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Estimating Half-Lives for Pesticide Dissipation from Plants

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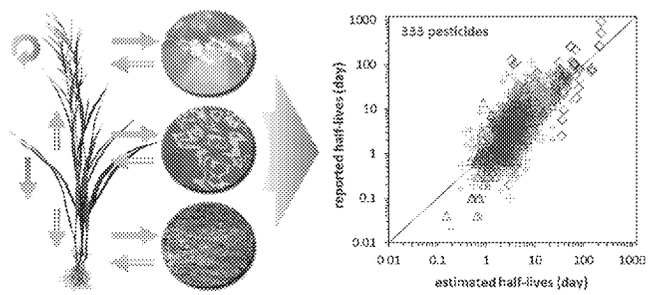
Supporting Information

ABSTRACT: Pesticide risk and impact assessment models critically rely on and are sensitive to information describing dissipation from plants. Despite recent progress, experimental data are not available for all relevant pesticide−plant combinations, and currently no model predicting plant dissipation accounts for the influence of substance properties, plant characteristics, temperature, and study conditions. In this study, we propose models to estimate half-lives for pesticide dissipation from plants and provide recommendations for how to use our results. On the basis of fitting experimental dissipation data with reported average air temperatures, we estimated a reaction activation energy of 14.25 kJ/mol and a temperature coefficient Q_{10} of 1.22 to correct dissipation from plants for the influence of temperature. We calculated a set of dissipation half-lives for 333 substances applied at 20 °C under field conditions. Half-lives range from 0.2 days for pyrethrins to 31 days for dalapon. Parameter estimates are provided to correct for specific plant species, temperatures, and study conditions. Finally, we propose a predictive regression model for pesticides without available measured dissipation data to estimate half-lives based on substance properties at the level of chemical substance class. Estimated half-lives from our study are designed to be applied in risk and impact assessment models to either directly describe dissipation or as first proxy for describing degradation.



1. INTRODUCTION

Phase partitioning, intermedia transport, and degradation mainly drive pesticide dissipation and relate to the magnitude of residues in agricultural food crops and other plants. In addition to experimental data, deterministic models can help to predict residues in harvested plants as an integral part of risk and impact assessments.[1−4] Assessment models critically rely on and are highly sensitive to estimates of rate coefficients or corresponding half-lives describing pesticide dissipation from plants.[5−8] Half-lives for individual fate processes in plants are often not available, but are instead aggregated into overall dissipation half-lives.[9] Such dissipation half-lives are either estimated from individual experimental data per pesticide or derived from other parameters, such as soil half-lives.[5] However, both approaches fail to provide estimates that account for the variability of dissipation from plants.

Measurement-based half-lives generally refer to overall dissipation from plants, with degradation and intermedia exchange, such as uptake from soil into roots, as contributing mechanisms. Two studies reviewed measured pesticide dissipation from plants. Willis and McDowell (1987)[10] reviewed half-lives reported for 79 pesticides in the leaves of 53 plant species. Fantke and Juraske (2013)[11] reviewed half-lives reported for 346 pesticides in various parts of 183 plant species. Nevertheless, experimental data are not available for all pesticide−plant combinations and, when available, show large variation in half-lives reported per pesticide. Accordingly, attempts have been made to predict dissipation from plants based on half-lives in soil using generic extrapolation factors across pesticides ranging from 2 to 16 depending on the assessed pesticide−plant combinations, that is, some pesticides are estimated to dissipate 2 to 16 times faster from certain plants than from soil.[5,12−14] In a more recent study, the accuracy of using generic coefficients was questioned and, instead, a regression model was proposed for comparing soil and plant surface half-lives for 53 substances.[15] However, soil half-lives may vary by several orders of magnitude as a function of site-specific conditions,[16,17] whereas variation of half-lives in plants under field conditions is reported to be less than a factor of 30 across 1489 pesticide−plant combinations.[11] Hence, only using soil half-lives is not necessarily appropriate to estimate

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plant half-lives. Instead, dissipation processes in plants depend on a combination of substance properties,[7,18] plant characteristics,[3,10,19] and environmental conditions,[10,20,21] of which temperature plays a predominant role.[19,20] Furthermore, half-lives might differ between plant types (trees, cereals, root/tuber crops, etc.) as well as between components, such as leaves (surface), fruit pulp (interior), and roots (interior, below-ground). Juraske et al. (2008)[5] for example show that for bromopropylate and α-cypermethrin applied to tomato, dissipation from the plant interior is four times faster than from the plant surface assuming that enzymatic activity in plants is quicker than photodegradation on plant surfaces. However, it remains unclear whether this holds true for other pesticide−plant combinations as both enzymatic transformation and photodegradation are influenced by highly variable aspects like substance vapor pressure, plant surface roughness, and air temperature.[19,22]

As a result, estimated dissipation from plants remains highly uncertain with dominant influences not being explicitly represented. In this study, we present a sophisticated method for estimating pesticide dissipation half-lives in plants from substance properties, plant characteristics, study conditions, and temperature with focus on three objectives: (1) to characterize 4442 measured pesticide dissipation half-lives in plants by describing their distribution and by estimating the influence of temperature from a subset of 1030 data points with reported growth season average air temperatures; (2) to estimate geometric means and 95% confidence intervals of dissipation half-lives at 20 °C under field conditions for 333 reported pesticides, using multiple imputations to accommodate missing temperatures associated with reported half-lives; (3) to propose a regression-based model to predict dissipation half-lives for pesticides as a function of temperature, chemical class, selected substance properties, and plant characteristics, and evaluate model prediction performance using sums of squares of leave-one-out residuals for excluded data points. Our models and estimated half-lives are designed to be ultimately used as input in risk and impact assessments models.

2. MATERIALS AND METHODS

2.1. Characterizing Reported Dissipation Half-Lives in Plants. As a starting point, we use a recently published review of measured dissipation half-lives for a combination of 346 pesticides and 183 plants.[11] Excluding substances where information regarding chemical substance class (e.g., carbamates, triazoles), substance properties, or plant type was insufficient or unclear, 4442 data points (*reported half-lives*) for 333 pesticides are used. We investigated the distribution of half-lives including testing for normality and log-normality for each pesticide. We calculated pesticide-specific geometric mean half-lives and analyzed their distribution across pesticides. The results are summarized in probability distribution and quantile-quantile plots. SAS 9.4 was used as statistical analysis software throughout.

We first assessed the widely recognized influence of temperature T (K) on dissipation half-lives $HL^{\circ}_{\text{reported},T}$ (day) based on a subset of data points with reported average air temperature during growth (n = 1030, indicated by superscripted open circle ○) using a log−linear regression model (Model I). Besides temperature, we included as predictors in this model indicator variables for substance, plant, and cold storage, where cold storage refers to reduced light and temperature conditions after harvest:

$$\log HL^{\circ}_{\text{reported},T} = \alpha^{\circ} + \beta^{\circ}_{\text{subst},i} \times X_{\text{subst},i} + \beta^{\circ}_{\text{plant},j} \times X_{\text{plant},j} + \beta^{\circ}_{\text{storage}} \times X_{\text{storage}} + \beta^{\circ}_{T} \times (T - T_{\text{ref}}) \quad (1)$$

with intercept α°, estimates to account for substances $\beta^{\circ}_{\text{subst},i}$, plants $\beta^{\circ}_{\text{plant},j}$, cold storage conditions $\beta^{\circ}_{\text{storage}}$, and air temperature β°_{T} (K^{-1}), where T_{ref} = 293.16 K (20 °C) is set as reference air temperature. $X_{\text{subst},i}$, $X_{\text{plant},j}$, and X_{storage} are categorical variables for substance, plant, and cold storage coded to represent two-category factors, $X \rightarrow \{0,1\}$,[23] with X_{storage} = 1 for data points referring to cold storage conditions and with X_{storage} = 0 for all other data points. Correspondingly, $X_{\text{subst},i}$ and $X_{\text{plant},j}$ are column vectors of zeros except for the ith substance and jth plant, respectively. Plant components (e.g., leaves) are tested, but finally excluded from the list of predictors because of nonsignificance and autocorrelation between certain plant−plant component combinations (see Supporting Information (SI), section S-3).

Of special interest is the temperature estimate β°_{T} (K^{-1}), which serves as input for analyzing the full set of reported dissipation half-lives in plants. We used β°_{T} to calculate three components used by several risk and impact assessment models, namely (a) reaction activation energy E_a (kJ/mol):

$$E_a = -\beta^{\circ}_{T} \times R \times (T \times T_{\text{ref}}) \quad (2)$$

where R (kJ/mol/K) is the molar gas constant, (b) temperature coefficient Q_{10}:

$$Q_{10} = \exp(-\beta^{\circ}_{T} \times \Delta T) \quad (3)$$

where ΔT = 10 K, and (c) Arrhenius temperature correction factor cf_T relating different dissipation rate constants to temperature:

$$cf_T = \exp(-\beta^{\circ}_{T}) \quad (4)$$

Details how to derive E_a, Q_{10}, and cf_T from β°_{T} are given in the SI, section S-1.

2.2. Temperature Imputation and 333 Recommended Half-Lives at Reference Conditions. Temperature is considered one of the main predictors for estimating dissipation half-lives in plants for all reported pesticides. Since air temperatures were reported for only 23% of the data, we imputed missing values using information on optimal air temperature ranges for reported plants and techniques to substitute temperatures of arbitrary missing pattern (see SI, section S-2). We used multiple imputations to replace each missing temperature with a set of 10 plausible values representing the uncertainty of the imputation.[24] The multiply imputed data sets were each analyzed using linear regression, and results were combined by taking the average of regression estimates and a combination of the within- and between-regression variances. This procedure yields valid statistical inferences properly reflecting the uncertainty due to missing temperature data.[24−26]

Each reported half-life is now associated with a specific temperature, either reported or imputed when missing, yielding a combined set of 4442 half-lives $HL^{\bullet}_{\text{reported},T}$ (indicated by superscripted filled circle, ●). To account for imputation-related uncertainty, we provide for each half-life the relative increase in variance due to missing temperatures. Our second regression model estimates the half-life of a pesticide along with

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00002

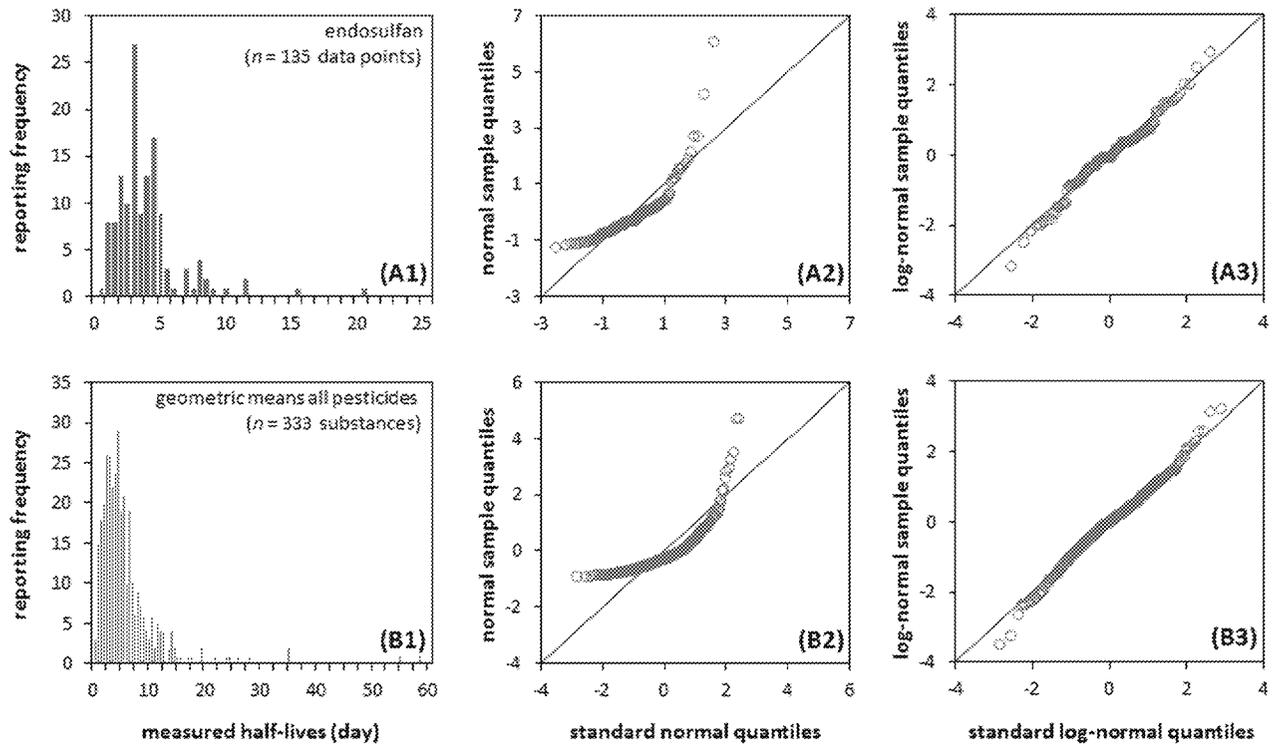

Figure 1. Probability distribution and quantile−quantile plots for assessing the normality and log-normality of the distribution of reported dissipation half-lives in plants for endosulfan (A1−A3) and of geometric means of reported dissipation half-lives in plants of all pesticides (B1−B3).

plant-specific and cold storage correction factors (Model II), where all coefficients are based on the combined data set:

$$\log HL^{\bullet}_{\text{reported},T} = \underbrace{\alpha^{\bullet} + \beta^{\bullet}_{\text{subst},i} \times X_{\text{subst},i}}_{\log HL^{\bullet}_{\text{ref},i}\ \text{(see Table 1)}} + \beta^{\bullet}_{\text{plant},j} \times X_{\text{plant},j} + \beta^{\bullet}_{\text{storage}} \times X_{\text{storage}} + \beta^{\bullet}_{T} \times (T - T_{\text{ref}}) \quad (5)$$

$HL^{\bullet}_{\text{ref},i}$ (day) denotes the geometric mean pesticide half-life corrected for an application to an average plant (average of 30 plant estimates) under reference conditions (field conditions, T_{ref} = 293.16 K), and $\beta^{\bullet}_{\text{plant},j}$, $\beta^{\bullet}_{\text{storage}}$, and $\beta^{\bullet}_{T} = \beta^{\circ}_{T}$ (K^{-1}) denote the multiplicative plant, cold storage, and temperature correction factors. As a sensitivity study, we tested our model with and without cold storage data to ensure that cold storage does not bias the regression. For each $HL^{\bullet}_{\text{ref},i}$ we also provide the 95% confidence interval obtained from t-distributions and standard error accounting for covariance between parameter estimates. Model prediction performance is evaluated using predicted leave-one-out residual sums of squared errors (PRESS), that is, each observation is predicted using all other $n - 1$ observations.[27]

2.3. Regression Model for Predicting Dissipation Half-Lives from Pesticide Properties. We finally want to predict half-lives of pesticides without reported dissipation data, based on their physicochemical properties. From the analysis of reported data, we hypothesize the following qualitative dependencies: (i) Higher temperature triggers microbial and chemical degradation in plants, thereby shortening half-lives.[10,18,20] (ii) Cold storage has additional effects beyond low temperature due to reduced light conditions, which diminishes dissipation.[28,29] (iii) Plant-specific characteristics like growth, transpiration, uptake mechanisms, location of fruits/tubers, and size and shape of leafy foliage are all likely to distinctly influence pesticide dissipation.[3,30,31] (iv) Molecular weight, phase partition coefficients, and saturation vapor pressure have been reported as important physicochemical properties affecting environmental fate processes of chemicals.[7,21,32] (v) Pesticides within the same substance class, such as carbamates, show similar patterns in terms of half-life dependencies. (vi) Finally, aspects like degradation in soil and air may additionally correlate with dissipation from plants,[10,18,31] along with ionization potential, polarity, stereoselectivity, and other substance-specific properties. We used this set of hypotheses in a final generic log−linear model (Model III) for predicting dissipation half-lives of pesticides belonging to a certain substance class (e.g., triazoles, carbamates) applied to a particular plant under specific conditions and temperature, $HL^{\circ}_{\text{predicted},T}$ (day), as

$$\log HL^{\circ}_{\text{predicted},T} = \alpha'^{\circ} + \beta'^{\circ}_{\text{subst-class},k} \times X_{\text{subst-class},k} + \beta'^{\circ}_{\text{plant},j} \times X_{\text{plant},j} + \beta'^{\circ}_{\text{storage}} \times X_{\text{storage}} + \beta'^{\circ}_{T} \times (T - T_{\text{ref}}) + \sum_{l=1}^{n} \beta'^{\circ}_{l} \times p_l \quad (6)$$

with intercept α'°, estimates for substance class $\beta'^{\circ}_{\text{subst-class},k}$, plant $\beta'^{\circ}_{\text{plant},j}$, cold storage conditions $\beta'^{\circ}_{\text{storage}}$, temperature $\beta'^{\circ}_{T} \triangleq \beta^{\circ}_{T}$ (K^{-1}; defined from temperature estimate of Model I), and substance-specific properties β'°_{l}, as well as variables with information about substance class $X_{\text{subst-class},k}$, plant $X_{\text{plant},j}$, and cold storage X_{storage}, average air temperature T (K), reference air temperature T_{ref} (K), and substance properties p_l.

Model Selection. We start from eq 6 to select and fit parameters of the final predictive Model III. Because of the difficulty of model selection in our settings, we used more than one selection method with the minimum Akaike information

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00003

criterion (AIC) statistic as major criterion,[33,34] and with separately considering significance levels. Model III is applied to the subset of 1030 data points with reported temperature. We only considered plants with $n \geq 8$ reported temperatures and substance classes with $n \geq 3$ reported substances per class to ensure reliability of estimates as a function of number of data points (see Figure 4). The minimum number of reported temperatures per plant is set to 8 to yield variances of less than a factor 2, whereas 3 substances per substance class are chosen as minimum in order not to overfit classes. Remaining plants and substance classes are pooled into categories *other plants* and *other substance classes*, respectively.

3. RESULTS

3.1. Variation of Reported Dissipation Half-Lives in Plants. Figure 1 (A1) presents the probability distribution of dissipation half-lives in plants for endosulfan as example substance with the largest number of data-points (n = 135). For endosulfan, 95% of half-lives fall in the range between 0.5 and 9 days. The shortest half-life (0.43 days) was reported in tomato foliage (27 °C, field conditions)[35] and the longest half-life (20.1 days) in cherry tomato fruits (17.5 °C, greenhouse).[36] Half-life distributions for other pesticides show skewed distributions similar to endosulfan. Figure 1 (B1) presents the distribution of geometric means of half-lives of all considered substances (n = 333). Interestingly, the variation across geometric means of half-lives of all substances is not much larger than the variation of reported half-lives for endosulfan, showing that 95% fall in the range between 0.5 and 13 days. This may be explained by the influence of limited dissipation through plant growth dilution, since plant mass can double within a few days. Half-lives greater than 30 days are mostly reported for cold storage conditions (not shown in Figure 1).[11] The large variation of reported half-lives for the same substance emphasizes the need for carefully analyzing these half-lives before reporting recommended values based on experimental data. Thus, we follow an iterative approach by introducing the different regression models (Models I, II, III). Figure 1 also shows quantile−quantile plots for normality (A2, B2) and log-normality (A3, B3) of reported half-lives, indicating a log-normal trend across data points per substance and across substances.

3.2. Coefficients for Temperature Correction. Qualitatively assessing the relationship between reported dissipation half-lives and air temperatures shows that they are inversely correlated (Figure 2). The remaining variability for each temperature is mainly influenced by substance properties, plant characteristics, and environmental conditions.

The first regression (Model I) on the subset of 1030 data points with reported air temperature supports that temperature and additionally study conditions (field, cold storage) and plant species are important predictors to estimate pesticide dissipation. Model I yields an adjusted coefficient of determination of R^2_{adj} = 0.638 and a root-mean-squared error of RMSE = 0.27 for a temperature estimate $\beta_T^\circ = -1.995 \times 10^{-2}$ K^{-1} (p-value < 0.0001) with estimated half-lives plotted against reported half-lives in Figure 3 (Model I). The RMSE of Model I corresponds to 95% of predicted half-lives falling within a factor 3 of reported half-lives. Excluding cold storage data influences β_T° less than 3%. The variance in dissipation half-lives is explained by each predictor individually and with combined predictors for temperature (23.2% individually/combined), substance (52% individually, 36% combined), plant (26.8% individually, 6.3% combined), and study conditions (21% individually, 5.2% combined). Parameter estimates, standard errors, and p-values used in Model I are given in Table S2 (SI). From β_T° we obtain a reaction activation energy E_a = 14.25 kJ/mol, a temperature coefficient Q_{10} = 1.22, and an Arrhenius temperature correction factor cf_T = 1.02.

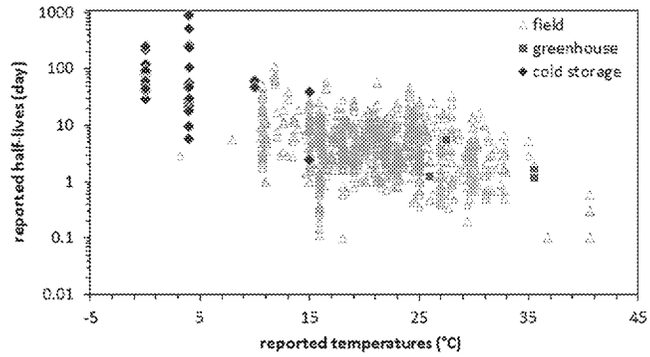

Figure 2. Reported pesticide dissipation half-lives in plants plotted as a function of reported average air temperatures (n = 1030), disaggregated according to field, greenhouse, and cold storage conditions.

3.3. Dissipation Half-Lives for 333 Reported Pesticides at 20 °C. Our regression Model II provides the corrected geometric means of half-lives for 333 pesticides applied to an average plant under reference conditions, $HL^{\bullet}_{ref,i}$. These half-lives calculated from intercept and substance estimates of eq 5 are listed in Table 1 and shown in Figure 3 (Model II). Half-lives range from 0.21 days for plant-derived pyrethrins to 30.8 days for dalapon with 95% of all half-lives falling in the range between 0.9 and 18 days. While on average 13 reported half-lives were available per substance with a maximum for endosulfan (n_{HL} = 135), imidacloprid (n_{HL} = 130), and methomyl (n_{HL} = 122), only 6 temperatures were reported on average per substance (n_T = 36 for endosulfan, n_T = 16 for imidacloprid, and n_T = 19 for methomyl, respectively). Across substances, temperature imputation contributes with 5.1% to standard errors of half-lives. Minimal contribution is found for substances with temperatures being reported for each half-life, such as for propamocarb hydrochloride, ziram, and fluazinam (contribution of temperature imputation to half-life standard errors < 0.1%), while maximal contribution is associated with substances, for which no reported temperatures are available, such as for propargite, nicotine, and dalapon (contribution to standard error between 1% and 17%). For calculating $HL^{\bullet}_{ref,i}$ from the combined data set of 4442 data points including imputed temperatures, we slightly increased the RMSE by 2% compared to the restricted data set with reported temperatures. The higher number of reported half-lives in the combined data set nevertheless enables a strong reduction in the uncertainty of our half-lives per substance as shown in Figure S3 (SI). Additionally, we consider more substances, plants, and data points per substance and plant in the combined data set and in Model II. Excluding cold storage from Model II yields an average deviation of less than 0.2%, and for 95% of the data yields deviations of less than 3.8%. For five substances, deviations are greater than 10% (indicated in Table 1); for those substances, half-lives estimated without cold storage data are given in Table S4 (SI).

Model II shows R^2_{adj} = 0.458 and a root-mean-squared error of RMSE = 0.3; that is, 95% of predicted half-lives fall within a

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00004

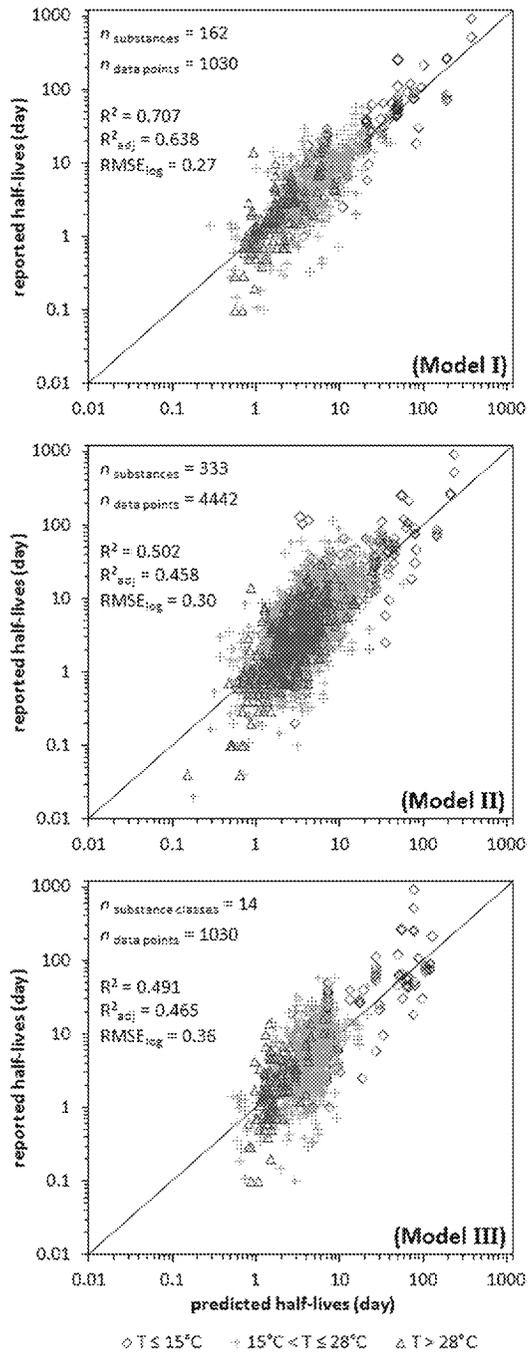

Figure 3. Reported vs predicted pesticide dissipation half-lives in plants disaggregated according to different temperature ranges: Model I, initial regression model fitted on $n = 1030$ data points with reported average air temperatures to yield temperature estimate; Model II, regression model fitted on $n = 4442$ data points to estimate dissipation half-lives under reference field conditions for 333 reported substances; Model III, predictive regression model to estimate dissipation half-lives for 14 substance classes from chemical properties of $n = 1030$ data points with reported average air temperatures. Root-mean-squared errors RMSE are given for the log of half-lives as fitted in Models I–III.

factor 3.4 of reported half-lives. In addition to dissipation half-lives for average plant and reference air temperature, Model II provides estimates for the influence of specific plants and for cold storage conditions (Table S3, SI). These estimates can be used to correct reference half-lives in Table 1 for pesticides applied to specific plants under given study conditions and temperature. For example, we can calculate the half-life of acetamiprid applied to Chinese cabbage under field conditions at 291.16 K (18 °C). We start from the log of the half-life for acetamiprid in Table 1, $\log HL_{\text{ref,acetamiprid}} = \alpha^{\bullet} + \beta^{\bullet}_{\text{acetamiprid}} = 0.595 + 0.16 = 0.755$. We then combine $\log HL_{\text{ref,acetamiprid}}$ in eq 5 with plant estimates taken from Table S3 (SI) and with temperature estimate $\beta^{\bullet}_{T}$ multiplied by the temperature difference to the reference air temperature, yielding $\log HL^{\text{Chinese cabbage,18 °C}}_{\text{acetamiprid}} = \log HL_{\text{ref,acetamiprid}} + \beta^{\bullet}_{\text{Chinese cabbage}} + \beta^{\bullet}_{T} \times (291.16\ \text{K} - T_{\text{ref}}) = 0.755 - 0.05 - 1.995 \times 10^{-2}\ \text{K}^{-1} \times (291.16\ \text{K} - 293.16\ \text{K}) = 0.745$, and thus a corrected half-life of $10^{0.745} = 5.56$ days. This half-life is 2.3% shorter than the reference half-life of 5.69 days at 20 °C (Table 1), which is explained by the influence of plant counteracting the influence of temperature in this example. Half-life corrections from plant correction parameter estimates range between $10^{-0.70} = 0.2$ days for zucchini and $10^{0.36} = 2.3$ days for lemon.

Standard errors of substance and plant parameter estimates (Figure 4) for 333 pesticides and 30 plants approximately follow a decrease proportional to $1/\sqrt{n}$. Both graphs in Figure 4 show that with a minimum of 20 data points, we get a standard error of 0.08, implying deviations between reported and predicted half-lives of a factor $10^{0.08 \times \text{Student's } t}$. For a Student's t-value of 2.086 corresponding to 20 data points as degree of freedom,[37] we get a factor 1.5 deviation. Standard errors are slightly higher for grass, wheat, and rice, which might be attributable to the wide range of grass species aggregated into a single plant estimate, the ubiquitous geographical distribution of wheat grown under diverse environmental conditions, and the different cultivation methods of rice (paddy fields, dryland farming, etc.).

3.4. Estimated Dissipation Half-Lives from Pesticide Classes and Properties. Model III aims at predicting half-lives of pesticides without measured dissipation data, as a function of their chemical class and properties. Stepwise regression is applied to the subset of 1030 data points with reported average air temperatures to ensure maximum accuracy, yielding the following final log–linear model (Model III):

$$\begin{aligned}\log HL^{\circ}_{\text{predicted},T} = {} & \alpha'^{\circ} + \beta'^{\circ}_{\text{subst-class},k} \times X_{\text{subst-class},k} \\ & + \beta'^{\circ}_{\text{plant},j} \times X_{\text{plant},j} + \beta'^{\circ}_{\text{storage}} \times X_{\text{storage}} \\ & + \beta'^{\circ}_{T} \times (T - T_{\text{ref}}) + \beta'^{\circ}_{MW} \times MW \\ & + \beta'^{\circ}_{K_{\text{OW}}} \times \log K_{\text{OW}} + \beta'^{\circ}_{P_{\text{V}}} \times P_{\text{V}} \qquad (7)\end{aligned}$$

with intercept α'°, estimates for substance class $\beta'^{\circ}_{\text{subst-class},k}$, plant $\beta'^{\circ}_{\text{plant},j}$, cold storage conditions $\beta'^{\circ}_{\text{storage}}$, temperature β'°_{T} (K^{-1}), substance molecular weight β'°_{MW} (mol/g), octanol/water partition coefficient $\beta'^{\circ}_{K_{\text{OW}}}$, and saturation vapor pressure $\beta'^{\circ}_{P_{\text{V}}}$ (Pa^{-1}), variables with information about substance class $X_{\text{subst-class},k}$, plant $X_{\text{plant},j}$, and cold storage X_{storage} with all $X \rightarrow \{0,1\}$, average air temperature T (K), reference air temperature T_{ref} (K), molecular weight MW (g/mol), octanol/water partition coefficient $\log K_{\text{OW}}$, and saturation vapor pressure P_{V} (Pa). All other tested substance properties (air/water and soil organic carbon/water partition coefficients, half-lives in air and soil) that are not included in Model III either did not significantly improve the model or showed strong correlation with another included variable. Model III shows $R^2_{\text{adj}} = 0.465$, an Akaike information criterion AIC = −960.4, and a root-mean-

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00005

Table 1. Target Class (TC), CAS Number, Predicted Geometric Mean of Dissipation Half-Life $HL^{\bullet}_{\mathrm{ref},i}$ (day), with Lower and Upper 95% Confidence Interval (CI) Limits in Parentheses, Relative Increase in Standard Error by Including Data with Imputed Average Air Temperatures θ_{imp} (%), Total Number of Reported Data Points n_{HL}, and Number of Data Points with Reported Average Air Temperatures n_T for 333 pesticides

pesticide	CAS RN	TC[a]	$HL^{\bullet}_{\mathrm{ref},i}$ (95% CI) (day)		θ_{imp}(%)	n_{HL}	n_T
2,4,5-T	93-76-5	H	9.90	(3.72−26.30)	1.75	3	2
2,4-D	94-75-7	H	2.92	(2.10−4.04)	1.68	9	5
2,4-D-dimethylammonium	2008-39-1	H	2.20	(1.06−4.56)	12.86	4	–
3-hydroxycarbofuran	16655-82-6	M	9.96	(6.13−16.17)	8.24	6	–
abamectin	71751-41-2	I	3.81	(3.59−4.04)	4.01	50	1
acephate	30560-19-1	I	4.64	(4.39−4.90)	4.85	54	14
acetamiprid	135410-20-7	I	5.69	(5.26−6.15)	6.57	38	14
acetochlor	34256-82-1	H	1.86	(0.90−3.86)	8.65	4	–
acrinathrin	101007-06-1	I	3.01	(1.98−4.56)	2.95	7	5
aldicarb	116-06-3	I	6.08	(5.12−7.22)	8.06	17	1
aldoxycarb	1646-88-4	I	10.28	(7.15−14.79)	5.05	8	–
aldrin	309-00-2	I	1.98	(0.11−36.30)	0.11	1	1
alpha-cypermethrin	67375-30-8	I	5.15	(3.73−7.12)	6.82	9	–
alpha-endosulfan	959-98-8	I	1.66	(1.30−2.12)	8.15	12	4
aminocarb	2032-59-9	I	4.34	(3.11−6.04)	1.76	9	7
amitraz	33089-61-1	I	3.83	(0.89−16.40)	7.60	2	–
anabasine	494-52-0	I	1.23	(0.59−2.53)	1.57	4	–
anilofos	64249-01-0	H	2.46	(1.37−4.42)	6.32	5	–
aramite	140-57-8	A	2.40	(1.16−4.96)	8.03	4	–
azadirachtin	11141-17-6	I	1.60	(1.50−1.69)	2.71	57	6
azimsulfuron	120162-55-2	H	3.35	(1.27−8.84)	9.44	3	–
azinphos-methyl	86-50-0	I	4.47	(4.21−4.74)	2.10	59	18
azoxystrobin	131860-33-8	F	3.53	(3.30−3.78)	8.35	42	5
benalaxyl	71626-11-4	F	2.26	(1.73−2.94)	4.03	11	–
bendiocarb	22781-23-3	I	5.70	(4.37−7.43)	9.64	11	–
benfuracarb	82560-54-1	I	4.64	(1.08−19.87)	8.09	2	–
benodanil	15310-01-7	F	6.99	(5.06−9.66)	6.60	9	–
benomyl	17804-35-2	F	4.26	(2.62−6.91)	1.27	6	4
beta-cypermethrin	65731-84-2	I	5.08	(1.92−13.38)	7.56	3	–
bifenthrin	82657-04-3	I	3.39	(3.22−3.57)	2.91	59	8
bitertanol	55179-31-2	F	4.90	(3.55−6.78)	9.40	9	1
boscalid	188425-85-6	F	6.63	(4.08−10.77)	4.09	6	–
bromomethane	74-83-9	I	16.91	(3.95−72.45)	2.44	2	–
bromopropylate	18181-80-1	A	4.01	(2.90−5.55)	4.73	9	–
bromoxynil	1689-84-5	H	1.97	(0.75−5.20)	0.26	3	3
bromoxynil octanoate	1689-99-2	H	2.85	(1.38−5.89)	4.38	4	1
buprofezin[b]	69327-76-0	I	6.42	(5.69−7.26)	1.61	24	13
cadusafos	95465-99-9	I	22.99	(11.11−47.56)	4.32	4	–
captan	133-06-2	F	4.45	(4.06−4.88)	3.61	33	16
carbaryl	63-25-2	I	4.05	(3.85−4.26)	4.44	59	6
carbendazim	10605-21-7	F	5.79	(5.32−6.29)	4.52	35	5
carbofuran	1563-66-2	I	4.62	(4.21−5.08)	3.52	31	4
carbophenothion	786-19-6	I	3.78	(3.23−4.42)	10.18	19	–
carbosulfan	55285-14-8	I	2.11	(1.85−2.42)	4.96	22	6
carfentrazone-ethyl	128639-02-1	H	6.31	(1.47−27.04)	4.55	2	–
chinomethionat	2439-01-2	F	3.58	(0.83−15.32)	0.28	2	2
chlorantraniliprole	500008-45-7	I	3.38	(2.23−5.12)	9.14	7	–
chlordane	57-74-9	I	5.74	(2.18−15.13)	11.68	3	–
chlordimeform	6164-98-3	A	0.91	(0.51−1.62)	1.65	5	2
chlorfenapyr	122453-73-0	I	3.38	(2.35−4.86)	5.23	8	1
chlorfenson	80-33-1	A	2.77	(0.15−50.64)	5.10	1	–
chlorfenvinphos	470-90-6	I	10.20	(4.92−21.16)	5.66	4	–
chlorfluazuron	71422-67-8	I	5.33	(4.09−6.95)	6.75	11	5
chlorimuron	99283-00-8	H	5.57	(2.11−14.73)	5.20	3	–
chlormequat	7003-89-6	P	6.05	(2.92−12.53)	7.01	4	–
chlorobenzilate	510-15-6	I	6.57	(5.15−8.40)	1.72	12	8
chloroneb	2675-77-6	F	16.87	(9.43−30.18)	6.66	5	–
chlorothalonil	1897-45-6	F	5.02	(4.74−5.31)	5.26	58	7

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00006

Table 1. continued

pesticide	CAS RN	TC[a]	$HL^{\bullet}_{ref,i}$ (95% CI) (day)		θ_{imp}(%)	n_{HL}	n_T
chlorphoxim	14816-20-7	I	1.31	(0.07−23.99)	4.22	1	–
chlorpyrifos	2921-88-2	I	4.01	(3.90−4.12)	7.20	107	29
chlorpyrifos-methyl	5598-13-0	I	3.27	(2.89−3.70)	12.73	24	–
chlorsulfuron	64902-72-3	H	0.30	(0.07−1.27)	4.99	2	–
chlozolinate	84332-86-5	F	7.93	(4.88−12.88)	9.85	6	–
cinerin I	25402-06-6	I	1.83	(0.43−7.82)	0.09	2	2
cinerin II	121-20-0	I	1.95	(0.46−8.35)	0.09	2	2
clopyralid	1702-17-6	H	4.15	(2.00−8.58)	3.23	4	–
clothianidin	210880-92-5	I	8.27	(5.46−12.54)	2.10	7	–
cyanazine	21725-46-2	H	3.36	(2.29−4.94)	10.50	8	–
cyanofenphos	13067-93-1	I	17.52	(0.96−320.98)	9.14	1	–
cyanophos	2636-26-2	I	1.46	(0.08−26.75)	2.87	1	–
cyantraniliprole	736994-63-1	I	10.84	(4.11−28.58)	0.30	3	3
cyazofamid	120116-88-3	F	8.16	(3.10−21.52)	9.68	3	–
cycloheximide	66-81-9	F	1.12	(0.54−2.32)	0.20	4	4
cycloprate	54460-46-7	A	8.18	(4.57−14.64)	11.20	5	–
cyfluthrin	68359-37-5	I	2.39	(2.24−2.54)	3.46	47	5
cyhalofop-butyl	122008-85-9	H	5.00	(1.17−21.38)	4.13	2	1
cyhalothrin	68085-85-8	I	4.77	(4.31−5.29)	5.40	29	4
cymoxanil	57966-95-7	F	1.61	(0.38−6.90)	9.13	2	–
cypermethrin	52315-07-8	I	4.24	(4.10−4.38)	5.84	108	53
cyproconazole	94361-06-5	F	6.28	(3.51−11.23)	4.15	5	–
cyprodinil	121552-61-2	F	5.70	(5.21−6.23)	5.70	33	13
cyromazine	66215-27-8	I	21.63	(10.45−44.78)	4.83	4	–
dalapon	75-99-0	H	30.80	(7.20−131.81)	15.21	2	–
DDT	50-29-3	I	10.78	(9.78−11.89)	3.32	30	4
deltamethrin	52918-63-5	I	3.76	(3.65−3.89)	8.30	93	25
demeton	8065-48-3	I	7.59	(5.83−9.89)	2.44	11	7
desmethylformamide pirimicarb	27218-04-8	I	1.37	(0.49−3.83)	5.02	2	–
diafenthiuron	80060-09-9	I	3.56	(2.47−5.13)	7.44	8	–
dialifos	10311-84-9	I	14.92	(10.79−20.62)	5.18	9	1
diazinon	333-41-5	I	2.16	(1.93−2.42)	1.82	26	10
dicamba	1918-00-9	H	6.30	(3.04−13.08)	2.94	4	2
dichlofluanid	1085-98-9	F	9.81	(6.04−15.95)	3.69	6	–
dichlorvos	62-73-7	I	1.12	(0.81−1.55)	4.32	9	–
diclofop-methyl	51338-27-3	H	1.53	(0.36−6.53)	2.64	2	–
dicloran	99-30-9	F	23.78	(14.61−38.70)	7.62	6	–
dicofol	115-32-2	A	3.45	(2.87−4.14)	5.45	16	–
dieldrin	60-57-1	I	5.13	(4.02−6.54)	10.48	12	2
diethofencarb	87130-20-9	F	2.55	(0.59−10.89)	0.10	2	2
difenoconazole	119446-68-3	F	5.02	(4.65−5.43)	8.55	38	2
diflubenzuron	35367-38-5	I	9.76	(7.99−11.92)	4.00	15	8
dimefox	115-26-4	I	12.92	(8.53−19.57)	8.70	7	–
dimetachlone	24096-53-5	F	6.85	(3.31−14.16)	9.09	4	–
dimethoate	60-51-5	I	3.61	(3.50−3.72)	6.52	94	20
dimethomorph	110488-70-5	F	5.14	(3.57−7.40)	6.97	8	1
diniconazole	83657-24-3	F	4.25	(2.38−7.61)	6.95	5	–
dinocap	39300-45-3	F	8.28	(5.09−13.46)	6.25	6	2
dinotefuran	165252-70-0	I	10.06	(3.82−26.53)	2.86	3	–
dioctyldiethylenetriamine	57413-95-3	B	9.75	(4.70−20.20)	4.41	4	–
dioxabenzophos	3811-49-2	I	1.38	(0.32−5.89)	3.50	2	–
dioxathion	78-34-2	I	2.82	(1.36−5.84)	6.89	4	–
dithianon	3347-22-6	F	11.65	(4.41−30.73)	0.19	3	3
dithiopyr	97886-45-8	H	5.03	(2.43−10.41)	3.42	4	–
dodine	2439-10-3	F	3.32	(1.85−5.95)	8.22	5	–
emamectin benzoate	155569-91-8	I	1.83	(1.21−2.77)	4.43	7	3
endosulfan	115-29-7	I	3.81	(3.72−3.89)	1.62	135	36
endosulfan sulfate	1031-07-8	M	3.69	(1.40−9.73)	8.28	3	–
endrin	72-20-8	I	2.48	(0.94−6.55)	3.40	3	1
EPN	2104-64-5	I	2.55	(2.00−3.25)	1.76	12	9
epoxiconazole	133855-98-8	F	13.31	(9.95−17.82)	5.07	10	4

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00007

Table 1. continued

pesticide	CAS RN	TC[a]	$HL^{\bullet}_{ref,i}$ (95% CI) (day)		θ_{imp}(%)	n_{HL}	n_T
esfenvalerate	66230-04-4	I	1.60	(0.77−3.31)	5.90	4	–
ethametsulfuron-methyl	97780-06-8	H	5.44	(3.04−9.73)	0.24	5	5
ethion	563-12-2	I	3.76	(3.25−4.35)	6.97	20	2
ethirimol	23947-60-6	F	5.02	(0.27−91.88)	7.72	1	–
Ethofumesate	26225-79-6	H	2.62	(0.61−11.20)	3.66	2	1
ethylenethiourea	96-45-7	F	8.44	(4.08−17.47)	9.57	4	–
ethylicin	682-91-7	F	2.56	(1.78−3.70)	8.29	8	–
etofenprox	80844-07-1	I	2.63	(1.62−4.27)	3.73	6	–
etoxazole	153233-91-1	A	3.77	(2.10−6.74)	5.59	5	–
famoxadone	131807-57-3	F	5.63	(4.32−7.35)	3.45	10	4
fenamidone	161326-34-7	F	7.14	(3.45−14.79)	2.50	4	1
fenamiphos	22224-92-6	N	6.74	(3.26−13.93)	8.77	4	–
fenarimol	60168-88-9	F	5.65	(0.31−103.43)	4.56	1	–
fenazaquin	120928-09-8	A	3.74	(3.07−4.54)	5.90	15	–
fenbuconazole	114369-43-6	F	7.75	(2.94−20.45)	2.31	3	–
fenhexamid	126833-17-8	F	7.70	(5.08−11.67)	4.55	7	–
fenitrothion	122-14-5	I	3.73	(3.49−4.00)	2.37	43	17
fenobucarb	3766-81-2	I	5.50	(3.07−9.85)	3.63	5	3
fenoprop	93-72-1	H	4.05	(0.94−17.42)	5.51	2	1
fenoxanil	115852-48-7	F	7.72	(5.08−11.74)	7.50	7	–
fenoxaprop-P-ethyl	71283-80-2	H	2.19	(1.44−3.34)	3.74	7	–
fenoxycarb	79127-80-3	I	7.48	(2.84−19.72)	2.00	3	–
fenpropathrin	39515-41-8	I	3.46	(2.28−5.24)	4.85	7	3
fenpropidin	67306-00-7	F	4.17	(0.97−17.89)	6.05	2	–
fenpropimorph	67564-91-4	F	1.59	(0.60−4.20)	1.05	3	2
Fenpyroximate	134098-61-6	A	1.51	(0.73−3.13)	5.35	4	–
fensulfothion	115-90-2	I	2.83	(0.66−12.11)	2.06	2	–
fenthion	55-38-9	I	2.49	(2.14−2.90)	2.92	20	5
fenthion sulfoxide[b]	3761-41-9	M	12.85	(7.17−23.02)	4.17	5	3
fenvalerate	51630-58-1	I	4.71	(4.50−4.94)	8.52	64	23
fipronil	120068-37-3	I	5.76	(4.74−6.99)	6.29	15	–
florasulam	145701-23-1	H	0.66	(0.37−1.19)	1.68	5	4
fluazinam	79622-59-6	F	3.77	(0.21−69.10)	0.06	1	1
flubendiamide	272451-65-7	I	2.25	(2.00−2.55)	7.12	24	–
flucythrinate	70124-77-5	I	5.78	(3.23−10.34)	4.87	5	2
fludioxonil[b]	131341-86-1	F	5.24	(4.55−6.03)	4.60	21	11
flufenoxuron	101463-69-8	I	15.32	(10.10−23.22)	2.97	7	2
fluometuron	2164-17-2	H	4.10	(2.52−6.65)	4.93	6	–
fluopicolide	239110-15-7	F	8.16	(1.91−34.94)	5.62	2	–
fluopyram	658066-35-4	F	5.51	(1.29−23.61)	5.55	2	–
fluoroxypyr	69377-81-7	H	6.08	(2.93−12.61)	4.99	4	–
fluoroxypyr-meptyl	81406-37-3	H	3.13	(1.19−8.27)	4.96	3	–
flurprimidol	56425-91-3	P	11.60	(4.40−30.56)	8.30	3	–
flusilazole	85509-19-9	F	4.89	(2.73−8.75)	5.35	5	–
fluvalinate	69409-94-5	I	2.92	(1.80−4.74)	2.86	6	–
folpet	133-07-3	F	5.19	(4.45−6.07)	1.14	19	17
forchlorfenuron	68157-60-8	P	3.28	(2.52−4.27)	11.40	11	4
formothion	2540-82-1	I	1.80	(0.42−7.69)	7.46	2	–
glyphosate	1071-83-6	H	3.98	(3.28−4.84)	5.24	15	–
halofenozide	112226-61-6	I	9.24	(3.51−24.36)	6.96	3	–
halosulfuron-methyl	100784-20-1	H	0.82	(0.19−3.52)	4.61	2	–
haloxyfop-P-methyl	72619-32-0	H	5.98	(0.33−109.44)	0.97	1	–
HCH	608-73-1	I	6.86	(5.13−9.18)	3.69	10	–
heptachlor	76-44-8	I	3.34	(2.49−4.47)	4.01	10	1
hexachlorobenzene	118-74-1	F	11.15	(7.73−16.10)	4.44	8	–
hexaconazole	79983-71-4	F	3.96	(3.45−4.56)	2.84	21	–
hexaflumuron	86479-06-3	I	4.08	(2.69−6.19)	1.36	7	4
hexazinone	51235-04-2	H	8.90	(4.30−18.40)	4.32	4	–
hymexazol	10004-44-1	F	3.56	(1.72−7.37)	2.42	4	–
imazalil	35554-44-0	F	5.72	(3.50−9.33)	0.71	6	6
imazapyr	81334-34-1	H	23.86	(5.58−102.09)	10.98	2	–

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00008

Table 1. continued

pesticide	CAS RN	TC[a]	$HL^{\bullet}_{ref,i}$ (95% CI) (day)		θ_{imp}(%)	n_{HL}	n_T
imazaquin	81335-37-7	H	10.72	(7.07−16.25)	5.83	7	–
imazethapyr	81335-77-5	H	15.51	(10.23−23.52)	2.10	7	5
imidacloprid	138261-41-3	I	3.70	(3.61−3.78)	1.62	130	16
imidaclothiz	105843-36-5	I	5.14	(1.95−13.56)	2.69	3	2
indoxacarb	173584-44-6	I	2.99	(2.35−3.82)	7.16	12	4
iodosulfuron-methyl-sodium	144550-36-7	H	5.49	(1.28−23.55)	8.88	2	–
iprodione	36734-19-7	F	6.92	(6.35−7.53)	5.37	34	6
iprovalicarb	140923-17-7	F	9.47	(5.83−15.38)	5.12	6	–
isazofos	42509-80-8	I	6.58	(4.58−9.47)	6.30	8	–
isocarbophos	24353-61-5	A	2.45	(1.92−3.12)	13.18	12	–
isofenphos	25311-71-1	I	3.75	(1.81−7.76)	3.96	4	–
isoproturon	34123-59-6	H	5.40	(2.04−14.29)	4.41	3	–
isoxaben	82558-50-7	H	5.92	(2.86−12.25)	8.58	4	–
jasmolin II	1172-63-0	I	3.51	(0.82−15.05)	0.09	2	2
kresoxim-methyl	143390-89-0	F	7.33	(5.90−9.11)	2.51	13	1
lambda-cyhalothrin	91465-08-6	I	2.86	(2.60−3.14)	11.75	30	6
leptophos	21609-90-5	I	6.78	(5.51−8.35)	5.43	14	–
lindane	58-89-9	I	3.68	(3.24−4.18)	5.27	23	5
lufenuron	103055-07-8	I	8.46	(6.31−11.34)	11.61	9	–
malathion	121-75-5	I	2.48	(2.40−2.56)	3.65	92	23
mancozeb	8018-01-7	F	4.69	(4.29−5.13)	2.80	33	12
mandipropamid	374726-62-2	F	3.46	(1.31−9.12)	8.33	3	–
maneb	12427-38-2	F	4.55	(3.57−5.80)	8.63	12	–
MCPA	94-74-6	H	3.89	(2.97−5.10)	5.49	11	–
mecarbam	2595-54-2	I	3.34	(2.61−4.27)	5.21	12	–
mecoprop	7085-19-0	H	3.89	(1.88−8.05)	9.43	4	–
mepanipyrim	110235-47-7	F	6.07	(2.30−15.99)	6.31	3	1
mepronil	55814-41-0	F	2.60	(1.45−4.66)	2.98	5	–
meptyldinocap	131-72-6	F	2.93	(2.02−4.24)	1.28	8	8
mesotrione	104206-82-8	H	1.13	(0.26−4.83)	6.15	2	–
metalaxyl	57837-19-1	F	5.81	(5.09−6.63)	5.14	22	5
metalaxyl-M	70630-17-0	F	3.74	(3.12−4.49)	7.75	16	2
metaldehyde	108-62-3	M	2.30	(1.52−3.50)	5.93	7	3
methamidophos	10265-92-6	I	5.28	(5.01−5.57)	5.19	55	18
methidathion	950-37-8	I	2.86	(2.43−3.37)	3.61	18	5
methiocarb	2032-65-7	I	3.68	(2.99−4.54)	1.08	14	11
methomyl	16752-77-5	I	2.04	(1.98−2.10)	5.92	122	19
methoprene	40596-69-8	I	1.88	(0.91−3.88)	4.36	4	–
methoxychlor	72-43-5	I	3.20	(0.75−13.67)	2.01	2	1
methoxyfenozide	161050-58-4	I	6.86	(1.60−29.36)	3.57	2	–
metolachlor	51218-45-2	H	19.91	(12.25−32.38)	2.14	6	4
metsulfuron-methyl	74223-64-6	H	5.75	(2.18−15.18)	5.53	3	–
mevinphos	7786-34-7	I	1.01	(0.78−1.32)	5.53	11	–
mexacarbate	315-18-4	I	1.58	(1.44−1.73)	9.08	33	–
milbemectin	51596-10-2	A	12.05	(5.82−24.94)	8.30	4	–
monocrotophos	6923-22-4	I	3.47	(3.00−4.01)	1.93	20	5
monosodium methylarsonate	2163-80-6	H	2.98	(2.07−4.29)	0.38	8	8
myclobutanil	88671-89-0	F	4.35	(4.05−4.68)	3.65	42	16
nicosulfuron	111991-09-4	H	0.64	(0.15−2.75)	11.59	2	–
nicotine	54-11-5	I	2.51	(1.88−3.36)	16.51	10	–
norflurazon	27314-13-2	H	7.91	(5.22−11.98)	3.99	7	–
nornicotine	494-97-3	I	2.19	(0.83−5.77)	6.05	3	–
novaluron	116714-46-6	I	2.15	(1.69−2.74)	4.07	12	–
oxadiazon	19666-30-9	H	2.45	(1.18−5.06)	3.40	4	–
oxamyl	23135-22-0	I	3.83	(1.45−10.09)	2.09	3	2
oxaziclomefone	153197-14-9	H	4.82	(1.13−20.65)	4.70	2	–
oxidemeton-methyl	301-12-2	I	4.43	(0.24−81.20)	6.00	1	–
oxyfluorfen	42874-03-3	H	1.27	(0.61−2.63)	8.09	4	–
paclobutrazol	76738-62-0	P	1.63	(1.21−2.18)	3.69	10	–
parathion	56-38-2	I	2.81	(2.73−2.91)	5.11	96	28
parathion-methyl	298-00-0	I	1.61	(1.51−1.72)	2.33	46	29

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00009

Table 1. continued

pesticide	CAS RN	TC[a]	$HL^{\bullet}_{ref,i}$ (95% CI) (day)		θ_{imp}(%)	n_{HL}	n_T
penconazole	66246-88-6	F	8.06	(6.44−10.08)	2.60	13	2
pencycuron	66063-05-6	F	2.91	(1.79−4.74)	0.38	6	6
pendimethalin	40487-42-1	H	6.73	(5.54−8.18)	4.65	15	−
penoxsulam	219714-96-2	H	1.56	(0.59−4.12)	0.15	3	3
pentachloroaniline	527-20-8	M	18.06	(11.10−29.40)	12.26	6	−
permethrin	52645-53-1	I	5.84	(5.20−6.57)	3.56	25	11
phenazine-1-carboxylic acid	2538-68-3	B	4.12	(1.99−8.52)	7.83	4	−
phenothrin	26002-80-2	I	4.93	(2.75−8.84)	3.40	5	2
phenthoate	2597-03-7	I	4.13	(2.99−5.72)	1.51	9	2
phorate	298-02-2	I	6.51	(4.53−9.36)	5.80	8	−
phosalone	2310-17-0	I	4.72	(3.62−6.15)	6.22	11	1
phosmet	732-11-6	I	2.10	(1.68−2.63)	4.87	13	−
phosphamidon	13171-21-6	I	3.96	(2.86−5.47)	6.06	9	−
phoxim	14816-18-3	I	1.61	(1.06−2.44)	9.08	7	−
picloram	1918-02-1	H	6.97	(3.36−14.46)	5.97	4	2
piperonyl butoxide	51-03-6	S	10.42	(5.82−18.67)	0.51	5	5
pirimicarb	23103-98-2	I	4.55	(3.90−5.31)	3.23	18	4
pirimiphos-methyl	29232-93-7	I	2.16	(1.87−2.51)	5.21	19	−
probenazole	27605-76-1	F	2.39	(0.56−10.22)	7.59	2	−
prochloraz	67747-09-5	F	5.81	(4.04−8.36)	4.39	8	3
procymidone	32809-16-8	F	9.89	(8.03−12.19)	7.81	14	6
profenofos	41198-08-7	I	2.24	(2.12−2.38)	1.84	51	3
propamocarb hydrochloride	25606-41-1	F	1.10	(0.06−20.06)	0.04	1	1
propargite	2312-35-8	A	3.77	(2.73−5.21)	17.35	9	−
propiconazole	60207-90-1	F	5.44	(4.75−6.22)	11.22	22	−
propineb	12071-83-9	F	3.50	(3.17−3.87)	6.87	30	4
Propisochlor	86763-47-5	H	2.95	(1.82−4.79)	10.69	6	−
propoxur	114-26-1	I	0.83	(0.31−2.19)	4.45	3	−
pymetrozine	123312-89-0	I	2.94	(2.19−3.93)	4.53	10	1
pyraclostrobin	175013-18-0	F	3.90	(1.88−8.07)	3.22	4	1
pyrazophos	13457-18-6	F	4.65	(3.37−6.43)	4.83	9	4
pyrethrin I	121-21-1	I	1.06	(0.59−1.89)	0.51	5	5
pyrethrin II	121-29-9	I	2.20	(1.23−3.94)	0.51	5	5
pyrethrins	8003-34-7	I	0.21	(0.10−0.44)	4.51	4	2
pyribenzoxim	168088-61-7	H	5.92	(2.24−15.64)	8.69	3	−
pyridaben	96489-71-3	I	4.84	(3.98−5.88)	1.70	15	7
pyridafenthion	119-12-0	I	8.06	(3.05−21.29)	0.20	3	3
Pyrimethanil	53112-28-0	F	5.35	(4.62−6.19)	5.59	20	2
pyriproxyfen[b]	95737-68-1	I	27.55	(16.93−44.82)	3.42	6	4
pyroxsulam	422556-08-9	H	1.03	(0.64−1.68)	6.35	6	−
quinalphos	13593-03-8	I	2.48	(2.19−2.82)	9.02	23	1
quinclorac	84087-01-4	H	3.42	(1.91−6.13)	5.79	5	−
quinoxyfen	124495-18-7	F	6.01	(0.33−110.11)	0.06	1	1
quintozene	82-68-8	F	14.74	(11.72−18.54)	7.85	13	−
quizalofop-ethyl	76578-14-8	H	1.30	(0.63−2.69)	0.89	4	4
quizalofop-P-tefuryl	119738-06-6	H	0.92	(0.44−1.90)	5.02	4	−
rimsulfuron	122931-48-0	H	0.75	(0.50−1.14)	9.64	7	−
rotenone	83-79-4	I	2.06	(1.27−3.36)	3.14	6	2
schradan	152-16-9	I	7.90	(3.82−16.35)	5.35	4	−
s-metolachlor	87392-12-9	H	9.63	(5.93−15.64)	9.09	6	−
spinosad	168316-95-8	I	4.42	(3.94−4.97)	5.63	24	4
spiromesifen	283594-90-1	I	2.75	(2.41−3.14)	2.71	22	4
spiroxamine	118134-30-8	F	10.20	(5.70−18.25)	0.89	5	4
sulfometuron-methyl	74222-97-2	H	6.18	(1.44−26.43)	4.79	2	−
sulfosulfuron	141776-32-1	H	4.42	(2.13−9.18)	1.05	4	4
sulfotep	3689-24-5	I	1.67	(0.39−7.17)	4.72	2	−
sulprofos	35400-43-2	I	1.30	(0.80−2.12)	1.62	6	5
Tau-fluvalinate	102851-06-9	I	1.87	(1.23−2.84)	0.45	7	7
tebuconazole	107534-96-3	F	7.67	(6.46−9.11)	4.23	17	5
tebufenozide	112410-23-8	I	17.73	(15.79−19.90)	1.91	26	24
tebufenpyrad	119168-77-3	A	5.09	(2.46−10.54)	3.36	4	2

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00010

Table 1. continued

pesticide	CAS RN	TC[a]	$HL^{\bullet}_{\mathrm{ref},i}$ (95% CI) (day)		θ_{imp}(%)	n_{HL}	n_T
teflubenzuron	83121-18-0	I	21.07	(10.19−43.60)	4.30	4	2
tepraloxydim	149979-41-9	H	3.43	(0.80−14.69)	11.74	2	–
tetraconazole	112281-77-3	F	5.27	(4.52−6.15)	3.31	19	7
tetradifon	116-29-0	A	6.13	(2.96−12.69)	3.45	4	–
thiacloprid	111988-49-9	I	3.76	(3.43−4.12)	3.71	32	7
thiamethoxam	153719-23-4	I	3.97	(2.76−5.72)	13.80	8	–
thidiazuron	51707-55-2	P	4.17	(0.98−17.87)	4.57	2	–
thifensulfuron-methyl	79277-27-3	H	11.11	(5.37−22.97)	4.89	4	–
thiophanate-methyl	23564-05-8	F	9.07	(7.25−11.35)	7.05	13	–
thiram	137-26-8	F	4.70	(3.10−7.12)	7.48	7	–
tolclofos-methyl	57018-04-9	F	1.81	(1.54−2.13)	3.59	18	–
tolylfluanid	731-27-1	F	6.85	(3.30−14.20)	0.53	4	4
toxaphene	8001-35-2	I	6.81	(5.53−8.39)	4.49	14	5
tralkoxydim	87820-88-0	H	6.34	(1.48−27.20)	8.65	2	–
tralocythrin	66841-26-7	I	1.64	(0.62−4.33)	9.71	3	–
tralomethrin	66841-25-6	I	5.37	(3.73−7.73)	8.68	8	4
triadimefon	43121-43-3	F	4.41	(3.99−4.88)	2.02	29	4
triadimenol	55219-65-3	F	10.42	(2.43−44.66)	0.28	2	2
triazophos	24017-47-8	I	6.05	(5.29−6.92)	7.00	22	–
tribufos	78-48-8	H	6.07	(4.00−9.20)	2.82	7	–
trichlorfon	52-68-6	I	2.71	(2.36−3.10)	1.65	21	2
triclopyr	55335-06-3	H	2.54	(1.23−5.26)	2.69	4	–
tricyclazole	41814-78-2	F	10.63	(4.03−28.06)	4.82	3	–
trifloxystrobin	141517-21-7	F	3.72	(1.80−7.70)	7.01	4	1
triflumuron[b]	64628-44-0	I	10.10	(7.27−14.03)	2.23	9	6
trifluralin	1582-09-8	H	2.66	(1.85−3.82)	3.12	8	–
triflusulfuron	135990-29-3	H	0.69	(0.39−1.24)	5.91	5	–
triforine	26644-46-2	F	4.36	(2.44−7.80)	2.46	5	–
vinclozolin	50471-44-8	F	3.03	(2.75−3.34)	3.21	31	26
zineb	12122-67-7	F	4.25	(3.72−4.85)	3.27	22	1
ziram	137-30-4	F	10.40	(0.57−190.43)	0.05	1	1

[a]Notation: A, acaricide; B, bactericide; F, fungicide; H, herbicide; I, insecticide; M, metabolite; N, nematicide; P, plant growth regulator; S, pesticide synergist. [b]Estimated half-lives differ > 10% when excluding cold storage data; half-lives without cold storage data are given in Table S4 (SI).

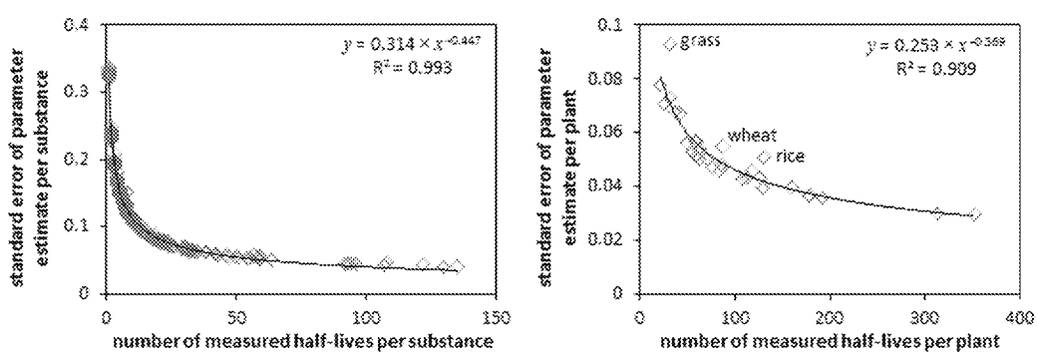

Figure 4. Standard errors of parameter estimates per substance (left) and plant (right) as a function of the number of reported dissipation half-lives for 333 pesticides and 30 plants (Model II).

squared error RMSE = 0.36; that is, 95% of predicted half-lives fall within a factor 4.5 of reported half-lives. From Model III we obtain estimates to correct for cold storage conditions, $\beta'^{\circ}_{\mathrm{storage}} = 0.69$, substance molecular weight, $\beta'^{\circ}_{MW} = -3.9 \times 10^{-4}$ mol/g, log K_{OW}, $\beta'^{\circ}_{K_{\mathrm{OW}}} = 0.02$, and saturation vapor pressure, $\beta'^{\circ}_{P_{\mathrm{V}}} = -0.09\ \mathrm{Pa}^{-1}$, (all p-values < 0.05). Estimates for 14 substance classes and 31 plants are given in Table 2 and resulting predicted half-lives plotted against reported half-lives are shown in Figure 3 (Model III).

Model III is designed to calculate from pesticide properties half-lives for individual pesticides, for which no reference half-lives are available (i.e., not in Table 1). This model targets all pesticides of substance classes listed in Table 2 and applied to specific plants under given conditions and temperatures. For example, except for some unpublished residue studies,[38] there is no reported dissipation half-life available for tefluthrin (CAS, 79538-32-2; substance class, pyrethroids) authorized in the European Union.[39] To calculate its half-life when applied to, for example, wheat at 295.16 K (22 °C) under field conditions, we use eq 7 with estimates for study condition, substance class, substance-specific properties, and plant taken from Table 2 along with β'°_{T}. We get log $HL^{\circ}_{\mathrm{predicted},T} = \alpha'^{\circ} + \beta'^{\circ}_{\mathrm{pyrethroids}} +$

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00011

Table 2. Predictor Variables with Associated Parameter Estimates (Intercept, α'°; Air Temperature, β'°_{T} (K^{-1}); Cold Storage Conditions, $\beta'^{\circ}_{storage}$; Substance Class, $\beta'^{\circ}_{subst\text{-}class,k}$; molecular weight, β'°_{MW} (mol/g); log K_{OW}, $\beta'^{\circ}_{K_{OW}}$; saturation vapor pressure, $\beta'^{\circ}_{P_V}$ (Pa^{-1}); and Plant, $\beta'^{\circ}_{plant,j}$; in Line with eq 7), Standard Errors SE, Numbers of Data Points with Reported Average Air Temperatures n_T for Substance Classes and Plants, and p-Values (Variable Significant, if p-Value <0.05) as Used in the Final Model to Estimate Dissipation Half-Lives for 14 Substance Classes Applied to 31 Plants (Model III)

symbol	variable name	parameter estimate	SE	n_T	p-value
α'°	intercept	1.35	0.11		<0.0001
β'°_{T}	air temperature	-1.995×10^{-2}	3.1×10^{-3}		<0.0001
$\beta'^{\circ}_{storage}$	study conditions (cold storage)	0.69	0.09		<0.0001
$\beta'^{\circ}_{subst\text{-}class}$	substance class: anilinopyrimidines	0.18	0.12	16	0.5005
	substance class: benzoylureas	0.07	0.11	27	0.0818
	substance class: botanical insecticides	−0.24	0.12	22	<0.0001
	substance class: carbamates	−0.19	0.08	66	<0.0001
	substance class: dithiocarbamates	0.03	0.11	19	0.0439
	substance class: imidazoles	0.30	0.14	10	0.7134
	substance class: morpholines	0.16	0.16	7	0.5403
	substance class: neonicotinoids	0.09	0.10	39	0.0954
	substance class: organochlorines	−0.05	0.08	65	0.0003
	substance class: organophosphates	−0.08	0.07	260	<0.0001
	substance class: pyrethroids	−0.08	0.08	158	<0.0001
	substance class: strobilurins	0.20	0.15	8	0.7262
	substance class: triazoles	0.25	0.07	43	<0.0001
	substance class: other substance classes[a]	0.09	0.07	290	0.0142
β'°_{MW}	molecular weight	-3.9×10^{-4}	1.7×10^{-4}		0.0228
$\beta'^{\circ}_{K_{OW}}$	log *Kow*	0.02	0.01		0.0127
$\beta'^{\circ}_{P_V}$	saturation vapor pressure	−0.09	0.03		0.0083
β'°_{plant}	plant: alfalfa	−0.04	0.10	19	0.6134
	plant: apple	0.07	0.08	29	0.4927
	plant: cabbage	−0.07	0.09	28	0.3172
	plant: cauliflower	0.04	0.13	9	0.8642
	plant: Chinese cabbage	−0.001	0.08	31	0.8593
	plant: cotton	−0.20	0.06	94	0.0006
	plant: cucumber	−0.06	0.13	10	0.5778
	plant: eggplant	0.07	0.07	42	0.4728
	plant: elm	0.20	0.09	32	0.0299
	plant: grape	0.13	0.08	108	0.0002
	plant: grass	0.27	0.13	10	0.0417
	plant: green bean	−0.25	0.09	22	0.0039
	plant: lemon	0.26	0.12	11	0.0397
	plant: lettuce	−0.33	0.12	12	0.0033
	plant: mango	−0.22	0.12	12	0.0455
	plant: okra	0.10	0.12	12	0.4526
	plant: olive	0.13	0.11	13	0.3232
	plant: onion	−0.53	0.12	11	<0.0001
	plant: orange	0.23	0.08	32	0.0075
	plant: peach	0.29	0.09	31	0.0015
	plant: pear	0.23	0.11	16	0.0363
	plant: pepper fruit	0.04	0.07	55	0.6328
	plant: potato	−0.23	0.15	8	0.1004
	plant: rice	0.12	0.07	36	0.1543
	plant: strawberry	−0.004	0.08	34	0.8263
	plant: tea	0.06	0.09	26	0.6191
	plant: tomato	−0.03	0.08	36	0.5345
	plant: wheat	−0.18	0.12	12	0.1096
	plant: white spruce	0.55	0.09	27	<0.0001
	plant: zucchini	−0.81	0.12	12	<0.0001
	plant: other plants[b]	−0.04	0.05	200	0.3128

[a]Substance classes with $n < 3$ reported substances per class. [b]Plant species with $n < 8$ reported average air temperatures.

CBD v. EPA (1:21-cv-00681-CJN) ED_005427A_00029970-00012

$\beta'^{\circ}_{\text{wheat}} + \beta'^{\circ}_{T} \times (T - T_{\text{ref}}) + \beta'^{\circ}_{MW} \times MW + \beta'^{\circ}_{K_{\text{OW}}} \times \log K_{\text{OW}} + \beta'^{\circ}_{P_{\text{V}}} \times P_{\text{V}} = 1.35 - 0.08 - 0.18 - 1.995 \times 10^{-2}\ \text{K}^{-1} \times (195.16\ \text{K} - 193.16\ \text{K}) - 3.9 \times 10^{-4}\ \text{mol/g} \times 418.7\ \text{g/mol} + 0.02 \times 6.4 - 0.09\ \text{Pa}^{-1} \times 8.4 \times 10^{-3}\ \text{Pa} = 1.014$, and thus a predicted half-life for tefluthrin dissipation from wheat at 22 °C of $10^{1.014}$ = 10.3 days. MW, K_{OW}, and P_{V} are taken from EFSA.[38] Half-life corrections from substance class parameter estimates of Model III vary between $10^{-0.24}$ = 0.58 days for botanical insecticides (e.g., pyrethrins, nicotine) and $10^{-0.19}$ = 0.65 days for carbamates (e.g., methomyl, pirimicarb) at the low end and $10^{0.25}$ = 1.78 days for triazoles (e.g., myclobutanil, difenoconazole) and $10^{0.3}$ = 2 days for imidazoles (e.g., prochloraz, imazalil) at the high end. Half-life corrections from plant parameter estimates vary between $10^{-0.81}$ = 0.15 days for zucchini and $10^{0.55}$ = 3.55 days for white spruce. For 8 out of 14 substance classes and for 14 out of 31 plants, p-values < 0.05.

4. DISCUSSION

4.1. Applicability and Limitations. We developed from Model II, which is controlled for plants, conditions, and temperature, a set of 333 comparative pesticide dissipation half-lives in plants under reference conditions represented by the average of 30 plant-specific parameter estimates, field conditions, and an air temperature of 20 °C (Table 1). For arriving at this set, just taking the geometric mean of all available measured half-lives per substance[11] would have led to biased estimates. Instead, correcting predicted half-lives for temperature, plant species, and study conditions, imputing missing temperatures, identifying a temperature coefficient in Model I, and finally studying the variability for each pesticide reduces the uncertainty around each presented half-life and much better reflects reference conditions. Half-lives in Table 1 are designed for direct use in risk and impact assessment models and can be combined with eq 5 to further correct for scenario-specific plants, conditions, and temperatures. Our final Model III (eq 7) is designed to estimate dissipation half-lives from properties of individual pesticides of 14 substance classes, for which no reference half-lives could be obtained in our study due to missing reported data.

We acknowledge that the predictive ability of leave-one-out cross validation can be too optimistic, since data perturbation may be insignificant when only omitting one data point.[40] Substance classes with less than three reported substances per class and plants with less than eight reported air temperatures were pooled into single categories *other substance classes* and *other plants*, respectively (see Table 2). Comparatively small standard errors for these two categories refer to the mean category estimate, whereas for each substance class and plant within these categories the standard error is generally higher. This is because these two categories are much more heterogeneous than categories for individual substance classes and plants (increases uncertainty per category component), but also contain more data points than individual substance classes and plants (decreases uncertainty of mean category estimate). Hence, half-lives based on these two categories generally have higher uncertainty than half-lives based on individual substance classes and plants.

4.2. Comparison with Other Temperature Coefficients. Temperature coefficients Q_{10} for metabolic processes in plants and other media are typically between 2 and 3,[41,42] while Q_{10} for physical processes are often much lower.[42−45] Our Q_{10} = 1.22 is at the very low end of Q_{10} for individual processes. This can be explained by the fact that there is more than one process contributing to overall dissipation from plants and that these processes go in a counter-direction. More specifically, overall dissipation results from the combination of temperature-dependent degradation as predominant removal process in plants as well as leaf and root uptake as temperature-dependent processes counter-acting removal from plants.[6,8] The combined effects of these processes with their distinct influence on residue dynamics[3,30,31] reduce the temperature-dependency of overall dissipation. This is in line with other studies, where a change from high to low Q_{10} has been observed when going from pure degradation to overall plant dissipation.[46,47]

4.3. Future Research Needs. Risk and impact assessment models often need information on individual processes contributing to dissipation. For that, additional research is required to systematically assess the contribution of processes like degradation or root uptake to overall pesticide dissipation from plants. Furthermore, reporting guidelines for measuring dissipation from plants need to be improved with respect to providing sufficient information on environmental study conditions (most importantly by reporting temperature, but also humidity, soil type, etc.) and residues in plants (providing enough data points to account for measurement variability and to effectively perform curve fitting for estimating dissipation kinetics). Finally, the specific and significant coefficient obtained for cold storage primarily shows that the temperature correction obtained on field crops cannot be directly applied for cold storage conditions. Targeted experiments are required to further analyze the influence of temperature and the characteristics of cold storage conditions on dissipation and contributing mechanisms, thereby increasing input data quality when using experimentally derived half-lives in modeling studies.

ASSOCIATED CONTENT

Supporting Information

Background material describes coefficients for temperature correction, optimal air temperature ranges for plants and statistics of our regression models. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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