2 Site conditions

The site itself can influence the meteorological conditions by affecting the turbulence. This is particularly important for determining dispersion of a spray plume. A high level of dispersion will cause the spray plume to spread faster and to a greater height. This can lead to higher environmental exposures at greater distances, but potentially for bystanders, can reduce exposures by reducing the airborne concentration.

Turbulence is affected by upwind terrain and by the crop and the downwind vegetation. The greater the height of the 'surface roughness', the greater the turbulence. Thus taller trees in an orchard will probably result in greater levels of dispersion than shorter grapevines. However, the level of dispersion cannot be determined from a single measurement of wind speed using a cup anemometer. A good drift experiment will use either an ultrasonic anemometer with three dimensions, or wind measurements at two heights or more.

There were no indications of the upwind terrain in the reports – unfortunately no photos were taken from the perspective of the bystanders, looking upwind beyond the crop.

The area downwind of the crop, where measurements were made, is in the lee of the crop, which as a rule of thumb remains for a distance of roughly 10 times the height of the crop. So for 2 m tall trees, the wind conditions will not settle to a typical atmospheric flow profile until 20 m downwind. The wind conditions in the 'bystander measurement zone' therefore are (a) rather unpredictable, and (b) strongly influenced by the crop itself.

While the standard for drift measurements should not be adhered to rigidly, there are useful guidelines in it which would have benefited the protocol:

Measurements shall be made at a downwind distance of at least four crop heights from the downwind edge of the sprayed area where appropriate. Measurements shall be at a height 1 m above the canopy and at least 2 m above the ground and at a frequency of least 0,1 Hz sampling rate.

We note that all wind measurements were made at 2 m, independent of the height of the crop, so the standard was unlikely to have been met. The location was only reported in two cases and appears to be (a) too close to the crop, and (b) near the edge of the crop where edge-effects could influence the measurement. For the other reports, supplementary data (source??) has indicated that the measurement was made at a considerable distance from the downwind measurement area (hundreds of metres) and could be within the crop, or within an adjacent crop, or somewhere else entirely. This is not good experimental practice and provides data which is of very limited value in understanding the influence of meteorology on exposure. Insufficient detail was given in the protocol, which only specified 'on-site'.

The dimensions of the site are also important. According to the standard, the length of each treated row needs to be great enough to ensure that the collectors are in the path of the spray plume even if the wind fluctuates by up to 45^0 and is not affected by what happens at the edge of each row. This ensures that each bystander represents a 'replicate' measurement, and variability is mostly due to the variability of the wind characteristics. The minimum length of row to be treated should be length of the row of collectors parallel to the tree row (= 50 m) plus the maximum distance downwind at each end, i.e. 80 m minimum. Not all of the sites achieved this, and therefore some of the variability in reported exposures will also have been caused by the shape of the treated area.

An alternative approach to the standard, appropriate for exposure assessment, would be to place collectors across the full length of each side of the field, including at the corners, to evaluate the full range of exposures for a wide range of field sizes and shapes. We assume this was not the intention of these studies, which will therefore result in a narrower range of worst-case exposures.

Similarly, the quantity of spray to which each collector is exposed is dependent on the integral over subsequent upwind passes, i.e. the number of rows sprayed, and this was very low in some cases. The protocol suggested a treated area of 100 m x 100 m, but this was not achieved in a number of cases and would therefore have affected the measured exposures. However, the first few rows are responsible for the majority of the exposure to spray, and therefore exposures are less likely to be sensitive to this parameter providing it is above a particular value (although it is not certain what this value is).

The upwind fetch over the treated area is even more important in determining the airborne concentration of vapours, and therefore an approximately square plot is necessary if all vapour samplers are to be considered equal.

Thus the area of the crop treated is not a relevant parameter itself, it is the two dimensions which are important for different, independent reasons.

Sites used in 7091, 7501 and 8027 had a short upwind fetch. Sites used in 7092, 7459 and 8028 had particularly short rows.

4. Experimental conduct

Applied volume rate is an important parameter for defining the applied dose rate, and therefore needs to be determined with some degree of accuracy and reliability. Volume rate varies over the treated area due to changes in speed – and this is the case even when the sprayer has a rate controller that adjusts pressure to compensate for changes in speed. No mention of rate controllers on the sprayers was made in the reports, but they are less common on airblast sprayers than on boom sprayers. Often for spray trials they would be disabled because we require operating pressure to be constant. Usually, the relevant volume rate relates to the main part of the application - i.e. while operating at the desired speed and pressure in a straight line, so would not include changes that might result from slowing down at the end of rows, switching off early or late at the end of rows, priming the pump and spray lines, which we could term 'edge effects'. The methodology specified in the protocol does not provide the necessary data as it includes these edge effects. It also relies on accurate measurement of the treated area and accurate measurement of the used liquid volume which is difficult with an unknown residual volume in the sprayer. Thus the volume measurements conducted in the trials do not accurately reflect the volume applied that we need for subsequent extrapolation of the data to other doses and volumes.

The calibration of the sprayer is the best method of determining the applied volume. Calibration of sprayer is required by good application practice and so it was surprising to see in the protocol that this was explicitly excluded. The ISO drift standard specifies tha*t nozzle type, operating pressure, **measured flow rate** and application rate in the directly sprayed area* are reported.

Calibration requires that the flow rate is measured at the set operating pressure and actual sprayer speed is measured during the central part of each row (in this case, the 80 m required in the experimental protocol). As this was not done, we have attempted to determine the applied volume from the details of nozzles, pressures and speeds provided but in many cases these are significantly

different from that reported (Table 2). Where the reported applied volume was slightly higher than our estimate, this was clearly because of the 'edge effects' and we can be confident that the estimate was a reasonable one. Where the report suggests a much lower volume than we have calculated, we have been unable to identify conclusively the cause. One suggestion was that the speed was inaccurately recorded, or only recorded for the first three rows and the speed then changed for the remaining rows. Another cause could have been inaccurate reporting of the sprayer flow rate, due to incorrect nozzles or pressures, or worn nozzles. Much of the information about the application was not provided as part of the main report, but in supplementary data, which has not been made widely available or subjected to the same quality checks. In a number of cases, the quality of the description of equipment was poor. This could potentially have been mitigated by clearer requirements in the protocol and a greater level of independent application expertise present for the trial.

Table 2 Reported and calculated application volumes from each trial

Trial	Water volume reported, L/ha	Water volume calculated by SSAU, L/ha	% difference
AC116-003	346	274	21%
AC116-005	610	577	5%
CEMR-7089	1091	1069	2%
CEMR-7090	597	691	-16%
CEMR-7091	962	633	34%
CEMR-7092	872	1129	-29%
CEMR-7456	1055	1069	-1%
CEMR-7457	150	1093	-629%
CEMR-7458	641	551	14%
CEMR-7459	569	1491	-162%
CEMR-7500	605	636	-5%
CEMR-7501	824	792	4%
CEMR-8025	530	716	-35%
CEMR-8026	140	718	-413%
CEMR-8027	122	266	-118%
CEMR-8028	213	452	-112%

Light green: A small difference - some or all of which could be explained by inaccuracies in vol measurement; could be regarded as a good estimate of application volume.

Dark green: A large difference - can't be explained by inaccuracies in vol measurement alone, but could be explained by variable speeds or the point at which spray stopped on turns

Red: A large difference that probably only be explained by different application conditions from that reported.

The methodology for measurement of sprayed volume was not explicitly specified in the protocol but relied on what was available on the sprayer and is likely to have only modest levels of accuracy. We have made some attempts to estimate the level of accuracy, based on measured quantities of residual volume (Debaer *et al*, 2008) and possible errors in sight gauges and flowmeters and these showed that measurement inaccuracies are highest for the lowest volume applications, but are unlikely to be

the cause of large differences between the applied volume reported and the applied volume calculated from application parameters.

There are four trials (CEMR-7089, 7500, 8026, 8028) which apparently had no liquid left in the sprayer at the end. This suggests that the sprayer ran out of liquid exactly at the point the operator would have switched it off for the last time. This would have been quite a coincidence to have happened once (and rather unnerving for those conducting the trial) but not really plausible that it happened four times. It is more likely that there was already a significant amount of volume in the sprayer before loading.

Once the volume is correctly quantified, the dose can be determined from the concentration of active substance in the spray liquid and the applied volume. Unfortunately, although tank samples were obtained, these were not analysed. The actual concentration of active substance cannot be determined with any accuracy from the quantities mixed of water and product because of the poor accuracy of measurement of water volume, and the unknown residual volume left in the sprayer (Debaer *et al*, 2008). The ISO standard requires that: *deposits on collectors or samplers should be calculated based on the calibration of the tracing technique, with* ***samples of the spray liquid taken from a nozzle at the time of the spraying.***

4. Comparison with other data

Existing published bystander exposure data relates to the quantity of spray liquid, measured using a tracer, rather then the quantity of active substance. Therefore the comparisons we have undertaken have been with spray liquid exposure, with new values taken from the database.

In order to compare field data, some normalisation is necessary. For a given set of application conditions, downwind exposures to spray liquid will be proportional to the applied volume. This has not always been recognised in regulatory risk assessments and can be difficult to identify from field data because volume cannot be changed without changing other parameters. However, it is based on the fundamental scientific principles and it would be perverse not to accept this. Therefore when data are compared, we consider only normalised values for exposure to spray liquid, i.e. we scale all data to 100 L/ha by dividing by the volume applied and multiplying by 100. For the following analysis, we have taken the value of volume given in the reports, rather than the volume we have estimated from application conditions.

4.1 Lloyd *et al*, 1987

Lloyd *et al* used three different spraying systems, but only one of these has been used to define the regulatory exposures because the other two would not be considered typical of orchard applications. The data on which the regulatory exposure is based is for an application of 470 L/ha.

Measurements were made at distances 8 and 50 m from outside line of tree trunks. The new studies measured from one half-row width beyond the centre of the last row, and therefore varied from trial to trial, depending on the row width. Distances were 5, 10 and 15 m. We therefore have no common distances for comparison, but the actual distance from the outside line of the trunks in these studies, which ranged from 6.25 to 7 m is close enough to 8 m to compare with Lloyd *et al*.

Although it is not completely clear from the Lloyd *et al* report, there is an indication that bystanders were sometimes placed on two sides of the treated area because of the fluctuating wind, and while wind directions were not accurately recorded, handwritten diagrams show that in some experiments, the wind was perpendicular to the rows, in some along the rows and in others at 45° to the rows.

Where the bystander was placed is not clear, other than 'downwind'. In some cases, it appears that the bystander would also be downwind of a windbreak.

The area treated was 110 x 80 m, similar to the proposed size for the new studies, but actually larger than many of them. Row width was 5 m and trees were 2.6 m tall. No details of when the experiments were conducted, nor the growth stage of the crop, were given, but met data suggest summer in the UK (sunshine and temperatures around 20 degrees), so full leaf is probable.

A comparison between the two datasets is given in Fig 1. The new data have a similar median value for pome fruit, but a much higher 75th percentile. This is not surprising because the Lloyd *et al* data were obtained on the same field each time with the same sprayer operating under the same conditions, so that only the meteorology was different

However, when split into dormant and full leaf, the new dormant data are much higher and full leaf are significantly lower. Full leaf data in new studies were undertaken at lower wind speeds (median wind speed 1.2 m/s, compared with 2.4 m/s for dormant; Lloyd *et al* median windspeed was 2.0 m/s). This lower windspeed, which might be typical of conditions during this part of the season, is likely to result in lower exposures.

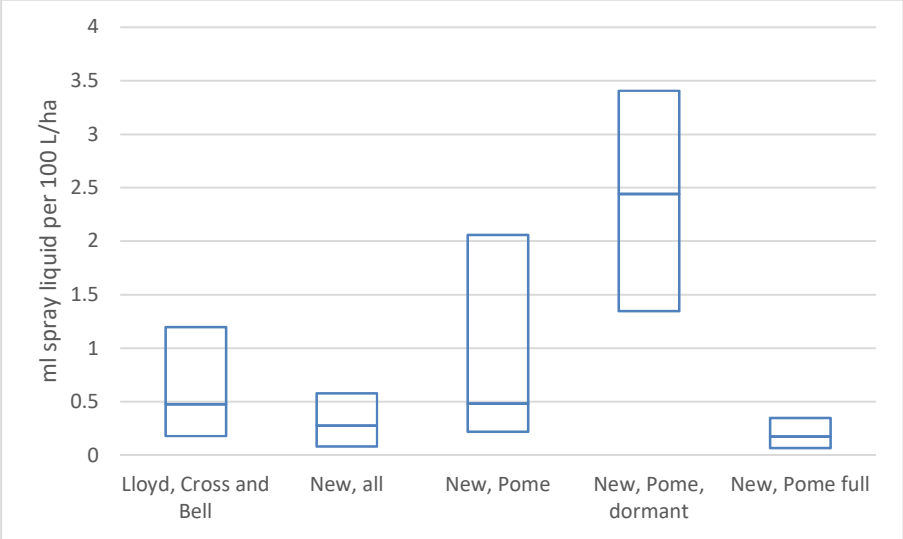

Figure 1. Comparison of new data with that for Lloyd *et al* (1987) Boxes indicate median, 25th and 75th percentiles of normalised data.

4.2 Butler Ellis *et al* (2014)

The data obtained by Butler Ellis *et al* (2014) was based on a similar sprayer to that used by Lloyd *et al* (1987), and spraying 5-6 spray passes (i.e. 5-7 tree rows) The purpose of these trials was not to provide exposure data *per se*, but to provide information relevant to modelling exposures, in particular the relationship between airborne spray concentrations and bystander exposure.

It can be seen that the new data are much higher than Butler Ellis *et al*. Butler Ellis *et al* used the same field and sprayer for all experiments, so it would be expected that the variability would be greater when undertaken across a range of sites and with different spraying operations. It was also a relatively small number of rows that were sprayed, which might also account for a lower level of exposure.

However, the difference is surprising, given that nominal wind conditions are similar. Separating out the early season trials shows this very strongly (Figure 3). Butler Ellis *et al* had insufficient data obtained at full leaf to enable a comparison between these data subsets.

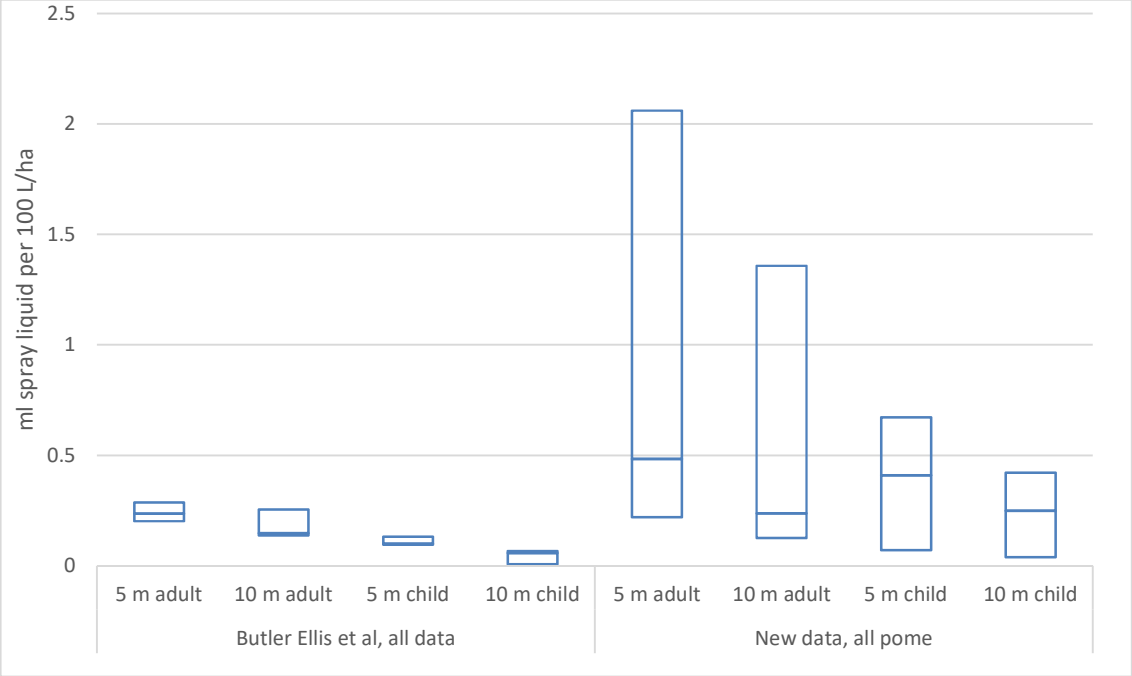

Figure 2. Median, 25th and 75th percentiles of normalised data for all pome data: comparison between new data and Butler Ellis *et al* (2014)

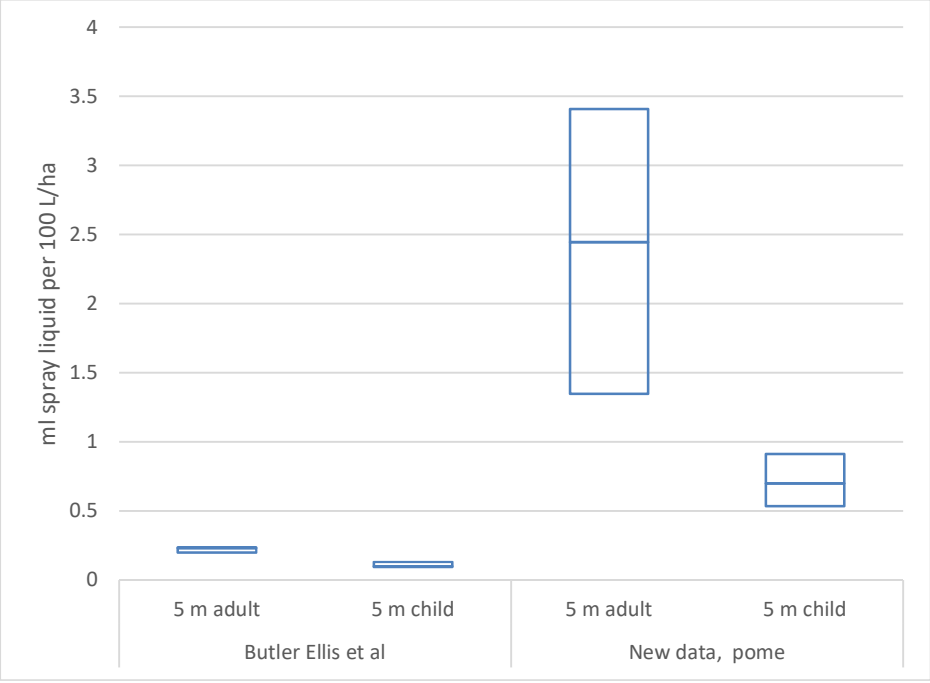

Figure 3. Median, 25th and 75th percentiles of data obtained early season: comparison between new data and Butler Ellis *et al* (2014).

4.3 Discussion of comparisons with existing data

The comparisons between these data and existing data suggest that the possible exposures are higher than previously measured, when a wider range of sites, conditions and sprayers are included. A review of the contextual data has identified no clear reason why these exposure values could be an overestimate, although the aim of the protocol was to provide worst case data only, whereas this is not necessarily true of the older studies.

When separating out grapes from pome fruit, exposures from applications to grapes are clearly much lower than pome fruit, and exposures from full leaf pome fruit are clearly much lower than exposures from dormant pome fruit applications. In fact there are three trials with dormant pome fruit which give very high values of exposure; the fourth, with much lower exposure, had a mean wind direction of greater than 45° to the sample line.

5. Comparison with models

5.1 BROWSE model

The BROWSE model was developed to provide a more detailed, realistic and flexible exposure assessment, which could potentially provide a higher-tier option. It can be used to specify wind speed, sprayer type (cross-flow or axial fan), crop growth stage (dormant, transition, full leaf) and application volume, and is based on a spray quality of 'very fine'. The model was run to predict the quantity of spray liquid deposited on a bystander at 5, 10 and 15 m downwind. A previous exercise (Butler Ellis *et al*, 2017) suggested that the BROWSE model underestimated field measurements.

Figure 4 shows the relationship between measured values, and the predicted values (median, 75th and 95th percentiles) from BROWSE. It can be seen that BROWSE predicts a much narrower range of exposures than were measured, and there appears to be little or no correlation between predictions and measurement. It is probable that the spray drift data on which BROWSE is based was obtained under a narrower set of conditions, possibly more closely aligned to the ISO drift standard. However, the level of conservatism of BROWSE appears to be about right, with 49% of new measurements > predicted median, 18% of measurements > 75th percentile and none > 95th percentile.

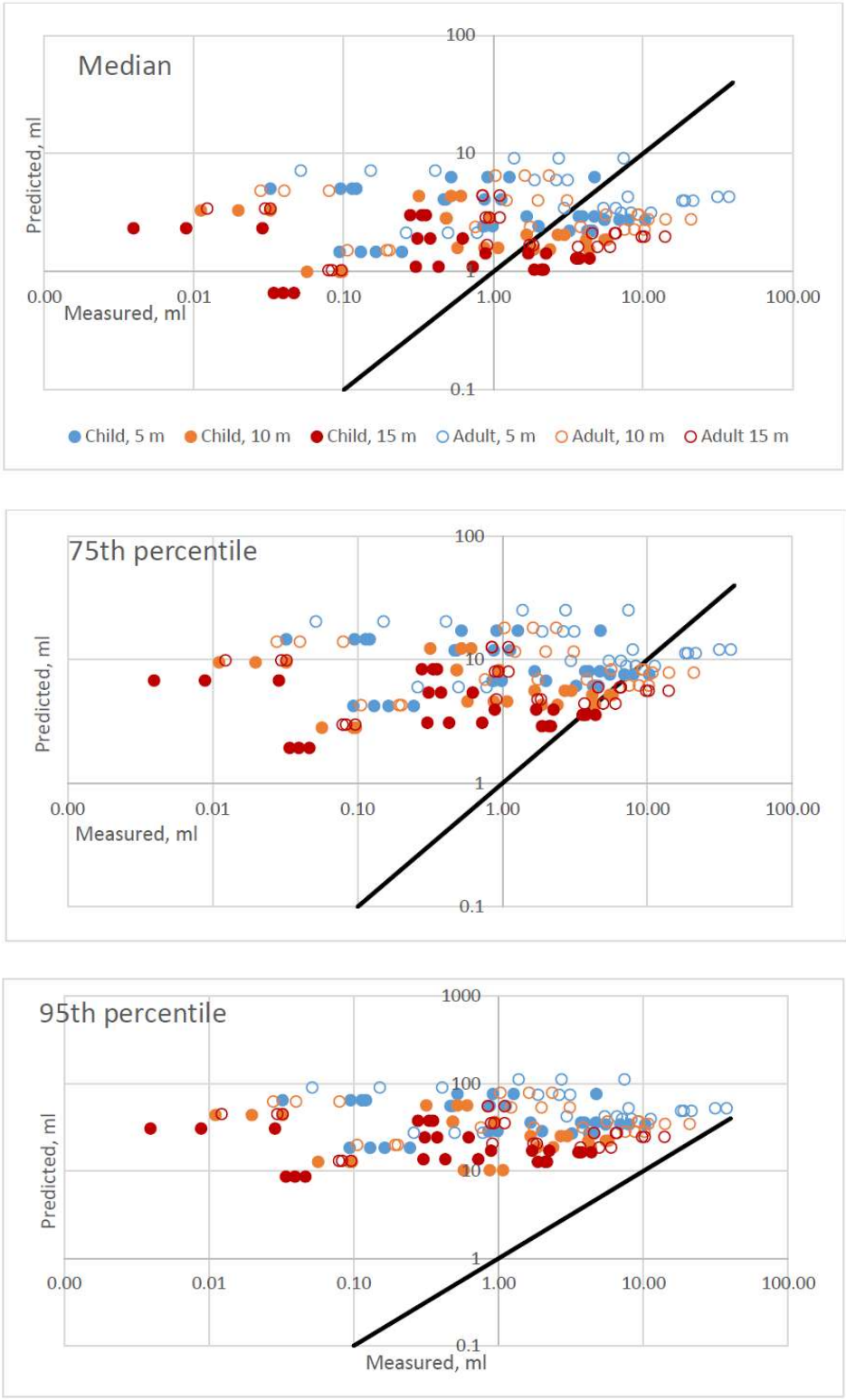

Figure 4. Comparison between BROWSE predicted values (median, 75th percentile and 95th percentile) and each individual value of potential bystander exposure, ml. The black line indicates perfect correlation.

5.2 Comparison with current regulatory assessment – the EFSA calculator

The EFSA calculator uses the same value of quantity of spray liquid for all scenarios, including both grapes and pome fruit, with differences between children and adults, and resident (75th percentile) and bystanders (95th percentile). These values are compared with the 75th and 95th percentiles for all the new data, grapes and pome, at 5 m downwind.

Table 3. Comparison between new exposure data and EFSA calculator, ml per person

	Adult		Child	
	New data	EFSA	New data	EFSA
75th	9.02	5.63	4.31	1.69
95th	29.88	12.90	7.81	3.87

This suggests that the existing exposure assessment might not be sufficiently protective, particularly for early season applications to pome fruit where exposures are highest. This would be expected for the high volume applications (>470 L/ha) since the current exposure assessment does not take into account the applied volume. If we normalise all the data to 100 L/ha (including the EFSA values, which are based on trials at 470 L/ha), then the EFSA calculator is slightly over conservative at 75% and slightly under-conservative at 95%.

Table 4. Comparison between normalised values of new exposure data and EFSA calculator, ml per person per 100 L/ha applied

	Adult		Child	
	New data	EFSA	New data	EFSA
75th	0.60	0.78	0.17	0.24
95th	3.37	2.74	0.91	0.82

6. Use of new data in regulatory exposure assessment

The availability of new data for regulatory exposure assessment is to be welcomed, because of the shortage of empirical data that has been available up until now. The technical quality of the data from each trial, compared with what was anticipated from the requirements in the protocol is summarised in Table 5. It can be seen that only five trials actually fully met the requirements of the protocol (and we have relaxed the wind direction requirement to ± 45° to achieve this), and only six had a reported applied volume that was consistent with the reported application equipment and conditions of use. Only two trials achieved both of these.

It should be emphasised that, while some of the problems which occurred could have been reduced or eliminated, the challenge which is posed by conducting such trials should not be underestimated, and these data should not be discarded solely because they did not meet some of the criteria. We recognise that the data which was used to develop the existing exposure assessment would not stand up any better to the scrutiny we have given these new trials, and would probably be worse in some cases.

Table 5. Comparison of trials against criteria. Two ticks means meets the criterion fully; one tick indicates only a slight shortfall

Trial number	Comment	Complies with protocol	Application practice validated by vol calculation
AC116-003	Slightly short row length	✓	✓
AC116-005	OK	✓✓	✓✓
CEMR - 7089	Average wind direction > 30°	✓✓	✓✓
CEMR - 7090	Slightly short row length; Volume suggests application inaccurately reported; very low wind speed		✓
CEMR-7091	Short upwind fetch. Volume suggests application inaccurately reported	✓	
CEMR-7092	Very short row length; Volume suggests application inaccurately reported		
CEMR-7456	Average wind direction > 45°		✓✓
CEMR-7457	Average wind direction > 30°; Volume suggests application inaccurately reported	✓✓	
CEMR-7458	Average wind direction > 30°; short upwind fetch	✓	✓✓
CEMR-7459	Volume suggests application inaccurately reported; very short row		
CEMR-7500	Very short upwind fetch		✓✓
CEMR 7501	Short upwind fetch	✓	✓✓
CEMR 8025	Volume suggests application inaccurately reported	✓✓	
CEMR 8026	Volume suggests application inaccurately reported	✓✓	
CEMR 8027	Volume suggests application inaccurately reported; slightly short upwind fetch	✓	
CEMR 8028	Volume suggests application inaccurately reported; very short row length.		

In particular, we do not believe that insisting on an average wind direction within 30 degrees of the sample line is necessary, as the wind rarely behaves that predictably and this provides an unrealistic constraint on the data. Any wind within ± 45° of the sample line will give realistic exposures which are highest on the side of the field where the drift samplers are located.

For example, for a treated field surrounded by bystanders, those within ±30° of directly downwind would represent the 83rd percentile. If we further take the 95th percentile of these data, we are probably ending up with something closer to the 99.99th percentile of possible exposures. If we recognise that any wind direction ± 45° is acceptable, this provides the 75th percentile of exposures around the whole field. Taking the 80th percentile of these data will lead to the 95th percentile of all exposures.

However, if we wish to better understand the full range of possible exposures, a different experimental protocol would be required, which would also include wind directions along the rows, with bystanders at a wider range of locations.

We do not believe that there are any reasons for recommending that any of the new data should be eliminated from further analysis to support developments in exposure assessment. However, it is important that any such analysis is done with the full knowledge of the contextual data and the limitations of the experiment, so that it is not extrapolated inappropriately.

7. Conclusions

Sixteen field trials have been reviewed and the protocol and experimental conduct have been assessed for their suitability for informing regulatory exposure assessment. The exposure data produced by these trials have been compared with existing data and model predictions.

The comparisons between these data and existing data suggest that the possible exposures are higher than previously measured, when a wider range of sites, conditions and sprayers are included. A review of the contextual data has identified no clear reason why these exposure values could be an overestimate, although the aim of the protocol was to provide worst case data only, whereas this is not necessarily true of the older studies.

When separating out grapes from pome fruit, exposures from applications to grapes are clearly much lower than pome fruit, and exposures from full leaf pome fruit are clearly much lower than exposures from dormant pome fruit applications.

Comparison with predictions from the BROWSE model shows that BROWSE gives a much narrower range of exposures than were measured, and there appears to be little or no correlation between predictions and measurement. However, the level of conservatism of BROWSE appears to be consistent with the new data.

If the EFSA calculator is adjusted to take account of applied volume correctly, then the current approach is slightly over conservative at 75% and slightly under-conservative at 95% when compared with the new data.

Few of the trials fully met both the requirements of the protocol and our own criteria for a ‘good’ trial. However, we do not believe that there are any reasons for recommending that any of the new data should be eliminated from further analysis to support developments in exposure assessment. It is important that any such analysis is done with the full knowledge of the contextual data and the limitations of the experiment, and using relevant expertise, so that it is not extrapolated inappropriately.

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Appendix D: Exposure data

The database consists of sixteen studies, spread across four scenarios: pome and vine crops in early and late growth stages. Each study measured exposures for three sets of adult and child mannequins at three distances downwind of the sprayed crop to provide exposure records comprising potential dermal, actual dermal and inhalation exposure for 18 "individuals". So, each scenario was supported by 72 individual exposure records and the total the database provides an overall total of 288 records. The contributions to the database, i.e. number of records associated with each scenario, are shown in Tables D1-4.

Table D1: Pome early scenario: summary of numbers of exposure records showing country location of studies, sprayer types, spray quality, and distributions of selected application, environmental, and exposure variables

Country	Sprayer category	Spray quality	Spray volume (l/ha)	Amount applied (g a.s.)	Area sprayed (ha)	Wind deviation from sample line (°)	Mean wind speed (m/s)	Mean air temp. (°C)	Wet Bulb Depression (°C)
France: 0	Radial: 54	Fine: 18	Min.: 530.0	Min.: 89.00	Min.: 0.780	Min.: -56.00	Min.: 1.450	Min.: 2.20	Min.: 1.400
Italy: 18	Sideways: 18	unknown: 0	1st Qu.: 590.0	1st Qu.: 91.25	1st Qu.: 0.945	1st Qu.: -29.00	1st Qu.: 1.742	1st Qu.: 2.95	1st Qu.: 1.475
Poland: 18		Very Fine: 54	Median: 625.5	Median: 94.00	Median: 1.050	Median: 6.00	Median: 2.405	Median: 8.40	Median: 2.300
Spain: 36			Mean: 709.0	Mean: 93.50	Mean: 1.080	Mean: -2.75	Mean: 2.433	Mean: 8.45	Mean:3.400
			3rd Qu.: 744.5	3rd Qu.:96.25	3rd Qu.: 1.185	3rd Qu.: 32.25	3rd Qu.: 3.095	3rd Qu.: 13.90	3rd Qu.: 4.225
			Max.: 1055.0	Max.: 97.00	Max.: 1.440	Max.: 33.00	Max.: 3.470	Max.:14.80	Max.:7.600

	Adults			Children			Adults			Children		
	PDE (ml spray)	ADE (ml spray)	PIE (ml spray)	PDE (ml spray)	ADE (ml spray)	PIE (ml spray)	PDE (µg active subst.)	ADE (µg active subst.)	PIE (µg active subst.)	PDE (µg active subst.)	ADE (µg active subst.)	PIE (µg active subst.)
Minimum	0.840	0.570	0.0014	0.280	0.170	0.000009	77.620	52.880	0.01	25.590	15.780	0.0001
1st Quartile	4.345	2.615	0.0027	1.570	0.880	0.00905	665.055	418.053	0.02125	210.368	122.283	0.0654
Median	8.090	4.215	0.00855	2.835	1.425	0.0146	1240.690	699.410	0.0505	430.450	217.125	0.1135
Mean	9.061	4.671	0.011758	2.958	1.511	0.017014	1347.394	694.505	0.084447	440.399	225.421	0.152461
3rd Quartile	10.453	6.070	0.01575	4.258	2.165	0.023275	1738.760	1033.598	0.11	606.625	316.003	0.22525
Maximum	37.520	11.280	0.0611	8.760	4.080	0.046	5365.440	1647.170	0.437	1279.340	595.120	0.424

Table D2: Pome late scenario: summary of numbers of exposure records showing country location of studies, sprayer types, spray quality, and distributions of selected application, environmental, and exposure variables

Country	Sprayer category	Spray quality	Spray volume (l/ha)	Amount applied (g a.s.)	Area sprayed (ha)	Wind deviation from sample line (°)	Mean wind speed (m/s)	Mean air temp. (°C)	Wet Bulb Depression (°C)
France: 0	Radial: 36	Fine: 0	Min.: 605.0	Min.: 96.0	Min.: 0.7800	Min.: -7.00	Min.: 1.100	Min.: 16.70	Min.: 4.000
Italy: 18	Sideways: 36	unknown: 0	1st Qu.: 769.2	1st Qu.: 112.5	1st Qu.: 0.8175	1st Qu.: 1.25	1st Qu.: 1.167	1st Qu.: 17.90	1st Qu.: 4.900
Poland: 36		Very Fine: 72	Median: 893.0	Median: 119.5	Median: 0.8350	Median: 15.00	Median: 1.245	Median: 18.35	Median: 6.050
Spain: 18			Mean: 870.5	Mean: 115.5	Mean: 0.8875	Mean: 14.25	Mean: 1.485	Mean: 21.40	Mean: 5.925
			3rd Qu.: 994.2	3rd Qu.: 122.5	3rd Qu.: 0.9050	3rd Qu.: 28.00	3rd Qu.: 1.562	3rd Qu.: 21.85	3rd Qu.: 7.075
			Max.: 1091.0	Max.: 127.0	Max.: 1.1000	Max.: 34.00	Max.: 2.350	Max.: 32.20	Max.: 7.600

	Adults			Children			Adults			Children		
	PDE (ml spray)	ADE (ml spray)	PIE (ml spray)	PDE (ml spray)	ADE (ml spray)	PIE (ml spray)	PDE (µg active substance)	ADE (µg active substance)	PIE (µg active substance)	PDE (µg active substance)	ADE (µg active substance)	PIE (µg active substance)
Minimum	0.010	0.010	0.0017	0.010	0.004	0.0011	1.530	0.890	0.01	0.660	0.430	0.01
1st Quartile	0.095	0.030	0.00485	0.058	0.020	0.004425	17.423	6.183	0.04	10.710	3.930	0.051
Medium	0.770	0.375	0.0094	0.270	0.125	0.00705	96.450	50.540	0.052	35.250	16.740	0.0625
Mean	1.271	0.691	0.008757	0.412	0.203	0.007914	164.581	89.468	0.059314	53.292	25.968	0.064333
3rd Quartile	1.850	1.070	0.0112	0.648	0.325	0.0105	249.608	134.345	0.0775	83.370	40.765	0.07525
Maximum	6.560	3.480	0.0193	1.990	0.940	0.0173	937.520	497.950	0.193	285.130	134.110	0.156

Table D3: Vine early scenario: summary of numbers of exposure records showing country location of studies, sprayer types, spray quality, and distributions of selected application, environmental, and exposure variables

Country	Sprayer category	Spray quality	Spray volume (l/ha)	Amount applied (g a.s.)	Area sprayed (ha)	Wind deviation from sample line (°)	Mean wind speed (m/s)	Mean air temp. (°C)	Wet Bulb Depression (°C)
France: 18	Radial: 54	Fine: 54	Min.: 140.0	Min.: 11.00	Min.: 0.780	Min.: -13.0	Min.: 1.370	Min.: 12.60	Min.: 3.200
Italy: 18	Sideways: 18	unknown: 18	1st Qu.: 147.5	1st Qu.: 22.25	1st Qu.: 1.012	1st Qu.: -2.5	1st Qu.: 1.812	1st Qu.: 14.40	1st Qu.: 3.725
Poland: 0		Very Fine: 0	Median: 248.0	Median: 34.50	Median: 1.145	Median: 7.5	Median: 2.040	Median: 15.85	Median: 5.000
Spain: 36			Mean: 301.2	Mean: 32.50	Mean: 1.165	Mean: 10.0	Mean: 2.197	Mean: 15.50	Mean: 4.850
			3rd Qu.: 401.8	3rd Qu.: 44.75	3rd Qu.: 1.298	3rd Qu.: 20.0	3rd Qu.: 2.425	3rd Qu.: 16.95	3rd Qu.: 6.125
			Max.: 569.0	Max.: 50.00	Max.: 1.590	Max.: 38.0	Max.: 3.340	Max.: 17.70	Max.: 6.200

	Adults			Children			Adults			Children		
	PDE (ml spray)	ADE (ml spray)	PIE (ml spray)	PDE (ml spray)	ADE (ml spray)	PIE (ml spray)	PDE (µg active substance)	ADE (µg active substance)	PIE (µg active substance)	PDE (µg active substance)	ADE (µg active substance)	PIE (µg active substance)
Minimum	0.020	0.010	0.0022	0.010	0.010	0.0019	1.220	0.990	0.0088	0.700	0.500	0.0088
1st Quartile	0.128	0.080	0.0032	0.050	0.038	0.002775	19.008	14.408	0.0125	7.400	5.833	0.0135
Medium	0.270	0.210	0.00345	0.145	0.105	0.0039	35.220	26.715	0.01675	18.025	11.215	0.0215
Mean	0.546	0.402	0.004139	0.267	0.198	0.004733	56.540	39.923	0.029553	27.248	19.621	0.040897
3rd Quartile	0.555	0.413	0.005225	0.290	0.213	0.00505	76.768	49.643	0.03675	34.758	26.203	0.0655
Maximum	3.020	2.190	0.0084	1.360	1.030	0.0211	232.630	168.660	0.112	102.170	77.110	0.153

Table D4: Vine late scenario: summary of numbers of exposure records showing country location of studies, sprayer types, spray quality, and distributions of selected application, environmental, and exposure variables

Country	Sprayer category	Spray quality	Spray volume (l/ha)	Amount applied (g a.s.)	Area sprayed (ha)	Wind deviation from sample line (°)	Mean wind speed (m/s)	Mean air temp. (°C)	Wet Bulb Depression (°C)
France: 36	Radial: 18	Fine: 18	Min.: 122.0	Min.: 47.0	Min.: 0.720	Min.: 11.00	Min.: 0.3000	Min.: 23.80	Min.: 4.70
Italy: 18	Sideways: 54	unknown: 18	1st Qu.: 190.2	1st Qu.: 60.5	1st Qu.: 0.735	1st Qu.: 22.25	1st Qu.: 0.9675	1st Qu.: 23.95	1st Qu.: 5.00
Poland: 0		Very Fine: 36	Median: 405.0	Median: 66.0	Median: 0.745	Median: 26.50	Median: 1.5000	Median: 25.35	Median: 5.70
Spain: 18			Mean: 451.0	Mean: 63.5	Mean: 0.825	Mean: 27.00	Mean: 1.5200	Mean: 26.60	Mean: 6.95
			3rd Qu.: 665.8	3rd Qu.: 69.0	3rd Qu.: 0.835	3rd Qu.: 31.25	3rd Qu.: 2.0525	3rd Qu.: 28.00	3rd Qu.: 7.65
			Max.: 872.0	Max.: 75.0	Max.: 1.090	Max.: 44.00	Max.: 2.7800	Max.: 31.90	Max.: 11.70

	Adults			Children			Adults			Children		
	PDE (ml spray)	ADE (ml spray)	PIE (ml spray)	PDE (ml spray)	ADE (ml spray)	PIE (ml spray)	PDE (µg active substance)	ADE (µg active substance)	PIE (µg active substance)	PDE (µg active substance)	ADE (µg active substance)	PIE (µg active substance)
Minimum	0.010	0.010	0.0011	0.010	0.004	0.0009	5.110	2.000	0.007	2.140	1.500	0.007
1st Quartile	0.068	0.040	0.004475	0.038	0.020	0.002875	9.898	5.808	0.054125	4.765	2.778	0.054975
Medium	0.195	0.115	0.0065	0.095	0.055	0.0043	33.260	18.735	0.06635	12.200	7.175	0.0664
Mean	0.287	0.164	0.008003	0.108	0.063	0.006019	47.781	25.956	0.066964	17.740	10.000	0.066361
3rd Quartile	0.478	0.283	0.011225	0.153	0.090	0.00865	55.038	29.793	0.081125	20.968	12.245	0.0833
Maximum	0.910	0.530	0.021	0.300	0.180	0.0151	285.920	166.750	0.1078	92.320	51.960	0.1456

Appendix E: Data visualisation and analysis

E1: Visualisation of data

The following four sets of plots (Figures E.1.1-E1.4) show, separately for and adult and child, frequencies of different wind deviation angles from sampling line; wind speeds; potential inhalation exposures; actual dermal exposures; and potential dermal exposures.

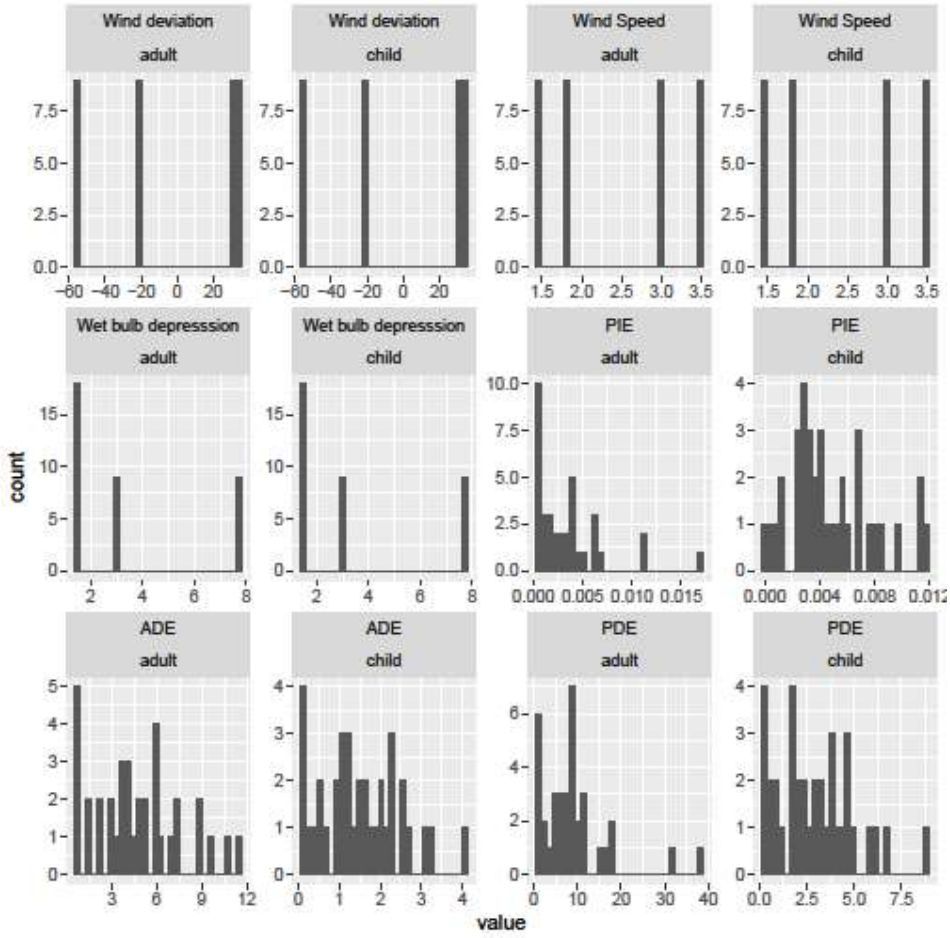


Figure E1.1: Pome Early Trials
(vertical axes, counts, horizontal axes: wind deviation, degrees; wind speed, m/s; wet bulb depression, degrees C; exposure values ml spray)

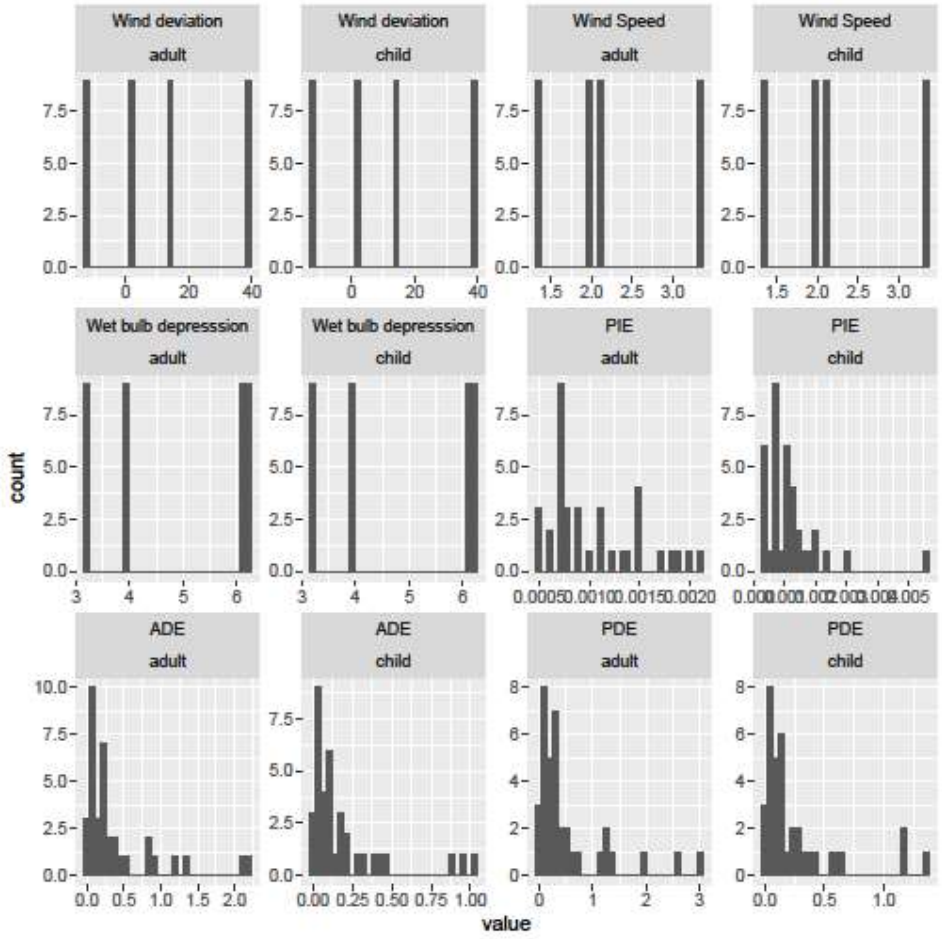


Figure E1.2: Vine Early Trials
(vertical axes, counts, horizontal axes: wind deviation, degrees; wind speed, m/s; wet bulb depression, degrees C; exposure values ml spray)

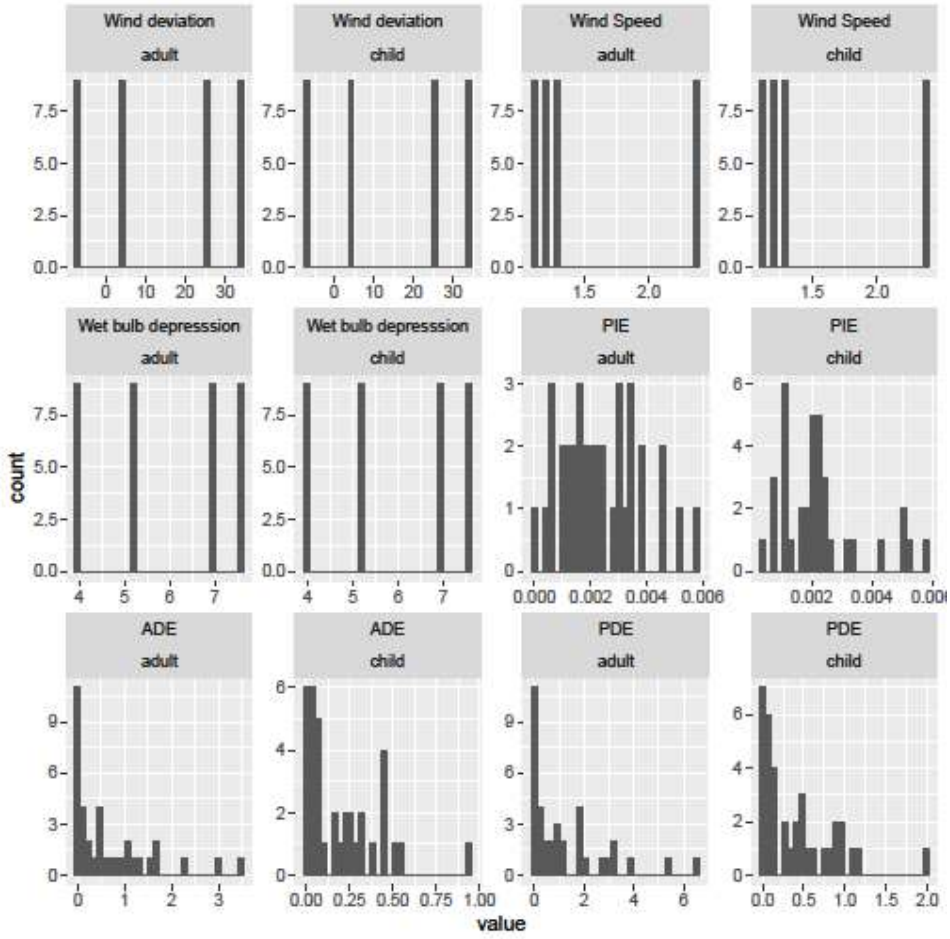


Figure E1.3: Pome Late Trials
(vertical axes, counts, horizontal axes: wind deviation, degrees; wind speed, m/s; wet bulb depression, degrees C; exposure values ml spray)

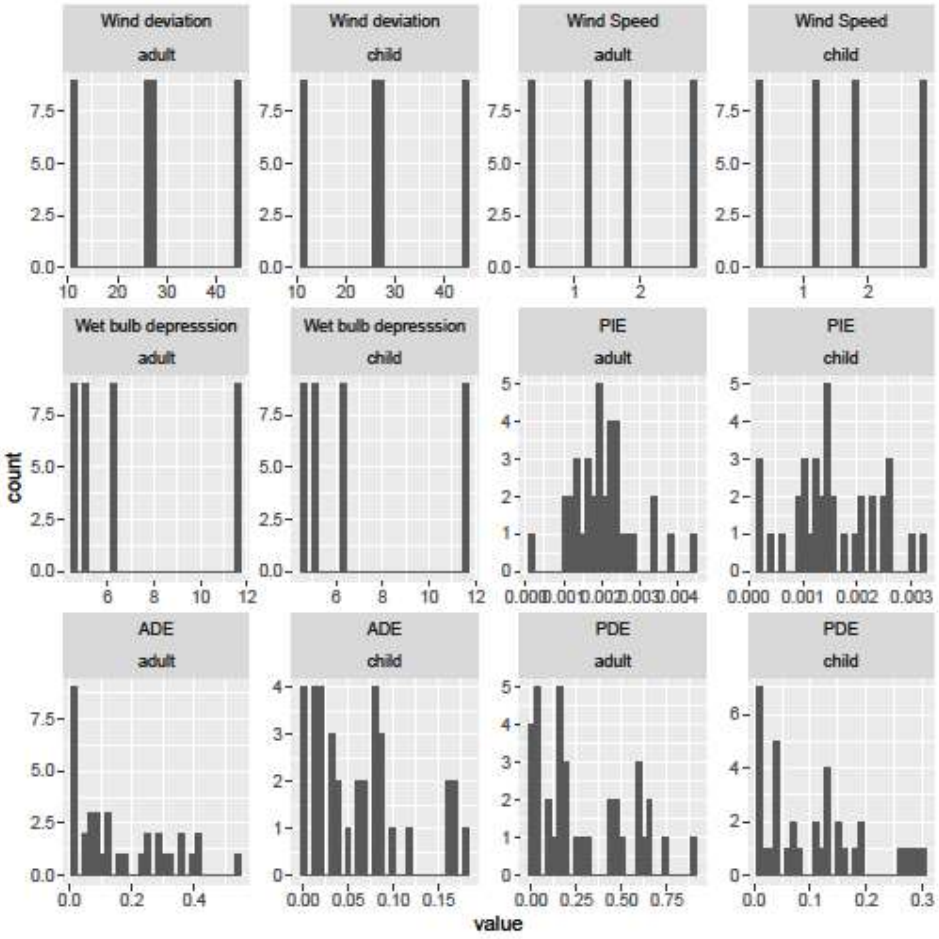


Figure E1.4: Vines Late Trials
(vertical axes, counts, horizontal axes: wind deviation, degrees; wind speed, m/s; wet bulb depression, degrees C; exposure values ml spray)

The following four sets of paired plots (Figures E1.5-E1.8) explore potential relationships between the following variables each set of the trials: wind deviation from the sampling line; wind speed; wet bulb depression; PDE; ADE; and PIE.

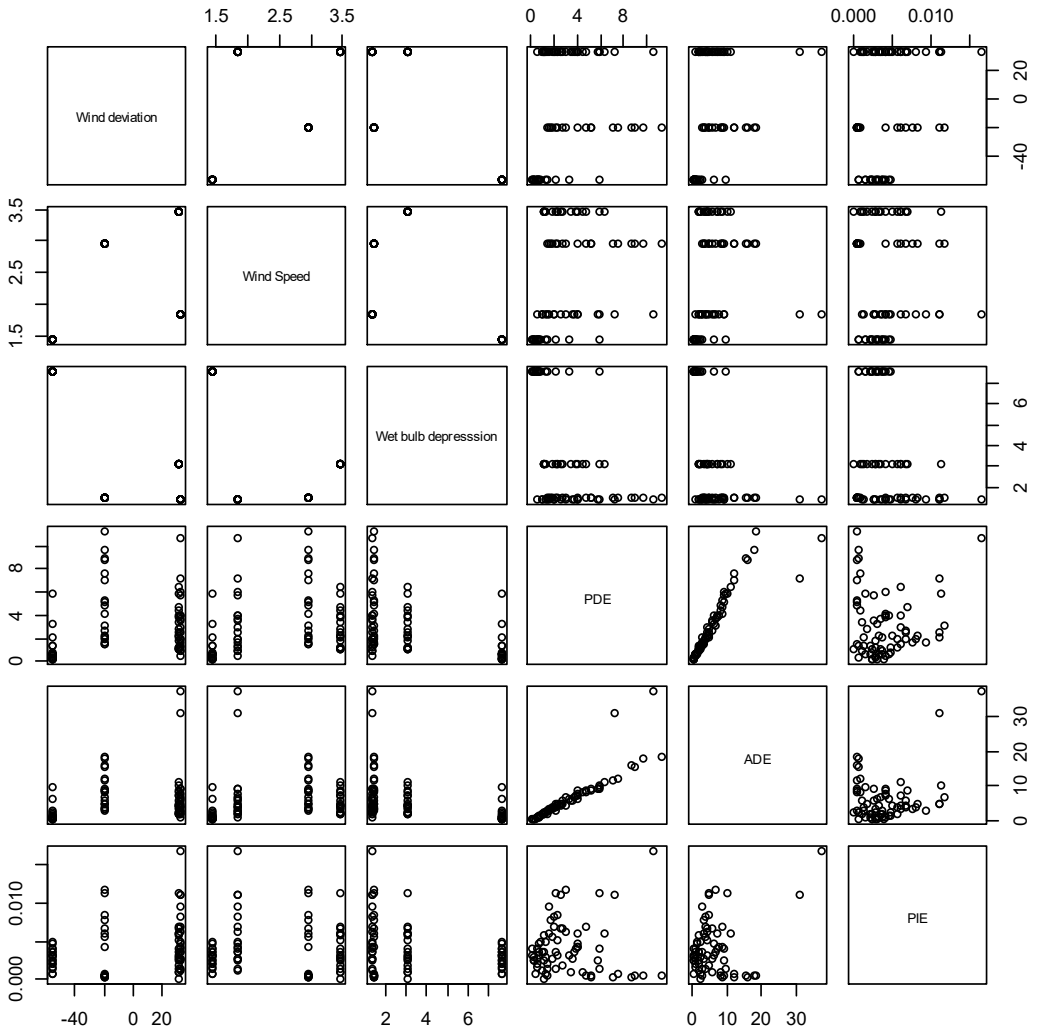


Figure E1.5: Pome Early Trials
(axes: wind deviation, degrees; wind speed, m/s; wet bulb depression, degrees C; exposure values ml spray)

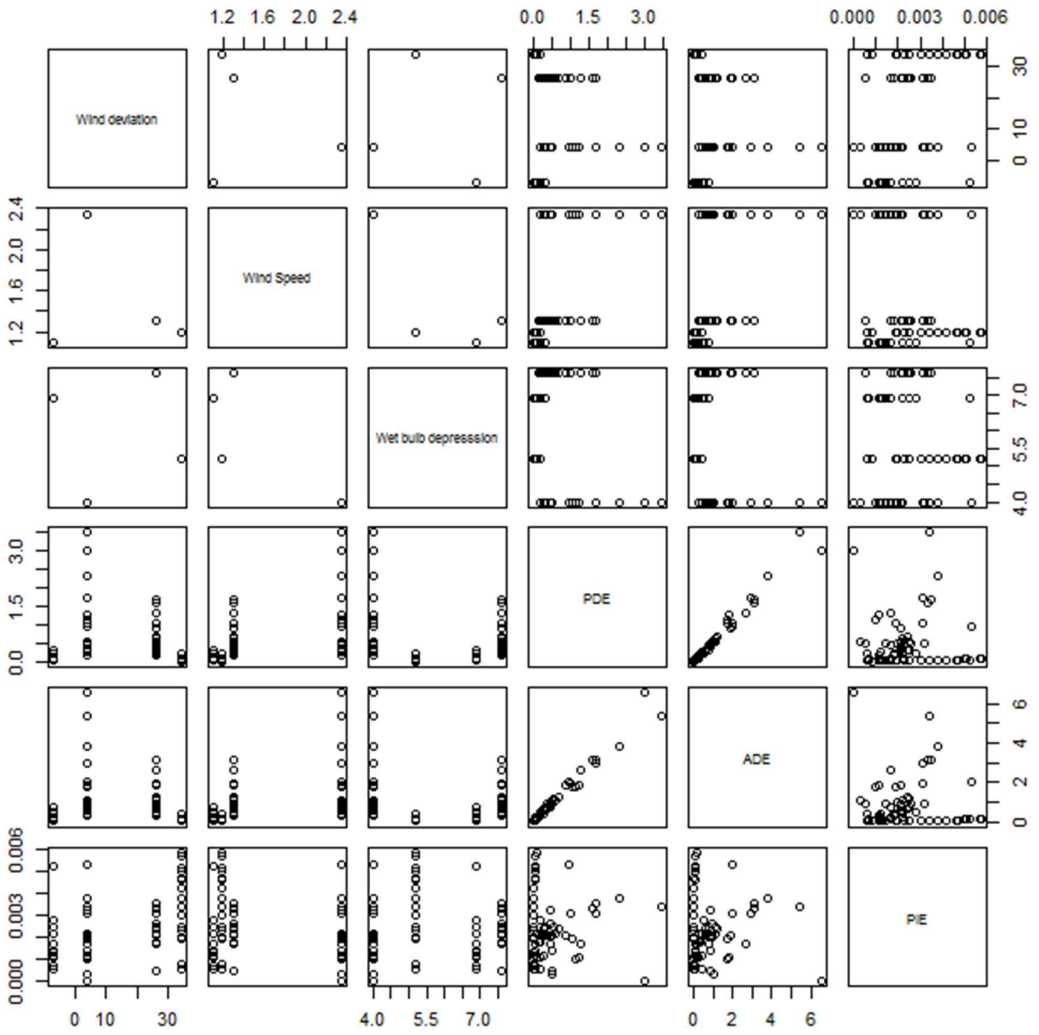


Figure E1.6: Pome Late Trials
(axes: wind deviation, degrees; wind speed, m/s; wet bulb depression, degrees C; exposure values ml spray)

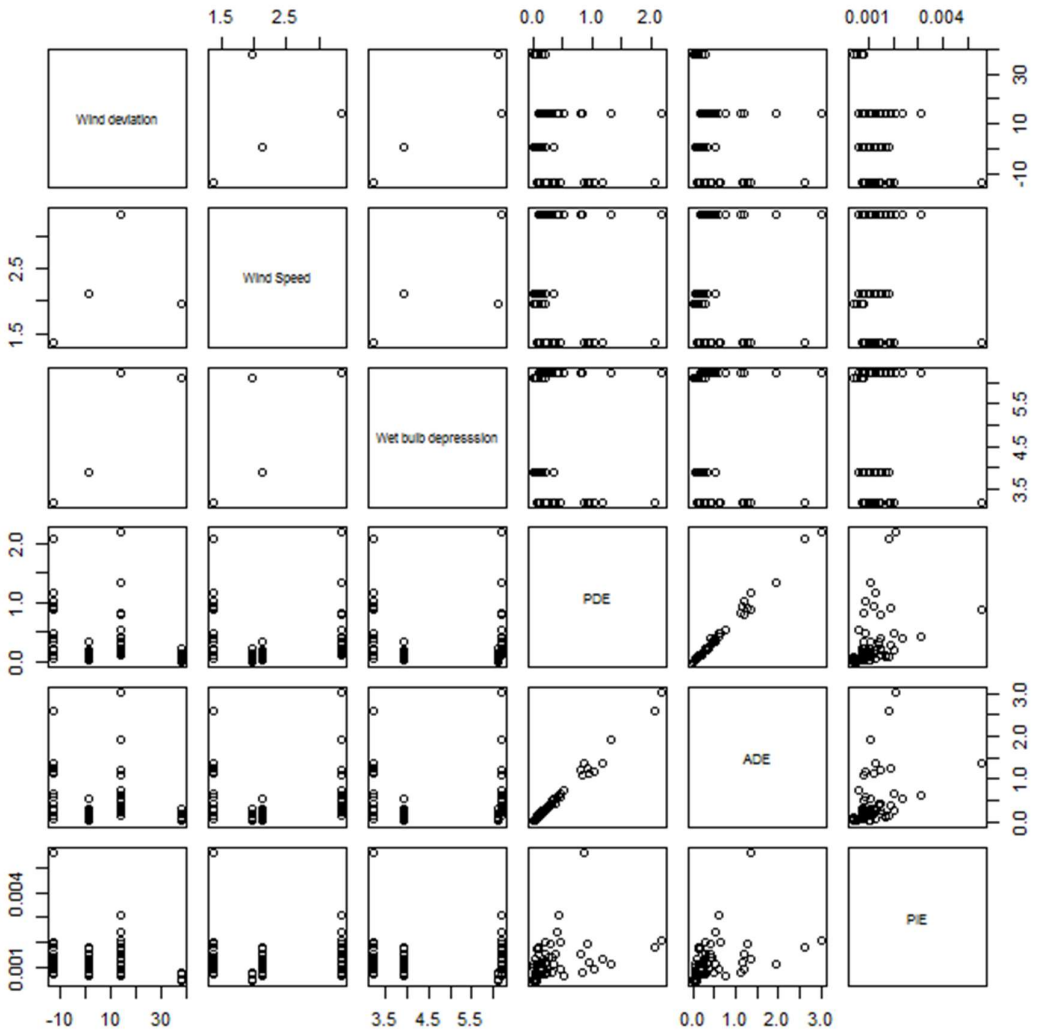


Figure E1.7: Vine Early Trials
(axes: wind deviation, degrees; wind speed, m/s; wet bulb depression, degrees C; exposure values ml spray)

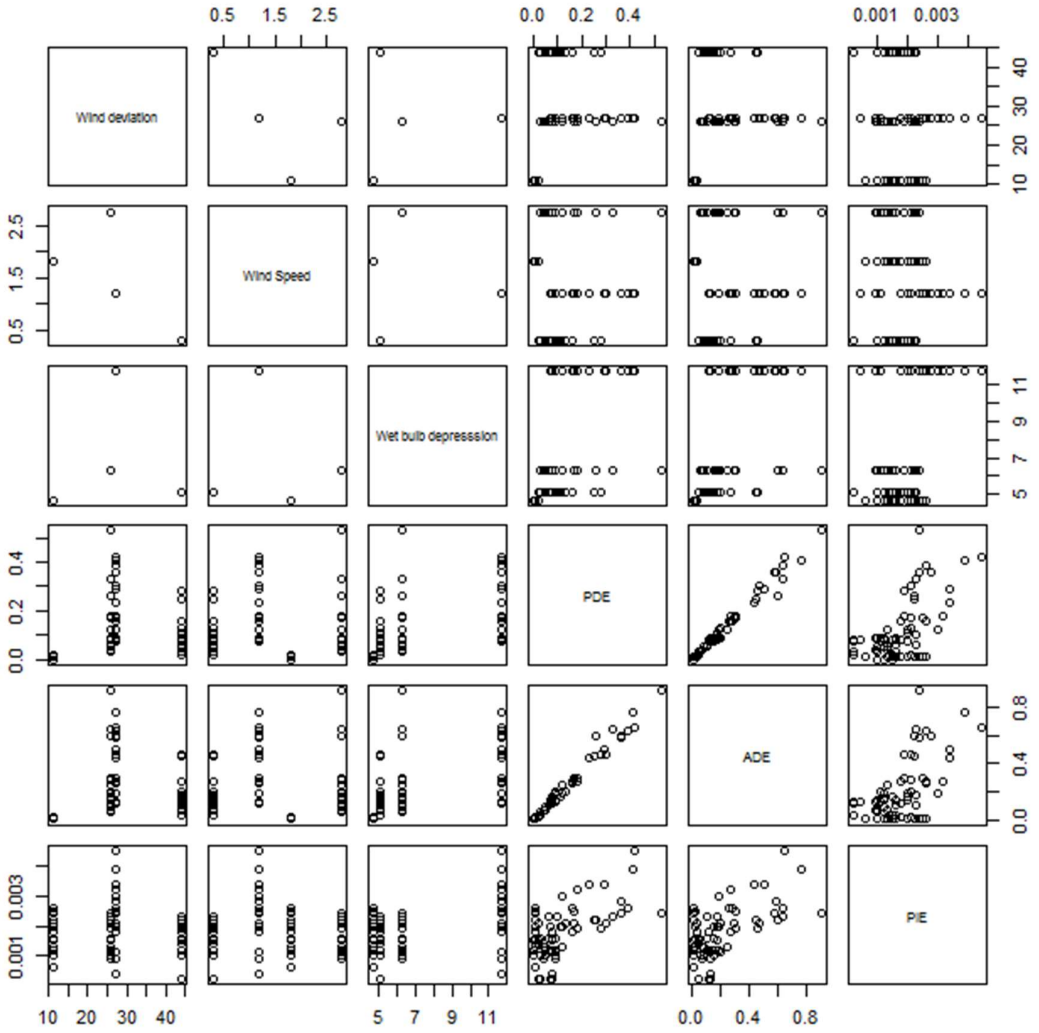


Figure E1.8: Vine Late Trials
(axes: wind deviation, degrees; wind speed, m/s; wet bulb depression, degrees C; exposure values ml spray)

E2: Factorial Analysis of Database

To explore possible interrelations among the combined numerical and categorical variables in the bystander database, as an aid to identification of any underlying structure of the variables, factorial analysis of the mixed data was done using R[i] and the PCAmixdata package[ii]. The PCAmixdata method essentially combines Principal Components Analysis (PCA) of quantitative variables and Multiple Correspondence Analysis (MCA) of qualitative variables. This was an attempt to reduce the data dimensionality, visualise any correlations between variables as well as to illustrate the observations in 3-dimensional space.

The factorial analysis was performed using a reduced set of variables, after removing obviously redundant variables, those variables which served as intermediates to calculate the exposure variables, and using simplified technical categorical variables to represent the essential differences between application equipment to reduce the complexity presented by detailed technical categorical variables to simpler factors which were considered to represent the essential differences between the various equipment used.

The selected variables, 16 numerical variables and 7 categorical variables, are shown in Table E2.1.

Table E2.1: Selection of Variables for factorial analysis

Database variable [Alternative name - for use in R]	**Name used in analysis**	**Rational for non-use in factorial analysis**	**Numerical/ categorical**
Study ID [study ID]		Study code used instead	
Study code [study.code]	**code**		Categorical
Application details:			
Country [country]	**country**		Categorical
Active substance [as]		Not varied within each crop	
Form. Type [formulation.type]		Not varied within each crop	
Sprayer type [sprayer.type]		12 different sprayers reported, replaced by two class generic sprayer category (see cat)	
Sprayer category [sprayer.cat]	**cat**		Categorical
Nozzle type [nozzle.type]		13 different combinations reported, with nozzle pressure replaced by two class spray quality categories (see quality)	
Nozzle number [nozzle.no]	**nozzle_no**		Numerical
Nozzle pressure (bar) [nozzle.pressure]		With nozzle type replaced by two spray quality class categories (see quality)	
Spray quality [spray.quality]	**quality**		Categorical
Forward speed (km/hr) [fwd.speed]	**speed**		Numerical
Spray volume applied (L/ha) [spray.volume.applied]	**vol**		Numerical
a.s.applied (g a.s./ha) [g.a.s.applied]	**amount**		Numerical
Spray conc. (g a.s./L) [spray.conc]	**conc**		Numerical
Crop details:			
Crop [crop]	**crop**		Categorical
Growth stage [growth.stage]		Replaced by two leaf cover class leaf categories	
Leaf cover [leaf.cover]	**leaf**		Categorical

[i] R Core Team (2017). R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. URL http://www.R-project.org/

[ii] Marie Chavent, Vanessa Kuentz, Amaury Labenne, Benoit Liquet and Jerome Saracco (2017). PCAmixdata: Multivariate Analysis of Mixed Data. R package version 3.0. https://CRAN.R-project.org/package=PCAmixdata

Database variable [Alternative name - for use in R]	Name used in analysis	Rational for non-use in factorial analysis	Numerical/ categorical
Crop height (m) [tree.height]	**tree_ht**		Numerical
Row spacing (m) [row.spacing]	**spacing**		Numerical
Downwind vegetation:			
At mannequins: Type [type]	(not used)	Downwind vegetation at mannequin, similar bare soil or low vegetation, considered to show little variation between trials	
At mannequins: Height (m) [height]	(not used)	Downwind vegetation height at mannequin considered to show little variation between trials	
Behind mannequins: Type [type]	(not used)	Downwind vegetation behind mannequin, considered to show little variation between trials	
Behind mannequins: Height (m) [height]	(not used)	Downwind vegetation height behind mannequin considered to show little variation between trials	
At distance: Type [type]	(not used)	Distant vegetation downwind of mannequin not included	
Trial details:			
Area sprayed (ha) [area.sprayed]	**area**		Numerical
Number of rows [no.rows]	**rows**		Numerical
Row length (m) [row.length]	(not used)	Not considered to be relevant given minimum length and wind directions	
Meteorological data:			
Location of weather station [weather.station.location]	(not used)	Not considered to be relevant	
Unobstructed area [unobstructed.area]	(not used)	Not considered to be useful	
Mean wind direction [mean.wind.direction]	(not used)	Not used directly, but used in deviation calculation	
Spray track direction [track.direction]	(not used_	Not used directly, but used in deviation calculation	
Required sample line direction [sample.direction]	(not used)	Not used directly, but used in deviation calculation	
Deviation from required direction [deviation.from.direction]	**deviation**		Numerical
Mean wind speed (m/s) [mean.wind.speed]	**wind**		Numerical
Height wind speed recorded (m) [height.wind.speed.recorded]	(not used)	All collected 2.0 m height	
Mean air temperature (^{o}C) [mean.air.temp]	(not used)	Not used directly, but used in wet bulb depression calculation	
Radiation (W.m-2) [radiation]	(not used)	Not measured at all sites, not used	
Rainfall (mm) [rainfall]	(not used)	Only experienced at one site (Study 4), so not used	
Humidity (%) [humidity]	(not used)	Not used directly, but used in wet bulb depression calculation	
Bystander:			
Child/adult [child.adult]	**subject**		Categorical
Mannequin [mannequin.id]	(not used)	Mannequin ID not used directly	
Height (m) [height]	(not used)	Mannequin height standardised for each type so not used	
buffer zone (m) [buffer.zone]	(not used)	Not used, as actual distance used	
Actual distance taking into account row width (m) [buffer.zone.row.width]	**distance**		Numerical
Inhalation exposure spray:			
Flow rate sampler (L/min) [mean.sampler.flow.rate]	(not used)	Not used directly but used to calculate PIE	
Sampling time (min) [sampling.time.spray]	(not used)	Not used directly but used to calculate PIE	
Air vol. reported (m^3) [sampling.volume.spray]	(not used)	Not used directly but used to calculate PIE	

Database variable [Alternative name - for use in R]	Name used in analysis	Rational for non-use in factorial analysis	Numerical/ categorical
a.s. measured (µg/specimen) [sampler.as.measured.spray]	(not used)	Not used directly but used to calculate PIE	
Recovery [recovery.filter]	(not used)	Not used directly but used to calculate PIE	
spray calculated (ml) [spray.dilution.calculated]	(not used)	Not used directly but used to calculate PIE	
PIE.R [PIE.R]	(not used)		
PIE.B [PIE.B]	(not used)		
PIE.B.15 [PIE.B.15]	(not used)		
PIE (µg a.s. sample tube)	**PIEmass**		Numerical
Dermal exposure: inner body dosimeter			
Adj inner body total (µg/specimen) [adj.inner.body.total]	(not used)	Not used directly but used to calculate ADE & PDE	
Lower arms (µg/specimen) [lower.arms]	(not used)	Not used directly but used to calculate ADE & PDE	
Recovery [recovery.dosimeter]	(not used)	Not used directly but used to calculate ADE & PDE	
Adj lower arms (µg/specimen) [adj.lower.arms]	(not used)	Not used directly but used to calculate ADE & PDE	
Torso & upper arms (µg/specimen) [torso.upperarms]	(not used)	Not used directly but used to calculate ADE & PDE	
Recovery [recovery.dosimeter]	(not used)	Not used directly but used to calculate ADE & PDE	
Adj torso & upper arms (µg/specimen) [adj.torso.upperarms]	(not used)	Not used directly but used to calculate ADE & PDE	
Lower legs (µg/specimen) [lower.legs]	(not used)	Not used directly but used to calculate ADE & PDE	
Recovery [recovery.dosimeter]	(not used)	Not used directly but used to calculate ADE & PDE	
Adj lower legs (µg/specimen) [adj.lower.legs]	(not used)	Not used directly but used to calculate ADE & PDE	
Waist & thighs (µg/specimen) [waist.thighs]	(not used)	Not used directly but used to calculate ADE & PDE	
Recovery [recovery.dosimeter]	(not used)	Not used directly but used to calculate ADE & PDE	
Adj waist & thighs (µg/specimen) [adj.waist.thighs]	(not used)	Not used directly but used to calculate ADE & PDE	
Head/neck (µg/specimen) [head.neck]	(not used)	Not used directly but used to calculate ADE & PDE	
Recovery [recovery.dosimeter]	(not used)	Not used directly but used to calculate ADE & PDE	
Adj head/neck (µg/specimen) [adj.head.neck]	(not used)	Not used directly but used to calculate ADE & PDE	
Dermal exposure: outer body dosimeter			
Adj outer body total (µg/specimen) [adj.outer.body.total]	(not used)	Not used directly but used to calculate PDE	
t-shirt (µg/specimen) [t.shirt]	(not used)	Not used directly but used to calculate PDE	
Recovery [recovery.dosimeter]	(not used)	Not used directly but used to calculate PDE	
adj t-shirt (µg/specimen) [adj.t.shirt]	(not used)	Not used directly but used to calculate PDE	
shorts (µg/specimen) [shorts]	(not used)	Not used directly but used to calculate PDE	
Recovery [recovery.dosimeter]	(not used)	Not used directly but used to calculate PDE	
adj shorts (µg/specimen) [adj.shorts]	(not used)	Not used directly but used to calculate PDE	
inner body (ml spray per person) [inner.body.spray]	(not used)	Not used directly but used to calculate ADE	
outer body (ml spray per person) [adj.outer.body.spray]	(not used)	Not used directly but used to calculate PDE	
ADE (µg a.s. per person) [ADE]	**ADEmass**		Numerical
PDE (µg a.s. per person) [PDE]	**PDEmass**		Numerical
Estimated Wet Bulb depression ΔT (°C) [DT]	**DT**		Numerical

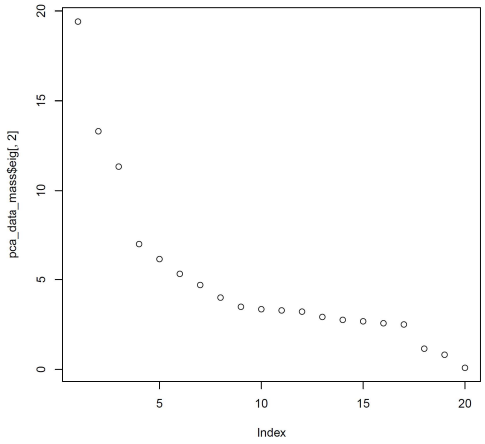


Figure 2.1: Visualisation of proportion of variation (y-axis, percent variation) associated with each of 20 individual dimensions (x-axis)

Figure E2.1 shows the percentage of variation associated with each of 20 individual dimensions. The first three dimensions were associated with 19.4, 13.3 and 11.3% of the total variation, so accounted for 44% of the cumulative total variation in the dataset. Although the first three dimensions only accounted for 44% of the variation in the dataset because the low incremental contributions from additional dimensions these were not considered to helpful to describing the dataset.

The squared loadings for the first 10 dimensions are presented in Table E2.2.

Table E2.2: Squared loadings for the 23 variables included in the factorial analysis

	dim1	dim2	dim3	dim4	dim5	dim6	dim7	dim8	dim9	dim10
nozzle_no	0.33	0.49	0.00	0.04	0.00	0.04	0.02	0.00	0.01	0.05
speed	0.09	0.30	0.02	0.22	0.22	0.00	0.01	0.02	0.01	0.01
vol	0.67	0.13	0.06	0.01	0.02	0.02	0.03	0.00	0.00	0.00
amount	0.75	0.11	0.05	0.02	0.00	0.00	0.02	0.01	0.00	0.00
conc	0.22	0.66	0.00	0.06	0.01	0.03	0.01	0.00	0.00	0.00
tree_ht	0.85	0.05	0.01	0.05	0.00	0.00	0.00	0.00	0.00	0.00
spacing	0.88	0.00	0.01	0.05	0.00	0.00	0.00	0.00	0.00	0.00
area	0.02	0.04	0.25	0.31	0.09	0.03	0.02	0.20	0.01	0.01
rows	0.52	0.00	0.02	0.01	0.01	0.10	0.31	0.00	0.00	0.02
deviation	0.03	0.00	0.08	0.09	0.60	0.01	0.01	0.08	0.09	0.00
wind	0.02	0.12	0.46	0.09	0.00	0.12	0.02	0.01	0.05	0.04
distance	0.00	0.00	0.01	0.00	0.01	0.02	0.00	0.00	0.05	0.02
PIEmass	0.05	0.13	0.10	0.01	0.05	0.13	0.02	0.00	0.00	0.00
ADEmass	0.14	0.03	0.52	0.00	0.06	0.01	0.00	0.02	0.07	0.03
PDEmass	0.13	0.03	0.45	0.00	0.08	0.03	0.01	0.01	0.07	0.03
DT	0.01	0.11	0.36	0.07	0.12	0.01	0.11	0.04	0.02	0.05
code	1.00	0.99	0.95	1.00	0.97	0.96	0.99	0.98	0.79	0.89
country	0.59	0.84	0.21	0.46	0.09	0.40	0.17	0.07	0.00	0.08
cat	0.04	0.70	0.09	0.01	0.01	0.04	0.05	0.02	0.03	0.00
quality	0.58	0.50	0.20	0.25	0.10	0.12	0.05	0.07	0.03	0.02
crop	0.85	0.04	0.02	0.02	0.01	0.00	0.00	0.00	0.03	0.00
leaf	0.01	0.07	0.65	0.04	0.00	0.06	0.02	0.05	0.00	0.01
subject	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.01	0.11	0.07

Table Table E2.2 shows a main output of the factorial analysis, the squared loadings for both the numerical and categorical variables. For the quantitative variables the values represent the squared correlation between the variable and the dimension. While for the qualitative variable the results represent the correlation ratio between the variable and the dimension.

From the Table, loadings for variables volume, amount, tree height, rows, quality, and crop are associated with dimension 1. Crop can be considered as the lead variable here as tree height, amount, and volume are determined by the crop. The associations for dimension 2 are concentration, category, and quality. While dimension 3 is associated with wind speed, ADEmass, PDEmass, and leaf. Further graphical representation of the analysis for the first three dimensions is shown below:

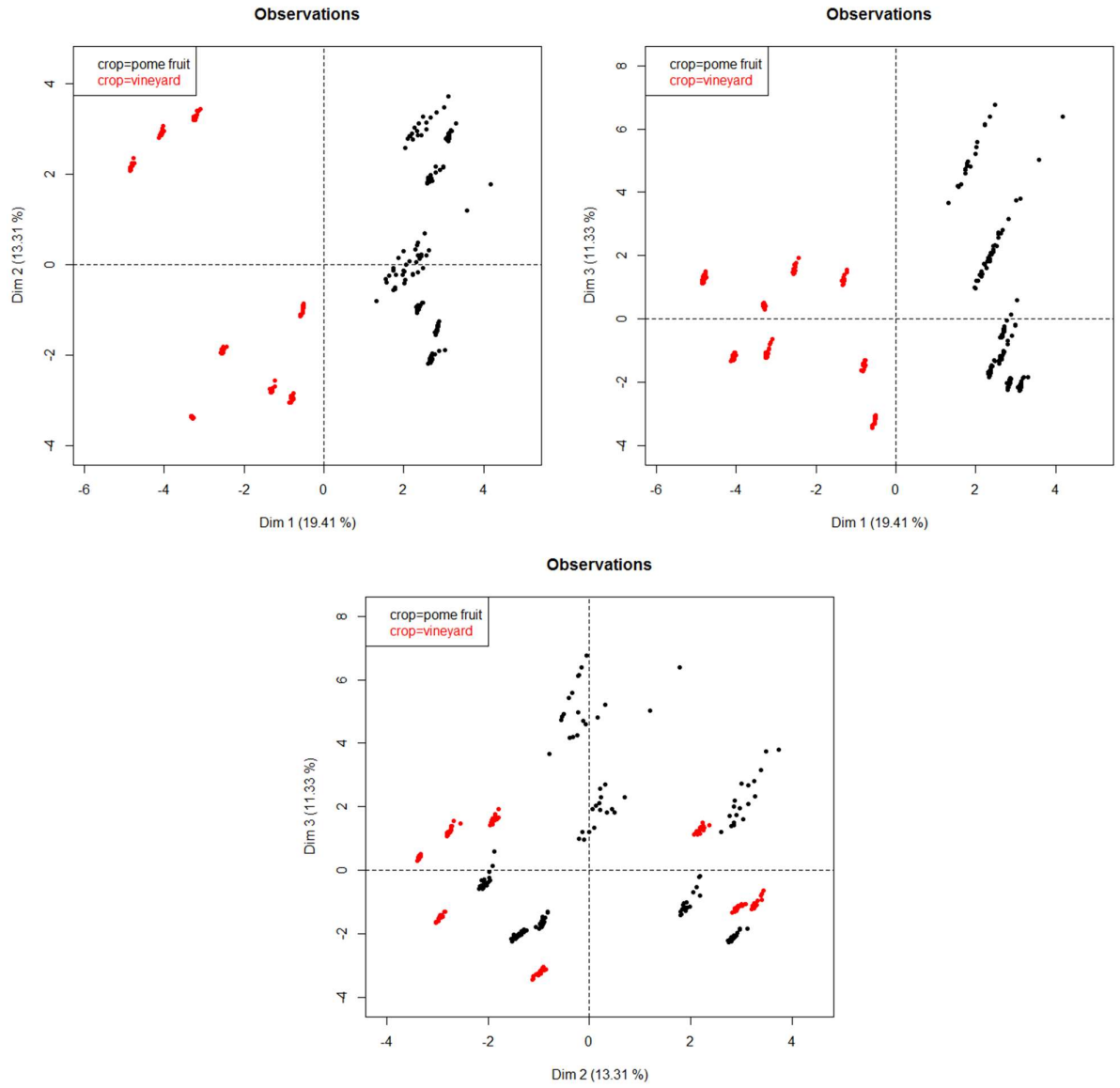


Figure E2.2 a, b, and c: Observations plotted on axes of first three dimensions, showing crop type (pome fruit or vineyard)

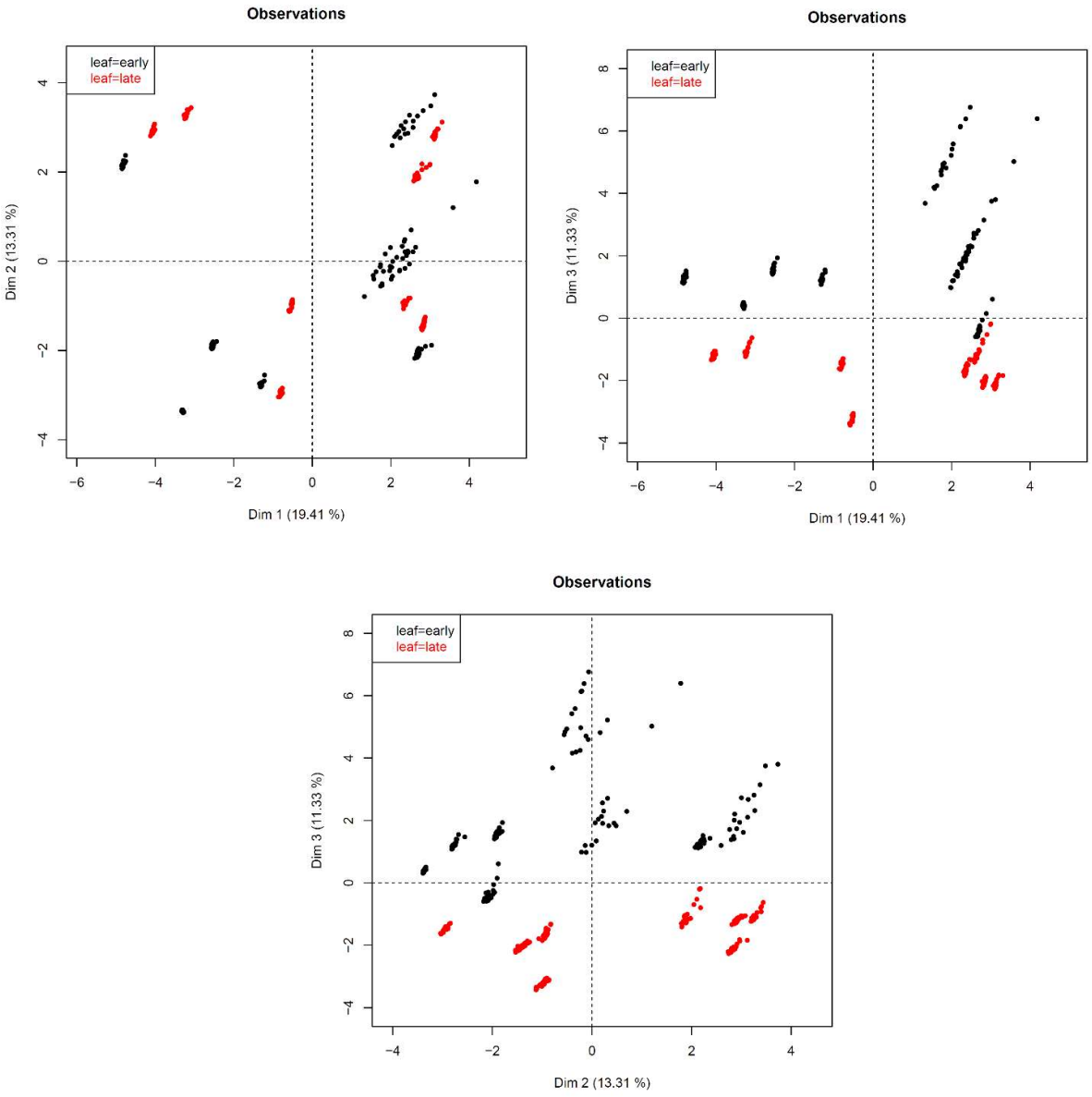


Figure E2.3 a, b, and c: Observations plotted on axes of first three dimensions, showing leaf cover (early = none, late = full)

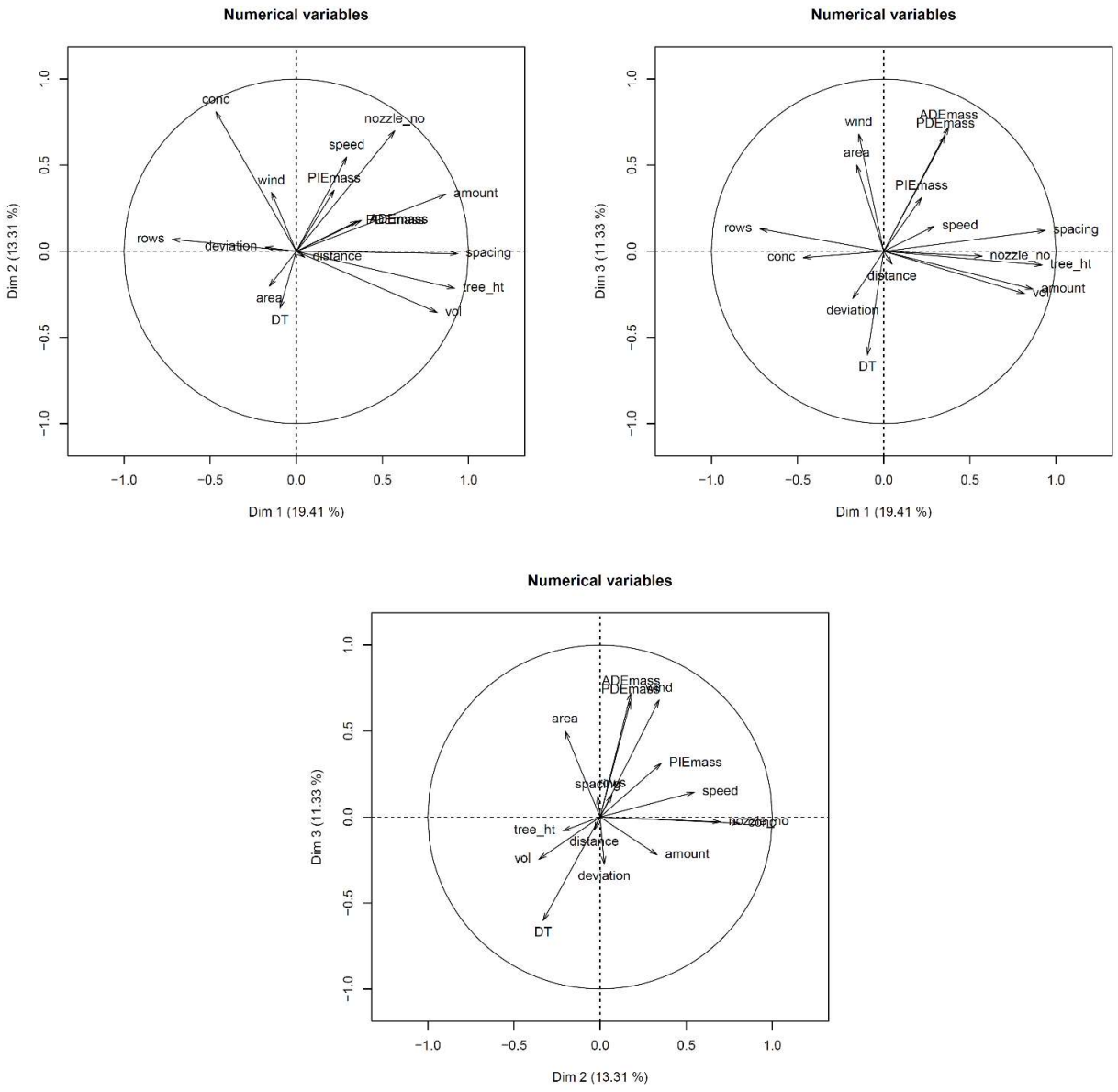


Figure E2.4 a, b and c: Correlation circles for numerical variables for first three dimensions

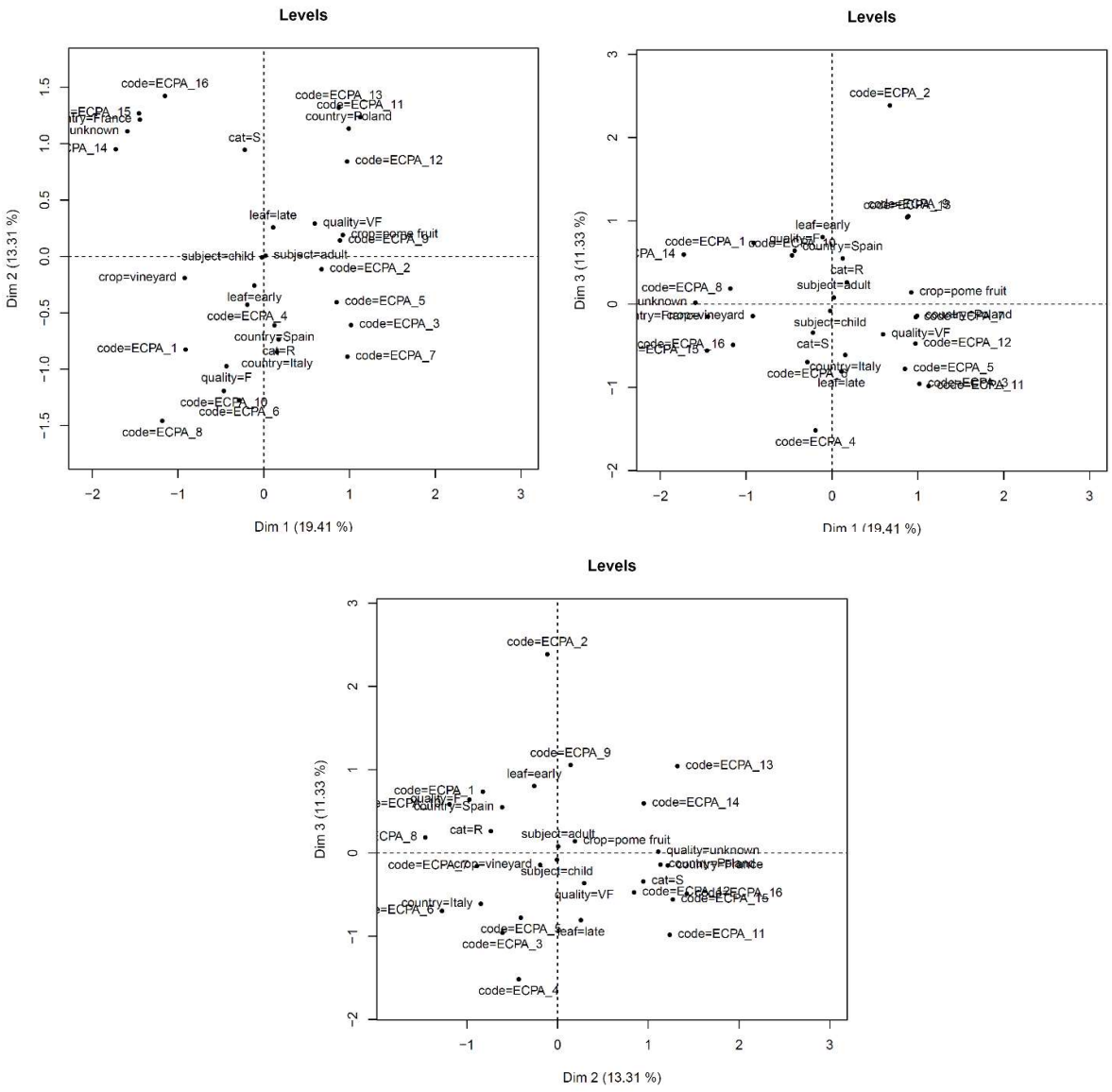


Figure E2.5 a, b, and c: Factor map for categorical variables

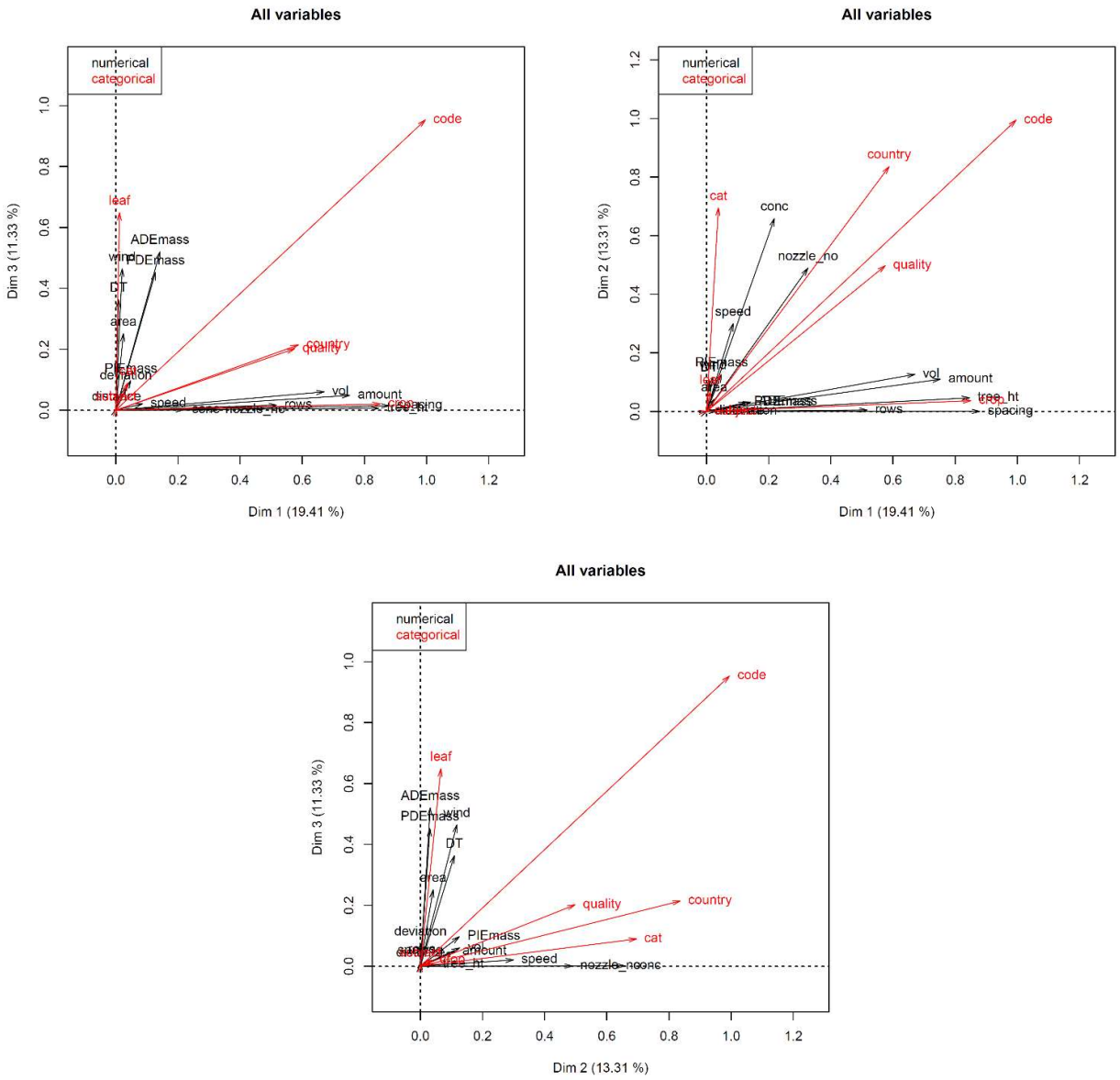


Figure E2.6 a, b, and c: Squared loadings for all variables for first three dimensions

These data suggest that separating the dataset into four subsets on the basis of crop type and leaf cover is reasonable, as these contribute most to the differences between the sampling units.

E3: Exploration of possible relationships between exposure and other variables

Linear regression was used to explore associations between measured exposures and other study variables. As the exposure data appeared to at least approximate lognormal distributions, and residuals from regressions using untransformed values were not normally distributed, linear regression fitting was done using log transformed exposure values (i.e. log10 values for PDE, ADE, and PIE) and untransformed predictor variables. The predictor values used, with abbreviated labels in square brackets, were: sprayer category [cat], nozzle number [nozzle_no], spray quality [quality], forward speed [speed], volume sprayed [vol], amount of active substance applied [amount], spray concentration [conc], tree height [tree_ht], row spacing [spacing], area sprayed [area], wind speed [wind], deviation from required wind direction [deviation], sample distance from spray line [distance], wet bulb and depression [DT]). This analysis was done using R and forward selection by AIC, allowing steps to be taken in both directions.

Two approaches were used. The first modelled the associations between the exposure mass (i.e. µg or mg active substance) reported and other variables, while the second approach looked at associations between exposure expressed as spray volume (ml). Summary results for these two approaches are shown in Table E3.1 and Table E3.2. Modelling was done separately for children and adults in the four separate scenarios: pome fruit early (i.e. no leaf cover); pome fruit late (i.e. full leaf cover); vine fruit early; and vine fruit late.

Table E3.1: Summary of regression results for log transformed exposure values where exposure was expressed as mass of active substance.
For each exposure the final model identified by the stepwise procedure and predictive variables, in descending order of statistical significance, are shown, along with the intercept and other coefficients, and the adjusted R^2 for the regression. The interquartile ranges of the predictive variables (untransformed values) are shown in parenthesis at each first occurrence of the variable.

Scenario / Exposure variable	Final predictive model	Intercept	1st variable	2nd variable	3rd variable	4th variable	Adjusted R squared
Pome Early child							
PDE (210-607)	~ vol + distance + speed	3.0661663	vol -0.0013966 (590-745)	distance -0.0457851 (7-17)	speed 0.1842799 (4.8-5.3)		0.8646
ADE (122-316)	~ vol + distance + speed	3.0502512	vol -0.0014652	distance -0.0425945	speed 0.1355651		0.8574
PIE (0.065-0.225)	~ distance + amount	6.08447	distance -0.05580	amount -0.06880 (91.3-96.3)			0.2255
Pome early adult							
PDE (665-1739)	~ DT + distance + tree_ht	4.534815	DT -0.134641 (1.5-4.2)	distance -0.040832	tree_ht -0.203915 (2.8-3.6)		0.8583
ADE (418-1034)	~ DT + distance + spacing	1.120306	DT -0.187459	distance -0.032771	spacing 0.678460 (3.7-4.0)		0.8713
PIE (0.021-0.110)	~ area + distance + wind	0.47660	area -1.61361 (0.9-1.2)	distance -0.02164	wind 0.09004 (1.7-3.1)		0.666
Pome late child							
PDE (10.7-83.3)	~ tree_ht + distance + amount + cat	9.215508	tree_ht -1.555877 (3.4-3.6)	distance -0.057255 (7-17)	amount -0.013900 (112.5-122.5)	catS -0.198490 (S:36 R:36)	0.8972
ADE	~ tree_ht + cat	9.673892	tree_ht	catS	distance		0.9042

Scenario / Exposure variable	Final predictive model	Intercept	1st variable	2nd variable	3rd variable	4th variable	Adjusted R squared
(3.93-40.8)	+ distance		-2.165644	-0.704822	-0.057815		
PIE (0.051-0.075)	~ distance	-0.957349	distance -0.023845				0.1696
Pome late adult							
PDE (17.4-250)	~ tree_ht + amount + distance + cat	10.793287	tree_ht -1.779299	amount -0.016860	distance -0.061469	catS -0.233823	0.9108
ADE (6.18-134)	~ tree_ht + cat + distance + speed	11.878644	tree_ht -2.705607	catS -1.016614	distance -0.058065	speed 0.043737 (4.75-6.0)	0.9112
PIE (0.040-0.078)	~ distance + speed	-1.20935	distance -0.02615	speed 0.03794			0.1705
Vine Early child							
PDE (7.40-34.8)	~ deviation + distance + wind	1.629681	deviation -0.024270 (-2.5-20.0)	distance -0.066635 (6-16)	wind 0.244612 (1.8-2.4)		0.8365
ADE (5.83-26.2)	~ deviation + distance + wind,	1.540769	deviation -0.023366	distance -0.067653	wind 0.222223		0.8249
PIE (0.014-0.066)	~ conc + vol + distance	-1.9544618	conc 3.0674701 (0.075-0.1475)	vol 0.0008738 (148-402)	distance -0.0277399		0.792
Vine early adult							
PDE (19.0-79.8)	~ amount + +speed + distance	0.886937	amount 0.021227 (22.25-44.75)	speed 0.156396 (2.75-4.62)	distance -0.061728		0.8298
ADE (14.4-49.6)	~ deviation + distance + wind	1.517526	deviation -0.023205	distance -0.060954	wind 0.339535 (1.8-2.4)		0.8118
PIE (0.013-0.037)	~ conc + vol + distance	-2.0280883	conc 2.6757190	vol 0.0005305	distance -0.0166376		0.8263
Vine late child							
PDE (4.77-21.0)	~ nozzle_no + vol + distance	2.534e-01	nozzle_no 9.111e-02 (8-11)	vol 7.845e-04 (190-665)	distance -4.523e-02 (7-16)		0.8917
ADE (2.78-12.2)	~ nozzle_no + vol + distance	4.590e-02	nozzle_no 8.536e-02	vol 7.568e-04	distance -4.051e-02		0.8724
PIE (0.055-0.083)	~ distance + conc	-1.11247	distance -0.03749	conc 1.18785 (0.11-0.33)			0.3789
Vine late adult							
PDE (9.90-55.0)	~ quality + amount + distance	6.652241	Quality-unknown -2.057684 (unknow:18 F:18 VF:36)	qualityVF 0.512039	amount -0.070876 (60.5-69.0)	distance -0.040622	0.8995
ADE (5.80-29.8)	~ quality + amount + distance	5.663666	Quality unknown -2.006407	qualityVF 0.409645	amount -0.059155	distance -0.040366	0.9055
PIE (0.054-0.081)	~ tree_ht + distance	-0.790812	tree_ht -0.145512 (0.6-1.95)	distance -0.020353			0.3708

Considering the results in Table E3.1, the adjusted r-squared values suggest that the models for potential dermal and actual dermal exposure are associated with reasonable proportions of variability in the data. Whereas, for inhalation exposures the r-squared values typically show that a large part of the variability is not explained by the models. Several variables were selected by the stepwise process as being useful determinants of exposures, but only bystander distance was consistently identified as being a statistically significant predictor. Other variables were not consistently selected across scenarios (e.g. spray volume was selected for pome fruit early child but

not for adult data in the same trials) or only appeared in isolated cases. Consideration of the magnitude of the coefficients and the variable interquartile ranges indicated that in most cases variation in distance was likely to be the variable with most influence on the regression. The repeated analysis where exposure was expressed as spray volume gave very similar results, Table E3.2. Distance again was the only consistently selected variable in all cases. Other variables selected in this second approach were often different to those variables selected for the same scenarios in the first approach. Finally, regulatory considerations suggest of the different variables identified as potential determinants only bystander distance would be practical variable to consider. Therefore, it was concluded that it was not possible to identify a practical regression model that would permit extrapolation of the observed results to other scenarios. Therefore, to use these the data it is considered appropriate to identify the required exposure statistics (i.e. 75th and 95th percentiles) from the observed normalised exposures. Given the uncertainty in the spray volume data the normalisation by the amount of active substance applied are the preferred values.

Table E3.2: Summary of regression results for log transformed exposure values where exposure is expressed as ml of spray solution. For each exposure the final model identified by the stepwise procedure and predictive variables, in descending order of statistical significance, are shown, along with the intercept and other coefficients, and the adjusted R2 for the regression.

Scenario / Exposure variable	"Best" predictive variables	Intercept	1st variable	2nd variable	3rd variable	4th variable	Adjusted R squared
Pome Early child							
PDE	~ DT + distance + spacing + cat	-2.365970	DT -0.171485	distance -0.045833	spacing 1.009477	catS -0.145050	0.8021
ADE	~ DT + distance + spacing	-1.373490	DT -0.142574	distance -0.042752	spacing 0.633429		0.7878
PIE	~ distance + cat	-1.17008	distance -0.05590	catS -0.45154			0.1968
Pome early adult							
PDE	~ DT + distance	1.721451	DT -0.129465	distance -0.040646			0.7933
ADE	~ DT + distance + speed	0.538751	DT -0.081689	distance -0.032741	speed 0.134097		0.8065
PIE	~ area + distance	-0.30112	area -1.46949	distance -0.02113			0.6474
Pome late child							
PDE	~ tree_ht + cat+ distance + vol	9.3030044	tree_ht -2.3079914	catS -1.1532134	distance -0.0551227	vol -0.0008463	0.9046
ADE	~ tree_ht + cat + distance	8.114843	tree_ht -2.294310	catS -0.990983	distance -0.054159		0.9117
PIE	~ conc + distance,	-1.342357	conc -3.902243	distance -0.023856			0.4105
Pome late adult							
PDE	~ tree_ht + cat + distance + vol	11.4981478	tree_ht -2.6859638	catS -1.3760462	distance -0.0626687	vol -0.0011810	0.9122
ADE	~ tree_ht + cat + distance + speed	10.544832	tree_ht -2.880064	catS -1.259828	distance -0.056396	speed 0.028713	0.9108
PIE	~ distance + amount	-1.210934	distance -0.026269	amount -0.005265			0.1509
Vine Early child							
PDE	~ vol + distance + tree_ht	-0.503314	vol 0.002529	distance -0.068410	tree_ht -0.277919		0.8416
ADE	~ vol + distance + speed	-1.257600	vol 0.002171	distance -0.067202	speed 0.089542		0.8455
PIE	~ deviation + distance + DT	-2.527121	deviation -0.013927	distance -0.027245	DT 0.118372		0.5489
Vine early adult							
PDE	~ vol + speed + distance	-0.9622978	vol 0.0016652	speed 0.1534782	distance -0.0612961		0.8176
ADE	~ vol + distance	-1.1182365	vol	distance	speed		0.8126

Scenario / Exposure variable	"Best" predictive variables	Intercept	1st variable	2nd variable	3rd variable	4th variable	Adjusted R squared
	+ speed		0.0018608	-0.0617499	0.1414686		
PIE	~ vol + distance	-2.3465707	vol 0.0004019	distance -0.0165348			0.3378
Vine late child							
PDE	~ quality + distance + spacing	0.967417	Quality unknown -1.403269	qualityVF -0.430376	distance -0.040147	spacing -0.410414	0.9182
ADE	~ quality + distance + vol	0.0015852	Quality unknown -1.5915244	qualityVF -0.6028264	distance -0.0438065	vol -0.0004971	0.9153
PIE	~ distance + vol	-2.1245197	distance -0.0374989	vol 0.0004780			0.3515
Vine late adult							
PDE	~ quality + distance + conc	0.14789	Quality unknown -1.79180	qualityVF -0.45790	distance -0.04431	conc 0.85284	0.9427
ADE	~ quality + distance	-0.118287	Quality unknown -1.488747	qualityVF -0.367230	distance -0.034930		0.9198
PIE	~ vol + distance	-2.236e+00	vol 6.548e-04	distance -2.043e-02			0.6008

E4: R Outputs

Individual regression summaries for final models

Regression using exposure masses

Pome Fruit Early Scenario

```
> summary(FinalPomeShort_early_childPDEmass)

Call:
lm(formula = log10(PDEmass) ~ vol + distance + speed, data = subset(PomeShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.31541 -0.08904 -0.01992  0.06213  0.74302

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  3.0661663  0.3945987   7.770 7.33e-09 ***
vol         -0.0013966  0.0002085  -6.699 1.46e-07 ***
distance    -0.0457851  0.0072960  -6.275 4.90e-07 ***
speed        0.1842799  0.0520323   3.542  0.00124 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1788 on 32 degrees of freedom
Multiple R-squared:  0.8762,          Adjusted R-squared:  0.8646
F-statistic: 75.48 on 3 and 32 DF,  p-value: 1.324e-14

> FinalPomeShort_early_childADEmass <- lm(log10(ADEmass) ~ vol + distance +speed, data = subset(PomeShort_early,subject=="child"))

> summary(FinalPomeShort_early_childADEmass)

Call:
lm(formula = log10(ADEmass) ~ vol + distance + speed, data = subset(PomeShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.33746 -0.10103 -0.00668  0.06027  0.72649

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  3.0502512  0.3846164   7.931 4.74e-09 ***
vol         -0.0014652  0.0002032  -7.211 3.44e-08 ***
distance    -0.0425945  0.0071114  -5.990 1.12e-06 ***
speed        0.1355651  0.0507160   2.673   0.0117 *
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1742 on 32 degrees of freedom
Multiple R-squared:  0.8696,          Adjusted R-squared:  0.8574
F-statistic: 71.14 on 3 and 32 DF,  p-value: 3.018e-14

FinalPomeShort_early_childPIEmass <- lm(log10(PIEmass) ~ distance + amount, data = subset(PomeShort_early,subject=="child"))

> summary(FinalPomeShort_early_childPIEmass)

Call:
lm(formula = log10(PIEmass) ~ distance + amount, data = subset(PomeShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-2.53117 -0.09228  0.13135  0.25507  0.76108

Coefficients:
            Estimate Std. Error t value Pr(>|t|)
(Intercept)  6.08447    2.68302   2.268   0.0300 *
distance    -0.05580    0.02241  -2.490   0.0180 *
amount      -0.06880    0.02858  -2.407   0.0218 *
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.549 on 33 degrees of freedom
Multiple R-squared:  0.2697,          Adjusted R-squared:  0.2255
F-statistic: 6.094 on 2 and 33 DF,  p-value: 0.005592
```

```
> FinalPomeShort_early_adultPDEmass <- lm(log10(PDEmass) ~ DT + distance +tree_ht, data = subset(PomeShort_early,subject=="adult"))

> summary(FinalPomeShort_early_adultPDEmass)

Call:
lm(formula = log10(PDEmass) ~ DT + distance + tree_ht, data = subset(PomeShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.30838 -0.11122 -0.00395  0.05920  0.53477

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  4.534815   0.217108  20.887  < 2e-16 ***
DT          -0.134641   0.015622  -8.619 7.54e-10 ***
distance    -0.040832   0.007592  -5.378 6.61e-06 ***
tree_ht     -0.203915   0.068762  -2.965  0.00567 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.186 on 32 degrees of freedom
Multiple R-squared:  0.8705,          Adjusted R-squared:  0.8583
F-statistic: 71.68 on 3 and 32 DF,  p-value: 2.719e-14

> FinalPomeShort_early_adultADEmass <- lm(log10(ADEmass) ~ DT + distance +spacing, data = subset(PomeShort_early,subject=="adult"))

> summary(FinalPomeShort_early_adultADEmass)

Call:
lm(formula = log10(ADEmass) ~ DT + distance + spacing, data = subset(PomeShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.28607 -0.11497  0.01237  0.07823  0.55793

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  1.120306   0.648533   1.727 0.093730 .
DT          -0.187459   0.014376 -13.040 2.38e-14 ***
distance    -0.032771   0.006581  -4.980 2.11e-05 ***
spacing      0.678460   0.176853   3.836 0.000553 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1612 on 32 degrees of freedom
Multiple R-squared:  0.8824,          Adjusted R-squared:  0.8713
F-statistic:    80 on 3 and 32 DF,  p-value: 5.867e-15

> FinalPomeShort_early_adultPIEmass <- lm(log10(PIEmass) ~ area + distance + wind, data = subset(PomeShort_early,subject=="adult"))

> summary(FinalPomeShort_early_adultPIEmass)

Call:
lm(formula = log10(PIEmass) ~ area + distance + wind, data = subset(PomeShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.62604 -0.14277 -0.00288  0.18098  0.56277

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  0.47660    0.24982   1.908   0.0654 .
area        -1.61361    0.19756  -8.168  2.5e-09 ***
distance    -0.02164    0.01080  -2.003   0.0538 .
wind         0.09004    0.05740   1.569   0.1266
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2647 on 32 degrees of freedom
Multiple R-squared:  0.6946,          Adjusted R-squared:  0.666
F-statistic: 24.27 on 3 and 32 DF,  p-value: 2.227e-08
```

Pome Fruit Late Scenario

```
> FinalPomeShort_late_childPDEmass <- lm(log10(PDEmass) ~ tree_ht + distance + amount + cat, data =
subset(PomeShort_late,subject=="child"))

> summary(FinalPomeShort_late_childPDEmass)

Call:
lm(formula = log10(PDEmass) ~ tree_ht + distance + amount + cat,
```

```
   data = subset(PomeShort_late, subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.43378 -0.11554  0.01365  0.13688  0.32794

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  9.215508   0.544035  16.939  < 2e-16 ***
tree_ht     -1.555877   0.221154  -7.035 6.72e-08 ***
distance    -0.057255   0.008609  -6.650 1.96e-07 ***
amount      -0.013900   0.005022  -2.768  0.00943 **
catS        -0.198490   0.160135  -1.240  0.22446
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2109 on 31 degrees of freedom
Multiple R-squared:  0.9089,          Adjusted R-squared:  0.8972
F-statistic: 77.33 on 4 and 31 DF,  p-value: 1.123e-15


> FinalPomeShort_late_childADEmass <- lm(log10(ADEmass) ~ tree_ht + cat + distance, data = subset(PomeShort_late,subject=="child"))

> summary(FinalPomeShort_late_childADEmass)

Call:
lm(formula = log10(ADEmass) ~ tree_ht + cat + distance, data = subset(PomeShort_late,
   subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.44248 -0.12923  0.02898  0.16687  0.34760

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  9.673892   0.523366  18.484  < 2e-16 ***
tree_ht     -2.165644   0.136819 -15.829  < 2e-16 ***
catS        -0.704822   0.096734  -7.286 2.79e-08 ***
distance    -0.057815   0.008376  -6.902 8.20e-08 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2052 on 32 degrees of freedom
Multiple R-squared:  0.9124,          Adjusted R-squared:  0.9042
F-statistic: 111.1 on 3 and 32 DF,  p-value: < 2.2e-16


> FinalPomeShort_late_childPIEmass <- lm(log10(PIEmass) ~ distance, data = subset(PomeShort_late,subject=="child"))

> summary(FinalPomeShort_late_childPIEmass)

Call:
lm(formula = log10(PIEmass) ~ distance, data = subset(PomeShort_late,
   subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.76247 -0.08236  0.04141  0.12254  0.31143

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -0.957349   0.104398  -9.170 1.02e-10 ***
distance    -0.023845   0.008353  -2.855  0.00729 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2047 on 34 degrees of freedom
Multiple R-squared:  0.1933,          Adjusted R-squared:  0.1696
F-statistic: 8.149 on 1 and 34 DF,  p-value: 0.007288


> FinalPomeShort_late_adultPDEmass <- lm(log10(PDEmass) ~ tree_ht + amount + distance + cat, data =
subset(PomeShort_late,subject=="adult"))

> summary(FinalPomeShort_late_adultPDEmass)

Call:
lm(formula = log10(PDEmass) ~ tree_ht + amount + distance + cat,
   data = subset(PomeShort_late, subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.47568 -0.12991  0.02681  0.12844  0.57439

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) 10.793287   0.574739  18.779  < 2e-16 ***
tree_ht     -1.779299   0.233636  -7.616 1.38e-08 ***
```

```
amount      -0.016860   0.005305  -3.178  0.00335 **
distance    -0.061469   0.009095  -6.758 1.45e-07 ***
catS        -0.233823   0.169172  -1.382  0.17680
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2228 on 31 degrees of freedom
Multiple R-squared:  0.921,             Adjusted R-squared:  0.9108
F-statistic: 90.32 on 4 and 31 DF,  p-value: < 2.2e-16


> FinalPomeShort_late_adultADEmass <- lm(log10(ADEmass) ~ tree_ht + cat + distance +speed, data =
subset(PomeShort_late,subject=="adult"))

> summary(FinalPomeShort_late_adultADEmass)

Call:
lm(formula = log10(ADEmass) ~ tree_ht + cat + distance + speed,
    data = subset(PomeShort_late, subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.48630 -0.12761  0.00579  0.12766  0.52998

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) 11.878644   0.619295  19.181  < 2e-16 ***
tree_ht     -2.705607   0.178208 -15.182 6.66e-16 ***
catS        -1.016614   0.163133  -6.232 6.36e-07 ***
distance    -0.058065   0.009356  -6.206 6.83e-07 ***
speed        0.043737   0.030559   1.431    0.162
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2292 on 31 degrees of freedom
Multiple R-squared:  0.9213,            Adjusted R-squared:  0.9112
F-statistic: 90.78 on 4 and 31 DF,  p-value: < 2.2e-16


> FinalPomeShort_late_adultPIEmass <- lm(log10(PIEmass) ~ distance + speed, data = subset(PomeShort_late,subject=="adult"))

> summary(FinalPomeShort_late_adultPIEmass)

Call:
lm(formula = log10(PIEmass) ~ distance + speed, data = subset(PomeShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.75932 -0.12492  0.01759  0.16374  0.59155

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.20935    0.19035  -6.353 3.92e-07 ***
distance    -0.02615    0.01077  -2.428    0.021 *
speed        0.03794    0.02245   1.690    0.101
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.258 on 32 degrees of freedom
  (1 observation deleted due to missingness)
Multiple R-squared:  0.2193,            Adjusted R-squared:  0.1705
F-statistic: 4.495 on 2 and 32 DF,  p-value: 0.01903
```

Vine Fruit Early Scenario

```
> FinalVineShort_early_childPDEmass <- lm(log10(PDEmass) ~ deviation + distance + wind, data = subset(VineShort_early,subject=="child"))

> summary(FinalVineShort_early_childPDEmass)

Call:
lm(formula = log10(PDEmass) ~ deviation + distance + wind, data = subset(VineShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.59211 -0.09873  0.02960  0.10884  0.43362

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  1.629681   0.161639  10.082 1.85e-11 ***
deviation   -0.024270   0.002117 -11.463 7.25e-13 ***
distance    -0.066635   0.009196  -7.246 3.12e-08 ***
wind         0.244612   0.055487   4.408  0.00011 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
```

```
Residual standard error: 0.2253 on 32 degrees of freedom
Multiple R-squared:  0.8506,          Adjusted R-squared:  0.8365
F-statistic: 60.71 on 3 and 32 DF,  p-value: 2.651e-13

> FinalVineShort_early_childADEmass <- lm(log10(ADEmass) ~ deviation + distance + wind, data = subset(VineShort_early,subject=="child"))

> summary(FinalVineShort_early_childADEmass)

Call:
lm(formula = log10(ADEmass) ~ deviation + distance + wind, data = subset(VineShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.62837 -0.07697  0.01904  0.11209  0.51615

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  1.540769   0.164451   9.369 1.09e-10 ***
deviation   -0.023366   0.002154 -10.847 2.99e-12 ***
distance    -0.067653   0.009356  -7.231 3.25e-08 ***
wind         0.222223   0.056453   3.936 0.000419 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2292 on 32 degrees of freedom
Multiple R-squared:  0.8399,          Adjusted R-squared:  0.8249
F-statistic: 55.98 on 3 and 32 DF,  p-value: 7.898e-13

> FinalVineShort_early_childPIEmass <- lm(log10(PIEmass) ~ conc + vol + distance, data = subset(VineShort_early,subject=="child"))

> summary(FinalVineShort_early_childPIEmass)

Call:
lm(formula = log10(PIEmass) ~ conc + vol + distance, data = subset(VineShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.28881 -0.13157 -0.01274  0.09564  0.40751

Coefficients:
              Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.9544618  0.1227421 -15.923  < 2e-16 ***
conc         3.0674701  0.2816402  10.891 2.69e-12 ***
vol          0.0008738  0.0001974   4.426 0.000105 ***
distance    -0.0277399  0.0071665  -3.871 0.000503 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1755 on 32 degrees of freedom
Multiple R-squared:  0.8099,          Adjusted R-squared:  0.792
F-statistic: 45.44 on 3 and 32 DF,  p-value: 1.22e-11

> FinalVineShort_early_adultPDEmass <- lm(log10(PDEmass) ~ amount + + speed + distance, data = subset(VineShort_early,subject=="adult"))

> summary(FinalVineShort_early_adultPDEmass)

Call:
lm(formula = log10(PDEmass) ~ amount + +speed + distance, data = subset(VineShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.57908 -0.14185  0.04518  0.14252  0.42331

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  0.886937   0.161016   5.508 4.52e-06 ***
amount       0.021227   0.002590   8.195 2.32e-09 ***
speed        0.156396   0.023611   6.624 1.81e-07 ***
distance    -0.061728   0.009497  -6.500 2.57e-07 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2327 on 32 degrees of freedom
Multiple R-squared:  0.8444,          Adjusted R-squared:  0.8298
F-statistic: 57.88 on 3 and 32 DF,  p-value: 5.039e-13

> FinalVineShort_early_adultADEmass <- lm(log10(ADEmass) ~ deviation + distance + wind, data = subset(VineShort_early,subject=="adult"))

> summary(FinalVineShort_early_adultADEmass)

Call:
```

```
lm(formula = log10(ADEmass) ~ deviation + distance + wind, data = subset(VineShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.58782 -0.12456 -0.01319  0.18494  0.33188

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  1.517526   0.169600   8.948 3.20e-10 ***
deviation   -0.023205   0.002221 -10.446 7.72e-12 ***
distance    -0.060954   0.009649  -6.317 4.34e-07 ***
wind         0.339535   0.058220   5.832 1.77e-06 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2364 on 32 degrees of freedom
Multiple R-squared:  0.828,             Adjusted R-squared:  0.8118
F-statistic: 51.34 on 3 and 32 DF,  p-value: 2.488e-12


> FinalVineShort_early_adultPIEmass <- lm(log10(PIEmass) ~ conc + vol + distance, data = subset(VineShort_early,subject=="adult"))

> summary(FinalVineShort_early_adultPIEmass)

Call:
lm(formula = log10(PIEmass) ~ conc + vol + distance, data = subset(VineShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.22902 -0.08076 -0.02549  0.06829  0.31281

Coefficients:
              Estimate Std. Error t value Pr(>|t|)
(Intercept) -2.0280883  0.0959771 -21.131  < 2e-16 ***
conc         2.6757190  0.2202262  12.150 1.58e-13 ***
vol          0.0005305  0.0001544   3.436  0.00165 **
distance    -0.0166376  0.0056038  -2.969  0.00562 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1373 on 32 degrees of freedom
Multiple R-squared:  0.8412,            Adjusted R-squared:  0.8263
F-statistic: 56.52 on 3 and 32 DF,  p-value: 6.947e-13
```

Vine Fruit Late Scenario

```
> FinalVineShort_late_childPDEmass <- lm(log10(PDEmass) ~ nozzle_no + vol + distance, data = subset(VineShort_late,subject=="child"))

> summary(FinalVineShort_late_childPDEmass)

Call:
lm(formula = log10(PDEmass) ~ nozzle_no + vol + distance, data = subset(VineShort_late,
    subject == "child"))

Residuals:
      Min        1Q    Median        3Q       Max
-0.264930 -0.054164 -0.003636  0.076461  0.294539

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  2.534e-01  1.128e-01   2.247   0.0317 *
nozzle_no    9.111e-02  6.094e-03  14.951 5.46e-16 ***
vol          7.845e-04  8.757e-05   8.959 3.11e-10 ***
distance    -4.523e-02  5.753e-03  -7.861 5.72e-09 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1409 on 32 degrees of freedom
Multiple R-squared:  0.901,             Adjusted R-squared:  0.8917
F-statistic:  97.1 on 3 and 32 DF,  p-value: 3.731e-16


> FinalVineShort_late_childADEmass <- lm(log10(ADEmass) ~ nozzle_no + vol + distance, data = subset(VineShort_late,subject=="child"))

> summary(FinalVineShort_late_childADEmass)

Call:
lm(formula = log10(ADEmass) ~ nozzle_no + vol + distance, data = subset(VineShort_late,
    subject == "child"))

Residuals:
      Min        1Q    Median        3Q       Max
-0.296419 -0.087452  0.000773  0.073310  0.292174
```

```
Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  4.590e-02  1.151e-01   0.399    0.693
nozzle_no    8.536e-02  6.218e-03  13.727 5.86e-15 ***
vol          7.568e-04  8.935e-05   8.469 1.12e-09 ***
distance    -4.051e-02  5.870e-03  -6.901 8.23e-08 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1438 on 32 degrees of freedom
Multiple R-squared:  0.8833,          Adjusted R-squared:  0.8724
F-statistic: 80.75 on 3 and 32 DF,  p-value: 5.139e-15


> FinalVineShort_late_childPIEmass <- lm(log10(PIEmass) ~ distance + conc, data = subset(VineShort_late,subject=="child"))

> summary(FinalVineShort_late_childPIEmass)

Call:
lm(formula = log10(PIEmass) ~ distance + conc, data = subset(VineShort_late,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.75978 -0.07443  0.04730  0.19193  0.36073

Coefficients:
            Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.11247    0.15805  -7.039  4.7e-08 ***
distance    -0.03749    0.01112  -3.370  0.00193 **
conc         1.18785    0.35330   3.362  0.00197 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2725 on 33 degrees of freedom
Multiple R-squared:  0.4144,          Adjusted R-squared:  0.3789
F-statistic: 11.68 on 2 and 33 DF,  p-value: 0.0001463


> FinalVineShort_late_adultPDEmass <- lm(log10(PDEmass) ~ quality + amount + distance, data = subset(VineShort_late,subject=="adult"))

> summary(FinalVineShort_late_adultPDEmass)

Call:
lm(formula = log10(PDEmass) ~ quality + amount + distance, data = subset(VineShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.24416 -0.09401 -0.01579  0.10956  0.29452

Coefficients:
                Estimate Std. Error t value Pr(>|t|)
(Intercept)     6.652241   0.582991  11.411 1.25e-12 ***
qualityunknown -2.057684   0.175133 -11.749 5.97e-13 ***
qualityVF       0.512039   0.081505   6.282 5.51e-07 ***
amount         -0.070876   0.008893  -7.970 5.35e-09 ***
distance       -0.040622   0.006160  -6.595 2.28e-07 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1509 on 31 degrees of freedom
Multiple R-squared:  0.911,           Adjusted R-squared:  0.8995
F-statistic: 79.33 on 4 and 31 DF,  p-value: 7.85e-16


> FinalVineShort_late_adultADEmass <- lm(log10(ADEmass) ~ quality + amount + distance, data = subset(VineShort_late,subject=="adult"))

> summary(FinalVineShort_late_adultADEmass)

Call:
lm(formula = log10(ADEmass) ~ quality + amount + distance, data = subset(VineShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.26728 -0.07680 -0.02601  0.09130  0.36445

Coefficients:
                Estimate Std. Error t value Pr(>|t|)
(Intercept)     5.663666   0.608655   9.305 1.74e-10 ***
qualityunknown -2.006407   0.182842 -10.973 3.33e-12 ***
qualityVF       0.409645   0.085093   4.814 3.66e-05 ***
amount         -0.059155   0.009284  -6.372 4.28e-07 ***
distance       -0.040366   0.006431  -6.277 5.59e-07 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
```

```
Residual standard error: 0.1575 on 31 degrees of freedom
Multiple R-squared:  0.9163,          Adjusted R-squared:  0.9055
F-statistic: 84.83 on 4 and 31 DF,  p-value: 3.056e-16

> FinalVineShort_late_adultPIEmass <- lm(log10(PIEmass) ~ tree_ht + distance, data = subset(VineShort_late,subject=="adult"))

> summary(FinalVineShort_late_adultPIEmass)

Call:
lm(formula = log10(PIEmass) ~ tree_ht + distance, data = subset(VineShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.72269 -0.05347  0.03060  0.07546  0.22296

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -0.790812   0.096922  -8.159 2.03e-09 ***
tree_ht     -0.145512   0.040008  -3.637  0.00093 ***
distance    -0.020353   0.006892  -2.953  0.00576 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1688 on 33 degrees of freedom
Multiple R-squared:  0.4067,          Adjusted R-squared:  0.3708
F-statistic: 11.31 on 2 and 33 DF,  p-value: 0.0001813
```

Regression using exposure ml spray

Pome Fruit Early Scenario

```
> FinalPomeShort_early_childPDE <- lm(log10(PDE) ~ DT + distance + spacing + cat, data = subset(PomeShort_early,subject=="child"))

> summary(FinalPomeShort_early_childPDE)

Call:
lm(formula = log10(PDE) ~ DT + distance + spacing + cat, data = subset(PomeShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.31351 -0.09225 -0.01847  0.05837  0.74105

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -2.365970   1.038951  -2.277  0.02982 *
DT          -0.171485   0.020570  -8.337 2.04e-09 ***
distance    -0.045833   0.007397  -6.196 7.03e-07 ***
spacing      1.009477   0.290275   3.478  0.00152 **
catS        -0.145050   0.102090  -1.421  0.16535
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1812 on 31 degrees of freedom
Multiple R-squared:  0.8247,          Adjusted R-squared:  0.8021
F-statistic: 36.47 on 4 and 31 DF,  p-value: 2.613e-11

> FinalPomeShort_early_childADE <- lm(log10(ADE) ~ DT + distance + spacing, data = subset(PomeShort_early,subject=="child"))

> summary(FinalPomeShort_early_childADE)

Call:
lm(formula = log10(ADE) ~ DT + distance + spacing, data = subset(PomeShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.33319 -0.10017 -0.00465  0.06588  0.73448

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.373490   0.709032  -1.937  0.06160 .
DT          -0.142574   0.015717  -9.071 2.33e-10 ***
distance    -0.042752   0.007195  -5.942 1.28e-06 ***
spacing      0.633429   0.193350   3.276  0.00253 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1762 on 32 degrees of freedom
Multiple R-squared:  0.806,           Adjusted R-squared:  0.7878
F-statistic: 44.32 on 3 and 32 DF,  p-value: 1.68e-11
```

```
> FinalPomeShort_early_childPIE <- lm(log10(PIE.B) ~ distance + cat, data = subset(PomeShort_early,subject=="child"))

> summary(FinalPomeShort_early_childPIE)

Call:
lm(formula = log10(PIE.B) ~ distance + cat, data = subset(PomeShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-2.47391 -0.15747  0.09715  0.27541  0.80484

Coefficients:
            Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.17008    0.29109  -4.020 0.000318 ***
distance    -0.05590    0.02277  -2.455 0.019531 *
catS        -0.45154    0.21473  -2.103 0.043194 *
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.5579 on 33 degrees of freedom
Multiple R-squared:  0.2427,          Adjusted R-squared:  0.1968
F-statistic: 5.289 on 2 and 33 DF,  p-value: 0.01018


> FinalPomeShort_early_adultPDE <- lm(log10(PDE) ~ DT + distance, data = subset(PomeShort_early,subject=="adult"))

> summary(FinalPomeShort_early_adultPDE)

Call:
lm(formula = log10(PDE) ~ DT + distance, data = subset(PomeShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.36821 -0.10374  0.00502  0.10043  0.52792

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  1.721451   0.105564  16.307  < 2e-16 ***
DT          -0.129465   0.012532 -10.331 7.12e-12 ***
distance    -0.040646   0.007724  -5.263 8.50e-06 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1892 on 33 degrees of freedom
Multiple R-squared:  0.8051,          Adjusted R-squared:  0.7933
F-statistic: 68.15 on 2 and 33 DF,  p-value: 1.919e-12


> FinalPomeShort_early_adultADE <- lm(log10(ADE) ~ DT + distance +speed, data = subset(PomeShort_early,subject=="adult"))

> summary(FinalPomeShort_early_adultADE)

Call:
lm(formula = log10(ADE) ~ DT + distance + speed, data = subset(PomeShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.29569 -0.08432  0.01400  0.08301  0.54941

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  0.538751   0.350831   1.536 0.134456
DT          -0.081689   0.018460  -4.425 0.000105 ***
distance    -0.032741   0.006652  -4.922 2.49e-05 ***
speed        0.134097   0.056890   2.357 0.024701 *
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.163 on 32 degrees of freedom
Multiple R-squared:  0.8231,          Adjusted R-squared:  0.8065
F-statistic: 49.63 on 3 and 32 DF,  p-value: 3.883e-12


> FinalPomeShort_early_adultPIE <- lm(log10(PIE.B) ~ area + distance, data = subset(PomeShort_early,subject=="adult"))

> summary(FinalPomeShort_early_adultPIE)

Call:
lm(formula = log10(PIE.B) ~ area + distance, data = subset(PomeShort_early,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.65012 -0.15996  0.01313  0.17321  0.54400
```

```
Coefficients:
            Estimate Std. Error t value Pr(>|t|)
(Intercept) -0.30112    0.24226  -1.243   0.2226
area        -1.46949    0.18638  -7.884 4.33e-09 ***
distance    -0.02113    0.01086  -1.946   0.0602 .
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.266 on 33 degrees of freedom
Multiple R-squared:  0.6676,          Adjusted R-squared:  0.6474
F-statistic: 33.13 on 2 and 33 DF,  p-value: 1.283e-08
```

Pome Fruit Late Scenario

```
> FinalPomeShort_late_childPDE <- lm(log10(PDE) ~ tree_ht + cat+ distance + vol, data = subset(PomeShort_late,subject=="child"))

> summary(FinalPomeShort_late_childPDE)

Call:
lm(formula = log10(PDE) ~ tree_ht + cat + distance + vol, data = subset(PomeShort_late,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.48621 -0.12949  0.00906  0.14505  0.31736

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept)  9.3030044  0.7456585  12.476 1.27e-13 ***
tree_ht     -2.3079914  0.1408244 -16.389  < 2e-16 ***
catS        -1.1532134  0.1698553  -6.789 1.33e-07 ***
distance    -0.0551227  0.0083476  -6.603 2.23e-07 ***
vol         -0.0008463  0.0003918  -2.160   0.0386 *
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2045 on 31 degrees of freedom
Multiple R-squared:  0.9155,          Adjusted R-squared:  0.9046
F-statistic:    84 on 4 and 31 DF,  p-value: 3.511e-16

> FinalPomeShort_late_childADE <- lm(log10(ADE) ~ tree_ht + cat + distance, data = subset(PomeShort_late,subject=="child"))

> summary(FinalPomeShort_late_childADE)

Call:
lm(formula = log10(ADE) ~ tree_ht + cat + distance, data = subset(PomeShort_late,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.41484 -0.13518  0.01653  0.12770  0.29953

Coefficients:
            Estimate Std. Error t value Pr(>|t|)
(Intercept)  8.114843   0.499762  16.237  < 2e-16 ***
tree_ht     -2.294310   0.130648 -17.561  < 2e-16 ***
catS        -0.990983   0.092371 -10.728 3.95e-12 ***
distance    -0.054159   0.007999  -6.771 1.19e-07 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1959 on 32 degrees of freedom
Multiple R-squared:  0.9192,          Adjusted R-squared:  0.9117
F-statistic: 121.4 on 3 and 32 DF,  p-value: < 2.2e-16

> FinalPomeShort_late_childPIE <- lm(log10(PIE.B) ~ conc + distance, data = subset(PomeShort_late,subject=="child"))

> summary(FinalPomeShort_late_childPIE)

Call:
lm(formula = log10(PIE.B) ~ conc + distance, data = subset(PomeShort_late,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.77792 -0.09564  0.04003  0.12738  0.29945

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.342357   0.165426  -8.115 2.29e-09 ***
conc        -3.902243   0.906246  -4.306  0.00014 ***
distance    -0.023856   0.008405  -2.838  0.00770 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1
```

```
Residual standard error: 0.2059 on 33 degrees of freedom
Multiple R-squared:  0.4442,          Adjusted R-squared:  0.4105
F-statistic: 13.19 on 2 and 33 DF,  p-value: 6.184e-05

> FinalPomeShort_late_adultPDE <- lm(log10(PDE) ~ tree_ht + cat + distance + vol, data = subset(PomeShort_late,subject=="adult"))

> summary(FinalPomeShort_late_adultPDE)

Call:
lm(formula = log10(PDE) ~ tree_ht + cat + distance + vol, data = subset(PomeShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.47391 -0.12326  0.03429  0.13465  0.58562

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) 11.4981478  0.8287203  13.875 7.68e-15 ***
tree_ht     -2.6859638  0.1565114 -17.161  < 2e-16 ***
catS        -1.3760462  0.1887761  -7.289 3.34e-08 ***
distance    -0.0626687  0.0092774  -6.755 1.46e-07 ***
vol         -0.0011810  0.0004354  -2.712   0.0108 *
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2272 on 31 degrees of freedom
Multiple R-squared:  0.9222,          Adjusted R-squared:  0.9122
F-statistic: 91.89 on 4 and 31 DF,  p-value: < 2.2e-16

> FinalPomeShort_late_adultADE <- lm(log10(ADE) ~ tree_ht + cat + distance +speed, data = subset(PomeShort_late,subject=="adult"))

> summary(FinalPomeShort_late_adultADE)

Call:
lm(formula = log10(ADE) ~ tree_ht + cat + distance + speed, data = subset(PomeShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.48228 -0.14900  0.00213  0.13525  0.53410

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) 10.544832   0.640531  16.463  < 2e-16 ***
tree_ht     -2.880064   0.184318 -15.625 3.02e-16 ***
catS        -1.259828   0.168727  -7.467 2.06e-08 ***
distance    -0.056396   0.009677  -5.828 2.00e-06 ***
speed        0.028713   0.031607   0.908    0.371
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.237 on 31 degrees of freedom
Multiple R-squared:  0.921,          Adjusted R-squared:  0.9108
F-statistic: 90.37 on 4 and 31 DF,  p-value: < 2.2e-16

> FinalPomeShort_late_adultPIE <- lm(log10(PIE.B) ~ distance + amount, data = subset(PomeShort_late,subject=="adult"))

> summary(FinalPomeShort_late_adultPIE)

Call:
lm(formula = log10(PIE.B) ~ distance + amount, data = subset(PomeShort_late,
    subject == "adult"))

Residuals:
    Min      1Q  Median      3Q     Max
-0.7061 -0.1478  0.0181  0.1844  0.5736

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.210934   0.445877  -2.716   0.0106 *
distance    -0.026269   0.010854  -2.420   0.0214 *
amount      -0.005265   0.003702  -1.422   0.1646
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.26 on 32 degrees of freedom
  (1 observation deleted due to missingness)
Multiple R-squared:  0.2009,          Adjusted R-squared:  0.1509
F-statistic: 4.022 on 2 and 32 DF,  p-value: 0.02766
```

Vine Fruit Early Scenario

```
> FinalVineShort_early_childPDE <- lm(log10(PDE) ~ vol + distance + tree_ht, data = subset(VineShort_early,subject=="child"))

> summary(FinalVineShort_early_childPDE)

Call:
lm(formula = log10(PDE) ~ vol + distance + tree_ht, data = subset(VineShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.53158 -0.11590  0.03399  0.12877  0.45099

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -0.503314   0.137964  -3.648  0.00093 ***
vol          0.002529   0.000220  11.493 6.78e-13 ***
distance    -0.068410   0.009098  -7.519 1.46e-08 ***
tree_ht     -0.277919   0.056664  -4.905 2.62e-05 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2229 on 32 degrees of freedom
Multiple R-squared:  0.8552,          Adjusted R-squared:  0.8416
F-statistic: 62.97 on 3 and 32 DF,  p-value: 1.612e-13


> FinalVineShort_early_childADE <- lm(log10(ADE) ~ vol + distance + speed, data = subset(VineShort_early,subject=="child"))

> summary(FinalVineShort_early_childADE)

Call:
lm(formula = log10(ADE) ~ vol + distance + speed, data = subset(VineShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.49117 -0.08717 -0.01150  0.10782  0.53909

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.257600   0.143871  -8.741 5.47e-10 ***
vol          0.002171   0.000207  10.489 6.95e-12 ***
distance    -0.067202   0.008804  -7.633 1.07e-08 ***
speed        0.089542   0.021764   4.114 0.000254 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2157 on 32 degrees of freedom
Multiple R-squared:  0.8588,          Adjusted R-squared:  0.8455
F-statistic: 64.87 on 3 and 32 DF,  p-value: 1.076e-13


> FinalVineShort_early_childPIE <- lm(log10(PIE.B) ~ deviation + distance + DT, data = subset(VineShort_early,subject=="child"))

> summary(FinalVineShort_early_childPIE)

Call:
lm(formula = log10(PIE.B) ~ deviation + distance + DT, data = subset(VineShort_early,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.25677 -0.11022  0.00505  0.07966  0.46865

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -2.527121   0.195260 -12.942 2.92e-14 ***
deviation   -0.013927   0.002958  -4.708 4.63e-05 ***
distance    -0.027245   0.006607  -4.123 0.000247 ***
DT           0.118372   0.041952   2.822 0.008144 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1619 on 32 degrees of freedom
Multiple R-squared:  0.5876,          Adjusted R-squared:  0.5489
F-statistic:  15.2 on 3 and 32 DF,  p-value: 2.532e-06


> FinalVineShort_early_adultPDE <- lm(log10(PDE) ~ vol + speed + distance, data = subset(VineShort_early,subject=="adult"))

> summary(FinalVineShort_early_adultPDE)

Call:
lm(formula = log10(PDE) ~ vol + speed + distance, data = subset(VineShort_early,
    subject == "adult"))

Residuals:
```

```
    Min      1Q  Median      3Q     Max
-0.60928 -0.12581  0.00631  0.14734  0.36941

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -0.9622978  0.1512886  -6.361 3.84e-07 ***
vol          0.0016652  0.0002177   7.650 1.02e-08 ***
speed        0.1534782  0.0228863   6.706 1.43e-07 ***
distance    -0.0612961  0.0092579  -6.621 1.82e-07 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2268 on 32 degrees of freedom
Multiple R-squared:  0.8332,          Adjusted R-squared:  0.8176
F-statistic:  53.3 on 3 and 32 DF,  p-value: 1.517e-12


> FinalVineShort_early_adultADE <- lm(log10(ADE) ~ vol + distance + speed, data = subset(VineShort_early,subject=="adult"))

> summary(FinalVineShort_early_adultADE)

Call:
lm(formula = log10(ADE) ~ vol + distance + speed, data = subset(VineShort_early,
    subject == "adult"))

Residuals:
    Min      1Q  Median      3Q     Max
-0.58076 -0.11897  0.01408  0.16620  0.35450

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -1.1182365  0.1578894  -7.082 4.93e-08 ***
vol          0.0018608  0.0002272   8.191 2.35e-09 ***
distance    -0.0617499  0.0096618  -6.391 3.52e-07 ***
speed        0.1414686  0.0238849   5.923 1.36e-06 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2367 on 32 degrees of freedom
Multiple R-squared:  0.8286,          Adjusted R-squared:  0.8126
F-statistic: 51.57 on 3 and 32 DF,  p-value: 2.342e-12


> FinalVineShort_early_adultPIE <- lm(log10(PIE.B) ~ vol + distance, data = subset(VineShort_early,subject=="adult"))

> summary(FinalVineShort_early_adultPIE)

Call:
lm(formula = log10(PIE.B) ~ vol + distance, data = subset(VineShort_early,
    subject == "adult"))

Residuals:
    Min      1Q  Median      3Q     Max
-0.24391 -0.08251 -0.01655  0.07946  0.35139

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -2.3465707  0.0730800 -32.110  < 2e-16 ***
vol          0.0004019  0.0001235   3.253  0.00263 **
distance    -0.0165348  0.0052953  -3.123  0.00372 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1297 on 33 degrees of freedom
Multiple R-squared:  0.3756,          Adjusted R-squared:  0.3378
F-statistic: 9.927 on 2 and 33 DF,  p-value: 0.0004215
```

Vine Fruit Late Scenario

```
> FinalVineShort_late_childPDE <- lm(log10(PDE) ~ quality + distance + spacing, data = subset(VineShort_late,subject=="child"))

> summary(FinalVineShort_late_childPDE)

Call:
lm(formula = log10(PDE) ~ quality + distance + spacing, data = subset(VineShort_late,
    subject == "child"))

Residuals:
      Min        1Q    Median        3Q       Max
-0.287195 -0.081967 -0.008488  0.114273  0.269832

Coefficients:
                Estimate Std. Error t value Pr(>|t|)
(Intercept)     0.967417   0.413190   2.341   0.0258 *
qualityunknown -1.403269   0.095742 -14.657 1.74e-15 ***
qualityVF      -0.430376   0.067700  -6.357 4.46e-07 ***
distance       -0.040147   0.005863  -6.848 1.13e-07 ***
```

```
spacing        -0.410414   0.135431  -3.030   0.0049 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1436 on 31 degrees of freedom
Multiple R-squared:  0.9276,          Adjusted R-squared:  0.9182
F-statistic: 99.26 on 4 and 31 DF,  p-value: < 2.2e-16


> FinalVineShort_late_childADE <- lm(log10(ADE) ~ quality + distance + vol, data = subset(VineShort_late,subject=="child"))

> summary(FinalVineShort_late_childADE)

Call:
lm(formula = log10(ADE) ~ quality + distance + vol, data = subset(VineShort_late,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.29716 -0.07840 -0.03549  0.11669  0.30490

Coefficients:
                Estimate Std. Error t value Pr(>|t|)
(Intercept)     0.0015852  0.1789694   0.009   0.9930
qualityunknown -1.5915244  0.1534125 -10.374 1.32e-11 ***
qualityVF      -0.6028264  0.1043847  -5.775 2.33e-06 ***
distance       -0.0438065  0.0060546  -7.235 3.88e-08 ***
vol            -0.0004971  0.0001821  -2.730   0.0103 *
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1483 on 31 degrees of freedom
Multiple R-squared:  0.925,           Adjusted R-squared:  0.9153
F-statistic:  95.6 on 4 and 31 DF,  p-value: < 2.2e-16


> FinalVineShort_late_childPIE <- lm(log10(PIE.B) ~ distance + vol, data = subset(VineShort_late,subject=="child"))

> summary(FinalVineShort_late_childPIE)

Call:
lm(formula = log10(PIE.B) ~ distance + vol, data = subset(VineShort_late,
    subject == "child"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.77539 -0.07268  0.05993  0.19550  0.33868

Coefficients:
             Estimate Std. Error t value Pr(>|t|)
(Intercept) -2.1245197  0.1484603 -14.310 1.04e-15 ***
distance    -0.0374989  0.0110840  -3.383  0.00186 **
vol          0.0004780  0.0001502   3.183  0.00317 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.2715 on 33 degrees of freedom
Multiple R-squared:  0.3886,          Adjusted R-squared:  0.3515
F-statistic: 10.49 on 2 and 33 DF,  p-value: 0.0002981


> FinalVineShort_late_adultPDE <- lm(log10(PDE) ~ quality + distance + conc, data = subset(VineShort_late,subject=="adult"))

> summary(FinalVineShort_late_adultPDE)

Call:
lm(formula = log10(PDE) ~ quality + distance + conc, data = subset(VineShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.25174 -0.10732  0.01385  0.09733  0.29011

Coefficients:
               Estimate Std. Error t value Pr(>|t|)
(Intercept)     0.14789    0.09218   1.604    0.119
qualityunknown -1.79180    0.13795 -12.989 4.44e-14 ***
qualityVF      -0.45790    0.08251  -5.550 4.44e-06 ***
distance       -0.04431    0.00618  -7.170 4.64e-08 ***
conc            0.85284    0.37968   2.246    0.032 *
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1514 on 31 degrees of freedom
Multiple R-squared:  0.9493,          Adjusted R-squared:  0.9427
F-statistic:   145 on 4 and 31 DF,  p-value: < 2.2e-16


> FinalVineShort_late_adultADE <- lm(log10(ADE) ~ quality + distance, data = subset(VineShort_late,subject=="adult"))
```

```
> summary(FinalVineShort_late_adultADE)

Call:
lm(formula = log10(ADE) ~ quality + distance, data = subset(VineShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.24790 -0.12835 -0.00881  0.12254  0.42811

Coefficients:
                Estimate Std. Error t value Pr(>|t|)
(Intercept)    -0.118287   0.098652  -1.199    0.239
qualityunknown -1.488747   0.080799 -18.425  < 2e-16 ***
qualityVF      -0.367230   0.069963  -5.249 9.63e-06 ***
distance       -0.034930   0.006994  -4.994 2.02e-05 ***
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1714 on 32 degrees of freedom
Multiple R-squared:  0.9266,          Adjusted R-squared:  0.9198
F-statistic: 134.7 on 3 and 32 DF,  p-value: < 2.2e-16


> FinalVineShort_late_adultPIE <- lm(log10(PIE.B) ~ vol + distance, data = subset(VineShort_late,subject=="adult"))

> summary(FinalVineShort_late_adultPIE)

Call:
lm(formula = log10(PIE.B) ~ vol + distance, data = subset(VineShort_late,
    subject == "adult"))

Residuals:
     Min       1Q   Median       3Q      Max
-0.77612 -0.03554  0.03348  0.08627  0.22083

Coefficients:
              Estimate Std. Error t value Pr(>|t|)
(Intercept) -2.236e+00  9.409e-02 -23.769  < 2e-16 ***
vol          6.548e-04  9.518e-05   6.880 7.43e-08 ***
distance    -2.043e-02  7.025e-03  -2.908  0.00646 **
---
Signif. codes:  0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' ' 1

Residual standard error: 0.1721 on 33 degrees of freedom
Multiple R-squared:  0.6236,          Adjusted R-squared:  0.6008
F-statistic: 27.34 on 2 and 33 DF,  p-value: 9.946e-08
```

E5: Quantile Regression

Following the above analysis, quantile regression was employed (R with the quantreg package) to estimate the 75th and 95th percentile functions verses distance from the sprayer. These functions were also plotted (using the ggplot package) and are shown below.

PDE, ADE & PIE vs crop type, leaf cover and distance

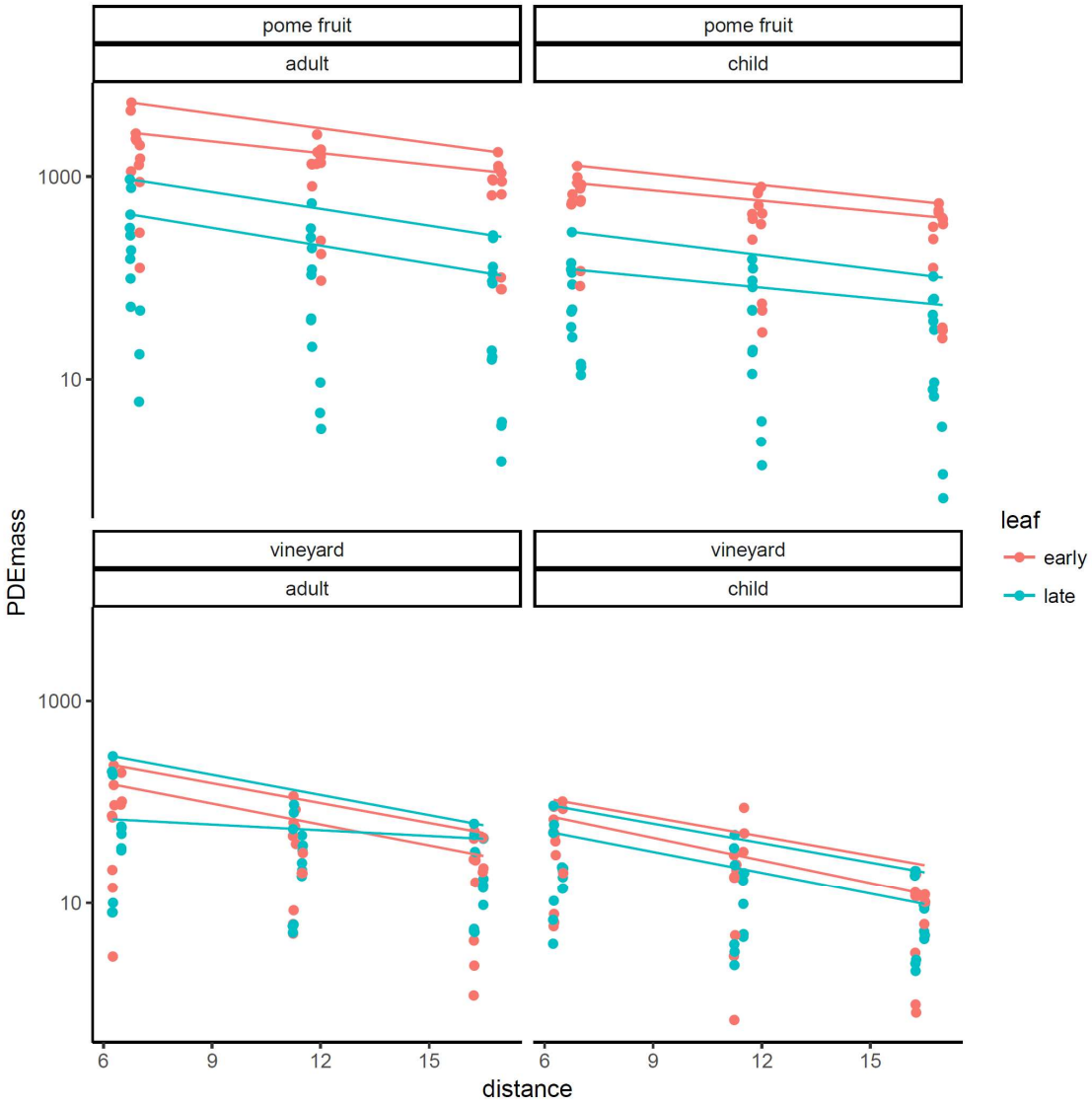


Figure E5.1: Potential dermal exposure (mass active µg/mannequin), fitted curves show quantile regression for 95th and 75th percentiles

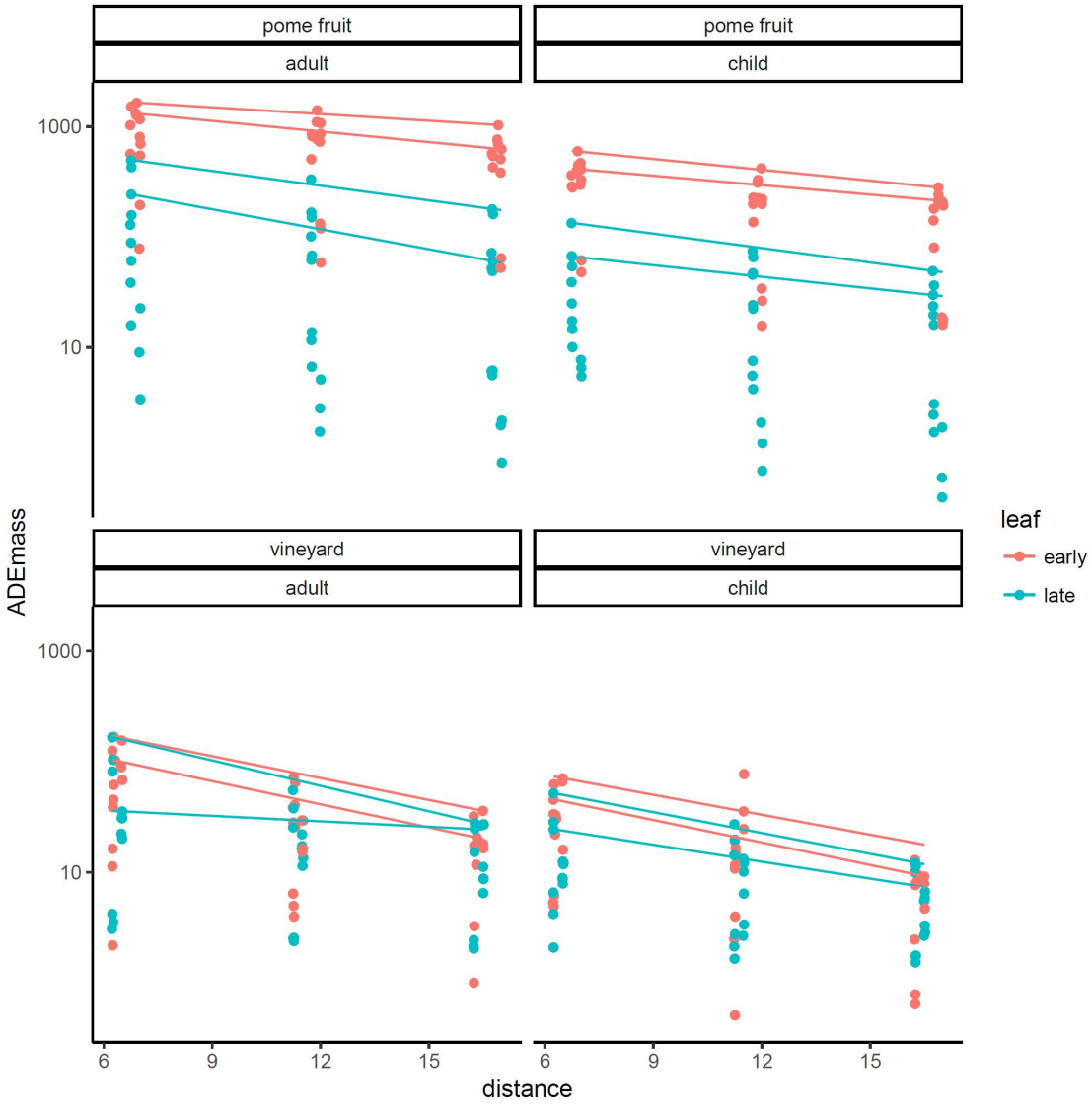


Figure E5.2: Actual dermal exposure (µg/mannequin), fitted curves show quantile regression for 95th and 75th percentiles

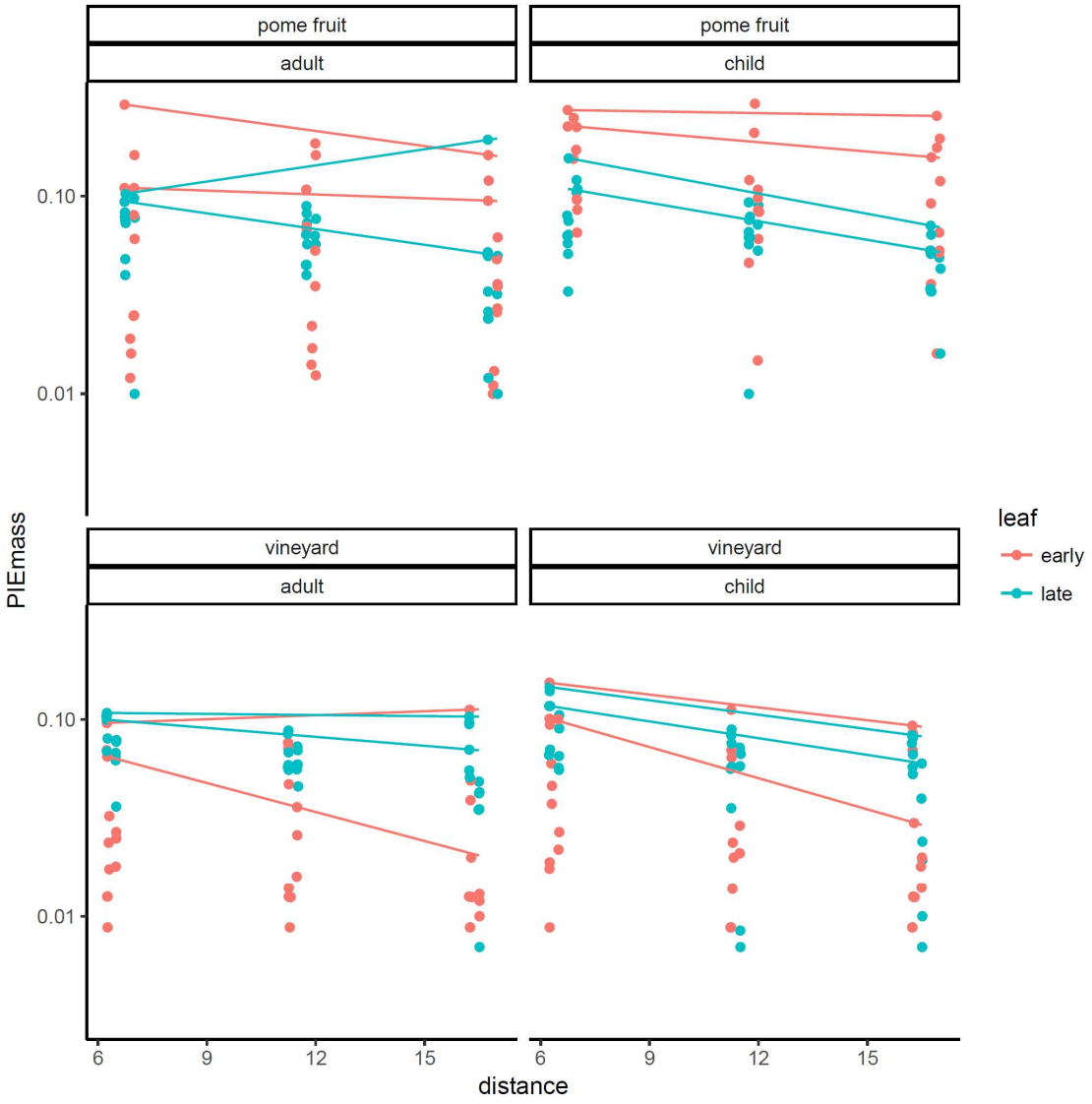


Figure E5.3: Potential inhalation exposure (µg/mannequin), fitted curves show quantile regression for 95th and 75th percentiles

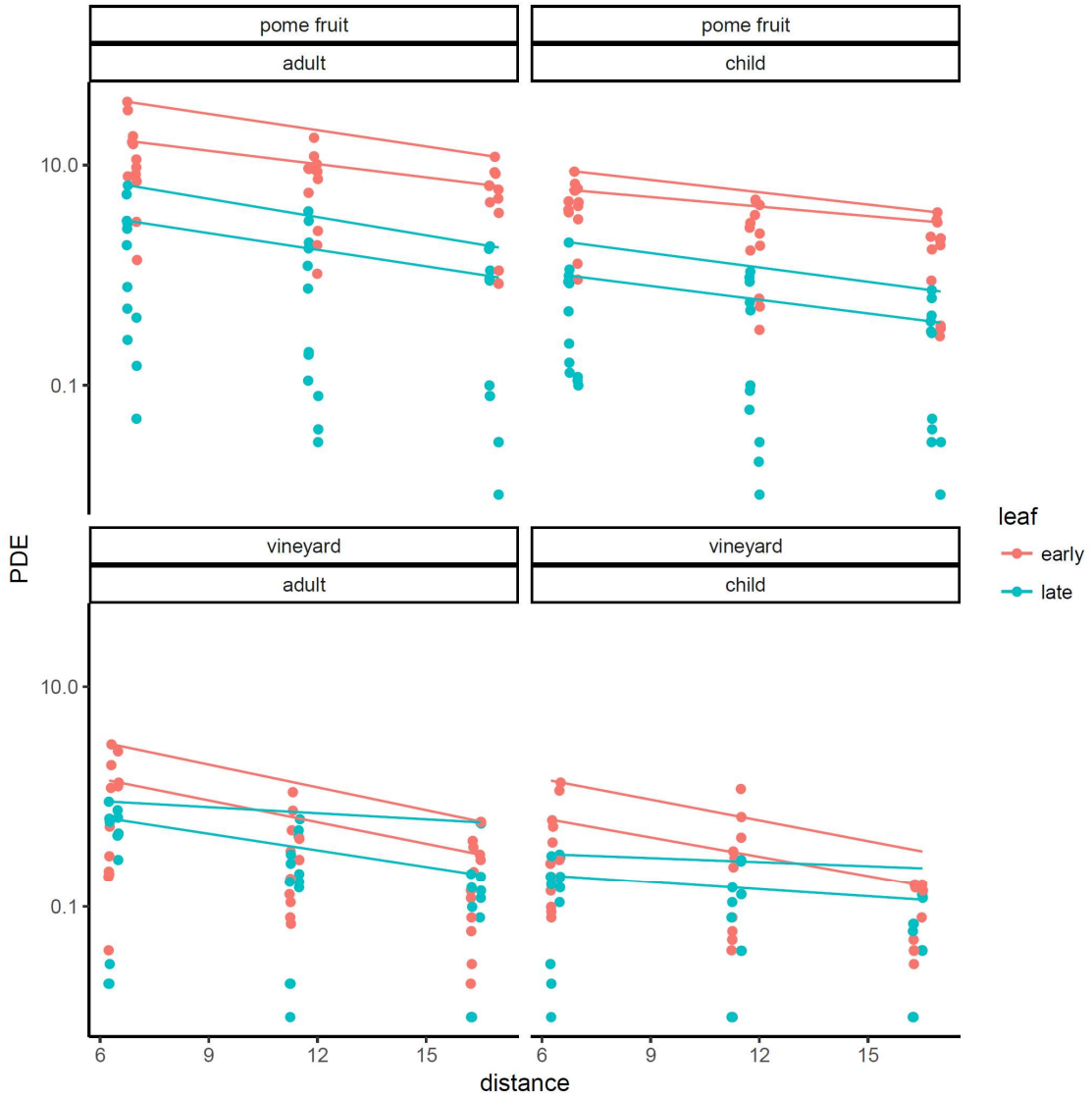


Figure E5.4: Potential dermal exposure (µg/mannequin), fitted curves show quantile regression for 95th and 75th percentiles

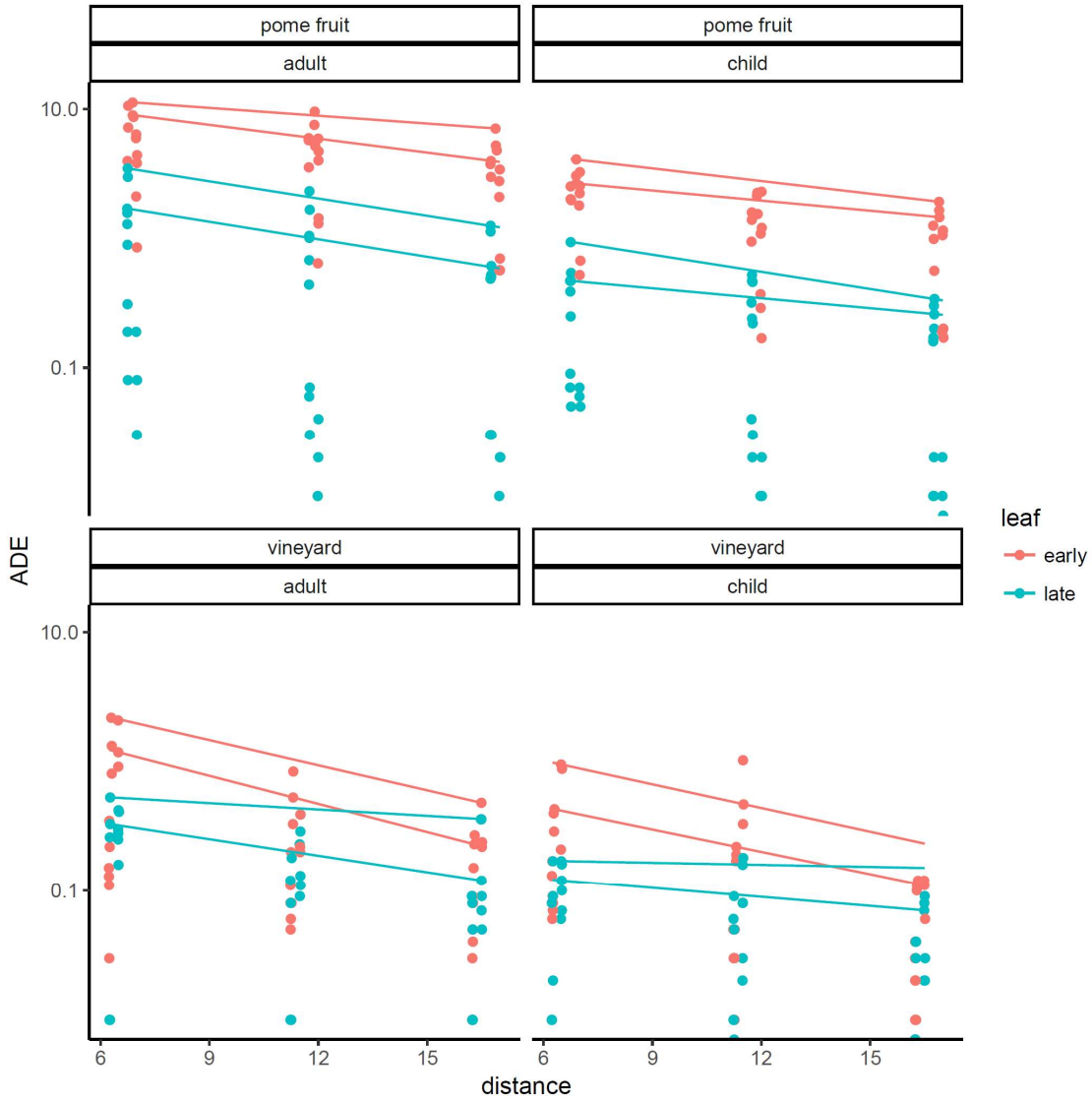


Figure E5.5: Actual dermal exposure (µg/mannequin), fitted curves show quantile regression for 95th and 75th percentiles

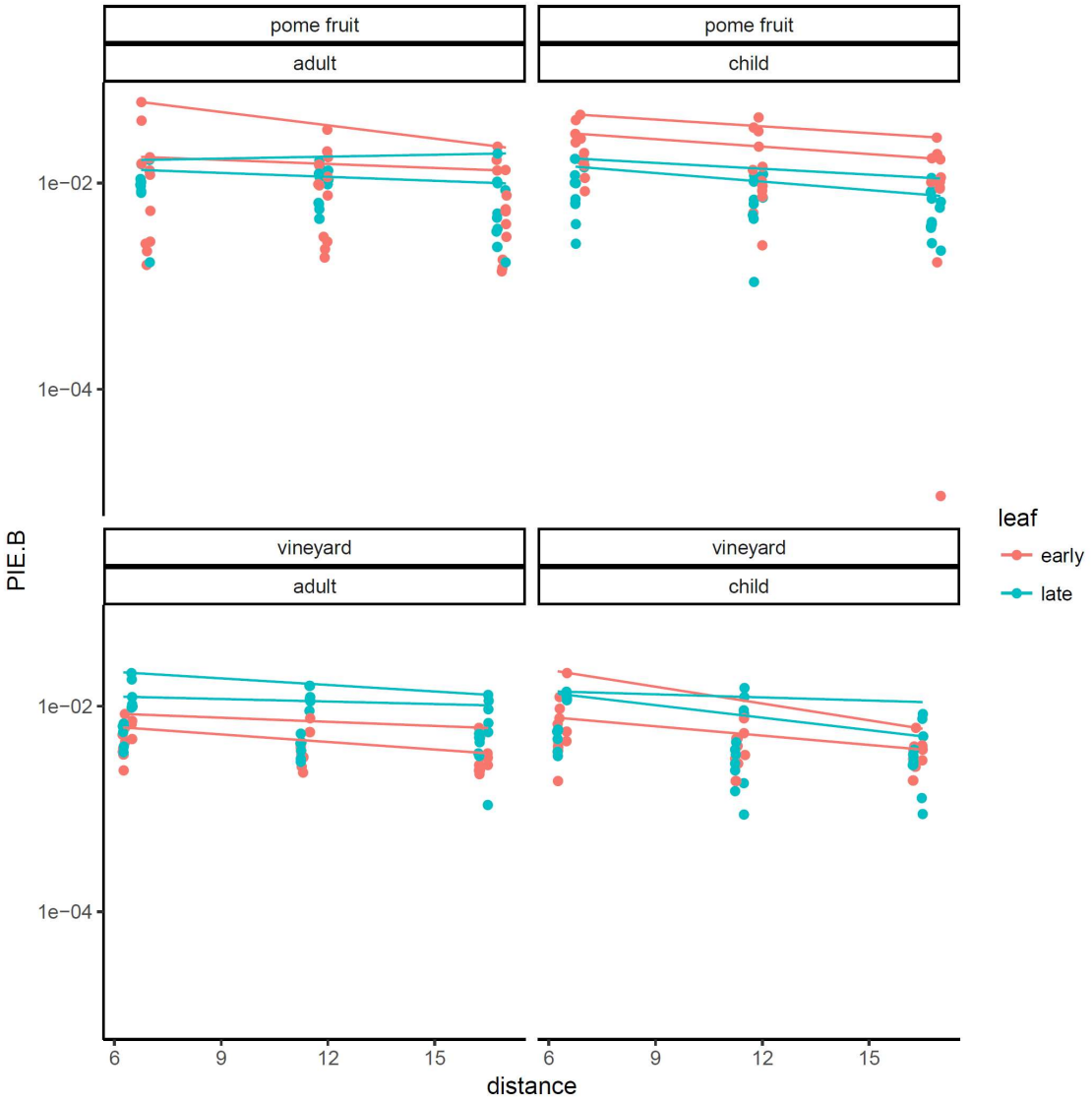


Figure E5.6: Potential inhalation exposure (µg/mannequin), fitted curves show quantile regression for 95th and 75th percentiles

PDE vs wind speed, wind direction, spray volume & a.s. applied

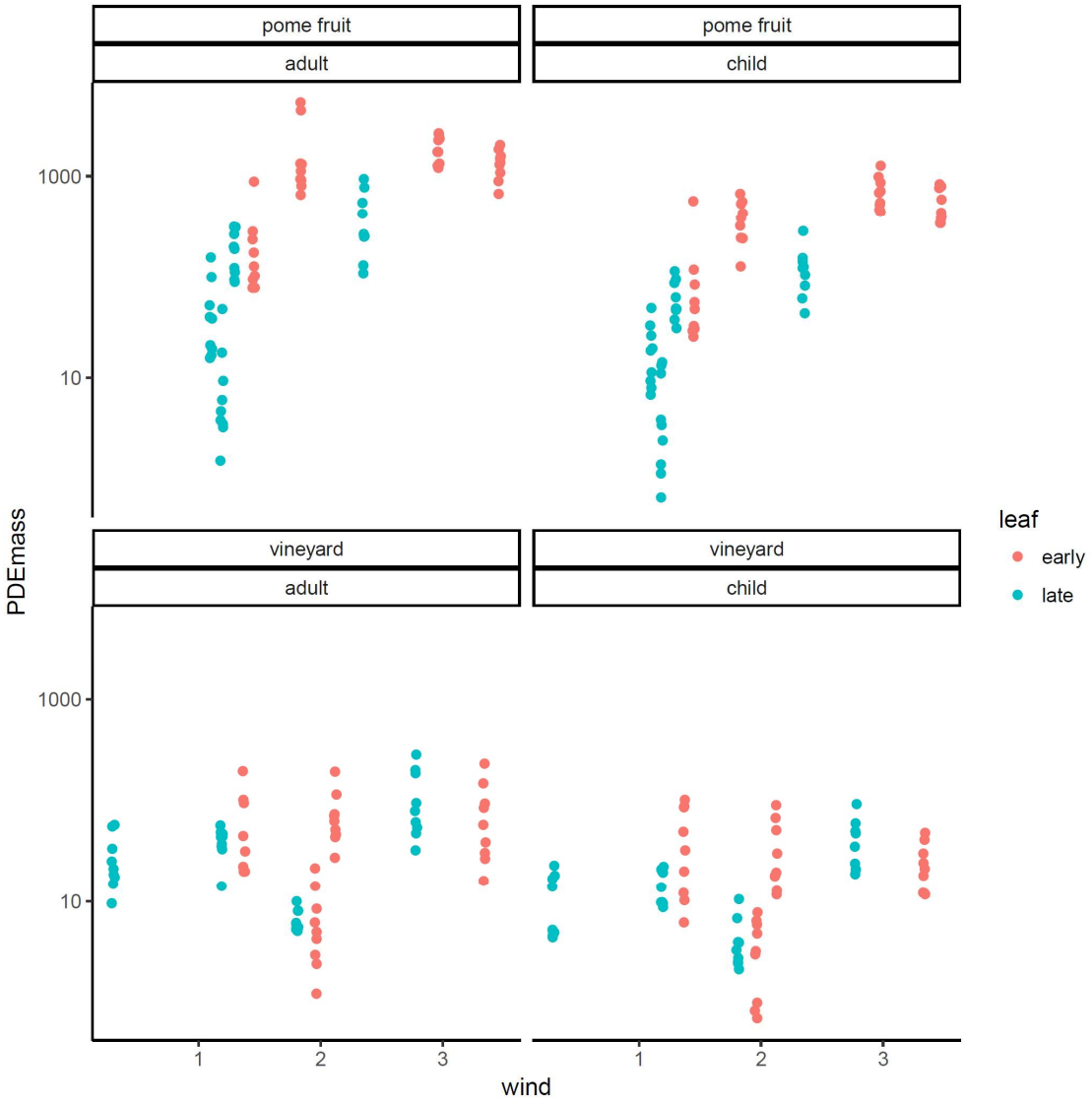


Figure E5.7: Potential dermal exposure and wind speed

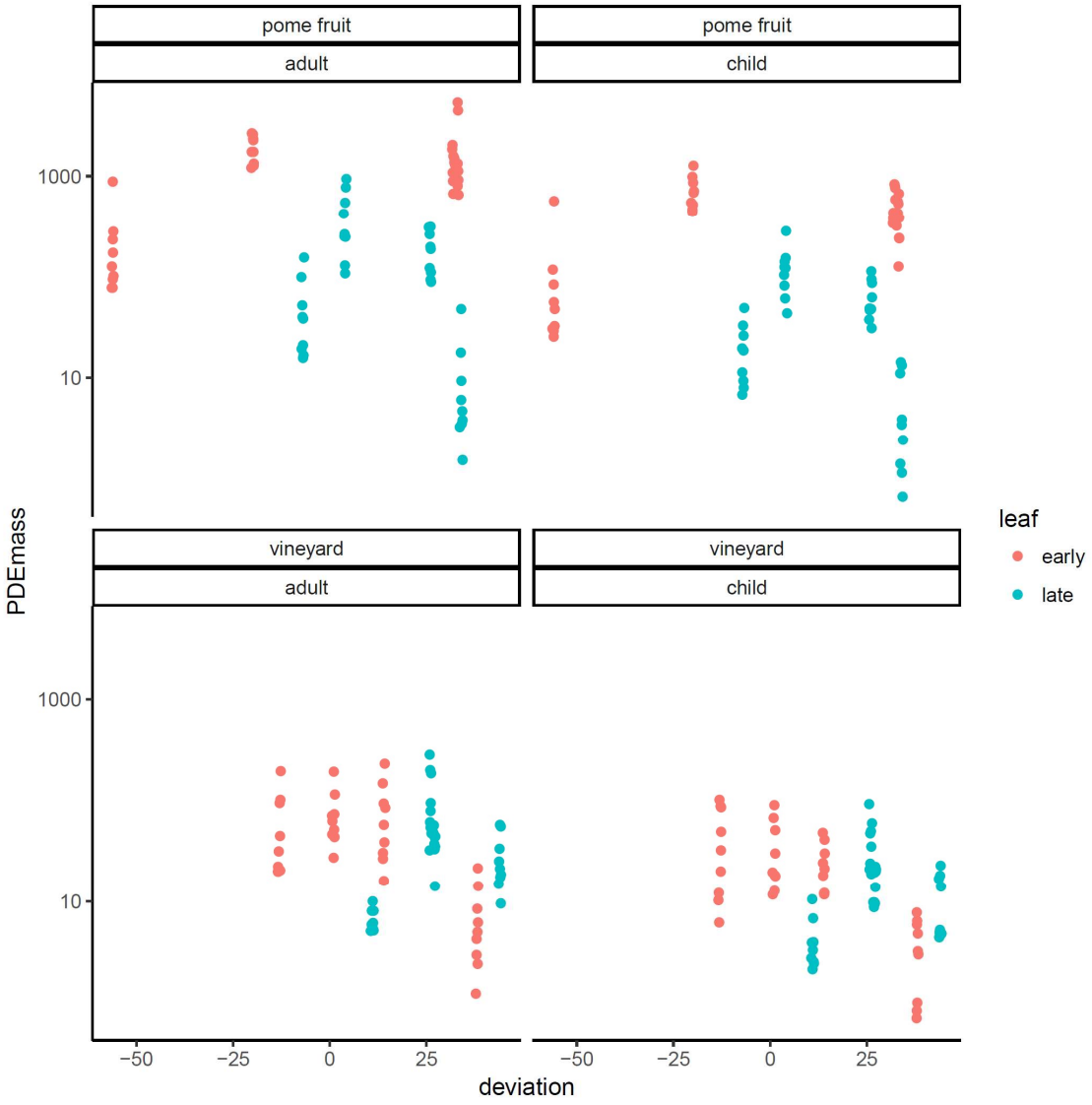


Figure E5.8: Potential dermal exposure and deviation in wind direction

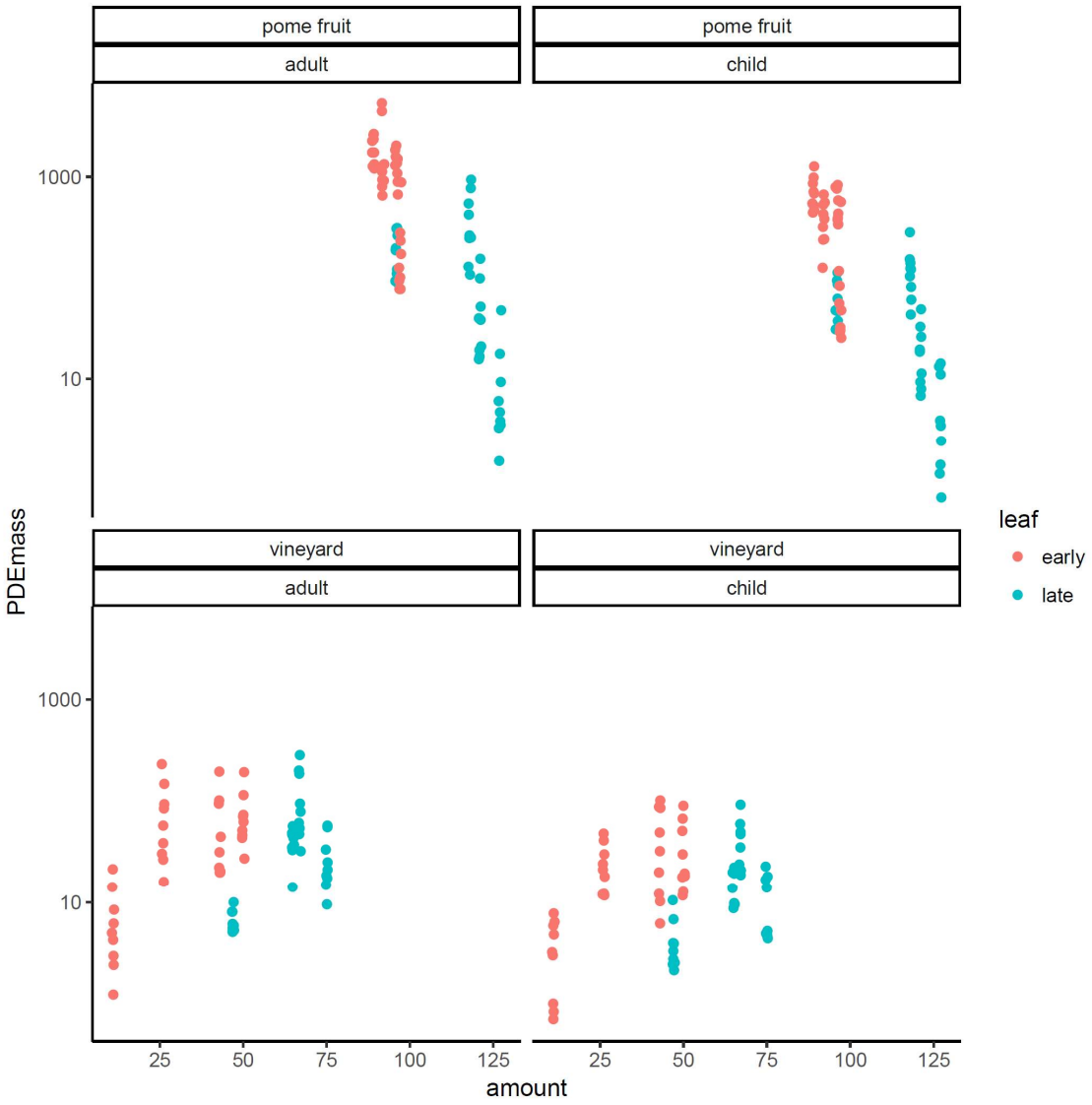


Figure E5.9: Potential dermal exposure and amount applied

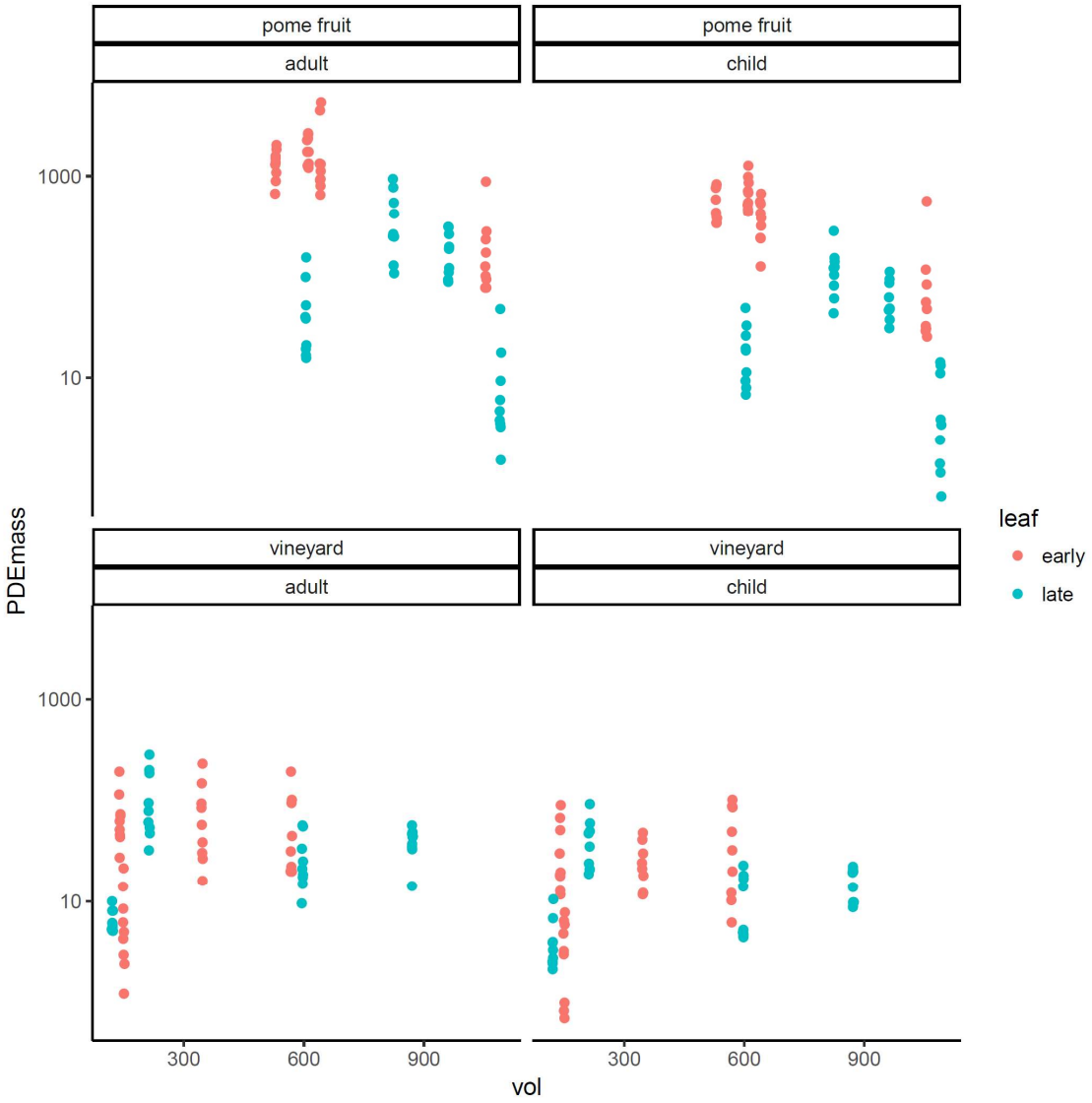


Figure E5.10: Potential dermal exposure and spray volume

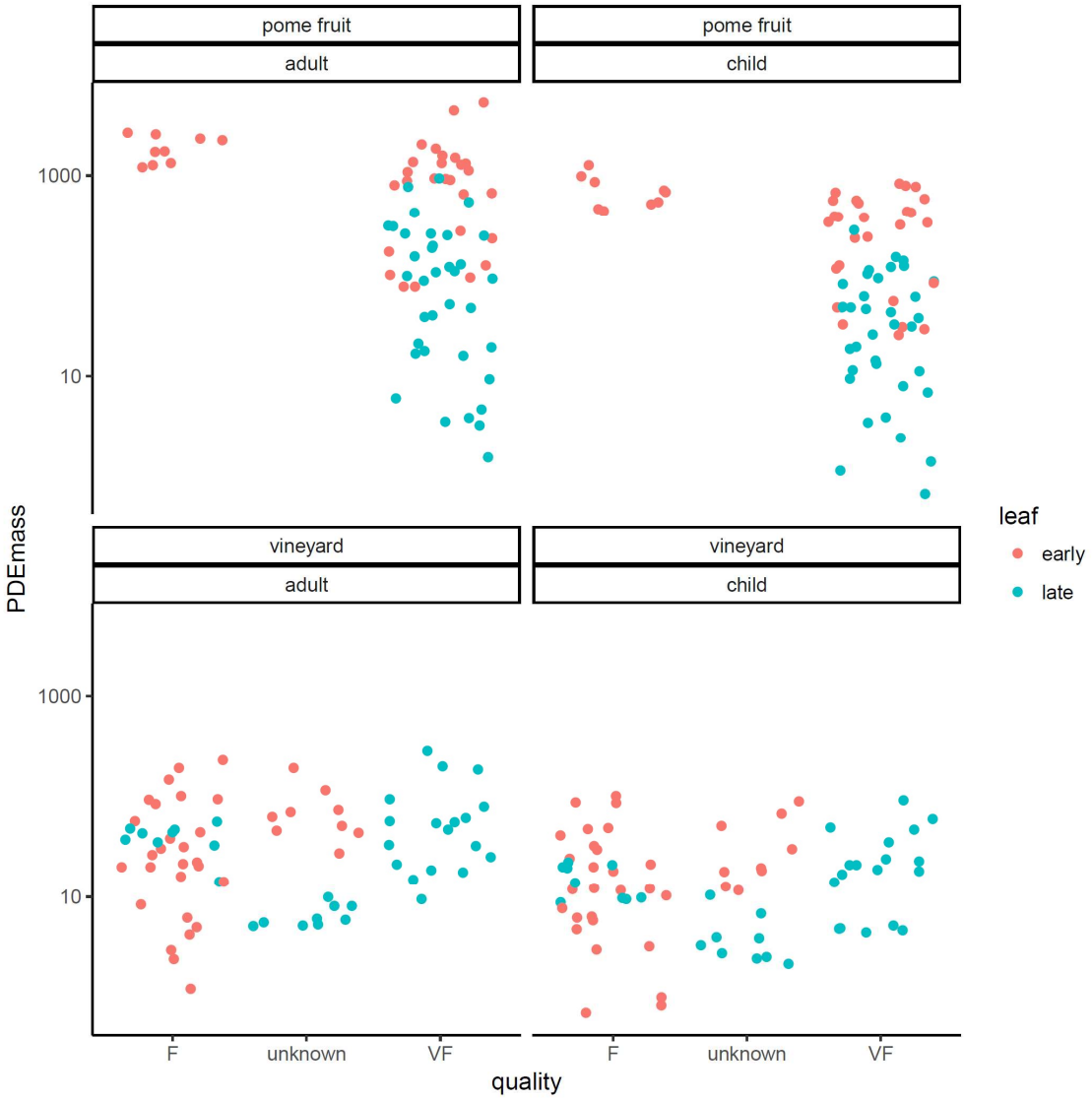


Figure E5.11: Potential dermal exposure and spray quality

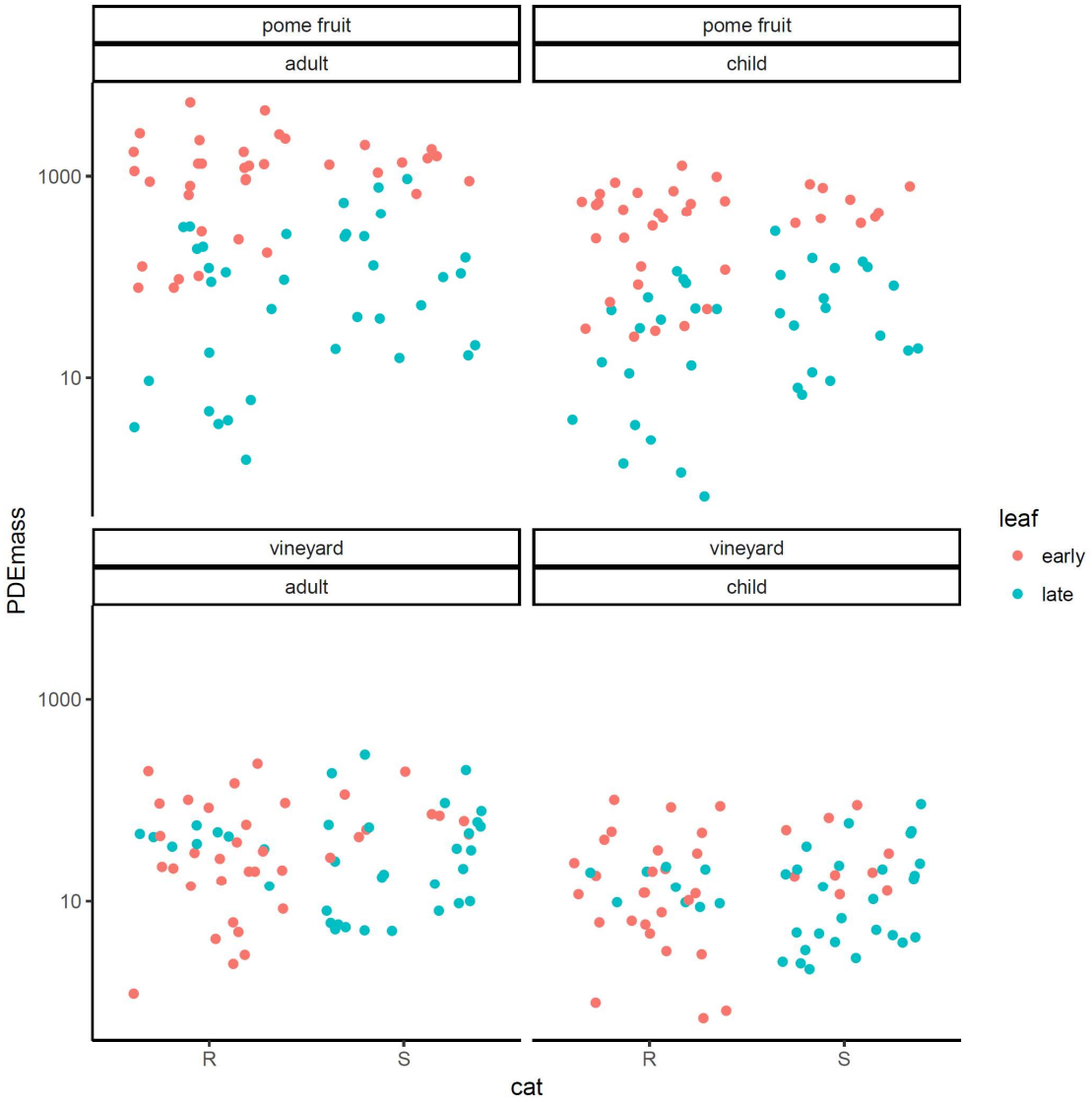


Figure E5.12: Potential dermal exposure and sprayer category

E6: Relationship between exposure expressed as volume of spray compared to mass of active substance

In Section 4, Study Design, the uncertainty regarding the measured amount of spray solution that was applied to the crop (and therefore the mannequins) relative to the target dose was discussed. Reported spray outputs were cross checked with theoretical spray output in Figure 9 and in eight of the studies there was a large difference that cannot be explained by inaccuracies in volume measurement; or by speed or other events. The differences between reported and calculated water volumes applied are summarised in Table E6.1.

Table E6.1: Summary of percentage difference in reported vs calculated water volume applied

Crop	Leaf cover	Differences between reported water volume and calculated water volume applied
Pome	No leaf	5%, -1%, 14%, -35%
Pome	Full leaf	2%, 34%, -5%, 4%
Vine	No leaf	21%, -629%, -162%, -413%
Vine	Full leaf	-16%, -29%, -118%, -112%

Numbers in red: there is a large difference that cannot be explained by inaccuracies in volume measurement; or by speed or other events.

On the basis of the comparison of the differences between reported water volume and calculated water volume applied it appears there is more uncertainty in the reported water volume from the vine data, with the greatest uncertainty with the vine no leaf scenario.

The impact of the discrepancies in water volume applied has been explored further by considering the relationship between the dermal and inhalation exposure estimates expressed as volume of spray compared to mass of active substance. The exposure estimates of volume of spray are calculated from the measured mass of active substance on dosimeters, the reported spray volume applied and concentration of active substance in the spray solution. Assuming the reported spray volume and concentration of active substance are correct there should be a consistent relationship between exposure volume and mass of active substance. The relationship between exposure estimates as a volume of spray compared to mass of active substance are shown in Figures E6.1 to E6.3, with graphs plotted for PDE, ADE and PIE for adults and children and for the different crops and leaf cover.

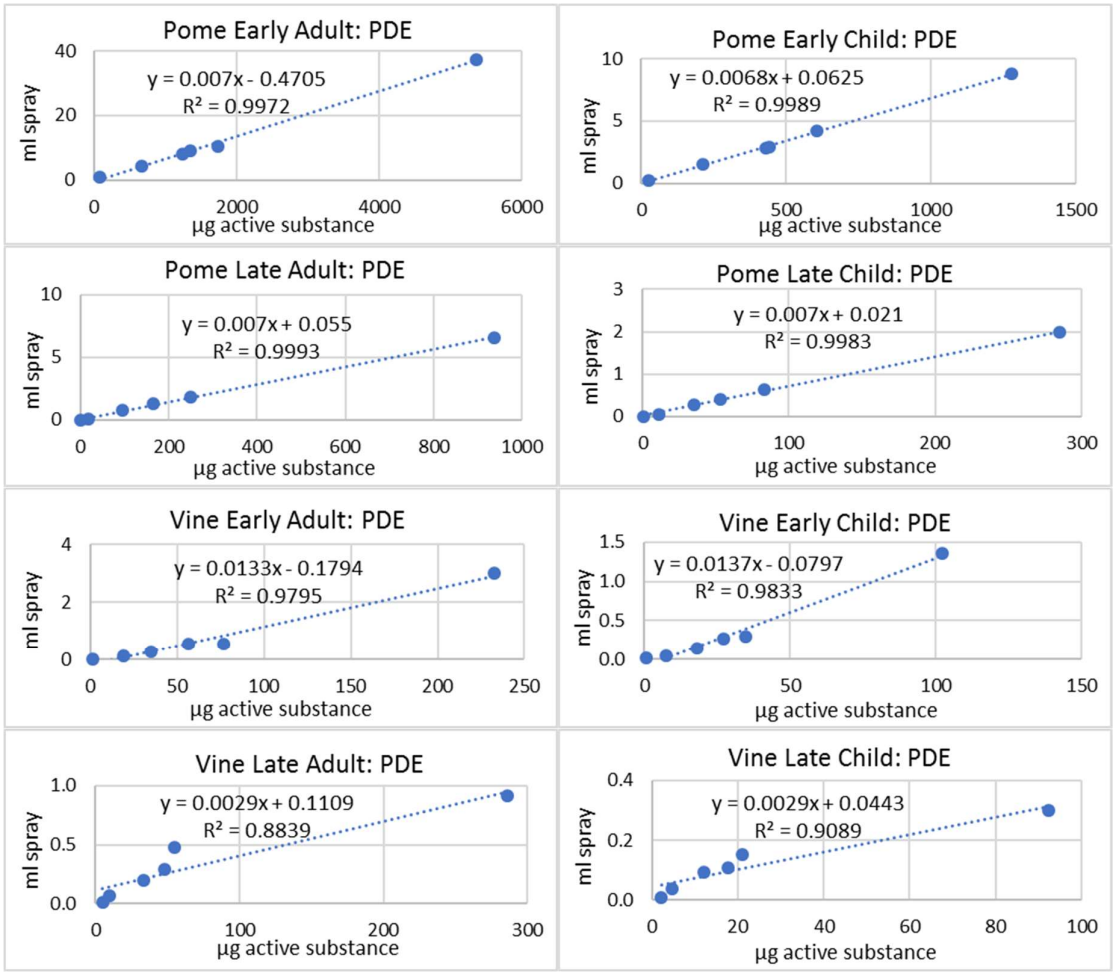


Figure E6.1: Potential dermal exposure – relationship between volume of spray (vertical axis) and mass of active (horizontal axis)

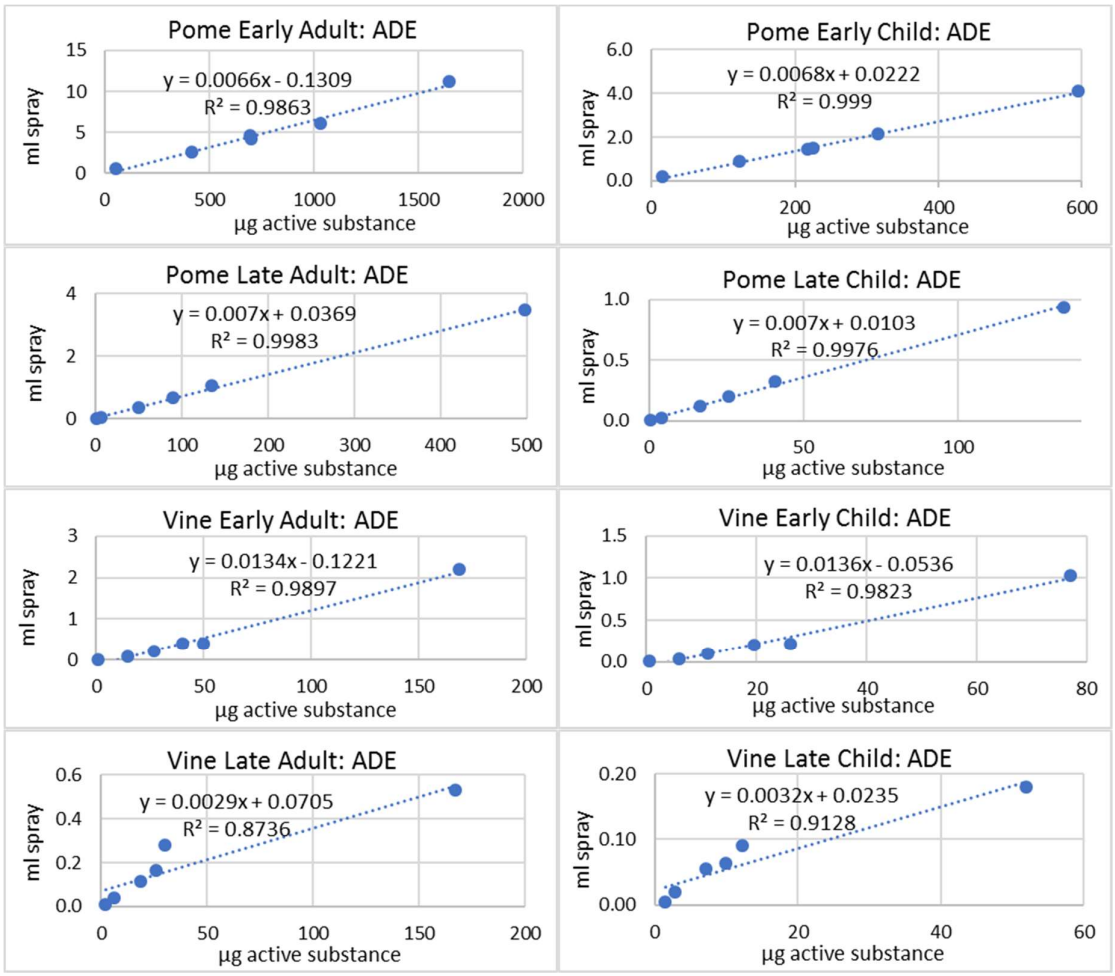


Figure E6.2: Actual dermal exposure – relationship between volume of spray (vertical axis) and mass of active (horizontal axis)

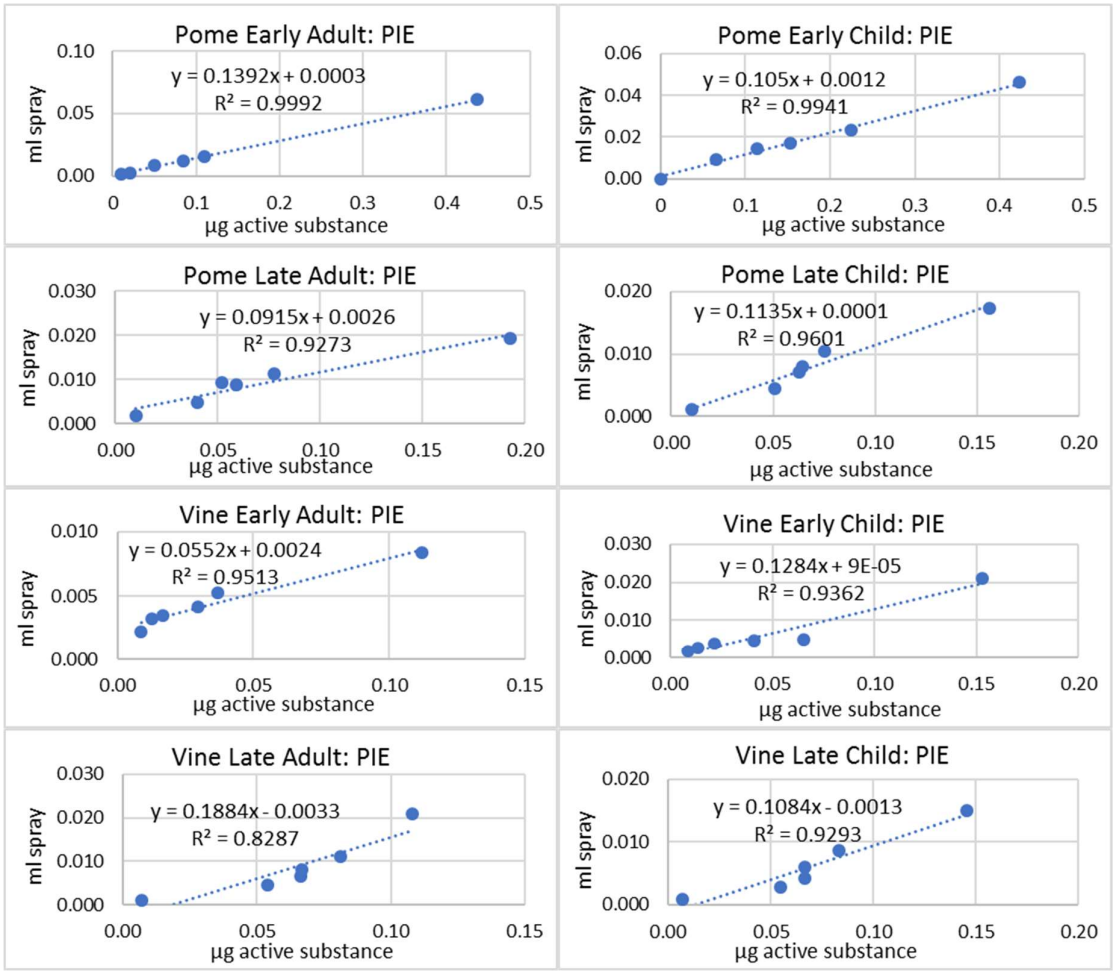


Figure E6.3: Potential inhalation exposure – relationship between volume of spray (vertical axis) and mass of active (horizontal axis)

Overall, the plots of dermal and inhalation exposure estimates expressed as a volume of spray compared to mass of active substance show good straight-line fits, with R squared values ranging from 0.83 to 0.99.

The pome fruit data shows very good correlation, with all the pome dermal data having an R squared of ≥0.99. Slightly more variation is seen in the pome inhalation data, with a minimum R squared of 0.93.

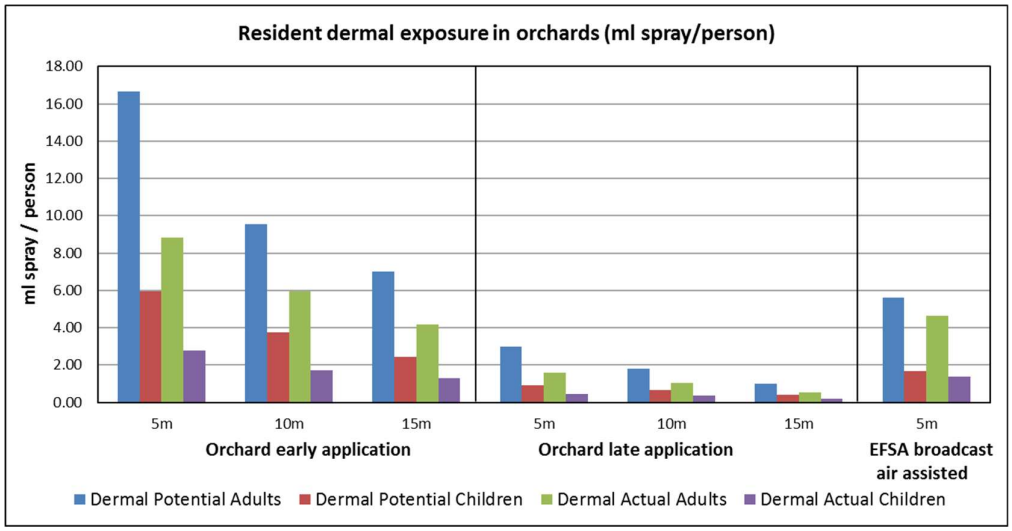
The vine early data is similar to the pome fruit data, with the dermal R squared values ≥0.98, and a minimum inhalation R squared value of 0.94. The vine full leaf data show most variation with the lowest R squared for dermal exposure being 0.87, and the lowest R squared for inhalation being 0.83. This differs slightly from the summary of percentage difference in reported vs calculated water volume applied (Table E3.2) where the vine no leaf data had the greatest uncertainty.

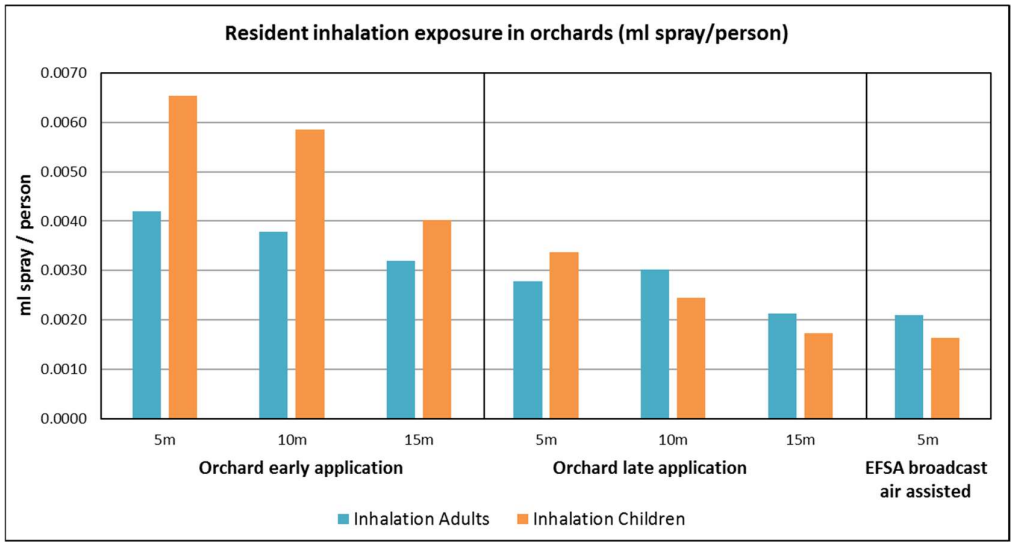
In conclusion, on the basis of comparison of the relationship between exposure as a volume of spray compared to mass of active substance, the expression of exposure as a volume of spray is considered to be reasonable, especially to facilitate comparisons with other data. However, the exposure expressed as mass of active is considered to be the definitive value.

Appendix F: Resident summary

Resident Summary: Orchards

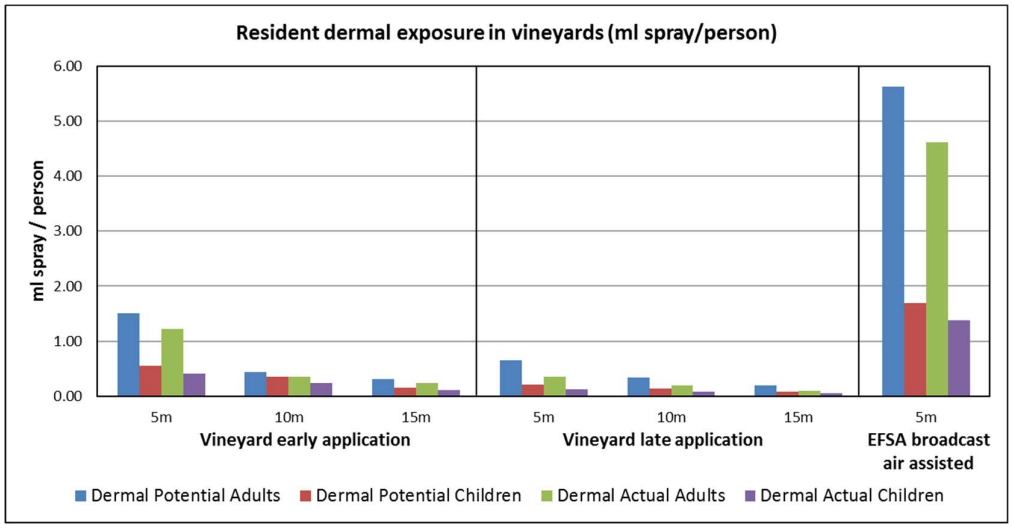
Crop / time of application	Distance	Resident 75th percentile exposure assuming average breathing rates (mL spray /person)					
		Dermal				Inhalation	
		Potential		Actual			
		Adults	Children	Adults	Children	Adults	Children
Orchard early application	5m	16.67	5.96	8.81	2.76	0.0042	0.0065
	10m	9.55	3.74	5.94	1.72	0.0038	0.0059
	15m	7.00	2.44	4.19	1.31	0.0032	0.0040
Orchard late application	5m	3.00	0.90	1.61	0.47	0.0028	0.0034
	10m	1.81	0.65	1.02	0.35	0.0030	0.0025
	15m	0.98	0.39	0.54	0.21	0.0021	0.0017
Broadcast EFSA	5m	5.63	1.69	4.62	1.38	0.00210	0.00164
	10m	5.63	1.69	4.62	1.38	0.00210	0.00164
	15m	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

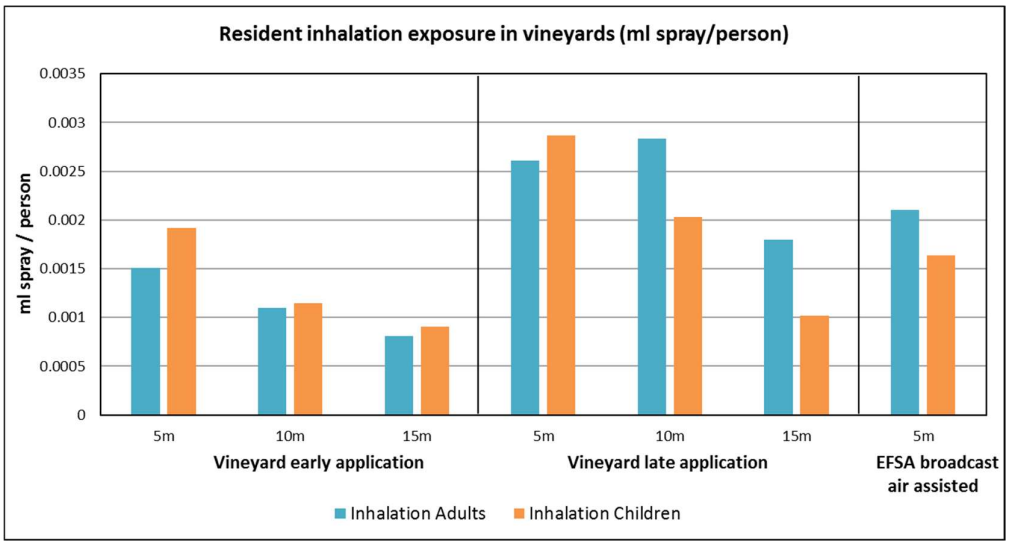




Resident summary: Vineyards

Crop / time of application	Distance	Resident 75th percentile exposure assuming average breathing rates (mL spray /person)					
		Dermal				Inhalation	
		Potential		Actual			
		Adults	Children	Adults	Children	Adults	Children
Vineyard early application	5m	1.51	0.56	1.22	0.41	0.0015	0.0019
	10m	0.44	0.35	0.35	0.24	0.0011	0.0011
	15m	0.31	0.15	0.24	0.11	0.0008	0.0009
Vineyard late application	5m	0.65	0.21	0.35	0.13	0.0026	0.0029
	10m	0.34	0.14	0.19	0.09	0.0028	0.0020
	15m	0.19	0.08	0.10	0.05	0.0018	0.0010
Broadcast EFSA	5m	5.63	1.69	4.62	1.38	0.00210	0.00164
	10m	5.63	1.69	4.62	1.38	0.00210	0.00164
	15m	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

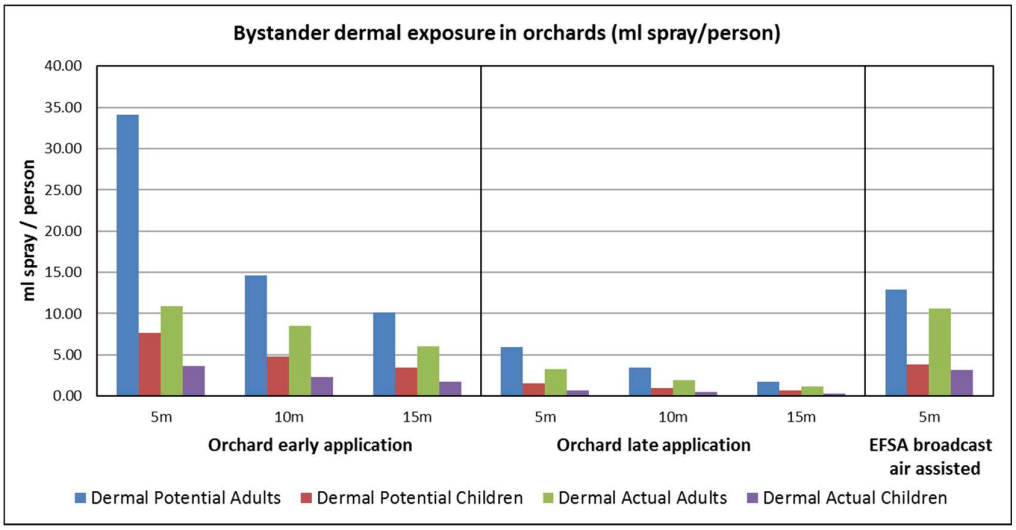


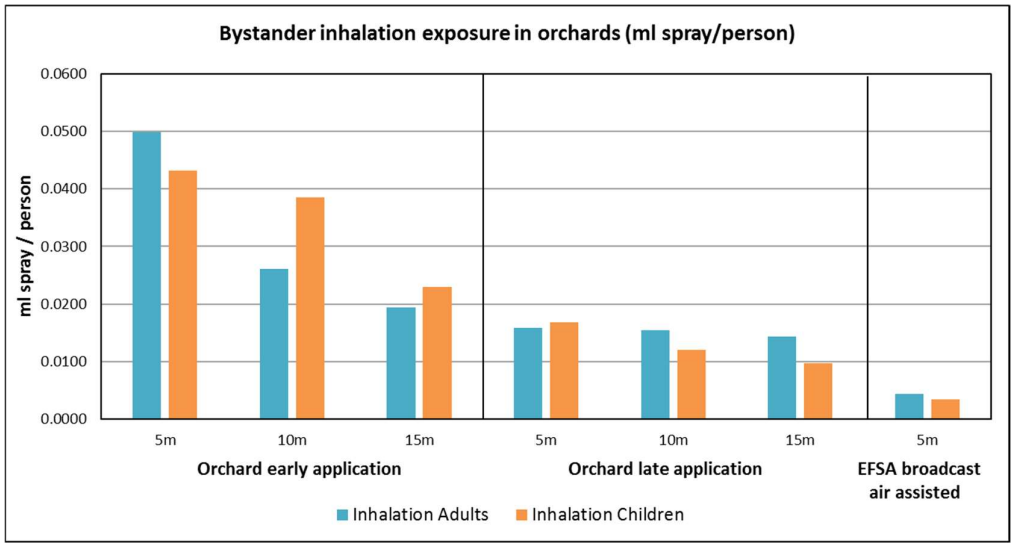


Appendix G: Bystander summary

Bystander Summary: Orchards

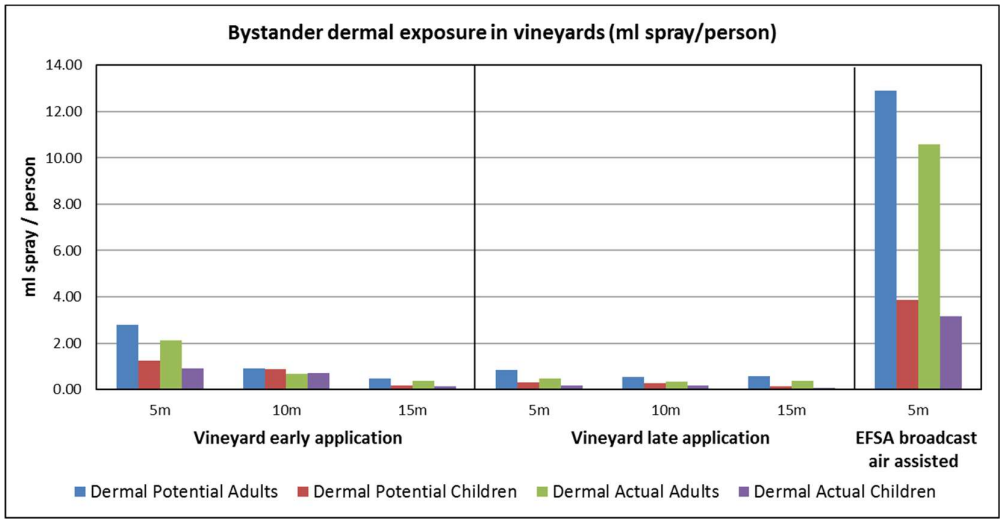
Crop / time of application	Distance	Bystander 95th percentile exposure assuming high intensity hourly breathing rates (mL spray /person)					
		Dermal				Inhalation	
		Potential		Actual			
		Adults	Children	Adults	Children	Adults	Children
Vineyard early application	5m	34.12	7.67	10.92	3.62	0.0499	0.0431
	10m	14.58	4.77	8.49	2.28	0.0261	0.0386
	15m	10.12	3.43	6.06	1.77	0.0194	0.0229
Vineyard late application	5m	5.93	1.52	3.22	0.72	0.0159	0.0168
	10m	3.43	1.00	1.96	0.49	0.0154	0.0121
	15m	1.79	0.67	1.19	0.32	0.0144	0.0097
Broadcast EFSA	5m	12.90	3.87	10.58	3.17	0.00440	0.00350
	10m	12.90	3.87	10.58	3.17	0.00440	0.00350
	15m	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

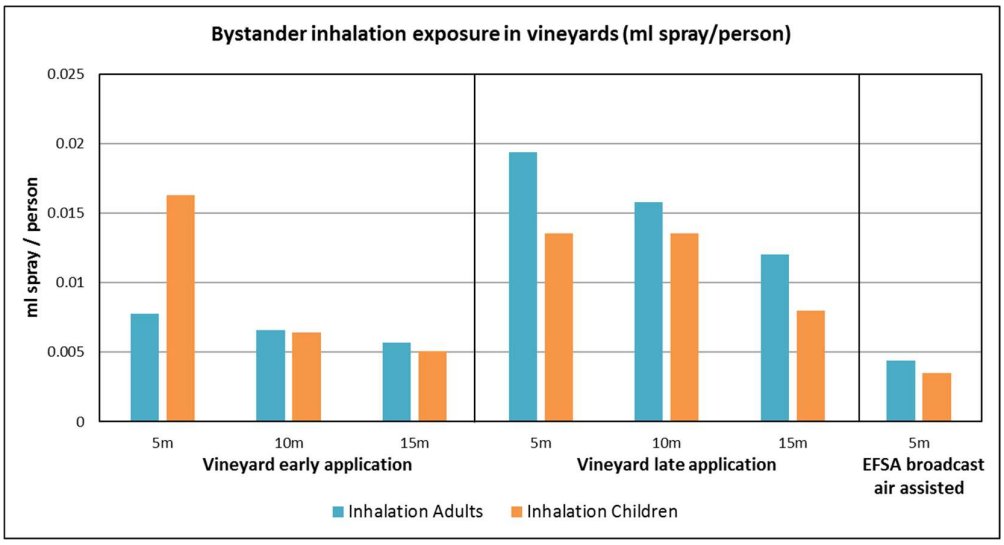




Bystander summary: Vineyards

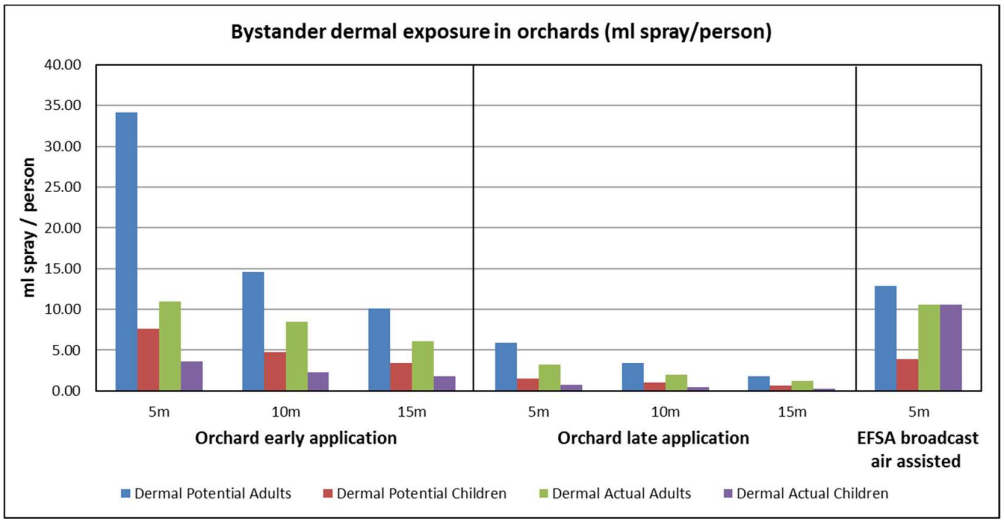
Crop / time of application	Distance	Bystander 95th percentile exposure assuming high intensity hourly breathing rates (mL spray /person)					
		Dermal				Inhalation	
		Potential		Actual			
		Adults	Children	Adults	Children	Adults	Children
Vineyard early application	5m	2.79	1.25	2.13	0.91	0.0078	0.0163
	10m	0.91	0.89	0.67	0.72	0.0066	0.0064
	15m	0.49	0.16	0.36	0.12	0.0057	0.0051
Vineyard late application	5m	0.83	0.30	0.47	0.17	0.0194	0.0135
	10m	0.56	0.26	0.34	0.17	0.0158	0.0135
	15m	0.59	0.13	0.36	0.08	0.0120	0.0080
Broadcast EFSA	5m	12.90	3.87	10.58	3.17	0.00440	0.00350
	10m	12.90	3.87	10.58	3.17	0.00440	0.00350
	15m	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

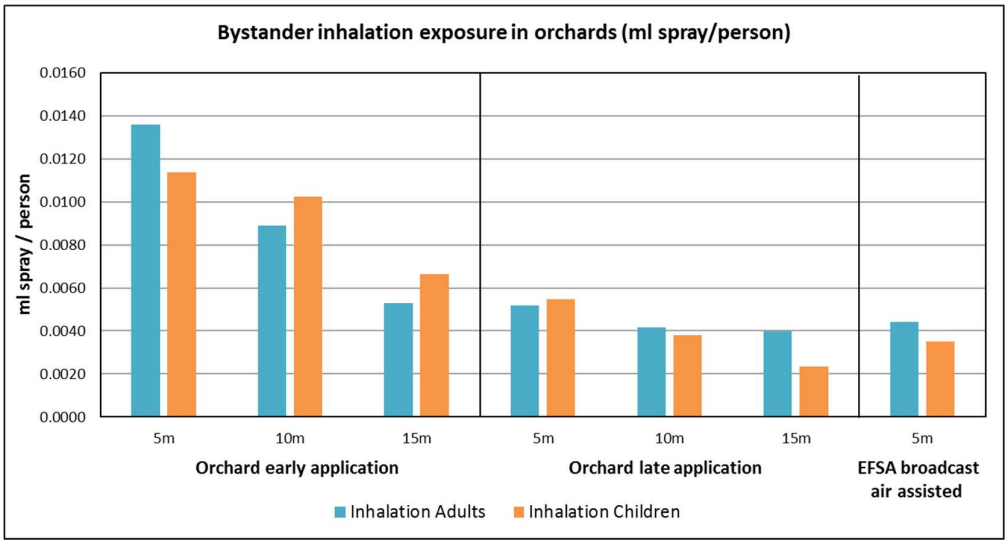




Bystander summary: Orchards (PIE 15 minutes)

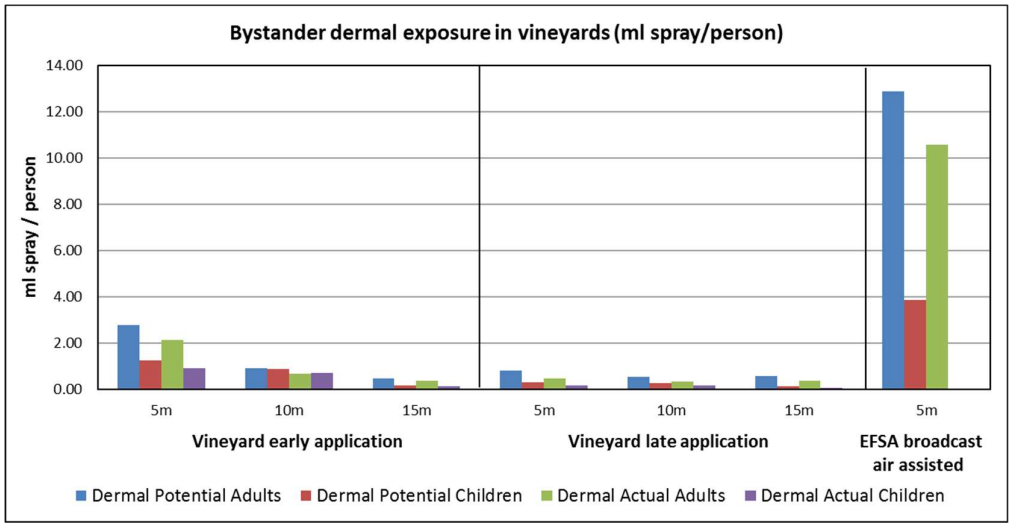
Crop / time of application	Distance	Bystander 95th percentile exposure assuming high intensity breathing rates (inhalation exposure duration normalised to 15 minutes) (mL spray /person)					
		Dermal				Inhalation	
		Potential		Actual			
		Adults	Children	Adults	Children	Adults	Children
Vineyard early application	5m	34.12	7.67	10.92	3.62	0.0136	0.0114
	10m	14.58	4.77	8.49	2.28	0.0089	0.0102
	15m	10.12	3.43	6.06	1.77	0.0053	0.0067
Vineyard late application	5m	5.93	1.52	3.22	0.72	0.0052	0.0055
	10m	3.43	1.00	1.96	0.49	0.0042	0.0038
	15m	1.79	0.67	1.19	0.32	0.0040	0.0024
Broadcast EFSA	5m	12.90	3.87	10.58	10.58	0.00440	0.00350
	10m	12.90	3.87	10.58	10.58	0.00440	0.00350
	15m	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

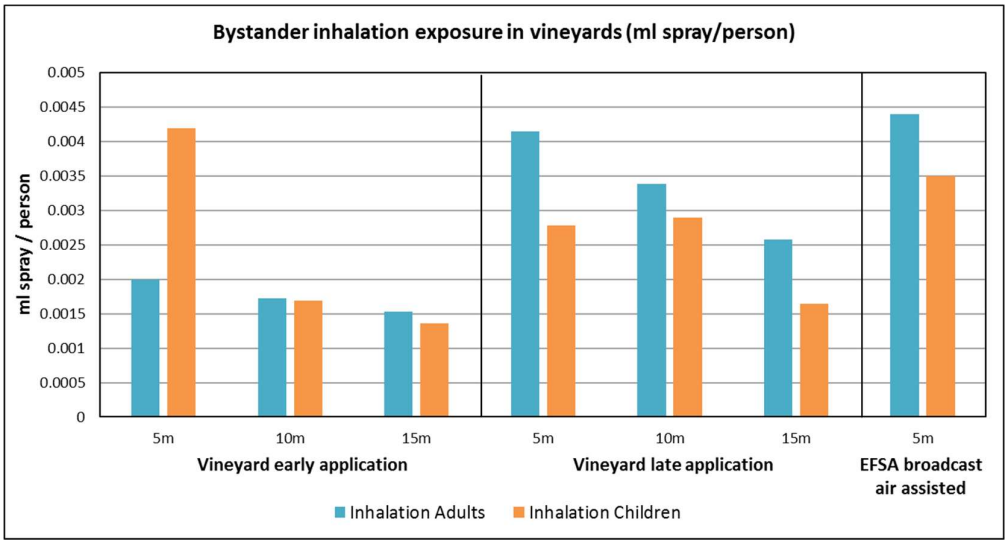




Bystander summary: Vineyards (PIE 15 minutes)

Crop / time of application	Distance	Bystander 95th percentile exposure assuming high intensity breathing rates (inhalation exposure duration normalised to 15 minutes) (mL spray /person)					
		Dermal				Inhalation	
		Potential		Actual			
		Adults	Children	Adults	Children	Adults	Children
Vineyard early application	5m	2.79	1.25	2.13	0.91	0.0020	0.0042
	10m	0.91	0.89	0.67	0.72	0.0017	0.0017
	15m	0.49	0.16	0.36	0.12	0.0015	0.0014
Vineyard late application	5m	0.83	0.30	0.47	0.17	0.0042	0.0028
	10m	0.56	0.26	0.34	0.17	0.0034	0.0029
	15m	0.59	0.13	0.36	0.08	0.0026	0.0016
Broadcast EFSA	5m	12.90	3.87	10.58	0.00	0.00440	0.00350
	10m	12.90	3.87	10.58	0.00	0.00440	0.00350
	15m	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.





Appendix H: Analytical methods

Determination of quinoxyfen residues on or in bystander and resident exposure dosimeters by LC-MS/MS: Analytical method CAM-0150/001

Principle of the method

Outer, inner and head/neck cotton dosimeter

Samples were cut into smaller sections and combined with the appropriate volume of methanol (full details in Table H.1) and mixed vigorously on a flatbed shaker for 2 hours and left to stand for 5 minutes. An aliquot of the sample was evaporated to dryness under nitrogen at 40 °C and reconstitute in 0.5 mL of methanol/water (50:50 v:v).

Air filters

10 mL acetone was added to the whole filter and shaken for 15 minutes followed by centrifugation for 5 minutes. After extraction, a 1 mL aliquot was taken. Evaporated to dryness under nitrogen at 40 °C and reconstitute in 1 mL of methanol/water (50:50 v:v).

Air tubes

10 mL of acetone was added to the filter paper and front section sorbent and shaken for 15 minutes followed by centrifugation for 5 minutes. After extraction, a 1 mL aliquot was taken. Evaporated to dryness under nitrogen at 40 °C and reconstitute in 1 mL of methanol:water (50:50 v:v).

Table H.1: Extraction procedure

Matrix	Extraction solvent	Volume of extraction solvent to add (mL)	Final analytical solution concentration (specimen/mL)
Outer dosimeter	Methanol	2500	0.004
Inner dosimeter (arms/legs)	Methanol	1000	0.004
Inner dosimeter (torso/waist)	Methanol	2000	0.004
Head/neck dosimeter	Methanol	1000	0.004
IOM air filter	Acetone	10	0.1
Air tubes	Acetone	10	0.1

Analysis was performed by LC-MS/MS using a Hichrom Ace C18, 2.1 x 50 mm, 3 µm column at 40 °C in positive ion mode for detection monitoring the following mass transitions: m/z 308 → 197 (quantification) and m/z 308 → 162 (confirmation). A gradient elution was used (Mobile phase A: water and 0.1% formic acid, Mobile phase B: methanol and 0.1% formic acid).

Stability of extracts

Stability of quinoxyfen residues in matrices were tested by determination of recovery at a fortification level of 0.1 µg/specimen for cotton dosimeters and 0.01 µg/specimen for air filters and tubes for 7 – 14 days stored between 2 – 8 °C.

Table H.2: Stability of extracts

Mass transition m/z 308 → 197				
Matrix	**Storage days**	**Fortification level (µg/specimen)**	**Recoveries % range (mean, n)**	**% RSD**
Outer dosimeter	11	0.1	90 – 96 (93, 6)	2.7
Inner dosimeter (arms/legs)	7	0.1	85 – 96 (91, 7)	5.2
Inner dosimeter (torso/waist)	8	0.1	81 – 98 (90, 7)	7.5
Head/neck dosimeter	8	0.1	78 – 102 (89, 7)	9.1
IOM air filter	14	0.01	97 – 106 (100, 7)	3.0
Air tubes	8	0.01	95 – 108 (101, 7)	4.4

Quinoxyfen was stable in all matrices for at least 7-14 days storage between 2 – 8 °C.

Stability of standards

Stability of quinoxyfen standard in methanol was determined after 112 days at 4 °C.

Table H.3: stability of standards

Matrix	Mass transition	% mean response factor in stored standards compared to freshly prepared standard solution
Stock standard in methanol	308 → 197	4.3

Stock standard solution in methanol was stable for at least 112 days stored at 4 °C.

Matrix effects

Matrix-matched standards were quantified against standards in methanol:water (50:50 v:v) at a concentration of 5 ng/ml. The matrix effect was calculated as a ratio of the mean peak area in matrix-matched standards to the mean peak area in solvent standards expressed as a percentage.

Table H.4: Matrix effects

Matrix	**Mass transition (m/z)**	**Matrix effect (%)**
Outer dosimeter	308 → 197	-3.1
Inner dosimeter (arms/legs)		-15.5
Inner dosimeter (torso/waist)		0.5
Head/neck dosimeter		-0.1
IOM air filter		-1.6
Air tubes		5.7

Matrix	Mass transition (m/z)	Matrix effect (%)
Outer dosimeter	308 → 162	-4.2
Inner dosimeter (arms/legs)		-16.6
Inner dosimeter (torso/waist)		-0.6
Head/neck dosimeter		-1.2
IOM air filter		-1.3
Air tubes		5.2

No significant matrix effects (>20%) were observed therefore calibration could be performed with standards in methanol:water (50:50 v:v).

Validation summary

LC-MS/MS is a highly specific technique and two mass transitions (3 ions) were monitored. Chromatograms of standard solutions, control samples and fortified samples have been presented showing no interferences >30% LOQ at the retention time of interest. Accuracy was assessed at 3 fortification levels for the analyte in each matrix of interest corresponding to the LOQ, 10xLOQ and 1000xLOQ; in all cases the mean recovery was within the acceptable range of 70 – 110%. To assess method precision, 7 determinations were made at each fortification level and the RSDs were within the acceptable limits of 20%. The overall RSDs were between 5.2 – 9.6%. The linear range is appropriate for the nominal test concentrations (allowing for necessary dilutions) and was determined using solvent-based standards as no significant matrix effects were observed. The LOQ of the method is 0.1 µg/specimen for cotton dosimeters and 0.01 µg/specimen for air filters and tubes. The method is satisfactorily validated in accordance with SANCO/3029/99 rev.4.

Table H5: Validation data

Matrix	Analyte (transition m/z)	LOQ (µg/specimen)	Recovery fortification level (µg/specimen)	% Recoveries range (mean, n)	Repeatability %RSD (n)	Linearity	Specificity
Outer dosimeter	Quinoxyfen (308 → 197)	0.1	0.1 1.0 100	82 – 112 (89, 7) 77 – 95 (89, 7) 74 – 102 (90, 7)	11.6 (7) 6.5 (7) 10.6 (7) Overall 9.3 (21)	0.12 – 100 ng/mL [approx. 0.03 – 25 µg/specimen] 9 standards, y = 8.5E10^{7}x - 1397, r^2 = 0.9989	Acceptable chromatograms presented for standard, control, fortified samples and reagent blank. No interference >30% LOQ at the retention time of interest (ca. 2.4 min). Identity confirmed by additional mass transition.
Inner dosimeter (arms/legs)		0.1	0.1 1.0 100	88 – 104 (96, 7) 80 – 92 (87, 7) 89 – 119 (98, 7)	6.2 (7) 4.8 (7) 10.6 (7) Overall 9.3 (21)	0.12 – 100 ng/mL [approx. 0.03 – 25 µg/specimen] 9 standards, y = 7.4E10^{7}x – 922, r^2 = 0.9986	
Inner dosimeter (torso/waist)		0.1	0.1 1.0 100	79 – 88 (85, 7) 71 – 85 (78, 7) 79 – 98 (88, 7)	3.7 (7) 5.8 (7) 9.3 (7) Overall: 8.1 (21)	0.12 – 100 ng/mL [approx. 0.03 – 25 µg/specimen] 9 standards, y = 7.1E10^{7}x + 832, r^2 = 0.9980	
Head/neck dosimeter		0.1	0.1 1.0 100	79 – 105 (92, 7) 90 – 103 (96, 7) 91 – 115 (103, 7)	9.3 (7) 4.2 (7) 7.8 (7)	0.12 – 100 ng/mL [approx. 0.03 – 25	

Matrix	Analyte (transition m/z)	LOQ (µg/specimen)	Recovery fortification level (µg/specimen)	% Recoveries range (mean, n)	Repeatability %RSD (n)	Linearity	Specificity
					Overall: 8.4 (21)	µg/specimen] 9 standards, y = 6.1E10^{7}x + 645, r^2 = 0.9979	
IOM air filters		0.01	0.01 0.1 10	101 – 105 (103, 7) 91 – 100 (95, 7) 85 -102 (94, 7)	1.5 (7) 3.2 (7) 6.0 (7) Overall: 5.7 (21)	0.12 – 100 ng/mL [equivalent to 0.0012 – 1 µg/specimen] 9 standards, y = 1.03E10^{8}x + 2026, r^2 = 0.9983	
Air sampling tubes		0.01	0.01 0.1 10	90 -103 (98, 7) 97 – 108 (103, 7) 80 – 110 (98, 7)	4.9 (7) 3.9 (7) 10.3 (7) Overall: 7.0 (21)	0.12 – 100 ng/mL [equivalent to 0.0012 – 1 µg/specimen] 9 standards, y = 6.9E10^{7}x – 643, r^2 = 0.9982	
Outer dosimeter	Quinoxyfen (308 → 162)	0.1	0.1 1.0 100	82 – 112 (89, 7) 77 – 96 (89, 7) 75 – 100 (89, 7)	11.9 (7) 6.5 (7) 10.1 (7) Overall: 9.3 (21)	0.12 – 100 ng/mL [approx. 0.03 – 25 µg/specimen] 9 standards, y = 5.7E10^{7}x – 811 r^2 = 0.9986	Acceptable chromatograms presented for standard, control, fortified samples and reagent blank.

Matrix	Analyte (transition m/z)	LOQ (µg/specimen)	Recovery fortification level (µg/specimen)	% Recoveries range (mean, n)	Repeatability %RSD (n)	Linearity	Specificity
Inner dosimeter (arms/legs)		0.1	0.1 1.0 100	81 – 94 (87, 7) 80 – 91 (86, 7) 89 – 118 (98, 7)	6.0 (7) 4.7 (7) 10.0 (7) Overall: 9.6 (21)	0.12 – 100 ng/mL [approx. 0.03 – 25 µg/specimen] 9 standards, y = $4.8E10^{7}$x – 306, r^2 = 0.9985	No interference >30% LOQ at the retention time of interest (ca. 2.4 min).
Inner dosimeter (torso/waist)		0.1	0.1 1.0 100	82 – 90 (86, 7) 71 – 86 (78, 7) 79 – 98 (87, 7)	3.6 (7) 6.2 (7) 9.1 (7) Overall: 8.3 (21)	0.12 – 100 ng/mL [approx. 0.03 – 25 µg/specimen] 9 standards, y = $4.7E10^{7}$x + 446, r^2 = 0.9965	
Head/neck dosimeter		0.1	0.1 1.0 100	74 – 101 (89, 7) 86 – 100 (94, 7) 90 – 113 (102, 7)	9.6 (7) 4.9 (7) 8.3 (7) Overall: 9.2 (21)	0.12 – 100 ng/mL [approx. 0.03 – 25 µg/specimen] 9 standards, y = $4.1E10^{8}$x + 318, r^2 = 0.9981	
IOM air filters		0.01	0.01 0.1 10	100 – 105 (103, 7) 92 – 100 (96, 7) 88 – 104 (97, 7)	1.6 (7) 3.2 (7) 6.6 (7)	0.12 – 100 ng/mL [equivalent to 0.0012 – 1 µg/specimen]	

Matrix	Analyte (transition m/z)	LOQ (µg/specimen)	Recovery fortification level (µg/specimen)	% Recoveries range (mean, n)	Repeatability %RSD (n)	Linearity	Specificity
					Overall: 5.2 (21)	9 standards, y = 6.8E10^{7}x + 1400, r^2 = 0.9980	
Air sampling tubes		0.01	0.01 0.1 10	89 – 102 (98, 7) 97 – 108 (103, 7) 80 – 109 (97, 7)	5.0 (7) 3.4 (7) 10.4 (7) Overall: 7.1 (21)	0.12 – 100 ng/mL [equivalent to 0.0012 – 1 µg/specimen] 9 standards, y = 4.5E10^{7}x – 579, r^2 = 0.9976	

Determination of kresoxim-methyl residues on or in bystander and resident exposure dosimeters by LC-MS/MS: Analytical method CAM-0149/001

Principle of the method

Outer, inner and head/neck cotton dosimeter

Samples were cut into smaller sections and combined with the appropriate volume of methanol (full details in Table H.6) and mixed vigorously on a flatbed shaker for 2 hours and left to stand for 5 minutes. A 0.5 mL aliquot of the sample extract was added to 0.5 mL of water and left to stand.

Air filters

10 mL acetone was added to the whole filter and shaken for 15 minutes followed by centrifugation for 5 minutes. After extraction, a 1 mL aliquot was taken. Evaporated to dryness under nitrogen at 40 °C and reconstituted in 1 mL of methanol:water (50:50 v:v).

Air tubes

10 mL of acetone was added to the filter paper and front section sorbent and shaken for 15 minutes followed by centrifugation for 5 minutes. After extraction, a 1 mL aliquot was taken. Evaporated to dryness under nitrogen at 40 °C and reconstituted in 1 mL of methanol:water (50:50 v:v).

Table H.6: Extraction procedure

Matrix	Extraction solvent	Volume of extraction solvent to add (mL)	Final analytical solution concentration (specimen/mL)
Outer dosimeter	Methanol	2500	0.0002
Inner dosimeter (arms/legs)	Methanol	1000	0.0005
Inner dosimeter (torso/waist)	Methanol	2000	0.00025
Head/neck dosimeter	Methanol	1000	0.0005
IOM air filter	Acetone	10	0.1
Air tubes	Acetone	10	0.1

Analysis was performed by LC-MS/MS using a Hichrom Ace C18, 2.1 x 50 mm, 3 µm column at 40 °C in positive ion mode for detection monitoring the following mass transitions: m/z 314 → 206 (quantification) and m/z 314 → 267 (confirmation). A gradient elution was used (Mobile phase A: water and 0.1% formic acid, Mobile phase B: methanol and 0.1% formic acid).

Stability of extracts

Stability of kresoxim-methyl residues in matrices were tested by determination of recovery at fortification level of 0.1 µg/specimen for cotton dosimeters and 0.01 µg/specimen for air filters and tubes for 7 – 11 days stored between 2 – 8 °C.

Table H.7: Stability of extracts

Mass transition m/z 314 → 206				
Matrix	**Storage days**	**Fortification level (µg/specimen)**	**Recoveries % range (mean, n)**	**% RSD**
Outer dosimeter	8	0.1	94 – 102 (100, 7)	3.1
Inner dosimeter (arms/legs)	8	0.1	102 – 115 (108, 7)	4.4
Inner dosimeter (torso/waist)	7	0.1	93 – 106 (97, 7)	5.0
Head/neck dosimeter	8	0.1	94 – 105 (100, 7)	3.6
IOM air filter	10	0.01	82 – 116 (97, 7)	11.5
Air tubes	11	0.01	90 – 100 (94, 7)	4.1

Kresoxim-methyl was stable in all matrices for at least 8 – 11 days storage between 2 – 8 °C.

Stability of standards

Stability of kresoxim-methyl standard in methanol was determined after 77 days and 96 days.

Table H.8: stability of standards

Matrix	Storage days	Mass transition (*m/z*)	% mean response factor in stored standards compared to freshly prepared standard solution
Stock standard in methanol	77	314 → 206	-6.9
	96		-8.2

Stock standard solution in methanol was stable for at least 96 days stored between 2 – 8 °C.

Matrix effects

Matrix-matched standards were quantified against standards in methanol/water (50:50 v:v) at a concentration of 0.5 ng/ml. The matrix effect was calculated as a ratio of the mean peak area in matrix-matched standards to the mean peak area in solvent standards expressed as a percentage.

Table H.9: Matrix effects

Matrix	**Mass transition (*m/z*)**	**Matrix effect (%)**
Outer dosimeter	314 → 206	6.7
Inner dosimeter (arms/legs)		4.8
Inner dosimeter (torso/waist)		9.9
Head/neck dosimeter		9.4
IOM air filter		-12.7
Air tubes		2.0
Outer dosimeter	314 → 206	7.9
Inner dosimeter (arms/legs)		7.2
Inner dosimeter (torso/waist)		8.2
Head/neck dosimeter		10.3
IOM air filter		-11.2
Air tubes		5.4

No significant matrix effects (>20%) were observed therefore calibration could be performed with standards in methanol/water (50:50 v:v).

Validation summary

LC-MS/MS is a highly specific technique and two mass transitions (3 ions) were monitored. Chromatograms of standard solutions, control samples and fortified samples have been presented showing no interferences >30% LOQ at the retention time of interest. Accuracy was assessed at 3 fortification levels for the analyte in each matrix of interest corresponding to the LOQ, 10xLOQ and 1000xLOQ; in all cases the mean recovery was within the acceptable range of 70 – 110%. To assess method precision, 7 determinations were made at each fortification level and the RSDs were within the acceptable limits of 20%. The overall RSDs were between 3.0 – 10.8%. The linear range is appropriate for the nominal test concentrations (allowing for necessary dilutions) and was determined using solvent-based standards as no significant matrix effects were observed. The LOQ of the method is 0.1 µg/specimen for dosimeters and 0.01 µg/specimen for air filters and tubes. The method is satisfactorily validated in accordance with SANCO/3029/99 rev.4.

Table H.10: Validation data

Matrix	Analyte (transition m/z)	LOQ (µg/specimen)	Recovery fortification level (µg/specimen)	% Recoveries range (mean, n)	Repeatability %RSD (n)	Linearity	Specificity
Outer dosimeter	Kresoxim-methyl (314 → 206)	0.1	0.1 1.0 100	89 – 105 (99, 7) 70 – 101 (91, 7) 104 – 114 (107, 6)	5.9 (7) 14.6 (7) 3.6 (6) Overall: 10.8 (20)	0.006 – 20 ng/mL [approx. 0.03 – 100 µg/specimen] 9 standards, y = 2.9E10^{8}x +1154, r^2 = 0.9998	Acceptable chromatograms presented for standard, control, fortified samples and reagent blank. No interferences >30% LOQ at the retention time of interest (ca. 2.7 min). Identity confirmed by additional mass transition.
Inner dosimeter (arms/legs)		0.1	0.1 1.0 100	92 – 103 (97, 7) 102 – 105 (103, 7) 99 – 101 (100, 7)	4.9 (7) 1.5 (7) 0.8 (7) Overall: 4.0 (21)	0.006 – 20 ng/mL [approx. 0.012 – 40 µg/specimen] 9 standards, y = 2.6E10^{8}x + 633, r^2 = 0.9975	
Inner dosimeter (torso/waist)		0.1	0.1 1.0 100	83 – 110 (97, 7) 97 – 103 (100, 7) 98 – 103 (101, 7)	9.2 (7) 1.9 (7) 1.6 (7) Overall 5.4 (21)	0.006 – 20 ng/mL [approx. 0.024 – 80 µg/specimen] 9 standards, y = 2.5E10^{8}x - 286, r^2 = 0.9996	
Head/neck dosimeter		0.1	0.1 1.0 100	86 – 102 (94, 7) 72 – 101 (95, 7) 100 – 109 (103, 7)	5.8 (7) 10.7 (7) 2.7 (7)	0.006 – 20 ng/mL [approx. 0.012 – 40	

Matrix	Analyte (transition m/z)	LOQ (µg/specimen)	Recovery fortification level (µg/specimen)	% Recoveries range (mean, n)	Repeatability %RSD (n)	Linearity	Specificity
					Overall: 8.0 (21)	µg/specimen] 9 standards, y = 2.5E10^{8}x – 4.38, r^2 = 0.9995	
IOM air filters		0.01	0.01 0.1 10	74 – 109 (96, 7) 96 – 108 (100, 7) 89 – 110 (98, 7)	12.9 (7) 3.8 (7) 7.4 (7) Overall: 8.5	0.12 – 100 ng/mL [equivalent to 0.0012 – 1 µg/specimen] 9 standards, y = 9.3E10^{7}x + 5513, r^2 = 0.9976	
Air sampling tubes		0.01	0.01 0.1 10	94 – 103 (98, 7) 73 – 105 (96, 7) 91 – 110 (101, 7)	3.3 (7) 12.0 (7) 5.5 (7) Overall: 7.8 (21)	0.12 – 100 ng/mL [equivalent to 0.0012 – 1 µg/specimen] 9 standards, y = 1.2E10^{8}x + 535, r^2 = 0.9989	
Outer dosimeter	Kresoxim-methyl (314 → 267)	0.1	0.1 1.0 100	86 – 96 (92, 7) 73 – 103 (92, 7) 104 – 114 (107, 7)	3.7 (7) 13.0 (7) 3.3 (7) Overall: 10.6 (21)	0.006 – 20 ng/mL [approx. 0.03 – 100 µg/specimen] 9 standards, y = 3.3E10^{8}x + 731 r^2 = 0.9997	Acceptable chromatograms presented for standard, control, fortified samples and reagent blank.

Matrix	Analyte (transition m/z)	LOQ (µg/specimen)	Recovery fortification level (µg/specimen)	% Recoveries range (mean, n)	Repeatability %RSD (n)	Linearity	Specificity
Inner dosimeter (arms/legs)		0.1	0.1 1.0 100	94 – 107 (101, 7) 104 - 107 (105, 7) 100 – 102 (101, 7)	4.0 (7) 1.1 (7) 0.7 (7) Overall: 3.0 (21)	0.006 – 20 ng/mL [approx. 0.012 – 40 µg/specimen] 9 standards, y = 2.9E10^{8}x + 522, r^2 = 0.9974	No interference >30% LOQ at the retention time of interest (ca. 2.7 min).
Inner dosimeter (torso/waist)		0.1	0.1 1.0 100	84 – 105 (96, 7) 95 – 103 (100, 7) 98 – 103 (101, 7)	8.7 (7) 2.7 (7) 1.6 (7) Overall: 5.4 (21)	0.006 – 20 ng/mL [approx. 0.024 – 80 µg/specimen] 9 standards, y = 2.8E10^{8}x – 1.27, r^2 = 0.9998	
Head/neck dosimeter		0.1	0.1 1.0 100	96 – 104 (98, 7) 72 – 102 (96, 7) 102 – 109 (104, 7)	2.7 (7) 11.1 (7) 2.3 (7) Overall: 7.0 (21)	0.006 – 20 ng/mL [approx. 0.012 – 40 µg/specimen] 9 standards, y = 2.9E10^{8}x + 319, r^2 = 0.9996	
IOM air filters		0.01	0.01 0.1 10	79 – 110 (98, 7) 98 – 109 (101, 7) 89 – 110 (98, 7)	12.2 (7) 3.7 (7) 7.2 (7)	0.12 – 100 ng/mL [equivalent to 0.0012 – 1 µg/specimen]	

Matrix	Analyte (transition m/z)	LOQ (µg/specimen)	Recovery fortification level (µg/specimen)	% Recoveries range (mean, n)	Repeatability %RSD (n)	Linearity	Specificity
					Overall: 8.1 (21)	9 standards, y = 1.0E10^{8}x + 6360, r^2 = 0.9971	
Air sampling tubes		0.01	0.01 0.1 10	92 – 106 (99, 7) 74 – 105 (97, 7) 91 – 110 (102, 7)	5.0 (7) 11.7 (7) 5.5 (7) Overall: 7.8 (21)	0.12 – 100 ng/mL [equivalent to 0.0012 – 1 µg/specimen] 9 standards, y = 1.3E10^{8}x + 3321, r^2 = 0.9983	